Report of the CCRI(II) Transfer Instrument Working Group

2011-2013

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Objective: Support to the BIPM in developing the SIR transfer instrument for short-lived radionuclides. Guidance in evaluating the best experimental setup and the most appropriate method of data analysis. Providing advice in defining comparison protocols.

Summary: The BIPM.RI(II)-K4.Tc-99m ongoing comparison is now running at a rate of two comparisons per year: NMIJ 2011 (result published), NIM 2012 (draft B), CNEA 2012 (draft A), LNMRI and IFIN-HH planned for 2013. The developments for the extension to ^{18}F are in progress.

1. TIWG meeting

In May 2012, the TIWG(II) met to discuss the present status and updates of the SIRTI equipment, the outcomes of the first three ^{99m}Tc comparisons (NIST, KRISS and NMIJ) and the extension to ^{18}F and ^{11}C.

1.1 Stability of the SIRTI

The ^{94}Nb measurement results are monitored over time in order to check the integrity of the detector following the many overseas travels. Each ^{94}Nb measurement is preceded by a threshold setting using the ^{93m}Nb x-ray peak and followed by a background measurement. The results are of course corrected for background and decay and thanks to the very long ^{94}Nb half-life (20 300 (1600) a, [1]), the uncertainty of the decay correction is negligible (and will remain so for at least the next 50 years). The results are plotted in figure 1 showing the long term stability of the SIRTI and no significant trend is presently observed. However the reduced chi-squared value is 2.7 showing that the quoted uncertainties are underestimated. As each ^{94}Nb measurement consists in the mean of 10 measurements of 700 s, and the measurement uncertainty is based on the standard deviation of these 10 measurements, fluctuations of the background and temperature during measurement should be covered by the standard deviation. In addition, a plot of the results versus temperature (figure 2) does not show any dependence. The reason for the chi-squared value larger than one is thus not clearly identified. Consequently, the external standard uncertainty of the weighted mean of the stability measurements of figure 1 is used as an additional uncertainty component of the SIRTI ^{99m}Tc measurement uncertainty.

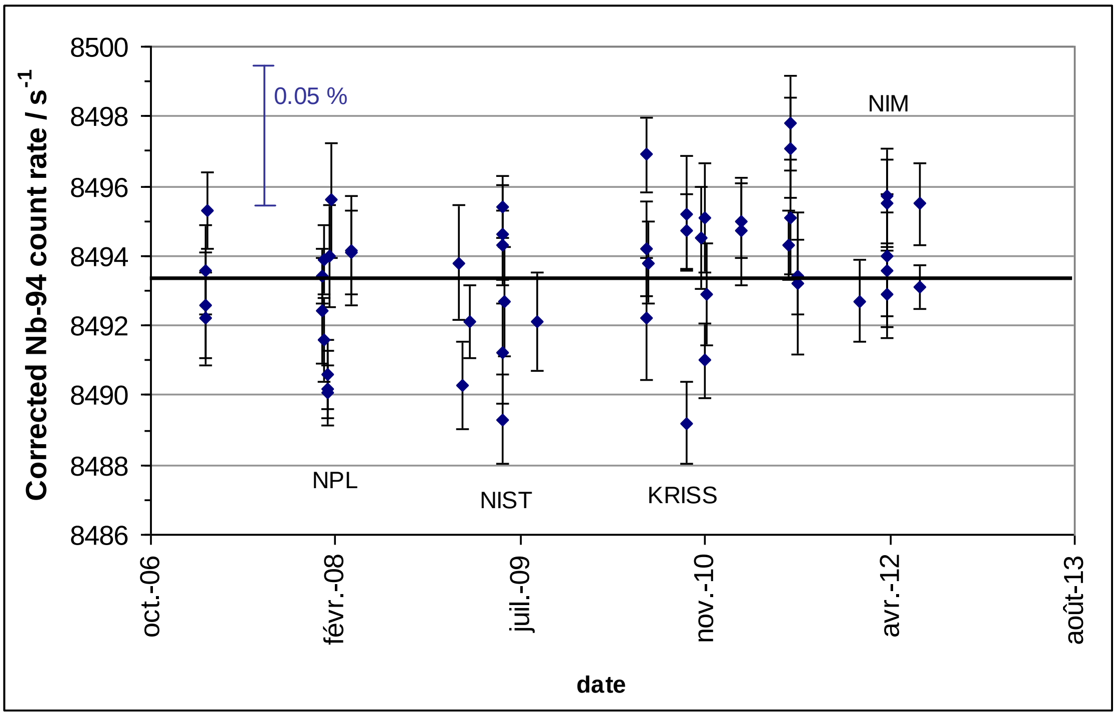

Figure 1. Long term stability measurements of the SIRTI. The line indicates the weighted mean value.

