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DDEP re-evaluation of the radioactive decay scheme of ^{137}Cs

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Abstract

A complete re-evaluation of the ^{137}Cs β^- decay scheme has been performed within the scope of the Decay Data Evaluation Project (DDEP), taking full advantage of the most recent measurements. An evaluated half-life of 30.018 (22) years is recommended, along with a full revision of the β^- decay branching ratios that also results in a re-evaluation of the absolute emission probability of the 662-keV gamma ray to give a value of 85.01 (20)%. Recommendations for new measurements are also suggested.

Keywords

Evaluated decay data, ^{137}Cs , Half-life, β^- decay, γ decay, DDEP

Highlights

- New recommendations for the ^{137}Cs decay data
- Decay Data Evaluation Project guidelines
- New measurements proposed to improve accuracy of recommended decay data.

I. Introduction

24 Initially discovered in 1941 by Margaret Melhase¹, ^{137}Cs has since become the prime
25 example of human made radioactivity. As one of the main products of the neutron induced
26 fission of ^{235}U (Plompen et al., 2020), ^{137}Cs is primarily related to the production of nuclear
27 energy. Created during reactor operation, ^{137}Cs plays an important role in decay heat
28 calculations, waste management (Wigeland et al., 2006) and decommissioning (IAEA, 1998).
29 However, for a vast majority of the public, radioactive cesium remains associated with
30 nuclear disasters after which radioactive contamination persists for several decades (Zhidkin
31 et al., 2020; Tajima et al., 2022). Beyond nuclear industry, ^{137}Cs has also been adopted for
32 direct use in a variety of applications: sealed radioactive sources in industrial radiography
33 (IAEA, 2011), brachytherapy (Ghilezan and Martinez, 2012), sterilization of medical tools
34 (Parsons, 2012), food irradiation (Bisht et al., 2021), and as a calibration standard for particle
35 detectors in research and industry. Decay parameters such as half-life or emission intensities
36 are essential for most applications and since the 1960s many evaluations have been tentatively
37 performed to provide a robust and consistent decay scheme accessible to non-specialists
38 (Matsumoto, 1965).

39 With an intermediate half-life of approximately 30 years and a single significant
40 gamma-ray emission, ^{137}Cs represents a readily available and well-defined source of gamma
41 radiation. The ^{137}Cs decays by β^- radioactivity to the ground and first two excited states of
42 ^{137}Ba mainly populating the second excited state that depopulates predominantly via an
43 intense 662-keV gamma ray. Even though the decay scheme of ^{137}Cs is simple, the evaluation
44 of the component decay data has long been problematic. Despite an abundance of
45 measurement campaigns, the recommended half-life has been assessed and discussed for
46 decades without resolution (Woods, 1990). The most recent half-life recommendations
47 performed by Helmer and Chechev (2006) and Browne and Tuli (2007) illustrate this

¹ undergraduate student at the University of California Berkeley (Patton, 1999),

48 complexity, with their reported uncertainties which are about ten times larger than reported in
 49 the most precise measurement of Dietz and Pachucki (1973). In addition to the half-life
 50 puzzle, the beta branching ratios have historically relied upon only a handful of measurements
 51 leading to various interpretations of the dataset (see Table 1).

52 Table 1: Recent evaluations of the main decay parameters of ^{137}Cs .

Evaluation	$T_{1/2}$ (years)	$^{137}\text{Cs } 7/2^+ \rightarrow ^{137}\text{Ba } 11/2^-$ β intensity (%)	$^{137}\text{Ba } 11/2^- \rightarrow ^{137}\text{Ba } 3/2^+$ γ intensity (%)	$^{137}\text{Cs } 7/2^+ \rightarrow ^{137}\text{Ba } 3/2^+$ β intensity (%)
Helmer and Chechev (2006)	30.05 (8)	94.36 (28)	84.99 (20)	5.64 (28)
Browne and Tuli (2007)	30.08 (9)	94.7 (2)	85.1 (2)	5.3 (2)
This work	30.018 (22)	94.57 (26)	85.01 (20)	5.43 (26)

53

54 A number of recent experimental studies support a re-evaluation of the ^{137}Cs decay
 55 scheme within the Decay Data Evaluation Project (Kellett and Bersillon, 2017). This exercise
 56 resulted in a newly recommended half-life and a re-balance of the beta branching ratios. A
 57 more extensive and exhaustive description of the analysis can be found in the Comments file
 58 of Leblond (2023).

