A Comparison of Techniques Used to Evaluate Low Level Radiochemical Data

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Abstract

Radiochemistry laboratories provide analytical data in the form of results, uncertainties and detection limits. When evaluating radiochemistry results, the user will eventually be confronted with the need to make a decision regarding the presence or absence of radioactivity. The method used to make this assessment must be carefully selected or the results can be misleading. This is especially true for low level determinations where the activity is difficult to distinguish from the background. Comparisons of three approaches used to assess radiochemical data are discussed to assist the data user.

Introduction

All analytical methods and systems have a certain level of 'noise' associated with them. This noise is due to random variations in the analytical and detection components of a system. In its simplest form a positive detection is a measurement "signal" distinguishable from the background "noise". When analyzing at low concentrations there is a point where the method's results can not be distinguished from the "noise" level of the analytical system. The terms used to describe the detection threshold are numerous and include minimum detectable activity, decision level, critical level, lower limit of detection, quantification limit, etc.

Because radiological measurements are random in nature, statistical evaluations are effective in calculating the sensitivity of a measurement. Radiation detectors demonstrate statistical fluctuations in that each time a background measurement is made, a different result is typically observed. Furthermore, the observed backgrounds can be assigned a mean and a variance. The statistical calculations improve as more background counts are measured. Detectors with higher background count rates actually follow the statistical models better than detectors with very low count rates. Therefore, counting backgrounds longer improves the performance of the statistical model. Most calculations used to determine a limit of detection are based on Poisson statistics and a 95% confidence interval marking the level where the signal is distinguishable from the noise. The above assumptions have held through radiological measurements since the 1960's with many lively debates in the ensuing years.

The lowest threshold used to distinguish a positive result is the decision level threshold (DLC) or sometimes called the decision level or critical level. The DLC is represented by Equation (1).

$$DLC = \frac{1.645 * \sqrt{B(ts)(1 + \frac{ts}{tb})}}{K * ts} \qquad \text{Eq. (1)}$$

where: 1.645 is a statistical value representing the 95% confidence interval

B is the gross background count rate (cpm)

ts is the sample count time (minutes)

tb is the blank count time (minutes)

K is a constant to convert counts to activity units

The DLC is an *a posteriori* (after the fact) measurement that can be compared with the result in order to make a judgment as to the presence or absence of activity in the sample. At the DLC one can say that there is a 95% confidence that the measured result is above the background. Stated differently, if twenty replicate measurements were made on a blank sample, one result would be expected above the DLC (also known as a false positive).

A second measure of sensitivity is the minimum detectable activity (MDA) Eq. (2). As the level of activity in a sample increases above the background (i.e. the sample is no longer a blank) a new distribution forms and at a certain level above the critical level the MDA is established. Although the MDA sounds like a level below which no activity will be detected, this is actually not the case. A laboratory can frequently detect activity below the MDA (approximately 50% of the time). This is a primary reason that it is not advisable to exclude data by reporting <MDA.

$$MDA = \frac{3 + 3.29 * \sqrt{B(ts)(1 + \frac{ts}{tb})}}{K * ts} \qquad \text{Eq. (2)}$$

Variables are defined are the same as referenced in Eq. (1)

The MDA is roughly equivalent to the term lower limit of detection (LLD) although various published calculations differ from Eq. (2). For example, some equations use 2.71 as opposed to 3 and others use 4.66 rather than 3.29. Most of these variations come from attempts to better represent the 95% confidence interval for low level counting or from simplifying the equations to assume identical background and counting times. From the equations above it is evident that counting a sample longer will improve the sensitivity of the measurement (Strom, Stansbury HPS 1992).

The method detection limit (MDL) is another term that should not be confused with DLC, MDA or LLD. The MDL is used for non-radiological measurements (metals, organics, general chemistry etc.) and is based on a 99% confidence interval. (Title 40 CFR Part 136, Appendix B). The procedure for determining the 99% confidence level for MDL involves running multiple standards near the detection limit and then applying a statistical calculation to determine the MDL.

