SPATIAL AND TEMPORAL VARIATIONS OF RADON AND RADON DAUGHTER CONCENTRATIONS WITHIN LIMESTONE CAVES

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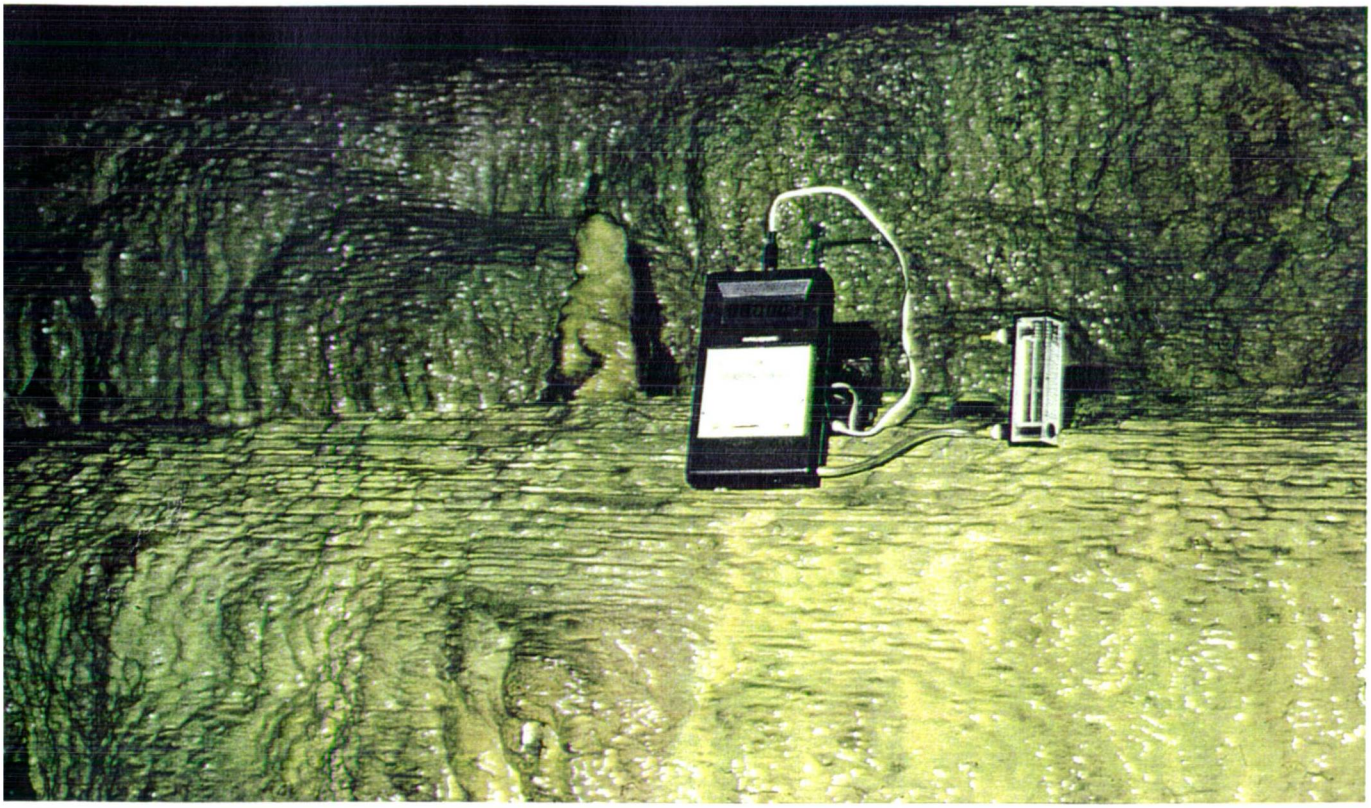
Frontispiece



Radon daughter measurements being undertaken in White Scar caves

ABSTRACT

This thesis outlines results from an investigation of radon and radon daughter concentrations in limestone caves, from a geographical and geological perspective Investigations were conducted at all geographical scales, ranging from a national investigation in the four major caving regions of England and Wales to a detailed survey within a single cave in the Peak District

The measured radon concentrations in some limestone caves in England and Wales are amongst the highest ever recorded in the world Significant spatial and temporal variations were recorded in concentrations at all scales, within a single cave, between caves in the same region and between different regions Additionally, seasonal and diurnal variations in concentrations were highlighted. External climatic variables and the cave radon budget were demonstrated to account for variations in cave radon concentrations

Within limestone caves seven primary sources of radon were identified and the relative importance of each to the overall radon budget was determined Sediments and the containing limestone rock were the major sources although in certain cases water and the soil were demonstrated to be influential

Models were developed to predict cave radon concentrations within a single cave However, these could not be transposed to predict radon concentrations in other caves in the same region or other regions

The users of limestone caves were identified, their potential exposure times were examined and legislation concerning their exposure was discussed Four groups were identified as being at risk from radon while underground, and three groups were identified as being at little risk Methods by which the risk from radon exposure can be reduced were examined

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CHAPTER ONE: INTRODUCTION

1.1 Background to the thesis

1 1 1 Project Definition

This thesis aims to investigate a radiochemical phenomenon, namely the existence of radon gas and its decay products in karst cave environments, from a geographical and geological perspective The geographical focus will examine and determine the controls on spatial and temporal variations of radon-222 and radon daughter concentrations Spatial variations will be considered at three levels within a single cave; between caves within the same geographical area, and between caves of different geographical regions Temporal variations will be examined on hourly, daily, monthly and annual timescales. The geological perspective will examine the mechanisms of, and controls on, radon production Two distinct but dependent processes can be identified firstly, radon emanation which is the release and migration of radon from production sites in the host material to internal voids, and secondly, radon exhalation, the movement of radon from pore spaces into the external atmosphere

1 1 2 Impetus for the thesis

Public awareness of hazards from radionuclides within everyday life and the wider terrestrial environment has increased in recent years This awareness promoted a study of radon gas in caves, and Breisch (1968) is often cited as the first study However, his work was preceded by an earlier thesis, by Fryer (1935) and a paper by Harris (1954) Breisch (1968) was followed by Ronaki (1972), Varbanov *et al* (1975), Trout (1975) and Van Cleave (1975) Trout's and Van Cleave's papers sparked interest in the United States National Park Service who sponsored projects by Yarborough *et al* (1976), Wilkening and Watkins (1976), and Beckman (1977, 1978) The initial research which determined the atmospheric concentrations of both radon-222 gas and radon daughters,

led to discussion of the associated health risks to cave users The majority of workers, from around the world, (e g Ahlstrand, 1976, 1980, Wilkening and Watkins, 1976, Yarborough, 1977, Yarborough and Ahlstrand, 1977, Seymore *et al*, 1980) have argued strongly that a health risk is evident and that people should be informed, but a minority (e g Cigna, 1986, Glanvill, 1989) suggest that there is little risk and that no further action should be taken During the initial period of research in America, a few measurements were being conducted in British caves, (primarily Show Caves) but no data were published

The impetus for further research in British caves came with the introduction of the Ionising Radiation's Regulations (IRR's) in 1985 These established legal limits on the levels to which workers in radioactive environments could be exposed The IRR's cover all forms of radiation, not just radon, and define three categories with respect to radon levels in the work place (see chapter two for definition of units)

1 Where concentrations in the work place are below 0 03 WL (Working Level, see section 2 4 3 for a definition of units) regular measurements are not required

2 Where concentrations are between 0 03 WL and 0 1 WL, the area must be designated a 'supervised area', and monitoring conducted to ensure that the average concentration over an 8 hour period does not rise above 0 1 WL

3 'Controlled areas' are designated if concentrations rise above an average of 0 1 WL for an 8 hour period during any part of the year In these, regular monitoring has to be undertaken

The IRR's also state that all work places where 'supervised area' or 'controlled area' status applies need to appoint a Radiological Protection Supervisor and a Radiological Protection Adviser Any workers entering these environments must be monitored and operate under an approved system of work (the IRR's will be covered in more detail in chapter ten)

Gunn (pers comm, 1993) has acted as Radiological Protection Adviser for several Show Caves across the country and as all were found to be 'controlled areas', (i.e above 0 1 WL on average over an eight hour period during the working day) at least for part of the year, regular monitoring was undertaken from 1989 onwards During this period measurements were also conducted in 'wild caves' and concentrations of radon and radon daughters were found to be higher than those in other countries (see chapter three). It was felt by Gunn and by the National Caving Association (NCA) that there was a potential health risk to cave users in Britain As a result, the study was widened and a Working Party established with the aim of gathering more information Measurements were made in both 'wild caves' and Show Caves in all areas of Britain, particularly in the Peak District where a research project was funded by Derbyshire County Council (see chapter eight) Concern was expressed regarding the well-being of some Show Cave guides and of children who enter the high radiation environments encountered in some caves Show Cave guides are particularly at risk due to the long periods of time that they spend underground, while children are thought to be more susceptible to radiobiological damage than older members of the population due to immaturity of their lungs (Cigna, 1987b, Cigna, 1989)

Through the work in Britain of Friederich (1981), Gunn *et al* (1989a and b, 1991), Mutter, (1987), Middleton (1988) and Williamson (1990), the fundamentals of the radon system became established, the conclusions being similar to those drawn in America during the late 1970's and early 1980's (see chapter three) Work by Strong *et al* (1975) demonstrated that radon concentrations in mine environments within limestone strata were higher than in mines in different geological settings, but the discussion did not account for the differences observed

Useful though this work was, it became clear that a more extensive and integrated investigation was necessary Hence, in October 1990 the author commenced the present study Subsequently, in October 1992, Workman commenced a Doctoral investigation of cave radon and radon daughter concentrations specifically in the North Pennines region Both projects involve integrated research where geographical, geological, sociological

and health perspectives are being investigated It is hoped that the present thesis will enhance knowledge of radon in caves and will be of benefit to all who enter these environments

1.2 Aims and Objectives

This investigation of radon concentrations within limestone caves in England and Wales is the largest carried out to date The main aim was to establish and account for spatial and temporal variations in radon concentrations within limestone caves The primary objectives were

1 To compare and contrast different methods for determining radon-222 and radon daughter concentrations within limestone caves From this, to develop appropriate measuring techniques

2 To determine the typical radon-222 concentrations within the main caving areas of England and Wales

3 To evaluate the controls on radon-222 concentrations within a single caving region

4 To determine the controls on radon-222 and radon daughter concentrations within a single cave environment

5 To quantify the extent of seasonal variations in cave radon gas concentrations

6 To evaluate the effects of cave morphology and geological setting on radon gas concentrations

7 To identify the relative importance of different sources of radon-222 to cave environments

8 To quantify the risks to cavers from radon gas

1.3 Environmental radiation

1 3 1 Sources of radiation

In the natural environment many sources of radiation exist Natural background radiation comes from six sources (figure 1 1) Radon-222 represents over 49 % of the background radiation dose, 13 % is derived from artificial radiation, primarily weapons testing, but also from medical uses including X-rays The other sources of natural background radiation dose include cosmic rays (9 %), radon-220 (thoron) (4 %), internal (11 %), and terrestrial gammas' (14 %) (NCRP, 1988) The dominance of radon-222 in the background radiation dose results from concentrations of around 50 Bq m^{-3} in the natural atmosphere (Hotzl and Winkler, 1987) When these are compared to concentrations in excess of 15,000 Bq m^{-3} (4 1 WL) which are regularly encountered in some British caves, the potential health hazard becomes obvious (see section 1 3 3 for a description of the health risks)

1 3 2 Radon isotopes and the environmental significance of Radon-222

Three isotopes of radon exist in the natural environment radon-222, produced within the decay series of uranium-238, radon-220 (thoron) formed as a product of the thorium-232 decay series and radon-219 formed from the uranium-235 decay series (figures 1 2, 1 3 and 1 4 respectively) This research is concerned primarily with radon-222 and the term radon will refer solely to radon-222, the other radon isotopes being differentiated by normal chemical notation where necessary

The focusing of research on to radon-222 and the relative exclusion of radon-220 (thoron) and radon-219 can be justified as the emanation / exhalation rate of the three radon isotopes varies within the natural environment, resulting in different environmental significance for radon-222, 220, and 219 Release rates of radon-220 (thoron) and radon-219 are generally low, reducing their importance within both the natural

environment and in cave atmospheres, while radon-222 has a relatively high release potential. The low release rates of radon-220 (thoron) and radon-219 are related to firstly, the depleted initial concentrations of the parent radionuclides (uranium-235 and thorium-232 respectively) within the Earth's crust, and secondly the short half lives of the radon isotopes (radon-220 and -219 have half lives of 55 seconds and 4 0 seconds respectively) The initial concentrations control the net production rate (Ball *et al*, 1991), while the half life affects the subsequent movement from production sites to the terrestrial environment (Semkow *et al*, 1991) The half life controls the time in which the radon isotopes can move from production sites to the atmosphere before decay occurs The longer the half life of the isotope involved, the greater the probability of release before decay (Ball *et al*, 1991) Thus, the production of radon-222 is greater than the other radon isotopes due to its relatively high initial concentration and its half life of 3 82 days

1 3 3 Definition and characterisation of radon and radon daughters

Radon is a colourless, odourless, and inert radioactive gas, produced within the uranium-238 decay series (NCRP, 1988)(figure 1 2) Uranium-238 is a ubiquitously distributed element within the earth's crust (Ivanovich and Harmon, 1982, p 56) The migration of radon involves two dependent but separate processes

1 emanation, which is the movement of radon from production sites in the mineral grains to the pore spaces (Jonassen, 1981, 1983, Semkow, 1990),

2 exhalation that is the movement from pore spaces to the terrestrial environment (Tanner, 1978, Semkow and Parekh, 1990)

Once in the atmosphere, radon decays releasing alpha particles with a half life of 3 82 days (figure 1 2) Upon decay, radon produces the four radon daughters polonium-218, lead-214, bismuth-214 and polonium-214 The daughters are solid radionuclides with short half-lives ranging from 26 8 minutes to 1 6 x 10^{-4} seconds (NCRP, 1988, p 16) Upon decay these emit a combination of alpha, beta and gamma emissions (figure 1 2)

The daughters are charged solid particles and not gaseous, which results in their attachment to either dust, aerosols or water particles in the atmosphere Once in the atmosphere, the radon and its daughters are available for inhalation by humans Because of its gaseous nature, radon is likely to be exhaled during respiration (Ball *et al*, 1991), while the solid attached radon daughter products easily become trapped on the lung lining (Eheman *et al*, 1991) The trapped particles then decay in-situ, resulting in irradiation of the surrounding lung tissue by alpha and beta particles While radon gas itself has relatively little associated health risk due to the large proportion that is exhaled, the daughter products have been linked to lung cancer and acute myeloid leukaemia (Eatough and Henshaw, 1990) In this thesis the term 'radon daughters' will be applied solely to the four decay products of radon-222, and not to the daughters of radon-220 (thoron) and radon-219

1.4 Karst Environments: Definitions

1 4 1 Karst

The term karst is thought to have been derived from the Indo-European word 'Kar' (rock) via the Slavian 'Krs', Slovenian 'Kras' (Ford and Williams, 1989) and Italian 'carso' (Sweeting, 1972) These terms originally defined geographic areas with bare, stoney ground, characteristic surface landforms (principally dolines and poljes) and predominantly underground drainage (Gams, 1991) However, their use within the modern scientific literature is not precisely defined and as such there is no universally accepted definition of what constitutes karst (Jennings, 1985) Limestone is the dominant lithology on which karst features develop but silicate and evaporite karst terrains are also recognised by some authors In this thesis reference will only be made to limestone karst terrains

1 4 2 Limestones

Limestones are sedimentary rocks, most commonly formed in shallow, warm, tropical seas, whose composition is predominantly of the carbonate mineral calcite Limestones vary in purity, impure strata containing less than 70 % carbonate From the perspective of this thesis it is the presence of cave passage within limestone beds that is important Impure beds do not generally allow for substantial cave development as they produce large quantities of insoluble residue It has been found that caves are more likely to develop in strata that contain over 70 % carbonate mineral (Ford, 1977) The distribution of limestones within Britain are outlined in figure 1 5

1 4 3 Caves

The definition of 'caves' is diverse within the speleological literature (Jennings, 1985) The definition adopted by the International Speleological Union defines caves as "solutionally enlarged voids, within limestone strata that can be penetrated by humans" (Ford and Williams 1989, p 242) The majority of caves have natural entrances but some were discovered during mining activities and may only be entered through artificial galleries In addition, miners have altered the dimensions of some natural caves This thesis recorded measurements made in three different cave environments entirely natural solutional caves, caves entered via mined levels, and totally mined levels within limestone strata The present research project was confined to caves over 20m in length or 10m in depth Caves of smaller dimensions were excluded because

1 The radon and radon daughter concentrations are likely to differ little, if at all, from those of the outside atmosphere

2 They attract few visitors and their significance to the caving population is relatively small.

3. Any caver visiting such a cave is unlikely, because of its dimensions, to spend long periods of time in the radiation environment, and therefore the associated health risk is small

The Great Britain Cave Database (Hardwick, 1994) indicates that caves less than 20 metres long or 10 metres deep represent over 51 % of known cave entrances but less than 3% of the total known cave passage length Therefore, whilst the exclusion criteria represent a large percentage of known cave entrances, in practice the vast majority of total cave passage was included in the investigation

1 4 4 Cave Microclimate

An initial literature review found that cave microclimate, and particularly air movements, has been highlighted as a dominant factor controlling radon and radon daughter concentrations within limestone cave environments (e g Wilkening and Watkins, 1976, Yarborough *et al*, 1976, Cunningham and LaRock, 1991, Gunn *et al*, 1991, Middleton *et al*, 1991)

Caves are commonly believed to be static environments, with constant temperatures, high relative humidity and low light intensities (Wigley and Brown, 1976) However, in most cases this is far from true, with measurable spatial and temporal variations in cave microclimate being detectable For example, Cropley (1965) detected the effects of external climatic changes at a distance of 1500 ft (460m)[1] away from an entrance in a cave in West Virginia

Cave microclimate is primarily a function of the relationships between passage morphology and the external atmosphere, the details of which have to be determined for each cave The features of an individual cave which control internal cave microclimate

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[1] In all cases where results are reported in non SI units within publications the original units will be reported and equivalent SI units reported in brackets

are the relationship between internal and external conditions, passage configuration, the rate of air circulation, and heat-transfer processes of conduction, evaporation and condensation (sublimation) Wigley and Brown (1976) state that air movements and speeds are the dominant controls on spatial and temporal variations in cave microclimate, primarily affecting sublimation and evaporation rates and temperature

Early investigations into radon and radon daughter concentrations in limestone caves (Wilkening and Watkins, 1976, Yarborough *et al*, 1976) established that air movements were the primary factor of cave microclimate that affected the spatial and temporal variation in the concentrations observed Therefore, the different processes and controls that operate to induce air movements within caves need to be established Wigley and Brown (1976) identify two basic cave types, with different controls on air movements caves with multiple entrances and single portal caves

In caves with multiple entrances, air currents are driven by thermally- or density-induced differences between cave endoclimate and exoclimate This has more recently been described as the 'chimney effect' (Atkinson, 1981, De Freitas *et al*, 1982, De Freitas and Littlejohn, 1987, Atkinson *et al*, 1983) or 'Upside down caves' (Yarborough *et al*, 1976) Air movement within chimney caves results from two or more entrances being at different altitudes These openings do not have to be penetrable by humans but must allow air movement through them. Air is forced to move through the cave as a result of thermally induced disequilibrium in air density between the cave and outside air Air will move in via the lower entrance during the winter months, while flow will reverse during the summer period In the autumn and spring months air will oscillate and both winter and summer systems may operate during one 24 hour period

Wigley and Brown (1976) stated that only small amounts of air move in single entrance caves systems However, this had previously been found to be untrue, for example, Tuttle and Stevenson (1975) demonstrated that significant volumes of air moved in single entrance caves Single entrance caves were renamed by Yarborough (1977) as 'right side up caves' Yarborough proposed that air can be induced to move in these systems as a

response to two processes firstly, thermal disequilibrium with the external atmosphere, and secondly, as a result of changes in atmospheric pressure (cave breathing) The movement of air in single entrance systems has been described by Geiger (1971), Cigna (1971) and Atkinson (1981) Letheren (1980) and Lewis (1981) describe the thermally induced movement in the entrance series of single entrance caves and identify two distinct circulation patterns

1 During the night / winter periods, warm, relatively less dense air rises out of the cave along the roof and this is replaced by cold, dense, external air flowing in along the floor of the cave

2 During the day-time /summer, the pattern reverses Relatively dense cold air drains out of the cave along the floor and is replaced by warm, less dense, air along the roof.

Cave breathing, where air movements within a single entranced cave, are controlled by changes in atmospheric pressure has been described by Andrieux, (1981), Conn (1966), Cigna (1971), and Smithson (1982 and 1985) This follows the universal gas law, which states that the volume of a gas is related to the pressure and temperature characteristics of the gas under study As temperature variations are small within cave systems, variations in temperature can be ignored during considerations of breathing caves (Coon, 1966) Changes in atmospheric pressure will result in changes in air volume within a cave system. An increase in outside air pressure will decrease the volume of the air within the cave and, therefore, the air will retract within the cave, resulting in air being drawn in from the external atmosphere Conversely, if outside pressure decreases then the volume of the air within the cave will increase resulting in a net exhalation of air from the cave

1.5 Structure of the thesis

Chapter Two presents a basic summary of those aspects of radiochemistry which are relevant to the thesis Chapter Three reviews the published cave radon literature and places the research undertaken during this project in a global perspective Chapter Four

presents a research design developed to meet the aims of the thesis as set out in section 1 2 Chapter Five compares different methods available for measuring radon-222 and radon daughter concentrations, and explains how appropriate techniques and methodologies were developed to investigate radon-222 and radon daughter concentrations in limestone caves. Chapter Six presents results from an investigation of radon-222 concentrations in limestone caves of England and Wales Chapter Seven aims to identify and quantify the different sources of radon-222 to the cave environment Chapter Eight presents the results of a detailed investigation of radon-222 concentrations within a single region, the Peak District of Derbyshire Chapter Nine examines the controls on temporal and spatial variations in radon-222 and radon daughter concentrations within a single cave This is achieved through weekly measurements of radon-222, radon daughter concentrations and cave microclimate at 28 sites within a single cave over a 56 week period Chapter Ten presents a brief summation of the Ionising Radiation Regulations (1985) and how they can be applied to cave environments in the light of this research and discusses the potential health risk to cave users Chapter Eleven presents conclusions and recommendations arising from the research project

CHAPTER TWO: ENVIRONMENTAL IONISING RADIATIONS AND PROPERTIES OF RADON-222

2.1 Introduction

The ionising radiation flux in the environment has two primary sources radioactive decay near the surface of the earth and cosmic rays of extra terrestrial origin impinging upon the earth In most situations the former is the dominant source of radiation and this chapter therefore will concentrate on this source However, it was felt that it might be useful to define and review those basic principles of radiochemistry and radioactivity that are pertinent to the research The aim is to provide those who are experienced in the geographical or geological sciences with sufficient background information to be confident in basic radiochemical theories In no way is it intended to be a complete summary of radiochemical theories The properties of atoms and development of radiochemical knowledge will be discussed in section 2 2, while the chemical and radiochemical properties of radon and radon daughters will be examined in section 2 3 A review of the developments of radiochemical units will be presented in section 2 4 finishing with definitions of the units that will be used in this thesis

Before this can commence these terms themselves need to be defined

1 Radioactivity results from the spontaneous decay of unstable atoms with the formation of daughter products from the original parent

2 Radiochemistry is the study of these processes

2.2 Radiochemical principles

2 2 1 Properties of atoms

Within the natural environment two types of atoms exist stable and radioactive Radioactive nuclei emit a variety of radiations, all with different properties The radiations are characteristic of the atoms from which they arise through the type of decay products and their associated energies (NCRP, 1988)

To fully understand the theories of radioactivity, it is first necessary to examine the structure of atoms, that is to say, the units from which all matter is made Atoms are microscopically small particles consisting of two principal components electrons and a nucleus Electrons are particles which have a negative electrical charge that orbit the nucleus in an "electron cloud" The nucleus consists of protons and neutrons protons are positively charged, this charge being equal in magnitude to that of the electron's negative charge, while neutrons have no electrical charge Protons and neutrons are similar in mass while electrons are approximately 2000 times smaller The nucleus is held together by high intensity forces that only exist over small ranges These forces lose strength as the number of protons increase, until a saturation point is reached

In stable atoms individual characteristics are distinguished by three standard chemical notations atomic mass, atomic number, and mass number Atomic mass is defined with reference to carbon-12, where a carbon-12 nucleus consists of six protons and six neutrons The atomic number is based upon the number of protons The atomic number positions the atom within the periodic table and can be combined with valency to determine chemical reactivity The mass number is the total sum of protons and neutrons within a nucleus, and is used in conjunction with the atomic number to define atomic characteristics

In radioactive particles spontaneous disintegration (or fission) of the nucleus occurs, therefore normal chemical determinates cannot be strictly applied to these atoms The

nature and properties of a radioactive atom are related to the type of particles emitted during decay and the associated energies of these particles There are three primary types of emissions alpha, beta and gamma particles

2 2 2 Radioactive decay

Alpha decay is the ejection of two neutrons and two protons from the nucleus in the form of a helium nucleus In general the energy associated with this type of decay increases with increasing atomic number Energies in a range of 1 8 - 11 7 MeV (Mega electron volts) have been observed during alpha decay Alpha particles can travel at high speeds but due to their relatively high charge are easily stopped by thin card Alpha decay is associated with a decrease in atomic number by two

Beta decay consists of the emission of positrons or negitrons from the nucleus These particles are usually emitted in a continuous stream from the decaying nucleus The particles emitted generally have the same mass/ charge ratios as ordinary atomic electrons With the loss of an electron from a neutron within the nucleus, the neutron is transformed into a proton Consequently, beta decay is associated with an increase in atomic number by one, while the mass number remains constant Energies associated with beta decay range from 0 to 16 4 MeV Beta particles through their low mass can travel up to 4 metres in open atmosphere but are stopped by thin sheets of metal. Beta decay can take the form of either positive (positrons) or negative (negatrons) beta particles Included within the broad definition of beta decay are EC-decay (electron capture) in which the nucleus captures an electron from the electron cloud surrounding the atom.

After both alpha and beta decay the nucleus can be left in an excited state, and through gamma decay these nuclei can return to a ground or stable state. To return to the ground state the nucleus may undergo numerous transformations, the most common of which is through the release of electromagnetic radiations commonly termed gamma rays The return to ground state may not occur through one process, with numerous intermediate

states existing Gamma rays are the most energetic of all emitted particles with energies between 10 KeV and 17 MeV Gamma decay results in a change in nucleus excitation, but no change in atomic number occurs Due to their high energies and zero mass these rays are extremely penetrating, and are not stopped by thin sheets of metal

2 2 3 Decay series

Within the natural environment three primary decay chains have been identified, these were originally referred to as the uranium, thorium and actinium chains but these atoms are now known by their modern names uranium-238, thorium-232 and uranium-235 respectively The head of each decay chain is termed the parent particle, these decay via a chain of daughters to produce stable end products The number of daughter products within different decay chains varies

Decay series do not need to be considered in their entirety and sections can be investigated in isolation For example, uranium-238 decays through six stages to produce radon Therefore, radon is a daughter product within the uranium-238 decay series Radon itself decays to produce eight daughter products four to lead-210 and four more to lead-206 before the chain ends Radon and its four daughters can also be considered separately from the rest of the uranium-238 decay series, usually assuming that the initial radon was in equilibrium with its parent, uranium-238 (see section 2 2 4 for a fuller explanation of radioactive equilibrium)

This thesis will primarily consider the decay series of uranium-238 and one of its daughter products, radon-222 Both the uranium-235 and thorium-232 decay series contain isotopes of radon, but these will not be considered in as much detail as the uranium-238 The reasons for their exclusion are given in section 1 3 2

Uranium-238 decays via fourteen transformations eight alpha and six beta decays, to the stable end product lead-206 Radon is the sixth decay product within this series (figure 1 2) With the production of radon-222 the characteristics of the decay series change

Prior to radon, most of the products have relatively long half lives ranging from 1 6 x 10^3 years to 4 47 x 10^9 years (the exceptions being thorium-234 and protactinium-234 which have short half lives), while radon has a half life of 3 86 days After radon the daughter products have relatively short half-lives ranging from 164 milliseconds to 22 3 years It is because of this and the chemical characteristics of radon (section 2 3 1) that the decay series can sometimes be split, so that studies are conducted on either the long-lived early section or separately on the short-lived radionuclides below and inclusive of radon All three decay series exhibit a similar split after the formation of a radon isotope

Figure 1 2 shows a schematic diagram of the uranium-238 decay series, illustrating the nature of decay and the relevant half-lives It can be seen that each atom in the decay series disintegrates by a set path which involves the release of a particular type of particle with associated energies This set decay route can be relatively straightforward for example, the decay of radon involves only the release of alpha particles, or more complicated, as in the case of protactinium-234 which, upon decay, results in a combination of alpha and gamma radiations being released

The routes by which each nucleus decays is set When the route involves two distinct disintegration paths the process is termed 'branching decay' An example of this within the uranium-238 decay series is thorium-234, the two decay paths consisting of 72% nuclei decay via alpha emissions of 4 77 MeV and 28% decay emitting alpha particles at 4 72 MeV The ratio 72 28 is termed the branching ratio In all cases the percentage which follows each route is constant, and a mixture of radioactive species develops Other atoms within the uranium-238 decay series also exhibit branching decay, examples being thorium-230, radium-226, and three of the radon daughters polonium-218, bismuth-214 and polonium-214

2 2 4 Radioactive equilibrium

Radioactive equilibrium is a description of the relations that exist between parent and daughters of radioactive assemblages within a set environment Radiochemists have

identified three different equilibrium states, each defining the relationships between parent and daughter products within a decay series The following description of these states (secular, transient and non equilibrium states) is based upon Readhead (1984)

Secular equilibrium describes the state that exists for a chain of radioactive atoms headed by a relatively long lived parent The only source is from the initial parent, and no secondary additions or redistribution occurs, with the only loss being radioactive decay The members of the series will reach an equilibrium status which is controlled directly by its predecessor in the chain Therefore, the activities of the daughter products and the parents are the same

Non equilibrium exists where the parent's half lives are shorter than that of the daughter Therefore, a permanent state of disequilibrium exists The parent decays completely away forming the daughter product that will remain unsupported The time before the daughters decay is controlled by their half-life

Transient equilibrium occurs when the parent's half life is much longer than that of the daughters Therefore, the parent's activity does not decrease noticeably during the course of many daughters' half lives

In theory the uranium-238 decay series generally exhibits secular equilibrium if no additions or removals are made to the system. This results in the activities of the daughter products being controlled directly by that of their parents As the primary concern here is radon, the following discussion will be applied to the section of the uranium decay series below and inclusive of radon, although the principles outlined can be applied to the rest of the radioactive decay series

If we assume that radon is the only long-lived parent present, the time it takes to reach an overall equilibrium depends primarily on the rate of loss of each chain member, determined by the radioactive half life of each individual. The times taken to reach secular equilibrium are shown in table 2 1 Table 2 1 includes only radon and its four

immediate daughters, lead-210 being excluded because of its relatively long half-life (22 3 years) All four daughters would reach equilibrium with the parent radon in approximately 4 hours

Radioactive equilibrium is a crucial factor in determining the potential hazard of radon contaminated atmospheres, as the health threat is from the daughters rather than radon itself. Although radon and its daughters will reach secular equilibrium if no additions or removals are made to the system, these strict rules cannot be guaranteed in the majority of situations Therefore, a non-equilibrium mixture will nearly always be present Changes in equilibrium status could be a consequence of either changes in production or removal rates Changes in production rates could occur as a result of variations in atmospheric pressure which may either inhibit or increase radon exhalation The removal of radon or radon daughters could occur as a consequence of either air movements or plate out[1]

2 2 5 Radioactive decay

The activity of a radioactive compound is not only dependent upon the quantity of material present but also the radioactive half-life or rate of decay, the half-life being the average time for the quantity of radioactive atoms to decrease by half This concept was originally defined by Rutherford in 1904 as

> " the activity decrease from any initial value to half that value in a given time, then in a further interval of time the activity would decrease by half again or to one-quarter of the initial activity" (NCRP 1988, p 9)

The decay constant, determines the rate at which radioactive particles decay and the half-life for any atom is constant and is related to the decay constant The decay constant

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[1] Plate out is the deposition of particles onto the floor or walls of a cave due to either natural settling or forced deposition by air movements

is an average and variations are evident due to the random nature of radioactive decay No normal experimental chemical conditions (temperature, pressure, gravitational and magnetic fields) have been able to change the decay constant for radioactive decay

2 2 6 Development and discovery of radiochemistry, with special reference to radon-222

The effects of radioactivity have been documented since at least the 15 century, one of the first accounts appearing in "De Rei Metallica" by G Agricola in 1596 This text describes the affects of "corrosive dusts that eat away at the lungs" (cited in Brenner, 1989, p 23) These effects were reported from silver mines in Germany and Czechoslovakia This report also mentions the hazards of "demons of ferocious aspects" but the underlying theme relates the radioactive working environment to increases in mortality rate for miners, and this has subsequently been linked with cancers (Eatough and Henshaw, 1991)

The scientific discovery of radon and radioactivity did not occur until the late 19^{th} and early 20^{th} century, the term radioactivity being first used by H Becquerel in 1886 (Brenner, 1989) This stemmed from work initiated by Roentgen in 1885 who discovered the penetrating effects of x-rays In 1886 Becquerel carried out experiments using uranium salts which showed that rays were emitted that persisted for long periods of time, discharged electrical charges on thin sheets of aluminium, and were deflected by magnets This he termed radioactive decay In 1899, the Curies discovered polonium and radium by chemical treatment of pitchblende Don continued the Curies' work discovering radon in 1900 E Rutherford and F Soddy laid down the foundations of radioactive theory, identifying, characterising and naming alpha and beta particles The period from 1895 - 1904 saw the pioneering of radiochemistry, and the achievements of this period with special reference to radon isotopes are summarised in table 2 2

2.3 Properties of radon and radon daughters

2 3 1 Properties of radon gas

Radon is a colourless inert radioactive gas with a density of 510 kg m^{-3}, making it the heaviest inert gas in the natural environment As with all inert atoms it possesses a stable closed electronic configuration resulting in relative chemical inactivity. However, due to the low activation energy of the inner orbit, reactions can occur with fluorides and chlorides Radon solubility in water increases with temperature, but decreases with both pressure and carbon dioxide content (NCRP, 1988). Its high density results in a low diffusion coefficient in free air (0 1 cm s^{-1}) A comprehensive review of radon's physical properties was conducted by Weigel (1978) while its chemical properties were described by Stein (1987) Radon as a radioactive gas is formed from radium-226 by alpha decay Radium-226 has a half life of 1600 years and releases 4 8 MeV during decay Radon has a half life of 3 86 days and releases 5 5 MeV solely as alpha particles during decay Upon decay radon produces four relatively shorted lived daughter products namely polonium-218, lead-214, bismuth-214 and polonium-214 Polonium-214 decays to form lead-210 with a half life of 22 3 years, and this is considered to be the end of the radon sub-decay series The uranium-238 decay series ends with the formation of lead-206 from lead-210 via bismuth-210 and lead-210 intermediates (see figure 1 2)

2 3 2 Properties of the short-lived radon daughters

The short-lived daughters of radon have received greater attention than radon gas due to their potential health threat (NCRP, 1988) The daughters rarely exist in sufficient quantities to allow for the physical and chemical properties of the elements polonium, lead or bismuth to be important Consequently, these chemical properties will not be discussed However, the radiochemical properties of the elements are vital particularly in understanding the associated health risks and these will therefore be discussed The radon daughters are linked together due to their associated health risk, related to their

relatively short half-lives, high attachment rate to atmospheric particles and the high energies released upon their decay These particles become attached to the solid fraction in the atmosphere and subsequently, upon inhalation by humans, become trapped in the lungs Due to their relatively short half lives these are likely to decay "in situ" before removal can occur The resulting radiations are active over a small area within the sensitive lungs The radon daughters finish with the formation of lead-210, which with a half life of 22 3 years is likely to be removed by bronchial processes prior to decay The radiochemical properties of the radon daughters, the associated energies released during decay, and the half-lives vary for each individual daughter

Polonium-218, the first radon daughter, is solely an alpha-emitting radionuclide, with a half-life of 3 05 minutes (0 0508 hours) This decay results in the release of 6 00 MeV Unlike its parent, polonium-218 is a solid element, usually in an ionic state, which results in a significant percentage becoming attached to particles in the atmosphere (see section 2 3 3 for further details of unattached and attached fractions)

Lead-214, the second and longest surviving radon daughter, is a beta-emitter with a half-life of 26 8 minutes (0 447 hours) Upon decay 0 87 MeV are released into the atmosphere

Bismuth-214, the third and second longest surviving radon daughter, is a beta-emitter with a half-life of 19 7 minutes (0 328 hours) Upon decay, between 1 54 and 3 27 MeV are released to the atmosphere

Polonium-214, the fourth radon daughter, is an alpha emitter with a half-life of 164 μsecond (4 56 x 10^{-8} hours) Upon decay 7 69 MeV are released to the atmosphere, resulting in the formation of lead-210 With the formation of lead-210 the radon daughter sub-sequence finishes, as lead-210 has a half-life of 22.3 years which effectively terminates the chain

2 3 3 Attached and unattached fractions

In the atmosphere radon and radon daughters can exist in two states, either attached to particulate matter or unattached Radon is usually found in an unattached state in the atmosphere due to its neutral electronic state and chemical non-reactivity Conversely, radon daughters through their ionised state and chemical reactivity are usually found in an attached state in the atmosphere The particles in the atmosphere that most readily react and combine with the radon daughters are water, aerosols and dust Formation usually occurs with the first daughter, polonium-218 which, when produced by the decay of radon, is highly electrically charged, i.e in an ionised state When the subsequent daughters are produced the energy associated with their formation is not sufficient to release them. Therefore, once radon daughters form compounds, they are unlikely to disintegrate (NCRP, 1988) In some environments a fraction of radon daughters can exist in their ionic state NCRP (1988) found that attachment rate was proportional to atmospheric aerosol content, and, as cave environments usually have high humidity levels, (between 80 and 100%) it has been assumed by many authors (e g Yarborough *et al*, 1976) that the majority of radon daughters in cave atmospheres will be attached due to the high moisture content

2 3 4 Equilibrium of radon and radon daughters

Radon and its daughters should be in secular equilibrium, where the activity of the resulting radon daughters is controlled by the decay of radon Equilibrium of radon with its daughters should be achieved in 4 hours (table 2 1) if no processes remove either constituent from the atmosphere under study Therefore, studies of radioactive equilibrium can give valuable information on the age of the air

The state of radioactive equilibrium of radon and its daughters is sometimes expressed as a set of ratios For example 1 0 0 9 0 7 0 6 would describe a mixture of the polonium-218, lead-214, bismuth-214 in ratios of 90%, 70% and 60% of the parent radon Usually polonium-214 is not included due to its short half-life Within this thesis,

equilibrium will be expressed as the sum of the radon daughters' activity to that of radon, a method that is commonly used when investigating accrued dose The equilibrium factor (F) can therefore be described as

> *"the ratio of the potential alpha energy concentrations in the existing mixture to that which would exist if all short lived daughters were in equilibrium with the radon present" (NCRP, 1988 page 18)*

and mathematically expressed as

$$F = \frac{A}{B} \times 3700$$

Equation 2- 1

Where F is the equilibrium factor, A radon daughter concentrations in Working Levels and B radon gas concentrations in Bq m^{3}

2.4. Units

2 4 1 Introduction

In any scientific investigation it is important to clearly define the units of measurement In the case of radioactivity units have been developed over time, often in isolation, and as such may seem to be obscure and overly complex In this thesis all measurements are expressed in the International System of Units (SI units) developed in 1970 and defined below Older units are also considered as it was necessary to review work by authors who used these

When any radioactive investigation is undertaken three main facets of a radioactive particle can be considered the rate of radioactive decay, the amount of energy released and the nature of the particles emitted from the nucleus

Each of these can be investigated quantitatively However, the effects of radioactivity on health are more difficult to quantify as they are related to a combination of factors, both radiobiological and epidemiological.

2 4 2 Radon units

2.4.2.1. Radon activity (Bq)

Initial determinations of radon usually take the form of determining the activity of radon within a particular environment Activity of a sample is generally taken to mean the rate of decay of that sample per unit time The SI unit is the Becquerel (symbol Bq) defined as 1 disintegration per second This is named after Henri Becquerel who was awarded the Nobel prize in 1903 for the discovery of radioactivity (with Marie and Pierre Curie) The older unit of activity is the Curie (Ci), named after the Curies This was historically based upon the activity of radium and is approximately equal to the decay of 1 gram of radium-226 As accuracy of measuring systems improved during the course of the 20th Century the exact figure changed, and this has now been set arbitrarily at $3\ 7 \times 10^{10}$ disintegrations per second Two other measures of activity have been used, but only for limited periods of time in local geographical areas, these being the Mache and Eman These units are only used in this thesis when contained in the original reference and in each case conversions to Bq are included

2.4.2.2. Equilibrium equivalent concentrations (EEC)

In most situations equilibrium does not exist between radioactive parents and progeny Therefore, a unit is needed to describe this non-equilibrium mixture This has been termed the EEC under SI units, and given the symbol Bq m^{-3} EEC is defined as

"a nonequilibrium mixture of short lived progeny in air is that activity concentration of the parent gas in radioactive equilibrium with the concentrations of its short lived progeny that has the same potential alpha energy concentration as the nonequilibrium mixture " (NCRP, 1988 p64)

Therefore, the EEC represents the activity of radon in equilibrium that would result in the same potential alpha energy being released as the initial non-equilibrium sample

2 4 3 Radon daughters

The units used to measure radon daughter concentrations have also developed over time and are more complex than those for radon as they may relate to the total or to individual activity of the constituent

2.4.3.1. Potential alpha energy

The potential alpha energy relates to the total alpha energy that could be released by an atom as it decays through its entire radioactive series The term 'potential' is used for two reasons firstly, lead-214 and bismuth-214 are beta and gamma emitters that ultimately decay to form polonium-214, which in turn liberates alpha particles Secondly, it is known that radioactive particles can move and be redistributed within the body once they have been deposited The unit of measurement primarily relates to the first stage in determining the health implications of entering a particular radiation environment This is another reason why only the alpha energy is included, as it is believed that alpha particles are the most important radiobiologically Due to the long half-life of lead-210 (22 3 years) this is not included in any calculations as it is likely to have been removed by mucus activity prior to decay

Potential Alpha Energy is normally reported in MeV, where an electron volt represents the energy lost as an electron falls through the potential difference of one volt The

potential alpha energy of individual atoms can be found in standard radiochemical texts (e g O'Riordan *et al*, 1987)

2.4.3.2. Potential alpha energy concentration (PAEC)

The PAEC represents the sum of all the potential alpha energies in a volume of air, divided by the volume of that air This is usually measured in J m^{-3} (or MeV l^{-1}), where by definition 1 MeV l^{-1} = 1 6 x 10^{-13} J

The most commonly used unit to describe the Potential Alpha Energy Concentration is the Working Level, first suggested in 1955 by representatives of several uranium mining states who had met to decide upon the safe exposure level to radon and formally defined in 1958 as

> *"any level of concentration or burden of radioactivity in a given air environment which produced a pre-specified potential alpha energy concentration. . Thus, related to environments containing radon and radon daughters, a Working Level is represented by any combination of short lived radon progeny in one litre of air that will result in the emission of 1 3 x 10^5 MeV of potential alpha energy from the radioactive decay of the progeny This would mean 100 pCil^{-1} (picocuries per litre) of each of radon-222, polonium-218, lead-214, bismuth-214, and polonium-214 " (NCRP, 1988 p74)*

They concluded that since the dominant danger was from the radon progeny, any standardisation needed to be based upon the concentrations of the first four radon daughters, with a single unit representing the concentration of all four They decided that the safe level for working (a Working Level) was 100 pCil^{-1} of each of the three daughters products, polonium-218, lead-214 and bismuth-214 This was felt to be one tenth of the German and Czechoslovakian mining levels, and over a working life time of forty hours a week no biological damage would arise as a consequence of exposure

This standard could more precisely be represented as 1 3 x 10^5 MeV of potential alpha energy per litre of air derived from the alpha energy released during the decay of the radon progeny in radioactive equilibrium with 100 pCi of radon-222 per litre of air This standard was then published in 1958 Public Health Report (NCRP,1988)

Due to the characteristics of the radon-222 decay series, over half of the contribution to the Working Level comes from lead-214 and virtually none from polonium-214 Thus the definition of a Working Level is a compromise based upon hypothetical equilibrium atmospheres The Working Level was initially defined only for the progeny of radon-222 but it can equally be applied to the progeny of other radon isotopes by allowing 1 WL to represent 1.37 X 10^5 MeV of potential alpha energy regardless of the source Thus 1 WL corresponds to an activity concentration of 7 43 pCi l^{-1} = 275 Bq m^{-3} for radon-220 (thoron) and of 161 pCi l^{-1} = 5960 Bq m^{-3} for radon-219 (NCRP, 1988)

2 4 4 Dose

Radiation dose can be calculated in three main ways as a function of exposure time in a radiation environment (potential alpha energy exposure), as a function of the absorption of radiation into the cells which receive the radiation (absorbed dose), or as a function of the absorption and radiobiological effects of the radioactive particles (dose equivalent) Dose is defined as a measure of the potential radiological damage arising from exposure to different types and amounts of radiation (NCRP, 1988)

2.4.4.1. Potential alpha energy exposure

The Potential alpha energy exposure is the time integral of the potential alpha energy concentration that a person receives, usually given in units of Working Level Months (WLM), or alternatively Bqh^{-1} m^{-3} (Becquerels per hour per cubic metre) or Jh^{-1} m^{-3} (Joules per hour per cubic metre) One WLM represents an exposure to concentrations of 1 WL for 1 working month (170 hours) 1 WLY equals 365 x 24 divided by 170 =

51.5 WLM PAE exposures can be investigated using WLH (Working Level Hour), WLM and WLY (Working Level Year)

2.4.4.2. Effective dose equivalents

Effective dose equivalents are obtained by multiplying the dose equivalent to various tissues and organs with a risk weighting factor applicable to each, and summing the products Therefore, the effective dose equivalent is an assessment of the actual radiobiological damage to the whole person and to individual organs being exposed to varying degrees of radiation Effective dose equivalents are represented by the unit Sievert (Sv) under the SI unit classification (O'Riordan *et al* , 1987)

2.5 Summary

The properties of radon and radon daughters and basic radiochemical principles have been discussed during this chapter with the intention of providing a basic overview for reference Units within radiochemistry are numerous and complex, having evolved over a period of time These have now been standardised and interrelations have been established with the introduction of SI units Two main facets of radiochemistry are usually determined These are the activity of the concentration under study, and the calculated activity if the radiochemical mixture were in equilibrium. These can then be related to the time spent in the particular environment to determine the potential risk to health, and a dose estimate made In the case of radon, these measurements can be made for both radon and radon daughter concentrations

Within this thesis radon activities are reported in equilibrium-equivalent radon concentrations (Bq m^{-3}), radon daughter concentrations in Working Levels (WL) and dose equivalents in millisieverts (mSv)

The application of these basic theories to the investigation of radon and radon daughter concentrations within limestone caves has highlighted three areas where further investigations are needed

1. The ratio of unattached to attached radon daughters within limestone cave atmospheres and the relationships with relative humidities and aerosol contents It has always been assumed that the high relative humidities and aerosol contents found in cave environments would lead to there being few unattached radon daughters present

2 The nature and effects of changes in exhalation rates and redistribution mechanisms on the equilibrium factor

3 The relationship between the equilibrium factor and the age of the air within the cave These data could be used to estimate and determine features of the cave microclimate such as air exchange rates and timing

CHAPTER THREE: RADON IN LIMESTONE CAVES: AN INTERNATIONAL LITERATURE REVIEW

3.1 Introduction

In this chapter the published literature on radon and radon daughter concentrations within limestone caves is reviewed The chapter is divided into three sections, the first two sections being based primarily upon summaries of the available literature in tabular form whilst the third presents a basic synopsis of the theories proposed to account for variations in the radon system. The cave radon system is a multivariate system with numerous interdependent parameters These however, have been treated in isolation to allow for the theories developed within the literature and the mechanisms to account for the radon system to be discussed

3.2 Historical summary of international radon literature

Most authors have credited Breisch (1968) as the first worker to publish results discussing radon within limestone caves (see table 3 1 for a historical list of radon references) However, Yarborough (1977) identified Reckmeyer (1962) as the primary authors Breisch (1968) refers to work by Harris (1957) where radon gas concentrations have been recorded in a limestone cave, but the present author could not trace this reference to validate the findings reported by Breisch (1968) However, the earliest published work of which the author is aware, is a Ph D thesis from 1935 by Fryer This was conducted in the Department of Physics, at Indiana University The work investigated the radioactivity of air, water, cave and soil gases Fryer's work in 1935 was not followed until Harris (1954), Geslin and Urbain (1962), Reckmeyer (1962), Breisch (1968) and Saumande and Renault (1971) It was Breisch's 1968 paper that renewed interest in the radon issue and promoted Van Cleave (1975) and Trout (1975) to undertake studies in the USA. Their work led to the involvement of the National Park Service (NPS) in the US who sponsored several projects from 1976 - 1980, primarily

those of Yarborough *et al* (1976) and Ahlstrand and Fry (1976) This work promoted interest in other areas of the world and research was undertaken in Japan (Ikeya, 1976, Miki and Ikeya, 1980), South Africa (Gamble, 1981), Poland (Ciezkowski, 1978), United Kingdom (Friedench, 1981, Middleton, 1988, Gunn *et al*, 1989a and b, Williamson, 1990), Canada (Atkinson *et al*, 1983), Spain (Fernandez *et al*, 1984), Hungary (Gadoros, 1985, Stelcl and Plachy, 1988, Geczy *et al*, 1989), Slovenia (Kobal *et al*, 1978, 1986, 1987 and 1988), France (Saumande, 1971, Saumande *et al*, 1988) and Italy (Cigna and Clemente, 1981, Cigna 1986, 1987a) The focus of research during the 1990's has shifted away from the USA and Hungary towards Europe and Australia, with work being conducted in Luxembourg (Massen *et al*, 1992 and Kies and Masson., 1993), England (Hyland *et al*, 1993, Hyland and Gunn, 1993) and Australia (Lyons *et al*, 1992) Tables 3 1 and 3 2 provide summaries of radon related works up to 1993

Table 3 2 presents a summary of published radon and radon daughter concentrations from limestone caves Where original units were not standard (i.e not SI units) they have been converted to SI units with radon gas concentrations, in Bq m^{-3}, and radon daughter concentrations, in WL The standardisation of the published data could lead to misinterpretations due to errors associated with conversion although every effort has been made to avoid this An attempt has been made to present mean concentrations where possible However, in some cases the mean is derived from published data, based upon summary data of maximum and minimum values for individual sites or caves, and not from the complete data set Consequently, the mean values may not be directly comparable

The highest published concentration is 190,000 Bq m^{-3} in Ireland by a Dublin High School (Kelly, 1994), previously the maximum had been 155,000 Bq m^{-3} recorded in Giants Hole, Derbyshire (Gunn *et al*, 1989 a and b) In most countries concentrations do not rise above 25,000 Bq m^{-3} Maximum concentrations recorded in the following countries are USA 13 4 WL (approximately 50,000 Bq m^{-3}, Yarborough and Ahlstrand, 1977), Greece 88,060 Bq m^{-3} (Papastefanou *et al*, 1986), Russia 68,110 Bq m^{-3} (Klimchouk, 1992), England 155,000 Bq m^{-3} (Gunn *et al*, 1989) The mean

concentration varies for each country but is generally in the range 1,000 - 5,000 Bq m^{-3} Exceptionally high concentrations have usually been accounted for by the existence of additional sources of uranium within the karst environments, primarily related to sediments within the cave environment (Bottrell, 1991, Gunn *et al*, 1989, Lenart *et al*, 1989, Navratil and Stelcl, 1990, Papastefanou *et al*, 1986, Surbeck and Medici, 1990, Yarborough *et al*, 1976)

3.3 The general cave radon system

The radon system within limestone caves has two primary features, namely that variations occur on a seasonal and diurnal basis All authors have recorded either one or both of these phenomena Generally, radon and radon daughter concentrations are highest during the summer months and lowest in the winter periods In autumn and spring concentrations are highly variable and may reflect either the summer or winter radon system on a daily basis Diurnal variations have been observed where concentrations are highest during the day and lower at night, considerable variations being exhibited between the day and night time periods Several theories have been proposed to account for these variations, and these are discussed below

3.4 Theories proposed to account for the cave radon system

3 4 1 Cave morphology

Yarborough *et al* (1976) were the first authors to suggest that seasonal variations in radon and radon daughter concentrations are based upon cave morphology Their theories were later developed and modified by Gamble (1981) and Atkinson *et al* (1983) Yarborough *et al* (1976) proposed two basic cave morphologies, these being Right Side Up (RSU) and Upside down (USD) caves (figure 3 1) Concentrations in each cave type experience an increase during the summer months and decrease during

the winter, assuming that the cave is left to ventilate naturally These increases and decreases were attributed to natural airflows within the cavern.

Yarborough *et al* (1976) define RSU caves as caves with the majority of passage below the cavern entrance (these were later termed sack caves by Gamble (1981)) USD caves were defined as those that have the majority of cave passage above the entrance (Gamble, 1981) Atkinson *et al* (1983) later termed these transient caves In summer, increases in radon concentrations in RSU caves can be related to stagnation of cave air, while in USD caves, increases are related to the origin of the cave air, air flowing out from the cave entrance in the summer results in mobilisation of radon from the further reaches of the cavern In both cases a decrease in concentrations is observed during the winter months as external air flows into the cave resulting in dilution and redistribution of cavern radon Concentrations observed during the spring and autumn months are highly variable and will approximate to either summer or winter conditions Yarborough *et al* (1976) suggest that temperature differentials between the internal cave air and external atmosphere are the primary driving force for the air movements and in a later paper the air movements were shown to relate to Newton's second law for the conservation of linear momentum (Yarborough, 1977) Yarborough *et al's* (1976) simplistic account does not fully explain the processes operating within the cave environment that generate the forces that result in the airflows producing seasonal variations in radon and radon daughter concentrations Yarborough *et al* (1976) suggest that density differences between the internal and external atmosphere result in air movements The density of air can be defined mathematically as

$$K = P - \frac{Mpw}{273\ 13 + \Theta} + Mkw$$

Equation 3- 1

Where K = the air density in kg m^3, Θ = air temperature in ℃, P = atmospheric pressure in millimetres of mercury, M = relative humidity, pw = partial pressure of water kw = density of water vapour in kg m^3

Equation 3 1 suggests that the greatest differences in air density between the internal cave air and the external atmosphere will result from differences in air temperature, and this is consequently the driving force behind air currents in the majority of caves Some authors (for example, De Freitas *et al*, 1982 and Atkinson *et al*, 1983) demonstrate that temperature differences between the two atmospheres can be used to determine the direction and strength of air currents instead of calculating air density

Differences in air density between two points result in a pressure gradient that will induce air to move from areas of relatively high pressure to zones of relatively low pressure Generally, caves have a stable internal temperature that is lower than the external temperature during the summer but above the external temperature during the winter months In RSU caves during the winter months, the internal cave air is warmer than the external air Using equation 3 1, the implication is that the internal cave air will have a relatively low density in comparison to the external air Therefore, air will flow into the cavern diluting and redistributing the cavern radon Smithson (1982, 1985) found that dense external air flowed into the caverns along the floor level and expelled cavern air along the roof. Conversely, during the summer months the outside air is warmer than the cavern air, thereby creating a low pressure zone outside the cavern into which the relatively high pressure air in the cavern automatically flows

In USD caves it is assumed that two entrances exist, even if one of these entrances is unknown and may not be penetrable to humans However, the unknown entrance will still allow for air movement Air in these caves is induced to flow as a result of air density differences when the volume of air inside the cave is compared with the density of the same volume of air outside Due to temperature, humidity and atmospheric pressure differences, the two air masses will have different densities The difference in air density between these two air masses can be defined by

$$K = \frac{3\,485P - 1\,31Kw}{1000T}$$

Equation 3- 2

Where K = air density, P = atmospheric pressure, Kw = vapour pressure and T = air temperature (in °Kelvin's)

The direction of air movement within the cave is determined by the difference between the mean density of air inside the cave (K_{int}) and that of the outside air density (K_{ext}) Air flows in an upward motion (by entering the cavern at the lower entrance) when $K_{int} < K_{ext}$, (normally associated with the winter months) This results in dilution and redistribution of radon within the cavern Conversely, when $K_{int} > K_{ext}$, air enters the upper portal and moves through the cavern and out of the lower entrance This is the usual system during the summer months, resulting in increased radon concentrations as radon is mobilised from the further reaches of the cavern and there is a decrease in the efficiency of dilution of the internal radon rich air by incoming radon free air It is interesting to note that De Freitas *et al*, (1982) demonstrate that reversal of air flow occurs when K_{int} and K_{ext} are identical, not when thermal conditions prevailing inside and outside the cavern are the same This implies that air temperature alone can not be used as an indicator of air movements, and where possible air density should be used

3 4 2 Temperature

Temperature differences between the internal cave air and external atmosphere have been suggested by numerous authors as the primary influence on observed radon and radon daughter concentrations within limestone caves (Ahlstrand, 1980, Gunn *et al*, 1989a, Middleton, 1988, Navratil and Stelcl, 1990, Quinn, 1988, Yarborough, 1981, Williamson, 1990) Ahlstrand (1980) suggested that the minimum external temperature is the primary control on radon concentrations, but this was in a RSU cave (as classified by Yarborough *et al*, (1976)) Yarborough (1981) demonstrated with data from the United States that temperature and pressure are influential in controlling cave radon concentrations along with cave morphology Temperature and pressure are important as they control air flows within the cavern Middleton (1988) demonstrated that airflows within Giant's Hole, in the Peak District, are dependent upon the external temperature The direction of air movement, either into or out from the cavern was demonstrated to influence radon concentrations Quinn (1988) demonstrated that external temperature influences radon concentrations in Lehman Cave, Nevada, USA, however, due to the large standard errors associated with the measurements, predictive models cannot be derived based solely upon temperature profiles Williamson (1990) demonstrated that, for a number of caves external temperature and pressure are influential in controlling observed radon concentration However, there has been little discussion of the interaction between external temperature and pressure and cave morphology Navratil and Stelcl (1990) are not as definitive in their conclusions as Yarborough *et al*, 1976 and instead of demonstrating that external temperature influences cave radon concentrations, they only link warm and cold periods of the year with variations in observed radon concentrations

Yarborough (1981) stated that the influences of air temperature on air movements within limestone caves and their importance on observed radon and radon daughter concentrations have to be investigated fully and jointly, with all research projects addressing all aspects of cave microclimate to explain the processes operating within the

cave radon system. Therefore, all theories relating the effects of temperature on cave radon also need to be related to cave morphology which not all workers appreciated Therefore, the processes discussed in section 3 4 1 need to be incorporated and are dependent upon the theories discussed in section 3 4 2

3 4 3 Pressure

A number of authors have demonstrated that changes in atmospheric pressure influence cave radon concentrations (Ahlstrand, 1980, Carson, 1981, Yarborough, 1981, Middleton, 1988, Gunn *et al*, 1989a and b, Navratil and Stelcl, 1990, Williamson, 1990) Shimo and Yamauchi (1980) demonstrated that atmospheric pressure was important in controlling radon concentrations in an industrial tunnel. In all published studies changes in atmospheric pressure have only been influential on radon and radon daughter concentrations within caves classified as RSU by Yarborough *et al*, (1976) To date, no published results have identified caves classified as USD as being affected by atmospheric pressure The volume of gas (air) within a cavern is controlled by Boyles law

$$\frac{P_1V_1}{T_1} = \frac{P_2V_2}{T_2}$$

Equation 3- 3

Where P_1 and P_2 represent the initial and final atmospheric pressures and V_1 and V_2 represent the initial and final volumes of gas and T_1 and T_2 the initial and final temperatures in degreees Kelvin

Therefore, if the atmospheric pressure changes, the volume of air within the cave will be affected This change in volume can result in the expansion of air if atmospheric pressure decreases or a decrease in the cave air volume if atmospheric pressure increases A change in atmospheric pressure by 10 millibars results in the volume of the cave air to change by 10% Conn (1966) states that atmospheric pressure changes will affect the volume of air in caves instantaneously Therefore, changes in atmospheric pressure can

either induce air to flow into the cave, thereby diluting and redistributing cavern radon and promoting a lowering in concentrations, or they can result in the expulsion of air from the cave, and hence result in radon mobilisation and consequent increases in radon concentrations

Yarborough (1981) established that changes in atmospheric pressure can influence observed radon concentrations Atmospheric pressure can have a twofold effect firstly, by influencing air flow and secondly by changing the rate of radon exhalation into the cave Changes in atmospheric pressure will alter the volume of cave air, in relation to the Boyles law Air flow within the cavern can either increase or decrease depending on whether the expansion or contraction of the cave air is in opposition or is superimposed on the natural air flow If atmospheric pressure increases then exhalation will decrease while conversely if atmospheric pressure decreases exhalation will increase This indicates that even though there are no published data describing the effects of atmospheric pressure changes in USD caves the effects of changes in exhalation rate could be an important variable in the radon system. The effects of pressure on exhalation rates will be considered further in section 3 4 7 and in chapter seven

3 4 4 Depth

Several authors have demonstrated that depth of cave passage from the entrance can influence observed radon concentrations (e g Lenart *et al*, 1989, Piller and Surbeck, 1989, Hyland and Gunn, 1993) The increase in concentrations could be related to two facets of the radon system. Firstly, further into the cavern the influences of the external atmosphere are more strongly buffered, and secondly, there might be a tendency for radon to 'sink' to the bottom of the cavern, due to the density of radon and its daughters However, since little detailed work has previously been undertaken or published on this subject, the possible controls on and processes operating will be discussed further in chapter six.

3 4 5 Equilibrium between radon and radon daughters

In their study Ahlstrand and Fry (1976 and 1977) found that the equilibrium factor between radon and radon daughters concentrations was always lower than expected (in all cases below 0 5) They suggested that this could be related to the high moisture content in caves which results in a higher percentage of daughters being attached to aerosols The high percentage of attached fraction results in a larger proportion of daughters being plated-out Cigna (1986) suggests that dose estimates for Show Cave guides in Italy would be underestimated if the low equilibrium factors were not accounted for during calculations Kobal *et al*, (1987) found that caves in Yugoslavia had highly variable equilibrium factors ranging from 0 21 - 1 0 These results imply that equilibrium factors cannot always be assumed to be low Burian and Stelcl (1990) demonstrated that the equilibrium factor is temporally as well as spatially highly variable and can be used to provide an indicator of the age of cave air

3 4 6 Radon-220 (Thoron)

Those authors who have undertaken radon-220 (thoron) measurements within limestone caves have all found the concentrations of radon-220 to be low (e g Yarborough *et al*, 1976, Ahlstrand, 1980, Kobal *et al*, 1978, 1988, Lenart *et al*, 1989) However, insufficient information is available to totally disregard radon-220 (thoron) as a hazard within the karst environment Therefore, limited measurements were undertaken within the karst systems of England and Wales during the course of this research

3 4 7 Sources of radon to limestone cave environments

Very few authors have identified or quantified the relative importance of different sources of radon to limestone cave environments The first author to suggest a source for the radon in caves was Breisch (1968), who proposed that uranium rich strata were influential in radon production Wilkening (1976) suggested that the radon concentration recorded in Carlsbad caverns could solely be accounted for by emanation from the

limestone bedrock Somogyi *et al*, (1989) suggested that the uranium concentrations within the parent limestone rock (2 2 ppm) were sufficient to maintain radon concentrations encountered in Hungarian caves. On the other hand, Lenart *et al* (1989) suggested that sediments are an important source of radon in Hungarian caves Surbeck and Medici (1990) demonstrated that the soil acts as the primary source for cave radon in Switzerland, and that the radon was transported into the cave via percolation water Conversely, Navratil and Stelcl (1990) suggested that the high radon concentrations encountered in Czechoslovakian caves could be accounted for by the addition of uranium to the cave environment through sediments within the system containing uranium-rich graywakies (*sic*) and shales Middleton (1988), Gunn *et al* (1989a and b) and Williamson (1990) all suggested that Namurian shales are the main sources of uranium and hence radon in cave environments in the English Peak District Bottrell (1991) demonstrated that the age and nature of sediments are influential in controlling the radon production in sediments from Speedwell Cavern, also in the Peak District Generally, older, fine-grained sediments had a greater radon production potential than younger coarser sediments

The sources of radon to the cave environment are considered further in chapter seven where the individual sources and their relative importance to the overall radon budget of a karst system are investigated

3 4 8 Variations of radon in relation to height within cave passages

Both Trout (1975) and Miki and Ikeya (1980) have investigated variations in radon concentrations in relation to height within a single passage Both authors undertook investigations in RSU caves and found that no variation existed However, as little attention has been given to this problem a detailed investigation was undertaken during this research project

3 4 9 Sunspot Activity

Somogyi *et al* (1989) presented average radon concentrations for a number of caves over a 10 year period. They demonstrated how yearly variations in average radon concentrations could be related to sunspot activity

3 4 10 Models developed to account for observed radon concentrations

The first author to develop models to predict radon concentrations within limestone caves was Wilkening (1976) He demonstrated that when a cave system was in equilibrium, the radon production rate from the walls was balanced by losses due to radioactive decay and removal by ventilation The ventilation would result in radon rich cave air being replaced by relatively radon free external air This process can be modeled by

$$SE = \lambda VC + Q(C - C_{od})$$

Equation 3 - 4

Where SE is the net inward flux of atoms per second for each square meter, λ the decay constant for radon, C and C_{od} are the radon concentrations within the cavity and outdoors repectively, V the volume of the cavern and Q the air flow

If the cavern air has relatively high radon concentrations and the external atmosphere has low concentrations, C_{od} can be neglected and therefore equation 3 4 can be solved to produce

$$C = \frac{SE}{(\lambda V + Q)}$$

Equation 3 - 5

Therefore, the radon concentration is inversely proportional to volume and the air exchange rate assuming perfect mixing on timescales less than 3 86 days (the half life of radon) This model describes the radon concentration within a simple cavern but cannot be widely used in anything other than the simplest of systems as the errors associated with determining the volume of the cavity and the air exchange rate are likely to be large As the flux of radon into the system is unlikely to be homogeneous and the effects of emanation from sediments and other sources cannot be accounted for within this model, it only allows for a basic understanding of the radon system.

Yarborough (1977) uses Newton's second law for the conservation of energy to describe the air flows in caves that control radon concentrations Newton's second law can be represented by

$$\sum \vec{F} = \frac{d(p\vec{v})}{dt} = p\vec{a}$$

Equation 3 -6

Where $\Sigma F^{\rightarrow}$ represents the summed vector of the major forces which result in a fluid's motion, the derivative $d(pv^{\rightarrow})/dt$ represents changes in air velocity over time

This being equal to the inertia force ($pa^{\rightarrow}$) needed to initiate motion The inertia force $pa^{\rightarrow}$ has to be overcome before air movement can occur and therefore the airflow within caves will follow the vector equation outlined

$$\sum \vec{F} = \vec{P} + \vec{G}t = \vec{p}a$$

Equation 3 - 7

Where the derivative $\vec{P}$ is produced by atmospheric pressure and $\vec{G}$ represents temperature gradients between the internal and external atmosphere

As both temperature and pressure affect the density of air ΣF can be represented by P + Gt which is equal to the inertia force $\vec{pa}$ From equation 3 7, Yarborough (1977) demonstrated that temperature gradients are the predominant driving force producing air flows within limestone caves in both RSU and USD cave types

Miki and Ikeya (1980) developed a model to predict the radon concentrations within different sectors of a single cave using distance from the entrance, exhalation from the cave walls, radius of the cave passageway, length of the cave, average velocity of the cave air, the decay constant of radon, and a correction factor dependent upon surface area This model is an advance upon Wilkenings (1976) model and accounts for variations in internal cave morphology and surface area A series of differential equations can then be solved to predict the radon concentrations at particular sites within the cavern It was found that nearly all measured values fall within the predicted values

Atkinson *et al* (1983) developed a theoretical model to predict radon concentrations within Castleguard Cave, Canada Under stable atmospheric conditions radon within the cavern will increase with distance from the entrance according to

$$A(x) = \frac{A_w}{\lambda}(1 - e^{\frac{-\lambda x}{v}})$$

Equation 3 - 8

Where A(x) = the radon concentration in the passage at distance x from the entrance, v= the velocity of air, λ = the decay constant of radon and A_w the exhalation of radon into the cave atmosphere

As lambda (λ) is small, radon should reach equilibrium (radon decay being equal to addition by exhalation) in approximately 30,000 m. Atkinson *et al* (1983) based their equation on Wigley and Brown's (1976) equation, for relaxation length, which accounts for the increase in cave air temperature with distance from the entrance The increase in cave air temperature can be accounted for by addition of geothermal energy and the processes of condensation and sublimation Atkinson *et al* (1983) proposed that the relaxation length, with reference to radon, is related to air velocity and the decay constant. Therefore, it would be expected that the radon concentration within the cave would gradually increase and reach a plateau However, their measurements demonstrated a gradual increase in concentrations for the first 2600 m followed by a sudden decrease to an overall minimum after which a gradual increase in concentrations was observed Atkinson *et al* (1983) suggested that these deviations from expected values indicated that sources of radon free air were entering the system.

Most models developed to account for variations in radon concentrations within limestone caves have been relatively simple and based upon limited data and therefore none have been entirely successful in fully accounting for the variations in radon concentrations observed

3 4 11 Management Techniques

In the United States, several authors have developed management techniques to allow caves owned and operated by the National Park service to be run efficiently and safely Yarborough (1976) and Yarborough and Ahlstrand (1977) were the first to publish management plans, which stated that

1 Monitoring should take place, on an annual basis, in caves where concentrations below 0 03 WL occur throughout the year Caves with concentrations between 0 03 - 0 1 WL should be investigated tri-monthly, and those caves with

concentrations above 0 1 WL should be investigated at least monthly, if not more often.

