

# NSDD-09 Action item #29

## Guidelines for evaluation of half-lives (ground states and long-lived isomers)

Preliminary draft:

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## Half-life compilations/evaluations available

### Independent:

- ENSDF (most comprehensive source)
- DDEP (for selected ~250 nuclei)
- Table of Isotopes (1978)

### Data extracted mainly from other compilations:

- Nuclear Wallet Cards (mainly ENSDF, other)
- NuDat (ENSDF / NWC)
- NUBASE-2003 (mainly ENSDF, NWC, other)
- Wall Chart of Nuclides-2002 (mainly ENSDF, NWC)
- Table of Isotopes (1996, 1999: mainly ENSDF)

# Half-life evaluation

1. Identify and accumulate ALL published measurements of the half-life of the ground state or isomer. (For literature prior to 1960, may have to search for references in sources other than NSR).
2. Ensure that all of the above half-life data and origins (reference keynumbers) are listed systematically in ***Adopted Levels, Gammas data set***. (Check quoted uncertainty:  $1\sigma$ ,  $2\sigma$  or  $3\sigma$ ; ENSDF uses  $1\sigma$ )
3. Consider any other features of each specific measurement for either rejection or increased preference, based on your own experience and subjective judgments. Examples include the following:
  - acceptance or rejection of *grey* references (publications that have not been fully peer reviewed: laboratory reports; conference proceedings; sometimes the journal issue of a set of conference papers),

# Half-life Evaluation (cont.)

- measurement technique (compared with others, the technique is judged or known to be more appropriate for the half-life being addressed),
  - recognized difficulties and complications (e.g. impact of impurities, detector limitations, background corrections, dead-time corrections, relative to a “known” half-life),
  - known reliability or improvements in a particular measurement technique (improvements might make the date of the measurements important),
  - regular and lengthy measurement program of specific half-lives for important applications (normally a policy instigated by national standards laboratories, but also observed to be undertaken by others) can result in accepting only the most recently reported value; complications can also arise when the laboratory changes equipment/technique,
    - if the same author(s) determine a particular half-life data based on the same measurement technique/apparatus, only consider the most recent value in deducing the recommended value,
- and various other imponderables..... statistical, systematic uncertainties,

## Half-life Evaluation (cont.)

4. Identify outliers, document and discard, based on the criteria adopted in least-squares analysis codes. Numerous averaging techniques have been proposed and developed (see AVETOOLS). Examples include:
  - weighted mean (WM);
  - limitation of the relative statistical weight (LRSW, LWM);
  - normalised residuals (NR);
  - Rajeval Technique (RT)
  
5. These techniques use different methods to handle the uncertainties, identify outliers, and derive the mean value and uncertainty. LRSW, NR and RT use the uncertainties and occasionally inflate them to accommodate discrepant data; all three of these methods should be used simultaneously to identify outliers (i.e. defined as such if at least two of the methods identify a data point as an outlier).

## Half-life Evaluation (cont.)

6. Other methods:
  - Bootstrap Median (BM): well-known in epidemiological and social sciences but not in physical applications.
  - Mandel-Paule (MP): maximum likelihood method, developed at NIST for inter-laboratory comparison of “standards” mainly chemical compounds.
  - Codes for both are available, methods include uncertainties; but no criterion for identification of outliers.
  - Seem less sensitive to a few values quoted with high precision. See for example attached example of Au-198 half-life analysis
7. All acceptable half-life data to be analyzed by means of these techniques
  - may need to define which method is the most appropriate –WM? LRSW? NRM? BM? Other? and so adhere to consistency in the selection of the recommended half-life value and uncertainty,
  - role of reduced  $\chi^2$  in such analyses needs to be better defined, implemented, and used to develop a more rigorous understanding of the data set adopted for full analysis.
8. Uncertainties of **0.01%-0.1%** quoted in a paper should be viewed with caution!

# Half-life Evaluation, Examples (cont)

## Co-62 half-life

### Co-62 half-life

Reference	Half-life (min)	Comments
1949Pa01	1.6 (2)	$\beta$ -decay curves followed over six half-lives; decay curve shown
1960Pr05	1.9 (3)	$\beta$ -decay curve not shown – only lists half-life
1962Va23	1.5 (1)*	$\beta$ -decay curve followed over four half-lives; no discussion of impurities
1969Wa16	1.50 (4) <sup>#</sup>	$\gamma$ - $\gamma$ coincidence and high energy $\beta$ ; decay curves not shown – only lists half-life
1970Jo12	1.4 (2)	1129-keV $\gamma$ decay followed for more than five half-lives; decay curves shown for several $\gamma$ rays
	<b>1.54(10)</b>	Recommended value (LRSW – Limitation of Relative Statistical Weights)

\* Uncertainty increased to  $\pm 0.2$  to reduce weighting to below 50%.

<sup>#</sup> Uncertainty increased to  $\pm 0.20$  to reduce weighting to below 50%.

### Co-62m half-life

Reference	Half-life (min)	Comments
1949Pa01	13.9 (2)	$\beta$ -decay curves followed over six half-lives; decay curve shown
1957Ga15	13.91 (5)*	$\gamma$ decay measured in well-type scintillation detector; minor Cu-64 and Ni-65 impurities present; no decay curves shown – only lists half-life
1960Pr05	13.8 (2)	$\beta$ -decay curve not shown – only lists half-life
1962Va23	13.9 (2)	$\beta$ -decay curve followed over about two half-lives; no discussion of impurities
1969Wa16	14.00 (24)	High energy $\beta$ and $\gamma$ decay; decay curves not shown - only lists half-life
1969Mo04	13.8 (5)	1163-, 1172-, 2003- and 2103-keV $\gamma$ decay followed for about six half-lives; decay curves shown for several $\gamma$ rays
1970Jo12	13.5 (3)	1163- and 1173-keV $\gamma$ decay followed for more than two half-lives; decay curves shown for several $\gamma$ rays
	<b>13.86 (9)</b>	Recommended value (LRSW)

\* Uncertainty increased to  $\pm 0.20$  to reduce weighting to below 50%.

