Soil radium, soil gas radon and indoor radon empirical relationships to assist in post-closure impact assessment related to near-surface radioactive waste disposal

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Abstract

Least squares (LS), Theil's (TS) and weighted total least squares (WTLS) regression analysis methods are used to develop empirical relationships between radium in the ground, radon in soil and radon in dwellings to assist in the post-closure assessment of indoor radon related to near surface radioactive waste disposal at the Low Level Waste Repository in England. The data sets used are (i) estimated ^{226}Ra in the <2mm fraction of topsoils (eRa226) derived from estimated uranium (eU) from airborne gamma spectrometry data, (ii) eRa226 derived from measurements of uranium in soil geochemical samples, (iii) soil gas radon and (iv) indoor radon data. For models comparing indoor radon and (i) eRa226 derived from airborne eU data and (ii) soil gas radon data, some of the geological groupings have significant slopes. For these groupings there is reasonable agreement in slope and intercept between the three regression analysis methods (LS, TS and WTLS). Relationships between radon in dwellings and radium in the ground or radon in soil differ depending on the characteristics of the underlying geological units, with more permeable units having steeper slopes and higher indoor radon concentrations for a given radium or soil gas radon concentration in the ground. The regression models comparing indoor radon with soil gas radon have intercepts close to 5 Bq m^{-3} whilst the intercepts for those comparing indoor radon with eRa226 from airborne eU vary from

about 20 Bq m^{-3} for a moderately permeable geological unit to about 40 Bq m^{-3} for highly permeable limestone, implying unrealistically high contributions to indoor radon from sources other than the ground. An intercept value of 5 Bq m^{-3} is assumed as an appropriate mean value for the UK for sources of indoor radon other than radon from the ground, based on examination of UK data. Comparison with published data used to derive an average indoor radon : soil ^{226}Ra ratio shows that whereas the published data are generally clustered with no obvious correlation, the data from this study have substantially different relationships depending largely on the permeability of the underlying geology. Models for the relatively impermeable geological units plot parallel to the average indoor radon : soil ^{226}Ra model but with lower indoor radon : soil ^{226}Ra ratios, whilst the models for the permeable geological units plot parallel to the average indoor radon : soil ^{226}Ra model but with higher than average indoor radon : soil ^{226}Ra ratios.

1 Introduction

The Low Level Waste Repository (LLWR; see http://www.llwrsite.com/ for further details) is the UK's principal facility for the disposal of solid low-level radioactive waste. The facility is located on the West Cumbrian coastal plain close to the village of Drigg and approximately 5 km south-east of the Sellafield nuclear site. The LLWR receives wastes from a range of consignors including from nuclear industry sites, defence establishments and users of radioactive materials, and from the clean-up of historically contaminated sites. The bulk of the volume and of total radioactive inventory received by the LLWR comes from facilities associated with the nuclear industry. Significant radiological impacts could arise, however, from the disposal of consignments of wastes from processing and use of naturally-occurring radionuclides, mainly ^{226}Ra and ^{232}Th and their progeny. These wastes consist of mineral sands, wastes from processing such minerals and also wastes from clean up of sites at which processing, manufacture or use of thorium and radium products took place, e.g. radium luminising facilities. The production of radon-222 in the waste is directly proportional to the inventory of its parent, radium-226, which itself is due to disposed ^{226}Ra (half-life 1600 years) plus in-growth from decay of disposed ^{234}U (half-life 250,000 years) via ^{230}Th (half-life 77,000 years).

A particular concern for the post-closure radiological environmental impact assessment is that an excavation into the engineered cap, or into the waste itself, could give opportunities for exposures to radon from radium-bearing wastes. The case that can give the highest dose is if it is assumed that a dwelling is constructed either directly above the radium-bearing waste or on spoil created by excavation of repository cover materials and waste, including radium-bearing waste. In the past, the case of a dwelling constructed on a degraded low level radioactive waste site or on excavated spoil was assessed making use of models that attempt to represent the entry and build up of radon within such a dwelling taking account of radon migration processes and the possible characteristics of the dwelling. This approach is subject to large uncertainties because the entry of radon and accumulation in a building is highly sensitive to ground conditions, building construction and ventilation, and because the characteristics of a future building are unknown. For the most recent assessments of the LLWR (Sumerling, 2008), a simpler approach was adopted using an empirical relationship between the concentration of radon in dwellings and the concentration of ^{226}Ra in soil, based on general data from UNSCEAR (2000).

In the UK, there are substantial data sets of measured radon in dwellings, naturally-occurring radionuclides in soil and radon in soil gas, all related to classification of local geology. This gives the opportunity to develop empirical relationships that are intrinsically matched to the average characteristics of UK houses and also, by choice of the geological association, that are appropriate to represent ground conditions more relevant to the radiological assessment cases. This paper describes work carried out by the British Geological Survey and the Health Protection Agency to analyse UK data and to develop empirical relationships between uranium and ^{226}Ra in soil, radon in soil and radon in dwellings. These relationships will assist in the post-closure assessment of radon related to near surface radioactive waste disposal as practiced at the LLWR.

2 Materials and methods

2.1 Introduction.

Four types of data were used in this study: (i) estimated ^{226}Ra in the <2mm fraction of topsoils (eRa226) derived from estimated uranium (eU) from airborne gamma spectrometry data, (ii) eRa226 derived from measurements of uranium in soil geochemical samples, (iii) concentration of radon in soil gas; and (iv) radon concentrations in homes. Not all types of data are available in all parts of the UK, and where they are available the numbers of results are variable. The amount of appropriate data available over artificial ground (assumed to be similar to the potential situation if the repository is later disturbed) is limited. As a consequence, this study focussed on data from high-permeability geological units with relatively high radon potential, to mimic the likely permeability of disturbed ground. It was known from previous investigations (Appleton et al., 2000; Miles and Appleton, 2005; Appleton and Miles, 2009; BGS and HPA unpublished data) that the following geological units were the most likely to provide appropriate data, in terms of permeability, for the current study: (i) Lower Carboniferous limestones (DINLM); (ii) Lower Carboniferous mudstone with subsidiary siltstone, sandstone, limestone (DINMDMIX); (iii) Jurassic Inferior Oolite limestone units (INOLMST); (iv) Jurassic Northampton Sand Formation (INONS); and (v) the Jurassic Marlstone Rock Formation (MRB). A number of geological units with moderate or low permeability were included in the study for comparative purposes. These were Triassic mudstones (TRIMD), Westphalian and Namurian mudstones with subsidiary siltstone, sandstone, and limestone (WESNAMMDMIX), Namurian and Westphalian sandstones (NAM&WESSD), Upper Lias mudstones (ULI), and Permian dolomites (PERDO).

2.2 Estimated uranium from airborne radiometric data

The High Resolution Airborne Resource and Environmental Survey (HiRES-1) of the English Midlands, including 1024 channel gamma spectrometry, is described in Peart et al., (2004). An equivalent uranium (eU) value is determined from the ^{214}Bi gamma peak assuming equilibrium between the measured gamma peak from ^{214}Bi (a radon decay product) and the parent natural U concentration in the ground. This assumed equilibrium, however, may not always hold true due to the differing geochemical behaviour of the members of the ^{238}U decay chain. The geochemical behaviour of ^{226}Ra is markedly different from U, which may lead to U being removed during weathering, leaving ^{226}Ra, although in the context of this study, the determination of ^{226}Ra is more important than the determination of U. Also the ^{214}Bi gamma peak effectively reports short-lived radon decay product concentrations in the top 30 cm or so of the ground, from which some degree of loss of radon may occur. Therefore the ^{214}Bi gamma peak could indicate higher or lower eU values than the ^{238}U levels actually present. In addition, the data from the airborne gamma spectrometry surveys are calibrated against ground gamma spectrometry data determined using instruments which themselves are calibrated using concrete pads with known U concentrations, for which it is assumed that there is equilibrium between ^{238}U and ^{214}Bi. There will be greater uncertainty attached to average eU data for urban areas (Appleton et al., 2008). The lack of a 1:1 relationship between airborne eU and U measured by XRF in the <2mm fraction of topsoils (unpublished data and Appleton et al., 2008) may be due to (i) radon loss (IAEA, 2003), (ii) varying levels of soil moisture (Grasty, 1997); (iii) radon decay products washed-out of the air by rain; (iv) possible calibration problems with the HiRES airborne gamma spectrometry data and/or (v) lack of a 1:1 relationship between (a) U in the whole surface (0-15 cm depth) soil, which is the dominant source of the ^{214}Bi signal for airborne gamma spectrometric determination of eU and (b) U determined by XRF in the <2mm fraction of topsoil collected from the 5-20cm depth interval. Data for the HiRES area appear to indicate that radon loss is not a major factor. This is because eU vs. U in <2mm surface soil regression lines are almost identical for (i) geological units likely to be characterised by coarse grained, permeable, less moisture retentive soils and (ii) fine grained, impermeable, moisture retentive soils. Whatever the reasons for the lack of the 1:1 relationship, the equation (Estimated <2mm topsoil U concentration = 1.72 x HiRES eU) is used in this study report to convert the HiRES eU data into estimated <2mm topsoil U concentrations, thereby facilitating comparison with regression models derived from the topsoil

(<2mm) U geochemical data. Estimated U in topsoil is then converted to estimated ^{226}Ra activity concentration (eRa226) for the regression analysis. Assuming U-natural (U-238) and ^{226}Ra are in equilibrium, estimated topsoil U derived from airborne gamma spectrometry surveys and U in soil samples determined by XRF analysis can be converted from mg kg^{-1} U to activity concentrations (Bq kg^{-1} of ^{238}U or ^{226}Ra) using the formula: 1 mg kg^{-1} U-nat = 12.35 Bq ^{238,234}U kg^{-1} = 12.35 Bq ^{226}Ra kg^{-1} if in equilibrium (IAEA, 1989).

2.3 Soil uranium geochemical data

The BGS regional and urban soil geochemical survey methods are described in Johnson et al. (2005) and Fordyce et al. (2005) and availability of soil U data in Beresford et al. (2007). Regional soil and urban samples are collected at a density of approximately 1 sample per 2 km^2 and 4 samples per km^2, respectively. Topsoil (A) soil samples collected from the 5–20 cm depth range, are sieved to pass <150 µm and subsurface (S) soil samples from 35- to 50-cm depth are sieved to pass a 2-mm mesh. In some cases this might be expected to enhance the U concentration in the size fraction analysed. In general U concentrations, determined in most of the samples by X-ray Fluorescence Spectroscopy (XRF), are approximately the same in the A and S samples, although there is variation at individual sites presumably reflecting a range of pedological, topographic and other factors. For the geological units of specific interest to the present study (DINLM, INOLMST and INONS), U is on average only ±5% different between the A (<2mm) and S (<150 micron) soils so these differences are unlikely to have a major impact when statistics and models are based on grouped data. Topsoil U data were used for the present study as there is likely to be a closer correlation between U in topsoils and the airborne radiometric data.

2.4 Soil gas and indoor radon data

The soil gas radon measurements used in the present study were made using a ‘Lucas cell’ type scintillation counter following extraction by pumping from a depth of 60-70 cm (see Ball et al., 1991 for further details). Uncertainties related to the measurement of radon in soil gas and the statistics derived from grouped data are discussed by Appleton et al (2000) and Emery et al. (2005). Indoor radon measurement methods and uncertainties for grouped indoor radon data are explained in Miles and Appleton (2005) and Hunter et al. (2005, 2009).

