

Occurrence of Radium-224, Radium-226, and Radium-228 in Aquifers Used Primarily for Drinking Water in the United States: Retrospective Survey of Results from 1987 to 2004

Zoltan Szabo, Eric Jacobsen,
Jeffrey M Fischer, Thomas F Kraemer,
and Vincent T dePaul

U.S. Geological Survey

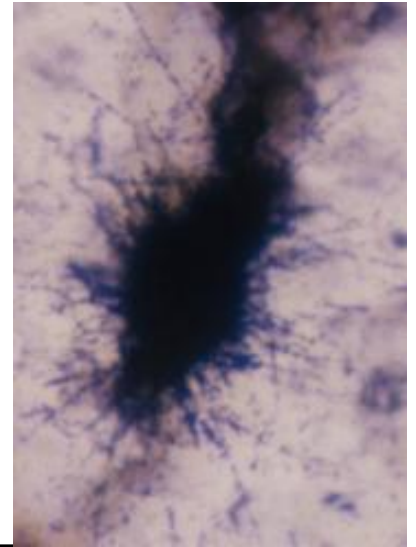


OBJECTIVES

- Where does Ra occur?
- Are there geochemical or geological features on National and local scales that help evaluate occurrence of Ra?
- Is measurement of gross alpha helpful?

Radionuclide Health Effects

MCLs: About 1 in 20,000 risk
Carcinogens (MCLG = 0)
Class 'A' = Humans
Linear dose, No threshold



Uranium

Bone cancer (50%)

Kidney toxicity (50%)

Radium

Bone Cancer (228>226>224)

Sinus Cancer (226)

Bone fracturing (224, 226)

Radon

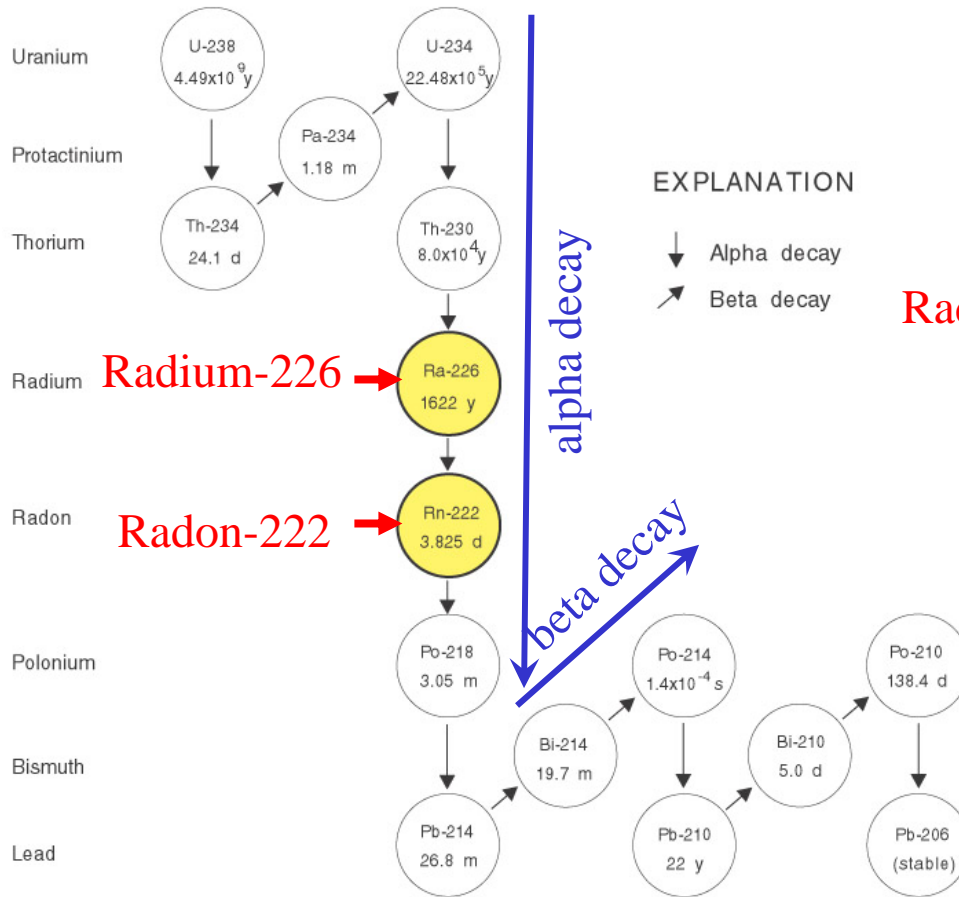
Lung cancer (>80%)

Stomach cancer (<20%)

Ionizing Radiation Damage = Long-term cancer risk
Tissue Retention Controls Dose (Time = Dose, Linear)

