# GEOLOGICAL CONTROLS ON RADON POTENTIAL IN ENGLAND

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## Abstract

Radon exposure is a chronic and serious geohazard but with the correct knowledge of its distribution provided by an accurate radon potential map, this risk to human health can be reduced through well directed radon testing programmes and building control regulations.

The radon potential map presented here, produced by mapping radon concentrations in homes, grouped by underlying geology, provides the most detailed and accurate assessment of radon in England.

Bedrock and superficial geology associated with the most radon prone areas are investigated using the joint HPA-BGS radon potential dataset, geological information and, where available, soil geochemistry, airborne radiometric or laboratory analysis.

Some of the geological units associated with high radon potential are well known, such as the granite intrusions in south west England, the Carboniferous limestones of Derbyshire and the Jurassic ironstones in Northamptonshire. This study provides a more comprehensive description of the main bedrock geological units associated with intermediate to high radon potential in England including: granites and associated uranium mineralisation in south west England; Devonian, Carboniferous, Permian and Jurassic limestones and dolomites; Devonian, Carboniferous, Jurassic and Cretaceous sandstones; Silurian, Devonian, Lower Carboniferous and Jurassic mudstones; Jurassic ironstones; and some Triassic breccias and conglomerates. Uranium in soil is elevated over many known radon-prone areas but also reflects the accumulation of U in organic-rich soil and peat. Near surface weathering, bedrock fracturing and former working of the ironstones in the English Midlands are all implicated in increased radon potential on these geological units.

Keywords: Geological radon potential, England, radon emanation, radon risk mapping

### 1. Introduction

The rationale for having a good understanding of the controls on radon potential, and for producing the most accurate maps possible is a simple one. Exposure to radon in homes is the second leading cause of lung cancer in the UK after cigarette smoking (AGIR, 2009). The risk increases with long term exposure so it is vital to identify those areas most at risk of high levels, in order to prevent members of the public receiving high exposures to radon.

Radon (<sup>222</sup>Rn) is a natural radioactive gas produced by the radioactive decay of radium (<sup>226</sup>Ra), which in turn is produced from uranium-238 (<sup>238</sup>U). Inhalation of the short-lived decay products of radon has been linked to an increase in the risk of developing lung cancer and has been shown to be responsible for about 2 % of all deaths from cancer in Europe (Darby et al., 2005). In the UK it is estimated that 1100 deaths a year from lung cancer are related to indoor radon (AGIR, 2009). The risk from radon is around 25 times higher for cigarette smokers than non-smokers.

Identifying those areas most at risk of high radon levels facilitates the use of building regulations to help prevent high radon levels in new buildings or to remediate existing buildings (Scivyer, 2007). The radon potential map presented here forms the basis on which building regulations for radon control are regulated in England. It also allows surveys of radon in existing houses to be directed to the highest risk areas. This system focuses efforts on radon reduction where it is most required (HPA, 2010), with the aim to reduce radiation exposure to the population.

Geology is the most important factor controlling the source and distribution of radon (Appleton and Miles, 2005; Appleton and Miles, 2010). Relatively high levels of radon emissions are associated with particular types of bedrock and unconsolidated deposits; for example some (but not all) granites, limestones, ironstones, phosphatic rocks, and shales rich in organic materials.

The release of radon from rocks and soils is controlled largely by the types of minerals in which uranium and radium occur. In magmas, the large, highly charged  $U^{4+}$  ion becomes concentrated in late-stage differentiates, often in accessory minerals such as zircon and allanite. In sedimentary rocks, phosphate and organic complexes may be a more important source of uranium than primary minerals. Once radon gas is released from minerals, its migration to the surface is controlled by the transmission characteristics of the bedrock and soil including: soil permeability, drainage and moisture content; the nature of the carrier fluids, including carbon dioxide gas

and groundwater; and meteorological factors such as barometric pressure, wind, relative humidity and rainfall (Ball et al., 1991; Appleton and Ball, 1995; Appleton and Ball, 2001; Appleton, 2005).

Radon released from rocks and soils is quickly diluted in the atmosphere so that concentrations in the open air are normally very low (about 4 Bq  $m^{-3}$  in the UK; HPA, 2010). Radon that enters buildings can reach high concentrations in some circumstances; the construction method and the degree of ventilation influence the degree of radon build-up. Soil gas radon enters buildings through the cracks and openings in the foundations, aided by the pressure differential there is between the soil and the building. Almost all buildings exert a negative pressure (suction) on the soil because of the natural stack effect - hot air rising and drawing replacement air from under the building - which will be more efficient on higher permeability ground. Wind travelling over the building also contributes to the negative pressure effect. Indoor radon levels fluctuate with outdoor temperature, and hence season (Miles et al., 2012). A person's exposure to radon will also vary according to how particular buildings and spaces are used (NRPB, 2000).

The average indoor radon concentration in the UK is 20 Bq m<sup>-3</sup>, with a range of 5 to 10,000 Bq m<sup>-3</sup>, or more. Based on the estimated risks of lung cancer from prolonged exposure, an Action Level (AL) for radon in homes of 200 Bq m<sup>-3</sup> was established in the UK (NRPB, 1990). The Health Protection Agency (HPA) has recommended that parts of the UK with 1 % probability or more of homes being above the AL should be designated as radon Affected Areas. By July 2010, around 52,900 homes in England had been identified from the national radon database held by the HPA as having radon levels at or above the Action Level of 200 Bq m<sup>-3</sup> (HPA, 2010). This same study identified 121,400 homes in England at, or above, the Target Level of 100 Bq m<sup>-3</sup>

In this paper the results are presented of detailed radon potential mapping for the whole of England carried out jointly by the British Geological Survey and the Health Protection Agency (Miles et al., 2007) focussing on the geological settings of the most distinct radon Affected Areas in England. Given the quantity and range of data available for England, it is possible in some cases to identify the likely cause of elevated radon potential using a range of investigation techniques. Therefore an analysis of the main geological and geochemical associations with intermediate to high radon potential in England are discussed and presented.

# 2. Methodology

## 2.1 Radon mapping methodology

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The cost-effective identification of (a) existing buildings with high radon and (b) those areas where protective measures need to be installed in new buildings depends on the existence of accurate radon potential maps. The probability of homes in the UK having radon concentrations above the UK Action Level is currently estimated by grouping in-house radon measurements by geological units and, within those units, by 1-km grid squares. This methodology is fully described in Miles and Appleton (2005). The Radiation Protection Division of the UK HPA maintains the UK national radon database, which contains the results of more than 500,000 indoor radon measurements. Using this extensive database and digital geological information (Figure 1), the HPA and the BGS collaborated on the production of joint radon potential information for England (Figure 2) and Wales (Appleton and Miles, 2005) which is published as an indicative atlas for England and Wales (Miles et al., 2007).

The geological simplification required for the radon potential mapping took the approximately 2850 named 1:50 000 scale bedrock units in England and reduced this to a simplified bedrock classification system comprising 224 units. The decisions made in the process of grouping geological units were driven by expert knowledge and additional geoscientific datasets (such as those outlined in Section 2.2). The process was, at times, iterative based on analysis of the indoor radon data itself. In addition, the 615 individually named 1:50 000 scale superficial geology units in England were grouped according to a simplified 10 unit system based on permeability and genetic type since these factors most influence radon potential (see Table 1 in Miles and Appleton, 2005). The main simplified superficial geology units are: clay-silt (mainly impermeable alluvium); diamicton (mainly glacial till, which is generally, though not always, relatively impermeable); sand and gravel (mainly permeable glaciofluvial deposits but also raised terrace, raised marine, marine beach and river terrace deposits), and peat. A total of 1285 bedrock-superficial geology combinations resulted from the simplified classification and were used for this mapping.

This type of radon potential map is the most effective and meaningful way of representing spatial variations in radon potential data (Miles and Appleton, 2005). Lateral variation in indoor radon potential within geological units is observed where lithological variations or gradients occur within mapped units, where uranium mineralisation occurs or where transported material was derived from uranium rich areas and is variably distributed. This intra-geological unit variation, where applicable, is reflected in the estimated radon potential for 1 km- geological combination polygons and results in a range of radon potential values for a specific bedrock-superficial combination. This level of detail allows Local Authorities to better target awareness

campaigns, measurement programmes and remediation assistance since the controlling factors for radon generation are better reflected in the map.

#### 2.2 Supporting technique methodologies

Capturing and analysing data on the radon-bearing properties of the rocks or soils themselves – as close to the source as possible – facilitates the investigation of the controlling factors on radon distribution. Laboratory investigation of radon-prone rock units sheds light on the mechanisms of radon emanation and the factors that control its movement in the environment. Studying other geochemical or geological information also allows the indoor radon results to be grouped most appropriately for the joint HPA-BGS radon potential mapping (Miles and Appleton, 2005). In addition, large-scale geochemical data, such as the airborne radiometric and soil uranium data presented here, allows the subtle chemical changes within a mapped geological unit to be detected since this may have a significant bearing on radon potential. Indeed airborne radiometric data and soil geochemistry have been used to compile radon hazard maps of Norway (Smethurst et al., 2008) and inform radon potential mapping in Ireland and the UK (Appleton et al., 2011a; Appleton et al., 2011b; Appleton et al., 2008; Scheib et al., 2006). Taking these factors into consideration, a range of supporting methodologies formed part of this study and are detailed below. The range of investigative techniques and data used in this study allows a more complete picture of the geological controls on radon to be built.

## 2.2.1 Uranium estimates by high-resolution airborne radiometric survey

Equivalent uranium (eU, determined from <sup>214</sup>Bi), equivalent thorium, (eTh, determined from <sup>208</sup>Tl) and potassium (K) were determined by an airborne gamma spectrometry survey, HiRES-1, in 1998 over 14 000 km<sup>2</sup> of Central England (Peart et al., 2004). Procedures for processing the airborne radiometric data were based on those described in the reference manuals of the International Atomic Energy Authority (IAEA, 1991, 2003) and the Australian Geological Survey Organisation (Grasty and Minty, 1995). Gamma radiation measured by the airborne system comes from a thin surface layer of about 30 cm in rock, rather more in less dense unconsolidated material such as mineral soil, to a maximum of a few metres in dry peat. A ternary diagram showing the relative abundance of K, eTh and eU is shown in Figure 3.

#### 2.2.2 Uranium in topsoil

Uranium analysis from two separate soil surveys are presented here, the most detailed of which is the Geochemical Baseline Survey of the Environment (G-BASE), the British Geological Survey's national

geochemical mapping. Over 43, 000 samples from both rural and urban settings, collected since 1986, were available at the time of writing. Soil samples were collected at a density of 1 every 2 km<sup>2</sup> and 4 per km<sup>2</sup> in rural and urban settings, respectively (Johnson et al., 2005). Uranium data from the topsoil sample (5 - 20 cm depth, <2 mm) is report here. Where topsoil data was unavailable, data from the profile sample (35 - 50 cm) has been levelled to a topsoil equivalent concentration. Covering the remainder of England, at a density of one site per 5-km grid square of the British National Grid, is the National Soil Inventory (NSI) data (Rawlins et al., 2012). Since both surveys (Rawlins et al., 2012; BGS, 2010) were analysed by the same laboratories and the analytical data underwent the same procedures, both sets of data are presented here as a combined inverse distance weighted grid (variable search radius, using 5 data points, power of 2; Figure 4).

