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Nuclear data for day-1 radionuclides

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Abbreviations, Participant short names

Abbreviations

NMI	National Metrology Institute
NDS	Nuclear Data Sheets
DDEP	Decay Data Evaluation Project
HPGe	High Purity Germanium
PET	Positron Emission Tomography
SPECT	Single Photon Emission Computer Tomography
TAGS	Total Absorption Gamma-ray Spectroscopy
TDPAC	Time Differential Perturbed Angular Correlation

Participant short names

CERN	European organization for nuclear research
NPL	National Physical Laboratory
PSI	Paul Scherrer Institut
CEA	Commissariat à l'énergie atomique et aux énergies alternatives
IST-ID	Associação do Instituto Superior Técnico para a IST-ID Investigação e Desenvolvimento
DTU	Danmarks Tekniske Universitet
CHUV	Centre hospitalier universitaire vaudois
GANIL	Grand Accélérateur National d'Ions Lourds
SCK CEN	Studiecentrum voor Kernenergie / Centre d'étude de l'énergie nucléaire
ARRONAX	Groupement d'intérêt public ARRONAX
ESS	European spallation source ERIC
TUM	Klinikum rechts der Isar der technischen Universität München
KULeuven	Katholieke Universiteit Leuven
MedAustron	Entwicklungs- und Betriebsgesellschaft MedAustron GmbH
SCIPROM	SCIPROM Sàrl
MUI	Medizinische Universität Innsbruck
ILL	Institut Max von Laue - Paul Langevin
JRC	JRC -Joint Research Centre- European Commission
NCBJ	Narodowe Centrum Badań Jądrowych
GSI	GSI Helmholtzzentrum für Schwerionenforschung GmbH
LU	Latvijas Universitāte

INFN | Istituto Nazionale di Fisica Nucleare
UiO | Universitetet i Oslo

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Summary

Nuclear decay data are fundamental constants of a radionuclide's decay process and are unique to that radionuclide. Nuclear decay data have been identified as being of importance to a wide range of activities in nuclear medicine, from the production of radionuclides to their use in nuclear medicine clinics. Accurate and precise data on nuclear decay is therefore necessary to ensure confidence in all activities undertaken throughout the process of using radiopharmaceutical products.

This report provides a review of the status of the nuclear decay data of the diagnostic or therapeutic radionuclides in the PRISMAP catalogue available at the start of the project in May 2021. There were eighteen original radionuclides available at the commencement of the project, which were identified as Sc-44, Sc-47, Cu-64, Cu-67, Ag-111, La-135, Tb-149, Tb-152, Tb-155, Tb-161, Er-165, Ho-166, Er-169, Yb-175, Pt-195m, Bi-213, At-211 and Ac-225. This review does not include a review of the radionuclides Dy-166 and Tm-165 that are used as generators to produce Ho-166 and Er-165 and has focused on these progenies instead. Whilst PRISMAP has extended their radionuclide catalogue since the initiation of the project to now encompass a total of twenty-six radionuclides at the time of publishing this report, these additional radionuclides have not been covered by this review. These may be included in a future review.

A summary of the current state of nuclear decay data of the initial eighteen radionuclides have been covered using the latest evaluations published by the Nuclear Data Sheets or the Decay Data Evaluation Project. Where recent studies have been published since the last evaluation a comparison to these new values have been included. Based on these reviews' recommendations, where the current literature is lacking or there is room for improvement, new nuclear decay data studies are needed or have been proposed.

1. Introduction

PRISMAP has brought together European nuclear infrastructures with the aim of providing a sustainable source of high purity grade radionuclides for medical research applications. The PRISMAP consortium offers a wide array of radionuclides which have been identified for potential use for either diagnostic or therapeutic applications in nuclear medicine, with 18 such radionuclides available from the initiation of the project and two further radionuclides used to generate medical radionuclides (Table 1). The PRISMAP collaboration has used state-of-the-art production technologies (such as mass-separation) to provide access to these radionuclides that could previously not be produced in the purity that would be required due to the co-production of the radioisotopes of the same element. These radionuclides are being made available to researchers to accelerate R&D for the diagnosis and treatment of cancer by the use molecular radiotherapy.

Table 1. PRISMAP day-1 radionuclides.

Radionuclide	Application	Imaging(I)/ Treatment(T)/ Generator(G)	Production reaction
Sc-44/Sc-44m	PET	I	$^{44}\text{Ca}(p,n); ^{44}\text{Ca}(d,2n)$
Sc-47	β^- therapy, SPECT	I/T	$^{46}\text{Ca}(n,\gamma)^{47}\text{Ca}(\beta^-)$
Cu-64	PET	I	$^{64}\text{Ni}(p,n); ^{64}\text{Ni}(d,2n)$
Cu-67	β^- therapy, SPECT	I/T	$^{68}\text{Zn}(p,2p); ^{70}\text{Zn}(p,\alpha)$
Ag-111	β^- therapy, SPECT, TDPAC	I/T	$^{110}\text{Pd}(n,\gamma)^{111}\text{Pd}((\beta^-); ^{110}\text{Pd}(d,n)$
La-135	Auger therapy	T	$^{\text{nat}}\text{Ba}(p,X)$
Tb-149	α therapy, PET	I/T	$^{\text{nat}}\text{Ta}(p,\text{spall})$
Tb-152	PET	I	$^{\text{nat}}\text{Ta}(p,\text{spall})$
Tb-155	Auger therapy, SPECT	I	$^{\text{nat}}\text{Ta}(p,\text{spall})$
Tb-161	β^- therapy, SPECT	I/T	$^{160}\text{Gd}(n,\gamma)$
Dy-166	Generator for Ho-166 (β^- therapy, SPECT)	G	$^{164}\text{Dy}(n,\gamma)(n,\gamma)$
Er-165	Auger emitter	T	$^{165}\text{Ho}(n,\gamma)$
Tm-165	Generator for Er-165 (Auger therapy)	G	$^{\text{nat}}\text{Ta}(p,\text{spall})$
Er-169	β^- therapy	T	$^{168}\text{Er}(n,\gamma)$
Yb-175	β^- therapy, (SPECT)	T	$^{174}\text{Yb}(n,\gamma)$
Pt-195m	Auger therapy, SPECT	I/T	$^{194}\text{Pt}(n,\gamma)$
Bi-213	α therapy	T	^{225}Ac generator
At-211	α therapy	T	$^{209}\text{Bi}(\alpha,2n)$
Ac-225	α therapy	T	^{229}Th generator; $^{232}\text{Th}(p,\text{spall})$

Nuclear decay data are fundamental constants of radioactive decay and whilst their study is of use to better understand the nature of radioactive decay and standard models, the data are also a critical component for the use of said radionuclides in the real world. Certain nuclear decay data, such as the radioactive half-life, decay branching ratios and gamma-ray emission intensities are used across many aspects of nuclear medicine, such as determining the activity, quantitative imaging, quality control and release of radiopharmaceutical products, and for dosimetry modelling. Therefore, having accurate and precise data for these radionuclides is of importance to the diagnosis and treatment of cancer.

