Bilateral comparison of ^{14}C activity measurements at the NCBJ RC POLATOM and the ENEA-INMRI

T. Ziemek[1], M. Capogni[2], R. Broda[1], P. De Felice[2], T. Dziel[1], A. Listkowska[1]

[1] National Centre for Nuclear Research RC POLATOM, Poland
[2] ENEA National Institute of Ionizing Radiation Metrology, Italy

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Outlines

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- Measurements methods
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The aim of the comparison

- To evaluate the performance of both Institutes for measurements of a medium-energy long-lived pure β-emitter ^{14}C
- To check the performances of the Triple-to-Double Coincidence Ratio (TDCR) counters available both at POLATOM and ENEA
- To compare results of on-line (in the FPGA digital board) and off-line (in the CAEN Desktop digitizer) analyses in the two TDCR systems
- ^{14}C standards are of particular importance in many applications:
  - in metrology for self-normalizing and calibration of LS counters
  - in radiometric dating
  - in assessing the impact of ^{14}C releases in the environment and setting-up and implementing technical and organizational procedures for ensuring public health
  - in measurements in coolant circuit of BWR and PWR reactor
  - in enabling accurate and precise measurements in plant and other facility at which fission reactions or spent fuel processing take place

^{14}C solution and LS-sources

- The ^{14}C solution used:

Chemical composition	Solubilization medium	Carrier conc. /(μg g^{-1})	Density (g cm^{-3})	Impurities
CH_3COONa	H_2O	CH_3COONa: 60	1.0	–

- A set of six ^{14}C sources was prepared at POLATOM:

  - 20 mL high-performance PerkinElmer glass vials
  - 10 mL of Ultima Gold liquid scintillator
  - mass of radioactive material from 135.7 mg to 138.6 mg

- The Mettler MT5 balance traceable to SI system of units through the national standard of mass in Poland was used

- All six sources were measured at POLATOM and three of them, ready-to-measure, have been sent and measured at ENEA-INMRI

Measurement methods

- **POLATOM**:

  TDCR method (4P-LS-BP-00-00-TD)

  - used in two LS-counters
  - counting efficiency was changed by photomultipliers defocusing
  - each source was measured during 30 min

  CIEMAT/NIST efficiency tracing method (4P-LS-BP-00-00-CN)

  - used in two scintillation counters
  - ^{3}H as a tracer
  - each source was measured during 15 min

- **ENEA-INMRI:**

  TDCR method (4P-LS-BP-00-00-TD)

  - used in two LS-counters
  - each source was measured during 20 min

Counters used at the RC POLATOM

TDCR counter:
3 PM-tubes RCA 8850,
MAC-3 module

TDCRG counter
(TDCR detector with γ-channel):
3 PM-tubes ET Enterprises 9214B,
NaI(Tl) 3”$\times$3” detector in γ-channel,
FPGA XILINX Spartan-3AN digital board

Ziemek, T., Jeczmieniowski, A., Cacko, D., Broda, R., Lech, E., **2016**. *A new 4π(LS)-γ coincidence counter at NCBJ RC POLATOM with TDCR detector in the beta channel* . Appl. Radiat. Isot. **109**, 290-295

Counters used at the RC POLATOM

Tri-Carb 2810 TR
scintillation counter

Wallac 1411
scintillation counter

Counters used at the ENEA-INMRI

Hidex 300SL

'Metro' version LS-counter

ENEA portable TDCR counter:

3 PM-tubes Hamamatsu R7600U-200,
CAEN Desktop digitizer DT5720

Capogni, M., De Felice, P., **2014**. *A prototype of a portable TDCR system at ENEA* . Appl. Radiat. Isot. **93** , 45-51

^{14}C activity calculation

- **POLATOM**:

  - **TDCRB-03** software for the TDCR method
    with kB = (0.010 ± 0.001) cm MeV^{-1}
  - **CN2005** software for the CIEMAT/NIST method
    with kB = (0.010 ± 0.001) cm MeV^{-1}

- **ENEA-INMRI:**

  - **TDCRB-07** software for the TDCR method (from CEA-LNHB)
    with kB = (0.011 ± 0.001) cm MeV^{-1}