#### 2.2.3 Ironstone bulk rock radon emanation analysis

Uranium concentration in bulk rock was determined using ICP-MS following a mixed acid digest (HF/HCIO4/HNO3) on ironstone samples collected at 10 sites spanning both the Northampton Sand and Marlstone Rock formations. This allowed the basic composition to be assessed as a factor in radon generation. In order to determine how efficient radon emanation was in each of the ironstones under investigation, selected samples underwent testing (e.g. Andrews and Wood, 1972). The rock samples were cut with a rock saw to give flat surfaces on all faces to enable calculation of surface area to the nearest 5 cm<sup>2</sup>. The dry sample volume and volume of the internal porosity of each sample to the nearest 1 ml was measured using water displacement. The rocks were then immersed in reverse osmosis water, sealed in glass Kilner jars and allowed to stand for 25 days to allow the radon to reach secular equilibrium. Care was taken to exclude any air space in the jars to maximise radon concentration in the water. After this period, duplicate 10 ml aliquots of the water were taken by syringe from the sample container, placed into a low background glass liquid scintillation counting vial containing 10 ml of toluene-based LSC cocktail and shaken. Liquid scintillation counting was performed on a LBK Wallac Rackbeta counter, including duplicates, standards and blank samples. The results were calibrated with an in-house radium standard solution. This provides a stable source of radon and had been compared to two certified standard radium solutions.

## 2.2.4 Ironstone petrographic studies: alpha radiography and SEM petrography

Autoradiographic registration of alpha-particle emission from geological materials makes it possible to locate sources of high radioactivity prior to identification of the source by chemical or mineralogical methods. With a view to better understand the source geometry of radon in ironstone, 37 samples from the Northampton Sand

Formation and the Marlstone Rock were sliced, polished and analysed by autoradiography prior to a detailed microscopic study.

Alpha-particle autoradiography was carried out following the technique described by Basham (1981), using 'CR39' plastic film. This is a highly sensitive thermoset polymer with particularly high optical qualities, with high track resolution and uniform response to alpha particles of energies up to 6 MeV. Each specimen slice was first cleaned with ethanol to remove all traces of preparatory residues (e.g. rock-saw lubricants etc.) that could contaminate the film. A piece of CR39 film was then placed on the smooth surface and the sample was packed tight in a protective parcel. All specimens were left undisturbed for 7 weeks to expose the film to radioactivity in the samples. After exposure, the film was carefully removed, cleaned with ethanol and etched by suspension in a beaker containing an aqueous solution of 6N NaOH\* at 80 °C. During etching the film was periodically removed, rinsed with water, and placed under an optical microscope to monitor track-etch development. After 4-5 hours, the tracks were visible under a microscope and the film was rinsed with water and cleaned again with ethanol. The radiographs were examined under an optical microscope to identify alpha-particle track marks and observe their spatial distributions. These were mapped out onto the film and matched back to the original slices. In order to confirm the petrographic and mineralogical identity of the radiographic hotspots, selected samples were subsequently prepared for more detailed examination by scanning electron microscopy (SEM). The areas selected for SEM analysis were prepared as much smaller (3 to 4 cm diameter) polished blocks, which were carbon-coated before examination in the SEM. A Leo 435 VP Scanning Electron Microscope was used, operated in Back-Scattered Electron Imaging mode, which produces images where the brightness is related to average atomic density, thus allowing regions and phases of differing chemical composition to be distinguished. In addition, semi-quantitative chemical analysis of individual phases was also performed using an ISIS 300 Energy-Dispersive Spectrometer.

#### 3. Results and discussion

A range of bedrock types contribute to the radon prone areas of England, shown in Figure 2. Following an initial overview of uranium in topsoil across England, bedrock lithologies associated with the most radon prone areas are investigated using the joint HPA-BGS radon potential dataset, geological information and, where available, soil uranium, airborne radiometric or laboratory analysis. We then consider the radon potential of superficial deposits, looking at the effect they may have on the radon potential of underlying bedrock units, and in their own right.

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#### 3.1 Uranium in topsoil across England

Uranium in topsoil across England demonstrates mean and median values of 2.16 and 2.11 mg kg<sup>-1</sup>, respectively (n= 48134, standard deviation 1.03). One-way analysis of variance (ANOVA) demonstrates that bedrock-superficial geology combination (the same groupings used for the HPA-BGS radon potential mapping) explains 26.6 % of the variance in topsoil uranium, which is approximately the same as for indoor radon (Appleton and Miles, 2010). Ninety per cent of the soil uranium data is below 3.01 mg kg<sup>-1</sup>, with only 1% of data exhibiting values above 4.80 mg kg<sup>-1</sup>. The majority of elevated concentrations relate to:

1. Carboniferous-Permian granite of SW England (DCFEIN and CARBGR, Table 1; Figure 4, No: 1) most likely due to higher uranium in the granite parent material combined with the affinity of U for organic rich (peaty), upland soils;

2. Triassic and Jurassic (Lias Group) mudstone, siltstone, limestone and sandstone sedimentary assemblages in Somerset (TRIMD and LLI, Table 1; Figure 4, No: 2);

3. Silurian mudstone and siltstones in Hereford and Worcester, particularly where covered with peat (SPRDMDMIX, Table 1 (up to a maximum of 88.1 mg kg<sup>-1</sup> U in topsoil with cover of peat); Figure 4, No: 3). Uranium is strongly bound and therefore can become concentrated by the organic compounds such as humic acid in peaty soils. This sample on peat with exceptionally high uranium also displays elevated selenium, molybdenum and vanadium, and warrants further investigation (Rawlins et al., 2012 );

4. Lower Cretaceous sandstones and clays of the Wealden Group in south-east England (WGS and WGM, Table 1; Figure 4, No: 4);

5. Holocene peats of the Fen Basin (e.g. AMKIM\PEAT and GLT\PEAT, Table 1; Figure 4, No: 5). This U anomaly corresponds closely to the boundary of exposed peat and the topography here would suggest that stream or groundwater flooding off the adjacent higher ground is being concentrated in the peat due to strong binding mechanisms (BGS, 2010);

6. Carboniferous limestone of the Peak District and north into Cumbria and Northumberland (DINLM and DINMDMIX, Table 1; Figure 4, No: 6); and

Volcanic rocks surrounding the Cheviot granite pluton in Northumberland (DLDMA, Table 1; Figure 4, No: 7).

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Low soil concentrations of uranium were found on: the high ground of the Pennines (e.g. NAMMDMIX, Table 1; Figure 4, No. 8); pebbly, gravelly sandstone of the Sherwood Sandstone Group (e.g. TRISD, Table 1; Figure 4, No. 9); soils of East Anglia over White Chalk or glacial till, sand and gravel (e.g. WCK, Table 1; Figure 4, No. 10); and, soils of southern England over the Lower Greensand and Chalk groups (e.g. WCK and LGS, Table 1; Figure 4, No. 11). Soils in the urban environment of the Greater London Authority area (GLA) are notably low in uranium (e.g. THAMC, Table 1; Figure 4, No. 12).

### 3.2 Igneous and metamorphic bedrock

## 3.2.1 Felsic igneous lithologies

The granites in SW England are characterised by: high uranium concentrations in rock (Dartmoor, 3.2-35.5; St Austell, 0.6-66.6; and Land's End, 2.6-38.2 mg kg<sup>-1</sup> U (Simpson et al., 1979)) and soil (Figure 4); a deep weathering profile; and uranium in a mineral phase which is easily weathered. Although the uranium may be removed, radium generally remains in situ which continues to produce radon gas (Ball and Miles, 1993). The late Carboniferous (Stephanian) Dartmoor granite intrusion (DCFEIN) exhibits the highest radon potential values for England with an average radon potential of 66.1 % (n= 1763, Table 2, Figure 2). The other extensive outcrops of Carboniferous-Permian granite on the SW peninsula (CARBGR) including the Bodmin, St Austell, Carnmenellis and Land's end intrusions, display a high average radon potential of 43.9 %>AL, ranging up to 89.1 %>AL (Table 2, Figure 2). These granites are uraniferous with concentrations ranging from 6- 25 mg kg<sup>-1</sup> although higher values occur rarely (Ball and Miles, 1993). The greatest proportion of the uranium in the granites occurs in the mineral uraninite, which is often weathered near the surface. Much of the uranium is removed by weathering but a large part of the radium remains in the relict minerals and, because of the greater specific surface area, these minerals in the weathered rock become more efficient radon generators than expected from the remnant uranium. In addition, uranium mineralisation has been known in the area for at least a century and the minerals were mined, originally for glass colouring, before reworking of some deposits for their radium content. Two types of uranium occurrences are recognised: (i) within high-temperature tin/ tungsten/ copper lodes and (ii) in later 'crosscourses'. Most, but not all, of the known uranium vein mineralisation occurs within about 2 km either side of the granite contact. Uranium mineralisation ranges from small, high-grade, to more disseminated occurrences (Simpson et al., 1979). Radon is easily emanated from the host rock and high values of radon have been measured in ground and surface waters (110-740 Bq/l; Burgess et al., 1982; Edmunds et al., 1984, 1987) and also in soil gas (frequently > 400 Bq/l; BGS unpublished data and Gregory, 1987).

There are extensive areas of alteration within the granites; tourmalinisation, greisening, haematisation, and kaolinisation all affect substantial volumes of rock. Of these, kaolinisation is the most important because it is very widespread and usually resulted in a substantial decrease in the uranium concentration (Ball and Miles, 1993). Of these granites the St Austell granite exhibits the most internal variation in radon potential (Figure 5); the western part is intensely altered (kaolinised) with much of the original uraninite removed, probably during Tertiary times (between 65 million and 1.8 million years ago) whilst the eastern portion remains relatively fresh but with surface alteration of the uraninite. The eastern portion of the granite is more radioactive than the western portion and there is a zone of higher radioactivity to the south west of the granite, which corresponds to an increase in uranium concentration in the country rocks associated with uranium vein mineralisation (Appleton and Ball, 1995).

#### 3.2.2 Mafic Igneous lithologies

Mafic igneous rocks have a fairly widespread distribution across England, but due to their nature do not generally occupy extensive areas. In addition, since they often form high or uneven ground, these areas are often not densely populated. In theory mafic igneous rocks are not associated with high radon potential. Houses with high radon assigned to mafic igneous lithologies may in fact be situated on a more radon prone rock type into which the dyke or sill has been intruded. The width of these narrow units becomes exaggerated on the 1: 50,000 scale digital geology resulting in some houses being wrongly assigned to the mafic igneous rock type.

The early Devonian Cheviot Volcanic Formation surrounding the Cheviot granite pluton in Northumberland, northern England, is made up of a thick pile of andesitic lava with associated pyroclastic and reworked volcanic rocks, subordinate biotite-phyric rhyolite and tuff (BGS, 2011; Stone, 2008). Due to the low population in this area, there are limited radon measurements on this unit. The average radon potential here is 3.5 %>AL (DLDMA, n=32, Table 2).

