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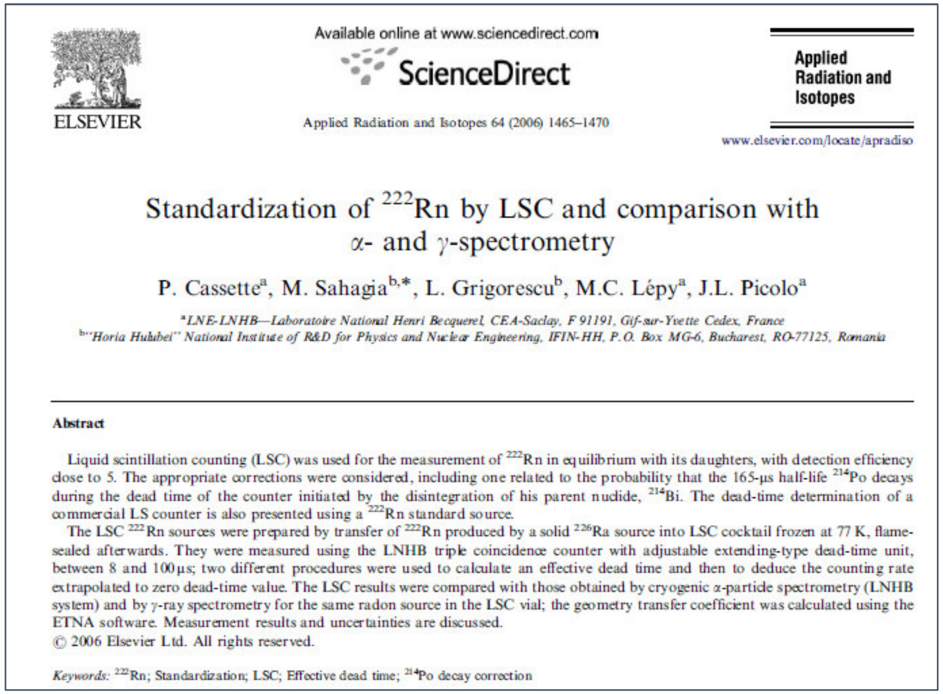
Previous work:

Simplifications
Detection efficiency of alpha radionuclides (except ^{214}Po) = 1

Detection efficiencies of ^{214}Pb and ^{214}Bi = 1

Correction of ^{214}Po losses due to dead-time create by ^{214}Bi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Standardization of ^{222}Rn by LSC and comparison with α- and γ-spectrometry

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Abstract

Liquid scintillation counting (LSC) was used for the measurement of ^{222}Rn in equilibrium with its daughters, with detection efficiency close to 5. The appropriate corrections were considered, including one related to the probability that the 165-μs half-life ^{214}Po decays during the dead time of the counter initiated by the disintegration of his parent nuclide, ^{214}Bi. The dead-time determination of a commercial LS counter is also presented using a ^{222}Rn standard source.

The LSC ^{222}Rn sources were prepared by transfer of ^{222}Rn produced by a solid ^{226}Ra source into LSC cocktail frozen at 77 K, flame-sealed afterwards. They were measured using the LNHB triple coincidence counter with adjustable extending-type dead-time unit, between 8 and 100 μs; two different procedures were used to calculate an effective dead time and then to deduce the counting rate extrapolated to zero dead-time value. The LSC results were compared with those obtained by cryogenic α-particle spectrometry (LNHB system) and by γ-ray spectrometry for the same radon source in the LSC vial; the geometry transfer coefficient was calculated using the ETNA software. Measurement results and uncertainties are discussed.

Keywords: ^{222}Rn; Standardization; LSC; Effective dead time; ^{214}Po decay correction

This work:

Calculation of the detection efficiencies of ^{214}Pb and ^{214}Bi with the free parameter model considering:

- Beta spectra of ^{214}Pb and ^{214}Bi using the Betashape code and PENELOPE Monte Carlo simulation

- Re-assessment of ^{214}Po counting losses by correction factor function of the base dead-time duration and by measurements with various dead-times and extrapolation to zero dead-time

^{222}Rn in water, LS measurement methods

1-Two-phases cocktail (ASTM, 1998)

- Injection of water under a purely organic LS cocktail
- Radon *is supposed* to migrate in the organic phase and interfering radionuclides *are supposed* not to be detected
- Calibration is achieved using a ^{226}Ra standard solution

2- Homogeneous source with water-miscible cocktail (Salonen 2010* and others)

- Intimate mixture of water and LS cocktail
- Blank made using degassed water

* L. Salonen, Comparison of two direct LS methods for measuring ^{222}Rn in drinking water using alpha/beta liquid scintillation spectrometry. Applied Radiat. Isot., 68, 2010

Two-phases cocktail (ASTM, 1998)

- Advantage: simplicity
- Drawbacks:
  - Imperfect radon extraction by the organic phase
  - Possible interference of ^{210}Po (*cf.* Salonen, 2010)
  - Detection of radionuclides remaining in the aqueous phase (e.g. high-energy beta with long range)
  - Non-trivial calibration using a ^{226}Ra solution (general problem of ^{210}Pb activity in the ^{226}Ra solution)

Homogeneous LS cocktail

- Advantages: simplicity, compensation of interfering radionuclides (e.g. ^{226}Ra and ^{210}Pb + daughters), possible absolute measurement using the TDCR method