59 II. Half-life evaluation

60 The ^{137}Cs half-life has been intensively studied in many countries during a period
 61 covering more than 70 years. A total of 56 publications from 1948 to 2020 have been
 62 identified in which a wide variety of measurement techniques were adopted: direct counting
 63 with ionization chambers, germanium detectors, liquid scintillation counters, mass
 64 spectrometry and specific activity with either proportional counters or liquid scintillators. This
 65 comprehensive dataset has been extensively refined in this work by following DDEP
 66 guidelines: only measurements with uncertainties are considered and only one value per

67 author and laboratory is used (the most precise is usually selected). A careful analysis of the
68 experimental details along with a full assessment of the uncertainty budget should also be
69 performed so as to discard any doubtful work. This form of analysis performed by Woods
70 (1990) provides valuable insights into many publications from the 1950s to the 1970s.
71 Applying this methodology to the dataset reduces it to only six publications, which are listed
72 in Table 2. The unit chosen to quantify the half-life is “year” for all references and if
73 necessary conversion was performed on the basis of 1 year = 365.242198 days. Similarly, the
74 uncertainty on the half-life is chosen to be given at the 68 % confidence level. If necessary,
75 conversions were performed to convert to the appropriate confidence level, and
76 symmetrisation was performed based on the procedure described in Kondev et al. (2021).

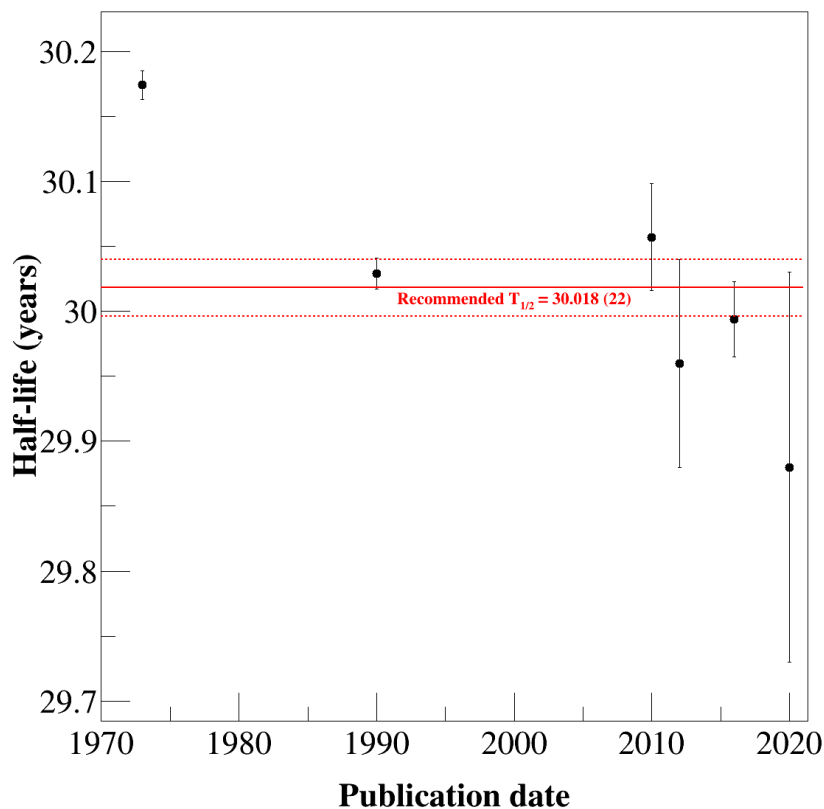
77 Table 2: Measurements considered in the evaluation of the ^{137}Cs half-life

Reference	Half-life (years)	Comments
Dietz and Pachucki (1973)	30.174 (11)	Discarded
Martin and Taylor (1990)	30.029 (12)	Uncertainty increased to 0.022 y
Schrader (2010)	30.057 (41)	
Bellotti et al. (2012)	29.96 (8)	
Juget et al. (2016)	29.994 (29)	
Unterweger and Fitzgerald (2020)	29.88 (15)	