Although some rigorously debate its merit, a common practice for radiological laboratories is to determine a sample specific MDA, also known as an *a posteriori* (after the fact) MDA. This calculation is useful in assessing the sensitivity achieved in the determination of a specific sample activity. The *a posteriori* MDA will account for the actual detector efficiency and volume used in the measurement while the *a priori* MDA must make assumptions that cannot be known until after a measurement is made. In reviewing the MDA, a data user can assess if the laboratory met the sensitivity requirements of a contractual or regulatory level. At the reported MDA the data user can also conclude that there is a 95% probability that the activity detected is above the DLC.

Evaluation of uncertainties has gained popularity as a means of determining the significance of a measurement. In this approach the data user compares the result to some multiple of the uncertainty in order to establish a threshold for detection. For example, comparing a result to 2 times the total propagated uncertainty (TPU) (MacLellan) has been proposed as an improved means of determining the critical level. The TPU is a calculation that approximates all sources of uncertainty in the analytical process.

Summary

The table below shows how low-level detection decisions are different for the three cases discussed. The DLC is clearly the most conservative approach, followed by the TPU and finally the MDA comparison. General Engineering Laboratories, LLC (GEL) provides data in formats that can be customized to meet the specific requirements of a given project. We select counting times and sample aliquots to match the sensitivity required. It is very important to communicate in advance any special sensitivities or calculations that are required for a project. GEL has remained flexible in reporting radiological data due to the need to service programs that are effectively “etched in stone”. For these reasons we resist the urge to suggest one particular approach, since the approach ultimately depends on what the data user is trying to accomplish.

Experimental

The table below provides a comparison of results calculated by DLC, MDA and 2x TPU. The results are based on a 10g sample, a 96% yield for plutonium analysis, 1000-minute background and sample count time and a 27% counting efficiency.

Isotope	Activity	TPU	DLC	MDA	Net Area	BKG Area	Activity > DLC	Activity > 2*TPU	Activity > MDA
Pu- 239	0.00	0.00	0.00	0.53	0	0			
	0.35	0.25	0.00	0.53	2	0	√		
	0.53	0.31	0.00	0.53	3	0	√		
	0.70	0.35	0.00	0.53	4	0	√		√
	0.88	0.39	0.00	0.53	5	0	√	√	√
	1.76	0.56	0.00	0.53	10	0	√	√	√
	2.64	0.68	0.00	0.53	15	0	√	√	√
Pu- 239	0.00	0.35	0.58	1.69	0	2			
	0.35	0.43	0.58	1.69	2	2			
	0.70	0.50	0.58	1.69	4	2	√		
	0.88	0.53	0.58	1.69	5	2	√		
	1.06	0.56	0.58	1.69	6	2	√		
	1.23	0.58	0.58	1.69	7	2	√	√	
	1.76	0.66	0.58	1.69	10	2	√	√	√
	2.64	0.77	0.58	1.69	15	2	√	√	√
Pu- 239	0.00	0.50	0.82	2.17	0	4			
	0.35	0.56	0.82	2.17	2	4			
	0.70	0.61	0.82	2.17	4	4			
	0.88	0.63	0.82	2.17	5	4	√		
	1.58	0.73	0.82	2.17	9	4	√	√	
	1.76	0.75	0.82	2.17	10	4	√	√	
	2.29	0.81	0.82	2.17	13	4	√	√	√
	2.64	0.84	0.82	2.17	15	4	√	√	√
Pu- 239	0.00	0.61	1.01	2.54	0	6			
	0.88	0.73	1.01	2.54	5	6			
	1.06	0.75	1.01	2.54	6	6	√		
	1.23	0.77	1.01	2.54	7	6	√		
	1.41	0.79	1.01	2.54	8	6	√		
	1.58	0.81	1.01	2.54	9	6	√		
	1.76	0.83	1.01	2.54	10	6	√	√	
	2.64	0.92	1.01	2.54	15	6	√	√	√
Pu- 239	0.00	0.79	1.30	3.12	0	10			
	0.18	0.81	1.30	3.12	1	10			
	0.88	0.88	1.30	3.12	5	10			
	1.41	0.93	1.30	3.12	8	10	√		
	1.76	0.96	1.30	3.12	10	10	√		
	2.11	1.00	1.30	3.12	12	10	√	√	
	2.64	1.04	1.30	3.12	15	10	√	√	
	3.52	1.11	1.30	3.12	20	10	√	√	√

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