Middle Devonian mafic igneous intrusive rocks crop out at Lizard Point on the extreme south-west of the SW peninsula with smaller scattered occurrences elsewhere on the peninsula. This generalised geological grouping encompassing, for example, peridotite and serpentinite of the Lizard complex (BGS, 2011), has an intermediate average radon potential of 8.4 %>AL (DCMAIN) despite the largest outcrop area being below the Radon

Affected Area threshold of 1%>AL. A further generalised grouping of Devonian age basaltic lava, tuff and pyroclastic rock displays a high average radon potential of 10 %>AL (DCMA, Table 2). This group crops out in a relatively sporadic and elongate manner on the SW peninsula from west of Paignton in Devon, across to around St Ives and Penzance in Cornwall.

Thin slithers of Carboniferous mafic igneous intrusive rock, such as quartz-microgabbro of the Whin Sill Complex occur across England, particularly in the North. This generalised geological grouping displays an average of 5.6 %>AL (CARBMAIN, Table 2) but this ranges up to 50.4 %>AL. This high degree of variation probably reflects the range of compositions this generalised grouping is likely to contain, combined with a higher degree of uncertainty when dealing with thin geological units (Hunter et al., 2009). For example, indoor radon measurements may have been incorrectly assigned to narrow mafic intrusions rather than the surrounding bedrock (such as Carboniferous limestone) due to the amplification of the width of narrow units on the 1: 50 000 scale geological map. Relatively narrow outcrops of metamicrograbbro to the north and east of the Dartmoor Granite intrusion frequently have >12% >AL. This is likely to reflect uranium-bearing metalliferous veins, which are relatively common in mafic rocks within the Dartmoor Granite aureole. Basaltic lava and tuff from the Lower Carboniferous Dinantian displays a high average radon potential of 11.1 %>AL (DINMA, Table 2). This unit is also distributed in thin, elongate structures and encompasses, for example, the Litton Tuff member of the Carboniferous Limestone Supergroup. This is dominated by volcanic breccias (agglomerate) with clasts which include some limestone and shows an elevated maximum eU recording of 7.1 ppm from the HiRES-1 airborne survey (Table 4).

### 3.2.3 Metamorphic lithologies

Two distinctive areas of Mica Schist occur on Lizard Point in south Cornwall and Start Point in south Devon (see Figure 1 for locations; Jackson, 2008). These Devonian metamorphic rocks collectively display an average radon potential of 7.4 %>AL (DCMETA). Small uraniferous mineral occurrences near Start Point are not related to the granite contact (Ball and Miles, 1993).

### 3.3 Sedimentary bedrock types

## 3.3.1 Limestone

There are a number of radon Affected Areas associated with limestone units in England – some of which are spatially extensive – ranging from Devonian to Jurassic in age. Although the concentration of uranium found in

limestone is generally fairly low, high emissions of radon can occur on limestone due to the high specific surface area of the uranium minerals and high bedrock and soil permeability. Uranium is often uniformly distributed within limestones (usually < 10 ppm U); being mostly associated with finely divided organic matter in the matrix of bio-classic limestones. The higher specific surface area of the U minerals in limestones, relative to resistate U minerals in granites for example, allows efficient release of radon into air and water (Appleton and Ball, 1995).

Lower Carboniferous Dinantian limestone crops out extensively, mostly in north England, and displays the highest recorded indoor radon measurement on limestone bedrock of 6252 Bq m<sup>-3</sup>. Over 13, 000 indoor radon measurements have been made on this group of limestone formations, with an average of 13.7 % of dwellings expected to exceed the Action Level (DINLM, Table 3). The HiRES-1 airborne radiometric survey of the Midlands shows that the highest eU measurements were recorded on this bedrock (Table 4). The Dinantian limestone is clearly elevated in uranium relative to surrounding rocks (Figure 3) although most of the eU signal detected by the airborne survey would reflect the nature of the soils overlying the limestone. Bedrock permeability is also likely to be high in this area due to the nature of limestone. There are also extensive areas of mixed, limestone dominated Dinantian sedimentary rock in northern England which are also radon affected (DINLMMIX, Table 3, Figure 3). This group comprises grainstone, oolite, calcarenite, dolostone, sandstone, siltstone, mudstone and conglomerate lithologies. Elsewhere bioclastic limestones alternate with coals typically in regular cyclothemic sequence (BGS, 2011; Stone, 2008).

Other extensive limestone areas include the Great Oolite Group limestones and the older Inferior Oolite Group limestone. Both from the Jurassic, they crop out extensively from around Kingston-Upon-Hull in the north east (see Figure 1 for locations), to south-west England. The Great Oolite Group contains argillaceous formations, is dominantly calcareous but actually rarely oolitic. It sits unconformably on top of the Inferior Oolite Group, which itself comprises limestones with thin marls and mudstones that may variably be fossiliferous, bioturbated, ooidal, cross-bedded and with shell-fragments (BGS, 2011). Many indoor radon measurements have been recorded on these limestones and results show a range of radon potential values with some instances of the maximum recorded indoor radon level exceeding the Action Level by more than a factor of 10 (prefix GOG codes and INOLMST, Table 3). The Inferior Oolite Group limestone, in particular, stands out as being elevated in uranium relative to potassium or thorium (deep blue unit labelled INO-LMST in Figure 3) although the

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arithmetic mean eU for this unit is well within the range of uranium values measured by the airborne survey across many different geological units, presented in Table 4.

Another Jurassic limestone occupying much smaller areas from the east of Oxford, south to the Isle of Portland, the Portland Group (Portland stone formation) limestone displays an average radon potential of 9.3 %>AL (PST, Table 3). The Portland Group is approximately 75 m thick; the upper portion comprising predominantly limestone (micrites, wackestones, packstones and grainstones) with the lower part being predominantly argillaceous in nature encompassing dolomitic sandstones, some mudstones, shales, and thin beds or nodular layers of micrite (Jackson, 2008). It was elevated indoor radon measurements on this bedrock unit, made as a result of the map presented here, which resulted in a school closure on the Isle of Portland, pending remedial action, in summer 2010

 $(http://www.dorsetecho.co.uk/news/8488994.Portland\_s\_Grove\_Infant\_School\_reopens\_after\_radon\_alert/\ ).$ 

The early Jurassic Lower Lias limestone and the Lower and Middle Jurassic limestone dominated sedimentary rocks are less spatially extensive than the Inferior and Great Oolite limestones but nonetheless have considerable numbers of indoor radon measurements recorded; and are radon affected (LLILMST and LMJ, Table 3).

An extensive swathe of Permian dolomite, the Cadeby and Brotherton formations, extends from Newcastle upon Tyne to Nottingham (see Figure 1 for locations) and displays an average radon potential of 1.6 %>AL but this ranges up to 64 % near Mansfield in Derbyshire (PERDO). Here, the Cadeby Formation consists of dolostone, commonly oolitic or granular, with subordinate mudstone, dolomitic siltstone and sandstone (BGS, 2011).

Older than all other limestones in England, but occupying a relatively restricted area in SW England, Devonian limestones (split into lower, middle and upper Devonian) are radon affected and display values well in exceedance of the Action Level (DCUDLM, DCMDLM and DCLDLM, Table 3).

#### 3.3.2 Sandstone and sandstone dominant lithologies

The Cretaceous Upper Greensand extends from south of Exeter, north-east to Luton with a further arcuate outcrop south of London. This Formation comprises fine-grained sand and sandstone and is variably silty, glauconitic and shelly. It displays an average radon potential of 1.7 %>AL but this ranges up to 41.7 %>AL in limited areas (UGS, Table 5).

The Upper Lias Bridport Sand Formation is made up of fine-grained and very fine-grained sand, is locally calcite-cemented and displays an intermediate average radon potential of 2.6 %>AL (ranging up to 13.1%>AL, ULIBS, Table 5).

More extensive across England than the younger sandstones described above, a group of Permian sandstone formations have a low average radon potential of 0.5 %>AL (PERSD, Table 5). The vast majority of this outcrop is not classed as radon affected but some areas on the SW peninsula display intermediate radon potential. The maximum recorded indoor measurement on this sandstone is 1545 Bq m<sup>-3</sup>, which is over 7 times greater than the current UK Action Level. The portion of this unit assessed by the HiRES-1 airborne survey looks to be dominated more by thorium than uranium (Figure 3, Table 4). This geological grouping includes: coarse-grained, cross-bedded aeolian sandstone in the north; very fine- to medium-grained sandstone, cross-stratified with subordinate beds of mudstone and conglomerate in the Midlands; and fine- to medium-grained, cross-stratified aeolian sandstone in west England (Shropshire and north to Merseyside and Greater Manchester). These units are largely not radon affected. In the southwest however, (largely in Devon) the sequence is increasingly dominated by breccias (sandstone, slate and quartz porphyry clasts), with subordinate sandstone, often arising from alluvial fan deposits (BGS, 2011; Jackson, 2008; Stone, 2008). Some of these areas are moderately radon affected.

Carboniferous Westphalian sandstone displays an average radon potential of 1.3 %>AL (WESDSD, n= 422, Table 5). The small portions of this unit that are radon affected, close to the Welsh border, have a more limestone or ironstone influenced composition than elsewhere in the outcrop. A sandstone group consisting of Duckmantian and Bolsovian age strata (also of the Westphalian) is not radon affected in the north where it outcrops close to Newcastle but has some radon affected areas on the West- South Yorkshire County border. This unit has an average radon potential of 1.7 %>AL but ranges up to 10.2 %>AL in some limited areas (WESDUSD, Table 5). Likewise, a further, generalised group of sandstones belonging to the Westphalian (Langsettian substage) has a lower radon potential in its northern outcrops; with intermediate to high radon potential from close to Sheffield, south to Ilkeston. There is considerable variation in this outcrop however (WESLASD, Ave 1.2 %>AL max. 35.9%, Table 5). This north to south increase in radon potential (and considerable spatial variation) may reflect changes in composition and permeability. The more northern outcrops are interbedded with mudstone and siltstone with coal seams in the upper part whilst further south the sandstone is fine-grained, micaceous and carbonaceous (BGS, 2011).

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The Namurian (Holsworthy Group) sandstone, which covers a wide area of Devon and the extreme north-Cornish coast, displays a relatively low average radon potential of 1.9 %>AL (NAMHSD) but ranges up to 30.9 %>AL, (Table 5) and comprises:

(a) thick-bedded, somewhat argillaceous and silty sandstones with additional mudstone interbeds; locally containing thin ironstone beds and black sulphurous shales with fossil-bearing calcareous nodules; and

(b) rhythmically bedded mudstones and subordinate sandstones and siltstones. Sandstone percentage varies from 20-75%, both vertically and laterally. Fossil-bearing calcareous nodules and scattered ironstone nodules are present in the upper part of the Formation (Jackson, 2008).