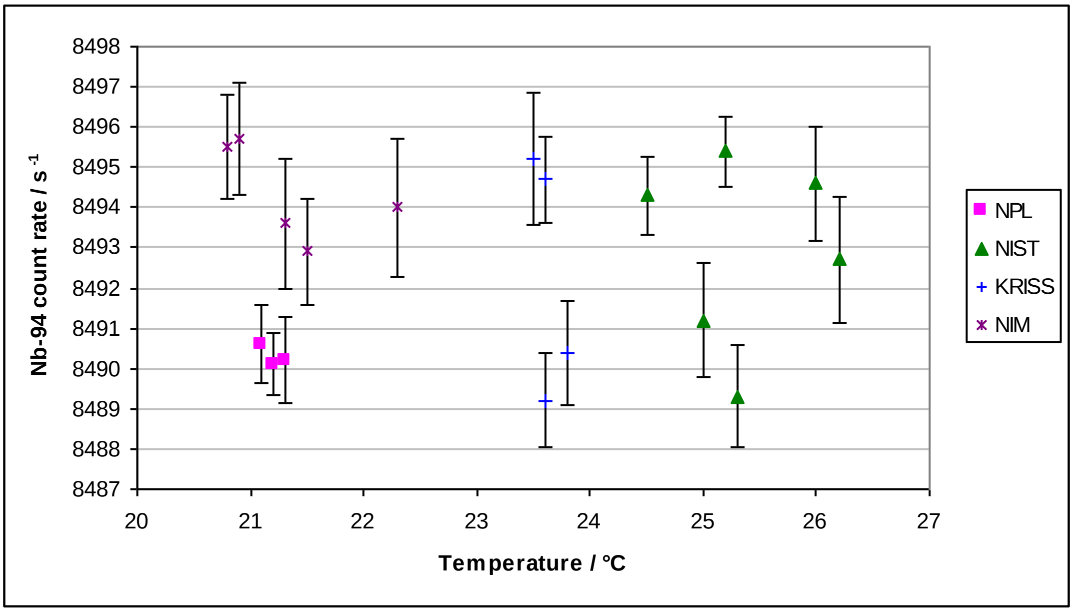

Figure 2. Reproducibility of the Nb measurements. The corrected ^{94}Nb count rate above the threshold is independent of temperature.

1.2 Dependence of the SIRTI versus count rate

The behaviour of the SIRTI versus the count rate was determined by measuring strong ^{99m}Tc sources and looking at the SIRTI measurement results as a function of the ^{99m}Tc count rate in the detector. Each series of results was renormalized to the KCRV in order to eliminate the influence of the ^{99m}Tc activity measurement by the NMI. The equivalent activity A_E results, which are corrected for decay, should be constant but this is not the case (see figure 3) and the deviation observed is identical for the two measurements of the NPL and LNE-LNHB ampoules. A relative effect of 1.8×10^{-3} is observed at 50 000 s^{-1}.

Underestimated A_E values correspond, by definition [2], to overestimated count rates and are thus consistent with a shift of the energy spectrum towards high energy (assuming a fixed threshold). Indeed the 140 keV γ-ray peak position increased from channel 813 to channel 832 when the count rate increased from 38 000 s^{-1} to 120 000 s^{-1}. Possible explanations could be, a priori, a drift of the PMT gain, a shift of the amplifier baseline related to a slow phosphorescent component of the NaI(Tl) light ("afterglow" [3]), the pile-up of below-the-threshold pulses or a bias in the MTR2 live-time correction. The latter two possible causes were investigated and quantified but do not seem to be able to produce such a large effect versus count rate. The former two are related to the peak shift observed (19 channels) and assuming the same shift takes place at the level of the threshold, the effect on the equivalent activity is calculated to be one order of magnitude less than the bias observed in figure 3.

