



Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by Battelle Since 1965

Metastable Radioxenon Verification Laboratory (MRVL) Year-End Report

November 2014

Matt Cooper
Jim Hayes
Lance Lidey



Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes **any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights.** Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PACIFIC NORTHWEST NATIONAL LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC05-76RL01830

Printed in the United States of America

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information,
P.O. Box 62, Oak Ridge, TN 37831-0062;
ph: (865) 576-8401
fax: (865) 576-5728
email: reports@adonis.osti.gov

Available to the public from the National Technical Information Service
5301 Shawnee Rd., Alexandria, VA 22312
ph: (800) 553-NTIS (6847)
email: orders@ntis.gov <<http://www.ntis.gov/about/form.aspx>>
Online ordering: <http://www.ntis.gov>



This document was printed on recycled paper.

(8/2010)

Contents

Metastable Radioxenon Verification Laboratory (MRVL).....2

1.1 Introduction2

1.2 Verification and Validation2

1.3 Bibliography.....4

Metastable Radioxenon Verification Laboratory (MRVL)

1.1 Introduction

The MRVL system is designed to measure multiple radioxenon isotopes (^{135}Xe , ^{133}Xe , $^{133\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$) simultaneously. The system has 12 channels to load samples and make nuclear measurements.

Although the MRVL system has demonstrated excellent stability in measurements of Xe-133 and Xe-135 over the year of evaluation prior to delivery, there has been concern about system stability over measurements performed on samples with orders of magnitude different radioactivity, and samples containing multiple isotopes. To address these concerns, a series of evaluation test have been performed at the end-user laboratory. The evaluation was performed in two separate phases. Phase 1 made measurements on isotopically pure Xe-133 from high radioactivity down to the system background levels of activity, addressing the potential count rate dependencies when activities change from extreme high to very low. The second phase performed measurements on samples containing multiple isotopes (Xe-135, Xe-133 and Xe-133m), and addressed concerns about the dependence of isotopic concentrations on the presence of additional isotopes. The MRVL showed a concentration dependence on the Xe-133 due to the amount of Xe-133m that was in the sample. The dependency is due to the decay of Xe-133m into Xe-133. This document focuses on the second phase and will address the analysis used to account for ingrowth of Xe-133 from Xe-133m.

1.2 Verification and Validation

There are two specific impacts of Xe-133m on Xe-133. First, the spectral region used in the beta-gamma detection of Xe-133m is in coincidence with a portion of the region from Xe-133. This overlap means that the portion of the signal that overlaps between Xe-133 and Xe-133m needs to be separated and accounted for in the software algorithm. The second impact of Xe-133m on Xe-133 is due to Xe-133m decaying into Xe-133, which means that a portion of the detected Xe-133 concentration does not originate from the Xe-133 that was present when the sample was produced, but is in fact ingrowth from the decay of Xe-133m.

The first impact of Xe-133m is accounted for in the current concentration calculation. This means, the area of overlap between Xe-133m and Xe-133 are subtracted out in the calculation of the Xe-133 concentration, so there will be no influence on Xe-133 concentrations from the number of detected decays of Xe-133m. The second impact of Xe-133m, the ingrowth of Xe-133, is not currently accounted for in the concentration calculations. The ingrowth of Xe-133 from Xe-133m can be accounted for by using a derived equation based on the Bateman equation (Bateman, 1910).

When the Xe-133 concentration, determined by the MRVL system, is plotted, it deviates from the expected decay curve for Xe-133. The deviation is due to the ingrowth of Xe-133 from Xe-133m, which is why this deviation is not observed in pure Xe-133 samples, but is observed in the mixed Xe-133/135 samples that contain Xe-133m as a contaminant. The deviation is shown in blue in *Figure 1* where the blue data show the concentration results for Xe-133 from the MRVL. The blue line is a quadratic fit to the data and is illustrative of the deviation from the expected horizontal straight line. The expected horizontal straight line is also shown, and in the case of the Xe-133m concentration reported by the MRVL are shown in green. Finally, when the

correction for ingrowth of Xe-133 from Xe-133m, derived from the Bateman equation, is applied the Xe-133 concentration, the data comes in line with the expected Xe-133 decay and can be fit with a horizontal straight line as seen in blue in *Figure 1*.