78

79 The most precise of the six measurements has been performed by Dietz and Pachucki
80 (1973) by means of mass spectrometry. They surveyed the ratios between the ^{137}Cs , ^{135}Cs and
81 ^{133}Cs concentrations over a period of 11 years, although only a limited amount of detail can be

82 found concerning the uncertainty budget. Four of the other measurements have been
83 performed by direct counting with ionization chambers over a period of time varying from 8
84 to 30 years (Martin and Taylor, 1990; Schrader, 2010; Juget et al., 2016; Unterweger and
85 Fitzgerald, 2020). Overall, these four measurements provide greater levels of detail
86 concerning the experimental conditions and their uncertainty budgets. The other remaining
87 experimental study was a direct measurement over a period of 217 days, performed by means
88 of a high-purity germanium detector located in an underground laboratory at the Gran Sasso
89 Science Institute (Bellotti et al., 2012). While the measurement technique is described in
90 detail, little information is provided regarding the uncertainty budget estimation. The half-life
91 values and the associated uncertainties from the six measurements are plotted in Fig. 1.



92

93 Figure 1: Measurements considered in the re-evaluation of the ^{137}Cs half-life – measured
94 values and associated uncertainties are represented as function of the publication date.

95 The plot clearly displays the difficulty encountered by previous evaluators to
96 recommend a half-life: the two most precise measurements are extremely discrepant. Prior to
97 2010 this situation was especially difficult to handle although four separate measurements at
98 later dates exhibit good agreement and reasonable overlap with Martin and Taylor (1990)
99 while being discrepant with respect to Dietz and Pachucki (1973). This significant
100 discrepancy could either be attributed to an unaccounted experimental bias in the mass
101 spectrometry measurement, or a substantial underestimation of the associated uncertainties.
102 Under these circumstances and the previously mentioned lack of details on the uncertainty
103 estimation, the study of Dietz and Pachucki (1973) has been removed from further
104 consideration in the present analysis.

