NPL REPORT CIRM 4

REPORT

6th NPL Environmental Radioactivity Intercomparison Exercise, 1996

Report on Workshop, 16 April 1997

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ABSTRACT

The minutes of the workshop for the 6th NPL environmental radioactivity intercomparison held on 16 April 1997 in the Stiles Conference Room, Building 23, at the National Physical Laboratory, are reported. This workshop detailed the results of the sixth in a series of environmental radioactivity intercomparisons organised by the National Physical Laboratory. It also discussed problems highlighted by this and previous exercises, and considered possible formats for the next NPL intercomparison.

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Approved on behalf of Managing Director, NPL,
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1 INTRODUCTION

As part of NPL's continuing support to the National Measurement System Policy Unit of the UK Department of Trade and Industry, NPL has conducted the sixth in a series of environmental radioactivity intercomparisons. This intercomparison used three sample types: (i) α-emitting and pure β-emitting radionuclides in an aqueous matrix (dilute nitric acid), (ii) γ-emitting radionuclides in an aqueous matrix (dilute hydrochloric acid), and (iii) γ-emitters in milk.

This report contains the minutes of the workshop, held at NPL on 16 April 1997, to discuss the results of this intercomparison exercise, problems arising from this exercise and future intercomparisons.

2 LIST OF PARTICIPANTS

AEA Technology	Ian Adsley
Babcock Rosyth	Jonathan Kemp
BNFL (Westlakes)	Jim Desmond, Michael Froggatt
Central Veterinary Laboratory	Keith Lewis, Arthur Lally
Devonport Management Ltd	Graham Shepherd
Defence Radiological Protection Service	Terry Gingell
Glasgow Scientific Services	Keith McKay
Inst. of Terr. Ecology	Arthur Sanchez
IRAS Limited	Adrian Clacher
Lancaster University	Keith Bradshaw
Magnox Bradwell	Tristan Godfrey, J Wyatt
Magnox Gloucs	John Martin, Kevin Odell
Magnox Oldbury	Phil Clarke
North West Water	Elaine Timms, Karen Tuke
Stanger	Stephen Day
UKAEA Dounreay	A Ross
Urenco (Capenhurst)	Bob Sharrock, B Gillespie
Vickers Shipyard Engineering Limited	John Ennis, John Lavender
NPL	Neil Bowles, Wendy Brosnan, Matthew Carter, Julian Dean, Piers de Lavison, Simon Jerome, John Keightley, John Makepeace, Adrian Marks, Mike Woods and Simon Woods.
UKAS	Roger Stillman

3 SESSION 1

Chair: **John Makepeace, NPL**

3.1 WELCOMING ADDRESS.

The Chairman introduced the Director of NPL's Centre for Ionising Radiation and Acoustics, **Adrian Marks,** whose welcoming address and introduction outlined the importance of these gatherings of the radioactive measurement community in ensuring a uniformity of approach and maintaining "best practice" in the assay of

radionuclides in environmental samples. Mr Marks also pointed out that the National Measurement System Policy Unit (NMS PU) is beginning the process of formulating its next three year Ionising Radiation Metrology programme, due to commence in October 1998, and that this would require input from the user community and the participants of this workshop.

3.2 INTRODUCTION.

Mike Woods (NPL) gave an introduction in which he expressed a desire for the workshop to facilitate dialogue, discussion and criticism as well as to improve the measurement capabilities of the participants.

3.3 SOURCE PREPARATION FOR α AND β EMITTERS.

Simon Jerome (NPL) explained the source preparation procedures for the α and pure β emitting radionuclides as listed in Table 2 of the NPL Report CIRM 3[1] (superseding NPL Report CIRA(EXT)023[2]). This included details of the standardisations of the stock material as well as the dilution processes and checks performed. He pointed out that the ^{238}U, ^{238}Pu and ^{239}Pu were traceable to the French Primary Standards at the Laboratoire Primaire des Rayonnements Ionisants (LPRI). A check on the LPRI results was performed by α spectrometry. The presence of the impurities ^{235}U and ^{234}U, in the ^{238}U, was noted. The ^{3}H was traceable to the US Primary Standard at the National Institute of Standards and Technology (NIST).

