Uncertainty Assessment in the Application of the Triple-to-Double Coincidence Ratio Method

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Also: P. Cassette, L. Mo, C. Wätjen, B. Simpson, W. van Wyngaardt, R. Broda, P. Arenillas, and T. Altzitzoglou
Introduction

• Data comparison study carried out under auspices of ICRM LSCWG

• Goals
  – Study differences in calculation results from different TDCR analysis programs using a single set of data
  – Investigate differences in analysis techniques and uncertainty assessment philosophies between laboratories using the TDCR method
  – Study effect of not taking asymmetry of PMT efficiencies into account on calculated activity
Protocol

- Participation solicited from all laboratories using/planning to use TDCR
- 11 positive responses, received 8 sets of results
- A real TDCR data set from a recent NIST standardization was distributed to all participants on 23 June 2008
- Participants were given all necessary information to analyze data set
- Information requested
  - Activity concentration of solution, with and without accounting for phototube asymmetry, if possible.
  - Uncertainty budget
  - Details of analysis methods, programs used, assumptions made, etc.
- Reporting date was 1 September 2008
Participants

- T. Altzitzoglou (IRMM)
- P. Arenillas (CNEA)
- R. Broda (POLATOM)
- P. Cassette (LNHB)
- L. Mo (ANSTO)
- B. Simpson & W. van Wyngaardt (NMISA)
- C. Wätjen (IFIN)
- B. Zimmerman (NIST)
Distributed $^{99}\text{Tc}$ data set

- Two LSC sources of $^{99}\text{Tc}$, each prepared in 10 mL of Hionic Fluor with about 0.6 g of $10^{-3}$ mol/L KOH
- Data acquired in NIST TDCR system using MAC3 with 45 $\mu$s deadtime, 1 kHz clock pulse
- 600 s counting time for each source
- Two counting cycles per source
- Each counting cycle consisted of 4 repeated measurements with
  - 4 grey filters
  - 2 different focusing voltages for each filter
  - 32 data points per counting cycle
- The masses of the added $^{99}\text{Tc}$ were provided
- Data for an equivalently-prepared background were also provided (counted for 1000 s)
- Data format

Start time/date; Counts A,B,C,# Clock pulses; Counts in AB,BC,AC; Counts T; Counts D
$^{99}$Tc decay scheme

- $E_{\beta,\text{max}} = 293.5$ keV
- Second forbidden, non-unique
- Shape factor?
  - $q^2 + (0.54 \pm 0.02)p^2$
  - $q^2 + (0.3211 \pm x)p^2$
  - $q^2 + (1.6 \pm x)p^2$ (??)
  - $q^2 + p^2$ (default shape for second forbidden, non-unique in many codes)
Programs used

- TDCR-01 (POLATOM, no asymmetry): 2
- TDCR-02 (POLATOM, asymmetry): 2
- TDCR-02p (POLATOM, asymmetry, Polya): 1
- TDCR-06b (LNHB): 1
- TDCR-7 (LNHB): 1
- TDCR-11 (LNHB): 1
- Other homemade: 3
Results – with phototube asymmetry

$^{99}$Tc, TDCR models assuming unequal phototube efficiencies

$(k=1$ uncertainties)
Results – with equal phototube efficiencies

$^{99}$Tc TDCR models assuming equal phototube efficiencies

$C_A$ (kBq/g) ~2% 

(k=1 uncertainties)
Effect of taking into account phototube efficiency asymmetry

- $\varepsilon_{LSD}$ ranged from about 92 to 82 % (NIST calculation)
- Typical relative efficiencies $\varepsilon_A: \varepsilon_B: \varepsilon_C$ were 1, 0.68, 1.039
- Ratio of activities ($A_{Sym}/A_{Asym}$) ranged from 1.0035 to 1.0086
- Average ratio 1.0065 ($s=0.16 \%$)
- Ignoring asymmetry in this case always results in higher activity, because average efficiency always low
- Example: for one counting point, NIST-calculated $\varepsilon_A: \varepsilon_B: \varepsilon_C$ were 0.1119, 0.076, 0.1163; average $\varepsilon$ becomes 0.1015
- For low-efficiency cases in NIST TDCR, slopes of $\Delta\varepsilon_A, \varepsilon_B, \varepsilon_C/\Delta$TDCR are not equal
Does the choice of program matter?

$^{99}$Tc, TDCR models assuming unequal phototube efficiencies

![Graph showing TDCR models assuming unequal phototube efficiencies](image)
Choice of program

• Three different philosophies
  – LNHB/POLATOM (different implementations, perhaps diverged?)
  – EFFY-based
  – Independent (though mostly LNHB-based)
• Not enough data to conclude whether program or analysis method led to observed differences, but initial response would be “no”
• TDCRB-02 code: better result observed when assuming Polya distribution instead of Poisson – is this real?
• How does each program deal with shape factor?
Influence of shape factor?

Responses somewhat misleading: no difference in activity whether $\lambda_2 = 0.54$ or 1.6?

Hard to believe! Need to see the spectra!

Other fitting parameters may be compensating for effect of choice of $\lambda_2$?

Data from L. Laureano-Perez (NIST), using modified version of TRACER code (Cassette)
Shape factor, cont’d

$^{99}$Tc, TDCR models assuming unequal phototube efficiencies

$\lambda_2 = 0.54$ (?) $\quad \lambda_2 = 0.32$ $\quad \lambda_2 = 1.6$

$\lambda_2 = 0.54$ $\quad \lambda_2 = 0.54$

$\lambda_2 = ?$ $\quad \lambda_2 = ?$

$\lambda_2 = ?$

C_A (kBq/g)

Laboratory
Source of discrepancy with Participants C, E, F could be due to use of incorrect (default, $\lambda_2 = 1$) shape factor for 2nd forbidden non-unique decay.

Need to follow up with participants for more information about how their codes were used.
## Uncertainty assessments, Pt. 1

Relative standard uncertainties (in %) reported by each participant

<table>
<thead>
<tr>
<th>Component</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
<th>F</th>
<th>G</th>
<th>H</th>
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<tr>
<td>&quot;Statistical model&quot;</td>
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## Uncertainty assessments, Pt. 2

Relative standard uncertainties (in %) reported by each participant

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<th>Component</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
<th>F</th>
<th>G</th>
<th>H</th>
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</table>
Observations from uncertainty tables

- Number, type of components seems correlated with degree of experience with the method
- Some components almost universally evaluated
  - Spectrum endpoint
  - Poisson counting statistics (with/without background)
  - Value of kB (not always evaluated, but at least considered)
Observations, cont’d

• Many differences in how uncertainties are assessed
  – Only one participant inquired as to the NIST uncertainty on the mass, but 2 other (non-NIST) participants reported mass uncertainties!
  – Details about how uncertainty components were evaluated were generally lacking.
How does TDCR compare to other methods?

$^{99}$Tc, TDCR models assuming unequal phototube efficiencies
Conclusions/Recommendations

• Results – general
  – Bimodal distribution of results observed – could be due to use of incorrect shape factor (needs to be checked).
  – For values using “real” shape factor, very good agreement with other techniques was observed.
  – Need to account for phototube efficiency differences was very evident – 0.6 % in this relatively high-efficiency case.

• Uncertainties reporting
  – Some fundamental uncertainty components, such as live timing, scintillator effects not addressed by some participants.
  – Details regarding evaluation methodologies for uncertainty components should be part of every budget
    • What data were used to estimate magnitude
    • Degrees of freedom (if available)
    • More complete explanation of what source of variability the component is actually quantifying.