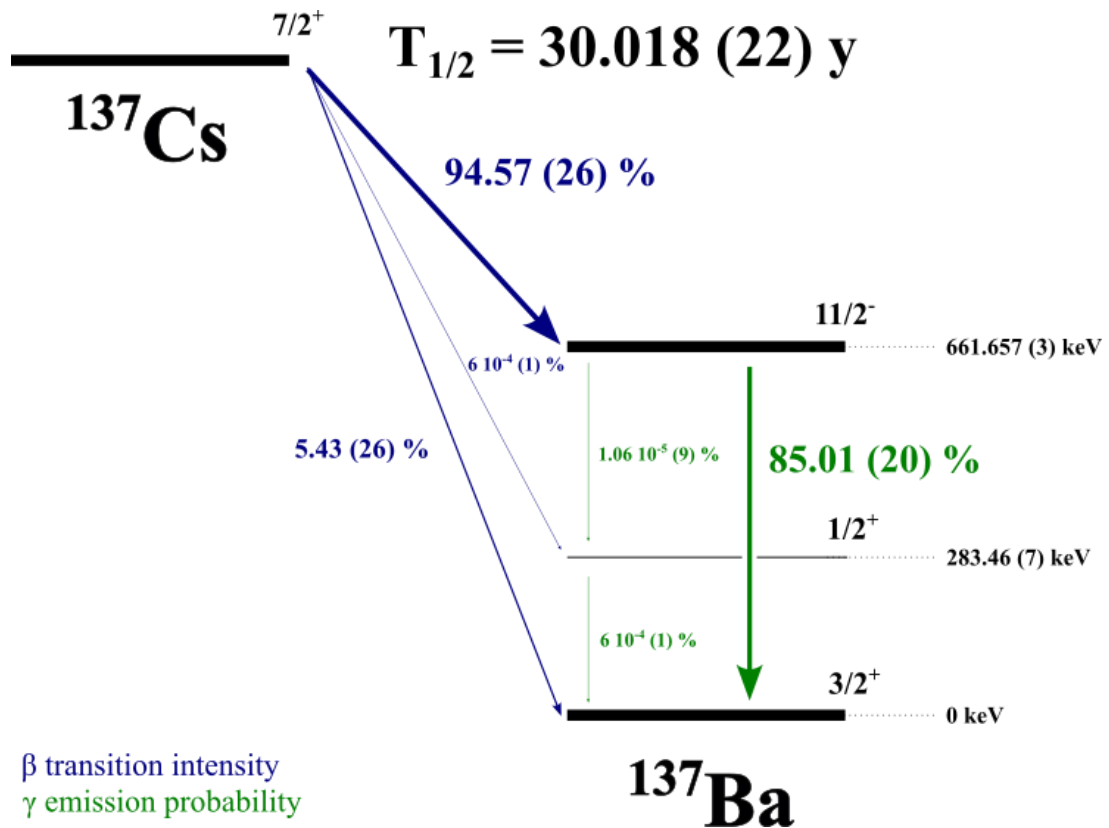
105 The resulting set of five measurements is consistent (Table 2), and was adopted to
106 calculate an unweighted average $T_{1/2} = 29.984$ (30) years and weighted-average
107 $T_{1/2} = 30.024$ (11) years. Both averaging methods provide compatible results, but deviate
108 slightly from each other, highlighting the impact that the inclusion of uncertainties has in
109 these calculations. Both of the studies of Unterweger and Fitzgerald (2020) and Bellotti et al.
110 (2012) have little influence of the final weighted average because of their relatively large
111 uncertainties. Furthermore, the uncertainty assigned by Bellotti et al. (2012) is of limited
112 value because of the short measurement time when compared to the expected half-life. As for
113 Unterweger and Fitzgerald (2020) the uncertainty was initially estimated to be much smaller
114 (as reported in Unterweger, 2002) but had to be increased after the discovery of an unforeseen
115 experimental bias. Approximately 78% of the total weighted-average is therefore associated
116 with the measurement of Martin and Taylor (1990) which strongly dictates the mean value.
117 However, why this measurement has an uncertainty significantly lower than the values
118 assigned in both Schrader (2010) and Juget et al. (2016) is unclear since all of them employ a
119 similar detection technique. Although the work of Martin and Taylor (1990) involves

120 arguably a longer measurement time, the statistical component of the uncertainty is not
121 dominant in any of the three studies. Furthermore, Martin and Taylor (1990) provide two
122 estimations of the uncertainty in their work: 4.5 days (preferred in the article) and 31 days (at
123 the 99% confidence level, and later referred to as "excessively pessimistic"). On the basis of
124 all of these considerations, the uncertainty of Martin and Taylor (1990) has been justifiably
125 expanded from 0.012 to 0.022 years in order to limit the impact of this reference on the final
126 recommended value. With this corrected dataset, the calculated weighted mean is
127 $T_{1/2} = 30.018$ (16) years, or $T_{1/2} = 30.018$ (22) years if the uncertainty is limited to no less than
128 the lowest uncertainty declared in the adopted dataset. The latter value is preferred for the
129 DDEP recommendation, and is compared with the evaluations of Helmer and Chechev (2006)
130 and Browne and Tuli (2007). The newly recommended half-life is consistent with previous
131 evaluations, but with a deduced uncertainty that is four times smaller and highlights the effect
132 of the latest measurements on the current recommendation.

133 **III. Decay scheme reconstruction**

134 The decay scheme of ^{137}Cs is relatively simple and involves only three states of the
135 ^{137}Ba (Fig 2), whereby the beta decay primarily populates the ground state ($3/2^+$) and the
136 second excited state ($11/2^-$). Beta decay to the first excited state ($1/2^+$) of ^{137}Ba is extremely
137 weak, and has never been observed experimentally. Once populated, the second excited state
138 of ^{137}Ba decays mostly by a gamma transition to the ground state. Recently, a second very
139 weak gamma transition to the first excited state has been observed (Moran et al., 2014). The
140 decay of the first excited state then is performed through a weak gamma transition which has
141 only been observed three time (Bikit et al., 1996; Wagner et al., 1997; Sergeev and Valiev,
142 2007). The ^{137}Cs $11/2^-$ state is known to be metastable and is a candidate for a double-gamma
143 emission decay. Such event has been measured to be extremely small at more than six orders

144 of magnitude less probable than direct single-photon emission (Merchan et al., 2015; Pietralla
 145 et al., 2016).



