GEOCHEMICAL AND HYDROGEOLOGICAL CONTROLS ON RADIUM AND URANIUM IN DEEP SANDSTONE AQUIFERS OF ILLINOIS

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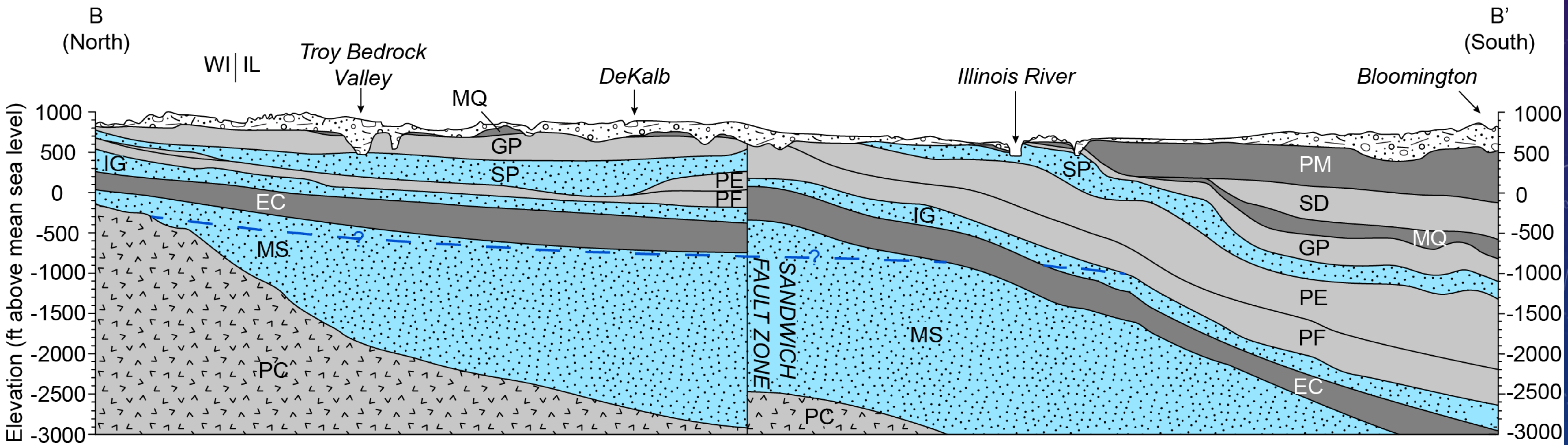
ILLINOIS BASIN: PALEOZOIC SEDIMENTARY ROCKS