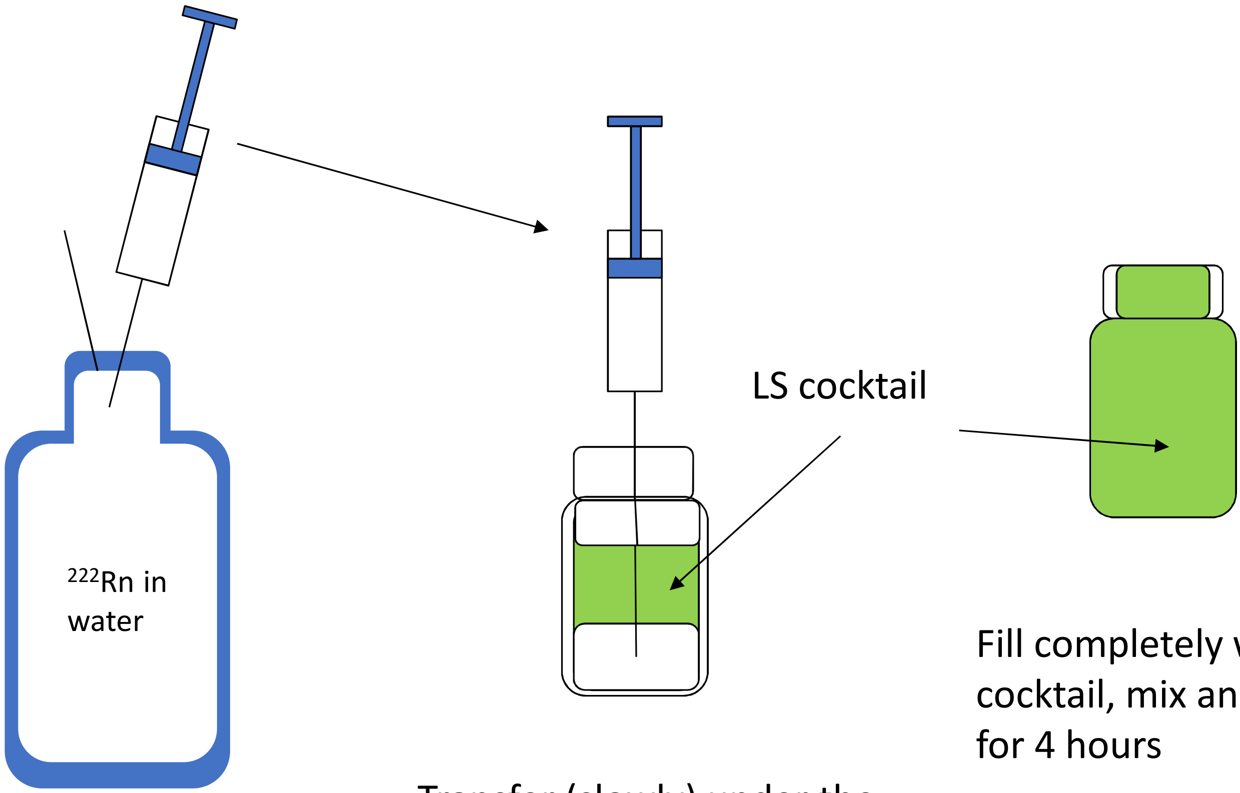
- Drawback: measurement of 2 samples instead of 1

Homogeneous LS cocktail

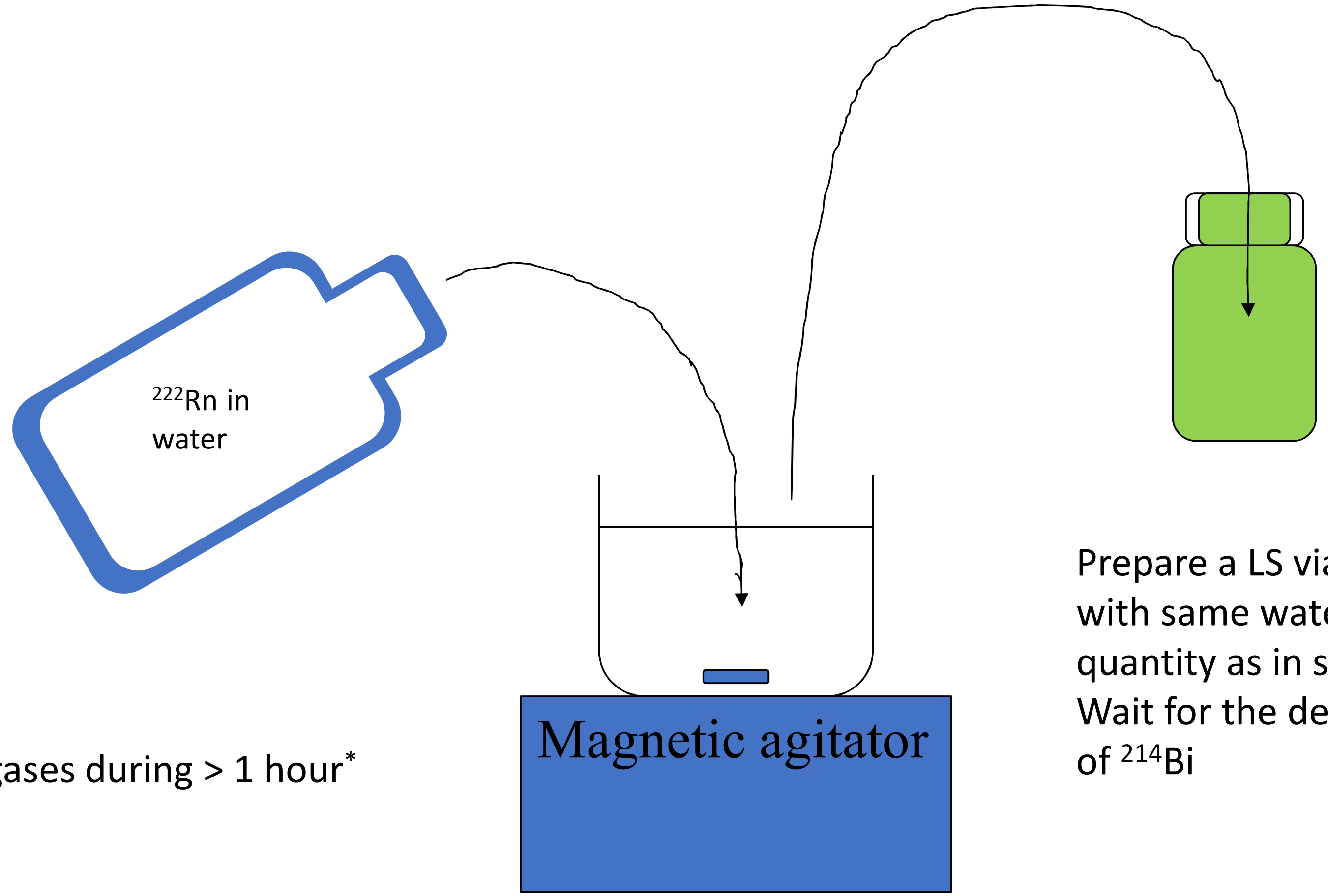
- Sampling the solution to measure using a syringe (slow sampling to avoid degassing)
- Injection of the solution under the LS cocktail
- Full filling of the LS vial*, mixture of the two phases, wait for secular equilibrium (e.g. 4 hours)
- Sampling the solution and vigorous degassing in an open vial (e.g. by magnetic agitation)
- Mixture with the LS cocktail (use the same amount of solution) and measurement in a LS counter. The sample could be re-measured after 15 days if the alpha activity in the blank is not negligible (possible presence of ^{226}Ra)
- Measurement of the radon-in-water source