3.4 RESULTS AND DISCUSSIONS FROM α AND β INTERCOMPARISONS.

Mike Woods (NPL) detailed the results for the α and β intercomparisons and the statistical tests carried out on the results. He reviewed the results from previous intercomparisons in this series and compared these against the current results. He pointed out that although most of the results were good, there appeared to be a problem with some participants quoting uncertainties less than those obtained by the national standards laboratories.

3.4.1 α intercomparisons.

The results for ^{238}Pu were about 5% low, compared with the NPL result, whilst those for $^{239}Pu/^{240}Pu$ gave good agreement. This was noted as a cause for some concern as similar techniques were used for both measurements. The results for ^{241}Am fell into two main categories, those measured using radiochemistry techniques (α-spectrometry) and those obtained by γ-spectrometry. Both of these techniques yielded results with a large spread about the NPL value. It was pointed out that in the next intercomparison exercise, NPL would check the correlation between these two methods in more detail, including a review of data from previous exercises, and publish the results. One participant measured ^{241}Am by mass spectroscopy, the result being about 40% high, but no reason for this anomaly was forthcoming.

In order to investigate the anomalies between the results for ^{238}Pu and $^{239}Pu/^{240}Pu$, it was suggested that NPL mix accurately weighed aliquots of each of the stock solutions, and determine the ratio between the activities of each by α-spectrometry. Unfortunately there was insufficient material remaining to do this: NPL will attempt to procure more samples from the same supplier batches, as used in this exercise, and determine this activity ratio.

3.4.2 β intercomparisons.

For the majority of ^{3}H results, the tritium was isolated by distillation, and all determinations of activity were made by Liquid Scintillation Counting. The results were encouraging as the number of discrepant results has fallen since the previous exercise, although it is felt that better performance is achievable. As only five results were reported for ^{35}S (two of which were discrepant), it was felt that more data is needed to give an overall picture of the measurement capability for this radionuclide. Several techniques were used in the assay of ^{90}Sr, and in all cases the principle involved allowing ^{90}Y to ingrow into purified ^{90}Sr and to separate and count the ^{90}Y. An improvement in measurement capabilities for this radionuclide was noted from previous exercises. Only 3 results were received for the assay of ^{99}Tc and as such it is impossible to form general conclusions, although the results received seemed encouraging.

Arthur Lally (CVL) pointed out that NPL does not measure the samples in the same manner as the participants, and as such, this exercise is not a true intercomparison. NPL dispenses a predetermined amount of activity, for each radionuclide, into each bottle, whereas the participants have to then assay these samples. He also suggested that an external body, capable of making accurate, traceable measurements on these samples could be asked to join in these intercomparisons as then the participants would be able to compare their measurement procedures and agreement with a laboratory that utilises similar techniques. He also suggested that NPL could send around one solution to each participant and then study the spread of results; this would not entail the standardisation of each of the radioisotopes.

Mike Woods (NPL) pointed out that using another laboratory in the manner suggested by Mr Lally would achieve little since that laboratory would be in no different position to any other participant. The NPL samples are produced from standards which are directly traceable to national standards of radioactivity and exclude any calibration bias inherent in the normal analytical procedures used by participants. The suggestion of distributing one sample to all participants, which had not been standardised, would only yield a distribution of the spread of results, and these would not be traceable to national standards or indicate accuracy.

3.5 SOURCE PREPARATION FOR γ-EMITTERS.

Julian Dean (NPL) detailed the source preparation procedures for the γ-emitting radioisotopes as listed in Table 2 of the NPL report CIRM 3[1]. This included the standardisations of the stock material as well as the dilution processes and checks performed. All of the stock solutions used were traceable to the UK Primary Standards held at NPL.

