Intercomparison of $^{67}$Ga solution sources in UK Hospitals, 1996

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ABSTRACT

During 1995, as part of a routine QA programme for radionuclide calibrators in the North West region of the UK, it was observed that a group of radionuclide calibrators gave consistently low readings for $^{67}$Ga when compared against the value predicted by a secondary standard calibrator. This shortfall ranged between 5.5 % and 6.7 %.

It was decided that a resolution of this problem could best be achieved by an intercomparison and an exercise was mounted between the National Physical Laboratory, Amersham International plc and the hospital physics community.

The conduct of that intercomparison is described here, the results are discussed and recommendations made.
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1 INTRODUCTION

Diagnostic nuclear medicine plays a critical role in the optimisation of healthcare. Whether or not the maximum benefit is achieved from its practice depends partly on the quality of the associated measurements and the ability to balance the level of useful information obtained against the detrimental effects of ionising radiation. This has led to the requirement that the total activity of any radioactive administration shall be known with confidence to a sufficiently high degree of accuracy and precision.

There are numerous techniques and facilities for determining activity but the most economic method is via the use of radionuclide calibrators. Their principle of operation is relatively simple and, used correctly, they will easily achieve the levels of accuracy required by the diagnostic community. Correct use implies certain pre-requisites such as trained operators, traceability, regular quality control procedures, control of operating conditions - in essence, a quality system.

A protocol for establishing and maintaining such a system has been recommended for use in UK hospitals(1). Amongst many of the quality aspects it addresses are those of traceable calibration factors and regular recalibration. It is recognised that calibration factors are calibrator dependent, radionuclide dependent, and vary with sample geometry and container. It is important, therefore, that individual calibration factors are checked and confirmed on a regular basis. Generally, recalibrations are conducted according to individual hospital schedules. Occasionally, however, general concerns arise which are best addressed by the conduct of intercomparisons across the whole measurement community. Such intercomparisons serve not only as a means of recalibration but also of identifying and focusing attention on particular problem areas. In addition, these exercises can provide an overall estimate of the performance level of the measurement community and the confidence which can be placed in it.

During 1995, as part of a routine QA programme for radionuclide calibrators in the North West region of the UK, it was observed that a group of radionuclide calibrators gave consistently low readings for $^{67}$Ga when compared against the value predicted by a secondary standard calibrator. This shortfall ranged between 5.5 % and 6.7 % and was reported in a Radionuclide Topic Group Newsletter(2) of the Institute of Physical Sciences in Medicine (now the Institute for Physics and Engineering in Medicine and Biology).

It was decided that a resolution of this problem could best be achieved by an intercomparison and an exercise was mounted between the National Physical Laboratory (NPL), Amersham International plc (AI) and the hospital physics community.

2 PARTICIPANTS

Participation was open to all UK hospitals and the exercise was publicised via the contact mailing lists of AI and NPL.

Several participants took the opportunity to share their sample by circulating it amongst several hospital departments in their region.

A list of participants is given in Appendix 1.
3 INTERCOMPARISON SAMPLES

A stock solution of $^{67}$Ga was accurately sub-divided into a series of 4 ml aliquots in P6 vials. These samples were made available to users. Three aliquots from the same stock solution were also despatched to NPL for activity determinations.

At midday on the date of receipt of the samples, the total activity in each vial was approximately 40 MBq. The samples comprised $^{67}$Ga in 0.1 M HCl with a carrier concentration of 0.1 mg g$^{-1}$ Ga.

4 NPL MEASUREMENTS AND INTERNATIONAL EQUIVALENCE

The samples sent to NPL were assayed using a sealed, high-pressure, re-entrant ionisation chamber. This chamber had been previously calibrated for $^{67}$Ga using solutions which had been standardised absolutely using the primary standardisation facilities and techniques available at NPL.

As part of NPL’s remit to intercompare and maintain equivalence with other national standards laboratories, NPL has in the past submitted samples of $^{67}$Ga to the international reference system (SIR) maintained by the International Bureau of Weights and Measures (BIPM) in Paris. The activity value submitted to BIPM was determined using the same high pressure ionisation chamber used in this exercise. From those comparisons, the NPL value was within 0.1% of the mean value determined from the values from all participating national standards laboratories: this is well within the statistical uncertainties.

