

Application of $^{129}\text{I}/\text{I}$ Ratios in Groundwater Studies Conducted at Los Alamos National Laboratory, New Mexico

**Patrick Longmire, Ph.D., Michael Dale,
Kim Granzow, and Stephen Yanicak**

**DOE Oversight Bureau,
New Mexico Environment Department
1183 Diamond Drive, Suite B, Los Alamos, NM 87544**



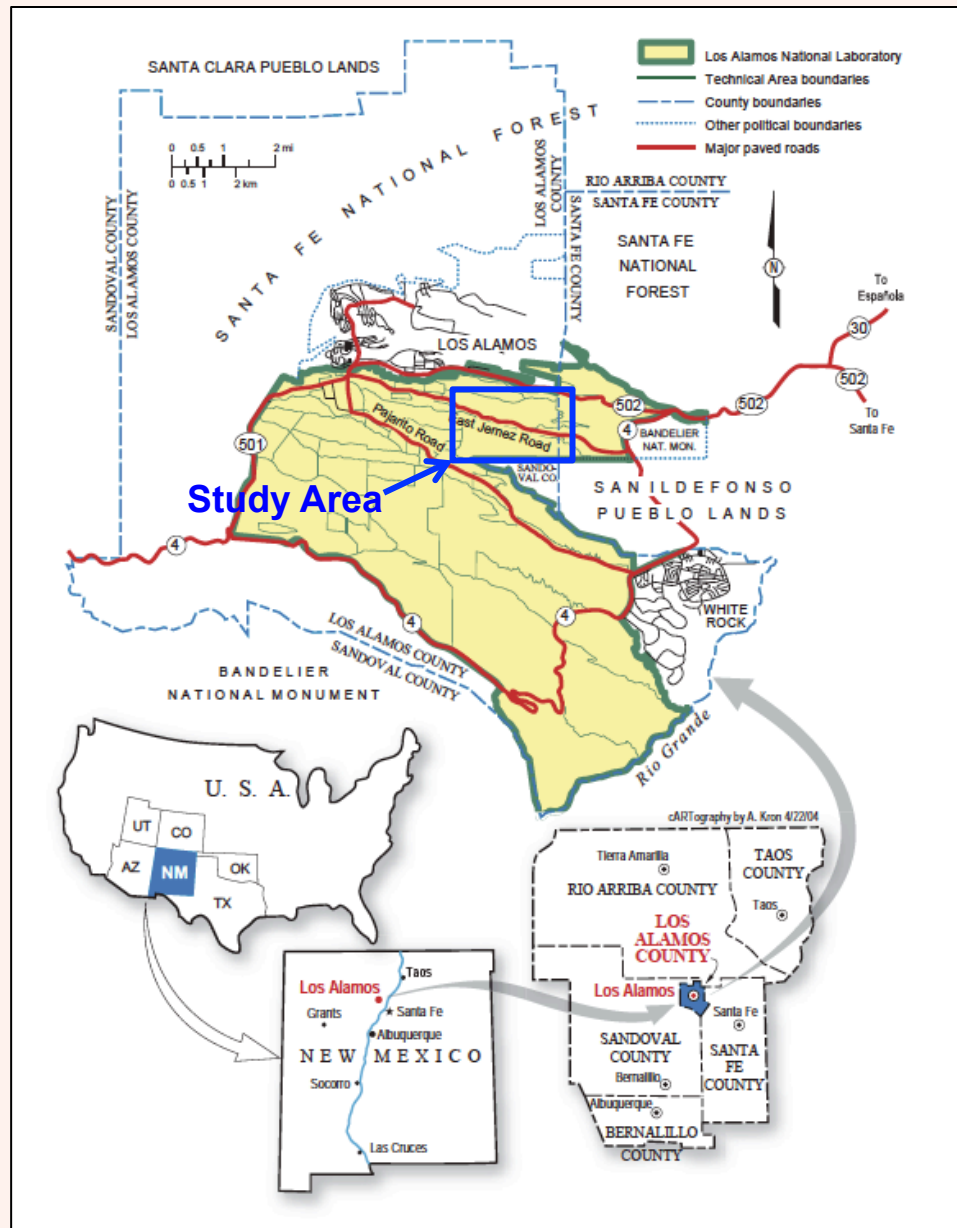
Application of $^{129}\text{I}/\text{I}$ Ratios in Groundwater Studies Conducted at Los Alamos National Laboratory, New Mexico

- **Natural and Anthropogenic Sources of ^{129}I Iodine**
- **Analytical Methods**
- **Hydrogeochemical and Hydrological Setting (groundwater mixing) at LANL**
- **Distribution of ^{129}I and $^{129}\text{I}/\text{I}$ ratios in groundwater**
- **Summary and Conclusions**

Acknowledgment: "This material is based upon work supported by the Department of Energy Office of Environmental Management under Award Number *DE-EM0002420*."

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 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. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof."

Los Alamos National Laboratory, New Mexico



Natural and Anthropogenic Sources of ^{129}I Iodine

Natural sources of ^{129}I include cosmic spallation of xenon and fission of uranium occurring in the subsurface.

Fission of uranium releases ^{129}I to groundwater and the atmosphere from volcanic emissions. Residence times for ^{129}I in the atmosphere and oceans are two weeks and 40,000 years, respectively.

Anthropogenic ^{129}I is a fission product of ^{235}U and ^{239}Pu processing at nuclear facilities. Isotope ratios of $^{129}\text{I}/\text{I}$ increased in some parts of the world during the 1960's resulting from atmospheric nuclear testing. Atmospheric $^{129}\text{I}/\text{I}$ ratios ranged from 10^{-7} to 10^{-4} in the past.

