A protocol for uncertainty assessment of half-lives

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Half-life Measurements

- Half-life determination by following the decay of a radioactive source
- The problem of data discrepancy; examples
- New procedure for uncertainty calculation
Example: decay of $^{55}$Fe

$$T_{1/2}=1005.0 \text{ d } \pm 1.4\text{d}$$
Half-life and uncertainty from least-squares fit

$<T_{1/2}>$(d) vs. time (d)

=> fit underestimates uncertainty!
Residuals $^{55}$Fe: blow up

Van Ammel et al., ARI (2006)

uncertainty only including counting statistics
$\Rightarrow$ does not fully account for spread of data
Autocorrelated data => not stochastic

autocorrelation plot of the residuals

\[ r_k = \frac{\sum_{t=1}^{N-k} (Y_t - \bar{Y})(Y_{t+k} - \bar{Y})}{\sum_{t=1}^{N} (Y_t - \bar{Y})^2} \]
High, medium, low frequency instabilities

- noise,
- counting statistics, …
- => random effects

- geometrical reproducibility,
- ‘seasonal’ effects,
- short-lived impurity, …

- dead time,
- detector/source degradation,
- background subtraction, …
Intermediate conclusion

• fitted parameters are only meaningful if the model rigorously applies to the data

• hence the model should include all possible medium and long-term instabilities

• common statistical tests (uncertainty!) require randomness of data; they do not apply to autocorrelated data

• the uncertainty derived from a fit is unreliable if these conditions are not fulfilled
Examples from literature
Half-life of $^{134}\text{Cs}$
Half-life of $^{109}\text{Cd}$

A selection of the 'best' data

- Vaninbrouck
- Lagoute
- Schrader
- Unterweger
- Martin
‘Trend analysis’ of residuals

short-term

medium-term

long-term
Residuals Ba-133

data scatter exceeds uncertainty
=> unidentified **HIGH FREQUENCY** component
Residuals Cs-134

MEDIUM FREQUENCY instability
=> fit underestimates uncertainty
Residuals Ba-133 and Eu-152

‘independent’ measurements, yet positively correlated

=> instability is not stochastical

NIM A390 (1997) 267-273
Residuals Ce-144

sign of LOW FREQUENCY deviation
=> fit tends to minimise it
Residuals $^{99}\text{Tc}^m$

(a) Systematic deviation for short times

(b) Electrometer range switching

ARI 60 (2004) 317-323
Residuals Y-88

only a few measurement data
=> LACK OF INFORMATION
An alternative procedure

- Quantify all sources of instability
- Determine their rate of change
- Apply uncertainty propagation
- Sum all components independently
An alternative data analysis method that should lead to a realistic uncertainty budget
Protocol for uncertainty assessment of half-lives

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(Received March 2, 2007)

The apparent tendency to underestimate the uncertainty of experimentally determined half-life values of radionuclides is discussed. It is argued that the uncertainty derived from a least-squares analysis of a decay curve is prone to error. As it is quite common for a series of activity measurement results to be autocorrelated, the prerequisite of randomness of data for common statistical tests to apply is not fulfilled. In this work, an alternative data analysis method is applied that leads to a more realistic uncertainty budget. The uncertainty components are being subdivided in three categories according to the relative frequency at which they occur, an appropriate uncertainty propagation formula applied and then the total uncertainty obtained from an independent sum. An attempt is made to apply the protocol to problematic cases in literature, yet it is clear that the reporting is usually incomplete for a full uncertainty analysis. Suggestions are made for a concise but more complete reporting style, for the sake of traceability.
consider START and STOP

\[ T_{1/2} = (T_2 - T_1) \frac{\ln 2}{\ln(A_1 / A_2)} \]

\[ \sigma(T_{1/2}) = \frac{1}{\lambda T} \sqrt{\frac{\sigma^2(A_1)}{A_1^2} + \frac{\sigma^2(A_2)}{A_2^2}} \]

\[ T = T_2 - T_1 \]
Uncertainty Propagation

• Quantify all sources of uncertainty, $\sigma(A)$, and their ‘frequency’ of occurrence, $n$

• Apply uncertainty propagation formula:

$$
\frac{\sigma(T_{1/2})}{T_{1/2}} \approx \left\{ \frac{2}{\lambda T} \sqrt{\frac{2}{n + 1}} \right\} \frac{\sigma(A)}{A}
$$

$n$= the number of occurrences of the effect

conservative value $n=1$ for medium and low frequencies

• Sum all components independently
Hypothetical case Fe-55

\[ T_{1/2} = 1005 \text{ d} \]
\[ T = 290 \text{ d} \]

\[ \lambda T = 0.2 \]

<table>
<thead>
<tr>
<th>type</th>
<th>( \sigma(A)/A )</th>
<th>n</th>
<th>factor</th>
<th>( \sigma(T_{1/2})/T_{1/2} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>high</td>
<td>1%</td>
<td>100</td>
<td>0.71</td>
<td>0.706%</td>
</tr>
<tr>
<td>medium</td>
<td>1%</td>
<td>5</td>
<td>2.90</td>
<td>2.897%</td>
</tr>
<tr>
<td>low</td>
<td>1%</td>
<td>1</td>
<td>5.02</td>
<td>5.018%</td>
</tr>
</tbody>
</table>
Realistic uncertainty

![Graph showing the decay of Fe-55 with time (d)].
Literature: lack of information

- most papers on measurement of $T_{1/2}$ contain insufficient information for a proper review
- sometimes result is given without description of experimental design
- too succinct reporting style even by reputed reference laboratories
- lack of transparency, traceability

$\Rightarrow$ need to redo undocumented experiments
$\Rightarrow$ incomplete report is lost information
What is essential information?

• description of the experiment

• any circumstance that is considered relevant for a traceable account of how the half-life value and its uncertainty were calculated

• result and uncertainty budget
What was measured and how?

• activity as count rate, current, ratio, …; which part of the decay (particle, energy range)?

• how many sources? which was their initial activity?

• which (type of) detector was used? at which efficiency?

• number of measurements performed?

• time period of measurement campaign?
How were the data analysed?

• **method of linearization**
  - e.g. by least squares fit: mention free and fixed parameters, explicitly state which statistical weights were assigned to the data!

• **which corrections were performed?**
  - radioimpurity correction
  - background subtraction
  - differentiate between stochastic uncertainty components and possible ‘systematic’ components (long-term component!)
Residual plot

- always present a detailed residual plot!
- avoid ‘cleanup’ of ‘outliers’; rather indicate them by a different symbol than the data that were used for the linearization process
- perform many measurement with excellent statistical accuracy, in order to increase the chance of observing and quantifying medium-term instabilities
Exhaustive uncertainty budget

• assemble a table with all identifiable sources of uncertainty

• separate components that should be visible in the residuals from the invisible ones

• the estimated amplitude of the short- and medium-term components should cover all visible deviations in the residuals

• be generous when estimating the (mostly invisible) effects of long-term instabilities
  – investigate thoroughly systematic effect by dead time
  – study and discuss possible non-linearity of detector response
Final Conclusions

Half-life measurements are **NOT TRIVIAL**!

- perform **many measurements** with good statistical accuracy and carefully study the **non-stochastic part** of the residuals

- identify and quantify **short, medium and low frequency instabilities** separately and apply a conservative propagation factor to $T_{1/2}$

- **report** in sufficient **detail**, for traceability