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Dose rate conversion parameters: Assessment of nuclear data

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1	Dose Rate Conversion Parameters: Assessment of Nuclear Data
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7	R
, 8	Abstract
U	
9	The dating of materials using stored dose methods requires accurate determination of the
10	environmental dose rate. The calculation of dose rates from radionuclide concentrations
11	requires conversion parameters derived from nuclear data (half life, decay energies and
12	intensities, and branching ratios). With the substantial body of primary data, it is convenient
13	to use data from evaluated libraries. These libraries show variations reflecting both newer
14	data unavailable to earlier evaluations and the relative importance given to different data sets
15	by the evaluators. Commonly used conversion parameters derive from the Evaluated Nuclear
16	Structure Data File (ENSDF), either directly or from secondary publications, with new
17	tabulations produced in recent years following revisions to this library. Other international
18	evaluations of nuclear data include the NEA/OECD supported JEF2.2 and JEFF3.11
19	evaluations, and the Decay Data Evaluation Project (DDEP). A technique comparing
20	different evaluations to identify data that can not be confidently used has been developed.
21	These differences have been investigated with an evaluation of underlying nuclear data.
22	Particular radionuclides of interest are discussed; ²¹⁴ Bi where recent evaluations depend on a
23	single high precision data set, ²²⁸ Ac where the decay scheme is incomplete and further
24	measurements are required, and 40 K where the mean beta energy has been calculated in the
25	evaluations using an incorrect shape factor. Revised dose rate conversion factors have been

26	produced, which are largely consistent with earlier values with the exception of the 40 K beta
27	parameter which is 4% higher than recent values but consistent with earlier calculations.
28	
29	
30	Keywords
31	Dose rate conversion factors; evaluated nuclear data; 214Bi; 228Ac; 40K
32	
22	
33	Research Highlights
34	• Innovative comparison of evaluated libraries to highlight lower confidence data
35	• Highlighted radionuclides with lowest confidence data - 214Bi, 228Ac, 40K
36	• 40K beta spectrum incorrect in all evaluated libraries, improved mean energy given
37	• Revised dose rate conversion factors using best available data given
38	
39	
40	1. Introduction
41	In luminescence or electron-spin resonance dating the age of a mineral is determined from the
42	ratio of the measured equivalent dose to the environmental dose rate the mineral had been
43	exposed to, including appropriate corrections for grain size and water content. In some
44	instances dose rates can be measured directly, using in-situ spectrometers or dosimeters, but
15	in money cases does not a calculated from measurements of the activity concentration (Da ba
45	in many cases dose rates calculated from measurements of the activity concentration (Bq kg
46	¹) or elemental concentrations (% or ppm) of natural radionuclides, or alpha and beta
47	counting rates, in samples returned to the laboratory for analysis are needed in the absence of
48	field measurements or to supplement other measurements. This calculation uses dose rate
49	conversion factors derived from nuclear data. The calibration of field instruments and

50 dosimeters often also relies on appropriate conversion factors.

51

52 These conversion factors have been calculated several times since the 1970's, following 53 revisions to the nuclear data used, as shown in Fig. 1. The values for these parameters are 54 tabulated in the supplementary material. These show some significant variation, but with recent estimates converging on a common value within a few % uncertainty. Nevertheless, 55 56 comparisons between dose rates determined by different methods and institutions continue to 57 show discrepancies, with the conversion factors used being one potential explanatory factor. 58 Therefore, there is reason to examine these conversion factors again. As the nuclear data used 59 to determine these factors is updated, it could be asked whether it is necessary to update the 60 conversion factors? Or, has the point been reached where those updates will not lead to 61 significant changes in the conversion factors? And, to what extent can we be confident in the 62 nuclear data used? It would be beneficial if variations in nuclear data had been settled to values that are very unlikely to change significantly with further measurement, from which 63 64 the stored dose dating community could derive an agreed set of conversion factors. It is the 65 aim of this work to examine whether this point has been reached, and if not what is needed to move closer to that position. 66

67

The nuclear data required for these calculations are the energies and intensities of discrete radiation (alpha, gamma and x-ray) and the intensities and mean energies of beta radiation, which requires both the decay endpoint and shape of the beta spectrum. When nuclide concentrations are expressed as elemental concentrations (ppm or %) then the half lives of the decays are also required.

