Cesium Environmental Radiochemistry:

Isotopic Signatures, Sediment Partitioning, & Environmental Cycling

Darin Snyder (INL) & Sue Clark (WSU)

Second International Symposium on the Fukushima Accident

June 7, 2013

www.inl.gov

Idaho National Laboratory

Washington State University

Acknowledgements

Jim Delmore (INL) – Oversight and mentorship

Nick Mann (INL) – Instrumental during sample preparation and chemical separation method development

Troy Tranter (INL) – Critical to the procedure development for chemical separation

John Olson (INL) – Led the thermal ionization mass spectrometer modifications and provided guidance during measurements

Mike Abbott (INL) – Provided valuable insight with respect to atmospheric transport and dispersal of fission products and access to key samples

Mathew Snow (WSU & INL) – Preparation and analysis of a variety of soil samples from containing Chernobyl debris as well as reprocessing contamination

We thank the U.S. Department of Energy NNSA and Office of Science for providing funding to support this work.

Outline

1. Cesium Isotopic Signatures
   a) Discriminating between recent and legacy cesium
   b) Elucidation of source
2. Cesium Cycling in Freshwater Systems
3. Summary, Observations, Recommendations

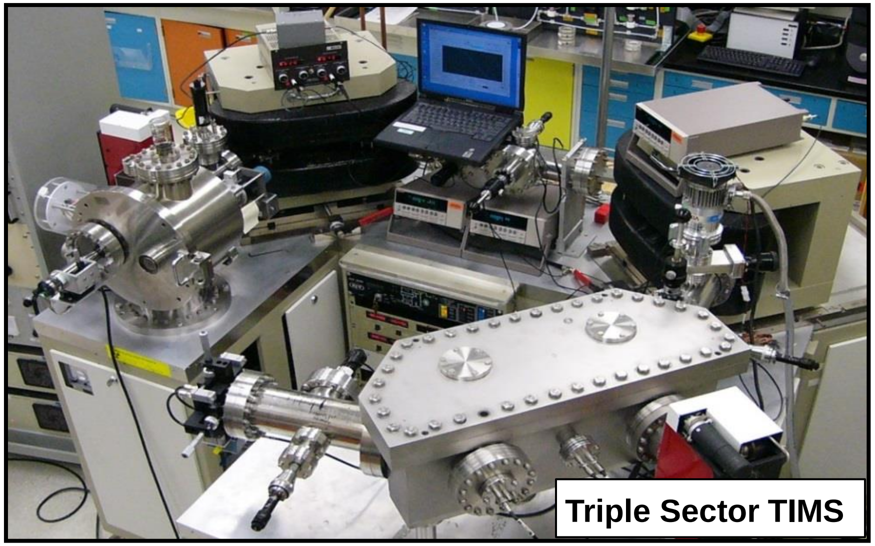
Outline

1. **Cesium Isotopic Signatures**
   a) **Discriminating between recent and legacy cesium**
   b) **Elucidation of source**
2. Cesium Cycling in Freshwater Systems
3. Summary, Observations, Recommendations

Cs Signatures

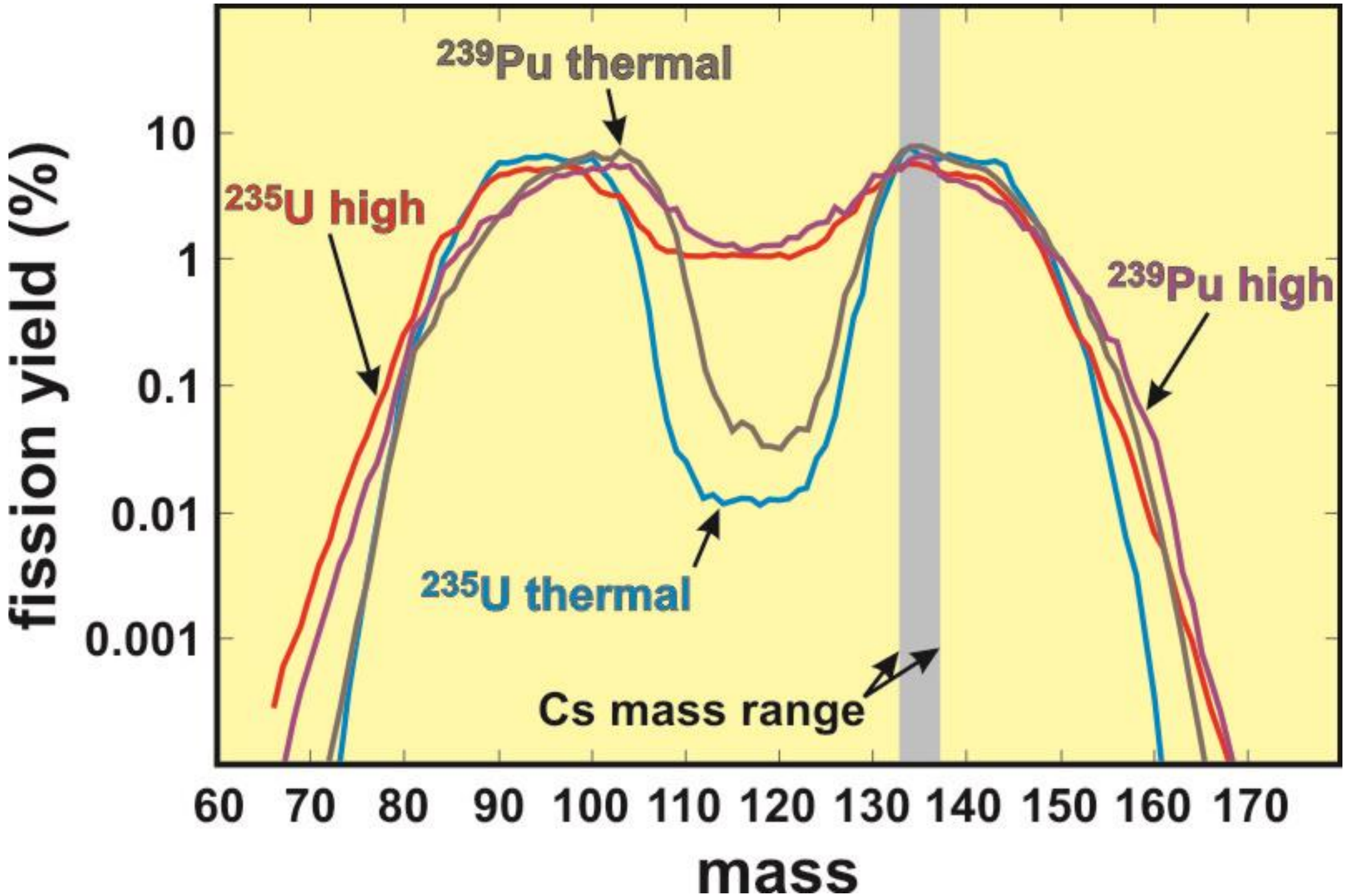
Allow for the differentiation among possible origins of fission product Cs:

- Is it old or new bomb debris, or reprocessing of old or new fuel from a power reactor?
- Mass spectrometric analysis of Cs can provide information to aid in answering these signature questions.