2.5 Regression analysis methods

Regression analysis based on the average values for spatially and geologically grouped data require that the data in each subset is approximately normally distributed so that the value used for the regression analysis is a robust central estimate. For regression analysis which incorporates a value for the uncertainty of the group average (i.e. standard deviation) then the distribution in each subset should be close to normal. It is well documented that indoor radon data are usually positively skewed and follow a generally lognormal distribution (Miles, 1998). Frequency distributions of skewness coefficients and Anderson-Darling normality tests were produced for a representative selection of data subsets (i.e. data grouped by 1-km or 5-km grid square and geology). The Anderson-Darling (AD) test compares the empirical cumulative distribution function of the data subset with the distribution expected if the data were normal. The AD test is especially effective at detecting departure from normality in the high and low values of a distribution. The AD tests indicated that regression analysis should be based on (1) arithmetic means for eRa226 derived from (a) HIRES airborne data and (b) U in <2mm surface soils; and (2) geometric means for soil gas radon and indoor radon data.

Three regression analysis methods which use very different algorithms have been applied and compared. These are: (i) Least squares (LS); (ii) Theil's method (TS) and (iii) Weighted total least-squares (WTLS). In LS analysis, the method makes the assumption that all the uncertainty is associated with y and that the y residuals (distances of y values from the calculated line) are normally distributed. If this is the case then the standard deviations on slope and intercept can also be calculated. The main drawbacks with the LS method for the data being considered here are: (i) there are significant uncertainties on both the x and the y data sets; and (ii) the results of the least squares method are not robust to outliers.

Theil's method (Theil, 1950) is a non-parametric approach to straight line fitting that can be expressed in the following algorithm: (i) the xy pairs are ranked according to their x value and split into two groups those above the median x value and those below (if the number of points is odd the median value is removed); and (ii) the slope of all combinations of xy pairs from the lower and upper sets is determined. For a given pair of points $(x_i, y_i), (x_j, y_j)$ where $x_j > x_i$

$$m_{ij} = \frac{(y_j - y_i)}{(x_j - x_i)}$$

The median value of all m_{ij} are then determined and used as the final slope value; and (iii) using the median value of m values for c_i the intercepts are estimated for each point using the equation:

$$c_i = y_i - mx_i$$

The median value for c_i is used as the final intercept value. The MATLAB implementation of this algorithm as described by Glaister (2005) has been used in this study. The advantages of this approach are: (i) it makes no assumptions about the errors being on the x or the y values; and (ii) it is robust to outliers. The disadvantage is that the method does not take account of the uncertainties on the x and y values.

A recent study has developed a new algorithm for fitting a straight line to data sets where there is uncertainty on both the x and the y axes (Krystek and Anton, 2007) called the total Weighted Total Least Squares Method (WTLS). Using this algorithm the problem is reduced to a one-dimensional search for a minimum. Global convergence and stability are assured by determining the angle of the straight line with respect to the abscissa instead of the slope. The complete uncertainty matrix is calculated, i.e. variances and covariance of the fitting parameters. The mathematical derivation and final equations for the model fitting are too lengthy to be included here and full details are given in Krystek and Anton (2007). The authors have written an implementation of the algorithm in the MATLAB programming language which has been used in this study. The advantage of this method compared to the LS is that the uncertainty on both the x and the y data are taken into account (although it is necessary to know what the uncertainties are on each x y point expressed as a standard deviation). Like LS, however, the method is not resistant to outliers in the data.

3. Results

3.1 Models derived from HiRES airborne data

Least squares (LS) regression equations, R^2 and significance data for three geological units with adequate data and representing different ground permeabilities are presented in Table 1 and illustrated in Figure 1. Plots of the regression models with and without the y axis intercept constrained to 5 Bq m^{-3} are illustrated in Figures 1 and 2. It is well established (see references in Scheib et al., 2006; Barnet et al., 2008; Kemski et al., 2005, 2006) that a specific ^{226}Ra or radon concentration in the ground will generally result in higher indoor radon concentrations when the ground has high permeability (for example over karstified limestones such as the Lower Carboniferous limestones of the English Midlands: DINLM) and low indoor radon concentrations when the ground is relatively impermeable (for example over the Triassic mudstones: TRIMD). Intermediate indoor radon concentrations will be associated with geological units that have moderate permeability (for example the Lower Carboniferous mudstone with subsidiary siltstone, sandstone, and limestone: DINMDMIX). For this reason, linear regression models for individual geological units with strongly contrasting permeability tend to be oriented above each other, as illustrated in Figure 1. Although no data are known to be available for the sedimentary terrains of the English Midlands, it is likely that the emanation coefficients of permeable soils and rocks (derived from sandstones, limestones and sand and gravel superficial deposits, for example) will be higher than emanation coefficients for fine-grained impermeable rocks such as mudstones. However, the dominant reason for the orientation of the regression models in Figure 1 is generally considered to be variations in the permeability of the ground. Multiple regression modelling using ground permeability data and soil variables (K and Th) which correlate with permeability in the Carboniferous, Permo-Triassic and Jurassic sedimentary terrains of Derbyshire tend to confirm this relationship (Scheib et al., 2006).

Regression lines for high permeability units intersected the y axis at high positive values (see for example Figure 1). These would imply unrealistically high contributions to indoor radon from sources other than the ground. The reason for the high intercepts may be related to the magnitudes of the errors on the input parameters and to the LS regression method. Initial studies of the impact of uncertainty of both the indoor radon and estimated soil ^{226}Ra data on the outcome of regression models showed that adding uncertainties on to data that is linear with a small positive intercept generally leads to a regression line with a significantly larger positive intercept than in the input data. It is possible to force a regression line to intercept the y axis at a particular value known to correspond to the physical situation. In an empirical model based on UNSCEAR data, Sumerling (2008) used a forced intercept of 16 Bq m^{-3} on the presupposition that 40% of the average indoor concentration is a constant related to outdoor air and building materials. UNSCEAR (1993) estimated the world mean outdoor radon concentration at 10 Bq m^{-3}, implying a world mean contribution from building materials of 6 Bq m^{-3}. However, an intercept of 16 Bq m^{-3} is not appropriate for the UK, where contributions from outdoor air and building materials are lower than the world mean. Wrixon et al (1988) showed that the mean outdoor radon concentration in the UK was much lower than the world mean, at 4 Bq m^{-3}. Gunby et al (1993) showed that the distribution of indoor radon concentrations in the UK was consistent with a lognormal distribution with a constant additional contribution of 4 Bq m^{-3}. Since any additional contribution from building materials would be expected to be normally, rather than lognormally, distributed, this implies that any contribution from building materials is very small on average. This conclusion is consistent with the results of measurements of radon emanation from UK building materials. A value of 5 Bq m^{-3} was assumed here for the contribution from outdoor radon and building materials together, and was used as a forced intercept in for the regression models indicated by the thin lines in Figure 1.

Comparison of the results of LS, TS and WTLS regression analysis based on eRa226 derived from HiRES airborne data and indoor radon data grouped by geology and 1-km grid square showed that 'All data', DINLM and DINMDMIX had slopes significantly different from 0 for both the WTLS and the LS methods (Figures 2-3, Table 2). The LS method showed a lower uncertainty than the WTLS method whilst the TS slope agreed with the LS and WTLS (within their uncertainties) for DINLM and DINMDMIX but not for 'all data' where it gave a lower slope. This suggests that for 'all data', outliers may have exerted an undue influence on the results.

For the LS method none of the groupings had an intercept significantly different from 1.61 (natural logarithm of 5 Bq m^{-3}, which is the average contribution to indoor radon from building materials and outside air), but the WTLS method showed significant intercepts for all groups (Figure 2 and 4; Table 2). The TS intercepts agrees quite well with the LS and WTLS intercepts.

3.3 Models based on soil geochemical data

The LS regression equations, R^2 and significance data for three geological units with adequate data and representing different ground permeabilities are presented in Table 3. Regression lines for geological units with different permeabilities are generally oriented above each other as illustrated in Figure 5 in which the high permeability Lower Carboniferous limestones (DINLM), Northampton Sand Formation (INONS) and Inferior Oolite limestones (INOLMST) lie above the relatively impermeable Westphalian (WESMDMIX) mudstone with subsidiary siltstone, sandstone units. Only the correlation coefficients for the INOLMST and 'All data' regression models were statistically significant ($p < 0.05$; Table 3). When the regression models were forced to intersect the y axis at 5 Bq m^{-3}, the regression lines for the permeable units (INONS, DINLM and INOLMST) were virtually coincident (Figure 5).

Although the regression models for most of the individual geological units were not statistically significant (probably due to the relatively small number of data points and the uncertainties in grouped indoor radon and soil U data used to produce the regression models), there was a logical relationship between the regression models for the permeable and impermeable geological units which is similar to the relationship observed for statistically significant regression models derived from the HiRES data. The slopes for the regression models forced to intersect the y axis at 5 Bq m^{-3} were similar for HiRES (Figure 1) and topsoil geochemical (Figure 5) data.

In the comparison of LS, TS and WTLS regression methods (Figures 6-7, 9; Table 4), the LS method showed that only 'all data' and INOLMST had a slope significantly different from zero whereas the WTLS showed that only 'all data' have a significant (but negative) slope. For 'all data' the TS method agreed with the LS method but not with the WTLS, whilst for the INOLMST, the TS slope agreed with the LS method slope (within the measured uncertainty). The LS method showed intercepts for all groups except TRIMD and WESDUMIX to be significantly greater than 1.61 (natural log of 5 Bq m^{-3}) whereas the WTLS method showed only 'all data' and DINLM to have intercepts significantly greater than this value (Figures 6, 8; Table 4). Taking into account the relatively large intercept standard deviations (Table 4), it is clear that the intercepts for INOLMST, INONS, TRIMD, ULI and WESDUMIX were not significantly different from 1.61 (natural log of 5 Bq m^{-3}).

3.4 Soil gas radon – indoor radon regression models

The LS regression equations, R^2 and significance data for five geological units with adequate data and representing different ground permeabilities are presented in Table 5. Plots of the regression models with and without the y axis intercept constrained to 5 Bq m^{-3} are illustrated in Figures 9 and 10. Statistically significant regression models for which the intercept is not fixed at 5 Bq m^{-3} were available for: (i) Northampton Sand Formation: 1-km grouped data (average SGRn); (ii) Northampton Sand Formation: 5-km grouped data; (iii) Lower Carboniferous limestones: 1-km grouped data collected in 2002-2004; (iv) Carboniferous and Permian data for Derbyshire and Nottinghamshire: 1-km grouped data; and (v) Carboniferous and Permian data for Derbyshire and Nottinghamshire: 5-km grouped data.

Only the third model of the above (Lower Carboniferous limestones in Derbyshire, 1-km grouping of 2002-2004 data) was statistically significant ($p < 0.05$) when the intercept was set to 5 Bq m^{-3}.

The slopes of the regression lines for the Carboniferous and Permian in Derbyshire and Nottinghamshire grouped by 1-km and 5-km grid square were likely to be steeper than regression lines for individual geological units. Unfortunately, sufficient data were not available to prove this. Regression models for other geological units grouped by 1-km and 5-km grid square were not statistically significant, probably largely due to the relatively small number of data points and the uncertainties in the grouped indoor radon and soil gas radon data used to produce the regression models.

It is difficult to recommend a single LS regression model for risk modelling and it is suggested that modelling be based on the two models that produce the lowest and highest indoor radon estimates as these probably provide a reasonable range for modelling. The linear regression models based on data from the UK are similar to those derived from arithmetic mean soil gas radon data in the Czech Republic (Barnet et al., 2008), but predict significantly higher indoor radon concentrations compared with models based on data from Germany where it is estimated that the ratio of indoor radon to soil gas radon ranges from about 0.002 to 0.0005 (Kemski et al., 2006), mainly because the German soil gas radon data are the maximum value of several measurements at one site whereas arithmetic means are used in the Czech Republic and GMs in the UK. Different national and regional house characteristics will also impact on indoor radon – soil gas radon regression models. Older buildings with ‘leaky’ floors are likely to be characterised by higher indoor radon for a specific soil gas concentration compared with buildings that have ‘gas-tight’ floors.