73

74 2. Nuclear Data Tables

With the very large quantity of nuclear data available, and the specialist nature of many
publications, it is often impractical to assimilate the primary data, and it is therefore
convenient to refer to evaluated libraries. In this work, three such libraries are considered.

The Evaluated Nuclear Structure Data File (ENSDF) was the first comprehensive nuclear 80 structure data base, developed by staff at Berkeley and Oak Ridge in the late 1970s while the 81 7th Edition of the *Table of Isotopes* was being compiled, with the first version of the data base 82 largely mirroring the *Table of Isotopes* 7th Edition. The ENSDF consortium developed, and 83 an international network of evaluators was established under the IAEA. The ENSDF database 84 85 is maintained by the US National Nuclear Data Center at Brookhaven National Laboratory 86 (NNDC, 2017). The database is under constant development, with updated publication of evaluated data in Nuclear Data Sheets at a rate of approximately every 7-11 years. The 8th 87 88 Edition of the Table of Isotopes was produced using ENSDF data.

89

90 The NEA and OECD coordinated evaluation projects with particular emphasis on nuclear 91 data relevant to reactor physics, which for decay data produced two evaluated libraries, the 92 Joint Evaluated File Version 2.2 (JEF2.2) in 1993 (NEA 2000) and Joint Evaluated Fission 93 and Fusion File Version 3.1.1 (JEFF 3.1.1) in 2007 (Kellett et.al. 2009). Both of these data 94 bases are fixed, without ongoing development and revision.

95

96 It has been noted (Helmer 1999) that a significant limitation on many evaluated libraries is 97 the lack of comments on the origin of the data and processing done, making it impossible for 98 others to judge the quality of the evaluations, and that values for quantities differ between 99 evaluated libraries. To address these concerns the Decay Data Evaluation Project (DDEP) 100 was initiated in 1995 (Helmer 1999, Helmer et.al. 2002) with the intention of giving the most

101 precise values that are justified, with a methodology that includes accounting for all

102 measurements of a quantity (either using or explicitly excluding each measurement),

103 providing written documentation of all the data used and the decisions made, and for each

104 evaluation to be reviewed by other members of the DDEP.

105

For luminescence dating, dose rate conversion factors were calculated in the 1970s and 80s 106 (Aitken 1974, 1983, Bell 1976, Sanderson 1987 and others) using various editions of the 107 Table of Isotopes and Nuclear Data Sheets. Several recent re-evaluations use ENSDF data 108 (Adamiec & Aitken 1998, Guerin et.al. 2011, Liritzis et.al. 2013), without any apparent 109 110 detailed consideration of the basis for the revisions to the evaluated library. As noted, these 111 conversion factors have converged to common values within a few percent. However, they have all been determined from different generations of the same group of evaluations, and 112 any variations using different evaluation procedures would result in a reduction in confidence 113 in these parameters. 114

115

The purpose of this paper is to examine the different evaluated nuclear data libraries for the 116 radionuclides of relevance to dosimetry from natural sources: the decay chains from ²³⁸U, 117 ²³⁵U and ²³²Th, and the decays of ⁴⁰K and ⁸⁷Rb. This will identify the individual radionuclides 118 where there is significant variability in the evaluated nuclear data, or where the uncertainties 119 are larger, which result in the most significant uncertainty in dose rate conversion factors. 120 121 From this examination, further details of existing data for those radionuclides identified as most significant are given. The influence of these variations on dose rate conversion factors 122 for the infinite matrix condition are then described. 123

124

125	3. Examination of evaluated data for naturally occurring radionuclides
126	
127	The data in the JEF2.2, JEFF3.11, ENSDF and DDEP libraries were interrogated to tabulate
128	values for the half lives, mean decay energies and intensities, and where appropriate
129	branching ratios, for all naturally occurring radionuclides. This data was tabulated for alpha,
130	beta and gamma decay, x-rays and conversion electrons. These tables, with a more extensive
131	discussion of the nuclear data, are given elsewhere (Cresswell et.al. 2018). Data were
132	downloaded from the ENSDF and DDEP libraries in March 2017. These values have been
133	used to calculate dose rate conversion factors for each of these evaluations, as shown in Fig. 2
134	and tabulated in the supplementary material.
135	
136	For the uranium series both ENSDF and DDEP generate significantly lower gamma
137	conversion factors and higher beta conversion factors compared to JEF2.2/JEFF3.1.1. These
138	are driven by data for ²¹⁴ Bi, with significant differences for both beta and gamma mean
139	energies between the evaluated libraries (Table 1) accounting for approximately 1.5% of the
140	total beta and 3.5% of the total gamma energies. The end-point energies, shapes and
141	intensities of beta decays to excited levels in ²¹⁴ Po are determined from analysis of the
142	gamma ray emission spectra. Thus differences in the evaluation of the gamma emission data
143	will account for differences in both the gamma and beta energies.
144	

