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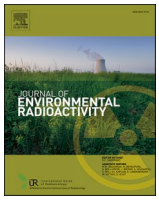

Gamma-radiation levels outdoors in Great Britain based on K, Th and U geochemical data

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ABSTRACT

Gamma-rays from naturally occurring radionuclides are a major component of background radiation. They are an important tool for geology and are also important for radiation protection. In this paper we use over a quarter of a million geochemical measurements of concentrations of potassium, thorium and uranium in soils and in stream sediments to estimate outdoor gamma-ray dose rates across Great Britain. The soil concentrations are generally at a depth of 5–20 cm with some at 35–50 cm. Soil measurements will give spatially relatively precise estimates, but as soil data are not available for much of Scotland, stream sediment data are used there. Kriging methods are used to estimate surface concentrations of K, Th and U and dose rates are imputed from these concentrations. Our results are compared with measurement surveys of both outdoor and indoor gamma-ray dose rates. Recently there has been interest in exploring the carcinogenic risks of low dose radiation by investigating associations between childhood cancer rates and doses from natural background gamma radiation. To achieve adequate statistical power, such studies must be so large that it is impractical to assess exposures by direct measurements in the homes of study subjects. Instead the exposures must be modelled. The results presented here will be an important input to such work.

1. Introduction

Gamma rays from naturally occurring radionuclides arise largely from potassium-40 and from the decay chains headed by thorium-232 and uranium-238. Gamma ray data can give important information on total gamma-ray dose rates (Beresford et al., 2007; Rawlins et al., 2012; Beamish, 2014; Kleinschmidt and Watson, 2016; Cinelli et al., 2019) sometimes focussing specifically on local geology (Beamish and White, 2011), radon potential (Appleton et al., 2008, 2011; Ferreira et al., 2018), radon flux (Manohar et al., 2013), doses to terrestrial wildlife (Beresford et al., 2008; Jones et al., 2009) and outdoor doses to people (Green et al., 1989). These gamma rays are also important for radiation protection. At a population level, natural radiation sources give the largest radiation doses, though for some individuals medical exposures may be higher.

Over the last few years, there has been interest in the possibility that epidemiological studies might be able to detect the effects of natural background radiation on childhood cancer rates. A number of such epidemiological studies have been carried out in recent years as reviewed by Mazzei-Abba et al. (2020). Such epidemiological studies must be very large if they are to be able to detect the very small effects expected (Little et al., 2010). This implies that it is, in practice, impossible to carry out direct measurements of gamma radiation levels for all study subjects. Instead modelling must be used to estimate radiation exposures.

In practice, measurements of outdoor gamma-ray dose rates will include a contribution from directly ionising cosmic rays. In this paper we are generally concerned with the gamma-ray doses from terrestrial radionuclides without the cosmic ray contribution; where they are included this will be made clear. Kendall et al. (2016) estimated the average outdoor dose rate from directly ionising cosmic rays to be 43 nGy/h and the average indoor dose rate to be 34 nGy/h. Both these values are taken as invariant across the populated area of Great Britain. The indoor dose-rate will, in fact vary significantly from one building to another depending on the shielding provided by the building materials. Cosmic ray dose-rates at sea level vary only slightly with latitude. The variation with altitude is potentially more significant. However, most of the GB population live at altitudes below 300 m at which height the increase in dose rate is only 3–4% (UNSCEAR, 2008).

For most people not living in timber houses, gamma dose rates are

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somewhat higher inside the home than outside (UNSCEAR, 2000), and people in urbanised countries spend more time indoors than out. In consequence, indoor gamma-ray dose rates are more likely to be directly linked to carcinogenic effects than outdoor dose rates. Nevertheless, as we shall show, outdoor dose rates (Green et al., 1989) correlate with indoor (Kendall et al., 2016). Moreover, outdoor dose rates may be easier to model than indoor dose rates. The former are determined entirely by geology, albeit that the very local environment is likely to be dominant. Attenuation of radiation from nuclides in soil is mainly controlled by soil moisture content, density and porosity. The majority of the dose is from the top 50 cm of the soil profile (Grasty and Minty, 1995; Beamish, 2014). In contrast, for indoor dose rates, the house construction materials play an important role, both because they provide shielding against gamma rays from outside and because radionuclides within the building materials provide another source of radiation (Kendall et al., 2018, 2021).

We noted above that gamma rays from naturally occurring radionuclides can give information about the local geology. Conversely, knowledge of the local geology, including geochemical and geophysical data, allows predictions of gamma-ray dose rates in the area. In this paper we shall adopt this approach to offer predictions of outdoor gamma-ray dose rates from K, Th and U geochemical data. A gamma dose rate map for Great Britain (GB: England, Scotland and Wales) forms part of the European Terrestrial Gamma Dose Rate Map (Cinelli et al., 2019) but this is very generalised being based on soil data from only about 100 sites extracted from the FOREGS (Steenfelt and Tarvainen, 2005) and GEMAS (Reimann et al., 2014a, b) data bases. Airborne gamma-ray spectrometric data has been used to estimate gamma dose rates (Beamish, 2014) but these data are available for relatively small sectors of GB so is not useful for national epidemiological studies.

A detailed evaluation of the relationship between gamma dose rate data derived from airborne geophysics gamma-ray spectrometry and soil chemistry is beyond the scope of the present study. Appleton et al. (2008) observed that airborne geophysics gamma spectrometry estimates of K_2O were about 20% less than soil K_2O concentrations determined by x-ray fluorescence spectrometry (XRF), with airborne estimated Th being about 30% less than soil Th and airborne estimated U only about a third of soil U. This relationship and the broadly linear correlation between soil and airborne geophysics K, Th and U estimates was confirmed by Beamish (2014), based on the Tellus Northern Ireland data. Similar relationships also characterise other UK airborne data sets (Emery et al., 2005; Beamish, 2014).

In this study, dose predictions based on geochemical data will be compared with indoor and outdoor measurement data. It is anticipated that these data will be included in future modelling of indoor gamma-ray dose rates in GB, thereby following, and extending, the French use of geogenic uranium potential (Ielsch et al., 2017; Warnery et al., 2015).