This report provides a review of these radionuclides to inform on the state of the nuclear decay data and inform on where there is a need for new investigations (excluding the ones being used as generators/parent radionuclides). The substantial review and recommendations by Nicholls published in 2022 provides a significant amount of detail on the state of many of these radionuclides and these recommendations have been incorporated into this review document (Nicholls, 2022). The evaluated values are primarily taken from the evaluations published in the NDS or in some cases in the DDEP depending on when the evaluations have been performed. The most recent evaluation has been typically taken.

2. Nuclear decay data review of day-1 radionuclides

2.1 Scandium-44

A new evaluation of Sc-44 was published in 2023, a summary of the evaluated nuclear decay data is provided in Table 2 (Chen and Singh, 2023). Scandium-44 decays via either β^+ or ε to Ca-44, predominantly via β^+ ($I_{\beta^+} = 94.278(11)$) to Ca-44. This decay ratio between the decay routes appears to be well known, with the I_{β^+} determined from the weighted average of two measurements of the ratio of ε/β^+ by anti-coincidence and coincidence counting (Baerg, 1983; Stocker and Baerg, 1976). It should be noted that these determinations have been performed at the same laboratory and an independent measurement may be useful to confirm this value. However, a separate measurement of the annihilation radiation intensity provides a less precise confirmation of the decay ratio (Schötzig, 1990).

The most recent evaluated half-life of Sc-44 of 4.0420(25) h is from a recent study in 2016 (García-Toraño et al., 2016). A further study in 2022, whilst noted in the evaluation has not been used to derive the half-life, with a value of 4.042(7) h provides evidence that the half-life is approximately 1.8 % longer than was previously reported (Chen et al., 2011). Further half-life studies may be beneficial to improve the currently available dataset, though it is unlikely that any further significant improvement to the precision will be found.

The main gamma-ray emission of the 1157.020(15) keV is suitably precise ($I_\gamma = 99.90(42)$ %) and the decay scheme is well defined. There is one uncertain gamma ray from the 3307.9 keV to 1157.039 keV excited states, having been only reported by one study. As the intensity of this gamma ray is insignificant for any dose calculations there is no imperative to confirm this beyond curiosity.

Table 2. Nuclear decay data of Sc-44 (Chen and Singh, 2023).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle Emissions	Principal gamma-ray emission
4.0420(25)	3652.5(18)	94.278(11) % β^+ 5.723(19) % ε	$E_{\beta^+,end} = 1473.7(18)$ keV	$E_\gamma = 1057.022(15)$ keV ($I_\gamma = 99.90(42)$ %)

2.2 Scandium-47

Scandium-47 undergoes β^- decays to the ground state of Ti-47. It has a single excited state which is populated 68.4 % of decays with the remainder decaying directly to the ground state. The nuclear decay data for this radionuclide appears to be complete with precise determinations. The recommended nuclear decay data for Sc-47 is almost exclusively based on a single high-precision study by two National Metrology Institutes and a European Commission's Joint Research Centre (Reher et al., 1986). This single study determined a precise half-life from six determinations by three independent techniques: NaI(Tl) integral counting, HPGe gamma-ray spectrometry, and Ionisation chamber measurements. These six determinations are all in agreement within their standard uncertainties, which does provide confidence in the result. Whilst there may be benefit to revisiting this half-life to confirm the value and using the updated uncertainty evaluation techniques it would be unlikely that any improvement in the precision could be found.

This study along with another in the 1950s using a beta spectrometer provide precise data for the beta decay endpoint energies of 439.0(13) keV and 600.3(14) keV. These beta decay branching ratios are taken from the Reher et al. (Reher et al., 1986) study, based on beta spectrometry measurements and the absolute gamma-ray intensities with relative uncertainties of 1-2 %. For the purposes of dose calculations, the precision on these values is likely adequate.

The emission intensity of the single gamma ray of 159.381 keV comes from Reher et al. (Reher et al., 1986) based on a weighted mean of two HPGe gamma-ray spectrometry determinations (67.8(5) % and 67.1(8) %) and two by NaI(Tl) spectrometry (68.7(4) % and 68.9(7) %). It is noted that there is a bias between the two detector types with an approximate relative difference between their arithmetic means of 2 %. It may be of interest to revisit the absolute gamma-ray emission intensity to resolve this bias.

A summary of the evaluated nuclear decay data is provided in Table 3.

Table 3. Nuclear decay data of Sc-47 (Nica, 2009).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
3.3492(6)	600.3(19)	100 % β^-	$E_{\beta^-,end} = 600.3(14)$ keV ($I_{\beta} = 31.6(6)$ %)	$E_{\gamma} = 159.4$ keV ($I_{\gamma} = 68.3(4)$ %)

2.3 Copper-64

The nuclear decay data for Cu-64 has undergone a recent evaluation in 2021 and has been published in the NDS and incorporates new studies that have been performed by many NMIs. A summary of the evaluated nuclear decay data is provided in Table 4 (Chen and Singh, 2021). Copper-64 decays through electron capture (44.03(25) %) or β^+ (17.49(51) %) to ^{64}Ni or β^- (38.48(26) %) to ^{64}Zn . The accuracy of the β^- value has been questioned by Bergeron et al. (Bergeron et al., 2018) due to discrepancies between techniques used to determine the activity. They concluded that the branching ratios would be worthy of further scrutiny. The decay scheme is well-defined.

