Application of MARSSIM and MARLAP Concepts to the Interpretation of Sample Data for Determining the Presence of Radioactivity

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ABSTRACT

The U.S. Army Corps of Engineers (USACE) conducts cleanups of radioactive wastes at radiologically contaminated sites around the country that are part of the Formerly Utilized Sites Remedial Action Program (FUSRAP). At these sites environmental media are surveyed or sampled, and the results are used to guide decision-making, e.g., to help define the extent of radiological contamination and to determine if remediated areas may be released. USACE routinely implements the guidance of the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) and the Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP) in its remediation work. Contractors and analytical laboratories frequently ignore the recommendations of MARSSIM and MARLAP for the interpretation of analytical data to determine the presence or absence of radioactive material. Many times radiological data are compared to a "detection level" (L_D or S_D) or their activity equivalent, the "minimum detectable activity" (MDA). However, current guidance uses a different criteria for comparison. MARSSIM and MARLAP suggest comparing radiological results to a "critical level" (L_C or S_C) and not to an L_D, S_D or MDA. In MARSSIM and MARLAP, a positive result for radioactivity is one exceeding the "critical level." If radiological data are compared to the L_D or MDA a "U" or "non-detect" qualifier may be placed on the data when it is possible that the "critical level" has been exceeded. In addition, decisions using the MDA may not meet the project's data quality objectives (DQOs). It may be important to re-evaluate the interpretation of radiological data so that consistent criteria are applied to remediation decision-making.

INTRODUCTION

An important decision following environmental survey or sample analysis is the determination of the presence of radioactivity. Many times this decision has used the detection level (L_D or S_D) or the MDA. For example, analytical labs routinely report and use MDAs for radioactive samples, and they assign data qualifiers based on that value. MARSSIM and MARLAP guidance suggests comparing radiological results to a "critical level" [L_C (MARSSIM) or S_C (MARLAP)] and not to an L_D, S_D or MDA. The question is, "Does the sample or surveyed material have a positive amount of the radioactive analyte?"

DEFINITIONS

Critical level - The critical level (L_C, MARSSIM) is the level, in counts, at which there is a statistical probability (with a predetermined confidence) of incorrectly identifying a measurement system background value as "greater than background." Any response above this level is considered to be greater than background. The critical level is also called the critical value or decision level (S_C, MARLAP). (See Figures 1 and 2.)

Detection limit - The detection limit (L_D, MARSSIM) is the net response level, in counts, that can be expected to be seen with a detector with a fixed level of certainty. The detection limit is an *a priori* estimate of the detection capability of a measurement system. The detection limit is also called the minimum detectable value (S_D, MARLAP). (See Figures 1 and 2.)

Minimum detectable activity - The minimum detectable activity (MDA) is the detection limit (counts) multiplied by an appropriate conversion factor. It is reported in activity units, e.g. pCi. The minimum detectable activity is also called the "minimum detectable amount" (MARLAP).

Minimum detectable concentration - The minimum detectable concentration (MDC) is the detection limit (counts) multiplied by an appropriate conversion factor. It is reported in concentration units, e.g. pCi/g. It is the minimum concentration of an analyte that must be present to give a specified power, 1 – β, where β is the probability of a Type II error.

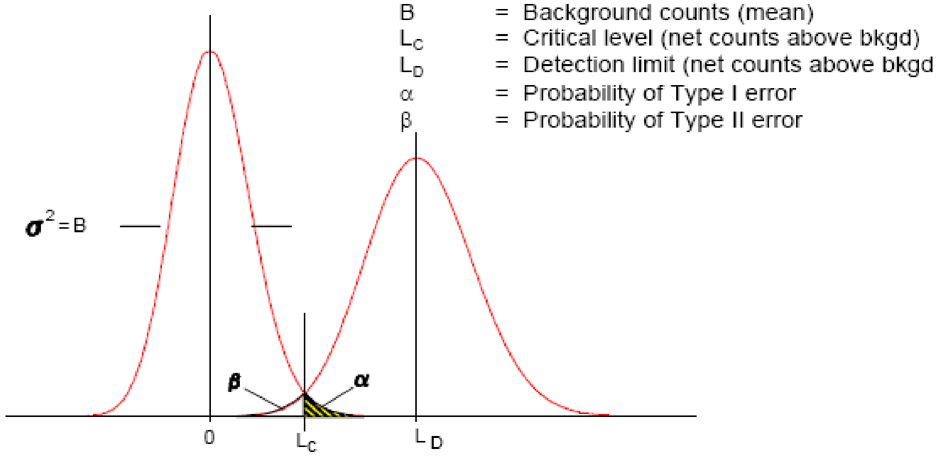

Fig. 1. Graphically represented probabilities for Type I and Type II errors in detection sensitivity for instrumentation with a background response (from MARSSIM Figure 6.2)
Note the Type I error (α), called a "false positive," is associated with the L_C.