A previous assessment of natural background radiation doses in GB (Jones et al., 2009) was based on stream sediment and soil geochemical data derived mainly from the Geochemical Baseline Survey of the Environment (G-BASE) conducted by the British Geological Survey (BGS). At that time, the G-BASE survey was incomplete so only partial coverage of GB was available. The BGS stream sediment survey has now been completed and the results available in the Geochemical Atlas (Everett et al., 2019; https://www.bgs.ac.uk/geology-projects/applied-geochemistry/g-base-regional-geochemistry/stream-sediment-geochemical-atlas/). Furthermore, BGS soil geochemical data produced for incorporation in a BGS Soil Geochemical Atlas are now available for the whole of England and Wales and a small sector of Scotland. These geochemical data are used in the present study to derive outdoor terrestrial gamma radiation dose rate estimates for the whole of GB.

2. Materials and methods

2.1. Measurements of indoor and outdoor gamma-ray dose rates

Measurements of indoor gamma-ray dose rates were made in the National Radiological Protection Board's National Survey of indoor natural radiation levels (Wrixon et al., 1988). Measurements were completed in 2283 dwellings. These have since been augmented with indoor gamma-ray dose-rate data for another 7916 dwellings from the United Kingdom Childhood Cancer Study (UKCCS) (UK Childhood Cancer Study Investigators, 2002). For both sets of measurements the assessment was almost always based on long term measurements from two thermoluminescent dosimeters placed in different rooms of the dwelling in question. The mean indoor dose-rate from terrestrial gamma-rays was found to be 62 nGy/h and the (additional) indoor contribution from directly ionising cosmic rays assessed as a constant 34 nGy/h.

Measurements of outdoor gamma-ray dose rates were made as part of a companion survey by the National Radiological Protection Board (Green et al., 1989). Over 3100 10-min measurements were made, following a standard protocol, using a vertical energy compensated Geiger-Müller tube 1m above the ground. The mean area-weighted outdoor dose from terrestrial gamma-rays was found to be 34 nGy/h; this excludes the dose from directly ionising cosmic rays, assessed as a constant 43 nGy/h across Great Britain. The results of this outdoor gamma ray survey were published as a double smoothed and infilled map of the 10 × 10km squares of the British National Grid. The spatial precision and granularity of this data was by its nature very generalised (see Figs. 3 and 4 in Green et al., 1989). More details of both indoor and outdoor measurements and of small extensions to the outdoor map are given by Kendall et al. (2016). In this study, as in Kendall et al. (2016), outdoor dose rates comprise the terrestrial gamma-ray dose rates published by Green et al. (1989) plus 43 nGy/h for outdoor cosmic radiation component.

In addition to these national surveys, RIMNET (the Radioactive Incident Monitoring NETwork) maintains about one hundred fixed monitoring stations across the United Kingdom in order to provide baseline data on outdoor gamma-ray dose rates against which releases from any future nuclear accident can be assessed. Data from these RIMNET stations have been published over many years and they provide an additional source of information on outdoor gamma-ray dose rates. These data are described more fully in Section 4.2.

2.2. Geochemical data

There are two main sources of geochemical data available in GB through the BGS:

1. Surface (5–20 cm depth, A) soil data for about 40 elements including K_2O, Th and U are available for all of England and Wales (Fordyce et al., 2005; Flight and Scheib, 2011) at variable sampling densities, and the Glasgow-Clyde valley area of Scotland (Fordyce et al., 2012, 2017). Data for deeper (S) soils (35–50 cm depth) are also available for some areas and were also used in this study where appropriate.
2. Stream sediment geochemical data (Johnson et al., 2017; Everett et al., 2019) are available for the whole of GB for K_2O and U and for most of England and Wales for Th.

The BGS soil and stream sediment geochemical data can be used to provide coverage for a national assessment of outdoor gamma radiation dose using K, Th and U geochemical data in the modelling process on the basis that ^{40}K, ^{238}U and ^{232}Th are the major natural contributors (roughly on equal terms) to terrestrial outdoor gamma dose rates (UNSCEAR, 2000).

Whereas the stream sediment geochemical data will provide a broad indication of the spatial variation of K, Th and U, the soil data should, in

theory, be spatially more accurate, especially in urban areas where higher density soil sampling was carried out. The less spatially connected stream sediment data for K and U has to be used to provide gamma radiation dose rate estimates for most of Scotland where no soil data are available.

The evaluation of background outdoor dose rates from naturally occurring radionuclides in GB reported here was based on this large number of surface soil, deeper soil and stream sediment samples, totalling 260,795 analytical determinations (Table 1).

The systematic collection of soils by BGS from rural areas varied slightly from area to area as the survey progressed but mainly comprised the collection and analysis of shallow sub-surface (5–20 cm depth) soil samples at a density of one sample every alternate 1 km square of the British National Grid. This became the standard G-BASE regional soil sampling density until the final years of the project when an alternative, statistically based sampling strategy was adopted to ensure representative coverage of the last remaining unsampled areas of southern England. Of GB urban centres, 22 have been sampled at a density of 4 samples per km^2 (Fordyce et al., 2005; Flight and Scheib, 2011; Ferreira et al., 2017). In addition, sub-samples held at Rothamsted Research from the National Soil Inventory (NSI) of England and Wales sample archive, National Soil Resources Institute, Cranfield University, collected at a density of 1 per 25 km^2 (McGrath and Loveland, 1992), were analysed by XRF at the BGS and included in the compilation of surface soil data used in this study. The variation in soil sample density within England and Wales and the collection and analysis of deeper (30–50 cm depth) samples reflects changes in the focus and objectives of the GBASE soil sampling programme and availability of funding.

The locations of available surface (A; n = 39,362) and deeper (S; n = 13,379) soil samples used to map K_2O concentrations in surface soils in England and Wales are shown in Fig. 1 together with the extents of GBASE geochemical atlas areas. Fewer deeper (S) soil samples are available for Th (n = 9256 in the Humber-Trent (HUMB) and Wales (WALE) geochemical atlas areas) and U (13,383 samples in the WALE, Tyne-Tees (TYNE) and Liverpool Bay (LIVB) atlas areas).