5 MEASUREMENT AND REPORTING PROTOCOL

Participants were invited to assay their P6 vial in each of their radionuclide calibrators and to report their results directly to NPL. Subsequently participants were informed of the traceable activity content of their sample based on the NPL standardisation. They were encouraged to derive as much useful information as possible from this exercise by making measurements using both pre-set facilities and manual settings, where available, and additionally by transferring activity to other containers (such as syringes) and reporting measurements on these. Estimates of measurement uncertainties were also sought. A standard reporting form, as reproduced in Appendix 2, was provided.

6 CONFIDENTIALITY

All results were reported directly to NPL. Each reported calibrator was given a Code Number by NPL and the results in this Report are tabulated against those Code Numbers. The correlation between Code Number and calibrator (and hence participant) remains confidential to NPL.
ANALYSIS OF RESULTS

Each result was decayed to a common reference time of 12.00 GMT on 27 March 1996, using a half-life of 3.261 days. In one case, the participant had made his own decay correction to the reference time. This had used a different half-life value but, from the data provided, it was possible to correct this using the NPL value.

It had been hoped that participants would estimate and report their uncertainties in the space provided on the reporting form: estimates could then be made of the significance or otherwise of differences between reported and NPL values. However, only a minority of participants provided such information and then sometimes incompletely or ambiguously.

From a knowledge of the total mass of solution dispensed to each vial (supplied by Al) and the activity concentration determined by NPL, the recorded activity values were compared with the correct values and the result recorded as a ratio.

In some cases, participants had subsequently transferred a weighed mass of solution to another container, such as a syringe. Both the solution in the new container and the residue in the P6 vial were assayed and activity values reported. The number of these additional measurements was small but some useful information was gleaned from them.

All the results are presented in Table 1. Summarised breakdowns by chamber type and container are given in Table 2. In these breakdowns, the results from syringes and residue vials have been separated from the original vial results. The results have also been displayed pictorially in histogram form in Figure 1.

A total of 118 independent results have been tabulated. Of this number, 11 relate to measurements made using syringes whilst 6 are for P6 vials containing less than the original 4 ml of solution (the "residue" containers).

DISCUSSION

8.1 ISOCAL II and PITMAN 238 SYSTEMS

The immediate objective was to resolve the concern surrounding the calibration of the ISOCAL II chamber. It should be noted that the ISOCAL II is the successor to the Pitman 238 chamber, being of similar design and response: hence, it is appropriate for these two chambers to be considered together. In the following discussion, references to ISOCAL II may be taken as applying to both chambers.

It is clear from the results that the calibration of the ISOCAL II is such that it will indicate activity values which, on average, are about 8 - 14% low, the single exception being that case where the response was 4% high. It should be noted that the ISOCAL II incorporates a small-volume chamber which is operated at atmospheric temperature and pressure. Correct operation depends on adjusting the electrometer gain to compensate for variations in the environmental variables. However, the data received does not necessarily suggest that is the cause of the problem. Because of the generally low readings, this suggests that the initial calibration was in error. Since this chamber is no longer available commercially, it is impossible to determine how the initial calibration was performed or, indeed, whether traceable sources were used.
8.2 OTHER SYSTEMS

If the ISOCAL II and Pitman 238 results are excluded, the overall picture is fairly good and, although there appears to be a slight bias towards underestimating the true activity, this is only of the order of 1 - 2%. There are two extreme outliers (Code Numbers 16 and 67 with ratio values of 0.80 and 0.58 respectively), both being for PRESET positions. It was reported by the participant that these systems had not been used for several years for the assay of $^{67}$Ga and as such their PRESET accuracies had not been checked. It is noteworthy that the DIAL SETTING option in both cases had produced acceptable results. This implies that a QA system may be being used which gives assurance for dial settings but does not for presets. Provided the operating protocol takes account of this, problems should not arise but it is perhaps a point that should be reviewed.

If all the P6 vial results for 4 ml of solution are considered, the mean ratio is 0.981 with a standard deviation of ± 0.054. Excluding the two outliers, the corresponding values are 0.986 and ± 0.030 respectively. Further, if the ISOCAL II/Pitman 238 results are removed from the calculation, the mean ratio becomes 0.990 with a standard deviation of ± 0.023. In each case, the mean reported activity is not statistically different from the NPL value.

8.3 SYRINGE DATA

Because of the thinner, lower-density container wall, it would be expected that material contained in a syringe would produce a higher response than that in a P6 glass vial. This is indeed the case: the results indicate that, if no allowance is made for this effect, activities would, on average, be overestimated by about 4%.