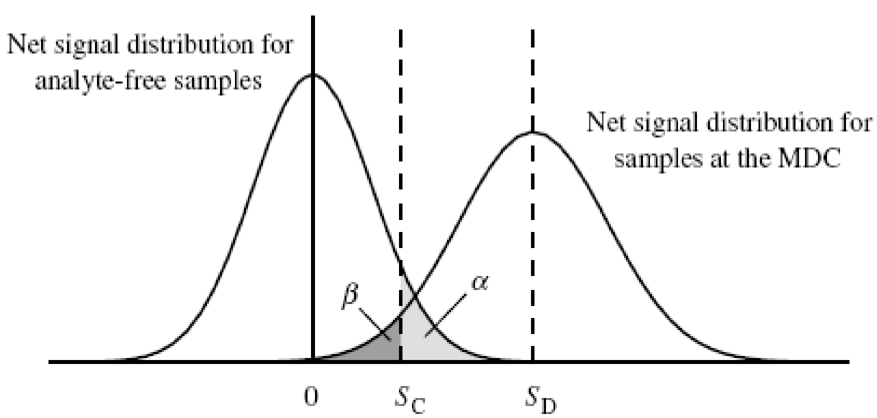

Fig. 2. The critical net signal, S_C, and minimum detectable net signal, S_D (from MARLAP Figure 20.1)

DISCUSSION

An important decision in radioactivity measurement is whether the measured value is sufficiently different from background levels to be considered a "detect." NBS Handbook 80 (NBS 1961), defined the minimum detectable activity as "…that amount of activity which, in the same counting time, gives a count which is different from the background count by three times the standard deviation of the background count." Subsequently a different formula, the Currie formula (Currie 1968) has been widely used to make detection decisions, i.e., $L_D = 2.71 + 4.65 \sqrt{B}$ and $MDC = k\,(2.71 + 4.65 \sqrt{B})$. Discussions about the application and accuracy of using the Currie formula have been taking place for over a decade. These discussions have resulted in revision of the Currie formula, and for positive detection decisions, replacement of its use with the critical level. ANSI N13.30 (ANSI 1996), a document on radioabioassay, discusses detection issues and recommends use of the critical level for radioabioassay samples.

Analytical laboratories and many decision makers currently use the Currie formula for their detection decisions and for assignment of qualifiers to the data. Many manufacturers of radiation detection equipment also retain and use the Currie formula in their data analysis software. The critical level is commonly ignored. Guidance quoted from MARSSIM and MARLAP supporting use of the critical level is presented below.

MARSSIM

Concepts for the use of the critical level for field measurements and instrumentation are primarily contained in MARSSIM Chapter 6, Section 6.7.1. Information and recommendations quoted from MARSSIM, Section 6.7.1. are as follows:

> The MDC is the *a priori* net activity level above the critical level that an instrument can be expected to detect 95% of the time. This value should be used when stating the detection capability of an instrument. The MDC is the detection limit, L_D, multiplied by an appropriate conversion factor to give units of activity. Again, this value is used before any measurements are made and is used to estimate the level of activity that can be detected using a given protocol.
>
> The critical level, L_C, is the lower bound on the 95% detection interval defined for L_D and is the level at which there is a 5% chance of calling a background value "greater than background." This value should be used when actually counting samples or making direct radiation measurements. Any response above this level should be considered as above background (i.e., a net positive result). This will ensure 95% detection capability for L_D.
>
> If the background of the detection system is not well known the equations are: (Currie assumed "paired blanks" (Currie 1968), which is interpreted to mean that the sample and background count times are the same).
>
> - $L_C = k \sqrt{2B}$
> - $L_D = k^2 + 2k \sqrt{2B}$
>
> Where:
> L_C = critical level (counts),
> L_D = detection limit (counts),
> k = Poisson probability sum for α and β (assuming α and β are equal)
> B = number of background counts that are expected to occur while performing an actual measurement