The origin of the effect observed in figure 3 is thus not completely understood and consequently the fitted line is not considered as sufficiently robust to be used to correct the A_E data. It was decided to rather use this fitted line to evaluate a type B uncertainty and to limit the count rate in all comparisons to 20 000 s^{-1}.

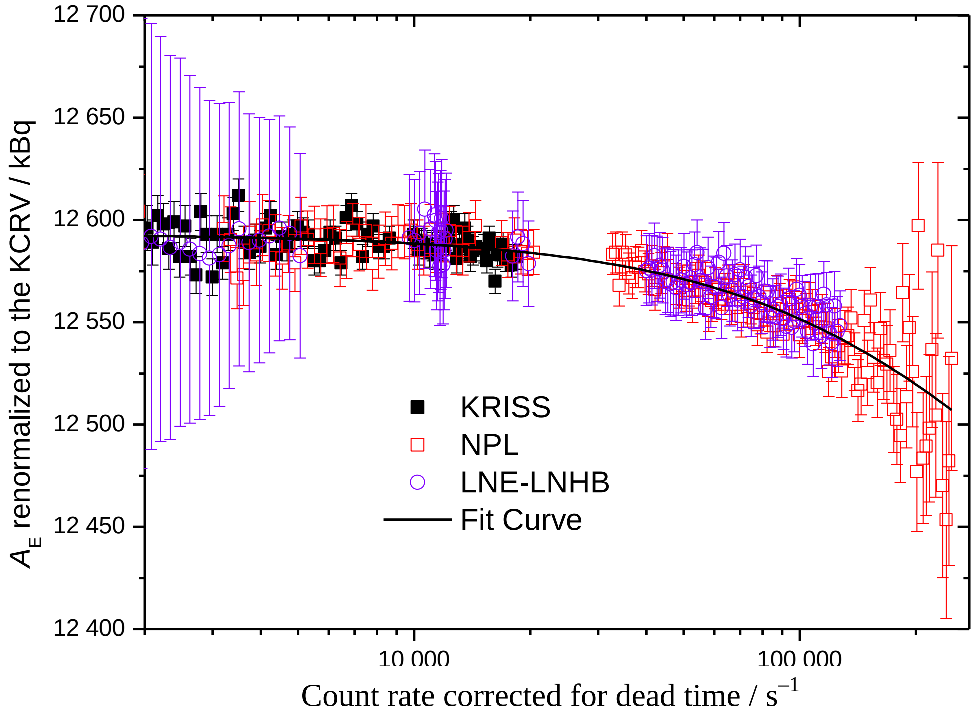

Figure 3. Dependence of the SIRTI response as a function of count rate.

1.3 Copy of the SIRTI NaI(Tl)

A copy of the SIRTI well-type NaI(Tl) has been purchased at the BIPM. In case of damage of the original detector the copy would allow to continue the comparison programme with no delay. The CIEMAT and the NPL volunteered to each send a ^{99m}Tc SIR ampoule to the BIPM that would allow calibrating the copy detector against the SIR.

1.4 Outcome of the SIRTI comparisons at NIST, KRISS and NMIJ

The results of the K4 comparisons are very satisfactory as shown in figure 4. An overview of the comparisons carried out at the NIST, KRISS, NMIJ and the NIM is given hereafter.