# Half-life Evaluation, Examples (cont)

## Cu-62 half-life

Cu-62 half-life		
Reference	Half-life (min)	Comments
1954Nu27	10.1 (2) <sup>*</sup>	Cu-62 milked from parent Zn-62
1965Eb01	9.76 (2)	Decay of positron annihilation radiation; Cu-64 impurity considered constant - no decay curves, only lists measured half-life
1965Li11	9.79 (6)	Decay of positron annihilation radiation corrected for Cu-64 activity, and fitting of excitation functions for Co-63(n,2n)Cu-62 reaction at En=12.6-19.6 MeV – lists half-life derived from these fittings
1969Bo11	9.7 (1)	Decay of positron annihilation radiation and fitting of excitation functions for Co-63(n,2n)Cu-62 reaction at En=13-18 MeV – lists half-life derived from these fittings
1969Jo07	9.73 (2)	Decay of positron annihilation radiation - no decay curves, only lists measured half-life
1975Ca40	9.80 (2)	γ-ray decay – no decay curves, only lists measured value
1997Zi06	9.68 (4)	4πβ liquid scintillation spectrometry, twelve independent measurements spanning two to four half-lives
	9.673 (26)	4πγ ionization chamber, two independent measurements spanning two to four half-lives
2002Un02	9.673 (8) <sup>*</sup>	Quote 1997Zi06, see above, but uncertainty seems statistical only.
	9.74 (6)	LRSW: weighted average of the above with uncertainty expanded so that range includes the most precise value (9.673 min); data set exhibits significant inconsistencies that mitigate against LSWM approach
1997Zi06	9.68 (4)	4πβ liquid scintillation spectrometry, twelve independent measurements spanning two to four half-lives
	9.673 (26)	4πγ ionization chamber, two independent measurements spanning two to four half-lives
2002Un02	9.673 (8) <sup>*</sup>	Quote 1997Zi06, see above, but uncertainty seems statistical only.
	9.675 (22)	Recommended value: from weighted average of two values in 1997Zi06. Uncertainty should be increased to 0.026.

<sup>\*</sup> Rejected as outlier, and not included in the data sets for LRSW analyses.  
<sup>\*</sup> Not included in averaging.



# Bi-207 half-life evaluation in NDS

**Bi-207 half-life: 2011Ko04 – F.G. Kondev, S. Lalkovski, NDS 112 (2011) 707-853**

**Recommended  $T_{1/2}$ : 31.55 y 4**

- $T_{1/2}$ : From [2002Un02](#), using  $4\pi\gamma$  pressurized ionization chamber; stat. uncertainty 0.025 y and syst. uncertainty 0.033 y. No impurities in the sources were observed using HPGe; decay has been followed over a period of  $t \approx 28$  years. The value agrees with that of 31.55 y 5 reported by the same group ([1992Un01](#)), when decay was followed over a period of  $t \approx 19$  years.  
**The value is superior to others described below.**

## **Others (not used in the present evaluation):**

- 32.7 y 8 ([1991Li10](#)) by measuring the activity of a calibrated  $^{207}\text{Bi}$  source ( $t \approx 17$  years after the source was calibrated) with a HPGe detector; value determined by averaging activities for 569 $\gamma$ , ( $I_\gamma = 97.75\%$ ), 1063 $\gamma$  ( $I_\gamma = 76.0\%$  14) and 1770  $\gamma$  ( $I_\gamma = 6.95\%$  13);  $T_{1/2} = 31.6$  y 7, when the activity was deduced using 569 $\gamma$  only. The quoted uncertainty is statistical only. A sizable syst. uncertainty can be expected, given the uncertainties in the nuclear data parameters used in the calibration of the source.
- 34.9 y 4 ([1990Al11](#)) using a gas-flow proportional counter system; the uncertainty is stat. only and quoted at 2s level; the source was produced by bombarding a Pb target with 22-MeV deuterons following chemical separation; the measurements were followed over a period of  $t = 3.4$  years. A break in the singles rates were observed around  $t = 1.7$  years after the beginning of the measurements, So the data were analyzed in two separate parts yielding  $T_{1/2} = 34.88$  y 21 from the first 27 points (up to  $t = 1.7$  y) and 35.2 y 9 from the next 8 points; the quoted  $T_{1/2}$  is higher than the adopted one. The quoted uncertainty is statistical only and large syst. uncertainty should be expected owing to sensitivity of the measurements to temperature and humidity changes. It is worth noting that  $T_{1/2} = 66.6$  y 16 was reported by this group ([1990Al11](#)) for  $^{44}\text{Ti}$ , which is higher than other precise measurements of 58.9 y 3 ([2006Ah10](#)) and 60.7 y 13 ([1999Wi01](#)).

## Bi-207 half-life evaluation in NDS (cont.)

33.4 y 8 ([1978Ya04](#)) deduced indirectly using the decay of a  $^{211}\text{At}$  source and knowledge of the  $\epsilon/\alpha$  branching ratio of  $^{211}\text{At}$  (0.583/0.417), the emission probability of 6.568 MeV  $\alpha$  to the 569.7 keV level of  $^{207}\text{Pb}$  (0.58% 1), the half-life of  $^{211}\text{At}$  (7.23 h 2) and the total emission probability of 569.7 $\gamma$  fed in  $^{207}\text{Bi}$   $\epsilon$  decay (99.85%). The quoted uncertainty is statistical only, but a large syst. uncertainty can be expected. The authors also quote a value of 32.2 y 13 using disintegration rate of  $^{211}\text{At}$  in a purified sample measured by the means of a liquid scintillation counter and by adopting the 569.7 $\gamma$  to determine the decay rate of  $^{207}\text{Bi}$ . A measurement performed after a complete decay of  $^{211}\text{At}$  yielded  $T_{1/2}=32.2$  y 37, whereby the large uncertainty was attributed to the poor detection efficiency of gamma counting of this particular sample.

38 y 4 ([1972Ru10](#)) using a  $^{207}\text{Bi}$  source by counting the 569.7 keV gamma ray, using a NaI scintillation spectrometer over a period of  $t=0.5$  years.

38 y 3 ([1961Ap01](#)) deduced indirectly using the decay of  $^{211}\text{At}$  source and knowledge of the  $\alpha$  branching ratio of  $^{211}\text{At}$  (40.9%), the half-life of  $^{211}\text{At}$  (7.214 h 35) and the total emission probability of 569.7 $\gamma$  that is fed in  $^{207}\text{Bi}$   $\epsilon$  decay (assumed 100% gamma-ray emission probability and 2.2% total  $\alpha$ ).

28 y 3 ([1959So12](#)) using the parent-daughter activity of  $^{207}\text{Po}$  and  $^{207}\text{Bi}$ .

# Bi-207 half-life evaluation

## by Alan Nichols

### 1978Ya04: Yanokura et al., Nucl. Phys. A229 (1978) 92-98

Three different approaches were taken to measure the half-life of Bi-207.