3.6 RESULTS AND DISCUSSIONS FROM γ-INTERCOMPARISONS.

Mike Woods (NPL) presented the results for the γ-intercomparisons. All of the participants used semiconductor detectors. Again, as in the case for the α and β emitters, there appeared to be a problem with some participants quoting uncertainties less than those obtained by the national standards laboratory, which often leads to the data being classed as discrepant via use of the u-statistic. It was encouraging that the use of nuclear decay data, as recommended by NPL[3], was very much in evidence. Most of the laboratories used efficiency calibration standards of the mixed radionuclide type, which would suggest that these laboratories should yield excellent

agreement with the NPL value for those radionuclides common to the mixed standards and this exercise. It was also noted that only about one in three laboratories made corrections for the problem of cascade summing (for ^{60}Co, ^{125}Sb, ^{134}Cs and ^{154}Eu).

The results for the aqueous and milk samples revealed a level of metrological performance which is comparable to previous exercises. It is of some concern that those nuclides which are used as calibration nuclides, (^{57}Co, ^{60}Co and ^{137}Cs), are still causing significant problems.

The ^{155}Eu results were of major interest, as both the aqueous and milk samples revealed a large low-bias. Possible reasons for this were discussed, ie: cascade summing and the fact that ^{155}Eu emits low energy γ-rays in the region of the calibration curve where the rate of change of detector efficiency with energy is large. It was decided that NPL would take another look at its results for this radionuclide as there may be a problem with the NPL quoted value.

IT WAS SUBSEQUENTLY DETERMINED THAT THE IMPURITIES (^{152}Eu AND ^{154}Eu) PRESENT IN THE ^{155}Eu STOCK SOLUTION WERE NOT PROPERLY ACCOUNTED FOR WHEN DETERMINING THE ^{155}Eu ACTIVITY BY ASSAY USING THE NPL SECONDARY STANDARD IONISATION CHAMBER. NPL REPORT CIRM 3[1] CONTAINS THE CORRECT NPL DATA AND SUPERSEDES NPL REPORT CIRA(EXT)023[2]. THE NPL DATA FOR ^{154}Eu WAS SIMILARLY AFFECTED BY IMPURITIES IN THE ^{154}Eu STOCK SOLUTION (^{152}Eu AND ^{155}Eu) AND THE NPL DATA FOR THIS RADIONUCLIDE HAS ALSO BEEN REVISED IN NPL REPORT CIRM 3[1], ALTHOUGH IN THIS CASE THE IONISATION CHAMBER RESPONSE WAS NOT SIGNIFICANTLY AFFECTED BY THE IMPURITIES.

4 SESSION 2

Chair: **Julian Dean, NPL**

4.1 UKAS UPDATE.

Roger Stillman (UKAS) gave an update on UKAS. In August 1995, NAMAS was privatised and merged with NACCB. The name NAMAS was not entirely lost as it now stands for National Accreditation of Measurement and Sampling. UKAS is effectively a monopoly organisation, which has no shareholders and 11 stakeholders (companies). It is operated in such a way to keep operating costs to a minimum. An important change is that accreditations now have associated expiry dates (4 years). UKAS has in total over 2000 accredited laboratories, but only 6 radiation measurement organisations are accredited as calibration laboratories and 11 are accredited as test laboratories.

Mr Stillman stressed the importance of proficiency testing schemes, both mandatory and voluntary. He also stressed the need to be able to demonstrate knowledge of how to determine uncertainties to an assessor.

4.2 UNCERTAINTY BUDGETS.

Mike Woods (NPL) referred to a series of guidance notes on uncertainty budgets [4-7], and gave an example for an uncertainty budget for a γ-spectrometry measurement. This example was well received by the participants, and specific requests were made for NPL to consider running uncertainty courses specifically focused on radioactivity measurements.