** Glass or polyethylene-PTFE vials, screw cap with aluminum foil liner*

Source preparation 1



Source preparation 2, blank



Degases during > 1 hour*

Prepare a LS vial with same water quantity as in step 1
Wait for the decay of ^{214}Bi

* *Removing radon from water is a long process!*

Measurement

Blank: measure after the decay of ^{214}Bi (4 h) and re-measure after 15 days (if necessary, for ^{226}Ra determination)

- Compensate for any activity present in the water, except radon
- Allows the determination of ^{226}Ra activity in the water by following the ^{222}Rn ingrowth

Source: wait for secular equilibrium (> 4 h) and measure

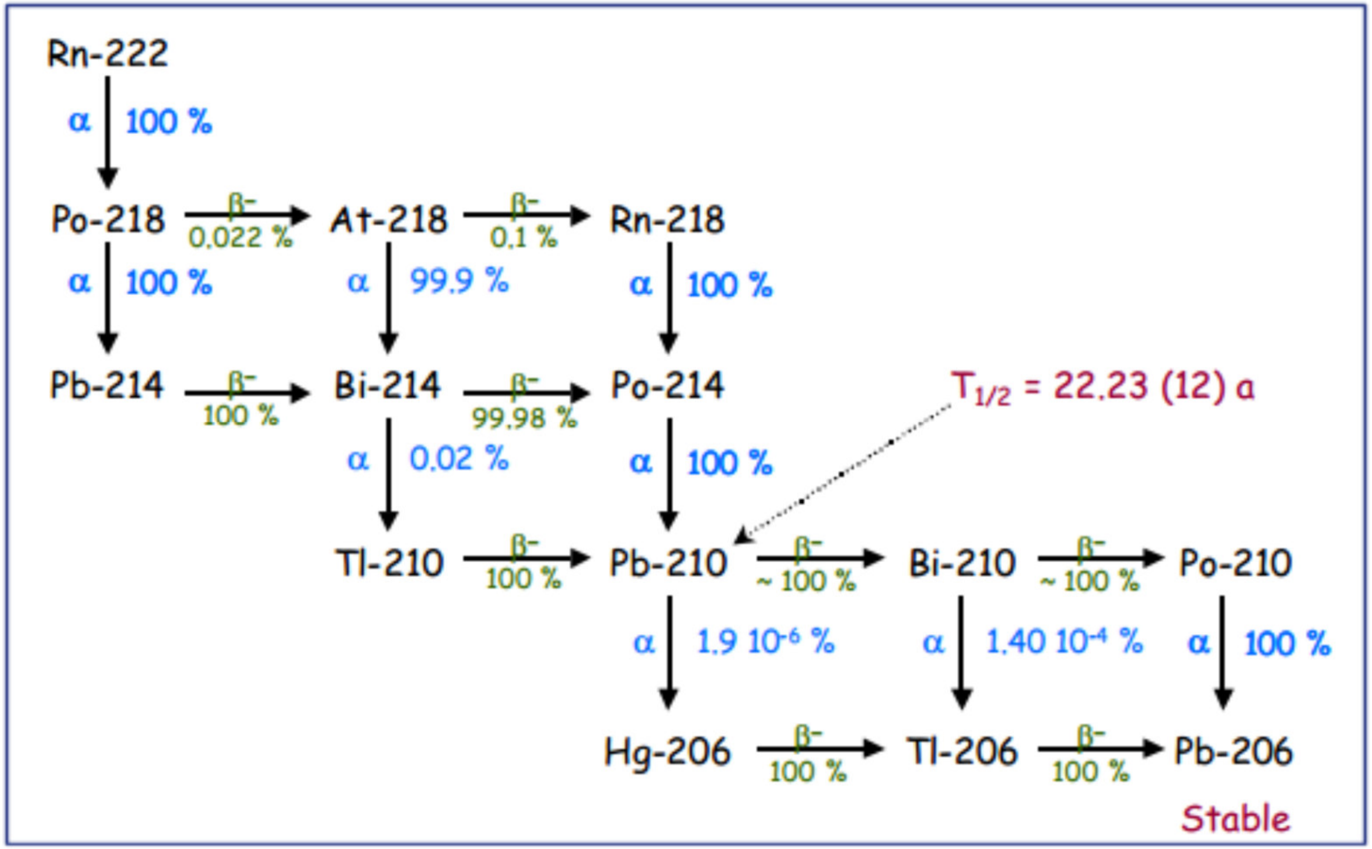
- Subtract the counting rate of the blank
- Calculate the (^{222}Rn + daughters) detection efficiency with the TDCR method