The intensity of the 1345.77 keV gamma-ray emission has been determined through multiple studies and has been determined with satisfactory precision (Bé et al., 2012; Luca et al., 2012; Pibida et al., 2017; Wanke et al., 2010; Yamazaki et al., 2018).

The half-life of Cu-64 has been measured extensively with over nineteen measurements by multiple techniques. Many of these have been performed in the last decade by NMIs. The weighted mean of these determinations has resulted in a half-life of 12.7006(20) h, with a relative uncertainty of 0.016 %.

Table 4. Nuclear decay data of Cu-64 (Singh and Chen, 2021).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
12.7006(20)	$Q(\epsilon) = 1674.62(21)$	61.52(30) % $\epsilon + \beta^+$	$E_{\beta^+,end} = 652.6(21)$ keV ($I_{\beta^+} = 17.49(15)$ %)	$E_{\gamma} = 1345.77$ keV ($I_{\gamma} = 0.472(4)$ %)
	$Q(\beta^-) = 579.6(6)$	38.48(26) % β^-	$E_{\beta^-,end} = 579.6(6)$ keV ($I_{\beta^-} = 38.48(26)$ %)	

2.4 Copper-67

The nuclear decay data for Cu-67 was most recently evaluated in 2005, A summary of the evaluated nuclear decay data is provided in Table 5 (Junde et al., 2005). Since then, a new study has proposed significant changes to the decay scheme (Chen et al., 2015).

Copper-67 decays by 100 % β^- emission to Zn-67, with a calculated Q-value of 561.7(15) which is in agreement with the measured value of 560.3(10) keV determined by (Chen et al., 2015).

A half-life of 61.83(12) h was derived from four determinations made in the 1960s and 1970s. Whilst these values are mostly in agreement, it would be beneficial to confirm the half-life and potentially improve the precision.

In the evaluated nuclear decay data, the intensity of the beta decay branches has been derived from the gamma-ray emission intensity balance. However, these gamma-ray intensities have only been taken from a single study from the 1970s (Meyer et al., 1978). In the more recent study, absolute gamma-ray emission intensities show significant differences from the previously reported values. These have been used to determine the beta branching ratios, which provide significant changes to these values. Considering the paucity of data for this radionuclide it would be of benefit for additional studies to corroborate the newest values and to improve their precision.

Table 5. Nuclear decay data of Cu-67 (Junde et al., 2005).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
61.83(12)	561.7(15)	100 % β^-	$E_{\beta^-,end} = 561.7(15)$ keV ($I_{\beta} \approx 20\%$)	$E_{\gamma} = 184.577$ keV ($I_{\gamma} = 48.7(3)$ %)

2.5 Silver-111

Silver-111 decays through 100 % β^- emission to Cd-111, with approximately 92% direct to the ground state, resulting in low intensities for the gamma-ray emissions. The last evaluation of this radionuclide was performed in 2009 and published in NDS, a summary of the evaluated nuclear decay data is provided in Table 3 (Blachot, 2009), since which new studies have been published that provide data that can significantly improve the nuclear decay data.

The evaluated half-life of Ag-111 is 7.45(1) d, determined from two studies performed in 1960 and 1974 (Baerg et al., 1960; Rothman et al., 1974). Two new studies in 2015 and 2023 by HPGe gamma-ray spectrometry have determined half-lives which are shorter by approximately 0.4% than previously reported (Collins et al., 2016; Dong et al., 2023a).

The evaluated beta decay branches are not currently known with a high precision, with a relative standard uncertainty of 5.4 % on the decay direct to the ground state. These have been deduced from the intensity balance using the gamma-ray intensities, which themselves are known with poor precision. New studies in 2013 and 2015 (Collins et al., 2014; Krane, 2015) have determined gamma-ray emission intensities with greater precision than previously available and confirmed or disproved the existence of certain gamma rays. These works also showed significant discrepancies with the values in the evaluations with 10% deviations of the 96.75 keV and 245.40 keV intensities. In the Krane study, new beta-branching ratios were derived from the gamma-ray intensity balance which, whilst in agreement with the evaluated values, provide greater precision.

Table 6. Nuclear decay data of Ag-111 (Blachot, 2009).

$T_{1/2}$	Q-value	Decay mode(s)	Principal particle	Principal gamma-ray
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$T_{1/2}$ /d	Q-value /keV	Decay mode(s)	emissions	emission
7.45(1)	1036.8(14)	100 % β^-	$E_{\beta^-,end} = 1035(2)$ keV ($I_{\beta} = 92(5)$ %)	$E_{\gamma} = 342.13$ keV ($I_{\gamma} = 6.68(33)$ %)

2.6 Lanthanum-135

Lanthanum-135 decays by almost 100% electron capture to the daughter nucleus of Ba-135. The decay scheme has primarily been derived from one study in 1971 (Bazan and Meyer, 1971). Though it is relatively well defined there are some uncertain excited states at higher energies and an uncertain gamma transition placement. The electron capture decay branching ratio to these excited states is small (< 0.0004 %).

The current evaluated half-life of ^{135}La , from 2008, is 19.5(2) h (Singh et al., 2008). This has been derived from the weighted mean of two studies, though there were a further four studies that were omitted from the evaluation. Since this evaluation there has been a new study of the half-life that has determined a half-life of 18.91(2) h (Abel et al., 2018). This is a significant change to the evaluated half-life and is worth corroborating with further studies.

30 gamma transitions that have been identified from different studies, though only 25 of these have been placed. As La-135 goes directly to the ground state of Ba-135 in 98.1 % of the decays the gamma emission intensities are small, with the principal 480.51 keV gamma ray having an intensity of 1.52 %. Due to a scarcity of studies of the gamma-ray emission intensities and the absolute intensity being based on the 98.1 % electron capture decay to the ground state, the precision of the absolute gamma-ray emission intensities is poor with a minimum standard uncertainty for an activity determination by gamma-ray spectrometry of 15.8 %. This is due to the 15.8 % uncertainty attributed to the normalisation scaling factor which is correlated across all the gamma rays and cannot be reduced by the use of multiple gamma rays to determine an activity. There is clearly a need here for the determination of the absolute intensity directly from an absolute activity standard derived by primary standardisation at an NMI coupled with HPGe gamma-ray spectrometry measurements using a well-defined efficiency calibration, which could potentially reduce the uncertainty by a factor of 30.

