Recommendations of the CCRI(II) Working Group on activity measurements, particularly for ^{204}Tl

1. Adsorption and Residual activity

To mimimize adsorption, the ampoules to be used should be conditioned with non-active solution of the same chemical composition.[1]

All residual activity in the ampoule should be accounted for in the final result whether it comes from surface film after the liquid is removed or because some of the activity is adsorbed. To avoid an unnecessary increase in the measurement uncertainty due to the correction for the residual activity, rinsings are not recommended unless liquid scintillation (LS) techniques are not available. For the assessment of residual activity using LS, it is better to use a water immiscible cocktail to keep the efficiency at around 50 % for the activity adsorbed on the walls and in the surface film, which will also minimize the uncertainty. In the event that a suitable method is not available for the direct measurement of residual activity in the ampoule, it is recommended to rinse the ampoule with an aggressive solution to remove most (if not all) the activity and prepare sources for measurement by another method e.g. proportional counting (PC).

2. Carrier concentration and acidity

To use carrier concentrations generally between 20 µg/g of the ion and 50 µg/g[2]; and for ^{204}Tl, 30 µg/g of Tl^{+}; for LS the carrier concentration might have to be adapted to maintain the stability of the solution; high concentrations do not appear to affect LS results for ^{204}Tl but they do cause problems in the production of solid sources used for proportional counting (PC) particularly through high self-absorption.

Any addition to the original solution should maintain the original solution pH so as to keep the ions in solution and not change the oxidation state of the radionuclide.

TlCl is photosensitive in much the same way as AgCl which might have consequences which are not currently known.

3. Chemical Composition

To keep $^{204}TlCl$ as the preferred chemical format; although using $^{204}Tl\ SO_4$ in the 1964 comparison apparently produced more coherent results, there is no fundamental reason why $^{204}TlCl$, a more common radiopharmaceutical form, should not produce equivalent results.

4. Impurities

The solution should be checked for impurities; impurities were not a problem in the activity measurements for ^{204}Tl.

5. Solid sources

Great care needs to be taken in preparing solid sources particularly for PC; film absorption is much less than self-absorption for a high carrier concentration and a non-

[1] Monographie BIPM-1 (1975)
[2] NCRP 58 has a compilation of guideline values

homogenous distribution. Correction factors greater than 10 % can be involved and the effect on the extrapolation curve for tracer activity measurements can be significant.

For weighing and deposition, a stable laboratory environment is important with humidity control between 50 % and 70 % to reduce static electricity problems.

6. Instrumentation

Instrumentation to measure absolute activity does not necessarily need more than one detector, depending on the method, but this needs to be adequate and used appropriately e.g. in LS counting equipment, anti-coincidence guards reduce detection efficiency and thresholds greater than 5 keV make it difficult to produce good results for ^{204}Tl; in LS with pulse durations of about 5 ns, coincidence resolving times greater than a few tens of nanoseconds need appropriate corrections.

7. Liquid scintillation techniques

LS techniques generally produce reliable and consistent results; the CIEMAT-NIST method seems to agree with the LS tracer efficiency methods. For comparison of the results, the appropriate quench parameter to use is the Figure of Merit expressed as e/keV. Quenching of the solution of radionuclide under investigation is not strictly necessary but does act as a quality control on the systems.

8. Proportional counter (PC) based tracer efficiency methods

The loss of high energy electrons must be corrected; tracer efficiency methods must take account of the PC gas pressures and PC dimensions to correct for losses of electron energy e.g. using an 8 cm diameter 15 cm long PC, at 1 MPa, 200 keV is totally absorbed but high energy electrons might produce little ionization because of the low linear energy transfer which will result in low beta efficiencies.

Other negative effects include the non–compatibility of beta energies and spectral shapes for the tracer and the radionuclide under investigation, and the possibility of non co-precipitation.[3]

Ideally the tracer activity should be in the same proportion as the solution activity but in any case not different by more than 20 % in the source to be measured.

Extrapolation methods used in the comparison were not always consistent with the theory; in all cases at each measurement point the tracer contribution should be subtracted from the total measured response, taking into account the different half lives, before the extrapolation function is established.[4]

9. Uncertainties

Care needs to be taken when producing uncertainty budgets and in combining uncertainties. Error propagation for each uncertainty component should follow the ISO recommendations.[5] Many inconsistencies have been noted, for example not all laboratories include uncertainties for the same components. Often the type B uncertainty is the largest component and indeed the limiting factor for the technique. The uncertainties on the tracers

[3] Lowenthal (?1987)
[4] Funck
[5] GUM

are much smaller than the discrepancies seen in the results and so do not appear to be the cause.

10. Future Work

Further experiments

The IRMM will study the effects of covered and non-covered sources in their PC at pressures up to 1.5 MPa.

The IRMM and the BNM-LNHB will assess the effects of photosensitivity in the ampoule. A non-active solution and chemical methods to check the thallium concentration can be used.

The NPL will study the corrections needed for solid sources. (This will involve putting a project proposal forward and will only be undertaken if the proposal is accepted within their budget.)

The IRMM will study self-absorption by changing carrier concentrations and confirm the absolute activity by LS methods.

Before August 2000, the BIPM will produce a batch of 40 ampoules of ^{204}TlCl at low carrier concentration for the various experiments which have been identified.

Further theoretical study

There is a need to simulate the spectra coming out of the PC and BNM-LNHB will extend their work to the higher energies at which gas ionization occurs. They are also studying the effect of using different gases in PCs.

BIPM will undertake to start work on theoretical simulations using the Monte Carlo code PENELOPE.

Comparison Protocol

It is recommended that the *Monographie BIPM-1* (1975) should be updated and extended by the CCRI(II).

The Extended SIR Working Group should take into account the *Monographie BIPM-3* (1980) when producing their new monograph on liquid scintillation techniques.

The BIPM will rewrite the comparison protocol taking into account the recommendations of the Working Groups.

Study trial comparison with invited participants

It is recommended that once the further experimental work has identified solutions to the problems encountered, a new trial comparison including at least the BNM-LNHB, NPL, IRMM and BIPM, all of whom will use at least two techniques, should be held. Two other participants using non-LS techniques could also be invited to take part.

Case study

A case study on the ^{204}Tl comparison illustrating the possible pitfalls in a key comparison would be a significant aid to future comparisons and for teaching purposes. IRMM could coordinate the production of such a report, which would be a reference work incorporating the earlier ^{204}Tl comparison of 1964.

Full scale comparison

The CCRI(II) should undertake a full-scale comparison of ^{204}Tl activity measurements once the trial comparison has produced consistent results across the different methods.