TDCR SL counter: mini TDCR from LNHB

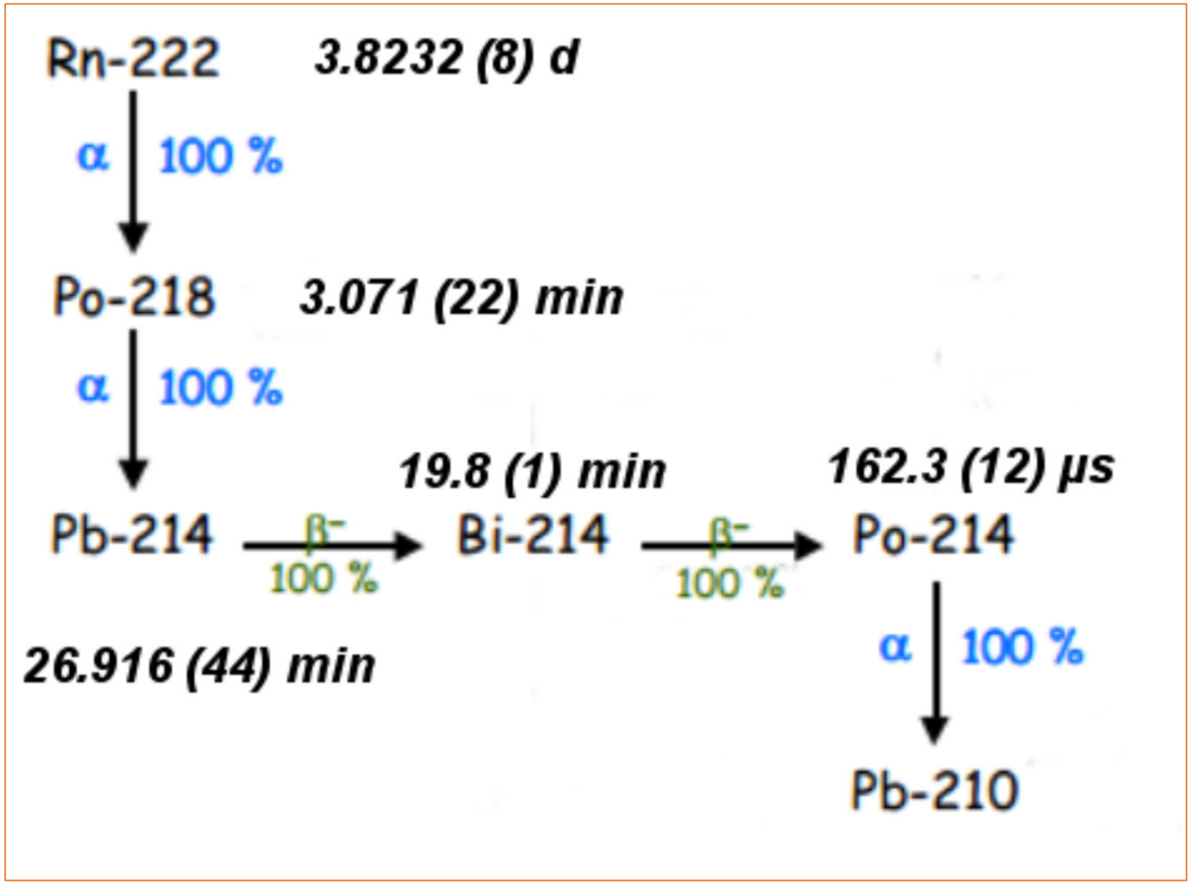
TDCR counter with online (NanoTDCR, Labzy) and offline (CAEN digitizer) acquisitions

Simultaneous measurements with various coincidence resolving time and dead-time durations

^{222}Rn decay scheme



^{222}Rn simplified decay scheme used



Secular equilibrium

From Bateman equations:

$$\frac{A_{Po-218}}{A_{Rn-222}} = 1.00055 \qquad \frac{A_{Pb-214}}{A_{Rn-222}} = 1.00545$$

$$\frac{A_{Bi-214}}{A_{Rn-222}} = 1.00910 \qquad \frac{A_{Po-214}}{A_{Rn-222}} = 1.00910$$

$$A_{total} = A_{Rn-222} + A_{Po-218} + A_{Pb-214} + A_{Bi-214} + A_{Po-214}$$

If ^{210}Pb activity is negligible (fresh ^{222}Rn source)

$$N_{total} = A_{Rn-222}(\varepsilon_{Rn-222} + 1.00055\, \varepsilon_{Po-218} + 1.00545\, \varepsilon_{Pb-214} + 1.0091\, \varepsilon_{Bi-214} + 1.0091 \varepsilon_{Po-214})$$

Detection efficiencies

$\varepsilon_{Rn\text{-}222}$ and $\varepsilon_{Po218} = 1$

ε_{Pb-214} , ε_{Bi-214} function of the figure of merit and spectra, can be deduced from the experimental TDCR value

$\varepsilon_{Po\text{-}214}$ function of the dead-time created by the decay of ^{214}Bi