Analytical Methods

¹²⁹Iodine and ³⁶Chlorine

Accelerator mass spectrometry

²³⁹Plutonium and Tritium

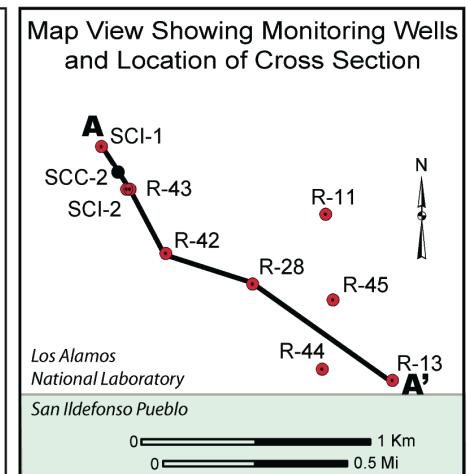
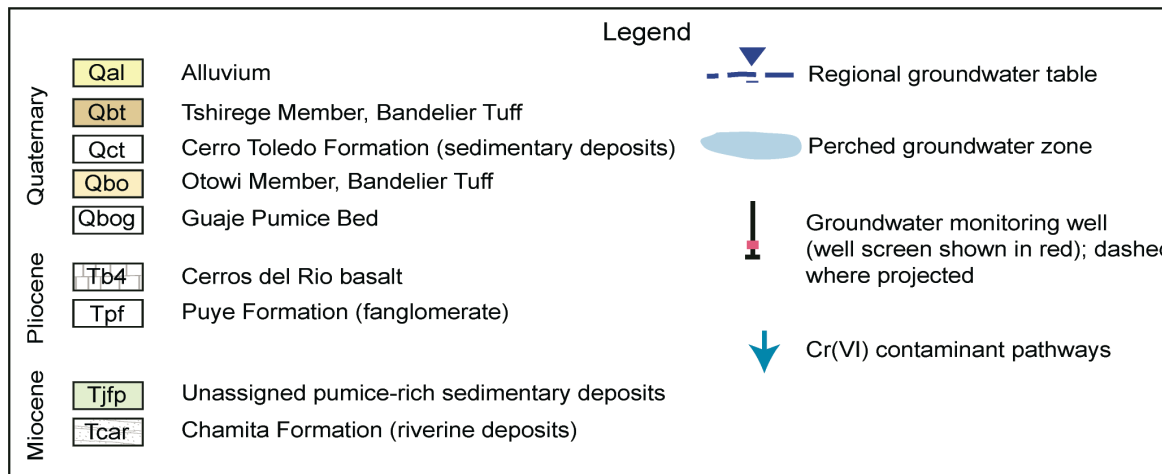
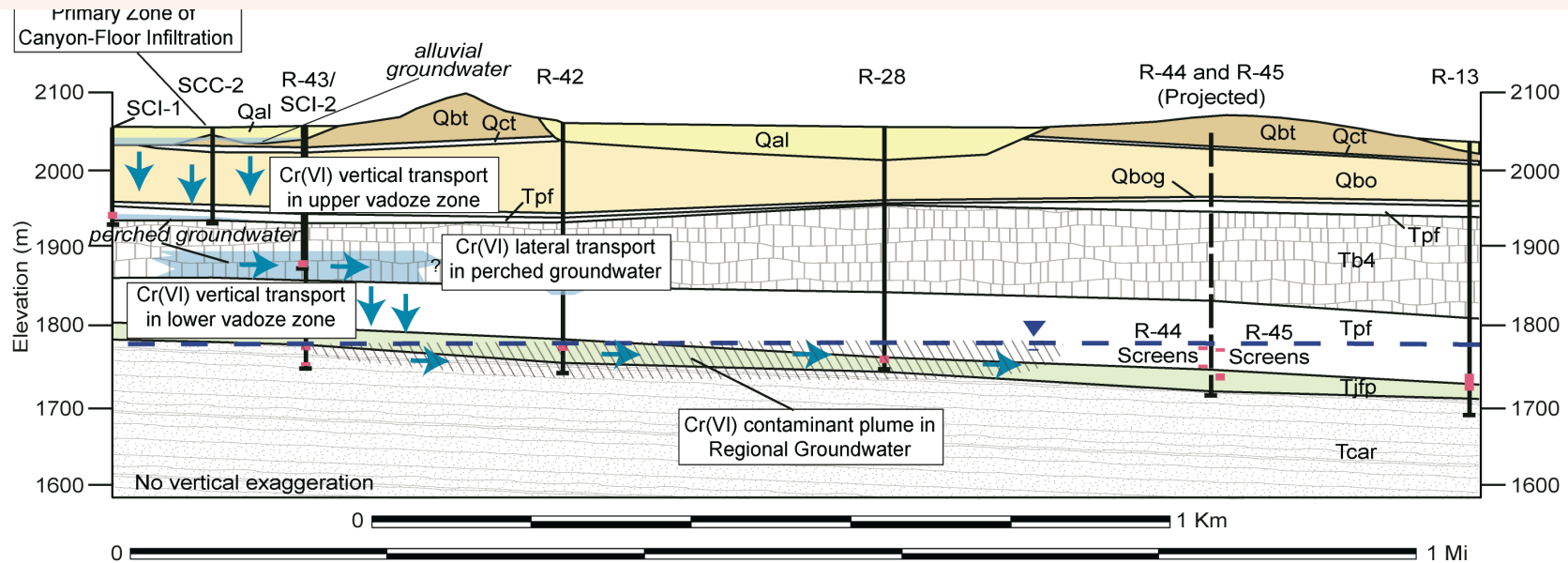
Alpha spectrometry

Electrolytic enrichment and liquid scintillation

Oxyanions

**Liquid chromatography/mass spectrometry-
mass/spectrometry**

Conceptual Model of Groundwater Movement Through the Vadose Zone to the Regional Aquifer, Los Alamos National Laboratory, New Mexico



Source: LANL 2012

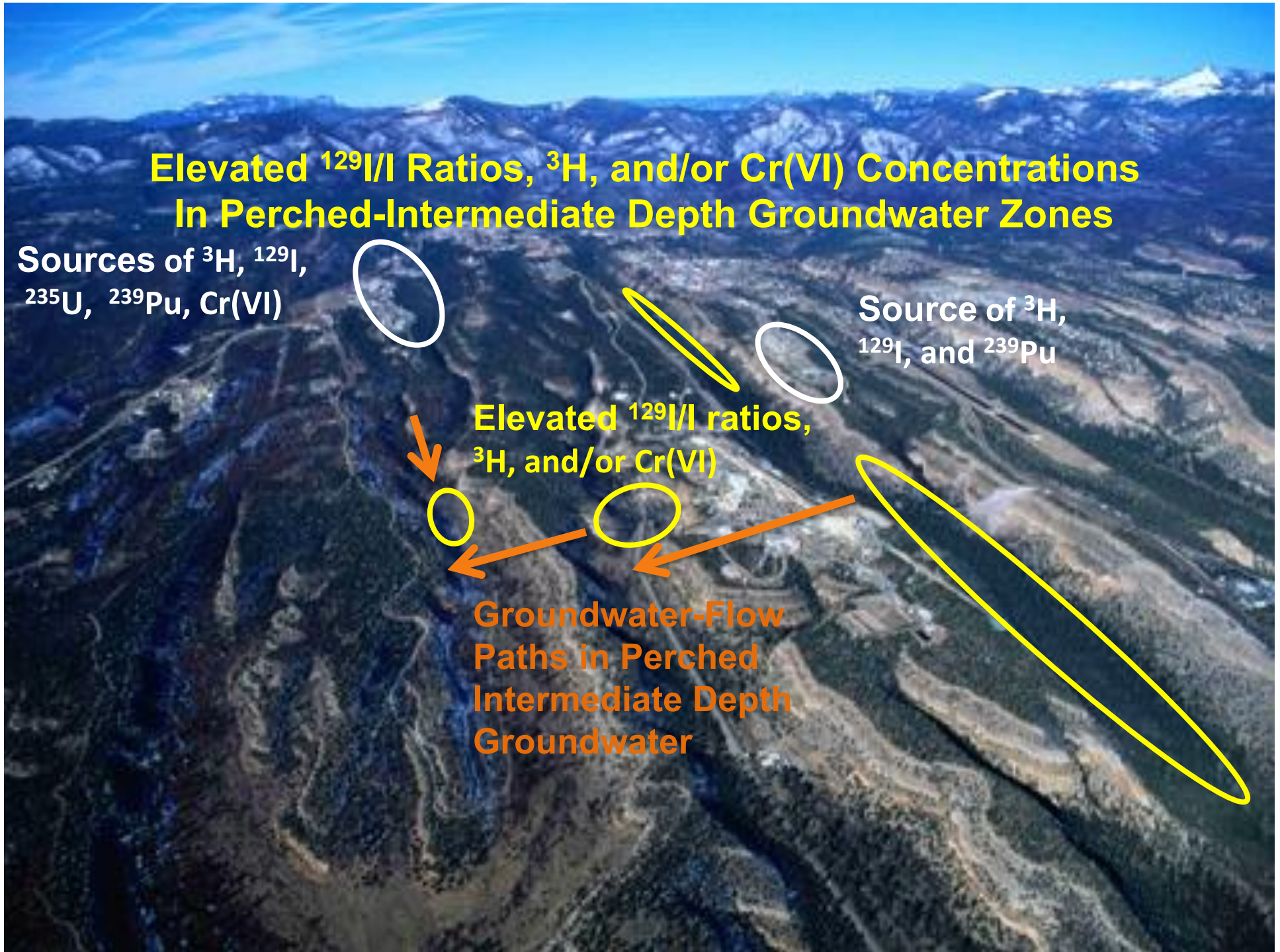
Elevated $^{129}\text{I}/\text{I}$ Ratios, ^3H , and/or Cr(VI) Concentrations In Perched-Intermediate Depth Groundwater Zones

Sources of ^3H , ^{129}I ,
 ^{235}U , ^{239}Pu , Cr(VI)

Source of ^3H ,
 ^{129}I , and ^{239}Pu

Elevated $^{129}\text{I}/\text{I}$ ratios,
 ^3H , and/or Cr(VI)

Groundwater-Flow
Paths in Perched
Intermediate Depth
Groundwater



Elevated $^{129}\text{I}/\text{I}$ Ratios, ^3H , and Cr(VI) Concentrations In the Regional Aquifer