(1). The absolute disintegration rate of At-211 in a purified sample was measured by means of a liquid scintillation counter, and a large volume of the same solution was used to study the gamma-ray decay of daughter Po-211 and Bi-207 with a heavily-shielded Ge(Li) detector, calibrated against IAEA standard  $\gamma$ -sources of Na-22, Mn-54, Co-57, Co-60, Ba-133 and Cs-137. The prominent 569.7-keV gamma ray was used to calculate the decay rate of Bi-207 (emission probability of 99.85% was used from Parsa and Markowitz, *J. Inorg. Nucl. Chem.* **36** (1974) 1429 (??)), with a theoretical total internal conversion coefficient of 0.0221 adopted for this E2 transition). Thus, the half-life value for Bi-207 was “evaluated” to be  $32.2 \pm 1.3$  years.

(2). A Bi-207 half-life of  $31.7 \pm 3.7$  years was determined from a source prepared for liquid scintillation counting, but after complete decay of At-211, whereby the large uncertainty was attributed to the poor detection efficiency of gamma counting this particular liquid sample (??).

(3). And finally, the half-life of Bi-207 was also determined from the EC/ $\alpha$  branching ratio, the emission probability of the 6868-keV  $\alpha$  transition from Po-211 to the 569.7-keV nuclear level in Pb-207, the half-life of At-211, and the decay probability of Bi-207 feeding the 569.7-keV nuclear level in Pb-207. A half-life value of  $33.4 \pm 0.8$  years was calculated via this method. The authors assigned the small uncertainty to the counting statistics involving the 569.7-keV gamma ray – this value was adopted as the definitive recommended half-life through rather nebulous reasoning (simply because the value was deemed to be the most accurate?).

Systematic uncertainties are ignored in this set of studies, and are difficult to extract from the contents of the paper. Furthermore, such issues as the data sources for the direct 569.7-keV gamma-ray study need to be re-assessed (emission probability and ICC(total)) to derive a new half-life value, rather than simply adopt the original value of  $32.2 \pm 1.3$  years. The half-life derived from the liquid sample should simply be discarded as seriously inaccurate. Finally, the half-life calculated from the EC/ $\alpha$  branching ratio and other derived nuclear data needs to be re-assessed (and discard if deemed inappropriate).

# Bi-207 half-life evaluation

by Alan Nichols

## 1990Al11: Alburger and Harbottle, *Phys Rev. C*41 (1990) 2320-2324

- An end-window gas-flow proportional counter was used to determine the decay of  $\beta^-$  radiation from two samples of Ti-44 and one sample of Bi-207. Consideration of the detailed and overall performance of this system can be found in Alburger *et al. Earth Planetary Sci. Letts.* **78** (1986) 168-176. Long-term drift in counter voltage was deemed to be of the order of less than 0.5 V (c.f. 25 V to achieve the equivalent of  $1\sigma$  statistical uncertainty); box pressure would have to vary by 0.15" compared with monitored changes of better than 0.03". Changes in temperature of 2°F would result in  $1\sigma$  standard deviation change in activity ratios, while a variation from 30% to 80% in the relative humidity would also cause a variance of  $1\sigma$  standard deviation. These latter parameters were only monitored close to the end of the earlier studies on Si-32/Cl-36 with the following observations: temperature fluctuated from 72.4 to 74.7°F, and average relative humidity varied between 35% to 76% - judged as unfortunate and important variations in any attempt to define SYSTEMATIC uncertainties. Fluctuations of the data points from a smooth exponential decay were observed that are approximately THREE times the statistical uncertainty, and the authors assigned this unusual behaviour to variations in the temperature and relative humidity. Uncertainties were also identified with the operating pressure for the system – judged by the authors as operational under somewhat lower conditions than optimum. Other considerations involved studies of restoration of operational stability (system required a week to re-stabilize of any power shut-down), and change to a new gas supply (no observable effect). One might judge an overall SYSTEMATIC uncertainty of the order of  $\pm 1.5$  for a value of 34.9 years, without consideration of source preparation, radionuclidic purity and stability.
- Clearly, the uncertainties quantified in this paper are only the STATISTICAL uncertainties from the relative activity measurements for Cl-36, Ti-44 and Bi-207 (Figs. 1, 2, 3, 4). A recommended value of 34.9(4) years is derived by the authors for the half-life of Bi-207.
- Consideration of a combination of systematic and statistical uncertainties could result in a significant adjustment to  $34.9 \pm 2.0$  years. However, there are a number of imponderables in this analysis that can be seen to justify the rejection of the half-life value from this particular study by the original 207 mass chain evaluators.

# Bi-207 half-life evaluation

by Alan Nichols (cont.)

## 1991Li10: Lin and Harbottle, *J. Radioanal. Nucl. Chem.* 153 (1991) 51

- Note same common author for 1990 and 1991 publications (Harbottle).
- An inadequate paper, with insufficient detail and lack of clear traceability. Used gamma-ray spectroscopy to monitor the disintegration rates of individual gamma rays, and calculated half-life data from a combination of these disintegrations rates, “known” gamma abundances and detector efficiency curve. Measured gamma-ray abundances are compared with equivalent data from the NBS certification of the Bi-207 source, and recommendations to be found in *Nucl. Data Sheets* **43** (1984) 383.
- Interestingly, three half-life values are quoted in this paper:
  - (1).  $31.6 \pm 0.7$  years from “only” the major 569-keV gamma line;
  - (2).  $32.7 \pm 0.7$  years from the 569- and 1063-keV gamma lines;
  - (3).  $32.7 \pm 0.8$  years from the 569-, 1063- and 1770-keV gamma lines.
- There is an argument to be made for just adopting the half-life value of  $31.6 \pm 0.7$  years, although a reasonable understanding of the recommended uncertainty is required (and is judged to be unrealizable).

# Bi-207 half-life evaluation

by Alan Nichol (cont.)

- **1992Un01: Unterweger et al., Nucl. Instrum. Methods Phys. Res. A312 (1992) 349-352**
- **2002Un02: Unterweger, Appl. Radiat. Isot. 56 (2002) 125-130**
- Represent a small part of a long-term NBS/NIST exercise to monitor, characterise and revise the decay half-lives of an extensive list of radionuclides maintained and stored within NIST. These studies have been ongoing for approximately five decades, based on measurements by means of  $4\pi\gamma$  pressurized ionization chambers and (more recently) high-resolution HPGe detectors.
- Both of these papers lack sufficient detail, but refer to detailed descriptions and equipment and techniques to be found in NBS Special Publication 626 (1982) 85 and NBS Special Publication 250-10 (1987). However, specific systematic uncertainties are noted, such as the lower response of the ionization chambers that was believed to arise from instabilities in the old battery pack, and improvements noted after the vibrating reed electrometer and capacitor bank were replaced with a multi-range electrometer. Other unexplained changes also occurred periodically in the response of the ionization chamber to radium references sources prior to 1973.
- The 1992 publication contains a recommended half-life for Bi-207 of  $11523 \pm 18$  days which is equivalent to  $31.55 \pm 0.05$  years (1 year (mean tropical year)  $\equiv$  365.2422 days), which had only been followed for 0.6 half-lives ( $\sim$  19 years). Uncertainties are quantified in terms of Statistical Uncertainty (10.0) and Other Uncertainty (16.0), although I am uncertain as to what these numbers really mean.
- The 2002 publication contains a recommended half-life for Bi-207 of  $11523 \pm 15$  days which is equivalent to  $31.55 \pm 0.04$  years (1 year  $\equiv$  365.2422 days), which had been followed for 0.9 half-lives ( $\sim$  28 years). Uncertainties are quantified in terms of Statistical Uncertainty (9) and Other Uncertainty (12), although I am uncertain as to what these numbers really mean.