As stated in Nichols (Nichols, 2022) there are also requirements for improved X-ray and Auger-electron data studies.

A summary of the evaluated nuclear decay data is provided in Table 7.

Table 7. Nuclear decay data of La-135 (Singh et al., 2008).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
19.5(2)	1200(10)	100 % $\epsilon + \beta^+$	-	$E_{\gamma} = 480.51$ keV ($I_{\gamma} = 1.52(24)$ %)

2.7 Terbium-149

Terbium-149 is a special radionuclide as it undergoes decay by either alpha-particle decay to the ground state of Eu-145 ($I_{\alpha} = 16.7(14)$ %) or by $\epsilon + \beta^+$ decay to the daughter nucleus of Gd-149 ($I_{\epsilon + \beta^+} = 83.3(17)$ %), with a β^+ emission being emitted in 7.1(3) % of all decays (Singh and Chen, 2022). Both Eu-145 ($T_{1/2} = 5.93(4)$ d) and Gd-149 ($T_{1/2} = 9.28(10)$ d) decay by electron capture to Sm-145 ($T_{1/2} = 340(3)$ d) and Eu-149 ($T_{1/2} = 93.1(4)$ d) respectively. As the decay progeny of Tb-149 have longer half-lives they can never achieve equilibrium and are in essence considered long-lived contaminants, though due to their half-lives being significantly longer and the decay branches, their activities will only be a small fraction of the starting activity of Tb-149. It is therefore useful to consider these decay progenies when considering the nuclear decay data of Tb-149.

The half-life of Tb-149 has been derived from a weighted mean of six determinations, with a relative standard uncertainty of 0.61 %. These determinations have all been made before 1970. There is a clear need for further studies to improve the precision of this half-life. The half-lives of the direct progeny of Eu-145 and Gd-149 have been derived from a small number of studies made over 40 years ago and have relative uncertainties of 0.67 % and 1.1 % respectively. These would also benefit from new precision studies to improve these values as these would be of importance for determining a more accurate alpha decay branching ratio.

Overall, whilst the alpha decay mode is simple the nuclear decay data relies mainly on a single study performed in 1972 (Vylov et al., 1972), though there are several additional studies performed in the 1960s. The alpha decay pathway to Eu-145 is dominated by decay straight to the ground state, with a small decay branch to the 329.5 keV excited state ($I_{\alpha} = 0.0050(17) \%$). There are no observed gamma transitions from this excited state to the ground state. The alpha decay branching ratio has a relative uncertainty of 8.4 %, which would benefit from further studies to improve the precision since it is of importance for dosimetry and for activity measurements.

The electron capture/positron decay branch to Gd-149 is far more complex than the alpha decay branch with over 70 excited states and over 300 gamma transitions. Nine of the gamma transitions placements are in doubt and one remains unplaced. The gamma-ray emission intensities have been derived from only one study (Jackson et al., 1978). Therefore, it would be beneficial to determine the intensities to confirm the accuracy of the values and potentially improve the precision.

The gamma-ray emission intensities of the gamma rays emitted following the decay of Eu-145 and Gd-149 have significant uncertainties. The principal gamma ray following the decay of each radionuclide has standard uncertainties of 6.3 % for the 893.73 keV gamma ray of Sm-145 and 6.3 % for the 149.73 keV gamma ray from Eu-149. It would be of use to improve the precision of these gamma-ray intensities for the determination of an improved alpha branching ratio for Tb-149.

A summary of the evaluated nuclear decay data is provided in Table 8.

Table 8. Nuclear decay data of Tb-149 (Singh and Chen, 2022).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
4.12(3)	$\alpha = 4077.5(22)$	16.7(14) % α	$E_{\alpha} = 3967(3) \text{ keV}$ ($I_{\alpha} = 16.7(14) \%$)	
	$\varepsilon + \beta^{+} = 3639(4)$	83.3(17) % $\varepsilon + \beta^{+}$	$E_{\beta^{+}, \text{end}} = 1411(4) \text{ keV}$ ($I_{\beta} = 3.95(11) \%$)	$E_{\gamma} = 352.24(2) \text{ keV}$ ($I_{\gamma} = 29.83(64) \%$)

2.8 Terbium-152

The decay scheme of Tb-152 is complex, decaying by either electron capture ($I_{\varepsilon} = 79.7(15) \%$) or positron emission ($I_{\beta^{+}} = 20.3(15) \%$) to Gd-152, with 111 proposed excited states in the daughter nuclei giving rise to 387 possible gamma transitions. Many of these proposed gamma transitions are unplaced with the possibility that some excited states have not been observed due to the small electron capture branching to these states and the high energy of the gamma ray making detection challenging by high-resolution gamma-ray spectrometry alone. This radionuclide has been identified as a suitable candidate for TAGS (Nichols, 2022). The positron branching ratio is also not well known, with a relative uncertainty of 7.6 % and would benefit from further measurements to determine this with greater precision.

The half-life of Tb-152 has been determined in two studies performed in the 1960s and currently has a large relative standard uncertainty of 0.57 %. There is a need for further determinations of the half-life to improve the precision.

The gamma-ray emission intensities of Tb-152 have not been extensively measured though they have been reported with reasonable precision, with a relative uncertainty of 2.7 % for the main gamma ray of 344.2785 keV. Further studies would be worth to confirm the accuracy of these emission intensities.

A summary of the evaluated nuclear decay data is provided in Table 9.

Table 9. Nuclear decay data of Tb-152 (Martin, 2013).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
17.5(1)	3990(40)	100 % $\varepsilon+\beta^+$	$E_{\beta^+,end} = 3990(4)$ keV ($I_{\beta} = 8.0(13)$ %)	$E_{\gamma} = 344.2785(13)$ keV ($I_{\gamma} = 63.5(17)$ %)

2.9 Terbium-155

Terbium-155 undergoes electron capture decay to Gd-155, to one of 22 excited states of the daughter nucleus and with over 100 gamma transitions from the de-excitation of these states. The nuclear decay data was recently evaluated in 2019, though primarily the data for the decay scheme comes from one study from 1976 with some additional data taken from a study in 1986 (Meyer et al., 1976; Schmidt et al., 1986). Whilst the decay scheme is mostly complete there are nine uncertain placements and 31 unplaced for the gamma transitions. Further high-resolution gamma-gamma measurements would be useful to resolve these placements.

