Soil uranium, soil gas radon and indoor radon empirical relationships in the UK and other European countries.

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Abstract
Least squares (LS) regression analysis is used to develop empirical relationships between uranium in the ground, radon in soil and radon in dwellings to assist in the development of a geogenic radon potential map for Europe. The data sets used are (i) estimated uranium in the <2mm fraction of topsoils derived from airborne gamma spectrometry data, (ii) U measured in the <2mm fraction of topsoil geochemical samples, (iii) soil gas radon and (iv) indoor radon data. Linear relationships between radon in dwellings and uranium 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 uranium or soil gas radon concentration in the ground. UK regression models are compared with published data for other European countries.

1 Introduction
The production of a European geogenic radon potential map was formulated in 2008 (IGC33, Oslo) as a contribution to the European Atlas of Natural Radiations, a project promoted by the Joint Research Centre (JRC). A classification scheme is required which can be applied in all the countries of Europe. One of the problems with developing the classification scheme is that each country has different types of information out of which a map has to be generated. The ultimate aim is to provide an indication of the concentration of radon in homes because it is well established that the main risk to human health is related to exposure to radon indoors. A radon potential classification scheme, based on combining layers of available input variables including indoor radon, soil gas radon, U/Ra in soil, rock, stream sediments requires an understanding of the empirical relationships between these variables. These relationships are explored for data available in the UK and compared with those based on data published for other European countries.

2 Data and methods
2.1 Introduction.
Four types of data were used in this study: (i) estimated U in the <2mm fraction of topsoils derived from estimated uranium (eU) from airborne gamma spectrometry data, (ii) uranium in soil geochemical samples, (iii) 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. This study focussed on data for geological units with a relatively wide range of permeabilities as it is known from previous work that permeability has a major influence on the relationship between indoor radon, soil gas radon and U in the ground (Miles and Appleton, 2005; Scheib et al., 2006).
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). The observed 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, (ii) varying levels of soil moisture; (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 Bi214 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 linear regression equation (Estimated <2mm topsoil U concentration = 1.7216 x HiRES eU) is used in this study 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.

2.3 Soil uranium geochemical data

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 Johnson et al. (2005). Topsoil (A) soil samples collected from the 5–20 cm depth range, are sieved to pass a 2mm mesh and subsurface (S) soil samples from 35-50 cm depth are sieved to pass <150µm. 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).

2.5 Regression analysis

Regression analysis based on the average values for spatially and geologically grouped data require that the distribution in each subset is approximately normally distributed so that the value used for the regression analysis is a robust central estimate. It is well documented that indoor radon data are usually positively skewed and follow a generally lognormal distribution (Miles, 1998). Anderson-Darling (AD) normality tests for a representative selection of data subsets (i.e. data grouped by 1-km or 5-km grid square and geology) indicate 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. The least squares (LS) regression analysis 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. However, the LS method has limitations because (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.

3. Regression models

3.1 Models derived from 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 U 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 stacked up above each other, as illustrated in Figure 1. 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 tends to confirm this relationship (BGS unpublished data and Scheib et al., 2006).

Regression lines for high permeability units intersect 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 uranium data on the outcome of regression models (HPA, unpublished data) showed that adding uncertainties on to model data that is linear with a zero intercept leads to a regression line with a positive intercept.

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. 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, a conclusion that is consistent with the results of measurements of radon emanation from UK building materials. A value of 5 Bq m^{-3} is assumed here for the contribution from outdoor radon and building materials together, and is used as a forced intercept in for the regression models indicated by the thin lines in Figure 1.

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

Data set	Model formula	No. of data points	R^2	Significance (p)
HiRES DINLM 1-km grouping	y = 26.526x + 52.465	51	0.2095	<0.05
HiRES DINMDMIX 1-km grouping	y = 16.252x + 17.461	39	0.2884	<0.05
HiRES all data 1-km grouping	y = 37.361x – 35.247	296	0.3244	<0.05
HiRES TRIMD 1-km grouping	y = 0.5536x + 20.193	35	0.0024	>0.05
Data set	Model with 5 Bq/m^3 indoor radon intercept			
HiRES DINLM 1-km grouping	y = 38.666x + 5	51	0.161	<0.05
HiRES DINMDMIX 1-km grouping	y = 19.956x + 5	39	0.2703	<0.05
HiRES all data 1-km grouping	y = 23.1584x + 5	296	0.2729	<0.05
HiRES TRIMD 1-km grouping	y = 7.1124x + 5	35	-0.3407	>0.05