The evaluation of the gamma decay scheme involves normalising available gamma spectrometry data to the 609 keV (1st excited state to ground state) transition, to allow the generation of a relative intensity level scheme. This is then normalised to an evaluated absolute intensity for the 609 keV transition. The absolute intensities for the 609 keV emission in the ENSDF and DDEP libraries are identical (45.49 \pm 0.19%), whereas the

150 intensity in the JEF2.2/JEFF3.11 library is higher (46.9 \pm 4.0%). The JEF2.2 evaluation for ²¹⁴Bi was not revised in the JEFF3.11 evaluation, and the ENSDF and DDEP libraries include 151 two measurements not available to the JEF2.2 evaluators. In particular, a high precision data 152 set reported by Morel et.al. (2004) using ²²⁶Ra sources certified to 0.2% precision, and 4 153 different HPGe detectors and 2 or 3 source geometries for each detector to check cascade 154 summing correction, with an absolute intensity for the 609 keV gamma ray of $45.57 \pm 0.18\%$. 155 To date, this dataset is the most precise measurement of the critical 609 keV absolute 156 intensity, and the relative intensities of the other emissions from ²¹⁴Bi decay. To tie down this 157 critical intensity more conclusively there would be value in independent measurements with 158 159 similar attention to detail to rule out potential bias in the source activity certification, detector 160 efficiencies and cascade summing corrections.

161

For the thorium series, the ENSDF and DDEP libraries produce lower gamma dose rate 162 conversion factors, with significantly improved precision, compared to JEF2.2 and JEFF 163 3.11. This is largely explained by differences in the ²²⁸Ac decay data (Table 2). Even with 164 more recent data improving the precision of the evaluation, the DDEP evaluation (Pearce 165 2010) notes that this decay scheme is incomplete. The effective O-value¹ calculated from 166 167 individual decay rates and intensities $(2010 \pm 100 \text{ keV})$ is low compared to the Q-value from mass differences (2123.8 \pm 2.7 keV). There is a ~7% discrepancy between beta and gamma 168 169 emissions, suggesting missing gammas. "Further measurements of the gamma data, 170 particularly at low energy, would be of benefit, as would coincidence studies to validate the placement of gammas in the level scheme" (Pearce 2010). Absolute gamma emissions were 171 normalised to the 463 keV emission, however it is noted that "this value is not consistent with 172 173 expected beta decay characteristics" (Pearce 2010). If the discrepancies in the decay scheme

¹ The Q-value is the difference in rest mass energy of the parent and daughter nuclides. This should equal the total energy released in the decay (the sum of all radiation and nuclear recoil).

are the result of unobserved gamma rays, with associated beta transitions, then it is expected
that total gamma and beta energies per decay would be larger than determined from the
ENSDF/DDEP libraries by a few percent.

177

For ⁴⁰K the half lives given in the evaluated libraries (Table 3) considered here carry 178 relatively large uncertainties, of 1.0-1.5% for the JEF2.2 and JEFF3.11 libraries, and 0.2% for 179 the ENSDF and DDEP libraries. In addition, the half lives vary significantly with the JEF2.2 180 181 value approximately 2.5% larger than that given in the ENSDF and DDEP libraries. This difference in half life is significant when ⁴⁰K activity is given in terms of elemental 182 composition (%K). In geochronology based on the decay of 40 K it is desirable to know the 183 ⁴⁰K to the greatest possible precision. Given the importance of the ⁴⁰K decay constants in 184 these communities, independent evaluations of the half life and branching ratios have been 185 conducted combining experimental nuclear physics data with Ar-Ar ages from independently 186 dated minerals. Half lives calculated by Min et.al. (2000) and Renne et.al. (2010) using these 187 approaches are also given in Table 3. 188