Radium Decay Series

Uranium 238



Thorium 232

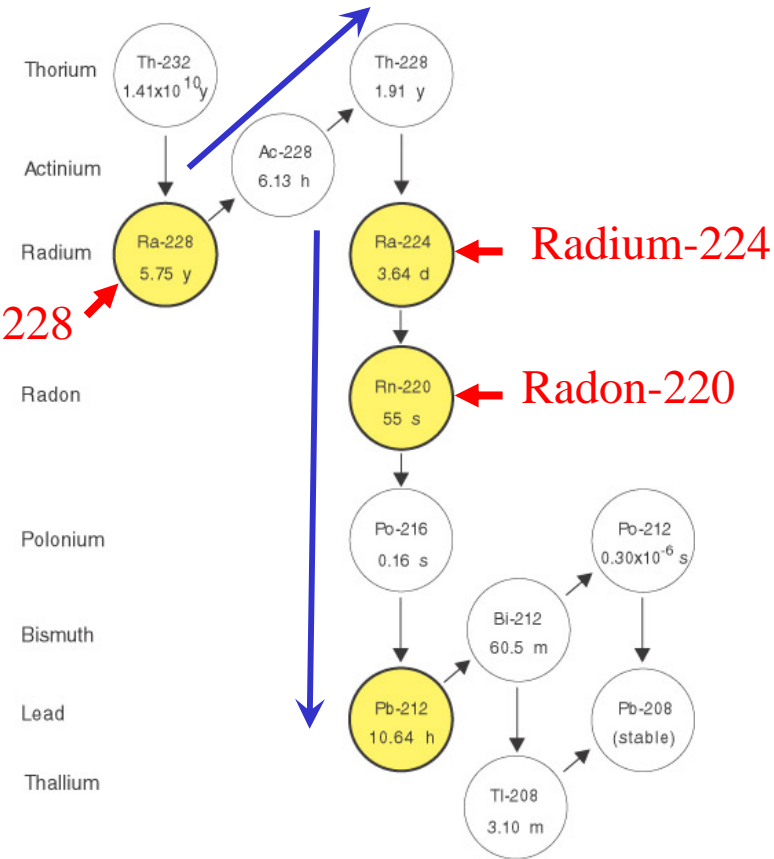
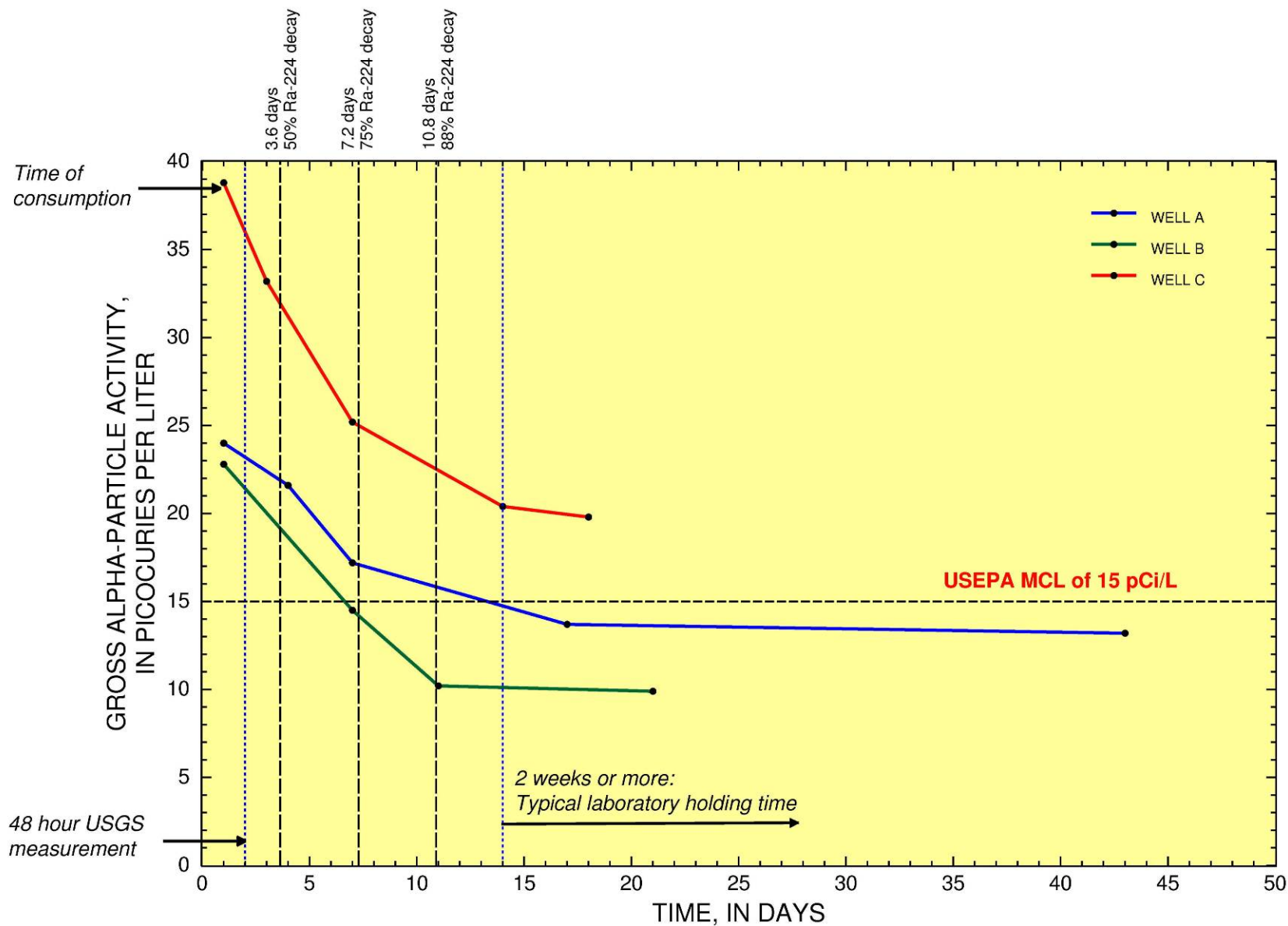


Figure 1. Diagram showing uranium-238 and thorium-232 radioactive decay series. (Radionuclides of interest in this study are shaded). [Times shown are half-lives: y, years; d, days; h, hours; m, minutes; s, seconds] (From Hall and others, 1985)

Gross alpha ≡ many individual isotope contributions!
Gross beta ≡ many individual isotope contributions!



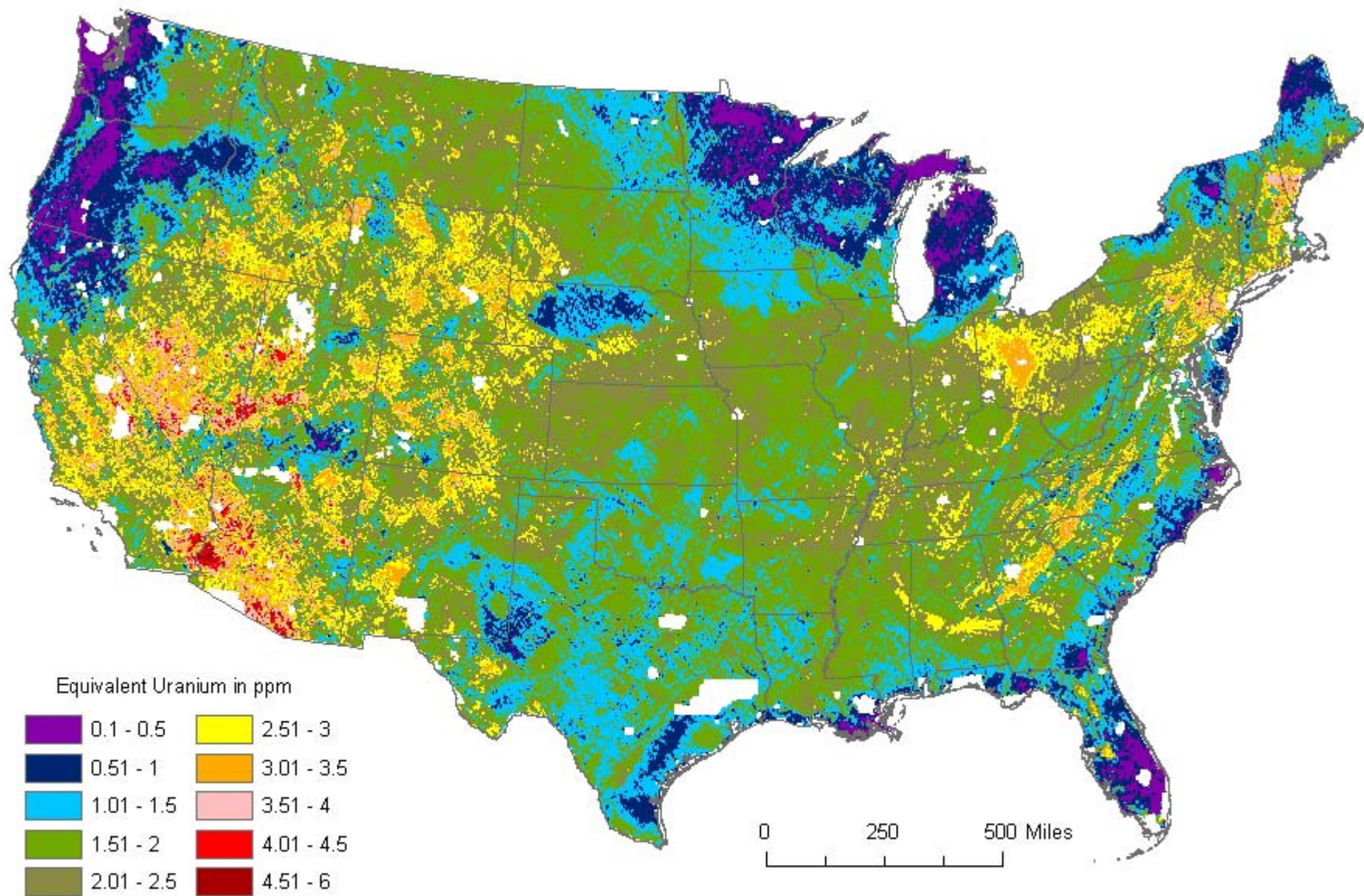
Decline of gross alpha-particle activity through time in samples from selected public-supply wells, southern New Jersey, 1997.