Sources of Cr(VI) ,
 ^3H , ^{129}I , and ^{239}Pu

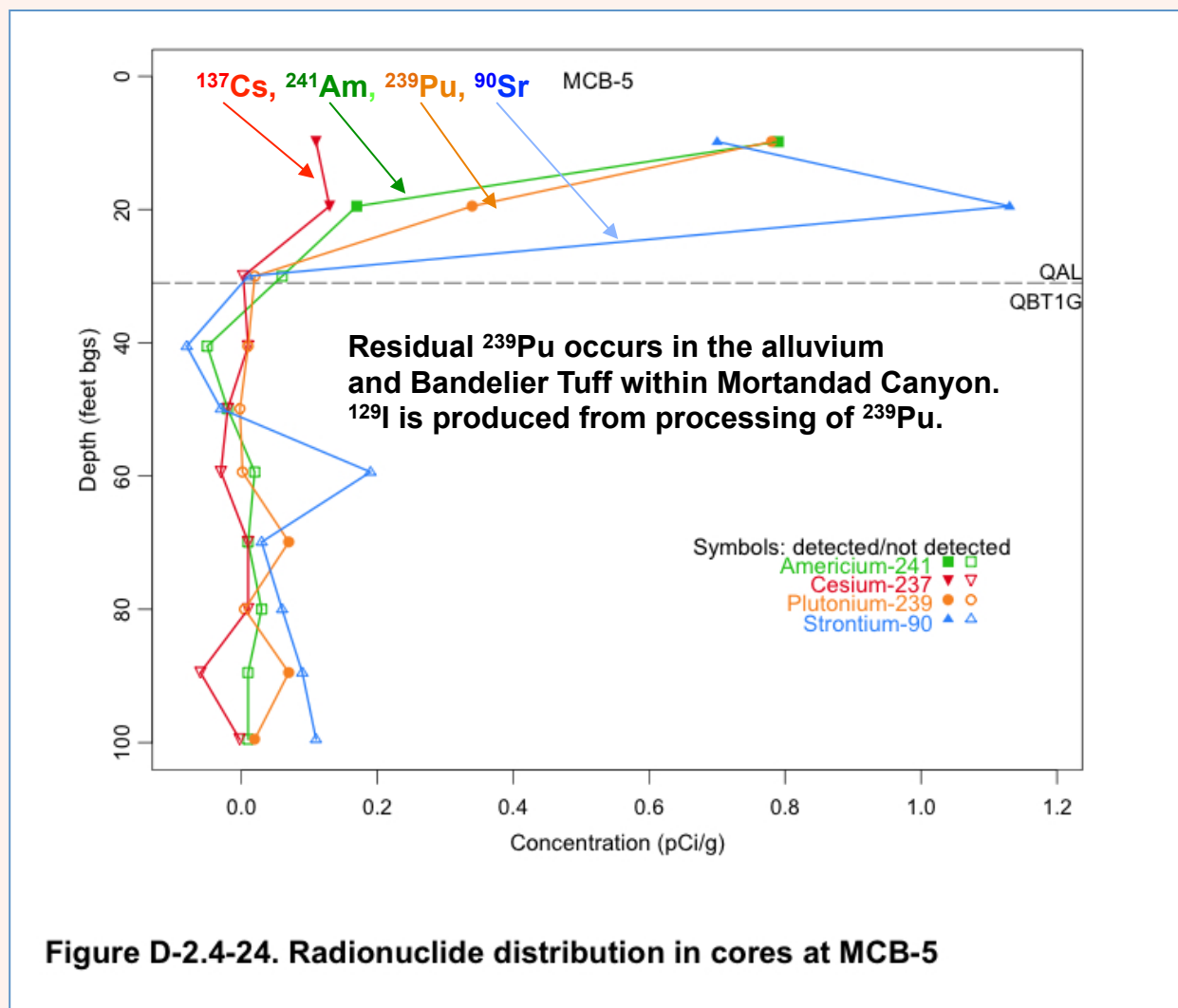
Major Source of Cr(VI)

Source of ^3H ,
 ^{129}I , and ^{239}Pu

Groundwater-
Flow Paths in the
Upper Portion of the
Regional Aquifer



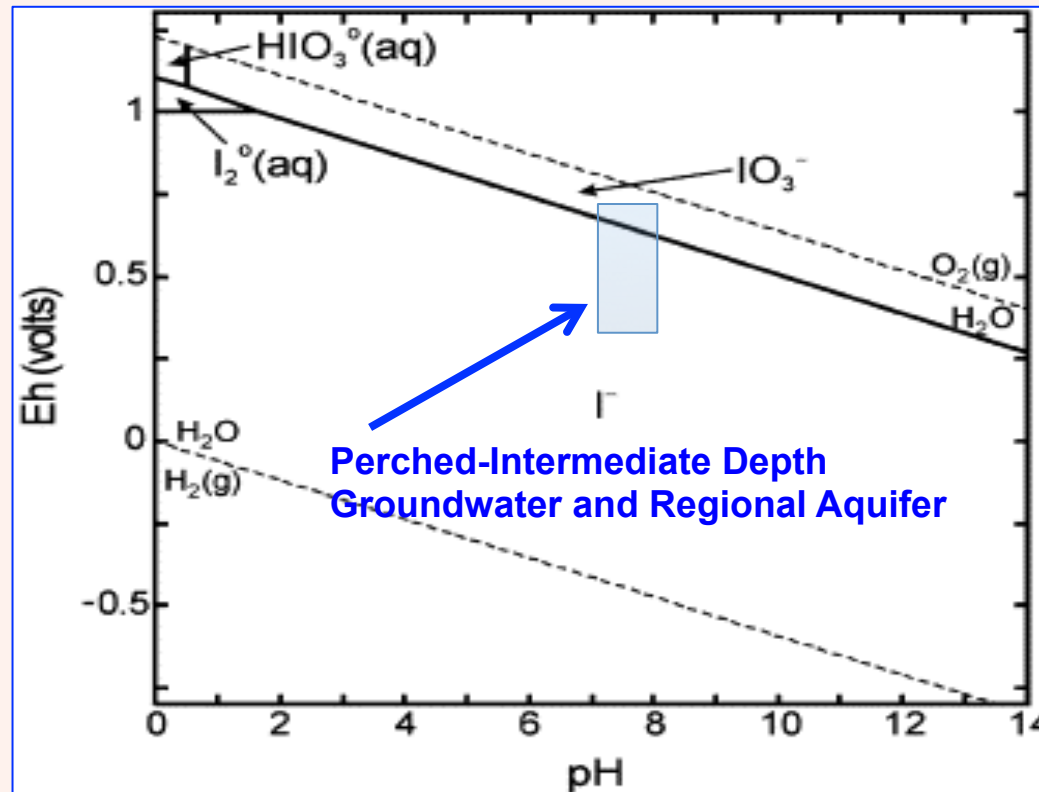
Distributions of Radionuclides in Corehole MCB-5, Mortandad Canyon, New Mexico



