GROSS a/ b MEASUREMENTS IN DRINKING WATERS BY LIQUID SCINTILATION TECHNIQUE: VALIDATION AND INTERLABORATORY COMPARISON DATA

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Combining and Reporting Analytical Results. The Role of (metrological) Traceability and (Measurement) Uncertainty for Comparing Analytical Results

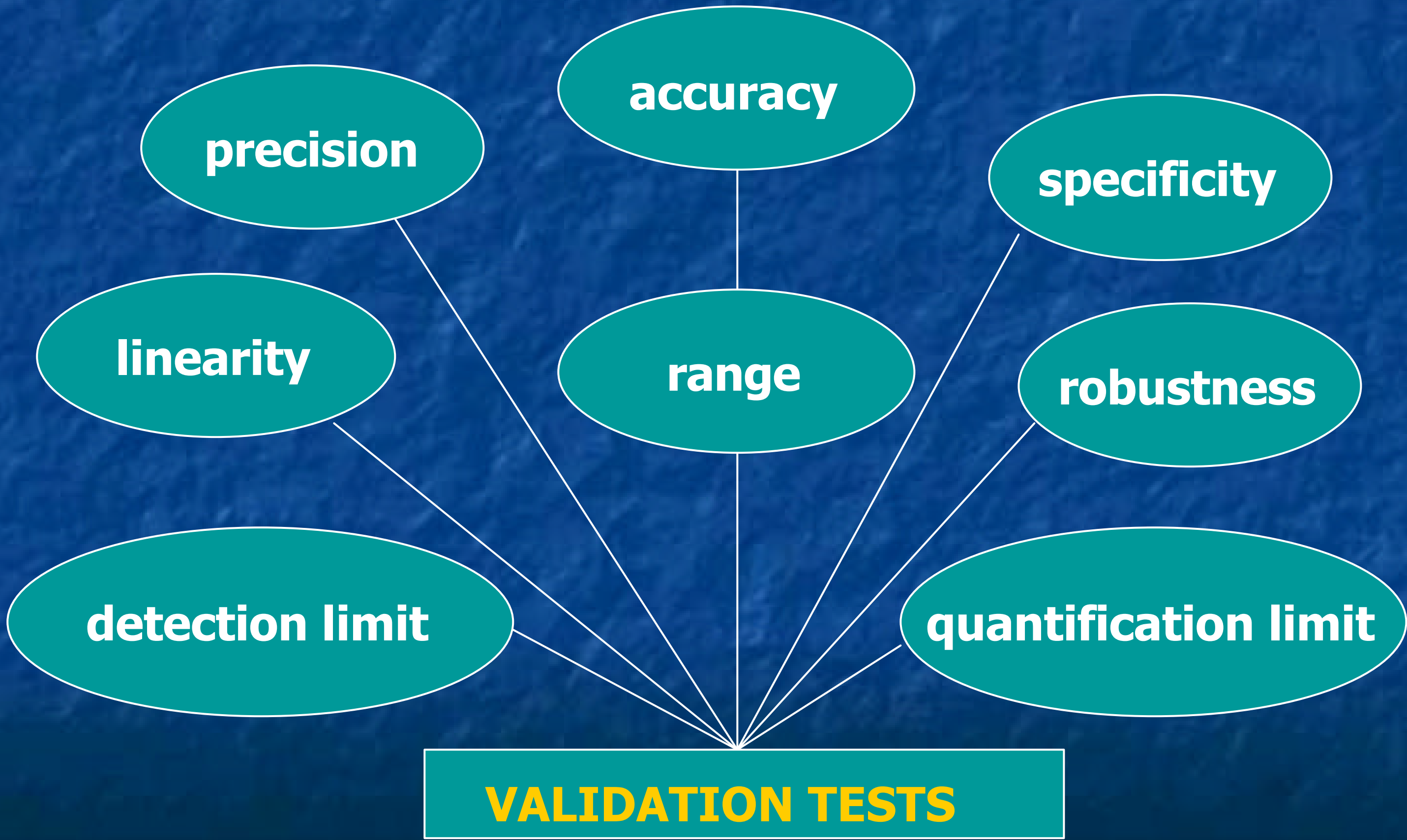
Quality assurance (QA) programme

- A quality assurance programme has been implemented at the *Department of Radiological Protection and Nuclear Safety* (DPRSN) of *Nuclear and Technological Institute* (ITN) in order to guarantee the quality of the results and for the further accreditation of the Environmental Radioactivity laboratories based on ISO 17025 standard.