Comparison of the LS, TS and WTLS regression methods showed that ‘all data’, BGS Derbyshire Carb. Lmst. 1km grouping (2002-04 data), BGS Derby-Notts Carb-Perm 5km grouping and the BGS Derby-Notts Carb-Perm 1km grouping all had slope values significantly greater than 0 for the LS method (Figures 11-12; Table 6) . The WTLS method , however, only showed that only ‘all data’, BGS INONS 1km grouping and BGS Derby-Notts Carb-Perm 5km grouping had slope values greater than 0. Of the three methods, the WTLS had higher slope values. The Theil’s method agreed more closely with the LS than the WTLS.

The LS method showed all groupings to have intercepts significantly different from 1.61 (natural log equivalent of 5 Bq m^{-3}) apart from the BGS Derby-Notts Carb-Perm 1km grouping and Carb. Lmst 1km grouping (2002-04 data) (Figures 11, 13; Table 6). In contrast none of the intercepts were significantly different from this value for the WTLS method. In most instances the Theil’s method agreed more closely with the LS method apart from BGS Derby-Notts Carb-Perm 1km grouping where it agreed more closely with the WTLS method.

4. Comparison with published soil ^{226}Ra – indoor radon data regression models

Sheppard et al., (2006) compiled data to facilitate the assessment of the environmental impact of radioactive waste and used this to derive an average indoor radon : soil ^{226}Ra ratio with a lognormal distribution (geometric mean (GM) 1.5 (indicated by the long-dash line in Figures 14-15), geometric standard deviation (GSD) 2.6). The GMs of the data used by Sheppard et al. (2006) to derive this model are illustrated in Figure 14. An earlier model by Amiro (1992) has similar characteristics (GM 1.7, GSD 3). Sheppard et al.’s GMs were mainly derived from large surveys of areas representing a wide range of geological terrains and normal levels of soil ^{226}Ra. Most of the data reported in Sheppard et al., (2006) are clustered with no obvious correlation (Figure 14) although Sheppard et al.

(2006) noted that a correlation can be assumed on theoretical grounds. The GM for a uraniferous granite area in Finland (F-UGran in Figure 14; data from Voutilainen et al., 1988) plots very close to the lognormal distribution proposed by Sheppard et al. (2006). The GM for the alum shale area in Sweden (S-AlSh in Figure 14; data from Stranden and Strand, 1988) falls just outside the uncertainty bounds based on a indoor radon: soil ^{226}Ra ratio of 1.5 and the average indoor radon GSD for England and Wales corrected for measurement uncertainty (2.27; 1 GSD and 2 GSD bounds in Figures 14 and 15 are indicated by short-dash lines). Also plotted in Figure 14 is the least squares regression model for the Oslo area (derived from Figure 6 in Smethurst et al., 2008). This has a slightly higher ratio of about 1.9 but this would be expected because the model was based on average rather than GM indoor radon and soil estimated ^{226}Ra concentrations. The least squares linear regression models for the relatively impermeable Westphalian (WESMDMIX) mudstones with subsidiary siltstones and sandstones from the English Midlands plot parallel to the Sheppard et al. (2006) model but below it, whilst the models for the very permeable Northampton Sand Formation (INONS) and Lower Carboniferous Limestone (DINLM) plot parallel but above the Sheppard et al. (2006) model. Most of the Sheppard et al. (2006) data and data from this study fall within 1 GSD of the GM. Although it is speculative to base a conclusion on two extreme points (derived by Sheppard et al (2006) from data for the alum shale area in Sweden (S-AlSh) and uraniferous granite in Finland (F-UGran)), these support fixing intercepts at 5 Bq m^{-3} indoor radon for models derived from eRa226 estimated from U in BGS <2mm topsoil samples (Figure 14). Slopes and intercepts for LS, WTLS and Theil models based on ^{226}Ra derived from soil U data are essentially the same for DINLM, INOLMST, INONS, TRIMD, ULI and WESDUMDMIX (Table 3).

The relationships between LS models based on BGS HIRES data and HPA indoor radon data for England with the model data published by Sheppard et al., (2006) (Figure 15) are similar to the models for BGS soil geochemical data and HPA indoor radon data described above. The extensions of the DINLM and DINMDMIX models without fixed intercepts pass closer to the F-UGran and S-AlSh points than the models with intercepts fixed at 5 Bq m^{-3}.

5. Discussion and conclusions

The LS regression analysis demonstrated that relationships between ^{226}Ra in the ground, radon in soil and radon in dwellings differ depending on the characteristics of the underlying geological units. The results are applicable to the analysis of possible future scenarios for the LLWR and provide a range of models for ground with different permeabilities. The LS regression models for geological units with high permeability, analogous to the disturbed ground of the repository wastes and capping materials, predict higher levels of indoor radon for a specific estimated ^{226}Ra concentrations in the ground or soil gas radon concentrations than regression models for generally impermeable geological units. That being the case, it would be appropriate to use the models for the high permeability units in post-closure assessments of radon related to the radioactive waste in the LLWR as this will tend to be cautious. The results are consistent with work elsewhere, and the range of empirical relations for different geologies and with and without forced intercepts probably represent the range of possible outcomes from the situations modelled.

Uncertainties related to the measurement methods and GMs for grouped data used to formulate the regression models were identified and these impact on the regression model slope and intercept uncertainties. Whereas the GM radon concentrations in homes are reasonably robust, being based on 30 or more measurements in each case, there is still significant uncertainty in these values. The TS method should better deal with the impact of outliers on the regression model whilst WTLS analysis takes into account uncertainties in both the 'x' and 'y' axes and should therefore be the optimum and most reliable and robust regression analysis method for the estimated slopes and intercepts and their associated uncertainties.

From the comparison of LS, TS and WTLS methods, it was concluded that models for indoor radon vs. eRa226 derived from U measured in <2mm soil samples seem to have the poorest support with none of the groupings giving slopes significantly different from zero according to the WTLS method.

For models between indoor radon and (i) eRa226 in <2mm soil derived from HIRES eU data and (ii) soil gas radon data, some of the geological groupings had significant slopes according to the WTLS method. For these groupings there was reasonable agreement in slope and intercept between the three regression analysis methods (LS, TS and WTLS).

The distribution of radon in homes appears to be lognormal because factors determining how much radon gets from the source into the home are multiplicative (Gunby et al, 1993). We need to determine whether the relationship between the source (or surrogate for the source, ^{226}Ra estimated from airborne gamma or U in soil) and the output (radon in homes) is linear. If the factors controlling the passage of radon from the source to the output were to vary from one area to another, we would expect to see significant variations in GSD from one area to another, since the GSD depends on those factors. In particular, if the factors controlling the passage of radon from the source to the output were to vary depending on the strength of the source, then we would expect to see systematic variation in GSD with GM. In fact we observe little or no variation in GSD with GM (Miles, 1998), implying that the factors controlling the passage of radon from the source to the output do not vary with GM. This in turn implies that the relationship between the source and the output should be linear, since in each case the same set of multiplicative controlling factors apply. Hence doubling the ^{226}Ra in the ground should double concentrations in homes, giving a lognormal distribution which, if plotted as a histogram on a logarithmic x axis, is just shifted to the right without changing shape. If the measurements of the source term (^{226}Ra) have normally distributed uncertainties, the appropriate variables to use for modelling would be arithmetic mean source value and GM radon in homes (Figures 1 and 5 above) . In contrast, the relationship between soil gas and indoor radon (Figures 9 and 10) is best defined using GMs on both axes because both variables are lognormally distributed. It therefore seems to be appropriate to use these models for predicting indoor radon levels which are likely to occur at the ^{226}Ra and soil gas radon concentrations found or predicted at near-surface radioactive waste disposal sites. These models should be used in preference to those based on the log (y)-linear (x) relationship between LnGM indoor radon and estimated ^{226}Ra (Figures 2 and 6), which was used in this study for the comparison between WTLS, LS and Theil regression models. The LnGM radon was used on the y axis because the uncertainty (expressed by the LnGSD) had to be symmetrically distributed either side of each data point for the WTLS regression analysis. There is little difference between the indoor radon concentrations predicted by the GM indoor radon – eRa226 and LnGM indoor radon – eRa226 regression lines at the upper limits of the data used to construct the models. However, at the higher concentrations which may in some circumstances be encountered above near-surface radioactive waste disposal sites, the LnGM indoor radon – eRa226 regression models predict higher radon concentrations than the GM indoor radon – eRa226 models, when the intercepts for both types of models are unconstrained.

Intercepts of 4-5 Bq m^{-3} would be expected for all regression analysis models as this is the average contribution to indoor air from radon in outdoor air and building materials for dwellings in the UK. The soil gas data vs. indoor radon regression models have intercepts of 4 Bq m^{-3} for both the INONS (1-km grouping) and the Derby-Notts Carb-Perm 5-km grouping. In contrast, the intercepts for the eRa226 (HIRES) vs. indoor radon models have intercepts of 18 Bq m-3 for DINMDMIX and 42 Bq m^{-3} for DINLM (1 Bq m^{-3} for 'all data').

A key issue is whether to use models with an intercept constrained for theoretical reasons to 5 Bq m^{-3}, which represents the average value for the contribution from outdoor radon and building materials), or unconstrained models based solely on the empirical data. Slopes of the fixed intercept models are generally steeper than the slopes for unconstrained models. Extrapolation of fixed intercept models to the likely ^{226}Ra and soil gas radon concentrations that may be encountered in certain circumstances in future over the LLWR will predict higher indoor radon concentrations than will extrapolation of unconstrained models. However, models with and without a constrained intercept for the value of indoor radon gave results that were generally within a factor of two of each other, suggesting that, allowing for uncertainties on the inputs, such extrapolation of the constrained models may be justified.

Comparison with published data used to derive an average indoor radon : soil radium ratio showed that the LS models for the relatively impermeable English geological units plot parallel to the Sheppard et al. (2006) model but below it (i.e. lower indoor radon : soil radium ratios), whilst the models for the permeable English geological units plot parallel but above the Sheppard et al. (2006) model (i.e. have higher indoor radon : soil radium ratios).

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References

Amiro, B.D., 1992. The Atmosphere Submodel for the Assessment of Canada's Nuclear Fuel Waste Management Concept. Report AECL-9889, COG-91-199. Atomic Energy of Canada Limited, Pinawa, Manitoba.

Appleton, J.D., Miles, J.C.H., Talbot, D, 2000. Dealing with radon emissions in respect of new development: Evaluation of mapping and site investigation methods for targeting areas where new development may require radon protective measures. British Geological Survey Research Report RR/00/12.

Appleton, J.D., Miles, J.C.H., Green, B.M.R., Larmour, R. 2008. Pilot study of the application of Tellus airborne radiometric and soil geochemical data for radon mapping. Journal of Environmental Radioactivity, 99, 1687-1697.

Appleton, J.D., Miles, J.C.H., 2009. A statistical evaluation of the geogenic controls on indoor radon concentrations and radon risk. Journal of Environmental Radioactivity doi:10.1016/j.jenvrad.2009.06.002 Available online 3 July 2009

Ball, T.K., Cameron, D.G., Colman, T.B., Roberts, P.D., 1991. Behaviour of Radon in the Geological Environment - a Review. Quarterly Journal of Engineering Geology. 24(2), 169-182.

Barnet, I., Pacherová, P., Neznal, M., Neznal, M., 2008. Radon in Geological Environment – Czech Experience. Czech Geological Survey Special Paper 19. Prague: Czech Geological Survey.