P: probability that the decay of ^{214}Po occurs during the dead-time created by the decay of ^{214}Bi: with the LS counter used, the dead-time is triggered by single or coincident events , thus the probability that the decay of ^{214}Bi starts the dead-time is higher than the probability of detection of ^{214}Bi in coincidences

$$P = 1 - \exp(-\lambda_{Po-214} T_m)$$

T_m: base duration of the dead-time (extending-type*)

* This only works with extending-type dead-time (with fixed dead-time, the live-time of ^{214}Po is biased)

Total counting rate

$$\boxed{N_{total} = A_{Rn-222}(\varepsilon_{Rn-222} + 1.00055\,\varepsilon_{Po-218} + 1.00545\,\varepsilon_{Pb-214} + 1.0091\,\varepsilon_{Bi-214} + 1.0091\varepsilon_{Po-214})}$$

With $\varepsilon_{Rn-222} = \varepsilon_{Po-218} = 1$, ε_{Pb-214} and ε_{Bi-214} function of the TDCR value

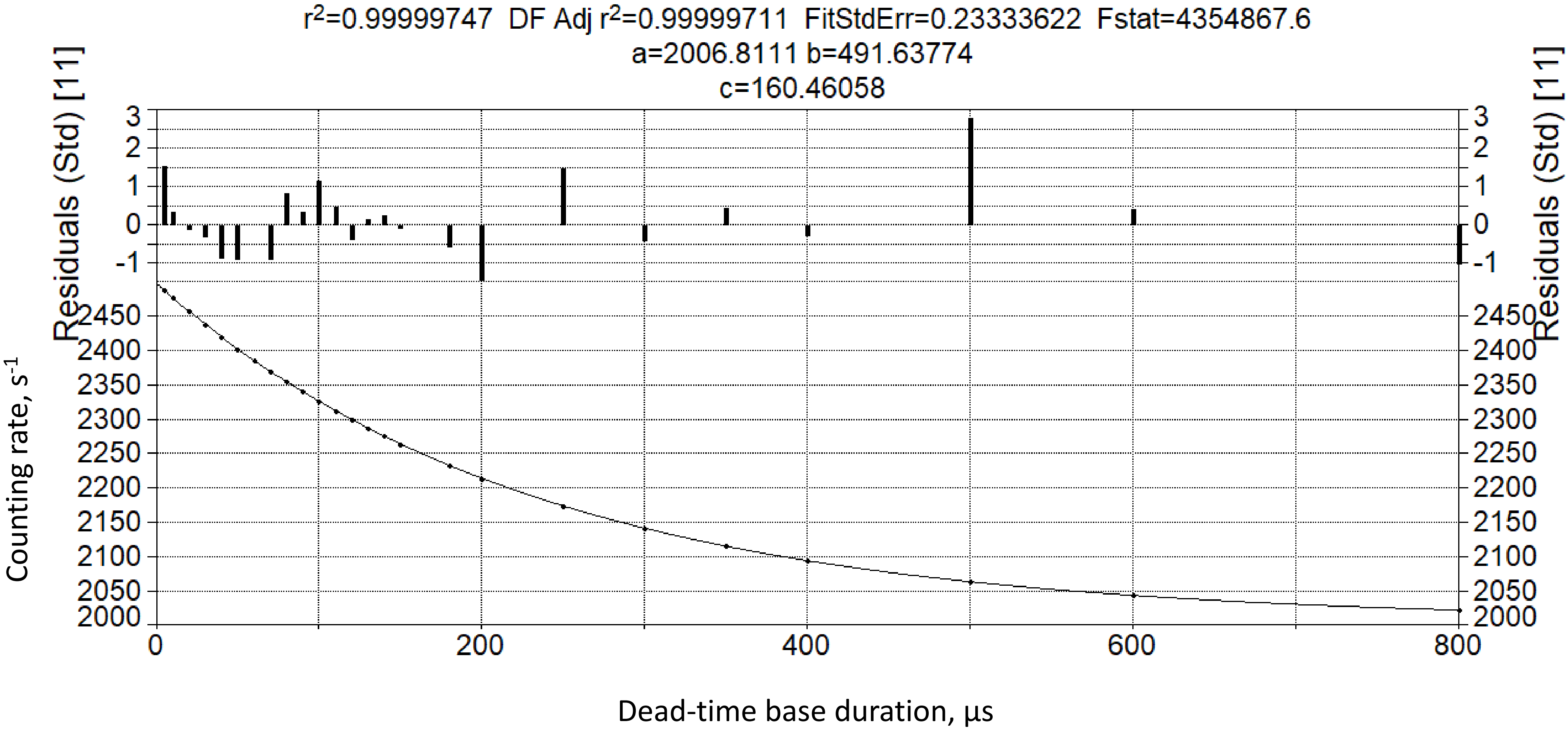
$$\varepsilon_{Po-214} = (1-\varepsilon^{*}_{Bi-214}) + \varepsilon^{*}_{Bi-214}\, exp(-\tau\, Ln(2)/T_{1/2\ (Po-214)})$$

Where τ is the dead-time duration and ε^{*}_{Bi-214} is the probability that the decay of Bi-214 triggers the dead-time circuit

Thus, $$N_{total} = a + b\, exp(-\tau\, Ln(2)/T_{1/2(Po-214)})$$

By varying the dead-time duration for the same measurement, the half-life of ^{214}Po can be deduced from the exponential fit of the curve (N_{total} *vs.* τ)

Example of variation of counting rate *vs.* dead-time duration



^{214}Po half-life

Three independent measurements with various counting rates

Experiment id	Counting rate @ 50 µs s^{-1}	^{214}Po Half-life fit µs	Standard uncertainty µs
A4	4327	160.52	0.18
C3	3545	160.01	0.98
S6	1421	160.37	0.15
Mean		**160.45**	**0.11**