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

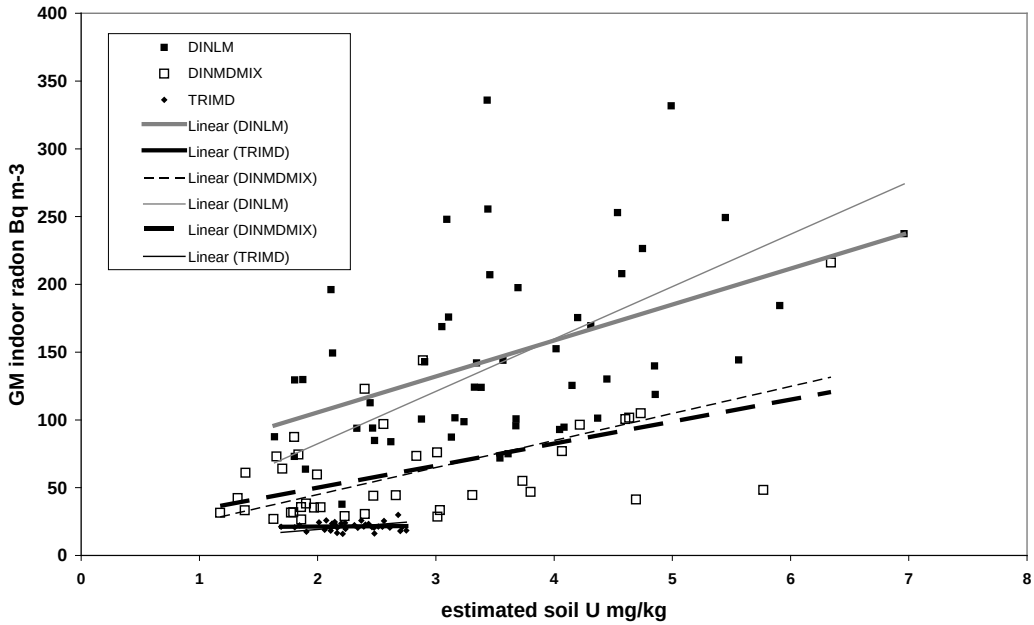

Figure 1. LS regression models between estimated soil U (derived from HiRES eU) and indoor radon with data grouped by 1km-geology (thin regression lines have intercepts set to 5 Bq m^{-3}).

The least squares regression model for the Oslo area (derived from Figure 6 in Smethurst et al., 2008) and the average model for N. Ireland (Appleton et al., 2008) are compared with the linear regression models for the English Midlands in Figure 2. The Oslofjord and N. Ireland models correspond fairly closely to that for the intermediate permeability group (DINMDMIX) although it should be noted that the Oslofjord model is based on average rather than GM indoor radon, which is likely to be significantly higher than the GM values used for the N. Ireland and English regression models. In the UK, arithmetic mean indoor radon is about 1.5 times higher than GM indoor radon.