Drinking Water MCLs

- Ra-226 + Ra-228 5 pCi/L
- Gross alpha 15 pCi/L
- Gross beta 4 millirems/yr
- Uranium 30 ug/L

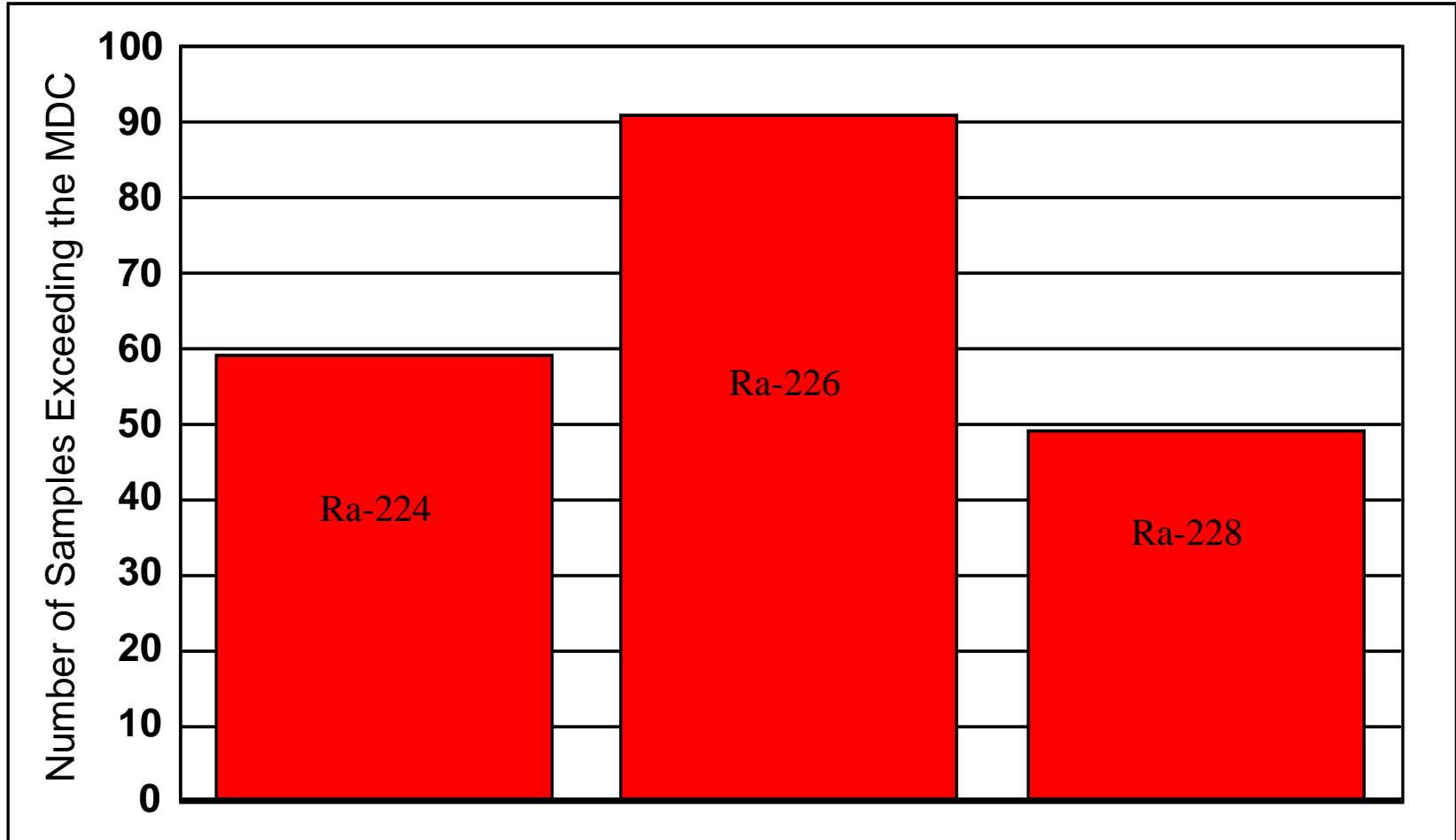
EPA recommends samples be collected and analyzed within 48 hours

Where does Ra occur?