Namurian sandstone of the Millstone Grit Group forms an important and extensive outcrop trending north-south down through northern England. This coarse-grained feldspathic sandstone displays an average radon potential of 1.0 %>AL, ranging up to 19.2 %>AL (NAMSD, Table 5). Dinantian sandstone occurs in narrow but widespread outcrops, particularly in north England, and has an intermediate average radon potential of 3.8 %>AL (DINSD, n=501, Table 5).

Occupying a discrete area in Cornwall on the SW peninsula (north and south-west of Falmouth Bay), mid-Devonian sandstone has a high average radon potential of 12.4 %>AL (DCMDSD, Table 5). This unit belongs to the Portscatho Formation and comprises interbedded sandstone beds with slaty mudstone. Also from the mid-Devonian and occupying a limited area on the north Devon Coast is the Hangman Sandstone Formation. This thickly bedded and laminated fine- to medium-grained sandstone displays an average radon potential of 6.1%>AL (ranges up to 20.1 %>AL, n=136, DDMDSD, Table 5).

Running in a narrow band from Brixham in Devon, south of Plymouth to north of Newquay in Cornwall, lower Devonian sandstone has a high average radon potential of 15.5 %>AL, ranging up to 47.6 %>AL (DCLDSD, Table 5).

Sandstone of the Bailey Hill Formation (Silurian, Ludlow) crops out close to the Welsh border and has an intermediate average radon potential of 7.1 %>AL (SLUDSDMIX, Table 5).

### 3.3.3 Mudstone and mudstone dominant lithologies

Jurassic Inferior Oolite Group (Grantham Formation) mudstone dominant sedimentary rocks display an average radon potential of 13.7 %>AL (INOGRF, Table 6), higher on average than the overlying Inferior Oolite

Limestone (see section 3.2.1, av. 9.9%, Table 3) and only slightly less than the ironstones of the underlying Northampton Sand Formation (av. 15.5%, Table 7). They occur in narrow outcrops and comprise a range of mudstones, sandy mudstones and argillaceous siltstone-sandstone, which is commonly ferruginous, contains abundant plant debris and may be locally carbonate-cemented (Jackson, 2008). It is possible that the radon is derived principally from the ferruginous matrix material of the Grantham Formation mudstone.

One of the most spatially extensive mudstone dominant sedimentary units is the Lower Lias extending from near Middlesbrough in the north, to the Taunton area in Somerset to the south. Over 21,000 indoor radon measurements have been made on this geological grouping which displays an average radon potential of 1.2 %>AL (LLI, Table 6). This geological grouping consists of laminated shales and mudstones with locally incorporated limestone beds comprising abundant argillaceous limestone, phosphatic or ironstone (sideritic mudstone) nodules in some areas (Jackson, 2008). In the Midlands this unit is largely not radon affected but some areas to the south-east of Bristol on the Somerset-Avon border where limestone beds are common, and to the south and east of Lutterworth in South Leicestershire a higher radon potential ranging up to 59.1 %>AL is displayed. The Jurassic Middle Lias (MLI) comprises a mixture of dark, silty and sandy mudstone with limited interbeds of ferruginous limestone (some ooidal) and sandstone (BGS, 2011). Elsewhere the MLI contains abundant, argillaceous limestone and phosphatic or ironstone nodules. The average radon potential is 3.4 %>AL (MLI, Table 6) but this does increase up to 42 %>AL in the Banbury – Chipping Norton area, where the Dyrham Formation is overlain by the Marlstone Rock which has been worked considerably in this area (Whitehead, 1952; Horton, 1987). The elevated radon recorded to the MLI here may suggest that there may possibly be basal ironstone remaining, post working, where the Dyrham Formation is mapped. Alternatively, increased radon on the narrow MLI units may also indicate that indoor radon measured over the Marlstone Rock (a radon affected unit, see section 3.3.4.2) may be assigned to the MLI grouping due to geological boundary and house location uncertainties, which are amplified on thin units.

Mudstone dominant sedimentary rocks from the Namurian (Holsworthy Group, principally Crackington Formation) occupy an extensive area of Devon and north Cornwall (average 5.2%>AL, NAMHMDMIX, Table 6). Radon potential greater than 6% and ranging up to 79.1 %>AL occurs both immediately to the north of the Bodmin granite and north and east of Dartmoor granite, where there is evidence of high uranium concentrations related to uranium vein mineralisation, most of which occurs within 2 km of the granite contact (Ball and Miles, 1993). High radon potential also characterises this bedrock to the north of Tiverton on the SW peninsula, where this generalised geological grouping encompasses a condensed marine succession, the Dowhills mudstone, that occurs diffusely just below the Crackington Formation. Condensed marine successions are known to contain enhanced levels of natural radioactivity (Appleton and Ball, 1995). The Holsworthy group comprises rhythmically bedded mudstones and subordinate sandstones and siltstones. Scattered ironstone nodules are present in the uppermost part of the Formation and elsewhere fossil-bearing calcareous nodules occur (Jackson, 2008).

Lower Carboniferous Dinantian mudstone and mudstone dominant sedimentary rocks, cropping out in the far north of England and in the Midlands surrounding the Derbyshire Dome, display an intermediate average radon potential of 4.5 %>AL (DINMDMIX, Table 6). This ranges up to 54 %>AL both in the north in the vicinity of the Cheviot Pluton and Volcanic Formation, and in the Midlands surrounding the Carboniferous limestones of the Derbyshire Dome. In the Midlands, the HiRES-1 airborne survey data shows this geological group, known locally here as the Bowland Shale, is clearly elevated in uranium relative to many other rock-units (Figure 3). In this area this fissile and blocky mudstone is weakly calcareous, with subordinate sequences of interbedded limestone and sandstone. In places it is organic-rich, marine bands are also present as well as volcaniclastic debris flows in part of the Formation. The depositional and diagenetic environment of many black shales leads to enrichment of uranium. It is normally located mainly in the fine grained mud matrix, where it may be present at levels up to 20 mg kg<sup>-1</sup> U, and also in organic-rich bands at concentrations up to 40 mg kg<sup>-1</sup> U. Uranium is rare in detrital phases and may also be remobilised and relocated on iron oxides (Appleton and Ball, 1995). The Bowland (formerly Edale) Shale in Derbyshire comprises uranium enriched black shales containing 5-10 mg kg<sup>-</sup> <sup>1</sup> U (Bottrell, 1993) whilst the basal mudstones in the Chapel en le Frith area have 10-60 mg kg<sup>-1</sup> U (Ball et al., 1992). In the Chapel en le Frith area, pre-glacial karst infill has introduced debris from the overlying uraniferous black shales in a disaggregated form deep into the underlying limestone. This may explain the high radon potential encountered in the apron reef limestone facies forming the border of the limestone mass in the Peak District (Derbyshire). It should be noted that apron reefs also contain frequent uranium concentrations in phosphatic nodules and in metalliferous veins and joint surfaces.

In the north, close to the border with Scotland, rocks of the Lyne and Ballagan formations represent cyclical sequences of sandstone, siltstone, mudstone and thin limestones, with localised dolomitic cement stones, gypsum and common desiccation cracks (BGS, 2011). These features contribute to the radon potential of this

unit by increasing bedrock permeability (e.g. desiccation cracks) and exhibiting a more limestone-rich composition.

A further two Dinantian mudstone dominant sedimentary geological groupings display high average radon potential:

 Largely outcropping to the NE of Preston, the Bowland Shale Formation comprises mudstone with subordinate and variable detrital limestone, siltstone and sandstone. This group also includes reef limestone in places – which is well known for its elevated radon potential – and displays an average radon potential of 17.6 %>AL (ranges up to 67.8 %>AL, DINMD, Table 6). Airborne survey measurements up to 10.8 ppm eU were recorded on this unit (Table 4).

2. The Teign Valley Group mudstone dominant sedimentary units crop out mainly near Launceston (Cornwall) and south of Buckfastleigh (Devon). This locally siliceous, mudstone with laminae and thin beds of siltstone displays an average radon potential of 23.0 %>AL (DINTVMDMIX, Table 6). Intermediate to high concentrations of U (5-21 mg kg<sup>-1</sup>) characterise this rock unit (Appleton and Ball, 1995), locally called the Meldon Shale and Quartzite Formation.

Extensive areas of Devonian mudstone make up large swathes of the SW peninsula; having been split into Upper, Middle, and Lower Devonian. These units display higher radon potential in Cornwall (DC prefix, Table 6) than in Devon (DD prefix, Table 5) although both are radon affected and display intermediate to high radon potential. The Upper Devonian slaty, silty mudstone which is locally interbedded with thin bioclastic limestone and dolomite beds, generally displays the highest radon potential in this grouping (Cornwall average: 16.8 %>AL, ranging up to 89.4 %>AL, DCUDMD, Table 6). Some of the highest recorded radon levels in England have been measured on these mudstone units; in places more than 70 times greater than the UK Action Level. High radon potential is generally associated with the zone of uranium mineralisation that occurs around the granites in Cornwall and Devon.

Displaying generally lower radon potential is the Silurian (Přídolí) mudstone dominant sedimentary group. Largely outcropping in Hereford and Worcester, this unit extends north into the Shropshire hills and south-west across Wales. This red and silty mudstone with calcretes and sandstones displays an average radon potential of 1.3 %>AL in England (ranging up to 29 %>AL, SPRDMDMIX, Table 6). Older still, the Silurian (Ludlow) mudstone dominant sedimentary rocks of the Lake District display a low average radon potential of 0.3 %>AL although this ranges up to 18.7 %>AL (SLUDKMDMIX, Table 6). This silty mudstone with subordinate siltstone and sandstone crops out in north-west England (Stone, 2008).

#### 3.3.4 Ironstones

Jurassic ironstones, consisting of several formations, crop out from Lincolnshire, through east Leicestershire, Northamptonshire and extend south-east, through Oxfordshire. These ironstones have been known for some time to be associated with elevated radon (Sutherland, 1991; Appleton and Ball, 1995; Sharman, 1995). Detailed investigations carried out on two of the ironstones, the Northampton Sand Formation and the Marlstone Rock Formation which are both significant in terms of radon potential in England, are reported below.

### 3.3.4.1 Northampton Sand Formation

The Jurassic Inferior Oolite Group, Northampton Sand Formation exhibits a high average radon potential of 15.5 %>AL (INONS, Table 7) and ranges up to 86.9 %>AL. Indoor radon measurements in excess of 4000 Bq m<sup>-3</sup> have been recorded on this bedrock formation. The Northampton Sand Formation consists of ferruginous sandstones and oolitic ironstone with a basal layer up to 30 cm thick containing phosphatic pebbles (BGS, 2011). Whereas the ferruginous sandstones and ironstones mainly contain low concentrations of U (< 3 ppm), the phosphatic pebbles contain up to 55 ppm with a mean of 20 ppm (Sutherland, 1991). However, it is probable that the bulk rock itself – which in many cases contains radium in a disseminated form – contributes more radon to the overall level of radon emissions than the thin U-enriched phosphate horizons. Radon gas can migrate to the surface relatively rapidly as the Northampton Sand Formation has extensive 'boxwork' style weathering in which Fe-rich clays and carbonates have been largely altered to a mixture of permeable goethite and haematite (Hodgkinson, 2012). The unit also includes lenses of mudstone and limestone in places.