2.3. Estimating outdoor terrestrial dose rates for England and Wales

The predicted concentrations of K_2O, Th and U in surface soils for the Soil Geochemical Atlas of the UK (In preparation; Bob Lister, BGS, pers. comm.) were derived by Ordinary Kriging in ArcGIS Geostatistical Analyst of log-transformed data to a 500 m grid and are based solely on surface soil data. As the data are not normally distributed, a log-transformation was applied and the output grid data were back-transformed to the original data units then exported as a raster grid. Ordinary kriging makes use of a variogram – a model of the spatial autocorrelation within the data – in order to optimise the weights given to surrounding sample points when interpolating concentrations to new locations (in this case the cell centres of the output grid). An exponential semivariogram model was generated for each variable. All default parameters were used apart from the maximum number of nearest neighbours, which was set to 8 in order to retain short-scale geochemical variation evident in the raw data. The search radius was set at 10,000 m and the minimum number of data points set to 5.

Table 1
Numbers of soil and stream sediment samples with K_2O, Th and U analytical data in England, Wales and Scotland used in this study.

Sample type	Element/ oxide	Number in England and Wales	Number in Scotland
Surface (A) soil	K_2O, Th, U	39362	3934
Deeper (S) soil	K_2O	13379	
	Th	9256	
	U	6507	
Stream sediment	K_2O		48901
	U		49514

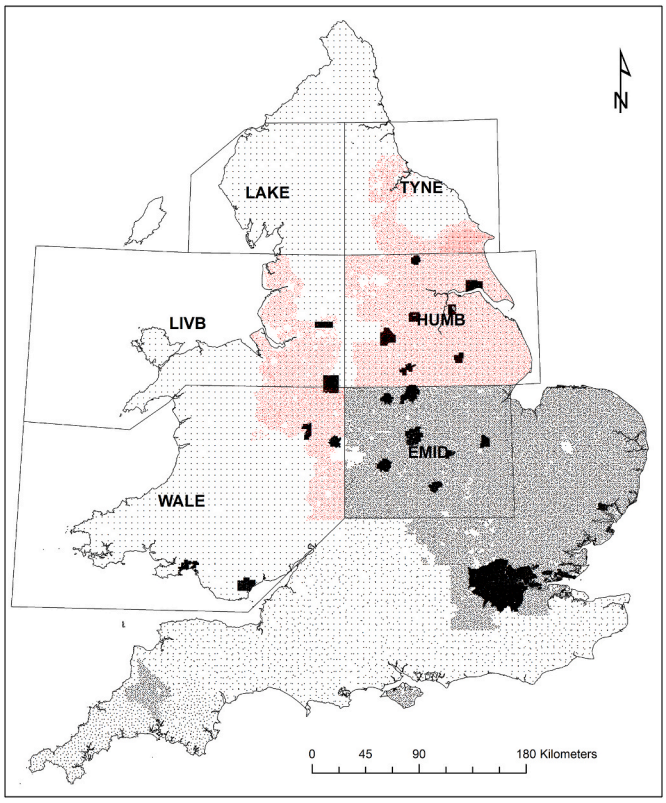

Fig. 1. Location of BGS surface (black dot) and deeper (red dot) soil samples used to map surface soil K_2O for England and Wales and extents of GBASE Geochemical Atlas areas. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Regression analysis of surface and deeper soil data (Table 2) at locations where both sample types are available was used to convert deeper soil analytical data to estimated surface soil concentrations prior to Ordinary Kriging in ArcGIS of the combined datasets. This procedure was used in previous studies of natural background radiation dose estimation in England and Wales (Jones et al., 2009). A range of sample groupings were evaluated and the regression models selected for this study, mainly on the basis of having the highest R-sq values, were used to convert the deeper (S) soil data to estimated surface (A) soil K_2O, Th and U concentrations (Table 2, Fig. 2 and SM-1 to SM-2). The various regression models based on different data subsets produce similar estimated surface K_2O, Th and U concentrations (Table 2).

The combined surface and deeper soil data for the sector of central and northern England and the Welsh Borders (including all or parts of the East Midlands (EMID), Wales (WALE), Liverpool Bay (LIVB) and Tyne Tees (TYNE) geochemical atlas areas) were re-interpolated to a 500 m grid by Ordinary Kriging in ArcGIS Geostatistical Analyst/Geostatistical Wizard based on the nearest 8 neighbours (minimum 5) with a search radius of 10,000 m. As would be expected, the combination of deeper and surface soil data resulted in more detailed maps of predicted surface soil K_2O, Th and U in those areas previously mapped only using data for the lower density (1 per 25 km^2) Rothamsted surface soil samples (McGrath and Loveland, 1992).

The predicted surface soil K_2O, Th and U concentration maps were converted into terrestrial gamma-ray dose rate (TDGR) maps (Figures SM-3 to 5) using the IAEA (2010) conversion constants 13.078 nGy/h per 1% K, 5.675 nGy/h per 1 mg/kg U and 2.494 nGy/h per 1 mg/kg Th. Slightly different conversion constants were used for the European Atlas of Natural Radiation (Cinelli et al., 2019) in which Terrestrial Gamma Dose Rate (nGy/h) = 13.052*% K + 5.682*mg/kg U

Table 2
Regression models for soil sample locations where both surface (A) and deeper (S) soil data are available for England and Wales. The regression models selected to convert the deeper soil data for HUMB, TYNE, LIVB and WALE to estimated surface soil K_2O (wt. %), Th (mg/kg) and U (mg/kg) concentrations are indicated in **BOLD** type.

					Estimated concentration in A soil for specified concentration in S soil			
	Number of sample sites	Sample group for regression	Regression Equation	R-sq %	S	Est. A	S	Est. A
K_2O	**10843**	**All available**	**A = 0.2414 + 0.7072 * S**	**74**	**1**	**0.95**	**4**	**3.07**
K_2O	7231	EMID area	A = 0.1345 + 0.7214 * S	73	1	0.86	4	3.02
K_2O	1607	Glasgow (GLAS) area	A = 0.3203 + 0.7153 * S	71	1	1.04	4	3.18
Th	10843	All available	A = 2.170 + 0.6526*S	59	10	8.70	40	28.27
Th	**7231**	**EMID area**	**A = 2.202 + 0.6590*S**	**63**	**10**	**8.79**	**40**	**28.56**
U	10337	All available	A = 1.099 + 0.4184*S	33	2	1.94	10	5.28
U	10314	Subset with U (S) < 10 mg/kg	A = 0–7296 + 0.5735*S	39	2	1.88	10	6.46
U	10243	Subset with U (S) < 6 mg/kg	A = 0.8051 + 0.5390*S	32	2	1.88	10	6.20
U	8124	All GLAS + HUMB	A = 1.138 + 0.4O80*S	34	2	1.95	10	5.22
U	**8102**	**Subset with U (S) < 10 mg/kg**	**A = 0.6993 + 0.5908*S**	**41**	**2**	**1.88**	**10**	**6.61**
U	8043	Subset with U (S) < 6 mg/kg	A = 0.8013 + 0.5445*S	33	2	1.89	10	6.25

