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How the choice of data reduction can strongly influence uncertainty assessment: A re-analysis of Mn-bath experiments

S. Croft*, A. Favalli

Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545, USA

HIGHLIGHTS

- Carefully assessing and reporting uncertainties in experimental work is vital.
- The uncertainty or bias introduced by the procedure adopted matters.
- Different analysis on the same data can give dramatically different uncertainty.
- We show a factor ten reduction in the standard deviation.

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ABSTRACT

The Mn-bath technique is widely used, especially by standardization laboratories, for the absolute determination of neutron emission rates. Understanding the limitations of the technique, and in particular the total measurement uncertainty, is crucial if quality results, fit for purpose, are to be reported. In this work, we show that the way in which the acquired data is analyzed can strongly influence the uncertainty assessment. We take a carefully performed set of Mn-bath measurements from the literature as our example and show that the same data when reanalyzed can be used to justify an uncertainty smaller by about an order of magnitude than was originally reported. This finding should caution all those involved in radiation measurements to critically assess their approach to data analysis and to perform a careful uncertainty analysis taking into account possible alternatives.

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1. Introduction

Hwang and Lee (H&L) (Hwang and Lee, 1988) describe an implementation of the Mn-bath technique and a series of experiments to determine the neutron emission rate of a ^{252}Cf source. These authors went to great pains to obtain a result which has a large apparent uncertainty. The emission rate obtained at the reference date has a reported relative standard deviation of approximately 13%. This is strikingly large compared to the claimed performance of the Mn-bath and related methods for absolute neutron emission rate determination that has been quoted in other experiments, which are currently in the range 0.3–1.3% taken across state of the art international facilities (Roberts et al., 2011). Although one might anticipate some advances to have taken place over the intervening period since H&L's work the improvement is modest in comparison. The uncertainty analysis provided by H&L is not very detailed, but most of their intermediate results are provided in the form of tables which allows for a review of their analysis in some detail. The purpose of reanalyzing this data is to demonstrate how different conclusions may be drawn depending on the method chosen to reduce the data. While the reanalysis of this data will not be wholly definitive due to the fact that not all of the information one would like to have is available, the reanalysis of the data does illustrate the salient aspects. There are small numerical inconsistencies between the various tables of results given by H&L but these are unimportant compared to the large canvas picture we shall paint.

That the uncertainties for both the neutron emission rate of their source and the hydrogen to manganese cross-section ratio, are unfavorably compared to what others were doing at the time and even many years previous, is not our focus. We only wish to use the data as a concrete example of critical thinking and analysis. We acknowledge the work of H&L as an important stepping stone establishing a source calibration facility which now achieves comparable results to others around the world.

* Corresponding author. Tel.: +1 505 606 2083; fax: +1 505 665 4433.
E-mail address: scroft@lanl.gov (S. Croft).

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2. Data analysis

We shall not repeat the clear and extensive description of their work provided by H&L but shall assume the reader has their paper to hand as we step through the data analysis.

As a preliminary experiment H&L determined the ^{56}Mn half-life during dilution experiments using both the main detector and the remote detector. Their results are summarized in Table 2 of Ref. (Hwang and Lee, 1988) with a mean value of 154.801 ± 1.112 min reported. Pooling the data from both detectors and performing an inverse variance weighted arithmetic mean, assuming the variance is dominated by counting precision the reanalyzed data yields a value of 154.81 ± 0.095 min. Here we report the uncertainty at the one external standard error (ext. SE) level derived from the scatter in the results. The internal SE, based on the assigned individual uncertainties, is larger at ±0.29 min suggesting the individual uncertainties in H&L's Table 2 may be overstated. Our analysis suggests the data of H&L would support a lower uncertainty on the half-life estimate than they claim.

Saturation activities (steady state after mixing and ingrowth) were estimated during both the growth phase and the decay phase. The values during the growth phase are systematically higher although no explanation is offered. The later analysis performed by H&L is performed solely on the growth phase data but the fact there is a difference could raise the question of whether an unrecognized systematic error exists and ought to be propagated based on the observed discrepancy. As to the magnitude of the difference H&L state that on the average the growth phase rate exceed the decay phase rates by a factor of 1.0128 ± 0.0009. Reanalyzing the data in H&L's Table 4 we obtain a weighted mean value of 1.0129 ± 0.0016, where again we have adopted the ext. SE. An unweighted analysis gives a similar result (1.0128 ± 0.0018). Both approaches give substantially larger uncertainty estimates than quoted by H&L although the magnitude of the effect is similar.