2 People should wear respirators if radon concentrations rise above 1 0 WL and no one should be allowed to enter areas with concentrations above 2 0 WL.

3 The Kusnetz method should be developed as an international standard for determining radon daughter concentrations within limestone caves

4 Show Cave guides should also include their recreational caving activity within their dose records

5 Air from Show Caves should not be used to ventilate office buildings

6 No smoking should be allowed in caves

7 One-off measurements should be discouraged and intensive monitoring programs encouraged

8 Forced ventilation should not be used within Show Caves as this would affect the fragile cave microclimate

9 The received radiation dose for all workers should be calculated

Subsequent management plans (e g Gamble, 1981, Gunn *et al*, 1989a and b) have generally followed the same format as those initially developed by Yarborough (1976) and Yarborough and Ahlstrand (1977) However, in certain British Show Caves where summer concentrations were often above 2 0 WL, forced ventilation has been introduced in an attempt to reduce radon daughter concentrations and hence reduce the long term impacts of exposure to radon daughters on Show Cave guides (Gunn, pers comms , 1993)

3 4 12 Health Risk

The majority of authors who have considered the health risk, relating exposure to radon and radon daughter concentrations, consider that there is a health risk associated with radon in limestone caves (e g Ahlstrand and Fry, 1976, Yarborough *et al*, 1976, Cigna and Clemente, 1981, Gunn *et al*, 1989a and b, Hyland and Gunn, 1993) However, a small minority of authors consider the risk to be very small (e g Aley, 1976, Beckman, 1977) The health risk is dependent upon the atmospheric concentration and the exposure period Therefore, guides within Show Caves may be at risk even in caves with low radon concentrations due to their long exposure periods whilst visitors to a Show Cave on a one off basis are at little risk due to the short exposure period. The risk faced by recreational cavers varies and has been shown to be significant in some areas of the world, such as Giant's Hole, in the Peak District (Gunn *et al*, 1989a and b) but insignificant in others (e g Italy by Cigna, 1987a) The risk to cavers health is considered in more detail in chapter ten

3.5. Summary

The review of the international literature has highlighted six areas of interest

1 Temporal and Spatial variations in radon and radon daughter concentrations occur at all scales of investigation from a single cave to international investigations

2 Cave morphology and air flow have been identified as the primary controls on recorded concentrations

3 External temperature and pressure influence air flows within caverns and are therefore of importance in any study of cave radon concentrations

4 Basic models have been developed to account for the concentrations of radon within limestone caves, but none of these withstand rigorous testing and tend only to be valid for the caves in which they were developed

5 A potential health risk has been identified in all countries although the estimated magnitude of this risk varies

6 Management plans have been written with the intention of reducing the exposure of persons entering limestone caves to radon and radon daughters

CHAPTER FOUR: RESEARCH DESIGN

4.1 Introduction

This chapter explains how a research project was designed to fulfill the aims outlined in section 1 2 From the literature review in chapter three it is clear that most investigations of radon in caves have been conducted on a small number of caves on a national or regional basis and that only a few detailed investigations have taken place One aspect of the radon phenomenon that has received little investigation, and for which little information is available, is the identification and quantification of different sources of radon to the karst environment Considerable, attention has been devoted to calculating the radiation doses received by guides within Show Caves but little consideration has been given to other users of limestone cave environments

4.2 Methods

Before any project can commence the various methods available to investigate the phenomenon under study need to be identified and compared In the case of radon measurements in caves no clear consistent sampling / analytical protocol had previously been internationally identified as a standard technique Therefore, a trial of different methodologies was carried out during a six month period at the start of the project, the results of which are reported in chapter five Standard methods for determining radon and radon daughter concentrations were established and these were then used during all field investigations

4.3 National project

To determine typical radon gas concentrations in the limestone caves of England and Wales a survey was carried out across the four main caving regions Passive radon gas detectors were placed at 250 sites within thirty-four caves and broad temporal variations

at all sites were established by sampling during each of the four seasons of the year The sampling periods were 2 -11 August 1991, 1 - 10 November 1991, 31 December 1991 - 9 January 1992, and 3 - 12 May 1992 The effects of cave morphology and geological setting were investigated by varying the detector location in different caves and within individual caves The data collected were also used to assess the potential threat to the health of cave users

4.4 Regional investigation

As expected from the literature review in chapter three, regional variations in radon and radon daughter concentrations were identified in the national investigation To understand variations in radon concentrations within a single region, a more detailed investigation was required Due to the high concentrations recorded and the potential risk to health, this was undertaken in the Peak District Derbyshire County Council sponsored an investigation of caves and abandoned mines used by recreational caving groups in this region during 1990 Twenty-six sites in eight caves were initially investigated for thirteen sampling periods of twenty-eight days and an additional three sites in a ninth cave were included for the last six of the twenty-eight day sampling periods The Peak District caving region was divided into five sub zones and caves used by outdoor educational departments were sampled to determine the integrated radon gas concentration over twenty-eight day sampling periods The locations of detectors within each cave were chosen so as to produce as representative a radon concentration for the cave as was possible given the constraints of time and resources together with the need to place detectors along the routes most commonly used by educational groups Therefore, the results might not represent the whole cave under investigation, but do represent concentrations along a heavily used route

Results from this project contributed towards the determination of typical concentrations of radon gas within caves in the Peak District together with the evaluation of seasonal

controls, the effects of cave morphology and geological setting on radon concentrations and the determination of the risk to cave users from radon gas within cave environments

4.5 Detailed Investigation

A detailed investigation of the controls on radon gas concentrations within a single cave was undertaken in Peak Cavern, in the Peak District Radon daughter, radon gas and meteorological data were collected weekly from twenty-four sites within the cave over a fifty-five week period Additional data were collected at individual sites on hourly and three hourly intervals External climatic data were obtained from a nearby automatic weather station, three hourly data from Manchester Airport and daily records from Buxton weather station

4.6 Emanation and Exhalation

A national survey of uranium-238 concentration within the main cave bearing limestone strata was undertaken to determine the net radon production rate of limestone (radon emanation) Exhalation tests were then conducted on individual beds to determine the percentage of radon produced within the rock matrix that is released into the atmosphere (radon exhalation) Radon emanation and exhalation were determined for samples in both the field and in controlled laboratory environments Selected samples were collected to encompass the range of sediments found within caves in the four main caving regions, and their radon emanation and radon exhalation rates were measured The relative importance of sediment and rock to the overall radon budget was determined. Samples were collected from sites within caves investigated during the national project (section 4 3)

4.7 Estimate of the potential health risk to users of limestone cave environments

The risk which radon poses to the health of cave users was examined using data collected during the national investigation of radon gas concentration in England and Wales The users of limestone caves were identified and the amount of time spent underground by each individual group was determined by a combination of telephone surveys and postal questionnaires These data are combined with the results from the national survey and the relative risk to the health of cave users determined Dosemeters were issued to a selected number of people, on a quarterly basis for four quarters, as a check on the estimates

4.8 Summary

This thesis contains results from seven sub projects that are linked in an attempt to present an integrated research project investigating the phenomenon of radon gas and radon daughter concentrations within limestone caves The overall project is the largest single investigation into radon and radon daughter concentrations within limestone caves conducted to date

The results from each sub project are presented in individual chapters and linked where possible during the course of the thesis The full extent of the interrelationships between chapters are developed in the final chapters where discussions and conclusions are drawn

The results from additional radon and radon daughter measurements which were undertaken during the course of this resarch but not included in the main discussion, are presented in an appendix. This will allow any users of cave environments who are concerned about the risks from radon daughters within limestone caves to take positive action to avoid over exposure

CHAPTER FIVE: METHODS FOR DETERMINING RADON AND RADON DAUGHTER CONCENTRATIONS

5.1 Introduction

Within health physics, numerous techniques have been developed to measure different radionuclides in diverse environments with varying degrees of accuracy At the onset of any research project the different methodologies available need to be scrutinised under preset constraints, depending upon the nature and aims of the individual project, to allow for a combination of appropriate techniques to be employed In this research project the following constraints particularly apply that any method employed be accurate, robust, light in weight and small in dimension Due to the sensitivity of most instruments, the latter three criteria were the hardest to fulfill. Due to the location of the research (often in wild caves), these were also the most important In order to fulfill these criteria, the methodologies employed were not the most sensitive available but their robust nature allowed for data to be collected under adverse conditions in hostile environments, thus allowing the investigation of radon and radon daughter concentrations in limestone caves to be undertaken

In any investigation of radon induced radioactivity, four parameters may be measured or estimated

1 Radon gas concentrations

2 The total radon daughter concentrations

3 The concentration of individual radon daughters

4 Radiation dose

The review below is based upon the discussion of the first two parameters as these are most directly relevant to the study Estimates of received radiation dose (dosimetry)

were undertaken for all users of limestone caves and are discussed in chapter ten Only limited investigations of the concentration of individual radon daughters have been undertaken, data being available from only one cave at hourly intervals for some 20 days

The chapter is split into three main sections, the first two review and compare techniques available for measurement of radon gas and radon daughter concentrations, and the third discusses sampling protocols developed to ensure methodological consistency and accuracy

5.2 Measurement of Radon gas concentrations

5 2 1 Introduction

The methods used to measure radon gas concentrations may be divided into two groups

1 Methods in which it is assumed that radon and radon daughters are in equilibrium during sampling

2 Methods in which the radon daughters are removed during sampling and subsequently allowed to ingrow before counting, equilibrium conditions being allowed to develop

All the methods reported below are based upon the removal of radon daughters during sampling It was felt that any method that assumed equilibrium between radon and its daughters during calculations would introduce errors, as the equilibrium factors are known to vary in mine and cave atmospheres (Yarborough, 1982, McFarlane, 1984) This was subsequently confirmed, equilibrium ratios of 0 05 to 0 98 being encountered during sampling

The timescale over which the radon concentrations are measured is very important Some techniques use a bulk sample of air collected over a period of 1 - 90 minutes and these are termed 'spot' measurements in this thesis The measurement is representative only of the time period during which gas was collected Alternatively, some methods sample over longer periods of time, from four hours to over 28 days These are termed 'integrated' techniques as the measurement obtained represents the mean for the sampling period The longer this period is the greater will be the 'smoothing' of the data For example, 24 hour integrated samples will smooth out variations between day and night while 28 day integrated samples may 'hide' substantial day to day variations

5 2 2 Time Integrated Measures

Two methods were used during this project track etch detectors for sampling 7 days and over, and charcoal canisters for investigations under 4 days

5.2.2.1 Track Etch Film

Track etch methods involve placing a sealed chamber containing a specially developed plastic film into the radon environment under study Detailed reviews of the different track etch methodologies are provided by Bartlett *et al*, (1988), Henshaw, (1989), Miles *et al*, (1983), Pfligersdorffer *et al*, (1981), Somogyi and Lenart, (1986) and Wrixon *et al* (1988) In summary, radon, due to its gaseous nature, enters the closed chamber, whereas radon daughters, being solid, are prevented from diffusing into the chamber The radon in the chamber decays resulting in the emission of alpha particles These particles impart their energy onto the plastic film, producing conical pits or tracks The detector equilibrates with the radon in the atmosphere under study in approximately 25 minutes (Bartlett *et al*, 1988, Khan *et al*, 1987) At the end of the sampling period the detectors are returned to the laboratory for development Many different permutations on the development technique exist, but all involve placing the film in 20% sodium hydroxide at constant temperatures (between 25°C and 89°C) for a fixed period

of time (Camplin *et al*, 1988, Green *et al*, 1981, NCRP, 1988) During the development process it is critical that the concentration of the sodium hydroxide solution remains constant (Amin and Henshaw, 1981, Henshaw 1981) Development etches pits or tracks on the film and these are counted under a microscope or with an image analyzing system (Fews and Henshaw, 1982) Through calibration in radon chambers the number of tracks can be directly related to radon activity and exposure time (NCRP, 1988) The error associated with these systems is approximately 15%, with 5% associated with counting and 10% calibration errors (Bartlett *et al*, 1988, Gilvin and Bartlett, 1988)

Two types of plastic film were used during this project poly ally diglycol carbonate (supplied by National Radiological Protection Board, (NRPB[1]) and by Nuclear Enterprise Technologies, (NET[2]))(Gilvin and Bartlett, 1988) and the polymer polydiglycol carbonate (developed by Tastrak Ltd [3])(Camplin *et al*, 1988) These films are only sensitive to alpha emissions Other films have been developed to measure beta and gamma emissions, but these are not be considered in this thesis

The NRPB and NET dosemeters were provided as complete units with the film being sealed in plastic chambers The Tastrak film was supplied separately, and sealed chambers had to be constructed This involved sticking the film to the base of a plastic cup (or yogurt pot) and sealing the top with clingfilm using an elastic band This was usually done at the sampling location, the alpha sensitive film being wrapped in tin foil to prevent contamination during transportation Exposure periods of between 6 days and 3 months were employed for NRPB and NET detectors, while Tastrak detectors were generally exposed for 7 day with a 14 day maximum. The results from all track etch methods give equilibrium equivalent radon gas concentrations in Bq m^{-3}

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[1]NRPB National Radiological Protection Board, Chilton, Didcot, Oxon, England

[2]NET Nuclear Enterprise Technologies, Chilton, Didcot, Oxon, England

[3]Tastrak Limited, HH Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol, BS8 1TL, England

Comparison of Track Etch Detectors

NRPB and Tastrak detectors were used during the nationwide cave radon survey, in which 250 detectors were placed at 3 monthly intervals in the main caving regions of England and Wales for sampling periods of seven days (see chapter six) The majority were Tastrak films, while additional NRPB monitors were placed at approximately 30 sites in the Peak District The only detectors to be tested intrinsically were Tastrak films, with duplicate detectors being placed at 20 sites and sets of 5 detectors being located at 10 sites during the first survey period (1/8/91 - 10/8/91)

Results from duplicate measurements of radon gas using Tastrak detectors are shown in figure 5 1 The null hypothesis that results from duplicate measurements of radon gas using Tastrak detectors were significantly different was tested using the ANOVA (analysis of variance) technique and rejected (F crit = 0 884, F calc = 3 86), at the 95% significance level. Hence, it is concluded that no statistical difference exits between duplicate measurements of radon gas using Tastrak detectors Figure 5 2 demonstrates the variation between results within the data set At concentrations below 20,000 Bq m^{-3} the variation is 10 %, above 20,000 Bq m^{-3} the amount of divergence decreases to less than 10% The ANOVA analysis demonstrates that variations in results from Tastrak detectors are a function of random counting errors and all results are within ± 10% (McLaughlin *et al*, 1992)

The relationship between results from Tastrak and NRPB detectors, exposed at the same site and covering radon concentrations between 0 and 50,000 Bq m^{-3} are shown in figure 5 3 Duplicate sets of detectors were placed at 30 sampling locations in Derbyshire during all four sampling periods Figure 5 4 indicates that differences in recorded concentrations between each method occur over the entire measurement range The null hypothesis that results from NRPB and Tastrak detectors differ significantly was tested by ANOVA and rejected (F crit = 0 65, F calc = 3 87) Hence, results gained from the National project (chapter six) may be compared with those collected from an earlier project in Peak District caves (in chapter eight) where only NRPB detectors were used

(Gunn and Hyland, 1991) Although the results from the two methods do not differ significantly variations between ± 10 - 15 % are observed. However, extreme variations in excess of 40% were recorded with a maximum of 300% These variations can be explained by either one or a combination of the following

1 Tastrak detectors were counted manually and as such are likely to have a greater degree of error than the machine counted NRPB detectors, especially when concentrations are high and pits are overlapping, thus making them hard to differentiate

2 When concentrations are high (in homes and work places concentrations in excess of 1,000 Bq m^{-3} are considered high), Tastrak detectors have in excess of 1,500 pits per 1 cm^2 This makes counting exceptionally difficult and as a consequence small areas were counted and assumed to be representative of the overall sample. Either 1/6 or 1/8 of a square centimeter were counted in these cases The results from the small 'representative area' were then extrapolated The counting methodologies could be improved if an image analyzing system was employed, like that used by the NRPB and NET However, image analyzing systems still have difficulties in determining the concentrations when pits are overlapping Tastrak Ltd (Henshaw, 1993, per comms) feel that results from Tastrak detectors are likely to underestimate the 'true' radon concentration due to manual counting errors Results from this project support this view

3 Due to the design of Tastrak detectors, errors can be introduced if the sampling chamber is not sealed effectively but this can not occur with NRPB detectors which are supplied ready assembled Tastrak detectors had to be constructed at the sampling location and inadequate sealing could allow radon daughters to enter the sampling chamber This would result in the formation of pits arising from radon daughters, leading to higher concentrations being observed than if pure radon had been sampled This problem can be overcome by manual counting, whereby pits associated with radon daughters can be differentiated

from those associated with 'pure' radon, due to their non-random nature In all cases where contamination could have occurred, Tastrak Ltd identified the detectors concerned when they were returned Upon receipt the author investigated and recounted any contaminated detectors If the results were within ± 10 % of the original then the average was taken and used However, if the count was not within ± 10 % of the original count then the piece of film was recounted a further four times and the mean taken of the counts made by the author

4 Both NRPB and Tastrak detectors were designed to monitor radon concentration in homes where concentrations are generally low, and as a consequence the detector calibration is more sensitive at lower concentrations The maximum calibration limit for the NRPB detectors is 5,000 Bq m^{-3} (Gilvin and Bartlett, 1988) Yonehora (1987) outlines improved calibration and counting techniques which as yet have not been implemented by either NRPB or Tastrak Ltd

5 Increased humidity levels during exposure could introduce errors as calibration of all track etch detectors occurred in relatively low humidity environments (Gilvin and Bartlett, 1988) However, the effects of exposure to high humidity levels would be similar in all detector designs and therefore unlikely to cause errors between different methodologies

Although, in general, statistical analysis has demonstrated that the differences between results from different track etch detectors are not significant and therefore no standardisation of results was undertaken other international and inter laboratory comparisons have also found a high degree of correlation between results from different track etch detectors (Djeffal *et al*, 1992, Gilvin and Bartlett, 1988, Yonehora 1987) Different authors have found errors of between 5 and 15 % for different detector types, and all authors feel that no standardisation between results need occur (Djeffal *et al*,

1992, Gilvin and Bartlett, 1988, McLaughlin *et al*, 1992, Miles and Stinnaeve, 1986, Yonehora 1987)

5.2.2.2 Charcoal Canisters.

Activated charcoal has been found to absorb many gases from the atmosphere and in 1900 Rutherford discovered that radon was among these gases (NCRP, 1988) Since then health physicists have used charcoal's properties to absorb radon from the atmosphere to their advantage A theoretical and mathematical consideration of the absorption process can be found in Cohen and Cohen (1983) The detection method involves exposing canisters full of activated charcoal to the radon environment under consideration Exposure periods of up to 168 hours can be used, during which time absorption of radon occurs from the atmosphere into the charcoal bed of the canister The canisters are then closed, sealed and returned to the laboratory for counting An eight hour delay after sealing allows for the ingrowth of radon daughters to equilibrium with the absorbed radon (George, 1984, George *et al*, 1983) The canisters are counted for 200 seconds on a gamma spectrometer, using a Gi crystal, a photomultiplier and a multi-channel analyzer The gamma's arising from the radon daughters of lead-218 and bismuth-214 are counted and from the activity of the lead and bismuth peaks, the equilibrium equivalent radon activity can be determined The detection limit is circa 0 12 pCi l^{-1} (4 44 Bq m^{-3}) while the sensitivity is ± 10% at the 95% confidence level (Budnitz, 1974, NCRP, 1988) Gamma spectrometers can be fitted with NaI (Ti) crystal detectors, although these do not allow for as accurate a peak distinction as Gi detectors, and so were not used in this research

The major problem with the method is the preferential absorption of water over radon which, prior to correction, can lead to errors of 50% (Pensko, 1983 in NCRP, 1988, Shimo *et al*, 1987) All calculations in this thesis account for water gain and the sensitivity of the method is as reported above Detailed explanations of correction formulae can be found in Jenkins (1991) Due to the absorption of water in humid cave environments, the sampling period was decreased from the maximum of 168 hours to 96

hours in an attempt to reduce this error However, even this was found to be too long and the maximum sampling period was further reduced to 72 hours

Comparison of results from Charcoal Canisters

Seventy sets of duplicate charcoal canisters were exposed in ten caves in the Peak District The null hypothesis, that results from these duplicate measurements differed significantly was tested using ANOVA analysis and rejected at the 95% significance level (F crit = 0 936, F calc = 3 87) Figure 5 5 illustrates the relationship between results obtained from the duplicate charcoal canisters and figure 5 6 graphs the difference between results in the range from 0 to 40,000 Bq m^{-3} In all ranges the difference between duplicate canisters are generally small, less than ± 30% Development of the charcoal canister method occurred for use with domestic environments, with low humidity levels Therefore, the high humidity encountered in caves and the consequent effects of preferential absorption of water over radon could be a larger source of error in limestone caves (Geiger, 1971, George, 1984, Shimo *et al*, 1987)

It may be concluded that the charcoal method generally gives reproducible time integrated results over short sampling periods (less than 72 hours), although the known diurnal variations in radon concentrations are removed However, charcoal canisters allow for comparison of results between numerous sites to be made easily, during a single sampling period, at selective sites within a single cave Charcoal canisters were placed at a site where the Portable Radiation monitor was set to determine radon gas concentrations using the passive radon detector (see section 5 2 4) It was found that results gained from charcoal canisters are slightly below the maximum radon gas concentration derived from the Portable Radiation monitor (Michaels *et al*, 1987) On all occasions the charcoal canister results did not conform to either the mean or median radon concentration The tendency for the charcoal canisters to be slightly below the maximum radon gas concentration for the period can be explained by two features of the absorption process described by Cohen and Cohen (1983)

1 Radon will continuously be absorbed from the atmosphere into the charcoal bed as long as the radon concentrations in the atmosphere are rising or stable Therefore, the canister will absorb radon until it is at equilibrium with the atmosphere

2 When the atmospheric radon concentration decreases the charcoal bed will act as a source of radon and therefore radon will be exhaled from the bed to the atmosphere

The first of these points explains why charcoal canisters tend towards the maximum, while the second point explains why the results are slightly below the maximum. Further work is required to determine the relationship between the decrease in radon concentrations from the maximum with time

5.2.2.3 Portable Radiation Monitor (AB - 5)

The portable radiation monitor comprises a portable photomultiplier tube which is coupled to a passive radon detector comprising of a scintillating tube that allows radon to diffuse into a chamber Radon daughters are excluded by a thin membrane The radon decay results in the production of alpha particles and the alpha energy is converted to light by a zincsulfide scintillator (ZnS(Ag)) coating the sides of the passive detector (Budnitz, 1974) The amount of light is determined over set sampling periods and the radon concentration calculated The results are generally integrated over preset time periods, between either one or two hours, although the instrument has the capability of determining radon concentrations for intervals from 1 minute to 24 hour. However, the biggest constraints on the sampling interval are the available memory capacity, battery life, and the statistical validity of radon concentrations determined from relatively few counts recorded during shorter sampling periods The portable radiation monitor, when coupled with a passive radon detector, can determine radon concentrations for up to 4 days using its own internal battery or up to 30 days, if externally powered, with a sampling period of one hour

The method has a reported sensitivity of 1 5 cpm/pCi l^{-1}, the minimum detection limit being 37 Bq m^{-3} (1 0 pCi l^{-1}) Results from the passive detectors are given as equilibrium equivalent radon (Bq m^{-3})

5 2 3 Spot Measurements of radon gas

5.2.3.1 Lucas cells

The simplest and most common method of measuring radon gas concentrations at specific points in time uses Lucas cells, which consists of small metal chambers These are between 100 and 300 ml in size, with two inlet valves on the top, and a flat bottom closed with clear plastic The insides of the chambers are coated with zincsulfide scintillator (ZnS(Ag)) Samples are taken by drawing filtered air (to remove radon daughters) into the chambers, approximately 10 liters per minute are drawn through for 5 minutes These are then stored for 8 hours prior to counting on a photomultiplier tube The 8 hour delay allows for

1 The radon to decay and reach equilibrium with its daughters

2 Daughter products that were not removed during filtration to decay away

3 The products from radon-220 (thoron) and actinium to decay, due to their short half lives

This ensures that equilibrium equivalent radon concentrations are determined A lower detection limit of 370 Bq m^{-3} (10 pCi l^{-1}) has been quoted (Budnitz, 1974), but more sensitive results can be gained through longer sampling periods, and Lucas (1957) reports 1 1 mBq as the lowest detection limit at 95% confidence interval (NCRP, 1988)

Comparison of results from Lucas cells

One hundred and ten duplicate measurements were undertaken using lucas cells to test the null hypothesis that radon gas concentrations derived from duplicate measurements

differ significantly This was tested using the ANOVA technique and rejected (F crit = 0 828, F calc = 4 07) at the 95% confidence level. Figure 5 7 clearly illustrates the relationships between results from different Lucas cells sampled at the same site during a single sampling period. Figure 5 8 demonstrates that even though results are not significantly different variations in results occur However, in all instances the difference is between ± 15%

Although duplicate measurements do not differ significantly it is evident that errors can be introduced with the continued use of Lucas cells (Knoll, 1979, Michaels *et al*, 1987, Lai, 1991) In this study the primary source of errors (contamination of the cells with long lived radionuclides), was avoided by not taking samples or cleansing the cells whilst in the Radiochemistry Departments (at either Manchester Metropolitan University or The University of Huddersfield) where the potential to encounter long lived radionuclides is high Another potential source of error is due to the inability to reduce the background count of individual cells to less than 2 counts per minute This can result from

1 Contamination with long lived radionuclides

2 Build up of lead-214 as a product of radon decay, if high activity radon samples are collected

3 Failure to purge the cells of radon after counting

To avoid increasing the background count from these sources a rigorous cleansing technique was employed This involved purging the cells immediately after counting by drawing 10 litres per minute of filtered air from a relatively radon-free environment through the cell for ten minutes The cells were then left for a short period of time before counting again to ensure the background count had been reduced If not, the process was repeated until the background was below two counts per minute

5.2.3.2 The Two-filter Method

This method involves air being drawn into a tube over two filters, 8 3 cm in diameter at a set distance of 120 cm apart, at a rate of 2 litres per minute for 10 minutes The first filter excludes all radon daughter particles but allows radon gas to penetrate into the tube As the radon gas moves along the tube's length, a small fraction decays, resulting in the deposition of polonium-218 on the second filter paper Due to its' short half life, of 3 02 minutes, this needs to be counted as soon as sampling finishes The method has a very high sensitivity of 0 1 $pCil^{-1}$ (3 7 Bq m^{-3}) but errors introduced due to the sampling technique can be as high as ± 25 % A further problem is that due to the short elapse period, counting has to be performed within the cavern, where the background count could be relatively high This is especially important as the count rates of the polonium-218 will be very low Hence, in limestone caves, the increased accuracy of the method is negated by the increased background count associated with the sampling location Finally, the equipment is very large and bulky, including carrying the portable radiation monitor which is very sensitive and fragile Hence, it was decided that the method was not appropriate for use during this project

5.3 Summary of radon gas measuring techniques

Results from Tastrak detectors have been found to be reproducible with a precision of ± 10% (McLaughlin *et al*, 1992, Shimo *et al*, 1987) In order to test their accuracy results from Tastrak and NRPB detectors were compared and found not to be significantly different Therefore, it was concluded that Tastrak and NRPB give statistically comparable results and direct comparison of results can be undertaken

Time-integrated radon measuring techniques are important in determining the 'average' radon concentration at a particular site during a selected exposure period and in comparing different sites during the same exposure period However, they are always 'average' results that do not account for the variations in radon concentrations that are

known to exist Due to the relative ease of measurement, several detectors can be placed during a single sampling period to allow for the determination of radon concentrations on a wide spatial basis

Where multiple sets of charcoal detectors have been exposed during a single sampling period, variations in results have been recorded in excess of 10,000 Bq m^{-3} in all ranges of concentrations Due to the errors encountered, charcoal canisters were only used for comparative purposes within a single cave during a single sampling period

Results from Lucas cells were found to be highly accurate and reproducible but, due to the sampling technique, the results obtained are limited to a single site at the time of sampling only (Lai, 1991) No extrapolation of data to other areas of the cave or over time can be undertaken Spot measurements of radon, using Lucas cells, initially seem very reliable, but this reliability can only be achieved through careful maintenance of the cells to prevent contamination Due to the nature of spot measurements, the data are spatially and temporally limited However, the primary purpose of determining spot measurements of radon concentrations is to allow for the equilibrium factor between radon and the radon daughters to be calculated (Shimo *et al*, 1987)

5.4 Radon daughter detection methods

5 4 1 Introduction

In order to measure radon daughter concentrations it is necessary to collect a sample from the environment under study This is usually performed using pumps and filters because of the solid charged nature of the daughters The activity on the filters is then determined and the radon daughter concentration calculated from the total number of counts related to the delay period and sampling rate Filter papers of less than 0 8 micron pore size are able to collect 98 % of the solid particles that try to pass through them so that lost particles represent an insignificant source of error (Budnitz, 1974, NCRP, 1988) All methods stipulate a pump rate at which air should be drawn, and if

this rate is not maintained then errors result Hence, the pumps used must have a manometer so that the pump rate can be accurately controlled The rate is set at the start of each reading to compensate for changes in battery charge, air density and atmospheric pressure (Beckman, 1975) All of the three methods discussed below involve drawing air samples at a fixed rate over a filter paper and errors associated with sampling are usually larger than those attributable to the methodologies *per se*

5 4 2 Spot Measurements of radon daughter concentrations

5.4.2.1 The Kusnetz Method

The most common method for determining the potential alpha energy concentration is the Kusnetz method (Kusnetz, 1956), which became internationally accepted in 1973 when it was recommended in the USA as the industrial standard (NCRP, 1988) A fixed sample of air (usually 100 litres drawn at a rate of 10 litres per minute for 10 minutes) is drawn over a glass microfibre filter with a pore size of 8 μ which collects solid radon daughter particles from the atmosphere The filters are allowed to equilibrate for 40 - 90 minutes and the radioactive particles on them are counted using either a rate meter or a zinc sulphide scintillation detector connected to a photomultiplier tube (Budnitz, 1974) The delay allows for the decay of polonium-218 so that only alphas arising from polonium-214 are counted The filters are counted for two minutes, and using the mid-sampling time and mid-count time, the delays are calculated These are then applied to an empirical formula and the Kusnetz correction factor is determined (table 5 1) The radon daughter concentrations, in Working Levels, are calculated based upon the number of disintegrations per minute and the Kusnetz factor (Beckman, 1975, Kusnetz, 1956)

$$Rd = \frac{C2M}{12K}$$

Equation 5 - 1

Where Rd = Radon daughter concentration (WL), C2M = Counts per two minutes, K = Kusnetz factor

The method benefits from relative insensitivity to equilibrium of the three radon daughters during sampling Errors in mine atmospheres have been found to be ± 15% at 0 3 WL, but the percentage error declines at higher working levels to ± 3% at 5 WL (Budnitz, 1974, NCRP, 1988) The increased error at low concentrations is probably associated with random statistical fluctuation at low counting levels Budnitz (1974) and the NCRP (1988) examined the precision of the Kusnetz method and demonstrated that the methodological error decreased with increasing radon daughter concentrations and due to the background electrical noise within the instruments the instrumental errors are larger than those associated with counting errors due to the statistical fluctuations associated with radioactive decay

Comparison of results from the Kusnetz method

An experiment was set up to test the conclusions of Budnitz (1974) that multiple determinations of radon daughter concentrations using the Kusnetz method produce statistically similar results The null hypothesis that duplicate determinations of radon daughter concentrations using the Kusnetz method differ significantly was tested using ANOVA and rejected at the 95% confidence level (F crit = 0 91, F calc = 5 12) The duplicate results are all within the reported sensitivity of ± 15 % at 0 3 WL and ± 3 % at the 5 WL As errors can be introduced due to deficiencies in counting as well as sampling, the efficiency of the detecting equipment (Portable Radiation Monitor) was determined annually by the NRPB They found that for a 10 minute sample at 10 litres per minute and a 40 minute delay from a radon chamber with radon daughter concentrations of 0 24 and 1 1 WL at an equilibrium factor of 0 97, the detector efficiency was + 0 030 and - 0 003 for detecting polonium-214 this demonstrates an overall efficiency of 94% Therefore, due to the determined sensitivity any variations observed are primarily due to sampling errors rather than counting errors (Khan *et al*, 1987)

5.4.2.2 The Rolle Method

The Rolle method is a refinement of the Kusnetz method (Rolle, 1972) The sampling procedures are the same, but the count times vary, with each filter being counted 4 35 minutes after collection The count time is increased, when compared to the Kusnetz method, to 5 minutes for a 10 minute sample rather than two minutes This procedure results in the intrinsic errors being reduced to ± 12% at 0 3 WL due to increased count rates arising from polonium-218 that have not been allowed to decay (Rolle, 1972) Although the Rolle method is slightly more accurate than the Kusnetz method, a portable rate meter, or zinc sulphide scintillation detector connected to a photomultiplier tube, must be carried so that filters can be counted within approximately 5 minutes This poses no problems in homes and laboratories, but it is not possible in the majority of wild caves Hence, the Rolle method has had limited use within this study, although it has been used in the entrance series of some of the more accessible Show Caves The results obtained may be less accurate than would normally be the case due to the higher than normal background count in cave environments

5.4.2.3 The Instant Working Level Meter (IWLM) or Radon sniffer

The IWLM is a portable, compact instrument developed by Thompson and Neilson[4], that measure the total potential alpha energy or Working Level (Thompson and Neilson, 1988) It draws a smaller quantity of air than either the Rolle or Kusnetz methods but has increased flexibility because the sampling periods are variable This allows the operator to sample until the errors associated with the statistical fluctuations of radioactive decay are reduced The IWLM draws 1 litre of air per minute over a 25 mm filter of pore size 8 µ for a pre-set time period, during which the total counts are recorded on a digital display The radon daughters are deposited upon the filter and the resulting energy from the decay of alphas is measured by the instrument The total

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[4] Mensura Technology Limited, 32 Greenfield Avenue, Parbold, Wigan, Lancs, WN8 7DH, England

counts for each sampling period may be converted into Working Levels, using the general formula

$$L = \frac{n}{(t - 0\ 5)C}$$

Equation 5 - 2

Where WL the radon daughter concentration in Working Levels, n is the total counts, t the sampling period and C the instrument calibration factor

During the course of the study two IWLM were used, each being independently calibrated with standard sources and hence having a different calibration factor For both instruments, equation 5 2 was used to calculated radon daughter concentrations in WL, for sampling periods in excess of 1 hour (Thompson and Neilson, 1988) Calibration factors of 8500 and 7100 were used for IWLM 6 8 and 7 1 respectively For shorter sampling periods (less than half an hour), radon daughter concentrations were calculated using equation 5 3 for IWLM 6 8 or equation 5 4 for IWLM 7 1

$$Rd = 0\,164t^{-1\,58C}$$

Equation 5 - 3

$$Rd = 0\,194t^{-1\,58C}$$

Equation 5 - 4

Where Rd = radon daughter concentrations (WL), t = sample time (minutes), C = total counts in time t

Equations 5 3 and 5 4 were derived by comparing results obtained from Instant Working Meters run for sampling periods of less than 30 minutes and comparing the results with

those derived from the same site during the same sampling period with radon daughter concentrations derived via the Kusnetz method

Both instruments have a reported accuracy of ± 20 % for sampling periods over 6 hours with atmospheric radon daughter concentrations of 5 mWL For shorter sampling periods, accuracy decreases to ± 25 % at atmospheric radon daughter concentrations of 20 mWL (Thompson and Nielsen, 1988) Calibrations were performed in radon chambers with maximum radon concentrations of 600 Bq m^{-3}, while radon daughter concentrations were varied between 0 01 - 0 06 WL (Bigu, 1987)

Comparison of results from IWLM

As the IWLM are light and compact they were used to obtain most of the measurements in the detailed investigation of spatial and temporal variations of radon daughter concentrations in wild caves Hence, it was important to compare the results obtained from the IWLM with those from more established methods, such as the Kusnetz or Rolle methods This was achieved during a detailed investigation of instrumentation over an initial three month period, although comparisons continued to be made during the course of the project to ensure no subsequent deviation occurred The two IWLM used during the course of the project were compared on a number of occasions and the repeatability of the results are illustrated in Figures 5 9 A null hypothesis that the radon daughter concentrations from the two IWLM differ significantly was tested using ANOVA and rejected (F crit = 0 764, F calc = 4 88) at the 95% significance level. Figure 5 10 demonstrates that at radon daughter concentrations below 2 WL the differences between the two instruments are usually relatively small and can be accounted for by radon variations (low count rates) Above 2 WL the absolute differences between instruments increase, but they remain small (less than ± 10%) and as the ANOVA results demonstrate the variations are not significant

Comparison of results from the Kusnetz method and Instant Working Level Meters

As the Kusnetz method has a reported sensitivity of ± 3 - 15 % which is lower than that of the IWLM (± 20%) it was felt that both methods should be compared and the results from the IWLM standardised to be 'Kusnetz equivalent' if necessary Comparisons were undertaken where both of the IWLM were run for 20 minutes and an average from the two machines was compared with the average of two ten minute Kusnetz results Figure 5 11 demonstrates that results from the Kusnetz method are similar to those from IWLM The null hypothesis that radon daughter concentrations measured using IWLM differ significantly from those measured using the Kusnetz method, was tested using ANOVA analysis and rejected at the 95% significance level (F crit = 0 764, F calc = 3 88) Figure 5 12 identifies variations of up to 0 8 WL between individual concentrations determined by the Kusnetz method and the IWLM However, it is felt that errors resulting from sampling are larger than methodological errors and this has been confirmed by a number of authors (for example, NCRP, 1988 , Weng *et al*, 1992) Errors associated with sampling can range from inconsistencies in the instrument pump rate to the influence of position and height of the instrument within the passage understudy due to small scale variation in radon concentrations

5 4 3 Continuous Measurements of Radon Daughter Concentrations

The only method available to the author by which continuous measurements of radon daughter concentrations could be made was by coupling a IWLM to a data logging system. Measurements were then made on different time intervals, from half an hour to 24 hour intervals The logging equipment has the capability to record up to 36 days at half hourly intervals but data was rarely collected for longer that 7 days from wild caves as a result either of instrument failure, resulting from high humidity levels encountered in the caves, or from battery failure, the maximum sampling period being about four days

on a single battery However, in Show Caves, data for the maximum 36 days sampling were collected

The coupling of an IWLM with a data logging system allowed for detailed investigation of temporal variations in radon daughter concentrations Continuous measurements of radon daughters were combined with results from the Passive Portable Radiation monitor, thus determining radon gas concentrations, and allowing the temporal variation of radon gas, radon daughter concentrations and equilibrium factors to be determined The instrument has the same sensitivity as the individual IWLM, described in section 5 4 3 2, at ± 20 % at 5 mWL and ± 25 % at 20 mWL

5.5 Summary of radon daughter measuring techniques

1 The Rolle method has the lowest reported associated error, and therefore in theory should have been used However, when used in limestone caves the sensitivity cannot be ensured due to high background counts Therefore, the method was only rarely used during this thesis

2 The Kusnetz method was used where possible, that is at sites where filters could be analysed within 90 minutes These sites were primarily in Show Caves or in the entrance series to Wild Caves

3 The majority of concentrations in Wild Caves were obtained using IWLM due to their compact, light and robust nature

4 The two IWLM produce comparable results (± 20%) which are reproducable to within (± 20%)

5 Differences between concentrations measured by IWLM and the Kusnetz method are particularly evident below 0 1 WL and above 2 WL. However, it has been demonstrated that these differences are not statistically significant.

5.6 Sampling protocol for radon and radon daughters in limestone caves.

To try to overcome and standardise any errors introduced through sampling, a rigid sampling protocol was devised and adhered to during the course of all measurements performed by the author The sampling protocol for the national survey of radon in caves (see chapter six for details), using Tastrak and NRPB monitors, was not as stringent due to the number of different people involved with the placement and removal of detectors One potential error concerns detector location and although it was stressed that detectors should be located at exactly the same site during each sampling period, this was not guaranteed

The author always ensured that all radon and radon daughter measurements satisfied the following criteria:

1 All measurements were taken at exactly the same site during different sampling runs

2. Measurements were taken as close to the ground as possible, generally within 30 cm.

3 Only filtered air was drawn into Lucas cells, the intake hose being located as close to either the IWLM or Kusnetz pumps as possible This increases the validity of calculated equilibrium factors

4 All pumps were positioned at right-angles to the passageways This made the determination of placement easier than if positions facing up or downdraft were used

5 IWLM were always run for a minimum of 15 minutes However, if 150 counts (an associated counting error of 8 2 %) had not been recorded in this period, sampling was continued until this figure was reached as a minimum.

6 All pumps were checked at the start and during each measurement period to ensure that the correct volume of air was sampled Adjustments were made as necessary

7 The IWLM were calibrated on a monthly basis using calibrated check sources provided by Thompson and Neilson

8 The Portable Radiation Monitor was calibrated annually

CHAPTER SIX: RADON CONCENTRATIONS WITHIN SELECTED LIMESTONE CAVES OF ENGLAND AND WALES

6.1 Introduction

Previous workers have shown that radon and radon daughter concentrations within limestone caves exhibit both temporal and spatial variations at a variety of scales (e g Gunn, *et al*, 1989a and b, 1991, Kobal *et al*, 1987, Wilkening and Watkins, 1976, Yarborough, 1977, 1982). Various explanations have been given to account for these variations, primarily stressing the interrelationships of radon and radon daughter concentrations with cave microclimates (see chapter three) However, prior to this thesis the variations in radon concentrations within the caves of England and Wales were only partially understood Hence, it was decided to undertake a survey of the main caving areas of England and Wales, with the following primary objectives

1 To establish typical radon gas concentrations

2 To examine the spatial variability in radon gas concentrations within each individual caving area

3 To determine the extent of seasonal variations in cave radon gas concentrations

4 To evaluate the effects of cave morphology and geological setting on radon gas concentrations

5 To estimate the radiation dose which may potentially be accrued by cave users

6.2 The measurement of radon and radon daughters in limestone caves

Having evaluated the various techniques for the determination of radon and radon daughter concentrations (chapter five) it was decided that track etch methods were the most appropriate for the survey Both Tastrak and NRPB detectors were used and it was established that there is no statistical significantly difference between the two techniques (chapter five)

To establish seasonal variations, sampling was undertaken over four periods, each of one week duration, during each of the four seasons of the year The sampling periods were 2 - 11 August 1991, 1 - 10 November 1991, 31 January - 9 February 1992 and 3 -12 May 1992 During each sampling period 250 Tastrak and 55 NRPB detectors were deployed The original intention was to place all the detectors in the study caves during the same weekend and to remove them all in the following weekend However, in practice this was not always achieved due to time pressures on the individuals aiding in the placement and removal of detectors Nevertheless, in all cases the detectors were only in place for 6 to 8 days, even if placement and removal was not during the preset dates

6.3 Location of Detectors

6 3 1 Geographical area

There are over 3,000 caves in Great Britain (Hardwick, 1994) but only 250 detectors were available for each sampling period Hence, considerable attention was given to designing a sampling strategy which would be as representative as possible of the regions under study The majority of accessible caves are located within seven regions and there is great variability in the type and nature of cave passage within each region (table 6 1, Hardwick, 1994) The North Pennines region contains the majority of both cave entrances and accessible passage (62% and 52 8% respectively) while the least number of entrances and smallest amount of accessible cave passage is in North Wales (2 6% and

1 7% respectively) There are also a small number of caves outside the main areas which are grouped under "other" and have 0 1% of entrances and total accessible passage

As Scotland's caves are distant and widely scattered, it was decided for logistical reasons to exclude this region from the survey The North Wales caves were also excluded as they form only a small percentage of the total and would have received only 1 or 2 detectors if allocated on a proportionate basis Finally, it was decided to exclude caves from lithologies other than the Carboniferous limestones, although subsequently a small number of caves in the Jurassic Limestone were included in the survey Therefore, the study initially concentrated on the four major caving regions the North Pennines, South Wales, the Mendip Hills and the Peak District These four contain over 86 7 % of the cave entrances and 93.7 % of the total accessible cave passage in Great Britain

6 3 2 Distribution of detectors within the four major caving regions

To determine how many detectors were to be allocated to each region it was necessary to derive an index that accounted for the spatial variability and different morphology of the caves in each area Three indices were considered

1 The total number of cave entrances

2 The total length cave passage

3 The mean passage length per entrance, (total passage length divided by the total number of entrances)

As indices 1 and 2 produce the same rank (table 6 1) it was decided to eliminate index 2 from further consideration Table 6 2 shows the mean passage length per entrance and number of cave entrances for the four caving regions under study The South Wales area has the highest mean passage length per entrance (0 949 km/ent) as the caves in this area are generally of an extensive nature, large amounts of passage being associated with each entrance Conversely, the North Pennine region has the lowest mean passage length per

entrance (0 190 km/ent) due to a predominance of relatively short often vertical caves The other two areas, the Mendip Hills and the Peak District, are very similar with mean passage lengths per entrance of 0 273 km/ent and 0 210 km/ent respectively

Given the range of values for entrances and mean passage length it became clear that if either index were used alone as a criterion for distributing detectors to caving regions, a skewed distribution would result, with either the South Wales or the North Pennine region receiving the majority of detectors (table 6 3) Using the number of cave entrances as a criterion, the majority of detectors (77 %) would be located in the North Pennine region, the South Wales region would receive only 7 %, and the Peak District and the Mendip Hills 8 % If mean passage length were to be used as a criterion, South Wales would receive the majority (58 %), the North Pennines the least (12 %), the Mendip Hills (17 %) and the Peak District (13 %) Therefore, to ensure the distribution of monitors was as representative as possible, the initial allocation to each region used the mean number of detectors derived from indices 1 and 3 (table 6 4)

As the author alone could not place and remove all the detectors during the predetermined sampling dates, local cavers were enlisted to help This was achieved through consultation with the regional caving bodies, three of whom proved extremely helpful. However, the Council of Southern Caving Clubs, representing the Mendip Hills region, offered only limited help Persons from this region who had initially expressed an interest in the project subsequently withdrew and the Council, acting on behalf of its members, finally decided that no radon surveys should be undertaken in the region's caves Therefore, the monitors from the Mendip Hills region were redistributed to other caving regions Primarily, the detectors were spread over the other three major caving regions, but a small number were allocated to the Portland region, which is situated in close geographical proximity to the Mendip Hills but is in Jurassic and not Carboniferous limestones The initial remit of the project was solely to investigate radon concentrations in caves within the Carboniferous limestones and therefore this additional area did not fall within the project definition However, the results were included as they provide an indication of the effect which the different emanation and exhalation rates of the two

limestones have on observed radon concentrations In addition, it was possible to allocate four detectors to a Show Cave in the Mendip Hills

6 3 3 Distribution of detectors within individual caving regions

Having established the number of detectors to be placed within each caving region it was necessary to determine which caves were to receive detectors and the number and location of detectors in each cave In order to reduce the sample size it was decided to focus on areas within each region which lay within the boundaries of cave Sites of Special Scientific Interest (SSSI) This criterion was used for three reasons

1 The forty eight British cave SSSI include within their boundaries over 32 % of the known cave entrances and, more importantly, over 75 % of known accessible cave passage

2 The majority of the longer and more popular caves are located within SSSI It was felt to be important to include the longer and more popular caving trips to facilitate the quantification of the potential radiation dose received by cavers from radon gas

3 To provide radiological information about the caves contained within the SSSI boundaries that can be related to their regional, geological and geomorphological context

The final choice of caves was also influenced by accessibility (with regard to ease of placement and collection of detectors) and popularity, to enable transposition of the results obtained onto the majority of the caving population For consistency, both the number of entrances and the mean passage length should have been used in conjunction with SSSI boundaries to locate detectors within each area However, only the length of passage was used in conjunction with SSSI boundaries for two reasons

1 Passage length includes 75 % of known accessible passage in Great Britain and is therefore more valid than the 32 % of cave entrances within the SSSI boundaries

2 Passage length gives a better indication of the likely sporting nature of a cave and hence can be used as a surrogate to determine the number of potential visitors This allowed a larger number of popular caves to be sampled

6.3.3.1 North Pennines

The North Pennines region includes the area from Alnwick in Northumberland in the northeast, across to Arnside in Cumbria to the west, and down to Grassington, North Yorkshire in the south (figure 6 1) There are twenty cave SSSI which encompass over 71% of the total accessible cave passage in the area These received 110 detectors (44 0 % of total number of detectors allocated) during each sampling period (table 6 5) It was felt that a minimum of three detectors per cave were needed to provide a representative sample of cave radon concentrations Hence, those SSSI which would have received less than three detectors on the basis of the passage length criteria were excluded from the survey The detectors which would have been allocated to these areas were then redistributed to the other areas in the region However, the Dow Cave SSSI, which should have been rejected on these grounds was included because an interested party was willing to place and remove the detectors from this location The Brants Gill catchment was excluded as volunteers could not be found who would place or remove detectors, primarily due to the dangerous nature of the major caves due to either their instability or their tendency to flood

Once the number of detectors to be allocated to each SSSI had been determined it was necessary to decide which individual cave would receive the detectors and where within the cave they would be located Individual caves within each SSSI were chosen to ensure that all of the major caves were sampled, this decision being based upon passage length and consultations with local cavers from the region Within each cave, detectors were

located on the most popular route followed by visitors Having taken all these factors into consideration, the caves in which detectors were located are listed in table 6 6

6.3.3.2 South Wales and the Forest of Dean

The South Wales caving region includes the fringes of the South Wales coalfields and the Welsh coastline, enclosing the area from Crickhowell in the south east to Kidwell in the south west down to Cardiff in the south (figure 6 2) The caves in the Forest of Dean region are generally included with those of South Wales (e g Ford, 1989) and this practice was continued in the radon survey, the combined area being referred to solely as the South Wales region There are five SSSI in the South Wales caving region and they contain over 87 % of the accessible cave passage in the area Table 6 7 lists the five areas, the accessible passage length, the theoretical distribution and actual number of detectors that were allocated Given the passage length associated with the Dan-yr-Ogof system it should have received eight detectors, 12 % of the South Wales total. However, two large fans have been installed to reduce radon concentrations in the Dan-yr-Ogof and Cathedral Tourist Caves, and these have been found to influence the radon concentrations throughout the cave systems (regular measurements undertaken in the Tourist Cave are included in appendix 1) Hence, the eight detectors were allocated to the other SSSI and the final distribution of detectors is shown in table 6 8 In total, nine caves were investigated, the largest number of detectors being placed in Ogof Agen Allwedd (20) in the Mynydd Llangattwg area, and the least in Tunnel Cave (3) in the Little Neath River Cave area Although results were gained from Little Neath River Cave during the first sample period (August 1991) there were problems with the 'volunteer helpers' and the detectors from subsequent sampling periods were either not placed or not returned Hence, this area was not included in the main discussion but the available results are included in Appendix 1

6.3.3.3 Mendip Hills

The Mendip Hills limestone outcrop extends from Frome in the east to Axbridge in the west, with isolated outcrops extending further west to Weston-super-Mare and north to the Bristol channel and the City of Bristol, including the Avon Gorge (figure 6 3) Based upon SSSI boundaries the region can be divided into five areas, which contain 80 % of the total accessible cave passage in the area In theory this region should have received 31 detectors during each sampling period but due to logistical problems only 4 detectors were placed Table 6 9 lists the 5 areas, the proposed study caves and the number of detectors that would have been allocated to each The four detectors placed in this region were concentrated in two caves in the Cheddar area Spot results obtained from some of the other caves outlined in table 6 9 are included in the following discussion, but the different sampling method used may mean they are not directly comparable. Most of the detectors that would have been allocated to the Mendip Hills were relocated to the other three main caving regions, but six were allocated to Portland, a small caving region situated close to the Mendip Hills

6.3.3.4 The Peak District

The Peak District limestone outcrop lies predominantly in Derbyshire together with small parts of Staffordshire Castleton marks the northerly border, Matlock the easterly and Ashbourne the southerly (figure 6 4) According to the protocol outlined in section 6 4 2, this region should have received 36 detectors during each survey period However, additional sites were investigated by placing NRPB monitors to supplement the Tastrak film bringing the total to 44 (table 6 10) Twenty three detectors were placed in the Castleton SSSI (Peak Cavern, P8, Giants Hole and Gautries Hole), eight detectors in Stoney Middleton Dale SSSI (Carlswark Cavern and Streaks Pot), five in Bradwell Dale SSSI (Bagshaw Cavern), four in the Upper Lathkill Dale SSSI (Hillocks Mine and Knotlow Mine), and four in Matlock SSSI (Devonshire Mine and Jug Holes Mine)

6.3.3.5 Portland

Due to the limited geographical extent of this region the same criteria as for the other regions could not be used for the placement of detectors, and sites were primarily decided upon by local cavers, according to the accessibility and popularity of the caverns Six detectors were placed during each sampling period As the detectors were redistributed from the Mendip Hills region, the first survey period was missed

6.4 Results

6 4 1 Introduction

Tables 6 11 and 6 13 summarise the results from the 1991 - 1992 national cave radon survey Of the 1000 Tastrak detectors placed during the four survey periods, results were gained from 820. Of the losses, forty seven can be accounted for by non-returns from the South Wales region while the rest are presumed to have been lost due to flooding or operator error (primarily not being able to re-find the sampling site)

The regions with the highest and lowest overall means are the Peak District (8,528 Bq m^{-3}) and Portland regions (454 Bq m^{-3}) respectively Between these range South Wales (2,601 Bq m^{-3}), the Mendip Hills (1,129 Bq m^{-3}) and the North Pennines (1,116 Bq m^{-3}) However, the limited number of detectors from the Mendip Hills and Portland regions limits the inferences that may be drawn As the mean concentrations from the Mendip Hills and North Pennines regions are close, the final positions in the ranking could be interchangeable Gunn *et al* (1991) and Reaich and Kerr (1991) found higher concentrations in the Mendip Hills region than those reported during this project Seasonal variations are also apparent in the results from all regions In all five regions the standard deviation is either close to or above the mean implying considerable variations in radon concentrations

6 4 2 International comparison

Table 6 12 outlines a selection of published radon concentrations from caves around the world (for more detailed discussion see chapter three) Mean, maximum and minimum radon concentrations (Bq m^{-3}) are tabulated together with the number of samples taken and whether the results are based upon spot or integrated measurements (see chapter five) In the present survey the maximum concentration integrated during a seven day period at a single site was from Giants Hole in the Peak District (46,080 Bq m^{-3}) Kelly (1994) reports that a survey undertaken by a Dublin High School recorded a concentration of 190,000 Bq m^{-3} from Dunmore cave in Ireland, the highest to date for any cave in the world However, the author has not been able to obtain a copy of the full report Previously the maximum reported concentration was by Gunn *et al* (1991) who recorded spot radon daughter concentrations in excess of 42 WL (circa 155,000 Bq m^{-3}) in Giant's Hole. Initially this was assumed to be an isolated occurrence, but during the summer of 1992 members of the British Geological Survey recorded radon concentrations in excess of 135,000 Bq m^{-3} on several occasions (Ball 1992, pers coms) Maximum concentrations in the same order of magnitude as the maximum from this project were measured by Pillar and Surbeck (1989) who recorded concentrations of 40,000 Bq m^{-3}, in caves in Switzerland, and by Stelcl, (1991) who recorded 40,000 Bq m^{-3} in Czechoslovakian caves. However, in both cases these were spot measurements, whereas the results from this project were time integrated and therefore not directly comparable

Three caves in the United States have spot maximum radon concentrations in excess of 20,000 Bq m^{-3}, Mammoth cave (33,226 Bq m^{-3}, Eheman *et al*, 1991), Bat cave (25,530 Bq m^{-3}, Yarborough, 1976), and Stanton cave (24,272 Bq m^{-3}, Yarborough, 1976) However, the mean of the reported maxima for the United States (recorded in table 6 12) is approximately 11,500 Bq m^{-3}. Other areas of the world have lower maximum radon concentrations, which range from 480 Bq m^{-3} in Italy (Cigna, 1986) to 14,000 Bq m^{-3} in Hungary (Somogyi *et al*, 1989) It is not possible to carry out a similar comparison for

mean concentrations as differences in the size of the data sets can lead to statistical errors and because American workers generally publish weighted average results that are not directly comparable with the means presented from this project Also, it is not always clear whether the reported mean values are based upon spot or integrated measurements which will give different results

In global terms, the maximum results from the other caving regions of England and Wales are not as extraordinary as those reported for the Peak District The site maxima from the North Pennine and South Wales regions (27,136 and 19,968 Bq m^{-3} respectively) tend towards the upper portion of those recorded in the United States and are higher than those recorded in other countries apart from Switzerland and Czechoslovakia Conversely, the results from the Mendip Hills and Portland regions are below the means for the United States, Spain (Fernandez *et al*, 1984), Hungary (Geczy *et al*, 1989, Somogyi *et al*, 1989), the former USSR (Gunn, 1991), and the former Yugoslavia (Kobal *et al*, 1987), but above those reported in South Africa (Gamble, 1981) and Italy (Cigna, 1986) The comparatively low results gained from the Mendip Hills region could be a consequence of detector location and it should be noted that relatively few results were obtained from this region Work by Reaich and Kerr (1991) and Gunn *et al* (1991) in this region indicate levels in excess of those encountered during this project Their results indicate that maximum concentrations encountered in the Mendip Hills region are around 19,000 Bq m^{-3}, which is similar to the concentrations observed in the North Pennine and South Wales regions during this project

6 4 3 Comparison of the five caving regions

The majority of workers have considered the temporal variations in radon and radon daughter concentration on either a diurnal or seasonal basis within individual caves (see chapter three) The present project is thought to be the first to investigate variations in cave radon concentrations at a national level, examining inter-regional variation, (using data from different caving regions) and intra-regional variation, (comparing data from different areas within each region) However, national comparisons based solely on the

number of samples may not always be valid as the geographical extent of a single caving region in the USA (Kentucky for example) may be larger than the whole of all the caving regions in the United Kingdom. Similarly Mammoth cave in the USA contains more passage than all the caves included in this project With these factors in mind this section considers inter-regional variations and the following section, discusses intra-regional variations

Four authors have undertaken country-wide reviews of cave radon concentrations Kobal *et al* (1988) discussed results from several Show Caves in Yugoslavia However, the authors considered only the potential threat to health and did not investigate the reasons why this threat varied from cave to cave or region to region Carson (1981) examined radon data from caves across the USA, most results being from caves operated as Show Caves by the National Park Service He provided a detailed discussion regarding Mammoth Cave, primarily directed towards the potential health implications Yarborough (1977, 1982) also discussed the problems of radon in National Park Service caves across the USA. He established a cave radon database but did not attempt to account for the regional variations that were apparent Considerable attention was put into accounting for spatial and temporal variations within individual caves and the majority of our knowledge on the relationships between cave radon concentrations and cave microclimate comes from this source

A null hypothesis that radon concentrations from different regions were not significantly different from each other was tested using Student T-Tests It was found that at the 95% probability level radon concentrations from the Portland, South Wales and Peak District regions are all from distinct populations Concentrations in the North Pennines and the Mendip Hills regions are not from distinct populations and can be grouped together The results from the North Pennines and Mendip Hills region are from a population that is different from the individual populations representing the Portland, South Wales and Peak District regions However, the number of results from the Mendip Hills region limits the conclusions that can be drawn for this region. The differences in radon concentrations will be discussed in the remainder of this section and the seasonal factors

which are also important in controlling mean concentrations are considered further in section 6 5 6 and chapters eight and nine

Analysis of cave radon concentrations by region has highlighted that significant variation occurs between results from different regions However, it is important to recognise that a region in the British context may be much smaller than a region in other countries For example, the Kentuck region of the USA has more cave passage than the whole of Great Britain Therefore, additional analysis was performed to identify, on a national scale, any other variables that might influence cave radon concentrations

Overall mean radon concentrations, and means by season were ranked in descending order and caves were classified into four major morphological categories Although the classifications of RSU and USD caves as proposed by Yarborough *et al* (1976) were used initially, it was felt that these two broad categories do not fully encompass the range of cave morphologies encountered Therefore, two additional categories were included, pots (caves with considerable vertical development, e g Lost Johns Caverns in the North Pennines region) and systems (caves with considerable amounts of passage, lateral and vertical development, and several entrances, e g Ease Gill Caverns in the North Pennines region and Ogof Ffynnon Ddu in the South Wales region) Additional characteristics noted included total passage length, total depth, altitude of the highest entrance, presence of an active streamway, and geology at the highest entrance Statistical analyses (in the form of Student T Tests) were undertaken to determine whether any of these variables influenced cave radon concentrations No relationships were found between any of the variables and either the overall or seasonal mean radon concentrations Therefore, even though there are considerable limitations on the analysis of radon concentrations by region it is the only variable by which a significant statistical relationship has been demonstrated for the data

Two mechanisms can be proposed to account for the inter regional variations in mean radon concentrations variations in the radon budget and variations in the effectiveness of

the dispersion / accumulation mechanisms In order to investigate these more thoroughly it would be necessary to

1 Identify those sources of radon within limestone caves that contribute to the overall radon budget The sources are thought to be the containing limestone rock and associated mineral veins, sediment, and water

2 Quantify variations in emanation rates for each different source in each area and how they affect the overall radon budget

The mean and maximum radon concentration from the Peak District is significantly higher than concentrations recorded in all other regions This could be a result of high uranium-238 concentrations in the basal Namurian shales which were deposited after the limestones Some of this sequence will have been removed by erosion and provide a source of uranium in cave sediments In contrast, apart from the Swindon Hill region (Wrixon *et al*, 1988), the allogenic sediments in the North Pennine region are not uranium-rich and the lower mean radon concentrations may be a result of an absence of uranium.

Even though maxima for the South Wales and North Pennines regions are broadly similar (19,968 Bq m^{-3} and 27,136 Bq m^{-3}, respectively) analysis using the ANOVA technique identified a statistical difference between these two regions (F crit = 3 86, F calc = 35 9 at the 0 005 significance level) This difference is reflected in the mean concentrations (South Wales 2,561 Bq m^{-3} and the North Pennines 1,115 Bq m^{-3}) The difference in observed concentrations in each region could be a result either of variations in the dispersion / accumulation processes or of differences in the radon budget

The effects of variation in the dispersion / accumulation mechanisms of radon can be seen by comparing the cave morphology and mean radon concentrations from the North Pennines and South Wales regions The caves from the South Wales region generally

have a high mean passage length per entrance (0 949 km/ent), while the caves from the North Pennines region have less passage associated with each entrance (mean passage length per entrance of 0 190 km/ent) in particular due to the larger percentage of vertical systems Variations in cave morphology control and alter the methods by which radon is either dispersed or accumulates within a cave Differences in either the dispersion or accumulation mechanisms in South Wales are a consequence of the relatively few entrances in comparison to the associated passage length, resulting in less air movement through the caves to remove the radon Circulation in the North Pennines region is likely to be relatively greater as there are more entrances to induce the air to move through the cave systems, reducing radon concentrations through dispersion Therefore, the mean radon concentrations in these two areas will reflect the amount of air moving through the associated systems, which is controlled by the cave morphology Yarborough *et al* (1976) showed that the radon concentrations is related to the quantity of air moving through a cave system. Seasonal variations, related to differences in air movement, both in the direction and quantity of air flowing within individual caves, have been found to affect radon concentrations (Yarborough, 1976, 1977, Wilkening and Watkins, 1976, Somogyi *et al*, 1989, Miki and Ikeya, 1980, Surbeck and Medici, 1990) Therefore, differences in general cave morphologies affect ventilation rates and hence mean and maximum radon concentrations within different regions

However, cave morphology is only one component of the multifaceted cave radon system, another influence being the emanating surface area of the cave The caves of South Wales have proportionally larger emanating surface than those of the North Pennines region due to their length and average passage size Therefore as air moves through the cave systems in South Wales it will be in contact with proportionally more sources of radon than in the North Pennines Therefore, the differences between concentrations in the two regions may be a function of

1 the emanating surface area in each region

2 air movements

3 the different rates of emanation

In theory, if the radon budget solely controlled observed radon concentrations in each geological region, then maxima would be expected to follow a similar pattern to those for the mean concentrations, higher release rates resulting in more radon generation and therefore higher mean and maximum radon concentrations Table 6 11 shows that this is not always the case and there is one noticeable exception the maximum for the North Pennine region is ranked second, while the ranking for the mean radon concentration for this area was fifth This implies that other factors must be operating in addition to the total radon budget to control the maximum radon concentrations observed Two factors could have resulted in the increase in the maximum.

1 Accumulation and dispersion mechanisms, where localised microclimates within a single cave system result in the redistribution and accumulation of radon at a single site thereby increasing observed concentrations

2 Localised radon sources, such as elevated uranium concentrations at a single site (perhaps related to differences in mineralisation) may result in an increase in the radon emanation rate

If a localised radon source were the cause of the elevated maximum radon concentrations they might only be evident during the summer sampling period as incoming relatively radon free air entering the cave during the winter periods would dilute the cavern radon concentrations As the observed maximum occurred at a site situated 4 km from any known entrance during the November 1991 sampling period, it seems more likely that localised dispersion / accumulation were responsible However, if the maximum concentration is controlled by localised radon sources then the maxima observed must reflect the total radon emanation In theory each section of cave passage will have a maximum radon concentration, dependent solely upon exhalation, if no additional radon is added or removed from the section of passage under investigation

Although there are only 18 readings, Portland does appear to have significantly lower concentrations than the other regions This is due to lower uranium-238 in both the limestone strata and the allogenic sediment contained within the cave (Ball, 1993 pers coms) When this is combined with the short nature and multiple entrances in the caves the radon concentrations would be expected to be low

The minimum concentrations shown in table 6 11 are a function of the distance of detectors from the entrance of the cave and relate to the effects of relatively radon free air flowing into the cave, primarily during the winter months The minimum recorded in the Peak District was from Peak Cavern, Castleton, the detector being situated approximately 12 m inside the large entrance of the cavern, while the minimum concentrations from the Mendip Hills and South Wales regions were from sites at approximately 35 and 120 metres respectively from the entrance The trend of the minimum radon concentrations to increase with increased distance into the cavern is discussed further in section 6 5 5

6 4 4 Intra-regional Variation

6.4.4.1 North Pennines

Tables 6 14 and 6 17 summarise the results from the 6 SSSI in the North Pennine region The mean results, from all four sampling periods, fall into 2 categories and the same general trend is portrayed in the results from individual sampling periods

1 Areas with overall mean radon concentrations above 1,500 Bq m^{-3} (Leck Beck Fell, Stump Cross and Upper Nidderdale SSSI)

2 Areas with mean radon concentrations below 800 Bq m^{-3} (Kingsdale, Ingleborough Hill and Dow Cave SSSI)

The maximum recorded radon concentrations for the Kingsdale, Ingleborough Hill, Stump Cross and the Upper Nidderdale areas are all similar (5,300 - 6,500 Bq m^{-3}) but

the maximum from Leck Beck Fell is approximately 4 times larger at 27,136 Bq m^{-3}, and that from Dow cave area is much lower (1,392 Bq m^{-3}) Three possible explanations may be put forward to account for the division of the mean radon concentrations for the whole sampling period into two sets

Explanation 1

The areas with a high mean radon concentration (Leck Beck Fell, Stump Cross and Upper Nidderdale) are located close to the edge of the limestone outcrop, which is controlled by the North and South Craven Faults and Dent Fault (see figure 6 1) The formation of the fault was accompanied by a large degree of tectonic activity with associated minor faults and planes of weakness in the surrounding rocks Caves in the area have either cut across or developed preferentially along these planes of weakness which may extend to considerable depth effectively increasing the emanating surface area of the cave that is available to contribute radon This will promote an increase in the radon budget and consequently the mean radon concentration Conversely, areas with lower mean cave radon concentrations are further from the fault, being situated towards the center of the limestone block, being affected by less intensive secondary faulting Therefore, the development of secondary planes of weakness will be reduced, the total surface area from which radon could emanate to the cave is less, being controlled solely by passage dimension and hence the mean cave radon concentration is lower Detailed investigations being undertaken in other caves in the North Pennines region support this hypothesis (Workman, 1994 pers coms)

Explanation 2

As one area is situated close to the margin of the limestone block and the other is situated further towards the centre there may have been differences in sedimentological conditions during deposition. Although not sufficient to represent a facies change these may have altered the characteristics of the beds sufficiently to produce differences in radon

emanation and exhalation characteristics Tanner (1978) suggests that the following rock characteristics may influence radon release

1 Uranium concentration, which controls the radon production rate Increased uranium concentrations increasing the potential for radon release .

2 Grain size, which controls the amount of radon that can be released into the pore spaces (emanating power) and which can subsequently move to the external atmosphere (radon exhalation)

3 Moisture content, which affects the emanating power and exhalation rate Increased water content increases emanating power while reducing exhalation rates (for more details see chapter seven)

4 Porosity and permeability control the speed by which the emanated radon can move through the pore spaces to the external atmosphere

5 Presence of carrier fluids Both air and water can act as a carrier fluid to move radon from emanation sites to the external atmosphere Air currents are induced to flow through rock masses due to variations in atmospheric pressure which alter the air volume

If any one or a combination of these properties of the surrounding rock were different between the two areas then the radon budget would be altered, potentially resulting in differences in the observed mean radon concentration for each area

Explanation 3

The majority of caves in the areas with high mean radon concentration (Leck Beck Fell, Stump Cross and Upper Nidderdale) show pronounced vertical development The caves in the areas with the lower mean radon concentrations (Kingsdale, Ingleborough Hill and Dow cave areas) exhibit both vertical and horizontal development with two or more entrances, one of which is at a different altitude to the other The caves in the Kingsdale,

Ingleborough Hill and Dow Cave SSSI all have multiple entrances at different altitudes which enhance air movement through the caves Yarborough *et al* (1976) demonstrate that the amount of air movement within a cavern influences cave radon concentrations, therefore, increased air movements in these caves will lead to dilution and redistribution and therefore a lowering of cave radon concentrations

During all survey periods, apart from May 1992, the Leck Beck fell area has a maximum 2 - 5 times higher than all other SSSI This could be a consequence of either localised radon sources or accumulation mechanisms operating within the cavern In Dow Cave, the mean, maximum and minimum results are surprisingly similar This suggests that concentrations in the cave are controlled by emanation and the effects of accumulation / dispersion mechanisms are less If residence times for air within the caves are small, less than 3 8 days variations will also be limited and variation small

6.4.4.2 South Wales

Tables 6 15 and 6 18 summarise the data for the South Wales region Analysis of the means using a students T-test indicates that the means are significantly different for the Ogof-Ffynnon-Ddu and Mynydd-Llangattwg SSSI area during all sampling periods Ogof-Ffynnon-Ddu and Mynydd-Llangattwg have lower mean radon concentrations (2,872 and 5,163 Bq m^{-3} respectively) than Otter Hole where the mean concentration, was 12,332 Bq m^{-3} during the August 1991 sampling period The same distribution can be seen in the maximum, minimum and standard deviation results from the SSSI within the South Wales region, although results from Otter Hole are only available from August 1991

The entrance of Otter Hole is in an estuary and is sealed by a sump that rises and falls with the tide, while the entrances to the other caves surveyed are permanently open. Therefore, the results from Otter Hole could effectively be those for a sealed chamber where the effectiveness of normal redistribution and dilution mechanisms have been reduced This conclusion is supported by the relatively high minimum (7,461 Bq m^{-3})

which implies that little dilution occurs in any part of the cave However, further work needs to be conducted in Otter Hole and / or other similar caves to test this theory and the following measurements are needed

1 Time integrated mean radon concentrations over 1 week periods to determine whether the radon concentration within the cavern relates to variations in atmospheric pressure, which will imply that emanation is the primary control on observed radon concentrations

2 Spot measurements of radon and radon daughter concentrations to determine if a state of equilibrium exists Equilibrium will only exist in a closed system where emanation is the only mechanism of addition to the system and radioactive decay, controlled by the half life of radon is the only removal mechanism.

