DDEP evaluation of Cs-137

Sylvain Leblond

CCRI Webinar *Nuclear data evaluation for radionuclide metrology*

5 December 2024

LnHB Laboratoire National Henri Becquerel

Laboratoire National de Métrologie et d'Essais LNE

université PARIS-SACLAY

1. Decay data of ^{137}Cs overview

Initiation of an evaluation project

- In 2021 a joined initiative was proposed to re-evaluate the ^{137}Cs decay data
  - Independent evaluation processes from ENSDF, DDEP and the CTBTO
  - Previous evaluation works were performed in 2006/2007
  - Data were known to be inconsistent
  - No consensual agreement was found between evaluators for decades

- As part of my training as new evaluator it was decided that I would take care of the DDEP evaluation
  - First independent evaluation work
  - Started late 2021, finalised in may 2023
  - Large support from the DDEP collaboration, especially Alan L. Nichols who performed the review

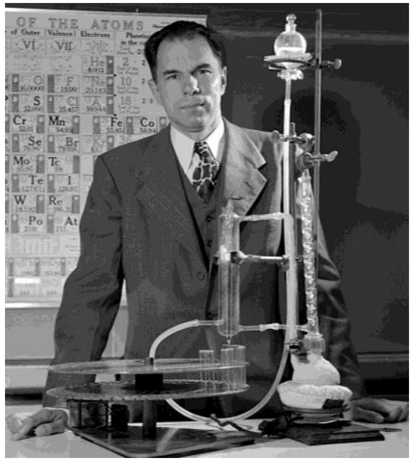
- The ^{137}Cs evaluation has been published
  - On the LNHB website in September 2023
  - In a peer-review journal
    DDEP re-evaluation of the radioactive decay scheme of ^{137}Cs, Applied Radiation and Isotopes 206, 111191 (April 2024)

Story of the ^{137}Cs discovery

- In 1941 at Berkeley
  - Margaret Melhase (undergraduate student) and Glenn Seaborg
  - First chemical extraction of radioactive Caesium
  - Produced by thermal neutron induced fission on ^{235}U

- Two different destinies
  - Glenn Seaborg shared 1951 Chemistry Nobel for ^{239}Pu discovery
  - Margaret Melhase could not pursue an academic career

(1941)



(1950)

- In 1969 Glenn Seaborg wrote to Margaret Melhase to ask for details on her chemical separation
  - *"My kids will never get over the thought that Glenn Seaborg had to ask anybody anything about radioisotopes – least of all their mother!"*
  - *"I hope you have succeeded in convincing your kids that you made an important contribution to the radioisotope field (resulting in many practical applications) during those days at Berkeley"*

Source D.D. Patton How cesium-137 was discovered by an undergraduate student J. Nucl. Med., 48 (1999), p. 18N

A very impactful discovery

- Since Margaret discovery
  - More than 80 000 publications in a wide range of applications



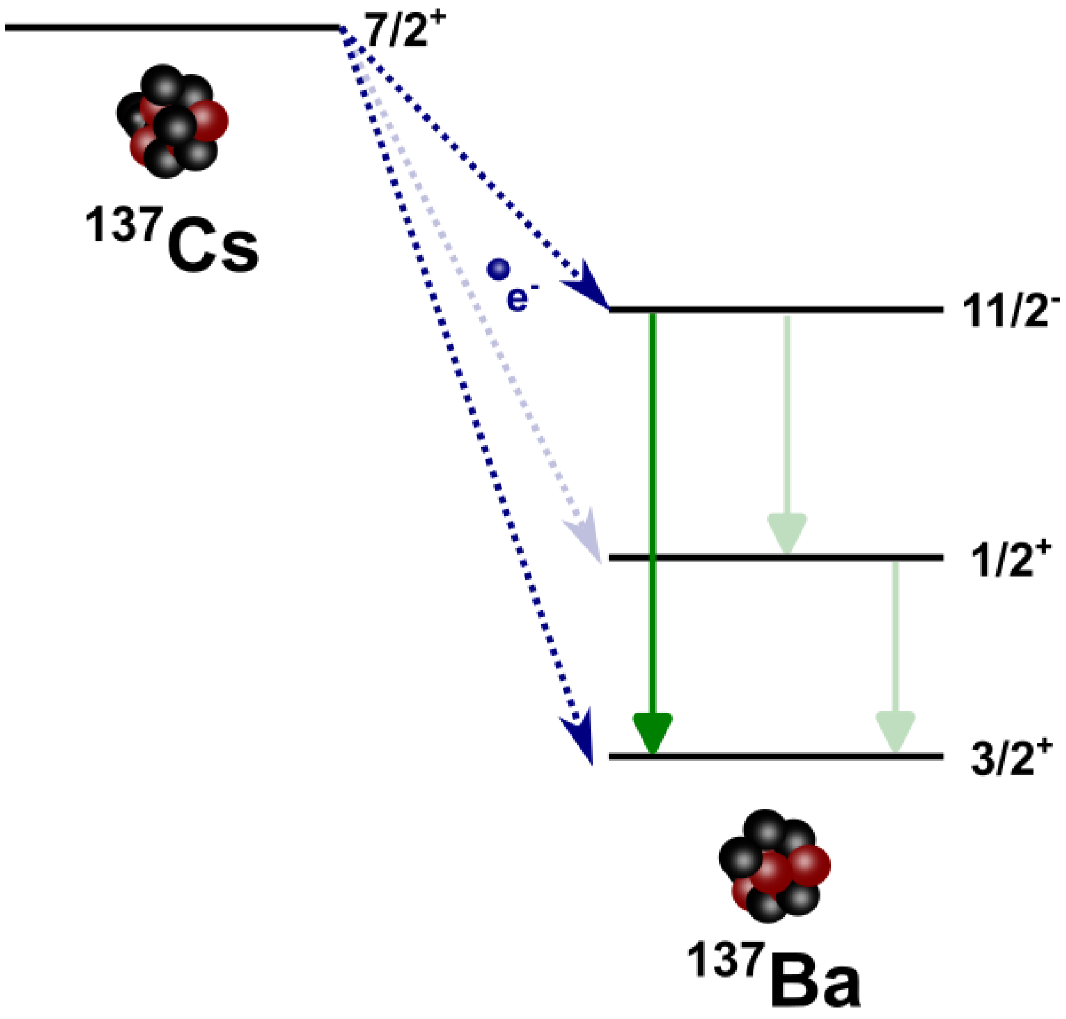
- One of the main fission products
  - Dominating medium-lived fission product
  - Primary source of penetrating gamma radiations from spent fuel
  - One of the most significant contamination radionuclide remaining after a nuclear disaster