189

190 There is also considerable variation in the mean beta energies reported in each library, ranging from ~450 keV to ~520 keV, corresponding to 5-10% variation in dose rate 191 192 conversion factors. The major difference to the mean energy is the shape of the betaspectrum assumed. To determine the mean beta decay energy, it is necessary to know the 193 194 shape and end-point energy of the beta spectrum. The end-point energy is well defined from the atomic mass difference between 40 K and 40 Ca, at 1311.07 \pm 0.12 keV. The shape of the 195 beta spectrum is proportional to a factor pWq^2 for sharing momentum between the leptons, 196 the Fermi function F(Z, W) and a shape factor C(W). 197

$$\frac{dN}{dW} \propto pWq^2 F(Z,W) C(W)$$

where W is the total beta energy, Z is the atomic number of the daughter, p is the momentum of the beta particle and q the momentum of the neutrino. Theoretical shape factors for allowed or forbidden unique transitions are given by:

$$C(W) = (2L-1)! \sum_{k=1}^{L} \lambda_k \frac{p^{2(k-1)}q^{2(L-k)}}{(2k-1)! [2(L-k)+1]!}$$

where $L=\Delta J$, and L=1 for $\Delta J=0$. The parameter λ_k cannot be calculated in a straightforward manner, and typically an assumption that $\lambda_k=1$ is used.

203

The decay from 40 K (ground state 4⁻) to 40 Ca (ground state 0⁺) is a third unique forbidden 204 205 (3U) transition. The commonly used LOGFT program for calculating the shape of the spectrum only calculates allowed and first and second unique forbidden (1U, 2U) transitions, 206 207 and when presented with any other transition defaults to an allowed transition shape. 208 Recalculations of the beta spectral shapes to evaluate the reliability of the $\lambda_k=1$ approximation (Mougeot 2015) have included the ⁴⁰K beta spectrum using an experimental 209 210 shape factor from Leutz et.al. (1965). The program BetaShape (Mougeot 2015, 2016) has 211 been used to generate spectra for different shape factors theoretically with the $\lambda_k=1$ assumption and for experimental shape factors reported in the literature. These spectra are 212 213 plotted in Fig.3, with the data included in the supplementary material. The mean beta 214 energies have been calculated for these, and given in Table 4 with corresponding mean 215 energies from the LOGFT program for the allowed and first and second unique forbidden transitions. The mean energy for the Leutz et.al. (1965) shape factor is identical to that 216 217 reported by Mougeot (2015). Recent high precision measurements of beta spectra have been conducted (Carles & Kossert 2007), proposing a shape factor of the form $C(W) = \lambda_1 p^6 + \lambda_2 p^6$ 218 $\lambda_2 q^6 + 7p^2 q^2 (p^2 + q^2)$ for ⁴⁰K, with $\lambda_1 = 1.8$ and $\lambda_2 = 1.23$, to resolve discrepancies between 219 cutoff energy yield and maximum point energy. However, the authors note that this form of 220

shape factor fails Cherenkov counting tests, and in correspondence stated that they consider
the spectrum generated by BetaShape using the Leutz et.al. (1965) shape factor is "the best
choice for K-40 at this moment" (Kossert pers. comm..)

224

Comparing the mean energy per decay in the different evaluations (Table 3) with the 225 calculated mean energies for different shape factors (Table 4) it appears that the JEFF 3.11 226 and DDEP evaluators have used the LOGFT program, letting the program default to an 227 228 allowed transition shape. The DDEP evaluators (Mougeot & Helmer 2009) state that the mean energy is given by the LOGFT program without further elaboration. The mean energy 229 230 reported by the ENSDF evaluation is consistent with forcing the LOGFT program to use a 2U 231 shape factor, which would be closer to the 3U value. And, the JEF2.2 evaluators have a value consistent with the Leutz et.al. (1965) shape factor, with a marginally larger endpoint energy. 232 It is noted that Lederer & Shirley (1978) also give a mean energy for ⁴⁰K beta decay of 233 583 keV, and this value is explicitly stated in the calculations of dose rate conversion factors 234 of Aitken (1983, 1985), Bell (1976), Nambi & Aitken (1986) and Sanderson (1987). 235 Subsequent calculations use the lower mean energy in the ENSDF library, Adamiec & Aitken 236 (1998) note that this leads to a 4% reduction in the 40 K beta conversion factor. 237 238 239 The beta spectrum is also required to calculate absorbed dose fractions. Mejdahl (1979)

recognised that this is a third unique forbidden transition, and used a spectrum transformed from a corresponding allowed shape using the method of Wu & Moszkowski (1966). Nathan et.al (2003) used a Fermi model spectrum with spectral factors from Behrens & Szybisz (1976), with the spectrum given in the supplementary information of Guérin et.al. (2012) having a mean energy of 508 keV, consistent with the use of an allowed spectral shape despite statements that spectral factors had been used.