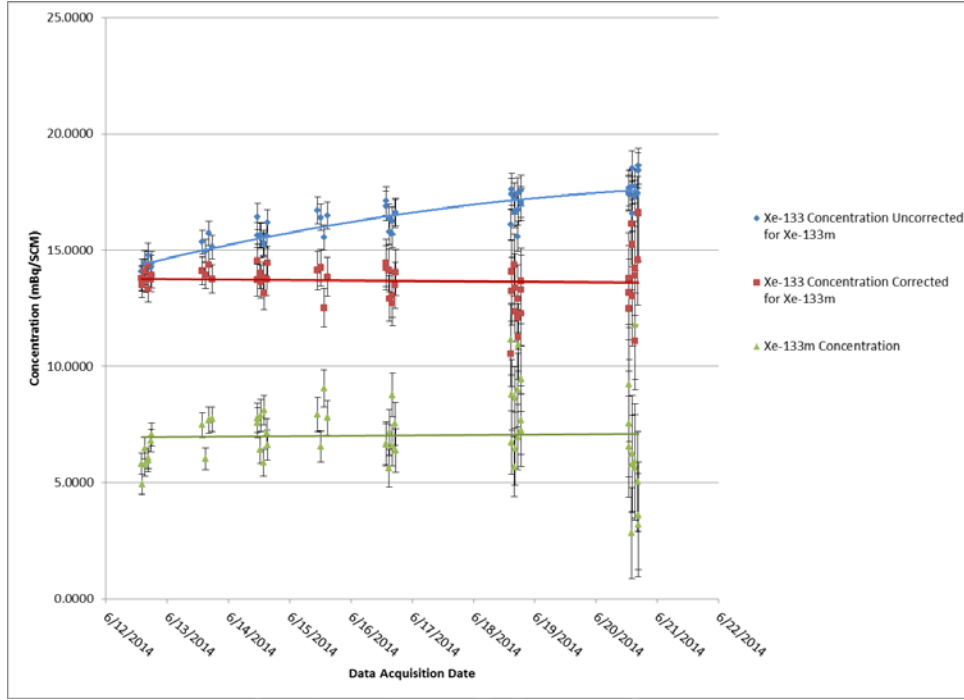


Figure 1. A plot showing the Xe-133 and Xe-133m results from the evaluation testing decay corrected to a common time, T_0 . In blue are the uncorrected Xe-133 results, which demonstrate the effect of ingrowth from Xe-133m, having an increasing concentration over time. The green data shows the results from Xe-133m, which has a linear fit. The linear fit is horizontal which corroborates that the samples are nearly identical and that decay and background corrections are performed correctly in the concentration calculation code. Finally, the data in red are Xe-133 that has been corrected to account for the ingrowth from Xe-133m. The corrected Xe-133 data also has a linear fit that is horizontal which demonstrates that the algorithm in the concentration calculation paired with the ingrowth correction, given in this paper, is accurate.

The equation used to perform the correction for the ingrowth of Xe-133 from Xe-133m is given by Eq. 1 (Ringbom, 2007) as a form of the Bateman equation. In Eq. 1, there are several terms that need additional explanation. C stands for concentration, where C_{Xe133m} is the concentration of Xe-133m reported by the MRVL and C'_{Xe133} is the concentration of Xe-133 reported by the MRVL. Second, there are six terms ($F_{C1}, F_{C2}, F_{P1}, F_{P2}, F_{A1}, F_{A2}$) that represent the isotopic decay for three times: collection, processing and acquisition. The collection time is set to 1 second since the systems decay-correct back to collection stop. The collection term remains in the concentration calculations to be consistent with the International Monitoring System (IMS) calculation method. The second time is the processing time, which is defined as the time between collection stop and acquisition start. The last time is the acquisition time, which in general is a 1000-minute count.

$$C_{Xe133} = C'_{Xe133} - \xi \cdot C_{Xe133m} \quad \text{Eq. 1}$$

Where,

$$\xi = \frac{\lambda_{Xe133}^2}{\lambda_{Xe133m}^2 \cdot (\lambda_{Xe133} - \lambda_{Xe133m})} \cdot \frac{F_{C1}}{F_{C2}} \cdot \left(\lambda_{Xe133} \cdot \frac{F_{P1} \cdot F_{A1}}{F_{P2} \cdot F_{A2}} - \lambda_{Xe133m} \right)$$

$$F_{C1} = 1 - e^{-\lambda_{Xe133m} \cdot T_C}$$

$$F_{C2} = 1 - e^{-\lambda_{Xe133} \cdot T_C}$$

$$F_{A1} = 1 - e^{-\lambda_{Xe133m} \cdot T_A}$$

$$F_{A2} = 1 - e^{-\lambda_{Xe133} \cdot T_A}$$

$$F_{P1} = e^{-\lambda_{Xe133m} \cdot T_P}$$

$$F_{P2} = e^{-\lambda_{Xe133} \cdot T_P}$$

The equation used to account for the Xe-133m decay, which results in biased Xe-133 activity concentration numbers, has been shown to remove the Xe-133 bias that is due to the Xe-133m decay. There is one outstanding issue remaining; where does the correction get applied? The equation can be implemented into the MRVL code that calculates the Xe-133 activity concentration or the equation can be applied at the end-user facility. The decision of where to apply the correction, i.e., in the MRVL system software algorithm or after the data is integrated into the Watchman software, will need to be made in conjunction with the user.

1.3 Bibliography

- Bateman, H. (1910). Solution of a System of Differeential Equations Occuring in the Theory of Radio-active Transfotmations. *Proceedings of the Cambridge Philosophical Society, Mathematical and physical sciences*, 423-427.
- Ringbom, A. (2007). *Derivation of the concentration equation assuming parent-daughter pair present in sample*. Stockholm, Sweden: Swedish Defence Research Agency (FOI).



Pacific Northwest
NATIONAL LABORATORY

*Proudly Operated by **Battelle** Since 1965*

902 Battelle Boulevard
P.O. Box 999
Richland, WA 99352
1-888-375-PNNL (7665)

U.S. DEPARTMENT OF
ENERGY

www.pnnl.gov