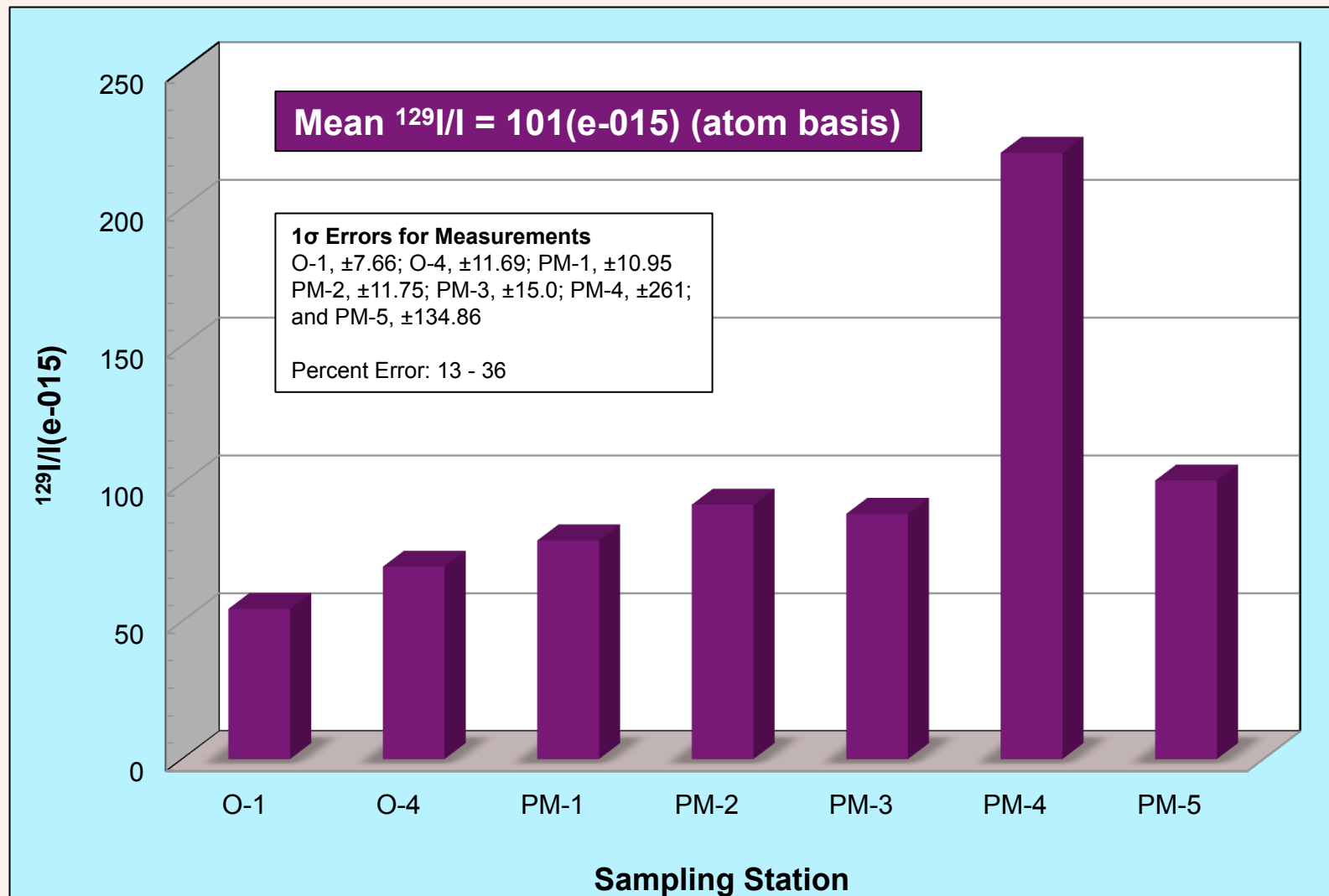
Source: Los Alamos National Laboratory, 2006, Mortandad Canyon Investigation Report, Environmental Restoration Project: Los Alamos National Laboratory, LA-UR-06-6752.

Eh-pH Diagram for Iodine at 25°C and 1 Bar

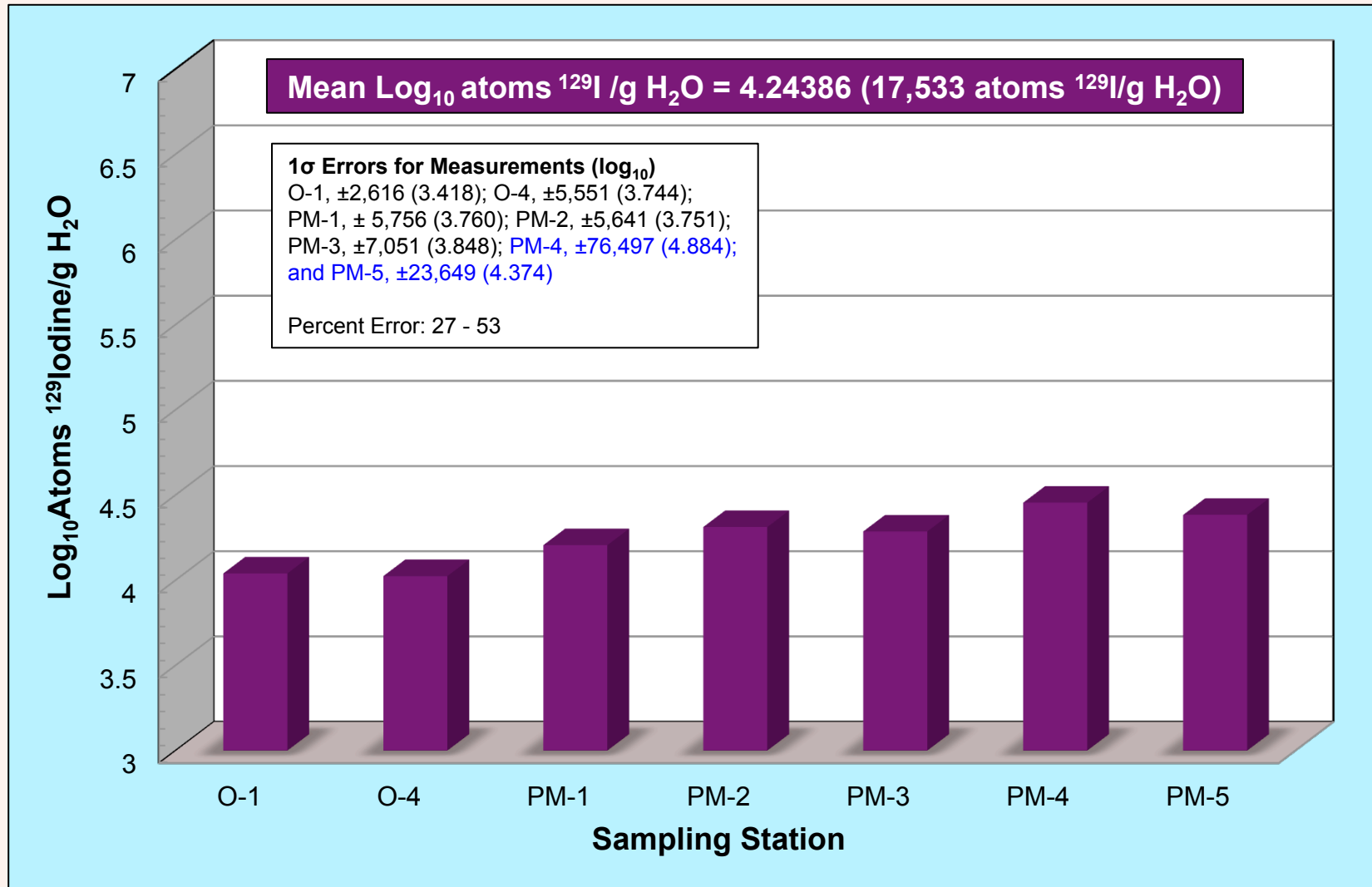
(Total dissolved I concentration = 10^{-8} mol/L. Source: Um et al., 2004)