Geologic Material

- Glacial Deposits
- Shale
- Carbonate
- Sandstone
- Crystalline Bedrock

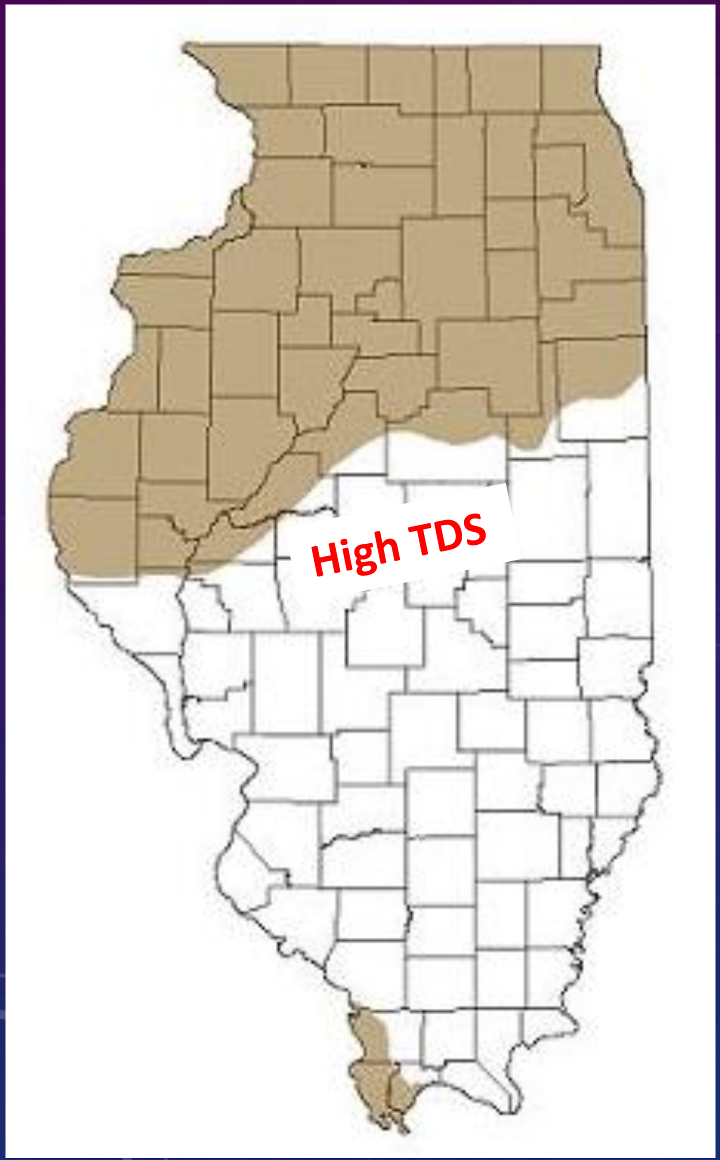
50X vertical exaggeration

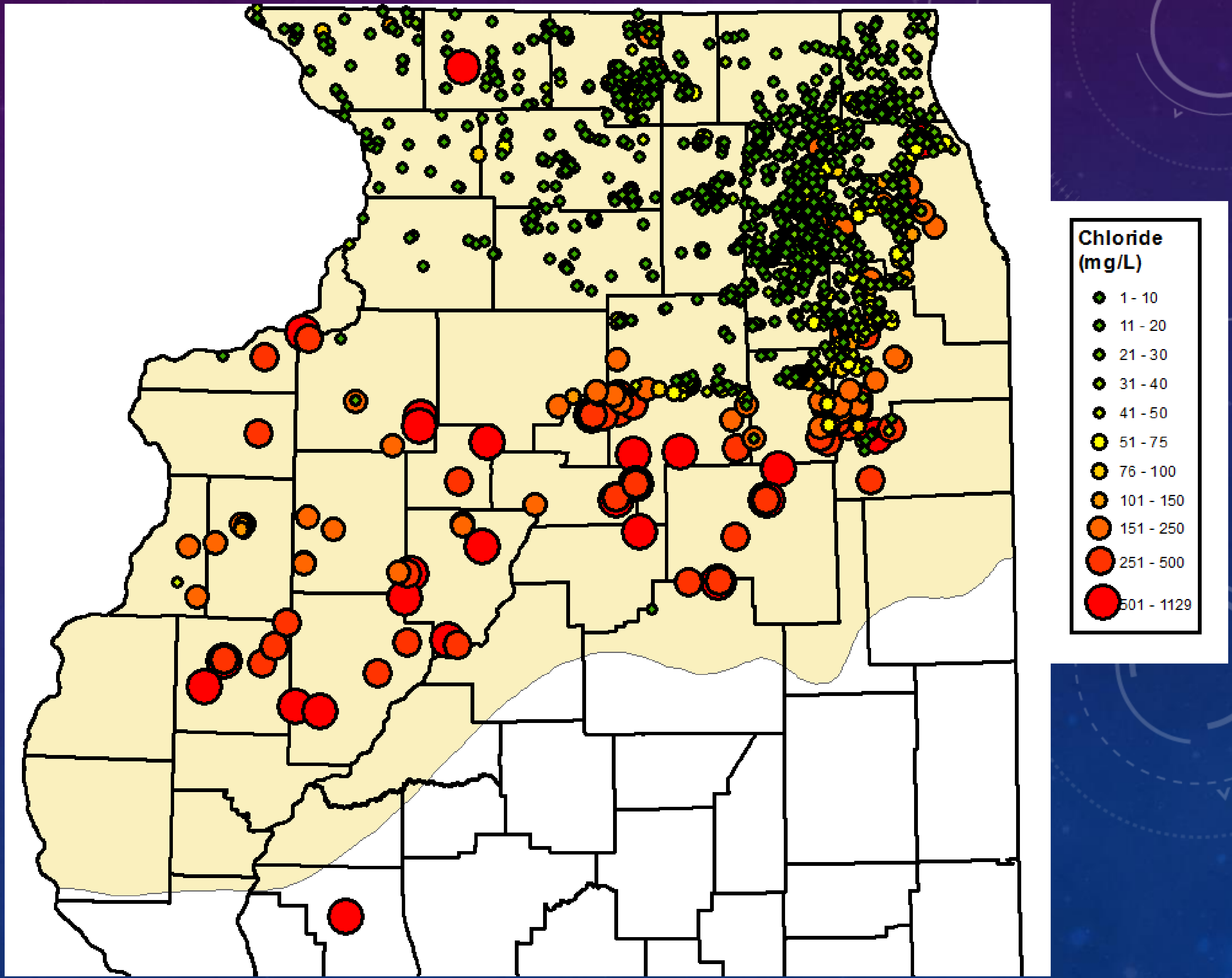
Hydrostratigraphic Units

PM	Pennsylvanian-Mississippian	PF	Potosi-Franconia
SD	Silurian-Devonian	IG	Ironton-Galesville
MQ	Maquoketa	EC	Eau Claire
GP	Galena-Platteville	MS	Mt. Simon
SP	St. Peter	PC	Precambrian
PE	Prairie du Chien-Eminence		