However, if it is assumed that

1 The radon concentrations in Otter Hole are from a sealed chamber

2 The radon concentrations in Otter Hole are solely a function of the radon budget

3 The radon budget for all caves in the South Wales region is similar

4 The radon concentrations in the other SSSI are a function of the radon budget and cave microclimate

Then by comparing the results from Otter Hole with the other areas in South Wales an estimation of the amount of redistribution / dilution of radon gas that occurs in limestone caves due to air movement can be made To do this effectively the maximum values were used as these are more likely to represent the radon concentration arising from a sealed chamber It can be seen that the maxima in the three SSSI in South Wales are quite varied, with the highest maximum being recorded in Otter Hole The maxima in the Ogof Ffynnon Ddu SSSI are approximately 0 31 of that from Otter Hole implying either that two-thirds of the total radon emanated is redistributed or that there is dilution in the Ogof

Ffynnon Ddu area In contrast, the results from Mynydd Llangattwg are approximately half the total Otter Hole radon maximum, implying that the radon concentrations have been diluted by 50 % Therefore it can be estimated, bearing in mind the assumptions outlined above, that between 30% to 50 % of the radon emanated into the South Wales cave systems is redistributed or diluted by internal air movements

6.4.4.3 Peak District

Tables 6 16 and 6.19 summarise the results from the Peak District region Analysis of the results using the Students t-Test for areas within the Peak District identifies that the results from this region can be divided into three broad groups

1 Castleton, Upper Lathkill Dale, and Bradwell with high mean radon concentrations (between 9,916 and 12,187 Bq m^{-3})

2 Stoney Middleton Dale with a mean concentration of 5,218 Bq m^{-3}

3 Matlock, with a low mean concentration (below 500 Bq m^{-3}) The mean and maximum concentrations from the Matlock SSSI are in some cases ten times lower than the other group

The low concentrations in the Matlock area could represent the influences of changes in the radon budget or cave morphology in comparison to the other areas within the Peak District region However, the caves in this area are relatively short and hence all detectors were located within 50 m of an entrance As a result the measured concentrations will reflect the influence of external (relatively radon free) air entering the cave

All areas, including the Matlock SSSI, are located close to the margin of the limestone massif Therefore, the position of the caves within the limestone mass does not affect cave concentrations However, different SSSI and caves are contained within different limestone beds which could result in variations, due to differences in the radon budget, in

concentrations in different SSSI. Additionally, sources of radon could be derived from the shales which surround the limestone, that have relatively high uranium concentrations (Ball *et al*, 1992, Bottrell, 1991) Hence, uranium enters the cave systems in allogenic sediments and the nature, type and distribution of the sediment within the individual caves influences their total radon budget This source could increase radon generation and elevate the observed radon concentrations Other possible localised sources of radon in the Peak District region are

1 Collophane nodules, wayboards and chert bands that have very high uranium concentrations A maximum of 80 ppm of Uranium has been recorded in the limestone sequence (Ball, 1992 pers coms, Peacock and Taylor, 1966) Even though the radon generation is high, due to the initial uranium concentrations, the release potential is low due to the grain size of the materials which reduces their exhalation rate

2 Hydrocarbons within the limestones whose formation has been associated with secondary uranium transportation and accumulation

3 Fluorite mineralisation, which is associated with secondary uranium movement and could produce zones of uranium accumulation

4 Deep seated radon sources associated with jointing

5 Reef facies limestones are known to contain high uranium concentrations (circa 7 - 30 ppm), compared with concentrations between 2 and 3 ppm in 'average' limestone beds (chapter seven)

The highest seven day concentration recorded during the project was from Giant's Hole, Castleton, a cave which had previously been identified as having very high radon concentrations (Gunn *et al*, 1991; Middleton *et al*, 1991) However, with the exception of the Matlock SSSI all other SSSI in the Peak District also have very high radon concentrations The seven day maxima in each SSSI are Bradwell Dale 31,817 Bq m^{-3} Upper Lathkill Dale 45,910 Bq m^{-3} and Stoney Middleton 39,047 Bq m^{-3} each of which is

very high in world terms Although the highest seven day maximum concentration was from the Castleton SSSI, the overall mean from this area is exceeded by two areas, Bradwell Dale and the Upper Lathkill Dale, apart from during the May 1992 sampling period (table 6 19)

6.4.4.4 Mendip Hills

Few conclusions can be drawn from the results from the Mendip Hills as only one area within the region has been studied in detail. Other one-off results (included in Appendix 1) and published work from the area (Reaich and Kerr, 1991) imply that the results obtained during this project are lower than would be expected and this could be a function of detector location in relatively short and well-ventilated caves

6.4.4.5 Portland

Due to the relatively small size of the Portland region it cannot be divided into areas However, the results imply that cave morphology greatly affects the observed concentrations Due to the limited size of the caves and the low uranium concentrations in the surrounding Jurassic limestones the mean radon concentrations are low

6 4 5 Variation within individual caves

Although data were obtained from 42 caves during the project only a selection which highlight salient points of interest will be discussed Figure 6 6 shows results from Carno Adit Only the last three survey periods are shown as an incomplete data set was collected during the first period Carno Adit is a man made adit used for water supply in the South Wales region Detectors 1 - 7 were placed in passages within the Dowlais limestones while the last detector (8) was placed in passages within the Llanelly shales (figure 6 13) From these results two interesting features can be seen

1 In all cases the radon concentrations recorded at site 8 are below that of site 7 This is thought to be a consequence of the different emanation / exhalation characteristic of the Dowlais limestones compared with the Llanelly shales In particular the radon budget of the Llanelly shales is likely to be lower than that of the Dowlais limestone due either to a decrease in grain size or a difference in the original uranium concentration

2. On all occasions the concentration at site 4 is higher than at sites 1 - 3 In 1992 a section of new natural cave, in excess of 2 5 km, was found leading from the Adit at this site (Gascoine, 1992) The increase in radon could therefore be a result of radon rich air being drawn from the cave system below into the Adit The concentrations at site 7 are also markedly higher than the sites before it in the Adit (1 - 6) However, the increase is larger and more obvious than that at site 4 The Adit survey shows a number of small side passages near site 7, but the presently known passage is much less extensive than near site 4 The increase in the observed radon concentration may indicate a considerable amount of as-yet-undiscovered cave passage in this area If this is shown to be the case then radon concentrations within limestone caves could be used in cave exploration to suggest areas where new passage is present, in a similar way to surface air temperatures being used in the Peak District in winter to locate new entrances

A selection of the results from Peak Cavern, Peak District are shown in figure 6 7 Site 5, Treasury Sump, always has considerably higher concentrations than those surrounding it, and while the other sites show considerable variation, the results from Treasury Sump are relatively constant (figure 6 7) The detectors were located close to the waters edge in the sump chamber that lies at the base of a tube which descends some 25 meters from the larger Treasury Chamber The water level in the sump varies, but the sump is continuously sealed The floor of the area is covered with sands and gravel at an angle of approximately 17 degrees The increased radon concentration at this site could be related to variations in the radon budget, but this is felt to be unlikely as the sands and gravels have been found to have a low original uranium concentration, and emanation /

exhalation rates are low (Bottrell, 1991) Therefore, the increase in concentration is more likely to be a result of the relatively dense radon draining down to the sump from the passageways above Once the radon has drained into the area there is very little air movement to redistribute or dilute the radon due to the sump being sealed It is therefore suggested that Treasury Sump is acting as a radon sump, where radon redistributed from other areas of the cave accumulates

Figure 6 8 shows the mean seven day radon concentrations for the four sampling periods for sites in Ireby Fell Cavern and Lost Johns Cavern in the North Pennines Both caves have significant vertical development, including a number of vertical shafts The sites are numbered consequently into the cave, each being at a greater depth Overall, concentrations increase with depth and the increase follows a number of steps which coincide with the tops and bottoms of pitches Three possible explanations may be put forward to explain the general relationship of increased radon concentration with depth

1 Relatively heavy radon atoms drain towards the bottom of cave systems If this is the case then the equilibrium between radon and radon daughter should also increase with depth At present insufficient data are available to test this hypothesis

2 The quantity of rock, sediment, water and other radon sources increases with depth

3 The diluting effect of any inflowing, relatively radon-free air, will reduce concentrations near to the entrance Conversely, further into the cave radon from the entrance might have been redistributed to the further reaches of the cavern and the diluting effect of incoming radon free air will be reduced

In each of the caves discussed above, radon concentrations increase with distance from the entrance (figure 6 6 - 6 8) However, the relationship between distance and radon concentration is controlled by numerous processes including cave morphology, season, time of day, location of radon sources and passage area In chapter nine, results from a

more detailed investigation of radon and radon daughter concentrations in Peak Cavern, are discussed and the relationship between radon and depth / distance are considered further

Figures 6 6, 6.7 and 6 8 all show results from caves with relatively simple morphologies and in these caves the relationships between observed concentrations at individual sites follow similar patterns irrespective of the season However, the results from Ease Gill (figure 6 9), are far more complex and the relative concentrations at individual sites vary seasonally This is thought to be a result of the cave having several entrances and a large amount of passage As a result it is unlikely that a single process would control the air movements and hence the accumulation / dispersion processes within the cavern Within complex cave systems, several microclimatic systems may operate to influence radon concentrations These different microclimatic systems are dependent upon a complex set of relationships between the internal cave atmosphere and the external atmosphere, coupled with cave morphology Within Ease Gill the radon concentrations at the majority of sites are very variable, but at site three concentrations are consistently higher than those surrounding it This could relate to either a radon sink similar to Treasury Sump in Peak Cavern, or a dead end site where increased concentrations are a result of local accumulation mechanisms controlled by cave microclimate

6 4 6 Temporal variations

6.4.6.1 General relationships.

Tables 6 13, 6 17, 6 18 and 6 19 outline seven day integrated radon concentrations for each region during individual sampling periods The variation between seasons can be explained, at least in part, by differences in cave microclimate Previous studies of radon and radon daughter concentrations in limestone caves which have concentrated upon accounting for temporal variations within single caves, are reviewed in chapter three

Figure 6 5 clearly demonstrates that in all areas, apart from the Mendip Hills, concentrations decrease from a 'summer' (August 1991) maximum through the 'autumn' sampling period (November 1991) to their low in the 'winter' (February 1992) and then rise again during the 'spring' sampling period (May 1992) Similar seasonal variations in concentrations have been recorded in nearly all investigations of radon concentrations within limestone caves (see chapter three for further details)

As a detailed discussion of the causes of seasonal variations in radon concentrations has already been undertaken in chapter three, only a brief synopsis is presented here Seasonal variations in concentrations can be primarily related to air movements between the cave atmosphere and the external air Yarborough, 1976 classified caves into two primary types, Right Side Up (RSU) and Up-side Down (USD), in both of which radon concentrations increased during the summer months and decreased during the winter period In RSU caves, the summer increase is due to stagnation and lack of incoming external radon free air to dilute and redistribute cavern radon, while in USD caves the summer increase is due to mobilisation of radon rich air within the cave During the summer months, air flows out from the cave resulting in mobilisation of radon In both cases, during the winter months air flows into the caverns from the external atmosphere resulting in dilution and redistribution of the cavern radon

Figure 6.5 shows that concentrations observed during 'spring' and 'autumn' months are closer to those in the 'winter' sampling period than the 'summer' Ahlstrand and Fry (1976) showed that spring and autumn concentrations may follow either summer or winter conditions depending upon the mean monthly temperature However, this assumes that the cavern air temperature is the sole control on air movements, whereas moisture and atmospheric pressure also play an integral part in the movement of air within caves

The mean internal cave temperature in the caves of England and Wales is approximately 8 5°C Hence, if air temperature is the sole control then when the mean external temperature is higher than 8 5°C air will flow out from the cavern and radon concentrations will generally be higher than those encountered during months when the

external atmospheric temperature is below 8 5°C During this investigation the temperatures in autumn and spring were on average below 8 5 °C. It is therefore likely that radon free air moved into the caves reducing the observed radon concentrations The spring and autumn months of the year exhibit the greatest variation in radon and radon daughter concentrations with large scale fluctuations occurring on both daily and weekly timescales In contrast, the summer and winter months are usually either higher or lower, respectively, with less variation apparent on all time scales

Sudden changes in atmospheric pressure may also affect the volume of air within a cave, decreasing pressure leading to an increase in the volume of air and increasing atmospheric pressure to a decreases in the volume of air within the cave During the winter, if pressure decreases when the temperature differential induced air movement is inward, then the two processes will act in opposition and potentially a slight increase in concentrations may result Conversely if an increase in pressure occurs during the summer months, when the natural temperature induced air movement is outwards, then the decreasing volume of air will result in air being drawn into the cavern which could promote a lowering in the cavern radon concentration In addition to its influences on cave microclimate, atmospheric pressure affects the emanation and exhalation rates of radon which vary inversely with pressure The influence of pressure changes on cave radon has not previously been the subject of detailed research and it is not possible to draw conclusions from data collected in the regional study as these were collected over 7 day periods thus smoothing out short term changes However, the influence of atmospheric pressure on cave radon concentrations is considered further in chapters eight and nine

If it is assumed that the mean radon concentrations observed during the 'summer' sampling period represent the bulk emanation rate, with little or no dilution, and that the mean 'winter' results represent the bulk emanation rate after a degree of dilution by in-flowing radon free air, then the dilution factor can be estimated in the broadest terms by comparing the 'summer' and 'winter' means Based upon table 6 13 the winter means for individual regions are 0 2 - 0 5 times those of the summer This implies that

approximately 2 - 5 times the volume of air that was originally within the cave is flowing into the cave during the winter periods to reduce the radon concentrations It is interesting to note that a dilution factor in the same order of magnitude was determined in section 6 5 3 when the results from South Wales were considered although more stringent restrictions were applied

6 4 7 Specific Examples and Anomalies

Although broad seasonal trends are apparent, the results from some caves can not be generalised Figures 6 9, 6 10 and 6 11 outline data from three caves in the North Pennine region. All three caves have different morphologies that affect the radon concentrations recorded Ease Gill can be described as a 'system', with numerous entrances and both vertical and horizontal development (figure 6 9) Kingsdale Master cave can be classified as a chimney cave, with both vertical and horizontal development and at least two known entrances, at different elevations (figure 6 10) White Scar can be generalised as a tube (USD), being a cave with preferential horizontal development over vertical At present there is no humanly penetrable connection to the hill side above, but a 10 cm diameter bore hole has been drilled into the Battlefield Chamber (figure 6 11) The concentrations in the three caves exhibit two particular interesting features

1 The concentrations in all three caves are very variable, with both differences between individual sites within a single cave and differences in the concentrations within each cave being observed Ease Gill exhibits the most variation while White Scar and Kingsdale Master cave show less variation The most likely explanation is microclimatic regime White Scar cave and the Kingsdale Master cave have simple microclimatic regimes as there are few entrances However, Ease Gill Caverns has several entrances and the interrelationships existing between different entrances under different external climatic conditions result in several microclimatic regimes operating within the cavern, each of which will

affect the radon concentration observed at different sites at various times of the year

2 The air movements in both White Scar cave and Kingsdale Master cave are largely controlled by the chimney effect, whereby the density differences between the cavern and external air promote air movement through the system. Generally air moves in through the top entrance and out through the lower one during the summer months and vice versa during the winter It would be expected that radon concentrations would increase downwind, as radon is redistributed through the system. The radon concentrations recorded in White Scar follow the predicted pattern as expected for both 'winter' and 'summer' sampling periods However, Kingsdale Master cave does not follow the predicted pattern, the concentrations implying that air moves in via the upper entrance and out through the lower portal, during both winter and summer This deviation from the normal could be a result of the stream that enters the Kingsdale Master cave at the upper entrance and flows down towards the lower portal. The velocity of the moving water might be enough to 'drag' air along with it and reverse the airflow In the summer months the stream will be working in conjunction with the normal air flow system and consequently the volume of air induced to flow through the system will be larger than expected The increased airflows in the summer months might be able to move and redistribute radon, potentially drawing radon from microfractures in the rock and sediment promoting an increase in radon In the winter months, the stream's power reverses the natural air flow The reversal was observed by the author during the February 1992 sampling period, with air entering the upper entrance in contrast to what would be expected from density induced air movements

The results from Heron Pot, in the North Pennines are shown in figure 6 12 Heron Pot has two known entrances, one of which is approximately 60 metres above the other A stream flows through the system and during the winter months the increased flow can result in the lower entrance 'sumping up' for periods of time During August 1991, radon

concentrations increased along the path of the air flow, as expected, but this was also the case in November 1991 and February 1992, the increase during the 'winter' sampling periods being larger than that experienced during the summer During the November 1991 sampling period, water levels were relatively high but had not totally sealed the lower entrance Consequently, the amount of air moving through the cavern was reduced in comparison to August 1991, and this probably explains the increase in concentrations observed, with less radon being diluted by in-flowing external radon free air During February 1992, the stream discharge was sufficient to totally seal the lower entrance This again reduced the amount of air moving through the system resulting in a large increase in radon concentrations The results again imply that the relatively heavy radon atoms are moving to the lower reaches of the cavern since the relative increase in the lower portion of the system is larger than that near to the entrance

6.5 Summary

This chapter has provided an overview of the variations in cave radon concentrations in the North Pennines, Peak District and South Wales regions, highlighting and identifying the fundamentals of the radon system. Limited results were also presented from the Mendip Hills and Portland regions The radon concentrations recorded in some Peak District caves are amongst the highest in the world, and cave radon concentrations in the North Pennines and South Wales regions are higher than in most areas of the world. Between and within region variations in cave radon concentrations are evident and the mechanisms which control these variations have been identified and discussed Seasonal variations in cave radon concentrations follow a similar pattern to that reported by Yarborough (1976, 1977, 1982) and other authors, winter concentrations being generally lower than those in the summer This can largely be accounted for by variations in the rate and direction of air movement within the cavern, the net inward movement of air in the winter resulting in a dilution of radon by 30 - 50% Some caves do not follow the general patterns and specific examples were used to highlight other mechanisms that can also influence radon concentrations It is felt that insufficient data exist to predict radon

levels in individual caves or areas Depending upon the scale of investigation, different mechanisms can be seen to control the radon concentrations observed within limestone caves From a regional perspective two primary interrelated controls can be identified firstly, the radon budget, and secondly, accumulation / dispersion mechanisms When results are compared on an intra-regional scale, these two processes are still evident but some factors that control these mechanisms become evident The radon budget is influenced by

1 Uranium concentrations in the rocks and sediments found within individual caves (Peak District)

2 The amount and intensity of faulting and joint development (North Pennines)

3 Sedimentological facies changes in the host limestone rock (North Pennines)

4 Atmospheric pressure changes

The accumulation / dispersion mechanisms are controlled by cave morphology and particularly

1. Development status (horizontal versus vertical)

2 Number of entrances

3 Cave Microclimate

4 Atmospheric pressure

The total emanating area influences results from individual caves, such as Carno Adit, South Wales, and the dispersion / accumulation mechanisms were influenced by 'radon sumps' and the complexity of the system.

Integrated weekly mean concentrations do not permit the influence of changes in atmospheric pressure on either the radon budget or air movements with the cavern to be

identified Similarly diurnal variations cannot be identified Hence, more detailed work has been undertaken in a single region (the Peak District) and within a single cave (Peak Cavern) and this is discussed in chapters eight and nine

CHAPTER SEVEN: SOURCES OF RADON IN LIMESTONE CAVES

7.1 Introduction

From the cave radon survey of England and Wales it is clear that identification and quantification of the sources of radon to the karst environment is an essential pre-requisite to an understanding of the cave radon system. In this chapter the processes governing the generation of radon within the karst system are discussed and potential sources of radon identified Methods by which the relative importance of each source may be quantified are identified and the relative importance of each source is then quantified During the process of radon liberation, to the terrestrial environment, from production sites within different sources, two dependent but separate processes occur, emanation and exhalation Emanation was defined by Tanner (1978)

> *"before an atom of radon can migrate, it must escape from the site of production, governed by the position of its parent radium-226 isotope within the lattice Under steady state conditions, the fraction of radon atoms formed in a solid that escape from the solid is defined as radon emanation" (Tanner, 1978 p 3)*

More simply, emanation can be defined as the percentage of radon produced within the mineral lattice that escapes into the pore space or surrounding voids that is able to subsequently migrate to the terrestrial environment The process of exhalation can be defined as

> *"the percentage of radon emanated into the pore spaces that escapes into the terrestrial environment to pose a threat to human health" (Tanner, 1978 p.11)*

7.2 Radon Emanation

7 2 1 Introduction

The definition presented in the previous section will be used during the thesis However, within the scientific literature different names have been used to describe the processes of emanation In the Soviet literature emanation is referred to as the coefficient of emanation while in Europe the terms escape ratio, escape to production ratio or percentage emanation have all been used (Tanner, 1978) The theory of emanation was first developed in the 1940's by Flugge and Zimens (cited by Tanner, 1978), who described the processes as emanating power but little further work was conducted until Wahl and Bonner (1951) Specific investigations include the treatment by Quet *et al* (1972) of emanation for isolated particles and the study by Andrews and Wood (1972) of the emanation processes of radon into ground water More recently Semkow (1990) studied the processes of release from mineral grains More applied work was then conducted investigating the effects of radium-226 distribution within mineral grains and porosity on emanation (Semkow and Parekh, 1990) and the effects of fractured surfaces on the emanation processes (Semkow *et al*, 1991)

7 2 2 Processes of radon emanation

When radium-226 decays, most of the energy associated with its decay is carried off by the alpha particle produced during decay, and the remainder, approximately 10^4 - 10^5 times greater than typical chemical bond energies, is carried by the resulting radon atom. Figure 7 1 illustrates the recoil process Due to the energy imparted to the radon atom during the decay process it has a recoil range of 20 - 70 μm for minerals of common density (Quet *et al*, 1972) A radon atom that is directed towards the grain surface potentially has sufficient energy to escape into the pore Those atoms that terminate their recoil paths in a pore are termed the direct recoil fraction If the pore is filled with gas, the range of the radon atom within the pore is equal to the remaining kinetic energy

multiplied by its recoil range Depending upon the travel distance within the production mineral, this is 20 - 63 μm (Flugge and Zimens, 1939, cited by Tanner, 1978)

Direct Recoil

Factors affecting the direct recoil are the size of grains and the distribution and dimensions of pore spaces within the mineral lattice It is estimated that the direct recoil fraction represents only 1 % in dry compacted materials However, if the pores contain a fluid the travel range is decreased due to the increased friction and their ranges are as low as 0 1μm. Therefore, the probability that the radon atom will remain within the pore space is increased

Indirect recoil

If the pores are air filled and the recoiling radon atom has sufficient energy to traverse the pore space it will collide with a joining mineral grain Upon impact the radon atom will penetrate into the mineral lattice and become embedded Once embedded the radon atom has the potential to diffuse from the impact site to the pore space Due to the diffusion coefficient of radon, the diffusion length for this process is limited by the mean half life of radon However, the low diffusion coefficient implies that this process can not operate for radon-220 or radon-219

7 2 3 Controls on radon emanation

Radon production rate:

Before the process of radon emanation can occur, the radon atom has to be generated, and this is controlled by its immediate precursor, radium-226 If radium-226 is absent then no radon will be produced Therefore, the initial concentration of radium-226 has a direct bearing on the process of radon emanation by controlling the overall radon

production rate The initial radium-226 concentration is linked through the decay chain with the original concentration of uranium-238 (Ball, 1994, Ball *et al*, 1991)

Mineral location:

As the distribution of uranium-238 within mineral lattices is not homogeneous, the location of radium-226 atoms is also not homogeneous Most of the uranium-238 in rocks is associated with discrete uranium-238 bearing minerals but, even then only a few parts per million of uranium-238 are present (Ball *et al*, 1991) Due to the gaseous nature of radon, with a limited half life, the potential for release from the mineral lattice is enhanced if the production sites are located close to the mineral surfaces Therefore, the location of production sites is crucial in controlling the final amount of radon emanated from the rock (Andrews and Wood, 1972, Ball *et al*, 1991)

Moisture:

Water present in the pores or capillaries increases the probability that radon atoms will terminate their recoil paths in the pore spaces, due to the increased friction associated with travel through water when compared to air (recoil distances are approximately 0 1μm in water compared to 20 - 63 μm in air (Tanner, 1978)) Due to this, hydrated surfaces tend to have greater emanations than unhydrated surfaces (Tanner, 1978)

Grain size:

The size and shape of grains will affect the location of radium-226 atoms in relation to the mineral surface, thereby affecting the percentage of radon atoms formed which, due to the energy imparted in them during decay, are going to be within recoil distance Generally the smaller the grain size, the more likely that a radon atom generated will be able to escape via the surface More angular particles allow for looser packing which increases the size of pore spaces within the strata compared with rounded particles Therefore, generally angular grains have a higher emanation coefficient

7.3 Radon Exhalation

7 3 1 Introduction

The process of exhalation concerns the movement of radon from pore spaces or voids within the production material to the atmospheric environment The primary constraint on this movement is the half life of radon Once the radon is within the pore spaces any subsequent movement occurs as a result of either diffusion or transportation in a carrier fluid. The carrier fluids can be either gaseous (for example air) or liquid (such as water) It has been established that concentrations of radon gas vary depending upon the phase of the system (Tanner, 1978) For example, at temperatures common on the Earth's surface radon concentrations will be greatest in the liquid organic phase (for example, suspended in hydrocarbons) in comparison to the water phase while the gas phase is between these two At increasing temperatures, concentrations in the gas phase increase at the expense of the liquid phase At temperatures commonly found in caves, and on the earth's surface, concentration in the liquid phase will generally be less than in the gas phase Therefore, at the initial interface zone between air and water radon will be actively degassed from the water into the air Diffusion of radon within a porous environment has been comprehensively reviewed by Curries (1961) who states that initial diffusion is influenced by porosity, packing, particle shape and size of the porous environment These factors influence the degree and ease of movement within the material. However, within the natural environment at no time will pure diffusion occur as a gradient of some description will always exist between conditions within the medium and the external environment that will lead to flow This flow will result in a carrier fluid, either air or water, being circulated within the medium resulting in the transportation of radon within the fluid

Changes in climatic conditions control temperature and pressure gradients with a pronounced seasonal pattern These generally increase exhalation rates during the summer in comparison to the winter months (Smith *et al*, 1976) Fleischer (1987)

demonstrated that moisture within the pore spaces increases exhalation but an optimum moisture level exists with respect to exhalation. Fleischer states that between 11 and 25% moisture content is the optimum, with levels below 11% or above 25% decreasing the exhalation rate The decrease in exhalation rate with increased moisture content can be related to the reduced effects of transport in the gaseous phase which contains more radon than the fluid phase As a consequence, Gingrich and Fisher (1976) demonstrate a generally negative relationship between rainfall and exhalation rates Nero and Nazaroff (1985) demonstrated that pressure induced flow, as a consequence of either temperature or pressure differentials was more important in controlling exhalation rates than diffusion Rama and Moore (1984) attempted to quantify the difference between diffusion and pressure induced flow, and concluded that pressure induced flow accounts for the majority of radon exhaled from all materials

7.4 Sources of radon to the karst environment

There has been little research into the sources of radon to limestone caves and the author is not aware of any integrated projects which have concentrated solely on investigating the sources of radon to caves However, a number of projects have identified and studied single sources of radon to the cave environment (for example, Bottrell, 1991, Surbeck and Medici, 1990), while other authors have inferred sources of radon from concentrations (for example, Gunn *et al*, 1989a and b, Lenart *et al*, 1989, Middleton, 1988, Williamson, 1990)

The initial primary source of radon to the karst environment is the limestone itself This source is often discounted due to the low initial uranium-238 concentrations, typically 1 - 3 ppm (Lenart *et al*, 1989) but a number of authors have suggested that even with low original uranium-238 concentrations, limestone could be important due to the surface area exposed within cave environments (Breisch, 1968, Somogyi *et al*, 1989, Wilkening, 1976) Other beds contained within the geological sequence can be locally important sources of radon to cave environments Peacock and Taylor (1966) identified collophane nodules containing high uranium-238 concentrations (30 - 300 ppm) in both

the Derbyshire and Yorkshire limestone sequences These can potentially produce localised sources of radon. Other potential sources within the geological sequence include vein material, tuffs, volcanic sills and dykes, toadstones and paleokarst horizons

Bottrell (1991) conducted a survey of sediments within Speedwell Cavern, Castleton He concluded that sediments of different ages had varying uranium-238 concentrations and therefore, different radon production potential. In general older sediments had a higher radon production potential than younger sediments The nature of these sediments also varied and Bottrell (1991) suggested that fine grained sediments had a higher production potential than coarse grained Although sediments within a cave have been observed to influence radon concentrations the sources of these sediments are important (Lenart *et al*, 1989) For example, sediments in the Castleton region derived from uranium-238 rich Namurian shales are likely to have a larger impact on the overall radon budget than sediments in other areas derived from non uranium-238 rich sources (Bottrell, 1991)

Speleothems within the cave environment often contain uranium-238 which may be used in radiometric dating and may act as a source of radon to the cave atmosphere Lyons *et al* (1989) studied New Zealand speleothems and found that little radon was released from 16 out of 19 samples Three samples exhibited radon release but were unusual with very finely divided crystals They concluded that in general speleothems provide a minimal source of radon to the cave environment

Radon is present in the soil atmosphere (Surbeck and Medici, 1990) and water percolating through the soil will become enriched in radon that will degas into the cave in a similar manner to carbon dioxide Surbeck and Medici (1990) demonstrated that all the radon observed in a number of caves in Switzerland could be accounted for by radon derived from the soil.

Ball (1993 pers comms) has shown that surface water entering Giants Hole can act as a sink for radon, with radon being absorbed from the air into the water In other cases

radon has been observed to be released from water into the cave The importance of water as a source or a sink of radon within caves depends on the source of the water and the comparative radon concentrations in water and air Friederich (1981) demonstrated for caves in the Mendip Hills that radon in percolation water could be used to identify the route via which the water had flowed and the transmission time The radon in the water was assumed to be derived from either the limestone, soil or overlying sandstone Therefore, percolation waters were identified as a source of radon to the cave environment

Joints and faults within the limestone mass may also act as either an additional sources or a means by which radon can be transported into the cave environment Two processes operate

1 Faults and joints effectively increase the surface area of the cave, increasing the area from which radon emanates / exhales

2 Faults and joints act as pathways by which radon generated from underlying rocks migrates to the cave environment If the underlying rocks are uranium-238 enriched (for example, igneous rocks) then this source may provide significant quantities of radon to the karst environment

Radon may also move from one part of a cave to another For example, during the summer, air from within a cave will usually move towards the external atmosphere This outward movement will mobilise radon produced in the further reaches of the cave resulting in increased concentrations being observed closer to the entrance Conversely, during the winter outside air moves into the cave resulting in the dilution and redistribution of internal radon, and reducing the concentrations

7.5 Methods for determining radon emanation and exhalation

Emanation

Very few methods directly measure radon emanation and instead it is estimated from the net radon production rate based either upon radium-226 or uranium-238 concentrations It is then assumed that the sample is in equilibrium within the decay chain and that all radon produced is emanated from the sample However, these assumptions are known not to be true (Semkow, 1990) although the errors introduced are small. A number of methods have been developed that compare radon production and exhalation rates to determine percentage emanation for individual samples (e g Austin, 1973, Khrishnaswami and Seidemann, 1988) Despite the known problems it was decided to estimate radon emanation from net radon production due to the simplicity of the method and the availability of equipment Original concentrations of uranium-238 were determined using a portable gamma spectrometer or a laboratory based gamma spectrometer (Exploranium GS, 1989, Hansen, 1975)

Exhalation

The majority of methods for determining radon exhalation use a derivative of the accumulation method outlined by either NCRP (1988) or Kearney and Krueger (1987) The accumulator method involves placing the sample under investigation in a closed and air tight chamber for approximately 30 days The sample is left to equilibrate, resulting in the radon concentration in the chamber being equal to that resulting from radon exhalation The radon in the chamber is then sampled to determine the radon exhalation rate Initially radon gas concentrations in the chamber were determined using charcoal detectors (Countess, 1976, Hinton and Whicker, 1985, Kauffman *et al*, 1987, Rogers *et al*, 1982) However, Samuelsson (1986) showed that this method could result in a number of errors In particular, due to the active absorption process of charcoal detectors, radon is removed from the sampling chamber during the experiment potentially forming a radon gradient within the sampling chamber and leading to

enhanced radon exhalation After Samuelsson's (1986) paper the charcoal method lost favor and other methods were used to determine the radon concentration in the chamber One common alternative was the use of Lucas cells (Giletti and Kulp, 1955, Ingersoll and Stitt, 1983, Jonassen, 1981, see also discussion in chapter five) The increased sensitivity of Lucas cells over charcoal detectors increases the overall sensitivity of determining radon exhalation However, it was also found that in order to remove a sample of air from the accumulator, two inlets were needed, one to remove the sample of air and the other to allow external, relatively radon free air, to enter the accumulator This resulted in dilution of the radon within the accumulator reducing the accuracy of the method (Pogorski *et al*, 1981a and b, Samuelsson and Petterson, 1984)

Track etch detectors (see chapter five) provide the most reliable method for the determination of radon gas concentrations within the accumulator Samples are sealed in chambers for 30 days to equilibrate and then track etch detectors are introduced to the sampling vessel. These are then left in place for 6 - 7 days The resulting radon concentrations can be related to the radon exhalation rate and exposure time (Megumi and Mamuro, 1972 and 1974, Rogers and Nielson, 1991; Savvides *et al*, 1985) In the present research, all radon exhalation measurements were undertaken using the accumulator method and radon gas concentrations were determined using track etch detectors The method was origmally developed for determining radon emanation and exhalation within the uranium prospecting industry, detailed explanations of the method involved can be found in Fleischer (1988) and Fleischer and Mogro-Campero (1981)

Radon in water can be determined either by liquid scintillation counting (Schonfer, 1992, Prime *et al*, 1991) or by degassing the radon within the sample into Lucas cells and counting on a Portable Radiation detector (Hoather and Rackham, 1963, Strutt, 1994, see also chapter five) In this thesis all radon in water measurements were undertaken using the second method

7.6 Sampling Locations

An attempt was made to identify and quantify the sources of radon to the cave environments of England and Wales, all of the potential sources outlined in section 7 4 being investigated. Detailed investigations on a national scale were conducted on limestone strata and sediments and localised studies into the importance of soil and water to the overall radon budget were conducted in Derbyshire It was not possible to undertake direct investigations of the effects of jointing / faulting or radon redistribution by air currents However, inferences on the importance of both these sources were made from analysis of direct observations of radon and radon daughter concentrations

7 6 1 Limestones

In total 70 samples of the main cave bearing limestone strata within England and Wales were collected from the sites outlined in tables 7 1 - 7 4 All samples were collected from freshly exposed strata In all cases multiple samples were taken from individual beds Where possible samples represented the bottom, middle and top of each bed During collection the total uranium-238 concentration was determined using a portable gamma spectrometer Upon return to the laboratory, rock samples were cut into standard cubes (5 - 7 cm long and deep) For each sample the weight, surface area and volume were determined and a subjective judgment was made on the degree of fracturing and development of stylolites in each sample Sampling sites and the relative geological position were determined using Farr, (1989), Ford, (1987 a and b), (1989), Gunn, (1992), Harrison *et al*, (1992), Lowe, (1986, 1989), Maynard, (1991), Smart and Christopher, (1989), Smart and Gardener, (1989) and Stevenson and Gaunt, (1971)

7 6 2 Sediments

Twenty caves were selected from amongst the 47 used during the national investigation of radon concentrations in England and Wales (see chapter six) and three sediment

samples were taken from each cave Samples ranging in age and composition were collected to provide a basis for estimating the overall percentage of the radon budget that can be attributed to sediments However, it is recognised that the sediments collected do not represent the entire range of sediment samples encountered within cave environments

All sediment samples were air dried and then crushed The samples were then split and half were analyzed as a bulk sample while the other half were sieved into clay, silt, sand, and cobble fractions Exhalation tests were then performed on each sub sample Radon emanation was initially determined by XRF at Leeds University based upon uranium concentrations However, due to the high lead and strontium peaks in the samples, the derived uranium-238 concentrations were masked and the results are not valid Therefore, all subsequent emanation determinations were undertaken using either a portable or a laboratory gamma spectrometer

7 6 3 Speleothems

The bulk emanation rates of a number of speleothems from England and Wales, collected by John Gunn during 1980's for uranium series dating, (table 7 6) were determined from their uranium-238 concentrations as measured by Noel Christopher and John Andrews using Alpha Counting (Gunn pers coms , 1994) Exhalation rates were determined by the accumulator method

7 6 4 Water

Water samples were collected from Giant's Hole and Peak Cavern to determine the influence of water on the radon budget Four samples were collected from Giants Hole, on a quarterly basis, under a range of flow conditions Simultaneous determinations of the airborne radon and radon daughter concentrations were undertaken. Additional water samples were collected from seven sites in Peak Cavern, Peak District to

determine the effects of flow conditions and sources of flow on radon in water (for sample locations see figure 7 2)

7 6 5 Overlying Soil

A limited number of measurements of soil radon concentration have been undertaken within the Peak District region. Fixed traverse lines were established and samples collected seasonally in conjunction with the British Geological Survey (table 7.7)

7.7 Results

7 7 1 Soil

Table 7 7 outlines soil radon gas concentrations from the Peak District region, indicating the rock type underlying each sampling area Mean soil radon concentrations range from 31,000 to 81,000 Bq m^{-3}, the highest concentrations being over the Longstone Mudstones and Apron Reef Facies limestones around Castleton This indicates that high uranium concentrations are important in controlling the release of radon from bedrock (Hyde, 1994, O'Connor, 1994, O'Connor *et al*, 1992 and 1993)

A number of workers have investigated the effects of radon gas in the soil and its influences on radon concentrations in homes, and as a consequence papers outlining the diffusion of radon in soil (Holkko and Liukkonen, 1992) and the geological and geochemical controls on radon generation (Kemski *et al*, 1992, Lindmark and Rosen, 1984, Markkanen and Arvela, 1992, Sextro *et al*, 1987) have been produced However, the only workers to directly consider radon in soil overlaying karst areas are O'Connor (1994), O'Connor *et al* (1993) and Surbeck and Medici (1990) The high radon concentrations encountered within soil implies that soil could act as a source of radon to the cave environment as proposed by Surbeck and Medici (1990) However, the importance of this source depends on a number of limiting processes Before soil radon can enter any cavity a mechanism of transport is needed Rainfall is the only transport

medium as diffusion processes are unlikely to operate Due to the half life of radon, transport has to occur within approximately 3 5 days before the gas decays, and hence the distance from the production sites within the soil to the karst cavity is important Hardwick (1993) suggests that macro pores act as preferential transport routes for caesium from the soil to karst cavities and similar mechanisms could potentially operate to transport radon These routes would allow for rapid transport and therefore might enable radon to move from the soil to the soil / bedrock interface within the 3 86 day half-life Once the radon has been transported through the soil profile to the soil / bedrock interface, the transport potential will decrease and preferential movement will occur along planes of weakness and joints within the limestone mass The time of travel from the bedrock interface to the karst cavity depends upon the distance of travel, the nature of the travel path and the transport mechanism. Despite all these limitations to radon transport, table 7 9 indicates that radon concentrations in percolation waters were between 36 and 79 Bq l^{-1} This implies that potentially significant quantities of radon are transported into the cave via this route Due to the partition coefficient between aqueous phase and gaseous phase, radon will actively move from the aqueous phase to the gaseous phase, thereby, increasing the radon concentration within the cave atmosphere

7 7 2 Water

The concentrations of radon in stream water and air from Giants Hole are given in table 7 8 In general, radon concentrations in both air and water are higher in the summer than in winter, a phenomenon that has been widely discussed in the literature (see chapters three and six) During the winter months, radon gas concentrations in water are higher at the entrance than further into the cave Therefore, the decrease in concentrations indicates that radon is actively being degassed from the stream to the air During the summer (June 1992) radon concentrations in the water increased with distance from the entrance, suggesting that radon was being absorbed from the air into the water Radon usually degasses from water with aeration, and this process is used in several methods to determine radon in water (Strutt, 1994) However, in Giants Hole during the summer

months when radon gas concentrations in the air are above 10,000 Bq m^{-3}, radon is actively absorbed into the water during aeration as the water descends Garland Pot (a waterfall of approximately 5 meters) Therefore, in Giants Hole water can be seen to act as both a source of radon to the cave air and a sink.

The concentrations of radon in water from the risings and main streamway in Peak Cavern are given in table 7 9 (see figure 7 2 for sampling locations) Generally, radon concentrations in water emerging from sumps within the cave have higher concentrations than the streamway As water moves downstream from the risings, radon concentrations decrease Radon concentrations at the upstream side of Buxton Water sump are lower than those at the downstream side The higher concentrations in both Far and Ink sump also suggest that the passage of water through a sump may increase radon concentrations. Due to the partition coefficient between aqueous phase radon and gas phase radon, radon will generally move from the aqueous phase to the gas phase Therefore, in a vadose streamway, radon released into the water from the rocks and sediments will in turn be released to the air, the processes being aided by the turbulent flow within the streamways Conversely, in phreatic passages all the radon released is retained within the water as there are no or few air spaces Additionally, radon will be absorbed into the water from the total surface area of the passageway, whereas in vadose passages, water will only receive radon emanating from a small percentage of the passage surface area

In all instances radon concentrations in water emerging from Far Sump were higher than those emerging from Ink Sump and this could be a result of the distance traveled within the sump, Far Sump being 385m long while Ink Sump is 192m. However, the increase in concentration could also be a function of the route by which water has flowed to reach the sump Concentrations recorded at Far Sump during July and September are higher than those recorded during other months and in both of these instances the water was either not flowing or was only just flowing This implies that the residence time of the water within the sump might also affect the radon concentrations However, if residence is in excess of 10 days the system will reach equilibrium and therefore the measured

radon concentrations would be equal to the radon being emanated from the passage Friederich (1981) suggested that radon concentrations in percolation waters could be used to differentiate between the different routes by which percolation water moved from the soil into the cave environment, while Gabechava *et al* (1979) used radioactivity in water to determine the degree and intensity of karst development, suggesting that higher radon concentrations are associated with more intense karst development Hoehn and Von Gunten (1989) used radon in groundwater as a tool to infer the infiltration rates of water in non karst areas, while Prime *et al* (1991) and Sidebotham *et al* (1994) both investigated radon in surface waters in the Peak District and suggested that water from mine drainage, springs and surface waters could be distinguished by their radon concentration In this study water has been found to act as both a sink and source of radon to the karst environment However, sufficient information is not available to allow radon concentrations in water to be used as an indicator of karst processes

7 7 3 Limestone Strata

The uranium concentrations in the main strata outlined in tables 7 1 - 7 4 were measured using a portable gamma spectrometer Total uranium concentrations were recorded and used to approximate the uranium-238 component This will result in little error as uranium-238 represents 99 3% of the total uranium (Bottrell, 1991) Overall, the uranium concentrations range from 0 1 - 24 ppm. However, concentrations within the Peak District range from 2 5 - 24 ppm, while concentrations in other regions range from 0 1 to 2 4 ppm (table 7.10) Based upon the recorded uranium concentrations, radon production rates were estimated, assuming 100% emanation and exhalation from the samples Radon production rates range from 31 3 to 300 $Bqm^{-3} kg^{-1}$ in the Peak District and 1 3 to 30 0 $Bqm^{-3} kg^{-1}$ in other regions It can clearly be seen that the Peak District limestones have a higher radon production rate due to the original uranium concentrations and are therefore likely to release more radon into karst cavities, assuming that the controls on radon release, outlined in section 7 2, are the same for all

regions Table 7 11 demonstrates that original uranium concentration is the primary control on radon release within limestone rocks Measured radon release rates by both surface area and weight indicate that limestones in the Peak District release approximately ten times more radon than limestones in other regions

Bottrell (1991) proposed an emanation coefficient of 85% for sediments from Speedwell Cavern, in the Peak District Using results in tables 7 11 and 7 12 the emanation coefficient was calculated for the limestone samples analysed during this project (table 7.12) Emanation coefficients for limestone seem to be significantly less, ranging from 14% to 83% Bottrell (1991) calculated that a minimum of 17 4 kg of sediment would be needed to raise 1 m^3 of cave passage to 1 WL The quantity of rock needed to raise 1 m^3 of cave passage to 1 WL ranges from 17 - 3808 kg (table 7 12) However, one constraint on these calculations is that exhalation distance is not accounted for It is usually assumed that radon will only be exhaled from the upper few millimeters of the rock, but if exhalation occurs from a greater distance / depth then the quantities of rock needed might be reduced Instances where exhalation could occur from depth are in areas where large numbers of faults / joints intersect the rock mass and provide transport routes These weakness lines would also increase the surface area which is available to exhale radon from.

7 7 4 Sediments

Sediments were collected from sites outlined in table 7 5 and divided into two major groups Coarse sediments derived from the base of active streamways and silt and clay samples which were collected from areas of either relict passage / overflow passages which contained flood deposits As no reliable measurements of uranium concentrations were undertaken, the radon production rate can not be estimated Due to the sampling location the portable gamma spectrometer could not be used to determine uranium concentrations, and due to the high background counts and low counting rates the laboratory gamma spectrometer did not provide reliable results and the associated error was to large However, radon exhalation rates were determined Radon exhalation varies

from 25 - 75 Bq m^{-3} kg^{-1} for coarse sediments and 37 5 - 350 Bq m^{-3} kg^{-1} for silts and clay sediments In samples from all regions, radon exhalation was greater for fine grained silt and clay samples than coarser grained sediments This is a result of the increased emanation potential for fine grained sediments as the production sites are likely to be closer to the surface of the grain than in coarser sediments Therefore, radon produced in fine grained sediments has a higher probability of reaching the pore spaces than in coarser grained sediments (Semkow, 1990, Tanner, 1978) However, any of the processes which govern radon emanation and exhalation (in section 7.2) could result in increased exhalation for fine grained sediments when compared with coarser grained sediments

Exhalation from the Peak District limestones was higher than all other regions and the same trend is also observed for sediments from the Peak District caves although the difference is less only 2 - 3 times higher Together, these explain why the overall radon budget is considerably higher in the Peak District and goes some way towards explaining the higher radon concentrations in this region (chapter six)

If radon exhalation rates for limestone rock (table 7 11) are compared with exhalation rates for sediments (table 7 13) it can be seen that in the South Wales, North Pennines and Mendip Hills regions sediments (if present) contribute more to the overall radon budget than limestone rock However, in the Peak District radon exhalation rates from both sediments and rocks are high so neither source dominates and both will control the overall radon budget However, these comparisons have been undertaken using exhalation rates based upon Bq m^{-3} kg^{-1} and the relative importance of each source will have to be determined for each cave dependent upon the nature and amount of sediment in comparison to the surface area of exposed rock.

The radon exhalation rates for sediments which have been divided and sieved into sub samples are given in table 7 14 In all instances silt and clay fractions release more radon than other fractions and therefore potentially contribute more to the overall radon budget than other fractions However, the significance of each fraction to the total radon

budget will be site specific and determined by the particle size distribution of the sediment Table 7 15 outlines the quantities of sediment needed to raise 1 m^3 of cave passage to 1 WL Bottrell (1991) proposed that 17 4 kg of fine grained sediment from Speedwell Cavern would be needed but the figures in table 7 14 suggest that between 24 and 118 kg of sediment would be needed, more coarse grained sediment being needed than fine Bottrell's figure was based upon the original uranium concentration of 20 ppm and it was stated to be a maximum estimate based upon exhalation coefficients of 85% It has been shown (section 7 7.3) that this is too high and is likely to be between 60 and 70% Therefore, the exhalation coefficient used seems to be the source of error

7 7 5 Speleothems

The radon exhalation rates of selected speleothems are given in table 7 16 The survey of speleothems is rather limited and biased to the South Wales region All speleothems were found to release only small quantities of radon gas, independent of radon production rates governed by the original uranium-238 concentration These results confirm earlier work by Lyons *et al* (1989) Due to the small percentage of speleothem within the majority of limestone caves in comparison to other sources such as rock or sediment, speleothems are considered to be of little significance to the overall radon budgets in all but exceptional cases

7 7 6 Faults and joints

The effects of joints and faults on the overall radon budget could not be directly investigated However, inferences drawn in chapter six, with reference to caves in the North Pennines region, suggest that faults may influence observed radon concentrations on a regional basis and are also locally important in accounting for high concentrations The importance of faults and joints may be enhanced if a carrier fluid, such as water, exists to transport radon from 'depth' into the cave environment

7 7 7 Redistribution

The theoretical processes by which radon is redistributed were discussed in chapter six and are considered in more detail in chapters eight and nine On a site basis, redistribution within a cave may operate as both a source and a sink of radon during different seasons of the year However, when the process of redistribution is considered on a larger scale, either regionally or nationally its importance is relatively small It is felt that redistribution is important in controlling radon at a site within a cave However, on the larger scale it is not important as it provides no additional source of radon to the karst environment

7.8 Summary

It has been demonstrated that, sediments are likely to provide the major source of radon to limestone caves where they are present, with fine grained sediments contributing more than coarser grained sediments However, in the Peak District the limestone rock is also a significant source and in some instances will contribute more to the overall radon budget than sediments Water may act as either a source or a sink to radon However, radon is generally released from water to the cave Phreatic passages and percolation water also contribute radon to the cave environment In the Peak District region, radon gas in the soil could potentially be transported to caves However, this source is constrained by the need for mechanisms of transport and the maximum time for transportation determined by radon's half-life of 3 86 days The processes by which faults and joints influence radon concentrations within a cave have not been examined in detail but could provide considerable sources of radon The effect of redistribution of radon was demonstrated in chapter six and where zones of accumulation occur this could pose a significant source of radon to individual sites However, overall redistribution is insignificant regionally or nationally as a source of radon

The overall importance of different sources within individual caves needs to be determined and will depend upon the proportion of each source contained within the

cave The total surface area of each source will be more important than the weight or mass of sediment and therefore even though limestone produces less radon per unit mass than sediment within individual caves, the limestone could still be the primary source due to the surface area exposed, especially where the cave is surrounded by interconnected fissures and voids

CHAPTER EIGHT: RADON GAS IN CAVES OF THE PEAK DISTRICT

8.1 Introduction

Chapter six outlined results from a national investigation of radon concentrations within the main caving regions of England and Wales, in which the primary processes which control radon within caves were identified In this chapter the processes that influence radon within caves solely in the Peak District are examined The investigation was undertaken as part of a project funded by Derbyshire County Council in an attempt to determine and quantify the risk from radon to caving instructors and pupils As a result the location of detectors is slightly biased, all detectors being placed in caves or sections of caves that are regularly used by groups from outdoor pursuits centres

8.2 Methods

A total of 27 sites in nine caves were investigated during the project (for number of detectors in each cave see table 8 1 and for site locations figures 8 1 - 8 9) Radon gas concentrations at each site were determined using NRPB passive track etch radon monitors (see chapter five) Detectors were placed for thirteen 28 day sampling periods, from 6 November 1989 to 5 November 1990 Detectors were placed to ensure that caves used regularly by outdoor pursuits establishments were sampled rather than using a stratified sampling design, such as that in the National survey (see chapter six)

8.3 Results

8 3 1 Summary of results

The highest mean and the highest maximum concentration were both recorded in Giants Hole (7,856 and 30,564 Bq m^{-3} respectively), the mean concentrations in

both Knotlow Caverns (7,728 Bq m^{-3}) and Gautries Holes (6,509 Bq m^{-3}) being almost as high (table 8 2) The maximum concentration in Giants Hole, 30,564 Bq m^{-3}, is substantially higher than that recorded in Gautries Hole (21,189 Bq m^{-3}) all other maxima being below 15,025 Bq m^{-3} (table 8.2)

The mean concentrations recorded in Axe Hole and Jug Holes were considerably lower than those from Giants Hole, and represent the other end of the spectrum, with mean concentrations of 442 Bq m^{-3} and 208 Bq m^{-3} respectively In both cases the maximum concentrations are below 2,000 Bq m^{-3} (table 8 2) In a number of cases (for example, Giants Hole, P8 (Jackpot) and Axe Hole) the standard deviation is larger than the mean which indicates that the spread of the data is very large Gautries Holes also has a large standard deviation which is just below its mean This implies that these caves all have widely fluctuating radon concentrations on a seasonal basis (table 8 2) Minimum concentrations vary between 45 and 1,886 Bq m^{-3}, and in each case these are from sites close to the entrance and / or influenced by incoming relatively radon free air No winter results were collected from Knotlow Cavern, therefroe the mean

and minimum values listed are higher than expected as results from the winter period would be likley to have reduced these figures

If results recorded in table 8 2, derived from the Peak District investigation, are compared with results in table 6 16 and 6 19, recorded during the national investigation, a similarity can be observed, even though the sampling sites were not directly comparable Mean concentrations recorded for individual caves and areas are consistent and within the same order of magnitude

8 3 2 General radon system

Figures 8 10 - 8 18 provide a summary of results for individual sampling periods for each cave investigated The following features are apparent:

1 Concentrations at sites near to the entrance, are lower than those further

into the cave (for example Giants Hole, figure 8 11)

2 Concentrations are generally higher during the months of May, June, July, August, September, October (for example Giants Hole, P8, Gautries Hole, figures 8 10, 8 11 and 8 16)

3 Concentrations are generally lower during the months of November, December, January, February, March and April (for example Giants Hole, P8, Gautries Hole, figures 8 10, 8 11 and 8 16)

4 In certain caves, for example Carlswark Cavern, concentrations at one or two sites are relatively high throughout the year These are usually in the deeper further reaches of the cave (figure 8 12).

The lower concentrations recorded at sites one and two compared with the rest of a cavern could be related to incoming relatively relatively radon free air that dilutes the observed radon concentrations close to the entrance Air will enter the cave for part of the day at all times of the year, even during the summer months Due to thermal differences air will generally be exhaled from cave systems during the summer and air will only move from the external atmosphere into the cave for a limited period of time, usually at night Conversely, during the winter air will generally flow into the cave for the majority of the day, thereby reducing radon concentrations at sites near to the entrance Concentrations increase further into the cave as radon from other areas of the cave will be redistributed to these sites

The increase in concentrations during the summer months and a decrease during the winter period has been well documented and a full summary of these theories are included in section 3 4 Most authors have discussed this problem but the theories to account for the variation were first proposed by Yarborough *et al* (1976) Other work to have been conducted in this area includes that by Gamble (1981), Atkinson *et al* (1983) and Yarborough (1977)

The relatively high concentrations recorded throughout the year at particular

sites within a cave, for example sites 5 and 6 in Carlswark Cavern (figure 8 12) can be accounted for by the cave morphology and sampling location The elevated concentrations recorded in Carlswark Cavern, were at sites in Stalactite Passage (figure 8 3) Two facets of the sampling location could account for the increased concentrations recorded at these sites

1 Due to the morphology of the system, the main air movement is along Eyam Passage, connecting the Gin Entrance with Eyam Dale Shaft (figure 8 3) Hence, Stalactite Passage naturally receives little air movement to dilute/ redistribute the radon in this region

2 Stalactite Passage is part of a series of passages on a lower level to the main section of cave, entrance to this section being protected by a 'sump', a water filled section of passage This could reduce air movement between the lower series and the main section of cave

3 The sump could be transporting radon with the water which is then liberated in to the passage

All of these features could result in elevated concentration being recorded at sites 5 and 6 in Stalactite Passage

Table 8 3 summarises results from individual caves by each season Winter included results from December, January and February, spring included results from March, April and May, summer included results from June, July and August, autumn included results from September, October and November Apart from Devonshire and Jug Holes caverns, mean concentrations during the summer months are higher than during the winter period Results from the autumn and spring periods are variable and lie between concentrations during the winter and summer months During this project, results from the spring and autumn months were more akin to the summer sampling period than the winter period Surprisingly the maximum result (30,564 Bq m^{-3} from Giants Hole) was recorded during the autumn rather than the summer Mechanisms to account for seasonal variations have been proposed by a number of authors (for example

Ahlstrand and Fry, 1976; Carson, 1981, Gamble, 1981, Miki and Ikeya, 1980, Yarborough *et al*, 1976) these have been summarised in chapters three and six.

Table 8 4 summarises results by area From earlier works (for example Gunn *et al*, 1989a) it would be expected that caves in the Castleton area would have the highest means and maxima However, caves in the Lathkill Dale area have the highest mean concentration, althought this can be accounted for by the absence of winter results from this region The second highest mean and the highest maximum was recorded in the Castleton area Comparatively, caves in other areas (Eyam Dale, Buxton and Matlock) have considerably lower mean and maximum concentrations

The null hypothesis that radon concentrations do not vary by area within the Peak District region was analysed using the ANOVA technique This demonstrated that, at the 95% significance level, no difference existed between results from individual regions (F calc 9 17x10^{-2}, F crit 2 86) However, when areas are grouped, results from Castleton and Lathkill Dale differ from those in Eyam which differ from Buxton and Matlock areas Therefore, three different radon systems can be seen to operate, these could be a consequence of differences in the radon budget in each area For example, the caves in Castleton and Lathkill Dale are in a limestone with a high exhalation coefficient or additional sources of radon are available to the karst environment either from sediments or water (see chapter seven) Conversely, caves in the Eyam Dale, Buxton and Matlock regions are in zones with a lower overall radon budget or no additional sources of radon are available to the system.

The lithology at the entrance to each cave was determined and used to categorise the lithology within the cave The assumption that the lithology at the entrance is representative of the entire cave cannot be universally applied as it is known that different limestone beds are encountered within a single cave (for example Giants Hole) However, a null hypothesis was proposed that lithology at the entrance to the cave had no effect on observed cave radon concentrations Analysis using the

ANOVA technique demonstrates that radon concentrations within caves in the Eyam limestones and Apron Reef facies are different from radon concentrations within caves in the Hoptonwood, Woo Dale and Matlock limestones at the 95% significance level. It must be noted that within the Peak District the majority of significant caves are found within Asbian stage limestones (Ford, 1987b) which contains the Bee Low, Apron Reef facies and the Monsal Dale limestones The difference in results due to lithology clearly demonstrates that emanation / exhalation is important in controlling the initial radon concentration within particular caves or areas In chapter seven it was shown that within the Peak District radon exhalation and emanation rates varied for different limestone beds, reinforcing the relationship outlined above However, it was demonstrated that both the containing limestone rock and sediments contained within the cave derived from the allogenic catchment provided significant components to the radon budget

Analysis was performed by cave type, as proposed by Yarborough *et al* (1976) but no relationships between cave type and radon concentrations were observed The additional categories of cave type, proposed in chapter six, were included and further analysis performed but no relationship was observed

8 3 3 Influence of the external climate on the cave radon system

To determine the influence of the external climate on the cave radon system statistical analysis was performed The correlation between radon concentrations for individual caves and various climatic parameters, determined at Buxton weather station during individual sampling periods, were calculated (tables 8 6 - 8 8) The validity of the correlation coefficients was tested using students' T-Tests The null hypothesis that external mean temperature has no influence on cave radon concentrations was rejected for seven of the nine caves (table 8 6) The remaining two caves demonstrate either zero or a weak negative correlation with mean temperature

Some authors suggest that mean temperature is the primary control on radon concentrations (for example Ahlstrand and Fry, 1977; Ahlstrand, 1980, Trout, 1975, Quinn, 1990 and 1992), others have suggested maximum temperature (for example Yarborough, 1976, Yarborough *et al*, 1977, Kobal *et al*, 1987), others have suggested mnimum temperature (for example Quinn, 1988) During this project it has been found that the importance of the relationship between mean, mean maximum and mean minimum temperature varies depending upon the cave under investigation and no generalisation can be established for all caves.

Two of the nine caves, Knotlow Cavern and Jug Holes Cavern, did not return a strong correlation with temperature Therefore, other processes must be operating to influence radon within these caverns Analysis of the correlation between mean radon concentrations and other climatic variables (such as total rainfall or atmospheric pressure) revealed that neither of these two parameters could be demonstrated to influence radon concentrations within the cavern The surveys of these caves reveals two features common to both caves both possess multiple entrances and a complex network of passages which in Knotlow cavern is developed over a considerable depth The radon detectors were placed relatively close to major entrances and therefore the concentrations recorded might not reflect the 'true' concentrations for the caverns.

Table 8 6 demonstrates that 4 caves (Knotlow Cavern, Hillocks Mine, Jug Hole Cavern and Axe Hole) have recorded significant correlation with atmospheric pressure There has been considerable debate concerning the influence of pressure on radon concentrations A number of authors feel that temperature is the critical factor (for example Yarborough *et al*, 1976) conversely some authors feel that pressure is important (Ahlstrand, 1980, Carson, 1981 and Navratil and Stelcl, 1990) Some authors feel that pressure at the time of the measurement is important (Ahlstrand, 1980 and Williamson, 1990) while other feel that the previous day's pressure or changes in pressure during the previous day / week are more important (Carson, 1981, Middleton, 1988, Yarborough, 1981) In all cases where pressure has been deemed to be important, the caves

have been classified as RSU by Yarborough *et al* (1976) (see Tables 8 6, 8 7, 8 8)

The only positive correlation between total rainfall and mean radon concentrations was at Devonshire Cavern This relationship was not very strong A slightly stronger correlation was observed with maximum radon concentrations at Knotlow cavern This implies that increased surface runoff, and consequently increased discharges within underground streams, could increase air movements within caves and therefore affect radon concentrations Cigna (1971) inferred that air movements within caverns could be influenced by water levels However, sufficient data does not exist to allow for the hypothesis to be tested further Additional analysis will be undertaken in chapter nine A weak negative relationship between total rainfall and mean, maximum and minimum radon concentrations was returned for all other caves This implies that mechanisms outlined by Surbeck and Medici (1990) (outlined in section 3 4 7) whereby soil acts as a source of radon to the cave environment could be operating

8 3 4 Influence of climatic variables on seasonal cave radon concentrations

Correlation analysis was also performed between climatic variables and radon concentrations that had been grouped into season Not all caves were grouped as caves with less than three sites could result in correlation coefficients being derived that were not statistically valid Therefore, five out of the nine caves, P8 cave, Giants Hole, Carlswark, Knotlow and Devonshire Caverns were grouped The statistical significance of the correlation coefficients was confirmed using students' T-Tests

When correlation coefficients were examined, by season, the influence of temperature on cave radon concentrations was clearly evident The correlation is statistically significant in P8 (a RSU cave) during the spring and autumn

sampling periods, in Giant's Hole (a USD cave) during the winter, spring and autumn, in Carlswark Cavern (a USD cave) during all sampling periods and in Devonshire cavern (a USD cave) during spring and summer sampling periods (table 8 9) Temperature is an important control on cave radon concentrations during all seasons but its importance varies throughout the year and depends upon the cave under investigation related to the cave's morphology However, other facets such as altitude, position, depth, presence of a stream might need to be included in the cave morphology definition, to allow for cave morphology to be used as a tool to account for the radon system.

Significant correlations between total rainfall and cave radon emerged when data were grouped seasonally Negative relationships were observed for P8 (in winter), Giant's Hole (in spring), Knotlow Cavern (in summer) and positive relationships in Devonshire Cavern (in winter) and P8, Carlswark Cavern and Giants Cave (during the autumn)

One possibility is that colinearity exists between temperature and total rainfall, either overall or when grouped seasonally, which would result in the mechanisms outlined to explain temperature related variations also being valid for variations attributed to total rainfall. Correlation analysis demonstrates that a weak colinearity does exist between temperature, total rainfall and pressure Therefore, the relationship observed between total rainfall, pressure and cave radon concentrations could be partly coincidental (accountable by temperature) and partly due to distinct processes operating

The negative relationships could be explained by increased rainfall resulting in more discharge entering and flowing through the cavern The moving water would both entrain air within it and, due to friction between the air and water interface result, in air movements However, more likely to increase the volume of air moving within the cavern is the pumping affect of water falling down waterfalls The increased air movements would result in more relativley radon free air entering the cave systems thereby promoting a lowering of radon concentrations by dilution, resulting in a negative correlation coefficient A

positive correlation coefficient could be returned if the rainfall transported radon from the soil profile into the cavern within percolated water Surbeck and Medici (1990) showed that the soil contains elevated radon concentrations, and the soil was proposed as a source of radon to the karst environment Any water moving through the soil will absorb radon and when the percolating water enters the cave the radon is degassed into the atmosphere Therefore, increased rainfall could result in increased radon transport, thereby resulting in a positive correlation coefficient being returned It is interesting to note that the highest correlation coefficients were returned during the autumn period During the summer the soil is relatively dry and radon concentrations in the pore spaces increases The increased rainfall in autumn could displace the increased radon accumulated in the soil during the summer Conversely, during the winter period due to the continued high rainfall the radon concentration within the pore spaces would not increase greatly, reducing the importance of the soil as a source of radon to the cave environment compared with the autumn months

When radon concentrations are grouped into seasons the null hypothesis that cave radon concentrations are not affected by atmospheric pressure is rejected at certain sites, both positive and negative correlation coefficients being returned (table 8 9) Positive correlation coefficients were obtained for Carlswark and Devonshire Caverns (during the winter) and Devonshire Caverns during the spring Negative relationships were returned for Knotlow and Devonshire Caverns (during the summer), Giants and P8 cave (during the autumn months) Previously authors had only reported a negative relationship with pressure for RSU caves, as defined by Yarborough *et al* (1976) Part of the relationship observed can be accounted for by the colinearity between temperature and pressure but distinct processes will also be operating

The negative relationship observed in RSU caves (P8) has been reported by numerous authors, the first account was provided by Yarborough *et al* (1976) and is summarised in chapter three

This project is the first to identify both positive and negative relationships between pressure and radon concentrations in USD caves Mechanisms will be proposed to account for these relationships In USD caves during the summer, air enters the cavern via an upper portal and moves through the system to a lower entrance Conversely, during the winter months air enters the cavern via the lower portal and leaves the system by the upper opening Therefore, the effects of pressure on radon within the cave will vary in relation to the monitoring site In the examples below the extremes will be presented with site one referring to a monitor located close to the lower entrance, site two is located close to the upper entrance It is assumed that the effects of pressure acts uniformly on the volume of air within the cave Therefore, if pressure decreases the volume of air within the cavern will increase uniformly throughout the cavern, which will result in a net expansion of air at both portals

If during the summer, when air is entering the cavern via the upper entrance, atmospheric pressure increases then the volume of air within the cavern will decrease This will result in a uniform contraction of air within the cavern resulting in air being drawn into the cavern At the lower portal, site one, the contraction of air will be in opposition to the natural air movement, which is outward This will result in the two forces acting in opposition and this could result in a decrease in the overall velocity of air leaving the cavern or a complete reversal. A decrease in the velocity of air at site one will result in a reduction in the effectiveness of the redistribution processes, promoting an increase in radon concentrations Therefore, at site one an increase in atmospheric pressure will result in an increase in radon concentrations which would return a positive correlation coefficient Conversely, at site two, at the upper entrance where air is actively being drawn into the system, the contraction of the cave air will supplement the natural air flow potentially resulting in an increase in air velocity resulting in increased dilution by incoming relativley radon free air Therefore, at site two, increases in pressure will result in decreases in radon, and therefore a negative correlation coefficient will be returned However, if you look at the cave overall these effects will be 'averaged' out and the overall effects of

pressure theoretically will be zero

If during the summer atmospheric pressure decreases then the volume of air within the cavern will expand. At site one, at the lower entrance, the expansion of the air will increase the velocity, and therefore increase the amount of dilution, thereby reducing the radon concentration Therefore, a decrease in pressure will result in a decrease in radon concentration and therefore a positive correlation coefficient will be returned. Conversely, at site two, at the upper entrance, the expansion of the cave air will be acting in opposition to the natural thermally induced air flow, thereby reducing the velocity of incoming relatively radon free air and consequently reducing the effectiveness of the dilution mechanisms This will result in an increase in radon concentrations with decreasing atmospheric pressure, thereby resulting in a negative correlation coefficient

During the winter months these processes will operate in reverse and negative correlation coefficients will be returned for sites close to the lower entrance, while positive correlation coefficients between pressure and radon concentrations will be returned from site two, near the upper entrance

8.4 Summary

1 Typical radon concentrations for caves in the Peak District, used by outdoor pursuits centers, were determined

2 The results show a clear seasonal trend This phenomenon has been previously noted by numerous authors (for more details see chapter three)

3 The results suggest that certain limestone beds have a higher emanation / exhalation coefficient or that radon generated from other sources might influence cave radon concentrations These results confirm the conclusions drawn in chapter seven

4 Over the twelve month sampling period there was a positive correlation between 28 day mean, maximum and minimum radon concentrations and external temperature

5 When the data are grouped into seasons, the importance of mean, minimum and maximum temperature on cave radon concentrations

remains However, the influence of total rainfall and mean atmospheric pressure on cave radon concetrations also becomes apparent

6 Ramfall could affect two mechanisms of the cave radon system. Firstly the soil could act as a source of radon to the karst environment during certain seasons and secondly, discharge in underground streams, related to rainfall, may influence air currents in caves that could influence cave radon concentrations

7 Both positive and negative relationships between cave radon concentrations and atmospheric pressure were observed Previously, in the literature only negative relationships in RSU caves had been recorded New theories were proposed to account for the relationships in USD caves

CHAPTER NINE: RADON AND RADON DAUGHTER CONCENTRATIONS IN PEAK CAVERN

9.1 Introduction

Chapters six and eight presented results from measurements of mtegrated radon and radon daughter concentrations wthm lmestone caves However, due to the scale of measurement, bemg mtegrated concentratons, the results do not allow for detailed analysis of the cave radon system and the development of models to predct cave radon concentrations Hence, a more detailed study was undertaken m a single cave, Peak Cavern, m the Derbyshire Peak Dstrct

9.2 Methodology

Radon and radon daughter concentrations were determned usng the techniques outlined in chapter five Radon gas concentratons were determned usmg Lucas cells and radon daughter concentratons were determned usmg IWLM Meteorologcal data were collected wthm the cavern usng thermometers (accurate to ± 0 2 °C) and a barometer Air drecton was determned using joss-sticks Attempts were made to determine air speed usmg a specially calibrated hot wire anemometer, with two calibraton ranges (0 - 0 5 ms^{-1} and 0 5- 5 ms^{-1}) However, due to the heat radiated by the operator, durng measurement, convection currents were established wthm the vcnty of the site preventng accurate measurement m the range 0 - 0 5 ms^{-1}

Data for the outside clmate were available from three sources

1. A contnuously recording meteorologcal staton, at Oxlow Farm (NGR 124821) approxmately 2 km from the entrance to Peak Cavern

2 Daily meteorological records from Buxton Weather staton,

approximately 20 km from the entrance to Peak Cavern.