246

247 **4. Discussion and Conclusions**

248 The estimation of dose rate from radionuclide concentrations requires accurate nuclear data 249 covering half lives, branching ratios, emission energies and intensities. The use of evaluated libraries provides a convenient means of accessing this data to calculate conversion ratios. 250 251 The evolution of the evaluated library reported in Nuclear Data Sheets and Table of Isotopes, and more recently formalised in the Evaluated Nuclear Structure Data File (ENSDF), has 252 253 resulted in a variety of dose rate conversion factors being determined. However, with two 254 additional sets of evaluations; the NEA/OECD JEF2.2 and JEFF3.11 libraries tailored for 255 nuclear reactor and nuclear medicine applications, and the Decay Data Evaluation Project 256 (DDEP); an assessment of the variations in relevant nuclear data across current versions of these different libraries has been conducted with the intention of identifying if the different 257 evaluation processes significantly bias the resulting evaluated data and to identify which 258 radionuclides might carry significant variations, and thus be needing further measurements to 259 260 refine the nuclear data.

261

In the uranium series, combining both ²³⁵U and ²³⁸U, the JEF2.2/JEFF3.11 gamma conversion 262 263 factors are 0.7% higher than the corresponding ENSDF/DDEP values, and conversely the beta conversion parameters are 1.5% lower. It has been shown that these differences are 264 mostly due to differences in the data for ²¹⁴Bi. These differences are due to the inclusion of a 265 single high precision data set (Morel et.al. 2004) in the more recent ENSDF/DDEP 266 evaluations. Although the absolute intensity of the 609 keV gamma ray from this is consistent 267 268 with other measurements since the 1980s, within a 1-2 σ limit, the ENSDF/DDEP evaluations 269 are heavily biased to this single data set. It would be of benefit if an independent high 270 precision data set confirmed the measurements of Morel et.al. (2004). It is considered that the

271 ENSDF and DDEP evaluations, incorporating the Morel et.al. (2004) data, are the best 272 available, with the differences between them insignificant. For the purpose of calculating 273 dose conversion factors, the mean of these is used with an uncertainty assigned that 274 encompasses the 1σ range of both values.

275

In the thorium series, the gamma conversion factors from the ENSDF/DDEP libraries are 276 ~3.5% lower than for the JEF2.2/JEFF3.11 libraries. It has been shown that these differences 277 are largely derived from differences in the ²²⁸Ac nuclear data. Although this has not been 278 examined in detail here, the DDEP evaluator (Pearce 2009) noted that the decay data for this 279 280 radionuclide is incomplete with an ~7% discrepancy between beta and gamma intensity data, 281 and recommended that further experimental gamma data be collected with particular emphasis on low energy gammas and coincidence measurements to confirm the level scheme. 282 It is considered that the ENSDF/DDEP evaluations are the likely to underestimate total 283 gamma and beta energies per decay by 2-4%, and for this work it has been assumed that the 284 ENSDF/DDEP values should be used with an additional 3% added to them, with the 285 uncertainty increased by 10%. 286

287

For ⁴⁰K and ⁸⁷Rb, there is considerable variation in both beta and gamma dose conversion
factors between the four contemporary evaluated libraries and the literature values from
different versions of the ENSDF library, with 3-12% variation in the beta conversion
parameters and 1-4% variation for the gamma conversion parameters. These variations reflect
differences in ⁴⁰K nuclear data in different evaluations; with 2-5% variation in half life
values, upto 15% variation in mean beta energy, and small variations in branching ratios.