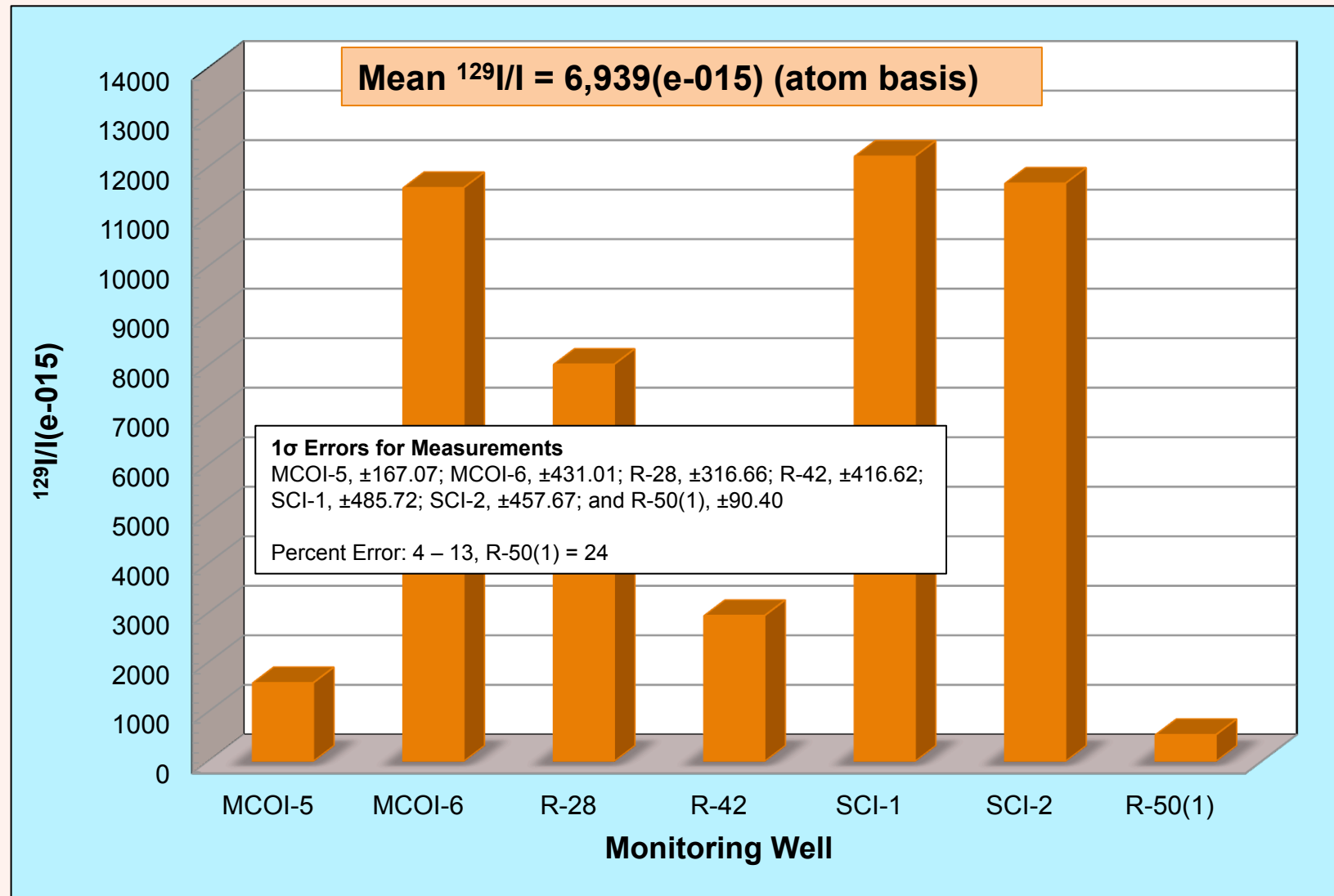
$^{129}\text{I}/\text{I}$ Ratios in Los Alamos County Supply Wells, New Mexico



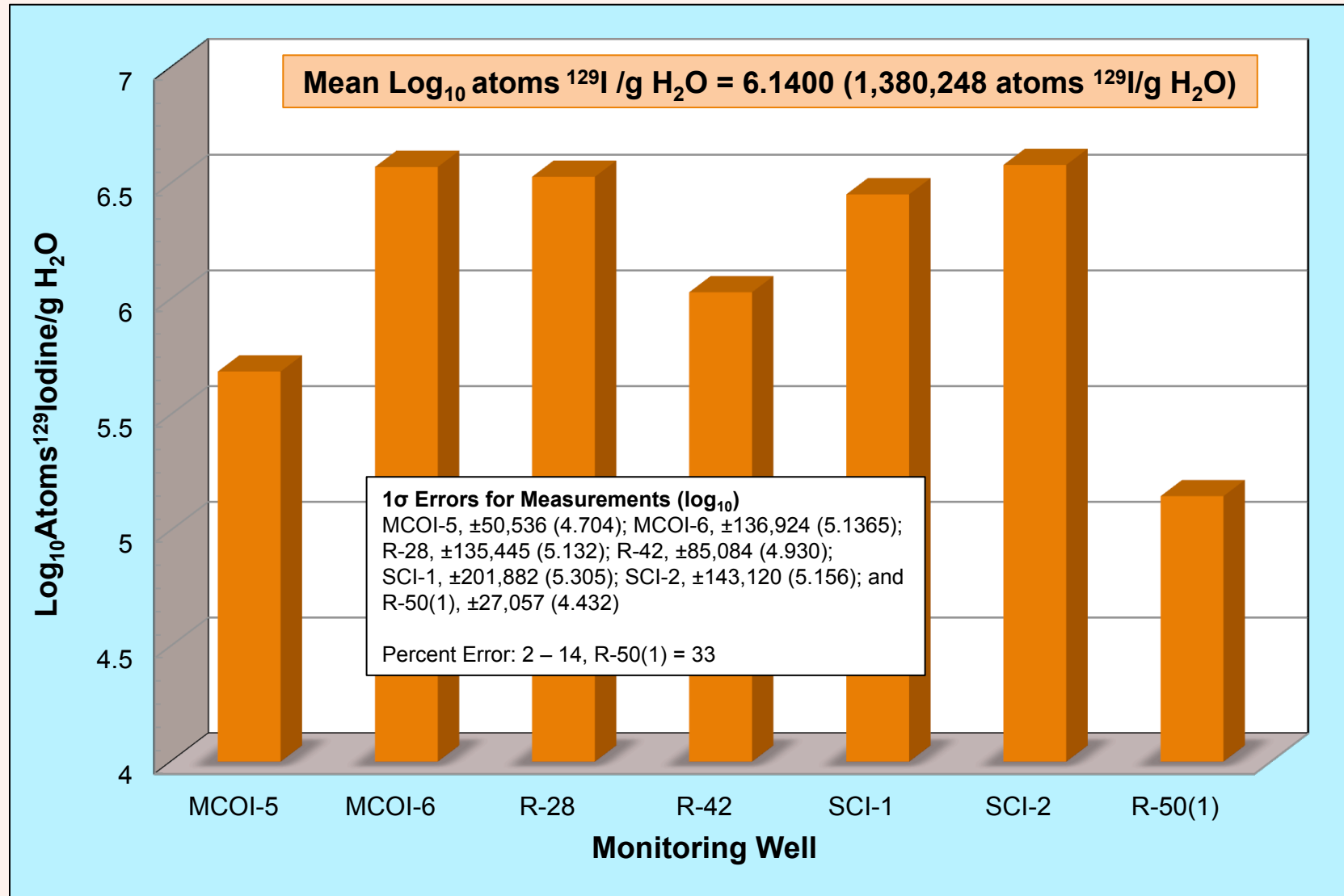
Atoms ^{129}I /g Water in Supply Wells, Los Alamos County, New Mexico



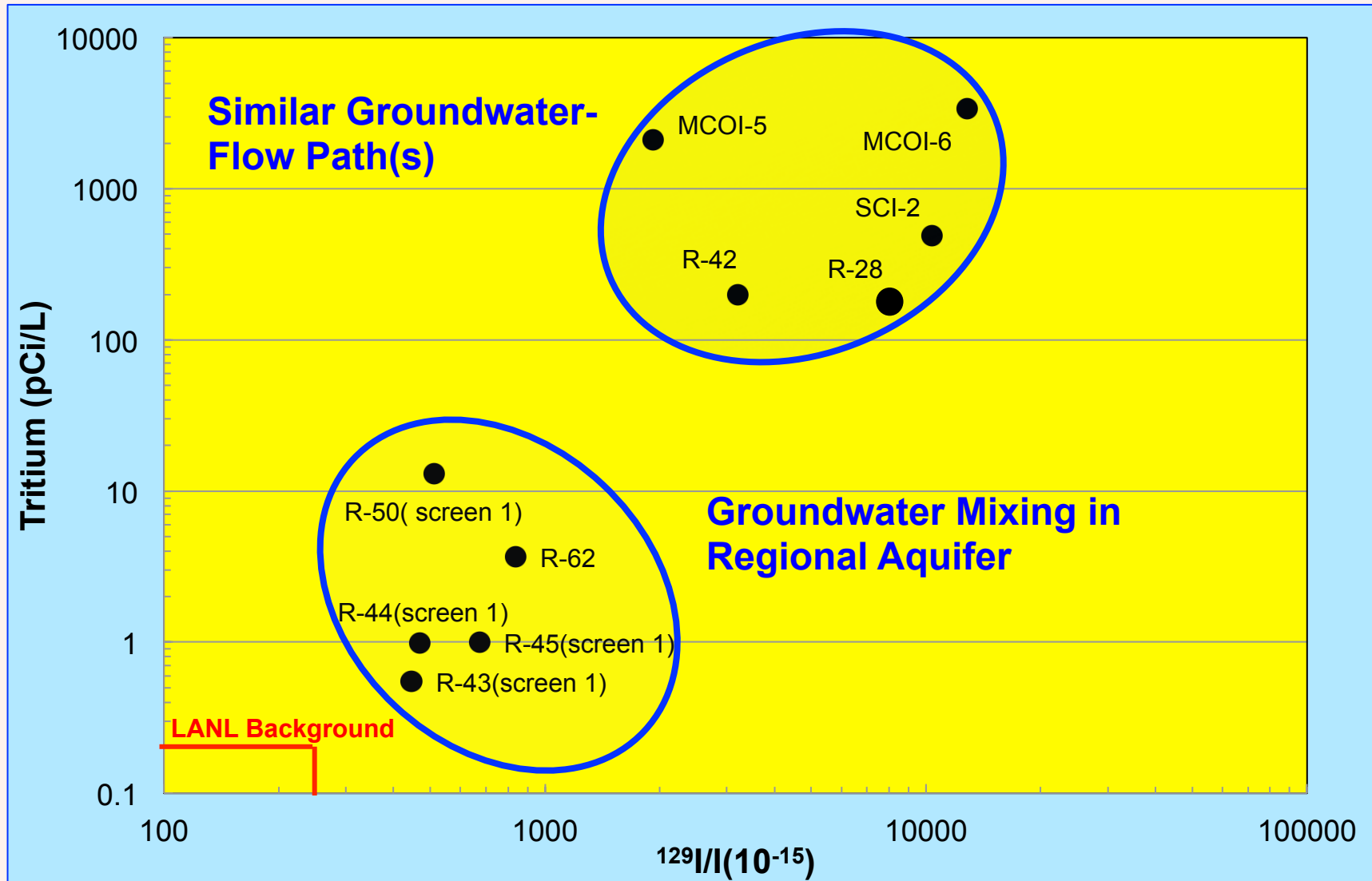
Average $^{129}\text{I}/\text{I}$ Ratios in Selected Monitoring Wells Downgradient From Sources of ^{129}I Iodine, Los Alamos National Laboratory, NM