Beresford, N.A., Appleton, J.D., Barnett, C.L., Bescoby, M.W., Breward, N., Jones, D.G., MacKenzie, A.C., Scheib, C., Wood, M.D., Thørring, H., 2007b. Assessment of naturally occurring radionuclides around England and Wales. Project SC030283. Environment Agency, Bristol. Available from: http://www.ceh.ac.uk/protect/pages/documents/AssessmentofnaturallyoccurringradionuclidesinEnglandandWales.pdf

Emery, C., Hodgkinson, E., Jones, D.G., Davis, J. Strutt, M., 2005. Investigation of airborne and ground-based radiometric data with soil gas radon to assess variability of indoor radon potential for the Carboniferous limestone of Derbyshire. British Geological Survey Internal Report, IR/05/061. .

Fordyce, F.M., Brown, S.E., Ander, E. L., Rawlins, B.G., O'Donnell, K. E., Lister, T.R., Breward, N., Johnson, C.C., 2005. GSUE: Urban geochemical mapping in Great Britain. Geochemistry: Exploration-Environment-Analysis 5(4), 325-336.

Glaister, P., 2005. Robust linear regression using Theil's method. Journal of Chemical Education, Vol. 82, 1472-1473.

Grasty, R.L., 1997. Radon emanation and soil moisture effects on airborne gamma-ray measurements. Geophysics. 62 (5), 1379-1385.

Gunby, J.A., Darby, S.C., Miles, J.C.H., Green, B.M.R., Cox, D.R., 1993. Factors affecting indoor radon concentrations in the United Kingdom. Health Physics 64, 2-12.

Hunter, N., Howarth, C.B., Miles, J.C.H., Muirhead, C.R., 2005. Year-to-year variations in radon levels in a sample of UK houses with the same occupants. in: McLaughlin, J.P., Simopoulos, E.S., Steinhäusler, F. (Editors), Seventh International Symposium on the Natural Radiation Environment (NRE-VII) Rhodes, Greece, 20-24 May 2002. Elsevier, pp. 438-447.

Hunter, N., Muirhead, C.R., Miles, J. C. H., Appleton, J. D., 2009. Uncertainties in radon related to house-specific factors and proximity to geological boundaries in England. Radiation Protection Dosimetry, Vol. 136 (1), 17–22.

IAEA, 1989. Construction and use of calibration facilities for radiometric field equipment. Technical Report Series No. 309, IAEA, Vienna.

IAEA. 2003. Guidelines for radioelement mapping using gamma ray spectrometry data. IAEA-TECDOC-1363. IAEA, Vienna

Johnson, C.C., Breward, N., Ander, E.L., Ault, L., 2005. G-BASE: Baseline geochemical mapping of Great Britain and Northern Ireland. Geochemistry: Exploration-Environment-Analysis 5, 1-13.

Kemski, J., Klingel, R., Siehl, A. Stegemann., R. 2005. Radon transfers from ground to houses and prediction of indoor radon in Germany based on geological information. Seventh International Symposium on the Natural Radiation Environment (NRE-VII), Rhodes, Greece, 2002. Radioactivity in the Environment, VII, 820-832.

Kemski, J., Klingel, R., Siehl. A., Valdivia-Manchego, M. R., 2006. Radon risk prediction based on gridded geological maps and soil gas measurements. in: Barnet, I., Neznal, M., Pacherova, P., Radon investigations in the Czech Republic XI & the Eighth International Workshop on the Geological Aspects of Radon Risk Mapping, Prague: Czech Geological Survey and Radon corp., pp. 139-156.

Krystek, M. Anton., M. 2007. A weighted total least-squares algorithm for fitting a straight line. Measurement Science and Technology, 18, 3438-3442.

Miles, J.C.H., 1998. Mapping radon prone areas by lognormal modelling of house radon data. Health Physics 74, 370-378.

Miles, J.C.H., Appleton, J.D., 2005. Mapping variation in radon potential both between and within geological units. Journal of Radiological Protection 25, 257-276.

Peart, R.J., Cuss, R. J., Beamish, D., Jones, D.G., 2004. The High Resolution Airborne Resource and Environmental Survey- (Phase 1) (HiRES-1): background, data processing and dissemination and future prospects. British Geological Survey, IR/03/112.

Scheib, C., Appleton, J.D., Jones, D.G., Hodgkinson, E., 2006. Airborne uranium data in support of radon potential mapping in Derbyshire, Central England. in: Barnet, I., Neznal, M. Pacherova, P., Radon investigations in the Czech Republic XI & the Eighth International Workshop on the Geological Aspects of Radon Risk Mapping, Prague: Czech Geological Survey and Radon corp., pp. 210-219.

Sheppard, S.C., Sheppard, M.I., Tait, J.C., Sanipellia, B.L., 2006. Revision and meta-analysis of selected biosphere parameter values for chlorine, iodine, neptunium, radium, radon and uranium. Journal of Environmental Radioactivity, 89 (2), 115-137

Smethurst, M.A., Strand, T., Sundal, A.V., Rudjord, A. L., 2008. Large-scale radon hazard evaluation of the Oslofjord region of Norway utilizing indoor radon concentrations, airborne gamma ray spectrometry and geological mapping. Science of the Total Environment, 407: 379-393.

Stranden, E., Strand, T., 1988. Radon in an alum shale rich Norwegian area. Radiation Protection Dosimetry 24, 367-370.

Sumerling T., 2008. LLWR Lifetime Plan: Performance Assessment Update for the LLWR, LLW Repository Ltd report 10005 LLWR LTP Volume 5 Issue 1, 30 April 2008.

Theil, H. 1950. A rank-invariant method of linear and polynomial regression analysis. Indagationes Mathematicae, 12, 85-91.

UNSCEAR, 1993 UNSCEAR, United Nations scientific committee on the effects of atomic radiation, Sources and effects of ionizing radiations, United Nations, New York .

UNSCEAR, 2000, Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly, Annex B: Exposure from Natural Sources. United Nations, New York.

Voutilainen, A., Castre'n, O., Mäkeläinen, I., Winqvist, K., Arvela, H., 1988. Radiological characteristics of a village on uraniferous granitic ground in Finland. Radiation Protection Dosimetry 24, 333-337.

Wrixon, A.D., Green, B.M.R., Lomas, P.R., Miles, J.C.H., Cliff, K.D., Francis, E.A., Driscoll, C.M.H, James, A.C., and O'Riordan, M.C., 1988. Natural radiation exposure in UK dwellings. NRPB R-190, May 1988.

Table 1. LS linear regression models derived from HiRES data converted to estimated Ra226 in <2mm fraction of topsoils

Data set	Model formula	No. of data points	R^2	Significance (p)
HiRES DINLM 1-km grouping	y = 2.15x + 52	51	0.21	<0.05
HiRES DINMDMIX 1-km grouping	y = 1.3x + 17	39	0.29	<0.05
HiRES all data 1-km grouping	y = 3.03x – 35	296	0.32	<0.05
HiRES TRIMD 1-km grouping	y = 0.04x + 20	35	0.01	>0.05
Data set	Model formula with 5 Bq/m3 indoor radon intercept			
HiRES DINLM 1-km grouping	y = 3.13x + 5	51	0.16	<0.05
HiRES DINMDMIX 1-km grouping	y = 1.62x + 5	39	0.27	<0.05
HiRES all data 1-km grouping	y = 1.88x + 5	296	0.27	<0.05
HiRES TRIMD 1-km grouping	y = 0.58x + 5	35	-0.34	>0.05

(p) <0.05 = R significant at the 95% confidence level; >0.05 = R not significant at the 95% confidence level

Table 2 Slope, intercept and associated uncertainties of log GM indoor radon against eRa-226 for models for a range of geological groups (based on HIRES data).

	Theil's method					
Grouping	Slope	Intercept				
All data	0.023	2.88				
DINLM	0.015	4.13				
DINMDMIX	0.017	3.17				
NAM & WESMDMIX	-0.025	3.53				
NAM & WESSD	-0.006	3.66				
TRIMD	-0.003	2.95				
PERDO	-0.021	4.06				
INOLMST	-0.044	5.51				
	WTLS method					
Grouping	Slope	Intercept	sd slope	sd intercept		
All data	0.051	2.00	0.04	0.21		
DINLM	0.022	3.89	0.02	0.45		
DINMDMIX	0.022	3.10	0.02	0.36		
NAM & WESMDMIX	-0.034	3.85	0.08	0.87		
NAM & WESSD	-0.010	3.78	2.03	1.11		
TRIMD	-0.003	2.92	0.09	1.23		
PERDO	-0.009	3.67	1.74	1.83		
INOLMST	-0.140	8.17	0.83	3.22		
	Least Squares					
Grouping	Slope	Intercept	95%ul slope	95%ll slope	95%ul intercept	95%ll intercept
All data	0.037	2.47	0.029	0.045	2.20	2.73
DINLM	0.016	4.16	0.008	0.024	3.78	4.54
DINMDMIX	0.018	3.28	0.008	0.028	2.89	3.67
NAM & WESMDMIX	-0.028	3.65	-0.046	-0.009	3.11	4.19
NAM & WESSD	-0.017	3.96	-0.054	0.020	3.03	4.89
TRIMD	0.002	2.80	-0.017	0.020	2.27	3.33
PERDO	-0.015	3.89	-0.064	0.033	2.45	5.33
INOLMST	-0.055	5.76	-0.110	0.000	4.18	7.35

sd = standard deviation; ul = upper limit; ll = lower limit

Table 3. LS linear regression models derived from U in topsoil samples converted to estimated Ra226 in <2mm fraction of topsoils

Data set	Model formula	No. of data points	R^2	Significance (p)
Topsoil DINLM 5-km grouping	y = 0.94x + 97	17	0.08	>0.05
Topsoil INONS 5-km grouping	y = 1.86x + 39	11	0.18	>0.05
Topsoil INOLMST	y = 2.57 + 7	10	0.62	<0.05
Topsoil all data 5-km grouping	y = 1.98x – 4	172	0.13	<0.05
Topsoil WESMDMIX 5-km grouping	y = 0.14x + 17	21	0.01	>0.05

Data set	Model formula with 5 Bq/m3 indoor radon intercept			
Topsoil DINLM 5-km grouping	y = 3.05x + 5	17	-0.37	>0.05
Topsoil INONS 5-km grouping	y = 3.3x + 5	11	0.06	>0.05
Topsoil INOLMST	y = 2.64x + 5	10	0.61	<0.05
Topsoil all data 5-km grouping	y = 1.68x + 5	172	0.13	<0.05
Topsoil WESMDMIX 5-km grouping	y = 0.48x + 5	21	-0.08	>0.05

Table 4 Slope, intercept and associated uncertainties of log GM indoor radon against eRa-226 for the different geological groupings (eRa226 derived from soil uranium data).