# Bi-207 half-life evaluation

by Alan Nichol (cont.)

## Concluding Remarks

I would recommend discarding:

- half-life (2) from 1978Ya04: Yanokura et al;
- half-life of 1990Al11: Alburger and Harbottle;
- half-lives (2) and (3) of 1991Li10: Lin and Harbottle;
- ignore 1992Un01 half-life of Unterweger *et al.* (replaced by recommended 2002Un02 value).
- Earlier (1972Ru10, 1961Ap01, 1959So12) half-life measurements are significantly less accurately characterised, and have not been discussed or assessed in this exercise.

Rework and accept half-lives (1) and (3) from 1978Ya04 (however, may still discard re-worked half-life (3)).

Accept half-life (1) of 1991Li10

Accept 2002Un02 half-life of Unterweger.

- Deduce “adopted” half-life from above three (or four) values: left as an exercise!

## Ba-139 half-life (in minutes)

84.547(15): 1989Ab05:  $\gamma$  counting  $t=20$  minutes ? (may be rejected) (Strasbourg)

84.44(22): 1985An25:  $\gamma$  counting,  $t\sim 6$  half-lives

84.63(34): 1972Ho01:  $\beta$  counting, prop. counter

85.2(8): 1969Su01

85(3): 1969Ke06

85(1): 1960Wi10, 1953Pa25

84.0(2): 1957Ba16

85.0(5): 1950Di04

84(1): 1948Sh27

85.6: 1937Po03

**83.06(28):** 1980Ge04:  $\gamma$  counting,  $t\sim 7$  half-lives: [most details \(Idaho Falls\)](#)

82.71(18): 1972Em01:  $\beta$  counting,  $t\sim 2.4$  half-lives, GM counter

82.9(2): 1962Fr04:  $\beta$  counting, scin. Counter

82.9(1): 1958Bu04

### **New (2010-2011) measurements:**

83.25(8):  $\gamma$  counting ([preprint from Ken Krane: March 22, 2011](#)) : several samples over  $t=4$  half-lives

83.6(6):  $\gamma$  counting (preliminary value at BARC, Mumbai, L. Danu et al., October 2010, analysis in progress) : experiment proposed by B. Singh based on the on-going  $A=139$  evaluation. Enriched Ba-138 material provided by McMaster. This experiment is expected to produce a better coincidence data.



1989Ab05: J. Radioanalytical and Nuclear Chemistry letters 135, 1  
(Strasbourg group)

PRECISION MEASUREMENTS OF THE HALF-LIVES OF NUCLIDES

TABLE 2

Half-lives of nuclides in this work compared with previous values

Nuclide	Half-life, present work	Number of runs	Gamma-ray analyzed, keV	Half-life, previous value	References
<sup>24</sup> Na	15.027±0.002 h	3	1369 2754	15.020±0.007 h	1
<sup>31</sup> Si	157.474±0.012 min	4	1266	157.3±0.3 min	1
<sup>49</sup> Ca	8.718±0.007 min	2	3085	8.715±0.023 min	2
<sup>52</sup> V	3.743±0.005 min	3	1434	3.75±0.01 min	3
<sup>139</sup> Ba	84.547±0.015 min	4	166	84.6±0.4 min	4
<sup>142</sup> Pr	19.140±0.002 h	4	1576	19.12±0.04 h	5
<sup>153</sup> Sm	46.70±0.05 h	5	103	46.7±0.1 h	6
<sup>159</sup> Gd	18.479±0.004 h	3	364	18.56±0.08 h	7
<sup>165</sup> Dy	2.334±0.001 h	3	95	2.334±0.006 h	8
<sup>166</sup> Ho	26.827±0.005 h	2	81	26.80±0.02 h	9
<sup>175</sup> Yb	4.185±0.001 d	2	283 396	4.19±0.01 d	10
<sup>187</sup> W	24.000±0.004 h	2	686	23.9±0.1 h	11
<sup>233</sup> Th	22.30±0.02 min	3	87	22.3±0.1 min	12
<sup>239</sup> U	23.44±0.02 min	3	75	23.50±0.05 min	13

ABZOUZI et al.: PRECISION MEASUREMENTS OF HALF-LIVES

## Au-198 half-life

- Discussed at NSDD-09
- Paper accepted in *App. Rad. & Isotopes*, see attached copy

# Author's Accepted Manuscript

Evaluation of half-life of  $^{198}\text{Au}$

Jun Chen, Scott D. Geraedts, Christian Ouellet,  
Balraj Singh

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Reference: ARI5355

To appear in: *Applied Radiation and  
Isotopes*

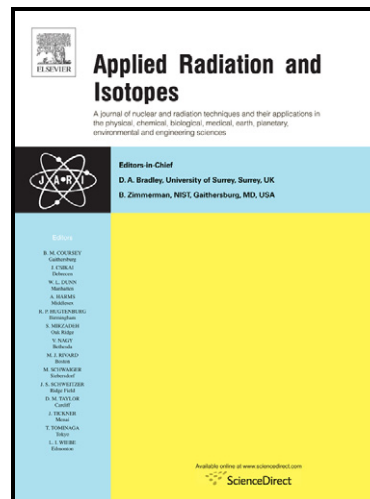
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# Evaluation of half-life of $^{198}\text{Au}$

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## Abstract

The half-life of the important radioisotope  $^{198}\text{Au}$  is evaluated by using several different statistical methods, such as Weighted Mean (WM), Limitation of Statistical Weights Method (LWM), Normalized Residuals Method (NRM), Rajeval Technique (RT), Bootstrap Median (BM) and Mandel-Paule Method (MP). After comparing and analyzing the results from these methods, a final value of  $2.6948 \pm 0.0012$  d is recommended based on the stability and sensitivity of the methods and the resulting uncertainties.