Decay overview of ^{137}Cs

- A β^- decay to ^{137}Ba
  - Populates ground & excited states
  - Half-life around 30 years
  - Total Q-value close to 1176 keV

- Emissions
  - Two main β^- branches
    - Ground state (E$_{max}$ ~ 1176 keV)
    - Second excited state (E$_{max}$ ~ 514 keV)

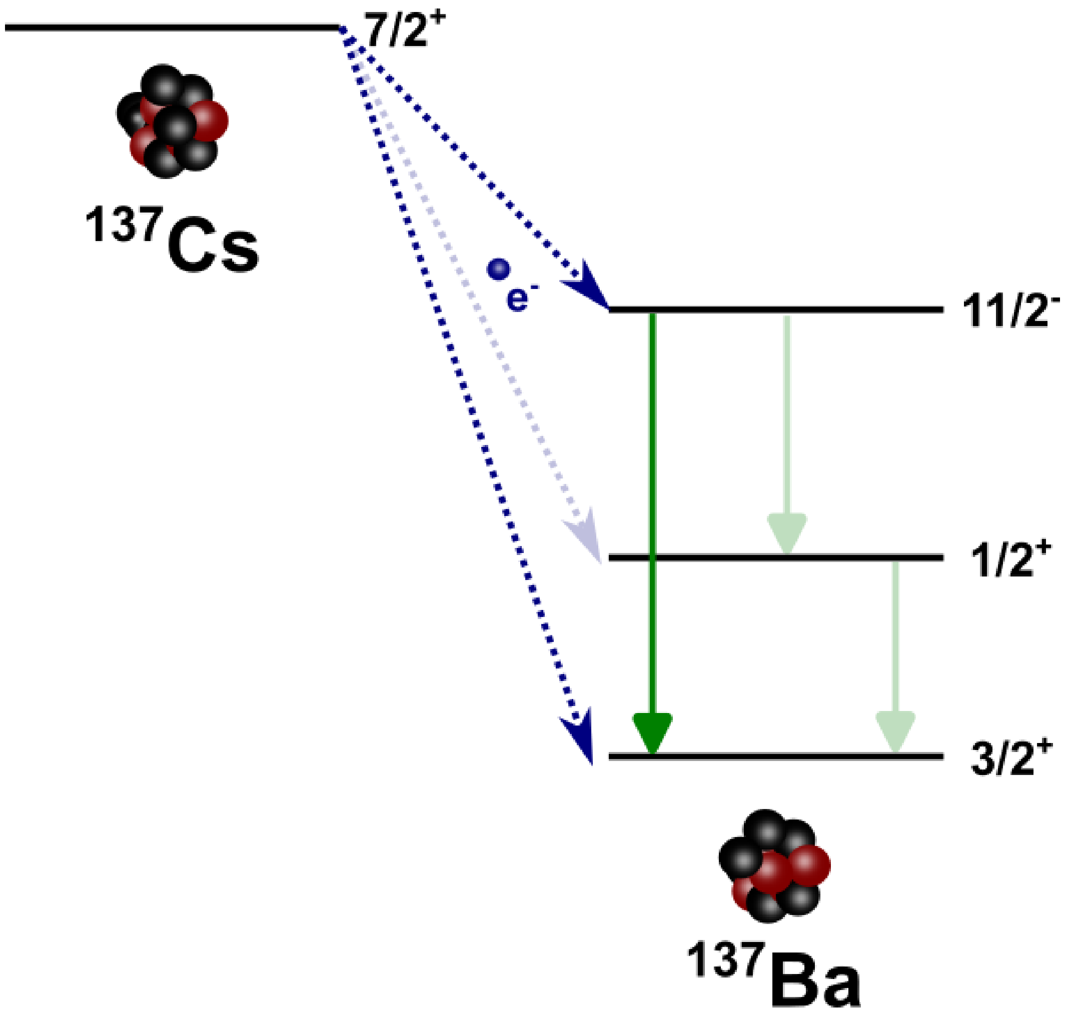
  - A very dominant 662 keV gamma emission
    - Good and easy-to-produce calibration source