	Theil's method					
Grouping	Slope	Intercept				
All data	0.015	3.05				
DINLM	0.007	4.51				
INOLMST	0.035	3.27				
INONS	0.033	3.42				
TRIMD	0.009	2.67				
ULI	0.018	2.83				
WESDUMDMIX	0.012	2.41				
	WTLS method					
Grouping	Slope	Intercept	sd slope	sd intercept		
All data	-0.085	6.12	0.025	0.77		
DINLM	0.011	4.34	0.030	1.35		
INOLMST	0.048	2.91	0.047	1.15		
INONS	0.031	3.49	0.052	1.25		
TRIMD	0.027	2.20	0.074	2.41		
ULI	0.025	2.58	0.035	1.12		
WESDUMDMIX	0.012	2.43	0.074	2.50		
	Least Squares					
Grouping	Slope	Intercept	95% ll slope	95% ul Slope	95% ll intercept	95% ul intercept
All data	0.027	2.81	0.01	0.043	2.22	3.39
DINLM	0.007	4.55	-0.01	0.018	4.00	5.10
INOLMST	0.041	3.11	0.02	0.066	2.46	3.76
INONS	0.027	3.60	-0.02	0.076	2.44	4.76
TRIMD	0.009	2.79	-0.05	0.070	0.80	4.78
ULI	0.022	2.70	0.00	0.043	2.02	3.38
WESDUMDMIX	0.011	2.46	-0.02	0.045	1.31	3.60

Table 5. LS linear regression models derived using soil gas radon data

Data set	Model formula	N	R^2	Significance (p)
Derby-Notts Carb-Perm 1km grouping	y = 1.97x + 56	13	0.25	<0.05
Derby-Notts Carb-Perm 5km grouping	y = 1.1x + 37	75	0.09	<0.05
INONS 5-km grouping	y = 2.0x + 53	10	0.35	<0.05
INONS 1-km grouping	y = 1.90x + 42	17	0.23	<0.05
Derby Carb. Lmst. 1-km grouping	y = 0.83x + 66	6	0.88	<0.05

Data set	Model formula with 5 Bq/m3 indoor radon intercept			
Derby-Notts Carb-Perm 1km grouping	y = 3.14x + 5	13	0.13	>0.05
Derby-Notts Carb-Perm 5km grouping	y = 2.31x + 5	75	-0.11	>0.05
INONS 5-km grouping	y = 4.49x + 5	10	-0.40	>0.05
INONS 1-km grouping	y = 3.52x + 5	17	0.01	>0.05
Derby Carb. Lmst. 1-km grouping	y = 1.15x + 5	6	0.68	<0.05

Table 6 Slope, intercept and associated uncertainties of log GM indoor radon against log GM soil gas radon for the different geological groupings.

	Theil's method					
Grouping	Slope	Intercept				
All data	0.455	2.73				
BGS INONS 1km grouping	0.358	3.26				
BGS INONS 5 km grouping	0.101	4.13				
BGS Derbyshire Carb Lmst 1km grouping (2002-04 data)	0.405	3.35				
BGS Derby-Notts Carb-Perm 5km grouping	0.364	2.68				
BGS Derby-Notts Carb-Perm 1km grouping	1.171	0.22				
	WTLS method					
Grouping	Slope	Intercept	sd slope	sd intercept		
All data	0.840	1.63	0.141	0.40		
BGS INONS 1km grouping	0.964	1.43	0.376	1.00		
BGS INONS 5 km grouping	0.254	3.85	0.590	1.40		
BGS Derbyshire Carb Lmst 1km grouping (2002-04 data)	0.568	2.46	0.526	2.41		
BGS Derby-Notts Carb-Perm 5km grouping	0.913	1.34	0.294	0.78		
BGS Derby-Notts Carb-Perm 1km grouping	1.315	0.04	0.705	2.42		
	LS					
Grouping	Slope	Intercept	95% ll slope	95% ul Slope	95% ll intercept	95% ul intercept
All data	0.438	2.81	0.308	0.569	2.45	3.18
BGS INONS 1km grouping	0.297	3.28	-0.124	0.718	2.18	4.39
BGS INONS 5 km grouping	0.112	4.15	-0.261	0.484	3.26	5.04
BGS Derbyshire Carb Lmst 1km grouping (2002-04 data)	0.453	3.01	0.114	0.793	1.50	4.51
BGS Derby-Notts Carb-Perm 5km grouping	0.376	2.80	0.185	0.566	2.30	3.29
BGS Derby-Notts Carb-Perm 1km grouping	0.783	1.94	0.153	1.412	-0.14	4.02

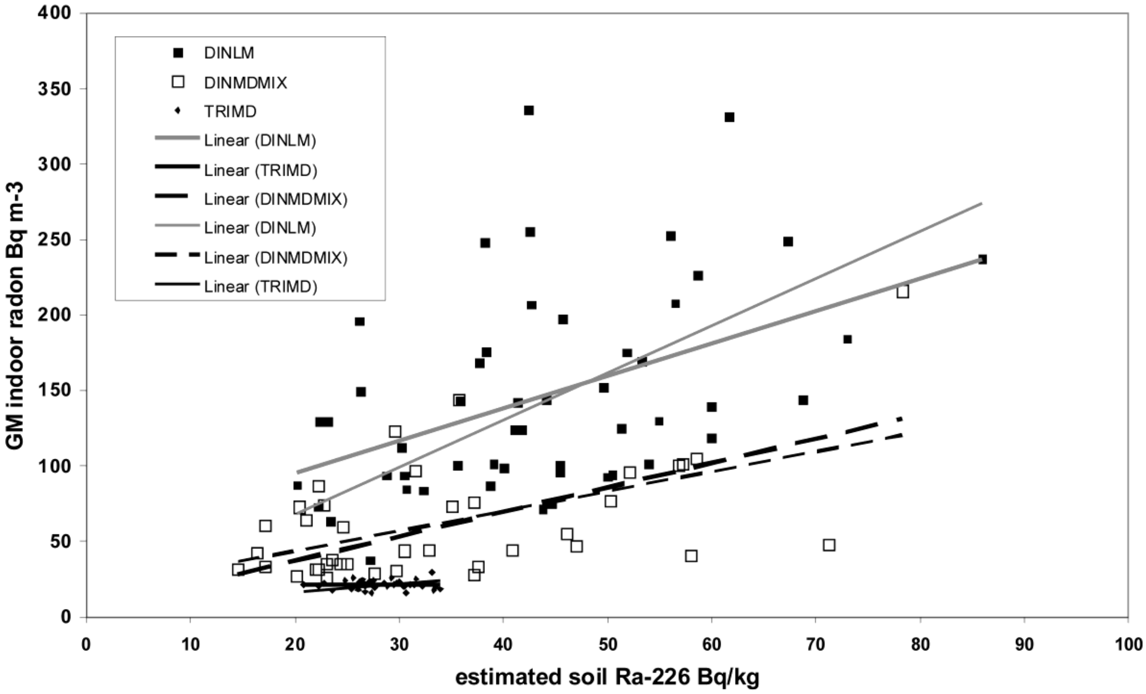

Figure 1. LS regression models between estimated soil ^{226}Ra (derived from HiRES airborne radiometric eU) and indoor radon with data grouped by 1km-geology (thin regression lines have intercepts set to 5 Bq m^{-3}).

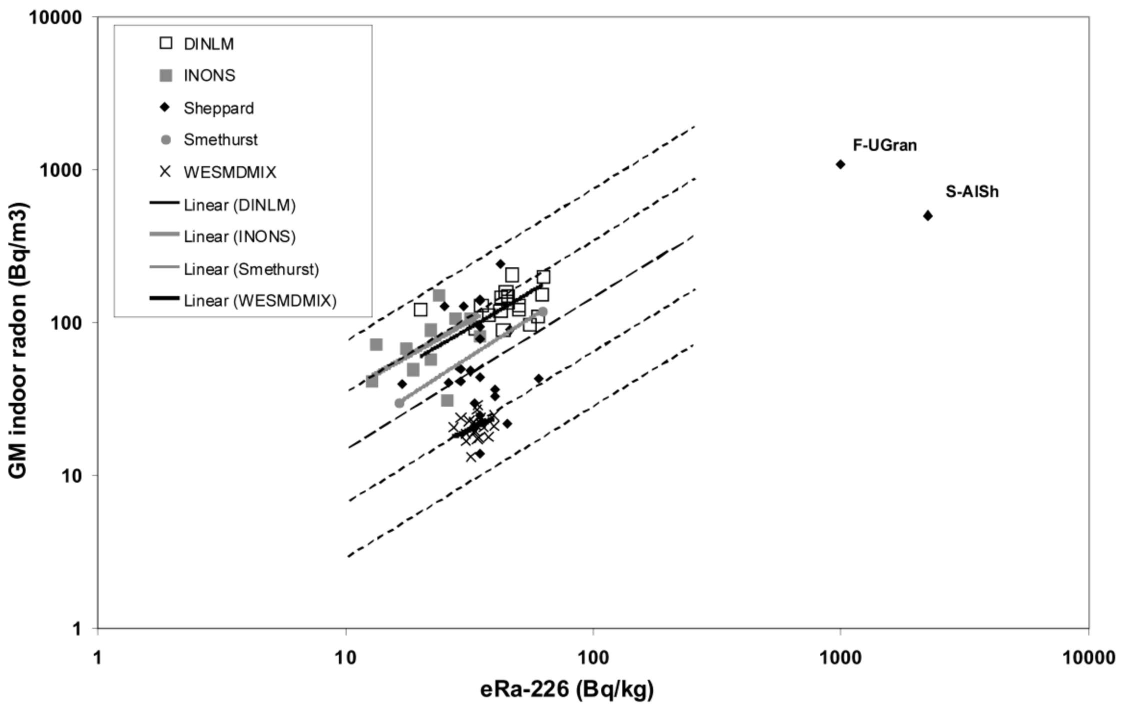

Figure 2 Plot of the $e^{226}Ra$ regression lines derived from HIRES airborne data

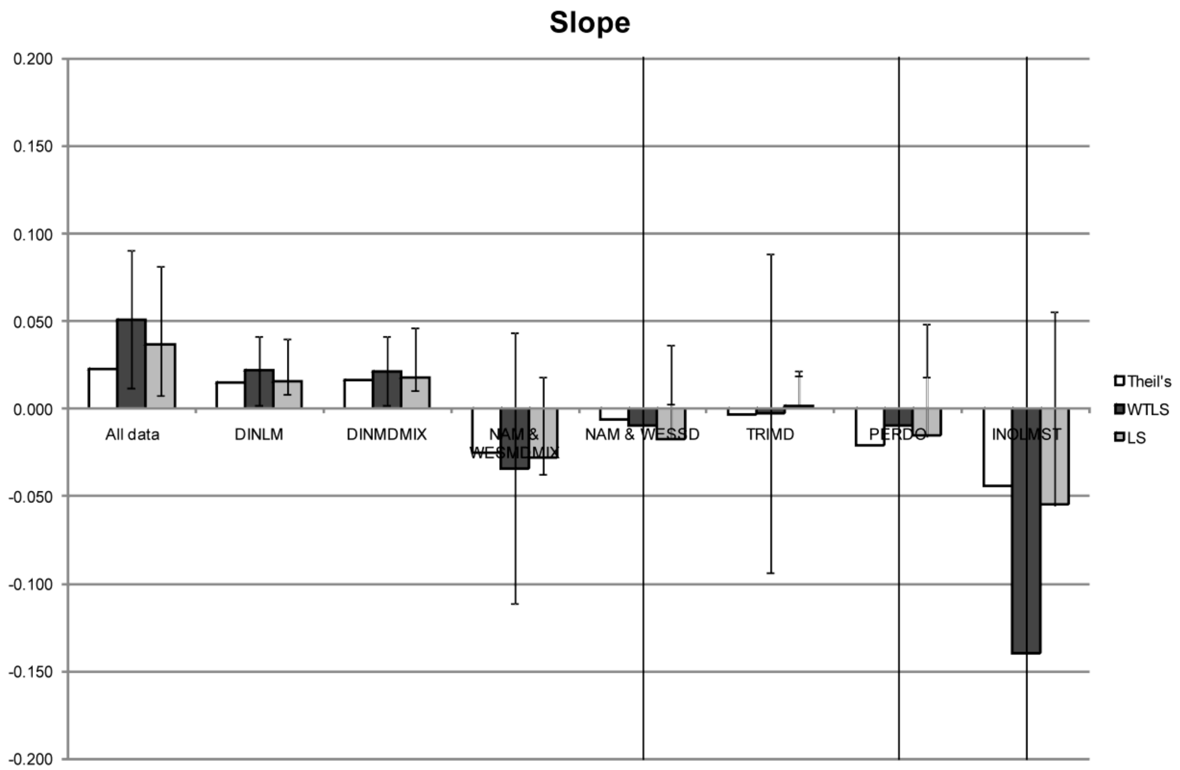

Figure 3 Comparison of the slopes (Log GM indoor radon/e^{226}Ra Bq kg^{-1}) and their 95% confidence limits for the regression lines derived from HIRES data.