The gamma-ray emission intensities are not well known, with large relative standard uncertainties of more than 5 %. The electron capture branching ratios to the excited states have been calculated from the gamma intensity balance; due to the poor precision, the uncertainties of these branching ratios are poor. High-precision measurements of these intensities are needed to improve the precision, ideally absolute gamma-ray emission intensities via an absolute activity standard.

The half-life of 5.32(6) d has been taken from a single study of the half-life published in 1970. A more recent study published in 2022 has determined a value of 5.2346(36) d, which is a relative difference of 1.6 % to the current evaluated value (Collins et al., 2022b). The precision quoted is also approximately an order of magnitude more precise. With only two half-life determinations and the large difference between their values and precision, it is considered worthwhile having further independent measurements.

A summary of the evaluated nuclear decay data is provided in Table 10.

Table 10. Nuclear decay data of Tb-155 (Nica, 2019).

$T_{1/2}$ /d	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma ray emission
5.32(6)	820(10)	100 % ε	-	$E_{\gamma} = 86.55(3)$ keV ($I_{\gamma} = 32.0(17)$ %)

2.10 Terbium-161

Terbium-161 decays via β^- emission to Dy-161. The decay scheme is well defined with 11 excited states and 37 gamma transitions, though four of those are uncertain as they have not been observed directly but are known from the ^{161}Ho electron capture decay. The intensity of the beta branches has been derived from the gamma intensity balance. However due to the poor precision ($u(I_{\gamma}) > 5$ %) of the gamma-ray emission intensities the standard uncertainties of the branching ratios have larger uncertainties. The evaluated gamma-ray emission intensities in NDS come from five studies published between 1965 and 1985 (Reich, 2011). A new determination of the gamma-ray emission intensities has since been published in 2021, which

as they have been derived from an absolute activity standard provide improved precision and suggest significant differences to many of the intensities (Juget et al., 2021). However, this work did not manage to determine an improved intensity for the 25.65 keV gamma ray which is required to determine the beta branching ratio to the ground state and the 25.65 keV excited state. Both states have a 100 % relative standard uncertainty.

The half-life of 6.89(2) d has been determined from the weighted mean of ten determinations from 1949 to 1989. Whilst these studies appear to be consistent and provide a relatively good precision three recently published values have questioned this half-life (Collins et al., 2022a; Dong et al., 2023b; Durán et al., 2020). These new determinations indicate a half-life that is approximately 1 % longer than previously considered, with a half-life of 6.9585(39) d, derived from the weighted mean of these three determinations. The precision from these new half-life values is far greater than the current evaluated half-life.

There are also additional requirements for X-ray and internal-conversion electron data. Tb-161 is also a significant Auger-electron emitter, there is also a need for measurement of this data as well.

A summary of the evaluated nuclear decay data is provided in Table 11.

Table 11. Nuclear decay data of Tb-161 (Reich, 2011).

$T_{1/2}$ /d	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
6.89(2)	593.0(13)	100 % β^-	$E_{\beta^-,end} = 522(-)$ keV ($I_{\beta} = 64(4)$ %)	$E_{\gamma} = 74.56669(6)$ keV ($I_{\gamma} = 10.20(54)$ %)

2.11 Holmium-166

Holmium-166 undergoes β^- decay to Er-166, with beta decay branches mainly to the ground state ($I_{\beta} = 48.8(12)$ %) or the 80.5775 keV excited state ($I_{\beta} = 49.9(12)$ %). These branching ratios have been determined from the gamma-ray intensity balance rather than from the values in the literature from 1955 to 1976 (Baglin, 2008a). The decay scheme for the ^{166}Ho decay is well defined with all gamma transitions placed with good certainty.

The limiting factor for the precision of the beta branching ratios is the gamma-ray emission intensities, with a relative standard uncertainty of 1.9 % on the 80.576 keV ($I_{\gamma} = 6.56(13)$ %) (Baglin, 2008a). The gamma-ray intensities for Ho-166 have been derived from nine studies published from 1962 to 1992. A more recent study by three NMIs published in 2019 has used absolute activity standards to determine the X-ray and gamma-ray emission intensities with a higher precision than from the previous studies (Bobin et al., 2019). For the 80.576 keV, the intensities determined were 6.61(7) %, 6.636(49) % and 6.618(51) % which are consistent with the current value but higher by approximately 0.8 %.

The half-life of 26.824(12) h for Ho-166 has been determined from the weighted mean of six studies, though in reality the one study far outweighs the others and dominates the half-life (Baglin, 2008a). This one dominant study comes from a paper where the standard uncertainty of the half-life is based purely on the least-squares fit, which is likely to have resulted in an underestimation of the uncertainty (Abzouzi et al., 1989). In the study by the three NMIs mentioned above, the half-life was also measured by each of the laboratories and provided a further five determinations, these values were all in agreement with the current half-life though systematically lower, with a half-life of 26.809(10) h derived from a weighted mean of the five determinations (Bobin et al., 2019).

A summary of the evaluated nuclear decay data is provided in Table 12.

Table 12. Nuclear decay data of Ho-166 (Baglin, 2008a).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
26.824(12)	1854.7(9)	100 % β^-	$E_{\beta^-,end} = 1773.1(14)$ keV ($I_{\beta} = 49.9(12)$ %)	$E_{\gamma} = 80.576(2)$ keV ($I_{\gamma} = 6.56(13)$ %)

2.12 Erbium-165

Erbium-165 undergoes a simple electron capture decay to the ground state of Ho-165 with no gamma-ray emissions. From the resulting electron capture, X-rays and Auger-electrons will be emitted. All of the studies for the nuclear and atomic data for this radionuclide have occurred primarily in the 1950s and 1960s, with the last significant study of the atomic data in 1980 (Gnade et al., 1980). These studies of the atomic data have provided the required data for the calculation of the Auger electron emission intensities though studies would be desirable to directly measure the Auger electron intensities to confirm the accuracy.