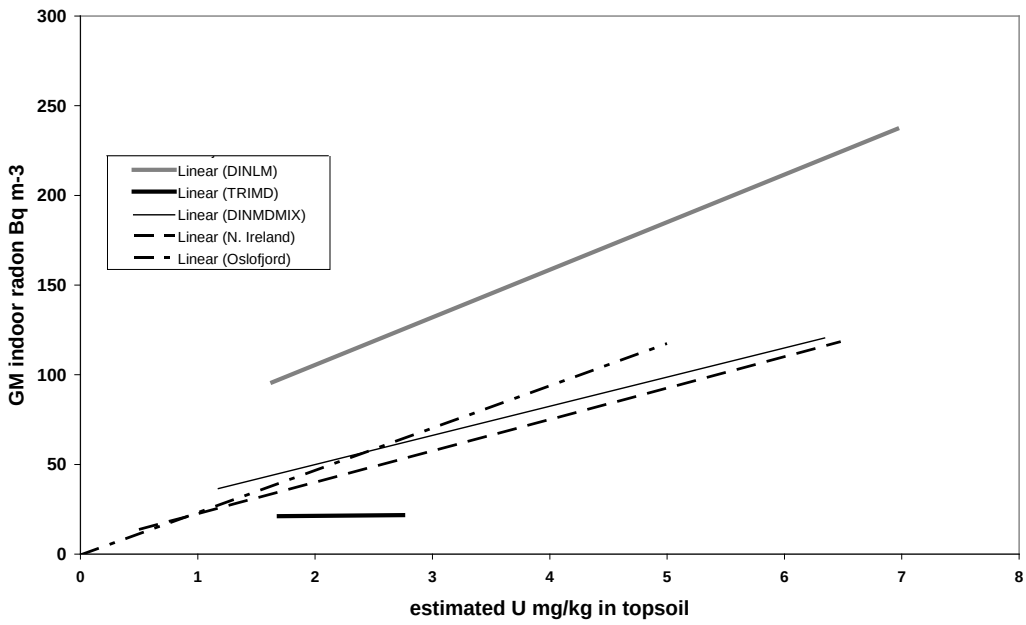

Figure 2. LS regression models between estimated soil U (derived from HiRES eU) and indoor radon with data grouped by 1km-geology compared with data for the Oslofjord region (Fig.6 in Smethurst et al., 2008) and N. Ireland (adapted from data in Appleton et al., 2008).

3.3 Models based on soil geochemical data

LS regression equations, R^2 and significance data for three geological units with adequate data and representing different ground permeabilities are presented in Table 2. Regression lines for geological units with different permeabilities are generally stacked up 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 are statistically significant ($p < 0.05$; Table 2). When the regression models are forced to intersect the y axis at 5 Bq m^{-3}, the regression lines for the permeable units (INONS, DINLM and INOLMST) are virtually coincident (Figure 3).

Although the regression models for most of the individual geological units are 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 is 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} are similar for HiRES (Figure 1) and topsoil geochemical (Figure 3) data.

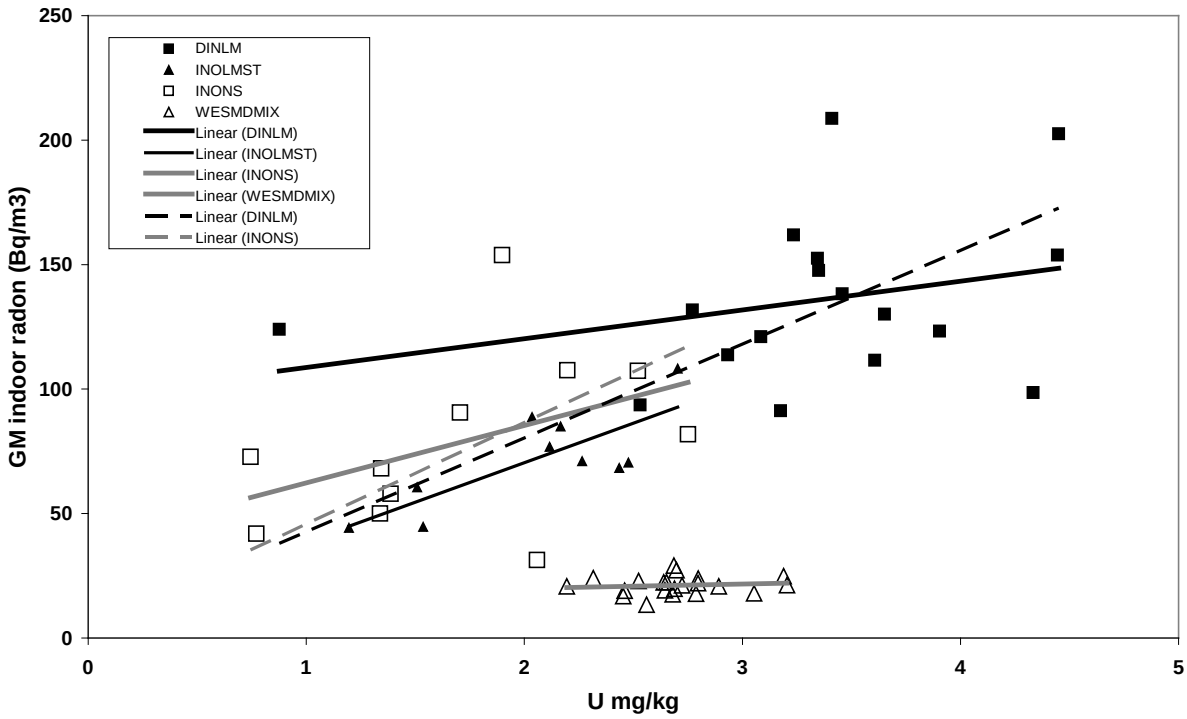

Figure 3. Regression models between U (mg/kg) in <2mm fraction of topsoils and indoor radon with data grouped by 5km-geology (dashed regression lines have intercepts set to 5 Bq m^{-3}).