3 Three hourly meteorological records from Manchester Airport, approximately 40 km from the entrance to Peak Cavern

9.3 Sampling

9 3 1 Sampling Locations

Gill and Beck (1991), Beck (1991) and Nash (1991) have all provided descriptions of Peak Cavern Although, the Peak - Speedwell cave system extends for approximately 14 km (5 5 miles), during this research project measurements were only conducted at 28 sites (table 9 1 and figure 9 1) within approximately 2 5 km of the Peak Cavern entrance Sampling was undertaken weekly for 56 weeks from June 1991 to July 1992 However, analysis is restricted to complete sets of measurements obtained during 47 of the sampling periods

Figure 9 2 outlines the broad relationship between passage development, geology and mineral veins The entrance to the cave, including the section used as a Show Cave during the summer months, is within the Apron Reef facies The majority of the remainder of the cave is contained within the Bee Low limestones Two mineral veins cut the cave, Faucet Rake near to the top of the Devil's Staircase and New Rake which controls development in the region around Galena Chamber (see Beck, 1980, Beck, 1991, Nash, 1991) For the purpose of this research the cave has been divided into five areas (outlined in table 9 1 and figure 9 1)

1 Entrance series comprising sites from the Entrance to the downstream end of Buxton Water Sump

2 The Upper Gallery sites from the downstream end of Buxton Water Sump to Treasury Chamber These connect through the Trenches to the White River Series and Speedwell Cavern although results from these

sections of cave are not included within this chapter (see appendix one)

3 Main Streamway the active streamway fed by Far Sump and Main Stream Inlet to the upstream end of Buxton Water Sump

4 Lake Passage the passage from Ink Sump that flows through Lake Sump into the Main streamway

5 Relict Passages these connect the Upper Gallery region, via Galena Chamber to the Main Streamway This series can be entered at Surprise View and joins the Main Streamway at Main Stream Inlet

9.4 Results

The Kolmogorov Smirnov test was used to determine if radon daughter concentrations and meteorological data fitted a normal distribution A significant fit was observed at the 95% level Therefore, parametric statistical tests can be used on all results

Table 9 2 outlines results by area for all sampling periods Overall concentrations range from 0 0001 to 22 41 WL, with a mean of 1 27 WL However, concentrations within the cave are not uniform and differences exist between areas This implies that different processes must operate to control concentrations The range of concentrations in areas one and two (0 0001 - 22 41 and 10 84 WL) are much greater than in areas three, four and five (0 13 - 6 41 WL) Table 9 3 and figure 9 3 outline mean radon daughter concentrations by individual sites Two sites, Treasury Sump (14) and the Great Cave (4), exhibit increased concentrations relative to those surrounding them. A mechanism was outlined in chapter six to account for the elevated concentrations at site 14, Treasury sump, where it was proposed that the site is acting as a radon sump Increased concentrations were also observed at site 4, the end of the Great Cave, during the summer months On 01/06/92 the highest concentration

recorded during this project (22 41 WL) was measured at this site. As demonstrated in chapter seven the Apron reef facies, in which this site is contained, have higher emanation / exhalation potential than other lithologies within the cave Therefore, the increased emanation / exhalation potential could result in elevated concentrations However, localised redistribution mechanisms account for the increased concentrations observed These can be explained by the differences in air density When the internal air density is higher than the external air density (generally during the summer months) air will flow out from the far reaches of the cave to the entrance Air will be drawn along the White River Series, the Trenches, the Upper Gallery and along the entrance series into the Great Cave As air flows along this path radon is mobilised and transported leading to radon rich air moving towards the entrance of the cave However, in the roof of the Great Cave a connection exists to the surface in Cave Dale This results, in a secondary air circulation pattern, with air entering the Great Cave from Cave Dale and flowing out of the entrance Where these two air masses meet, thermal differences exist and stagnation of air flowing from the far reaches of Peak Cavern occurs, resulting in elevated concentrations Due to the thermal characteristics of the two air masses, the location and characteristics of the stagnation zone vary and when the mixing zone does not coincide with the sampling site pronounced elevation (as occurred on 01/06/92) is not recorded Mutter (1987) also recorded elevated concentrations within the Great Cave but no mechanisms were proposed to account for them.

Table 9 4 and figure 9 4 outline variations in mean radon daughter concentrations by sampling period A seasonal trend in the results can clearly be seen, the highest concentrations being recorded during the months of May 1992 to August 1992 However, occasional increases in concentrations are observed during September 1991, November 1991 and January 1992 The processes that induced seasonal variations in radon daughter concentrations have been outlined in chapters six and eight and the elevated concentrations recorded in September and November 1991 can be accounted for primarily by thermal differences between the internal cave atmosphere and external air However, the high

concentrations in January 1992 are not related to thermal differences but are a result of flooding Radon daughter concentrations were recorded at half-hourly intervals during this period using the IWLM and a data logger (figure 9 5) During the three days prior to measurement 40 mm of rain fell, with a further 20mm during the measurement period This resulted in a flood within Peak Cavern. The increased discharge had two affects within the cave

1 There was a net expulsion of air from the cavern which may have reversed the thermally induced air movement or reduced its effectiveness

2 Sediments within the cavern were reworked In chapter seven it was demonstrated that sediments within limestone caves are a source of radon As the sediments were disturbed, radon contained within the pore spaces will have been released into the water and subsequently liberated to the air due to the turbulent flow

At present there are insufficient data to determine which factor is most important in influencing radon daughter concentrations during a flood events However, it is felt that the increased exhalation rate would be more important, but in all instances both processes will operate

Chapters six and eight provided basic summaries of the general cave radon system, and therefore little further attention will be devoted to discussing the general cave radon system within Peak Cavern The emphasis of this chapter will therefore be directed at developing and testing detailed models to account for the cave radon system in Peak Cavern

9.5 Development of Models

Models have been developed to predict mean cave radon concentrations based upon the meteorological indices outlined in table 9 5 All models were developed using SPSS for Windows version 6 0 Models were developed using data from sampling periods 1 - 23 and verified using data collected during sampling periods

24 - 47 As outlined in chapters six and eight distinct seasonal variation is apparent within cave radon data and therefore the use of data from sampling periods 1 - 23 to develop the models and verification using data from sampling periods 24 - 47 could result in errors, as each data set represents part of different seasons However, this source of error, due to the size of the data set, will be relatively small and valid predictive models will still be able to be developed

It has been proposed by Atkinson *et al* 1983, Quinn, 1990 and 1992, Yarborough *et* al. (1976) that either a single variable or a limited number of variables can be used to account for the majority of the variation observed in the cave radon data set As a consequence, the initial stage was to undertake correlation analysis between the radon daughter concentration at each site during each sampling period and all the corresponding meteorological variables It was demonstrated that no single variable was found to be significantly related to radon daughter concentrations Therefore, any model developed would need to be based upon a combination of variables rather than a single index. The determination of correlation coefficients allowed for colinearity between variables to established (table 9 6)

Subsequently multiple regression analysis was undertaken and of the 42 variables included in the analysis, only 23 were included within the final multiple regression equation (outlined in table 9 7) However, the practical use of a multiple regression equation with 23 variables included has to be questioned Therefore, even though the model is statistically valid, and accounts for 47 9% of the variation in mean radon concentration for the whole cave, it is not practical for modeling purposes

Stepwise multiple regression was then performed on all the cave radon data from Peak Cavern In total, all 42 variables outlined in table 9 5 were included Of the 42 variables entered, 18 were demonstrated to significantly contribute to variations in cave radon concentrations within Peak Cavern In total these accounted for 46 1% of the variation Only the five most significant variables are included and these account for 35 1% of the variation Table 9 8 outlines

the variables used in the equation to develop figure 9 6 which demonstrates the relationship between predicted and mean recorded radon daughter concentrations The order of the variables, listed within the tables, relates to the calculated statistical significance Therefore, in table 9 8 the previous week's minimum external air temperature is more significant in controlling concentrations than the previous week's maximum air pressure Figure 9 6 clearly demonstrates that predicted values tend towards the mean recorded concentration for individual periods However, when extreme concentrations are recorded (above 10 WL) the predicted concentrations clearly do not relate to recorded concentrations and at low concentrations negative values are returned Deviation is expected as the proposed model only accounts for 35 1% of the observed variation Therefore, to allow for improved prediction another variable might be needed to be added into the equation

It has already been indicated that different areas within the cave have different processes operating to control concentrations and therefore if components to represent different areas within Peak Cavern are introduced, the accuracy of predicting mean cave radon concentrations might be increased Therefore, further stepwise multiple regression analysis was undertaken using data from each of the five areas within Peak Cavern All 42 variables were included in the calculations and between 17 and 21 variables were demonstrated to be important In the models presented below only the five most significant parameters are included, listed in the tables in the order of statistical significance The regression equations and the parameters from which they are derived are included for each area in tables 9 9 - 9 13 The previous model accounted for 35 1% of the variation within the cave radon data set, these models account for between 38 2 and 65% of the variation within the cave radon data set Figures 9 7 to 9 11 outline predicted and mean radon concentrations for individual areas within Peak Cavern In all models for individual areas the predicted cave radon concentrations are within an acceptable margin (betweeen 38 2% and 65%) of recorded cave radon concentrations, allowing for cave radon concentrations within Peak Cavern, to be predicted from meteorological data One major

variable has been omitted from the models, the emanation / exhalation coefficient for either the entire cave or areas within it The inclusion of this variable would allow for increased accuracy of prediction However, overall the models developed do allow for prediction of radon concentrations within Peak Cavern

9.6 Application of models to other cave systems

The models outlined in tables 9 9 - 9 13 and figures 9 7 - 9 11 can be used to predict general concentrations within Peak Cavern However, the validity of the models proposed had previously only been confirmed for Peak Cavern Therefore, the models were tested by using data from two Show Caves The first, Treak Cliff Cavern, is within 1 5km of the entrance to Peak Cavern, while the second cave, Cheddar Show Caves is in a different geographical region, the Mendip Hills Radon daughter concentrations have been recorded monthly at both caves using the Kusnetz method (see chapter five), meteorological data at individual sites within both caves were collected during sampling Additional meteorological data is available for Cheddar Show Caves from Bristol Airport, while all the data sources outlined in section 9 2 are applicable to Treak Cliff Cavern

Figure 9 12 outlines recorded and predicted mean radon daughter concentrations for Treak Cliff Cavern, based upon the model derived for all areas of Peak Cavern Figure 9 12 clearly demonstrates that the model derived for Peak Cavern can not be transposed onto Treak Cliff Cavern Predicted values do not trend towards the mean concentration recorded Therefore, additional variables need to be included within the model to account for concentrations Figure 9 2 outlines one initial source of error, the model derived for Peak Cavern was based upon radon budgets controlled by cave development in the Apron Reef facies, Bee Low and Eyam limestones whereas Treak Cliff cavern is contained solely within Apron Reef facies limestone This difference in lithology was demonstrated in chapter seven to influence the radon budget The Apron Reef facies limestones were shown to have a higher release potential than other

limestones from the Peak District (see chapter seven) Therefore, the increased radon release rates in Treak Cliff Cavern explain why recorded concentrations are higher than predicted concentrations An additional source of error is related to cave morphology Peak Cavern can be defined as a large cave system with considerable amounts of cave passage (in excess of 5 miles) conversely Treak Cliff Cavern is relatively small (less than 300m long)(figure 9 12) As suggested in chapter six and eight, the difference in morphology will affect redistribution and dilution mechanisms operating within the respective caverns

Figure 9 13 demonstrates that differences exist between predicted radon concentrations and means and maximum concentrations for Cheddar Show Cave As demonstrated in chapter six, regional variations are apparent in cave radon concentrations, and therefore the difference observed between predicted and recorded concentrations in not surprising In chapter seven it was demonstrated that the radon budget was different for each caving region based upon emanation / exhalation from both limestones and sediments Generally, predicted concentrations (figure 9 13) are overestimates of concentrations recorded in Cheddar Show Caves This suggests that radon generation in the Peak District is higher than in the Mendip Hills, as demonstrated in chapter seven

As was demonstrated in chapter seven the radon budgets contributed significantly to the overall radon system and therefore any model developed to account for radon in caves generally will need to consider the radon budget within the model otherwise, as with the model developed above, they will only be valid for the cave or caves in which the data were collected However, limited models can be developed to account for spatial and temporal variation within specific caves which can be used to predict concentrations with a degree of accuracy Evidence from this study indicate that predicted concentrations for any site are within 15% of recorded concentration

9.7 Factors which influence cave radon concentrations

In earlier chapters it was hypothesised that single climatic variables were influential in controlling cave radon concentrations As outlined above a single variable can not be identified, using multiple regression, that controls cave radon concentrations within Peak Cavern the Peak District In an attempt to determine if variables can be combined as factors which operate in conjunction to control cave radon concentrations, factor analysis was undertaken The use of factor analysis to reduce data is widely accepted, especially within the fields of archeology, while Christopher and Wilcock (1981) used factor analysis to reduce a data set related to their investigation of the geochemistry of Derbyshire groundwaters in an attempt to explain the variation observed It was felt that similar analysis would be beneficial in aiding the interpretation of cave radon data, and therefore factor analysis was undertaken on the meteorological data set using SPSS for Windows Factor analysis examines the data set and uses correlation coefficients to establish variables within a data set which may be linked These combined indices can then be used as a single index to account for the variation in the data set being examined Factor analysis resulted in 34 factors being identified that account for all the variations observed in the cave radon data (table 9 14) However, only nine factors have eigen values over one and are deemed significant (Davis, 1973) and therefore these will be discussed in detail. The remaining factors with eigen values less than one which represent approximately 15% of the variation within the cave radon data will not be considered further Tables 9 15 - 9 23 outline the variables that have been combined to create factors one to nine The primary variables that each factor represents can be summarised as follows

1 Factor one accounts for changes in cave radon concentrations that are dependent upon temperature

2 Factor two relates to atmospheric pressure

3 Factor three represents small changes in atmospheric pressure (± 1 Mb)

4 Factor four represents larger scale changes in pressure (± 2 or 3 Mb)

5 Factor five represents the effects of external wind direction

6 Factor six relates to increases in atmospheric pressure by 2 Mb

7 Factor seven relates to the influence of the previous weeks rainfall.

8 Factor eight represents the influences of temperature dependent air movements

9 Factor nine represents large falls in atmospheric pressure (-3 Mb)

It is clear from tables 9 15 - 9 23 that temperature and pressure (both at the time of measurement and changes in pressure prior to undertaking the readings) are key factors influencing cave radon, as has been demonstrated in chapters six and eight and by previous work, which was summarised in chapter three Due to the colinearity between temperature variables either, mean, maximum or minimum temperature cannot be isolated and attributed as being the dominant control, since all are important Both static pressure (at the time of measurement) and changes in pressure over time prior to measurement are important and again one can not be used in isolation

Factor analysis has also demonstrated that two additional variables also exert an important influence on cave radon concentrations Firstly, external wind direction which presumably influences the amount of incoming radon free air being forced in the cave and therefore adds an additional mechanism by which concentrations can be lowered by dilution Although, the influence of external wind direction has been proposed by a number of authors (e g Atkinson, *et al*, 1983, Ahlstrand, 1976, Yarborough *et al*, 1976) it has never been considered as seriously as temperature or pressure The second variable is rainfall on a short term basis (based upon weekly figures) This implies that transport of radon in the karst environment, as proposed by Friederich (1981) and Surbeck and

Medici (1990) might be more important than initially thought These processes are considered in more detail later in this chapter

In all of the models developed using stepwise multiple regression (outlined in tables 9 8 - 9 13), a variable dependent upon temperature and pressure (either static or related to the amount of pressure change during either the previous day or week) is included Therefore, the processes outlined in chapters six and eight and those widely accepted within the literature (outlined in chapter three) can be accepted to explain variations in cave radon concentrations observed However, in all instances external wind direction is demonstrated as being important along with either monthly rainfall or weekly rainfall or a combination of the two Therefore, the two new variables highlighted by factor analysis have also been deemed significant during multiple regression analysis

The effects of the external wind direction were demonstrated to be important in all models outlined in table 9 8 - 9 13 and by factor analysis The effects of the external wind direction on recorded cave radon concentrations is related to the situation and morphology of the cavern Peak Cavern is situated at the far end of the Hope Valley, circa 15km long When the wind is blowing in the right direction (from the north or south), the valley channels air movement within it, resulting in wind moving predominantly either up or down valley The entrance to Peak Cavern is contained within a narrow gorge angled approximately north/south resulting in further channeling of air movement This results in air being channeled directly towards the Peak Cavern entrance when the external wind conditions are in the right direction When air is being forced directly at the entrance, which is approximately 30m wide by 20m high, the net effect is to force radon free air into the cavern. This may occur during any season of the year, and results in increased dilution of cave radon Figure 9 3 clearly demonstrates that the overall mean for sites one, two and three is lower in comparison to other sites within the cavern During the winter, air forced into the cavern will combine with the naturally thermally induced incoming air to further decrease radon concentration However, during the summer when the thermally

mduced air movement will be out from the entrance, the effects of the external wind direction will oppose the natural air movement and the overall dilution effect will less pronounced. Figure 9 15 shows results from the entrance series on 08/06/92, when air was naturally moving out from the cavern and air was flowing towards the entrance Concentrations can be seen to increase from Buxton Water to the Great Cave where a maximum is recorded However, at the entrance site and Lumbago Walk concentrations have clearly decreased One possible explanation is air flowing up the valley and gorge are being forced into the cave, resulting in a decrease in concentrations at the first two sites

The other new variable identified as significant is rainfall at both weekly and monthly intervals Surbeck and Medici (1990) proposed that the soil contains high levels of radon and that this could therefore be transported into the cave by percolation water controlled by rainfall. However, no indication was given of the time scale involved Shimo and Yamauchi (1980) demonstrated that precipitation influenced radon concentrations in an industrial tunnel, in a non karst area However, they did not propose any mechanisms to account for the relationship observed Andrews and Wood (1972) recorded radon concentrations in springs on the Mendip Hills and found a seasonal pattern, summer concentrations being lower than winter, completely opposite to the cave radon system. Water percolating through the rock mass will take approximately 25 days to reach equilibrium with radon and a value of 35 pCi l^{-1} (1300 Bq l^{-1}) would be expected (Andrews and Wood, 1972, Friederich, 1981) Bottrell and Atkinson (1992) propose three reservoirs in the unsaturated zone in the Carboniferous limestone around White Scar Cave the first less than three days, second from 30 - 70 days and the third from 150 - 170 days Due to the fracturing within the epikarst Friederich (1981) proposed that this region would be the major source of radon However, it is felt that the influence of soil radon must not be overlooked especially when soil radon concentrations recorded in table 7 7 (between 30,000 - 78,000 Bq m^{-3}) are considered

Rainfall on a monthly basis was demonstrated to be influential for the entire cave

and in areas 2, 3, 4 and 5, while rainfall on a weekly interval was influential for the entire cave and in areas 1, 2 and 5 These results imply that two separate processes operate to control cave radon concentrations Rainfall data on a monthly basis represents percolation water in equilibrium, with radon derived from the surrounding rock, which has taken in excess of 25 days to reach the cave The majority of radon will be derived from the surrounding rock mass and the influences of the epikarst and the soil will have been reduced due to the residence time This rate relates to Bottrell and Atkinson's (1992) second reservoir of 30 - 70 days Rainfall on a weekly basis represents water that enters the karst system via relatively quick routes, and therefore will not be in equilibrium. Friederich (1981) defined this route as shaft flow or it could relate to the first reservoir proposed by Bottrell and Atkinson (1992) In theory, as the water has passed rapidly to the karst system it will contain little radon derived from the rock mass Due to the short residence time, the majority of radon in these waters will be derived from the soil and the epikarst Beck (1980) while discussing the speleogenisis of Peak Cavern refers to

> *"the Cave dale lava/ Nettle tuff overlies much of the Peak / Speedwell cave system The tuff acts as a aquiclude, preventing a large number of small percolation inlets to the cave from developing Instead, there are a few streamlets which have passed through the tuff where it is breached by faults or major joints" (Beck, 1980 p147)*

This will allow for a route by which water can rapidly move from the surface to the cave increasing the influence of rainfall on a weekly basis The tuff, due to its volcanic nature, contains uranium, and initial limited measurements indicate concentrations ranging from 25 - 35 ppm. This is significantly higher than the surrounding limestone and therefore will act as another source of radon in conjunction with the soil and epikarst

The importance of rainfall on a weekly basis was demonstrated for areas one, two and five, while no relationship was demonstrated for areas three and

four Beck (1980) continues to state that the 'umbrella' effect of the tuff is not extensive over the entire system and certain sections within the Main Streamway are not covered by the Tuff (for example Pickering's Series) The Main Streamway is within area three and rainfall on a weekly basis was not demonstrated to be influential. Therefore, the Cave Dale lava/ Nettle tuff must affect the process by which rainfall on a weekly basis enters Peak Cavern Either the presence of the Tuff allows for the development of shaft flow, promoting rapid transport of radon from the soil and epikarst or the Tuff is significantly contributing radon to water which is represented by weekly rainfall. At present the exact extent of the Tuff overlying the Peak /Speedwell cave system is not known

9.8 Summary

The general radon system has been outlined for Peak Cavern, using detailed results collected on a weekly basis The mean concentration for the cavern during 1991 - 1992 was 1 27 WL However, individual site concentrations ranged from 0 0001 to 22 41 WL Spatial and temporal variations in concentrations were found within the cave Meteorological variables, as outlined in chapter three were used to account for the majority of variation in cave radon concentrations However, floods were shown to account for sudden and short lived increases in radon concentrations

A single model for the entire cave and five sub models for individual areas within the cavern were developed using stepwise multiple regression Only the five most significant variables were included in the models which accounted for 38 2 - 65% of the variation observed

In four of the models developed, the previous week's minimum temperature was the most dominant factor while external wind direction was the most dominant in the remaining models Pressure, either at the time of measurement or changes in pressure were included in all models Additional variables included were previous

week's rainfall and monthly rainfall. Mechanisms were proposed to account for the influence of these on cave radon concentrations

The models were tested using data from two Show Caves, and it was clearly demonstrated that models developed to predict concentrations within Peak Cavern could not be used to predict concentrations within other caves More sophisticated models would need to include data on cave morphology, the radon budget and the caving region concerned.

CHAPTER TEN: RISK ASSESSMENT FOR PERSONS ENTERING LIMESTONE CAVES

10.1 Introduction

This thesis has been a scientific investigation of radon within limestone caves from a geographical and geological prospective However, it has a practical application in that any visitor to a cave will receive a radiation dose and in some cases this may be sufficiently high to represent a threat to health Hence, this chapter briefly examines the related health issues

The health risk associated with the decay of radon and its daughters relates to the proven link between radon and both lung and skin cancers (e g Eatough and Henshaw, 1990, BEIR IV, 1988) All caves contain radon gas at concentrations ranging from those in the outside atmosphere to several thousand times the background At the lower end of the range the risk to health from spending a few hours underground will be no different from the risk of walking on the surface above the cave However, as concentrations increase the risk becomes proportionately higher As yet there is no indisputable proof that cavers' health has been adversely affected but research in uranium and coal mines in the United States of America and in England and Wales has shown that a risk exists in these environments (Camplin *et al*, 1988, Davies, 1989, Morrison *et al*, 1988) and in homes (Miles, 1991, Miles *et al*, 1981 and 1992, NRPB, 1989, Puskin and Nelson, 1989, Read, 1990) As no detailed risk assessment has been performed for limestone caves, the guidelines set out in the Ionising Radiation Regulations (1985) (IRR's) will be used as a basis for comparison These were drawn up in 1985 to safeguard the health of workers exposed to radionuclides in all environments (Age and Smith, 1992)

10.2 The Health and Safety at Work Act (1974)

Until 1985, legislation concerning ionising radiations was very restricted in its application This was because the legislation was made under the power of the Factories Act (1961) and hence only applied to the factories and other premises which the act covered This limitation was eventually dealt with when, using powers of the Health and Safety at Work Act (1974), the old legislation was repealed and a new piece of legislation, the Ionising Radiations Regulations (IRR's), was introduced Because the Health and Safety at Work Act covers all types of work, the new IRR's were drafted to cover all work situations In accordance with the underlying principles of the Health and Safety at Work Act, the new IRR's were also drafted to cover not only employees working with ionising radiations, but also other persons who are exposed to ionising radiations whilst at work

The IRR's created a special definition of work because of the nature of radon In essence the IRR's regulate exposure to radon if there is any 'work' in which there is exposure of 'a person' to an atmosphere containing radon above a concentration of 0 03 WL In this context, the definition of 'a person' covers not only an employee but also any other person The IRR's also state that a self employed person is to be treated as an employee for the purpose of controlling exposure to radiation

10 2 1 The Ionising Radiation Regulations (1985)

The IRR's are the regulations that control the exposure of employees to ionising radiations in the United Kingdom. They are based upon the Unsealed Sources Regulations (1968) and the Sealed Sources Regulations (1969) which were developed into the IRR's following recommendations by the ICRP (International Commission on Radiological Protection) which had been adopted by the NRPB and proposed to the UK government All of the aims outlined by the ICRP have been included and developed by the IRR's

The IRR's (1985) came into force on 1 October 1985 and were designed to introduce conditions whereby doses of radiation would be maintained at an acceptable level,

and well below the threshold for ill-health The IRR's are only concerned with exposure to ionising radiations arising during work activity The central principle is that all necessary steps shall be taken to reduce, so far as is reasonably practicable, the extent to which people are exposed to ionising radiations

10 2 2 Approved Code of Practice (ACOP part 1 and 2)

This was issued by the HSE to come into force on the same date as the regulations The ACOP details acceptable methods of meeting the regulations, but allows for the use of newer methods that may be developed if these are as good as, or better than, those described in the ACOP The ACOP (parts 1 and 2) deals comprehensively with the requirements for sealed sources of ionising radiations

10 2 3 Approved Code of Practice (ACOP part 3)

This is entitled 'Exposure to Radon' and gives specific guidance on certain work activities involving exposure to radon These work places are mainly in mines, but they can also occur in tourist mines, caverns, civil engineering work, underground tunnels, and in certain basements of buildings The ACOP part 3 came into operation on 4 April 1988 and was drawn up by a group of experts representing the CBI, TUC, HSE and NRPB

10 2 4 ACOP advice on control (Regulation 6)

The code provides advice on how exposure to radon daughters can be restricted by minimising the entry of radon to the work place, by good ventilation and by limiting exposure durations

Control of radon at source can be improved by effectively sealing off old workings, including bore holes, planning ventilation paths to avoid contamination from old workings, minimising the exposure of rock by concentrating workings, removing ore crushing and handling operations from intake airways, and pumping water if it passes

through the mine As dissolved radon gas is released rapidly from old mine water it is recommended that turbulence and water exposure is avoided

Limiting exposure times for employees is recommended, but implementation is left to the employer This occurs as a result of advice from the Radiological Protection Adviser (RPA) who determines received doses on a monthly or three monthly basis during the year When received doses are approaching the maximum allowed, restrictions must be placed upon the employee to ensure doses are kept within the regulations

All advice given in the ACOP is directed towards mine environments where ventilation exists No specific advice is give for the other environments especially in relation to underground tourist mines and caverns

10 2 5 Controlled and Supervised areas (Regulation 8)

The ACOP explains how controlled and supervised areas have to be designated in mine workings but no guidance is given for other environments If the potential alpha energy concentration, shown by measurements taken over any eight hour continuous period, exceeds 0.1 WL then controlled areas status applies Above 0 03 WL supervised areas status applies In some cases the whole workplace is a controlled area, but in others only some parts are controlled.

The boundaries of controlled areas have to be marked so that persons are aware that they are entering them. Only classified persons who are under medical supervision should enter controlled areas unless written permission is given by the employer The employer must designate, as classified persons, the employees who are likely to receive a dose exceeding three tenths of the dose limit, and must inform those employees that they are so designated Classified persons must be over 18 years old, and certified medically fit

Supervised areas must be designated where persons are likely to be exposed to doses of one third of those likely to occur in a classified area, i.e one tenth of the maximum

dose limit This is also the level of exposure (0 03 WL = 0 4 WLM/y) at which the employer must notify the HSE that work with ionising radiations has commenced.

10 2 6 Radiation Protection Advisers (RPA) and Radiation Protection Supervisors (RPS)

RPA's must be appointed by the employer who must also notify the HSE of their appointment An acknowledgment of this appointment must be made by the HSE The appointee must be suitably qualified and must be available for comment when required They must be knowledgeable in the principles of protection from ionising radiations and be aware of the legal requirements, and they must be properly informed and equipped by the employer (Regulation 10)

RPS's must be appointed, in writing, if the area is supervised Their names must be included, by the employer, in written Local Rules that have been prepared to ensure compliance with the IRR's They should be able to exert direct personal supervision on work places and they must understand the IRR's, Local Rules and principles of radiation protection (Regulation 11)

10 2 7 Dosimetry

Received radiation doses have to be determined for classified workers entering classified and supervised areas It is recommended that received doses be determined by personal dosimetry instead of area monitoring (Regulation 13)

If area monitoring is being used to determine received doses, the employer and the RPA have to determine the frequency and sites of monitoring within the work place The measurements should enable a realistic estimate to be made for each individual worker To achieve this it will be necessary to draw on existing knowledge of concentrations, the variations in these and the likely movements of the persons involved

10 2 8 Area Monitoring

The purpose of area monitoring is to determine the extent of a controlled or supervised area, to demonstrate the effectiveness of control measures taken to reduce concentrations and to a lesser extent to determine estimates of radiation doses (Regulation 24)

Regulation 24 stipulates the regularity of measurements in different radiation environments Between 0 03 and 0 05 WL, measurements need to be taken at 3 monthly intervals, or if consistent averages are established at 6 monthly intervals Between 0 05 and 0 15 WL, concentrations need to be determined at monthly intervals or if consistent averages are recorded at 3 monthly intervals Above 0 15 WL concentrations need to be determined at monthly intervals

10 2 9 Summary of IRR's

The IRR's provide guidelines to which all employers have to work in order to ensure that their employees do not receive radiation doses above the legal limit However, the regulations were developed for use in mine environments where forced ventilation can easily be introduced and exposure times limited to ensure that received radiation doses are kept to a minimum.

10.3 Users of Limestone caves

Seven major group of people who enter limestone caves can be identified the general public, guides and other employees in show caves, professional instructors, volunteer instructors; instructed groups, recreational cavers and speleologists

10 3 1 The General Public

The public may enter one of the fourteen show caves and limestone mines in England and Wales as paying visitors This group spend only a limited time underground, the average show cave trip being 30 minutes and trips in individual caverns 15 - 60

minutes in duration. Consequently, the radiation dose received is likely to be very small and significantly below the average yearly background radiation dose The general public are probably not covered by the IRR's as they are not at 'work' If the regulations did apply then they could enter controlled and supervised areas through a written system of work (local rules) and their annual received dose would be below the average yearly background radiation dose

10 3 2 Guides or other employees working in show caves

Guides are employees operating within a show cave, usually undertaking guided tours Other employees could include shop workers and maintenance persons The shop workers would be at little risk from exposure to radon, unless air from the cavern is drawn into the shop. Guides and maintenance workers may spend long periods of time underground and this could potentially result in exposure to high radiation doses Due to the nature of the work, the longest periods of time spent underground are during the summer months when concentrations are usually highest

Guides working within the Show Cave are clearly covered by the IRR's, as the exposure to radon is as a direct consequence of their occupation However, the nature of employment within Show Caves means that the majority of staff are seasonal and short term workers, such as students, and therefore, the length of employment is short The majority of workers are only employed during one season, while a minority are employed for three or more Consequently, their total five year doses will usually be considerably below the 75 mSv limit outlined in the IRR's (regulation 7) and the risk to health is probably quite small. However, a small minority of workers are employed on permanent contracts and may be exposed to high radiation doses for long periods of time In these cases the five-year total dose could approach or exceed 75 mSv with a consequent risk to health The majority of show cave operators are aware of the potential health threat associated with radon and steps are being taken to ensure that atmospheric concentrations are lowered and that exposure rates are kept to a minimum. As the majority of Show Cave guides are temporary / seasonal staff and are rarely employed for more than three years their lifetime radiation dose as a result of

exposure to radon is likely to be very much less than workers in other high radon environments (e g Metalliferious mines) who remain in the same job for over thirty years During their 2 - 3 years employment, guides could potentially receive a fairly high radiation dose which would be followed, in virtually every case by a longer period of exposure to low radon concentrations However, those guides who are employed on a more permanent basis could potentially receive very high life time doses

In conclusion, it is clear that, in order to operate within the IRR's, all Show Caves and Mines need to develop a written system of work, appoint RPA's and RPS's and undertake regular monitoring, either on a monthly or three monthly timescale Estimated radiation doses need to be calculated for all employees

10 3 3 Professional caving instructors

This group includes persons employed as professional instructors and based at outdoor education centres, teachers and self-employed instructors, all of whom take groups of people underground in "wild" caves as part of their employment Potentially, instructors could accrue high radiation doses during a single year Instructors are also under a moral and legal obligation to inform all persons in their care of the risks from radon in the same way that they inform people about the other risks faced while underground Although instructors are clearly covered by the IRR's as they receive a radiation dose as a consequence of their occupation, very few dose estimates have been calculated for instructors A few employers (primarily within the Army) are actively monitoring their instructors but this has only been ongoing since the start of 1993 One other group of instructors, from the White Hall Center, Derbyshire, are known to have been monitored In both cases positive action was taken to limit exposure times and to target caves which were known to have low radon and radon daughter concentrations. As a consequence, received doses were below 5 mSv To comply with the IRR's, individual instructors or institutions would need to develop a written system of work to allow instructors to enter controlled and supervised areas However, with the data available, at present, it would not be possible to delimit all controlled and supervised areas with any degree of accuracy Hence, it is felt to be

desirable that monitoring of received doses be undertaken to ensure that received doses are below limits set within the IRR's

Due to the nature of instructed caving, the guides are likely to spend variable amounts of time in a variety of caves Hence, it is difficult to estimate their personal radiation dose Two methods are available for determining radiation doses Firstly, individual instructors can be monitored through personal dosimetry This allows an estimate of the overall radiation dose to be made but the relative composition of the source of the individual components cannot be determined Therefore, effective action to decrease received dose cannot easily be taken. Also, it is difficult to ensure that dosemeters are worn correctly, potentially reducing the validity of the results gained. Secondly, potential radiation doses may be estimated by a combination of area monitoring and recording of exposure time To ensure that the results obtained by this method are as reliable as possible, more measurements would need to be obtained from caves which are popular with instructed groups This would have the added advantage of increasing the size of the data set available to the caving population as a whole If this information is passed onto recreational cavers, it would allow other parties to reduce their received doses through positive action by avoiding caves which are known to have high radon levels It is felt that a combination of both personal dosimetry and area monitoring should be undertaken Area monitoring should firstly be used to establish the likely received dose If this is above a predetermined threshold (5 mSv for example) then personal dosimetry should be undertaken However, if the likely dose is below 5 mSv then no further action is necessary, although initially personal dosimetry may be useful to confirm the absence of risk. In theory all persons entering cave environments as part of their contract of employment should be able to alter their caving practices to reduce their received dose to below 5 mSv per year

10 3 4 Volunteer leaders

Volunteer leaders are people such as scout masters who take groups underground in "wild" caves but who do not receive payment for doing so The situation with respect to the IRR's is unclear as officially they are not at 'work' as they are not being paid for

their duties However, they are also not actmg as 'recreatıonal cavers' as they have an official capacity and responsibility durmg the time that they are underground Potentially their received doses are the same as for the instructors outlined above

10 3 5 Instructed groups

Instructed groups consist of novices who are taken underground by more experienced leaders on adventure caving trips If the mstructor is a paid employee then the IRR's could be held to apply to the instructed group who would be m a similar position to Show Cave guides In both cases it is unlikely that a significant radiation dose would be received unless many visits to high radiation caves were undertaken

10 3 6 Recreational cavers

The majority of cavers m England and Wales visit "wild" caves for pleasure and sport Potentially, this group could receive very high radiation doses m any one year, dependmg upon the caves visited and the time spent underground The average recreational caver spends approximately 100 hours underground durmg any one year, but more dedicated individuals spend up to 900 hours underground and may accrue a far larger radiation doses

Gunn *et al* (1989) expressed an mitial view that recreational cavers are probably not covered by the IRR's as they are undertakmg a recreational activity and are not at work However, if a 'goodwill fee' is paid to the farmer / land owner for access to the cave, for parkmg, or in compensation for trespassing then the IRR's may apply In this instance, strict compliance with the IRR's would be virtually impossible and there is a danger that many land owners would close their caves to the public rather than take the risk This would provoke a very strong reaction from the cavmg community

An additional complexity relates to access control The control of access to an underground cavity is normally regarded as beng with the person controllmg access across the surface from a public right of way to an entrance mto the cavern However, this may not be the sole determining factor Ownership of an underground cavity

lies with the owner of the land above it unless this has been legally transferred, as for example in the case of mining rights Therefore, three categories of people could be deemed to have the right to control access firstly, the persons who own the land from the public right of way to the entrance of the cavity, secondly, the person in control of the entrance to an undetermined distance into the cavity, and thirdly, the persons whose land lies above the cavity Given that these persons or groups hold the rights to control access to only part of an underground cavity, then it would seem that they can only lawfully control access to that limited part and thus only enforce legal requirements within that part This gives rise to the potential of a caver being subject to the IRR's in one part of an underground cavity but not another because the access rights are held by two different persons

In view of these problems, it is felt that recreational cavers be clearly identified as being outside the scope of the IRR's If recreational cavers were deemed to be covered by the IRR's where 'goodwill fees' have been paid there is no practicable way of reducing their exposure short of preventing access to the caves Radon has been removed from Show Caves by the installation of ventilation fans However, this could not be implemented in 'wild caves' due to the much greater length of these systems and consequent high cost which landowners would not bear In addition, the installation of ventilation within 'wild caves' would alter many of the unique facets of the underground environment and might contravene the legislation that has designated a large proportion of caves as Sites of Special Scientific Interest

Although recreational cavers must logically fall outside of the scope of the IRR's, it is necessary to consider the potential health risks to recreational cavers More data need to be collected so that those sites that contain high radon concentrations may be identified and any seasonal variations quantified In addition it is necessary to increase awareness of the risks within the caving community

10 3 7 Speleologists

Speleologists, are scientists who are undertaking research on some aspect of the cave environment Due to the nature of their work, involving the collection of samples

over extended periods of time, the exposure rates may be exceptionally high For example, in one case it has been calculated that an individual received a radiation dose approaching 50 mSv during 12 days' field work. However, the majority of speleologists would spend considerably less time underground than cave guides or workers in mines, so their life time dose may not be excessive This group clearly falls within the scope of the IRR's but, as noted above, there is no logical means whereby the caves they enter could be designated as supervised or controlled areas Similarly, their radon concentrations cannot be reduced by ventilation Instead it is suggested that a combined approach be adopted. In the first instance there is a need for more data so that informed decisions can be made regarding risk. Secondly, estimates of received radiation dose should also be made through personal dosimetry to ensure that legal dose limits are complied with

10 3 8 Summary

The users of caves have been divided into 7 primary groups This division may appear to be somewhat artificial, but it is important, both in terms of the health implications (by identifying groups at risk) and for the enforcement of the Ionising Radiation Regulations (1985)(determining whether parties should be operating under the IRR's) Of the seven major users of limestone caves it can be seen that potentially five groups should be operating to the IRR's and that a potential health risk exist for these groups

10.4 The designation of supervised and controlled areas in limestone caves

All of the caves surveyed during this project have some sites which exceed the thresholds for the designation of supervised and classified areas for at least part of the year, although sites close to the entrance may fall below these thresholds due to the influence and dilution of relatively radon free external air Based upon figures in chapter six the highest mean concentration recorded (8,528 Bq m^{-3}) in a Peak District cave is approximately 24 times the limit for a controlled area and the lowest mean (454 Bq m^{-3} from a cave the Portland region) is still above the controlled area limit The

maximum (46,080 Bq m^{-3}) from a cave in the Peak District is approaching 128 times the limit for a controlled area

It has been suggested that persons entering a cave who are not undertaking any form of 'work' should be considered to be outside the scope of the IRR's However, if persons enter limestone caves as part of their work, then their employers could potentially be in breach of the Ionising Radiation Regulations (1985) in relation to controlled and supervised areas, even though they may not have the authority to designate such areas if they do not own the cave or access to it Moreover, the case of limestone caves in respect to the IRR's is potentially unique two parties could be in the same cave at the same time, one being an instructed group operating within the IRR's and the other consisting of individual recreational cavers not covered under the IRR's Two sets of rules would need to be applied to the same area at the same time By developing local rules for instructed groups this could feasibly occur However, to ensure that the IRR's are effective, dose estimates would need to be made for instructors These can then be monitored to ensure that the received radiation doses are below the legal limit for each year

In view of these problems it is proposed that all caves be considered to be controlled areas unless it can be clearly demonstrated that radon concentrations never exceed 0 1 WL This would obviate the need to delimit 'controlled areas' as could perhaps be done in more regular workplaces Any persons entering a cave as part of their work should operate under a written system of work and that 'local rules' should be developed for cavers as a whole

10 4 1 Potential dose estimates for cavers in England and Wales

Method of Calculation

Dose rates have been estimated based upon area monitoring using radon concentrations and exposure times The observed concentrations were derived from weekly integrated sampling during the different seasons of 1991 - 1992 from a limited number of caves, outlined in chapter six. Exposure times have been derived from

questionnaires sent to individual cavers from each group The selection of interviewees was not conducted scientifically and as a consequence the exposure times may not be truly representative of the caving population as a whole It has to be remembered that although several authors have reported errors of up to ± 50% in dose rates derived from area monitoring, the errors associated with dosimetry are up to ± 25% (Wilkening and Watkins, 1976, Yarborough, 1981) Despite these limitations, it is felt that the results presented will provide an initial estimate of the potential doses that could be received, thus allowing groups at risk to be identified

Table 10 1 outlines the range of times spent underground by the 7 major users of limestone caves Potentially the longest periods of time are spent underground by recreational cavers (maximum of 900 hours) However, it is rare that the maximum is achieved and an average of 100 hours is usual On average, guides within Show Caves spend the most time with 200 hours Visitors to show caves spend the least, usually between 15 and 60 minutes

Table 10 2 outlines the potential annual radiation doses for each group, in milli-Sieverts (mSv) (see chapter two) Minimum doses have been calculated using the average radon concentrations and minimum exposure times, maximum doses are based upon maximum exposure times and average radon concentrations and mean doses are based upon average exposure times and mean radon concentrations Minimum doses are all below 0 06 mSv and are a consequence of short exposure times It is felt that these doses will only be received by visitors to Show Caves and participants on one-off caving trips All other users of limestone caves will receive higher doses Potential maximum yearly doses may exceed 50 mSv although the number of cavers sufficiently active to receive these doses is probably very small Nevertheless, it is interesting to note that the maximum calculated dose for speleologists (57 9 mSv) is very similar to the dose recorded by personal dosimetry for the author during 1992 Average yearly doses range from 0 03 to 12 8 mSv In this case the highest doses are received by professional instructors Three groups could potentially receive doses below the natural background radiation level (volunteer leaders, members of instructed groups, and visitors to Show Caves) Table 10 3 outlines the potential radiation doses (mSv)

for each group by region, based upon the average exposure period and average regional radon concentration Doses range from 0 1 to 39 7 mSv in the Peak District, 0 03 to 12 1 mSv in South Wales and 0 01 to 5 2 mSv in the North Pennines In all regions a number of groups that are at relatively little risk, can be identified visitors to Show Caves, volunteer instructors and members of instructed groups Conversely, Show Cave guides, professional instructors, recreational cavers and speleologists can be identified as groups at risk The estimated radiation doses may not be truly representative of the actual received doses, and are likely to be overestimates Through the project design all cave types were included in the survey Therefore, estimated radiation doses will have been calculated for Show Cave staff using data from wild caves, while estimated radiation doses for instructors will have been calculated based upon data from all caves and not those that are known to be preferentially used by recreational groups Average concentrations were used to calculate estimated radiation doses to ensure that radon concentrations were representative of the area To allow more accurate estimations of received radiation dose, more detailed, targeted, measurements would need to be undertaken in caves that are used by each group

Seasonal variations in estimated radiation doses have been calculated using average exposure periods and mean radon concentrations for individual sampling periods (table 10 4) Estimated radiation doses during the summer range from 0 01 to 41 2 mSv while winter doses range from 0 02 to 8 5 mSv This implies that caving during the winter months is likely to result in a lower received radiation dose Spring and Autumn months are lower than the summer months (with doses ranging from 0 04 to 14 4 and 0 03 to 10 8 in Spring and Autumn respectively)

An initial investigation of recreational caving habits indicated that the average caving trip lasts for around four hours Estimated radiation doses have been calculated for individual regions using average radon concentrations and four hour exposure periods (table 10 5) Radiation doses for individual trips range from 0 04 to 4 29 mSv depending on whether the mean or maximum concentrations are used If the mean radon concentration is used then the estimated received doses, for all regions, do not exceed the natural annual background radiation dose of 0 79 mSv However, if the

maximum concentration is used then in three regions the calculated radiation dose is in excess of the average annual background radiation dose Using the means from the Peak District and North Penniens, the 15 mSv dose limit set by the Ionismg Radiation Regulations (1985) would be reached in approximately 76 (14) and 600 (24) hours of cavmg respectively (figures in brackets are based upon maximum concentrations) These calculated dose estimates are probably overestimates but can be used as an indicator of the length of time people should plan to spend underground each year The range of times needed to be spent underground to reach the 15 mSv limit would be between 14 and 1500 hours, but the average would be approximately 1000 hours

10.5 Summary

In this chapter the operations of the IRR's have been outlined and the primary users of limestone caves in England and Wales identified Of the seven groups, five should operate within the IRR's In certain cases a sixth group (recreational cavers) could be in a situation whereby they should comply with the IRR's However, it has been argued that positive action should be taken that would allow this group to operate outsides the IRR's

As the link between radon and various cancers is now well established (Henshaw *et al*, 1990), it can be assumed that any persons exposed to high radon concentrations, as a consequence of caving activities, could potentially be at risk Estimated radiation doses were calculated for each group and from these it was concluded that three groups were at relatively minor risk (General public, volunteer instructors and members of one-off instructed cavmg trips) However, four groups were identified as having an increased risk (Guides in show caves, Professional instructors, Recreational cavers and Speleologists) It is important to remember that an individual can be a member of two groups durmg the course of one year

It is proposed that

1 All caves should be regarded as being potentially controlled and supervised

areas for at least part of the year, until proven otherwise Written systems of works should be developed to allow entry into these areas

2 Cavers, especially those leading instructed groups, be advised to review their caving activities and to reduce or eliminate visits to caves which are known to have high radon concentrations

3 Instructed groups should be encouraged to cave during the winter months when concentrations are generally lower

4 Exposure times for groups with a high risk be reduced to a minimum.

5 The radiation doses received by employees be monitored This could be best achieved through a combination of personal dosimetry, area monitoring and the logging of exposure times in individual caves

6 Instructors to inform all persons in their care of the potential risk from radon

In addition, as recreational cavers are in the 'at risk' category then sufficient data should be collected and made available to allow individuals to decide on where they want to cave and what risks they are prepared to accept However, no action should be taken within the IRR's to interfere with their recreational caving habits It is interesting to note that some cavers have been active for in excess of 40 years (longer than the radon problem has been known about) and no ill effects have been observed The author is not aware of an increased incidence of lung cancer or other radon related problems within the 'active caving population,' but this aspect has not been investigated in detail In general, recreational cavers feel that the other risks faced while underground are more serious than radon

More data needs to be collected from all caving regions within the country to ensure that informed decisions can be made by individuals to reduce their received radiation doses The collection of data needs to be organised in a way that will ensure that all caving groups will benefit The highest priority for data collection are those caves used by instructors, instructed groups and novices Medium priority should be given to

caves which are popular with recreational cavers or those in which long and extended trips are undertaken (in excess of 6 hours) Finally, and of lowest priority are those caves that are rarely visited or those that are extreme in nature To ensure that the data are representative of actual radon concentrations, data need to be measured during at least four survey periods, which reflect the different seasons of the year

Notices informing people of the risks should be placed in all caves where very high radon concentrations (in excess of 50,000 Bq m^{-3}) have been recorded The aim would be to increase awareness and to ensure that received radiation doses are kept to a minimum by reducing the time spent underground

CHAPTER ELEVEN: CONCLUSIONS AND RECOMMENDATIONS

11.1 Conclusions

In section 1 2 the aims and objectives of the thesis were outlined. This chapter reviews the work undertaken and presents conclusions with reference to each initial aim.

Aim 1 To compare and contrast different methods for determining radon-222 and radon daughter concentrations within limestone caves From this, to develop appropriate measuring techniques

Chapter five discusses methods available to determine radon and radon daughter concentrations Three makes of integrated passive track etch detectors are shown to give comparable results when determining radon concentrations Charcoal detectors were highlighted as unreliable, due to the preferential absorption of water Spot measurements of radon-222 can be accurately undertaken using Lucas cells The Kusnetz method, the standard proposed in the United States, was demonstrated to be most accurate for the determination of radon daughter concentrations in accessible areas of Show Caves Concentrations in the further reaches of 'wild caves' were accuatly recorded using Instant Working Level Meters (IWLM) It is suggested that the Rolle method is unreliable for determination of radon daughter concentration in caves due to the elevated background counts To ensure sampling errors are kept to a minimum a rigid sampling protocol must be developed and followed The protocol followed in this thesis outlined, in chapter five, is a good working example

Aim 2 To determine the typical radon-222 concentrations within the main caving areas of England and Wales

A national investigation was undertaken during 1991 - 1992 to determine typical

concentrations using passive integrated track etch detectors It was demonstrated that concentrations in the Peak District, especially the Castleton area, were higher than other regions within England and Wales and other countries in the world Intra and inter regional variations in concentrations were highlighted and accounted for by variations in the radon budget and differences in the accumulation and dispersion mechanisms Seasonal variations in concentrations were apparent, with concentrations generally higher in the summer than the winter sampling periods Variations in external climate were demonstrated to influence concentrations, with external temperature and atmospheric pressure being the dominant controls Other controlling influences on concentrations were cave morphology (such as the number of entrances, development status, altitude and orientation of the entrances) and the total emanating surface area

Aim 3 To evaluate the controls on radon-222 concentrations within a single caving region.

Chapter eight outlined results from nine caves, used by outdoor education centers in the Peak District during 1989 - 1990 Concentrations varied within the region and selective areas were demonstrated to have elevated concentrations in comparison to others Mechanisms, primarily relating to the radon budget were proposed to account for these differences Generally external temperature is the most dominant control on concentrations However, no single temperature variable can be highlighted as the dominant controlling factor and the relative influence of maximum, mean or minimum temperature within a single cave varies during a year. Seasonal trends were highlighted which were demonstrated to be controlled by external meteorological factors, primarily temperature and pressure Previously pressure had only been demonstrated to be influential in RSU caves However, during this project both negative and positive relationships were demonstrated for changes in radon concentrations within USD caves with atmospheric pressure Mechanisms were developed to account for the relationship observed Additionally rainfall was also demonstrated to be an important control on radon concentrations in both USD and RSU caves

Mechanisms which allowed for rainfall to transport radon into the cave environment were proposed

Aim 4 To determine the controls on radon-222 and radon daughter concentrations within a single cave environment

Chapter nine outlines results from a detailed investigation conducted within Peak Cavern, in the Peak District Spatial and temporal variations in concentrations were demonstrated and external meteorological conditions were demonstrated to be important in controlling concentrations Short-lived increases in radon concentrations were related to flood events It was proposed that additional radon was released into the cave environment through the reworking of sediments Statistical models for the cave and areas within the cave were developed to allow for concentrations to be predicted It was shown that predictive models were only valid for the cave in which they were developed To allow for more general predictive models, factors representing the radon budget, location and morphology would need to included The development of the models and factor analysis allowed for the primary variables that control radon to be identified External temperature and pressure were the dominant controls, but total rainfall on both a weekly and monthly basis were also demonstrated to influence concentrations Mechanisms by which rainfall transports radon to the cave environment over these two time scales were proposed The influence of weekly rainfall represents radon transported from the soil, while rainfall on a monthly basis represents percolation water in equilibrium with the limestone Additionally, at all sites within the cavern, external wind direction influenced concentrations due to the effects of dilution by incoming radon free air

Aim 5 To quantify the extent of seasonal variations in cave radon gas concentrations

Chapter three reviews published literature on radon in caves and outlines mechanisms that have been proposed to account for seasonal variations in radon concentrations Chapters six, eight and nine all examine radon concentrations

and highlight that seasonal variations are apparent Generally summer concentrations are higher than winter, while spring and autumn tend towards summer concentrations rather than winter The primary controls on concentrations are external meteorological conditions, with temperature and pressure being the most significant factors No single temperature variable can be highlighted as the dominant control and the influence varies between regions, caves and during the seasons of the year Static pressure (at the time of the measurement) is the dominant pressure variable in some caves whereas changes in pressure during the previous day or week are the dominant controls in other caves

Aim 6 To evaluate the effects of cave morphology and geological setting on radon gas concentrations

Both variables were demonstrated during chapters six, eight and nine to influence concentrations The cave morphology, in conjunction with the external climate, influences the accumulation and dispersion mechanisms operating within a single cave to determine the concentrations recorded The geological setting affects the radon budget and controls the amount of radon released into the cave Variations in the radon budgets are apparent regionally, both within regions and within single caves

Aim 7 To identify the relative importance of different sources of radon-222 to cave environments

Chapter seven outlined the mechanisms by which radon is released from the six sources proposed for the cave environment It was demonstrated that sediments were potentially the greatest sources of radon to the cave However, due the relative surface area of limestone rock exposed, even though limestone rock releases less radon, the total generated by this source could be the major source of radon. The relative importance of each source to the overall radon budget would need to be determined for each individual cave / sampling site in order to accurately determine the radon budget The soil and water were

demonstrated to be secondary sources of radon that generally contribute small amounts to the overall radon budget Speleothems were demonstrated to make an insignificant contribution to the overall radon budget Faults and joints were proposed as sources of radon, but it was not possible to conclusively demonstrate their influence

Aim 8 To quantify the risk to cavers from radon gas

Chapter ten reviewed the users of limestone caves and proposed that seven groups could be identified, of these five could potentially be covered by the IRR's (1985) Four groups (guides in Show Caves, Professional cavers, Recreational cavers and Speleologists) were shown to potentially be at risk from radon while underground Methods were proposed by which affected groups could reduce their received dose

11.2 Recommendations

1 Standard methods should be established for monitoring radon gas and radon daughter concentrations within limestone caves Integrated determinations of radon gas should be undertaken using track etch methods, such as those supplied by Tastrak Ltd or the NRPB, spot determinations of radon gas should be made with Lucas cells Radon daughter concentrations in accessible areas of caves should be determined using the Kusnetz method while IWLM should be used in the further reaches

2 The national project, outlined in chapter six, highlighted variations in concentrations between regions Additional data should be collected from other regions which contain caves which were not included within the survey, for example Scotland and North Wales Due to the limited number of measurements recorded in the Mendip Hills and its importance as a caving region, a specific investigation should be undertaken within this region

3 The fundamentals of the cave radon system are well established and have

been widely published (see chapter three) However, as demonstrated in chapter six, variations away from the normal do occur These should be investigated further to determine other influences on the cave radon system.

4 Chapters six, eight and nine highlighted that cave radon concentrations are linked with cave microclimate In the future, all investigations should undertake joint microclimate and radon studies, allowing for a more detailed interpretation to be developed

5 Determination of equilibrium between radon and radon daughter concentrations should be undertaken The differences in equilibrium could then be used to estimate the age of the air and the relative importance of different microclimatical processes operating

6 Chapters six and seven indicated that bulk determinations of the radon budget could be undertaken in sealed sections of caves, for example those contained between two sumps This work will additionally allow for the effects of changes in atmospheric pressure on the exhalation rate to be determined

7 Detailed modeling should be undertaken using data collected continuously at a number of sites within the cave and the external microclimate to allow for further development of the cave radon system.

8 Further investigations on the radon budget within a cave need to be undertaken, specifically to determine the exhalation depth and the determination of the actual importance of each source material to the overall radon budgets

9 The importance of transport of radon from deep seated sources via faults and joints to the cave environment should be investigated Potentially the use of helium isotopes could be used to aid the interpretation of radon movement

from these source

10 Once accurate determinations of the radon budget have been made for either caves, areas or regions, more detailed modeling can be undertaken allowing radon concentrations to be more accurately modeled

11 The influence of rainfall on cave radon concentrations should be investigated further and its importance as a controlling factor examined

12 Chapter ten has highlighted that cavers are at risk from radon within caves Through the caving press they should be informed and once sufficient information is available recreational cavers will make an informed decision However, the National Caving Association must develop guidelines for professional cavers ensuring that all aspects of the IRR's (1985) are covered and that all persons who enter caves as part of their work are adequately supervised

13 The proportion of unattached to attached radon daughters should be determined This would allow for inferences on the ionic state of particles in cave air to be made

14 Detailed analysis of cave radon concentrations will allow for modeling of cave microclimate, in particular air movements, allowing for the cave environment to be further understood

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APPENDIX ONE

Table APP 1 1 Summary of radon daughter measurements (WL) from the Peak District Region.

Cave	Mean	Min	Max	No	Method Spot/ Int
Axe Hole	0 067	0 010	0 218	13	Int
Carlswark Cavern	0 396	0 015	1 41	78	Int
Devonshire Cavern	0 07	0 004	0 326	36	Int
Gautries Hole	0 904	0 253	2 94	24	Int
Giant's Hole	1 087	0 026	4 101	75	Int
Hillocks Mine	0 667	0 090	2 079	26	Int
Jug Holes	0 045	0 090	0 813	26	Int
Knotlow Caverns	1 07	0 262	1 714	18	Int
P8 Jackpot	0 345	0 027	1 38	37	Int
Oxlow Caverns	5 85	0 18	16 66	15	Spot
Poole's Cavern	0 46	0 01	0 73	200	Spot
Bagshaw Cavern	1 56	0 67	3 23	12	Spot
Peak Cavern	4 00	0 01	25 00	1040	Spot
Streaks Cave	0 5	0 21	0 89	10	Spot
Speedwell Mine	1 7	0 01	8 50	130	Spot
Blue John Caverns	0 935	0 01	5 66	252	Spot
Rutland Cavern	0 053	0 01	0 673	135	Spot
Masson Cavern	0 056	0 01	0 177	81	Spot
Treak Cliff Cavern	0 454	0 01	2 1	240	Spot

Black Rabbit Cave	0 675	0 05	1 89	10	Spot
Christmas Swallet	2 45			1	Spot
Maskhill Mine	6 38			1	Spot
Cascade Cavern	2 45			1	Spot
Lathkill Head Cavern	0 89			1	Spot
Merlin's Mine	0 76	0 02	2 56	6	Spot

Table APP 1 2 Summary of radon daughter measurements (WL) from the North Pennines Region.

Cave	Mean	Min	Max	No	Method Int / Spot
Ingleborough Cave	0 92	0 23	1 22	20	Spot
White Scar Cave	0 18	0 01	0 606	108	Spot
Manchester Hole	0 84	0 68	1 46	5	Spot
Birks Gill Cave	1 34	0 04	1 87	5	Spot
Sleets Gill Cave	0 84			1	Spot
Shatter Pot	0 45	0 02	1 56	6	Spot
November Hole	0 24	0 01	0 42	10	Spot
Diccan Pot	0 31	0 01	0 54	5	Spot
Juniper Gulf	0 68			1	Spot
Bar Pot	0 78			1	Spot
Tatham Wife Hole	0 94	0 04	1 63	4	Spot
Great Douk	0 45	0 03	0 78	5	Spot

Table APP 1 3 Summary of radon daughter measurements (WL) from the Mendip Hills Region.

Cave	Mean	Min	Max	No	Method Spot/ Int
Goatchurch Cavern	0 086			2	Spot
St Cuthbert's Swallet	0 689			2	Spot
Cuckoo Cleeves		3.25	5 13	12	Spot
Box Stone Mine	2 22			2	Spot
Rod's Pot	0 65	0 05	1 08	6	Spot
Sidcup Swallet	0.58			4	Spot
Swildon's Hole	1 45	0.03	1 52	29	Spot
Gough's Cave		0.01	1 447	351	Spot
Coxes Cave		0 01	0 34	56	Spot

Table APP 1 4 Summary of radon daughter measurements (WL) from the South Wales Region.

Cave	Mean	Min	Max	No	Method Spot / Int
Cathedral Cave	0 33	0 15	1 76	32	Spot
Dan-yr-Ogof	0 60	0 01	1 42	60	Spot
Bone Cave	0 02	0 01	0 05	6	Spot
Quarry Cave	0 03	0 01	0 04	3	Spot
Ogof Capel	2 67	1 78	3 56	12	Int
Porth-yr-Ogof	0 45	0 03	0 78	4	Spot

Table APP 1.5 Summary of radon daughter measurements (WL) from the North Wales Region.