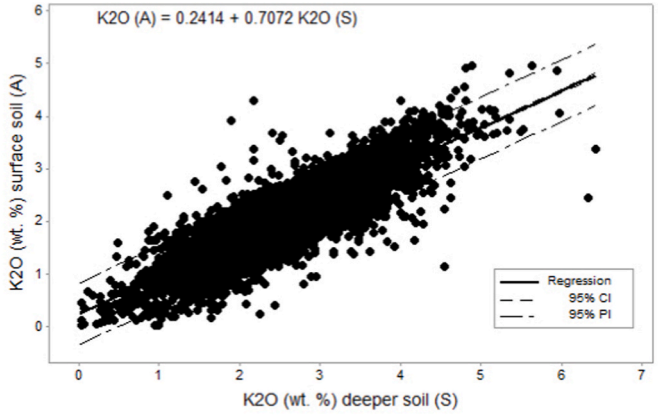

Fig. 2. Regression model used to convert K_2O in deeper soil (S) to estimated K_2O in surface soil (A) (R-sq = 73.7%) for England and Wales.

+ 2.69*mg/kg Th. Both of these conversion formulae assume that the decay series are in equilibrium.

2.4. Estimating outdoor terrestrial dose rates for Scotland

Mapping terrestrial gamma-ray dose rates in Scotland was based on (1) K, Th and U data for 3943 surface soil samples from the Clyde Basin, being a mixture of high-density urban sampling for Glasgow and relatively low-density rural soil sampling from the River Clyde drainage basin (Fordyce et al., 2017) and (2) K and U data for approximately 49, 500 stream sediment samples (Fig. 3). No stream sediment data are available for Th.

K_2O, Th and U surface soil maps for the Clyde Basin were produced using Ordinary Kriging in ArcGIS. The geochemical maps in the BGS Stream Sediment Atlas (Everett et al., 2019) are based on analyses of the <150 μm size fraction of sediment samples collected from first- and second-order (i.e., small) streams. Ordinary kriging was used to interpolate to a 500 m grid the mean concentrations based on the untransformed values for the nearest 9 samples. Variogram model parameters are documented in Everett et., (2017, Table 9 and Fig. 5). The K_2O stream sediment atlas map was used for this study because K_2O data are normally distributed. A new U map was produced using log-transformed data because the U distribution is strongly skewed (skewness +89 reducing to −0.10 when log-transformed).

In order to predict K_2O and U surface soil concentrations for areas where there are no soil data, the relationship between predicted soil and sediment K_2O and U concentrations was investigated for 8556 indoor dose-rate measurement locations (Kendall et al., 2016) where both predicted soil and sediment data are available (i.e., for England and

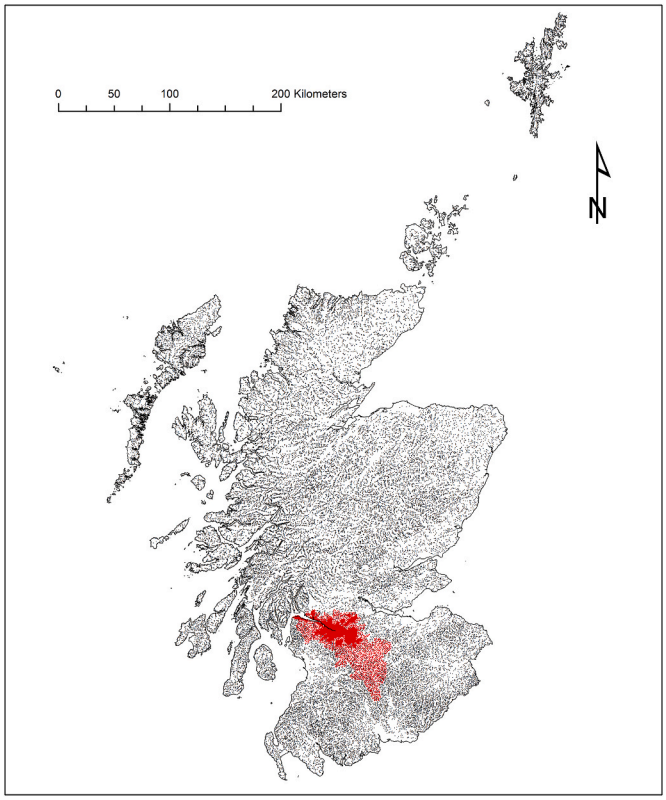

Fig. 3. Location of BGS stream sediment (black dot) and surface soil (red dot) samples in Scotland. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Wales, and the Clyde Basin in Scotland). Regression models for normally distributed K_2O (Fig. 4) and log-normally distributed U (Fig. SM-6) show that (1) estimated K_2O soil values generated from sediment data are on average 25% lower than the sediment concentrations, and (2) estimated U in soil derived from sediment U data is approximately the same at the 2 mg/kg U in sediment level, but less than half in soil at the 10 mg/kg U sediment level. The higher concentrations in stream sediment compared with soil occur principally in areas underlain by granitic rocks where U in sediment occurs predominantly in relatively dense, residual U-bearing minerals which tend to be concentrated in stream sediment compared with soil. There will also be instances where U in soil is higher than in sediment if the organic content of the soil is high. This is because available U is usually strongly adsorbed onto organic material. In contrast, K is usually found in micas, clay minerals and potassium

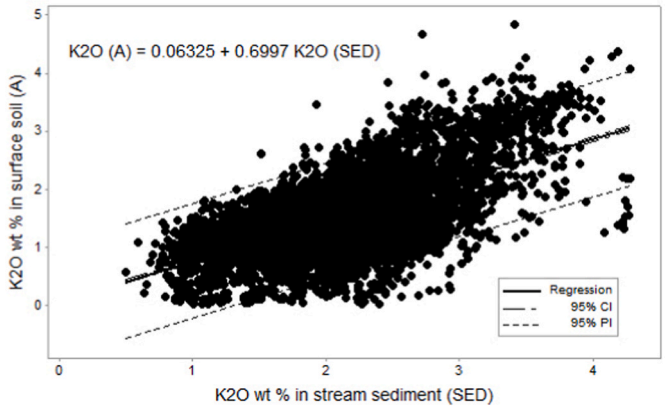

Fig. 4. K_2O (wt. %) stream sediment (SED) – surface soil (A) regression model (R-sq 37.5%) for Scotland.

feldspars which are less likely to be influenced by secondary adsorption or heavy mineral concentration. This could explain the greater dispersion/scatter of data points (R-sq 14.5%) in the U regression model (Figure SM-6) compared with the K_2O regression model (R-sq 37.5%; Fig. 5).