The ⁴⁰K half life is also critical to geochronology, and independent evaluations of nuclear 295 296 data and Ar-Ar measurements of known age minerals by the geochronology community has produced half-life values consistent with the latest ENSDF and DDEP evaluations with 297 298 similar precision of ~0.2%. The mean of the ENSDF & DDEP values for the half life has 299 therefore been taken as the best value currently available, with uncertainties to encompass the 300 1σ range of both values. The dominant difference between evaluated libraries is the mean beta decay energy, which reflects the shape of the decay spectrum used. The LOGFT 301 302 program most commonly used to calculate mean energies and branching ratios does not include the 3U transition needed to model the ⁴⁰K decays to ⁴⁰Ca and the ⁴⁰Ar ground state. 303 304 Thus, approximations are made in the calculations of mean energy and branching ratio, the 305 difference in mean energy reflects the difference between using a 2U or an allowed transition 306 as an approximation to the 3U transition. The use of an alternative program, BetaShape, that 307 does allow for 3U shape factors results in an increase in the calculated mean energy. At present, experimental shape factors for the ⁴⁰K decay are poorly measured, especially in the 308 low energy (<100 keV) region, and additional precision measurements of the 40 K beta 309 310 spectrum would be of considerable benefit in more accurately determining experimental shape factors. However, it would be surprising if future experimental data results in a mean 311 energy significantly different from 585 ± 5 keV, with the best current measurement giving a 312 313 mean energy of 583.98 ± 0.10 keV (Leutz et.al. 1965, Mougeot 2015) which includes an uncertainty twice that given by this shape factor. 314

315

Based on our assessment of what are currently the best nuclear data as reviewed here, dose rate conversion factors can be calculated (Table 5). In most cases, these are consistent with values currently regularly used (Adamiec & Aitken 1998, Guérin et.al. 2011, Liritzis et.al. 2013). The exception is the K+Rb beta parameter which is approximately 4% larger, in line

with earlier calculations using the 583 keV mean energy of Lederer & Shirley (1978),
including the calculations of Aitken (1983) which had been widely used prior to adoption of
the newer values. It should be noted that although these conversion factors are determined
using our assessment of the best available data, there are still serious doubts regarding that
data. As such, these factors are provisional subject to clarification of the inconsistencies in
the nuclear data.

326

These conversion factors have been used to calculate dose rates for a standard mineral composition following the assumptions of Adamiec & Aitken (1998) for alpha efficiency, beta attenuation and cosmic contributions. These are given in Table 6 with values given by Adamiec & Aitken (1998) for comparison. In all instances, the total dose rates are slightly larger than those calculated by Adamiec & Aitken (1998), and generally in agreement with those of Nambi & Aitken (1986) except for ⁴⁰K (where the current work predicts higher dose rates) and Th gamma.

334

It is clear that the choice of dose rate conversion factors to be used is still to be fully resolved. 335 The values suggested here may be used, but without resolution of significant doubts in the 336 337 nuclear data it may be preferable to maintain continuity with prior work by not changing the values used with each new revision. Whatever values are used, it should be recognised that 338 propagating uncertainties in the evaluated libraries would underestimate the true 339 340 uncertainties. In this work, uncertainties in the parameters are in the range of 1-3%, previous 341 studies (Aitken 1985, following Bell 1979) have noted that 5% is the maximum error likely to 342 occur. In addition, the values used should be clearly stated when reporting dose rates. 343

344

	ACCEPTED MANUSCRIPT
345	
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474 Figure 1: Dose rate conversion factors for the uranium series ($^{235}U + {}^{238}U$), Th series (^{232}Th)

- 475 and 40 K taken from the literature. The control lines show the robust mean ± 1 standard
- 476 deviation.

477



478 Figure 2: Dose rate conversion factors for the uranium series $(^{235}U + ^{238}U)$, Th series (^{232}Th)

- 479 and 40 K calculated from four evaluations. The control lines show the robust mean ± 1
- 480 standard deviation from previous literature values (Fig. 1).
- 481



Figure 3: Beta spectra calculated by the BetaShape program (Mougeot 2015, 2016) using theoretical shape factors, with the $\lambda_k = 1$ assumption, for allowed and first, second and third unique forbidden transitions, and for experimentally derived shape factors reported by Leutz et.al. (1965) and Carles & Kossert (2007). All spectra are calculated using an endpoint energy of 1311.07 keV.

490 Table 1: Mean gamma and beta energies per decay of ²¹⁴Bi calculated from different

491 evaluated libraries

	Mean	energy per decay (keV)
	Gamma	Beta
JEF 2.2/JEFF 3.11	1536.9 ± 30.3	613.7 ± 15.5
ENSDF	1474.1 ± 1.6	640.0 ± 3.3
DDEP	1467.8 ± 1.9	645.8 ± 3.7
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494 Table 2: Mean gamma energies per decay of ²²⁸Ac calculated from different evaluated