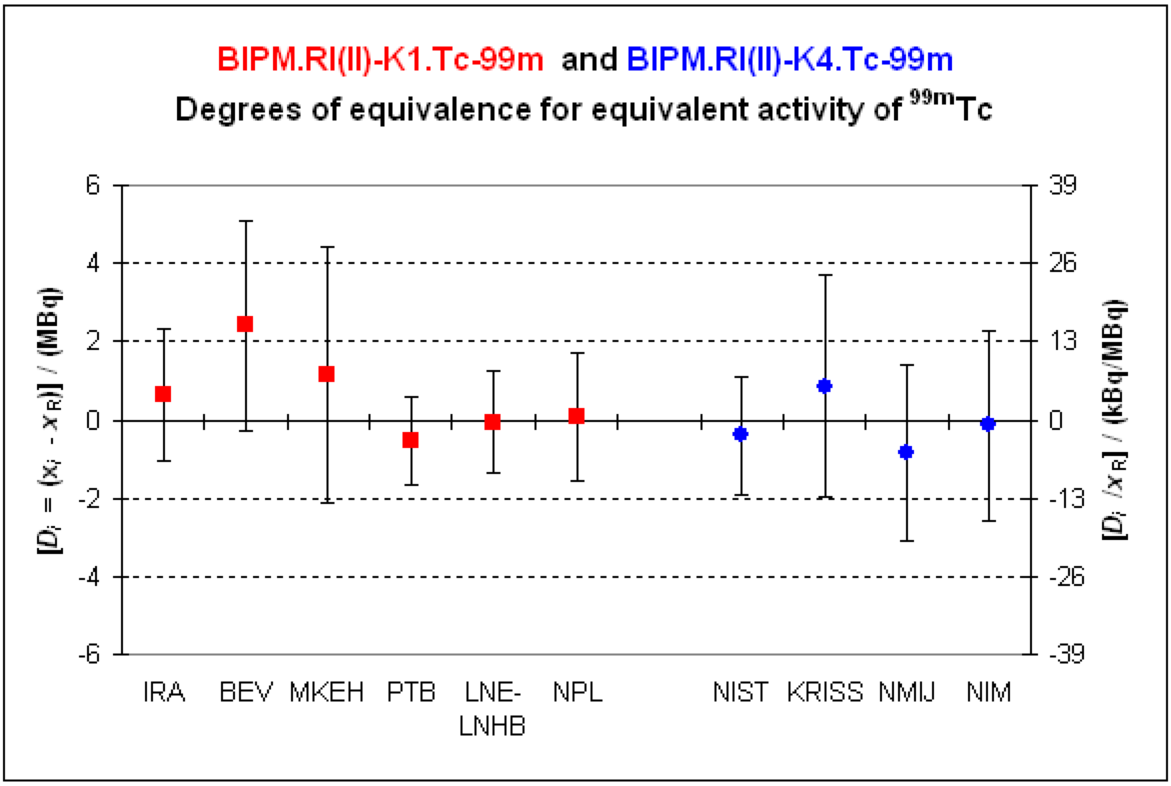

Figure 4. Degrees of equivalence for the SIR (red) and SIRTI (blue) comparisons

NIST 2009

There was a significant effect (0.2%) from drops forming in the top of the ampoule, confirmed by Monte-Carlo simulations. Centrifugation of the ampoule could solve the problem.

KRISS 2010

- The background level was twice that measured at the BIPM. A possible reason for this might be from high radon level present at the KRISS. The background was stable however so the measurements proceeded.
- There was no droplet in the ampoule (unique case in the SIRTI comparisons to date) probably because the KRISS rinsed the ampoule with a siliconizing solution to limit adsorption as part of their usual procedure.

NMIJ 2011 (during ICRM2011).

- The NMIJ did not perform a dilution of the solution, but waited several days for the activity to decay. However no ^{99}Mo impurity interfered with the ^{99m}Tc measurement.
- There were some small drops in the top of the ampoule but having this time a negligible effect on the results as shown by Monte-Carlo simulations.
- Problems with the MRT2 module gave an extra uncertainty of 5×10^{-4} to the NMIJ result. In the further SIRTI comparisons an expert in electronics from the BIPM, M. Nonis,

participated in the measurements and a second MTR2 module was taken to the participating laboratory.

- The measurement procedure identified a decreasing trend in the background which was linked to a ^{99}Mo source (300 MBq) that was stored in a neighbouring room.
- There has been no problem from an earthquake (magnitude 5.3) that occurred during measurements thanks to extra stability added to the instrument in preparation of such an event – the whole comparison having been delayed due to the massive earthquake earlier in 2011.