Table 2. LS linear regression models derived from U in <2mm fraction of topsoils

Data set	Model formula	No. of data points	R^2	Significance (p)
DINLM 5-km grouping	y = 11.547x + 97.14	17	0.0835	>0.05
INONS 5-km grouping	y = 23.030x + 39.299	11	0.1842	>0.05
INOLMST	y = 31.735 + 6.9356	10	0.6153	<0.05
All data 5-km grouping	y = 24.456x – 4.1043	172	0.1318	<0.05
WESMDMIX 5-km grouping	y = 1.6749x + 16.639	21	0.0136	>0.05
Data set	Model formula with 5 Bq/m^3 indoor radon intercept			
DINLM 5-km grouping	y = 37.678x + 5	17	-0.3696	>0.05
INONS 5-km grouping	y = 40.7797x + 5	11	0.0599	>0.05
INOLMST	y = 32.637x + 5	10	0.6148	<0.05
All data 5-km grouping	y = 20.723x + 5	172	0.1284	<0.05
WESMDMIX 5-km grouping	y = 5.9551x + 5	21	-0.0759	>0.05

3.4 Soil gas radon – indoor radon regression models

LS regression equations, R^2 and significance data for five geological units with adequate data and representing different ground permeabilities are presented in Table 3. Plots of the regression models with and without the y axis intercept constrained to 5 Bq m^{-3} are illustrated in Figures 4 and 5. Statistically significant regression models for which the intercept is not fixed at 5 Bq m^{-3} are 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; (v) Carboniferous and Permian data for Derbyshire and Nottinghamshire: 5-km grouped data. Only the third model above (Lower Carboniferous limestones in Derbyshire, 1-km grouping of 2002-2004 data) is statistically significant ($p < 0.05$) when the intercept is 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 are likely to be steeper than regression lines for individual geological units. Unfortunately, sufficient data is not available to prove this. Regression models for other geological units grouped by 1-km and 5-km grid square are 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.

Table 3. LS linear regression models derived using soil gas radon data

Data set	Model formula	N	R^2	Significance (p)
Derby-Notts Carb-Perm 1km gp.	y = 1.9692x + 55.971	13	0.2469	<0.05
Derby-Notts Carb-Perm 5km gp.	y = 1.077x + 37.281	75	0.0932	<0.05
INONS 5-km grouping	y = 2.019x + 53.222	10	0.3458	<0.05
INONS 1-km grouping	y = 1.8982x + 41.841	17	0.2278	<0.05
Derby Carb. Lmst. 1-km grouping	y = 0.833x + 66.101	6	0.8831	<0.05
Data set	Model formula with 5 Bq/m^3 indoor radon intercept			
Derby-Notts Carb-Perm 1km gp.	y = 3.143x + 5	13	0.1262	>0.05
Derby-Notts Carb-Perm 5km gp.	y = 2.3125x + 5	75	-0.1075	>0.05
INONS 5-km grouping	y = 4.4942x + 5	10	-0.3982	>0.05
INONS 1-km grouping	y = 3.5237x + 5	17	0.0041	>0.05
Derby Carb. Lmst. 1-km grouping	y = 1.154x + 5	6	0.6755	<0.05