K_2O and U concentrations predicted from stream sediment data were converted to surface soil equivalent values so these 'equivalent surface soil' data for most of Scotland can be combined with the surface soil data for the Clyde Basin. Estimated surface soil K_2O and U concentration maps for Scotland were converted into TGDR maps (Figures SM-7 to SM-8) using the IAEA (2010) conversion coefficients.

The relationship between Th dose with K dose, U dose and combined K + U dose for the 9337 indoor dose rate locations where Th data are available was evaluated to see if it was possible to estimate Th dose for Scotland for areas where no soil or stream sediment Th data are available (Figs: 5 and SM-9). It was judged that the best results were obtained using the K + U dose relationship: Th dose = 0.6082 K + U dose.

3. Results

Total estimated terrestrial dose rates derived from K, Th and U soil data for England and Wales are shown in Fig. 6 and total estimated terrestrial dose rates for Scotland, based on soil and stream sediment data, in Fig. 7. These were produced in ArcGIS Statistical Analyst by summing the dose values of the K, Th and U dose rate raster files. Corresponding single element maps for K, Th and U are in Supplementary Material SM-3 to SM-5 for England and Wales and in SM-7 to SM-9 for Scotland.

Table 3 gives a summary of the components of dose rates for Great

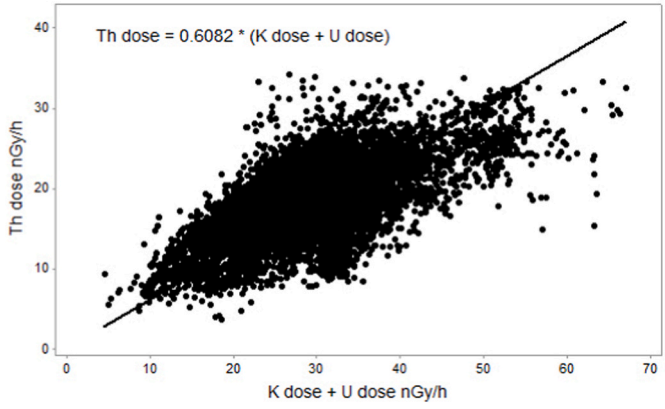

Fig. 5. Th dose: K + U dose relationship (Th dose = 0.6082 K + U dose; intercept set at zero) for Scotland.

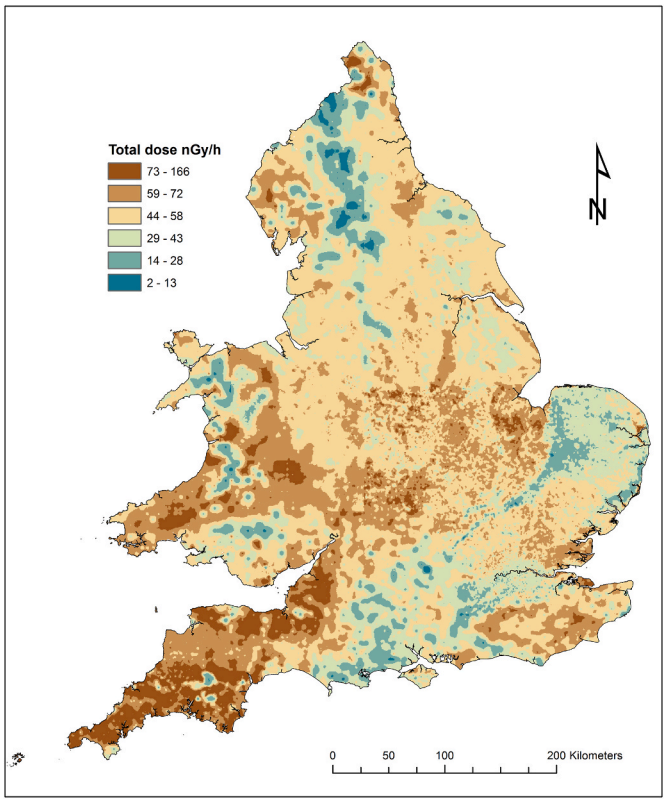

Fig. 6. Estimated Terrestrial Total (K + Th + U) dose rate (nGy/h) for England and Wales derived from soil K, Th and U geochemical data (classes based on standard deviations from the mean 50 nGy/h of 602,925 500 m grid values).

Britain. Corresponding data for England and Wales and for Scotland separately are in Supplementary Tables SM-1 and SM-2. On average for GB the contributions to the total dose from K, Th and U are 35, 38 and 27% respectively (Table 3) with similar amounts in England and Wales (Table SM-1) and Scotland (Table SM-2). Mean and median values are similar (Table 3). These results are consistent with the UNSCEAR rule of thumb that K, Th and U make roughly equal contributions to the outdoor gamma-ray dose rate (UNSCEAR, 2008).

4. Discussion

4.1. Relationship to bedrock geology

In England and Wales, the highest total dose rates are associated with the Carboniferous-Permian granitic igneous intrusions (median 78 nGy/h) and the mudstones, siltstones and sandstones of the Middle and Upper Devonian, Teign Valley Group and Holsworthy Group (68–78 nGy/h) in SW England (Supplementary Table SM-3). Above average dose rates also characterise the Ordovician and Silurian mudstones, siltstones and sandstones of Wales and the English Lake District (median 60–68 nGy/h). Extensive areas with average dose rates are underlain by (a) Jurassic Lias Group mudstones, the Kellaways and Oxford Clay formations; sandstones, limestones and argillaceous rocks of the Great Oolite Group (median 54–58 nGy/h); (b) mudstones, siltstones and sandstones of the Cretaceous Wealden Group (median 59 nGy/h), and (c) Triassic mudstones, siltstones and sandstones (median 56 nGy/h). The lowest total doses are associated with the Cretaceous Chalk and Lower Greensand (median 36–41 nGy/h), Neogene gravels, sands and clays (median 40 nGy/h) and the Carboniferous Yoredale Group limestones with subsidiary sandstones, siltstones and mudstones (median 28–38 nGy/h).