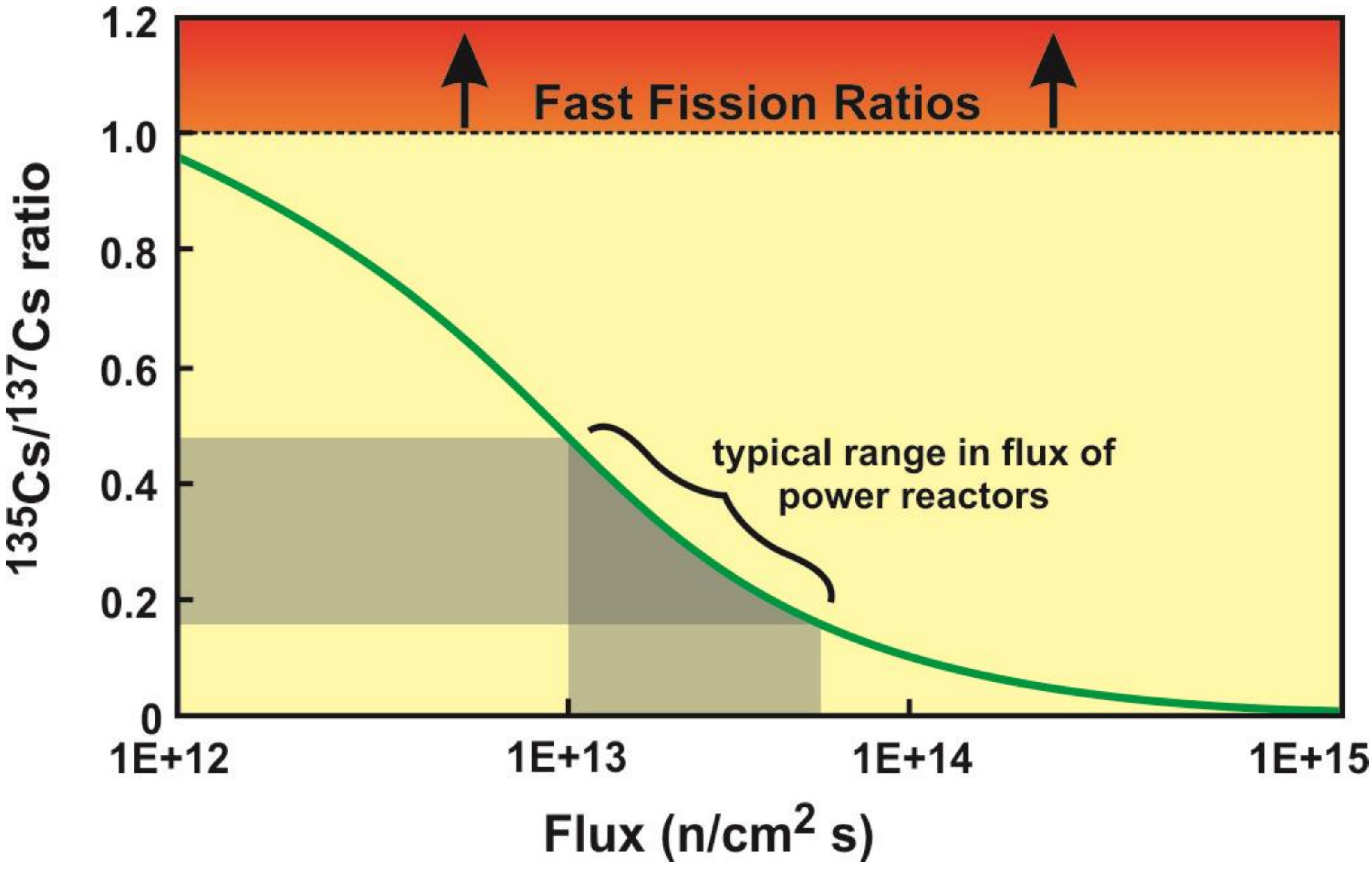


Triple Sector TIMS

Cesium Fission Yields



Ratio vs. Neutron Flux



Sources of Fission Product Cs

Potential sources of fission product Cs in the environment include: aged bomb debris, Chernobyl and Fukushima debris, effluents from an operating power reactor or reprocessing facility, and effluents from a recent detonation

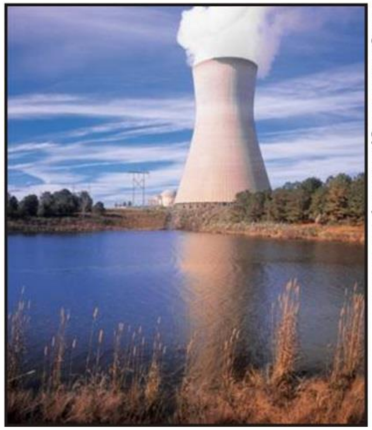
aged bomb debris

DOE

Chernobyl or Fukushima debris

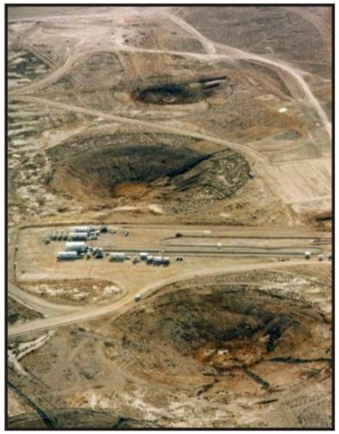
atomicarchive.com

operating thermal reactor or reprocessing of fuel



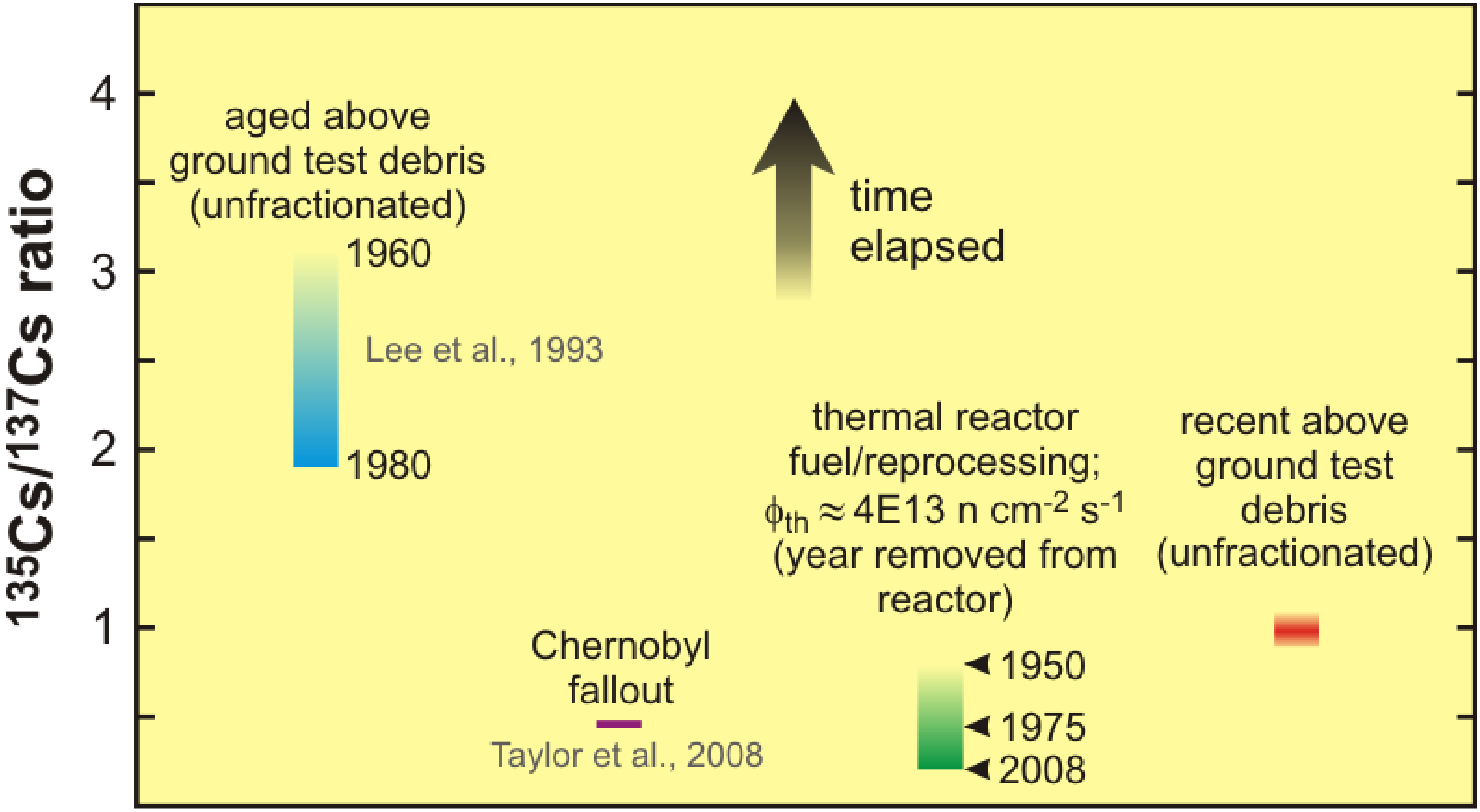
Progress Energy, Murray & Assoc.