Laboratory investigation of the Northampton Sand Formation (22 rock samples) revealed an average uranium concentration of 2.24 ppm (range 0.60 - 5.01 ppm, n=22, analysed by ICP-MS) and an average <sup>226</sup>Ra activity of 30.9 Bq kg<sup>-1</sup> (range 7.5 – 73 Bq kg<sup>-1</sup>, n=22, analysed by Liquid Scintillation Counting). U and <sup>226</sup>Ra correlate very strongly in this rock unit suggesting there is no obvious disequilibrium (r=0.879, p<0.0005).

3.3.4.2 Marlstone Rock Formation

The Middle Lias Marlstone Rock Formation, which is an ironstone with a basal phosphatic pebble bed, also displays a high average radon potential of 15.8 %>AL (Table 7), ranging up to 90.8%>AL. This unit occurs in Lincolnshire, Northamptonshire and Gloucestershire but is more extensively developed in Leicestershire, and also Oxfordshire around Banbury (Hodgkinson, 2012). Elevated eU, and in particular elevated eTh, is associated with the Marlstone Rock (Figure 3 & Table 4).

Laboratory investigation of the Marlstone Rock Formation (17 rock samples) revealed an average uranium concentration of 2.56 ppm (range 1.17 - 5.40 ppm, n=17, analysed by ICP-MS) and an average <sup>226</sup>Ra activity of 33.1 Bq kg<sup>-1</sup> (range 11.5 - 83.0 Bq kg<sup>-1</sup>, n=17, analysed by Liquid Scintillation Counting). U and <sup>226</sup>Ra also correlate very strongly in this rock unit suggesting there is no obvious disequilibrium (r=0.839, p<0.0005).

#### 3.3.4.3 SEM, Autoradiography and XRD investigation of ironstones

The ironstones consist mostly of varying proportions of carbonates, detrital silicates and Fe-rich phases. Where the iron-rich phases are primary, they are present as Fe-rich clay ooliths and matrix; but both these phases are often altered to fine-grained Fe oxides, especially towards the surface and where they have been exposed for longer. XRD analysis identified the major Fe oxide phase as goethite.

The carbonates, whether they occur as coarse bioclasts (common) or as sparry matrix, are clearly not a source of alpha particles. The carbonate cements all appear to greatly reduce both porosity and matrix permeability in the rocks and are therefore likely to have a significant effect on the ability of radon to migrate. In the Northampton Sand Formation, carbonate cement is restricted to limited areas of the lower levels but in the Marlstone Rock Formation, it is ubiquitous (although developed to varying degrees across the sampled outcrop). Sparry calcite cement was also observed to be a common feature in Marlstone Rock samples examined (Figure 7). Poorly developed porosity results from this feature.

The detrital silicate-based components include detrital zircons and occasionally monazites, and these act as small, intense point sources of alpha particles. However, these are not particularly concentrated, no more so than in many other sedimentary rocks, and are not likely to be the main source of radon.

The fine grained Fe-rich matrix in these rocks acts as a diffuse source of alpha particles (Figure 8). The detected alpha particle density from the matrix is far lower than from the detrital zircons and monazites, but volumetrically it represents a far greater alpha source. Furthermore, this matrix is fine grained and has a high porosity and is therefore more relevant as a source of radon in the disperse phase than point sources in the rock.

The matrix consists mostly of iron rich clays (chlorites and possible Fe-rich swelling clays) altered in places to goethite. Because of the fine grained matrix it was not possible to identify which phase gave rise to the alpha particles. However the Fe-rich clays, when present within ooliths, do not appear to be a significant identified source of alpha particles, so it is likely that most of the alpha particles originate from goethite.

Phosphate is present and is always associated with alpha emissions. Sedimentary phosphatic minerals are known to have the potential to accumulate uranium during deposition or diagenesis. It occurs mainly as pellets but also forms a minor matrix cement and rarely a veinfill. The pellets are rare and tightly cemented (Figure 9), and so are not likely to be the main contributor to radon, when compared to the more volumetrically significant fine-grained matrix. Moreover, the phosphate was not identified by SEM as a widespread matrix cement, and therefore it is not likely be the dominant source of the observed, matrix radioactivity. In most samples this phase was pure Ca phosphate, but an Al-bearing phosphate was also rarely observed.

Radon can travel by diffusion in solid matter but is extremely limited in its range, thus only emitters very close to pore space will contribute to the bulk radon emission of the rock mass. In addition, unless the porosity is interconnected, the radon will not be able to travel out of the parent rock and to the surface (either as a gas or dissolved in the groundwater). Inter-granular films precipitated onto grains surfaces are much more likely to release radon into the pore space than well-crystallised minerals (Appleton and Ball, 1995; Semkow, 1990). Surface weathering of the ironstones is therefore probably very important in enhancing their radon potential.

In summary, the detrital monazites and zircons are unlikely to be a significant source of radon because they are few in number and are present as dense single grains. The phosphates have lower alpha track densities than the detrital grains but are volumetrically more significant; however they occur mostly as tightly cemented sparry cement which would have little opportunity to release radon into the pore spaces. Some of the fine grained phosphatic cement within the matrix may be more significant but this occurs only infrequently in the Marlstone Rock. The iron-rich matrix has a lower alpha-track density than the other two sources but takes up a far higher volume. It also is very fine grained and has a high permeability in most locations (Figure 9). It is therefore likely to be the major source of the radon that escapes from the ironstone formations.

#### 3.3.4.4 Relationship between U and radon emanation

There is a strong linear correlation between bulk uranium concentration and the measured radon emanation levels observed in the Northampton Sand Formation ( $r^2=0.7961$ , p<0.0005, Figure 6). There is no significant

equivalent relationship observed in the Marlstone Rock Formation ( $r^2$ =0.099, p=0.219, Figure 6). Furthermore, even though both formations have similar concentrations of uranium, the amount of radon escaping from rock specimens of the Northampton Sand is higher than that escaping from the Marlstone Rock specimens at the locations sampled.

Radon emanation reaches a maximum of 232.6 Bq m<sup>-2</sup> in the Northampton Sand, compared to only 155.6 Bq m<sup>-2</sup> in the Marlstone Rock. This lower rate of radon emanation from the Marlstone Rock, compared to the Northampton Sand is likely due to differences in permeability. Petrographic observations have shown that the Marlstone Rock is tightly cemented by sparry carbonate throughout most of its extent, whereas the Northampton Sand has a fine grained, more permeable and often more altered matrix. Although it can be concluded that at this scale the escape of radon in the Marlstone Rock is impeded due to the lower permeability, the indoor radon data still shows a higher probability radon risk associated with this unit. We suggest that this relates to the high degree of fracturing in the Marlstone Rock Formation at a larger scale, although this was not measured in this current study. These fractures may control large-scale permeability and hence radon migration rather than matrix permeability. Near surface weathering of the ironstones is likely to be very important in enhancing radon potential since weathered mineral phases emanate radon more efficiently.

## 3.3.5 Chalk

The White Chalk Subgroup forms an extensive and important outcrop across England. It is often covered by superficial deposits (see section 3.4) but appears as bedrock at the surface across large swathes of the country. Chalk is a pure, white limestone that accumulated from the skeletal remains of tiny coccoliths and foraminifera, and coarser fragments of larger invertebrate animals in warm, shallow seas. Flint occurs as nodules and thin seams; the silica probably having been derived from biogenic sources such as sponges, radiolarian and diatoms. There was little input of terrigenous material; marl beds seem to occur in the White Chalk but are more common in the lower part of the Chalk Group. The majority of the White chalk bedrock is not radon affected (with an average of 0.49 %>AL) but locally this ranges up to 6.8 %>AL (WCK, Table 7). Indoor radon measurements of up to 445 Bq m<sup>-3</sup> have been recorded on this bedrock.

Also outcropping in narrow swathes across eastern and southern England, is the Cretaceous Grey Chalk subgroup. Like the White Chalk, this unit has a low average radon potential of 0.5 %>AL but this ranges up to 17.1 %>AL (GCK, Table 7). This group comprises blocky chalk with rhythmic alternations of marls; marly chalks with firm white chalk; as well as marly chalk with hard grey limestone.

#### 3.3.6 Breccia and conglomerate

Triassic breccias and conglomerate belonging to the Mercia Mudstone Group Marginal Facies occur in Avon and north Somerset and display an average radon potential of 3.0 %>AL (TRIBCO, Table 7). The thickest deposits lie unconformably on Carboniferous limestone and often clasts within the conglomerate are derived locally from rocks lying immediately below. The rock matrix usually consists of finer-grained rock fragments or, less commonly, siltstones, sandstones or micritic limestones (BGS, 2011; Jackson, 2008). Where these deposits overlie Carboniferous limestones, both the matrix and the limestone clasts are commonly dolomitised and the bedrock is known as 'Dolomitic Conglomerate'. It is this association with limestone or dolomitised material that results in the intermediate to locally high radon potential (max. 17%>AL, Table 6).

#### 3.3.7 Clay dominant sedimentary

Forming a limited outcrop on the SW peninsula (in and to the north-west of Newton Abbot, Devon) is the Tertiary Bovey formation. This siliciclastic argillaceous rock displays an average radon potential of 1.4 %>AL (BOF, Table 7) but locally ranges up to 19.2 %>AL near to the boundary with the uranium mineralised Dartmoor granite, from which the Bovey Formation sediments are partly derived.

#### 3.3.8 Shale and Chert

Slate belonging to the Dinantian Teign Valley Group crops out predominantly in north and east Cornwall on the SW peninsula and displays a high average radon potential of 24.9 %>AL (DINTVSL, Table 7). This overarching group contains a range of mudstone with: laminae and thin beds of siltstone and sandstone; scattered packets of coarse grained greywacke sandstone with interbedded mudstone; scattered sideritic, carbonaceous silty sandstone and argillaceous limestone nodules, and locally distributed units of tuff and basaltic lava (BGS, 2011). Elsewhere scattered thin calcareous siltstones and lenticular units of locally shelly limestone occur within the mudstone (Jackson, 2008).

Also belonging to the Dinantian Teign Valley Group are the chert beds of Devon and east Cornwall which exhibit a high average radon potential of 38.6 %>AL (DINCHRT, Table 7). These thin to medium bedded chert units are generally separated by siliceous mudstone beds. Also present are bluish black mudstone and thin to medium-bedded argillaceous limestones. Various volcanic rocks are present in this sequence in the Teign Valley and Ilsington area, south Devon and in the St Mellion area of east Cornwall. They include quartz-keratophyre tuffs, basic spilitic, and felsitic lavas. In places, local limestones and mudstones are dominant and reported to occur in distinctive lenticular units (BGS, 2011). This suggests that some cherts might be silicified limestones. DINTVSL, DINCHRT and the associated mudstones of the Brendon Formation (DINSL, average radon potential 5%>AL, max. 21.5%>AL, Table 7) predominantly crop out within the uranium mineralised zone surrounding the Bodmin and Dartmoor granites, which, along with their own compositions, probably explains their high radon potential.