NIM 2012

The preliminary comparison result calculated at NIM during measurements did not pass the Normalized Error test at the $k = 4$ level. Following the SIRTI protocol, the NIM was immediately informed of the problem, without being informed of the result it-self. The NIM could investigate the problem and found a typo in their calculation of the activity. The corrected activity result provided then by the NIM was very satisfactory.

1.5 Extension to ^{18}F and ^{11}C

a. A plastic liner has been machined at the BIPM to replace the brass liner used for ^{99m}Tc. This would prevent positrons reaching the detector while minimizing bremsstrahlung.
b. Monte-Carlo simulations show that the threshold setting is sensitive for ^{18}F, while this was not the case for ^{99m}Tc. If a larger well detector is used, the effect would be reduced, but not sufficiently so the original detector was decided to be sufficient. Indeed the ^{18}F measurements would be quicker and so less sensitive to daily temperature changes that influence the threshold position.
c. Results from the measurement of a ^{18}F ampoule from LNE-LNHB (2010) showed a 9×10^{-4} relative shift for the second measurement series that could not be explained. It should be noted however that the reduced chi-squared value is lower than one. See figure 5. Further tests with a ^{137}Cs source to mimic ^{18}F are in progress (see section 2).
d. The question of using a digital data acquisition system was raised. C. Bobin is developing a digital system at LNE-LNHB with off-line analysis and dedicated for NaI(Tl) detectors. CAEN has systems with triangular shaping that can undertake high-rate digital sampling that might be worth trying. ULS (Korea) is another provider of digital systems. For the ^{18}F comparison, analogue and digital systems could be used in parallel. However, the accuracy of the dead-time correction should be investigated in detail.
e. Once the SIRTI is validated for ^{18}F, no additional developments are necessary for the extension to ^{11}C and ^{64}Cu.
f. ^{18}F comparison scheme: the possibility to group the SIRTI comparisons of several European countries in a host laboratory has been discussed. This would enable to reduce the queue for participants but would need that the participating NMIs move their equipment to the host laboratory. A further discussion at the level of the KCWG concluded that very few NMIs could participate in such a comparison scheme and the project of group comparison was abandoned.