effluents from a recent bomb test



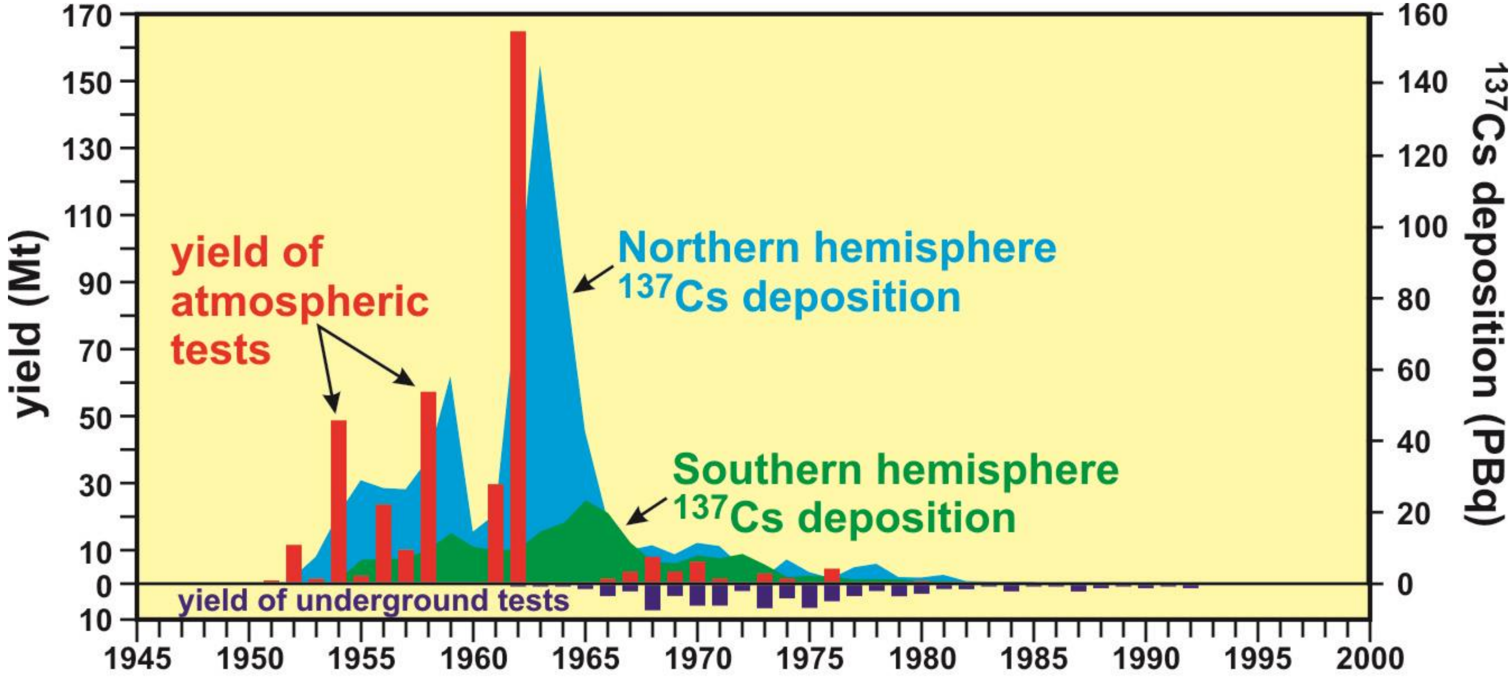
nuclearweaponsarchive.org

Anticipated Cs Isotope Ratios



Global Fallout Record

- Estimated annual global yield for both atmospheric and underground nuclear tests
- 1963 fallout peak results from the spike in atmospheric yield in 1962

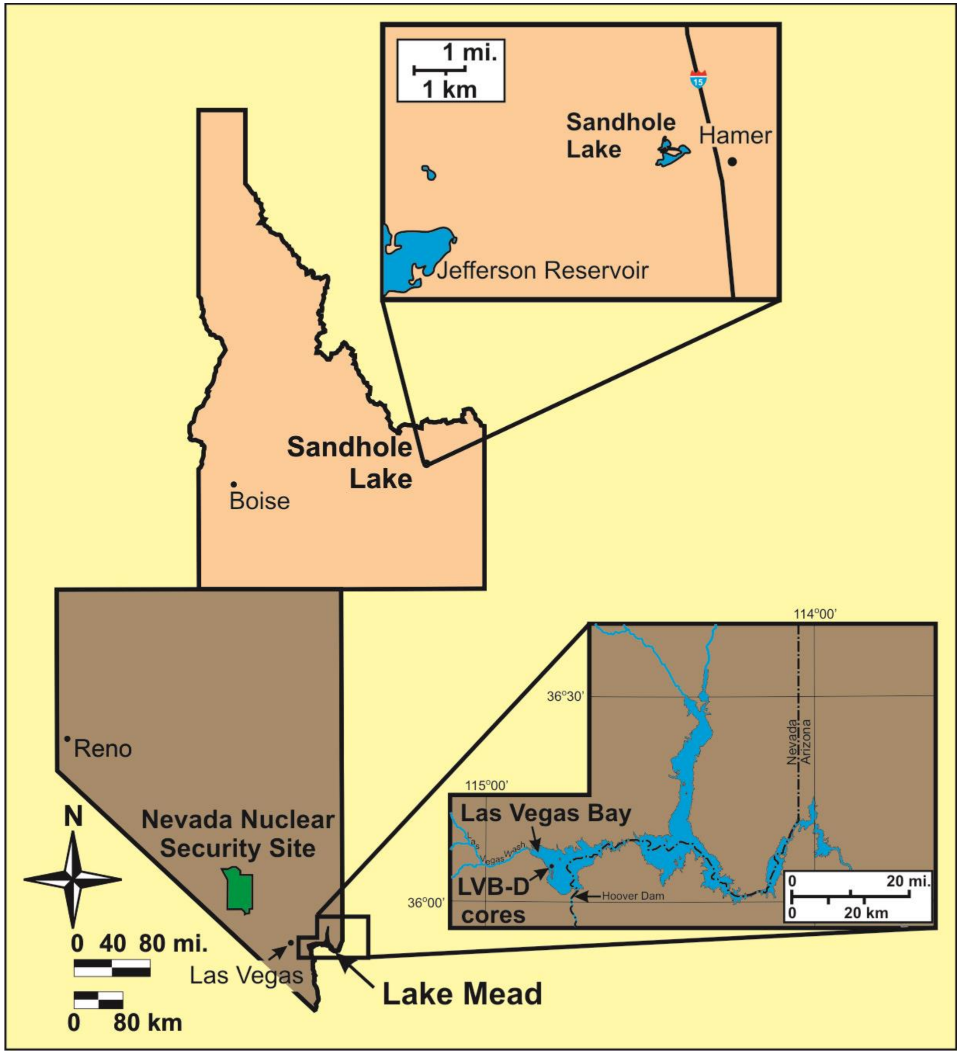


UNSCEAR, 2000.

Atmospheric Fallout Samples

Sediment samples for this work come were obtained from Sandhole Lake, ID and Lake Mead, NV.