Cave	Mean	Min	Max	No	Method Int / Spot
Cabin Cave	0.033	0.02	0.06	3	Int
Bog Cavern	0.9	0.65	1.15	2	Int
Belgrave Cavern	0.08	0.05	0.14	3	Int
Nadolig Cavern	0.91	0.73	1.116	3	Int
OHF	0.28	0.20	0.36	3	Int

Table APP 1.6 Summary of radon daughter measurements (WL) from Ireland

Cave	Mean	Min	Max	No	Method Int/ Spot
Marble Arch Cave	0.24	0.03	0.38	20	Spot
Prods Pot	0.34	0.13	0.67	5	Spot
Ailwee Cave	0.44	0.001	0.67	10	Spot
Crag Cave	0.34	0.001	0.74	5	Int

Table APP 1 7 Summary of radon daughter measurements (WL) from the South West England

Cave	Mean	Min	Max	No	Method Spot / Int
Kent's Cavern	0 158	0 01	0 383	172	Spot
Kitley Caverns	0 04	0 01	0 116	35	Spot
Reed Cavern	0 017			2	Spot
Bakers Pit	0 086			2	Spot

SPATIAL AND TEMPORAL VARIATIONS OF RADON AND RADON DAUGHTER CONCENTRATIONS WITHIN LIMESTONE CAVES

BY ROBERT QUENTIN THOMAS HYLAND

A thesis submitted to the University of Huddersfield in partial fulfillment of the requirements for the degree of Doctor of Philosophy

January 1995

Volume Two

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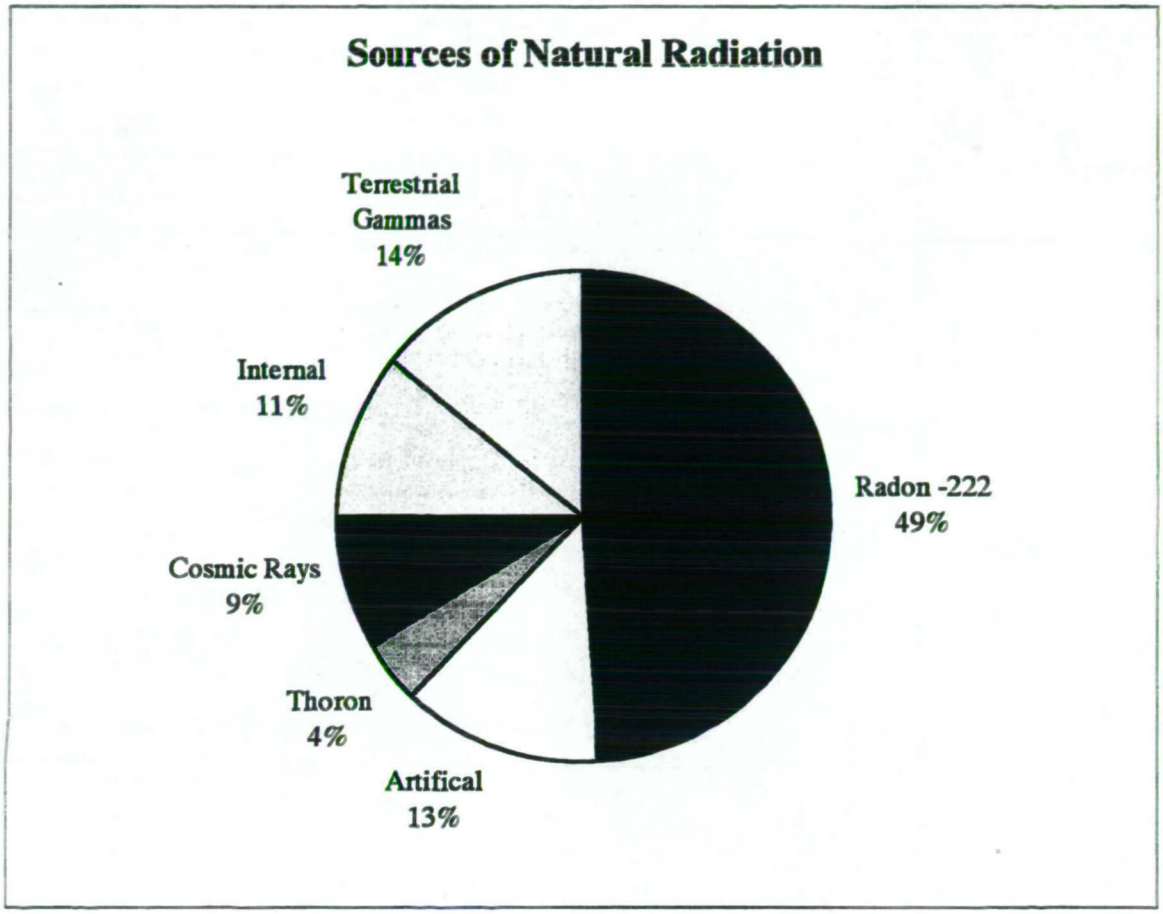

Figure 1.1 Sources of natural radiation

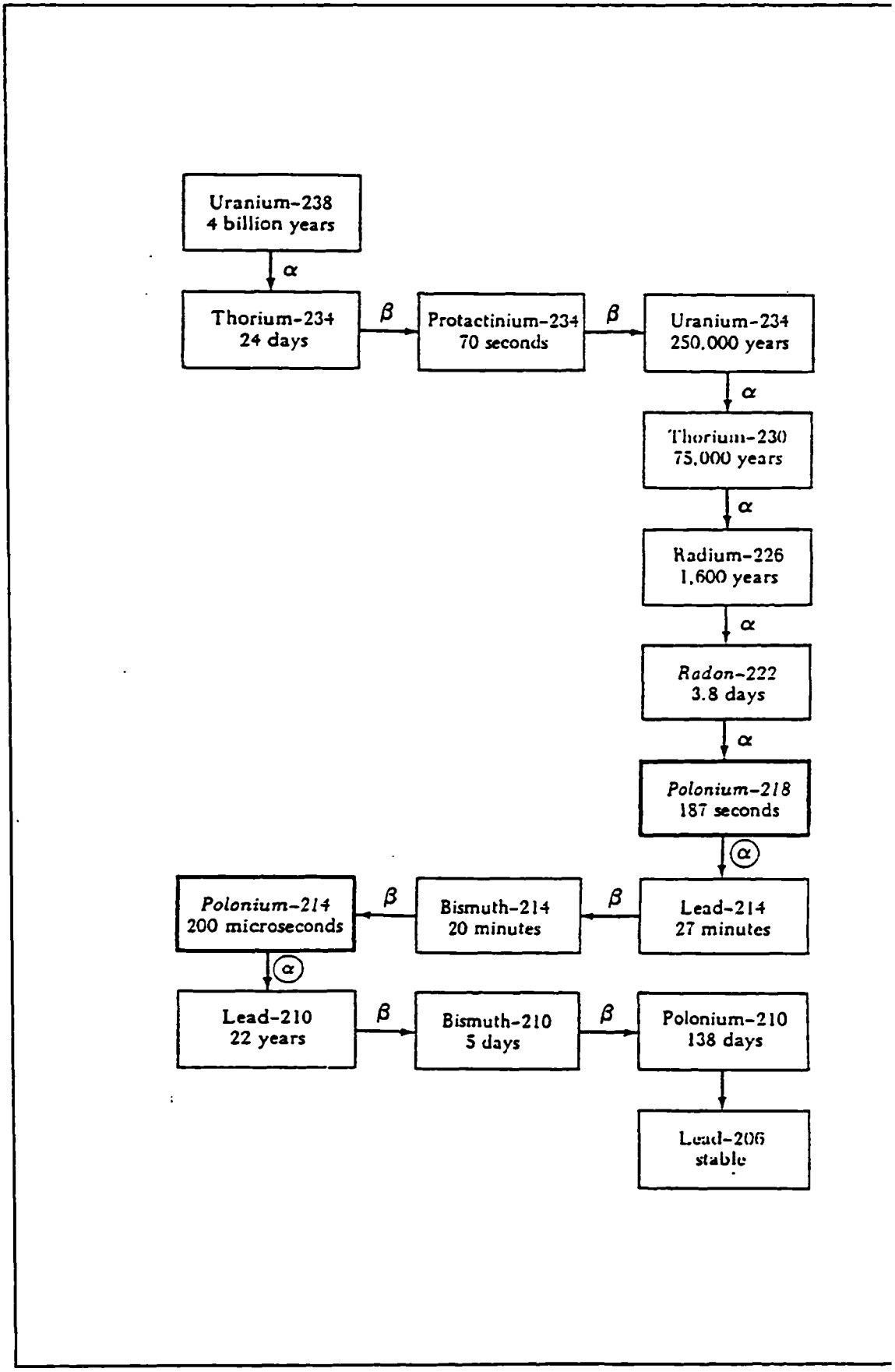

Figure 1.2 Principle decay scheme of the Uranium-238 decay series

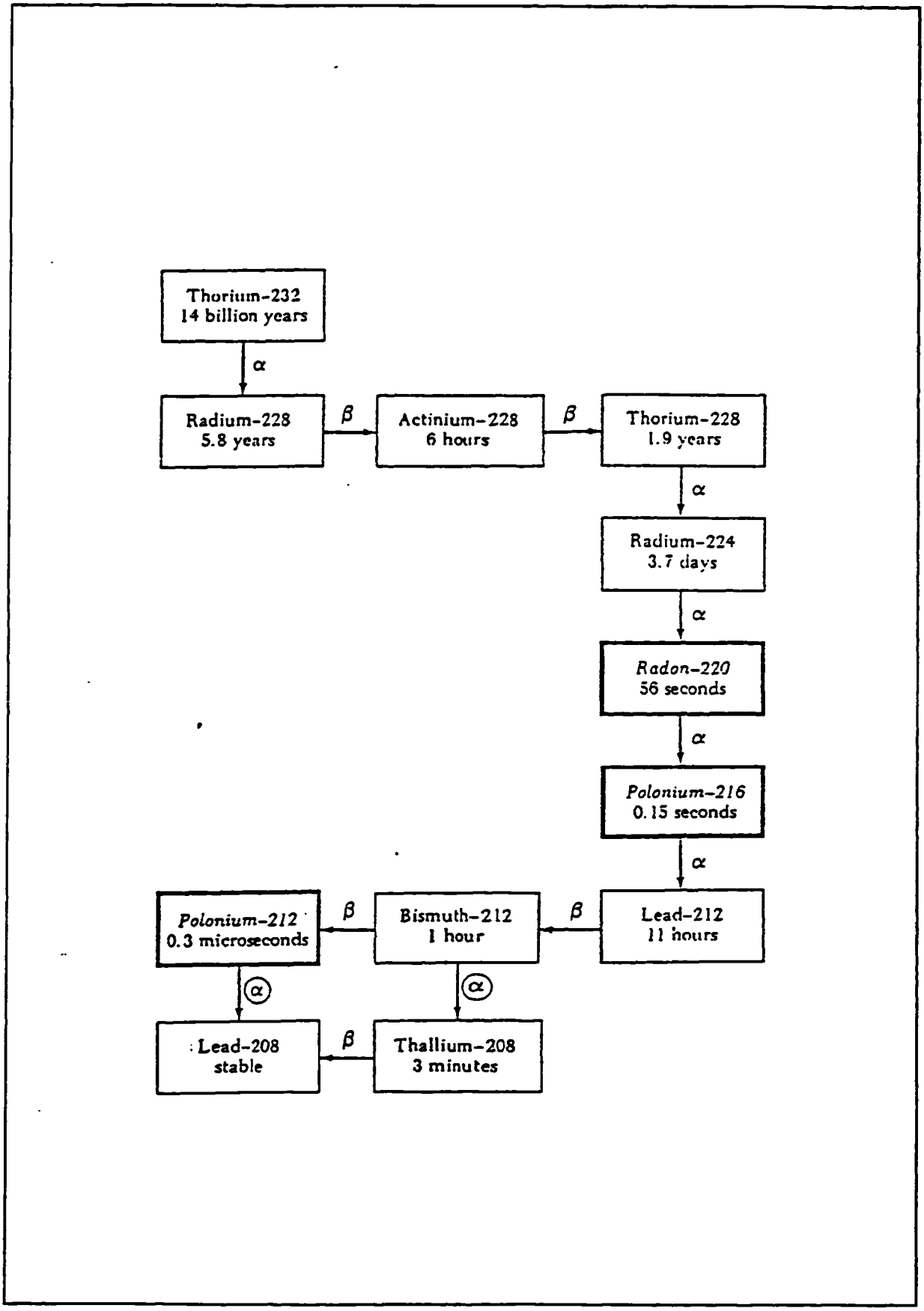

Figure 1.3 Principle decay scheme of the Thorium-232 decay series

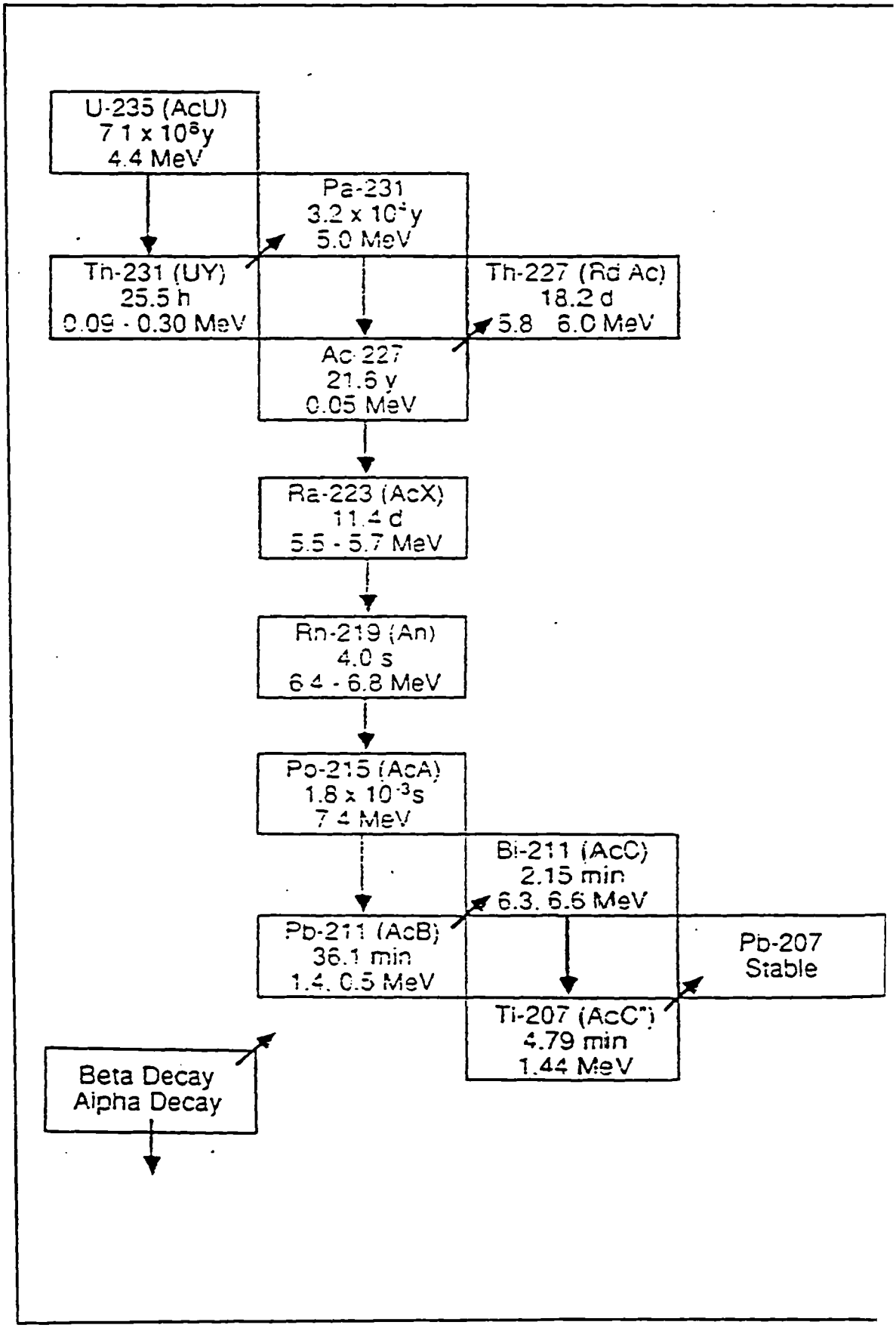

Figure 1.4 Principle decay scheme of the Uranium-235 decay series

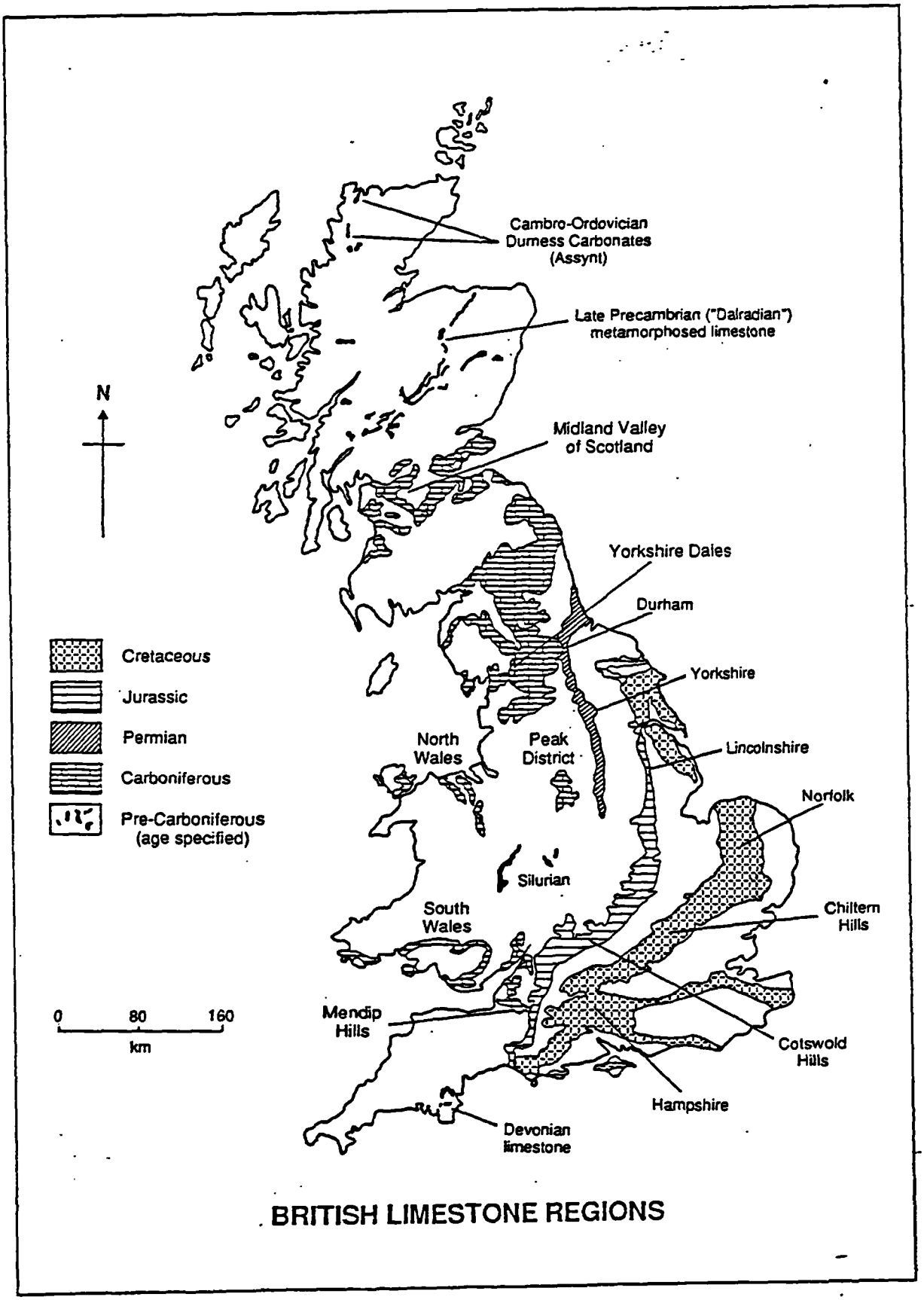

Figure 1.5 Distribution of limestone within Britain (From Gunn, 1994)

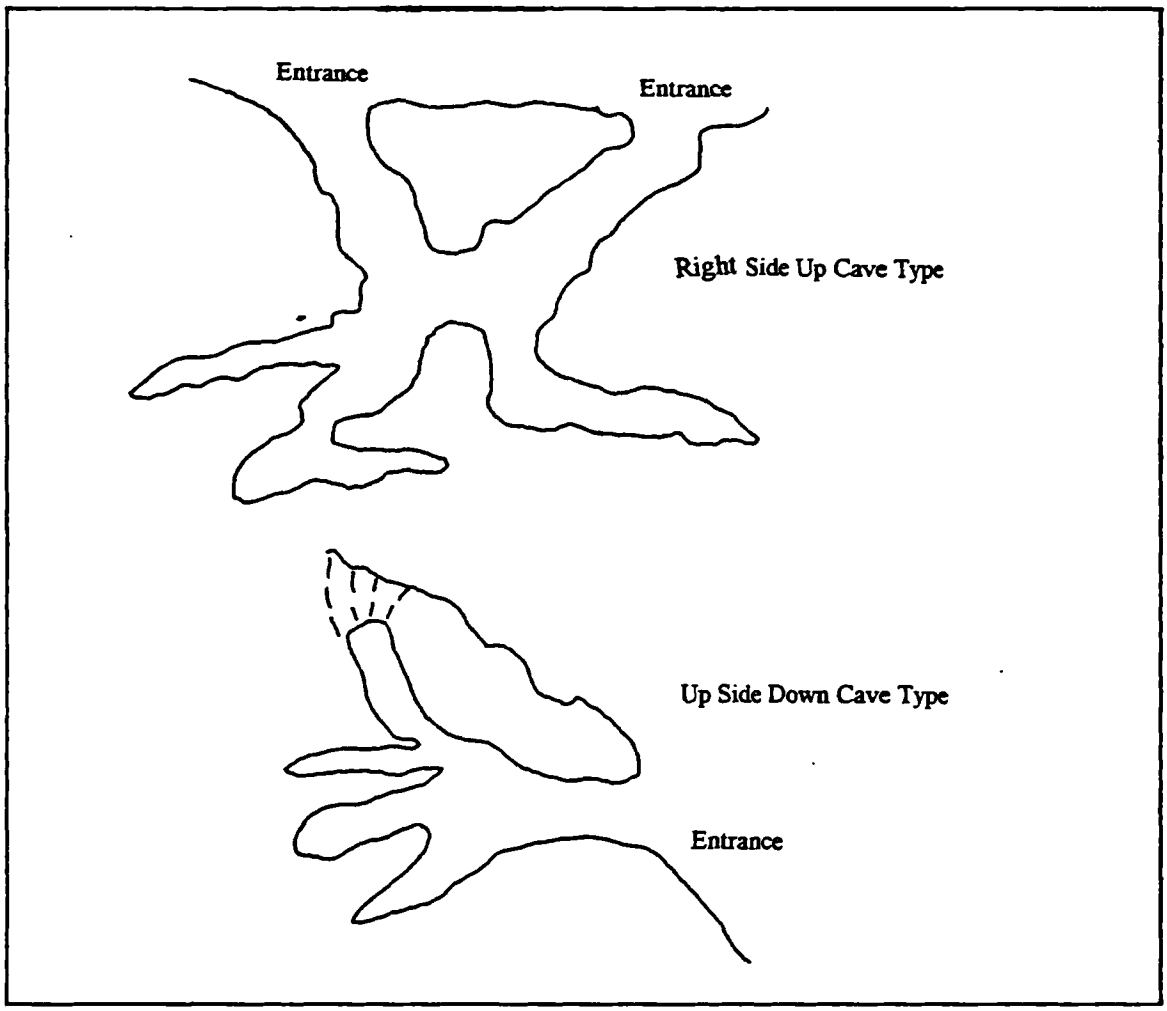

Figure 3.1 General cave configuration types, determining natural airflows. (From Yarborough, 1977).

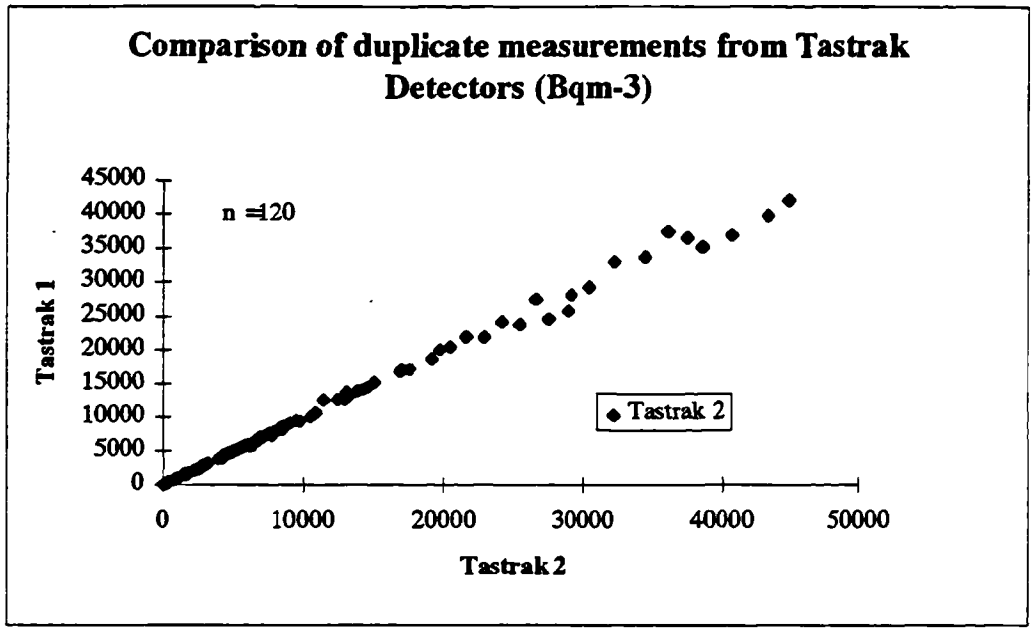

Figure 5.1 Comparison of duplicate measurements from Tastrak Detectors (Bq m^{-3})

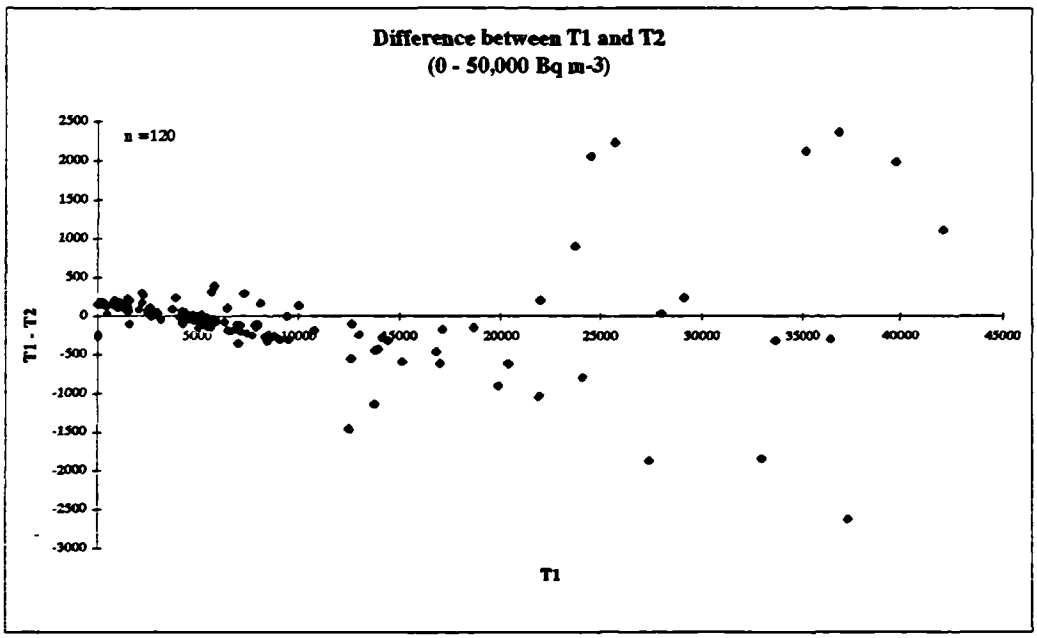

Figure 5.2 Difference between results from duplicate measurements using Tastrak detectors (Bq m^{-3})

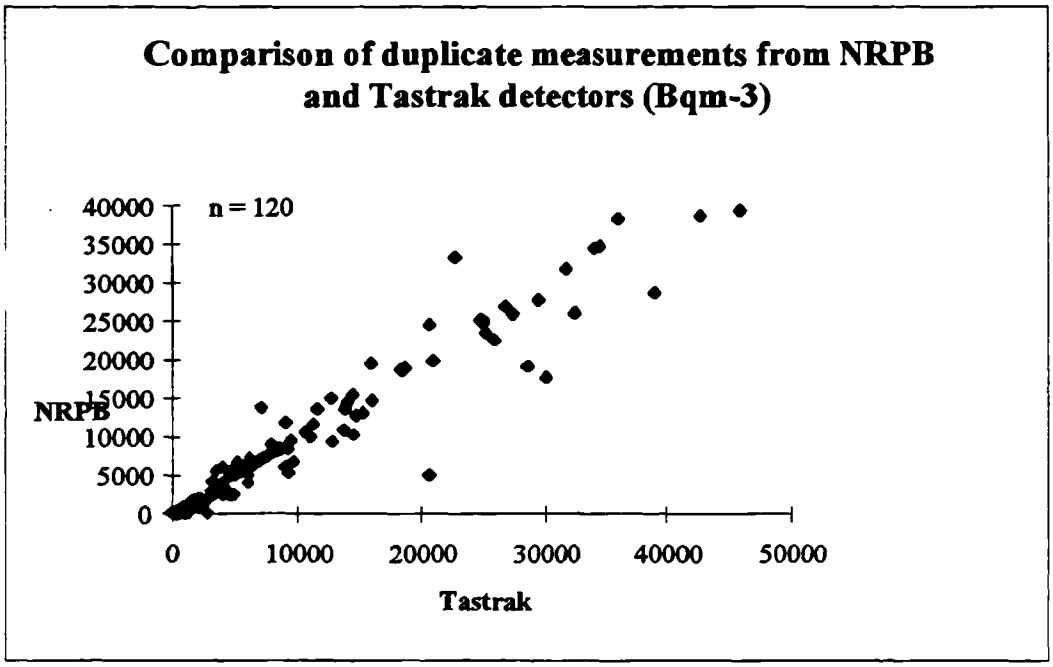

Figure 5.3 Comparison of duplicate measurements from NRPB and Tastrak detectors (Bq m^{-3})

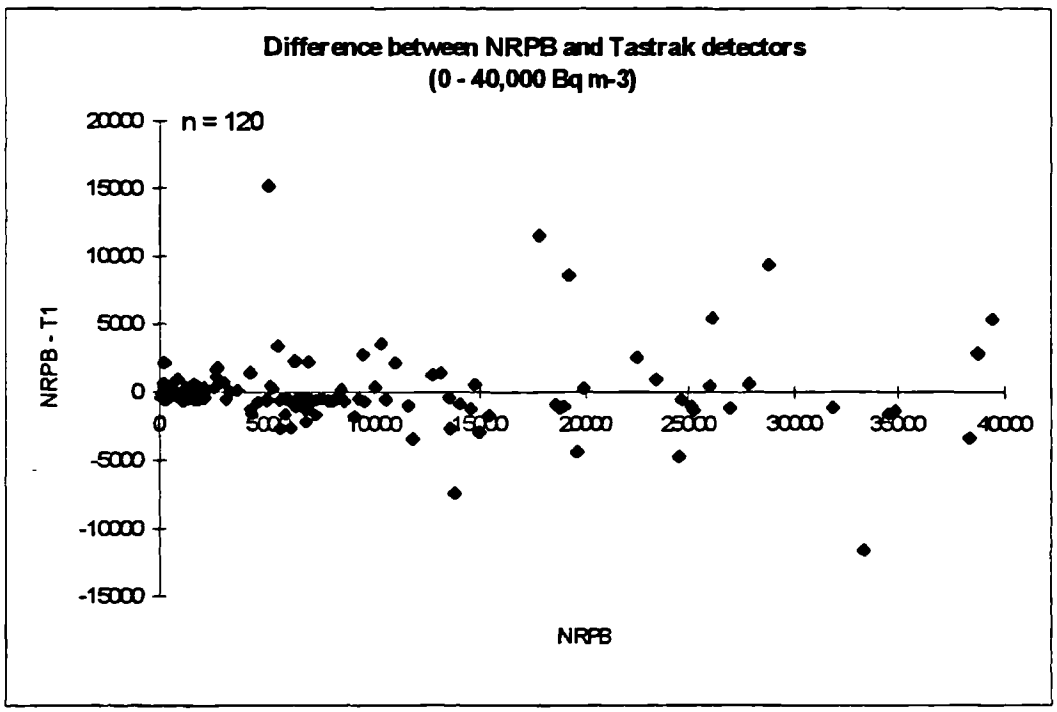

Figure 5.4 Difference between results from duplicate measurments using NRPB and Tastrak detectors (Bq m^{-3})

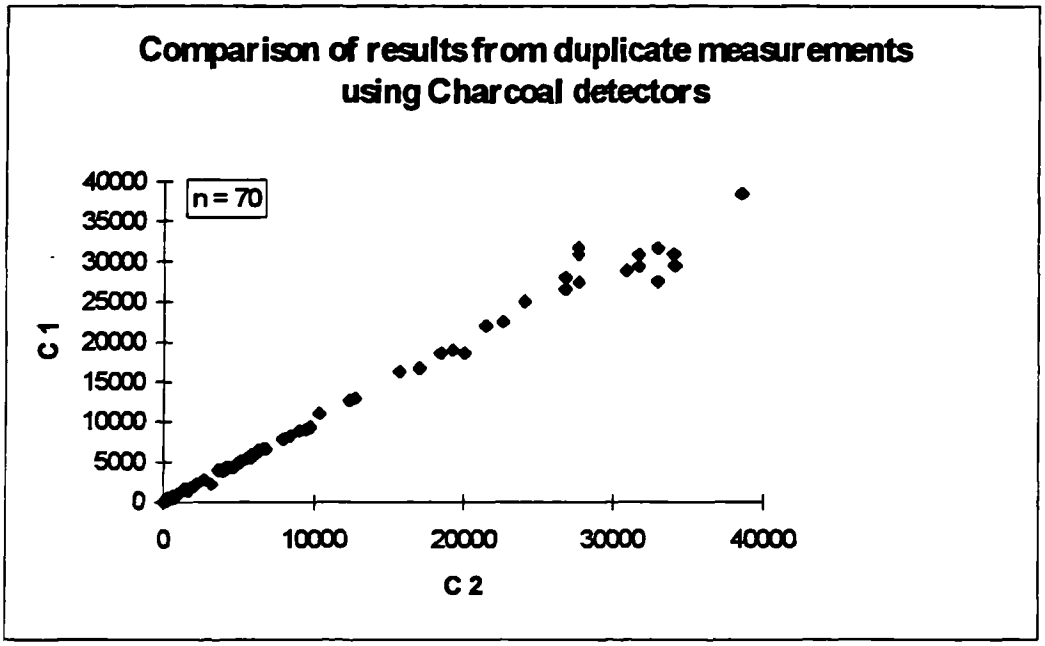

Figure 5.5 Comparison of results from duplicate measurements using Charcoal detectors (Bq m^{-3})

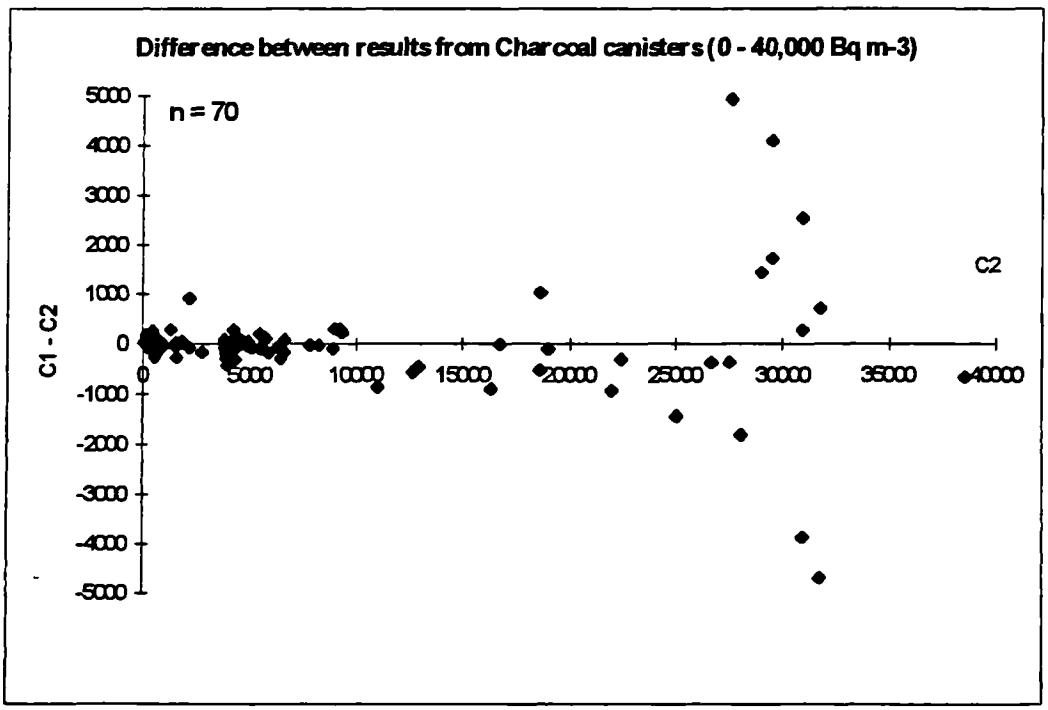

Figure 5.6 Difference between duplicate measurements using Charcoal canisters (Bq m^{-3})

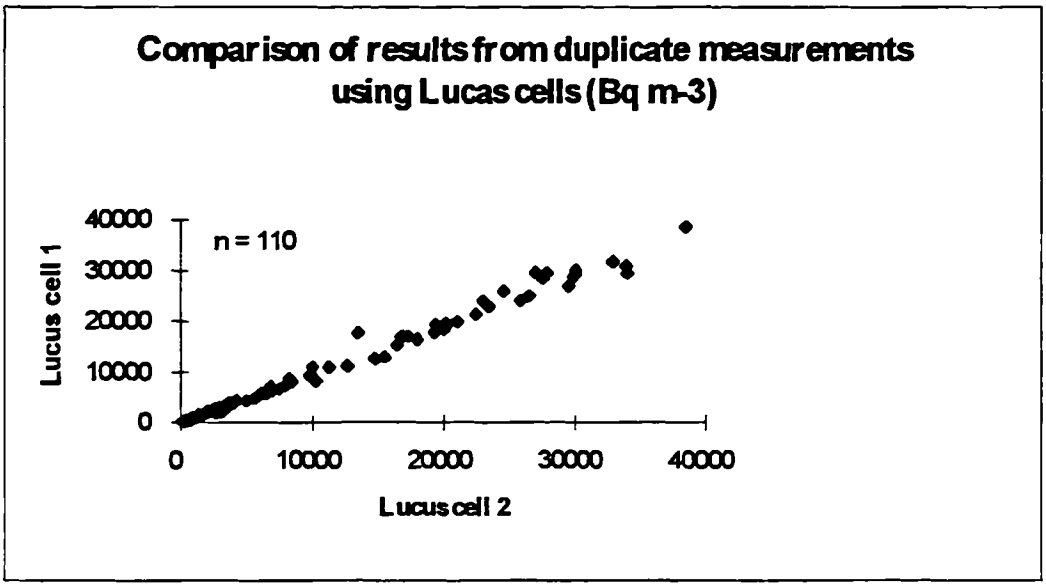

Figure 5.7 Comparison of results from duplicate measurements using Lucas Cells (Bq m^{-3})

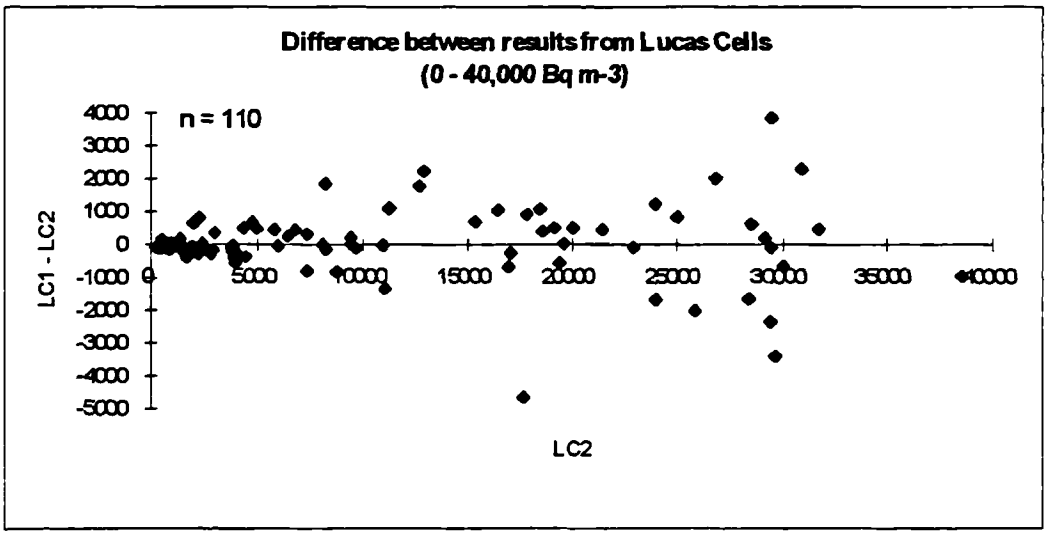

Figure 5.8 Difference between duplicate measurements using Lucas Cells (Bq m^{-3})

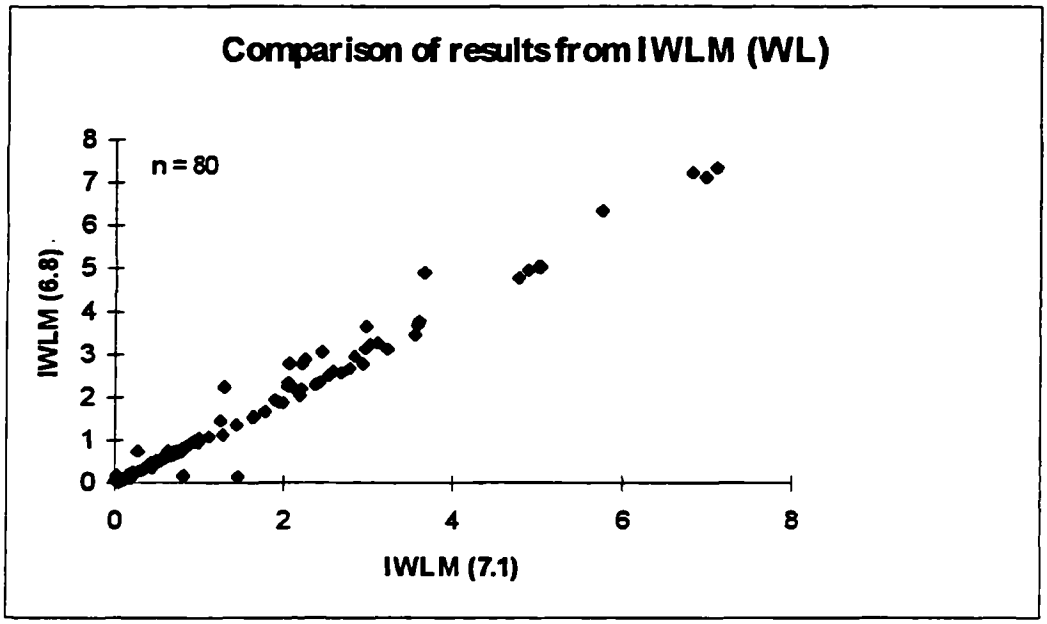

Figure 5.9 Comparison of duplicate measurements using Instant Working Level Meters (WL)

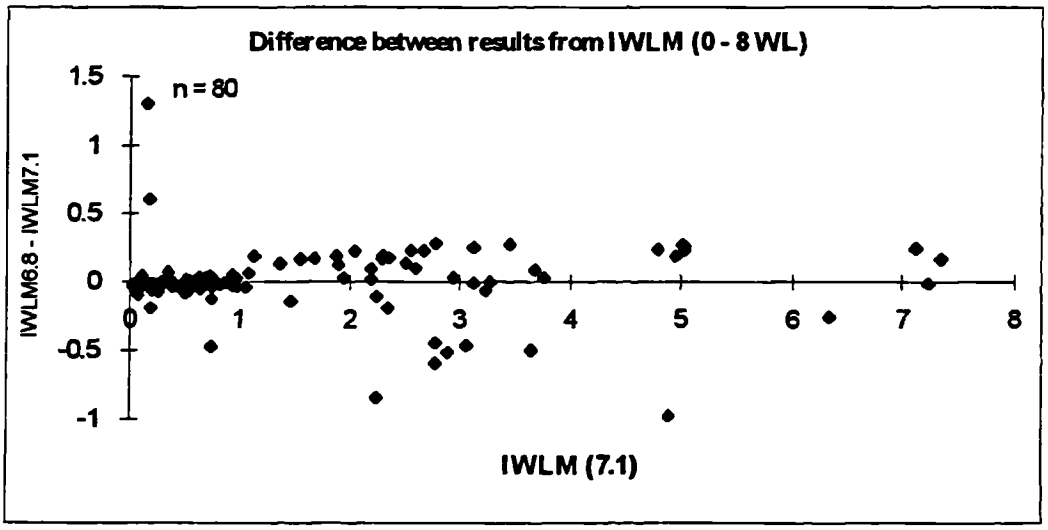

Figure 5.10 Difference between duplicate measurents using Instant Working Level Meters (WL)

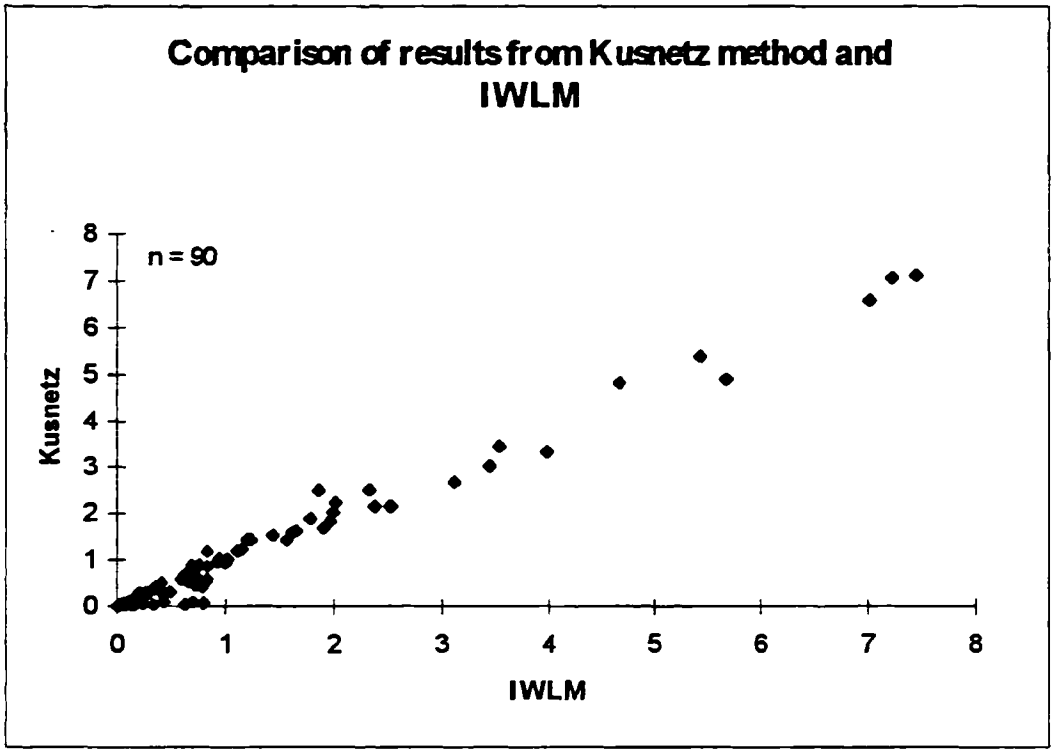

Figure 5.11 Comparison of results from the Kusnetz method and Instant Working Level Meters (WL).

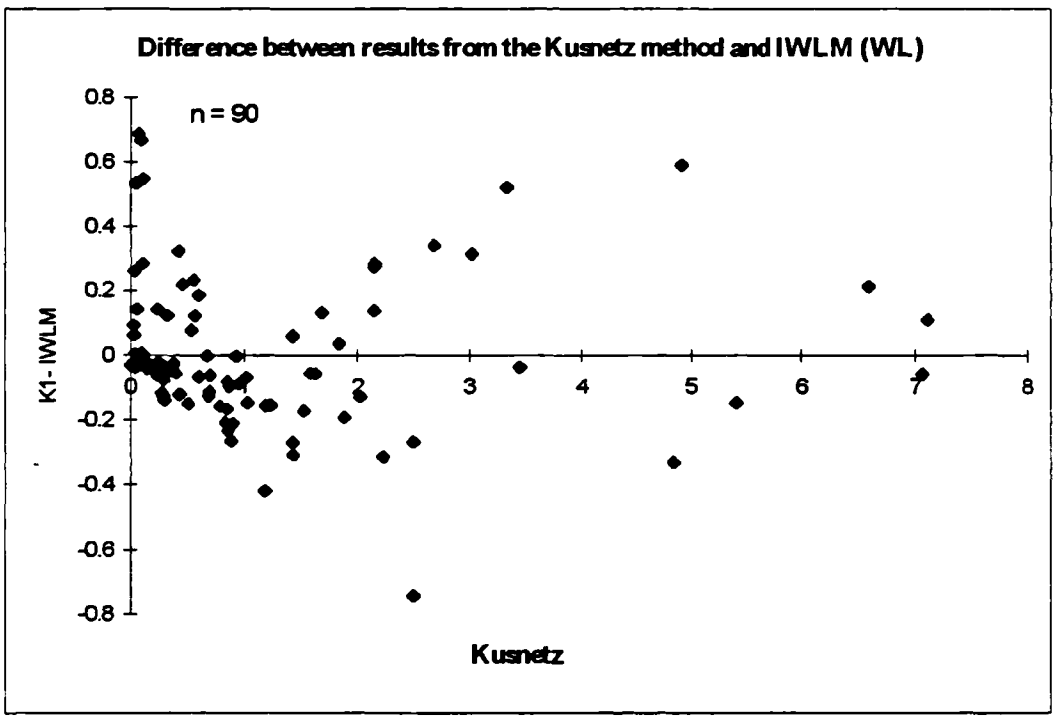

Figure 5.12 Difference between duplicate measurements from the Kusntez method and the Instant Working Level Meters (WL).

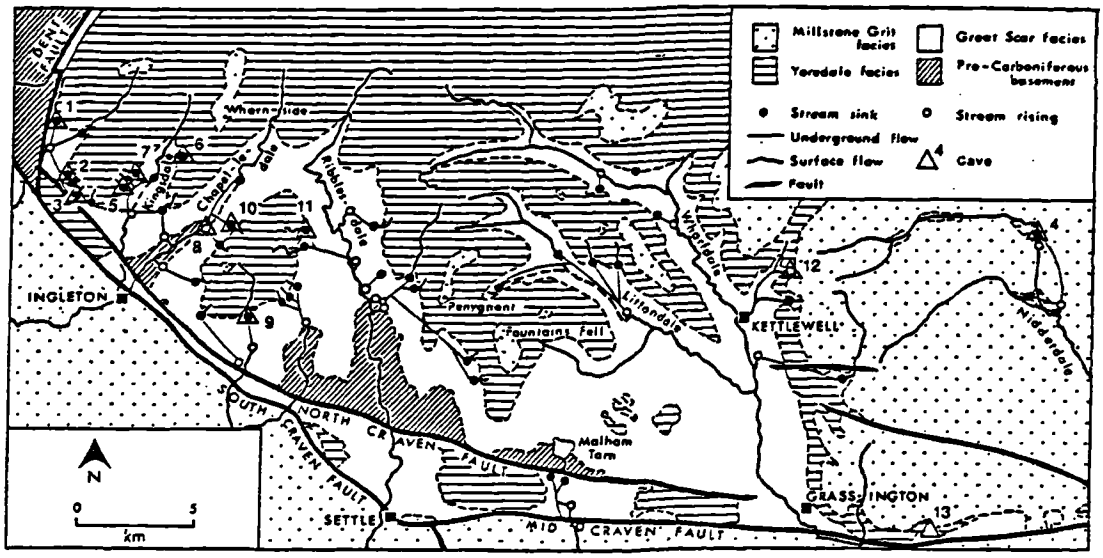

Figure 6.1 Location of caves within the North Pennines Region. 1. Ease Gill Caverns, 2. Lost John's Cavern, 3. Ireby Fell Cavern, 4. Goydon Pot, 5. Kingsdale Master Cave, 6. Heron Pot, 7. Yordas Cave, 8. White Scar Cave, 9. Gapping Gill, 10. Sunset Hole, 11. Upper Long Churn, 12. Dow Cave, 13. Stumps Cross Caverns.

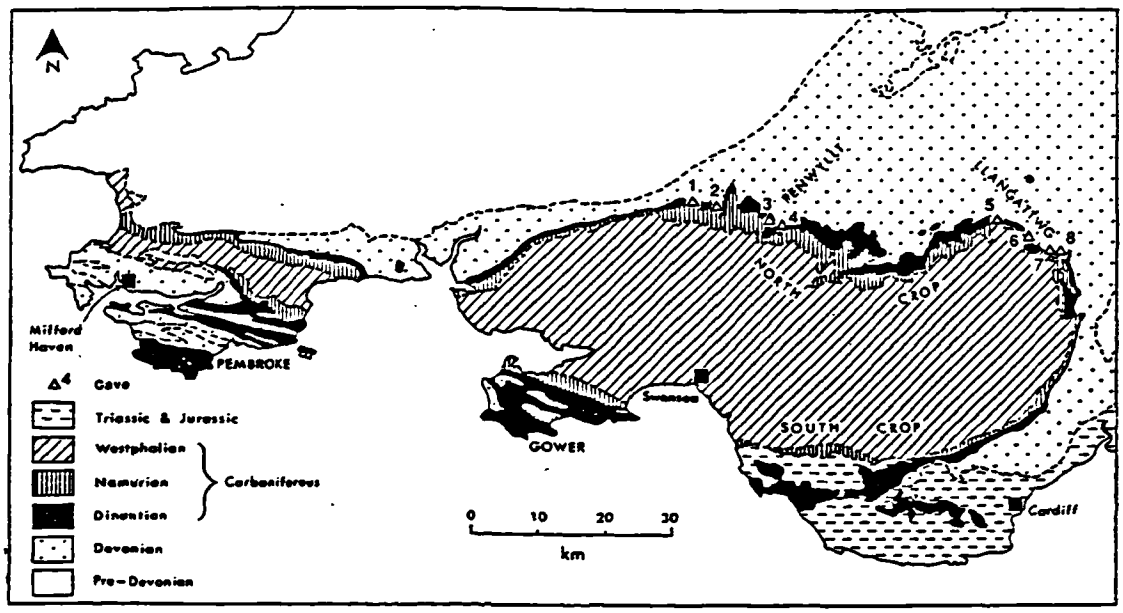

Figure 6.2 Location of caves within the South Wales Region. 1. Ogof Ffynnon Ddu, 2. Cwm Dwr, 3. Little Neath River Cave, 4. Tunnel Cave, 5. Carno Adit, 6. Ogof Agen Allwedd, 7. Ogof Darren Cilau, 8. Ogof Craig a Ffynnon.

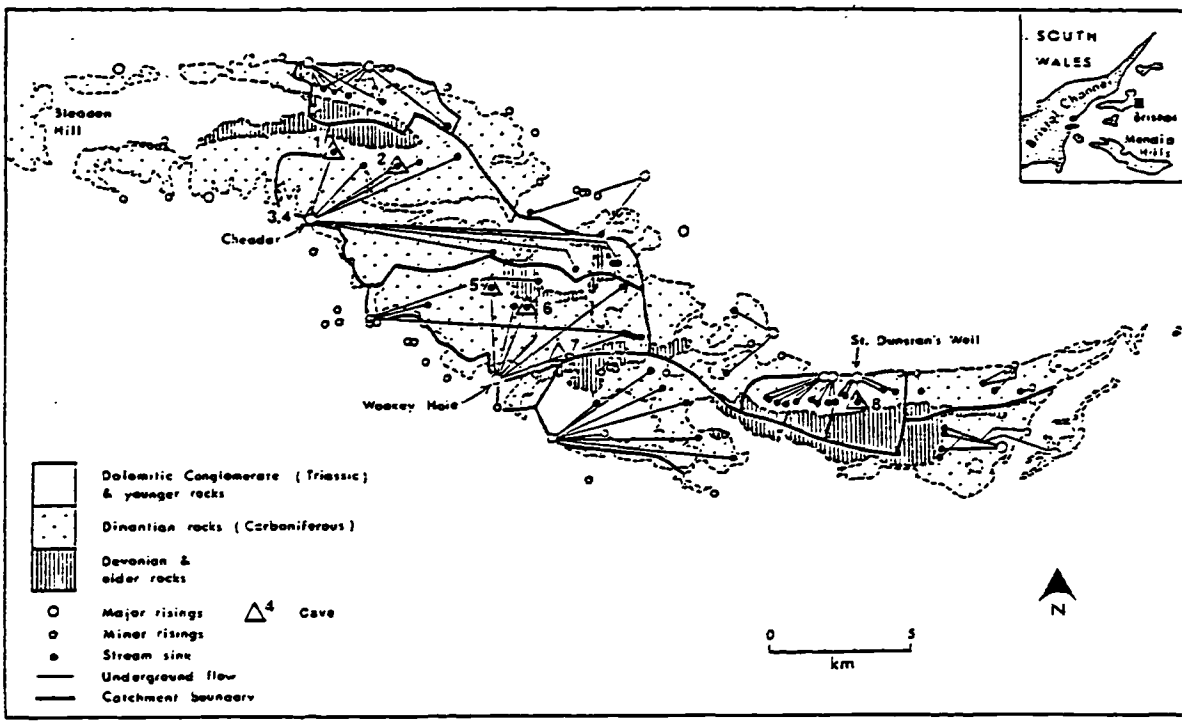

Figure 6.3 Location of caves within the Mendip Hills Region. 1. Charterhouse Cave, 2. Black Moor Swallet, 3. Goughs Cave, 4. Cox's Cave, 5. Swildons Hole, 6. St Cuthbert's Swallet, 7. Cuckoo Cleeves, 8. Fairy Cave.

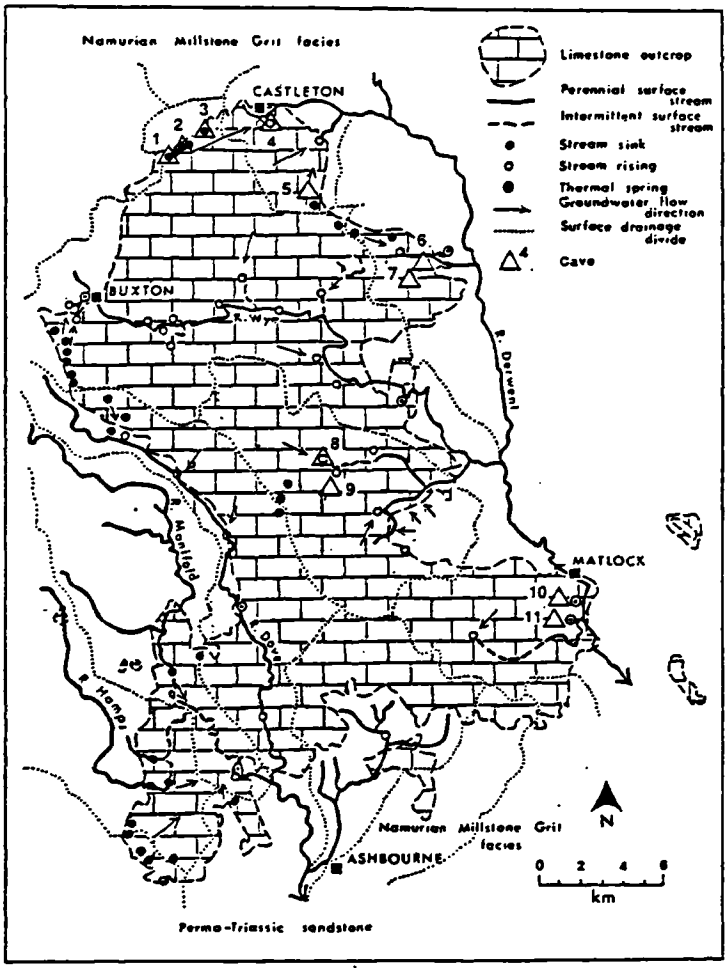

Figure 6.4 Location of caves within the Peak District. 1. Gautries Hole, 2. P8 (Jack Pot), 3.Giant's Hole, 4. Peak Cavern, 5. Bagshaw Cavern, 6. Carlswark Cavern, 8. Knotlow Cavern, 9. Hillocks Mine, 10. Jug Holes, 11, Devonshire Cavern.

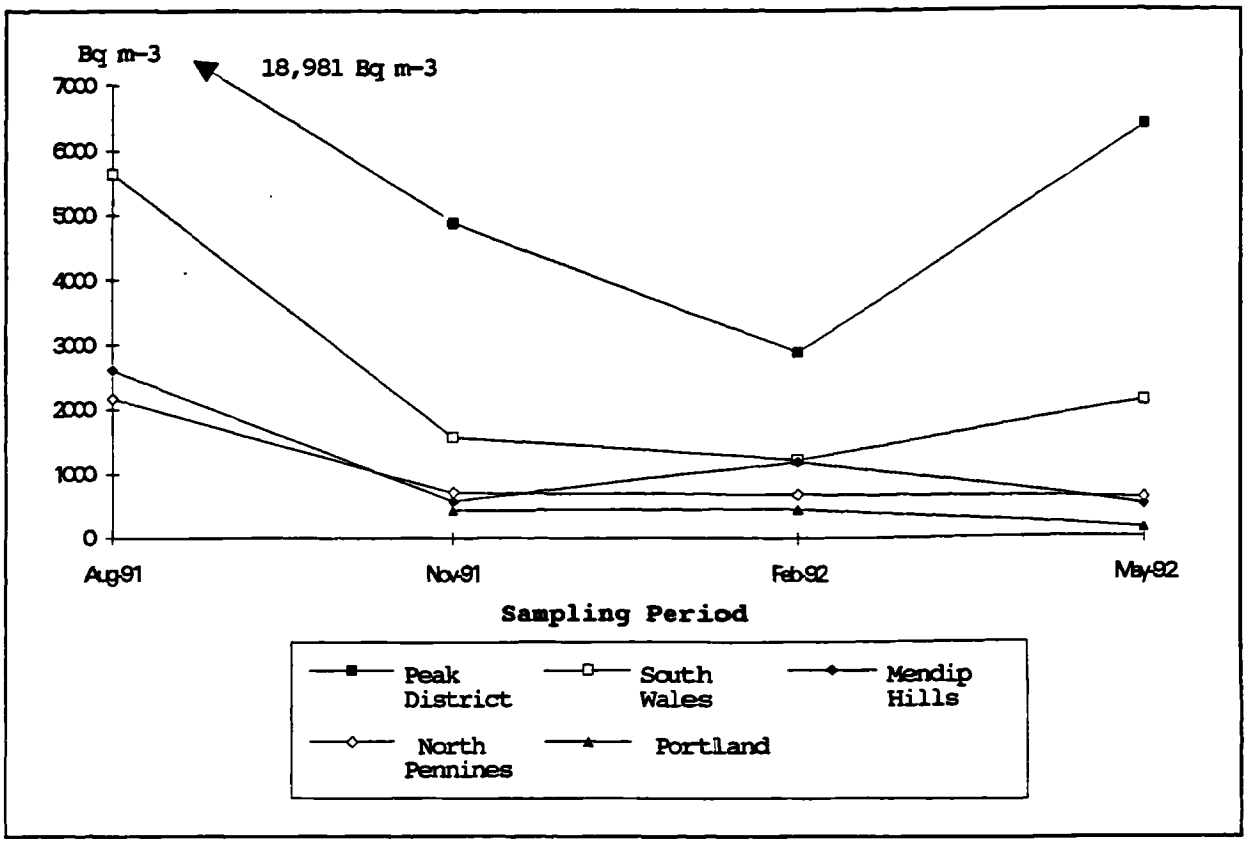

Figure 6.5 Seasonal variations in mean radon concentrations (Bq m^{-3}) by region

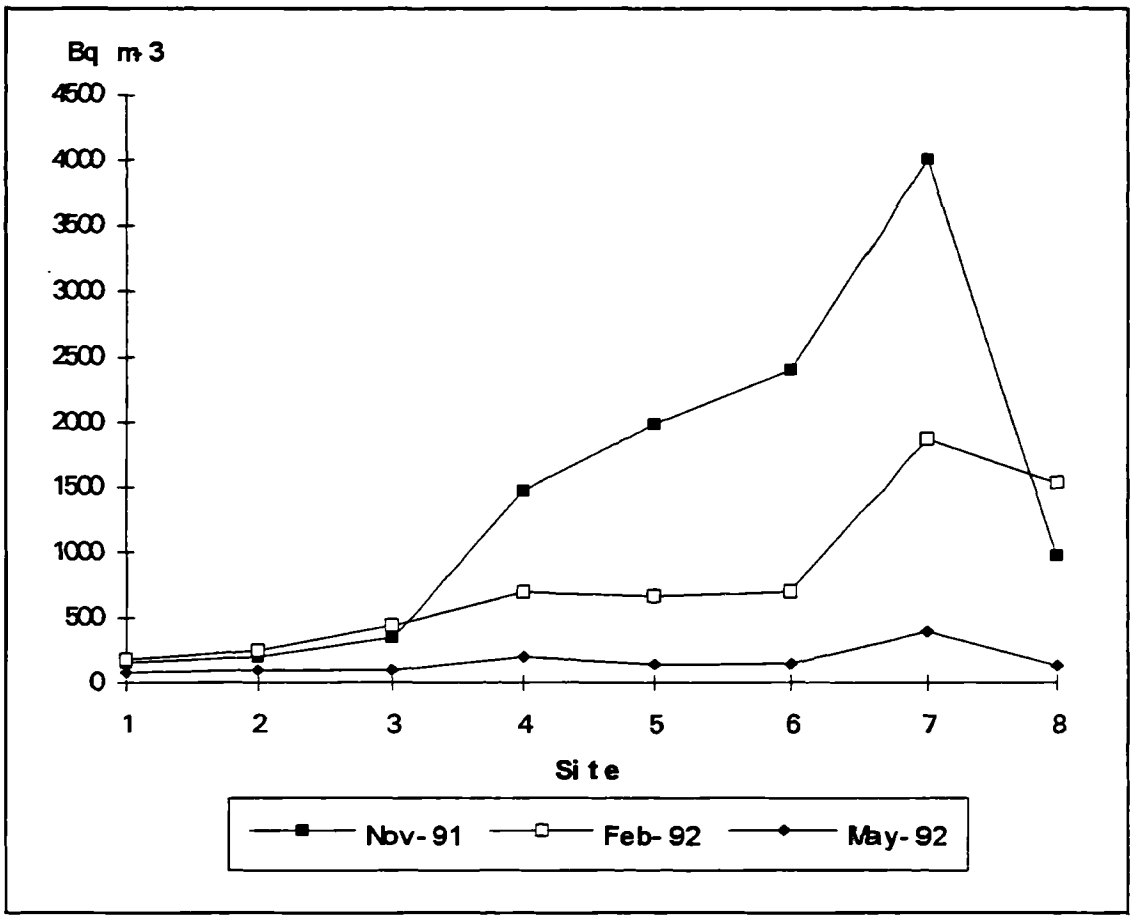

Figure 6.6 Variations in Radon Concentrations (Bq m^{-3}), Carno Adit, South Wales (for sample locations see figure 6.13)

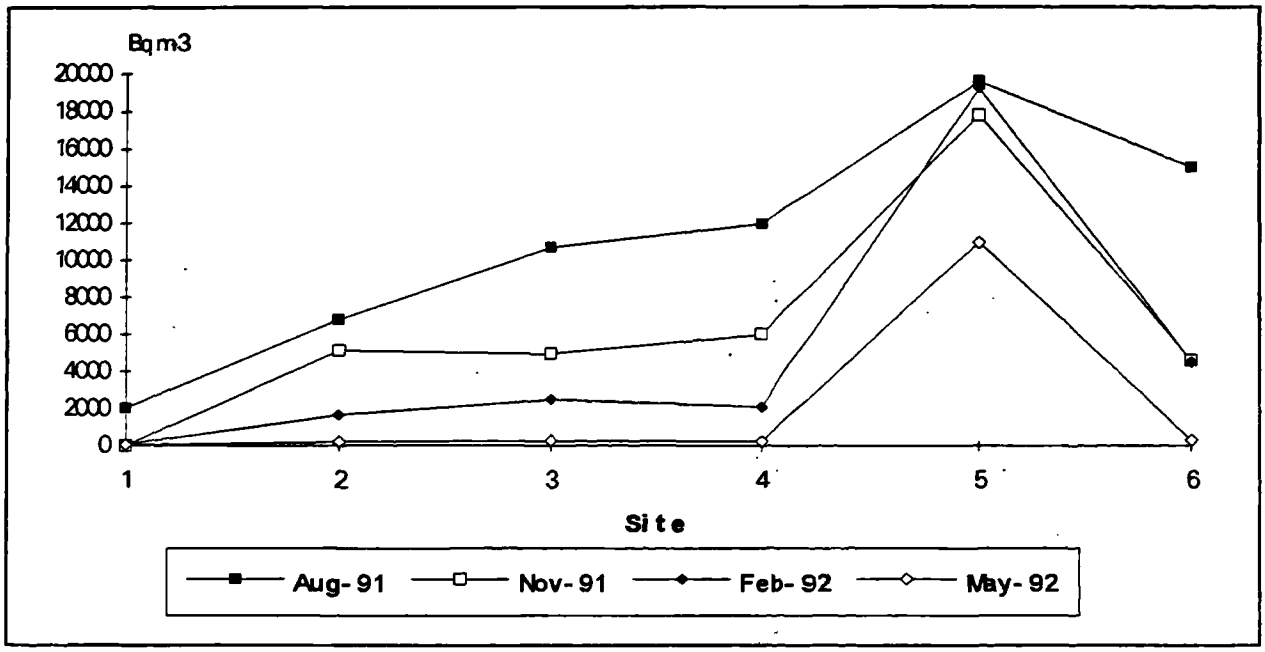

Figure 6.7 Variations in Radon Concentrations (Bq m^{-3}), Peak Cavern, Peak District (for sample locations see figure 6.14).

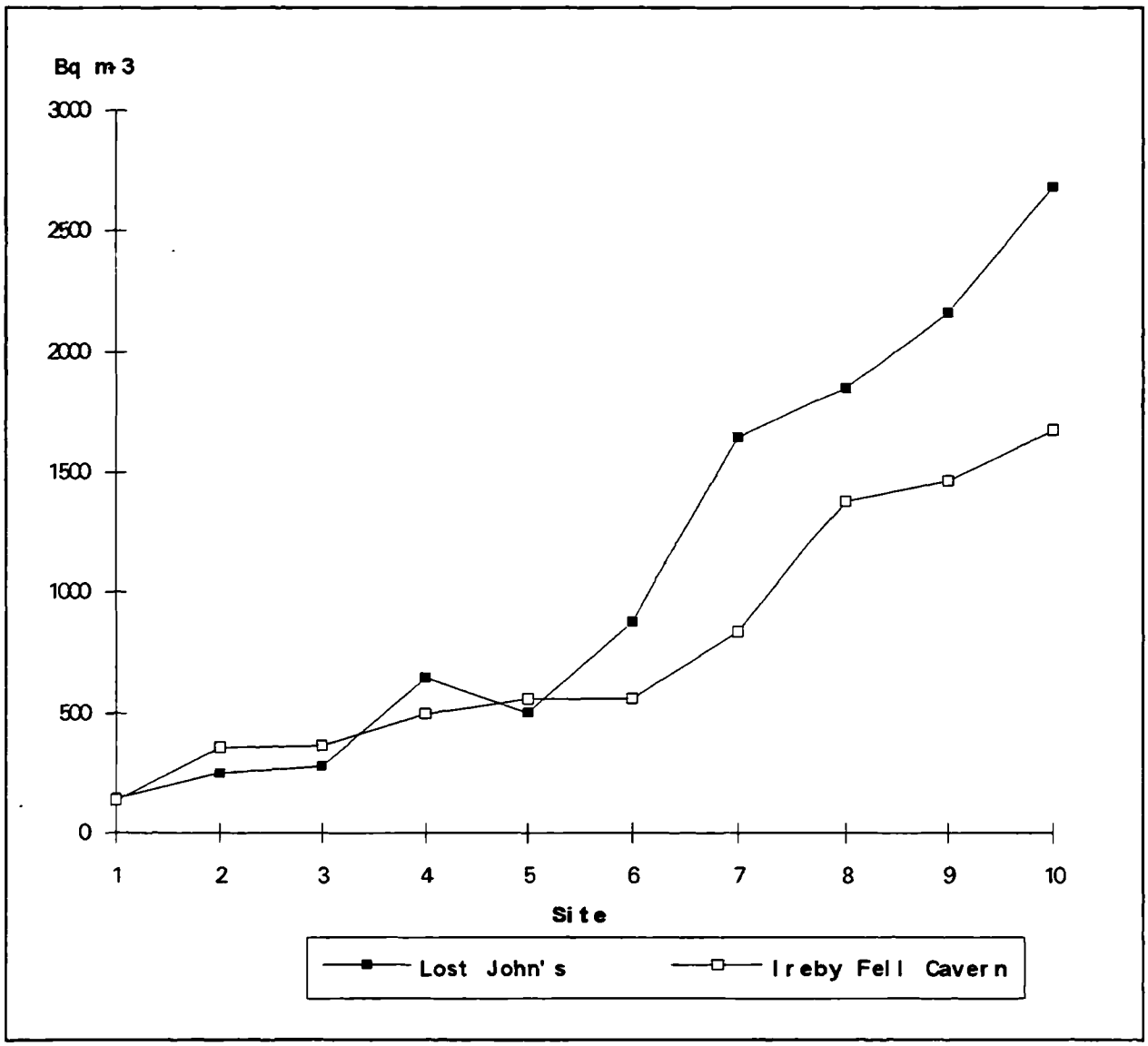

Figure 6.8 Mean Radon Concentrations (Bq m^{-3}), Ireby Fell and Lost John's, North Pennines (for sample locations see figures 6.15 and 6.16).

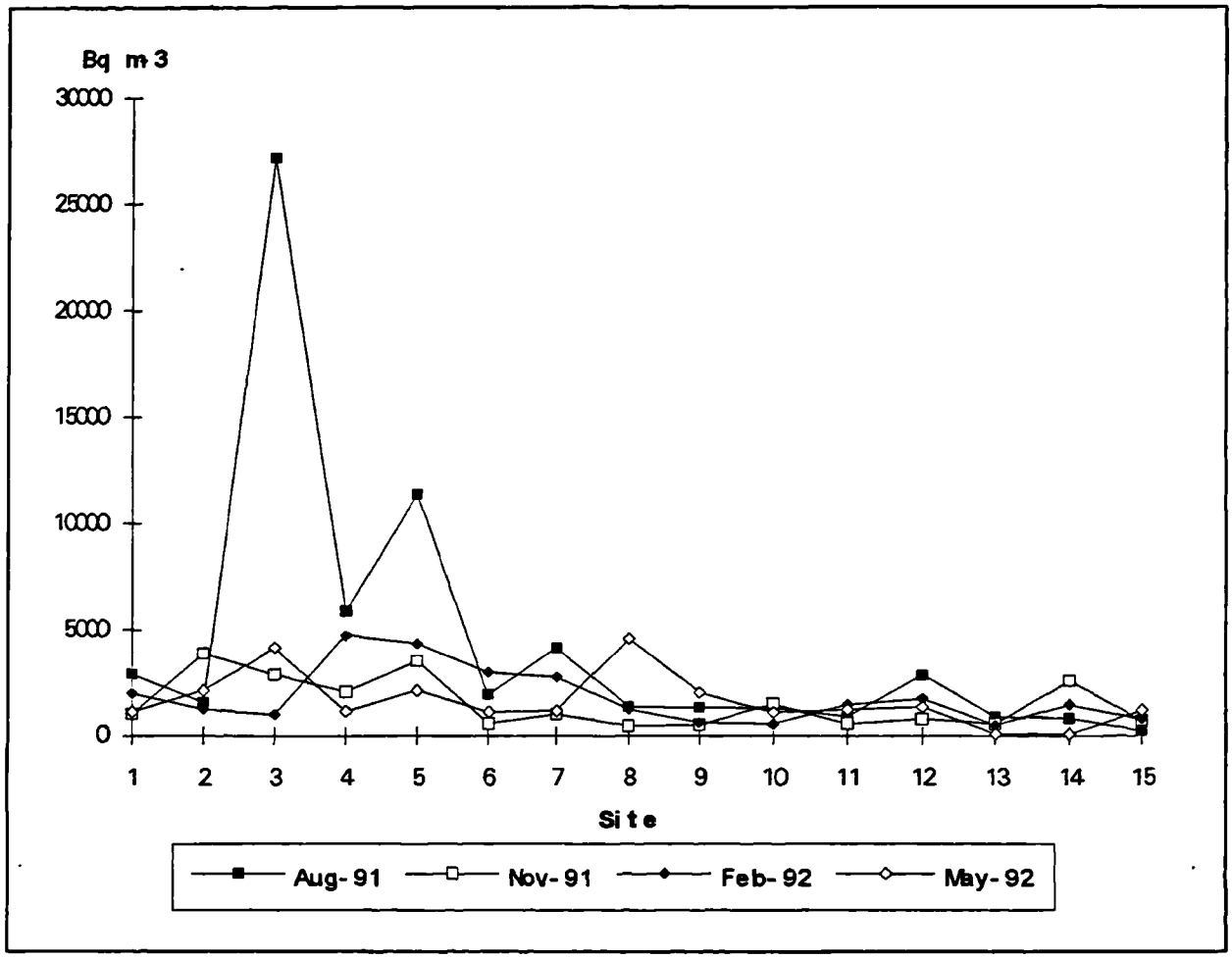

Figure 6.9 Seasonal Radon Concentrations (Bq m^{-3}), Ease Gill Caverns, North Pennines (for sample locations see figure 6.17).