- Some management and technical requirements have been carried out. New methodologies have also been developed and their validation performed to verify if those methods are acceptable for our intended purpose.

Validation of methods

- The validation requirements depends on the type of methodology and is an interactive process.



Accuracy of the method

- **The accuracy of the method can be assessed by analyzing a sample of known concentration and comparing the measured value to the true value, using reference materials (CRM);**

- **It can also be evaluated through the participation in interlaboratory exercises;**

- **It can be assessed by comparing test results from the new method with the results from an existing alternative method that is known to be accurate.**

Liquid Scintillation Counting (LSC) technique

Liquid Scintillation Counting (LSC) technique is one of the most practical methods for determination of gross alpha and gross beta activities, in water samples. **[1-5]**

Advantages:

- ✓ **Minimal sample preparation time;**
- ✓ **Small sample size;**
- ✓ **Reduced counting time;**
- ✓ **High detection efficiency ;**
- ✓ **No self-absorption problem.**

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[3] W. C. Burnett, J. Christoff, B. Stewart, T. Winters, P. Wilbur, *Radioact. and Radiochem.*, **1999**, *11*, 26.

[4] R. I. Kleinschmidt, *Appl. Rad. Isot.*, **2004**, *61*, 333.

[5] C. T. Wong, V. M. Soliman, S. K. Perera, *J.Rad. Nucl. Chem.*,**2005**, 264, 357.

Determination of Gross Alpha/ Beta

- Water samples were pre-concentrated by slow evaporation.

- One aliquot of 10 ml was withdrawn and added to 10 ml of cocktail.

- Cocktail: Ultima Gold LLT (Packard).

- Equipment: Tri-Carb 3170 TR/SL (Packard) with the ability to discriminate between alpha and beta particles by pulse decay analysis (PDA).

Optimization of the Pulse-Decay Discriminator (PDD)

▪ The optimization of PDD was made measuring separately an α emitter (^{241}Am; 24 Bq L^{-1}) and a β emitter (^{90}Sr/ 90Y; 23 Bq L^{-1}) standards, prepared at the same conditions of the samples.

The minimum interference occurred for a pulse-delay discrimination parameter set at 127.

▪ To study the influence of the quench on the optimum PDD, standards of ^{241}Am and ^{90}Sr with the same activity quenched were prepared with different amounts of CCl_4 (10 µl to 80 µl)).

Figure 1 shows the optimum PDD value was changed from 127 (0µl CCl_4) to 96 (80µl CCl_4).

The decreasing of the optimum PDD value with the increasing of the quenching is shown in Figure 2.