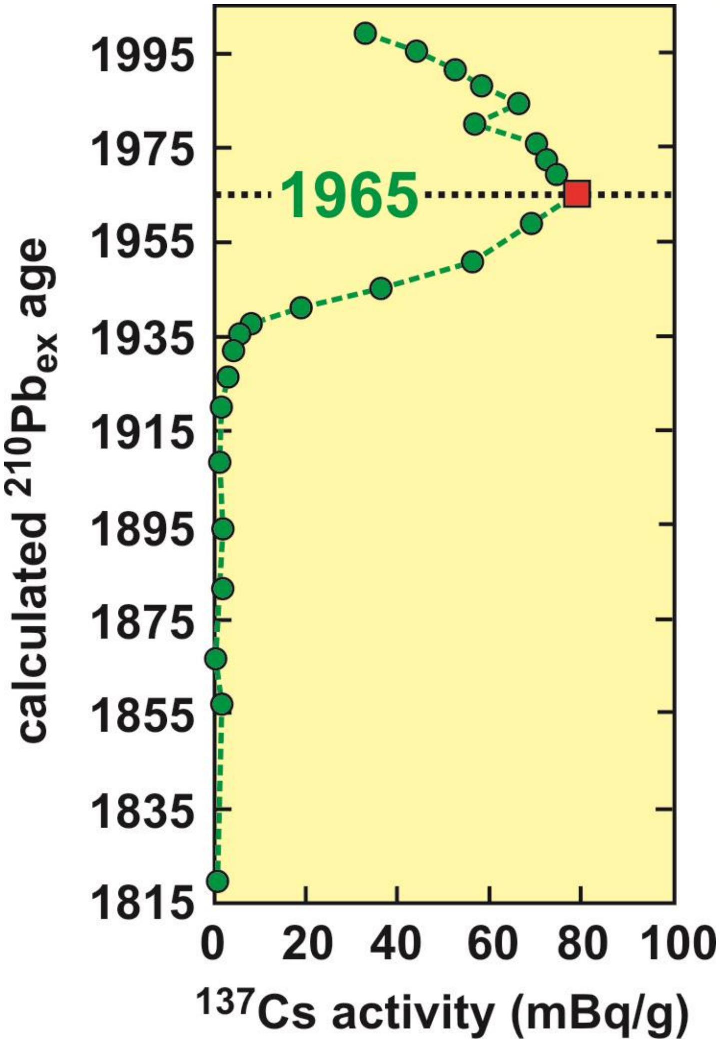
Sandhole Lake is located ~850 km NNE of NNSS, whereas Lake Mead lies ~130 km SE of NNSS.



Global Fallout Signature

Sandhole Lake, ID

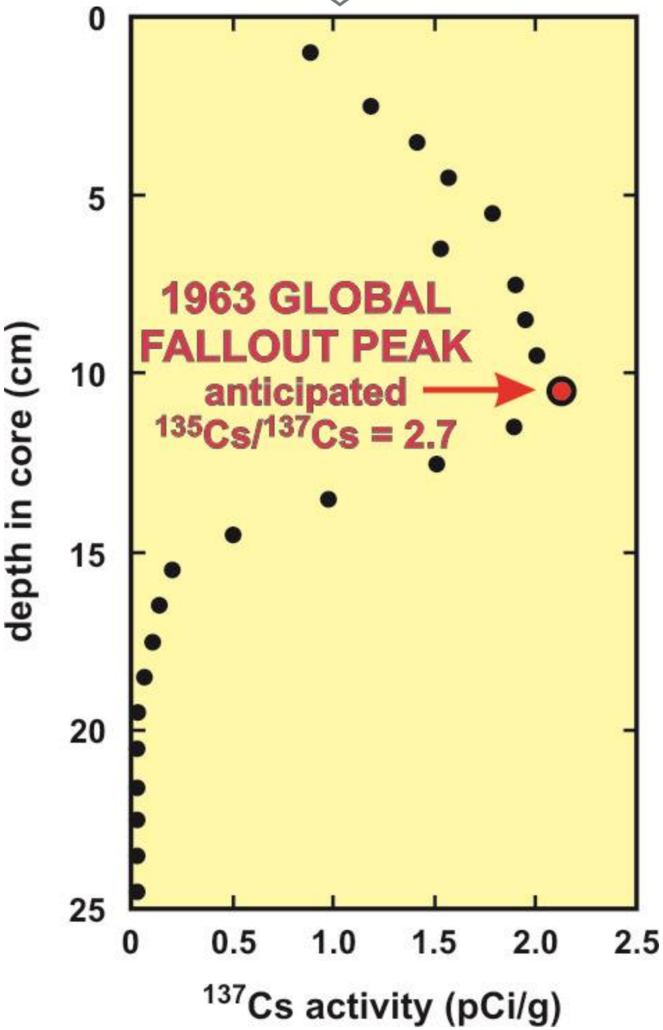
- ^{137}Cs depth profile exhibits a classic global fallout distribution
- Calculated $^{210}Pb_{ex}$ age is consistent with the ^{137}Cs distribution
- Sample is an ideal candidate to evaluate the $^{135}Cs/^{137}Cs$ ratio chronometer



Global Fallout Signature

Sandhole Lake, ID

- A spring-fed lake, dominated by aeolian sedimentation
- The maximum ^{137}Cs activity (2.13 pCi/g at 10.5 cm depth) is assumed to represent the 1963 global fallout peak
- Global fallout of this age should have a $^{135}Cs/^{137}Cs$ ratio of ~2.7



Snyder et al., 2012, J. Env. Radio., 110, 46-52.

Sandhole Lake Results

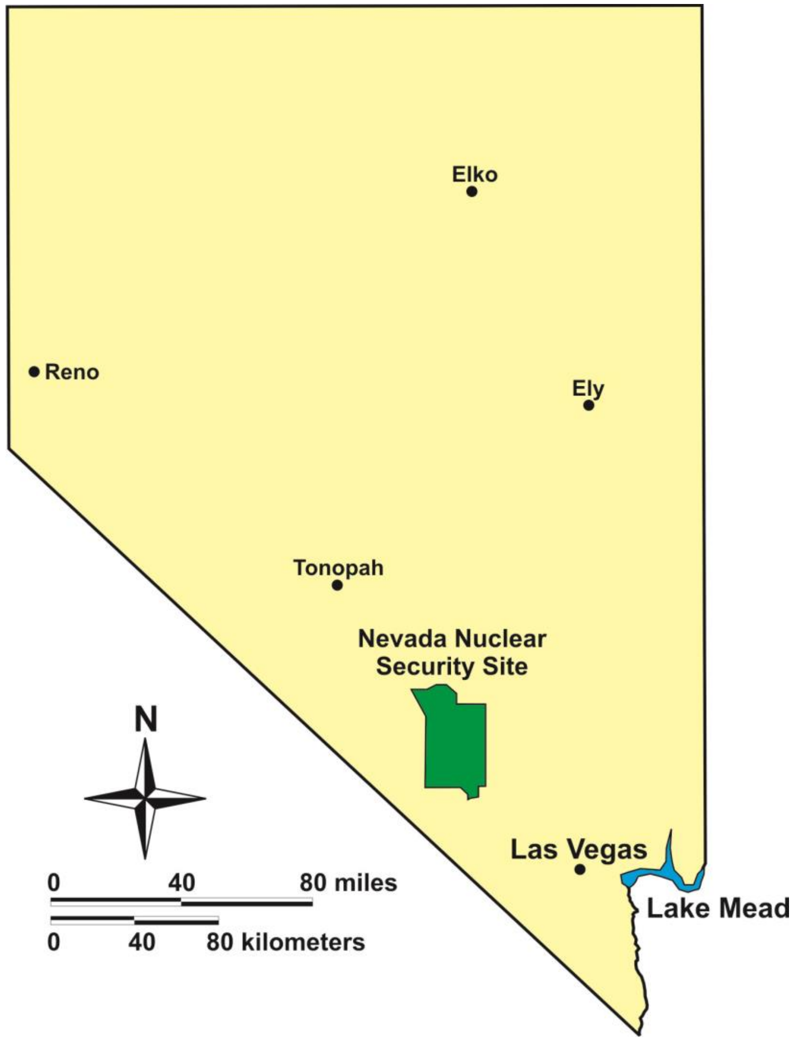
- The theoretical initial (1963) $^{135}Cs/^{137}Cs$ ratio of globally derived fallout is 1
- The measured $^{135}Cs/^{137}Cs$ ratio of the Sandhole Lake sample is 2.7 ± 0.5; this corresponds to an age-corrected, initial $^{135}Cs/^{137}Cs$ ratio of 1
- Consideration of the uncertainty of the measured ratio provides an age range of the fission product Cs of 35 – 51 y (1958 – 1974); (measured in 2009)
- Independent of the Cs results, USGS analysis of excess ^{210}Pb provides an age of 43 y for this sample

Regional Fallout Signature

Lake Mead lies approximately 130 km southeast of Frenchman Flat, NNSS.

