2004 CCRI(II) K2.I - 125 (2)  
Comparison ,  
2008 Re evaluation of IFIN-HH  
Data and Uncertainties  

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2004 comparison report

Radionuclide Metrology Laboratory (RML), IFIN-HH used the

One Na(Tl) detector: diameter 40 mm; height 2.5mm
Windows: A₁ (single peak ) 12…58 keV ; A₁ (sum peak) 58…85 keV
Dead time, non-extending, automatically corrected - live time clock
No Pile-up rejector

Resolving time: $\tau_r = (0.33 \pm 0.16) \mu$s. Measurement -Extrapolation

Distance source - detector : $x = 5 \text{ mm}$, choice
A second degree polynomial type $N_0 = \frac{(A_I + 2A_{II})^2}{A_{II}}$ as function of the

ratio $A_{II}/A_I$ was represented for various values $0< x < 30 \text{ mm}$. $N_0$ had the maximum value for $A_{II}/A_I = 0.0677$, at $x = 5 \text{ mm}$.

- Reported IFIN-HH value on the reference date (2004-06-01, 00 h UTC) was: $A_{\text{IFIN}} = [323.2 \pm 9.0 (2.8\%)] \text{ kBq g}^{-1}$
2008 critical re-evaluation of corrections and uncertainties

The conclusions of the Draft A report "International comparison of activity measurements of a solution of $^{125}\text{I}$", by G. Ratel, were:

(i) IFIN-HH result was +7.02 % higher than the mean $A_s = (302 \pm 0.98)$ kBq g$^{-1}$

Difference lies at +3.7$\sigma$, IFIN-HH considered as outlier.

New mean was: $A_s = (301.83 \pm 0.65)$ kBq g$^{-1}$;

(ii) Evaluation of uncertainties mentions:
- resolving time uncertainty: “normally the contributions remain low, but the IFIN ascribed - 1.6 %”
- possible sublimation of $^{125}\text{I}$ - contribution 1.0 %.
Comparison conclusions and study of published papers

- Look for the source of errors, by:
  - reconsidering the measurements
  - re-evaluating the processing of experimental data and uncertainties.
- We had reported the maximum $N_0$ value - measurement in the geometry “1”, 0.5 cm source – detector distance.
- We believe that we did not take into account the pile-up effects.
New processing of data

- **Follow the procedure applied at ENEA -Italy**
  
  *M. Capogni, A. Ceccatelli, P. De Felice, A. Fazio.*
  

- **Re-processing the data by an extrapolation procedure**, to apply both, pile-up and coincidence resolution time corrections:
  - take into account the $N_0$ value extrapolated for the $(A_{II}/A_I)=0$, instead of its maximum reported value.
  - 4 sources were measured in four geometries (“0”-source on the detector, “1”- at 0.5 cm, “2” and “3”).
  - were calculated expressions type:
    
    $$N_0/m = (N_{0 \text{ extrapolated}} /m) [1+ a(A_{II}/A_I) +b(A_{II}/A_I)^2 ]$$
Figure 1. $N_0/m$ versus $A_{II}/A_I$
## Extrapolation result

<table>
<thead>
<tr>
<th>Source</th>
<th>((N_0/m)_{\text{geometry}} \quad &quot;1&quot; \quad \text{s}^{-1}/ \text{mg})</th>
<th>a</th>
<th>b</th>
<th>1+ a((A_|/A_\perp)) + b((A_|/A_\perp)^2)</th>
<th>((N_0/m)_{\text{extrapolated}} \quad \text{s}^{-1}/ \text{mg})</th>
</tr>
</thead>
<tbody>
<tr>
<td>W181</td>
<td>463.51</td>
<td>2.062±0.54</td>
<td>-(15.2±3.6)</td>
<td>1.0689</td>
<td>433.64±6.08</td>
</tr>
<tr>
<td>W092</td>
<td>455.69</td>
<td>1.056±0.35</td>
<td>-(10.1±2.4)</td>
<td>1.0258</td>
<td>444.23±3.87</td>
</tr>
<tr>
<td>W081</td>
<td>459.24</td>
<td>2.058±0.43</td>
<td>-(14.3±2.8)</td>
<td>1.0759</td>
<td>427.42±4.95</td>
</tr>
<tr>
<td>W123</td>
<td>443.74</td>
<td>2.13±1.00</td>
<td>-(15.0±2.0)</td>
<td>1.0596</td>
<td>418.78±29.1</td>
</tr>
<tr>
<td>Mean</td>
<td>455.55±4.25</td>
<td>1.83±0.26</td>
<td>-(13.7±1.2)</td>
<td>1.058±0.022</td>
<td>430.87±5.40</td>
</tr>
</tbody>
</table>
Sources measured only in geometry “1”, 0.5 cm