→ The results are coherent within uncertainties

Even with different counting rates, the results are in very good agreement

Literature survey of ^{214}Po half-life determination

Publication	Half-life with standard uncertainty (µs)
Von Dardel (1950)	163.7 (18)
Ballini (1953)	158 (2)
Ogilvie (1960)	159.5 (30)
Debrowolski (1961)	164.3 (18)
Erlik (1971)	165 (3)
Zhou (1993)	160 (12)
DDEP recommended value (2008)	162.3 (12)
Suliman (2012)	164.2 (6)
Alexeyev (2013)	163.5 (8)
Bellini (2013)	163.6 (3)
This work	**160.45 (11)**

Our value is significantly lower than the last three measurements. We looked for possibilities of errors but up to now, no cause of bias was found

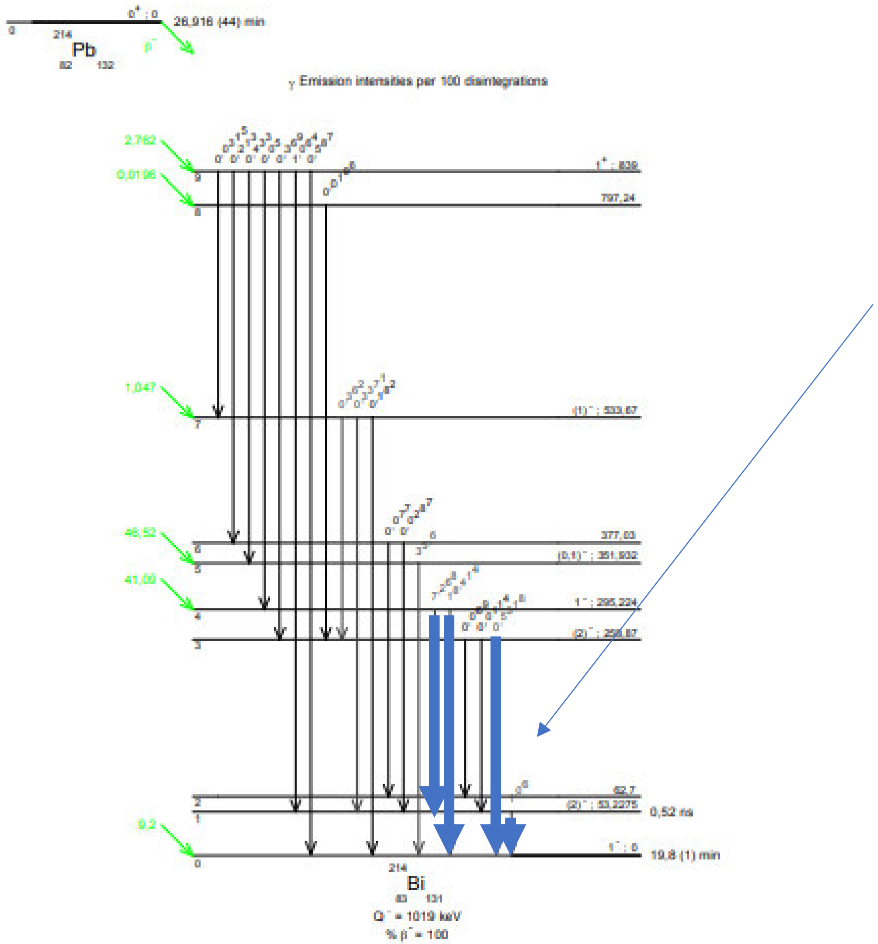
We are working on a Monte Carlo generation of synthetic dataset to check the results

The radon222 code

- Global TDCR code to calculate the detection efficiency of ^{222}Rn in equilibrium with progeny
- Input: *T*, *D*, base dead-time duration, relative quantum efficiency of each PMT, ^{214}Bi and ^{214}Pb energy spectra transferred to the scintillator
- Calculation for *kB*=0.01 cm/MeV (negligible influence of the kB value)
- *D* is the weighed sum of *D* from each radionuclide
- *T* is the weighed sum of *T* from each radionuclide
- Detection efficiency of alpha radionuclides = 1
- Detection efficiency of ^{214}Pb and ^{214}Bi calculated from the TDCR value (free parameter model)
- Detection efficiency of ^{214}Po taking into account the base dead-time (exponential law)

Output: global detection efficiency, detection efficiencies of ^{214}Pb, ^{214}Bi and ^{214}Po and figure of merit for beta radionuclides (e^-/keV)