In Scotland the highest total dose rates are associated with Silurian-Devonian granitic and syenitic igneous intrusions (median 69 nGy/h),

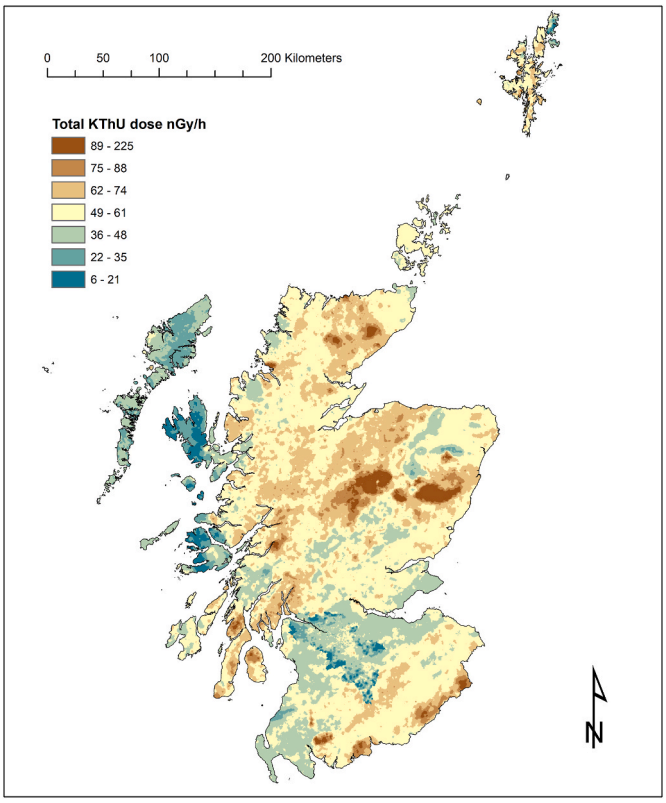

Fig. 7. Estimated Terrestrial Total (K + Th + U) dose rate (nGy/h) for Scotland derived from surface soil and stream sediment K, Th and U geochemical data (classes based on standard deviations from the mean 56 nGy/h of 294,551 500 m grid values).

Neoproterozoic Grampian Group migmatitic rocks, psammites and pelites (median 65–66 nGy/h), Loch Eil Group psammites (median 62 nGy/h), Torridon Group sandstones and mudstones (median 59 nGy/h), and South Highland Group psammites and pelites (median 59 nGy/h). Extensive areas with average total dose levels are associated with the Silurian Harwick and Gala Group greywackes (median 55–56 nGy/h), Devonian and Old Red Sandstone conglomerates, siltstones and mudstones (median 54–56 nGy/h), Carboniferous Inverclyde Group sandstone, siltstone, mudstone sequence (median 54 nGy/h), Permian Strathmore Group sandstone, conglomerate, siltstone, mudstone (median 52 nGy/h), and Silurian-Devonian mafic lavas and tuffs (median 52 nGy/h). The lowest total dose rates are associated with sedimentary rocks of the Carboniferous Clackmannan Group and Scottish Coal Measures Group (median 46 nGy/h), Carboniferous Dinantian mafic lavas and tuffs (median 43 nGy/h), with a substantially lower median (22 nGy/h) characterising the Palaeogene mafic lavas and tuffs of the Tertiary igneous province (Supplementary Table SM-4).

4.2. Relationship between terrestrial, indoor and outdoor dose rates

In this section we compare the terrestrial gamma-ray dose rates estimated above with the measured outdoor dose rates of Green et al. (1989) and with the indoor measurements of Kendall et al. (2016). For the comparison with the outdoor measurements we add 43 nGy/h for the directly ionising component of cosmic radiation to the Total (K + Th + U) dose rates derived from concentrations of K, Th and U in surface soil. Comparing data for the 10 km grid squares containing the 10,199 indoor measurement locations (Kendall et al., 2016) there appears to be a relatively good correlation between the two outdoor dose estimates (Fig. 8), especially considering that the outdoor dose rate estimates in Green et al. (1989) are smoothed 10 km grid values. The relationship is broadly the same for England and Wales (based solely on soil data) and Scotland (based largely on stream sediment data) (Supplementary Figures SM 10 and 11). It would be very interesting to compare the present predicted outdoor terrestrial gamma-ray dose rates derived from geochemical data with the approximately 3100 individual outdoor measurements (Green et al., 1989), but the latter are unfortunately not available. However, the mean and median values from the Green et al. (1989) outdoor data are 16 nGy/h lower than the BGS TGDR estimates based on K, Th and U geochemical data (Table 4). Possible reasons for this are discussed below.

Normal ambient gamma radiation dose rates (AGDR) at the 82 RIMNET fixed monitoring sites (UK Department for Business, Energy and Industrial Strategy, 2022) were compared with estimated terrestrial gamma-ray dose rates (TGDR) at these sites derived from K, Th and U geochemical data to which was added the dose from directly ionising cosmic rays, assessed as a constant 43 nGy/h across Great Britain (Green et al., 1989). AGDR at the RIMNET sites is on average about 12% higher than the TGDR estimates. The AGDR-TGDR Pearson correlation

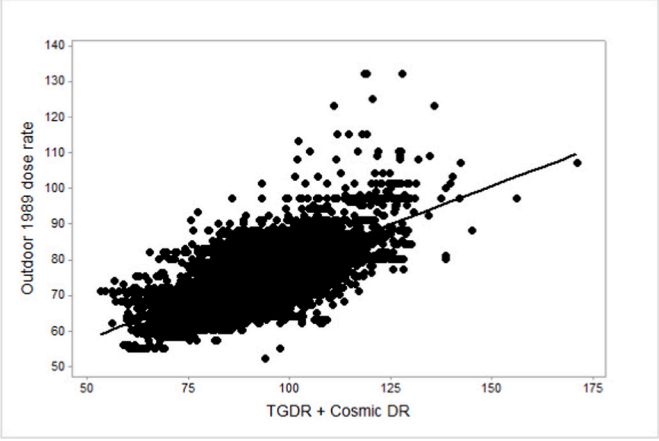

Fig. 8. Relationship between Outdoor Gamma dose rates (Green et al., 1989) and BGS Terrestrial gamma-ray dose rates (TGDR) derived from estimated K, Th, and U soil geochemical data plus cosmic radiation (43 nGy/h) at 10,199 childhood cancer study sites (Kendall et al., 2016).