By using the mean coefficients, \((a)_{\text{mean}}\) and \((b)_{\text{mean}}\), above Table, the corresponding extrapolated values, for the remaining six sources measured only at 0.5 cm (geometry “1”), were calculated.
Calculation of extrapolated values

<table>
<thead>
<tr>
<th>Source</th>
<th>((A_{II}/A_I))</th>
<th>(1 + 1.83(A_{II}/A_I) - 13.7 (A_{II}/A_I)^2)</th>
<th>((N_0/m)_{\text{geometry &quot;1&quot;}})</th>
<th>((N_0/m)_{\text{extrapolated}})</th>
</tr>
</thead>
<tbody>
<tr>
<td>W083</td>
<td>0.06687</td>
<td>1.0610</td>
<td>457.69</td>
<td>431.38</td>
</tr>
<tr>
<td>W080</td>
<td>0.0584</td>
<td>1.0600</td>
<td>451.97</td>
<td>426.39</td>
</tr>
<tr>
<td>W114</td>
<td>0.06166</td>
<td>1.0606</td>
<td>469.84</td>
<td>442.98</td>
</tr>
<tr>
<td>W089</td>
<td>0.06337</td>
<td>1.0608</td>
<td>471.84</td>
<td>444.79</td>
</tr>
<tr>
<td>W110</td>
<td>0.06553</td>
<td>1.0610</td>
<td>443.94</td>
<td>418.42</td>
</tr>
<tr>
<td>W091</td>
<td>0.05908</td>
<td>1.0607</td>
<td>464.41</td>
<td>437.82</td>
</tr>
<tr>
<td>Mean</td>
<td></td>
<td></td>
<td>459.95 ± 4.41</td>
<td>433.63 ± 4.16</td>
</tr>
</tbody>
</table>
New activity calculated

- \( (N_0_{\text{extrapolated}}/m) \) means for the two groups:
  \((430.87 \pm 5.40) \text{ s}^{-1}/\text{mg} \) and \((433.63 \pm 4.16) \text{ s}^{-1}/\text{mg} \)
agree within their standard deviations;
- mean value of all the 10 measured sources:
  \( (N_0_{\text{extrapolated}}/m) = (432.55 \pm 3.14) \text{ s}^{-1}/\text{mg} \), on 01.05.2004, 0:00h UTC

Recalculated radioactive concentration on 01.06.2004, 0:00h UTC:

\[ a = \frac{[432.55 \times 0.9983 \text{ (decay scheme)}]}{[1.001 \text{ (buoyancy)} \times 1.43558 \text{ (decay)}]}; \]

- \( a_{\text{IFIN}} = 300.49 \text{ kBq g}^{-1} \).
### New uncertainty budget

<table>
<thead>
<tr>
<th>Uncertainty</th>
<th>Type</th>
<th>Value, %</th>
<th>Calculation mode</th>
</tr>
</thead>
<tbody>
<tr>
<td>Statistics, $u_A$</td>
<td>A</td>
<td>0.72</td>
<td>Includes extrapolation uncertainties</td>
</tr>
<tr>
<td>Weighing</td>
<td>B</td>
<td>0.1</td>
<td>From experience</td>
</tr>
<tr>
<td>Background</td>
<td>B</td>
<td>0.1</td>
<td>Uncertainty propagation</td>
</tr>
<tr>
<td>Adsorption</td>
<td>B</td>
<td>0.1</td>
<td>Evaluated</td>
</tr>
<tr>
<td>Impurities</td>
<td>B</td>
<td>0.1</td>
<td>Detection limit</td>
</tr>
<tr>
<td>Decay scheme</td>
<td>B</td>
<td>0.13</td>
<td>Nuclear data uncertainties</td>
</tr>
<tr>
<td>Half life</td>
<td>B</td>
<td>0.1</td>
<td>Uncertainty propagation</td>
</tr>
<tr>
<td>$A_I$</td>
<td>B</td>
<td>0.52</td>
<td>Peaks threshold and deformation</td>
</tr>
<tr>
<td>$A_{II}$</td>
<td>B</td>
<td>1.0</td>
<td>Threshold between peaks influence</td>
</tr>
<tr>
<td>Combined uncertainty $u_c$</td>
<td></td>
<td>1.36</td>
<td>Quadratic sum of all uncertainties</td>
</tr>
</tbody>
</table>
Uncertainties in 2004 and in 2008

(i) Statistical uncertainty in 2008 is higher, due to extrapolation;

(ii) Uncertainties due to: coincidence resolution time and variation with source-detector distance vanished, due to extrapolation procedure.