8.4 UNCERTAINTIES

The response to the request for estimates of uncertainty was somewhat disappointing. There is an increased emphasis nowadays on such matters and indeed there has been a significant advance in the quality and depth of advice in this area over the last few years\(^{(4,5,6)}\). However, only one third of the participants made such estimates. For those who did, almost every possible reporting scenario occurred as may be seen from the summary of individual responses in Table 3.

For the estimation of random uncertainty, participants invariably determined the standard deviation of a number of consecutive readings. It was not clear whether this had been performed with the $^{67}$Ga source supplied for this exercise or whether it was based on previous experiences with this or other nuclides. At the level of activity used in this exercise, it would be reasonable to expect relatively small, non-random uncertainties. If the source had decayed significantly before measurements were made, this might result in a larger uncertainty estimate. The value of 1.7%, however, is worryingly large and might suggest that the system has excessive background noise.

In respect of the non-random uncertainty estimates, their derivation was not always explained. Where it was, participants either had used the value given by the manufacturer in the calibration certificate or had adopted a value equal to the difference observed between the certificated activity of a traceable standard and the value indicated by the chamber when that source was assayed.
Only four estimates were made of overall uncertainty and there is a significant
difference between them. No other comment is made here but it may be that the user
community may wish to consider the importance, or otherwise, of these differences.

8.5 COMPARISON WITH PREVIOUS EXERCISES

The two similar exercises that have been conducted, both by NPL, in the UK took
place in 1980 and 1986. The first used $^{57}$Co and $^{129}$I as the intercomparison
radionuclides whilst the second used $^{99m}$Tc and $^{131}$I. It is not possible to make exact
comparisons between the results of those exercises and the present one because of the
differences in such aspects as radionuclides, container formats and distribution of
calibrators. Nevertheless, some fairly crude comparisons can be made by looking at
the distribution of all the reported results in each of the exercises: these are shown in
Table 4.

As the table shows, there has been a continued improvement in the number of results
falling within both $\pm 5\%$ and $\pm 10\%$ of the NPL value. Because of the differences
between the intercomparisons, there may be other reasons for this apparent
improvement but it is comforting to believe that this is, in part, due to improvements
in quality assurance that participants have introduced into their measurement
facilities.

8.6 ESTIMATE OF CONFIDENCE IN USER MEASUREMENTS

The overall uncertainty estimated by NPL on the activity concentration of the $^{67}$Ga
stock solution is $\pm 2.6\%$ at the 95% confidence level. The distribution of results shows
that 95% lie within $\pm 10\%$ of the NPL value. The difference between these two
uncertainty bounds may be attributable to a variety of effects such as background
levels, electronic noise, variations in containers and their respective calibration factors,
electronic drift in the current measuring system and variations between production
chambers. It is not possible to determine easily whether any or all of these effects may
be the cause of the results distribution. Indeed, it can be argued that these differences
are not critical. However, it is reasonable to state with confidence that 95% of all
assays of $^{67}$Ga will produce activity values which are within $\pm 10\%$ of the true value.

9 CONCLUSIONS

The exercise has clearly been beneficial in confirming the observations that were made
in 1995; that is that the ISOCAL II and Pitman 238 systems are likely to underestimate
significantly the activities of $^{67}$Ga solutions. Indeed, the evidence here suggests that
the potential underestimation may be greater than originally suspected. In the course
of the intercomparison and the analysis of results, several other aspects have been
noted and addressed in the discussion section above. In the opinion of the authors,
several conclusions may be drawn.

(a) Calibration factors for $^{67}$Ga for the ISOCAL II and Pitman 238 systems are
suspect and are likely to lead to significant underestimates of activity.

(b) Users of these systems should recalibrate using sources of $^{67}$Ga which are
directly traceable to national standards.

(c) Users of these systems should consider the advisability of checking the
calibration accuracy for other radionuclides.
(d) Calibration factors for syringes are significantly different than those for P6 vials and users should consider the implications of this in respect of the accuracy of activity measurements.

(e) The accuracy of both DIAL SETTINGS and PRESET facilities should be checked regularly and confirmed against national standards.

(f) Excluding the ISOCAL II AND Pitman 238 systems, the overall accuracy of measurements is good and indicates a continuing improvement in quality.

(g) The reporting and understanding of uncertainties is poor and the user community should consider the significance of this.

(h) The exercise has been beneficial and similar intercomparisons should be considered for other specific radionuclides.

10 ACKNOWLEDGEMENTS

The authors are indebted to their colleagues at AI, in particular Penny Bird, Western Case and Steven Judge, for supplying the $^{67}$Ga stock solutions and for dispensing and distributing the intercomparison samples.