146

147 Figure 2: Recommended decay scheme for ^{137}Cs – transitions between states are coloured
 148 with respect to their types, and their relative probabilities are indicated by the extent of their
 149 line widths.

150 The decay scheme is reconstructed in two successive and independent steps to account
 151 properly for the large spread between the different transition probabilities. Only the ground
 152 state and the second excited state of ^{137}Ba are initially considered in order to estimate the beta
 153 branching ratios and the 662-keV gamma-ray emission probability. The first excited state is
 154 then added to this decay scheme and the remaining intensities are deduced from the intensity
 155 balance. Given the extremely low probability of the double-gamma emission probability, this
 156 particular decay mode has not been included in the tabulated recommendations.

157 During the first stage of the analysis, the contribution of the first excited state of ^{137}Ba
 158 is neglected. Assuming the balance of the decay scheme, the two beta branching ratios and the
 159 662-keV gamma-ray emission probability are thus linked by the following equation:

$$160 \quad 100 - I_{\beta}^{3/2^+} = I_{\beta}^{11/2^-} = I_{\gamma}^{662} (1 + \alpha_T^{662}) \quad (1)$$

161 where $I_{\beta}^{3/2^+}$ is the beta branching ratio to the $3/2^+$ ground state, $I_{\beta}^{11/2^-}$ is the beta branching
 162 ratio to the $11/2^-$ second excited state, I_{γ}^{662} is the gamma emission intensity from this state to
 163 the ground state and α_T^{662} is the total internal conversion coefficient associated with the 662-
 164 keV gamma transition. Assuming the total internal conversion coefficient of the 662-keV is
 165 known, the complete decay scheme of the ^{137}Cs can be reconstructed from the measurement
 166 of either of the beta branching ratios, or from the gamma-ray emission probability. The usual
 167 evaluation practice is to rely only on the gamma-ray measurements as they are generally
 168 considered more accurate and less sensitive to experimental bias than their beta-spectrum
 169 counterparts. However, all available studies have been taken into account for this analysis to
 170 mitigate for the limited amount of data.

171 A total of 22 measurements have been identified with this form of analysis, among
 172 which seven have been undertaken to determine the single gamma-ray emission probability,
 173 while 15 focused on the beta branching ratios. Similar to the analysis performed for the half-
 174 life recommendation, the dataset has been refined further by applying DDEP guidelines and
 175 corrections. The remaining measurements of suitable merit are all listed in Table 3.

176 Table 3: Measurements considered in the evaluation the absolute intensity of the 662-keV
 177 gamma ray.

Reference	Measurement type	Intensity measured (%)	Corresponding gamma emission intensity (%)	Comments
-----------	------------------	------------------------	--	----------

Hsue et al. (1966)	Beta branching ratios	94.0 (5)	84.50 (5)	Discarded
Hansen et al. (1969)	Beta branching ratios	94.6 (3)	85.0 (3)	
Legrand et al. (1973)	662-keV gamma emission intensity	85.3 (3)	85.3 (3)	Uncertainty corrected
Gromov et al. (1978)	Beta branching ratios	93.6 (5)	84.1 (5)	Discarded
Merritt and Gibson (1978)	662-keV gamma emission intensity	84.7 (7)	84.7 (7)	
Behrens and Christmas (1983)	Beta branching ratios	94.43 (7)	84.89 (15)	Uncertainty extended to 0.20%