fresh/saline groundwater contact

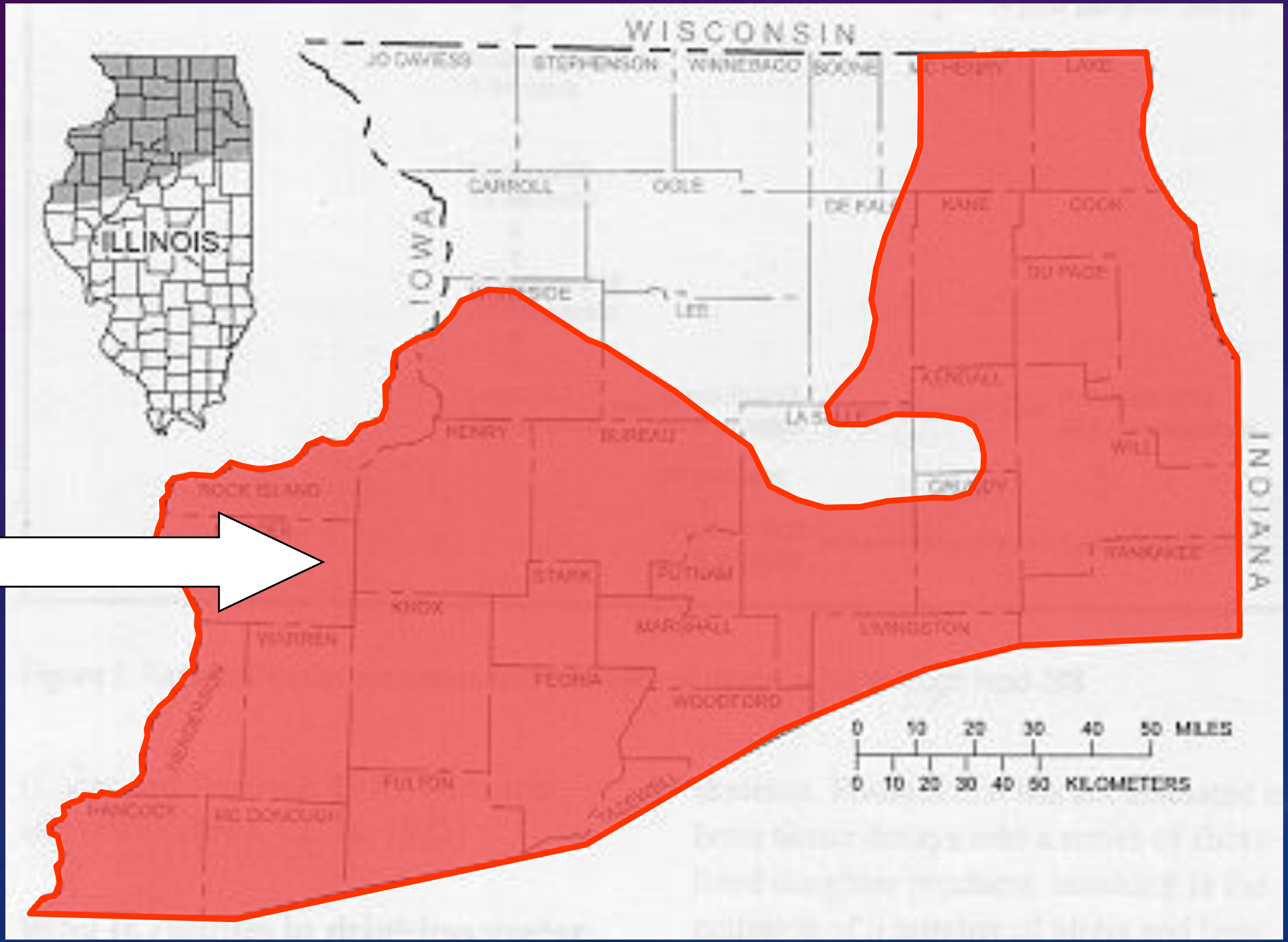
WATER QUALITY ISSUE: ELEVATED TDS





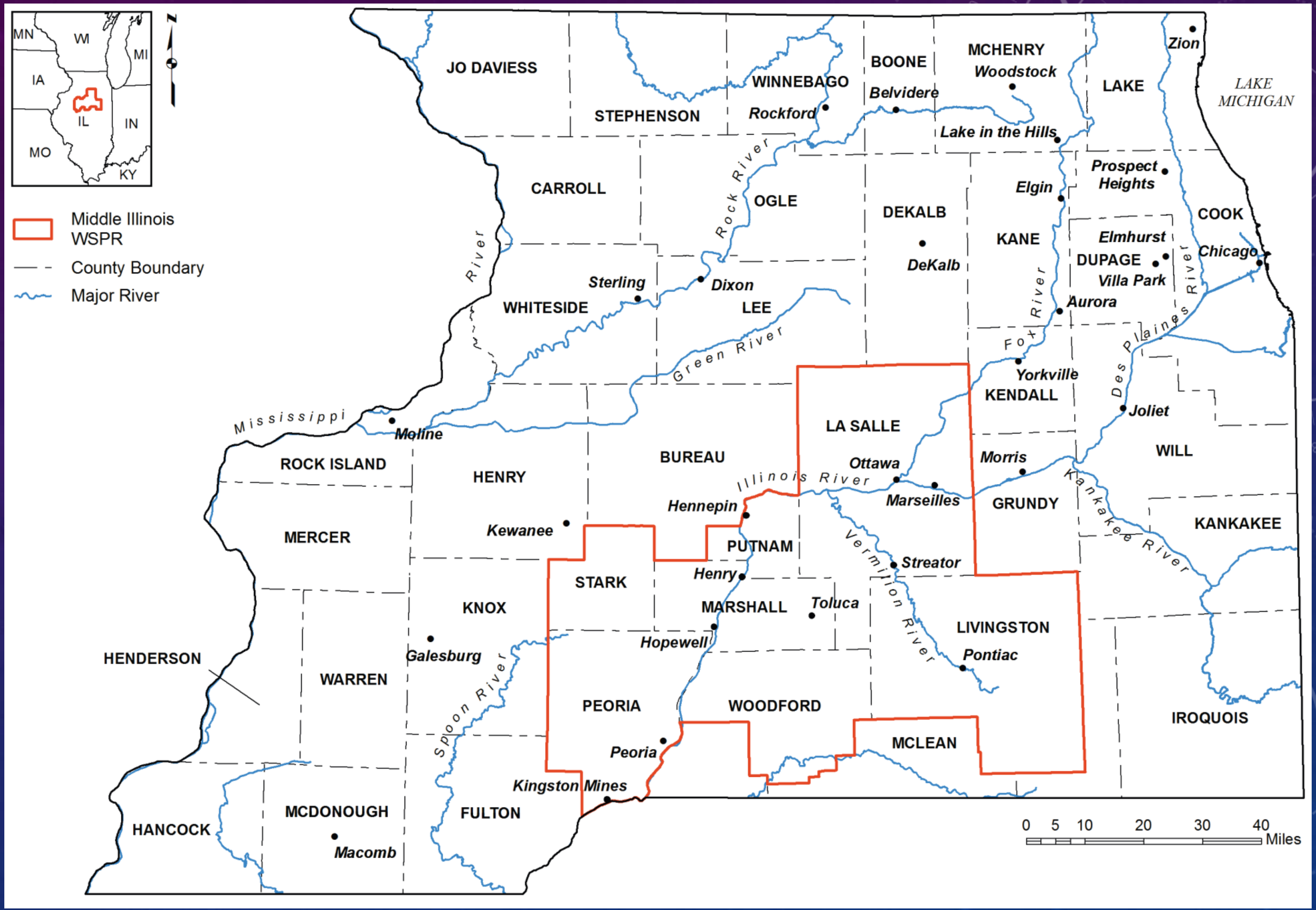
WATER QUALITY ISSUE: RADIUM (RA)