There has been a total of 13 publications containing a half-life determination for Er-165; these have all been published in the 1960s and 1950s. In the evaluation published in NDS in 2006 (Jain et al., 2006), the half-life was derived from the weighted mean of four of these studies. This provided a half-life of 10.36(4) h. It would be beneficial for a new determination of the half-life to take advantage of the advances in half-life metrology and to confirm the accuracy of this value.

A summary of the evaluated nuclear decay data is provided in Table 13.

Table 13. Nuclear decay data of Er-165 (Jain et al., 2006).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
10.36(4)	376.3(20)	100 % ϵ	-	-

2.13 Erbium-169

The evaluation of Er-169 has most recently been performed by the DDEP and has been used to review the nuclear decay data (Bé et al., 2016). Erbium-169 decays by 100% β^- to Tm-169 with a simple and well-defined decay scheme, with only two excited states. The β^- decay branches are dominated by a 56(5) % population of the ground state and a 44(5) % population of the 8.4102(1) keV excited state. There have only been two experimental studies of the beta transition to the 8.4 keV level, which results in the poor precision of the probabilities. Due to a high internal conversion factor for the 8.41017 keV gamma transition there are only low-intensity gamma-ray emissions. Furthermore, more detailed K X-ray and internal conversion electron data for the direct population of the 8.41017 keV state and the transition to the ground state have been recommended (Nichols, 2022). The Auger electron emissions have been calculated using various atomic data sources, with 203.3 electrons per 100 decays.

A recent study has determined the gamma-ray emission intensities and the K X-rays with far greater precision than available in the DDEP evaluation (Talip et al., 2021). For the 109.78 keV and 118.19 keV gamma rays, a relative decrease of approximately 30 % to the emission intensity was observed along with an order of magnitude improvement in the precision, from approximately 20% to 1-3%. However, as noted in the publication the intensities determined are only approximately 8 % greater than those from the NDS evaluation (Baglin, 2008b). For the K X-rays, the emission intensities were found to be an order of magnitude greater than previously calculated, with standard uncertainties of approximately 2 %. Considering such a significant difference it would be beneficial for confirmatory studies to be performed.

There have been seven publications of the half-life of Er-169 (which include an uncertainty). The half-life of 9.38(2) d has been derived from a simple mean of two of these studies (Myers, 1977; Schrader, 2004) as the use of the full dataset resulted in a discrepant dataset. Further measurements would be beneficial to improve the half-life. It should be noted that the evaluation in NDS used a weighted mean of the same two studies which resulted in a slightly more precise half-life value (Baglin, 2008b).

A summary of the evaluated nuclear decay data is provided in Table 14.

Table 14. Nuclear decay data of Er-169 (Bé et al., 2016).

$T_{1/2}$ /d	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
9.38(2)	353.0(12)	100 % β^-	$E_{\beta^-,end} = 353.0(12)$ keV ($I_{\beta} = 56(5)$ %)	$E_{\gamma} = 109.77930(14)$ keV ($I_{\gamma} = 0.0045(9)$ %)

2.14 Ytterbium-175

Ytterbium-175 decays to the ground state of Lu-175 via 100 % β^- emission, primarily directly to the ground state ($I_{\beta} = 72.9(5)$ %) with smaller beta transitions to three excited states. The de-excitation of these excited states can result in six gamma transitions, with the most intense gamma rays coming from the 396.328 keV state ($I_{\gamma}(396.329 \text{ keV}) = 13.15(25)$ %; $I_{\gamma}(282.522 \text{ keV}) = 6.128(84)$ %). A further 'strong' transition arises from the 113.805 keV state to the ground state ($I_{\gamma}(113.805 \text{ keV}) = 3.866(47)$ %) which is dominated by internal conversion electron emission (Basunia, 2004). The decay scheme is well-defined.

There have been four studies published on the gamma-ray emission intensities from 1966 to 1994, which have resulted in seemingly well-known intensities for all the main gamma-ray emissions. This has resulted in a good understanding of the beta branching ratios.

Until recently, the half-life of Yb-175 was reliant on a single value with a high precision quoted for the half-life (Abzouzi et al., 1989). This provided a half-life of 4.185(1) d. However, a recent study has called that study into question. This new study determined the half-life using three different detector systems which provided a consistent half-life that was significantly lower than previously published (Durán et al., 2021). A new half-life of 4.1615(39) d was determined, which is 0.56 % lower than Abzouzi et al. (1989).

A summary of the evaluated nuclear decay data is provided in Table 15.

Table 15. Nuclear decay data of Yb-175 (Basunia, 2004).

$T_{1/2}$ /d	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma ray emission
4.185(1)	470.1(13)	100 % β^-	$E_{\beta^-,end} = 470.1(13)$ keV ($I_{\beta} = 72.9(5)$ %)	$E_{\gamma} = 396.329(20)$ keV ($I_{\gamma} = 13.15(25)$ %)

2.15 Platinum-195m

The potential therapeutic Pt-195m decays by 100 % IT to stable Pt-195. The decay scheme is well defined with five excited states depopulated by nine gamma transitions, with the most intense gamma ray coming from the 98.90 keV state ($I_{\gamma} = 11.70(84)$ %). Whilst the decay is well defined, the gamma-ray emission intensities themselves have a poor precision (> 7 %) and appear to only come from one study in 1972, though this comes from a private communication. Further measurements are absolutely required, preferably with the absolute intensities derived from an absolute activity standard.

From the decay of Pt-195m, there is a high probability of internal conversion electrons arising from the gamma transitions from the 259.29 keV state through the 129.5 keV transition ($\alpha_T = 1135(19)$) and from the 129.79 keV state through the 30.89 keV transition ($\alpha_T = 37.6(7)$). It has been recommended that further data for the internal conversion electron probabilities would be beneficial (Nichols, 2022).

The half-life of 4.0104(47) d has been taken from one study from samples produced by photoactivation and measured by HPGe gamma-ray spectrometry (Mohr et al., 2000). New studies of the half-life using high-purity samples from PRISMAP would be beneficial to confirm this half-life.