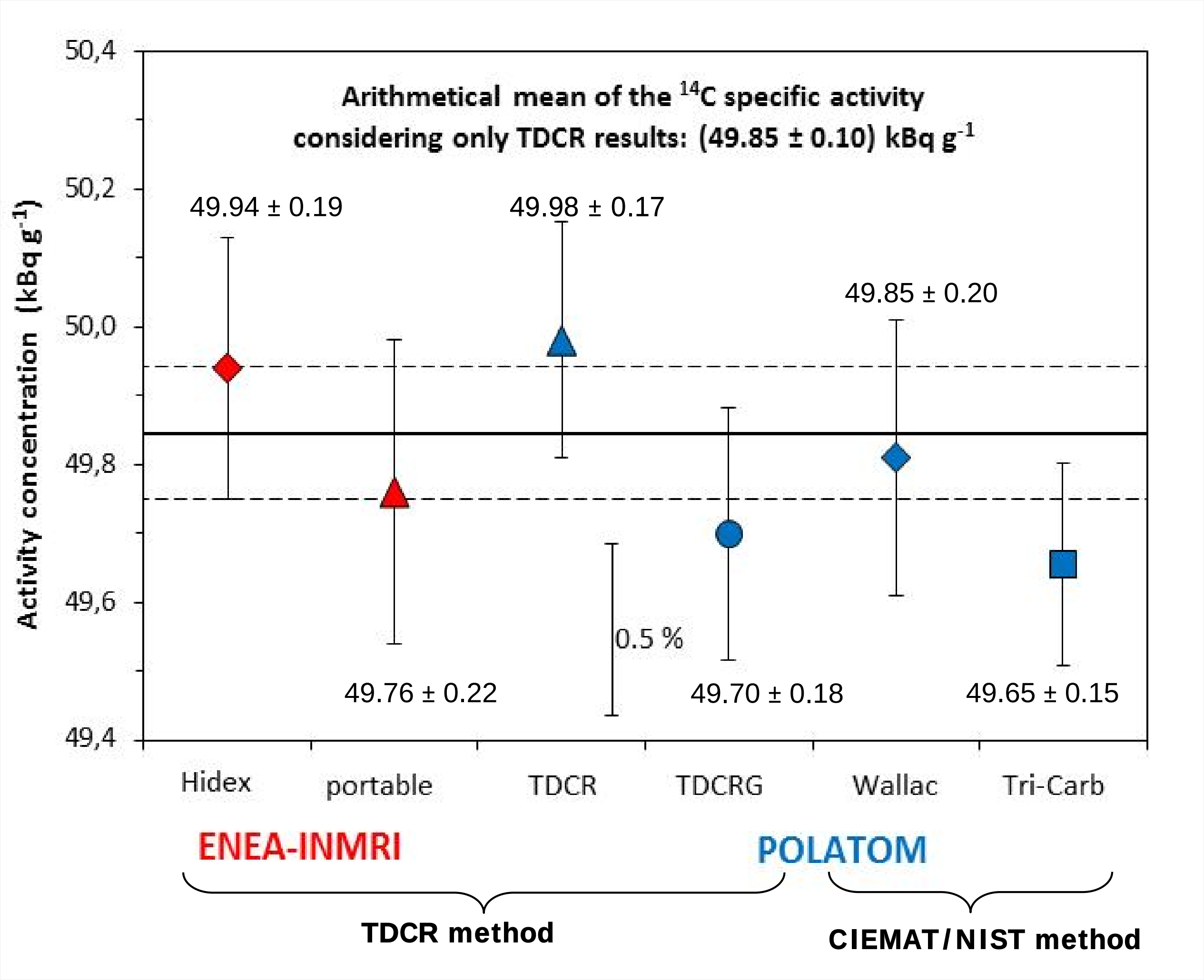
- The kB parameter value was selected with regard to the source activity independence of the detection efficiency in the triple counter.

- Data adopted by both laboratories (BIPM Monographie 5, 2013):

  $E_{\beta\ max}$ = 156.476 (4) keV

  $T_{1/2}$ = 5700 (30) y

Results of the ^{14}C bilateral comparison



Arithmetical mean values of all counting points of all sources measured in various LS-counters in both laboratories.