Region where ^{226}Ra + ^{228}Ra concentrations > 5 pCi/L have been detected



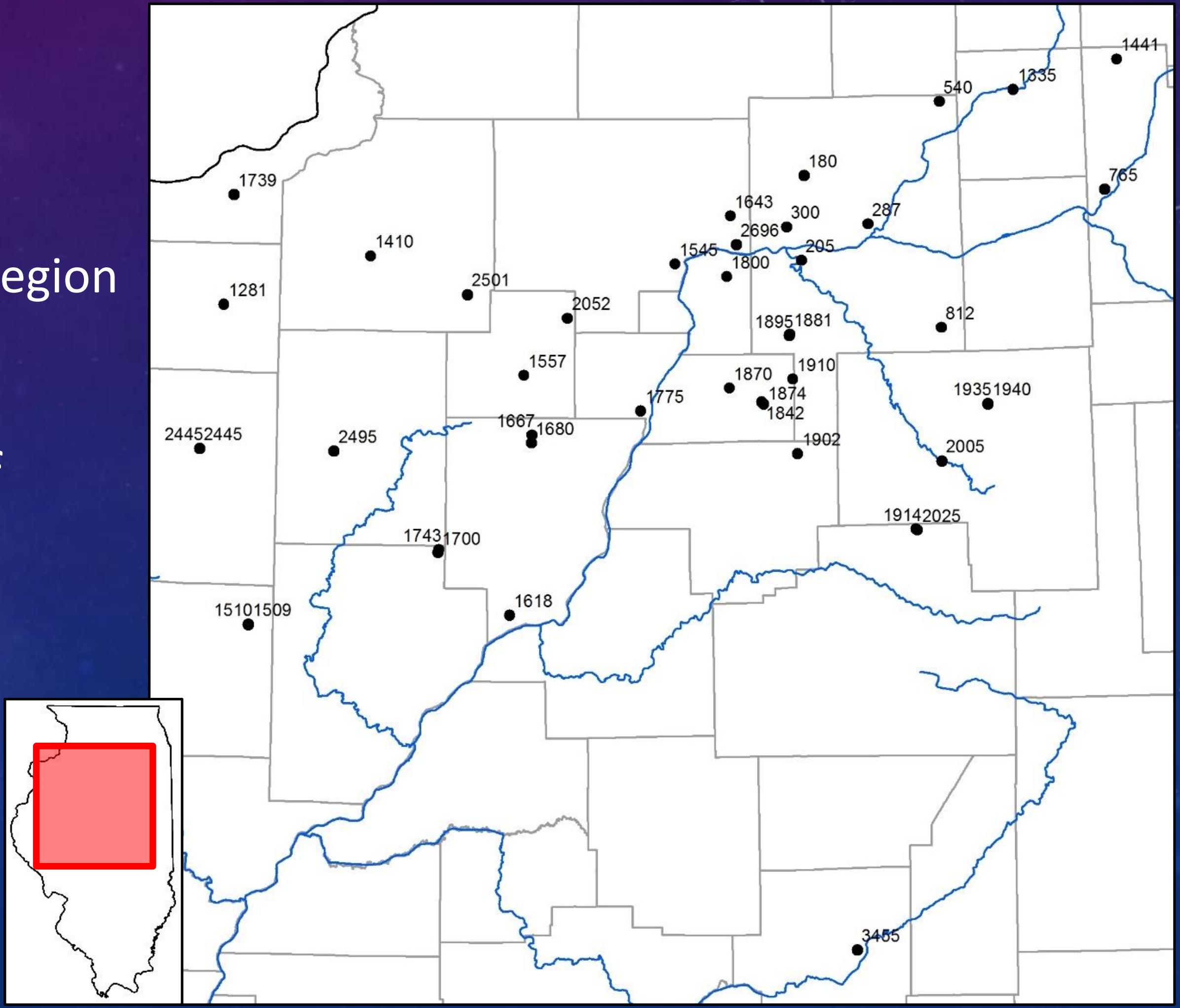
USEPA Drinking Water Standard = 5 pCi/L

MIDDLE ILLINOIS RIVER WATER SUPPLY PLANNING REGION



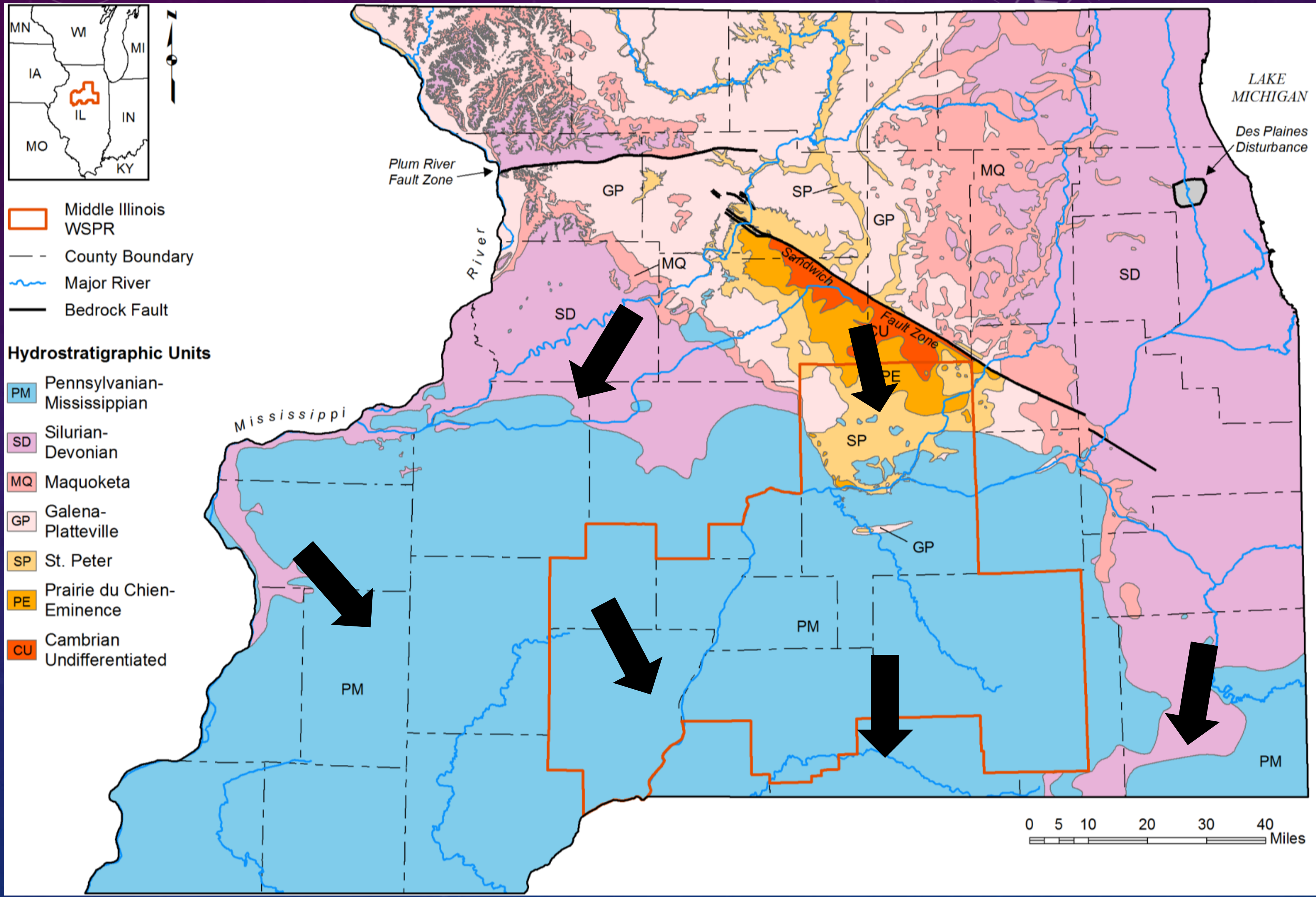
WATER CHEMISTRY SAMPLING IN THE ST. PETER SANDSTONE: MIDDLE ILLINOIS RIVER REGION