495 libraries

	Mean energy per decay (kev	V)
JEFF 3.11	955.6 ± 133.8	
ENSDF	864 ± 10	
DDEP	864 ± 10	
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	Half life	End point	Mean energy	Branching %	Mean energy
	(x10 ⁹ a)	(keV)	/ beta decay		/ 40K decay
			(keV)		(keV)
JEF2.2	1.280 ±	1311.6 ± 0.5	584.9 ± 0.6	0.893 ±	521.48 ±
	0.010			0.001	0.02
JEFF 3.11	$1.265 \pm$	$1311.04 \pm$	$508.29~\pm$	89.15 ± 0.13	453.16 ±
	0.020	0.12	0.05		0.66
ENSDF	$1.248 \pm$	$1311.07 \pm$	560.18 ±	89.14 ± 0.18	499.3 ± 1.0
	0.003	0.12	0.05		
DDEP	$1.2504 \pm$	$1311.07 \pm$	508.32 ±	89.25 ± 0.17	$453.68 \pm$
	0.0030	0.12	0.05		0.87
Min etal	$1.269 \pm$				
(2000)	0.025				
Renne etal	$1.2479 \pm$				
(2010)	0.0024				

499	Table 3: ⁴	⁴⁰ K half lives,	beta-	branching	ratios a	nd energie	s from	different	evaluations
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502 Table 4: Mean beta energies calculated for an endpoint energy of 1311.07 keV using the

- 503 BetaShape program (Mougeot 2015, 2016) for theoretical shape factors, assuming $\lambda_k = 1$, and
- 504 experimentally measured shape factors. With mean energies from the LOGFT program for
- 505 comparison.

		C(W)	Mean beta energy (keV)			
			BetaShape	LOGFT		
	Allowed	1	507.83 ± 0.05	508.31 ± 0.05		
	1U	$p^2 + q^2$	536.08 ± 0.05	534.04 ± 0.05		
	2U	$p^4 + q^4 + \frac{10}{3}p^2q^2$	563.01 ± 0.05	560.18 ± 0.05		
	3U	$p^6 + q^6 + 7p^2q^2(p^2 + q^2)$	587.89 ± 0.05			
	Leutz et.al. 1965	$0.95p^6 + 1.05q^6 + 6.3p^2q^4 + 6.25q^2p^4$	583.98 ± 0.05			
	Carles &	$1.8p^6 + 1.23q^6 + 7p^2q^4 + 7q^2p^4$	569.26 ± 0.05			
	Kossert 2007					
506						
507						
508						
	The second se					

509	Table 5: Dose rate conversion factors based on current best nuclear data. The da	ata used are
510	from the DDEP library with the exception of ²¹⁴ Bi (mean of ENSDF and DDEF	2), 228 Ac (the
511	ENSDF and DDEP values increased by 3%) and 40 K mean beta energy (from the	e Leutz et.al.
512	1965 shape factor), and ⁸⁷ Rb, ²³⁰ Th, ²²⁸ Ra and ²²⁷ Th which are not currently inc	luded in the
513	DDEP library (ENSDF data used for these).	$\boldsymbol{\mathcal{A}}$