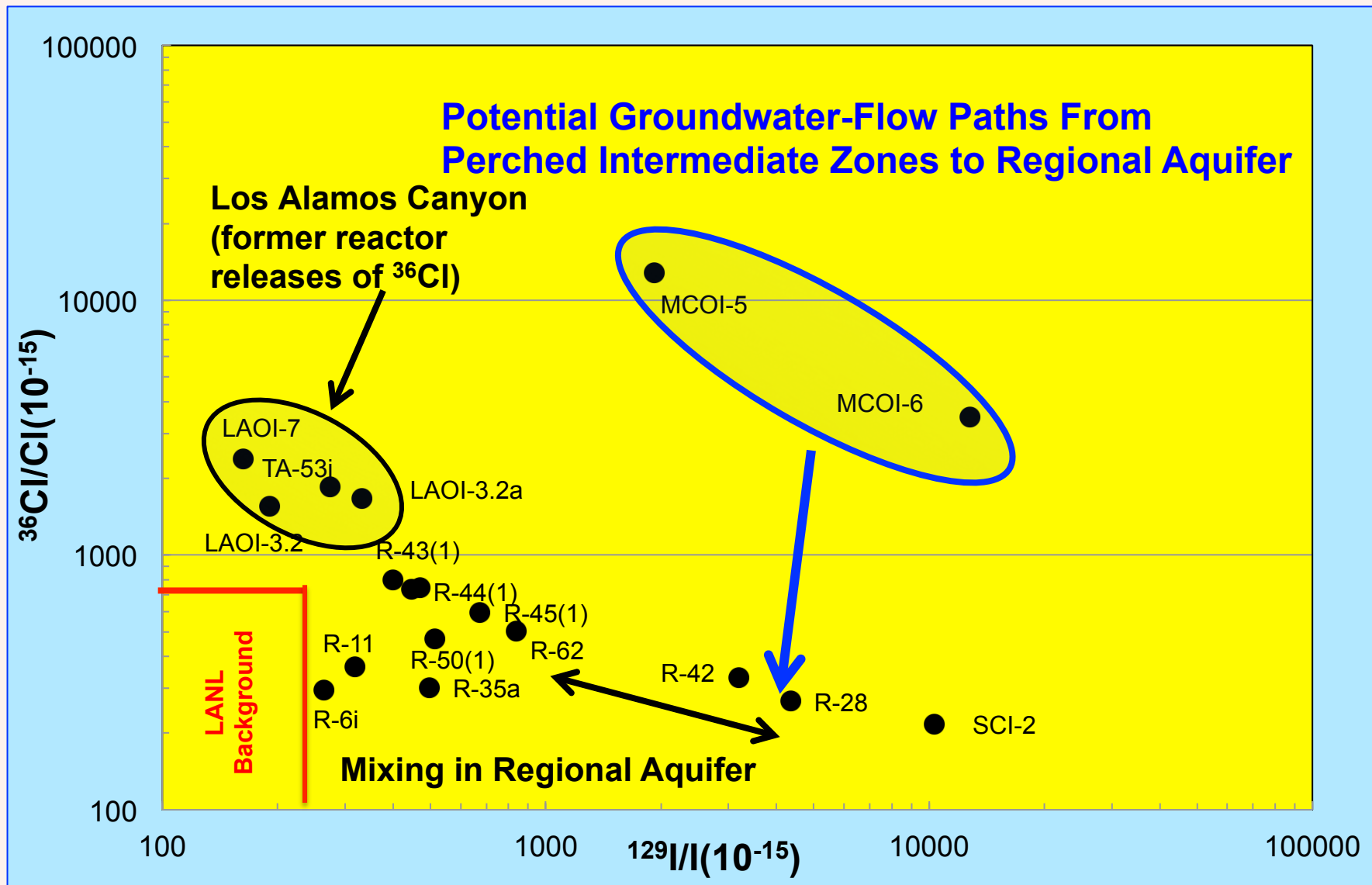
Atoms ^{129}I /g Water in Selected Monitoring Wells Downgradient From Sources of ^{129}I Iodine, Los Alamos National Laboratory, NM



$^{129}\text{I}/\text{I}$ Ratios Versus Tritium in Groundwater, Los Alamos National Laboratory, New Mexico



$^{129}\text{I}/\text{I}$ Ratios Versus $^{36}\text{Cl}/\text{Cl}$ Ratios in Groundwater, Los Alamos National Laboratory, New Mexico