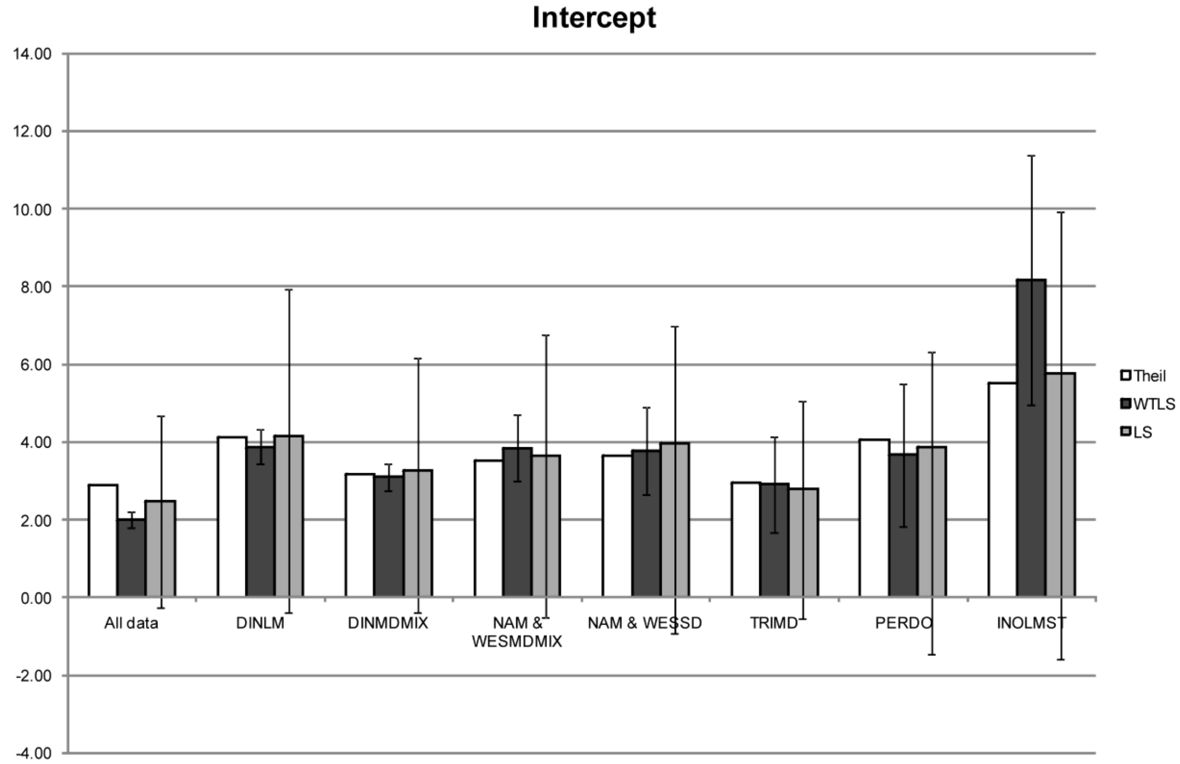

Figure 4 Comparison of the intercepts (Log GM indoor radon) and their 95% confidence limits for the regression lines derived from the HIRES airborne radiometric data.

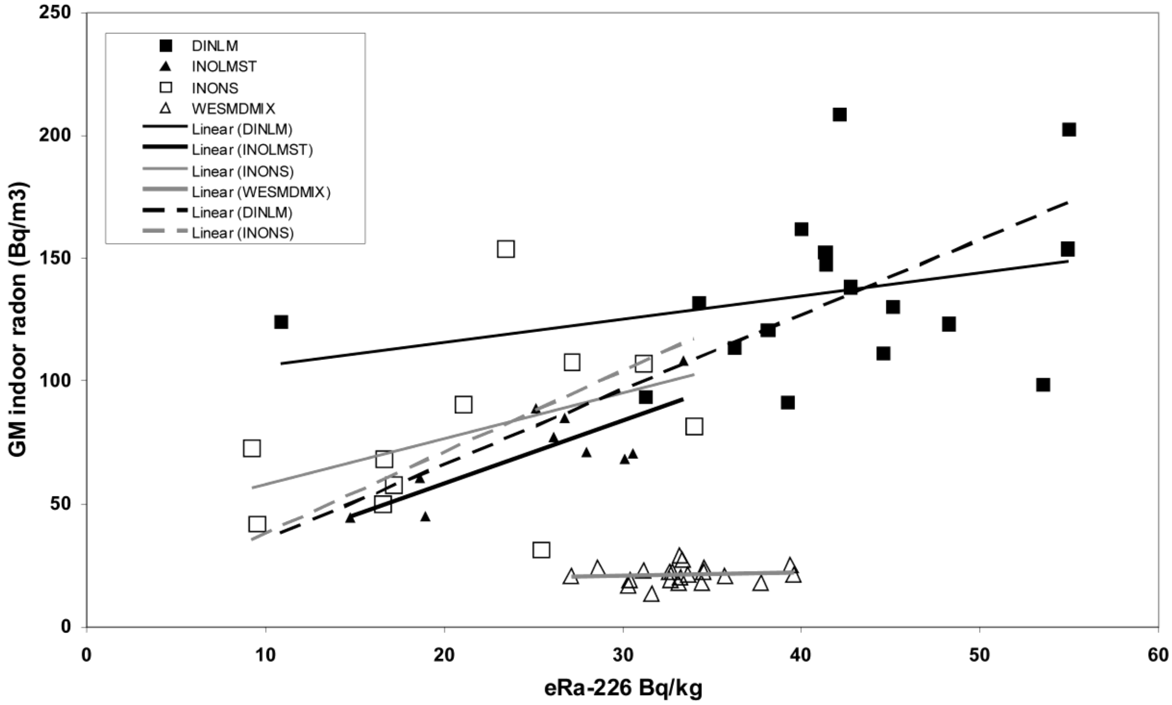

Figure 5. Regression models between $e^{226}Ra$ (derived from U in topsoil samples) and indoor radon with data grouped by 5km-geology (dashed regression lines have intercepts set to 5 Bq m^{-3}).

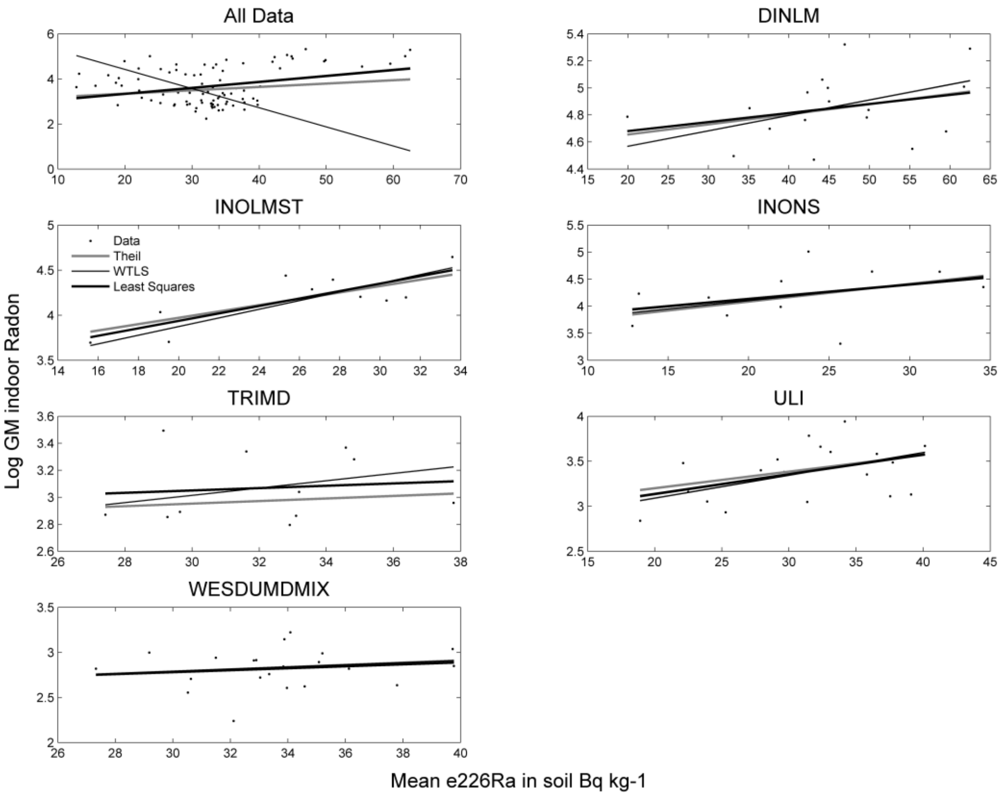

Figure 6 Plot of the $e^{226}Ra$ regression lines derived from soil U data.

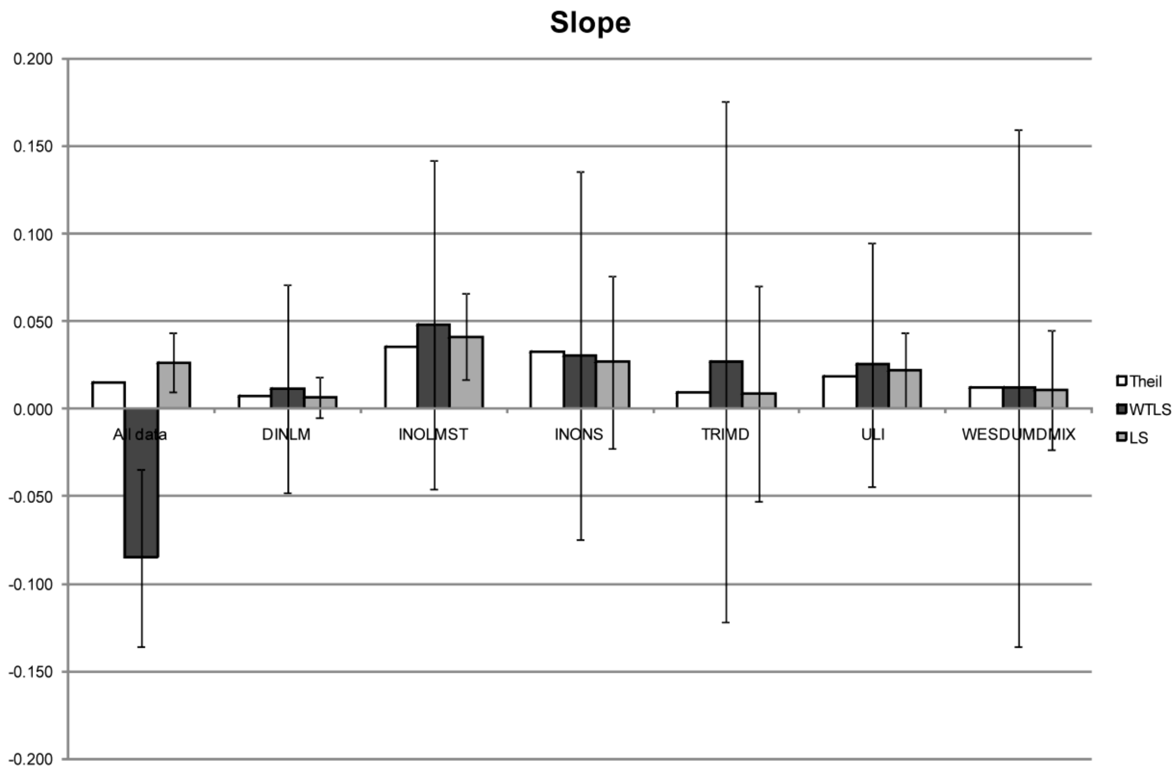

Figure 7 Comparison of the slopes (Log GM indoor radon/mean e^{226}Ra) of the regression lines and their 95% confidence limits for models derived from soil U geochemical data.