Gundersen and Szabo, 1995

Percent Detection



Distribution of NAWQA Radium Sampling

**114 sites
sampled in 2004**

**>200 sites
sampled in 1998-9**

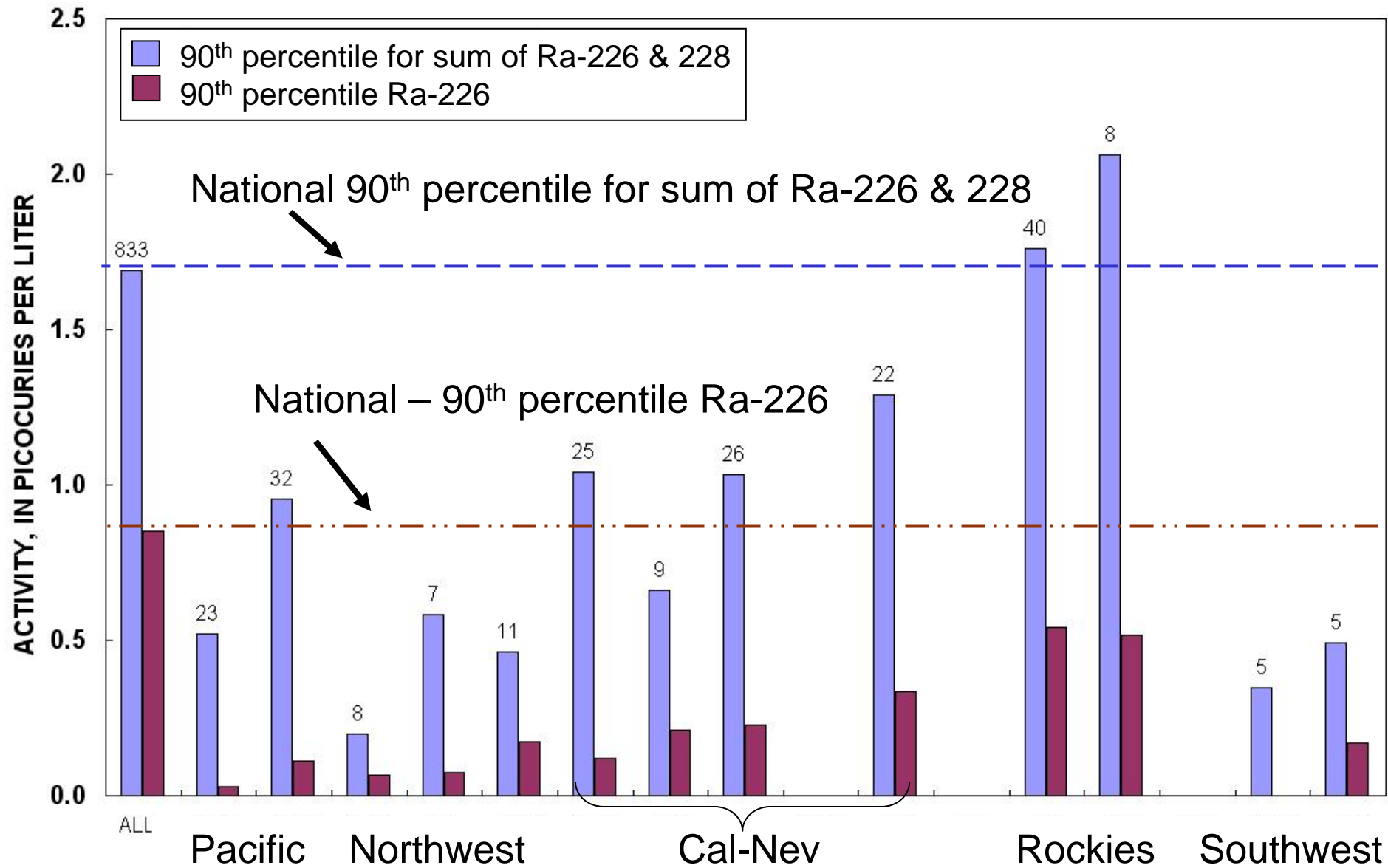
**Other sites
sampled in 1996 -2001**

Concentration, in **Picocuries/liter**

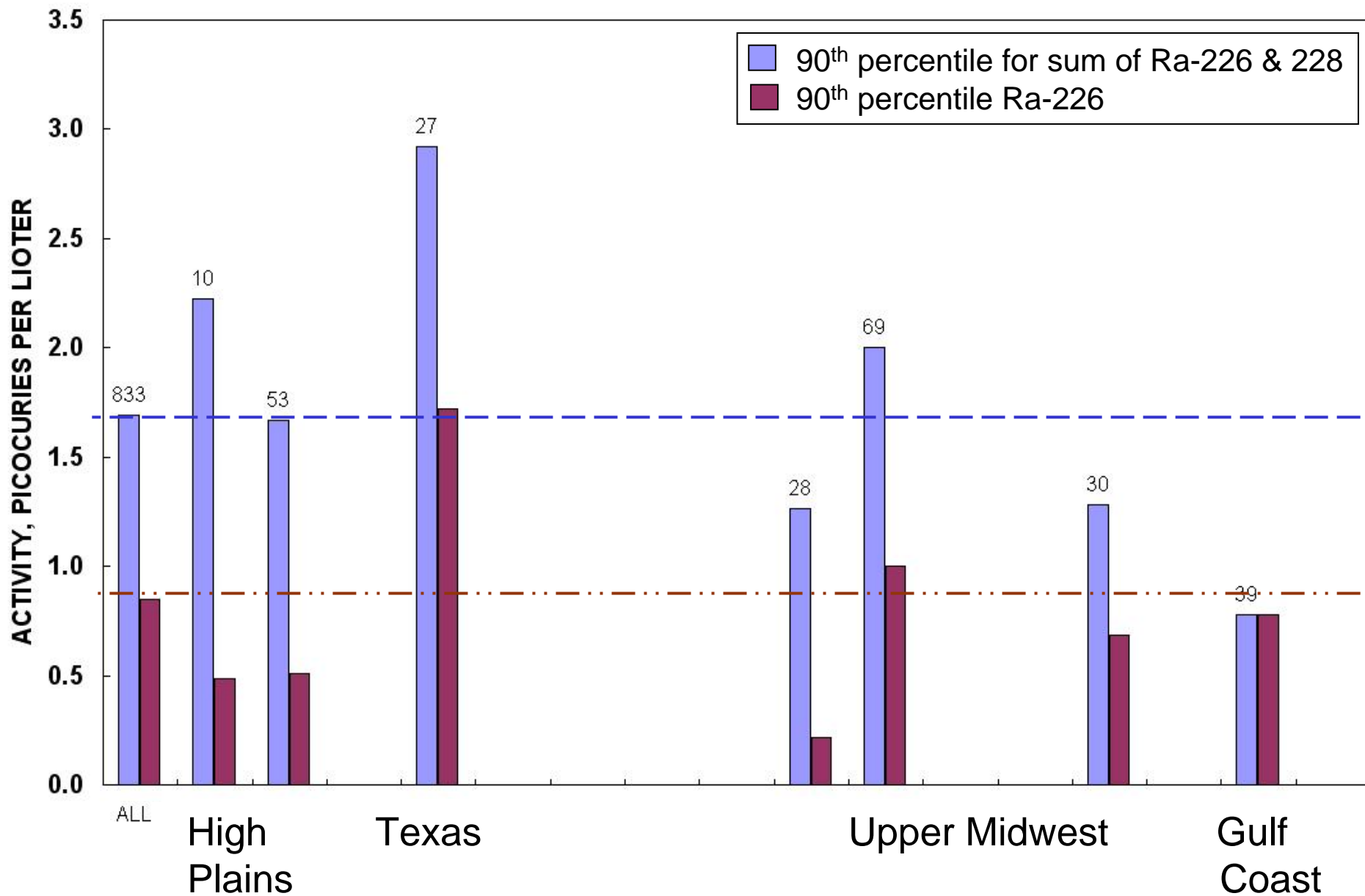
- LE 0.33
- GT 0.33 AND LE 0.57
- GT 0.57 AND LE 0.98
- GT 0.98 AND LE 1.04
- GT 1.04