Gp. = grouping; Lmst. = Limestone

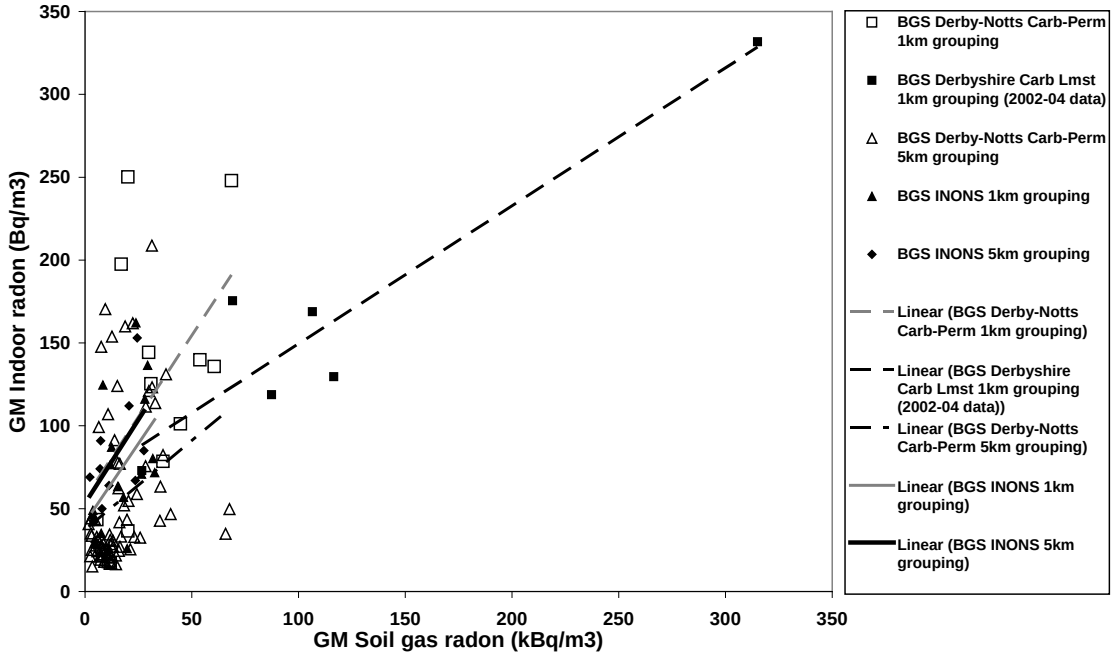

Figure 4. Relationship between GM soil gas radon (n>4) with GM indoor radon (n>19) with data grouped by 1-km or 5-km grid square and geology: data for Carboniferous and Permian of the English Midlands and the Northampton Sand Formation.

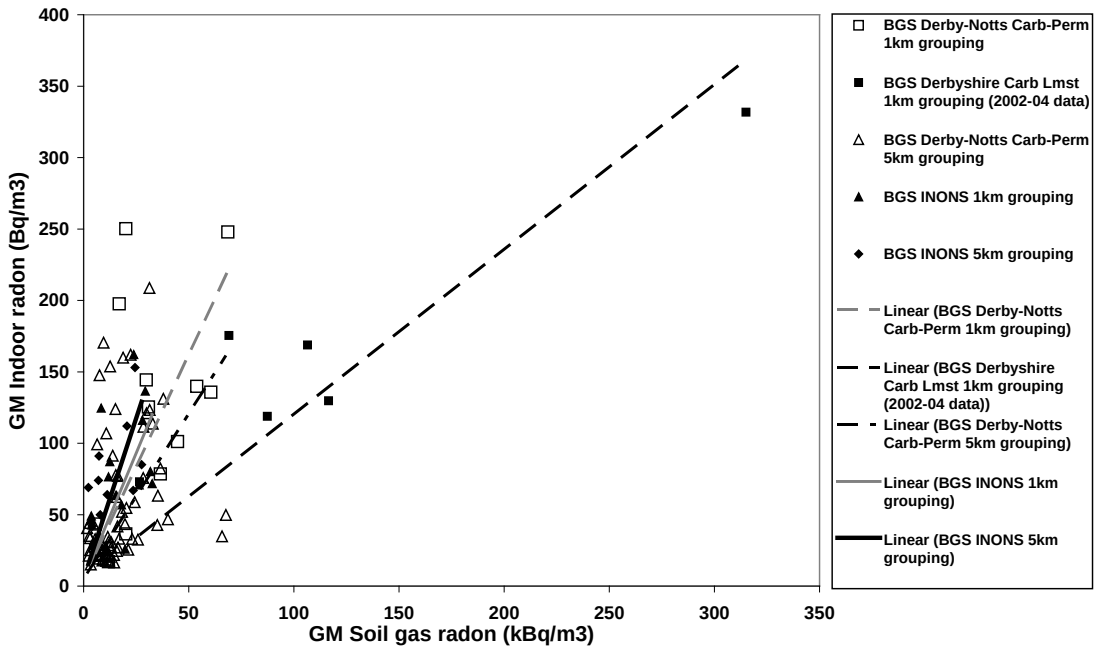

Figure 5. Relationship between GM soil gas radon (n>4) with GM indoor radon (n>19) with data grouped by 1-km or 5-km grid square and geology: data for Carboniferous and Permian of the English Midlands and the Northampton Sand Formation (intercepts set at 5 Bq m^{-3})