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## 1. Introduction

$^{198}\text{Au}$  is an important radioisotope for medical physics and basic physics research. Radiation treatment of cancer through injection of  $^{198}\text{Au}$  seeds requires precise and accurate knowledge of the half-life for therapeutic dosage. Additionally the  $\gamma$ -line from the decay of  $^{198}\text{Au}$  is the gold standard for  $\gamma$ -ray energy calibration, known to an extreme precision of  $411\,802.05 \pm 0.17$  eV (Helmer and van der Leun (2000)). Recent studies (Raiola et al. (2005), Spillane et al. (2007), Jenkins et al. (2009), Jenkins et al. (2010)) have shown dependence of the half-life of isotopes that decay by  $\alpha$ ,  $\beta^+$ ,  $\beta^-$ -decay and electron capture on temperatures, host materials and the earth-to-sun distance.  $^{198}\text{Au}$  was quickly understood as an ideal radioisotope for precision

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measurements of potential temperature related effects (Goodwin et al. (2007); Spillane et al. (2007); Kumar et al. (2008); Ruprecht et al. (2008); Fortak et al. (2010)). Ruprecht et al. (2008) investigated the predictions of temperature dependence of the decay half-life of  $^{198}\text{Au}$  and concluded with the remark: “(...) show measurements of the last 40 years with an error less than 0.005 d, so it seems a re-evaluation of the recommended half-life is called for.” In response to above comment by Ruprecht et al. (2008), the present re-evaluation of the half-life of  $^{198}\text{Au}$  was carried out which also demonstrates the use of a variety of widely used statistical methods for dealing with a large set of independent measurements, some of which may be discrepant.

## 2. Data Selection and Statistical Methods

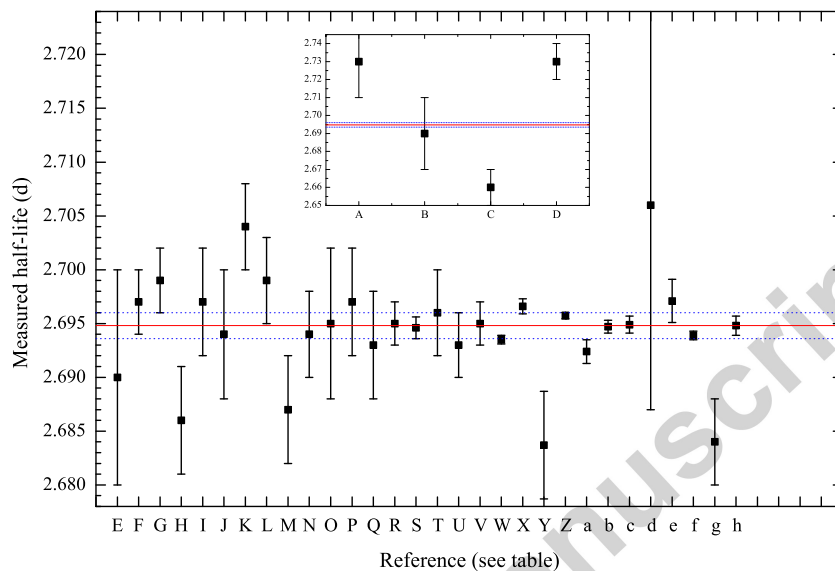
Table 1 gives a detail of the experimental measurements made of the half-life of  $^{198}\text{Au}$  over the past 75 years. All literatures on half-life measurement of  $^{198}\text{Au}$  have been scanned and studied carefully. Ultimately measurements were omitted only on the criteria that the same experimental group made a more precise measurement using the same apparatus (removing correlations), or the data were not assigned an uncertainty which many of the statistical data analysis techniques require, or the assigned uncertainties are unrealistically low or high. In particular, we omitted the values quoted by Saxon (1948) and Unterweger et al. (1992) because the authors of those papers later published updated values (Saxon and Heller (1949) and Unterweger and Lindstrom (2004)), using the same technique. The measurements used in our analysis are shown in Figure 1.

Ideally one would want to have the true value of a physical constant, whereas in practice experimenters make increasingly accurate and precise measurements approaching that true value. There is nonetheless some variation in the measurements, and occasionally a discrepancy but by and large measured values tend to converge. When the discrepancies are few, the weighted average is the established appropriate technique to use, specifically if the reduced chi-squared approaches unity ( $\chi^2/(N - 1) \approx 1$ ). For the purposes of determining whether data are discrepant the reduced chi-square of the weighted average must be significantly larger than 1.

The statistical techniques used in this paper and listed here are the same as those used by MacMahon et al. (2004) for half-life evaluations of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . A brief description of each method follows: an unweighted mean, a weighted mean, limitation of relative statistical weights method (LWM),

Reference	$T_{1/2}$ (d) ( $\Delta T_{1/2}$ )	Technique (instrument)
Amaldi et al. (1935)	2.7*	$\beta$ counting
McMillan et al. (1937)	2.7*	$\beta$ counting
Pool et al. (1937)	2.5*	$\beta$ counting
Sherr et al. (1941)	2.7*	$\beta$ counting
Ward et al. (1946)	2.73(2)	ionization chamber
Seren et al. (1947)	2.7*	$\beta$ counting, Geiger counter
Saxon (1948)	2.66(1)*	$\beta$ counting, magnetic spectrometer
Saxon and Heller (1949)	2.69(2)	$\beta$ counting, magnetic spectrometer
Steffan et al. (1949)	2.7*	$\beta$ and $\gamma$ counting
Cavanagh et al. (1951)	2.66(1)	$\beta$ counting, Geiger Counter
Sinclair and Holloway (1951)	2.73(1)	$\beta$ counting, liquid counter, electroscope
Silver (1951)	2.69(1)	ionization chamber, electrometer
Lockett and Thomas (1953)	2.697(3)	ionization chamber
Bell and Yaffe (1954)	2.699(3)	ionization chamber
Tobailem (1955b)	2.686(5)	$\gamma$ counting, ionization chambers
Tobailem (1955a)	2.686 (5)*	$\gamma$ counting, ionization chambers
Johansson (1956)	2.697(5)	$\gamma$ timing, NaI(Tl) detector
Sastre and Price (1956)	2.694(6)	Geiger counter
Keene (1958)	2.704(4)	Ionization chamber
Robert (1960)	2.699(4)	Calorimetry on gold samples
Starodubtsev et al. (1963)	2.687(5)	$\beta$ counting, $\beta$ spectrometer
Anspach et al. (1965)	2.694(4)	$4\pi$ , $2\pi$ ionization chambers, proportional counter
Goodier (1968)	2.695(7)	$4\pi$ ionization chamber
Lagoutine et al. (1968)	2.697(5)	ionization chambers, $4\pi$ $\beta$ counters, $\gamma$ counter
Reynolds et al. (1968)	2.693(5)	end window Geiger counter
Vuorinen and Kaloinen (1969)	2.695(2)	$4\pi$ $\beta$ - $\gamma$ coincidence counter
Cabell and Wilkins (1969)	2.6946(10)	$\gamma$ counting
Costa Paiva and Martinho (1970)	2.696(4)	$\gamma$ counting, NaI(Tl) detector
Debertin (1971)	2.693(3)	$\gamma$ counting, Ge(Li) detector
Hoppes et al. (1982)	2.695(2)	$4\pi$ ionization chamber, $\gamma$ counting
Rutledge et al. (1982)	2.6935(4)	$\gamma$ counting, GeLi detector
Abzouzi et al. (1990)	2.6966(7)	$\gamma$ counting, GeLi detector
Unterweger et al. (1992)	2.69517(21)*	$4\pi$ ionization chamber
Mignonsin (1994)	2.6837(50)	$\gamma$ counting, GeLi detector
Unterweger and Lindstrom (2004)	2.69573(30)	$4\pi$ ionization chamber
Lindstrom et al. (2005)	2.6924(11)	$4\pi$ ionization chamber, $\gamma$ counting, Ge detector
Novkovic et al. (2006)	2.6947(6)	$\gamma$ counting, Ge detector
Goodwin et al. (2007)	2.6949(8)	$\gamma$ counting, Ge detector; 2.6953(8) at 19 K
Spillane et al. (2007)	2.706(19)	$\gamma$ counting, Ge detector; 2.802(20) at 12 K
Kumar et al. (2008)	2.6971(20)	$\gamma$ counting, Ge detector; 2.6976(23) at 12.5K
Ruprecht et al. (2008)	2.6939(4)	$\gamma$ counting, 2 Ge detectors; 2.6935(5)-10K
Fortak et al. (2010)	2.684(4)	$\gamma$ counting, Ge detector; 2.687(5)-12K
Goodwin et al. (2010)	2.6948(9)	$\gamma$ counting, Ge detector
Lindstrom et al. (2010)	2.6910(4)*	$\gamma$ counting, Ge detector

