Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.37852

Advances in Thermal Ionization Mass Spectrometric Analysis of $^{135}Cs/^{137}Cs$ at Ultra-trace Levels and Extreme Dynamic Ranges

ALLISON A PRICE, HAYDEN MILLER, ANDREW A REINHARD, CHARLES LEWIS, JEREMY INGLIS, KIMBERLY A HINRICHS, KASSANDRA L CHRISTENSEN, SHELBI HRKACH, RONALD S AMATO, KATELYN GREEN, ROBERT E STEINER AND STEPHEN P LAMONT

Los Alamos National Laboratory

Cesium is produced as a fission fragment by a variety of nuclear processes. Given cesium is monoisotopic (stable ^{133}Cs) in natural samples, radiocesium offers a valuable nuclear nonproliferation monitoring tool. While ^{137}Cs (half-life = 30.07 years) is routinely measured in a variety of environmental matrices by gamma-spectrometry, analyses of additional Cs radioisotopes are required for discrimination between different nuclear sources. With a long half-life (2.3×10^6 years), anthropogenic ^{135}Cs, in ratio to ^{137}Cs, is an excellent candidate to distinguish between, and trace the transport and mixing of, various source terms. However, analysis of $^{135}Cs/^{137}Cs$ ratios by mass spectrometry presents several challenges. The high abundance of stable ^{133}Cs relative to trace-level ^{135}Cs and ^{137}Cs requires the ability to perform analyses at extreme dynamic ranges and leads to potential abundance sensitivity limitations. Isobaric interferences arise from stable barium, and stable rubidium can suppress Cs ionization. Both elements occur in high concentrations in environmental matrices and require thorough separation. Furthermore, the absence of a commercially available $^{135}Cs/^{137}Cs$ reference material has historically hindered method validation.

Here we describe our approach to the preparation and analysis of $^{135}Cs/^{137}Cs$ ratios at extreme dynamic ranges ($^{135,\ 137}Cs/^{133}Cs < 1 \times 10^{-8}$) and ultra trace levels ($^{135,\ 137}Cs$ ~10 fg) by thermal ionization mass spectrometry (TIMS). We first demonstrate an optimized chemical separation of Cs from interfering elements. Then, using a novel Nuclear Forensics Reference Material (NFRM) with an informative $^{135}Cs/^{137}Cs$ ratio, we evaluate the performance of a TIMS configuration equipped with multiple ATONA Faraday cups, a secondary electron multiplier (SEM), and a Wide Aperture Retarding Potential (WARP) system for ultra–high-dynamic-range measurements of radiocesium in environmental samples. Particular attention is given to strategies for managing the intense ^{133}Cs ion beams (>3 nA) derived from the inherent stable Cs in the samples, which can promote ion scattering and compromise measurement accuracy. Utilizing this information, we analyze mixes of the $^{135,137}Cs$ NFRM with ^{133}Cs single element standard and USGS BCR-2. With these analyses we are, for the first time, able to assess the precision and accuracy of trace level $^{135}Cs/^{137}Cs$ analyses of environmental samples for nonproliferation monitoring. LA-UR-26-21356