4. Comparison of regression models based on UK, Czech Republic and German data

Soil gas radon vs. indoor radon linear regression models for 1-km and 5-km grouped data from Derbyshire-Nottinghamshire Carboniferous-Permian data above are compared with data from Germany (Kemski et al., 2005, 2009; Barnet et al., 2006b) and the Czech Republic (Barnet, 2004; Barnet et al. 2003, 2005, 2008) in Figure 6. The German and Czech indoor radon data are generally

ground floor arithmetic mean concentrations so the UK data are converted from GM average house radon concentrations by multiplying GM indoor radon by 1.55 to convert from GM to AM and by 1.21 to convert from average house to ground floor radon concentrations (f = 1.8755). AM:GM ratios of 1.24-1.6, 1.25-1.49, 1.4-2.1 have been recorded in Spain, Croatia and Bratislava, respectively (Quindós et al., 2006; Radolić et al., 2006; Vicanova et al., 2006). In the UK, the corresponding factor for converting AM soil gas to GM soil gas is 1.38 and ratios of 1.2-1.34 have been reported for Spain (Quindós et al., 2006).

The linear regression models based on data from the UK are similar to those derived from indoor radon soil gas data in the Czech Republic 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 (Barnet et al., 2006b; Kemski et al., 2006, 2009), mainly because of different house characteristics. 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 (Kemski et al., 2009). The ratios between mean indoor and soil gas radon concentrations are higher in the Czech Republic than recorded in Germany and Barnet (2004) suggests that this may be explained by (a) differences in building quality (e.g. less efficient sealing of the basement against radon ingress into Czech houses) and (b) because the radon detectors are located in the rooms most likely to be affected by high radon in the Czech Republic whilst in Germany, as in England, detectors are located to determine the average radon concentration in the house. Kemski et al. (2009, Figure 8) recorded higher indoor/soil gas ratios for counties in the eastern sector of Germany (the old German Democratic Republic).

Some regions in the Czech Republic, such as Příbram, have a higher indoor radon/soil gas ratio than average (Figure 6). Barnet (2004) observed that lower indoor/soil gas ratios are associated with magmatic rocks, medium ratios with metamorphic and the highest ratios with sedimentary rocks. He suggested that differences in weathering styles resulted in differences in soil permeability, this being lowest over magmatic and highest over sedimentary strata. This in part controls the fraction of the soil gas radon that enters houses. This relationship is also observed in the data for England, where the most highly permeable bedrocks (Lower Carboniferous 'karst' limestones) are characterised by the highest indoor/soil gas ratios whilst impermeable mudstones and other argillaceous rocks are characterised by the lowest ratios. The linear regression models based on Italian data (Garavaglia et al., 2006) fall within the range of models for England, Germany and the Czech Republic, as does the model for Croatia (Radolić et al., 2006) (Figure 7).

As noted previously, most of the linear regression lines intersect the 'y' (indoor radon) axis above zero. In Germany the intersect is at 37 Bq m-3 and Kemski et al. (2009) suggested that this represents the non-geogenic contribution from building materials.

5. Discussion and conclusions

LS regression analysis demonstrates that relationships between uranium in the ground, radon in soil and radon in dwellings differ depending on the characteristics of the underlying geological units. The results provide a range of models for ground with different permeabilities. Uncertainties related to the measurement methods and GMs for grouped data used to formulate the regression models impact on the regression model slope and intercept uncertainties. Whereas the geometric mean 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.