Over 800 total sites, about 450 domestic wells

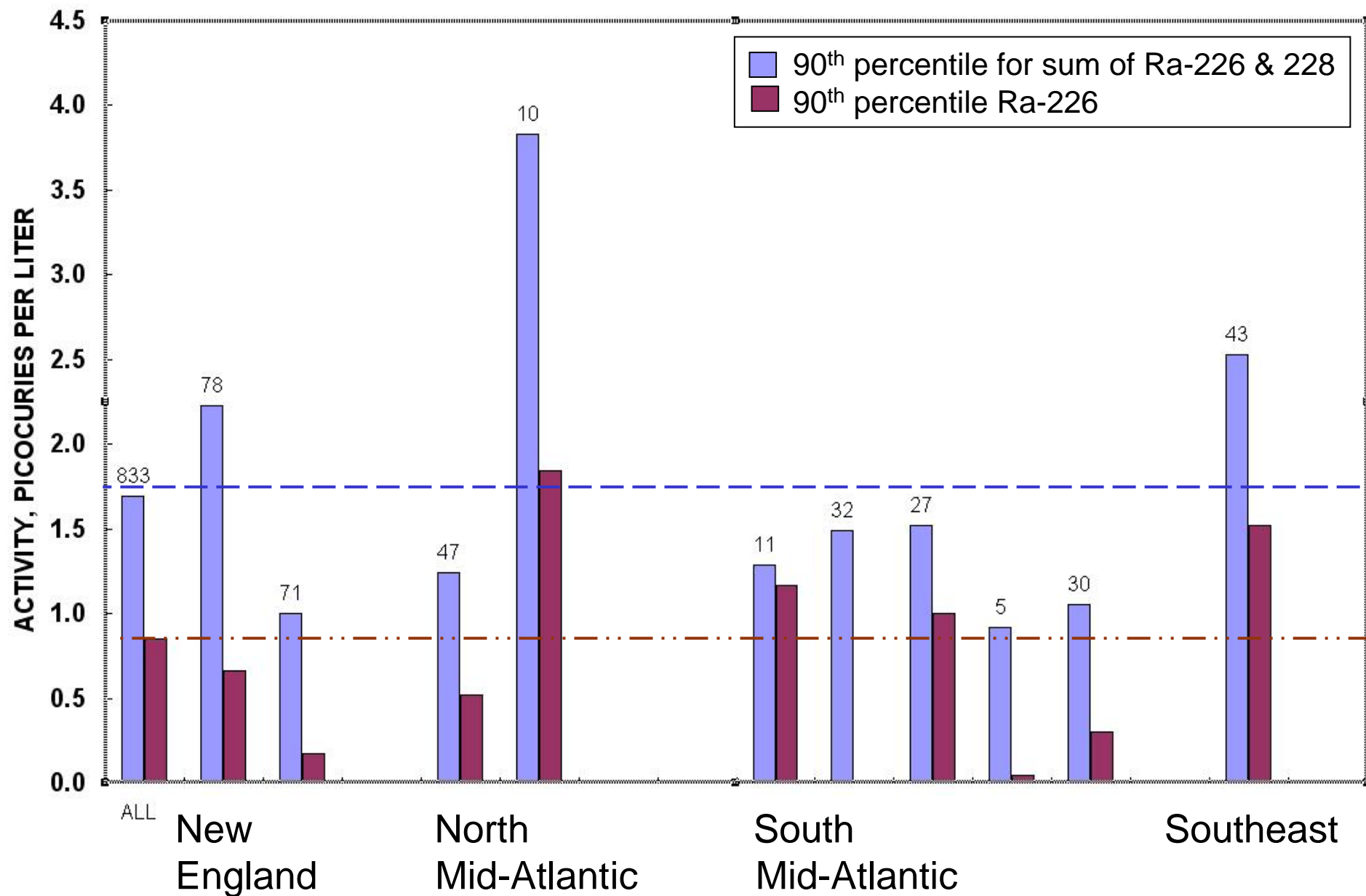
Western U.S., draft NAWQA data



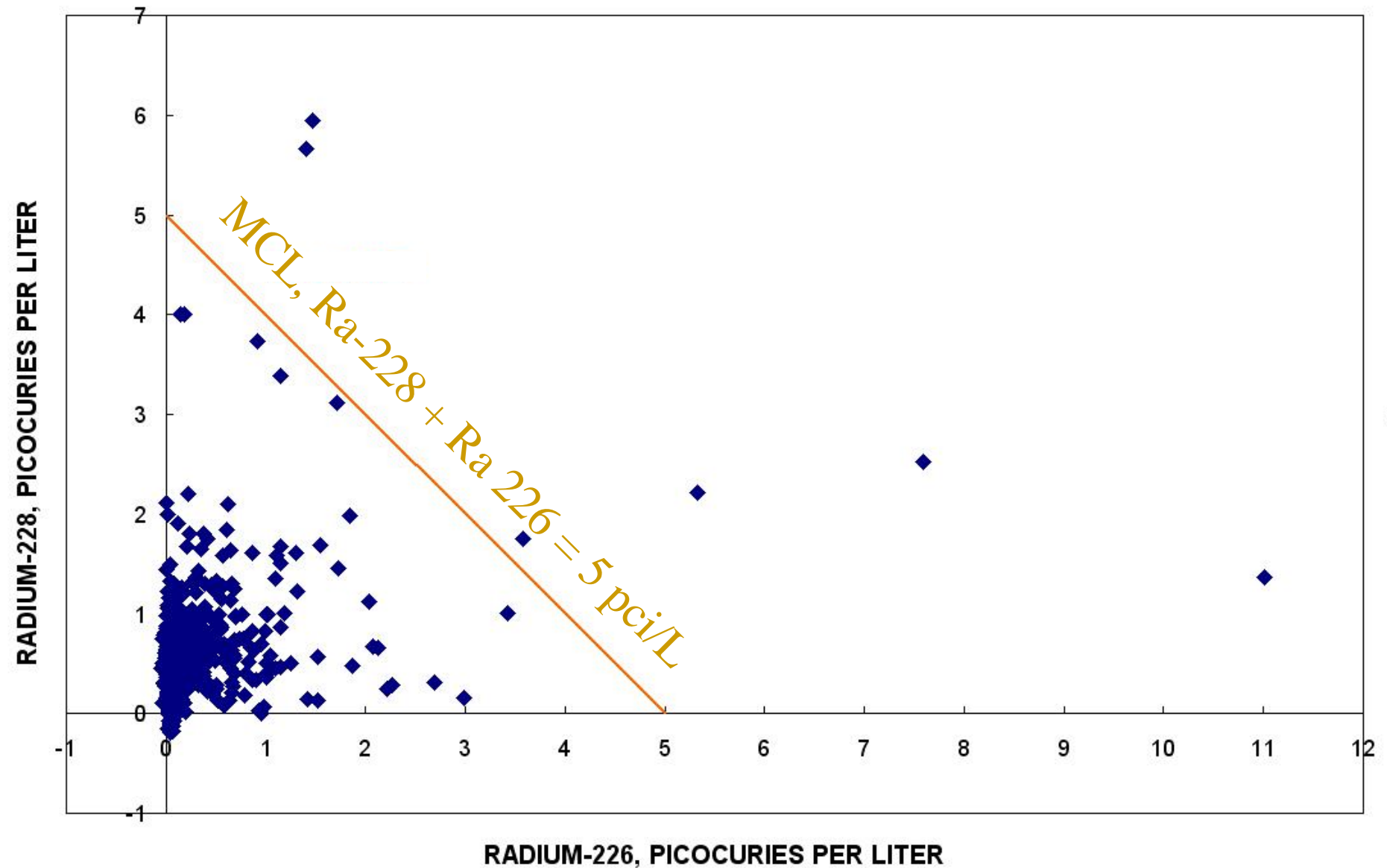
Central U.S., draft NAWQA data



Eastern U.S., draft NAWQA data