A further word on the difference between the growth phase and decay phase data is in order. The ^{56}Mn activity recorded may be thought of as coming from a fully mixed fraction of solution detected with efficiency ε and a component, too recently produced to allow it to be fully integrated into the volume of the solution in the bath, which is detected with a reduced efficiency $\varepsilon(1-\delta)$ (Smith and King, 1991). Analysis of the Mn-bath data therefore contains two mixing parameters, τ, the effective time to achieve complete mixing and, δ, the fractional difference in counting efficiency. H&L state the values of τ and δ used, 900 s and 0.3 respectively, but do not explain why these values were selected. It must be understood that τ and δ are parameters characteristic of the particular system and as such the values would have needed to be optimized empirically (Smith and King, 1991). Also, it must be appreciated that τ and δ are not independent. Having set an approximate value for τ the value of δ is generally picked so that the saturated activity estimated from the growth phases and decay phase agree (Smith and King, 1991). The systematic difference observed in the data of H&L could most likely be removed if an adjustment of the mixing parameters used in the data reduction was made. The difference between the two phases will not affect our conclusions as we shall see.

To determine the neutron source strength, Q, H&L used the following basic Eqs. (2.1)–(2.3) which we present in the form of a linear relation $y = ax$ with the terms defined as follows:

$$y = \frac{\varepsilon C}{A} = ax + b = \frac{1}{Q}\frac{\sigma_H}{\sigma_{Mn}}x + \frac{1}{Q}\left(1 + \frac{\sigma_S/\sigma_{Mn}}{(1+\overline{G\bar{r}s})_{Mn}}\right) \tag{2.1}$$

where C and x are given by:

$$C = (1-L)(1-O)(1-S) \tag{2.2}$$

$$x = \frac{N_H/N_{Mn}}{(1+\overline{G\bar{r}s})_{Mn}} \tag{2.3}$$

From relationship (2.1) there are three ways to calculate the source emission rate. From the linear fit the slope can be used:

$$Q = \frac{1}{a}\frac{\sigma_H}{\sigma_{Mn}} \tag{2.4}$$

or the emission rate can be obtained from intercept as:

$$Q = \frac{1}{b}\left(1 + \frac{\sigma_S/\sigma_{Mn}}{(1+\overline{G\bar{r}s})_{Mn}}\right) \tag{2.5}$$

or, a value for each of the six solution concentrations can be extracted separately using:

$$Q = \frac{1}{y}\left[\frac{\sigma_H}{\sigma_{Mn}}x + \left(1 + \frac{\sigma_S/\sigma_{Mn}}{(1+\overline{G\bar{r}s})_{Mn}}\right)\right] \tag{2.6}$$

Experimentally ε, C and A differ for every point and are subject to random uncertainty affecting y. Uncertainties in A are provided by H&L but not used in their data analysis. The fractional uncertainties are rather small and do not vary much across the six experiments increasing from 0.029% for experiment number 1 to 0.039 for experiment number 6. Unfortunately uncertainties in ε and C are not discussed. We assume that the uncertainty in ε is dominated by systematic uncertainty so that it will have negligible effect on the random fluctuation of a given measurement. Based on the state of the practice the accuracy of the 4π β-γ technique that underpins the ^{56}Mn calibration solution a small (in the context of our discussion) systematic uncertainty of about ±0.1% common to all six experiments can be estimated based on our understanding of present capabilities. The uncertainty in C is difficult to assess but fortunately the value of C is almost constant across the six experiments with a mean value of about 0.9924. If an arbitrary uncertainty of 5% is used in the deviation of C from unity then the fractional uncertainty in C turns into an assumed mostly systematic uncertainty of only about ±0.038%. H&L provide the nuclear data values they used along with uncertainties in the case of thermal cross sections. Uncertainties on the individual $(1+\overline{G\bar{r}s})$ values are not given but the range of values across the six experiments is not large, falling in the interval from 1.0133 to 1.0113 from experiment 1 to 6 with a mean of 1.0121 and a SE of ±0.00032 which was used as indicative of the uncertainty in both the individual values and also on the mean value $(1+\overline{G\bar{r}s})_{Mn}$. H&L list both N_H/N_{Mn} and x but do not discuss the uncertainties in these quantities nor do they make use of the uncertainties. However, the paper does give the formula used to calculate N_H/N_{Mn} in terms of the gravimetric and volumetric measurements and it is possible to propagate the uncertainties in these quantities to get an estimate of the uncertainty in N_H/N_{Mn}. The fractional uncertainties are significant and vary somewhat across the six experiments 1 to 6 as follows in %, 0.74, 0.40, 0.76. 0.64, 1.04, 0.33.

Despite the fact that each experiment has its own unique statistical worth the initial analysis by H&L of the data was based on an unweighted least squares fit. If there are large unrecognized random variations lurking in the data this would not systematically bias the results. However, assuming the uncertainty analysis on each point is valid, a weighted fit with uncertainties in both directions would seem more appropriate. This point will be discussed later in this paper. A slight complication arises in that the data are correlated through the embedded nuclear data. The random and systematic errors (due to nuclear data) are comparable. The correlation could be handled formally but is not germane to the purpose of this effort. Reworking the original data and