Table 1:  $^{198}\text{Au}$  half-life data at room temperature from literature. \* indicates the value omitted from the current analysis. Uncertainty in brackets is on last digit(s).



Label	Reference	Label	Reference
A	Ward et al. (1946)	R	Vuorinen and Kaloinen (1969)
B	Saxon and Heller (1949)	S	Cabell and Wilkins (1969)
C	Cavanagh et al. (1951)	T	Costa Paiva and Martinho (1970)
D	Sinclair and Holloway (1951)	U	Debertin (1971)
E	Silver (1951)	V	Hoppes et al. (1982)
F	Lockett and Thomas (1953)	W	Rutledge et al. (1982)
G	Bell and Yaffe (1954)	X	Abzouzi et al. (1990)
H	Tobalem (1955b)	Y	Mignonsin (1994)
I	Johansson (1956)	Z	Unterweger and Lindstrom (2004)
J	Sastre and Price (1956)	a	Lindstrom et al. (2005)
K	Keene (1958)	b	Novkovic et al. (2006)
L	Robert (1960)	c	Goodwin et al. (2007)
M	Starodubtsev et al. (1963)	d	Spillane et al. (2007)
N	Anspach et al. (1965)	e	Kumar et al. (2008)
O	Goodier (1968)	f	Ruprecht et al. (2008)
P	Lagoutine et al. (1968)	g	Fortak et al. (2010)
Q	Reynolds et al. (1968)	h	Goodwin et al. (2010)

Figure 1: Half-life measurements used in this evaluation. Our accepted value is represented by the solid horizontal line, and its uncertainty is represented by the dotted lines. The references in the figure are labeled with letters, their corresponding references listed in the table. (*color online*)

normalized residuals method (NRM), Rajeval technique (RT), the bootstrap median (BM) and the Mandel-Paule Method (MP). The weighted and unweighted means can be found in any textbook on the subject. For  $N$  data points  $x_i$  with standard deviation  $\sigma$ :  $x_u = \sum x_i / N$ ,  $\sigma_u = \sqrt{\frac{\sum (x_i - x_u)^2}{N(N-1)}}$ , and  $x_w = \frac{\sum x_i w_i}{\sum w_i}$ ,  $\sigma_w = \sqrt{\frac{1}{\sum w_i}}$  where  $w_i = 1/\sigma_i^2$ .

LWM is a procedure adopted by the IAEA in the Coordinated Research Program on X-ray and  $\gamma$ -ray decay data standards (Nichols (2004)). It is designed to prevent one very precise measurement from dominating the calculation of the mean, which is possible in a normal weighted mean algorithm. A value which has a statistical weight  $\frac{w_i}{\sum w_i}$  greater than 0.5 within a dataset is identified, and the uncertainty of this value is increased until the relative weight has dropped to 0.5. The procedure is also used to compare the unweighted mean with the weighted mean and if they overlap ( $|x_u - x_w| \leq \sigma_u + \sigma_w$ ), the weighted mean is adopted as the recommended value. If they do not overlap, the unweighted mean is adopted. In either case the uncertainty of the adopted value is increased to cover the most precise value in the data.

NRM is a prescription that increases and decreases the weight of individual data based on how closely separated they are from the bulk of the data, effectively increasing the uncertainty of outliers (James et al. (1992)). A normalized residual for each value is calculated as follows:  $R_i = \sqrt{\frac{w_i W}{(W - w_i)}} (x_i - \bar{x})$ , where  $W = \sum w_i = \sum 1/\sigma_i^2$ . If any  $R_i$  is greater than  $R_0 = \sqrt{1.8 \ln N + 2.6}$  the weight of the value with largest  $R_i$  is reduced until it equals  $R_0$ . Once no value has  $R_i > R_0$  then a weighted mean is calculated with the adjusted weights.

RT is an involved technique similar to the NR method (Rajput and MacMahon (1992)) that begins by rejecting outliers.  $y_i = \frac{x_i - x_{ui}}{\sqrt{\sigma_i^2 - \sigma_{ui}^2}}$  is calculated for each value where  $x_{ui}$  and  $\sigma_{ui}$  are the unweighted mean and standard deviation of the whole dataset excluding the value  $i$  in question. Any value with  $|y_i| > 5.88$  is rejected as an outlier. The technique then searches for discrepant data, calculating for each value the standard deviate  $Z_i = \frac{x_i - x_w}{\sqrt{\sigma_i^2 - \sigma_w^2}}$  and the probability integral  $P(Z_i) = \int_{-\inf}^{Z_i} \frac{1}{\sqrt{2\pi}} \exp(-\frac{t^2}{2}) dt$ . The absolute difference between  $P(Z_i)$  and 0.5 is the central deviation and if  $|P(Z) - 0.5| > (0.5)^{N/(N-1)}$  the value is deemed discrepant. The uncertainties of discrepant values are then adjusted to  $\sigma_i = \sqrt{\sigma_i^2 + \sigma_w^2}$ . A weighted mean



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9 is calculated once all discrepant data have had their uncertainties adjusted.