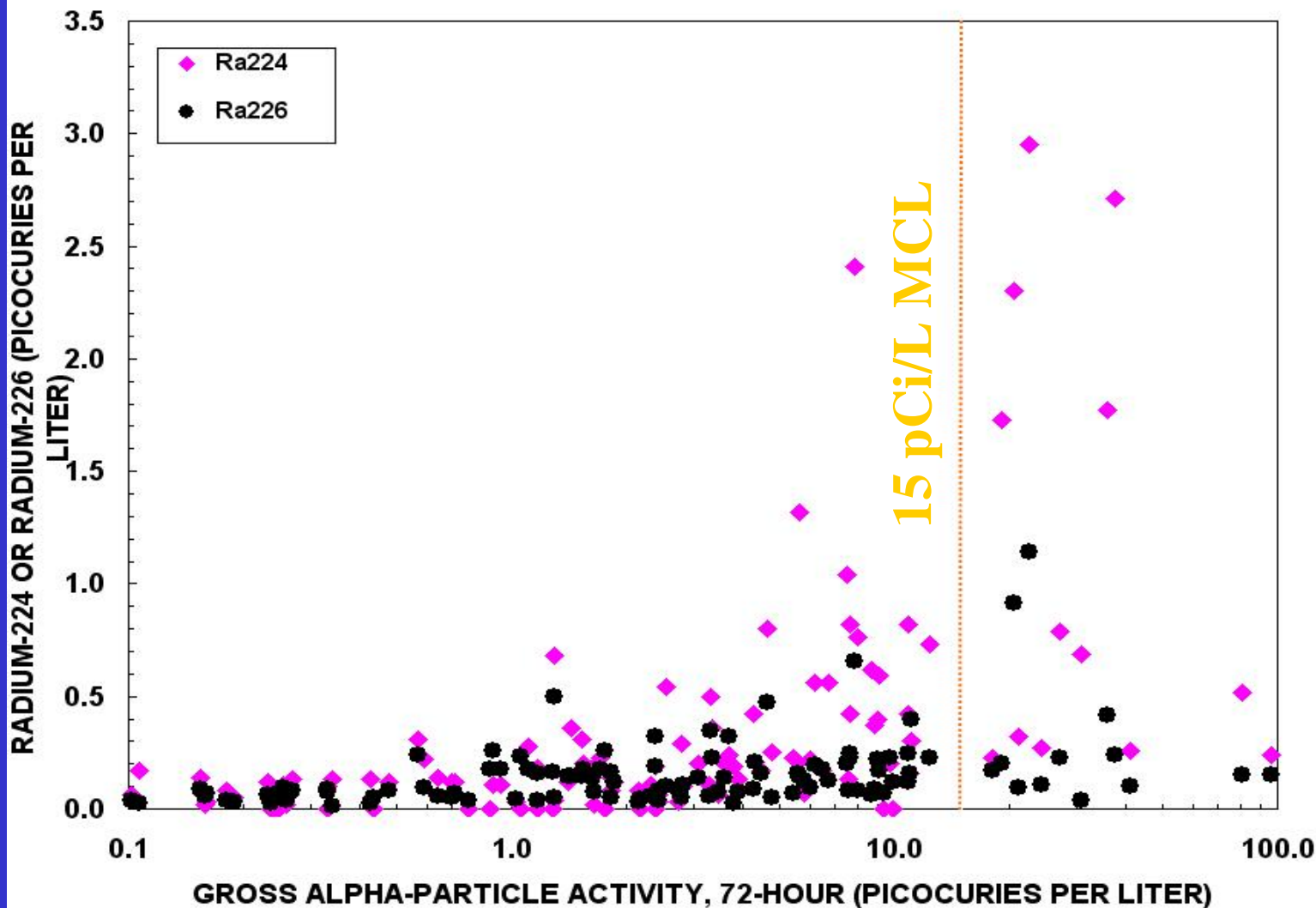
Radium 228 & 226 co-occurrence



Draft NAWQA data

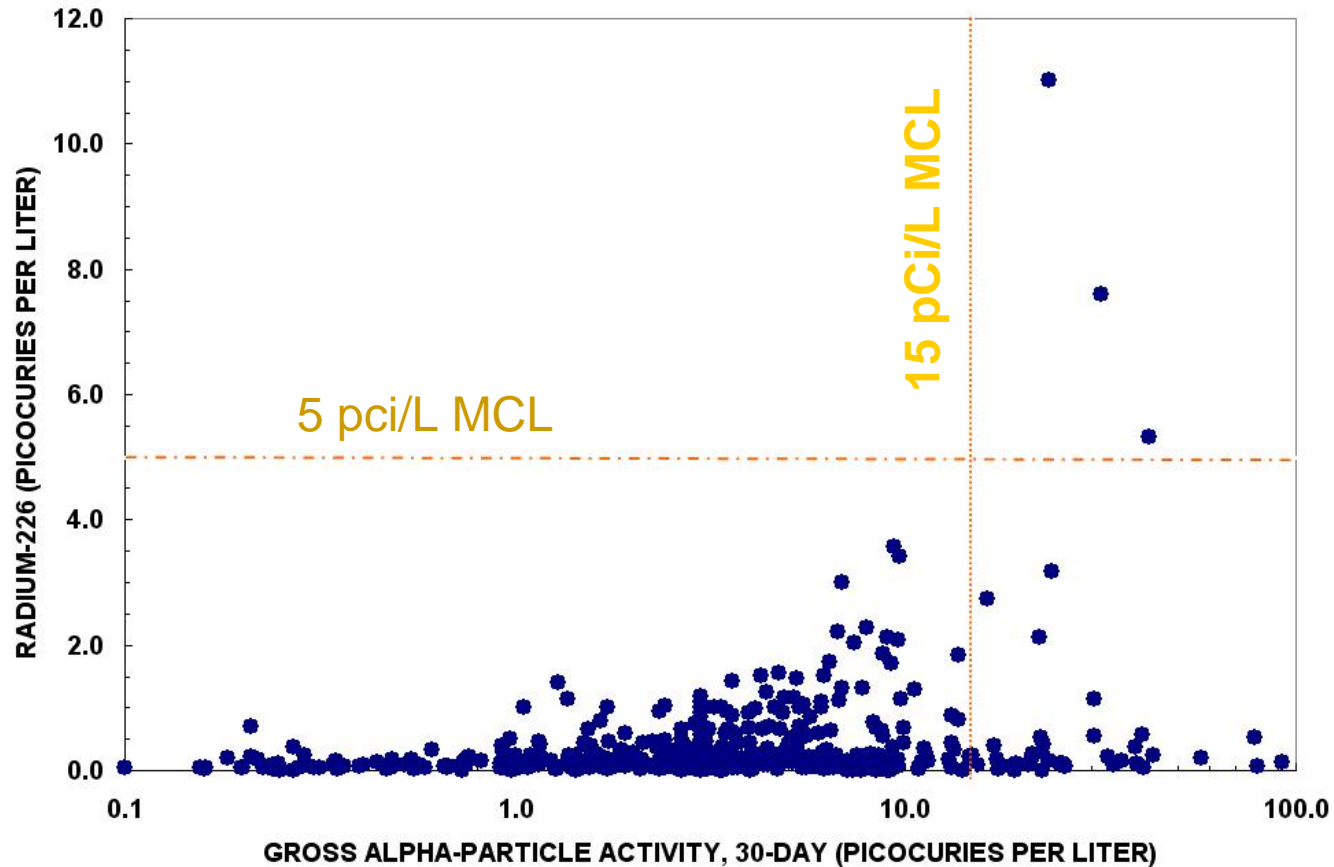
72 Hour Gross Alpha

Ra-224 and Ra-226 concentration as a function of 72-hour alpha activity



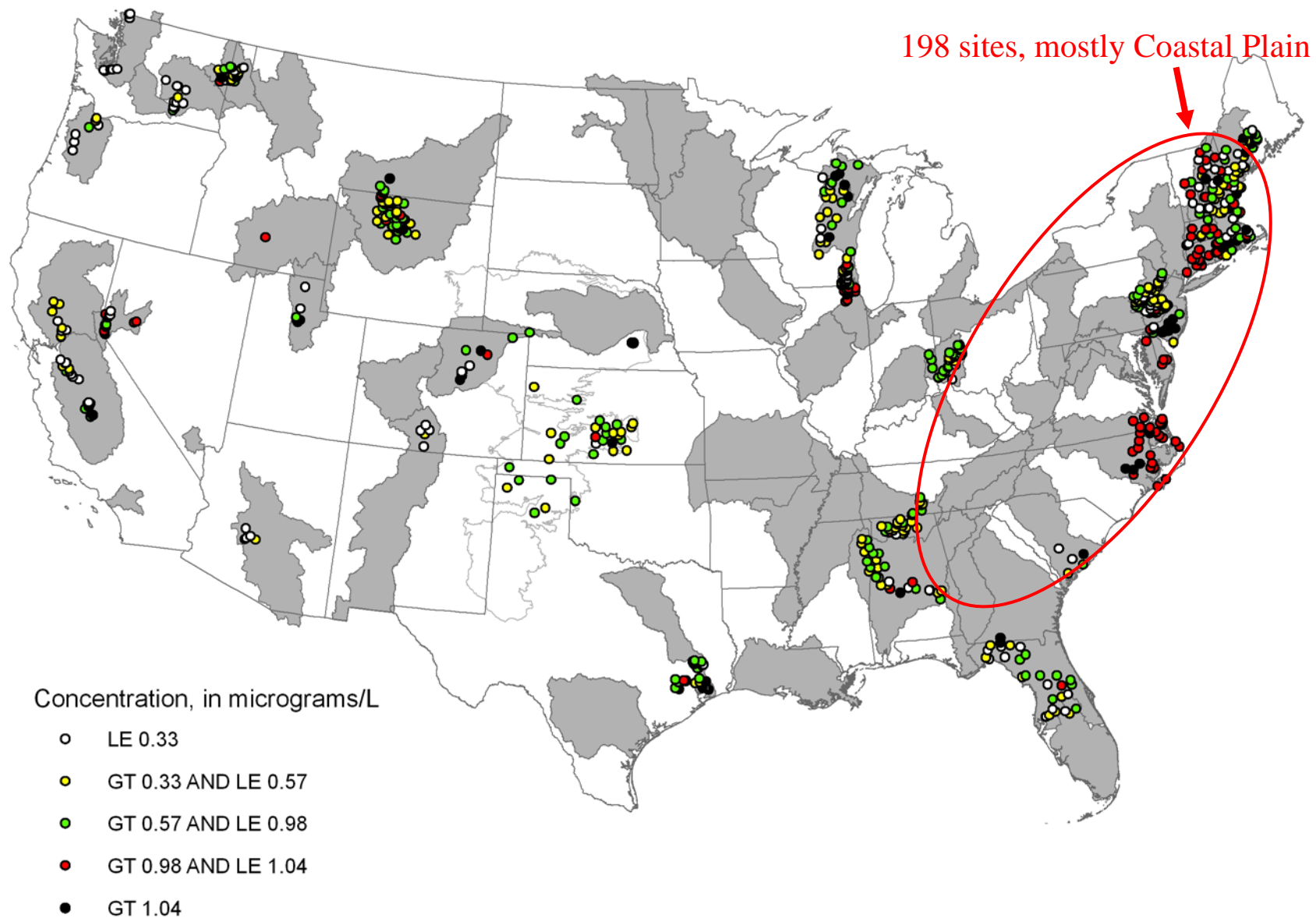