A summary of the evaluated nuclear decay data is provided in Table 16.

Table 16. Nuclear decay data of Pt-195m (Huang and Kang, 2014).

$T_{1/2}$ /d	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
4.0104(47)	-	100 % IT	-	$E_\gamma = 98.90(2)$ keV ($I_\gamma = 11.70(84)$ %)

2.16 Astatine-211

Astatine-211 decays by either electron capture ($I_\epsilon = 58.20(8)$ %) to Po-211 or α decay ($I_\alpha = 41.80(8)$ %) to Bi-207. There are six weak gamma transitions originating from either decay route as the decay branches are dominated by transitions direct to the ground state.

The half-life of 7.214(7) h derived in the most recent evaluation in NDS has been taken from one study in 1961, with a further four studies identified from 1956 to 1978 but not used (Singh et al., 2013). The main study has derived the half-life from the weighted mean of nine determinations using the uncertainty of the fit as the weight (Appelman, 1961). The uncertainty of this half-life does not account for any systematic effects and further studies are recommended to confirm the half-life.

A summary of the evaluated nuclear decay data is provided in Table 17.

Table 17. Nuclear decay data of At-211 (Singh et al., 2013).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
7.214(7)	$Q_\alpha = 5982.4(13)$	41.80(8) % α	$E_\alpha = 5869.5(22)$ keV ($I_\alpha = 41.80(8)$ %)	-
	$Q_\epsilon = 785.3(25)$	58.20(8) % ϵ	-	

2.17 Bismuth-213

A re-evaluation of the nuclear decay data has recently taken place including new studies for the half-life and absolute gamma-ray emission intensities (Basunia, 2022). A summary of the evaluated nuclear decay data is provided in Table 18. The decay scheme is well-defined, with one small uncertain excited state and one uncertain gamma transition. Bismuth-213 decays by α -particle ($I_\alpha = 2.140(10)$ %) and β^- emission ($I_\beta = 97.860(10)$ %) to Tl-209 and Po-213 respectively. The Po-213 decays relatively promptly ($T_{1/2} = 3.706(1)$ μ s) by α -particle decay. The branching ratio of the α branch is sufficiently well known. There are 18 gamma transitions through the β^- decay path, though there is only one significant gamma-ray emission of 440.45 keV ($I_\gamma = 25.90(20)$ %). The absolute gamma-ray intensities come from determinations by three NMIs using absolute activity standards, which show good consistency and sufficient precision.

The half-life of 45.59(6) min has been derived from a weighted mean of six studies, which form a consistent dataset (Basunia, 2022). Three of these determinations have been performed since 2013, which have been instrumental to confirming this half-life (Marouli et al., 2013; Suliman et al., 2013; Takács and Kossert, 2021).

Table 18. Nuclear decay data of Bi-213 (Basunia, 2022).

$T_{1/2}$ /min	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
45.59(6)	$Q_{\alpha} = 5988(4)$	2.140(10) % α	$E_{\alpha} = 5875(4)$ keV ($I_{\alpha} = 91.55(11)$ %)	$E_{\gamma} = 440.45(1)$ keV ($I_{\gamma} = 25.90(20)$ %)
	$Q_{\beta} = 1422(6)$	97.860(10) %	$E_{\beta} = 1422(6)$ keV ($I_{\beta} = 66.8(5)$ %)	

2.18 Actinium-225

Actinium-225 decays by alpha particle emission to Fr-221, which undergoes further alpha decay to At-217 followed by an alpha decay to Bi-213. Actinium-225 has a complex decay scheme, with over 100 gamma transitions which are all low intensity, with 14 unplaced and a further 20 observed in one study excluded. 50.7(15) % of the alpha decays go direct to the ground state. More extensive gamma emission intensity studies are required along with γ - γ coincidence studies are recommended (Nichols, 2022).

The half-life of 10.0(1) d previously recommended has been significantly improved with a half-life of 9.920(3) d determined from six measurement techniques (Pommé et al., 2012). A further study of the half-life has been published in 2020 providing a corroborating value of 9.9179(30) d (Kossert et al., 2020). Further measurements are unlikely to improve the half-life by any significant amount.

A summary of the evaluated nuclear decay data is provided in Table 19.

Table 19. Nuclear decay data of Ac-225 (Kumar Jain et al., 2007).

$T_{1/2}$ /h	Q-value /keV	Decay mode(s)	Principal particle emissions	Principal gamma-ray emission
9.920(3)	$Q_{\alpha} = 5935.1(14)$	100 % α	$E_{\alpha} = 5830(2)$ keV ($I_{\alpha} = 50.7(15)$ %)	$E_{\gamma} = 99.8(1)$ keV ($I_{\gamma} = 1.00(11)$ %)

3. A note on Auger-electron data

Auger electrons are an attractive tool for targeted radiation therapy as they deposit their energies over short distances (nm to μ m). The initial radionuclide catalogue of PRISMAP contains promising Auger-electron emitting radionuclides (namely Tb-161, Er-165, Pt-195m) that are suitable for targeted radiotherapy. Auger-electron energies and emission probabilities have been predominantly calculated due to a paucity of experimental data, mainly due to the difficulty in preparing ultra-thin sources with atomic scale thicknesses. The calculations of these Auger-electron yields and energy distributions rely on adequate atomic (X-ray and internal conversion electron) data and nuclear data to calculate them with confidence. However, this atomic and nuclear decay data is not always adequate and can be resulting in large errors. To develop a greater understanding of the effects of Auger-electrons for targeted cancer therapy, through accurate nanodosimetry calculations, accurate experimental data of the Auger-electron energy and emission intensities are required for the potential Auger-electron emitting radionuclides that have been identified. It is recognised that this would be a challenging experimental programme due to the difficulties in preparing the sources and the development of the measurement system.

4. Summary of the status of the nuclear decay data

Throughout the review in section 2, this report has identified where it considers the current evaluated nuclear decay data to be sufficient and has made subjective recommendations for where new nuclear decay data studies would be beneficial have been made from the assessments of the current literature. For ease of reference, these have been summarised in Table 20.