Decay data status

- Latest ^{137}Cs decay data evaluations
  - **DDEP**
    - R.G. Helmer and V.P. Chechev (2006)
  - **ENSDF**
    - E. Browne and J.K. Tuli (2007)

	DDEP	ENSDF
$T_{1/2}$ (y)	30.05 (8)	30.08 (9)
$I_\beta(11/2^-)$ (%)	94.36 (28)	94.7 (2)
$I_\beta(3/2^+)$ (%)	5.64 (28)	5.3 (2)
I_γ (%)	84.99 (20)	85.1 (2)

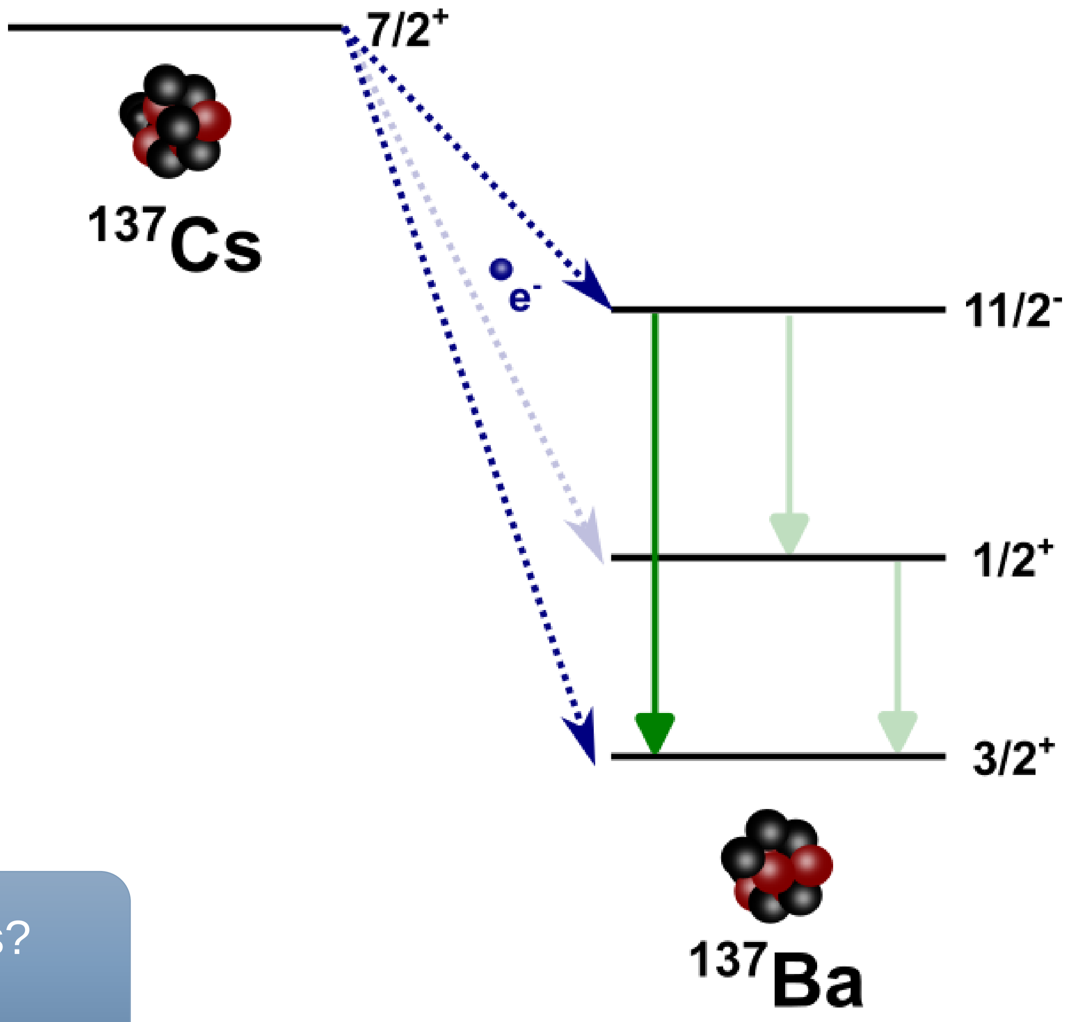


Decay data status

- Latest ^{137}Cs decay data evaluations
  - **DDEP**
    - R.G. Helmer and V.P. Chechev (2006)
  - **ENSDF**
    - E. Browne and J.K. Tuli (2007)

	DDEP	ENSDF
$T_{1/2}$ (y)	30.05 (8)	30.08 (9)
$I_{\beta}(11/2^{-})$ (%)	94.36 (28)	94.7 (2)
$I_{\beta}(3/2^{+})$ (%)	5.64 (28)	5.3 (2)
I_{γ} (%)	84.99 (20)	85.1 (2)

Can we improve the recommendations?
(focus on the half-life)



2. Evaluation of ^{137}Cs Half-life

At the beginning: collecting data

- Abundant bibliography
  - More than 75 publications
  - From 1948 to 2016
  - Different experimental methods
  - Many languages

At the beginning: collecting data

- Abundant bibliography
  - More than 75 publications
  - From 1948 to 2016
  - Different experimental methods
  - Many languages

- How to deal with a large dataset?
  - Should all the references be considered?
  - Are all reported uncertainties comparable?
  - Are all publications independant?