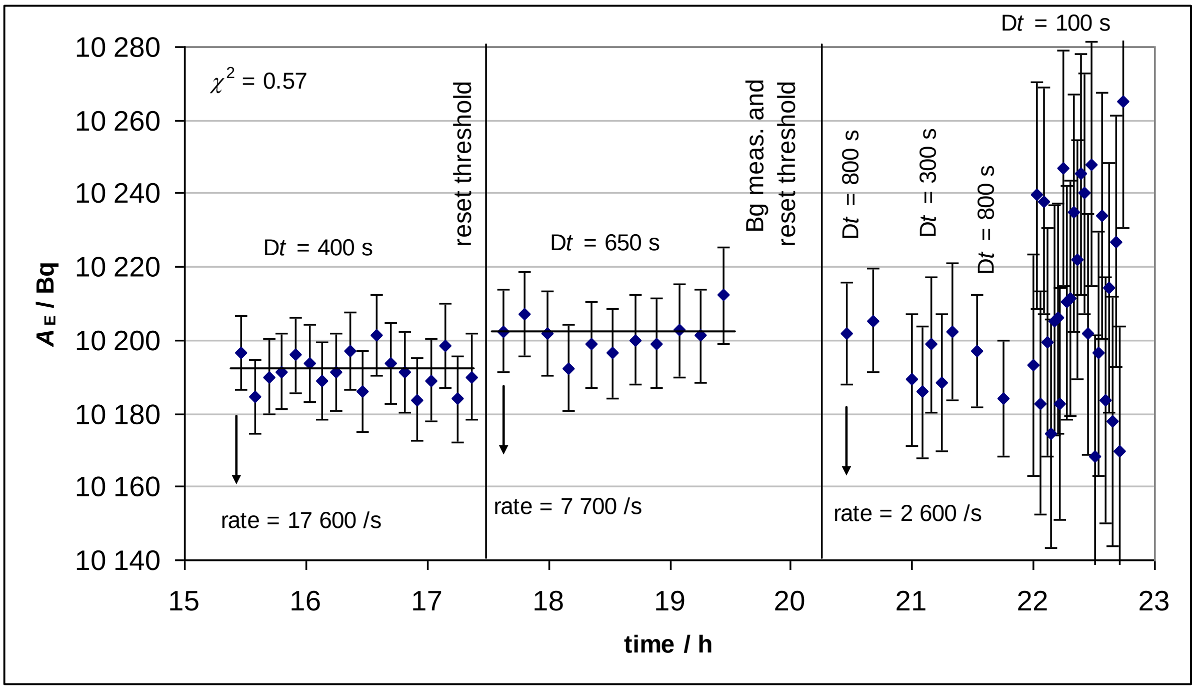

Figure 5. Trial ^{18}F measurements in the SIRTI. A 9 × 10^{-4} relative shift between the first two series of measurements is observed. D*t* is for measurement duration.

2. Progress since the last TIWG meeting

Hereafter, achievements about the SIRTI obtained since the last TIWG(II) meeting in May 2012 are reported.

- The SIRTI is now part of the BIPM quality management system and passed an external audit in September 2012.

- New USB scalers from National Instrument (USB-6341) running under LabView were setup at the BIPM by M. Nonis. They were tested using a 1 MHz frequency from the time department. They will replace the ORTEC scalers used until now and which are not robust (unstable connection with the computer).

- The MCA from Amptek has been upgraded to be compatible with Windows 7.

- Tests were carried out with a ^{137}Cs source in a brass holder in order to mimic ^{18}F. Repeatability was measured where each ^{137}Cs is preceded by the threshold setting using the Nb reference source, and followed by a background measurement. The results are consistent with uncertainty ($\chi^2 = 1.3$) as shown in figure 6.

- Monte Carlo simulations of the SIRTI for ^{18}F

The geometry for the simulations has been modified to replace the brass holder used for ^{99m}Tc by a plastic (PVC) holder for beta emitters. The simulations using Penelope 2008 confirmed that the β^+ emitted by ^{18}F ($E_{\beta\text{max}}$ = 634 keV) are stopped by the PVC while β^+ particles with an energy of 1 MeV reach the Al detector walls of the detector but not the sensitive part of the NaI(Tl). The allowed beta spectrum of ^{18}F has been calculated using the BIPM program Simpbeta [4] and is given in the input file of the Penelope program. Calculation of the detection efficiency for ^{18}F is in progress.