30 Day Gross alpha from Uranium Decay

Ra226 as a function of 30 day alpha activity

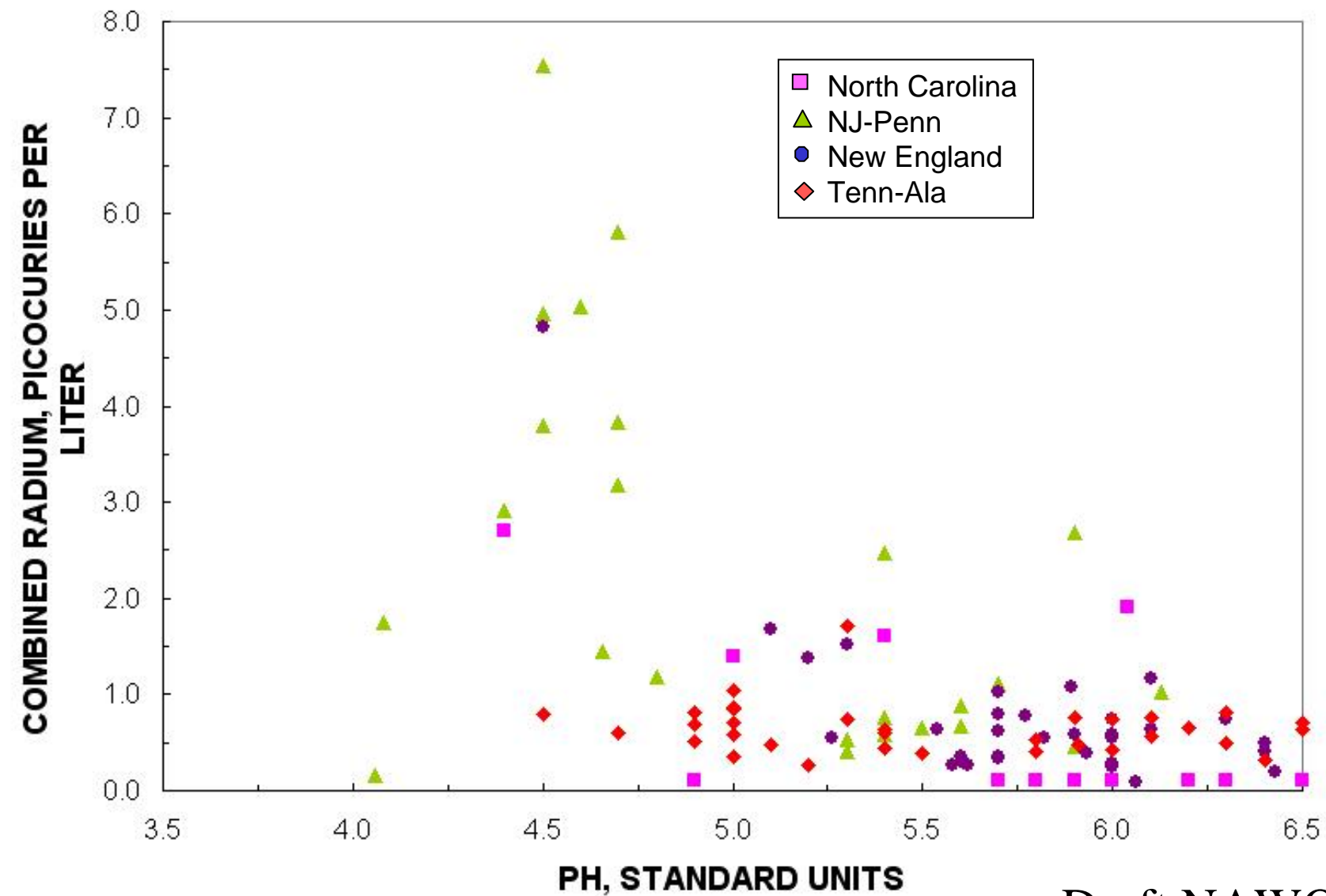


Are there geochemical or geological features on National (NAWQA) and local scales that help evaluate occurrence of Ra?