(iii) Possible $^{125}$I sublimation did not occur, as the IFIN-HH result was bigger than the comparison mean.

- Re-evaluated-2008, $u_c$ (1.4%) is half of the 2004 evaluated value (2.8%)
Agreement with the comparison mean

The IFIN-HH re-evaluated value is:

\[ a_{IFIN, \text{sum peak, re-eval}} = (300.49 \pm 4.09) \text{ kBq g}^{-1} \]

Differences from the comparison mean:

\[ D_{IFIN} = (300.49 - 301.83) = -1.34 \text{ kBq g}^{-1} \]

Uncertainty

\[ U_{IFIN} = 2\sqrt{4.09^2 + 0.65^2} = 8.28 \text{ kBq g}^{-1} \]

Conclusion: IFIN-HH value agrees with the mean of comparison
X – X, gamma Coincidence method

- The X-X, gamma coincidence method - applied for the standardization of a $^{125}$I solution prepared in our laboratory.

- The system, containing two identical, thin NaI(Tl) crystals and the standardization method were presented in the paper

M. Sahagia, C. Ivan, E. L. Grigorescu, A .C. Razdolescu
“Standardization of $^{125}$I by the Coincidence Method and Practical Applications”
Comparison of the two methods results, validation of the coincidence method

- Taking as standard the IFIN-HH coincidence measured solution, with radioactive concentration on measurement date, \( a = [604.1\pm3.6 \ (0.6\%)] \) kBq g\(^{-1}\), the comparison solution was relatively standardized.

Two vials containing about 0.2 g of each solution were prepared.

A Multi-gamma counter type LKB 2104, was used.

- The activities were calculated from the activity concentrations on the measurement date, and the mass of solution in the vials.

- Supplementary uncertainties were due to counting statistics, decay corrections and weighing.
Result of relative measurement

*IFIN solution vial:*

\[ A_{\text{IFIN,coinc}} = [4707 \pm 38 (0.8\%)] \text{ Bq on March 2-nd, 2006:} \]

Ratio of the counting rates of the vials:

\[ R = \left( \frac{N_y^{BIPM}}{N_y^{IFIN}} \right) = [0.007607 \pm 0.000085 (1.12\%)] \]

*BIPM solution prepared vial determined at IFIN:*

\[ A_{\text{BIPM,IFIN}} = [35.804 \pm 0.51 (1.41\%)] \text{ Bq.} \]

Radioactive concentration for March 2-nd, 2006:

\[ a = \frac{0.035804}{0.21060} = [0.17007 \pm 0.00241 (1.42\%)] \text{ kBq. g}^{-1}. \]

*Value for the comparison time, June 1-st, 2004, 00hUTC*

\[ a_{\text{BIPM,IFIN}} = [296.4 \pm 4.2 (1.42\%)] \text{ kBq. g}^{-1} \]
Agreement of results

- Difference from CCRI(II) comparison value:
  \[ D_{\text{IFIN coinc.}} = 296.4 - 301.83 = -5.43 \text{ kBq. g}^{-1} \]
  Uncertainty \[ U_{\text{IFIN}} = 2\sqrt{4.2^2 + 0.65^2} = 8.50 \text{ kBq. g}^{-1} \]

- Comparison of the IFIN-HH methods
  \[ D_{\text{coinc.-D sum peak}} = 296.4 - 300.5 = -4.1 \text{ kBq. g}^{-1} \]
  Uncertainty \[ U_{\text{IFIN}} = 2\sqrt{4.2^2 + 4.09^2} = 11.72 \text{ kBq. g}^{-1} \]
Conclusions

1. The source of errors in our result was due to neglecting the pile-up correction.

2. Correction was done by the extrapolation of activity to value of the ratio: $A_{\parallel}/A_{\perp} = 0$.

3. The re-evaluated value is in line with the other participants’ results;
   The uncertainty value was reduced at half initial value and it is consistent with the deviation from the mean.

4. Independent relative determination, with a solution standardized by the coincidence method;
   The value agrees with the mean comparison value and with the IFIN-HH sum-peak re-evaluated one.