Sediment samples from Lake Mead provide the opportunity to measure the $^{135}Cs/^{137}Cs$ ratio of locally derived fission product.

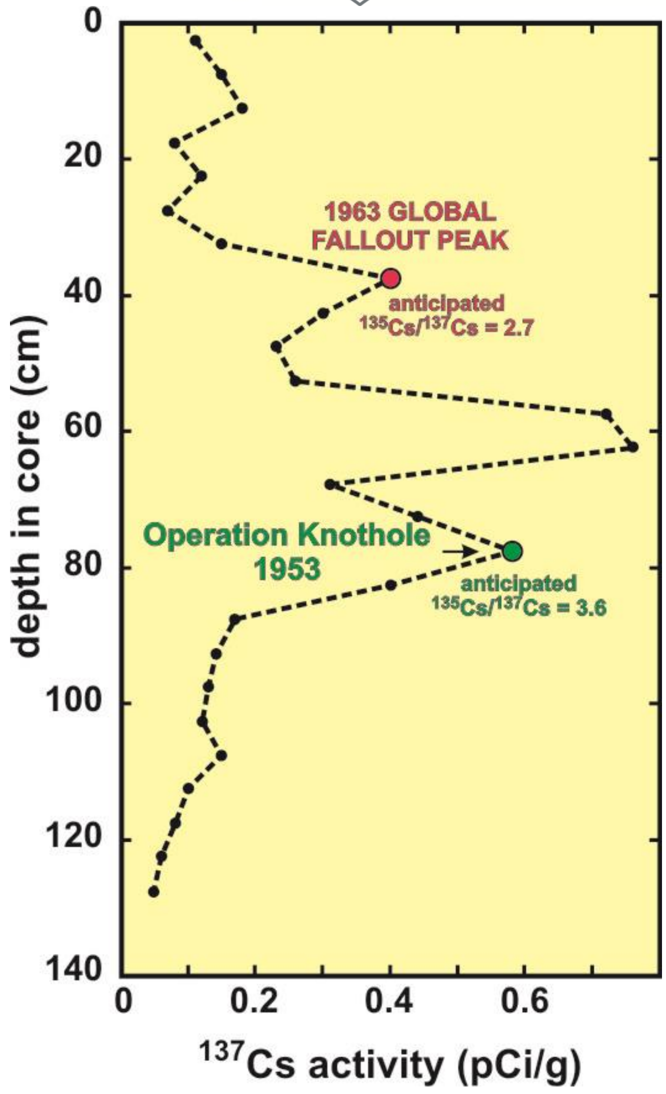
The USGS has made available lake core samples from previously conducted studies.



Lake Mead Samples

Van Metre et al. (2004)

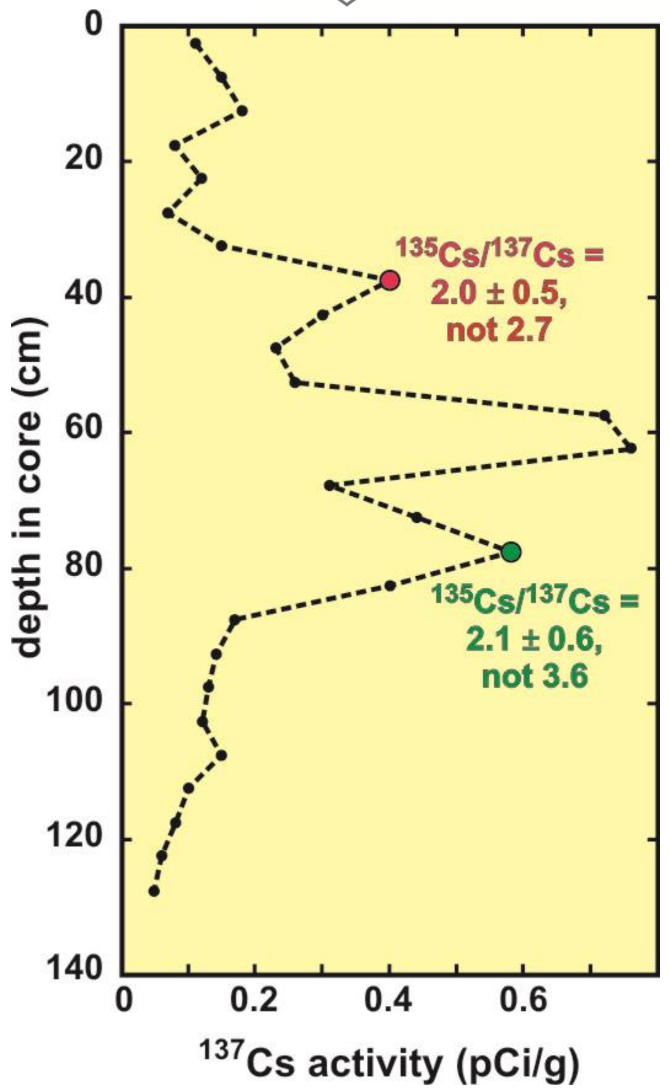
- individual ^{137}Cs peaks were ascribed to individual test shots as well as the 1963 global fallout peak
- measurement of the $^{135}Cs/^{137}Cs$ ratios of these samples can be used to evaluate these ages



Lake Mead Results

Our measured Cs ratios are inconsistent with the Van Metre et al. (2004) interpretation.

- the uppermost sample in the core does *not* represent global fallout
- the lowermost sample does not represent unfractionated, fission product from the Operation Knothole test series



Snyder et al., 2012, J. Env. Radio., 110, 46-52.