Distribution of NAWQA Ra-228 Data



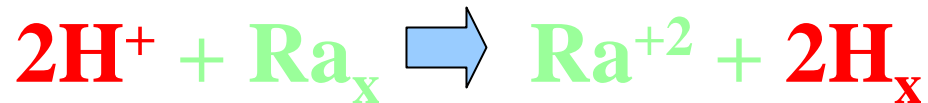
COMBINED RADIUM VS. PH



Draft NAWQA data

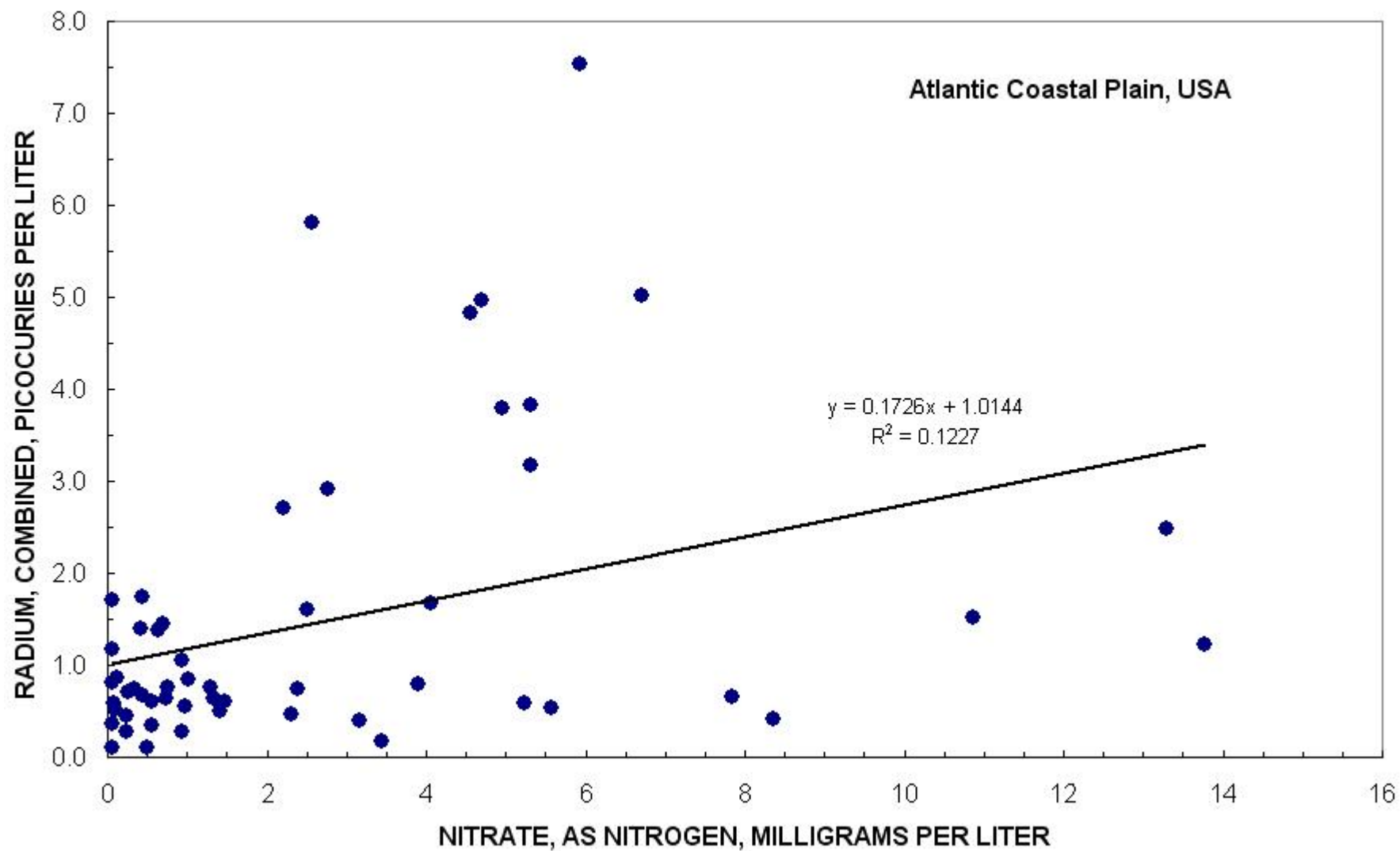
Hypothetical Chemical Reactions by which Hydrogen Ions in Water Increase Radium Mobility

➤ Ion-exchange, desorption reactions:



- Ra is preferentially removed from solid.
- Hydrogen ion is a master variable that exerts significant control over these reactions.
- Hydrogen ions with high concentrations of other cations preferentially remove Ra from solid.

Desorption in the presence of Nitrate



Draft NAWQA data

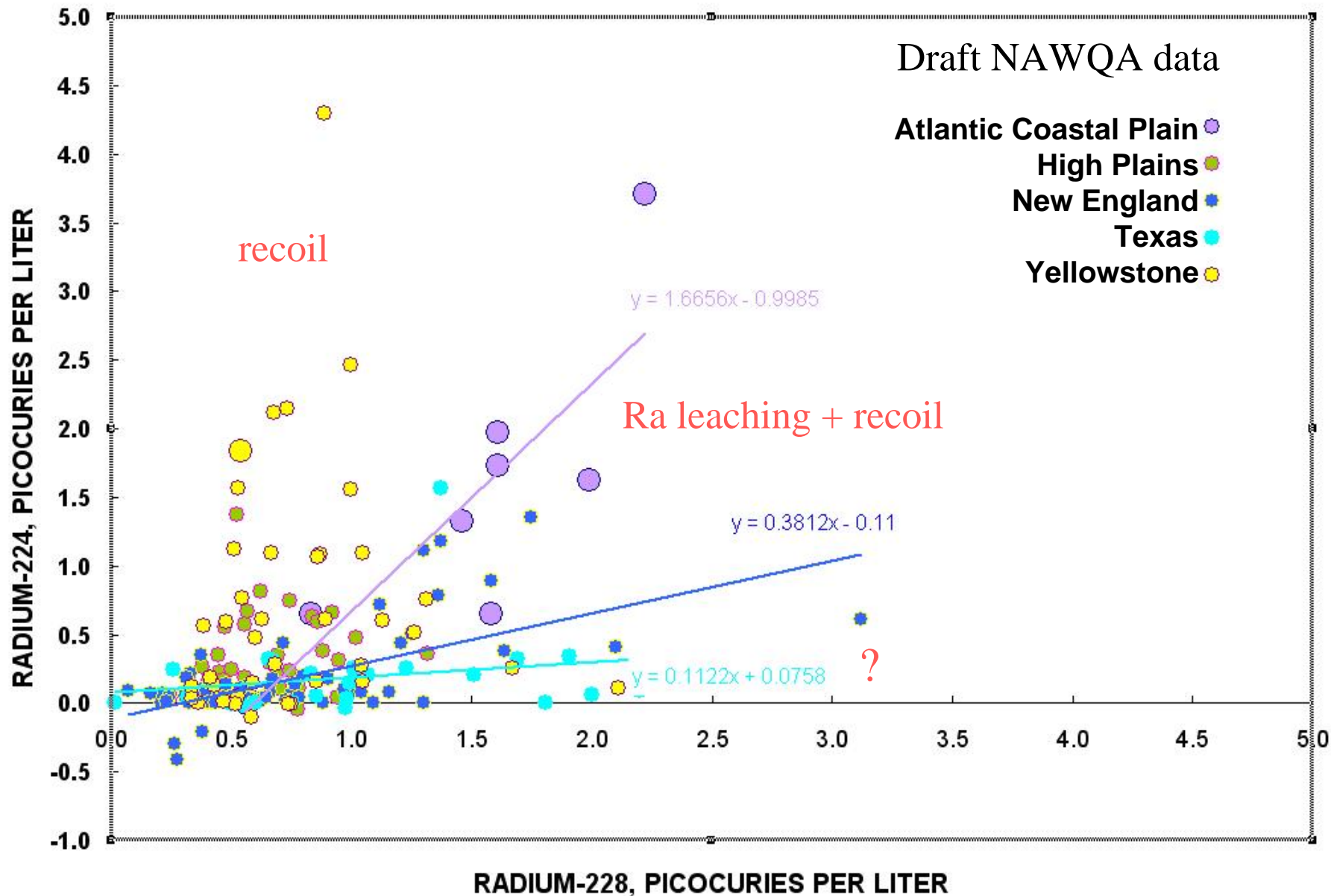
Hypothetical Chemical Reaction That Produces Hydrogen Ions in Water and Increases Radium Mobility

➤ Redox reactions:

Nitrification:



Chemical and Physical Processes in Different Regions



CONCLUSIONS

1. Higher concentrations of Ra occur primarily in unconsolidated aquifers - usually in acidic regions of the Eastern US
2. Ra-224 likely source of gross alpha in Eastern US
2. Ra-224 is likely to occur where other radium isotopes are present & correlates with Ra-228.
3. Ra-224 may be a substantial source of alpha activity in western US, but mobilization mechanism not understood (recoil?).

CONCLUSIONS

4. Further characterization of geochemical implications for monitoring programs and treatment applications is warranted!

More data collection and analysis is planned

Contacts:

Zoltan Szabo ZSzabo@USGS.GOV 609-771-3929

Jeff Fischer Fischer@USGS.GOV 609-771-3953