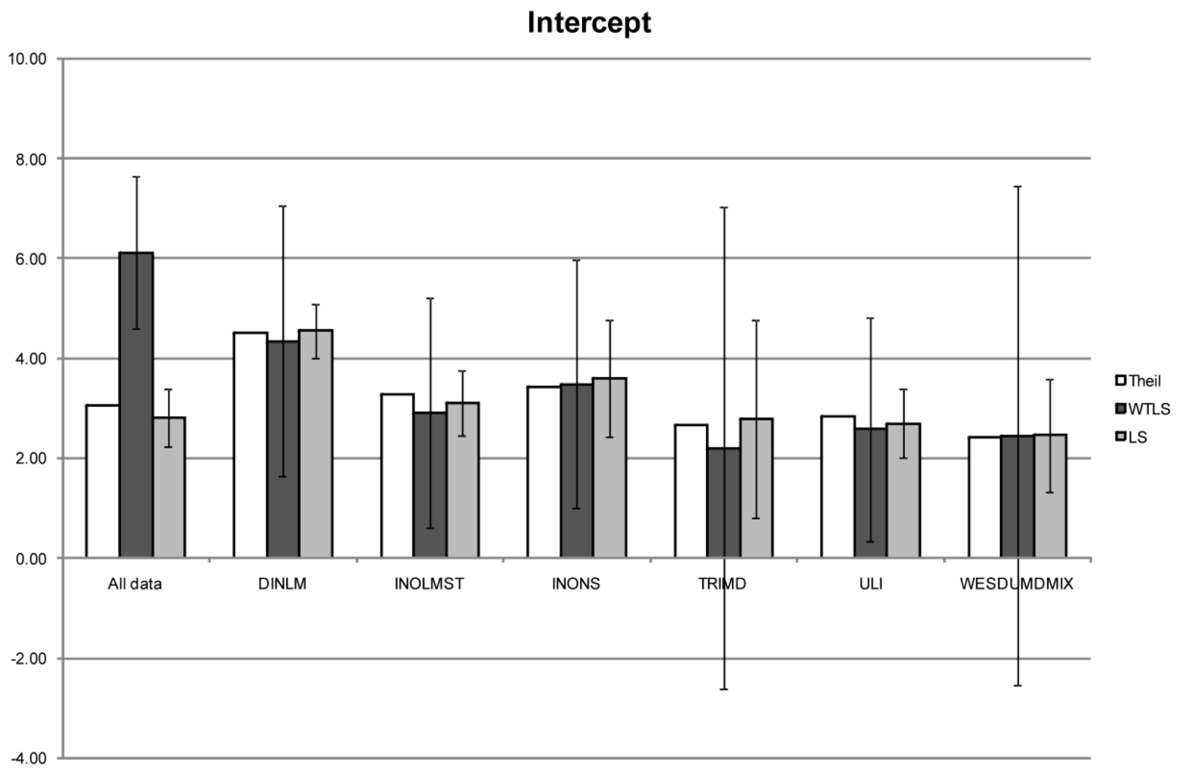

Figure 8 Comparison of the intercepts (Log GM indoor radon) of the e^{226}Ra regression lines and their 95% confidence limits for regression models derived from soil U geochemical data.

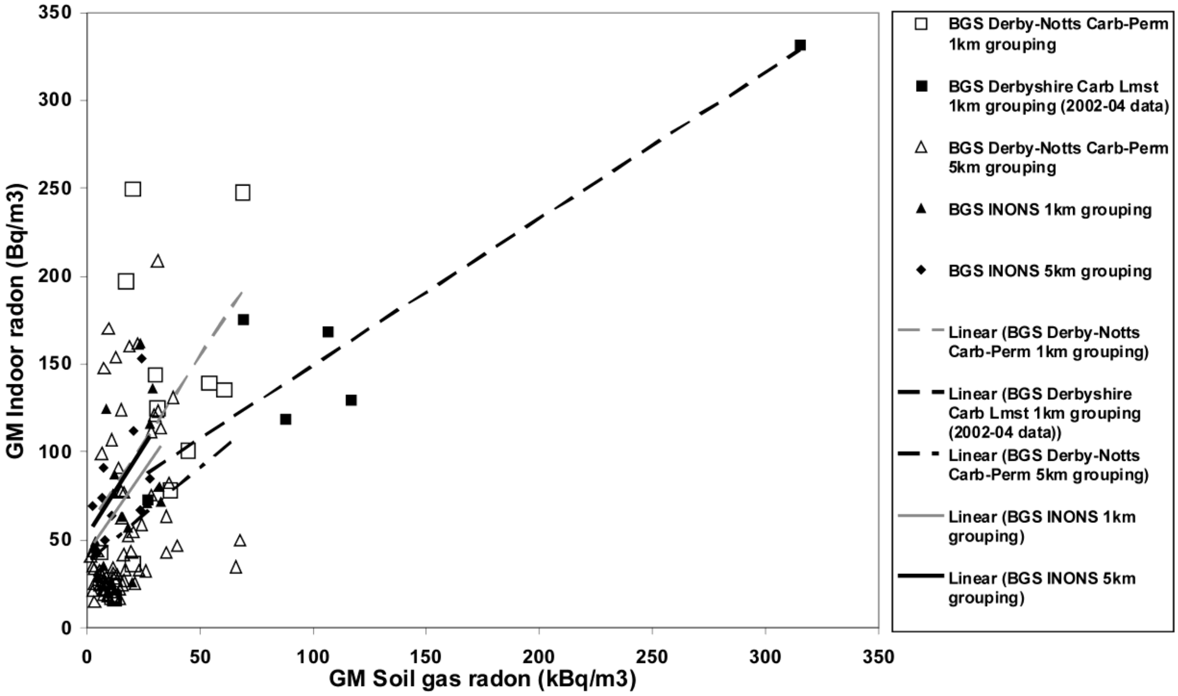

Figure 9 Relationship between GM soil gas radon (n>4) with GM indoor radon (n>19) with data grouped by 1-km or 5-km grid square and geology: data for Carboniferous and Permian of the English Midlands and the Northampton Sand Formation.

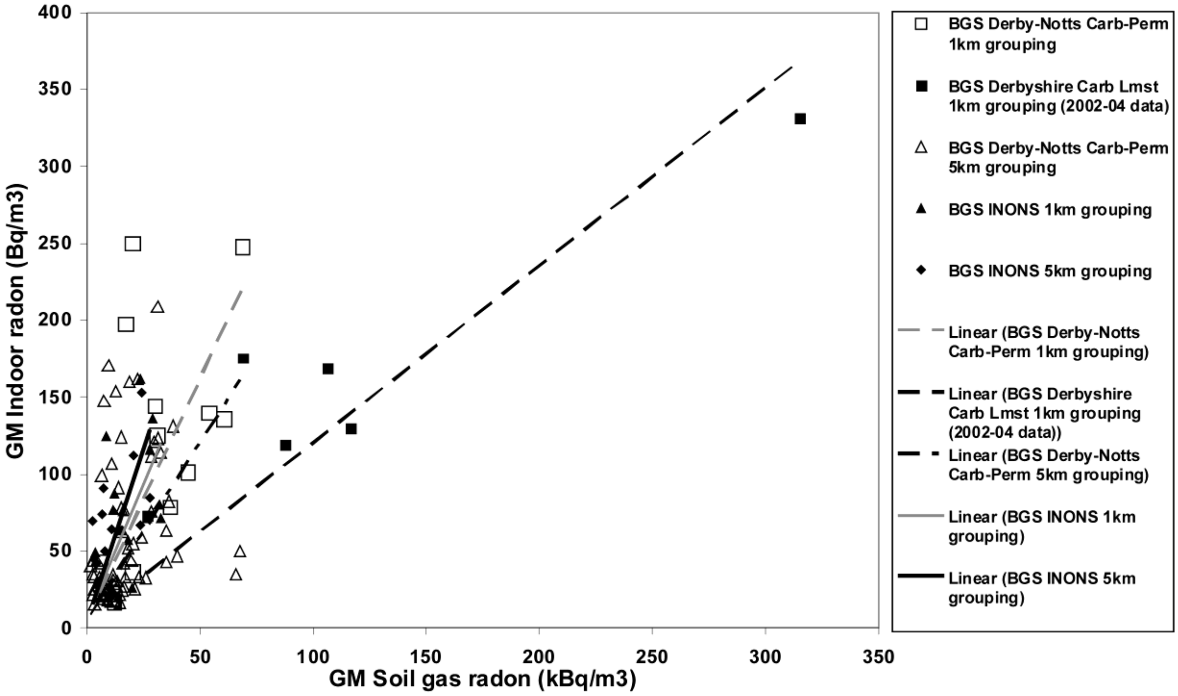

Figure 10. Relationship between GM soil gas radon (n>4) with GM indoor radon (n>19) with data grouped by 1-km or 5-km grid square and geology: data for Carboniferous and Permian of the English Midlands and the Northampton Sand Formation (intercepts set at 5 Bq m^{-3})

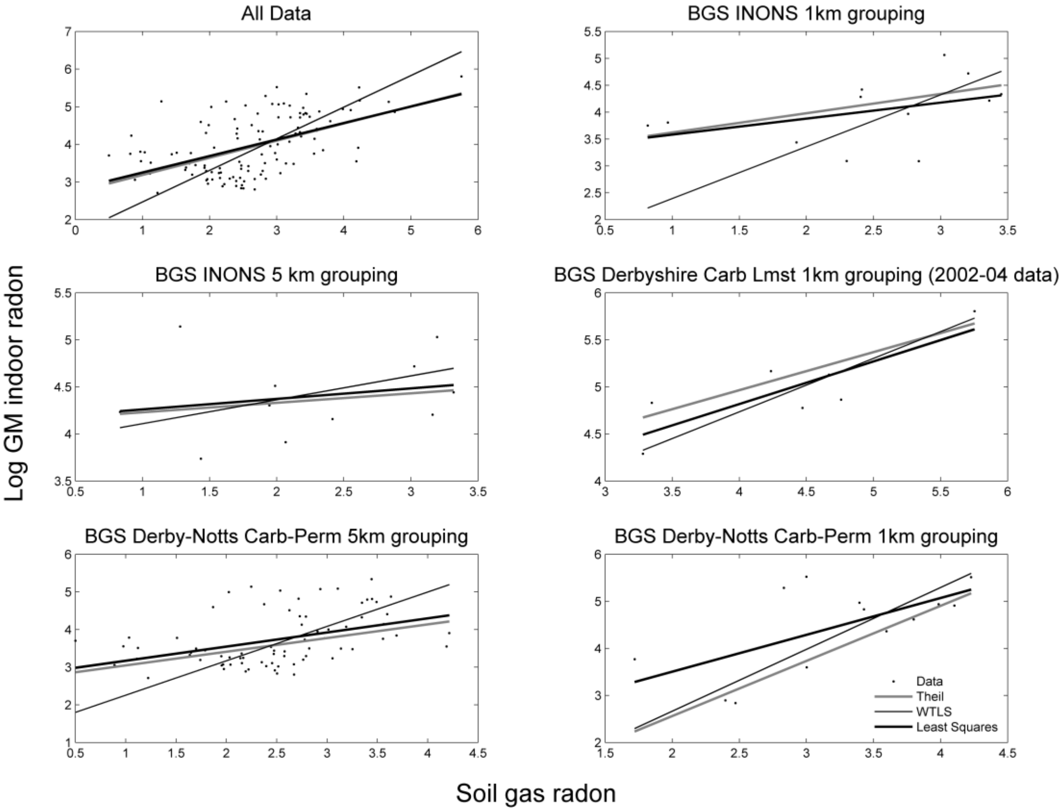

Figure 11 Plot of the regression lines based on soil gas radon data (Log scale) .