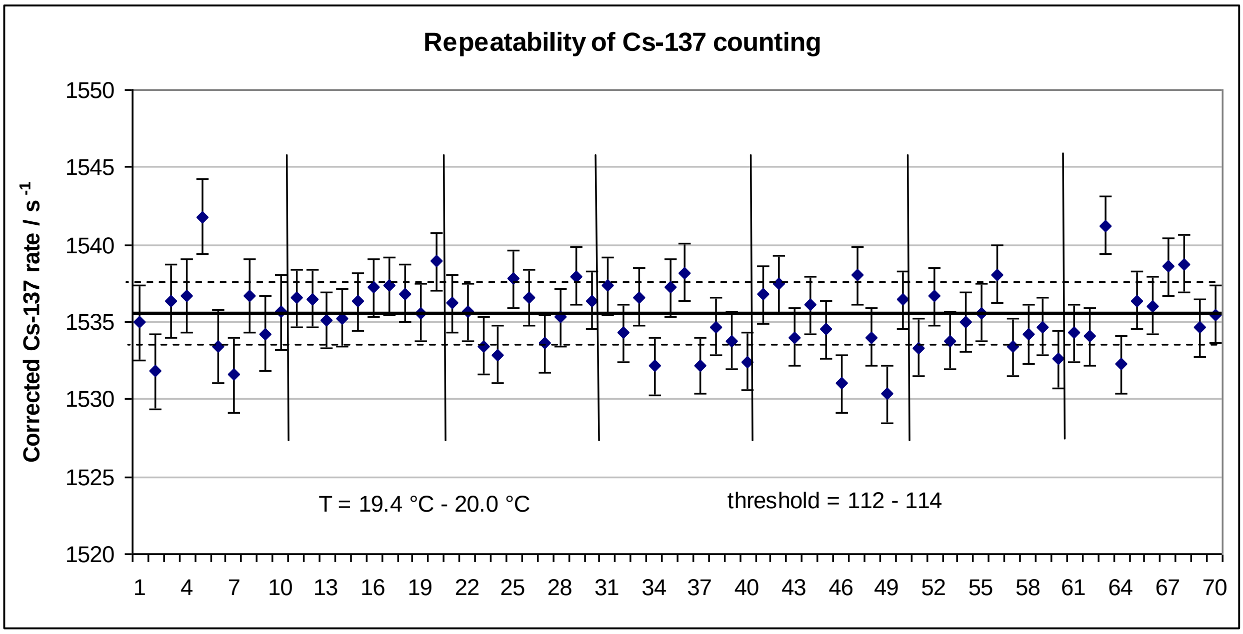

Figure 6. Repeatability of ^{137}Cs counting in a brass holder to mimic ^{18}F. The full horizontal line corresponds to the weighted mean and the dotted lines to the standard deviation. The vertical lines separate the different measurement series.

3. Future plans

The development of the SIRTI for ^{18}F will continue first by studying the behaviour of the SIRTI at high count rate when bipolar signals are used at the amplifier output instead of unipolar ones. Secondly, the SIRTI uncertainty budget will be evaluated for ^{18}F. This means that Monte-Carlo simulations need to be carried out to quantify the influence of the thickness of the ampoule walls, the solution volume and density. This was already done for ^{99m}Tc (γ-ray of 140.5 keV) but should be repeated for ^{18}F (β^+ spectrum). The program STEFFY [5] from S. Pommé could also be used.

By the end of 2013, the SIRTI should be ready for calibration against the SIR. The LNE-LNHB and the NPL volunteered to send ^{18}F SIR ampoules to the BIPM for this purpose. A trial ^{18}F K4 comparison could then be organized at a European laboratory already having results in the SIR for this nuclide.

4. Other news about the SIRTI

- The SIRTI is presently in Argentina awaiting customs issues to be solved. The next comparison is planned for June 2013 at LNMRI, Brazil, and a copy of the SIRTI equipment (including the new NI USB scalers) has been assembled and tested before exportation to

Brazil. Further planned K4 comparisons for ^{99m}Tc are IFIN-HH (2013), VNIIM (2014) and NMISA (2015). Other NMIs which showed interest are the BARC, ANSTO and NRC.

- In June 2012, a change of the BIPM policy entitled “BIPM measurement and consultancy services policy” took place for comparisons necessitating the travelling of the BIPM staff to the participating NMI: the participants are now requested to pay the local accommodation of the BIPM staff during the comparison. This is reflected in the latest version of the protocol of the K4 comparison on the KCDB. However this does not apply to laboratories that registered to the participants list before June 2012.

- Planned ^{18}F K4 comparisons are ENEA in 2014 (together with a ^{99m}Tc comparison) and the NIST (2015). An inquiry on the interest of NMIs in the SIRTI comparisons showed that 12 NMIs would like to participate for ^{18}F but only three of them declared to have a primary standardization method. Three other laboratories would like to take part but are situated sufficiently close to the BIPM for sending an ampoule to the SIR by road. Finally, 12 laboratories showed interest in a ^{11}C K4 comparison; five of them would have a primary standardization method. The ^{18}F and ^{11}C comparisons should be organized jointly in order to minimize the costs.

References

[1] NUDAT2.5, National Nuclear Data Center, Brookhaven National Laboratory, based on ENSDF and the Nuclear Wallet Cards.

[2] Michotte C., SIR Transfer Instrument, Protocol for the ongoing comparison of ^{99m}Tc on site at the NMI, BIPM.RI(II)-K4.Tc-99m, 2012, v2.9.

[3] Knoll G.F., Radiation detection and measurement, 1979, John Wiley & Sons ed., p. 258 and 293.

[4] Michotte C., Calculation of the shape of beta spectra based on the tables of Behrens & Jänecke, BIPM report in preparation.

[5] Pommé S., STEFFY - software to calculate nuclide-specific total counting efficiency in well-type gamma-ray detectors, Appl. Radiat. Isot., 2012, **70** (9), 2070-2074.