Thanks are also due to all the participants for their cooperation and openness in reporting.
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(6) The Expression of Uncertainty and Confidence in Measurement for Calibrations.

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M J Woods.

(8) Intercomparison of $^{99m}$Tc and $^{131}$I by Radionuclide Calibrators in UK Hospitals, 1986.
M J Woods.
Table 1. Reported results

Code numbers were allocated to individual calibrators. Subsets (e.g. 2(a), 2(b), 2(c)) refer to measurements made with the same calibrator.

<table>
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<tr>
<th>Code</th>
<th>Calibrator Manufacturer</th>
<th>Model</th>
<th>Dial setting</th>
<th>Reported activity (MBq)</th>
<th>NPL activity (MBq)</th>
<th>Preset/NPL</th>
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Table 1. Reported results

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## Table 1. Reported results

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Letters (e.g. d) indicate individual participants
Numbered subsets (e.g. d1, d2) indicate different calibrators for the same participant
Table 4. Distributions of results from past and present intercomparisons (expressed as percentage of results within given range of NPL value)

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Figure 1  Distribution of results (4 ml solution only)

No of results

= all systems

= ISOCAL II/Pitman 238

Reported value/NPI value
## APPENDIX 1

### PARTICIPANTS

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<tr>
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</tr>
<tr>
<td>S Dave</td>
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</tr>
<tr>
<td>M Evans</td>
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## APPENDIX 2

### AMERSHAM INTERNATIONAL/NATIONAL PHYSICAL LABORATORY

**INTERCOMPARISON OF $^{67}$Ga ACTIVITY - MARCH 1996**

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</tbody>
</table>

<table>
<thead>
<tr>
<th>Vial identifier</th>
<th>Nominal volume: 4.0 ml</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chem. form:</td>
<td>$0.1 \text{ mg/g Ga in 0.1 M HCl}$</td>
</tr>
</tbody>
</table>

### DETAILS OF RADIONUCLIDE CALIBRATOR (If more than one calibrator is used, please photocopy this reporting form and use a separate form for each calibrator)

<table>
<thead>
<tr>
<th>Manufacturer</th>
<th>Model/Type</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Serial No</td>
<td></td>
</tr>
</tbody>
</table>

### Measurement in original container (P6 vial)

<table>
<thead>
<tr>
<th>Calibration factor/dial setting used</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time of measurement (GMT)</td>
</tr>
<tr>
<td>Measured activity (after bgd subtraction)</td>
</tr>
</tbody>
</table>

If you used a preset setting, it would help to repeat the reading on the manually adjustable setting (if available) using the figure recommended by the manufacturer. Please record below.

<table>
<thead>
<tr>
<th>Calibration factor/dial setting used</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time of measurement (GMT)</td>
</tr>
<tr>
<td>Measured activity (after bgd subtraction)</td>
</tr>
</tbody>
</table>

---

P.T.O.
Assays are often made in different containers (e.g. syringe). If you do this routinely, it would help to make a similar measurement here. In order to check the accuracy of such an assay, you need to know how much solution was transferred from the P6 vial to the new container. This could be determined by weighing the new container empty and then after the solution has been added. An additional check would be to re-measure the P6 vial after the transfer.

**Measurement in different container (please give details)**

- Mass/volume transferred ........................................... g or ml
- Calibration factor/dial setting used
- Time of measurement (GMT)  Month ...... Day ...... Hour ...... Min ......
- Measured activity (after bgd subtraction)  .......................... MBq

If you used a preset setting, it would help to repeat the reading on the manually adjustable setting (if available) using the figure recommended by the manufacturer. Please record below.

**Calibration factor/dial setting used**

- Time of measurement (GMT)  Month ...... Day ...... Hour ...... Min ......
- Measured activity (after background subtraction)  .......................... MBq

**Remeasurement of P6 vial after removal of solution**

- Calibration factor/dial setting used
- Time of measurement (GMT)  Month...... Day.......Hour.......Min......
- Measured activity (after background subtraction)  .......................... MBq

**Uncertainties**

- Please provide an estimate of the uncertainty (at the 1σ level) on your measurements
- Random (Type A) ± ..........,%  
- Non-random (Type B) ± ..........,%  
- Overall ± ..................%

It would be useful if you could indicate below how these values were estimated

Please return this form (together with any additional comments you would like to add) to:

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Centre for Ionising Radiation and Acoustics  
National Physical Laboratory  
Teddington  
Middlesex TW11 0LW  
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