Some answers in DDEP guidelines

Constructing a meaningful dataset

- Private communications are discarded

- Treatment of experimental uncertainties
  - Measurement without uncertainty are discarded
  - Uncertainty should be reported at 1 σ confidence level
  - Uncertainty should be symmetric
  - Quadratic sum of Type A and B is considered

6.1 Rules for evaluation

All intermediate stages in the compilation and evaluation of a decay parameter are not presented in detail in order to avoid unnecessary complexity. The main stages comprise the following:

- critical analysis of published results and, if necessary, correction of these results to account for more recent values hitherto unavailable to the original experimentalists; as a rule, results without associated uncertainties are discarded, and the rejection of values is documented;
- data obtained through private communications are only used when there is no published article available;
- adjustments may be made to the reported uncertainties;
- only one result, generally the latest one, is taken into account per laboratory (or author);
- recommended values are derived from an analysis of all available measurements (or theoretical considerations), along with the standard deviations corresponding to the 1 σ confidence level.

Constructing a meaningful dataset

- Private communications are discarded

- Treatment of experimental uncertainties
  - Measurement without uncertainty are discarded
  - Uncertainty should be reported at 1 σ confidence level
  - Uncertainty should be symmetric
  - Quadratic sum of Type A and B is considered

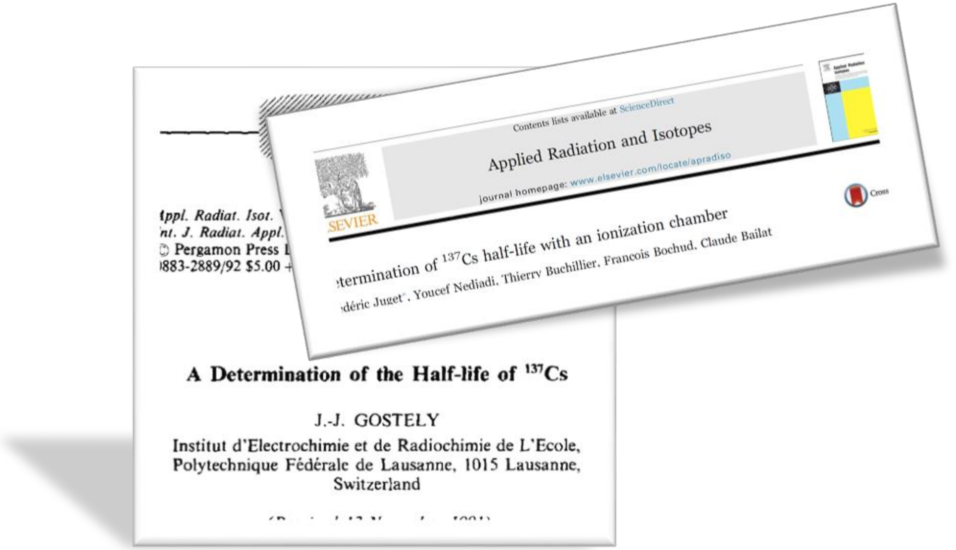
- Only one publication per author / laboratory is considered
  - To avoid systematic biases (analysis, source contamination...)
  - Requires to identify / construct the most reliable value
  - Important: identify the correlation between reported measurements

- Look for dependancies of measurements on physical constants
  - Correct them if possible with latest recommendations

6.1 Rules for evaluation

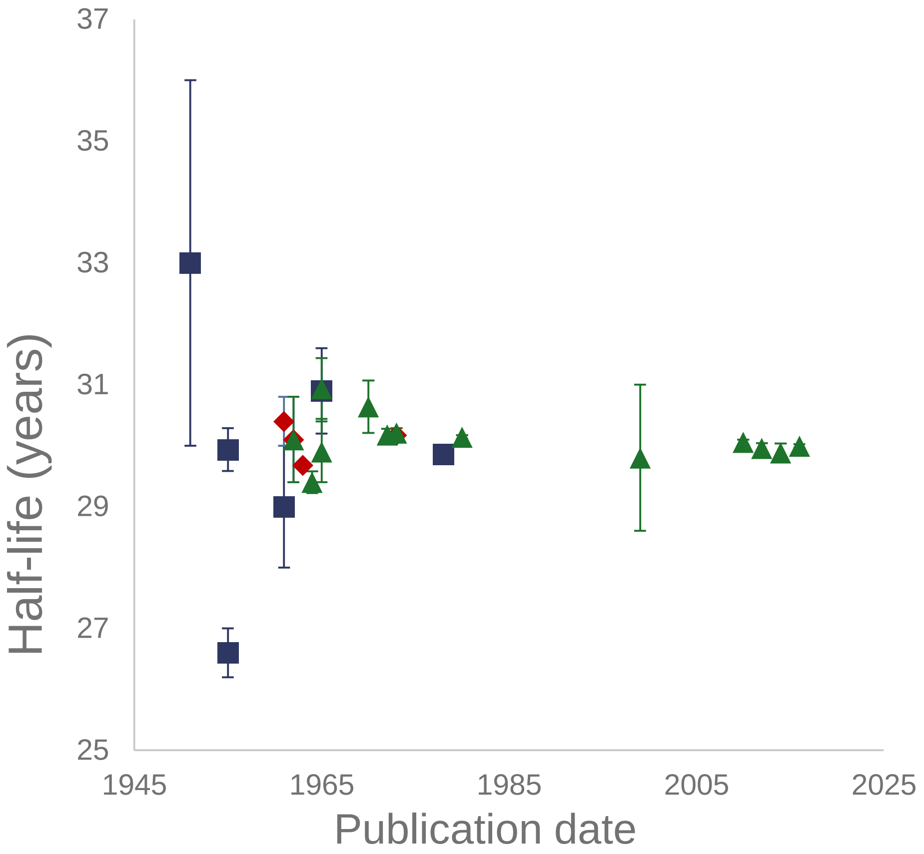
All intermediate stages in the compilation and evaluation of a decay parameter are not presented in detail in order to avoid unnecessary complexity. The main stages comprise the following:

- critical analysis of published results and, if necessary, correction of these results to account for more recent values hitherto unavailable to the original experimentalists; as a rule, results without associated uncertainties are discarded, and the rejection of values is documented;
- data obtained through private communications are only used when there is no published article available;
- adjustments may be made to the reported uncertainties;
- only one result, generally the latest one, is taken into account per laboratory (or author);
- recommended values are derived from an analysis of all available measurements (or theoretical considerations), along with the standard deviations corresponding to the 1 σ confidence level.



A first dataset

- Following all the previous recommendations
  - From initially 75 to 23 references selected
  - $T_{1/2}$ varying from 26 to 33 years
  - Uncertainties from 0.011 to 3 years



A first dataset

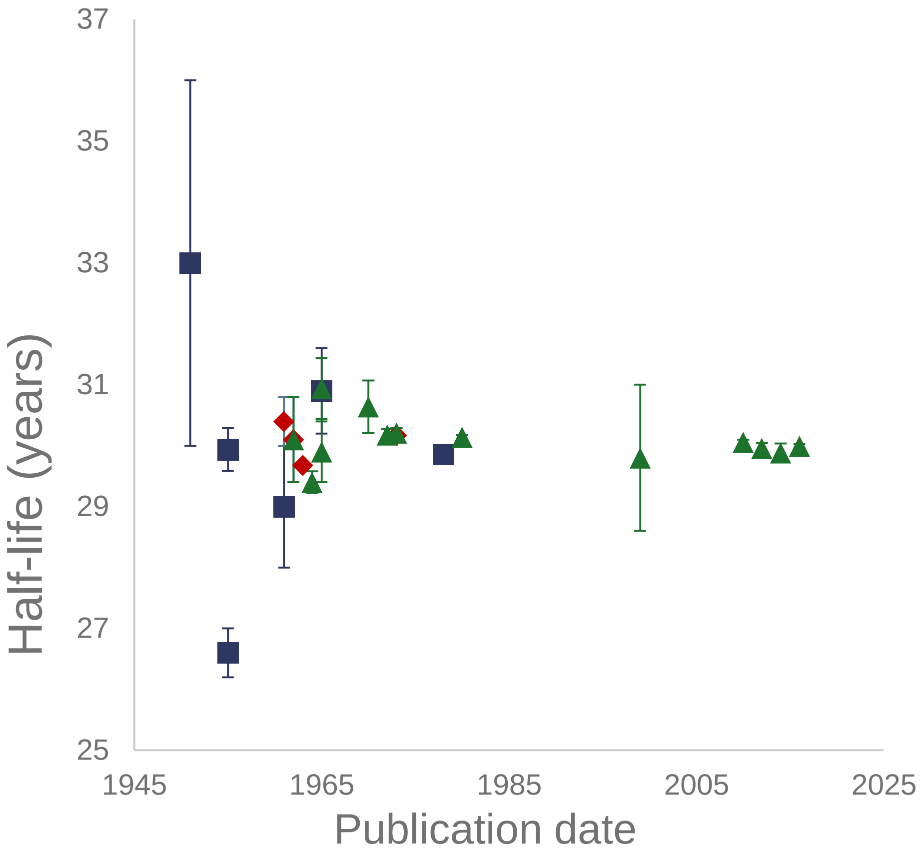
- Following all the previous recommendations
  - From initially 75 to 23 references selected
  - $T_{1/2}$ varying from 26 to 33 years
  - Uncertainties from 0.011 to 3 years

- Statistical test of the distribution

$$\frac{\chi^2}{(n-1)} = \frac{u_{ext}(\bar{M})^2}{u_{int}(\bar{M})^2}$$

$$\bar{M} = \frac{\sum {m_i}/{u_i^2}}{\sum {1}/{u_i^2}} \qquad u_{int}(\bar{M}) = \sqrt{\frac{1}{\sum {1}/{u_i^2}}} \qquad u_{ext}(\bar{M}) = \sqrt{\frac{\sum {(m_i - \bar{M})^2}/{u_i^2}}{(n-1)\sum {1}/{u_i^2}}}$$

  - $\chi^2 \sim 11 \gg \chi^2_{crit} \sim 1.9$
    - Discrepant dataset
    - At least one of the reported uncertainty cannot be trusted