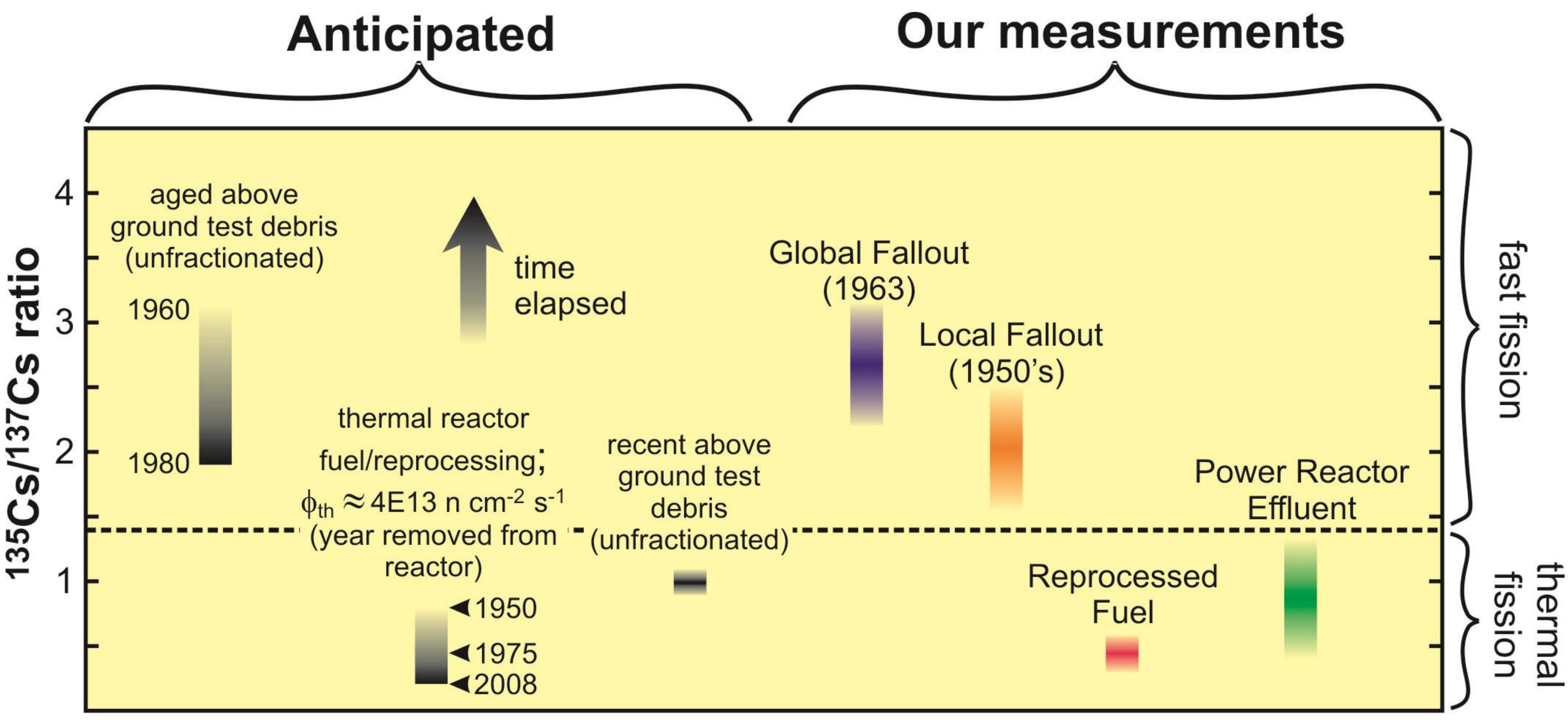
Lake Mead Interpretation

Assuming an initial $^{135}Cs/^{137}Cs$ ratio of 1 for these samples, the measured Cs ratio suggests that the fission product (in BOTH samples) is only about 30 years old.

However….

The last above ground nuclear test at the NNSS was conducted in July of 1962, therefore there hasn't been a source of a significant quantity of fission product for more than 50 years. It's more likely that these samples consist of fractionated, mixed-age fission product Cs. Redistribution of contaminated soil by wind and surface water have strongly influenced the geographical distribution of Cs.

Measured Signatures



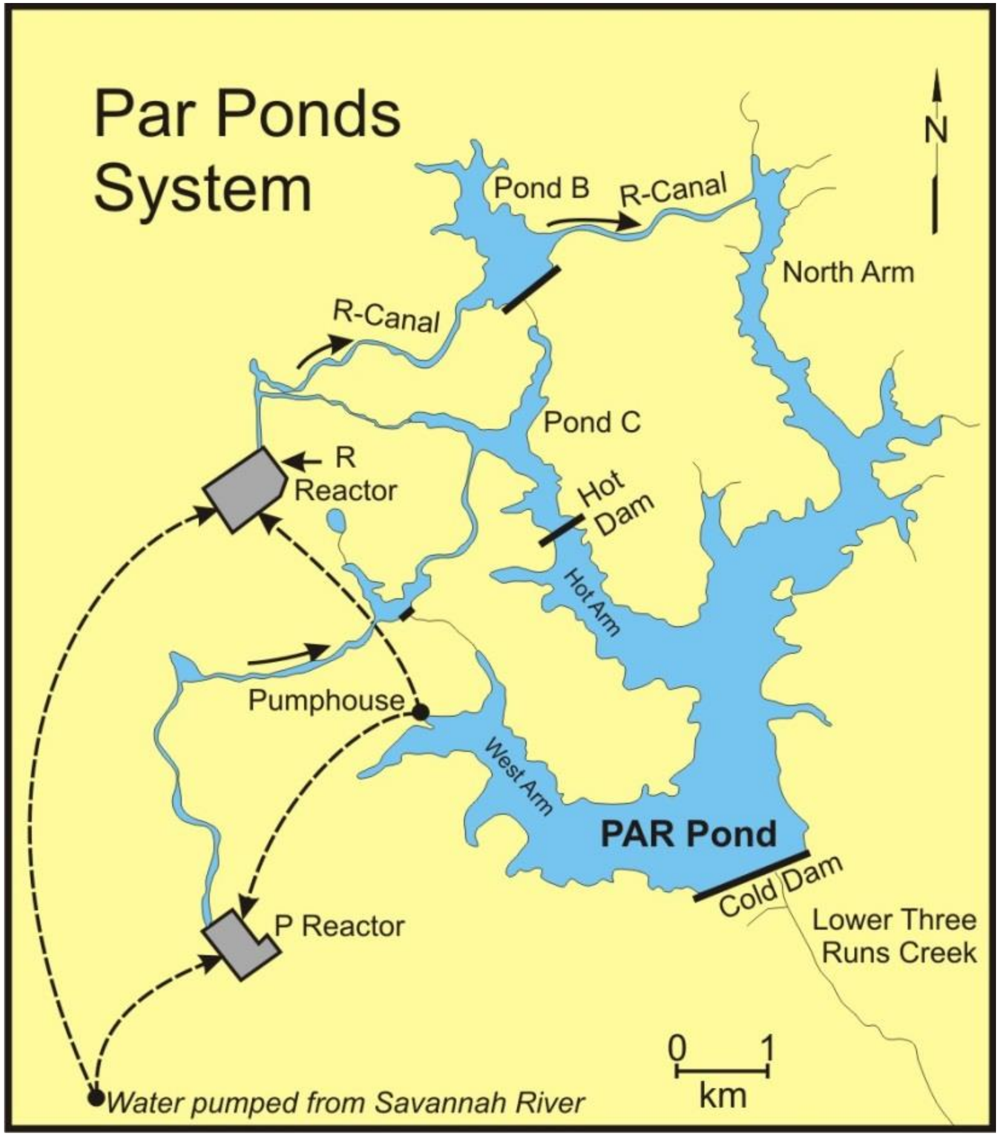
Isotopic Signatures Summary

- Demonstrated ability to measure $^{135}Cs/^{137}Cs$ ratios on environmental matrix at < 100 fg (< 10 pCi) ^{137}Cs level
- Developed a library of measured signatures → fast vs. thermal fission
- Demonstrated ability to constrain the age of fission product Cs
- Lake Mead *and* power reactor samples suggest 135 – 137 fractionation can occur and is measureable

Outline

1. Cesium Isotopic Signatures
   a) Discriminating between recent and legacy cesium
   b) Elucidation of source
2. **Cesium Cycling in Freshwater Systems**
3. Summary, Observations, Recommendations