### 3.4 Effects of superficial deposits on radon potential

Some bedrock units do not appear at the surface because they are always covered by superficial deposits, whilst other units do crop out as bedrock, as well as covered with a range of superficial deposits. The radon potential of these unconsolidated deposits reflects either their permeability or composition (and in some cases, both), in addition to the potential influence of the bedrock lying beneath. Taking impermeable clays, for example, they tend to exhibit low radon potential mainly because low permeability restricts the flow of radon-bearing air from the ground into buildings. In contrast, permeable sand and gravel and river terrace deposits often display higher radon potential relative to bedrock because high permeability and surface area facilitates both the release of radon and its flow from the ground into buildings. Chemical composition also plays a role, for example if fragments and mineral grains of uranium-rich granites are included in superficial material, this will lead to high radon in soils and dwellings, especially when these deposits are highly permeable sands and gravels (Appleton, 2005; Scheib et al., 2009). Such transportation of U-rich alluvium can be seen in Figure 3, where the rivers draining the U-rich high-ground of the Peak District (Derbyshire Dome, location labelled on Figure 1), show as blue on the ternary airborne image.

The behaviour of radon in the geological environment is very complex and this applies also to where superficial deposits are present. Therefore the effects of superficial deposits need to be considered on a regional, rather than national scale. To this end, the data presented in Table 8 are broken down by 100-km grid square. In very general terms though, where a bedrock type exhibits a high radon potential, cover of superficial deposits often lowers the radon potential. Also, superficial deposits of clay-with-flints or head often increase radon potential over bedrock. Examples of the effects observed include:

i. Superficial deposits of alluvium do not significantly change the radon potential relative to bedrock on the SW Carboniferous-Permian granite group since they are likely to be made up of locally derived material

(CARBGR, Table 8). Other types of superficial deposits do, in general, lower the radon potential relative to granite bedrock but there are regional variations: on the Land's End granites (grid-square SW) superficial deposits of Head decrease radon potential whereas in the Dartmoor area (SX) it appears to increase the radon potential (although measurements numbers are low here). Small superficial deposits of sand and gravel on the Land's End and Carnmenellis intrusions seem to substantially decrease radon potential (grid-square SW, Table 8).

ii. A regional variation is observed in the Dinantian limestone outcrops; the Derbyshire area (grid-square SK, DINLM, Table 8) exhibits elevated radon potential compared to where this bedrock crops out in Cumbria – North Yorkshire (grid-square SD). This relates to the U-rich reef limestones found in Derbyshire (e.g. The Bee Low Limestone Apron Reef), often with associated U mineralisation (Emery et al., 2005). Superficial deposits generally decrease the radon potential with the exception of the alluvium. Alluvium in this area includes transported U-rich material as demonstrated by the airborne data (Figure 3). Sand and gravel deposits and the glacially derived diamicton decrease radon potential substantially.

iii. Superficial cover of clay-with-flints increases the radon potential of the Cretaceous Upper Greensand (UGS, Table 8). This association of increased radon coinciding with clay-with-flints has been observed elsewhere (Killip, 2004), possibly reflecting phosphate-rich components with high uranium contents derived from the chalky source rock that the superficial deposits originate from (Killip, 2004).

iv. Middle Devonian mudstone of Cornwall exhibits higher radon in the Land's End area (grid-square SW) than further east (grid-square SX), but superficial deposits uniformly decrease radon potential relative to bedrock (DCMDMD, Table 8).

v. Superficial deposits upon Dinantian mudstone and mudstone dominant sedimentary rocks demonstrate a complex picture, illustrated in Table 8 for the Derbyshire area (grid square SK, DINMD and DINMDMIX, Table 8).

vi. Radon potential is decreased where alluvium and Head cover bedrock of mudstone dominant
 sedimentary rocks of the Namurian regional stage (Holsworthy Group) in north Devon and Cornwall
 (NAMHMDMIX, Table 8). The more permeable deposits of sand and gravel exhibit the same radon potential as
 the underlying bedrock. Cover of sand and gravel also exhibits high radon potential on Namurian mudstone

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dominant sedimentary (NAMMDMIX, Table 8) in the Staffordshire-Cheshire area (grid-square SJ), where radon levels are higher than in the Derbyshire area (grid-square SK).

vii. Although there are not a large number of indoor radon measurements where the Marlstone Rock Formation is covered by superficial deposits, it appears that radon potential is decreased substantially (MRB, Table 8). This decrease where covered by superficial deposits is mirrored on the Northampton Sand Formation (INONS, Table 8). In both cases, where the bedrock is covered with glacial till (diamicton), there is a greater decrease in radon potential than where there is a cover of sand and gravel. This probably reflects the relative permeability of the superficial geology units; sand and gravel being of higher permeability than the till (DMTN) and therefore impeding the flow of radon less than till. In contrast, where the Northampton Sand has been worked, radon potential increases (INONS\WGR, Table 8) relative to bedrock. This may reflect the phosphatic nature of the remaining basal beds (Hollingworth and Taylor, 1951) or the permeable nature of any backfill used after working ceased. During the Hi-RES-1 airborne survey, anomalous eTh values were observed over worked ground on the Northampton sand (Hodgkinson, 2012).

viii. Radon potential is higher on the younger White Chalk in the East Sussex area compared to the White Chalk outcrops in West Sussex-Hampshire (WCK, Table 8). This finding is in agreement with a detailed study of the area by Killip in 2004. Where the chalk is covered by clay-with-flints deposits, the radon potential is increased; as was observed on the Upper Greensand. Radium absorbed to the clay minerals and secondary iron hydroxides in the clay-with-flints is likely to be responsible for the increased radon potential (Killip, 2004). Increased fissures within the chalk, as well as shrinkage cracks in summer will all also contribute to the release and transport of radon.

The variation in effects observed in these examples highlights the complex nature of the natural environment and serves to emphasise the need for accurate, detailed data and interpretation.

## 4. Conclusions

Radon exposure is a chronic and serious geohazard but with the knowledge of its distribution provided by an accurate radon potential map, this risk to human health can be reduced through well directed radon testing programmes and building control regulations.

This study has demonstrated that a wide range of bedrock types contribute to the radon prone areas of England and provides a preliminary analysis of some of the likely geological controls. Uranium in soil data has

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highlighted organic binding mechanisms, for example in Holocene peats of the Fen basin and demonstrated higher concentrations over known radon-prone units such as the granites of SW England, Carboniferous limestone of the Peak District as well as over volcanic rocks surrounding the Cheviot granite pluton in Northumberland. The utility in the soil chemistry and gamma spectrometry data is to enhance understanding on the distribution of radon's precursor and to provide an evidence base to support the grouping of geological units used in the radon potential mapping methodology.

Some of the geological units associated with high radon potential are well known, such as the granite intrusions in south west England, the Carboniferous limestones of Derbyshire and the Jurassic ironstones in Northamptonshire. This study provides a more comprehensive description of the bedrock geological units associated with intermediate to high radon potential in England which encompass:

- 1. Granites and associated uranium mineralisation of south west England;
- Carboniferous (Dinantian) limestones throughout England; Jurassic limestones (especially those in the Inferior Oolite, Great Oolite and Portland Groups, and the Lower Lias in Somerset and Lincolnshire);
   Permian dolomites in the Midlands; and, Devonian limestones in SW England.
- Sandstones including the Cretaceous Upper Greensand, Jurassic Upper Lias Bridport Sand Formation, Permian sandstones in SW England, Carboniferous (Westphalian, and less commonly Namurian) sandstones especially in Yorkshire; and, Devonian sandstones in Cornwall.
- 4. Mudstone and mudstone dominated units including the Jurassic Grantham Formation in the Midlands; Carboniferous Crackington Formation in Devon and Cornwall; Lower Carboniferous (Bowland Shale Formation) especially in Derbyshire and the Craven Basin; Meldon Shale and Quartzite Formation in Cornwall; Devonian mudstones in Devon and Cornwall; and, Silurian mudstones in the Welsh borders and Lake District.
- 5. Ironstones of the Jurassic Northampton Sand Formation and Marlstone Rock. New uranium, SEM, autoradiograph and XRD investigations have provided a deeper understanding of the likely sources and controls of radon from these ironstones. Whereas the iron-rich matrix has a lower alpha-track density than other sources, such as phosphate-rich pellets, it is very fine grained and has a high permeability in most locations so is likely to be the major source of the radon that escapes from the ironstone formations. Emanation studies indicated a lower rate of radon emanation from the Marlstone Rock, compared to the Northampton Sand and this is likely due to differences in permeability: the Marlstone

Rock is mainly tightly cemented by sparry carbonate, whilst the Northampton Sand has a fine grained, more permeable and often more altered matrix. High radon potential associated with the Marlstone Rock is likely to be related to the high degree of fracturing which may control radon migration rather than matrix permeability. Weathering may also have a significant impact on radon release from the ironstones.

- 6. The Cretaceous Chalk is generally characterised by low radon potential but zones of moderately high potential may be related to phosphate-enrichment. However, this is not mapped at the 1:50 000 scale, and requires further investigation.
- Breccias and conglomerates of the Triassic and Mercia Mudstone Group marginal facies in the Somerset area where clasts of Carboniferous limestone and high permeability are likely to be the cause of high indoor radon concentrations.
- 8. Shales and cherts of the Dinantian Teign Valley Group in Cornwall, although these crop out predominantly within the uranium mineralised zone surrounding the Bodmin and Dartmoor granites, which probably explains the high radon potential associated with these units.

Superficial deposits have an important impact on the radon potential of bedrock so it is vital that they are considered in radon mapping exercises. The source, composition and permeability of the superficial deposits determine how they impact on bedrock radon potential or indeed contribute to radon potential themselves. In areas of relatively U-rich bedrock, for example the SW granites or Carboniferous limestone in Derbyshire, transported material such as alluvium often exhibits a similar radon potential as the parent-material or local bedrock. Worked ground on the Northampton Sand Formation exhibits increased radon potential relative to bedrock, possibly reflecting the phosphatic nature of the remaining basal beds. Also contributing to increased radon potential are superficial deposits of clay-with-flints, present in southern England across the Upper Greensand and the White Chalk Group, reflecting their source composition. Most other superficial deposits reduce radon potential relative to bedrock since they reduce permeability but variation is often complex and locally controlled. Impermeable clays tend to exhibit low radon potential mainly because low permeability restricts the flow of radon-bearing air from the ground into buildings. In contrast, permeable sand and gravel and river terrace deposits often display higher radon potential relative to bedrock because high permeability and surface area facilitates both the release of radon and its flow from the ground into buildings.

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Whilst reducing radon risk to people in homes remains the main motivation for investigating the distribution and geological controls on radon by radon potential mapping, long-term (3 month) measurements are the best way to assess the risk to people in individual homes and buildings. Geological information, soil geochemistry, mineralogical studies and airborne geophysical data all can contribute by informing the radon potential mapping process, particularly where measurement numbers are low. The variation in radon observed here, highlights the complex nature of the geological and pedological environment and emphasises the need for accurate, detailed data and interpretation. The level of detail contained in the radon potential map presented here has, since its publication in 2007, resulted in the identification of a substantial number of dwellings with radon above the Action Level as a result of follow-up measurement campaigns carried out in the areas of high radon potential identified by the joint HPA-BGS radon map (Miles et al., 2007).