TotalAlpha 2.79 ± 0.0 Beta 0.142 ± 0 Gamma 0.112 ± 0 $^{-1}$ The columns labelleinclude the full 235 U ofAdamiec & Aitken 19	Pre-Rn ¹ 2 1.27 \pm 0.01 2 0.056 \pm 0.001 2 0.0037 \pm 0.0001 3 'Pre-Rn' give values ecay series due to the	Total 0.738 ± 0.004 0.028 ± 0.001 $1 0.0489 \pm 0.000$ es for 100% escape e short half life of ²¹	Pre-Rn ¹ 0.309 ± 0.002 0.0096 ± 0.0007 $3 0.0188 \pm 0.0002$ of radon for ²³⁸ U and ¹⁹ Rn, following the ap	mGy a^{-1} % ⁻¹ 0.854 ± 0.008 0.248 ± 0.003 ²³² Th, but
TotalAlpha 2.79 ± 0.0 Beta 0.142 ± 0 Gamma 0.112 ± 0 ¹ The columns labelleinclude the full 235 U ofAdamiec & Aitken 1	Pre-Rn ¹ $2 1.27 \pm 0.01$ $2 0.056 \pm 0.001$ $2 0.0037 \pm 0.0001$ $3 0.0037 \pm 0.0001$ $3 0.0037 \pm 0.0001$ $3 0.0037 \pm 0.0001$	Total 0.738 ± 0.004 0.028 ± 0.001 $1 0.0489 \pm 0.000$ es for 100% escape e short half life of ²¹	Pre-Rn ¹ 0.309 ± 0.002 0.0096 ± 0.0007 $3 0.0188 \pm 0.0002$ of radon for ²³⁸ U and ¹⁹ Rn, following the ap	0.854 ± 0.008 0.248 ± 0.003 ²³² Th, but
Alpha 2.79 ± 0.0 Beta 0.142 ± 0 Gamma 0.112 ± 0 1 The columns labelleinclude the full 235 UAdamiec & Aitken 1	2 1.27 ± 0.01 2 0.056 ± 0.001 2 0.0037 ± 0.0001 3 'Pre-Rn' give values ecay series due to the	0.738 ± 0.004 0.028 ± 0.001 $1 0.0489 \pm 0.000$ es for 100% escape e short half life of ²¹	0.309 ± 0.002 0.0096 ± 0.0007 $3 0.0188 \pm 0.0002$ of radon for ²³⁸ U and ¹⁹ Rn, following the ap	0.854 ± 0.008 0.248 ± 0.003 232 Th, but
Beta 0.142 ± 0 Gamma 0.112 ± 0 ¹ The columns labello include the full ²³⁵ U of Adamiec & Aitken 1	$0.002 0.056 \pm 0.001$ 0.0037 ± 0.0001 $1 \text{ 'Pre-Rn' give values}$ ecay series due to the	0.028 ± 0.001 1 0.0489 ± 0.000 es for 100% escape e short half life of ²¹	0.0096 ± 0.0007 3 0.0188 ± 0.0002 of radon for ²³⁸ U and ¹⁹ Rn, following the ap	0.854 ± 0.008 0.248 ± 0.003 ²³² Th, but
Gamma 0.112 ± 0 ¹ The columns labelled include the full ²³⁵ U of Adamiec & Aitken 1	$\begin{array}{c} 0.0037 \pm 0.0001 \\ \hline 1 \text{ 'Pre-Rn' give values} \\ \hline \text{ecay series due to the} \end{array}$	$\frac{1}{2} 0.0489 \pm 0.000}{2}$	3 0.0188 ± 0.0002 of radon for ²³⁸ U and ¹⁹ Rn, following the ap	0.248 ± 0.003 ²³² Th, but
¹ The columns labelle include the full ²³⁵ U of Adamiec & Aitken 1	d 'Pre-Rn' give value ecay series due to the	es for 100% escape e short half life of ²¹	of radon for ²³⁸ U and ¹⁹ Rn, following the ap	²³² Th, but
include the full ²³⁵ U Adamiec & Aitken 1	ecay series due to the	e short half life of ²¹	¹⁹ Rn, following the ap	proach of
Adamiec & Aitken 1	-			
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- 519 Table 6: Dose rates (mGy a⁻¹) for a standard concentration from Nambi & Aitken, 1986
- 520 (N&A 86), Adamiec & Aitken 1998 (A&A 98) and the current work, assuming zero moisture
- 521 content.

		1% K	50 ppm Rb	3 ppm Th	1 ppm U	Total
Effective	N&A 86			0.190	0.222	0.413
alpha ¹	A&A 98			0.183	0.218	0.401
	This work			0.190	0.223	0.413
Beta	N&A 86	0.814	0.023	0.086	0.147	1.071
	A&A 98	0.782	0.019	0.082	0.146	1.029
	This work	0.835	0.019	0.084	0.142	1.080
Gamma ²	N&A 86	0.243		0.156	0.114	0.693
	A&A 98	0.243		0.143	0.113	0.679
	This work	0.247		0.147	0.112	0.686
Fine-	N&A 86	1.058	0.023	0.433	0.489	2.18
grain total	A&A 98	1.025	0.019	0.408	0.477	2.11
	This work	1.082	0.019	0.421	0.477	2.18
Coarse	N&A 86	0.976	0.018	0.234	0.246	1.65
grain	A&A 98	0.947	0.014	0.217	0.244	1.60
total ³	This work	0.999	0.014	0.223	0.240	1.66

522 Effective alpha dose rates are derived using k=0.1 and $k_{eff} = 0.86$ (Th) and 0.80 (U),

523 following Adamiec & Aitken (1998).

- ² Total gamma includes 0.180 mGy a⁻¹ as a cosmic component, following Adamiec & Aitken
 (1998).
- 526 ³ Beta attenuation factors for coarse grains taken as 0.90, except for Rb which is taken as
- 527 0.75, following Adamiec & Aitken (1998).