A first dataset

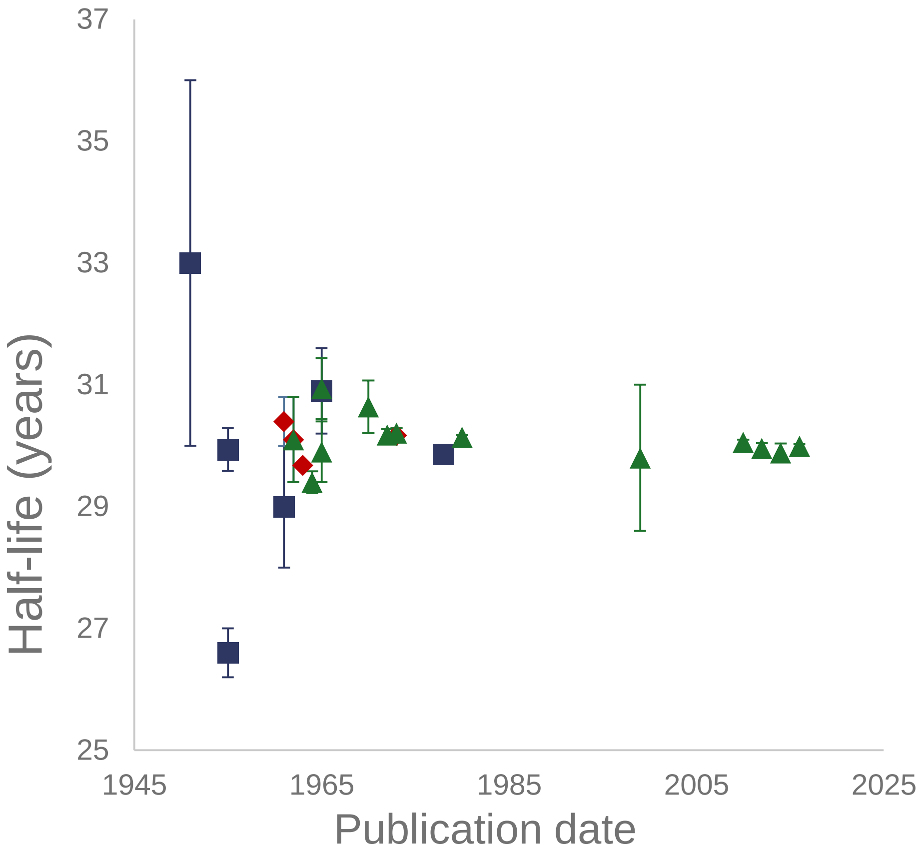
- Following all the previous recommendations
  - From initially 75 to 23 references selected
  - $T_{1/2}$ varying from 26 to 33 years
  - Uncertainties from 0.011 to 3 years

- Statistical test of the distribution

$$\frac{\chi^2}{(n-1)} = \frac{u_{ext}(\bar{M})^2}{u_{int}(\bar{M})^2}$$

$$\bar{M} = \frac{\sum {m_i}/{u_i^2}}{\sum {1}/{u_i^2}} \qquad u_{int}(\bar{M}) = \sqrt{\frac{1}{\sum {1}/{u_i^2}}} \qquad u_{ext}(\bar{M}) = \sqrt{\frac{\sum {(m_i - \bar{M})^2}/{u_i^2}}{(n-1)\sum {1}/{u_i^2}}}$$

  - $\chi^2 \sim 11 \gg \chi^2_{crit} \sim 1.9$
    - Discrepant dataset
    - At least one of the reported uncertainty cannot be trusted



Unweighted average $T_{1/2}$ = 30.09 (8) a

	DDEP	ENSDF
$T_{1/2}$ (a)	30.05 (8)	30.08 (9)

How to proceed further?

Dataset refinement

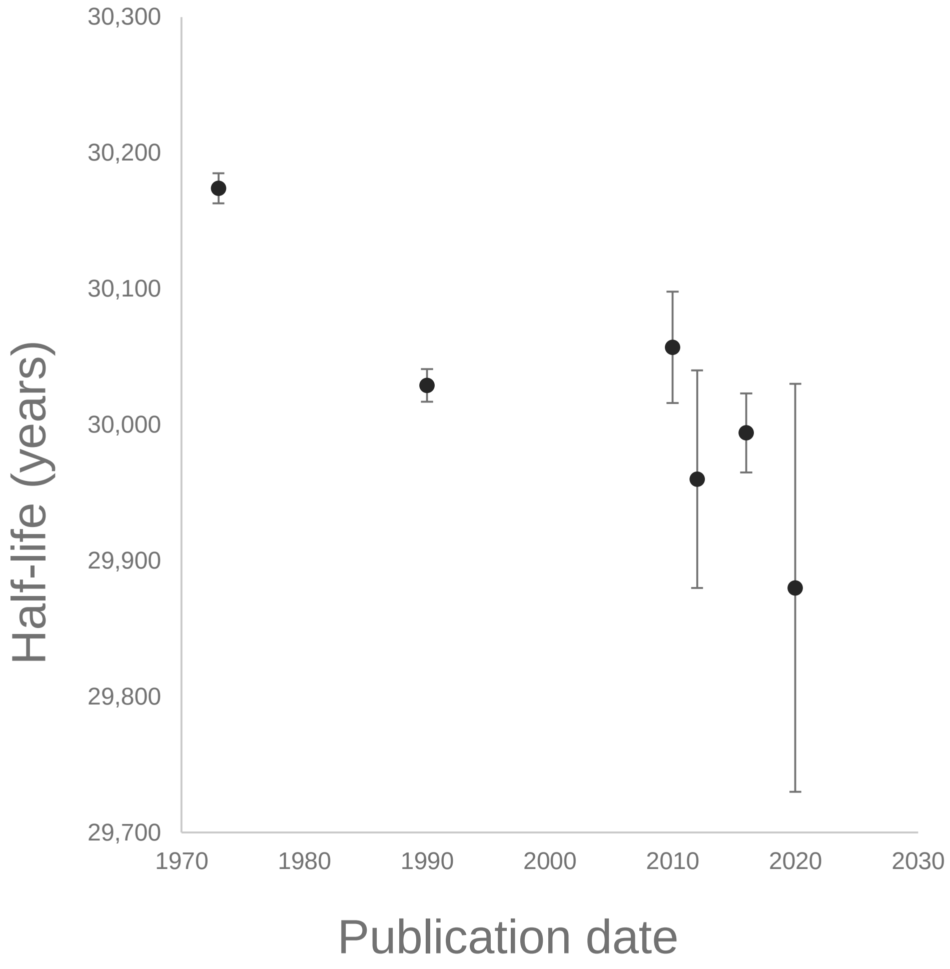
- Critical analysis of each publication
  - Evaluate possible experimental bias
  - Evaluate uncertainty budget assessment