Savannah River Site, USA



Located in the southeastern US

Warm, humid climate

Series of man-made ponds used as cooling system for reactors

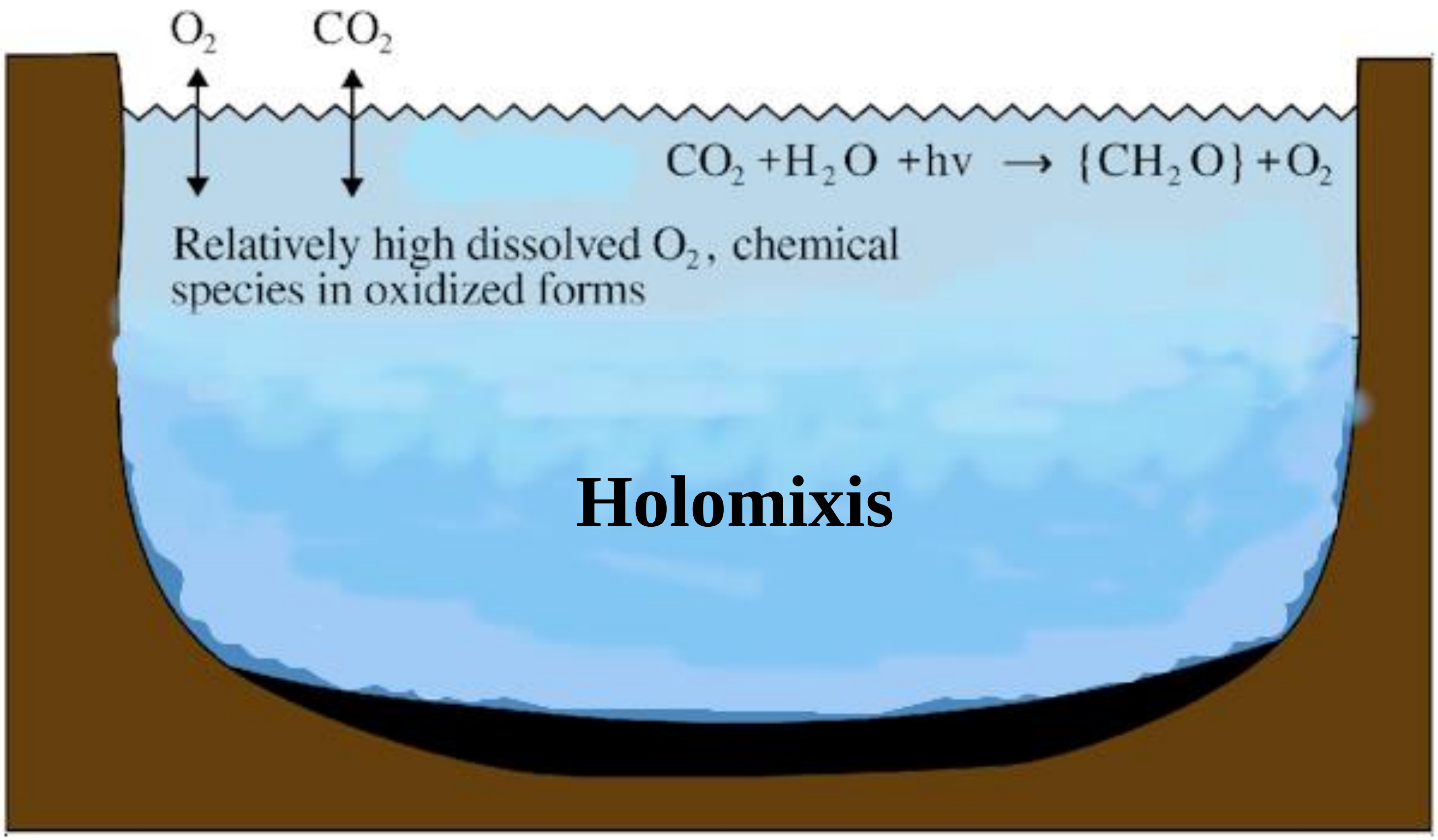
Reactor operations ceased in 1980's; legacy fission product signatures remain

Pond B is a shallow, monomictic lake

Pond B Field Site

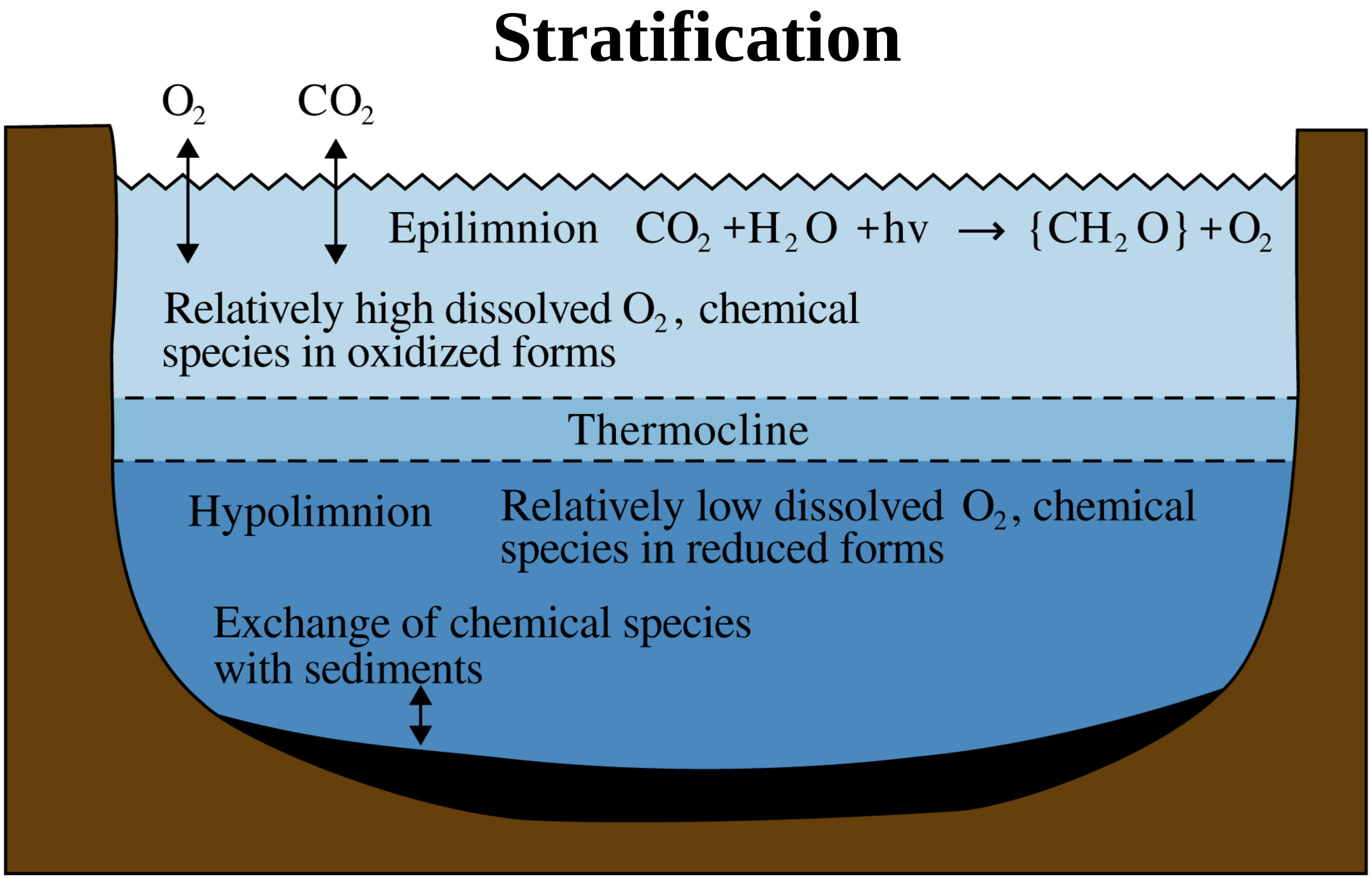
- Pond B was a part of the PAR Pond cooling system for the Savannah River defense materials production reactors.
- Pond B is a monomictic limnological system
  - Thermal stratification in mid-February
  - Lower anoxia develops about 30 days later
  - Thermally-driven system turnover in late October
- Water column and sediment samples collected seasonally

Seasonally Driven Stratification



Manahan, Stanley E., *Environmental Chemistry*, 6th edition, 1994.