If values of 0.05 for both α and β are selected as acceptable, then k = 1.645 (from MARSSIM, Appendix I, Table I.1) and

- $L_C = 2.33 \sqrt{B}$
- $L_D = 3 + 4.65 \sqrt{B}$
- and the MDC $= C \times (3 + 4.65 \sqrt{B})$

The following example illustrates the calculation of an MDC in Bq/m^2 for an instrument with a 15 cm^2 probe area when the measurement and background counting times are each one minute:

B = 40 counts
C = (5 dpm/count)(Bq/60 dpm)(1/15 cm^2 probe area)(10,000 cm^2/m^2) = 55.6 Bq/m^2-counts
MDC $= 55.6 \times (3 + 4.65 \sqrt{40}) = 1{,}800$ Bq/m^2 (1,100 dpm/100 cm^2)
$L_C = 2.33 \sqrt{40} = 15$ counts

If a person asked what level of contamination could be detected 95% of the time using this method, the answer would be 1,800 Bq/m^2 (1,100 dpm/100 cm^2). When actually performing measurements using this method, any count yielding greater than 55 total counts, or greater than 15 net counts (55-40=15) during a period of one minute, would be regarded as greater than background.

The following MARSSIM table presents L_C, L_D, and MDC sensitivities for several survey instruments.

Detector	Probe area (cm^2)	Background (cpm)	Efficiency (cpm/dpm)	Approximate Sensitivity L_C (counts)	L_D (counts)	MDC (Bq/m^2)[a]
Alpha proportional	50	1	0.15	2	7	150
Alpha proportional	100	1	0.15	2	7	83
Alpha proportional	600	5	0.15	5	13	25
Alpha scintillation	50	1	0.15	2	7	150
Beta proportional	100	300	0.20	40	83	700
Beta proportional	600	1500	0.20	90	183	250
Beta GM pancake	15	40	0.20	15	32	1800

Fig. 3. Examples of estimated detection sensitivities for alpha and beta survey instrumentation (from MARSSIM Section 6.7.1).

MARLAP

Concepts for the use of the critical level for analytical sample analysis are mainly presented in MARLAP Attachment 3B, Section 8.3.3, Glossary, Chapter 20, and Attachment 20A. Information and recommendations quoted from MARLAP are as follows:

> In the context of analyte detection the null hypothesis states that there is no analyte in the sample; so one presumes that no analyte is present unless there is sufficient analytical evidence to the contrary.
>
> (MARLAP Sections 3B.1)

> To determine how large the instrument signal for a sample must be to provide strong evidence for the presence of the analyte, one calculates a threshold value for the net signal, called the *critical value*, which is sometimes denoted by S_C. If the observed net signal for a sample exceeds the critical value, the analyte is considered detected; otherwise, it is not detected.
>
> (MARLAP Section 3B.2)

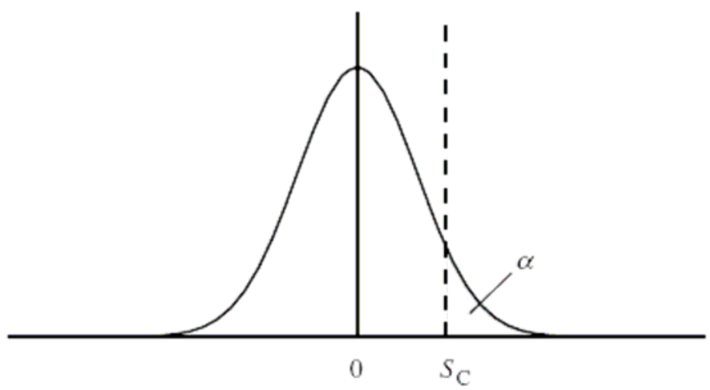

Fig. 4. The critical value of the net signal (MARLAP Figure 3B.1)

> The probability of a Type I error is often denoted by α. Before calculating the critical value one must choose a value for α. The most commonly used value is 0.05, or 5 percent.
>
> (MARLAP Section 3B.2)

> The MDC concept has generated controversy among radiochemists for years and has frequently been misinterpreted and misapplied.
>
> (MARLAP Section 20.2.4)

> MARLAP recommends that when a detection decision is required, the decision should be made by comparing the measured value (e.g., of the net instrument signal) to its critical value - not to the minimum detectable value.
>
> (MARLAP Section 3B.3)

> …the common practice of comparing a measured concentration to the MDC to make a detection decision is incorrect.
>
> (MARLAP Section 20.2.4)

> In radiochemistry, the minimum detectable value may be called the *minimum detectable concentration* (MDC), *minimum detectable amount* (MDA), or *minimum detectable activity* (also

abbreviated as MDA). MARLAP generally uses the term "minimum detectable concentration," or MDC. Unfortunately, it is also common to use the MDC incorrectly as a critical value, which it is not. It is difficult to imagine a scenario in which any useful purpose is served by comparing a measured result to the MDC. Nevertheless such comparisons are used frequently by many laboratories and data validators to make analyte detection decisions, often at the specific request of project planners.