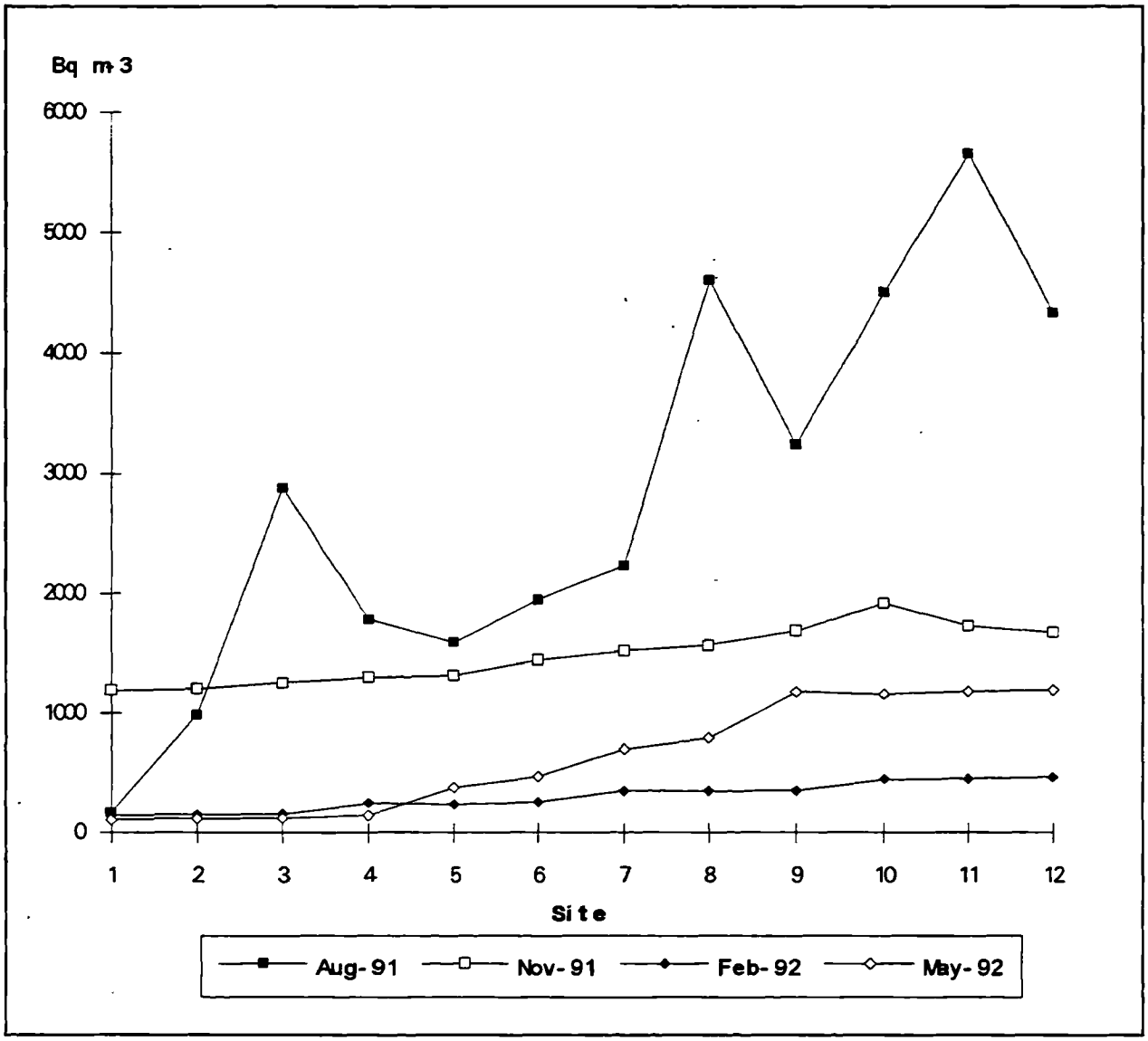

Figure 6.10 Seasonal Radon Concentrations (Bq m^{-3}), Kingsdale Master Cave, North Pennines (for sample locations see figure 6.18).

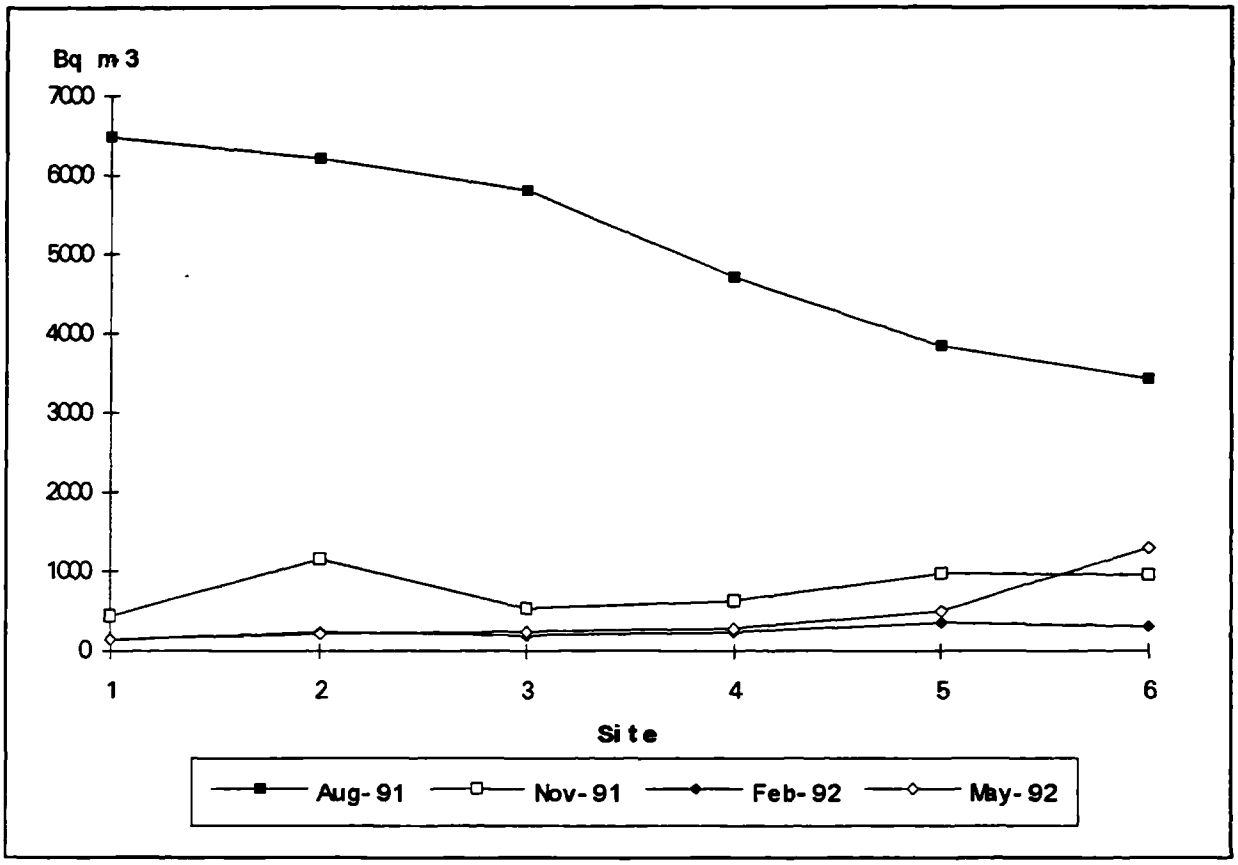

Figure 6.11 Seasonal Radon Concentrations (Bq m^{-3}), White Scar Cave, North Pennines (for sample locations see figure 6.19).

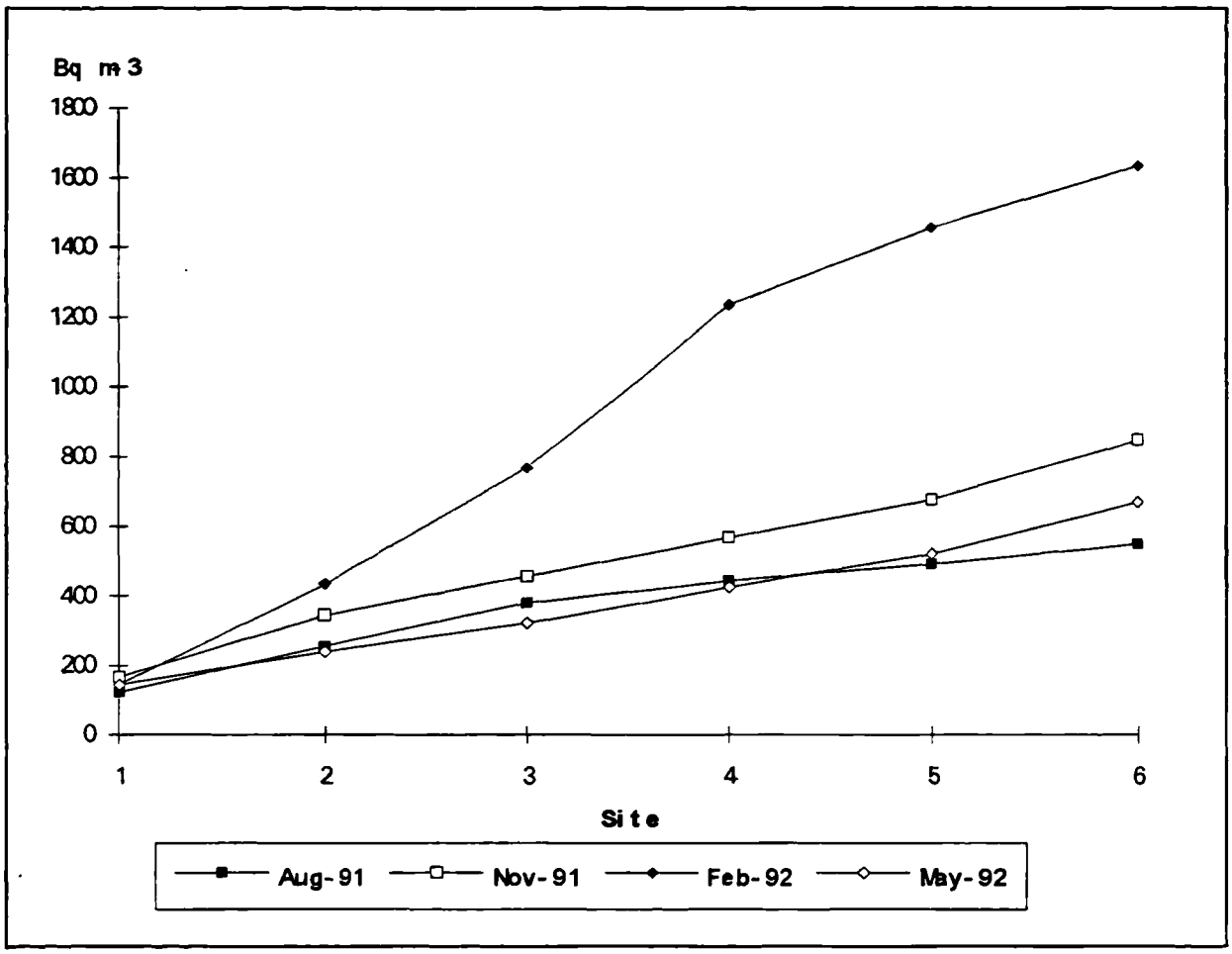

Figure 6.12 Seasonal Radon Concentrations (Bq m^{-3}), Heron Pot, North Pennines.

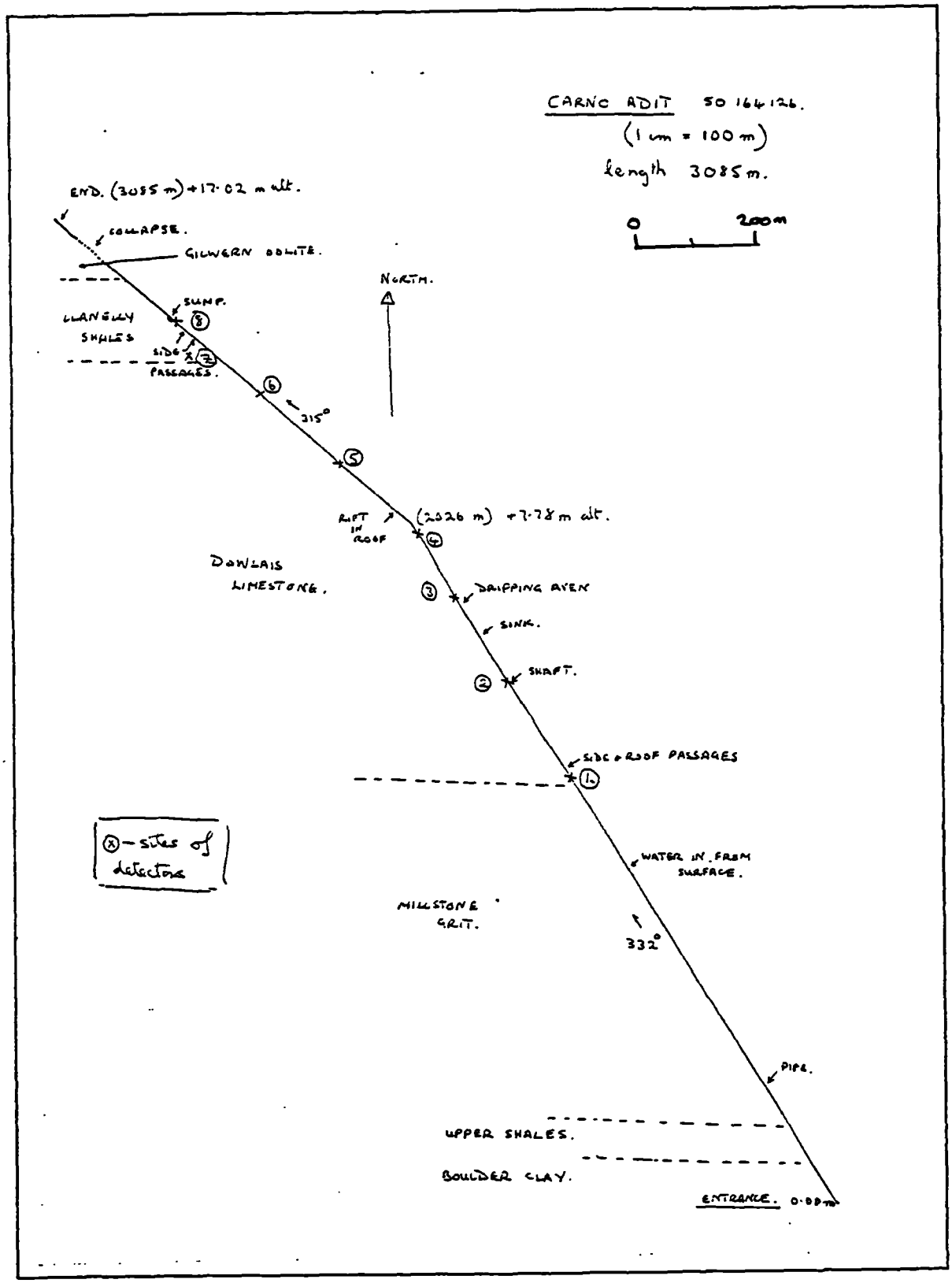

Figure 6.13 Generalised plan showing sampling locations and geology of Carno Adit, South Wales.

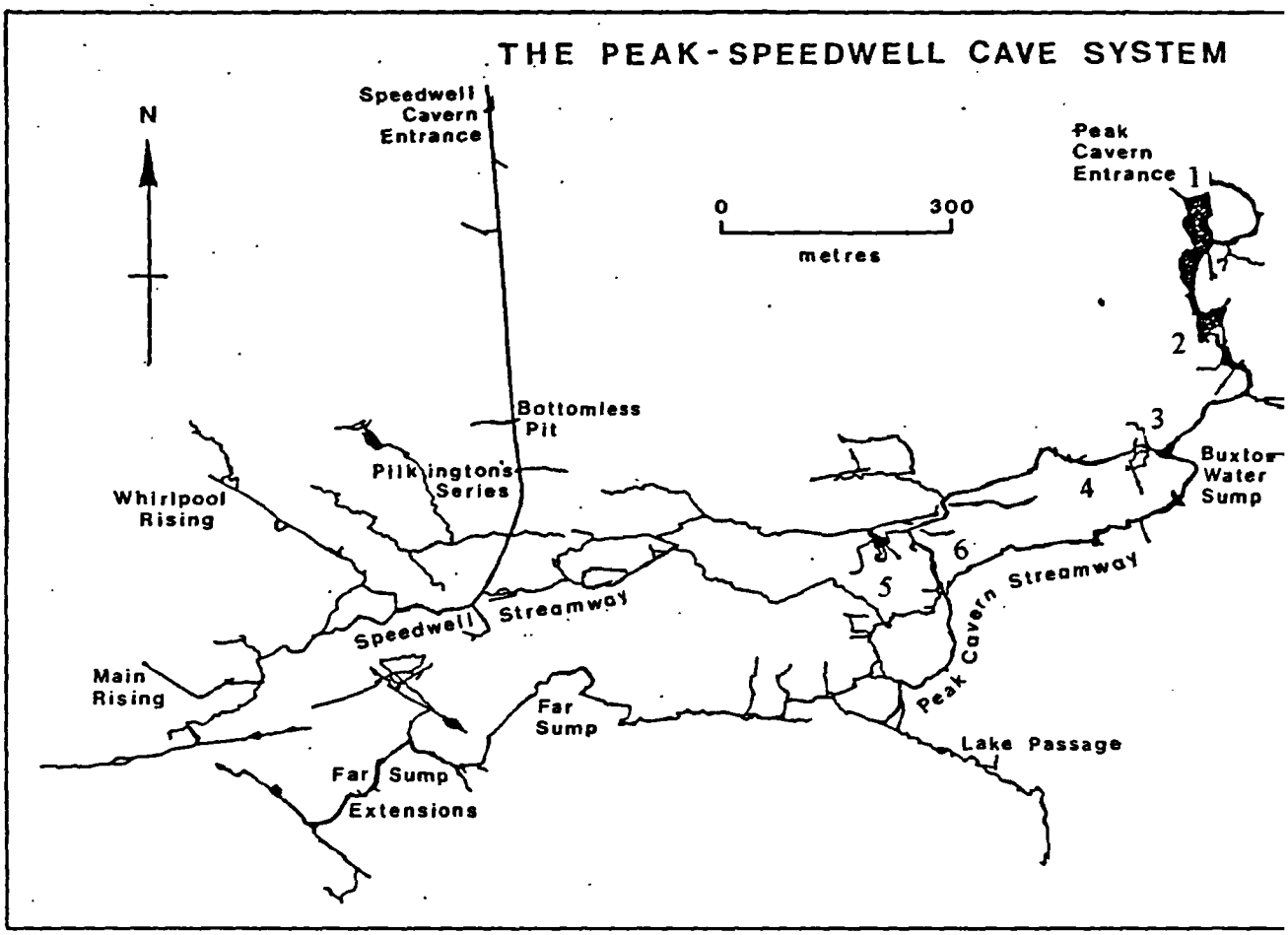

Figure 6.14 Sampling sites in Peak Cavern.

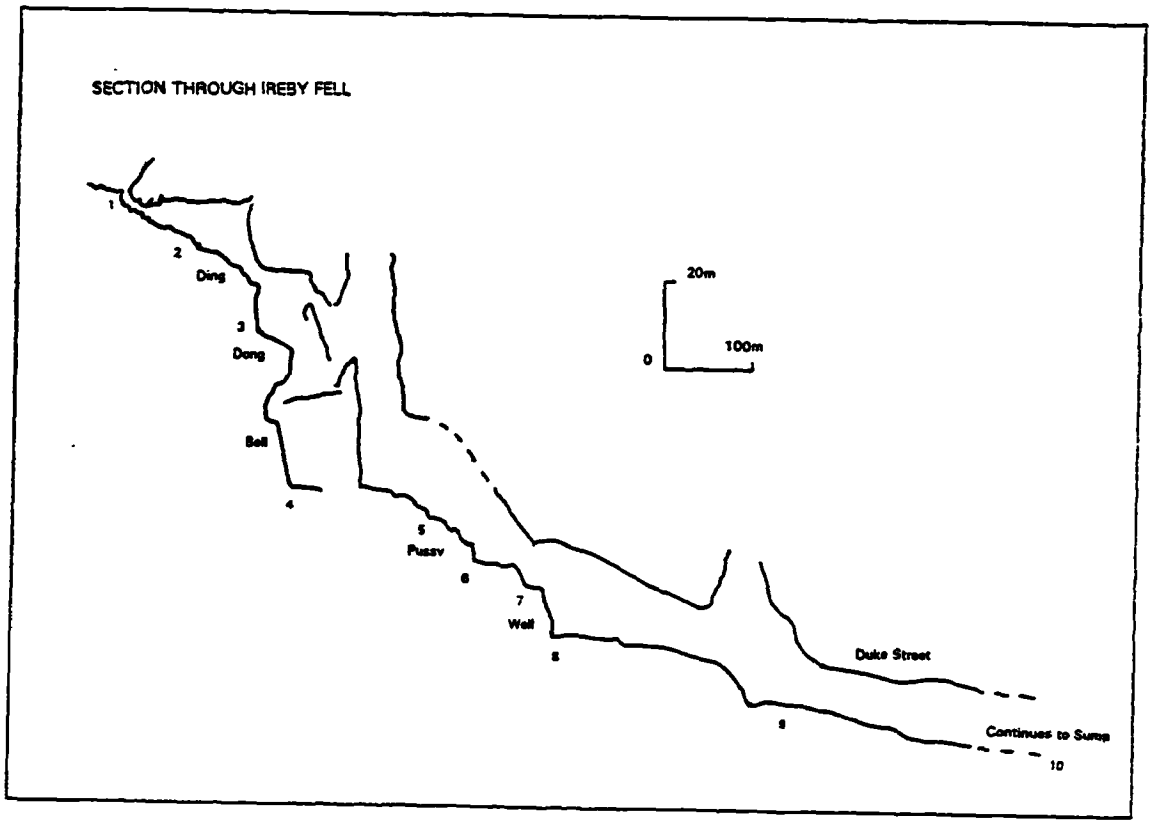

Figure 6.15 Location of monitoring sites in Ireby Fell Cavern.

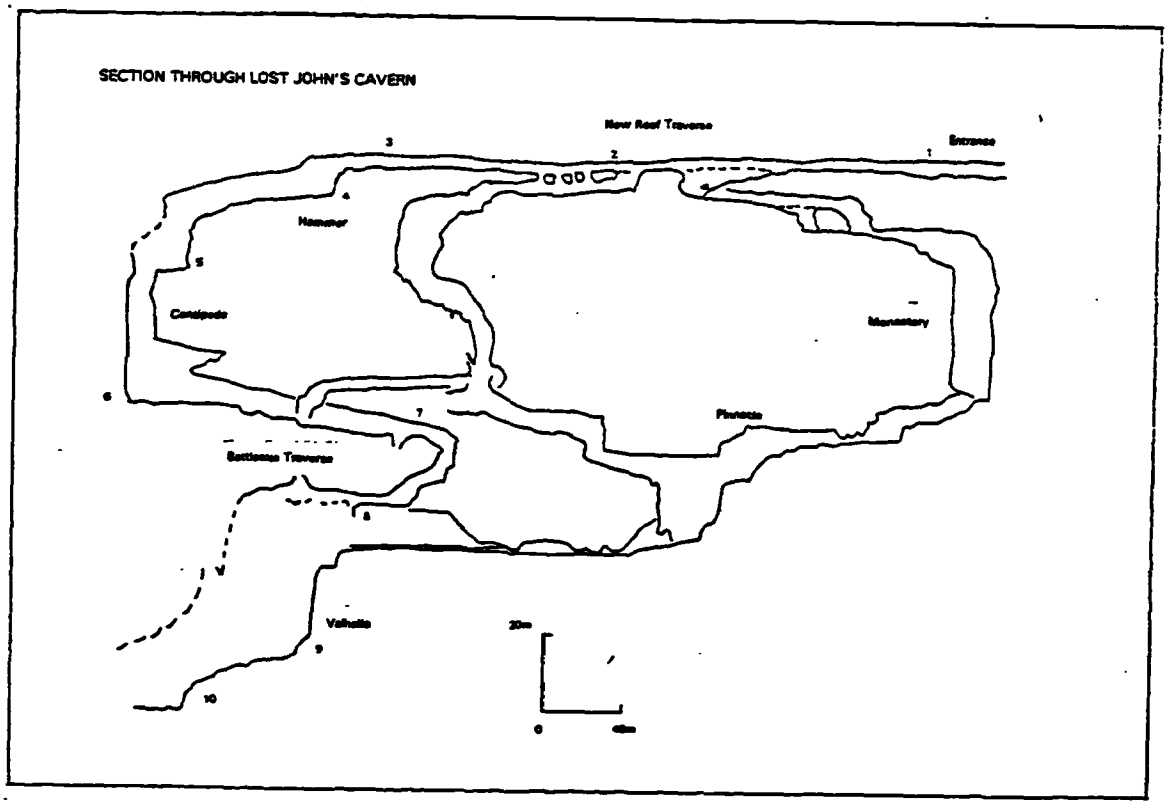

Figure 6.16 Location of monitoring sites within Lost Johns Caverns.

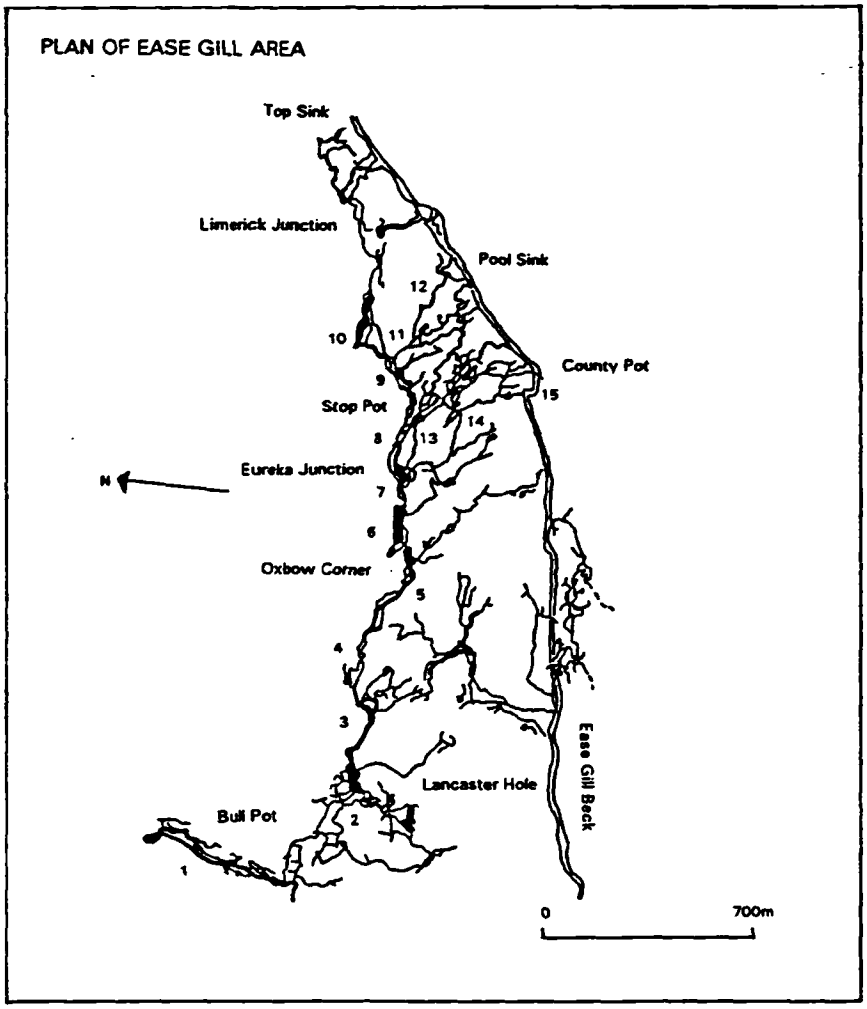

Figure 6.17 Location of monitoring sites within Ease Gill Caverns.

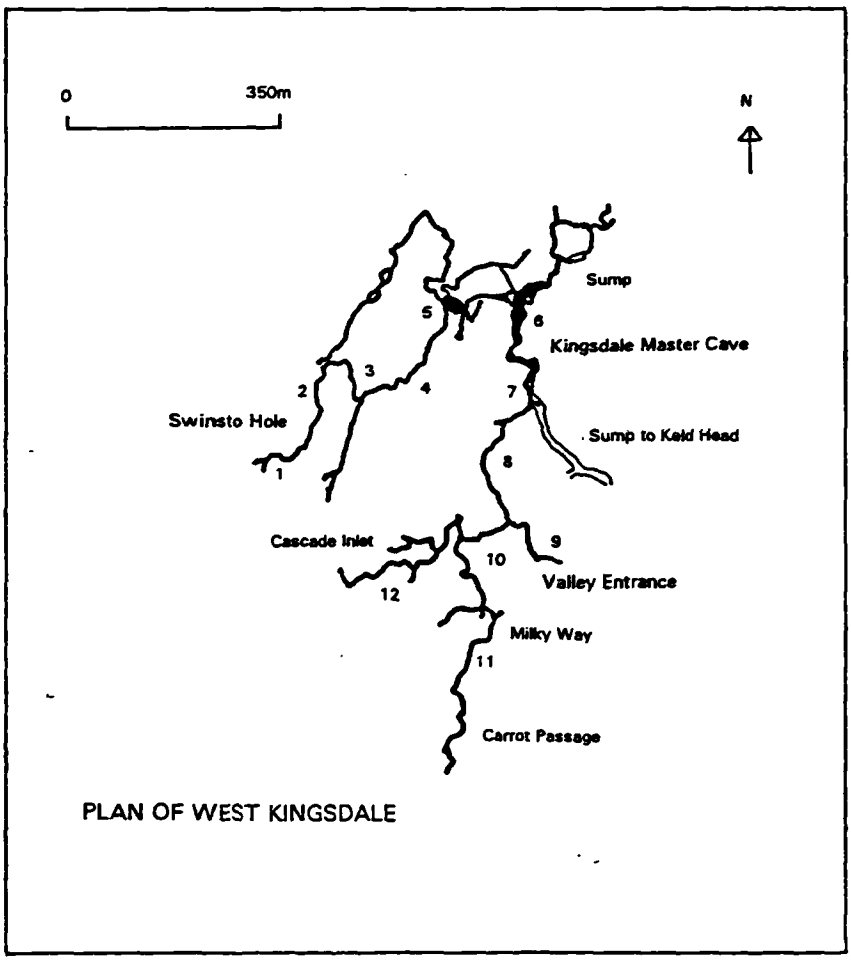

Figure 6.18 Location of monitoring sites within Kingsdale Master Cave.

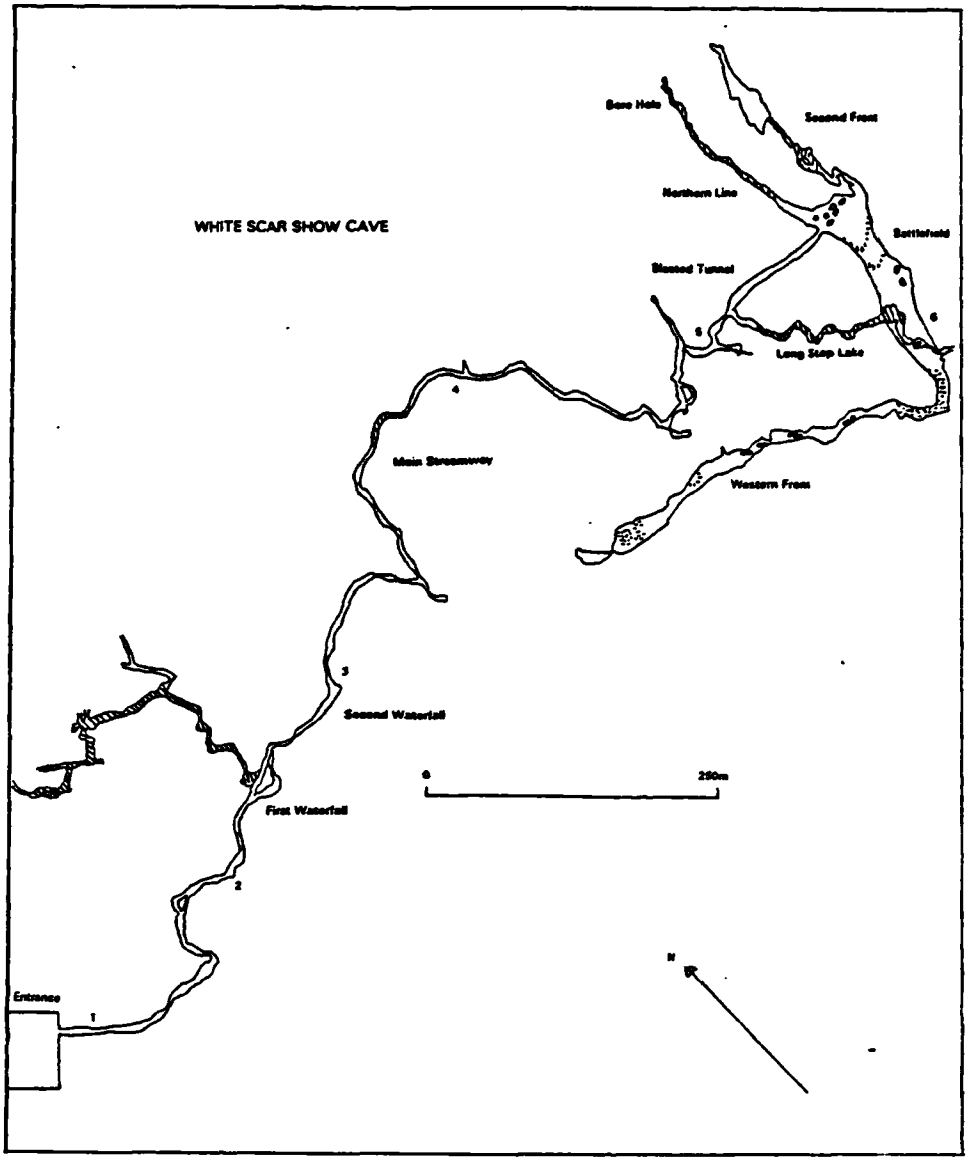

Figure 6.19 Location of monitoring sites within White Scar Cave.

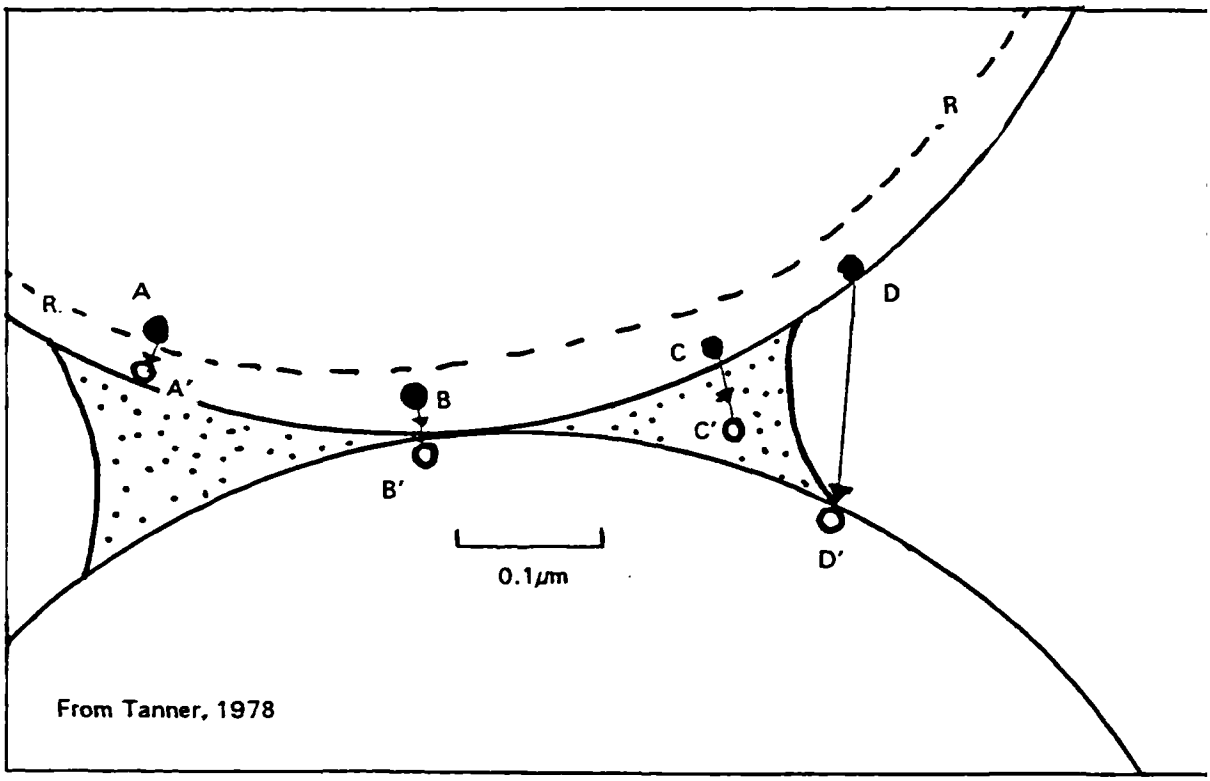

Figure 7.1 Schematic diagram of emanation processes. Two fine spherical grains are in contact, near to B. Water is present in the stippled zone of the pore; the open zone is air filled. Radium-226 atoms (closed circles) decay in the upper grain, each yielding an alpha particle and a radon-222 atom (open circles). Atom A lies at a greater depth within the grain than the recoil range, R; recoil radon-222 atom A' is contained within the grain. Atom B' escapes from the upper grain but buries itself in the lower grain. After escape from the upper grain, atom c' losses the remained of its recoil energy in the water and is free to diffuse through the pores. Atom D' losses little of its recoil energy in the air and buries itself in the lower grain. Atoms B' and D' may escape from their recoil pockets by diffusion before condensation of the excited atoms of the grain (indirect-recoil effect). With a mean half life of radon-222 its diffusion distance in solid grain is less than the width of any line in the figure.

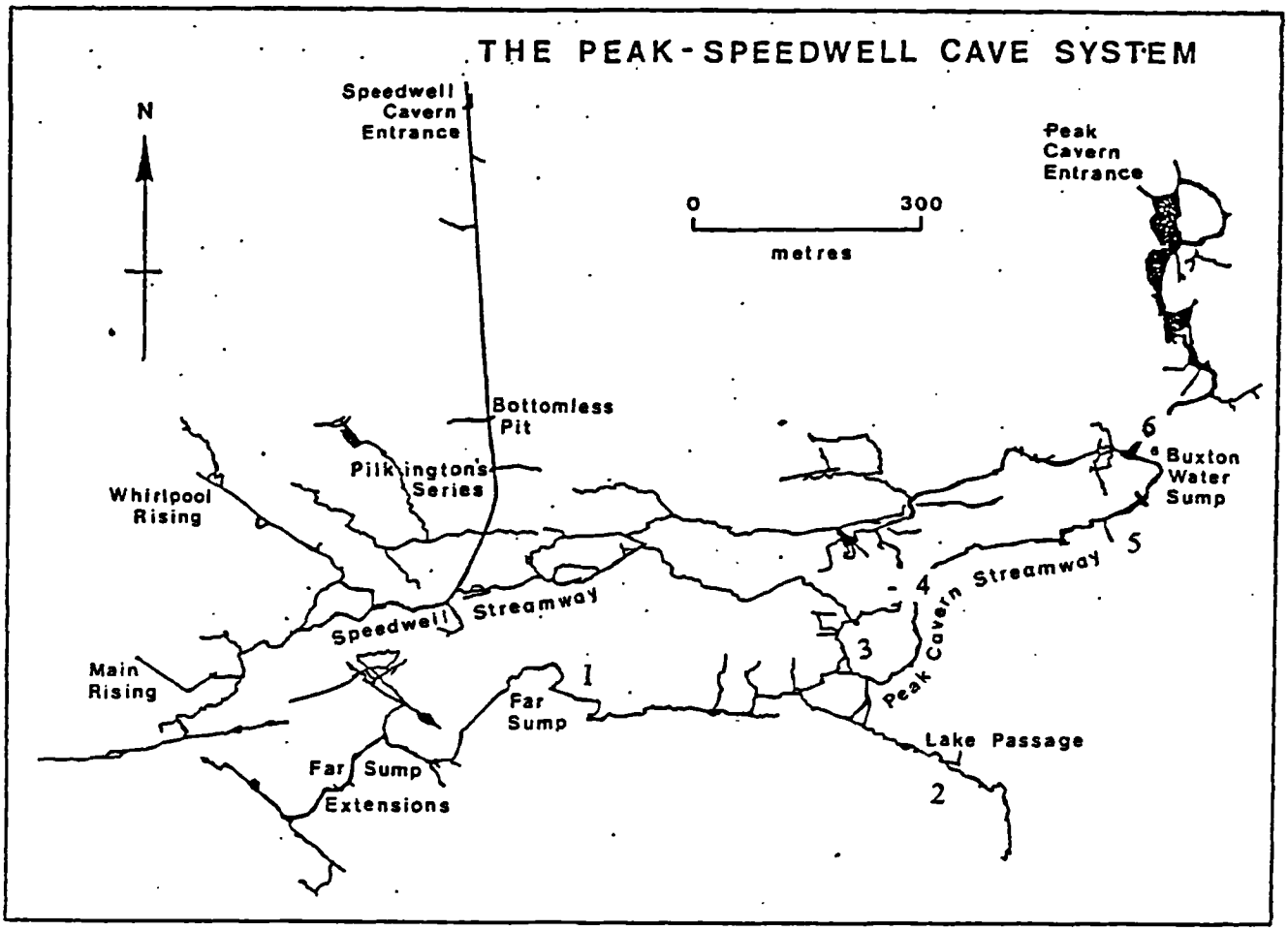

Figure 7.2 Water sampling sites in Peak Cavern

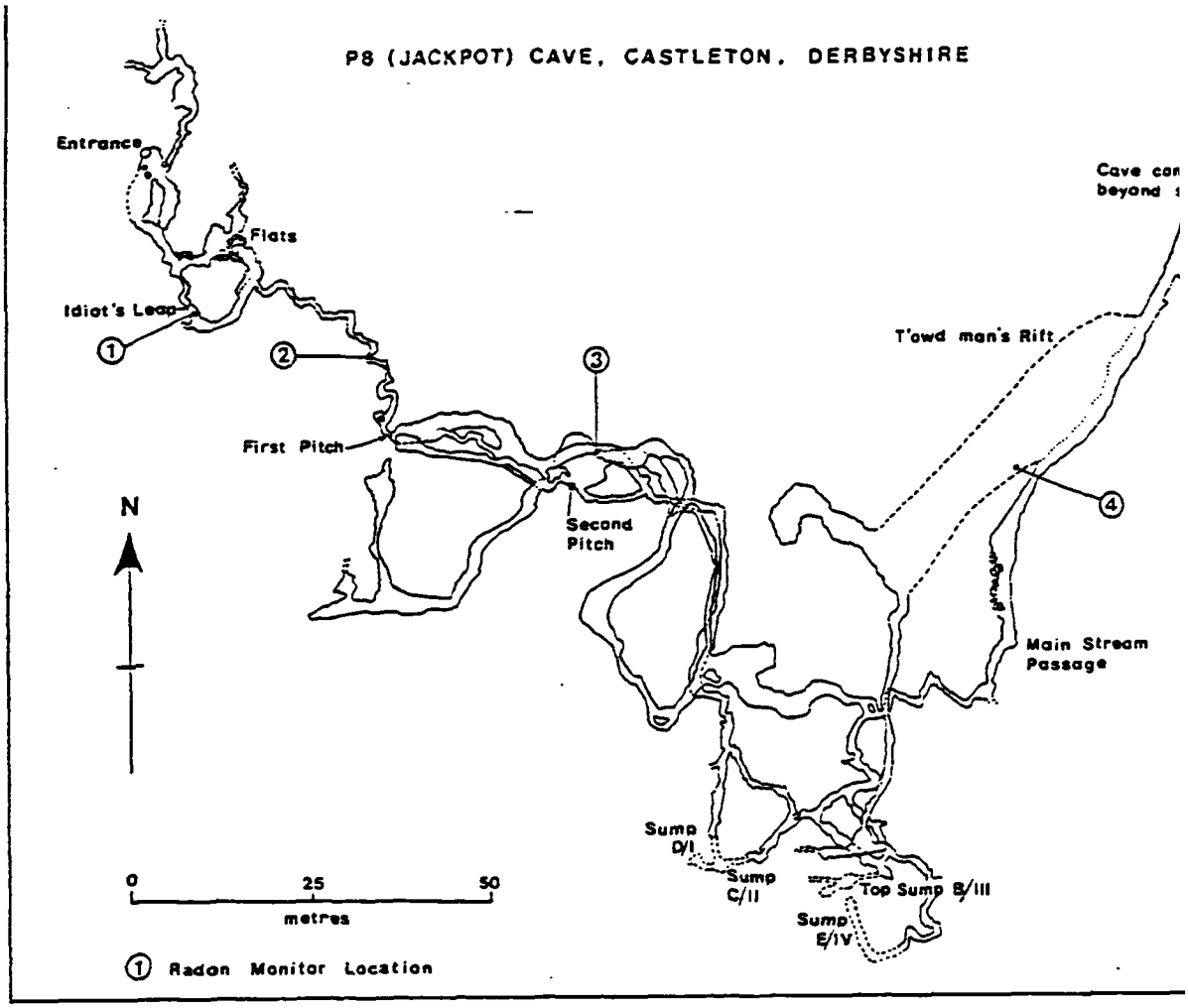

Figure 8.1 Location of monitors in P8 (Jack Pot) cave.

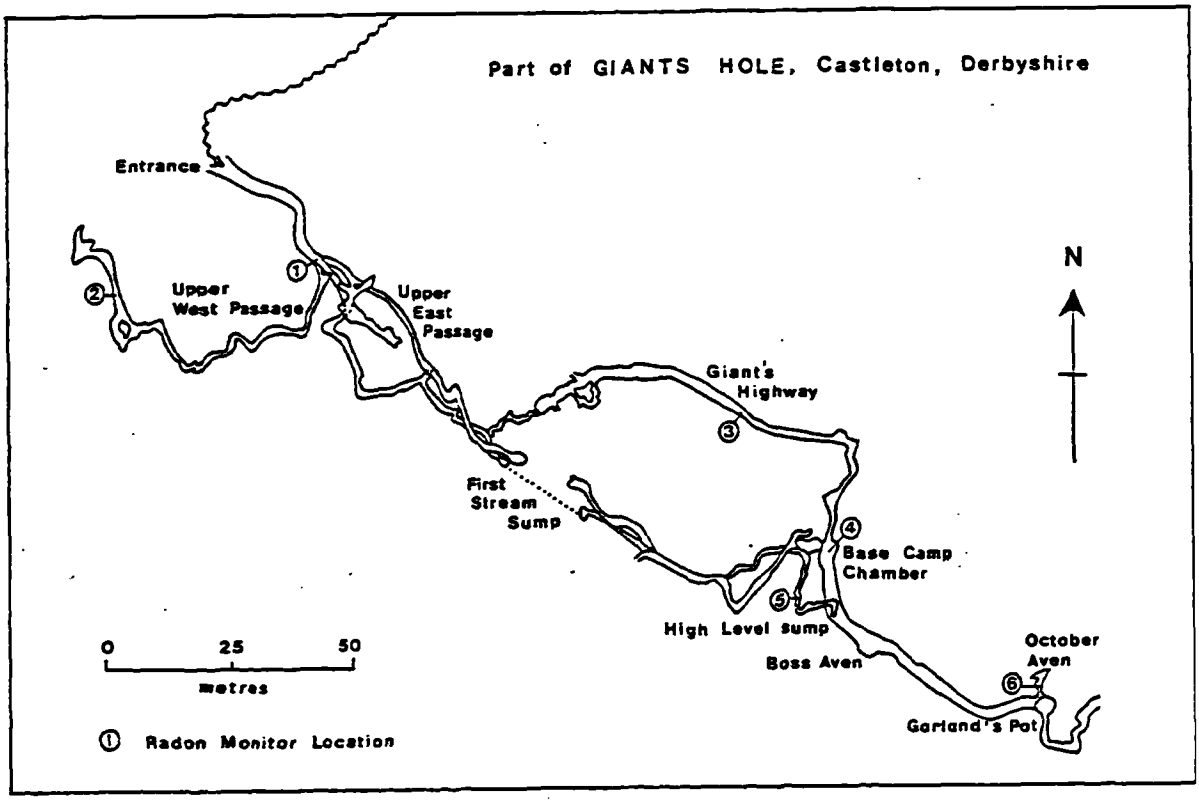

Figure 8.2 Location of monitors in Giant's Hole

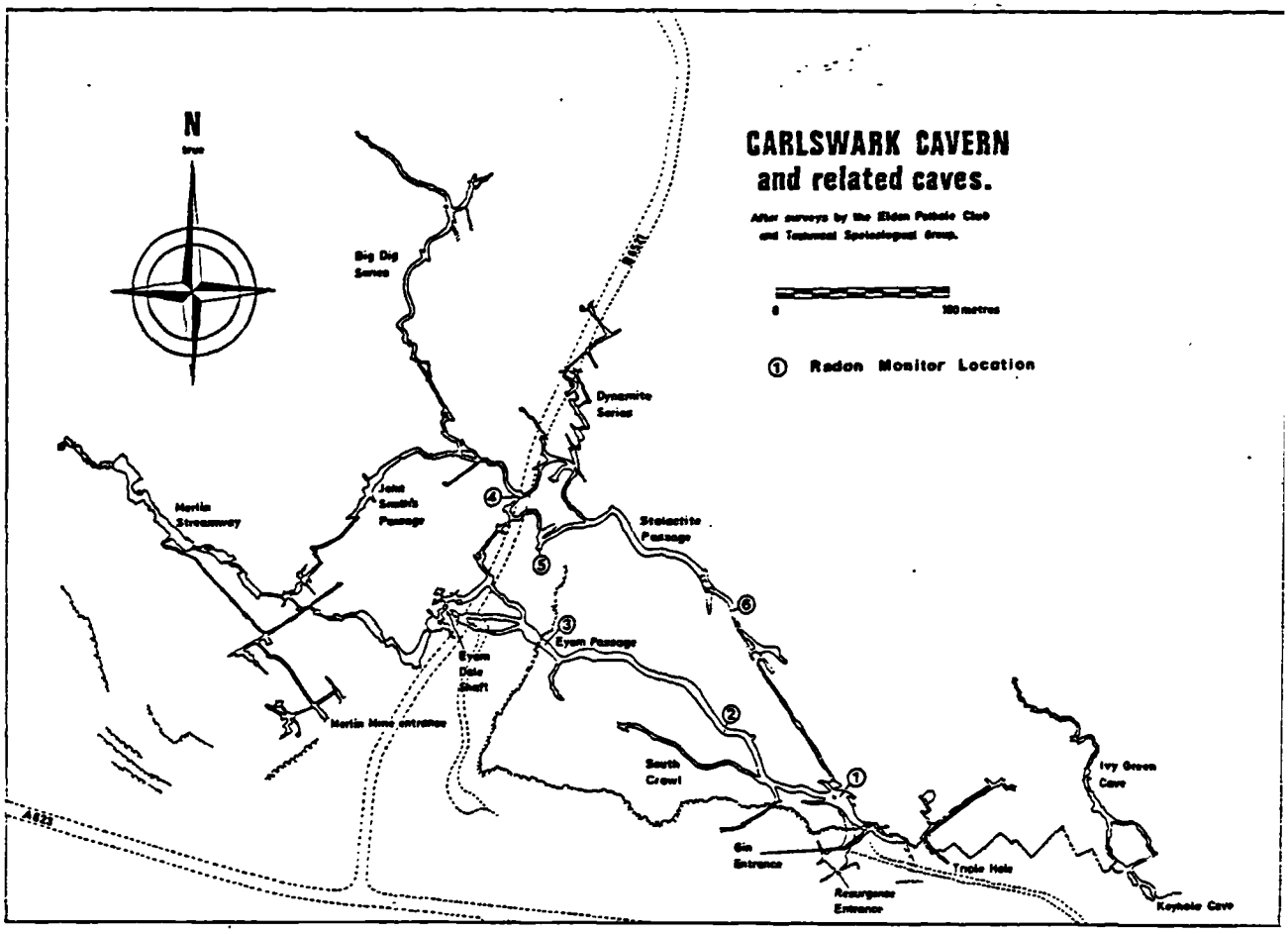

Figure 8.3 Location of monitors in Carlswark Cavern.

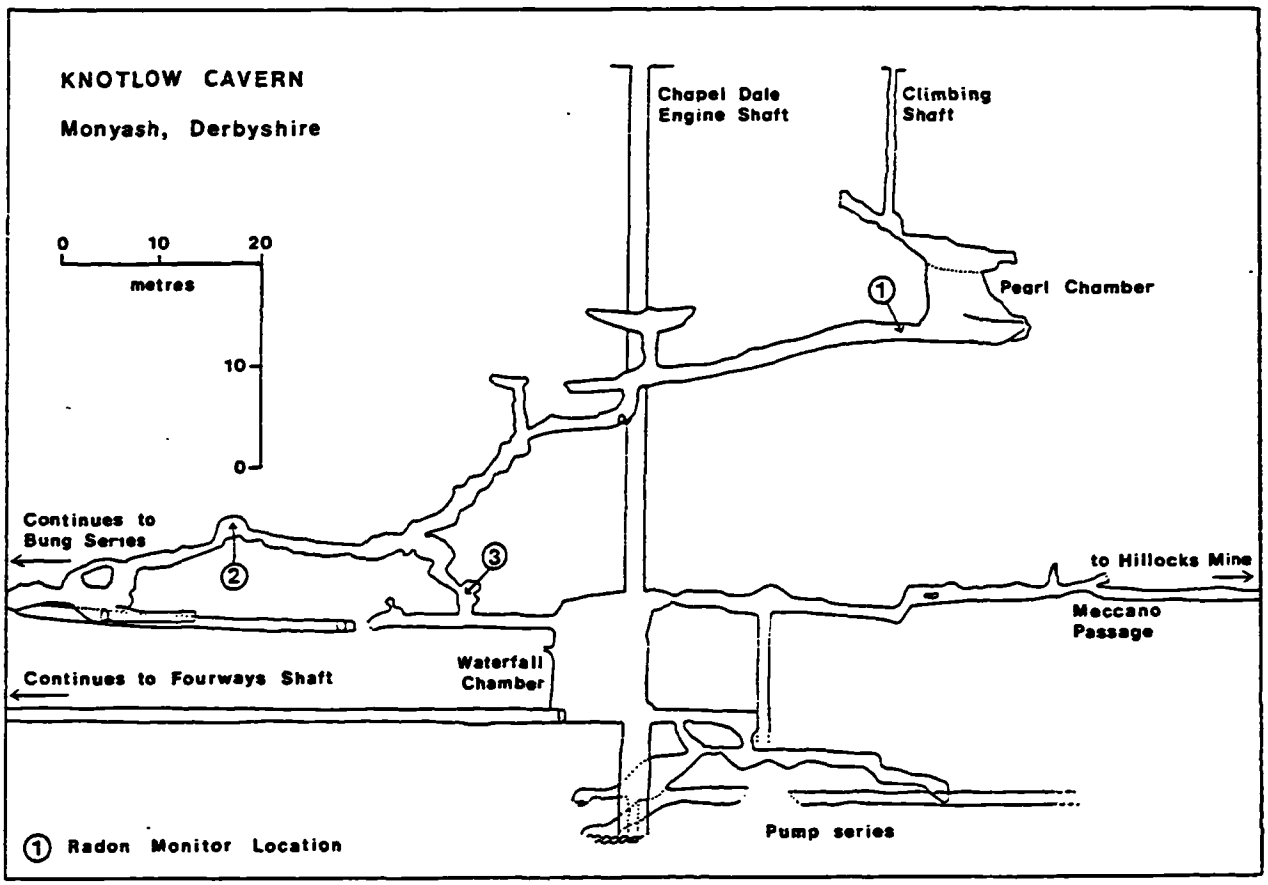

Figure 8.4 Location of monitors in Knotlow Cavern (section).

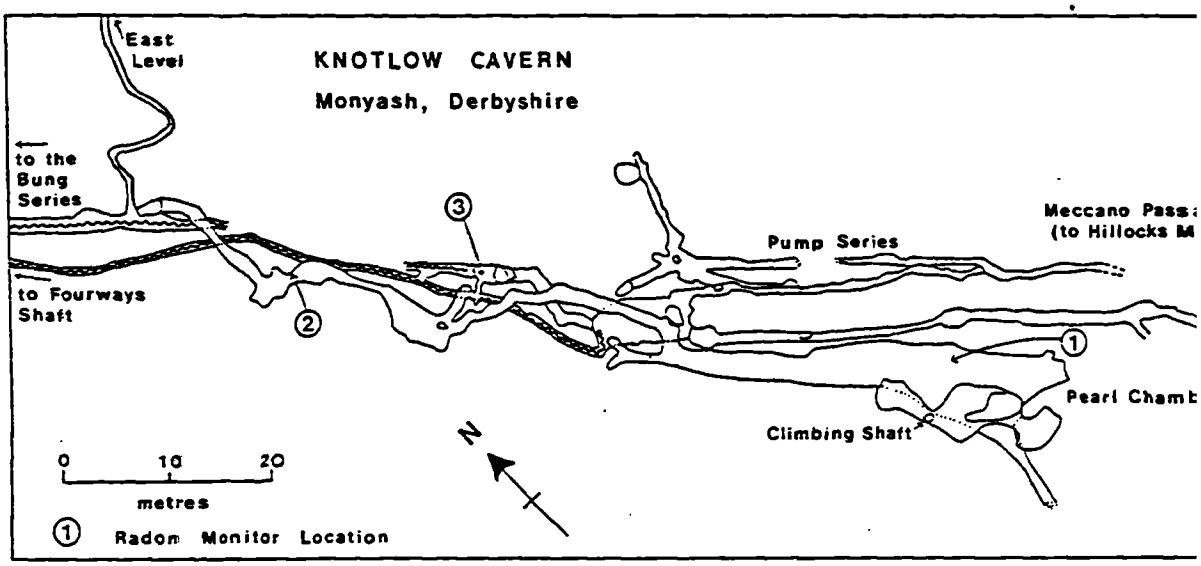

Figure 8.5 Location of monitors in Knotlow Cavern (plan).

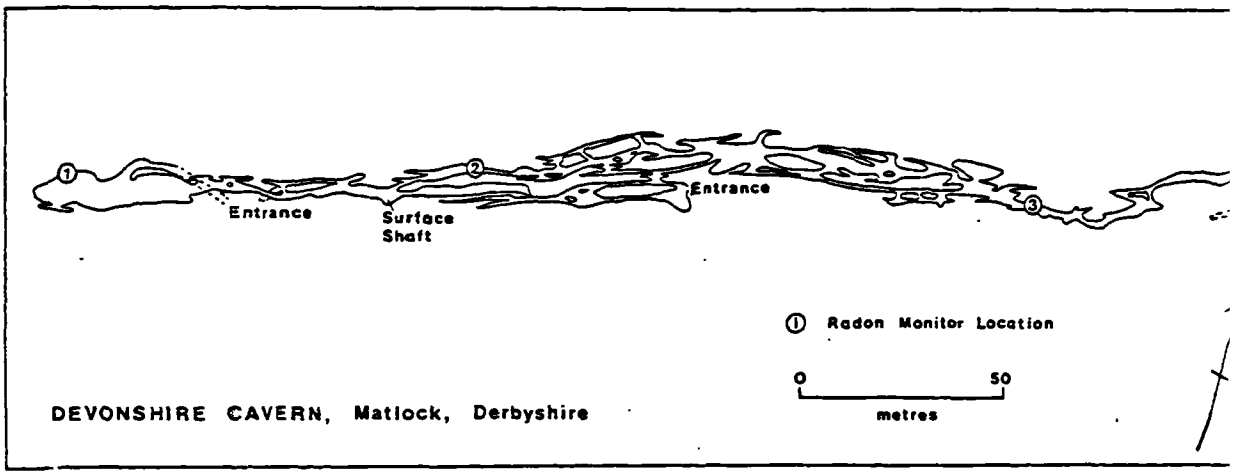

Figure 8.6 Location of monitors in Devonshire Cavern.

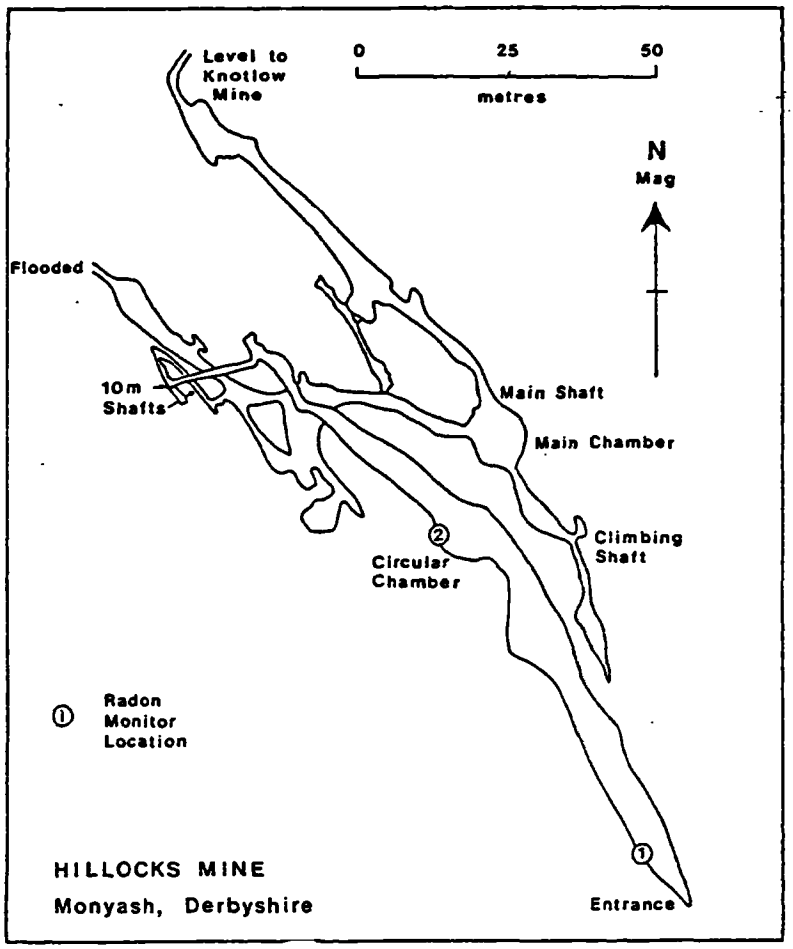

Figure 8.7 Location of monitors in Hilocks Mine.

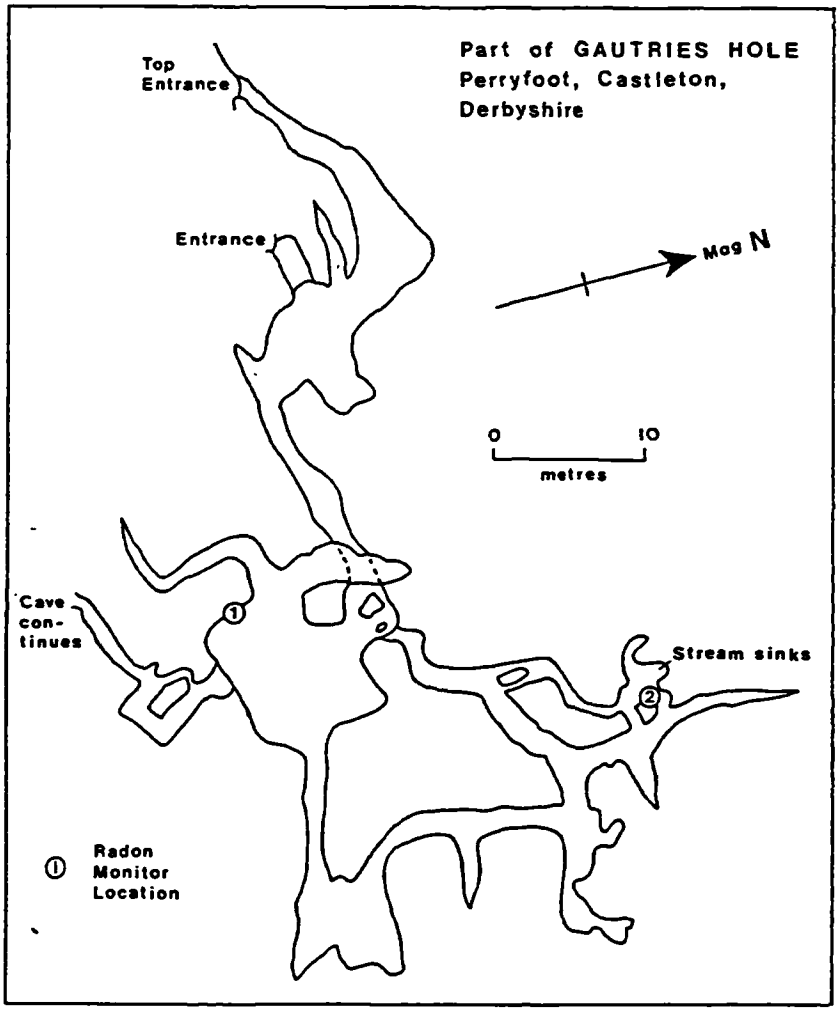

Figure 8.8 Location of monitors in Gautries Hole

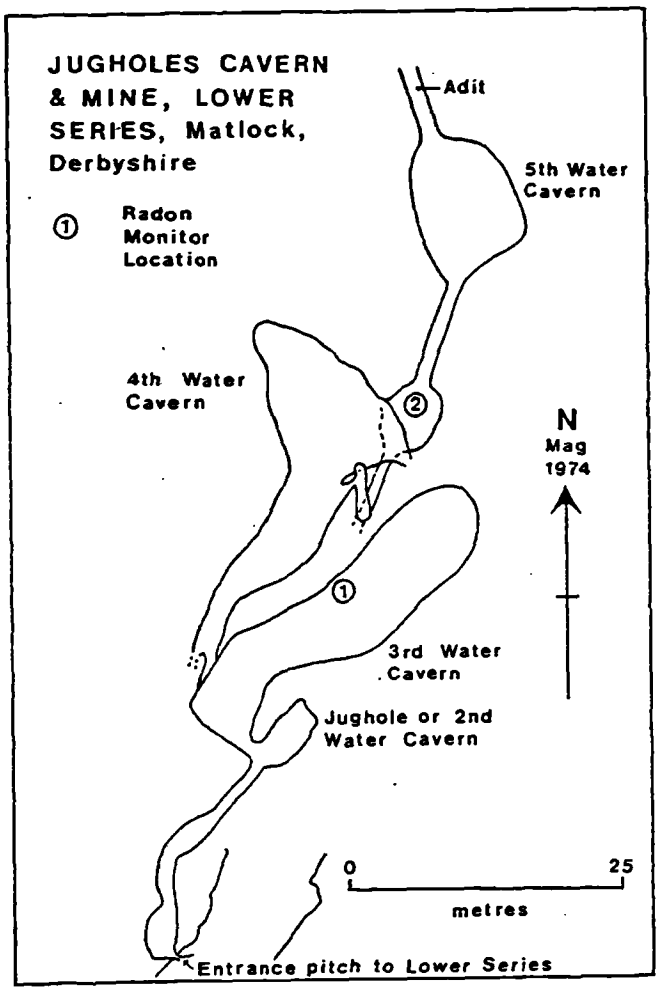

Figure 8.9 Location of monitors in Jug Holes Cavern and Mine

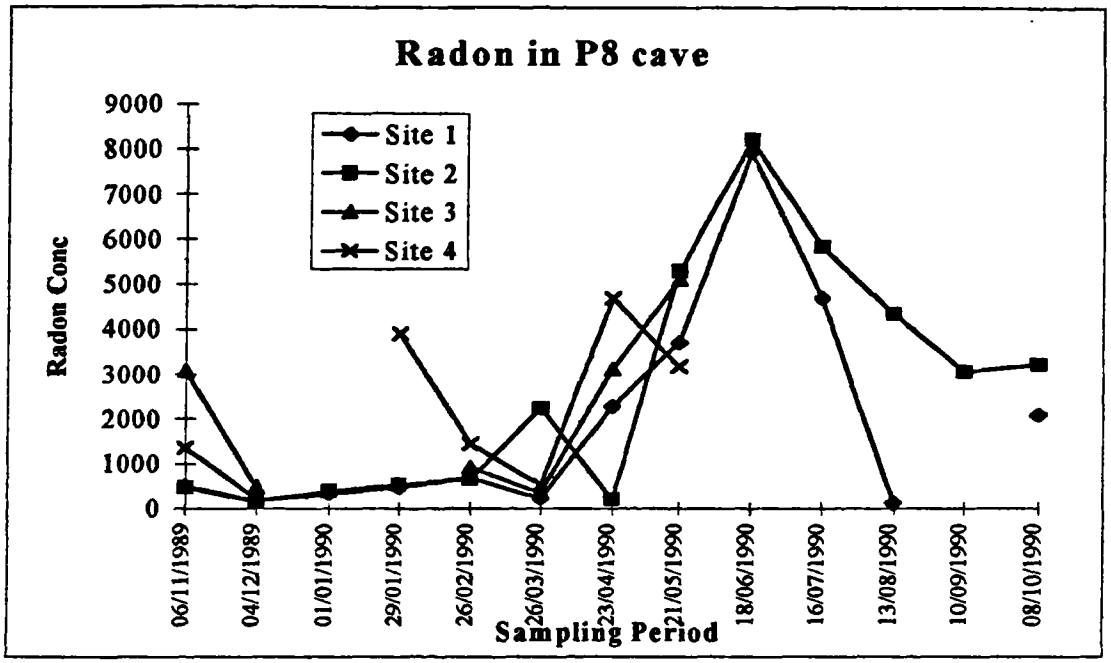

Figure 8.10 Radon concentrations in P8 cave (Bq m^{-3}).