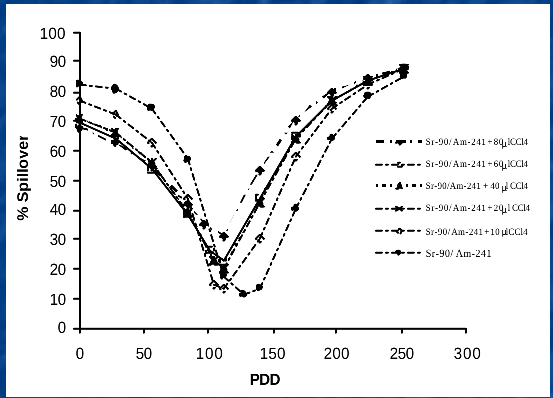

Figure 1 – *Total interferences versus PDD value*

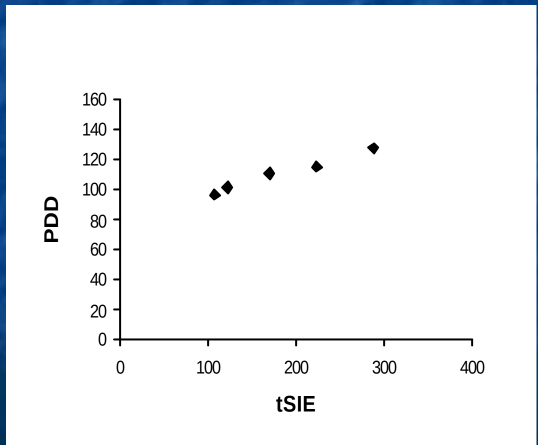

Figure 2 – Optimum *PDD value versus tSIE quenching parameter*

Counting Efficiency

- Alpha (^{241}Am) and beta (^{90}Sr/ ^{90}Y) standard sources were prepared with activities ranging from 1 to 25 Bq L^{-1}, using 10 ml of Ultima Gold LLT scintillation cocktail and 10 ml of 0.1M HCl distilled water, in order to achieve the same chemical and physical conditions as the environmental samples.

The mean value of the counting efficiency for ^{241}Am and ^{90}Sr/ ^{90}Y sources was 95.8% and 90.5%, respectively, using the PDD value set at 127.

- The influence of the quenching level on the counting efficiency was also studied. The alpha (^{241}Am; 24 Bq L^{-1}) and beta (^{90}Sr/ ^{90}Y; 23 Bq L^{-1}) standards, with different CCl_4 amounts, were prepared.

The volume of CCl_4 used (from 10 µl to a maximum of 90 µl) produced tSIE range values between 300 and 100. The alpha particle efficiency remains higher at lower quench and drop off towards lower values as the quench increases. The effect of quenching on the beta particle efficiency is less pronounced.

Validation Test

✓ Validation test based on the recovery of spiked samples was performed using the same procedure as in the sample analyses.

Tap water samples spiked with ^{241}Am and ^{90}Sr were prepared (with activities ranging from 1 to 25 Bq L^{-1}) using Ultima Gold LLT scintillation cocktail.

✓ The validation test results show a good agreement between the spiked samples recovery and the nominal activities.

Table 1 - *Alpha and beta activities (Bq L^{-1}) for water samples spiked with ^{241}Am and ^{90}Sr*

Alpha Activity (^{241}Am)	
Known[(a)]	**Measured[(b)]**
48.3	46.7 ± 2.4
24.1	23.9 ± 0.8
12.1	12.4 ± 0.5
5.8	4.9 ± 0.2
0.097	0.11 ± 0.04
Beta Activity (^{90}Sr)	
Known[(a)]	**Measured[(b)]**
22.5	20.2 ± 1.7
11.3	10.0 ± 2.2
5.5	5.0 ± 0.3
1.8	1.7 ± 0.1

(a) Uncertainty of the nominal value of activity is 0.47 % and 0.88% for ^{241}Am and ^{90}Sr, respectively.
(b) Average of the measurement of three replicate samples with the same nominal activity.

Intercomparison Data

➢The DPRSN, participated in an international analytical intercomparison exercise, for environmental radioactivity laboratories, organized by the CSN, Spain.

A drinking water sample (1000 ml) was pre-concentrated (80 ml) by slow evaporation on a hot plate. Five aliquots of 10 ml of this aqueous concentrate was withdrawn and added to five vials (glass vial, Packard) with 10 ml of Ultima Gold AB (Packard).

➢The intercomparison trial indicated that the LSC method provides acceptable result (Z = 2.21) for alpha activity and more accurate result for beta activity (Z= 1.62).

Table 2 – *CSN/ CIEMAT intercomparison results*

	Gross Alpha Activity ($Bq\ L^{-1}$)	**Gross Beta Activity ($Bq\ L^{-1}$)**
DPRSN/ ITN results by LSC[(a)]	**0.135 ± 0.017**	**1.698 ± 0.055**
CSN/ CIEMAT reference values	**0.091 ± 0.020**	**1.333 ± 0.227**

(a) Mean value

CONCLUSIONS

- The use of a low background liquid scintillation counting system with pulse shape analysis enables the simultaneous determination of alpha and beta activities in water samples. A suitable quenching correction should be performed to obtain the optimum PDD value and a good a/ β separation.

- The validation test results show a good agreement between the spiked samples recovery and the nominal activities. Acceptable results were obtained on the intercomparison exercise using the cocktail Ultima Gold AB.

- More accurate data will be expected in further intercomparison exercises using the optimization performed with cocktail Ultima Gold LLT.

- Further studies will be necessary in order to examine thoroughly the accuracy of this methodology and to test others validation requirements.