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Marker	Rock Code at	Rock description at location of soil sample	Bedrock Age	No.	Mean	Min.	Median	Max.
Fig. 4	location of							
	soil sample							
1	DCFEIN	Dartmoor Granite	Carboniferous	22	5.98	1.33	4.60	27.37
1	CARBGR	SW Granite Group (excluding Dartmoor	Carboniferous	23	5.49	2.63	4.00	33.29
		Granite)						
2	TRIMD	Mudstone	Triassic	1959	2.34	0.32	2.31	15.34
2	LLI	Mudstone dominant sedimentary	Jurassic (Lower Lias)	1067	2.43	0.27	2.47	6.05
3	SPRDMDMIX	Mudstone dominant sedimentary	Silurian (Pridoli)	73	2.09	0.45	2.10	4.11
4	WSG	Wealdon Group sandstone	Cretaceous	105	2.22	0.27	2.39	3.99
4	WGM	Wealdon Group mudstone	Cretaceous	100	2.73	0.52	2.92	4.18
5	AMKIM\PEAT	Ampthill and Kimmeridge mudstone dominant	Jurassic (bedrock)	179	4.08	1.72	3.91	7.59
		sedimentary covered by peat						
5	<b>GLT\PEAT</b>	Gault Clay mudstone covered by peat	Cretaceous	47	6.00	1.36	4.60	13.00
6	DINLM	Limestone	Dinantian	255	3.54	0.00	3.19	13.65
6	DINMDMIX	Mudstone dominant sedimentary	Dinantian	69	4.49	0.62	3.71	14.55
7	DLDMA	Maifc Igneous (Cheviot area)	Lower Devonian	10	3.13	0.63	3.42	4.96
8	NAMMDMIX	Mudstone dominant sedimentary	Namurian	211	2.08	0.00	2.09	5.10
9	TRISD	Sandstone (Sherwood Sandstone Group)	Triassic	1099	1.80	0.23	1.80	6.00
10 & 11	WCK	White Chalk	Cretaceous	2375	1.76	0.00	1.72	5.69
10	WCK\DMTN	White Chalk covered with Diamicton	Cretaceous	2448	1.93	0.27	1.92	6.69
10	WCK\SAGR	White Chalk covered with sand and gravel	Cretaceous	677	1.63	0.25	1.62	4.00
11	LGS	Lower Greensand Sandstone	Cretaceous	167	1.62	0.40	1.56	4.10
12	THAMC	Thames Group including London Clay	Tertiary	3093	2.00	0.10	2.10	6.45

Table 1 Uranium (mg kg<sup>-1</sup>) in topsoil for selected geological groupings. No. = number of soil measurements; Mean, minimum, median and maximum U in mg kg<sup>-1</sup>. See Figure 4 for generalised locations.

	Bedrock Geology			Ra	don statis	tics	
Bedrock Code	Bedrock description	Age	GM	Max.	Ave.	Max.	No.
			Bq m⁻³	Bq m <sup>-3</sup>	%>AL	%>AL	
DCFEIN	Dartmoor Granite	Carboniferous	285	2756	66.1	100.0	1763
CARBGR	SW Granite Group (excluding Dartmoor	Carboniferous	177	7484	43.9	89.1	7817
	Granite)						
DLDMA	Maifc Igneous (Cheviot area)	Lower Devonian	47	141	3.5	3.5	22
DCMAIN	Mafic Igneous Instrusive (Cornwall)	Mid Devonian	55	1905	8.4	68.4	1264
DCMA	Mafic Igneous (Cornwall)	Devonian	65	2985	10.0	58.3	6560
CARBMAIN	Mafic Igneous Instrusive	Carboniferous	69	1265	5.6	50.4	191
DINMA	Mafic Igneous	Dinantian	67	2926	11.1	37.5	1105
DCMETA	Metamorphic – Mica Schist	Devonian	49	6378	7.4	40.1	1287

Table 2 Summary radon statistics for felsic or mafic igneous and metamorphic bedrock with moderate and high radon potential.

Table footnote (to apply to Tables 2, 3, 5, 6 and 7):

GM = geometric mean indoor radon; Max. = maximum recorded indoor radon (temperature corrected), Ave. %>AL = average percentage of homes estimated to exceed the radon Action Level (%>AL), Max. %>AL = maximum percentages of homes estimated to exceed the radon Action Level (%>AL); No. = number of indoor radon measurements.

	Bedrock Geology			F	Radon Stat	istics	
Bedrock code	Bedrock description	Age	GM Bq m <sup>-3</sup>	Max. Bq m <sup>-3</sup>	Ave. %>AL	Max. %>AL	No.
DINLM	Limestone	Dinantian	107	6252	13.7	79.4	13097
DINLMMIX	Limestone dominant sedimentary	Dinantian	50	2648	4.3	64.1	484
GOGLMST	Great Oolite Group limestone	Jurassic	37	2596	2.8	28.1	14088
GOGFE	Great Oolite Group (Fullers Earth) Limestone with mudstone	Jurassic	38	1265	2.1	22.4	1020
GOGCB	Great Oolite Group (Cornbrash) limestone	Jurassic	36	875	0.3	27.1	1201
GOGFMLMST	Great Oolite Group (Forest Marble) limestone	Jurassic	40	477	1.1	10.4	431
INOLMST	Inferior Oolite Group limestone	Jurassic	70	2926	7.8	73.6	13247
PST	Portland Group Limestone dominant sedimentary	Jurassic	94	1384	9.3	63.7	294
LLILMST	Limestone	Jurassic- Lower Lias	80	4919	2.9	58.7	2126
LMJ	Sedimentary	Lower and Middle Jurassic	42	2840	4.1	22.4	3511
PERDO	Dolomite	Permian	39	2897	1.6	64.8	3866
DCUDLM	(Cornwall) limestone	Upper Devonian	57	1304	3.7	4.8	1058
DCMDLM	(Cornwall) limestone	Middle Devonian	54	2811	7.2	49.2	7778
DCLDLM	(Cornwall) limestone	Lower Devonian	60	1624	14.2	40.9	554

Table 3 Summary radon statistics for limestone or limestone dominant sedimentary lithologies with moderate and high radon potential.

	Bedrock	Airborne gamma ray spectrometry data								
Code	Description	No:	AM eU	Max eU	AM K	Max K	AM eTh	Max eTh		
CARBMAIN	Carboniferous mafic intrusive	225	1.1	2.9	0.3	0.8	2.3	6.0		
DINLM	Dinantian limestone	23374	2.0	21.0	0.3	1.7	3.8	16.7		
DINLMMIX	Dinantian limestone dominant	3985	2.0	5.2	0.5	1.7	4.0	9.8		
DINMA	Dinantian mafic	1016	1.5	7.4	0.3	1.2	2.9	8.3		
DINMD	Dinantian mudstone	476	3.2	10.8	0.4	1.2	4.0	9.4		
DINMDMIX	Dinantian mudstone dominant	10166	1.9	13.3	0.4	1.8	4.2	12.1		
DINSD	Dinantian sandstone	1199	1.4	4.9	0.2	0.8	2.7	6.1		
GOGCB	Great Oolite Group (Cornbrash) limestone	2812	1.5	3.3	0.5	1.2	5.9	12.5		
GOGLMST	Great Oolite Group limestone	2259	1.3	2.8	0.5	1.2	4.6	9.9		
INOGRF	Inferior Oolite Group mudstone dominant	505	1.5	3.0	0.5	1.3	5.2	10.7		
INOLMST	Inferior Oolite Group limestone	20001	1.4	3.1	0.4	1.3	3.8	10.6		
INONS	Northampton Sand Formation ironstone	1278	1.6	3.2	0.4	1.1	6.6	14.4		
LLI	Jurassic (Lower Lias) mudstone dominant	22788	1.4	4.6	0.8	2.0	6.0	22.0		
LLILMST	Jurassic (Lower Lias) limestone	2719	1.6	3.3	0.9	1.5	6.7	10.8		
MLI	Jurassic (Mid Lias) mixed sedimentary	1814	1.6	3.2	0.6	1.2	8.7	21.1		
MRB	Marlstone Rock ironstone dominant	4036	1.7	3.6	0.4	1.1	12.0	23.7		
NAMMDMIX	Namurian mudstone dominant	12685	1.1	9.3	0.3	1.6	3.4	11.1		
NAMSD	Namurian sandstone	16303	0.8	3.3	0.4	1.5	3.2	9.4		
PERDO	Permian dolomite	8300	1.4	3.7	0.6	2.3	4.6	17.6		
PERSD	Permian sandstone	5372	1.0	3.0	0.7	1.9	2.9	12.3		
WESDSD	Westphalian sandstone	626	1.4	3.3	0.6	1.2	4.5	9.1		
WESDUSD	Westphalian (Duckm. & Bols.) sandstone	2982	1.4	3.7	0.7	1.9	5.2	21.2		
WESLASD	Westphalian (Langsettian) sandstone	9432	1.1	4.0	0.5	1.9	4.4	15.4		
Mean values:			1.52		0.49		4.85			

 Table 4 Summary statistics from the HiRES-1 airborne gamma ray spectrometry survey for selected Radon Affected units.

Footnote to Table 4: No. = number of airborne measurements; AM eU = arithmetic mean equivalent uranium (ppm); Max eU = maximum recorded equivalent uranium (ppm); AM K = arithmetic mean potassium (%); Max K = maximum potassium (%); AM eTh = arithmetic mean equivalent thorium (ppm); Max eTh : maximum equivalent thorium (ppm).

	Bedrock			Ra	don Statis	stics	
Bedrock code	Bedrock description	Age	GM	Max.	Ave.	Max.	No.
			Bq m <sup>-3</sup>	Bq m⁻³	%>AL	%>AL	
UGS	Upper Greensand	Cretaceous	38	1441	1.7	41.7	2131
ULIBS	Bridport sand formation	Jurassic (U. Lias)	36	1047	2.6	13.1	3829
PERSD	Sandstone	Permian	32	1545	0.5	9.0	12707
WESDSD	Sandstone	Westphalian D	34	1090	1.3	18.4	315
WESDUSD	Duckmantian and Bolsovian sandstone	Westphalian	28	1058	1.7	10.2	1092
WESLASD	Langsettian sandstone	Westphalian	39	1560	1.2	35.9	5282
NAMHSD	(Holsworthy Group) sandstone	Namurian	40	1624	1.9	30.9	3544
NAMSD	Sandstone	Namurian	36	1794	1.0	19.2	2757
DINSD	Sandstone	Dinantian	75	1291	3.8	42.9	438
DCMDSD	Sandstone (Cornwall)	M. Devonian	85	4109	12.4	48.0	4617
DDMDSD	Sandstone (Devon)	M. Devonian	49	732	6.1	20.1	136
	Hangman Sandstone Fm						
DCLDSD	Sandstone (Cornwall)	L. Devonian	57	1794	15.5	47.6	2475
SLUDSDMIX	Sandstone dominant sedimentary	Silurian (Ludlow)	55	637	7.1	16.9	217

Table 5 Summary radon statistics for sandstone or sandstone dominant sedimentary lithologies with moderate and high radon potential.