Combined standard uncertainties for k = 1.

Uncertainties for the ^{14}C measurements

Uncertainty component	Assess-ment type	TDCR method				C/N method	
		POLATOM		ENEA-INMRI		POLATOM	
		TDCR counter	TDCRG counter	Hidex system	portable TDCR	Wallac 1411	Tri-Carb 2910 TR
Counting statistics	A	0.04	0.13	0.25	0.28	0.27	0.02
Weighing	B	0.1	0.1	0.10	0.10	0.1	0.1
Background	A	0.01	0.03	0.01	0.05	0.01	0.01
Counting time	B	0.001	0.001	0.01	0.01	0.001	0.001
Discriminator threshold setting	B	0.15	0.15				
Dead-time	B			0.10	0.15		
Coincidence resolving time	B			0.15	0.20		
Theoretical model (kB selection, Birks function, energy spectrum)	B			0.20	0.20		
^{3}H efficiency quenching function	B					0.1	0.1
Tracer	B					0.096	0.082
kB selection	B	0.15	0.15			0.02	0.02
Counting model *)	B	0.25	0.25			0.25	0.25
Combined uncertainty (as quadratic sum of all uncertainty components)	A	0.04	0.13	0.25	0.28	0.27	0.02
	B	0.34	0.34	0.29	0.34	0.30	0.29
	total	**0.34**	**0.37**	**0.38**	**0.44**	**0.40**	**0.30**

*) Comprises an influences of the stopping power *dE/dX* model selection, the β-spectrum calculation, the integration method for the Birks function and for the counting efficiency

Results of the ^{14}C bilateral comparison

Laboratory / Method / Counter	Measurement results		Final results	
	A_i (kBq g^{-1})	u_i (%)	$\bar{A}_i$ (kBq g^{-1})	u_i (%)
POLATOM (TDCR)				
TDCR counter	49.98 ± 0.17	0.34	**49.84 ± 0.13**	**0.25**
TDCRG counter	49.70 ± 0.18	0.37		
POLATOM (CIEMAT/NIST)				
Wallac 1411	49.85 ± 0.20	0.40		
Tri-Carb 2910 TR	49.65 ± 0.15	0.30		
ENEA-INMRI (TDCR)				
Hidex 300SL	49.94 ± 0.19	0.38	**49.85 ± 0.14**	**0.29**
ENEA portable TDCR	49.76 ± 0.22	0.44		

A_i - activity concentration

u_i - standard uncertainty (k = 1)

$\bar{A}_i$ - one result per laboratory as the outcome of the ^{14}C comparison (the mean arithmetical of the TDCR method measurement results)

Laboratories performances

- The quantity E_n (European Standard EN ISO/IEC 17043:2010):

$$E_n(k=2) = \frac{(x_i - x_j)}{\sqrt{(2u_i)^2 + (2u_j)^2}}$$

where x_i, u_i – the ^{14}C activity concentration and its standard uncertainty (k = 1) obtained at ENEA-INMRI
x_j, u_j – values obtained at POLATOM

- For any pair of results **0.08** < $|E_n|$ < **0.59** so $|E_n| < 1$
and this confirms that all results obtained by both laboratories are in good agreement.

- The degree of equivalence of the participated laboratories (employing the convention used in CCRI(II)):

$$D_{ij} = x_i - x_j \quad \text{with} \quad U_{ij} = 2\sqrt{(u_i)^2 + (u_j)^2}$$

where x_i, x_j – the founded activities $\bar{A}_i$ (one per laboratory)

D_{ij} = **-0.08** kBq g^{-1} U_{ij} = **0.38** kBq g^{-1} (k = 2)

Conclusions

- All results of ^{14}C measurements carried out at POLATOM and at ENEA-INMRI using the TDCR and CIEMAT/NIST method in six various counters are in a very good agreement
- Arithmetical mean of the TDCR method results were taken as final results of comparison measurements by both laboratories. The E_n parameter of the two final results is $|E_n| = 0.03$

Thank you for attention !