10 All of the above methods attempt to use the mean of a set of data to  
11 determine the actual value of the quantity which has been measured. How-  
12 ever, there is another measure of the actual value, the median, which can  
13 also be applied to a set of data. When all the data points are sorted from  $x_1$   
14 to  $x_i$ , the median is the central value  $x_{i/2}$  for an odd number of points, and  
15  $\frac{x_{i/2} + x_{i/2+1}}{2}$  for an even number of data points. The median is less sensitive  
16 to outliers than the mean, but it also has several drawbacks. It does not  
17 take into account uncertainties on the input data points, nor does it have an  
18 easily-computable uncertainty. In addition, the value of the median is usually  
19 limited to being the same as the value of one of the input data points. We  
20 can overcome these obstacles by employing a bootstrap method, as described  
21 by Helene and Vanin (2002) and Efron and Gong (1983). This method has  
22 been well known in epidemiology and textbooks on statistics in medical and  
23 social sciences, but has been relatively unknown in physics data analysis.  
24 The method of Helene and Vanin (2002) does not include uncertainties on  
25 the data points. Therefore we have written a code with the uncertainty of  
26 each data point taken into account based on an algorithm as follows. For  
27 each input data point, we generate a large array of values which fits a normal  
28 distribution. This distribution is centered around the input data point, and  
29 its width is determined by the uncertainty on the data point. By combining  
30 these distributions together, we obtain a large array of possible data points.  
31 We then select N data points, with replacement, from this large dataset. We  
32 compute the median of these N points. By repeating this procedure a large  
33 number of times (in this case 800,000 times), we obtain a distribution of me-  
34 dian measurements. The bootstrap median and its uncertainty can then be  
35 calculated by finding the average and standard deviation of this distribution.

36 There is another method for estimation of a common mean named Mandel-  
37 Paule Method (Rukhin and Vangel (1998)), which is a simpler procedure of  
38 the Maximum Likelihood Estimation and has been used in inter-laboratory  
39 comparisons of reference standards. This approach uses weights of the form  
40  $w_i = \frac{1}{y+t_i^2}$  where  $y$  is the estimator of the variance and  $t_i$  refers to the standard  
41 error associated with each input data which is replaced by the uncertainty  
42 in our case. The essential in this procedure is to solve  $y$  from the equation  
43  $\sum_i^p w_i(x_i - \tilde{x})^2 = p - 1$  where  $p$  is the total number of data points. The  
44 Mandel-Paule mean is the weighted mean with the weights calculated from  
45  $y$  and experimental uncertainties and its uncertainty as the square root of  $y$ .

We have written a computer code for this method based on the algorithm in Rukhin and Vangel (1998).

The computer codes for all of these techniques except for the Bootstrap Method and Mandel-Paule are freely available on the internet in the form of a program avetools at <http://people.physics.anu.edu.au/~txk103/> courtesy of T. Kibedi.

### 3. Results and Discussion

The final values for each technique can be seen in Table 2 as well as graphically in Figure 2. In Table 3 are the values from the Evaluated Nuclear Structure Data File (ENSDF) (Huang (2009)), the NUBASE evaluation of decay properties (Audi et al. (2003)), the Nuclear Wallet Cards (Tuli (2005, 2010)) and the Decay Data Evaluation Project (Schönfeld and Dersch (2004)). NUBASE and 2010 Wallet Cards are based on a measurement by Unterweger et al. in 1992, the wallet card on measurements up to 2005 and the DDEP value is derived from 20 values but only considering data up to 1994. For comparison, recent values from some low-temperature measurements are given in Table 4, essentially to show that there is no effect of temperature on the half-life.

Statistical Method	$T_{1/2}(d)(\Delta T_{1/2})$	$\chi_{reduced}^2$
Present evaluation		
Unweighted Mean	2.6954(20)	
Weighted Mean	2.6948(3)	2.79
Limitation of Statistical Weights	2.6948(10)	2.84
Normalized Residuals Method	2.6946(3)	2.43
Rajeval Technique	2.6944(2)	1.85
Bootstrap Median	2.6948(7)	2.85
Mandel-Paule Approach	2.6945(55)	2.90

Table 2: Values from various techniques.

We can see that the different statistical methods of arriving at an accepted value all produce slightly different, though similar results. We believe that the Unweighted Mean is not as accurate as the other methods. The essential

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 Previous Evaluations/Compilations
 

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ENSDF (Huang (2009))	2.6947(3)
NUBASE (Audi et al. (2003))	2.69517(21) <sup>a</sup>
Wallet Card(published, Tuli (2005))	2.6956(3)
Wallet Card(online, Tuli (2010))	2.69517(21) <sup>a</sup>
DDEP (Schönfeld and Dersch (2004))	2.6944(8)

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<sup>a</sup> from Unterweger et al. (1992).

Table 3: Some recent evaluations of the half-life

difference between our methods is how they deal with the uncertainties of the input data points. This difference is illustrated in Table 5. We can see that inclusion of the most precise point (2.69573(30), given by Unterweger and Lindstrom (2004)) has varying effects on the different methods. Some

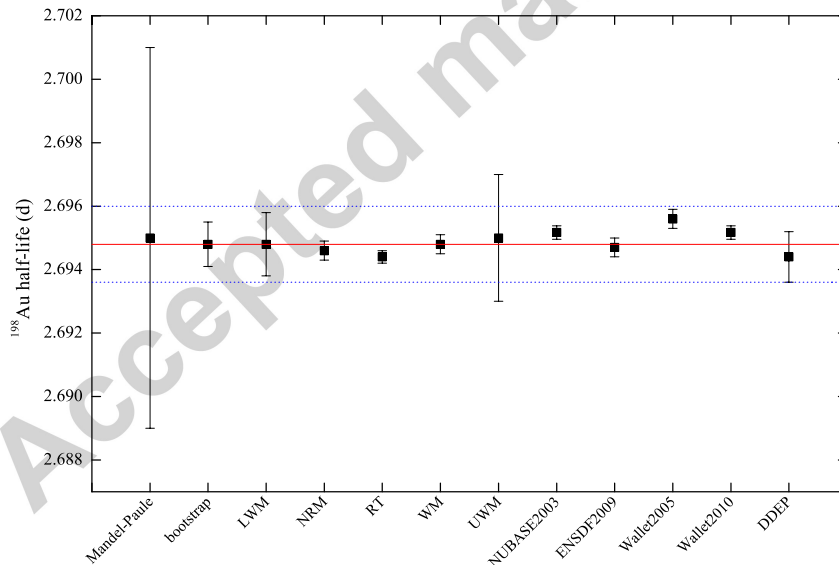


Figure 2: The results of the different statistical methods used in the current work and compared with the recent evaluations. Our recommended value is represented by the solid line, with the lines above and below this value representing the uncertainty. (*color online*)