Pb-214 spectrum

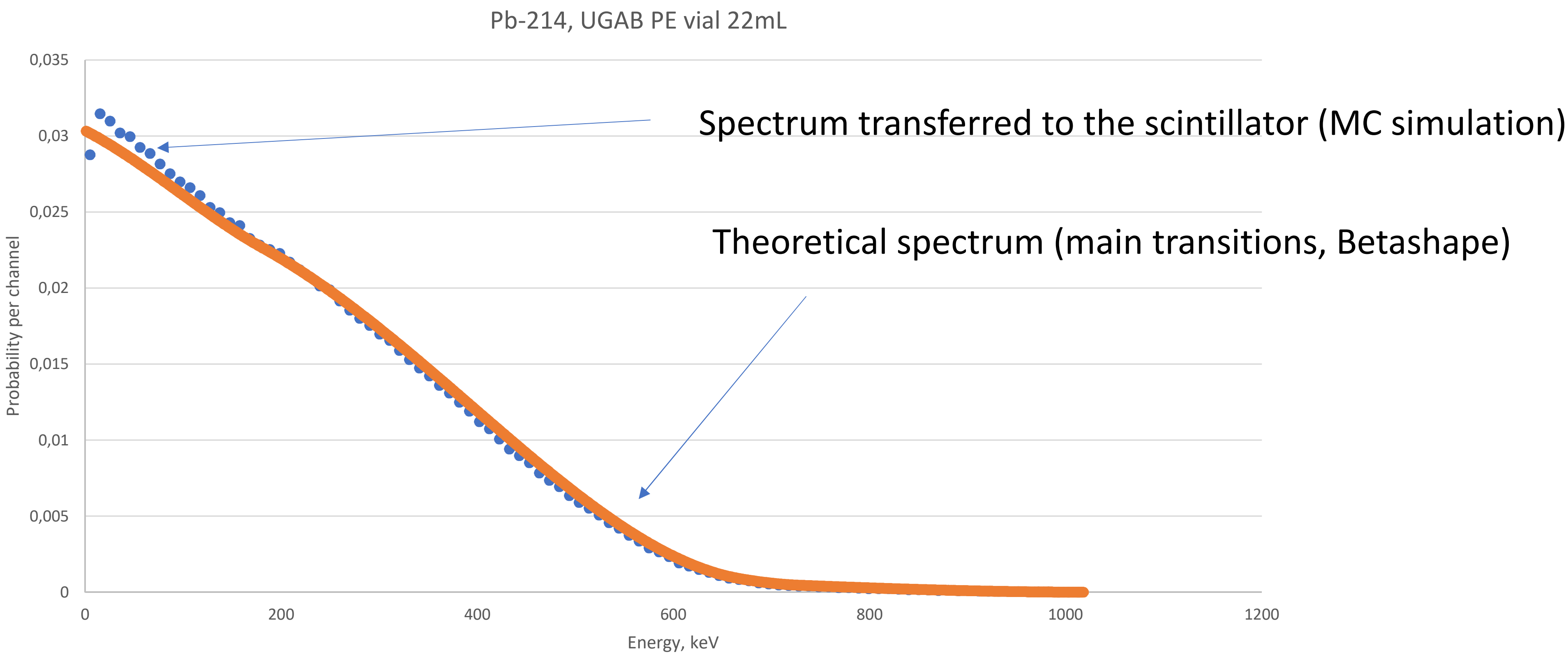


5 main beta transitions

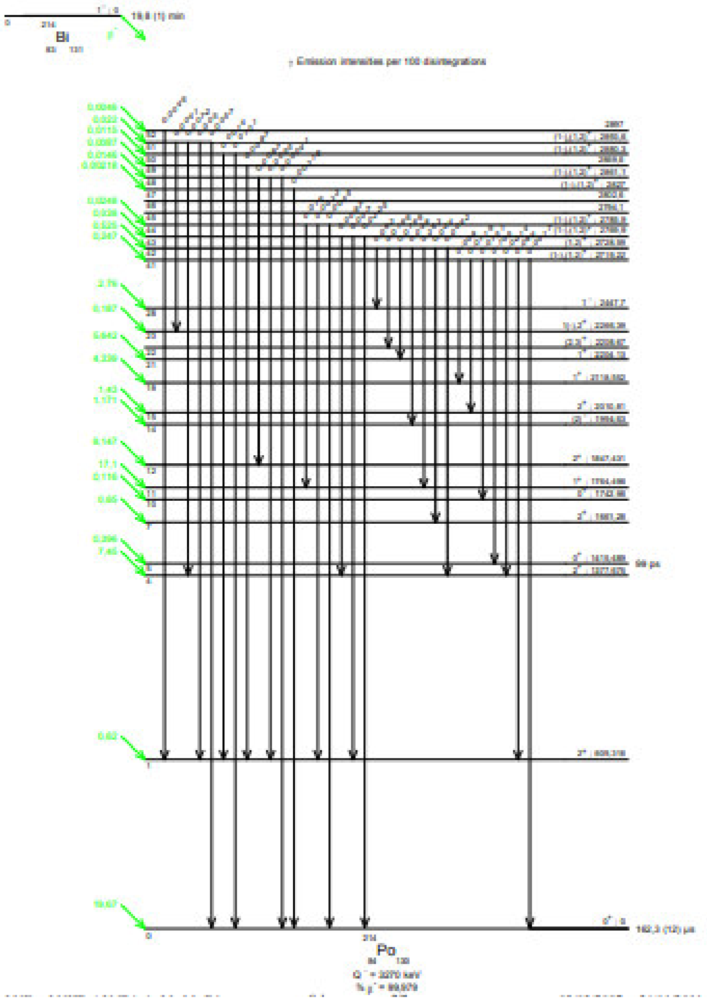
Conversion electrons (intensities > 1%)

- Calculation with Betashape code of the main transitions
- Calculation of the energy spectrum absorbed in the scintillator with PENELOPE 2018 code

