# Future CCRI(II) Key Comparisons – A Discussion

**M J Woods (NPL)**

The choice of radionuclides to be the subject of BIPM CCRI(II) key comparisons is decided by a consensus at the biennial meetings of CCRI(II). In the past these choices have been influenced primarily by the desire to select radionuclides that exhibit particular decay characteristics and associated challenges to the standardisation techniques available to NMIs. The choices, therefore, have been driven by metrological considerations.

Over the past decade or so, there has been a significant increase in the number of radionuclides for which standards have been required by the various user communities (nuclear, environmental, medical, etc). In addition, for a variety of reasons, the priorities for standards have changed and it is only recently that CCRI(II) key comparisons have started to reflect these changes. It is important that future comparisons make due recognition of these changes in user-priorities whilst still seeking to improve the basic metrological techniques.

It is proposed therefore that CCRI(II) should review its decision-making process and seek to better accommodate the various, and sometimes, conflicting metrological and user demands.

## Metrological Considerations

The need to continue to address the metrological issues is perhaps best reflected by the results of existing key comparisons that, of course, include the entries to the SIR database. The existing data for these are readily available and it is debatable whether these indicate whether there has been any significant improvement over the years in the achieved (and achievable) accuracy of absolute standardisations. The ratios of the external to internal uncertainties in the data tables can give some indication of the problems surrounding the standardisations of particular radionuclides. In the ideal situation, this ratio should be one. If the ratio is larger than one, this suggests that the uncertainties on individual values may be unrealistically low or that there are significant differences between the results from different NMIs and/or techniques. There are a significant number of radionuclides for which the ratio is equal to or greater than two but it should be noted that, in some cases, there are some obvious outliers in the data sets that need to be excluded and, once excluded, this may result in a much improved ratio. It would be premature therefore to use these ratios until a critical analysis of the key comparison data has been conducted. It is recommended that this analysis should be initiated as soon as possible.

**User-Community Considerations**

In respect of user-community requirements, it is useful to examine the various sectors that use radionuclide standards and to prioritise those needs. Table 1 provides a tentative list for consideration. This list has been separated into several categories and the following is a discussion of each.

*Gamma Spectrometry and Ionization Chamber Calibration Sources*

The list of 13 radionuclides represents those that are most commonly included in the mixed-radionuclide solutions and solid matrices that NMIs and secondary standards laboratories provide for the calibration of gamma spectrometry and ionization chamber systems. A cursory examination of the SIR database indicates that $^{57}$Co and $^{65}$Zn are those radionuclides for which the agreement between NMIs may be most problematic but the previous comments in respect of outliers should be borne in mind. These two radionuclides also play particularly important roles in the determination of the efficiency calibration curves for both gamma spectrometers and ionization chambers.

The 122 keV photon from $^{57}$Co is in the energy region where the calibration curve is changing rapidly and the accuracy of the calibration standard is particularly influential. It is also in the energy region which is preferred by the medical community for diagnostic examinations.

For $^{65}$Zn, the 511 keV photon emissions resulting from the positron emission has long been a cause for concern. The production of calibration curves, both for gamma spectrometers and for ionization chambers, has been complicated by the poor fitting that has been indicated for the 511 keV photon from $^{65}$Zn. Recent work at LNHB has suggested that the existing emission probability values may be in error by one or two percent and proposals are expected to be made by EUROMET to initiate a project to re-investigate this data. Such investigations will rely heavily on the ability to provide accurate, absolutely standardised solutions of this radionuclide. It might be appropriate to harmonise these researches by a coordinated approach which encompasses a CCRI(II) key comparison of the activity standardisation and a EUROMET-led determination of the 511 keV photon emission probability. As an additional (and metrological) interest, $^{65}$Zn is a radionuclide that decays by two different routes, namely electron capture and positron emission. Current experiences with other multiple decay mode radionuclides (such as $^{152}$Eu, $^{192}$Ir and $^{204}$Tl) have demonstrated the need to seek improvements in the ability of absolute measurement techniques to standardise such radionuclides.

*Medical User Standards*

For the medical user community, by far the most commonly used radionuclide is $^{99m}$Tc. Although the entries to the SIR database indicate that there is no significant problem, the number of entries is small and only represents what must be a very small fraction of the countries (and medical communities) which employ this radionuclide. This small number is no doubt influenced by the short half-life and the problems of transporting it to BIPM in a reasonable period of time. Given the predominance of this radionuclide, it is important that more NMIs provide entries to the SIR database but a key comparison, as normally envisaged, seems to be unrealistic.