- Support from previous works

  M.J. Woods, *The Half-life of ^{137}Cs – A critical Review,* NIM A286 (1990)

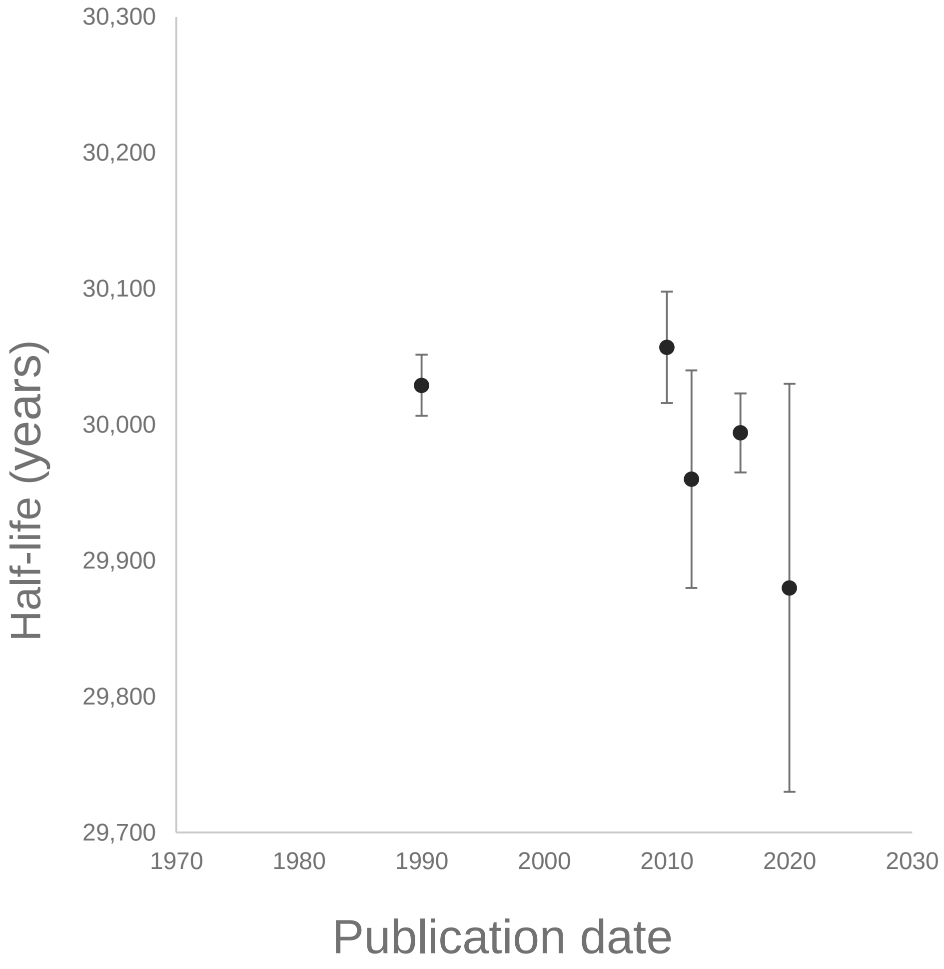
- Selected publications

Dietz et al., (1973)	GEC	$T_{1/2}$ = 30.174 (11) a
Martin et al., (1990)	CRNL	$T_{1/2}$ = 30.029 (12) a
Schrader et al., (2010)	PTB	$T_{1/2}$ = 30.06 (4) a
Bellotti et al., (2012)	INFN	$T_{1/2}$ = 29.96 (8) a
Juget et al., (2016),	IRP	$T_{1/2}$ = 29.994 (29) a
Unterweger et al., (2020)	NIST	$T_{1/2}$ = 29.88 (15) a



Final ajustements

- Removal of Dietz *et al.* (mass spectrometry)
  - Smallest reported uncertainty yet discrepant with other results
  - Possible contaminants are discussed but not quantified
- Increase of the uncertainty of Martin *et al.* (ionization chamber)
  - Very small uncertainty with regards to *Schrader and Juget*
  - Longer measurement time but the uncertainty dominated by other components
- Final dataset considered
  - ~~*Dietz et al.*, (1973) GEC $T_{1/2}$ = 30.174 (11) a~~
  - *Martin et al.*, (1990) CRNL $T_{1/2}$ = 30.029 (22) a
  - *Schrader et al.*, (2010) PTB $T_{1/2}$ = 30.06 (4) a
  - *Bellotti et al.*, (2012) INFN $T_{1/2}$ = 29.96 (8) a
  - *Juget et al.*, (2016), IRP $T_{1/2}$ = 29.994 (29) a
  - *Unterweger et al.*, (2020) NIST $T_{1/2}$ = 29.88 (15) a