	Bedrock			Ra	don Statis	tics	
Bedrock code	Bedrock description	Age	GM Bq m <sup>-3</sup>	Max. Bq m⁻³	Ave. %>AL	Max. %>AL	No.
INOGRF	Inferior Oolite Group Mudstone dominant sedimentary	Jurassic	57	1608	13.7	37.1	1403
LLI	Mudstone dominant sedimentary	Jurassic (Lower Lias)	47	3266	1.2	59.1	21506
MLI	Mudstone, sandstone, ironstone	Jurassic (Middle Lias)	36	2063	3.4	42.1	6076
NAMHMDMIX	Holsworthy Group mudstone dominant sedimentary	Namurian	44	3433	5.2	79.1	7917
DINMDMIX	Mudstone dominant sedimentary	Dinantian	56	7048	4.5	54.0	4243
DINMD	Mudstone	Dinantian	99	6128	17.6	67.8	537
DINTVMDMIX	Teign Valley Group mudstone dominant sedimentary	Dinantian	100	1963	23.0	54.8	944
DCUDMD	Mudstone (Cornwall)	Upper Devonian	84	14622	16.8	89.4	38655
DDUDMD	Mudstone (Devon)	Upper Devonian	48	1357	5.9	26.1	2244
DCMDMD	Mudstone (Cornwall)	Middle	64	11388	12.9	75.2	24582
		Devonian					
DDMDMD	Mudstone	Middle	52	2519	5.5	16.6	1611
		Devonian					
DCLDMD	Mudstone (Cornwall)	Lower Devonian	77	8870	15.4	92.1	22533
SPRDMDMIX	Mudstone dominant sedimentary	Silurian (Pridoli)	324	777	1.3	29.8	400
SLUDKMDMIX	Mudstone dominant sedimentary (Lake District)	Silurian (Ludlow)	43	977	0.3	18.7	827

Table 6 Summary radon statistics for mudstone or mudstone dominant sedimentary lithologies with moderate and high radon potential.

	Bedrock			Ra	don Stati	stics	
Bedrock code	Bedrock description	Age	GM	Max.	Ave.	Max.	No.
			Bq m <sup>-3</sup>	Bq m <sup>-3</sup>	%>AL	%>AL	
INONS	Northampton Sand Formation (Ironstone dominant sedimentary)	Jurassic	67	4726	15.5	86.9	27032
MRB	Marlstone Rock (Ironstone dominant sedimentary)	Jurassic	85	2596	15.8	90.8	5311
WCK	White Chalk	Cretaceous	26	445	0.49	6.8	1822
GCK	Grey Chalk	Cretaceous	31	884	0.5	17.1	622
TRIBRCO	Breccias and conglomerate	Triassic	53	986	3.0	17.1	1568
BOF	Bovey Formation (mudstone dominant sedimentary)	Tertiary	38	424	1.4	19.2	1174
DINSL	Teign Valley Group (Brendon Formation) slate	Dinantian	68	636	5.0	21.5	192
DINTVSL	Teign Valley Group slate	Dinantian	96	2280	24.9	74.2	912
DINCHRT	Chert	Dinantian	129	2445	38.6	83.9	492

Table 7 Summary radon statistics for ironstone, chalk, breccias, conglomerate, slate and chert lithologies with moderate and high radon potential.

			100	100 K grid square			( grid sq	uare
Rock Code	Bedrock description & Section	Superficial deposits	Grid	No.	Ave.	Grid	No.	Ave.
			Square		%>AL	Square		%>AL
CARBGR		None	SX	1321	37.8	SW	6264	46.6
CARBGR\CLSI	SW Granite Group	Clay-silt (mainly alluvium)	SX	150	38.3	SW	200	45.3
CARBGR\H-CLSI	(refer to section 3.2.1)	Head (mainly clay-silt)	SX	38	59.2	SW	114	37.3
CARBGR\SAGR		Sand and gravel	SX	-	-	SW	73	11.8
DINLM		None	SD	4455	13.1	SK	7477	34.3
DINLM\CLSI	Limestone – Dinantian	Clay-silt (mainly alluvium)	SD	250	14.2	SK	256	32.0
DINLM\DMTN	(refer to section 3.3.1)	Diamicton	SD	831	6.0	SK	47	8.8
DINLM\SAGR		Sand and gravel	SD	941	5.1	SK	-	-
UGS	Lippor Groopsand - Crotacoous	None	ST	1577	4.3	-	-	-
UGS\C-DMTN	(refer to section 2.2.2)	Clay-with-flints	ST	271	8.1	-	-	-
UGS\H-CLSI	(Teter to section 5.5.2)	Head (mainly clay-silt)	ST	330	5.2	-	-	-
DCMDMD		None	SX	16,503	8.0	SW	8079	17.8
DCMDMD\CLSI	Mudstone (Cornwall) – Devonian	Clay-silt (mainly alluvium)	SX	728	3.9	SW	582	8.4
DCMDMD\H-CLSI	(refer to section 3.3.3)	Head (mainly clay-silt)	SX	1352	4.9	SW	130	9.3
DCMDMD\SAGR		Sand and gravel	SX	-	-	SW	295	6.8
DINMD		None	SK	488	25.6	-	-	-
DINMD\CLSI	Mudstone – Dinantian	Clay-silt (mainly alluvium)	SK	50	3.7	-	-	-
DINMD\DMTN	(refer to section 3.3.3)	Diamicton	SK	156	8.9	-	-	-
DINMD\H-CLSI		Head (mainly clay-silt)	SK	43	33.0	-	-	-
DINMDMIX		None	SK	4135	8.2	-	-	-
DINMDMIX\CLSI	Mudstone dominant sedimentary –	Clay-silt (mainly alluvium)	SK	594	7.4	-	-	-
DINMDMIX\DMTN	Dinantian	Diamicton	SK	478	1.3	-	-	-
DINMDMIX\H-CLSI	(refer to section 3.3.3)	Head (mainly clay-silt)	SK	1403	1.9	-	-	-
DINMDMIX\SAGR		Sand and gravel	SK	32	20.5	-	-	-
NAMHMDMIX	Holeworthy Crown mudstone dominant	None	SX	5185	9.2	SS	2706	2.2
NAMHMDMIX\CLSI	noisworthy Group mudstone dominant	Clay-silt (mainly alluvium)	SX	299	3.9	SS	162	1.0
NAMHMDMIX\H-CLSI	(rotor to section 2.2.2)	Head (mainly clay-silt)	SX	160	1.2	-	-	-
NAMHMDMIX\SAGR		Sand and gravel	SX	216	9.2	-	-	-

			100	K grid squ	Jare	100 K grid square		
Rock Code	Bedrock description & Section	Superficial deposits	Grid	No.	Ave.	Grid	No.	Ave.
			Square		%>AL	Square		%>AL
NAMMDMIX		None	SJ	182	13.2	SK	1425	0.7
NAMMDMIX\CLSI	Mudstone dominant sedimentary –	Clay-silt (mainly alluvium)	SJ	-	-	SK	82	0.9
NAMMDMIX\DMTN	Namurian	Diamicton	SJ	390	4.1	SK	749	0.02
NAMMDMIX\H-CLSI	(refer to section 3.3.3)	Head (mainly clay-silt)	SJ	-	-	SK	175	0.8
NAMMDMIX\SAGR		Sand and gravel	SJ	315	15.2	SK	-	-
MRB	Marletona Back Ironstana Iurassia	None	SP	4613	19.2	SK	626	18.7
MRB\DMTN	(refer to section 2.2.4)	Diamicton	SP	53	0.35	-	-	-
MRB\SAGR	(1919) to section 5.5.4)	Sand and gravel	SP	47	1.9	-	-	-
INONS	Inferior Oplite Crown (Northematon Cond	None	SP	25,633	12.9	SK	1211	17.9
INONS\DMTN	Interior Conte Group (Northampton Sand	Diamicton	SP	1065	2.4	SK	-	-
INONS\ SAGR	(refer to section 2.2.4)	Sand and gravel	SP	343	4.5	SK	59	0.27
INONS\WGR	(refer to section 3.3.4)	Worked Ground*	SP	1332	21.6	SK	42	46.5
WCK	White Chalk Crotacoous	None	TQ	329	1.0	SU	819	0.7
WCK\C-DMTN	(refer to section 2.2.5)	Clay-with-flints	ΤQ	96	1.8	SU	184	0.7
WCK\CLSI	(refer to section 3.3.5)	Clay-silt (mainly alluvium)	ΤQ	59	1.65	SU	-	-

Table 8 The effects of superficial deposits on radon potential on selected units broken into regional areas by 100 km grid-square (No. = the number of indoor radon measurements; %>AL = the percentage of homes estimated to exceed the radon Action Level).

\* Worked Ground was the only artificial geology code used as a pseudo-superficial geology code since worked ground is extensive on the Northampton Sand, and it had a bearing on the radon potential.



Figure 1 Simplified bedrock geology map of England displaying selected locations detailed in text.



**Figure 2** Radon potential map of England showing the percentage of dwellings estimated to exceed the radon Action Level of 200 Bq m<sup>-3</sup> (%>AL). Extent of HiRES-1 survey shown in Figure 3 outlined by box.



Figure 3 Ternary image of the HiRES-1 airborne radiometric data showing the relative abundance of potassium (K, in red tones), equivalent thorium (eTh in greener tones) and equivalent uranium (eU, in blue tones). For bedrock code information see Tables 1 - 7. Linework represents 1: 625, 000 scale bedrock geology polygons. The extent of this survey is outlined in Figure 2.



Figure 4 Topsoil uranium (mg kg<sup>-1</sup>). Numbers indicate features described in the text (see section 3.1).



Figure 5 Radon potential map of the St Austel granite; displaying the percentage of dwellings estimated to exceed the radon Action Level of 200 Bq m<sup>-3</sup> (%>AL).



**Figure 6** Relationship between radon emanation measurements (radon per unit surface area, Bq m<sup>-2</sup>) and uranium concentration as determined by ICP-MS of bulk rock samples for the Northampton Sand and Marlstone Rock formations.

# Radon emanation Vs U concentration (ppm)



**Figure 7** Marlstone Rock Formation showing calcite cement Oolite of upper part of formation. Sample from depth of 1.1 m at Sauvey Castle, 15 km east of Leicester.



**Figure 8** Northampton Sand Formation enlargement of typical ooids with empty centres surrounded by surviving Fe-rich clay, while the matrix is almost entirely replaced by Fe oxide. Sample from depth of 0.1 m at Tywell, 5 km ESE of Kettering.



**Figure 9** Northampton Sand Formation showing a rare and tightly cemented Ca-phosphate pellet. Sample from depth of 0.6 m from Pitsford quarry, North of Northampton.



**Figure 10** Northampton Sand Formation showing replacement of ooliths and matrix by Fe oxides, and Fe oxide fracture mineralisation which is very pronounced towards the top of the Formation. Sample from depth of 0.1 m from Tywell, 5 km ESE of Kettering.