178

179 Three of the six selected measurements were performed by using magnetic
180 spectrometers to determine the beta branching ratios of the ^{137}Cs decay (Hsue et al., 1966;
181 Gromov et al., 1978; Behrens and Christmas, 1983). The last of these three studies is actually
182 an adjustment of the work of Christmas and Cross (1978) whereby the theoretical shape of the
183 beta spectrum has been revised. Among the remaining measurements, both Hansen et al.
184 (1969) and Legrand et al. (1973) determined the beta branching ratios by means of a
185 proportional counter, and this intensity was corrected for internal conversion on the basis of
186 an independent gamma-ray measurement of the same source by means of NaI(Tl) detectors.
187 Finally, Merritt and Gibson (1978) have measured the absolute intensity of the 662-keV
188 gamma-ray emission from a ^{137}Cs standard based on an accurate knowledge of the response
189 function of a proportional counter. All of these measurements can be compared by calculating
190 the corresponding 662-keV gamma-ray emission probability from each of the measured beta
191 branching ratios by means of Equation (1). BrIcc software was used to calculate the total
192 internal conversion coefficient by adopting the frozen orbital approximation and assuming a

193 pure M4 transition (Kibédi et al., 2008). The calculated value of $\alpha_T = 0.1124$ (16) is consistent
194 with the available experimental measurements (as detailed in Leblond, 2023).

195 As noted in Table 3, the measurements of Hsue et al. (1966) and Gromov et al. (1978)
196 have been discarded from the adopted dataset, after considering the lack of characterization of
197 the radioactive sources in both studies that also raises questions concerning unaccounted
198 contamination by ^{134}Cs . Thus, by far the most precise of all the remaining acceptable studies
199 is the measurement performed by Behrens and Christmas (1983) that has impacted
200 significantly on the recommendations of previous evaluations. While adequate details are
201 given concerning the experimental set-up, spectrum analysis and assessment of the
202 uncertainty budget, doubts have arisen as to the overall quality of some of their resulting data.
203 The internal conversion coefficients of the 662-keV gamma transition as adopted by Behrens
204 and Christmas (1983) disagree significantly with existing literature values (as described in
205 greater detail by Leblond, 2023). As the same magnetic spectrometer was used to determine
206 the branching ratios and the internal conversion coefficients, the existence of an unexpected
207 experimental bias and/or the underestimation of the final uncertainty can be questioned.
208 Besides the doubts regarding the most precise measurement, the confusion identified with the
209 uncertainties within the study of Legrand et al. (1973) is also a cause for concern. Based upon
210 the calculation undertaken by the authors, the uncertainty was adjusted in the current analysis
211 from the 99% to 68% confidence level. However, this correction does not appear in previous
212 evaluations, which have seemingly misinterpreted the convention adopted by the French
213 group.

214 An unweighted average of $I_\gamma^{662} = 84.98$ (13) % and a weighted average of $I_\gamma^{662} =$
215 84.98 (12) % were calculated for the gamma-ray emission probability, after accepting or
216 adjusting the quoted uncertainties as noted in Table 3. The measurement of Behrens and
217 Christmas (1983) accounts for 65% of the total weighting average, and therefore this

218 uncertainty was increased to $\pm 0.20\%$ in order to limit this final weighting to no more than
 219 50%. After extending the overall uncertainty to the smallest experimental value in the
 220 accepted dataset, the recommended absolute emission probability and uncertainty of the 662-
 221 keV gamma ray was determined to be $I_{\gamma}^{662} = 85.01 (20) \%$. A balanced decay scheme was
 222 subsequently derived in which the beta intensities to the $11/2^-$ and $3/2^+$ states of ^{137}Ba were
 223 calculated by means of Equation 1.

224 Once the main emission intensities had been deduced, the transitions associated with
 225 the weakly populated $1/2^+$ excited state were considered. The beta branching ratio was
 226 determined from the following equation:

$$227 \quad I_{\beta}^{1/2^+} + I_{\gamma}^{378} (1 + \alpha_T^{378}) = I_{\gamma}^{283} (1 + \alpha_T^{283}) \quad (2)$$

228 where $I_{\beta}^{1/2^+}$ is the branching ratio to the $1/2^+$ state, I_{γ}^{283} is the gamma emission intensity from
 229 this state to the ground state and I_{γ}^{378} is gamma emission intensity from the $11/2^-$ state to the
 230 $1/2^+$ state. α_T^{283} and α_T^{378} are the total conversion coefficient associated with the
 231 corresponding gamma transitions as calculated by means of the BrIcc code. The 283-keV
 232 gamma-ray emission probability was determined to be the weighted average of the available
 233 measurements (Bikit et al., 1996; Wagner et al., 1997; Sergeev and Valiev, 2007) while the
 234 378-keV gamma-ray emission probability was adopted directly from the only known
 235 measurement (Moran et al., 2014). The complete decay scheme constructed in the manner
 236 described above is presented in Fig. 2.