Summary and Conclusions

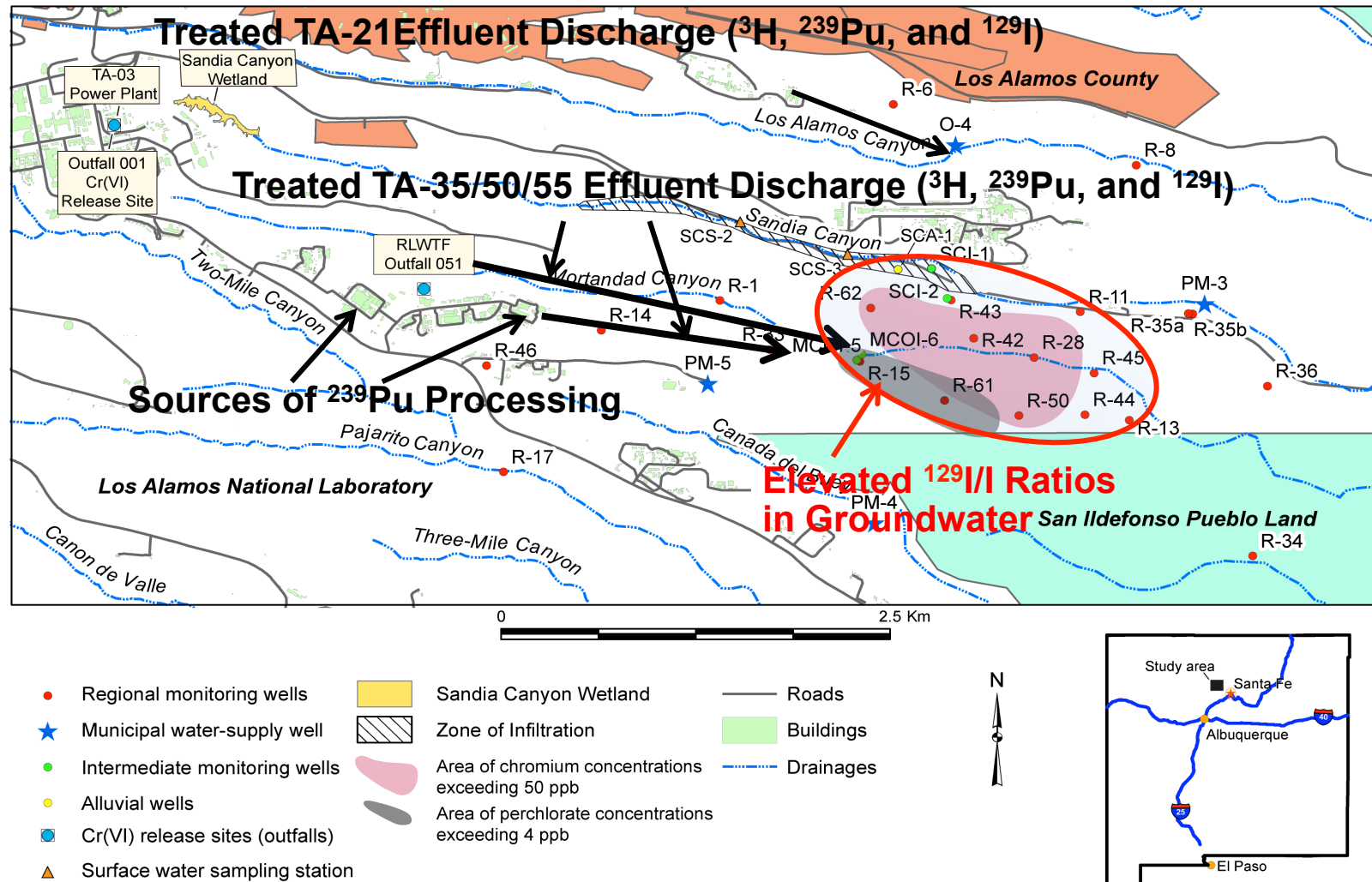
- The radioisotope ^{129}I ($T_{1/2} = 15.7$ Myrs) derived from ^{235}U and ^{239}Pu processing at Los Alamos National Laboratory is locally detected in groundwater above background ^{129}I activities.
- This isotope provides a unique tracer for groundwater investigations conducted at LANL that helps to identify source releases linked to groundwater-flow paths in aquifers.
- Aquifer systems are subject to binary and ternary mixing of natural- and industrial-derived waters containing iodate, chromate, and other chemicals.
- Local background ratios of $^{129}\text{I}/\text{I}$ vary from 54×10^{-15} to 220×10^{-15} in the regional aquifer (supply wells).

Summary and Conclusions

- Anthropogenic ratios of $^{129}\text{I}/\text{I}$ range from $1,252 \times 10^{-15}$ to $17,367 \times 10^{-15}$ within perched-intermediate depth groundwater in Mortandad Canyon.
- Anthropogenic ratios of $^{129}\text{I}/\text{I}$ range from $2,690 \times 10^{-15}$ to $11,688 \times 10^{-15}$ within the regional aquifer in Mortandad Canyon (centroid of chromium plume).
- Variability in $^{129}\text{I}/\text{I} \times 10^{-15}$ ratios and concentrations of anthropogenic iodate is controlled by non-uniform source releases of this isotope and iodate over time and non-uniform mixing (ternary) of groundwater in different aquifers.

Supplemental Slides

Plume Map of Total Dissolved Chromium and Elevated Above Background $^{129}\text{I}/\text{I}$ Ratios within the Regional Aquifer, Los Alamos National Laboratory