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Low-temperature measurements

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Spillane et al. (2007) at 12K	2.802(2)
Goodwin et al. (2007) at 19K	2.6953(3)
Ruprecht et al. (2008) at 10K	2.6935(5)
Kumar et al. (2008) at 12.5K	2.6976(23)
Fortak et al. (2010) at 12K	2.687(5)

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Table 4: Recent low-temperature measurements, though these are not included in the averaging procedures used in the present work, show that there is no significant change in the half-life with temperature. The value of Spillane et al. (2007) shows a significant effect which is believed to be unreliable, however, their work prompted several other studies.

of the methods exhibit a change that is larger than the uncertainty on the value. This is a problem in our situation because different groups often use different methods to calculate uncertainties. Though uncertainty is an important measure of how accurate a data point is, in situations where we are comparing measurements taken by a variety of different groups we may not want to give too large a weight to a particular value just because it is very precisely quoted. We can see that both the Bootstrap Median method and Mandel-Paule are less sensitive to the inclusion or exclusion of the very precise measurement. This sensitivity is also exhibited in Figure 3. This shows the output of the various averaging methods as more data points are included chronologically. We can see that when we have not included several of the most recent points, the results from these two methods are already quite close to its final value. The other methods continue to change significantly as more precise data points are included. We see that in this case, the Bootstrap Median and Mandel-Paule are the more stable method, and they have the greatest ability to predict the final value of a set of data when only a part of the set of data is studied. However, it can be seen that the uncertainty given by the Mandel-Paule method is unrealistically high, compared with the values from other methods as well as with some recent precise measurements. Therefore, the result from the Bootstrap Median is recommended.

Based on above analysis, we have chosen an accepted value and uncertainty of  $2.6948 \pm 0.0007$  d from Bootstrap Median with the  $\chi^2_{reduced} = 2.85$  for the fit to the data points, which also encompasses the values produced by the other methods. Following the policy of Particle Data Group (Nakamura (2010)), a scaling factor of  $\sqrt{\chi^2_{reduced}}$  is introduced to the uncertainty when

$\chi_{reduced}^2 > 1$ , which therefore increases our uncertainty to 0.0012. Nyikos et al. (1973) found a 0.0101(3) % change due to the influence of chemical environment on the half-life of  $^{198}\text{Au}$ . Recently an upper limit of  $<0.05\%$  was proposed by Goodwin et al. (2010), which combines the effects on the half-lives due to temperatures and host materials. Our recommended value of  $2.6948 \pm 0.0012$  d encompasses the change of 0.05%, which is represented by a horizontal line in Figure 1 and Figure 2 and overlaps with all the results from precise measurements as seen in Figure 1.

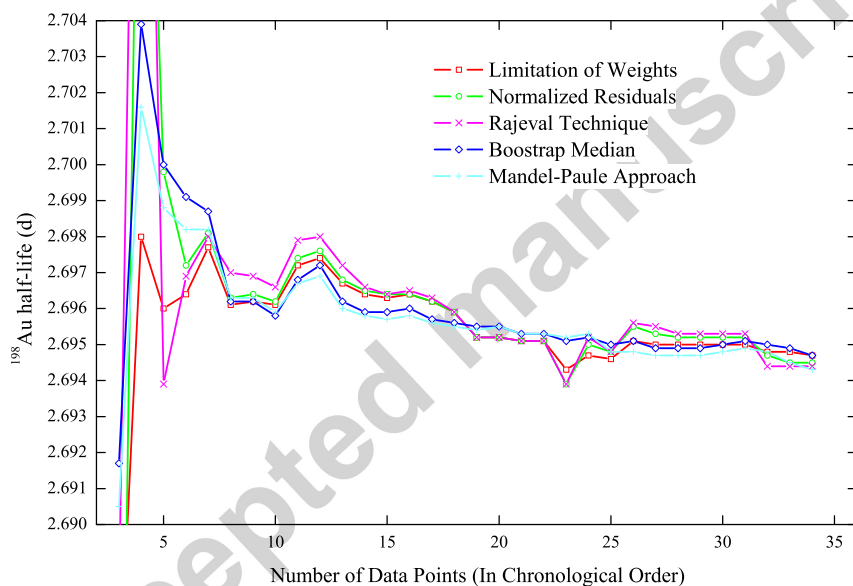


Figure 3: The output of the various statistical methods as more data points are included. We see that the Bootstrap Median and Mandel-Paule average are less sensitive to the effects of precise data points than the other methods. (*color online*)

The ENSDF (Huang (2009)) value of 2.6947(3) d is in agreement with ours, except for the uncertainty. We believe that the uncertainty in ENSDF is too low to be realistic in view of possible chemical and other effects. Moreover, ENSDF does not have the four most recent measurements of Kumar et al. (2008), Ruprecht et al. (2008), Goodwin et al. (2010) and Fortak et al. (2010). The ENSDF evaluation also used only the Rajeval technique. Using

our set of measurements, the Rajeval technique returns 2.6944(2) d, where we believe that the uncertainty of about 0.001% is unrealistically low.

Method	Value including the most precise data point	Value excluding the most precise data point	Difference
Weighted Mean	2.69475(29)	2.69429(33)	0.00046
Limitation of Statistical Weights	2.69475(98)	2.69429(80)	0.00046
Normalized Residuals	2.69455(29)	2.69424(31)	0.00031
Rajeval Technique	2.69438(21)	2.69417(21)	0.00021
Bootstrap Median	2.69485(68)	2.69475(72)	0.0001
Mandel-Paule Approach	2.69452(554)	2.69446(577)	0.00006

Table 5: The output of the various statistical methods, including and excluding the very precise measurement given by Unterweger and Lindstrom (2004). The different methods are seen to have different sensitivities to this measurement, and the Bootstrap and Mandel-Paule are the least sensitive methods.

#### 4. Conclusion

We have analyzed all the measurements of the half-life of  $^{198}\text{Au}$  using a variety of statistical methods. Based on our results, we recommend a value of  $2.6948 \pm 0.0012$  d for this half-life which overlaps the values from all the precise measurements. Of the various methods, we found that the Bootstrap Median method is robust and least sensitive to the inclusion of very precise data points and gives the realistic uncertainty. This procedure could be advantageous in situations where one does not have complete information about the assignment of uncertainties in the experimental data points used in the analysis.

#### 5. Acknowledgements

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