- 32 wells sampled, primarily public supply or industrial wells
  - Some open to deeper units
  - Also 1 sample from borehole near Decatur south of region
- Complete inorganic chemistry
- Stable isotopes (δD, $\delta^{18}O$, $\delta^{13}C$, $\delta^{34}S$ and $\delta^{18}O$ of SO_4^{2-}, $^{87}Sr/^{86}Sr$)
- Radioactive isotopes (^{226}Ra, ^{228}Ra, ^{222}Rn, ^{238}U, $^{234}U/^{238}U$)
- Supplemented with previously collected data, primarily from USGS and Illinois EPA



HYDROSTRATI-GRAPHIC UNITS PRESENT AT THE BEDROCK SURFACE

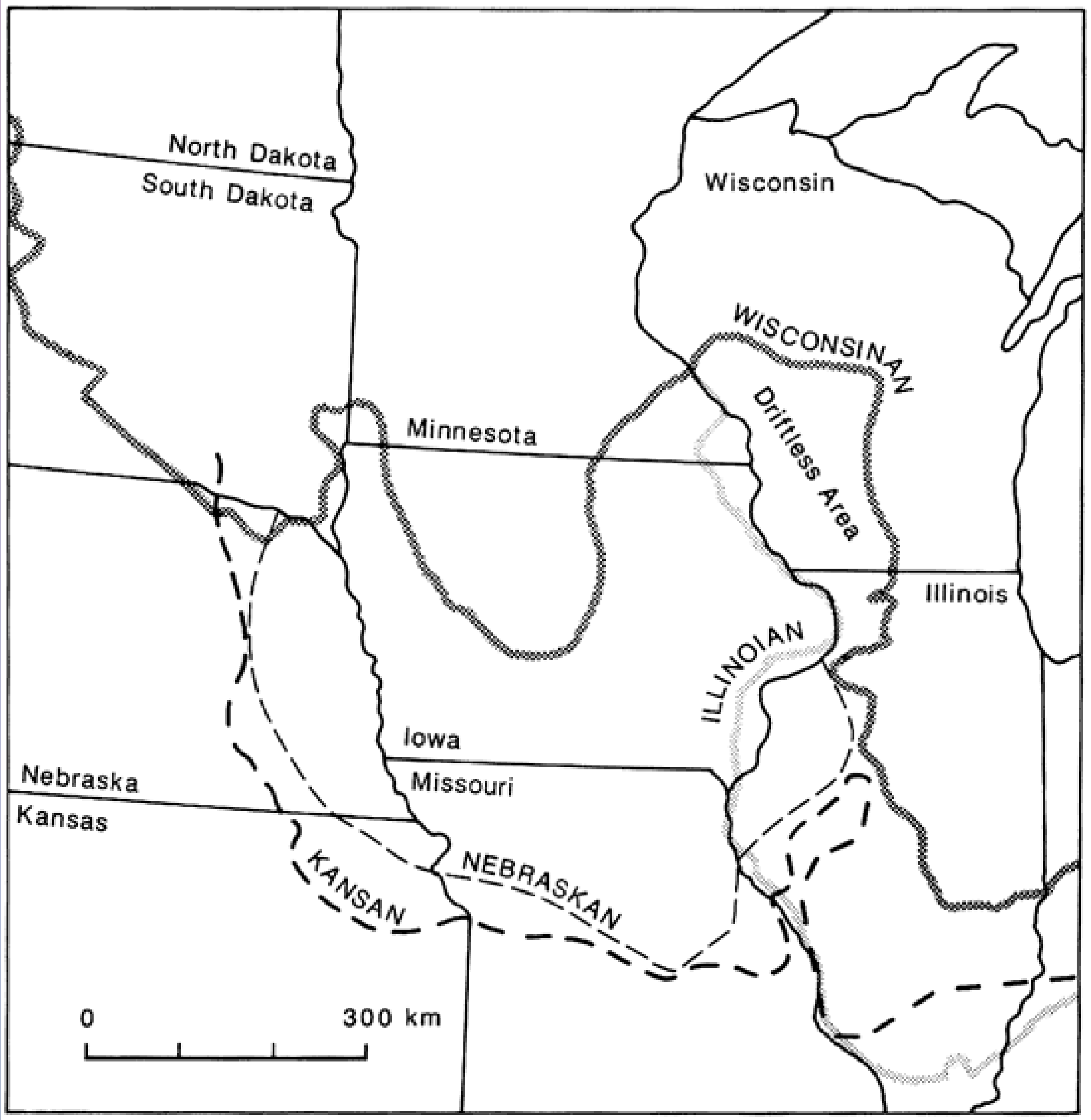
Predevelopment Flow Directions



PLEISTOCENE GLACIATION ALTERED HYDROGEOLOGY

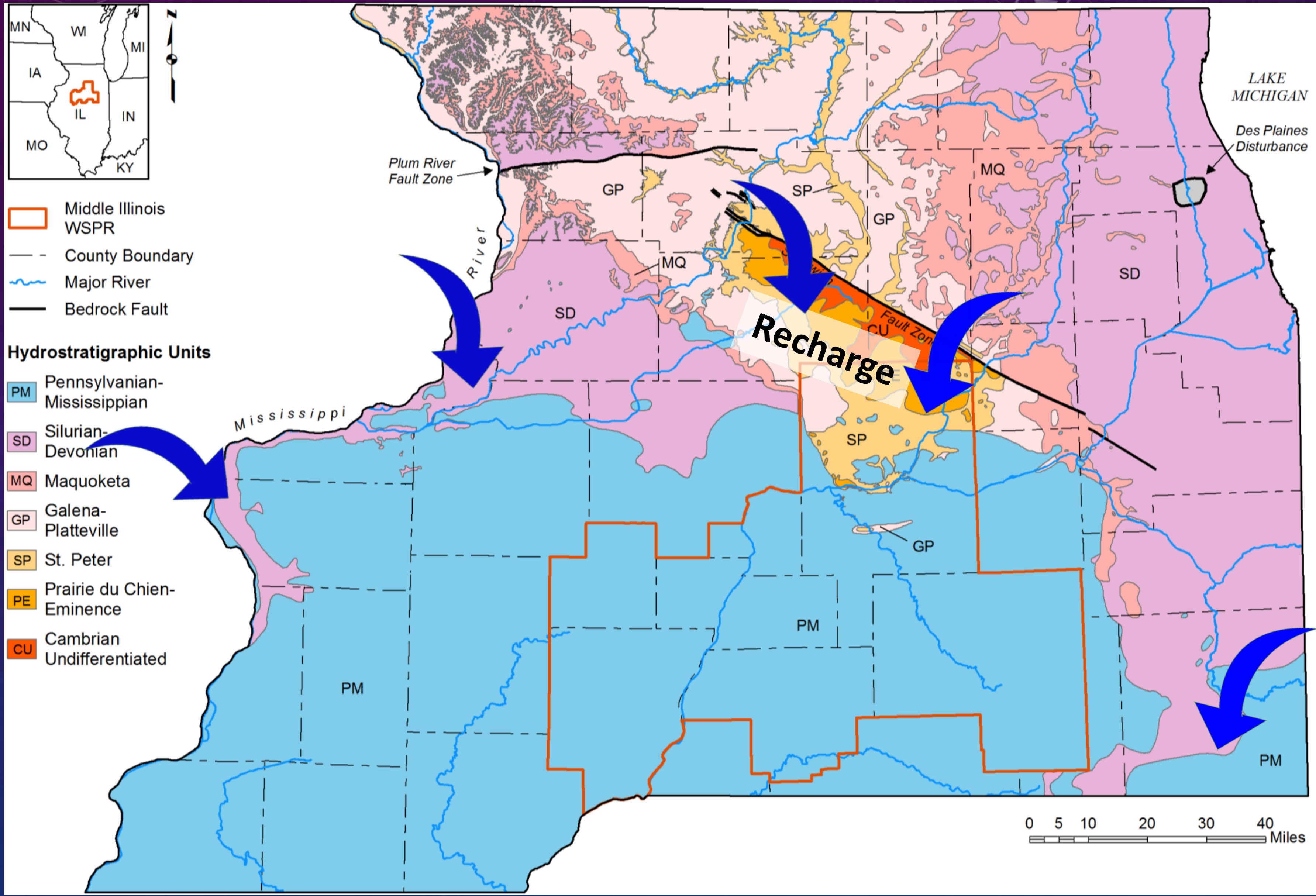
- Glaciers ebbed and flowed
- Changes in pressure, amounts of liquid water, groundwater flow directions

http://discoveringantarctica.org.uk/oceans-atmosphere-landscape/glaciation/what-are-glaciers/