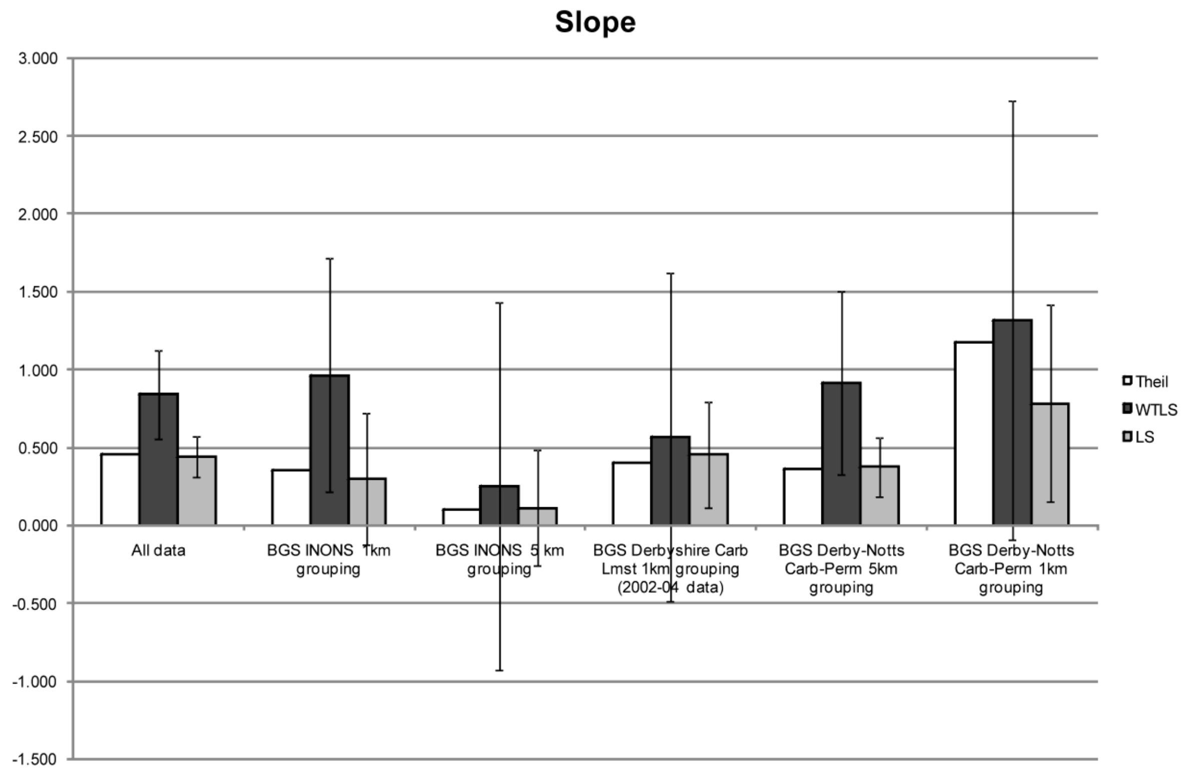

Figure 12 Comparison of the slopes (Log GM indoor radon/Log GM soil gas radon) and their 95% confidence limits for regression models derived from soil gas radon data.

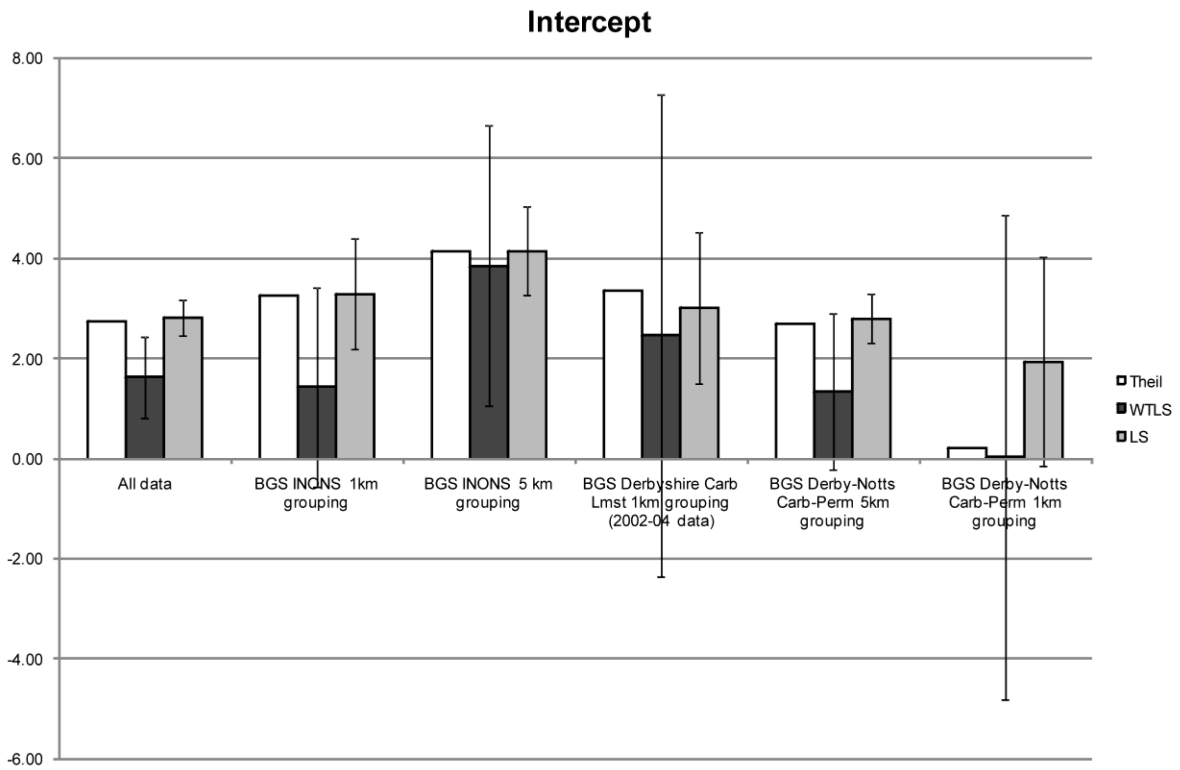

Figure 13 Comparison of the intercepts (Log GM indoor radon) and their 95% confidence limits for regression models derived from soil gas radon data.

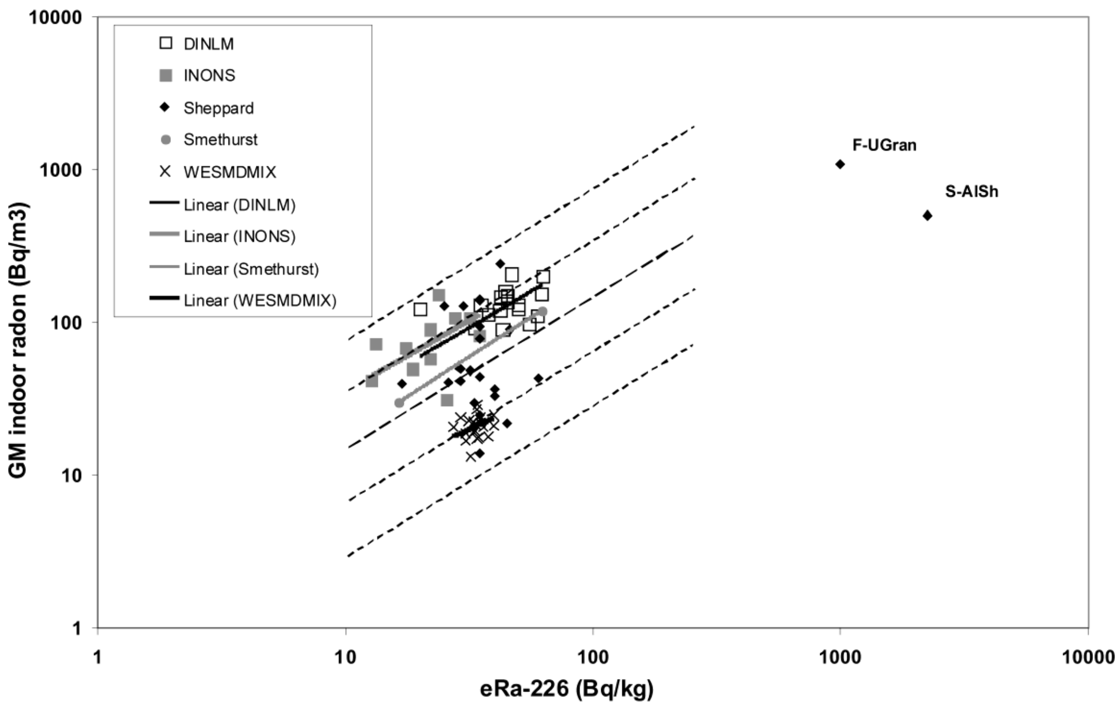

Figure 14 Relationship between average e^{226}Ra derived from U in <2mm soil and average indoor ^{222}Rn for the English Midlands (intercepts set to 5 Bq m^{-3} indoor radon) compared with published data.

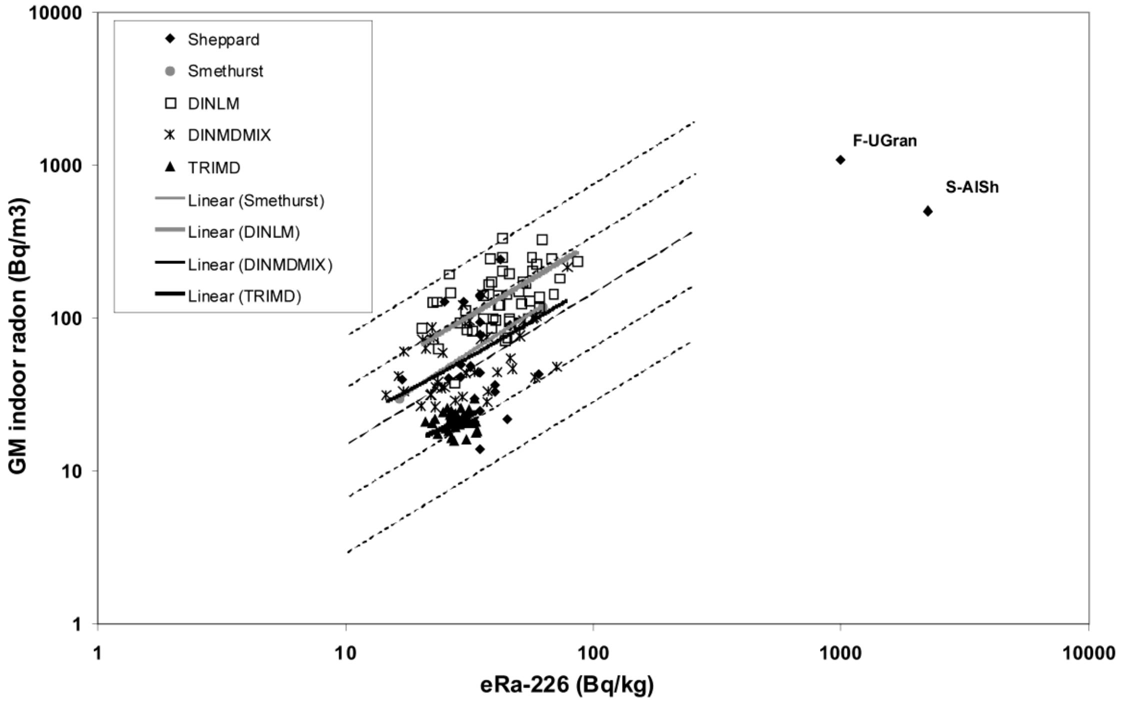

Figure 15 Relationship between average $e^{226}Ra$ in <2mm soil derived from HIRES data and average indoor ^{222}Rn for the English Midlands (intercepts fixed at 5 Bq m^{-3}) compared with published data.