237 The results obtained in this work are consistent and comparable to the previous
 238 evaluations, as listed in Table 1. Even though a complete re-analysis of the decay scheme has
 239 been performed, the uncertainties of the main emission probabilities could not be improved
 240 because of a lack of any new measurements. Nevertheless, the sum of all the intensities to the

241 ground state is effectively 100.0006 (2600) % in the recommended decay scheme. This small
242 imbalance arises inherently from the evaluation procedure, in which the decay scheme has
243 been reconstructed in two separate steps that are independent from each other. Alternative
244 methodologies lead either to the complete disappearance of all the transitions involving the
245 $1/2^+$ excited state, or to an unphysical constraint on the uncertainties of the main transition
246 probabilities. The recommended emission probabilities in this work are considered to be the
247 best compromise between the coherence and completeness of the decay scheme. If data users
248 require a fully balanced decay scheme, all the emissions identified with the $1/2^+$ excited state
249 of ^{137}Ba should be neglected.

250 **IV. Conclusions**

251 A re-analysis of the beta decay of ^{137}Cs has been performed within the collaborative
252 efforts of the DDEP in order to take account of recent relevant measurements along with
253 various improvements in some of the processing codes, most notably BetaShape (Mougeot,
254 2019) and BrIcc (Kibédi et al., 2008). New half-life measurements have helped in addressing
255 the long-known inconsistencies in the earlier studies. The DDEP recommendation half-life for
256 ^{137}Cs beta decay is thus updated to $T_{1/2} = 30.018 (22)$ years, with an uncertainty four times
257 lower than previous evaluations. However, doubts still remain regarding the low uncertainty
258 estimated by Martin and Taylor (1990) in their studies, which represents a large weighting
259 and domination of the averaging procedure. A number of further precise measurements are
260 required that would hopefully reduce the dependency of the evaluation on this specific work.
261 The analysis of existing known but unpublished half-life measurements mentioned by Pommé
262 et al. (2016) would be warmly welcomed at the beginning of a proposed international effort to
263 resolve this important issue of interlaboratory consistency.

264 A revision of the methodology used to evaluate the ^{137}Cs decay scheme has been
265 applied, leading to a re-evaluation of the main gamma-ray emission probability $I_{\gamma}^{662} =$
266 85.01 (20) %. This result is consistent with previous evaluations that also possess
267 comparable uncertainties. The evaluator plays an important role in the treatment of the
268 measured data to be found in Legrand et al. (1973) and Behrens and Christmas (1983). These
269 two sources of decay data are among the most precise of the datasets, but questions remain as
270 to the validity of their accuracy. The proposed inclusion, rejection and modification of their
271 uncertainties has a significant impact on the decay scheme formulated. Recommendations
272 proposed in this work still rely on the Behrens and Christmas (1983) measurement which
273 dominates the averaging procedure. Therefore, a series of new measurements of the beta
274 branching ratios and the main gamma-ray emission would provide valuable help to
275 reconstruct a more robust and unbiased decay scheme.

276 Finally, the β^{-} decay to the ground state of ^{137}Ba is a 2nd non-unique forbidden
277 transition, which is difficult to estimate from a theoretical perspective (Sadler and Behrens,
278 1993). Given the doubts concerning the most precise shape measurement performed by
279 Behrens and Christmas (1983), a new experimental determination of the shape would
280 therefore be of great interest and value.

281 The ^{137}Cs remains a prime example of a radionuclide with widespread applications
282 whose decay properties depend significantly upon a handful of measurements. Hopefully, the
283 realization of a strong need for more reliable and precise measurements will provide the
284 opportunity for evaluators to dispel the remaining lack of confidence surrounding the
285 complete decay scheme of this caesium radioisotope, more than 80 years after its discovery.

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291 **Declaration of interests**

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

294 The authors declare the following financial interests/personal relationships which may be
295 considered as potential competing interests:

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