Table 3
Summary Statistics for K, Th, U, K + U and Total (K + Th + U) dose rates (nGy/h) derived from BGS geochemical data, compared with indoor (Kendall et al., 2016) and outdoor (Green et al., 1989) dose rates at 10199 childhood cancer case locations in Great Britain (SE = standard error of mean; SD = standard deviation).

Variable	Mean	SE	SD	Minimum	Median	Maximum	Skewness
Total (KThU) dose	48.8	0.11	11.33	10.3	48.9	128.0	0.36
K dose	17.1	0.06	6.05	0.2	16.6	50.6	0.80
Th dose	18.8	0.05	4.62	3.6	18.9	66.3	0.30
U dose	13.0	0.04	3.56	0.7	13.2	43.5	0.11
KThU + cosmic	91.8	0.11	11.33	53.3	91.9	171.0	0.36
Outdoor 1989	75.6	0.08	7.66	52.0	76.0	132.0	0.51
Indoor	95.6	0.22	22.65	25.3	95.3	277.6	0.30

Table 4
Summary statistics for indoor dose rate, outdoor gamma dose rate (Green et al., 1989) and outdoor dose rate (nGy/h) derived from BGS geochemical data (plus 43 nGy/h cosmic dose) at 10,199 locations with indoor dose rate data (SE = standard error of mean; SD – standard deviation).

	Mean	SE	SD	Min.	Median	Max.	Skewness
Indoor dose rate	96	0.2	23	25	95	278	0.30
Outdoor dose rate (Green et al., 1989)	76	0.1	8	52	76	132	0.51
Outdoor dose rate from BGS geochemical data + cosmic dose (43 nGy/h)	92	0.1	11	53	92	171	0.36

coefficient is 0.595 (n = 82; $p < 0.001$). The AGDR-TGDR Pearson correlation coefficient for England and Wales (55 sites) where all TGDR estimates are derived from soil geochemical data is 0.619 ($p < 0.001$) and for the group of 34 sites in England and Wales where TGDR estimates are derived from the relatively high-density soil geochemical data, the Pearson correlation coefficient is 0.656 ($p < 0.001$). For the 27 sites in Scotland where the TGDR estimates are derived from stream sediment geochemical data, there is, as would be expected, a less significant correlation coefficient between AGDR and TDGR (r = 0.568, p = 0.002). Normal RIMNET AGDR values are quoted in the 2022 data set to the nearest 10 nGy/h. In some earlier RIMNET data sets, such as 2010, annual mean AGDR data are quoted to the nearest 1 nGy/h. The Pearson correlation coefficient between mean 2010 AGDR and soil derived TGDR is slightly higher (0.625; $p < 0.001$) compared with the Normal 2022 RIMNET AGDR – soil derived TGDR correlation (0.586; $p < 0.001$) for the 76 sites with 2010 and 2022 data.

Mean and median data for indoor dose rates at the 10,199 indoor measurement locations are virtually the same as the terrestrial + cosmic dose rates derived from BGS geochemical data (Table 3). These relationships for mean and median are similar when the data are split into England and Wales (n = 8964) and Scotland (n = 1235) (Supplementary data tables SM-5 and SM-6). It should be remembered that indoor dose rates differ from those outdoors because of the shielding of the building materials and because of radiation from radionuclides within them. The close similarity of the mean and median values is likely to be fortuitous.

The Pearson correlation coefficient between indoor dose rate and outdoor gamma dose rate (from Green et al., 1989) is higher than between indoor dose rate and terrestrial Total (K + Th + U) + cosmic dose rates (Table 5 and Table 6). When the data are split into England and Wales (n = 8964) and Scotland (n = 1235) the Pearson correlation correlations for England and Wales are virtually the same as for GB (Table 5). In contrast, whereas the correlation coefficient between outdoor and terrestrial + cosmic dose rates is still significant ($p < 0.001$) for the Scotland subset, the correlation coefficients between indoor dose rate and (a) outdoor dose rate (Green et al., 1989) or (b) Total (K + Th + U) terrestrial + cosmic dose rate derived from BGS sediment and soil geochemical data are not significant (Table 5).This probably reflects the smaller ranges and lower number of locations in Scotland compared with England and Wales (Supplementary Figures SM-10 and SM-11).

In the case of the indoor dose rate vs outdoor (Green, 1989) relationship (Fig. 9), tens to hundreds of indoor measurements have been assigned the same "infilled and doubly smoothed" value for the 10 km grid square in which the indoor measurement points are located (Fig. 4 in Green et al., 1989). It would be interesting to assess the relationship between the indoor dose rate data and the 'raw' outdoor dose rate data but, as noted above, the 'raw' data are unfortunately not available.

Table 6
Pearson correlation coefficients between indoor, outdoor (Green et al., 1989) and terrestrial K, Th, U, K + U and K + Th + U dose rates derived from BGS geochemical data at 10,199 childhood homes (Kendall et al., 2016) (all correlation coefficients have P values of <0.001).

	Indoor	Outdoor	K	Th	U	KU
Outdoor	0.312					
K	0.121	0.560				
Th	0.063	0.468	0.512			
U	0.154	0.464	0.282	0.516		
KU	0.163	0.643	0.900	0.630	0.672	
KThU	0.138	0.635	0.832	0.843	0.675	0.949

Outdoor gamma-ray measurements (Green et al., 1989) were made 1 m above the ground generating values representing a relatively large area with a radius of tens to up to 100 m (Cinelli et al., 2019). In contrast the soil samples were composites based on 5 sub-samples taken from a depth of 5–20 cm at the centre and four corners of a 20 m square. The soil-sampling measurements are likely to reflect a more accurately random sampling of the total area than the outdoor measurements, which were the result of ad-hoc measurements. Dose rates estimated in this study from soil and soil equivalent geochemical data are 5–20 cm depth soil dose rates which are likely to be higher than the dose rates measured at 1 m above the surface (Green et al., 1989), which they are by an average of 16 nGy/h (17%) (Table 4 and SM-3, SM-4).