^{214}Pb, UGAB, PE vial



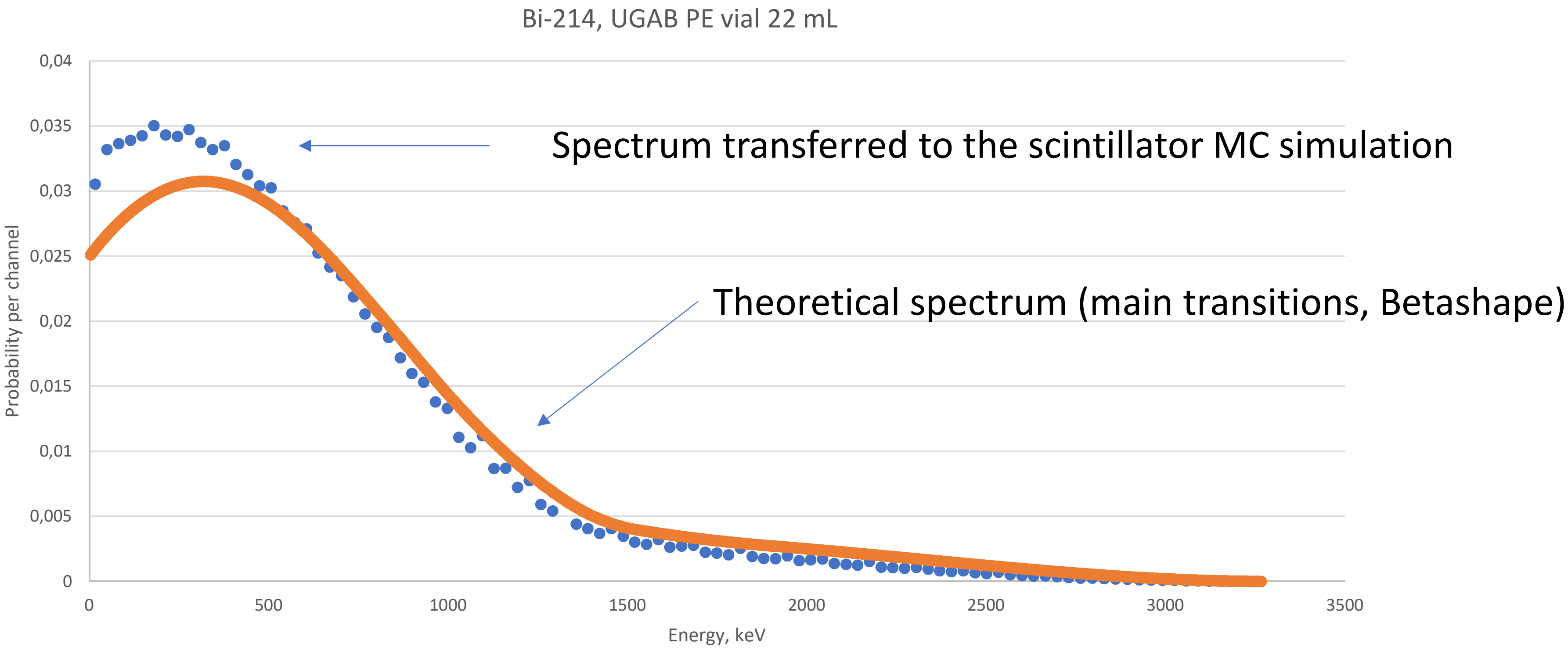
Bi-214 (sample)



- Alpha transitions neglected (very low probabilities)
- 14 beta transitions with P > 1 %
- No conversion electron emission > 1%

- Calculation with Betashape of the main transitions
- Calculation of the energy spectrum absorbed in the scintillator with PENELOPE 2018 code

^{214}Bi, UGAB, PE vial



Example of measurement

- Rn in water sample, miniature LNHB TDCR counter (B. Sabot *et al.,* Performance of portable TDCR systems developed at LNE-LNHB, NIM A, 2022)

- CAEN digitizer (1 GHz sampling rate) offline analysis

- Acquisition with various base dead-time durations (extending-type dead-time, from 5 µs to 2 ms)

- Reference value: radon activity measured with cryogenic ASD and quantitative transfer to the water (LNHB radon in water generator)

Example of activity measurement *vs.* base dead-time duration (20 to 200 µs)

TDCR calculation (5-200 µs)	
Mean activity	336.25 Bq
Standard deviation	0.21 Bq
Relative standard deviation	0.06%
Extrapolation to zero dead-time	
Mean activity	335.82 Bq
Uncertainty (fit model)	0.56 Bq
Relative uncertainty	0.17%
ASD reference value	
Activity of the sample	335.1 (14) Bq (0.4 %)

Relative difference: 0.13%

Relative difference: 0.3 %

Conclusions

- TDCR activity measurement of radon in water sample made simple and accurate using water miscible LS cocktail
- The analysis of the relation between the experimental count rate and the base of the extending-type dead-time allows the determination of the ^{214}Po half-life
- The dominant correction factor is due to ^{214}Po counting losses, when it decays during the dead-time generated by the decay of ^{214}Bi
- A simple exponential correction, considering the base dead-time duration seems to be accurate enough for base dead-time durations lower than 200 µs
- Results are coherent with those obtained by extrapolation to zero dead-time, but with better uncertainty
- In-depth analysis of the ^{214}Po half-life determination is ongoing

Thank you for your attention

DT (µs)	D corr	εD	Activity, Bq
5	2511,75373	4,9861	499,05
10	2501,17117	4,965	499,04
20	2481,10923	4,9243	499,10
30	2461,96921	4,8854	499,16
40	2443,54069	4,8481	499,21
50	2426,00403	4,8124	499,28
60	2409,42939	4,7781	499,40
70	2393,10527	4,7453	499,43
80	2378,13398	4,7139	499,58
90	2363,27293	4,6838	499,63
100	2349,33381	4,655	499,73
110	2335,67527	4,6274	499,76
120	2322,48772	4,6009	499,79
130	2310,21208	4,5756	499,87
140	2298,37736	4,5514	499,93
150	2286,91342	4,5282	499,97
180	2255,47778	4,4643	500,10
200	2236,5583	4,426	500,16
250	2196,90559	4,3438	500,51
300	2163,92687	4,2775	500,58
350	2137,93278	4,224	500,78
400	2116,5784	4,1809	500,87
500	2086,63641	4,118	501,26
600	2066,09411	4,0773	501,24
800	2044,36007	4,0323	501,47
1000	2037,75776	4,0152	501,94

From 5 µs to 1000 µs
Mean activity: 500.0 Bq
Standard deviation: 0.8 Bq (0.16 %)

From 5 µs to 100 µs
Mean activity: 499.33 Bq
Standard deviation: 0.24 Bq (0.05 %)

Extrapolation to zero dead-time:
497.28 Bq