

High-precision stable xenon isotope ratio measurements of atmospheric samples

H. Miller, A. Reinhard, D. Eldridge, and T. Rahn

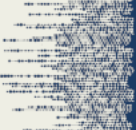
Los Alamos National Laboratory



INTRODUCTION AND MAIN RESULTS

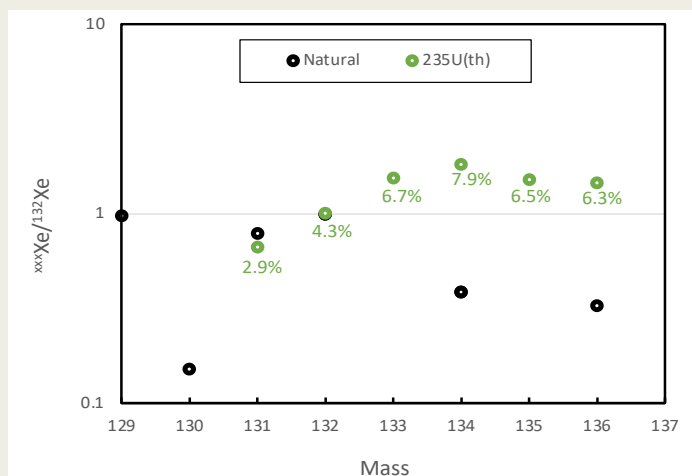
This presentation outlines the utility of stable xenon as a persistent signature of illicit nuclear activities that can be employed over timescales beyond that of radioxenon. We outline a new methodology, employing a Thermo 253 Ultra dual-inlet dynamic mass spectrometer, that can measure stable xenon isotope ratios with 10 permeg precision. In addition, we will discuss our sample purification technique.

**Note: Presenting author Hayden Miller *not* attending in-person.
Please direct questions to hmler@lanl.gov**



Introduction

Nuclear explosion monitoring techniques such as those used by the International Monitoring System (IMS), setup as a verification tool for the Comprehensive Nuclear-Test-Ban Treaty (CTBT), rely primarily on radioxenon isotopes to detect illicit nuclear activity. However, nuclear power plant operation and isotope production facilities produce large amounts of the radioxenon isotopes making distinguishing background from potential illicit activity extremely difficult. Stable xenon offers an additional constraint on the nature of fission producing events.



Xe isotope ratios for natural and ^{235}U thermal fission sources. Percent cumulative fission yields for each isotope are in green below the ratio. Data from ENDF-349.

Mass Spectrometry

Dual-inlet gas-source isotope ratio mass spectrometry

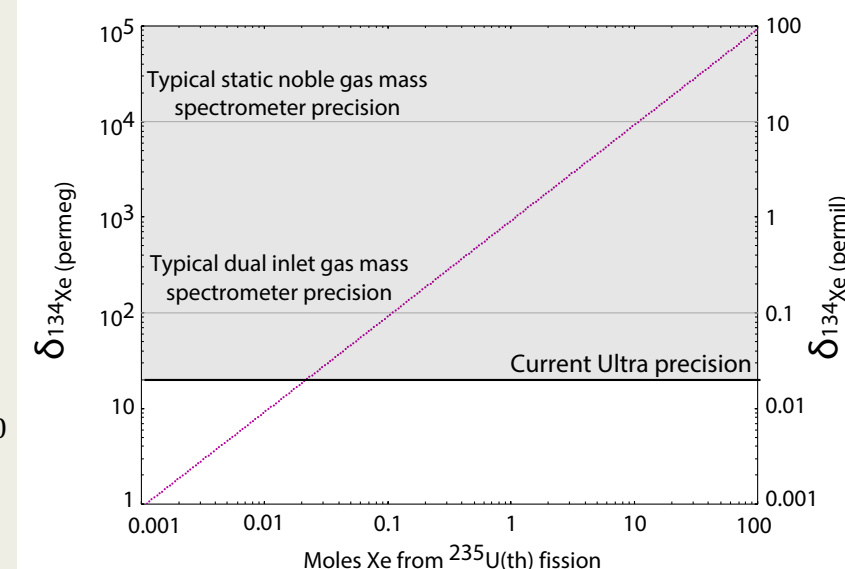
- Extremely high-precision isotope ratio determinations
- Large samples by mass spectrometry standards
- Requires close chemical and isotopic sample – standard matching
- Isotope ratio data reported in delta notation, where 1 permil = 1000 permeg:

$$\delta^{13x}\text{Xe}(\text{permil}) = \left[\left(\frac{(^{13x}\text{Xe}/^{132}\text{Xe})_{\text{measured}}}{(^{13x}\text{Xe}/^{132}\text{Xe})_{\text{atmosphere}}} - 1 \right) \times 1000 \right]$$



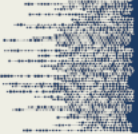
Thermo 253 Ultra Gas Source IRMS

Results



Future Work

- Characterize natural background (currently ongoing)
- Collect air samples where stable xenon perturbations are expected
- Sample processing methodology can be applied to water samples → apply to suite of National Nuclear Security Site (formerly Nevada Test Site) groundwater samples



Simplified Sample Purification Procedure

400 mL STP air → 4 mL STP pure Ar + Kr + Xe

- 1) Connect sample flask and pump line to high vacuum.
- 2) Isolate line, expand sample to preconditioned getter for 5 minutes to begin to react away active species.
- 3) Cryo-focus gas onto Si gel trap submerged in liquid-N₂ bath (77 K).
- 4) Isolate getter side of line, warm Si gel trap.
- 5) Getter additional 15 minutes, trap onto Si gel for 15 minutes while open to sample side of line again
- 6) Isolate Si gel trap, open line to turbo (pumps He, H₂)
- 7) Warm Si gel trap, open to getter side of line for additional 2.5 hours
- 8) Cryo-focus onto detachable sample tube