Perhaps the major factor to consider is that the surface (5–20 cm depth) soil samples were analysed by XRF on disaggregated, <2 mm sieved, dried soil. Deeper (30–50 cm depth) soils and stream sediment samples were sieved to pass a 150 μm mesh prior to analysis by XRF. K, Th and U in surface (5–20 cm depth) soils would in general be expected to be artificially enhanced in the analysed fraction of the soil samples due to the removal of the coarse >2 mm size fraction. No data are readily available to permit this to be corrected for. In addition, soil moisture content, density and porosity can combine to attenuate gamma radiation from sub-surface (5–20 cm depth) soils by factors in the range 0.6 (0–10 cm) to 0.3 (10–20 cm) (Grasty, 1997; Beamish, 2014, 2015). Attenuation could be particularly significant over wet and organic soils when gamma-ray dose rate measurements are made using total count Geiger-Muller counters or multichannel scintillation gamma-ray spectrometers.

4.3. Comparison with other approaches

Estimates of outdoor gamma-ray dose rates for use in epidemiology have been made in Finland (Nikkilä et al., 2016) and in Switzerland (Rybach et al., 2002; Folly et al., 2021). Predicted geogenic uranium potential was also used in France (Ielsch, 2010, 2017; Warnery et al., 2015). Both the Finnish work of Nikkilä and the Swiss approach of

Table 5
Pearson correlation coefficients (R^2) between indoor dose rate, outdoor dose rate (Green et al., 1989) and terrestrial + cosmic dose rate derived from BGS geochemical data at locations with indoor dose rate data (all correlation coefficients have P values of <0.001 except those marked **).

	GB (n = 10,199)		E&W (n = 8964)		Scotland (n = 1235)	
	Indoor dose rate	Outdoor dose rate	Indoor dose rate	Outdoor dose rate	Indoor dose rate	Outdoor dose rate
Outdoor dose rate (Green et al., 1989)	0.312		0.351		−0.011**	
Terrestrial + cosmic dose rate derived from BGS geochemical data	0.138	0.635	0.158	0.638	−0.065**	0.616

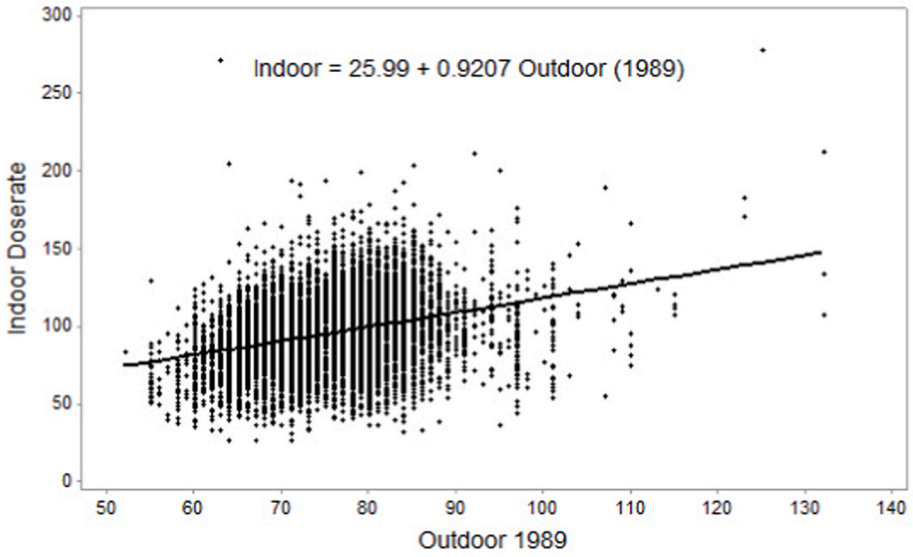

Fig. 9. Relationship between measured indoor dose rate for 10,199 locations and outdoor dose rate derived from 10 km grid square data in Green et al. (1989).

Rybach essentially involved interpolating measurements of various kinds. Folly et al. (2021) used airborne gamma-ray spectrometry data with geological and land coverage information in a Bayesian spatial modelling approach to predict terrestrial gamma radiation in Switzerland. These data were used in childhood cancer epidemiological studies in Switzerland (Mazzei-Abba et al., 2021).

In the context of the present work, it is worth contrasting the approach to estimating levels of uranium (and of thorium and potassium) described in this paper with that of Ielsch and co-workers (Ielsch et al. 2010, 2017). Ielsch et al. set out to investigate the uranium concentration in bedrock of the generalised geological units which were shown on the 1:1,000,000 geological map of France. The main source of information was 5092 geolocated rock analyses plus some hundred non-geolocated analyses. These were combined with various other sources of information, specified in the original papers. The geological units were finally assigned to five categories of "uranium potential" based on their mean uranium content.

This map was used by Warnery and co-workers (Warnery et al., 2015) in conjunction with indoor gamma-ray measurements at 17,404 locations in multi collocated cokriging to predict mean indoor gamma dose rates in cells of 1 × 1 km sq. Warnery et al. (2015) also employed ordinary kriging using the same measurement set. The introduction of the uranium potential data resulted in a modest improvement in the mean square error from 409 to 407 $(nSv/h)^2$. It will be interesting to see whether the present more detailed mapping of U, Th and K in surface soils allows more precise predictions of indoor gamma-ray dose rates than does the consideration of bedrock at a scale of 1:1,000,000.

5. Conclusions

Based on geochemical data we calculate that the mean outdoor gamma-ray dose rates from K, Th and U are 17.1, 18.8 and 13.0 nGy/h, in accordance with the UNSCEAR rule of thumb that the contributions are roughly equal (UNSCEAR, 2008).

The distributions of terrestrial K, Th, U, and total (K + Th + U) gamma-ray dose rates are approximately normal (Figure SM-12).

The terrestrial total (K + Th + U) gamma-ray dose rates (TDGR) predicted from soil and soil-equivalent K, Th and U geochemical data, correlate reasonably well (Pearson $r = 0.635$; $p < 0.001$; $n = 10,199$) with the outdoor measurement data reported by Green et al. (1989). The scope for closer agreement may be limited by the smoothing of the outdoor measurement data (Green et al. (1989). Correlation between TDGR and normal ambient gamma-ray dose rate (AGDR) data at 82 RIMNET sites is also significant ($r = 0.595$; $p < 0.001$).

The new gamma-ray dose rate data derived from geochemical data should provide useful input to the modelling of indoor gamma-ray dose rates.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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