4.3 CURRENT NPL PROGRAMME.

John Makepeace (NPL) detailed the relevant current work at NPL. This included details of the ^{210}Pb, ^{210}Po, ^{237}Np and ^{233}Pa standardisations, decay data measurements and the production of standard γ-ray spectra. The results of the recent international tritium intercomparison were discussed. John pointed out that formulation of the new NMS three year Ionising Radiation Metrology programme is currently underway, and he urged participants to communicate to the NPL any of their measurement requirements.

4.4 NUCLEAR DATA UPDATE.

Simon Woods (NPL) addressed the participants with regards to the importance of using validated or recommended nuclear data for the accurate determination of radioactivity levels. Due to problems highlighted from previous intercomparison exercises with the measurement of ^{155}Eu by γ spectrometry, he presented the main γ-ray emission probabilities (P_γ) for ^{155}Eu from a variety of nuclear data databases, and compared those from the Evaluated Nuclear Structure Data File (ENSDF) [8] with those measured at NPL.

Dr Woods issued some warnings for users of γ-spectrometry analysis software. He warned against automatically believing that the software is performing correctly, ie: to visually check results and to manually adjust peak fitting parameters where required. He recommended the practice of updating nuclear data libraries with the latest evaluated or recommended nuclear data and urged participants to consult expert bodies (such as the NPL) if they had any doubts as to the reliability of their nuclear data.

5 SESSION 3

Chair: **Mike Woods, NPL**

5.1 DISCUSSION ON FUTURE INTERCOMPARISONS.

5.1.1 ^{238}Pu and $^{239}Pu/^{240}Pu$.

The problems surrounding the current differences in ^{238}Pu and $^{239}Pu/^{240}Pu$ activity levels were raised again. The agreed action is detailed in section 3.4.1.

5.1.2 ^{3}H.

There was some concern expressed about the accuracy of the ^{3}H half life. NPL will keep a watching brief on this. The effects of isotopic fractionation in the distillation of tritiated water were noted and participants were encouraged to submit any available experimental data on this subject to NPL who would in turn disseminate this to the user community.

5.1.3 ^{155}Eu.

It was agreed that ^{155}Eu would not be included in future intercomparisons until the persisting problems have been resolved. Refer to section 3.6 for more details.

5.1.4 Standard γ-ray spectra.

Subject to the satisfactory conclusion of the current NPL project examining commercial software packages for γ-spectrometry, it was agreed that the participants welcomed the proposal that sample spectra be distributed for analysis. It was further agreed that the radionuclide composition of the spectral data should be revealed to participants. In designing such an exercise, NPL should give consideration to such matters as the use of pre-existing library nuclear data versus NPL recommended nuclear data; the reporting of results both before and after fine-tuning of user systems; and the reporting of experience gained from this exercise.

5.1.5 Uncertainties.

NPL should consider the benefits of running focused uncertainty courses, and investigate the feasibility of including these in the next NMS three year programme.

5.1.6 Next intercomparison.

After lengthy discussion, the general consensus of the participants was that the next exercise should follow the same general pattern as previous exercises, but the γ-emitter solution should be offered at the current activity levels and also at a lower level. NPL agreed to consider the production of a dense matrix sample. A number of γ-emitters were proposed as potential constituents of the sample. The next intercomparison and associated workshop will be completed before the end of September 1998.

5.1.7 Summary of responses to delegate questionnaires.

Only seven participants returned questionnaires, the only significant criticism was that the seating arrangements could be better optimised to offer a more informal environment and to provide improved visibility of the overhead projections.

There was a general feeling that the opportunity to interact and discuss problems of mutual concern was perceived to be of great benefit to the participants.

Several participants voiced the opinion that there were benefits to be had from encouraging small working groups to be formed to discuss specific problem areas. There was also a general call for more advice and guidance on the estimation of uncertainties. NPL was encouraged to address this in the next NMS three year programme.

6 ACKNOWLEDGEMENTS

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