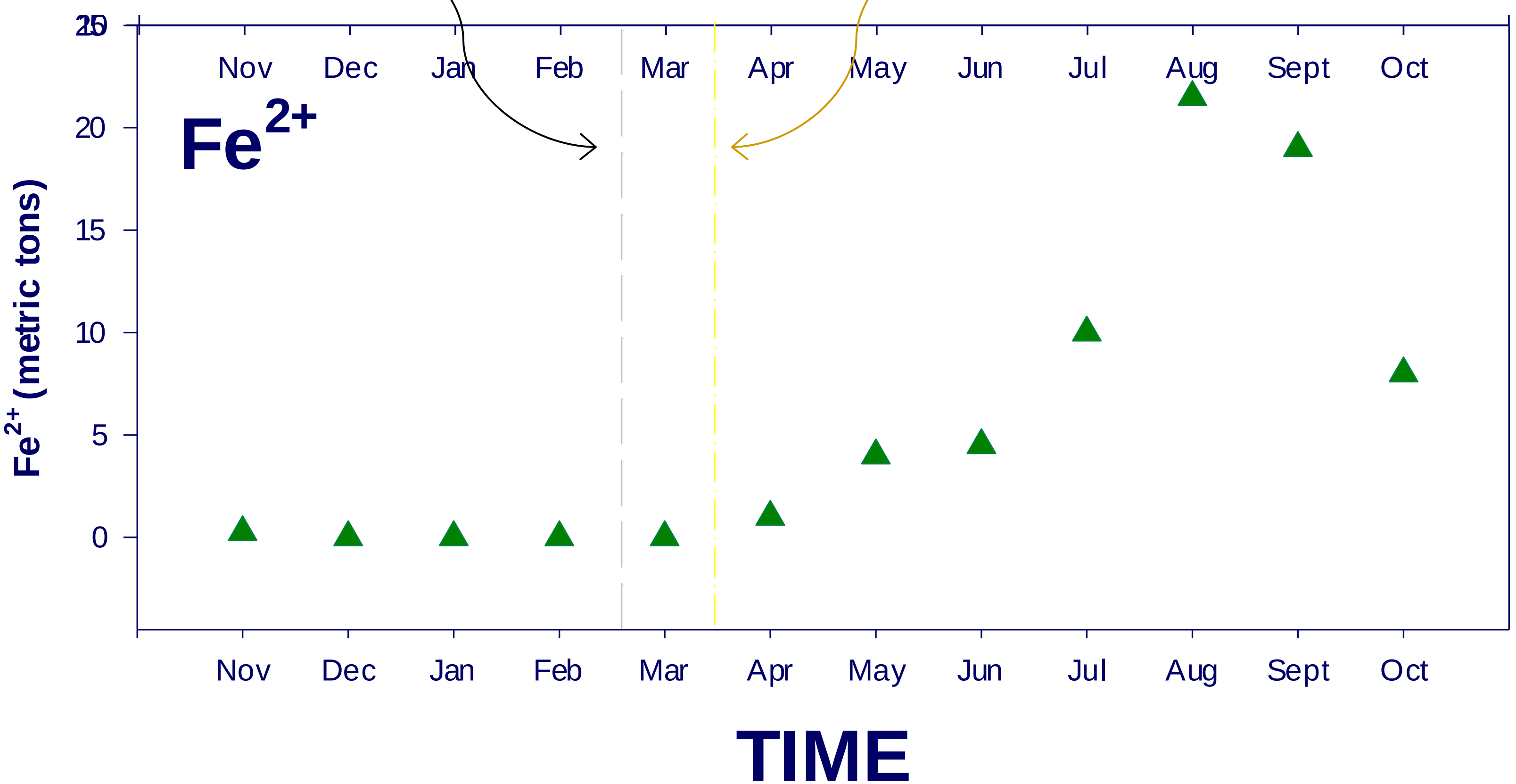
Seasonally Driven Stratification



Manahan, Stanley E. *Environmental Chemistry.* Sixth Edition. 1994.

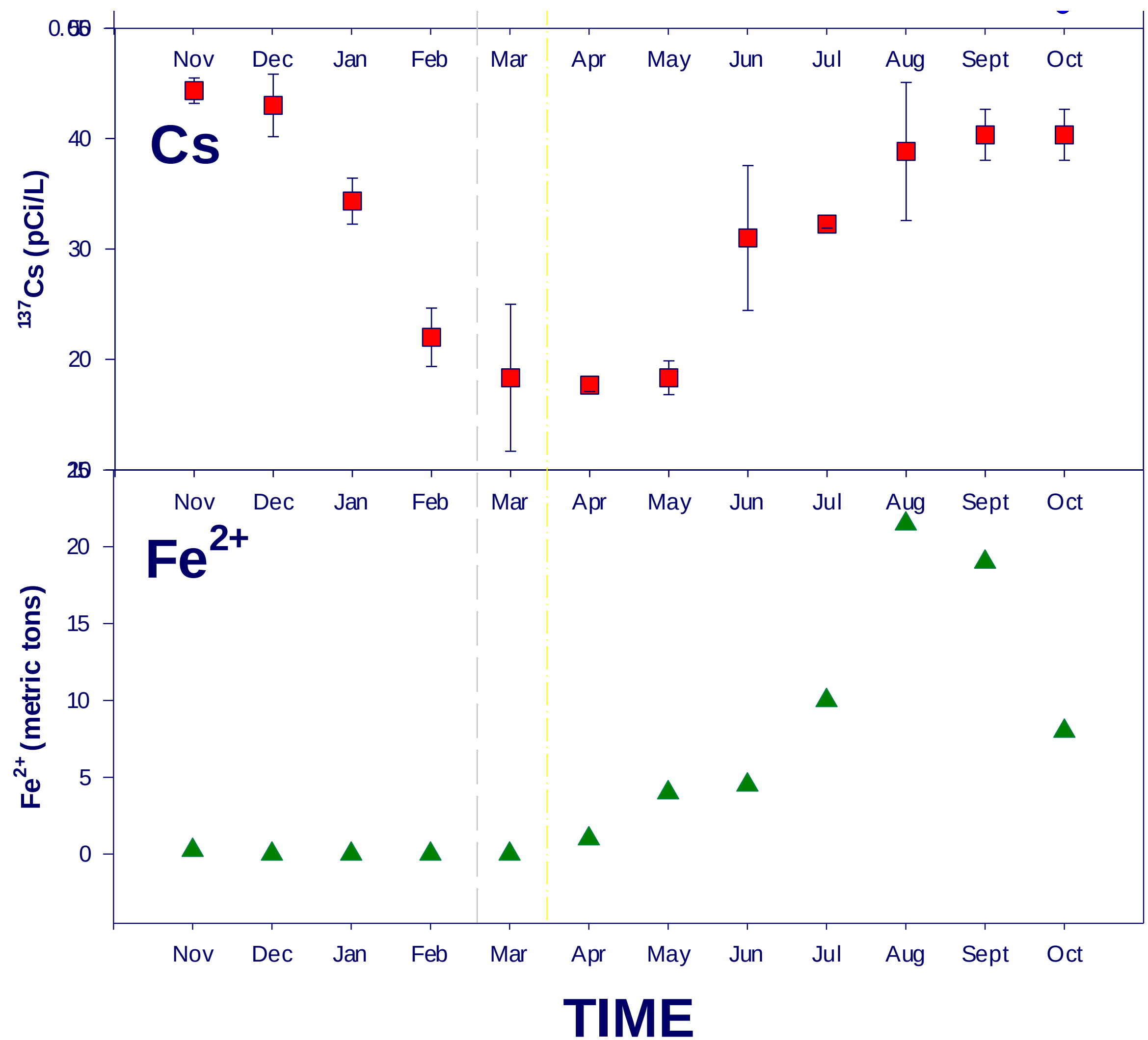
Iron Cycling

Onset of stratification Onset of anoxia



Cesium Cycling

Onset of stratification Onset of anoxia



Cycling - Summary

- The majority of the ^{137}Cs remains partitioned to the lake sediments.
- Measureable quantities of ^{137}Cs are available to cycle seasonally into the water column.
- The cycle is related to development of seasonal anoxia in the lower water column.
- Thermally-driven lake turnover results in seasonal distribution of ^{137}Cs to the upper water column.

Outline

1. Cesium Isotopic Signatures
   a) Discriminating between recent and legacy cesium
   b) Elucidation of source
2. Cesium Cycling in Freshwater Systems
3. **Summary, Observations, Recommendations**

Overall Summary

1. Technical basis exists to discriminate between legacy fission product cesium and new inputs of cesium from the Fukushima accident.
2. Contaminant cesium released to the environment has high potential for partitioning to soils and sediments.
3. Cesium partitioned to soils and sediments may be seasonally bioavailable, depending on natural cycles of the local environment.

Recommendations

1. Account for the redistribution of contaminated sediment (aeolian and/or fluvial) as this can be a critical process affecting remediation.
2. Develop a characterization plan that includes discrimination between recent and legacy fission product cesium.
3. Remediation plans must consider possible affects of seasonal influence on local geochemistry.
4. If fission product cesium is the primary contaminant of concern, soil treatments based on ion exchange should be considered.

Thank you for your time