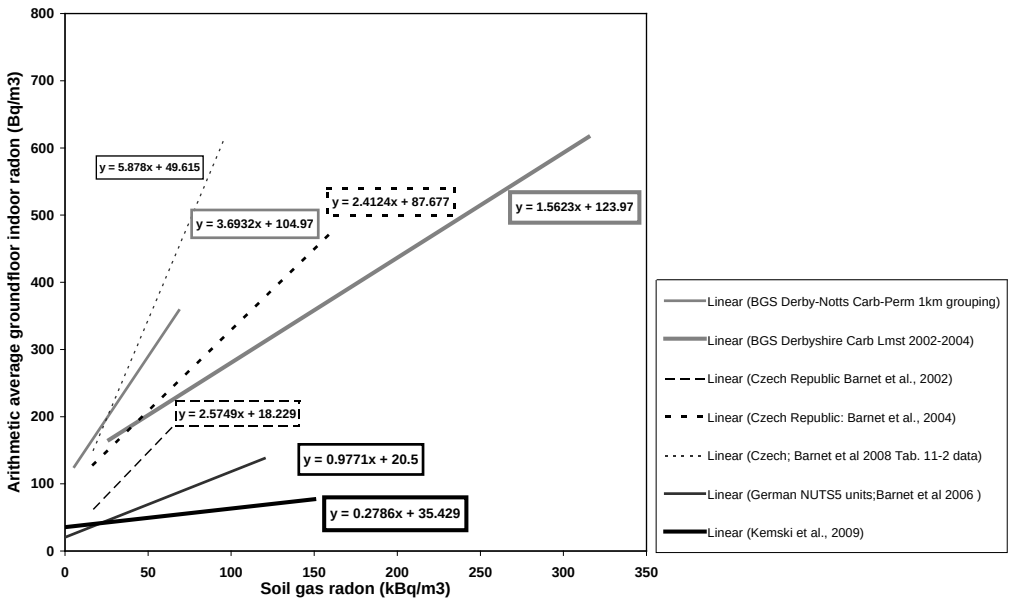

Figure 6. Comparison of soil gas radon vs. estimated arithmetic mean indoor radon linear regression models for England, Czech Republic and Germany.

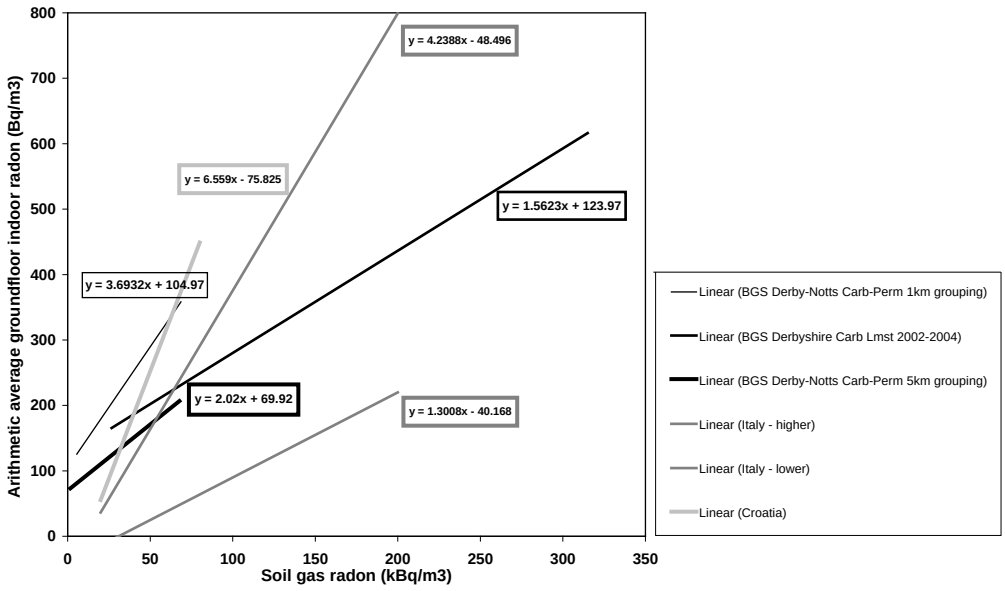

Figure 7. Comparison of soil gas radon vs. estimated arithmetic mean indoor radon linear regression models for England, Italy and Croatia.

The assumption must be that the relationship between the surrogate for the source of radon (i.e. airborne gamma or U in soil) and radon in homes is linear because any other relationship would imply that the factors affecting how radon gets into homes are correlated with the source term. There does no appear to be any such correlation, because if it was, GSD of indoor radon would vary systematically with GM, which it does not. The distribution of radon in homes is lognormal because of the intervening multiplicative factors determining how much radon gets into the home. Doubling the uranium in the ground should double concentrations in homes. If the measurements of the source term (uranium) have normally distributed errors, the appropriate variables to use for modelling would be arithmetic mean source value (i.e. U) and GM radon in homes. The relationship between soil gas and indoor radon is best defined using GMs on both axes because both variables are lognormally distributed.

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.

It is clear that additional research will be required to generate empirical relationships between the range of input factors proposed for the production of a Geogenic Radon Map of Europe.

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