It is expected that there will be an increase in the application of positron emission tomography (PET) in coming years and this technique relies on the use of relatively short-lived positron-emitting radionuclides (such as $^{11}$C, $^{18}$F, $^{68}$Ga and $^{124}$I). With the exception (perhaps) of $^{124}$I, it is unrealistic to expect that sources can be transmitted from NMI to BIPM and an alternative approach needs to be considered if equivalence is to be established between NMIs. There is currently an ICRM-led equivalence exercise for $^{18}$F that is exploring the options for an alternative approach using the NPL secondary standard radionuclide calibrator, whereby equivalence may be established remotely and transferred to the SIR system using an NMI that has close access to BIPM. It is suggested that the results of this exercise are awaited before any decisions on short-lived medical radionuclides are taken. Should it prove worthwhile, protocols would need to be established to ensure the robustness of such a system.

*Environmental Standards*

Although there are many radionuclides which are of importance in environmental monitoring, the isotopes of Pu, U, Th, Am and Np still hold and will continue to hold a significant importance in this area. In general, these radionuclides are alpha-particle emitters with minimal photon emission. There are some saving graces, however, in that, if one of the isotopes of a particular element can be standardised, it may well be possible to achieve sufficient secondary standardisation accuracy of the other isotopes using relative measurements via mass- and/or alpha-spectrometry. The requirement therefore is to establish equivalence between NMIs for at least one of the isotopes of each element. For Am, SIR and key comparison data exist for $^{241}$Am whilst for Pu, a key comparison of $^{238}$Pu is currently in progress. $^{241}$Pu is a special case because of its low energy beta emission. The final report of a EUROMET project on $^{237}$Np is about to be published and, within that exercise, some of the stock solution was submitted to the SIR system. For U and Th, the position is less satisfactory and an isotope of each of should be considered for key comparison purposes. It should be noted that these radionuclides also present some interesting and challenging metrological challenges that arise from the decay chain progeny that may or may not be in secular equilibrium with the parent.

**Recommendations**

There is a need for CCRI(II) key comparisons to address both the metrological and the user community needs. Where possible, it would be advantageous to address both issues within the same comparison exercise. The priorities for different NMIs will depend heavily on the demands from within their own national communities and the CCRI(II) key comparisons must try to balance these. It is clear however that there are some radionuclides which have a universal appeal and the discussion above relates to that. It is recommended that the following radionuclides be regarded as priorities for future comparisons:

$^{65}$Zn (gamma spectrometry standard, two decay modes, apparent poor agreement now) *(with consideration of harmonising the comparison with a parallel project to improve the accuracy of the absolute photon emission probabilities)*;

$^{57}$Co (gamma spectrometry standard, poor agreement now);

$^{228}$Th (environmental standard, decay chain complications, relative standard for other Th isotopes);

$^{232}$U (environmental standard, decay chain complications, relative standard for other U isotopes);

$^{99m}$Tc (medical standard, few SIR entries)
*( await the results of the ongoing, ICRM remote $^{18}$F comparison)*

Table 1 Radionuclide standards required by user communities

| |
|---|
| Calibration standards for gamma spectrometers and ionization chambers |
| $^{51}Cr$ $^{54}Mn$ $^{57}Co$ $^{60}Co$ $^{65}Zn$ $^{85}Sr$ $^{88}Y$ $^{109}Cd$ $^{113}Sn$ $^{137}Cs$ $^{139}Ce$ $^{203}Hg$ $^{241}Am$ |
| |
| Calibration standards for medical community |
| $^{32}P$ $^{89}Sr$ $^{90}Y$ $^{67}Ga$ $^{81m}Kr$ $^{99m}Tc$ $^{111}In$ $^{123}I$ $^{125}I$ $^{131}I$ $^{153}Sm$ $^{186}Re$ $^{188}Re$ $^{192}Ir$ $^{201}Tl$ |
| |
| Calibration standards for environmental monitoring community |
| $^{226}Ra$ $^{228}Ra$<br>$^{228}Th$ $^{229}Th$ $^{230}Th$ $^{232}Th$ $^{234}Th$<br>$^{232}U$ $^{233}U$ $^{234}U$ $^{235}U$ $^{236}U$ $^{238}U$<br>$^{237}Np$<br>$^{238}Pu$ $^{239}Pu$ $^{240}Pu$ $^{241}Pu$ $^{242}Pu$<br>$^{241}Am$ $^{243}Am$ |