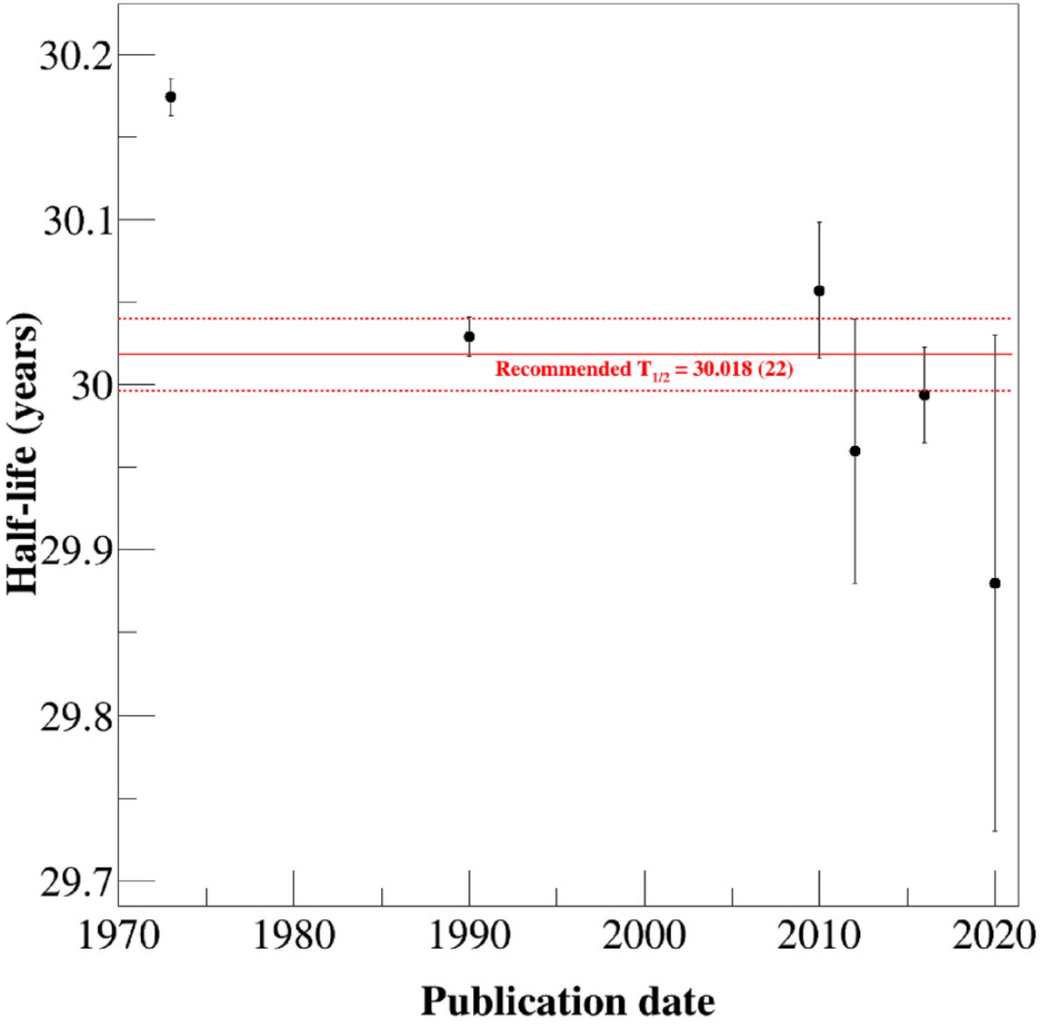


New DDEP recommendation

- A final dataset of 5 measurements
  - Consistent: $\chi^2 \sim 0,8 < \chi^2_{crit} \sim 3.3$
  - Unweighted mean $T_{1/2}$ = 29.984 (30) a
  - Weighted mean $T_{1/2}$ = 30.018 (16)$_{int}$ (14)$_{ext}$ a
  - Recommended uncertainty extended to match *Martin et al.* (1990)

$T_{1/2}$ = 30.018 (22) a

	DDEP	ENSDF	This work
$T_{1/2}$ (a)	30.05 (8)	30.08 (9)	**30.018 (22)**



3. Conclusions

(and outlooks)

Re-evaluation of ^{137}Cs decay data

- A complete re-evaluation of ^{137}Cs decay data was performed
  - A two years “*training*” project
  - Following the DDEP evaluation methodology
  - Improvements on the half-life and branching ratio evaluation
  - Involves arbitrary choices based on evaluator opinion
  - Complete analysis available online on LNHB website (41 pages)

http://www.lnhb.fr/home/nuclear-data/nuclear-data-table/

Comments on evaluation **^{137}Cs**

^{137}Cs - Comments on evaluation of decay data

May 2023

- Outlooks
  - Recommendation for new measurements
    - Half-life
      - Decrease the dependency on a single measurement (Martin *et al.*)
    - Emission intensity of 662 keV gamma
      - Improve the uncertainty on the recommended value (small dataset)