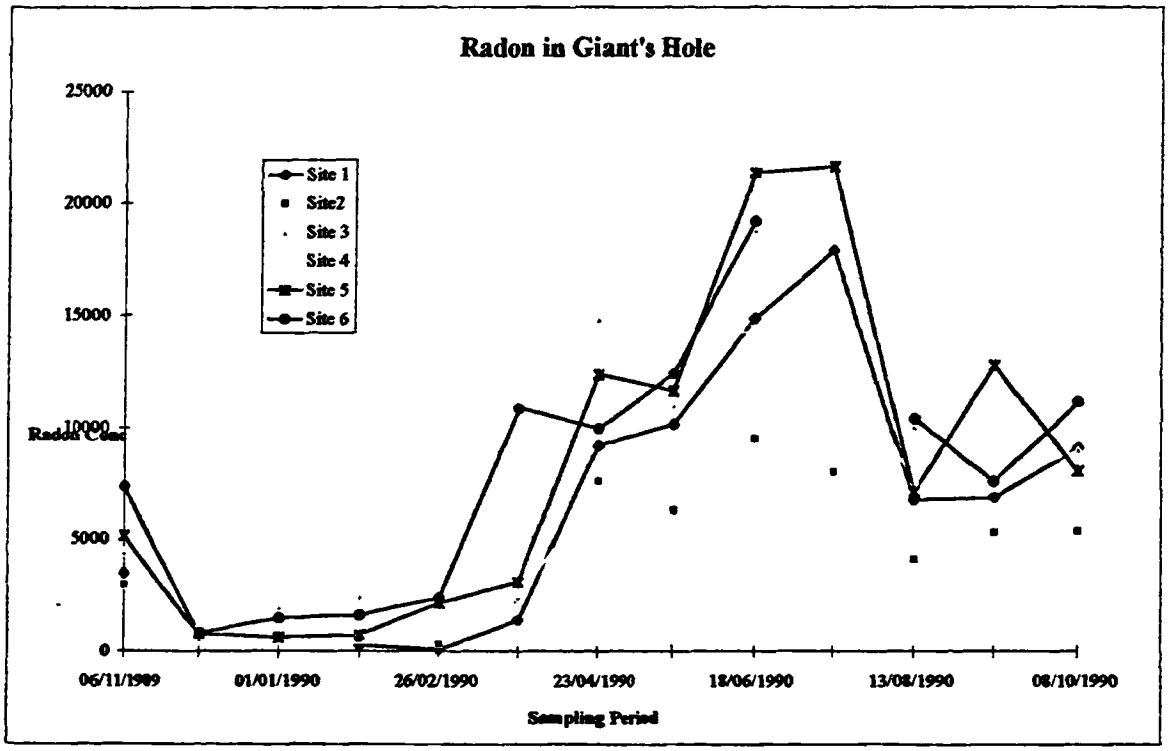

Figure 8.11 Radon concentrations in Gaint's Hole (Bq m^{-3}).

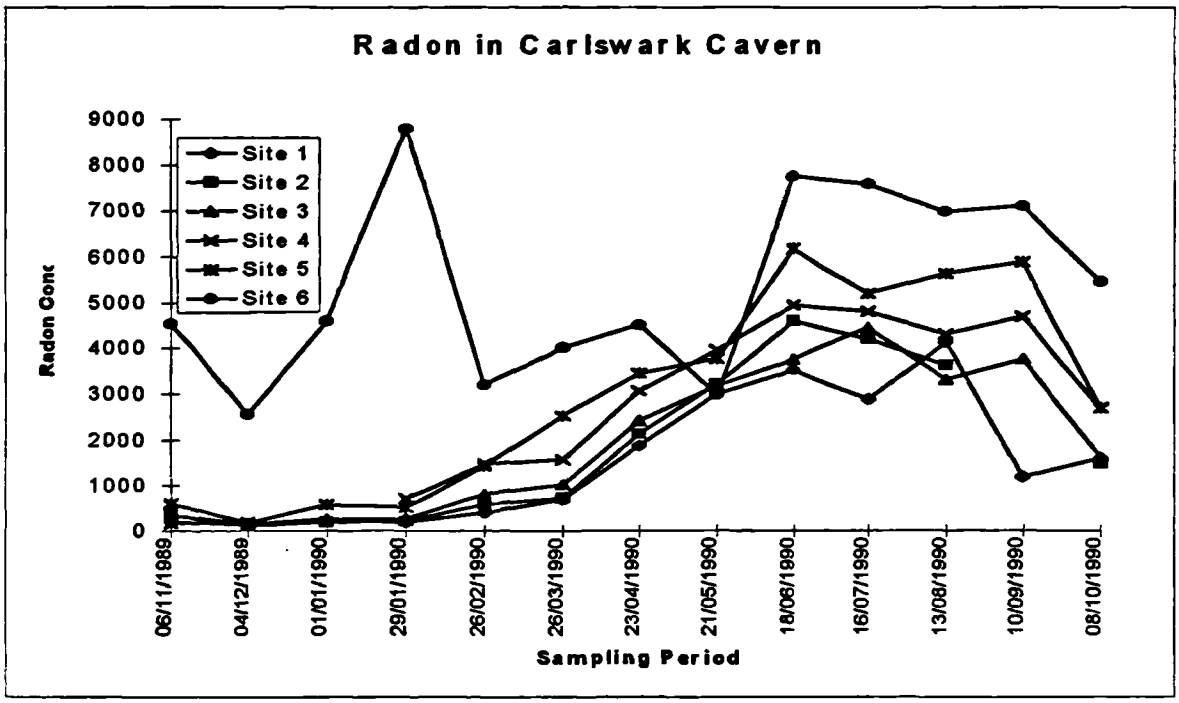

Figure 8.12 Radon concentrations in Carlswark Cavern (Bq m^{-3}).

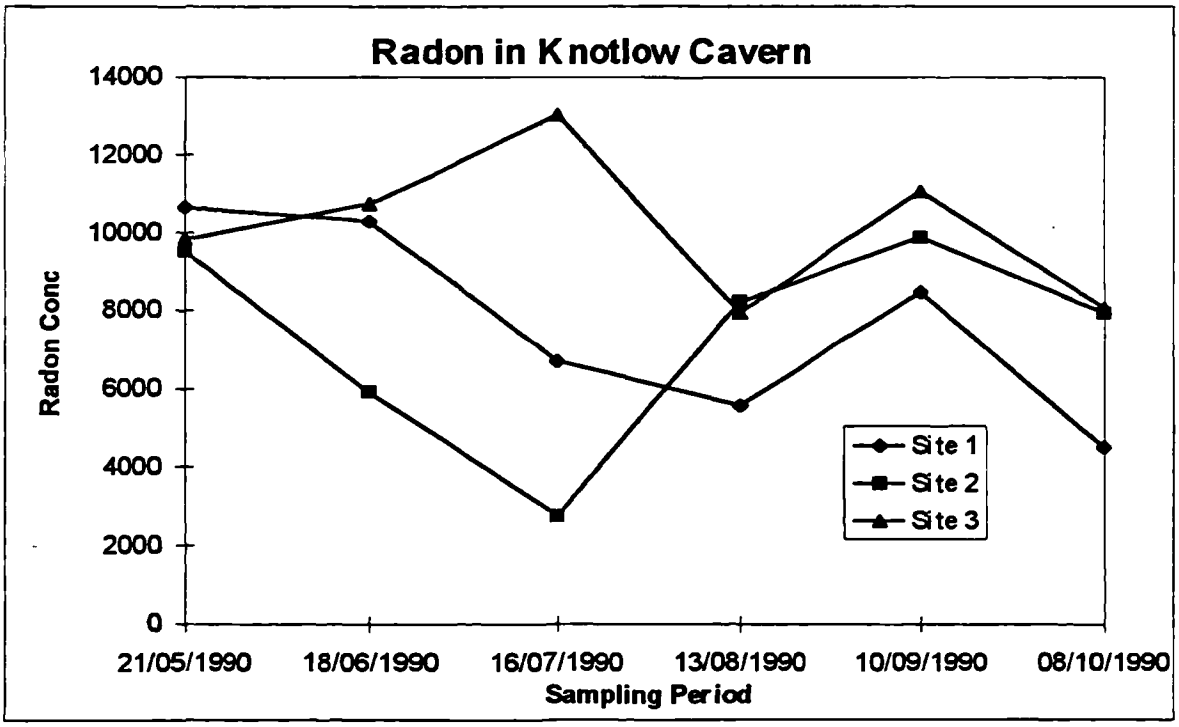

Figure 8.13 Radon concentrations in Knotlow Cavern (Bq m^{-3}).

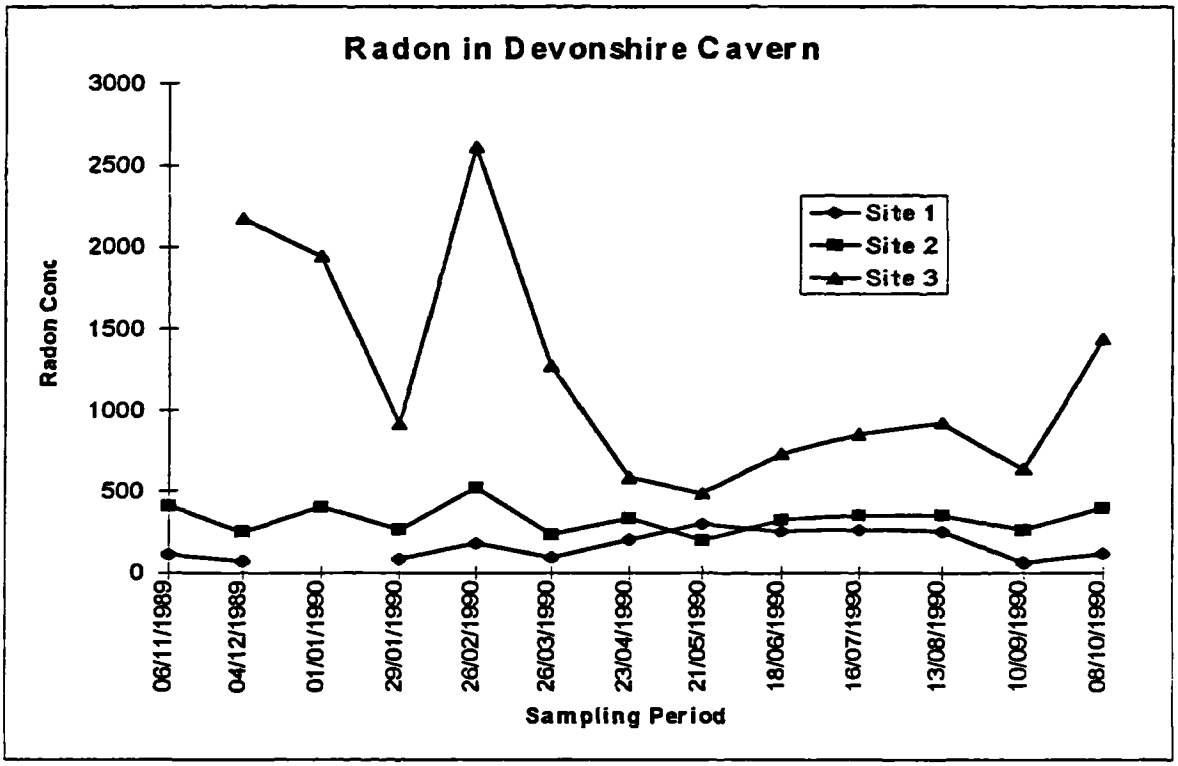

Figure 8.14 Radon concentrations in Devonshire Cavern (Bq m^{-3}).

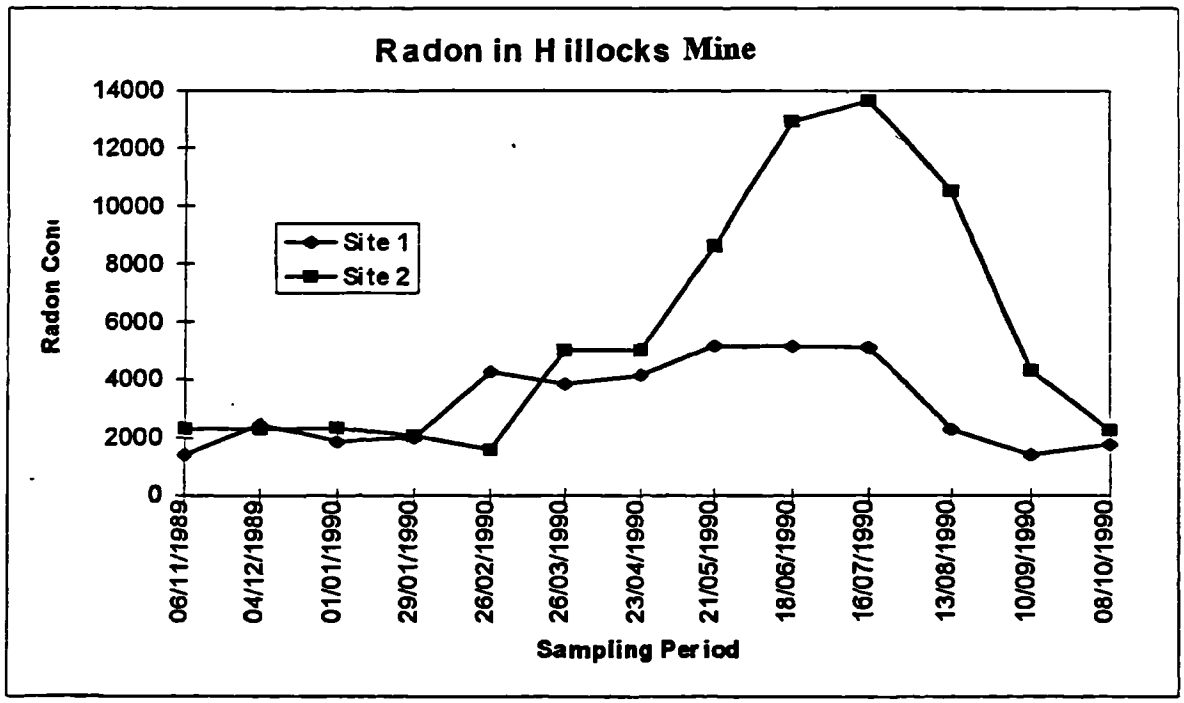

Figure 8.15 Radon Concentrations in Hillocks Mine (Bq m^{-3})

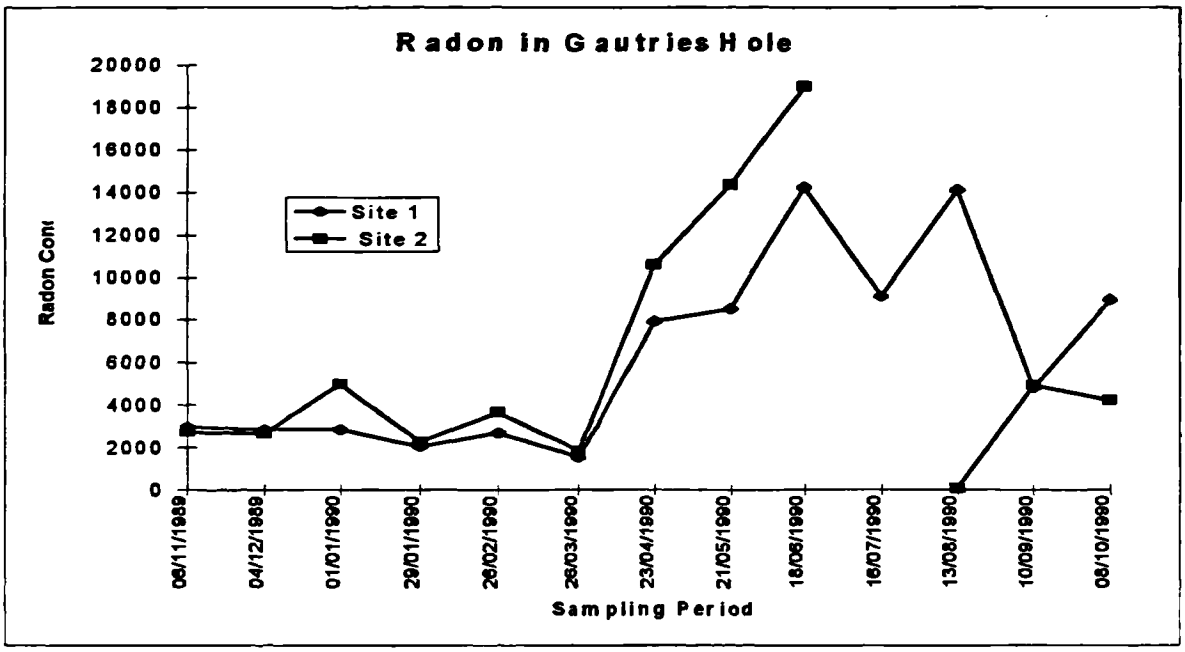

Figure 8.16 Radon concentrations in Gautries Hole (Bq m^{-3}).

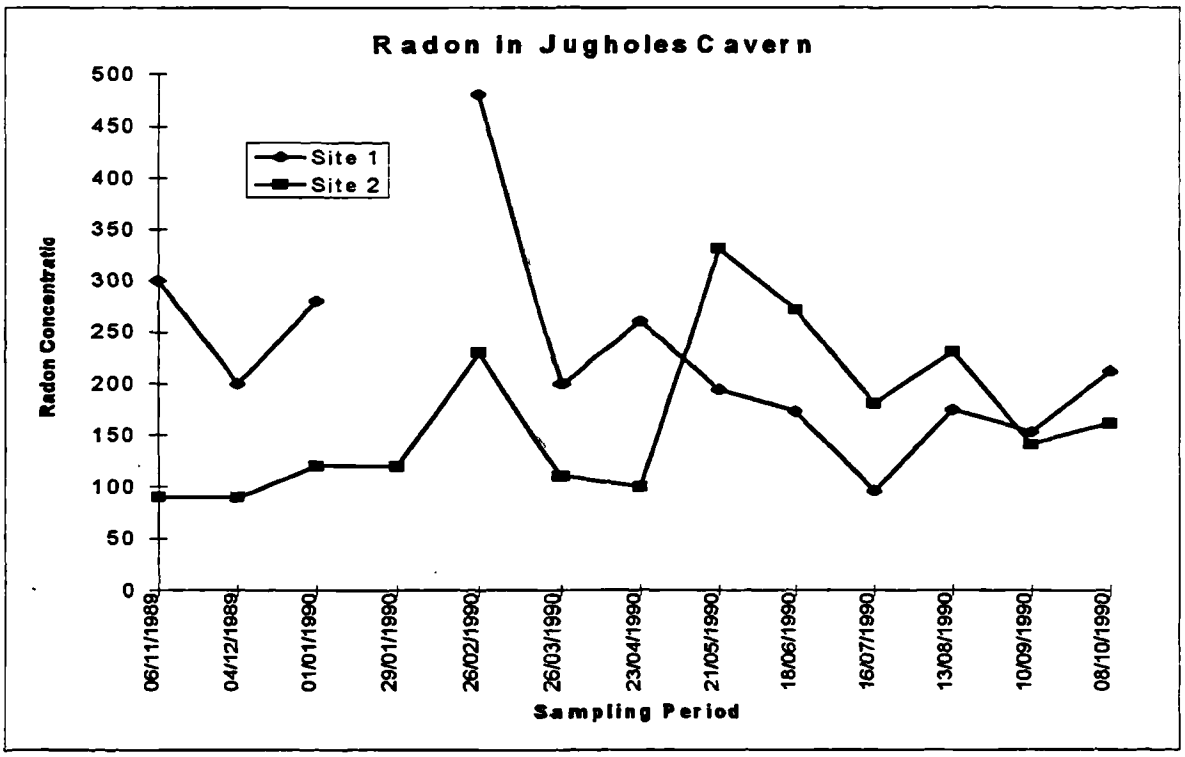

Figure 8.17 Radon concentrations in Jugholes Cavern (Bq m^{-3}).

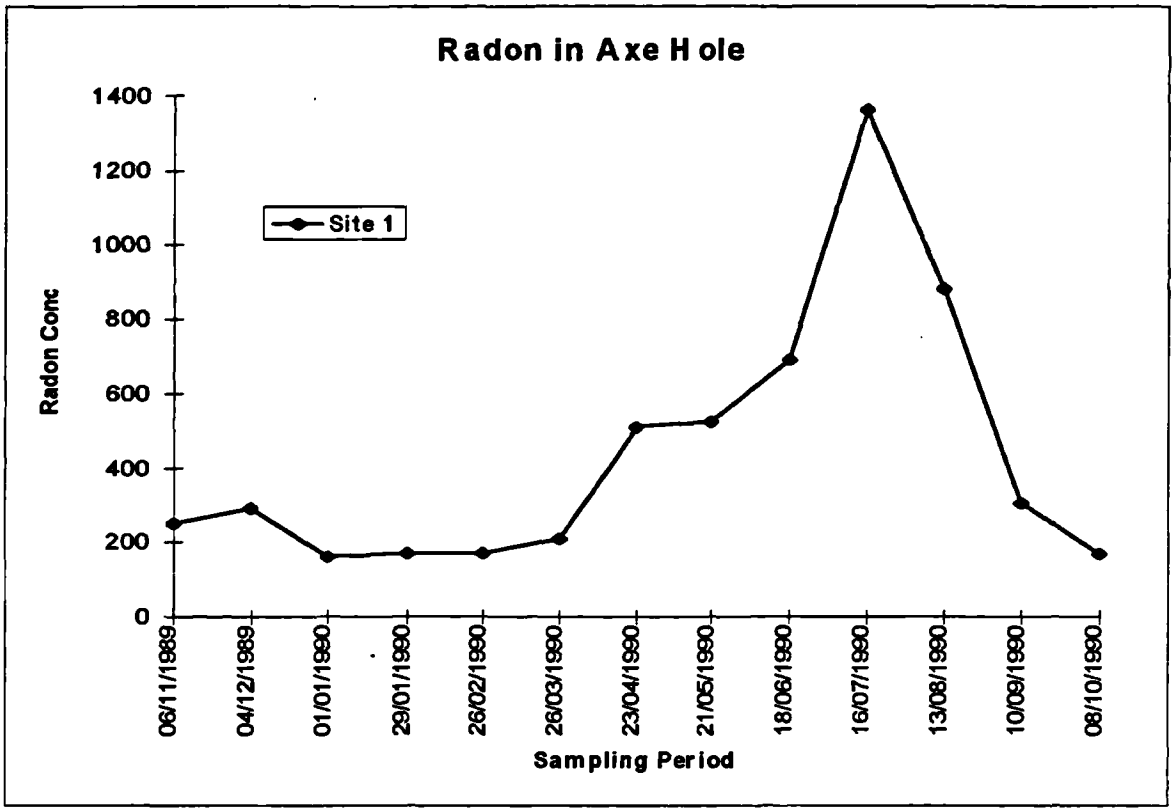

Figure 8.18 Radon concentrations in Axe Hole (Bq m^{-3}).

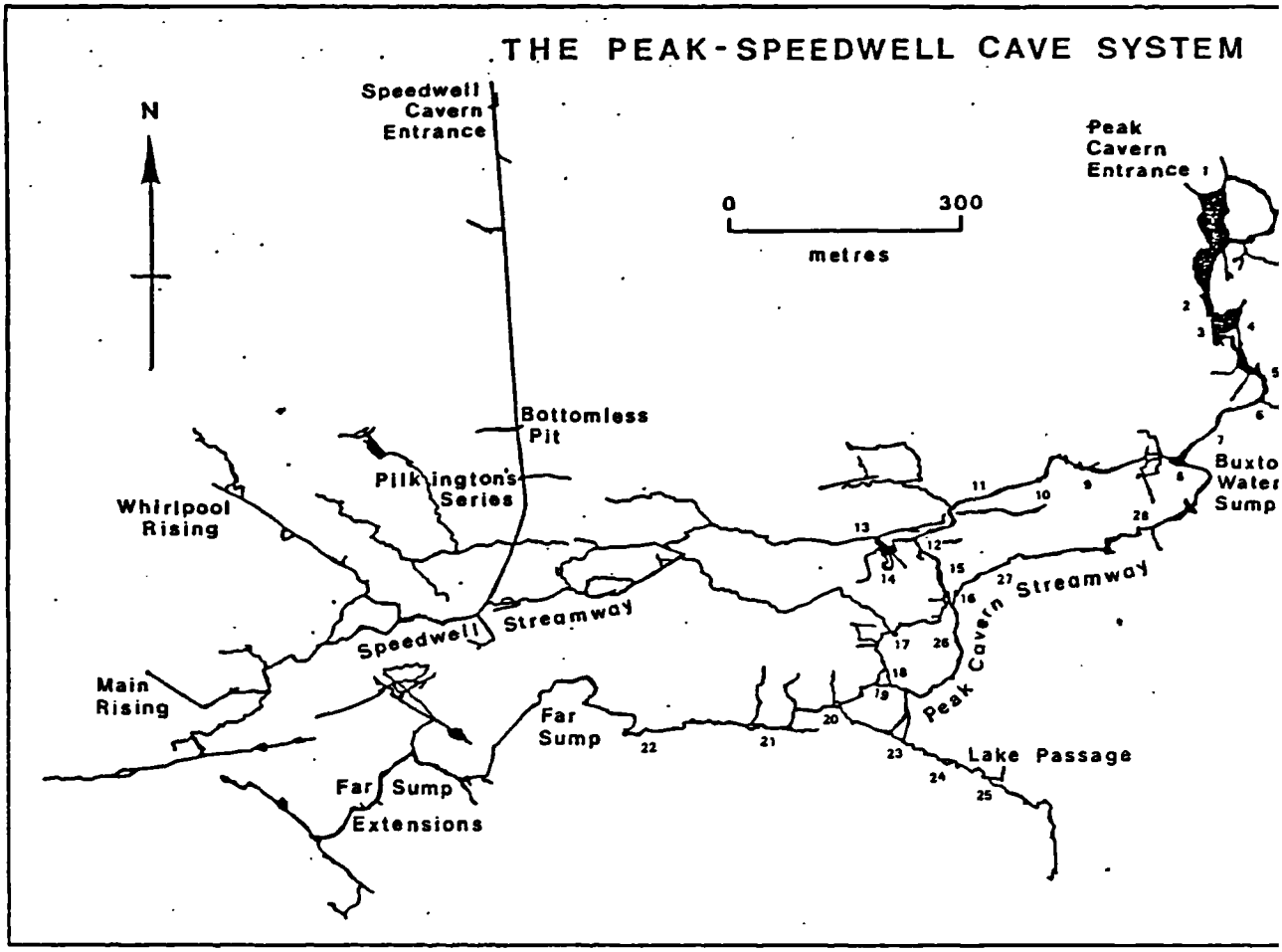

Figure 9.1 Sampling sites in Peak Cavern, see table 9.1 for site names

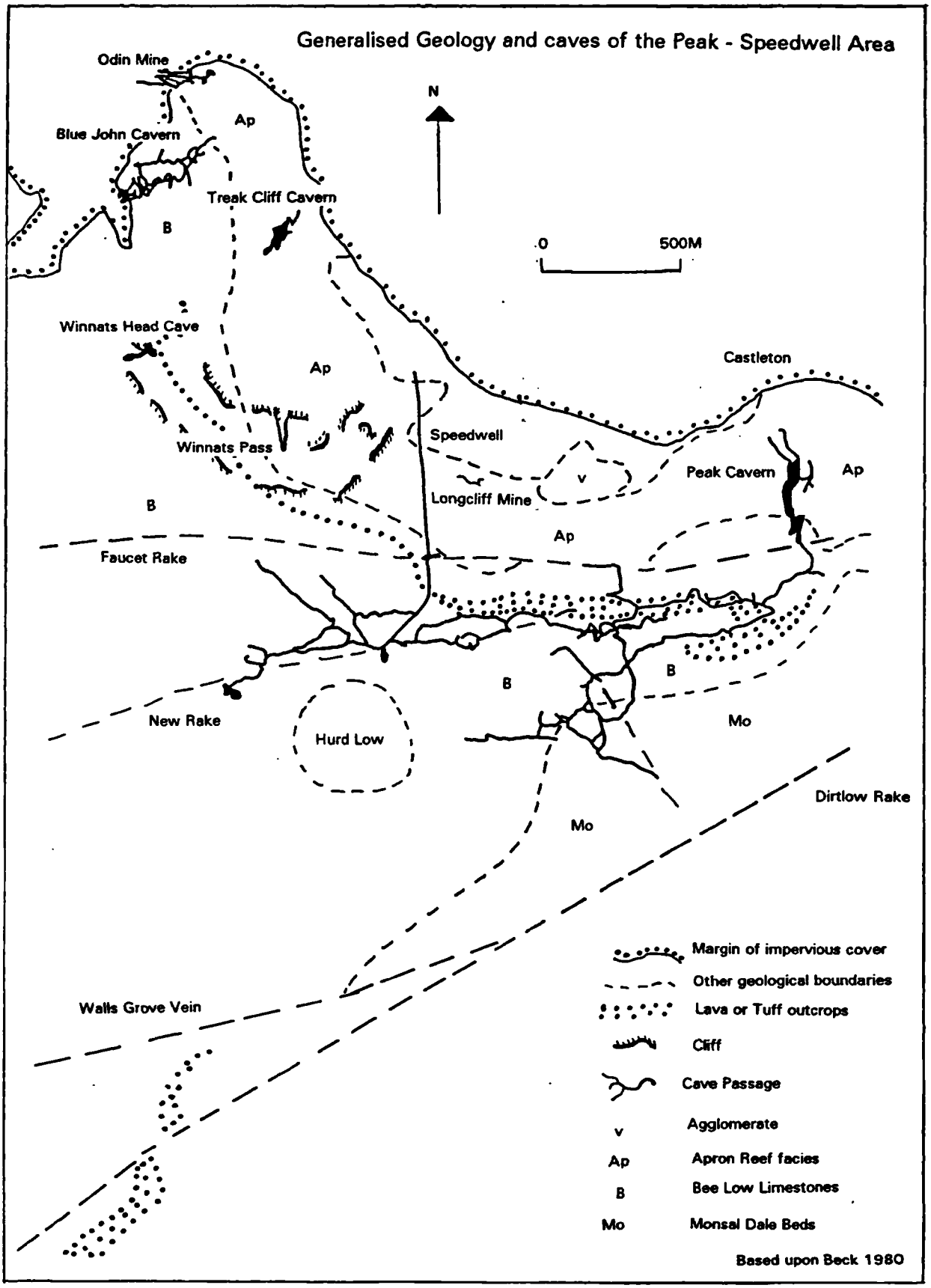

Figure 9.2 Relationship between passage development and geology in the Castleton region.

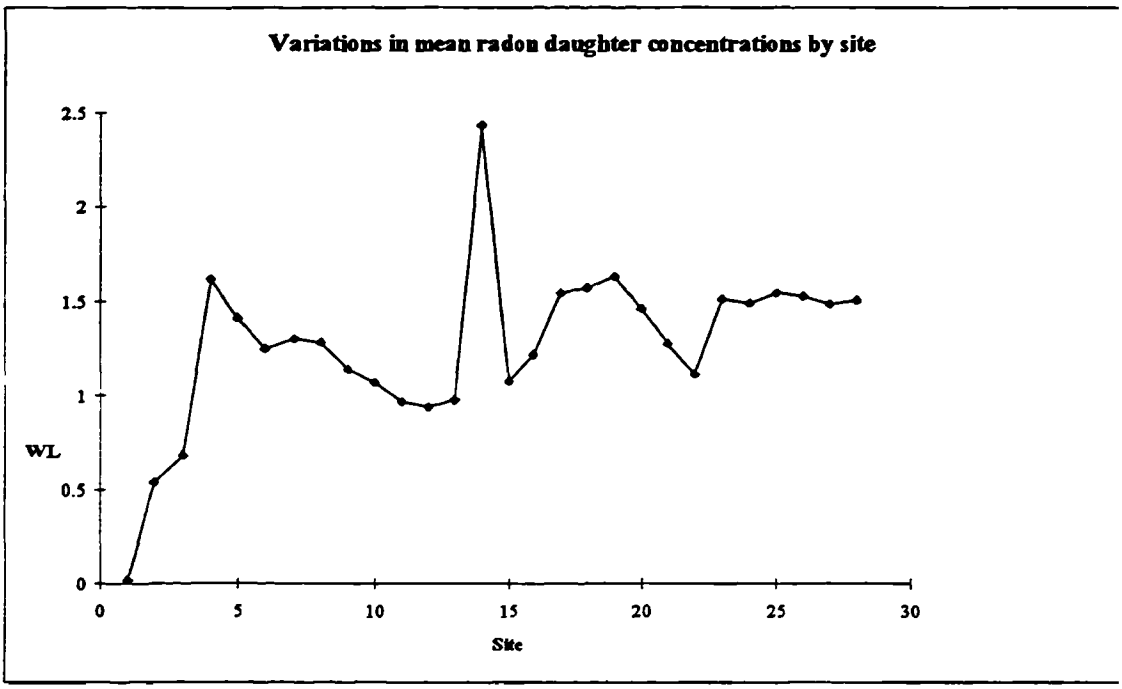

Figure 9.3 Variations in mean radon daughter concentrations by site, Peak Cavern 1991 -1992. Site locations are shown in figure 9.1 and table 9.1.

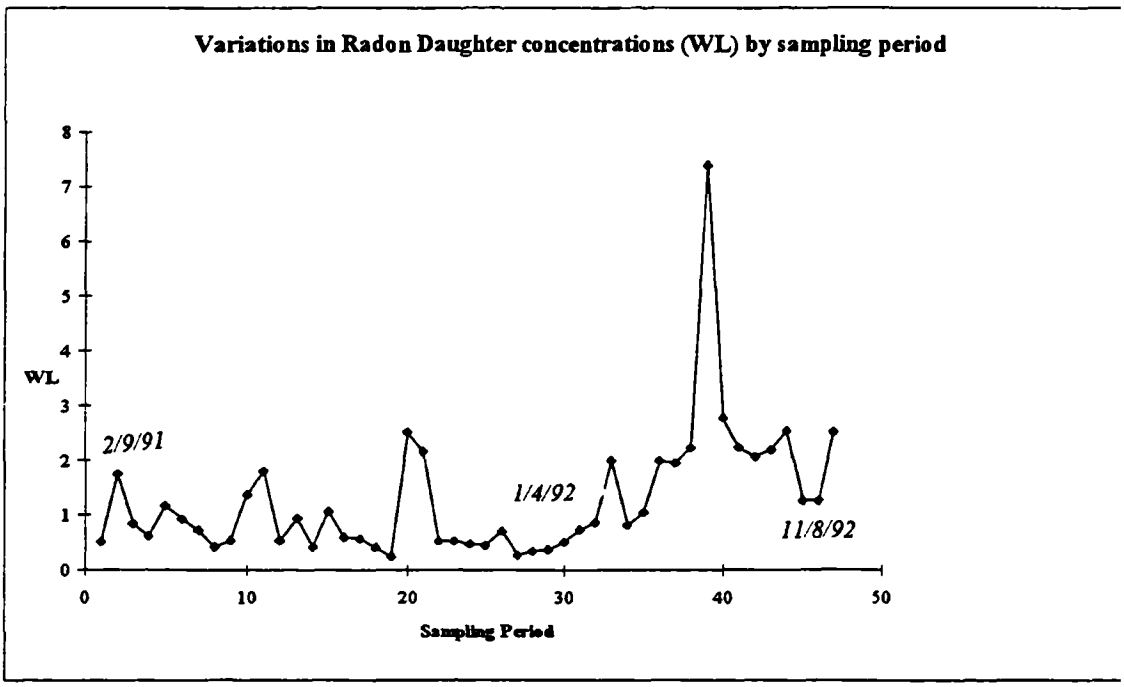

Figure 9.4 Variations in Mean Radon Daughter concentration (WL) by sampling period for all sites in Peak Cavern 1991 - 1992. For sampling dates, maximum and minims see table 9.4.

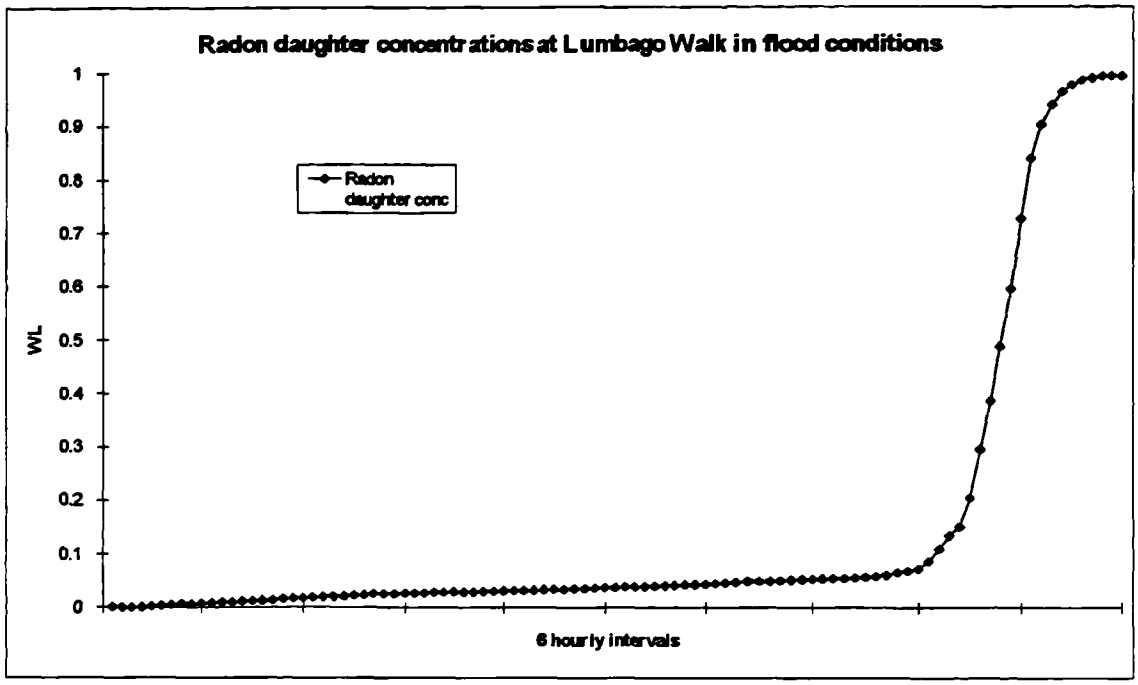

Figure 9.5 Radon daughter concentrations at Lumbago Walk in flood conditions

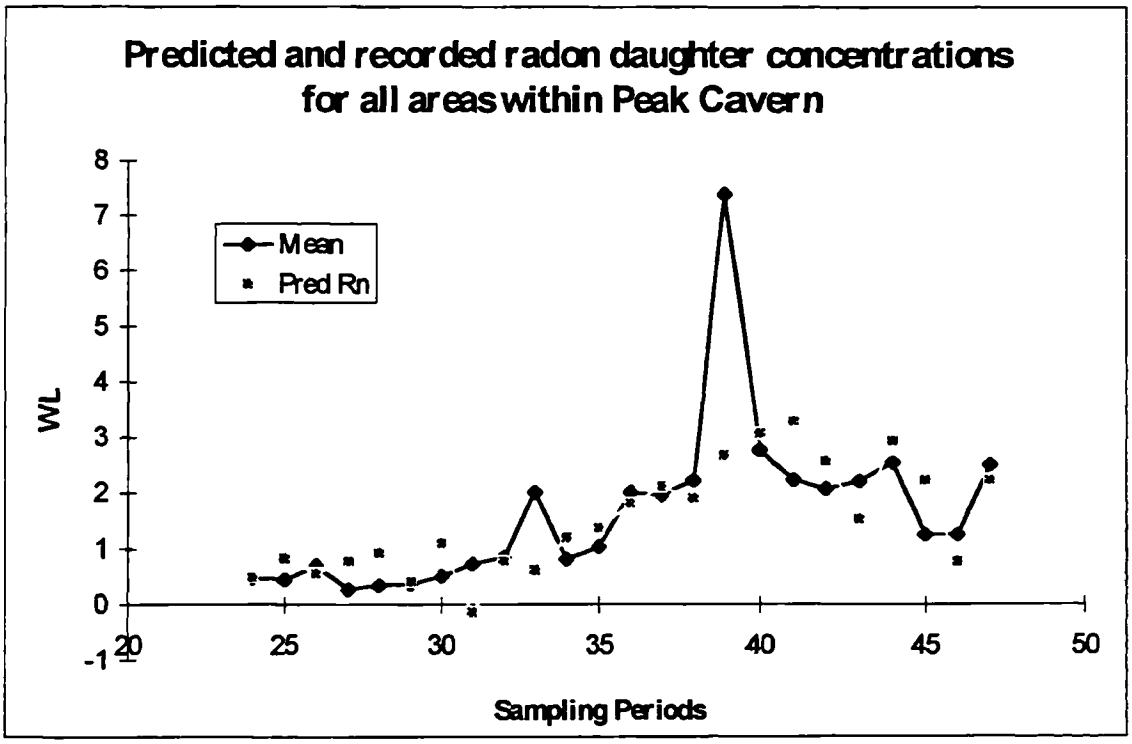

Figure 9.6. Predicted and recorded radon concentrations using a model based upon stepwise multiple regression for all sites within Peak Cavern. Developed using model outlined in table 9.8, dates of sampling periods in table 9.4.

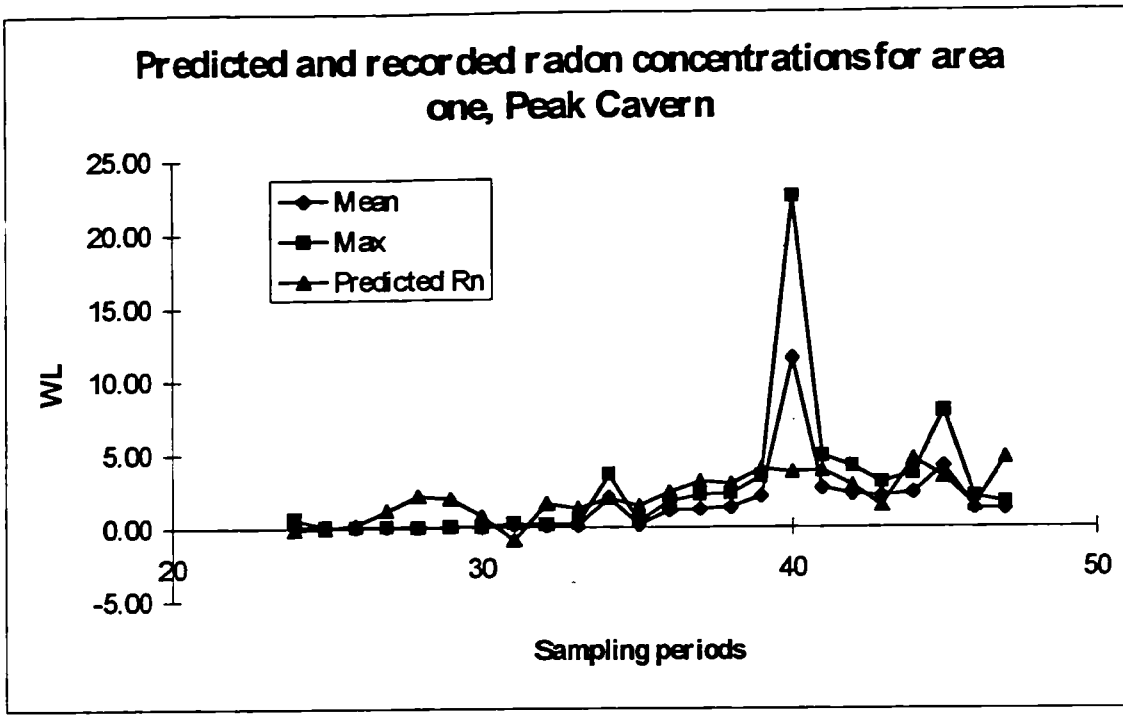

Figure 9.7 Predicted and recorded radon concentrations for area one within Peak Cavern. Developed using stepwise multiple regression outlined in table 9.9, dates of sampling periods in table 9.4.

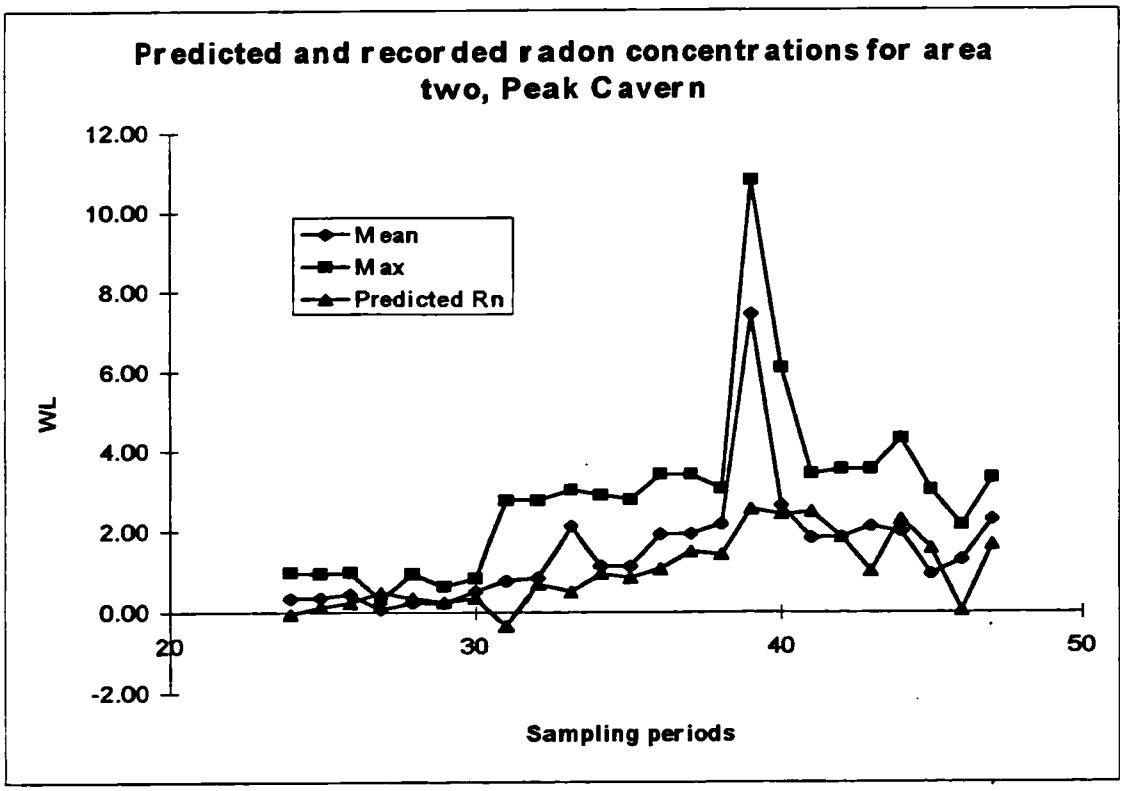

Figure 9.8 Predicted and recorded radon concentrations for area two, Peak Cavern Based upon a model developed from stepwise multiple regression outlined in table 9.10, dates of sampling periods in table 9.4.

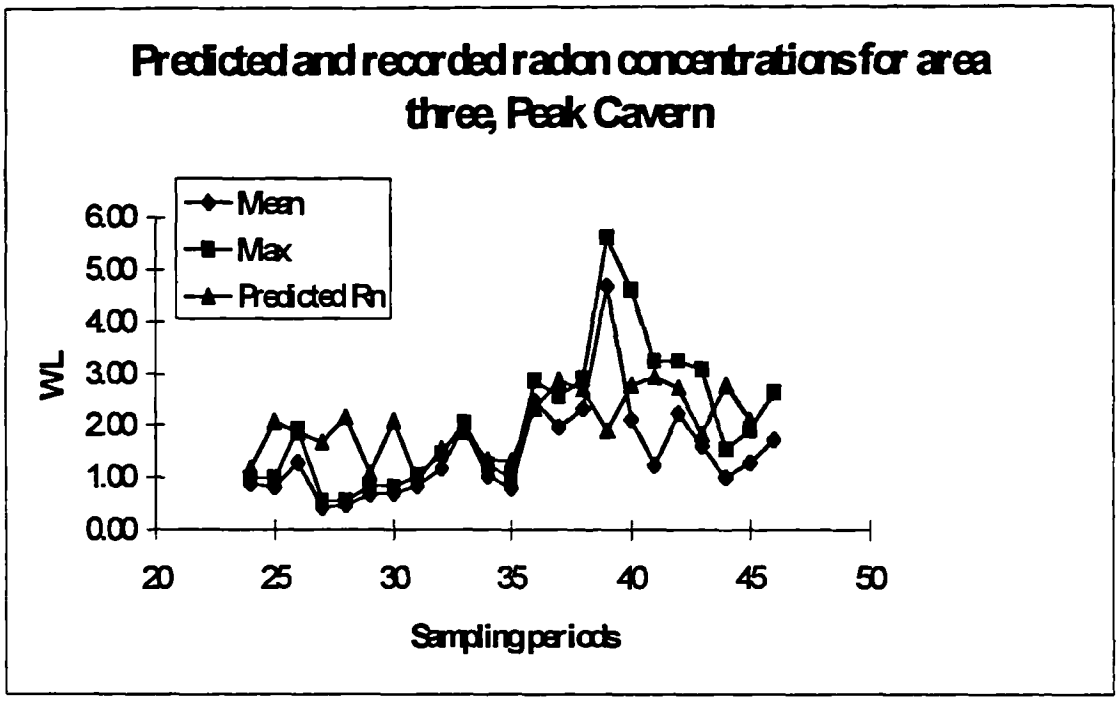

Figure 9.9 Predicted and recorded radon concentrations for area three , Peak Cavern, Based upon a model developed from stepwise multiple regression outlined in table 9.11, dates of sampling periods in table 9.4.

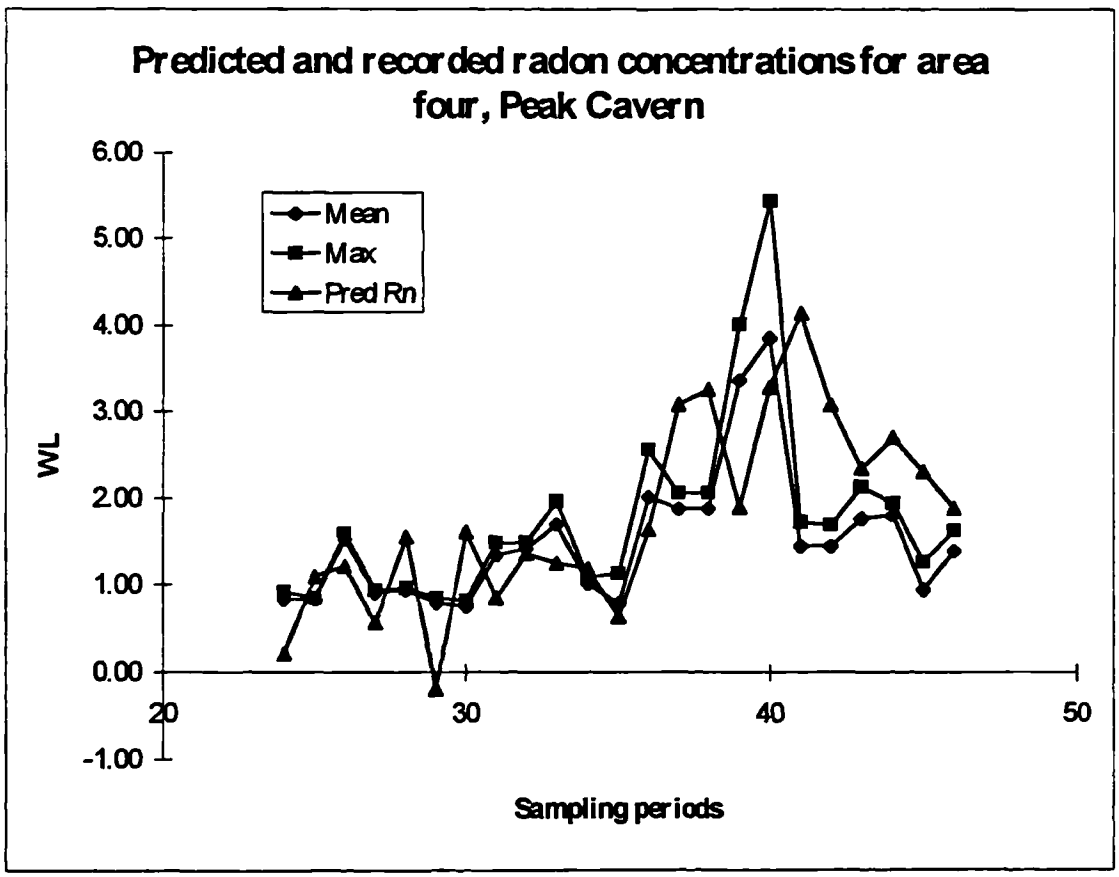

Figure 9.10 Predicted and recorded radon concentrations for area four, Peak Cavern. Based upon a model developed from stepwise multiple regression outlined in table 9.12, dates of sampling periods in table 9.4.

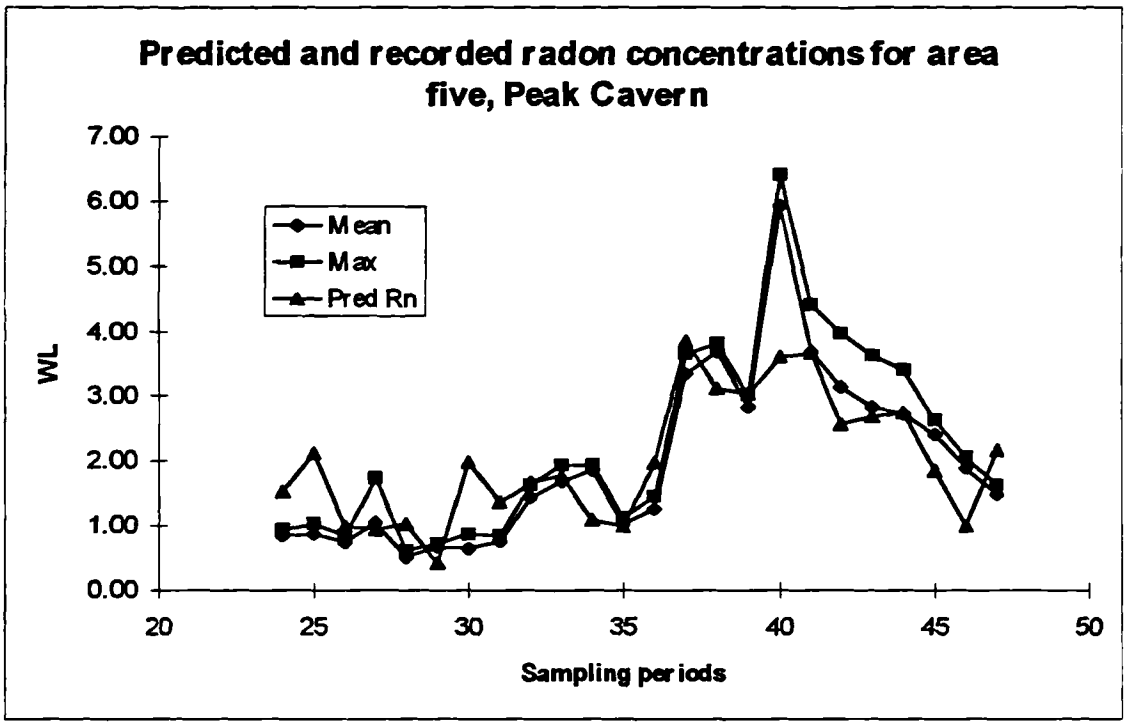

Figure 9.11 Predicted and recorded radon concentrations for area five, Peak Cavern. Based upon a model developed from stepwise multiple regression outlined in table 9.13 dates of sampling periods in table 9.4.

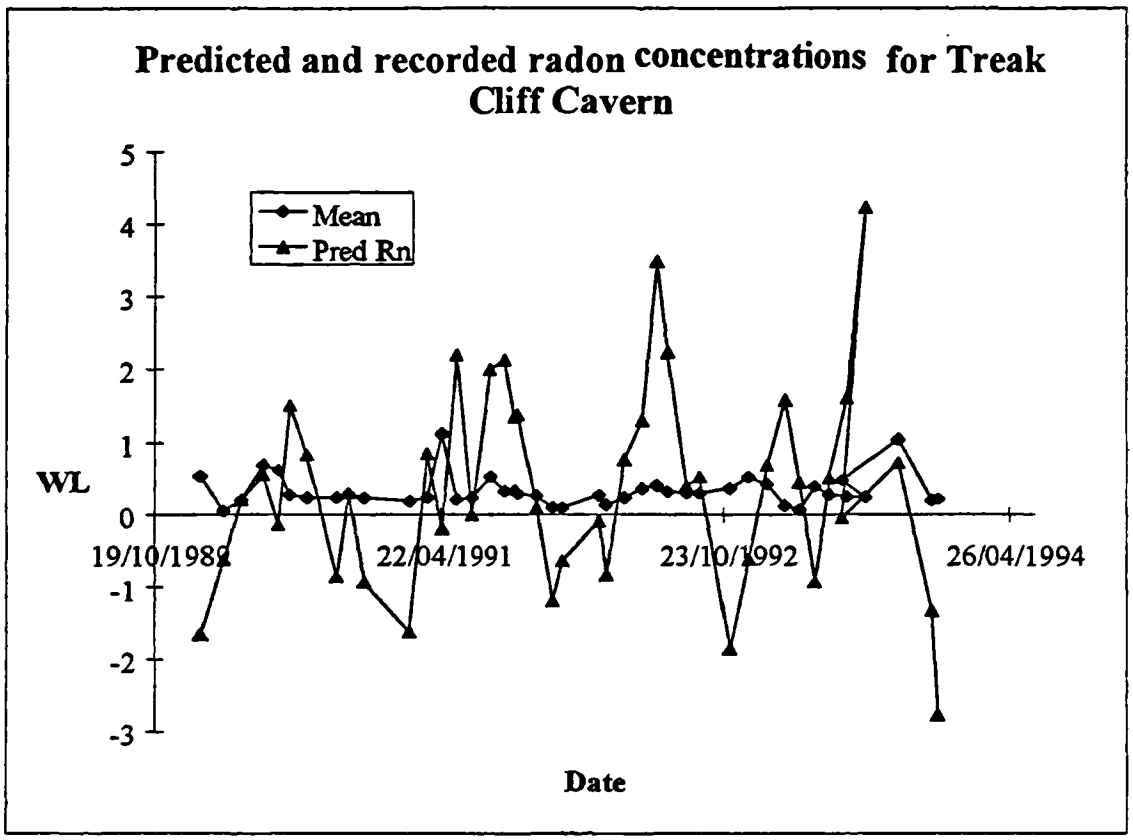

Figure 9.12 Predicted concentrations using models (outlined in table 9.8) developed for Peak Cavern compared with recorded concentrations in Treak Cliff Cavern 1989 -1994.

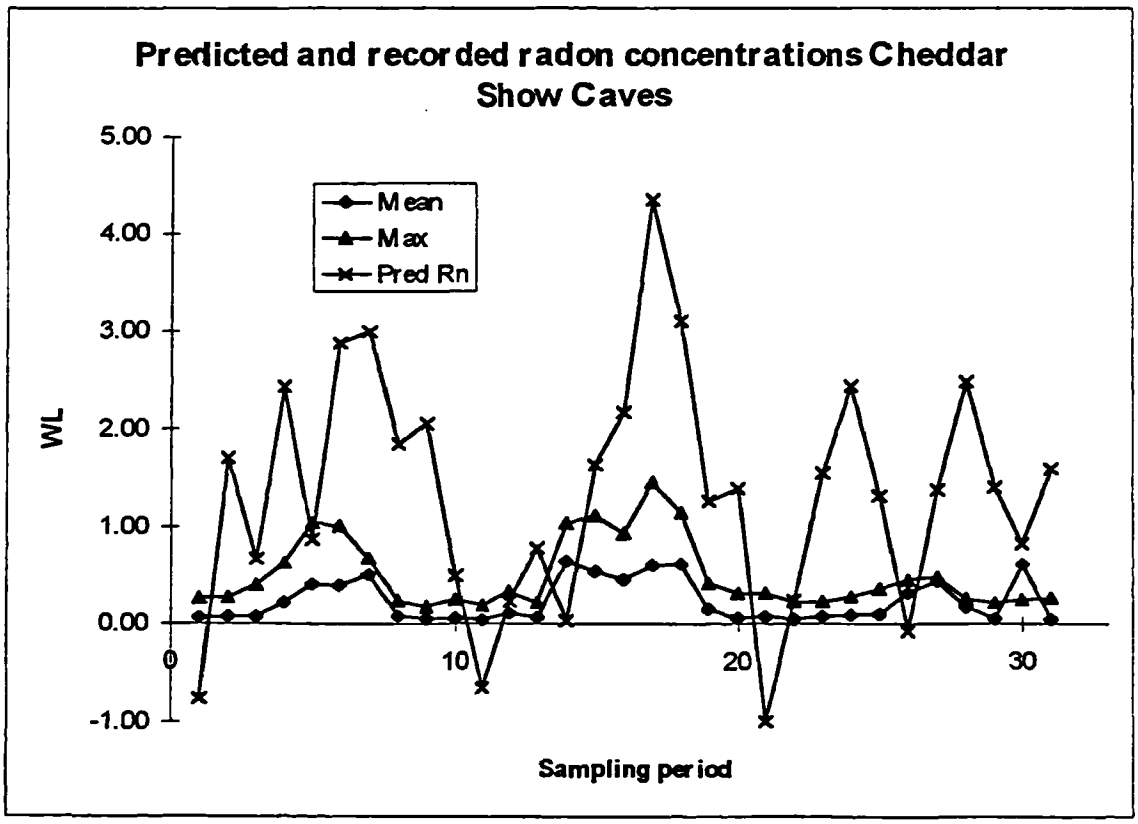

Figure 9.13 Predicted concentrations using model (outlined in table 9.8) developed for Peak Cavern compared with recorded concentrations in Cheddar Show Caves 1989 -1994

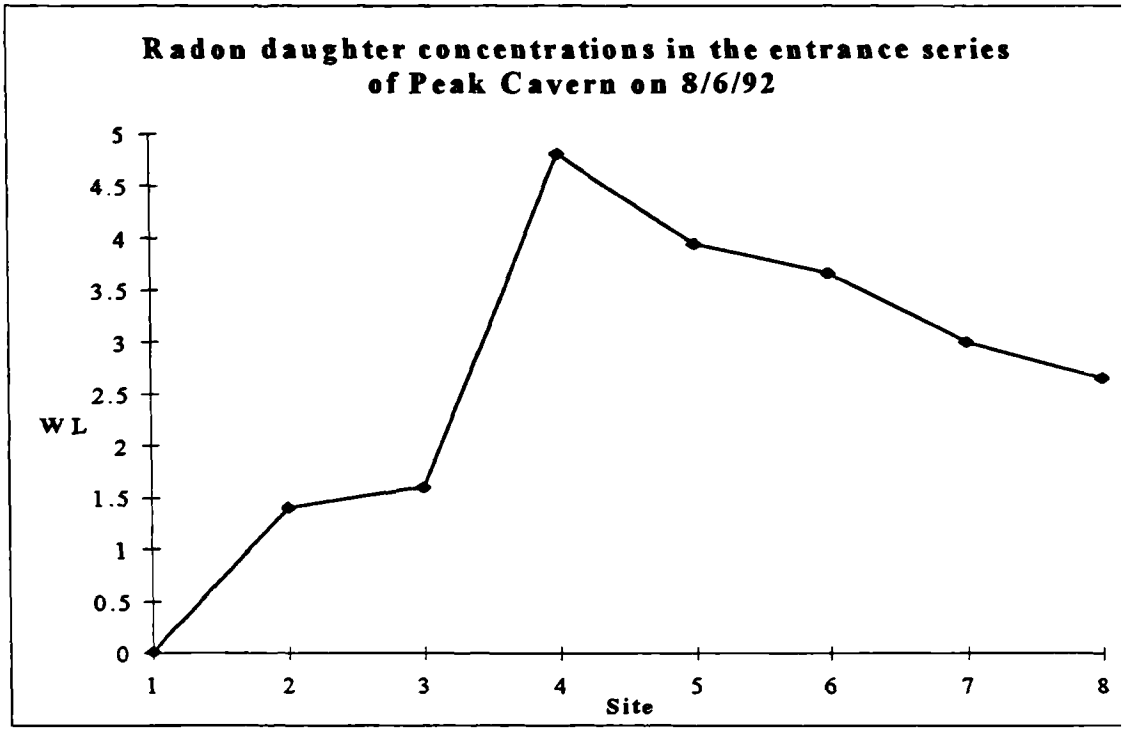

Figure 9.14 Radon daughter concentrations in the entrance series on Peak Cavern on 8/6/92. See figure 9.1 for location of sites and table 9.1 for site names.

TABLES

Table 2. 1 Approximate time for radon daughters to equilibrate with radon-222 (in minutes).

Time to Indicated Equilibrium (%)					
Daughter	Half-life	25 %	50 %	90 %	99 %
Po- 218	3 min	1	3	10	20
Pb- 214	27 min	16	31	95	180
Bi- 214	20 min	6	60	135	230
Po- 214	<< 1 sec	36	60	135	230

Table 2. 2 Radiochemical Advances in the Period 1895 - 1984, with special reference to radon isotopes. (Based upon NCRP, 1988).

1597	Agricola noted early death rates in miners from Germany and Czechoslovakia.
1896	Becquerel discovers radioactivity of uranium.
1888	The Curies and Schmidt discover radioactivity of thorium and also the elements radium and polonium.
1889	Rutherford discovers alpha and beta particles.
1899	Thomson and Rutherford demonstrate that radioactivity causes ionisation.
1899	Rutherford discovers thoron and calls it emanation
1900	Don discovers the emanation in the uranium- 238 series, which is now called radon.
1901	Rutherford and Brooks demonstrate that radon is radioactive and gaseous.
1901	Rutherford and Soddy discover transmutation.
1902	Thomson discovers radon in tap water.
1903	Rutherford and Soddy develop equations describing radioactive decay.
1904	Geisel and Debierne discover actinon.
1913	Arnstein identifies squamous cell carcinoma in autopsy of miner.
1913	Fajans discovers group displacement laws.
1914	First medical use of radon.
1925	First mention of the name radon in the literature.
1940's	Causal link shown between radon and lung cancer.
1941	National Bureau of Standards advisory committee adopts an air radon standard of 370 Bq m^{-3}.
1955	Concept of a Working Level first suggested.
1957	Development of the lucas cell.
1957	Rediscovery of radon in tap water in Maine.
1984	Discovery of high levels of radon in homes in Reading and focus of radon as an environmental hazard.