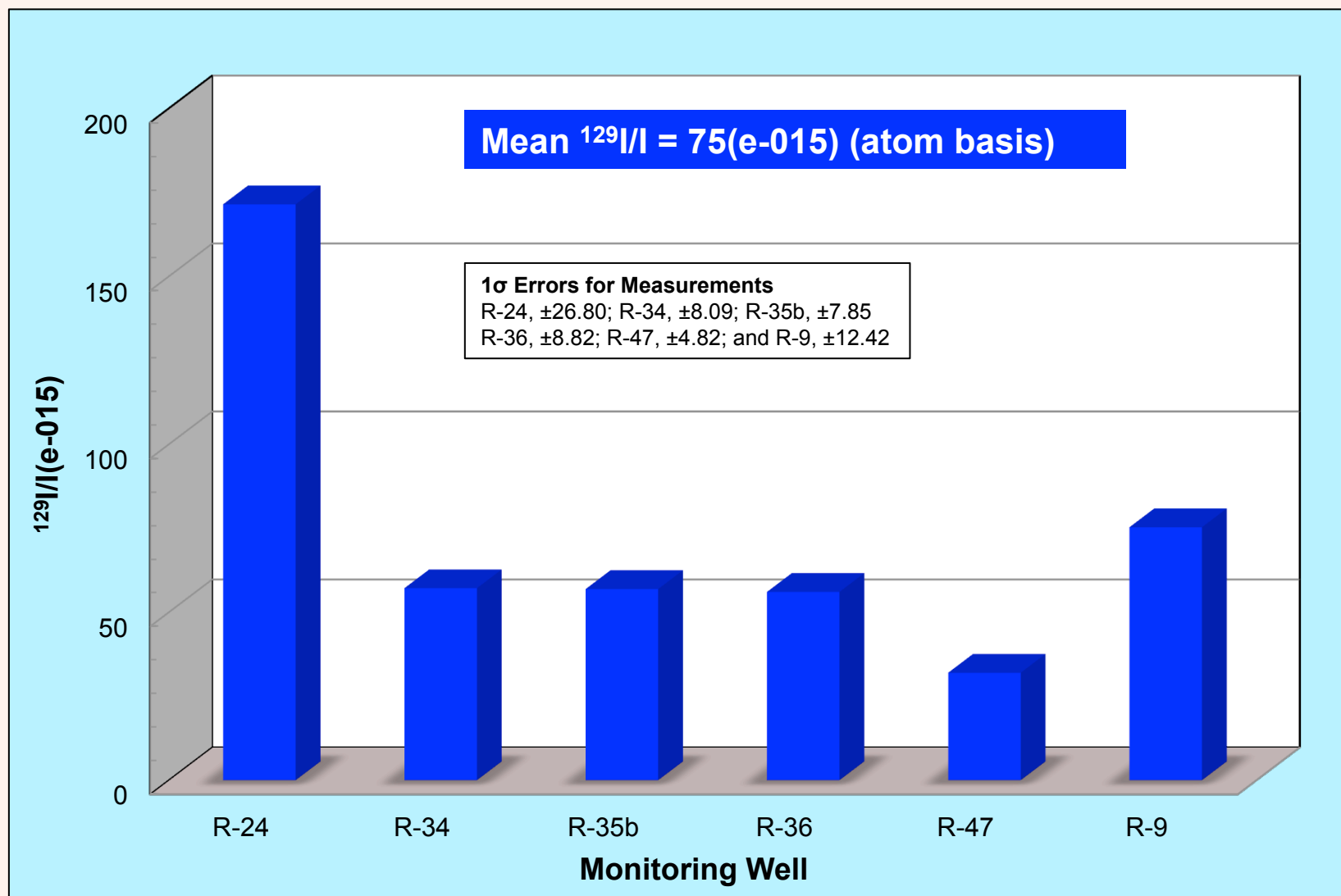


¹²⁹Iodine Yield, Percent Per Fission

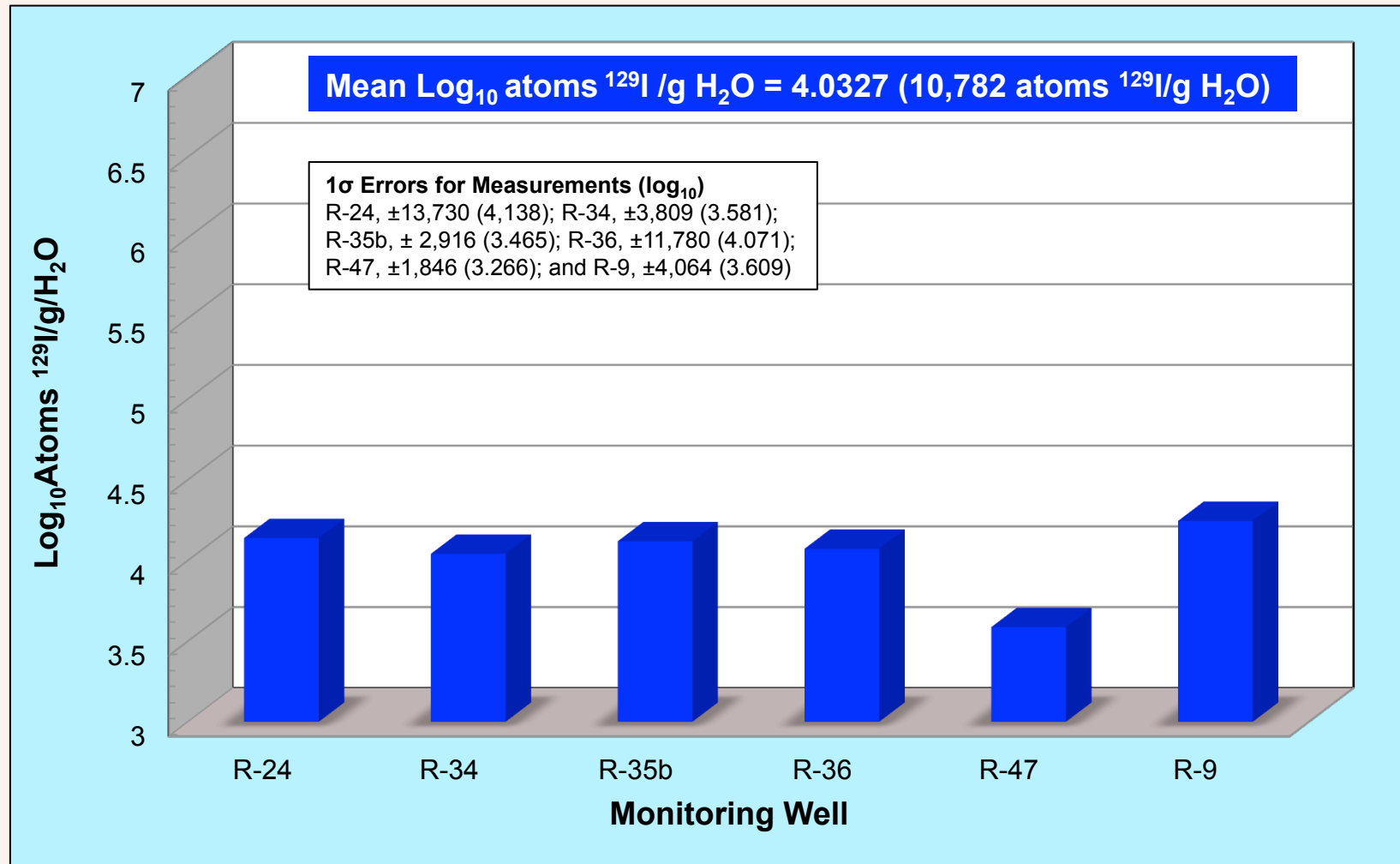
(<http://www-nds.iaea.org/sgnucdat/c3.htm>)

Isotope	Thermal	Fast
²³² Th	not fissile	0.431 ± 0.089
²³³ U	1.63 ± 0.26	1.73 ± 0.24
²³⁵ U	0.706 ± 0.032	1.03 ± 0.26
²³⁸ U	not fissile	0.622 ± 0.034
²³⁹ Pu	1.407 ± 0.086	1.31 ± 0.13
²⁴¹ Pu	1.428 ± 0.36	1.67 ± 0.36

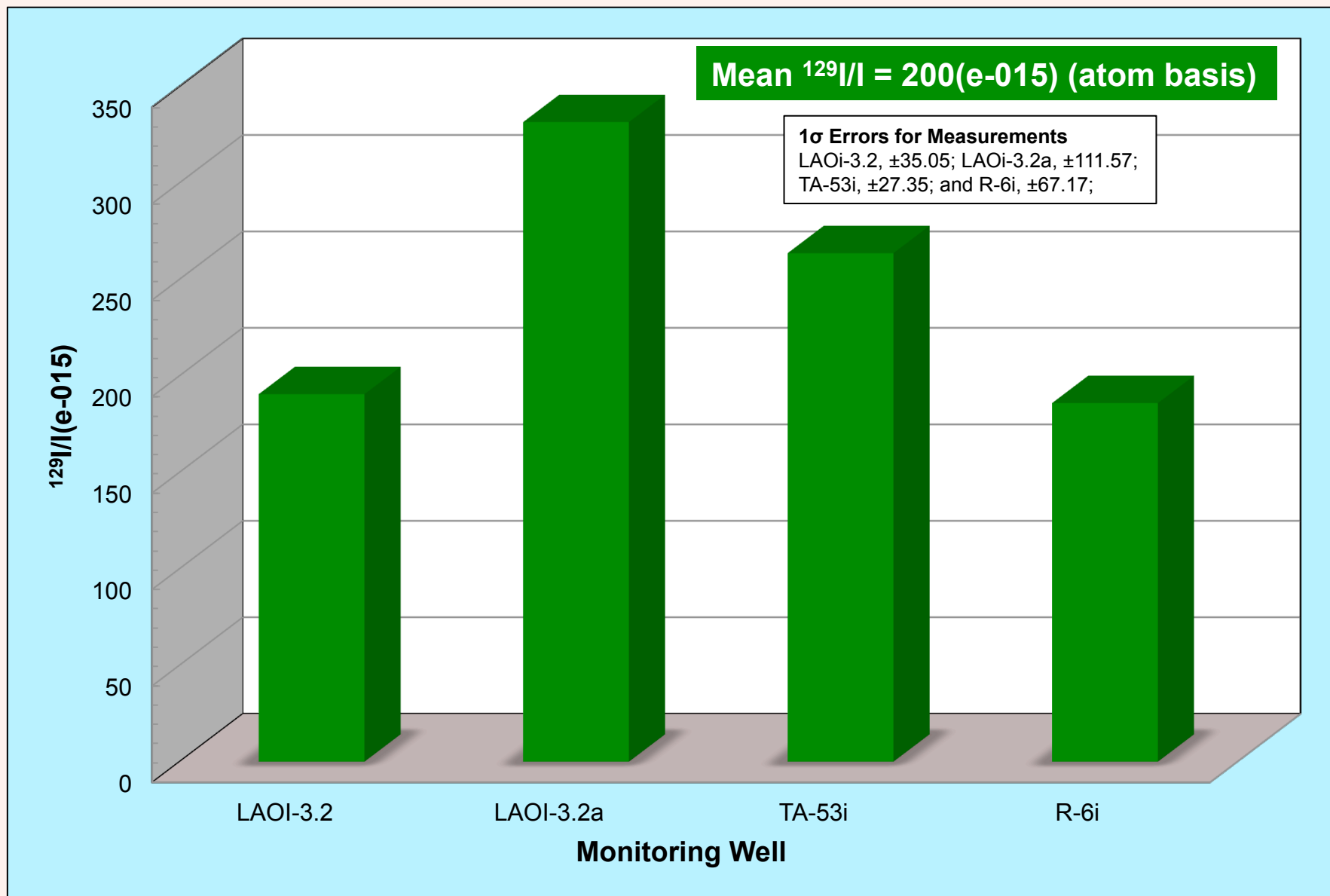
$^{129}\text{I}/\text{I}$ Ratios in Upper Portion of the Regional Aquifer, Los Alamos National Laboratory, New Mexico



Atoms ^{129}I /g Water in the Upper Portion of the Regional Aquifer, Los Alamos National Laboratory, New Mexico



$^{129}\text{I}/\text{I}$ Ratios in Selected Monitoring Wells Near or in Los Alamos Canyon, Los Alamos National Laboratory, New Mexico



Atoms ^{129}I /g Water in Selected Monitoring Wells Near or in Los Alamos Canyon, Los Alamos National Laboratory, New Mexico