Table 20. Summary of nuclear decay data needs for the PRISMAP day-1 radionuclides.

Radionuclide	Recommendations for future studies
Sc-44	<ul style="list-style-type: none"> ▪ New studies of the decay branching ratio of the ε/β^+ decay routes. ▪ Further studies of the half-life may be beneficial to improve the current evaluation dataset.
Sc-47	<ul style="list-style-type: none"> ▪ Absolute gamma-ray emission intensity studies of the 159.381 keV would be of interest to revisit to resolve a bias between values determined from different measurement systems.
Cu-64	<ul style="list-style-type: none"> ▪ Further studies of the decay branching ratio of the β^- decay route is worthy of further scrutiny.
Cu-67	<ul style="list-style-type: none"> ▪ New studies of the half-life would be beneficial to confirm the accuracy of the half-life and to improve the precision. ▪ A new decay data evaluation is required.
Ag-111	<ul style="list-style-type: none"> ▪ A new decay data evaluation is required.
La-135	<ul style="list-style-type: none"> ▪ Further studies of the half-life are needed. ▪ Additional γ-γ coincidence studies would be of use to complete the placement of the gamma transitions in the decay scheme. ▪ Absolute gamma-ray emission intensity studies derived from an absolute standard are needed to improve the precision of these values. ▪ Requirements for improved X-ray and Auger-electron data studies.
Tb-149	<ul style="list-style-type: none"> ▪ Precision measurements of the half-lives of Tb-149 and its decay progenies (Eu-145 and Gd-149) are needed. ▪ New studies are required to improve the precision of the alpha decay branching ratio. ▪ There is a requirement for new studies of the gamma-ray emission intensities to confirm the accuracy of the single study and to improve the precision. ▪ There is also a requirement to improve the gamma-ray emission intensities of the decay progenies.
Tb-152	<ul style="list-style-type: none"> ▪ New γ-γ coincidence and TAGS studies are needed to complete the decay scheme and to confirm the highest energy transition states. ▪ New half-life measurements are needed to confirm the accuracy of two studies in the 1960s and to improve the precision. ▪ Absolute gamma-ray emission intensity measurements are needed.
Tb-155	<ul style="list-style-type: none"> ▪ Further γ-γ coincidence measurements are needed to resolve the placement of 40 gamma transitions. ▪ Absolute gamma-ray emission intensities are required to improve the electron capture branching ratios. ▪ Further studies of the half-life are warranted to expand the evaluation dataset.
Tb-161	<ul style="list-style-type: none"> ▪ Further studies are required of the gamma-ray emission intensities, especially the 25.65 keV gamma ray to improve the beta branching ratio values to the ground state. ▪ A new evaluation of the half-life is required. ▪ There are requirements for further X-ray and internal conversion electron data. ▪ Studies of the Auger-electron emission data is required.
Ho-166	<ul style="list-style-type: none"> ▪ A new evaluation is required to account for new data.
Er-165	<ul style="list-style-type: none"> ▪ Direct measurement of the Auger-electron energies and intensities are required.

Radionuclide	Recommendations for future studies
	<ul style="list-style-type: none"> ▪ A modern measurement of the half-life would be desirable to confirm measurements made in the 1950s and 1960s.
Er-169	<ul style="list-style-type: none"> ▪ More detailed K X-ray and internal conversion data for the direct population of the 8.41 keV state and the transition to the ground state are recommended. ▪ Further studies of the half-life are recommended.
Yb-175	<ul style="list-style-type: none"> ▪ Further studies of the half-life are desirable to increase the evaluation dataset.
Pt-195m	<ul style="list-style-type: none"> ▪ Definite requirement for absolute gamma-ray emission intensities to improve the precision and confirm the accuracy. ▪ Further data for the internal conversion electron probabilities would be of benefit. ▪ New studies of the half-life using high-purity samples would be beneficial. ▪ Direct measurement of the Auger-electron energies and intensities are required.
At-211	<ul style="list-style-type: none"> ▪ New half-life determinations with complete uncertainty evaluation are required.
Bi-213	<ul style="list-style-type: none"> ▪ No recommendations.
Ac-225	<ul style="list-style-type: none"> ▪ Extensive gamma-ray emission intensity studies and γ-γ coincidence studies are recommended are required.

The status presented here, including the evaluated data as well as the recent complementary data, will be collated in an information portal on the PRISMAP website in collaboration with the Online Documentation task within the PRISMAP Training Office. This portal can then be further updated as new data is collected according to the recommendations above. Furthermore, online documentation will provide an easy access to this information, as well as the latest investigations in an easily accessible, centralised database.

5. Conclusions

The current state of eighteen radionuclides from the PRISMAP day-1 radionuclides has been reviewed using the most up-to-date evaluations and a review of literature published after those evaluations had taken place. Recommendations for areas of improvement have been identified or where further measurements would be beneficial to confirm a small dataset. In some cases, such as Tb-161, where the half-life was ‘known’ due to large datasets it has been noted that significant differences have been observed. This raises a particular issue where researchers may ignore measuring nuclear decay data for a radionuclide due to the impression that the data is already “good enough”, especially where the data was determined many decades previously. It is therefore just as important to confirm nuclear decay data, where it appears to be adequate, using the state-of-the-art capabilities available now as it is to determine interesting new data.

In several cases, for example, when reviewing the half-lives, where the dominant values are pre-2007 it may be advised that new determinations are needed. For half-lives this represents a paradigm shift in the metrology for half-life determinations led by the seminal paper by Pommé (Pommé, 2007). This has led to a significant revision in the importance of uncertainty evaluation and the biases that can arise.

Within PRISMAP, many actions are being undertaken to address some of the identified open questions from Table 20. In particular, the Networking Activity on Standardisation and Harmonisation is performing interlaboratory comparisons on the isotope Cu-64 through which opportunities to investigate its nuclear decay are being identified. Furthermore, two Joint Research Activities are dedicated to complement this information, one specifically on developing translational data, with an emphasis on the traceability of the data, and another activity specifically aimed at investigating the radiolanthanides, which represent half of the isotopes listed in Table 20. It is furthermore expected that this report will inspire the PRISMAP consortium, as well as its user forum, to further investigate those nuclear decay data.

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