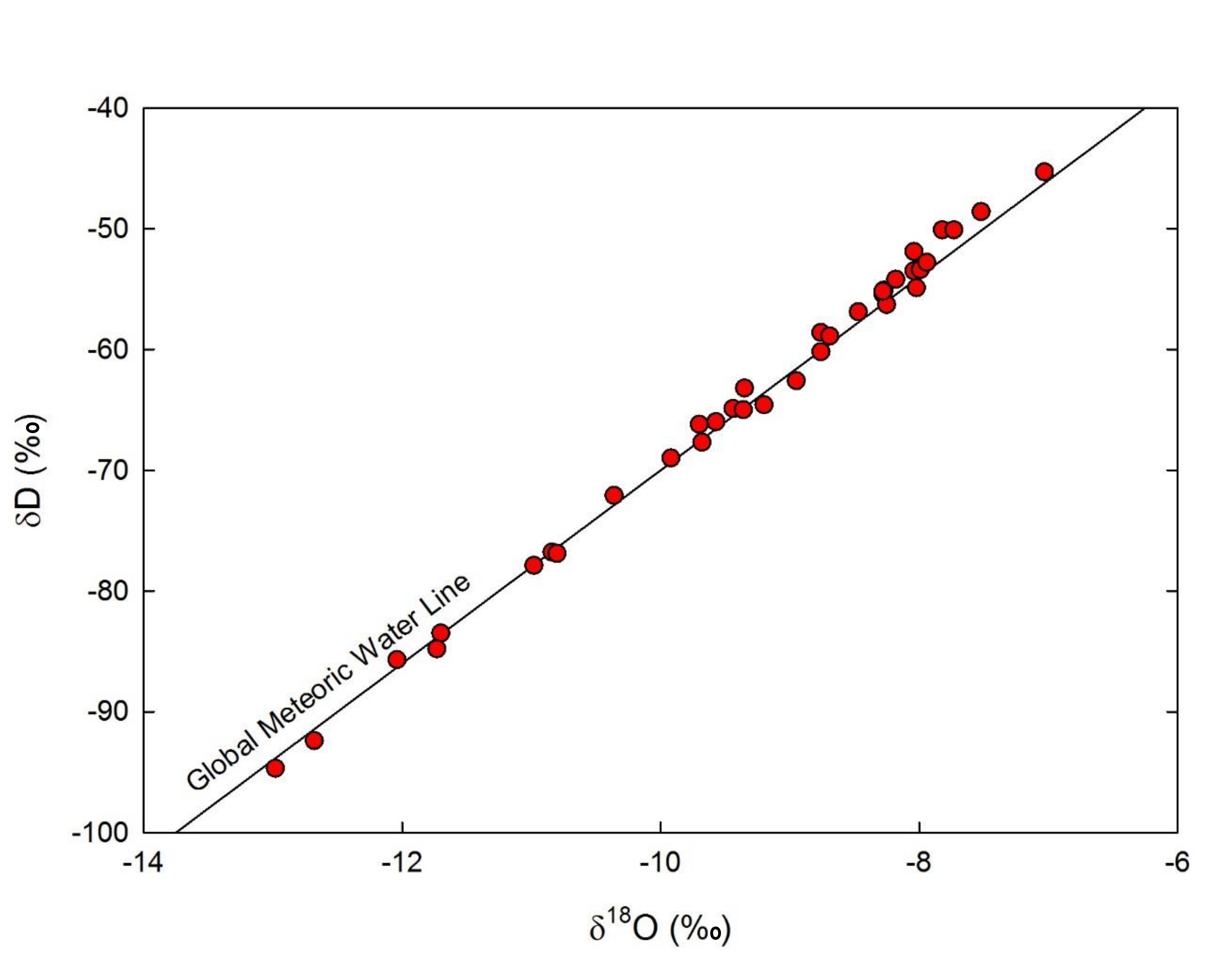
PLEISTOCENE RECHARGE

- When glaciers were melting, enhanced recharge into St. Pater and other bedrock units



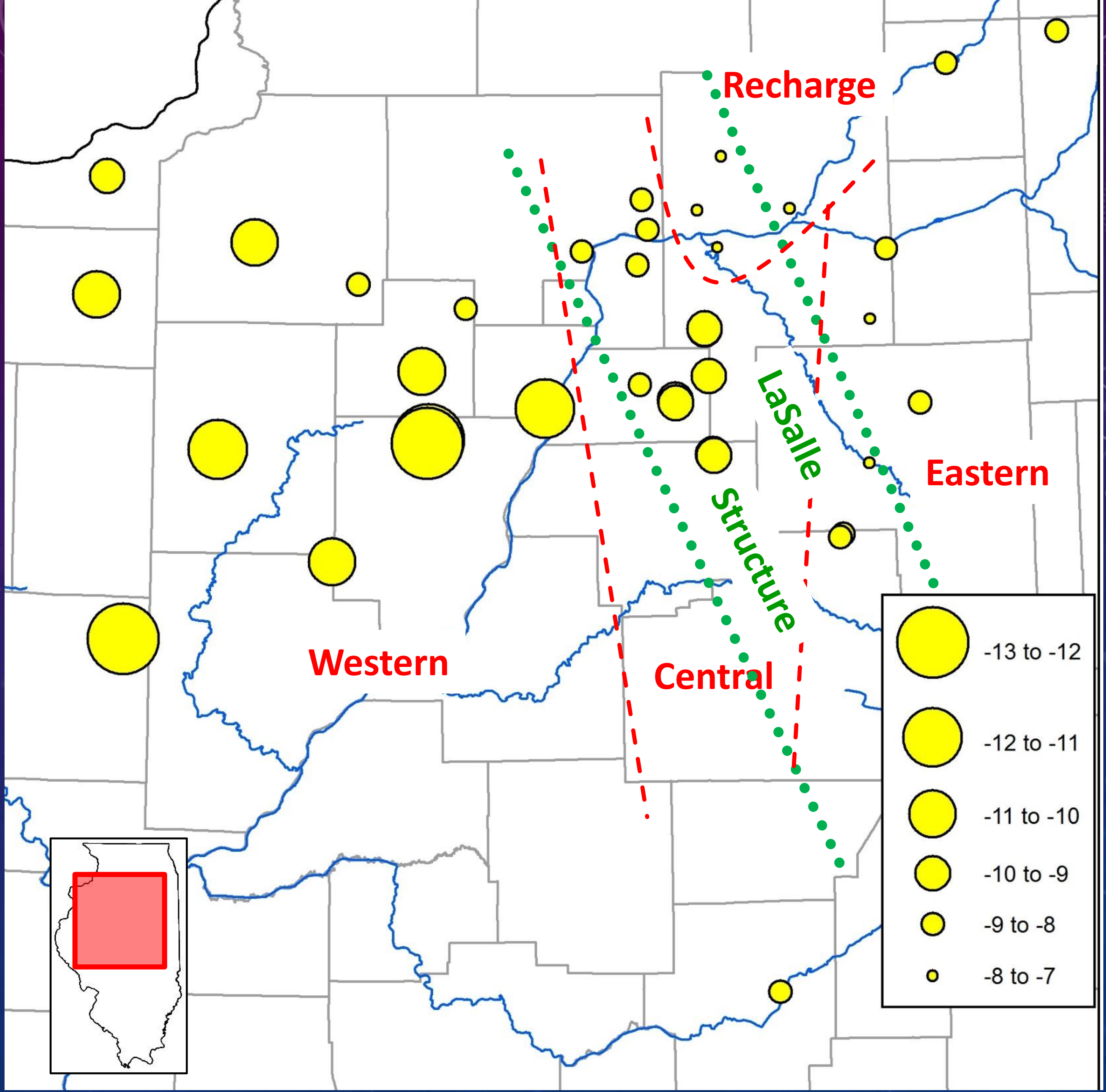
PLEISTOCENE RECHARGE

- Isotopically light water indicates recharge of relatively cold water
- No deviations from GMWL