Table 3. 1 Historical summary of international cave radon research

Name	Date	Country	No of Caves	Comment
Fryer	1935	USA	-	Identification of the initial problem
Harris .	1954	USA	1	Primary work in mines but one limestone cave investigated
Geslin and Urbain	1962	France	1	Investigation of radioactivity in cave sediments
Reckmeyer	1962	USA	-	Credited with first work by Yarborough et al., 1976
Breisch	1968	USA	1	Low Rn concs recorded; no sources identified
Saumande and Renault	1971	France	6	
Ronaki	1972	Hungary	1	Suggestion that air currents could affect observed Rn concs
Prime	1974/ 1975	England	1	Measurements performed in Show Caves, high levels encountered, borehole drilled to increase air movement
Van Cleave	1975	USA	-	Outline of work to be done in NPS caves in the US
Varbanov et al.	1975	Bulgaria	9	Regional variations highlighted
Trout	1975	USA	2	Increased Rn with distance from entrance; soil gas concs higher than air ∴ sed suggested as a source; changes in external temperature could influence observed Rn concs
Wilkening and Watkins	1976	USA	1	First mathematical model developed; first published dose estimates; seasonal variations
Yarborough et al.	1976	USA	25	Relationship with microclimate, primarily airflows; little thoron; cave morphology could influence observed Rn concs; two cave types identified: RSU and USD

Ahlstrand	1976	USA	1	Government involvement
Ahlstrand and Fry	1976	USA	1	Potential risk to guides identified; equilibrium factor lower than expected; seasonal and diurnal variations
Ikeya	1976	Japan	1	First measurements of Rn dose in Japanese caves
McFarlane	1976	UK	1	Work in mines within limestone and other strata
Oldham	1976	UK		Review of work in US
Aley	1976	USA		Used Yarborough's data and predicted little/ no risk to cave users
Yarborough	1976	USA	31	Developed management programs for Show Caves; proposed the Kusnetz method as a standard; seasonal and diurnal variations; two cave morphologies influence observed concs; don't use cave air for air conditioning
Knutson	1977	USA	-	Concentrated on health aspects, little risk to cave users
Yarborough and Ahlstrand	1977	USA	11	Proposed management of caves on three tier system below 0.03 WL; 0.03 -0.1 WL; and above 0.1 WL
Beckman	1977	USA	-	Determined health risk for Guides
Ahlstrand and Fry	1977	USA	1	Identified seasonal variations influenced by external temperatures; diurnal variations dependent upon pressure changes
Yarborough	1977	USA	4	Used Newton's law to demonstrate that external temperatures control airflows in caves; seasonal variations; cave morphology (RSU and USD)
Beckman	1978	USA	-	Concentrated on health risk
Gascoyne	1978	International	-	Basic review of published work
Ciezkowski	1978	Poland	11	Initial findings
Young	1979	UK	-	Reviewed procedure to ensure

				radiation doses are kept as low as reasonably possible
Miki and Ikeya	1980	Japan	1	Seasonal variations; vertical difference; Rn actively accumulating in caves
Ahlstrand	1980	USA	2	Observed concs affected by temperature and pressure
Aley	1980	USA	-	Review of USA data; no threat to cavers' health
Cigna and Clemente	1981	Italy	1	First work in Italy; risk to Show Cave guides
Gamble	1981	South Africa	12	First work in South Africa; effects of cave morphology on concs, two types: sack and transit; seasonal variations
Yarborough	1981	USA	9	Relation between Rn and airflows; airflow related to external temperature, pressure and cave morphology; Rn can be used to study cave airflows
Friederich	1981	UK	-	Studied percolation waters but proposed high levels in caves
De Bellard-Pietri	1981	Venezuela	2	Gamma readings only done so far. Concs in caves higher than outside and homes in Caracus City
Carson	1981	USA	1	Winter/ Summer concs relatively stable either low or high respectively; spring and autumn highly variable; pressure as important control; risk to guides
Yarborough	1982	USA	10	Two types of cave RSU and USD; seasonal and diurnal variation; recommends no smoking in caves; all workers should have health checks and then annual checks
Atkinson et al.	1983	Canada	1	First work in Canada; results in track densities not Rn concs; Rn controlled by dilution not redistribution; Rn concs influenced by air direction due to

				chimney effect
Fernanderz et al.	1984	Spain	1	First work in Spain; dose too high to be acceptable for long term guides ∴ forced ventilation introduced
McFarlane	1984	UK	1	Results from mine in grits and shales; radon concs attributed to bituminous limestones
Gadoros	1985	Hungary	3	Rn concs can be used to determine cave microclimate; activity in air is attributable to Ur content of limestone
Amano et al.	1985	Japan	1	Man made cave in dolerite; uniform exhalation rate; temp and pressure dependence of Rn concs
Cigna	1986	Italy	1	12 month averages ,error in results due to non equilibrium; no risk to recreational caver
Papastefanou et al.	1986	Greece	1	First work in Greece. High concentrations recorded (60,000 Bq m^{-3})
Kobal et al.	1987	Yugoslavia	11	First published work from Yugoslavia in international journals; no seasonal variation apparent; equilibrium factor large range; high doses could be received
Cigna	1987	Italy		Review paper; no risk to recreational cavers
Clark	1987	USA		Basic; no scientific review; no risk to recreational cavers
Pitts	1987	USA		Basic; no scientific review; risk to recreational cavers
Mutter	1987	UK	1	Spatial and temporal variations; increase in concs with distance from the entrance; increased passage size decreases observed Rn conc
Kobal et al.	1988	Yugoslavia	1	Seasonal variations apparent; health risk to those exposed for longer periods of time

Saumande et al.	1988	France	18	Concs recorded in pits and caves; clay sediments could be source of elevated concs
Middleton	1988	UK	1	Airflow, not depth, influences Rn conc; diurnal variations relate to temp; decreasing pressure increases Rn emanation; no sources identified.
Stelcl and Plachy	1988	Hungary	3	Seasonal and diurnal variations
Quinn	1988	USA	1	Tried to predict Rn concs from climatic data. Able to use previous days' mean and min temperature to predict if concs are likely to be above 0.03 WL
Geczy et al.	1989	Hungary	1	Used Rn to determined cave microclimate; seasonal and temporal variations; long term changes related to changes in cave volume due to inflilling of micro fractures; hysterisis can be seen in Rn concs
Somogyi et al.	1989	Hungary	16	Rn concs solely dependent upon emanation from limestone; short term variations related to external climate; long term variations related to sunspot activity
Lenart et al.	1989	Hungary	5	Yearly and seasonal variations; increased concentrations with depth; 3 radon zones related to cave microclimate; sediments and water proposed as sources
Piller and Surbeck	1989	Switzerland	2	Link established between cave Rn concs and concs observed in homes; increased concs with depth
Gunn et al. (a)	1989	UK	8	Spatial and temporal variations; health risk evident
Hakl et al.	1989	Hungary	1	Results from Karstic well demonstrate that water could be a source of radon in caves
Webster and Crawford	1989	USA	3	Rn from caves could influence Rn in homes
Gunn et al.(b)	1989	UK	8	Basic summary for British cavers;

				spatial and temporal variations highlighted; health risk identified
Surbeck and Medici	1990	Switzerland	1	Limestone not the only source of radon in caves; soil and deep-seated sources proposed
Burian and Stelcl	1990	Czechoslovakia	4	Seasonal and yearly variations; equilibrium factor relates to age of air; homogeneously mixed within a passage; little radon-220 (thoron)
Navratil and Stelcl	1990	Czechoslovakia	5	Main source of Rn from fluvial deposits; seasonal and diurnal variations; Pressure affects observed concs
Ball	1990	UK	-	Uses Gunn's data to predict health risk for cave users in S.Wales. Little risk over a cavers' lifetime
Williamson	1990	UK	6	Seasonal, spatial and temporal variations; pressure effects
Gunn	1990	UK	8	Spatial and temporal variations highlighted; health risk identified
Gunn and Hyland	1991	UK	9	Spatial, temporal and seasonal variations identified; health risk outlined; management plan proposed to reduce instructors' dose
Gunn et al.	1991	UK	8	Data from Wild and Show Caves implies considerable health risk to cave users
Taylor	1991	UK	1	Spatial and temporal variations; health and legal aspects outlined
Hakl et al.	1991	Hungary	13	Seasonal variations; water and sediments possible sources of Rn to caves
Quinn	1991	USA	2	Relation with cave morphology; airflow control on Rn concs
Aley	1991	USA	-	Open discussions and review of radon in caves
Workman	1991	UK	1	Spatial and temporal variations; health risk

Webster	1991	USA	4	Rn from caves can influence homes; spatial and temporal variations
Solomon et al.	1991	Austria	1	Seasonal and diurnal variations in Rn concs; health risk to guides
Stelcl	1991	Czechoslovakia	12	Health risks to long term guides, no risk of average guide or visitor
Eheman et al.	1991	USA	11	Health risks outlined and discussed
Cunningham and LaRock	1991	USA	1	Used Rn to determine cave microclimate
Middleton et al.	1991	UK	1	Airflow not depth influences Rn conc; diurnal variations relate to temp; decreasing pressure increases Rn emanation; no sources identified.
Prime and O'Hara.	1991	UK	1	Relationship with internal air movements, external wind direction and speed
Reaich and Kerr	1991	UK	1	High doses could be received by recreational cavers
Gunn	1991	Russia	11	First published work from USSR; generally concs low but could result in a significant dose
Massen et al.	1992	Luxembourg	1	Integrated study of Radon and cave microclimate
Workman	1992	UK	1	Identified seasonal variations
Lyons et al.	1992	Australia	20	Only limited samples from each cave
Klimchouk	1992	Russia	7	Limited samples

Gammage et al.	1992	USA	2	Rn transported from caves to houses due to secondary permeability developed in Karst regions
Kies and Masson	1993	Luxembourg	1	Seasonal variations; correlation with carbon dioxide; increase with distance from the entrance
Roberts	1993	UK	5	Rn concs affected by pressure but not temperature
Hyland and Gunn	1993	GB	42	Largest single project to-date identifies spatial and temporal variations on national and regional scales

Table 3. 2 Summary of international radon and radon daughter concentrations within limestone caves

Name	Date	Country	Num of Caves	Range (either Bqm^{-3} or WL)	Mean	Num of results
Fryer	1935	USA				
Harris	1954	USA	1	20 - 150*		
Breisch	1968	USA	1	37*	37*	2
Ronaki	1972	Hungary	1	1 - 111*	55*	15
Prime	1974/ 1975	England	1	Up to 11 WL		
Varbanov	1975	Bulgaria	9	4 - 126*	11*	9
Trout	1975	USA	2	0 - 1,905*	888*	30
Wilkening and Watkins	1976	USA	1	481 - 2,035*	980*+	12
Yarborough et al.	1976	USA	25	0.1 - 3.45 WL	0.496 WL+	
Ahlstrand and Fry	1976	USA	1	0.09 - 0.50 WL*		860
Yarborough	1976	USA	31	0.01 - 3.28 WL	0.547 WL+	
Yarborough and Ahlstrand	1977	USA	11	0.003 - 13.37 WL	0.333 WL+	
Ahlstrand and Fry	1977	USA	1	0.15 - 1.03 WL	0.385 WL+	
Yarborough	1977	USA	4	0.39 - 1.02 WL	0.687 WL+	
Ciezhouski	1978	Poland	11	740 - 5,143*	3,378*	22
Miki and Ikeya	1980	Japan	1	70 - 20,461*	5,476*	5
Ahlstrand	1980	USA	2	740 - 2,350*	1,480*	
Cigna and Clemente	1981	Italy	1	518 - 740*	437*	17
Gamble	1981	South Africa	12	0.003 - 0.62 WL	0.057 WL	63
Yarborough	1981	USA	9	0.27 - 1.2 WL	0.53 WL	

Carson	1981	USA	1	Mx 1.35 WL	0.749 WL	
Yarborough	1982	USA	10		1.04 WL+	
Fernander et al.	1984	Spain	1	1,110 - 5,920 *		
McFarlane	1984	UK	1	0.001 - 1.18 WL	0.242 WL	
Gadoros	1985	Hungary	3	Mx 12,000		
Amano et al.	1985	Japan	1	210 - 370	325	4
Cigna	1986	Italy	5	100 - 346	226	23
Papastefanou et al.	1986	Greece	1	185 - 88,060	25,340	6
Kobal et al.	1987	Yugoslavia	11	75 - 5,920	778	
Mutter	1987	UK	1			
Kobal et al.	1988	Yugoslavia	1	65 - 22,165	2,718	
Saumande et al.	1988	France	18	127 -10,876	2,450	
Middleton	1988	UK	1	2.13 - 22.16 WL	9.3 WL	70
Stelcl and Plachy	1988	Hungary	3	340 - 1,245	798	
Somogyi et al.	1989	Hungary	16	500 - 14,000	3,300	
Lenart et al.	1989	Hungary	5	100 - 12,000	706	
Piller and Surbeck	1989	Switzerland	2	2000 - 4,700		
Gunn et al.	1989	UK	8	0.01 - 24.3 WL	2.59 WL	
Seymore et al.	1989	USA	1	0.008 - 0.25 WL	1.58 WL	
Webster and Crawford	1989	USA	3	2.85 - 5.4 WL	4.4 WL	
Gunn et al.	1989	UK	8	2.1 - 24.3 WL		
Burian and Stelcl	1990	Czechoslovakia	4	0.1 - 21.7 WL	1.38 WL	156

Navratil and Stelcl	1990	Czechoslovakia	5	500 - 4,500		15
Williamson	1990	UK	6	0.01 - 41.9 WL	2.14 WL	86
Gunn	1990	UK	8	0.02 - 1.41 WL	0.197 WL	125
Gunn and Hyland	1991	UK	9	0.002 - 4.25 WL	0.557 WL	319
Gunn et al.	1991	UK	8	0.01 - 42 WL		
Taylor	1991	UK	1	70 - 252	104	14
Hakl et al.	1991	Hungary	13	200 - 14,000		
Jovanovic et al.	1991	Slovenia	1	700 - 7,500	2,640	
Quinn	1991	USA	2	0.05 - 0.13 WL	0.074 WL	15
Workman	1991	UK	1	422 - 1,475	942	4
Webster	1991	USA	4	0.01 - 5.36 WL	1.78 WL	21
Solomon et al.	1991	Austria	1	83 - 1,370	487	23
Stelcl	1991	Czechoslovakia	12	0.06 - 0.29 WL	0.139 WL	
Eheman et al.	1991	USA	11	370 - 9,460	2,580	335
Cunningham and LaRock	1991	USA	1	185 - 3,515	1,007	31
Middleton et al.	1991	UK	1	2.13 - 22.16 WL	9.3 WL	70
Prime et al.	1991	UK	1	0.01 - 0.73 WL	0.46 WL	200
Reaich and Kerr	1991	UK	1	12,000 - 17,000		
Gunn	1991	Russia	11	0.02 - 1.73 WL	0.332 WL	
Massen et al.	1992	Luxembourg	1	2,500 -6,200		
Workman	1992	UK	1	518 - 1,009	865	6
Lyons et al.	1992	Australia	20	0.01 - 0.90 WL	0.175 WL	71

Klimchouk	1992	Russia	7	156 - 68,110		
Gammage et al.	1992	USA	2	100 - 2,200		
Massen et al.	1993	Luxembourg	1	500 - 8,000		
Hyland and Gunn[a]	1993	UK	42	9 - 46,080	2,907	703
Roberts	1993	UK	5	0.02 - 1.16 WL	0.49 WL	14
Hyland and Gunn[b]	1993	GB	42	9 - 46,080	2,825	819
* Original concentrations reported in pCi l^{-1}, converted to Bq m^{-3} by multiplying by 37						
+ Mean concentration derived from summary data contained within paper; means based upon reported Maximums and Minimums						

Table 5.1 Kusnetz factors based upon delay after sampling

Delay after Sampling	Kusnetz Factor
40	150
45	140
50	130
55	120
60	110
65	100
70	90
75	82.5
80	75
85	67.5
90	60

Based upon Kusnetz, 1956

Table 6.1 Distribution of cave entrances and passage length within the seven major caving regions of Great Britain. (Data from Hardwick, 1993)

REGION	Number of caves	% of number	Cave length (m)	% of length
North Pennines	1,886	63.0	362,783	52.8
South Wales	299	10.8	177,500	25.8
Mendip Hills	233	7.7	54,926	8.0
Peak District	220	7.2	48,662	7.1
Scotland	190	4.2	12,787	1.9
Devon	133	4.4	17,942	2.6
North Wales	77	2.6	11,875	1.7
Other	4	0.1	906	0.1
TOTAL	3,042	100.0	687,381	100.0

Table 6.2 Mean passage length (km/ent) and total number of cave entrances for the four major caving regions. Data from Hardwick, 1993

REGION	Mean passage length (km/ent)	%	Number of cave entrances	%
North Pennines	0.190	12.7	1,886	71.5
South Wales	0.949	59.5	299	11.3
Mendip Hills	0.273	14.8	233	8.8
Peak District	0.210	13.0	220	8.4
TOTAL		100.0	2,638	100.0

Table 6.3 Distribution of detectors to each region if either number of caves or mean passage length are used as the criteria for determining location

REGION	% of detectors using number of cave entrances	% of detectors using mean passage length
North Pennines	77	12
South Wales	7	58
Mendip Hills	8	17
Peak District	8	13
TOTAL	100	100

Table 6.4 Number of Detectors to be placed in each Caving Region

REGION	% Number of detectors	Theoretical number of detectors	Final number of detectors
North Pennine	44.5	111	110
South Wales	32.5	81	86
Mendip Hills	12.5	31	NONE (4)
Peak District	10.5	27	44
Portland	0	0	6
TOTAL	100.0	250	250

Table 6.5 Theoretical distribution of detectors within the North Pennine Region if Passage length is used as criteria for location. (Passage length data from Hardwick, 1993)

SSSI	Total passage length (m)	Theoretical number of detectors	Notes
Upper Dentdale	3,863	1.7	*
Short Gill	781	0.4	*
Leck Beck Head	70,505	31.5	
Kingsdale	25,442	11.4	
Ingleborough	53,000	23.9	
Birkwith Caves	5,558	2.5	*
Brants Gill Catchment	14,089	4.3	+
Pikedaw Cavern	1,021	0.5	*
Sleets Gill Cave	3,071	1.3	*
Boreham Cave	3,137	1.4	*
Strans Gill	427	0.2	*
Birks Fell Cave	5,261	2.4	*
Dow Cave	4,414	1.9	
Black Keld Catchment	24,260	10.8	
Stump Cross	6,068	2.8	
Upper Nidderdale	9,520	4.5	
Crackpot Cave	549	0.3	*
Fairy Hole	3,479	1.5	*
Knock Fell Cavern	4,000	1.8	*
Hale Moss Caves	500	0.2	*
Areas with * were excluded to ensure that sufficient number of detectors were placed to ensure a representative sample is maintained. The area marked with + was excluded due to lack of resources.			

Table 6.6 Final location of detectors in the North Pennine Area.

SSSI	Cave	Number of Detectors
Leck Beck Head	Lost John	10
Leck Beck Head	Ireby Fell	10
Leck Beck Head	Ease Gill	15
Upper Nidderdale	Goydon Pot	6
Kingsdale	Kingsdale Master Cave	12
Kingsdale	Heron Pot	6
Kingsdale	Yordas Cave	4
Ingleborough Hill	White Scar Cave	6
Ingleborough Hill	Gaping Gill System	16
Ingleborough Hill	Sunset Hole	4
Ingleborough Hill	Upper Long Churn	10
Dow Cave	Dow Cave	6
Stump Cross	Stump Cross Caverns	5
	TOTAL	110

Table 6.7 Theoretical and actual distribution of detectors in the South Wales and Forest of Dean Caving Regions by SSSI's. (Passage length data from Hardwick, 1993)

SSSI	Passage Length	Theoretical number of Detectors	Number of Detectors
Dan-yr-Ogof	17,138	11	None (fan)
Ogof Ffynnon Ddu	38,313	24	27
Little Neath River Cave	9,020	6	6
Mynydd Llangattwg	66,781	43	48
Otter Hole	3,352	2	5
	TOTAL	86	86

Table 6.8 Final distribution of detectors within the South Wales and Forest of Dean Caving Regions

SSSI	Cave	Number of detectors
Ogof Ffynnon Ddu	Ogof Ffynnon Ddu	17
Ogof Ffynnon Ddu	Cwm Dwr	10
Mynydd Llangattwg	Ogof Agen Allwedd	20
Mynydd Llangattwg	Ogof Craig-a-Ffynnon	10
Mynydd Llangattwg	Ogof Darren Cilau	9
Mynydd Llangattwg	Camo Audit	9
Little Neath River Cave	Little Neath River Cave	3
Little Neath River Cave	Tunnel Cave	3
Otter Hole	Otter Hole	5
	TOTAL:	86

Table 6.9 Suggested allocation of detectors in the Mendip Hills Region

SSSI	Cave	Number of Detectors
Charterhouse	Charterhouse	3
Charterhouse	Blackmoor Swallet	2
Cheddar	Gough's Cave*	2
Cheddar	Cox's Cave*	2
Priddy Caves	Swildons Hole	5
Priddy Caves	St Cuthberts Swallet	7
Wookey Hole	Cuckoo Cleeves	5
St Dunstan's Well	Fairy Cave	5
	TOTAL	31
* These detectors were the only ones actually placed.		

Table 6.10 Distribution of detectors in the Peak District Region

SSSI	Cave	Number of detectors
Castleton Caves	Peak Cavern	10
Castleton Caves	Giant's Hole	4
Castleton Caves	P8 (Jackpot)	6
Castleton Caves	Gautries Hole	3
Stoney Middleton Dale	Carlswark Cavern	5
Stoney Middleton Dale	Streaks Pot	3
Bradwell	Bagshaw Cavern	5
Upper Lathkill Dale	Hillocks Mine	2
Upper Lathkill Dale	Knotlow Mine	2
Matlock	Devonshire Mine	2
Matlock	Jug Holes Mine	2
	TOTALS	44

Table 6.11 Regional cave radon concentrations (Bqm^{-3}) for England and Wales 1991 - 1992

REGION	Mean Rn conc	Max	Min	SD	No of readings
Portland	454	974	10	326	18
North Pennines	1,116	27,136	14	2,089	370
Mendip Hills	1,129	3,621	99	1,057	15
South Wales	2,601	19,968	127	2,773	249
Peak District	8,528	46,080	9	10,724	167

Table 6. 12 Summary of international cave radon concentrations (only the primary references from each country included)

County	Num	Mean	Max	Min	Method	Source
Switzerland	6	35,000	40,000	2,000	Spot	1
Greece	6	25,340	88,000	3,700	Spot	2
Greece	6	16,280	22,422	3,700	Spot	2
Japan	5	13,149	20,017	204	Spot	3
Hungary	25	3,300	14,000	500	Int 30	4
Yugoslavia	220	2,713	22,000	740	Int 30	5
USA	90	2,544*	3,774	1,443	Spot	6
Hungary	37	2,468	13,200	200	Int 30	7
USSR	14	2,390	8,550	373	Int 7	8
USA	12	1,776*	2,035	444	Spot	9
USA	60	1,235*	21,000	200	Spot	9
Czechoslovakia	60	1,235	21,000	200	Spot	10
USA	11	1,222*	4,255	185	Spot	11
USA	4	1,137*	2,349	743	Spot	12
Czechoslovakia	25	1,000	40,000	400	Int 30	13
Czechoslovakia	60	760	1,100	200	Spot	14
South Africa	63	212*	2,294	37	Spot	15
USA	33		12,765*	148	Spot	16
USA			9,620*		Spot	17
USA			33,226*		Spot	18
USA			24,272*		Spot	19
USA			25,530		Spot	19

* Results based upon radon daughter concentrations, converted to radon gas concentrations assuming an equilibrium factor of 0.5.

Sources: 1. Piller and Surbeck, 1989; 2.Papastefanou *et al.*, 1986; 3. Miki and Ikeya, 1989; 4. Somogyi *et al.*, 1989; 5. Kobal *et al.*, 1988; 6. Yarborough, 1977; 7. Lenart *et al.*, 1989; 8. Gunn, 1991; 9. Ahlstrand and Fry, 1976; 10. Burian and Stelcl, 1990; 11. Yarborough and Aley, 1977; 12. Ahlstrand and Fry, 1980; 13. Stelcl, 1991; 14. Burian and Stelcl 1990; 15. Gamble, 1981; 16. Yarborough *et al*,. 1976; 17. Carson, 1981; 18. Eheman *et al.*, 1991; 19. Yarborough, 1976.

Table 6.13 Cave Radon Concentrations in England and Wales, 1991 - 1992.

Sampling Periods	Para-meter	Peak District	South Wales	Mendip Hills	North Pennines	Portland
August	Num	43	65	3	109	
1991	Mean	19,413	4,939	2,610	2,172	
	SD	14,040	3,852	1,145	3,341	
	Max	46,080	19,968	3,621	27,136	
	Min	208	380	1,008	35	
November	Num	42	58	4	100	6
1991	Mean	4,776	1,530	575	694	443
	SD	4,868	1,163	329	1,207	357
	Max	17,774	4,973	905	9,876	974
	Min	9	156	127	27	154
February	Num	43	63	4	84	6
1992	Mean	3,389	1,390	1,191	697	464
	SD	3,622	919	826	831	292
	Max	19,200	4,169	2,400	4,767	960
	Min	77	127	173	10	141
May	Num	39	63	4	77	6
1992	Mean	6,234	2,388	510	626	445
	SD	6,546	1,704	285	540	342
	Max	26,990	6,802	809	1,993	985
	Min	281	394	99	14	10
Overall)	Total No	167	249	15	370	18
1991 -	Mean	8,528	2,601	1,129	1,116	454
1992	SD	10,724	2,773	1,057	2,089	326
	Max	46,080	19,968	3,621	27,136	974
	Min	9	127	99	14	10

Table 6.14 Summary of cave radon results for the North Pennines, 1991 - 1992.

SSSI	Mean	Max	Min	SD	Num
Leck Beck	1,504	27,136	14	3,047	128
Kingsdale	800	5,856	27	1,112	84
Ingleborough	610	6,480	60	1,200	100
Dow Cave	527	1,392	122	444	24
Stump Cross	2,418	5,568	782	1,321	19
Upper Nidderdale	2,244	5,312	233	1,865	15

Table 6.15 Summary of cave radon results for the South Wales and Forest of Dean caving regions, 1991 - 1992.

SSSI	Mean	Max	Min	SD	Num
Ogof Ffynnon Ddu	1,605	6,144	134	1,132	96
Mynydd Llangattwg	2,919	9,523	127	2,464	148
Otter Hole	12,332	19,968	7,461	4,381	5

Table 6.16 Summary cave radon results for the Peak District region, 1991 - 1992.

SSSI	Mean	Max	Min	SD	Num
Castleton	9,916	46,080	9	11,319	87
Stoney Middleton	5,218	39,047	178	10,061	32
Bradwell	12,187	31,817	234	7,396	16
Upper Lathkill Dale	12,019	45,910	1,117	11,339	16
Matlock	459	1,478	195	320	16

Table 6.17 Cave radon results for the North Pennines by area and sampling period

Sampling Period	Parameter	1*	2*	3*	4*	5*	6*
August	Num	35	21	36	6	5	6
1991	Mean	2,946	1,746	1,140	1,282	4,342	4,421
	SD	5,103	1,770	1,839	79	842	669
	Max	27,136	5,856	6,480	1,392	5,568	5,312
	Min	123	35	60	1,168	3,008	3,552
November	Num	35	21	32	6	5	1
1991	Mean	1,035	397	353	271	2,332	233
	SD	1,710	248	391	136	987	-
	Max	9,876	1,456	1,506	603	3,840	233
	Min	86	27	63	122	783	233
February	Num	30	21	18	6	5	4
1992	Mean	1,111	378	321	238	1,659	454
	SD	1,151	353	269	63	450	214
	Max	4,767	1,456	1,015	325	2,345	823
	Min	178	62	68	126	946	288
May	Num	28	21	14	6	4	4
1992	Mean	709	680	203	315	1,070	1,272
	SD	417	610	143	142	308	250
	Max	1,783	1,993	645	559	1,961	1,522
	Min	14	55	97	144	1,152	1,015
Overall	Num	128	84	100	24	19	15
1991 -	Mean	1,504	800	610	527	2,418	2,244
1992	SD	3,047	1,112	1,200	444	1,321	1,865
	Max	27,136	5,856	6,480	1,392	5,568	5,312
	Min	14	27	60	122	782	233
	1* Leck Beck, 2* Kingsdale, 3* Ingleborough, 4*Dow Cave, 5* Stump Cross, 6* Upper Nidderdale						

Table 6.18 Summary Cave radon results for South Wales by area and season

Sampling Period	Parameter	Ogof-Ffynnon-Ddu	Mynydd-Llangattwg	Otter Hole
August 1991	Num	22	38	5
	Mean	2,872	5,163	12,332
	SD	1,864	3,029	4,381
	Max	6,144	9,523	19,968
	Min	1,207	380	7,461
November 1991	Num	23	35	
	Mean	1,264	1,705	
	SD	573	1,398	
	Max	2,781	4,532	
	Min	199	156	
February 1992	Num	26	37	
	Mean	1,043	1,634	
	SD	473	1,382	
	Max	2,153	6,244	
	Min	134	127	
May 1992	Num	25	38	
	Mean	1,389	3,045	
	SD	189	2,106	
	Max	1,760	6,802	
	Min	1,088	424	
Overall 1991 - 1992	Num	96	148	5
	Mean	1,605	2919	12,332
	SD	1,132	2,464	4,381
	Max	6,144	9,523	19,968
	Min	134	127	7,461

Table 6.19 Summary of cave radon results for the Peak District by area and sampling period

Sampling Period	Parameter	1*	2*	3*	4*	5*
August	Num	23	4	4	8	4
1991	Mean	22,031	27,204	24,471	14,974	392
	SD	14,105	4,715	18,394	10,375	305
	Max	46,080	31,817	45,910	39,047	922
	Min	2,062	20,846	1,117	6,199	208
November	Num	22	4	4	8	4
1991	Mean	5,122	13,714	5,650	961	688
	SD	4,447	1,525	4081	1,017	685
	Max	17,774	15,579	11,630	2,578	1,214
	Min	9	11,849	2,907	178	264
February	Num	23	4	4	8	4
1992	Mean	4,074	4,364	6,161	1,089	305
	SD	3,998	636	2,874	930	108
	Max	19,200	8,100	8,448	2,523	425
	Min	77	234	2,427	202	195
May	Num	19	4	4	8	4
1992	Mean	7,872	3,464	11,795	3,849	450
	SD	8,356	983	3,987	1,663	253
	Max	26,990	6,000	16,677	6,363	891
	Min	579	2,000	7,570	1,755	281
Overall	Num	87	16	16	32	16
1991	Mean	9,916	12,187	12,019	5,218	459
-1992	SD	11,319	7,396	11,339	10,061	320
	Max	46,080	31,817	45,910	39,047	1,478
	Min	9	234	1,117	178	195
	1* Castleton, 2* Bradwell Dale, 3* Upper Lathkill Dale, 4* Stoney Middleton Dale, 5* Matlock.					

Table 7.1 Location of sampling sites for determination of emanation and exhalation of radon from main cave bearing limestone strata from South Wales.

Approximate Coral - Brachiopod zones	Regional Stage	Lithostratigraphy	Sample Collection Sites	Notes
D_2	Brigantian	Oystermouth Beds, Penwyllt Limestone, Oxwich Head Limestones		Insignificant amounts of cave passages developed in these strata
D_1	Asbian	Oxwich Head, Penderyn Oolite, Honeycombed Sandstones, Greenhall Limestones,		Insignificant amounts of cave passages developed in these strata
S_2	Holkerian	Dowlais Limestone, Cil-yr-Ychan Limestone, Hunts Bay Oolite	Ogof Ffynnon Ddu, Dan yr Ogof, Pwll Gwynt, Pwll-y-Pasg	Cave bearing Strata
C_2, S_1	Arundian	High Tor Limestone, Llanelly Formation, Gilwern Oolite	Llanelly Quarry Pot	
C_1	Chadian	Caswell Bay Oolite, Gilwern Oolite, Clydach Beds, Blaen Onneu Oolite	Ogof Darren Cilau, Agen Allwedd, Prices Dig	Cave bearing Strata
K,Z	Courceyan	Penmaen Burrows Limestone, Blaen Onnean Oolite, Pwll y Cwm Oolite, Lower Limestones	Ogof Chapel, Fell Swoop, Ogof Craig a Ffynnon	Cave bearing Strata

Table 7.2 Location of sampling sites for determination of emanation and exhalation of radon from main cave bearing limestone strata from North Pennines

Approximate Coral - Brachiopod zones	Regional Stage	Lithostratigraphy	Sample Collection Sites
D_2	Pendleian		No major caves developed within this strata
D_1	Brigantian	Wensleydale Group, Upper Alston Group	Wensleydale
S_2	Asbian	Kingsdale Formation, Greenhow limestone, Malham Formation	Kingsdale Valley, Malham Cove, Greenhow Quarry
C_2, S_1	Holkerian	Malham Formation, Fawes Wood Limestone, Horton Limestone, Stump Cross Limestone	Stump Cross Caverns, Horton in Ribblesdale
C_1	Arundian	Kilnsey Formation	Kilnsey Quarry

Table 7.3 Location of sampling sites for determination of emanation and exhalation of radon from main cave bearing limestone strata from the Peak District

Approximate Coral - Brachiopod zones	Regional Stage	Lithostratigraphy	Sample Collection Sites	Notes
D_2	Brigantian	Eyam Limestones, including reef facies, Monsal Dale Limestones	Stanlow Dale, Carlswark Cavern, Streak Pot, Bagshaw Caverns, Speedwell Quarry	
D_1	Asbian	Bee Low Limestones, including Apron Reef Facies and Monsal Dale Limestones	Treak Cliff Cavern, Oxlow Caverns, Giants Hole, Eldon Hole, Laneside Farm Quarry, Butterford Cross, Calver	Main Cave Bearing Strata
S_2	Holerian	Woo Dale Limestones	Laneside Farm, Dale Head Farm,	

Table 7.4 Location of sampling sites for determination of emanation and exhalation of radon from main cave bearing limestone strata from the Mendip Hills

Approximate Coral - Brachiopod zones	Regional Stage	Lithostratigraphy	Sample Collection Sites
D_2	Brigantian	Hotwell Limestone	Wellington Farm, Cheddar Gorge.
D_1	Asbian	Cliffton Down Group	Wells, Cheddar Gorge,
S_2	Holkerian	Black Rock Limestone	Priddy Village, Burrington Combe, Hunters lodge Inn

Table 7.5 Sediment samples colledted for determination of emanation and exhalation rates.

Region	Cave
North Pennines	Goydons Pot
	Dow Cave
	Upper Long Churn
	Kingsdale Master Cave
	Sunset Hole
South Wales	Ogof Ffynnon Ddu 1
	Ogof Ffynnon Ddu 2
	Cwm Dwr
	Carno Audit
	Ogof Agen Allwedd
Mendip Hills	Goughs Cave
	Swildons Hole
	St Cuberts
	Goatchurch Caverns
	Rods Pot
Peak District	Giants Hole
	Peak Cavern
	Carlswark Caverns
	Knotlow Caverns
	Devonshire Mine

Table 7.6 Collection sites of speleothem samples analysed for emanation and exhalation rates

Cave	U ppm	Sample Location
Ogof Ffynnon Ddu	2.6	Rawl Series
Ogof Ffynnon Ddu	0.13	Cathedral Passage
Ogof Ffynnon Ddu	0.66	Top Entrance Series
Ogof Ffynnon Ddu	1.7	Small Gnome
Powells Cave	2.09	Wall of Powells Cave
Dan-yr-Ogof	0.97	Red Chamber

Table 7.7 Soil radon gas (Bq m^{-3}) concentration by underlying geology.

Bedrock underlying soil sample Location	Number of Samples	Mean	Standard Error
Namurian Shales	279	31,000	3,000
Longstone Mudstone	3	81,000	30,000
Eyam limestone	47	30,000	20,000
Monsal Dale Limestones	83	40,000	13,000
Bee Low Limestones	75	55,000	5,000
Apron Reef Facies	42	78,000	8,000
Millersdale Limestones	38	33,000	14,000

Table 7.8 Radon Concentrations in both air (Bq m^{-3}) and water (Bq l^{-1}) from Giants Hole, Peak District.

Sampling Period	Entrance		Base Camp		Top of Garlands Pot		Bottom of Garlands Pot	
	Water	Air	Water	Air	Water	Air	Water	Air
Sept 91	11.0	342	32.0	12,342	26.0	11,032	38.0	10,023
Dec 91	8.2	175	1.1	243	0.7	345	0.4	654
Feb 92	10.5	124	2.4	994	1.4	1,163	0.6	1,199
Apr 92	6.9	245	1.7	9,567	2.1	8,653	0.6	6,743
Jun 92	11.8	356	23	21,954	46.0	17,653	54.0	14,854
Sept 92	10.5	431	6.3	5,789	2.0	6,793	1.5	6,873

Table 7.9 Concentrations of radon gas in stream and percolation water (Bq l^{-1}) from Peak Cavern, Castleton.

Sampling Location	Far Sump	Ink Sump	Main Stream Inlet	Surprise View	Upstream Buxton Water	Downstream Buxton Water	Percolation Water
Jan 91	53.8	42.3	27.4	18.4	16.4	21.5	35.7
Apr 91	56.3	45.6	30.4	20.2	16.5	21.9	65.4
July 91	68.7	48.9	32.3	24.6	18.5	24.7	73.3
Sep 91	60.3	42.5	29.4	22.3	17.4	22.7	79.3
Jan 92	59.9	43.3	35.6	21.6	19.3	22.5	61.4

Table 7.10 Uranium-238 concentrations and estimated maximum radon exhalation potential.

Region	Regional Stage	Uranium concentration (ppm)			Estimated net radon production rate (Bq m^{-3} kg^{-1})		
		Min	Mean	Max	Min	Mean	Max
Peak District	Brigantian	2.5	7	12	31.3	87.5	150.0
	Asbian	6	15	24	75.0	187.5	300.0
	Holkerian	4	11	14	50.0	137.5	175.0
North Pennines	Brigantian	0.2	0.6	2.1	2.5	7.5	26.3
	Asbian	0.1	0.5	1.9	1.3	6.3	23.8
	Holkerian	0.3	0.8	1.3	3.8	10.0	16.3
	Arundian	0.2	0.4	0.8	2.5	5.0	10.0
South Wales	Holkerian	0.1	1.0	2.1	1.3	12.5	26.3
	Chadian	0.5	1.4	2.4	6.3	17.5	30.0
Mendip Hills	Brigantian	0.1	1.9	2.1	1.3	23.8	26.2
	Asbian	0.3	1.6	1.8	3.8	20.0	22.5
	Holkerian	0.2	1.3	1.9	2.5	16.3	23.8

Table 7.11 Radon Exhalation Rates by regional geological stage.

Region	Regional Stage	Radon release rate ($Bqm^{-3}m^{-2}$)			Radon release rate ($Bq\ m^{-3}\ kg^{-1}$)		
		Min	Mean	Max	Min	Mean	Max
Peak District	Brigantian	79.0	170.0	260.0	26	72.0	117.0
	Asbian	25.0	29.0	45.0	19.0	125.0	218.0
	Holkerian	15.0	27.0	51.0	24.0	78.0	94.0
North Pennines	Brigantian	1.2	4.3	12.1	1.4	5.6	12.4
	Asbian	0.9	3.8	13.8	1.0	4.1	5.1
	Holkerian	0.6	2.9	9.6	1.2	2.1	3.6
	Arundian	0.1	3.6	5.4	1.1	1.4	2.1
South Wales	Holkerian	1.1	2.4	3.6	3.2	6.0	8.9
	Chadian	1.1	3.5	5.3	2.8	7.7	11.1
Mendip Hills	Brigantian	0.6	2.1	7.1	0.6	5.9	8.6
	Asbian	0.9	2.5	6.8	1.0	6.3	9.4
	Holkerian	0.7	2.3	6.2	1.2	6.1	8.9

Table 7.12 Percentage radon emanation and theoretical calculations of the quantity of rock needed to raise 1 m^3 of cave passage to 1 WL.

Region	Regional Stage	Radon emanation (%)			Quantities of rock needed to raise $1m^3$ of cave passage to 1WL		
		Min	Mean	Max	Max*	Mean*	Min*
Peak District	Brigantian	78	82	83	142	51	32
	Asbian	63	66	72	195	30	17
	Holkerian	48	51	53	154	47	39
North Pennines	Brigantian	47	49	56	2643	661	298
	Asbian	21	65	76	3700	902	725
	Holkerian	22	27	31	3808	1762	1027
	Arundian	14	28	56	2643	2623	1762
South Wales	Holkerian	33	40	48	1156	616	416
	Chadian	37	44	44	1321	481	333
Mendip Hills	Brigantian	33	25	48	616	623	427
	Asbian	26	32	41	3795	578	401
	Holkerian	37	37	48	3083	615	421

* The quantity needed is determined by 3700/ A, where A the Radon release rate is determined by $A = 12.5 \times [u] \times \phi$, [u] uranium concentration and ϕ the emanation coefficient.

Table 7.13 Radon Exhalation Rates for Sediments (Bq m^{-3} kg^{-1})

Region	Sample	Mean	Minimum	Maximum	No
Peak District	Coarse Sediments	42	25	75	5
	Silts and Clays	230	150	350	5
North Pennines	Coarse Sediments	37.5	25	50	5
	Silts and Clays	87.5	37.5	150	5
South Wales	Coarse Sediments	31.3	25	37.5	5
	Silts and Clays	100	50	150	5
Mendip Hills	Coarse Sediments	31.3	25	50.0	5
	Silts and Clays	100	62.5	162.5	5

Table 7.14 Radon exhalation rates(Bqm^{-3} kg^{-1}) for sub-samples from regional sediments samples.

Region	Sample	Mean	Minimum	Maximum	No
Peak District	Bulked	175.0	25.0	350.0	10
	Coarse Sand	37.5	25.0	62.5	10
	Fine Sands	50.0	37.5	87.5	10
	Silts and Clays	162.5	112.5	387.5	10
North Pennines	Bulked	62.5	25.0	150.0	10
	Coarse Sand	43.8	25.0	75.0	10
	Fine Sands	62.5	50.0	112.5	10
	Silts and Clays	75.0	37.5	150.0	10
South Wales	Bulked	47.5	25.0	150.0	10
	Coarse Sand	42.5	25.0	50.0	10
	Fine Sands	75.0	62.5	112.5	10
	Silts and Clays	87.5	62.5	150.0	10
Mendip Hills	Bulked	50.0	25.0	162.5	10
	Coarse Sand	31.0	25.0	37.5	10
	Fine Sands	45.7	37.0	100.0	10
	Silts and Clays	94.5	75.0	162.5	10

Table 7.15 Quantity of sediment (kg) needed to raise 1 m^3 of cave passage to 1WL.

Region	Sediment Type	Quantity of sediment (kg)
Peak District	Coarse Sands	88
	Silts and Clays	24
North Pennines	Coarse Sands	99
	Silts and Clays	42
South Wales	Coarse Sands	118
	Silts and Clays	37
Mendip Hills	Coarse Sands	118
	Silts and Clays	59

Table 7.16 Radon emanation and exhalation rates for speolethems (Bq m^{-3} kg^{-1})

Cave	Sample Location	U ppm	Exhalation assuming 100% emanation	Recorded exhalation rates
Ogof Ffynnon Ddu	Rawl Series	2.6	32.5	0.31
Ogof Ffynnon Ddu	Cathedral Passage	0.13	1.2	0.11
Ogof Ffynnon Ddu	Top Entrance Series	0.66	8.3	0.06
Ogof Ffynnon Ddu	Small Gnome	1.7	21.3	0.14
Powells Cave	Wall of Powells Cave	2.09	26.2	0.43
Dan-yr-Ogof	Red Chamber	0.97	12.1	0.21

Table 8.1 Number of detectors placed in each cave investigated

Cave Name	Number of detector locations
P8 Cave (Jackpot)	4
Giant's Hole	6
Carlswark Cavern	6
Knotlow Cavern	3
Devonshire Cavern	3
Hillocks Mine	2
Gautries Hole	2
Jugholes Cavern	2
Axe Hole	1

Table 8.2 Radon concentrations in selected Peak District caves 1991 - 1992 (Bq m^{-3}).

Cave	No of results	Mean	Maximum	Minimum	SD	No of missing results
Giant's Hole	75	7,856	30,564	648	8,162	3
Knotlow Cavern	18	7,728	15,025	1,886	4,063	0
Gautries Hole	24	6,509	21,189	45	6,164	2
Hillocks Mine	26	4,388	13,626	648	3,656	0
Carlswark Cavern	75	2,189	10,166	100	2,179	3
P8 Cave (Jackpot)	37	1,845	8,192	120	2,666	15
Devonshire Cavern	36	586	2,610	80	310	3
Jugholes Cavern	25	208	792	96	155	1
Axe Hole	13	442	1,570	160	451	0

Table 8.3 Radon concentrations in Peak District Caves, by season, 1991 - 1992 (Bq m^{-3}).

		CAVE								
Season	Var	1	2	3	4	5	6	7	8	9
Winter	Mean	646	948	465		792	2,358	2,990	230	198
	No	13	23	23		10	8	5	8	4
	Max	4,514	8,525	10,166		2,610	3,593	3,442	346	288
	Min	252	266	108		72	1,620	2,340	72	194
Spring	Mean	2,154	6,335	2,061		582	4,700	6,362	238	353
	No	12	18	18		8	6	8	6	3
	Max	5,422	17,748	5,854		1,714	5,825	12,240	418	590
	Min	591	2,721	354		76	1,261	1,974	88	97
Summer	Mean	2,591	12,721	4,354	7,613	476	8,261	10,974	188	977
	No	7	17	17	9	6	6	5	6	3
	Max	8,192	27,684	8,964	15,025	986	13,626	21,189	792	1,570
	Min	4,450	5,494	3,319	3,182	230	5,500	9,806	108	605
Autumn	Mean	1,985	7,070	1,877	8,339	495	2,234	4,756	176	241
	No	4	18	17	9	9	6	5	6	3
	Max	4,838	30,564	8,064	9,208	1,066	13,111	15,732	267	1,094
	Min	900	2,304	533	1,886	288	648	1,822	65	72

1 = P8 Cave, 2 = Giants Hole, 3 = Carlswark Cavern, 4 = Knotlow Cavern, 5 = Devonshire Cavern, 6 = Hillocks Mine, 7 = Gautries Hole, 8 = Jugholes Cavern, 9 = Axe Hole

Table 8.4 Radon concentrations in Peak District caves, grouped by region, 1991 - 1992 (Bq m^{-3}).

Region	Num	Mean	Maximum	Minimum
Lathkill Dale	44	6,953	13,626	648
Castleton	136	5,158	30,564	252
Eyam Dale	75	2,181	10,166	108
Buxton	13	437	1,570	72
Matlock	61	381	2,610	65

Table 8.5 Radon concentrations in Peak District caves grouped by geology at the entrance 1991 -1992 (Bq m^{-3}).

Geology	Num	Mean	Maximum	Minimum
Eyam Limestones	119	6,954	13,626	1,400
Apron Reef Facies	136	4,413	30,564	45
Hoptonwood Limestones	13	569	2,610	58
Woo Dale Limestones	36	437	1,361	160
Matlock Limestones	25	193	480	90

Table 8.6 Correlation coefficients (r) for radon concentrations for individual sampling periods with meteorological parameters.

Cave	Mean Temperature	Total Rainfall	Mean Pressure	Mean Minimum Temperature	Mean Maximum Temperature
P8 Cave (Jackpot)	0.79^1	-0.41^1	0.01^2	0.76^1	0.79^1
Giants Hole	0.83^1	-0.54^1	0.04^2	0.82^1	0.89
Carlswark Cavern	0.91^1	-0.41^1	0.01^2	0.92^1	0.93^1
Knotlow Cavern	-0.34^2	0.06^2	0.21^1	-0.37^1	-0.13^2
Devonshire Cavern	-0.41^1	0.42^1	0.12^2	-0.36^1	-0.41^1
Hillocks Mine	0.87^1	-0.61^1	-0.24^1	0.76^1	0.88^1
Gautries Hole	0.73^1	-0.34^2	0.16^2	0.72^1	0.79^1
Jug Hole Cavern	-0.04^2	-0.19^2	0.23^1	0.06^2	0.04^2
Axe Hole	0.88^1	-0.56^1	0.34^1	0.80^1	0.87^1
x^1 denotes significant at the 95% confidence level x^2 denotes not significant at the 95% significance level					

Table 8.7 Correlation coefficients (r) for maximum radon concentrations for individual sampling periods with meteorological parameters.

Cave	Mean Temperature	Total Rainfall	Mean Pressure	Mean Minimum Temperature	Mean Maximum Temperature
P8 Cave (Jackpot)	0.79^1	-0.50^1	-0.1^2	0.73^1	0.81^1
Giants Hole	0.85^1	0.54^1	-0.01^2	0.84^1	0.91^1
Carlswark Cavern	0.86^1	-0.38^1	0.06^2	0.88^1	0.86^1
Knotlow Cavern	0.33^2	0.71^1	-0.55^1	0.29^2	0.60^1
Devonshire Cavern	-0.50^1	0.40^1	0.03^2	-0.45^1	-0.51^1
Hillocks Mine	0.92^1	-0.57^1	-0.25^2	0.82^1	0.89^1
Gautries Hole	0.78^1	-0.25^2	0.17^2	0.78^1	0.80^1
Jug Holes Caverns	-0.14^2	-0.25^2	0.12^2	0.08^2	0.06^2
Axe Hole	0.87^1	-0.55^1	0.02^2	0.80^1	0.86^1
x^1 denotes significant at the 95% confidence level x^2 denotes not significant at the 95% significance level					

Table 8.8 Correlation coefficients (r) for minimum radon concentrations for individual sampling periods with meteorological parameters.

Cave	Mean Temperature	Total Rainfall	Mean Pressure	Mean Minimum Temperature	Mean Maximum Temperature
P8 Cave (Jackpot)	0.68^1	-0.28^2	0.07^2	0.64^1	0.63^1
Giants Hole	0.78^1	-0.50^1	0.16^2	0.76^1	0.85^1
Carlswark Cavern	0.92^1	-0.45^1	0.05^2	0.90^1	0.93^1
Knotlow Cavern	-0.58^1	0.38^1	0.50^1	-0.54^1	-0.43^1
Devonshire Cavern	-0.38^1	0.05^2	0.04^2	0.35^1	0.41^1
Hillocks Mine	0.58^1	-0.63^1	-0.04^2	0.43^1	0.61^1
Gautries Hole	0.53^1	-0.39^1	0.12^2	0.50^1	0.60^1
Jug Holes Caverns	0.20^1	-0.01^2	0.21^2	0.19^2	0.25^2
Axe Hole	0.87^1	-0.55^1	0.23^2	0.28^2	0.86^1

x^1 denotes significant at the 95% confidence level
x^2 denotes not significant at the 95% significance level

Table 8.9 Summary of Correlation coefficients (r) for mean radon concentrations for sampling periods grouped into seasons with meteorological parameters

Season	Cave	Mean Temp	Total Rainfall	Mean Pressure	Mean Min Temp	Mean Max Temp
Winter	P8 Cave (Jackpot)	0.326^1	-0.78^1	0.23^2	0.16^2	0.15^2
	Giants Hole	0.99^1	0.39^1	0.13^2	0.94^1	0.96^1
	Carlswark Caverns	0.79^1	-0.26^2	0.72^1	0.78^1	0.81^1
	Knotlow Caverns	023^2	0.34^1	0.54^1	0.23^2	0.45^1
	Devonshire Caverns	0.19^2	0.98^1	0.94^1	-0.09^2	-0.01^2
Spring	P8 Cave (Jackpot)	0.96^1	-0.14^2	0.41^1	0.89^1	0.88^1
	Giants Hole	0.99^1	-0.83^1	0.65^1	0.86^1	0.96^1
	Carlswark Caverns	0.99^1	-0.44^1	0.67^1	0.82^1	0.93^1
	Knotlow Caverns	0.34^2	0.43^1	0.02^2	0.12^2	0.34^1
	Devonshire Caverns	0.82^1	0.23^2	0.87^1	0.78^1	0.79^1
Summer	P8 Cave (Jackpot)	-0.44^1	-0.18^2	0.49^1	0.69^1	0.72^1
	Giants Hole	0.34^2	0.57^1	0.07^2	0.38^1	0.32^1
	Carlswark Caverns	0.91^1	0.06^2	-0.39^1	0.81^1	0.81^1
	Knotlow Caverns	0.13^2	-0.81^1	-0.91^1	0.23^2	0.33^2
	Devonshire Caverns	0.93^1	0.08^2	-0.91^1	0.97^1	0.38^1
Autumn	P8 Cave (Jackpot)	0.97^1	0.99^1	-0.84^1	0.76^1	0.85^1
	Giants Hole	0.95^1	0.96^1	-0.87^1	0.38^1	0.94^1
	Carlswark Caverns	0.86^1	0.88^1	0.28^2	0.42^1	0.41^1
	Knotlow Caverns	0.32^2	0.34^1	-0.42^1	0.56^1	0.41^1
	Devonshire Caverns	0.56^1	0.54^1	0.02^2	0.93^1	0.84^1

x^1 denotes significant at the 95% confidence level
x^2 denotes not significant at the 95% significance level

Table 9.1 Sampling locations and designation of areas within Peak Cavern. See figure 9.1 for location of sampling sites.

Sampling Location Number	Sampling Location	Area
1	Entrance	1
2	Start of Lumbago Walk	1
3	Start of Great Cave	1
4	End of Great Cave	1
5	Top of Devils Staircase	1
6	Bottom of Devils Staircase	1
7	Five Arches	1
8	Downstream end of Buxton Water Sump	1
9	Before Muddy Ducks	2
10	Razor Blade Avon	2
11	B Cubed	2
12	Start of Treasury Passage	2
13	Treasury Chamber	2
14	Treasury Sump (at waters edge)	2
15	First Aid Post	2
16	Top of Surprise View	2
17	Galena Chamber	5
18	Main Stream Inlet	5
19	Squaws Junction	3
20	Bottom of Puckerings Passage	3
21	Top of slope leading to Boulder Hall	3
22	Downstream end of Far Sump (waters edge)	3
23	Start of Lake Passage	4
24	Before Lake Sump	4
25	Downstream end of Ink Sump (waters edge)	4
26	Main streamway above Surprise View (30m)	3
27	Main streamway below Surprise View (30m)	3
28	Upstream of Buxton Water Sump (15m)	3

Table 9.2 Summary of Radon Daughter concentration (WL) for all sampling periods.

	No	Mean	Standard Deviation	Min	Max
Overall	1316	1.27	1.60	0.0001	22.41
Area 1	376	1.01	2.28	0.0001	22.41
Area 2	376	1.23	1.39	0.006	10.84
Area 3	282	1.40	1.01	0.189	5.61
Area 4	141	1.52	0.94	0.483	5.61
Area 5	141	1.59	1.25	0.13	6.41

Table 9.3 Summary of Radon Daughter concentrations (WL) in Peak Cavern by site. See figure 9.1 for site locations and table 9.1 for site names. (47 measurements at each site).

Site	Mean	SD	Min	Max
1	0.02	0.09	0.0001	0.63
2	0.54	0.84	0.0002	3.2
3	0.68	1.09	0.0008	5.36
4	1.62	3.65	0.0038	22.41
5	1.41	3.17	0.0037	20.21
6	1.25	2.04	0.002	11.3
7	1.30	2.57	0.005	16.41
8	1.28	2.10	0.005	12.68
9	1.14	1.74	0.006	10.84
10	1.07	1.45	0.008	8.63
11	0.97	1.27	0.010	7.41
12	0.94	1.12	0.012	6.41
13	0.98	1.09	0.130	6.03
14	2.44	1.67	0.717	9.03
15	1.08	1.04	0.150	5.04
16	1.22	1.00	0.130	6.01
17	1.55	1.34	0.130	6.31
18	1.58	1.25	0.180	6.41
19	1.64	1.19	0.243	5.06
20	1.47	1.18	0.201	4.91
21	1.28	1.04	0.189	4.42
22	1.12	0.89	0.201	1.31
23	1.52	0.90	0.432	4.32
24	1.50	0.90	0.501	4.40
25	1.55	1.04	0.491	5.61
26	1.53	0.96	0.413	5.61
27	1.49	0.98	0.334	5.43
28	1.51	0.99	0.214	5.01

Table 9.4 Summary of Radon Daughter concentrations (WL) by sampling period (results from all 28 sites included in each period).

Sampling Period	Mean	SD	Min	Max	No
(02/09/91) 1	0.50	0.32	0.003	0.93	28
(09/09/91) 2	1.74	0.60	0.008	3.31	28
(17/09/91) 3	0.83	0.56	0.006	2.04	28
(23/09/91) 4	0.60	0.57	0.001	1.89	28
(25/09/91) 5	1.16	0.80	0.004	4.64	28
(30/09/91) 6	0.92	0.63	0.006	3.64	28
(07/10/91) 7	0.72	0.47	0.0003	2.36	28
(17/10/91) 8	0.42	0.49	0.001	1.97	28
(23/10/91) 9	0.53	0.51	0.007	1.42	28
(31/10/91) 10	1.37	0.71	0.009	2.61	28
(07/11/91) 11	1.8	0.71	0.005	3.25	28
(13/11/91) 12	0.53	0.61	0.0003	1.64	28
(20/11/91) 13	0.94	1.07	0.0008	3.10	28
(25/11/91) 14	0.41	0.46	0.0002	1.38	28
(29/11/91) 15	1.06	0.64	0.016	1.96	28
(02/12/91) 16	0.59	0.61	0.016	1.94	28
(09/12/91) 17	0.56	0.65	0.0008	2.01	28
(12/12/91) 18	0.40	0.45	0.004	1.63	28
(22/12/91) 19	0.24	0.24	0.0006	0.89	28
(09/01/92) 20	2.51	1.59	0.0004	4.39	28
(13/01/92) 21	2.16	1.76	0.0003	5.13	28
(20/01/92) 22	0.51	0.54	0.0008	2.30	28
(28/01/92) 23	0.52	0.53	0.002	2.31	28
(03/02/92) 24	0.47	0.41	0.002	1.03	28
(10/02/92) 25	0.44	0.39	0.0008	0.98	28
(16/02/92) 26	0.70	0.64	0.0005	1.91	28
(23/02/92) 27	0.26	0.31	0.0003	0.93	28
(09/03/92) 28	0.34	0.34	0.0004	0.96	28
(16/03/92) 29	0.36	0.34	0.003	0.87	28
(01/04/92) 30	0.50	0.29	0.003	0.84	28
(07/04/92) 31	0.72	0.67	0.003	2.79	28
(15/04/92) 32	0.85	0.73	0.003	2.88	28
(24/04/92) 33	2.00	0.61	0.011	3.59	28
(05/05/92) 34	0.81	0.58	0.002	2.92	28
(12/05/92) 35	1.04	0.48	0.022	2.79	28
(18/05/92) 36	2.00	0.92	0.003	3.66	28
(26/05/92) 37	1.95	0.91	0.003	3.81	28
(29/05/92) 38	2.22	0.80	0.003	3.43	28
(01/06/92) 39	7.39	5.18	0.003	22.40	28
(08/06/92) 40	2.77	1.47	0.009	6.08	28
(12/06/92) 41	2.23	1.28	0.007	5.61	28
(17/06/92) 42	2.07	0.89	0.003	3.63	28
(22/06/92) 43	2.2	0.90	0.003	3.64	28
(06/07/92) 44	2.54	1.71	0.003	7.84	28
(13/07/92) 45	1.25	0.72	0.011	3.04	28
(04/08/92) 46	1.25	0.44	0.006	2.17	28
(11/08/92) 47	2.51	1.82	0.010	8.41	28

Table 9.5 List of Variables included in model development.

Number	Variable
1	Direction of air movement at measurement site
2	Internal cave air temperature at measurement site (°C)
3	Internal cave air pressure at measurement site (mb)
4	External air temperature (°C)
5	External air pressure (mb)
6	Difference between internal and external air temperature (°C)
7	External Air Direction
8	Number of one hour intervals where no pressure changes occurred during a day
9	Number of one hour intervals where pressure increases by 1 mb during a day
10	Number of one hour intervals where pressure increases by 2 mb during a day
11	Number of one hour intervals where pressure increases by 3 mb during a day
12	Number of one hour intervals where pressure decreases by 1 mb during a day
13	Number of one hour intervals where pressure decreases by 2 mb during a day
14	Number of one hour intervals where pressure decreases by 3 mb during a day
15	Previous days mean external temperature (°C)
16	Previous days minimum temperature (°C)
17	Previous days maximum temperature (°C)
18	Previous days mean external pressure (mb)
19	Previous days minimum external pressure (mb)
20	Previous days maximum external pressure (mb)
21	Previous days rainfall (mm)
22	Previous weeks mean external temperature (°C)
23	Previous weeks minimum external temperature (°C)
24	Previous weeks maximum external temperature (°C)
25	Previous weeks mean external pressure (mb)
26	Previous weeks minimum external pressure (mb)
27	Previous weeks maximum external pressure (mb)
28	Previous weeks rainfall (mm)
29	Total number where no pressure changes occur during an hour during a week
30	Total number pressure increases by 1 mb during an hour during a week
31	Total number pressure increases by 2 mb during an hour during a week
32	Total number pressure increases by 3 mb during an hour during a week
33	Total number pressure decreases by 1 mb during an hour during

	a week
34	Total number pressure decreases by 2 mb during an hour during a week
35	Total number pressure decreases by 3 mb during an hour during a week
36	28 day mean external temperature (°C)
37	28 day minimum external temperature (°C)
38	28 day maximum external temperature (°C)
39	28 day mean external pressure (mb)
40	28 day minimum external pressure (mb)
41	28 day maximum external pressure (mb)
42	28 day rainfall (mm)

Table 9.6 List of variables that exhibit significant colinearity (number refers to table 9.5)

	Variable Number															
	1	15	16	17	18	19	20	22	23	24	25	26	27	36	37	38
1		◆	◆	◆				◆	◆	◆				◆	◆	◆
15	◆		◆	◆				◆	◆	◆	◆	◆	◆		◆	◆
16	◆	◆		◆					◆			◆	◆			◆
17	◆	◆	◆					◆						◆	◆	◆
18								◆								
19								◆								
20								◆								
22	◆	◆		◆	◆	◆	◆							◆	◆	
23	◆	◆	◆							◆						◆
24	◆	◆							◆			◆			◆	◆
25		◆														
26		◆	◆							◆						
27		◆	◆													
36	◆			◆				◆								
37	◆	◆		◆				◆		◆						◆
38	◆	◆	◆	◆					◆	◆					◆	

Table 9.7 'B' values and significance levels for multiple regression equation based upon all variables for the entire cave (variable numbers based upon table 9.5)

Multiple R = 0.692, R squared = 0.479, Signif F = 0.0000		
Variable Number	'B' value	Significance level
1	0.053	0.01
8	-0.174	0.00
12	0.0385	0.00
7	$-7.1E^{-4}$	0.23
3	0.018	0.00
2	0.212	0.00
38	0.159	0.00
37	0.139	0.00
42	-0.045	0.00
15	0.021	0.52
25	-0.058	0.00
22	0.215	0.00
21	-0.068	0.00
19	-0.166	0.00
20	0.125	0.01
26	-0.088	0.00
28	0.075	0.00
27	0.081	0.26
29	0.014	0.00
30	0.062	0.00
32	0.332	0.00
33	0.069	0.00
35	0.570	0.00
Constant	-49.900	0.00

Table 9.8 Results of stepwise multiple regression for all areas of Peak Cavern (only the first five variables are included).

Multiple R = 0.593, R squared = 0.351, Significance level = 0.000		
Variable	'B' value	Significance level
Previous weeks Min external air temperature	0.1429	0.000
External Wind Direction	-0.004	0.000
Monthly Rainfall	-0.0204	0.000
Previous weeks rainfall	0.0359	0.000
Previous weeks maximum air pressure	0.0392	0.000
Constant	-38.066	0.000

Table 9.9 Results of stepwise multiple regression for area one within Peak Cavern.

Multiple R = 0.618, R squared = 0.382, Significance level = 0.000		
Variable	'B' value	Significance level
Previous weeks Min temperature	0.194	0.000
External wind direction	-0.005	0.000
Previous weeks rainfall	0.062	0.000
Internal cave air pressure	0.033	0.000
Number of pressure changes of magnitude +2 in the previous day	-0.183	0.000
Constant	-32.203	0.000

Table 9.10 Results of stepwise multiple regression for area two within Peak Cavern.

Multiple R = 0.678, R squared = 0.460, Significance level = 0.000		
Variable	'B' value	Significance level
Previous weeks Min temperature	0.115	0.000
External wind direction	-0.003	0.000
Previous weeks rainfall	0.038	0.000
Monthly Rainfall	-0.021	0.000
Internal Pressure	0.019	0.000
Constant	-17.66	0.001

Table 9.11 Results of stepwise multiple regression for area three within Peak Cavern.

Multiple R = 0.668, R squared = 0.4472 Significance level = 0.000		
Variable	'B' value	Significance level
External wind direction	-0.004	0.000
Previous weeks Min temperature	0.172	0.000
Monthly mean temperature	-0.131	0.000
Monthly rainfall	-0.013	0.001
Previous weeks Max external pressure	0.025	0.002
Constant	-21.57	0.016

Table 9.12 Results of stepwise multiple regression for area four within Peak Cavern.

Multiple R = 0.731, R squared = 0.534, Significance level = 0.000		
Variable	'B' value	Significance level
External wind direction	-0.005	0.000
Previous days Min temperature	0.233	0.000
Monthly rainfall	-0.022	0.000
Difference between internal and external air temperatures	-0.130	0.000
Number of weekly changes in pressure (+2)	0.047	0.011
Constant	2.024	0.000

Table 9.13 Results of stepwise multiple regression for area five within Peak Cavern.

Multiple R = 0.806, R squared = 0.650, Significance level = 0.000		
Variable	'B' value	Significance level
Previous days Max temperature	0.207	0.000
External wind direction	-0.004	0.011
Monthly rainfall	-0.032	0.000
Monthly Max temperature	-0.108	0.000
Previous weeks rainfall	0.024	0.001
Constant	3.611	0.000

Table 9.14 Proposed factors, eigen values, percentage and cumulative percentage variation explained

Factor	Eigen Value	Variation (%)	Cumulative(%)
1	10.45	29.9	29.9
2	6.94	19.8	49.7
3	2.77	7.9	57.7
4	2.00	5.7	63.4
5	1.51	43	71.9
6	1.45	4.1	75.5
7	1.25	3.6	78.9
8	1.21	3.5	81.9
9	1.04	3.0	84.6
10	0.92	2.6	86.9
11	0.796	2.2	89.1
12	0.787	1.8	90.9
13	0.644	1.8	92.7
14	0.615	1.5	94.2
15	0.535	1.2	95.4
16	0.417	1.0	96.4
17	0.348	0.9	97.3
18	0.314	0.5	97.8
19	0.185	0.5	98.3
20	0.170	0.4	98.8
21	0.153	0.3	99.1
22	0.104	0.3	99.3
23	0.093	0.2	99.5
24	0.071	0.1	99.7
25	0.48	0.1	99.8
26	0.036	0.1	99.9
27	0.25	0.1	99.9
28	0.19	0.1	100.0
29	0.01	0.0	100.0
30	0.01	0.0	100.0
31	0.00	0.0	100.0
32	0.00	0.0	100.0
33	0.00	0.0	100.0
34	0.00	0.0	100.0

Table 9.15 Correlation coefficient used to determine components which are combined within Factor 1, these represent temperature dependent variables.

Variable	Factor 1
Previous days maximum temperature	0.946
Previous weeks mean temperature	0.939
Previous days mean temperature	0.937
Previous weeks maximum temperature	0.920
Monthly mean temperature	0.917
Difference between the internal and external air temperatures	0.913
Previous days minimum temperature	0.905
Monthly minimum temperature	0.895
Previous weeks minimum temperature	0.894
Monthly maximum temperature	0.874

Table 9.16 Correlation coefficient used to determine components which are combined within Factor 2, these represent atmospheric pressure (static pressure at the time of measurement) dependent variables.

Variable	Factor 2
Previous days pressure	0.943
Previous days minimum pressure	0.941
Previous weeks mean pressure	0.915
Previous days maximum pressure	0.903
Previous weeks minimum pressure	0.806
Internal cave air pressure	0.671

Table 9.17 Correlation coefficient used to determine components which are combined within Factor 3, these represent the number of changes in atmospheric pressure of ± 1 mb.

Variable	Factor 3
Number of pressure changes during the previous week (0)	-0.844
Number of pressure changes during the previous week (-1)	0.718
Number of pressure changes during the day (+2)	-0.615
Number of pressure changes during the day (+1)	-0.532

Table 9.18 Correlation coefficient used to determine components which are combined within Factor 4, these represent the number of atmospheric pressure changes of ± 2 and 3 mb.

Variable	Factor
Number of pressure changes during the previous week (+2)	0.547
Number of pressure changes during the day (-3)	0.535
Number of pressure changes during the previous week (+3)	0.409

Table 9.19 Correlation coefficient used to determine components which are combined within Factor 5, which represents the effects of the external wind direction.

Variable	Factor 5
External Wind Direction	0.614
Number of pressure changes during the day (0)	0.569

Table 9.20 Correlation coefficient used to determine components which are combined within Factor 6, which represent increase in atmospheric pressure of 2 mb.

Variable	Factor 6
Number of pressure changes during the day (+2)	0.635

Table 9.21 Correlation coefficient used to determine components which are combined within Factor 7 which represent the effects of the previous weeks rainfall.

Variable	Factor 7
Previous weeks rainfall	-0.437
Number of pressure changes during the previous week (+3)	0.421
External Wind Direction	0.409

Table 9.22 Correlation coefficient used to determine components which are Combined within Factor 8, which represents the effects of the movement of air within Peak Cavern.

Variable	Factor 8
Direction of air movement at measurement site	0.722
Internal Temperature	0.700

Table 9.23 Correlation coefficient used to determine components which are combined within Factor 9, which represent decreases in pressure by 3 mb.

Variable	Factor 9
Number of weekly pressure changes (-3)	0.509

Table 10. 1 Range of hours spent underground by the different users of limestone caves.

Group	Minimum	Maximum	Mean
1. General Public	0.25	1	0.5
2. Guides in Show Caves	1	700	200
3. Instructors	1	400	100
4. Volunteer Instructors	1	100	20
5. Members of instructed groups	1	10	4
6. Recreational Cavers	1	900	100
7. Speleologists	1	600	150

Table 10. 2 Estimated annual radiation dose (mSv) for users of limestone caves. Based upon mean radon concentrations for England and Wales.

Group	Range of Hours	Average Hours	Min Dose [*1]	Max Dose [*2]	Mean Dose [*3]
1	0.25 - 1	0.5	0.01	0.06	0.03
2	1- 700	200	0.06	45.1	12.8
3	1- 400	100	0.06	25.8	5.5
4	1 -100	20	0.06	6.4	1.3
5	1 - 10	4	0.06	0.6	0.3
6	1 - 900	100	0.06	57.9	6.4
7	1 - 600	150	0.06	38.6	9.6

[*1] Doses based upon minimum exposure times and average radon concentrations
[*2] Doses based upon maximum exposure times and average radon concentrations
[*3] Doses based upon mean exposure times and average radon concentrations

Table 10. 3 Estimated annual radiation doses (mSv) for users of limestone caves by region. Based upon mean exposure times and radon concentrations.

Group	Range of Hours	Average Hours	Peak District	South Wales	North Pennines
1	0.25 - 1	0.5	0.1	0.03	0.01
2	1- 700	200	39.7	12.1	5.2
3	1- 400	100	19.8	6.0	2.6
4	1 -100	20	4.0	1.2	0.52
5	1 - 10	4	0.8	0.2	0.10
6	1 - 900	100	19.9	6.0	2.6
7	1 - 600	150	29.8	9.1	3.9
Doses based upon mean exposure times for each group and mean regional radon concentration.					

Table 10. 4 Estimated annual radiation doses (mSv) for users of limestone caves. Based upon mean exposure times and seasonal mean radon concentrations for England and Wales

Group	Range of Hours	Average Hours	Summer	Winter	Spring	Autumn
1	0.25 - 1	0.5	0.1	0.02	0.04	0.03
2	1- 700	200	41.2	8.5	14.4	10.8
3	1- 400	100	20.5	4.2	7.2	5.4
4	1 -100	20	4.1	0.9	1.4	1.1
5	1 - 10	4	0.8	0.2	0.3	0.2
6	1 - 900	100	20.5	4.2	7.2	5.4
7	1 - 600	150	30.8	6.4	10.7	8.2
Doses based upon mean seasonal radon concentrations and exposure times						

Table 10. 5 Potential radiation doses (mSv) for a 4 hour caving trip based upon maximum and mean radon concentrations determined in 1991 - 1992.

Region	Based upon means	Based upon maximums
Peak District	0.79	4.29
South Wales	0.24	1.89
Mendip Hills	0.11	0.33
North Pennines	0.10	2.52
Portland	0.04	0.09