(MARLAP Section 3B.3)

This common but incorrect practice of comparing the measured result to the MDC to make a detection decision produces the undesirable effect of making detection much harder than it should be, because the MDC is typically at least twice as large as the concentration that corresponds to the critical value of the instrument signal.

(MARLAP Section 3B.3)

…when the MDC is used for the detection decision, the probability of detection is only about 50 percent, because the measured concentration is as likely to be below the MDC as above it. When an analyte-free sample is analyzed, the probability of a Type I error is expected to be low (usually 5 percent), but when the MDC is used for the detection decision, the probability of a Type I error is actually much smaller - perhaps 0.1 percent or less.

(MARLAP Section 3B.3)

The lab qualifier "U" is defined in MARLAP as "a normal, not detected (< critical value) result."

(MARLAP Section 8.3.3)

Critical value (S_C) (3B.2): In the context of analyte detection, the minimum measured value (e.g., of the instrument signal or the analyte concentration) required to give confidence that a positive (nonzero) amount of analyte is present in the material analyzed. The critical value is sometimes called the critical level or decision level.

(MARLAP Glossary)

Many experts strongly discourage the reporting of a sample-specific MDC because of its limited usefulness and the likelihood of its misuse.

(MARLAP Section 20.2.4)

The minimum detectable value (MDC or MDA) should be used only as a performance characteristic of the measurement process.

(MARLAP Section 20.3)

A measurement result should never be compared to the minimum detectable value to make a detection decision.

(MARLAP Section 20.3)

No single equation is appropriate for the critical level for all circumstances. Several equations are therefore, presented in the MARLAP guidance. MARLAP Section 20.4 presents equations for "simpler methods," including normally distributed signals, and for the Poisson-Normal Approximation. Equations for "low-background detection issues" that are more generally applicable even when background is high are presented in Attachment 20A. Many useful examples are also included. Attachment 20A.2.2 presents very important recommendations for choosing an equation. The reader of MARLAP could benefit by reviewing the recommendations in Attachment 20A.2.2 before reading about the preceding equations.

CONCLUSIONS

MARSSIM and MARLAP recommend the use of the critical level to define detected radioactivity from survey and sample results. The significance is that even small positive detections can affect project decisions, such as the designation of MARSSIM non-impacted areas, the presence of a nuclide in media, such as Pu-239 in groundwater or transuranic nuclides in waste profiles, or in the assignment of data validation qualifiers. It may be necessary to require the analytical lab and/or the contractor to implement the MARSSIM and MARLAP guidance regarding critical level in advance of the analysis so that the recommended guidance can most easily be applied to raw data at the time of analysis. In this regard, the scope of work for contractor and lab should include requirements to use MARLAP and MARSSIM guidance for the detection of radioactivity.

REFERENCES

1. American National Standards Institute. 1996. *Performance Criteria for Radiobioassay*, HPS N13.30-1996, New York, May.

2. Currie, L. A. 1968. *Limits for Qualitative Detection and Quantitative Determination*. Anal. Chem. 40(3):586-593.

3. Currie, L. A. 1984. *Lower Limit of Detection: Definition and Elaboration of a Proposed Position for Radiological Effluent and Environmental Measurements.* NUREG/CR-4007, Washington, D.C.

4. DOD, DOE, EPA, and NRC. 2000. *Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM)*, Revision 1, NUREG-1575, DOE/EH-0624, EPA 402-R-97-016, Washington, D.C., August.

5. DOD, DOE, EPA, DHS, FDA, USGS, NIST, and NRC. 2004. *Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP)*, Revision 0, NUREG-1576, NTIS PB2004-105421, EPA 402-B-04-001A, Washington, D.C., July.

6. U.S. Department of Commerce, National Bureau of Standards. 1961. *A Manual of Radioactivity Procedures*, NBS Handbook 80, Washington, D.C., November.