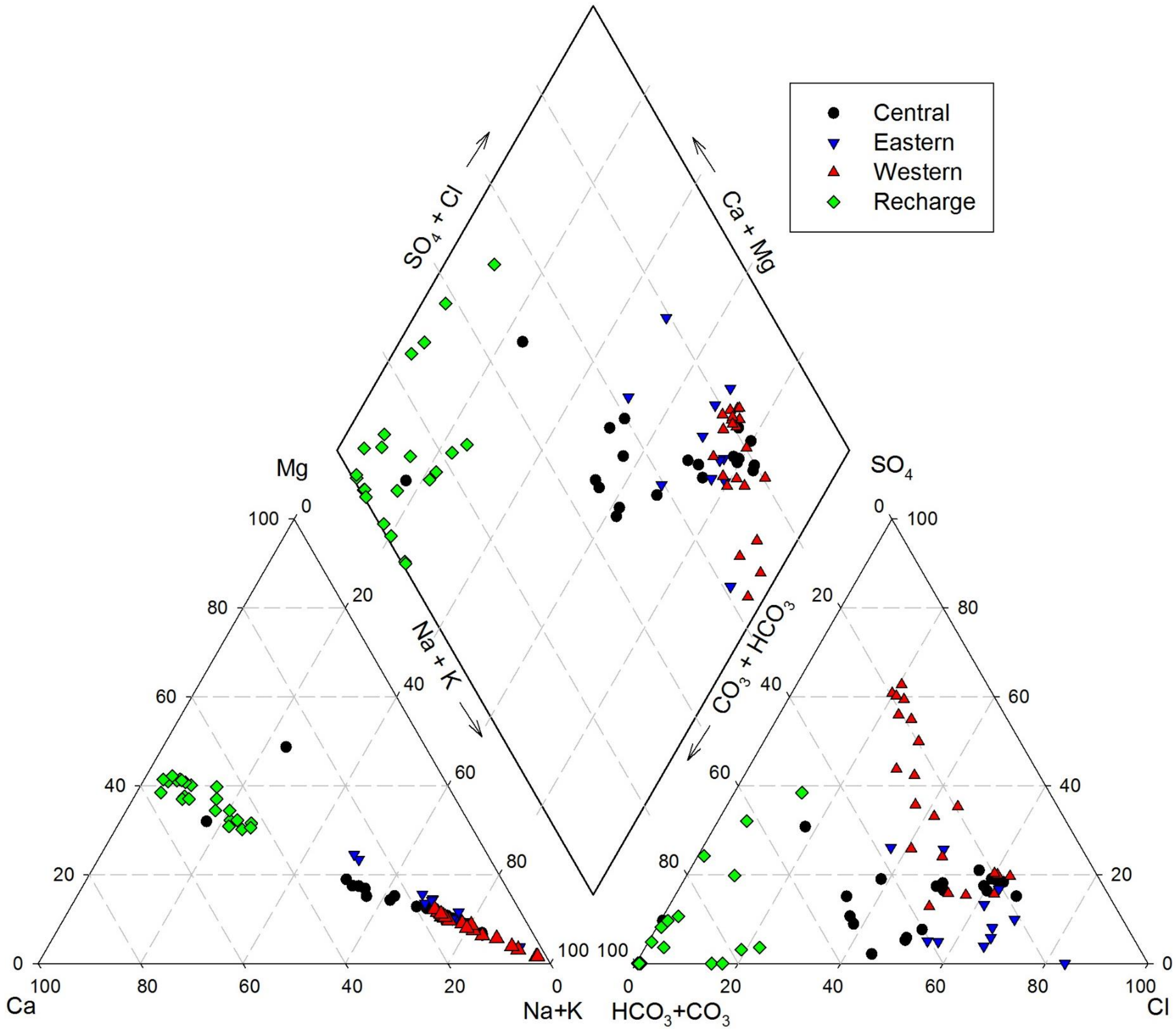
δ^{18}O VALUES

- Pleistocene recharge traveling > 100 miles from recharge zones
- Spatial differences
- Multiple flowpaths? LaSalle Structure appears to be playing a role
- Multiple end members?
- Pleistocene recharge displacing high TDS waters. Density effects?



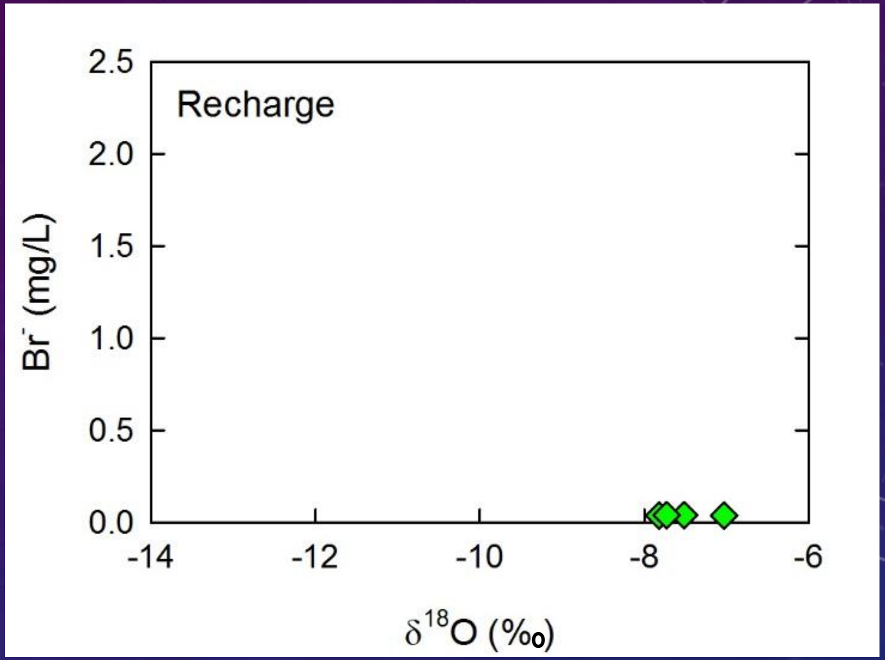
PIPER DIAGRAM

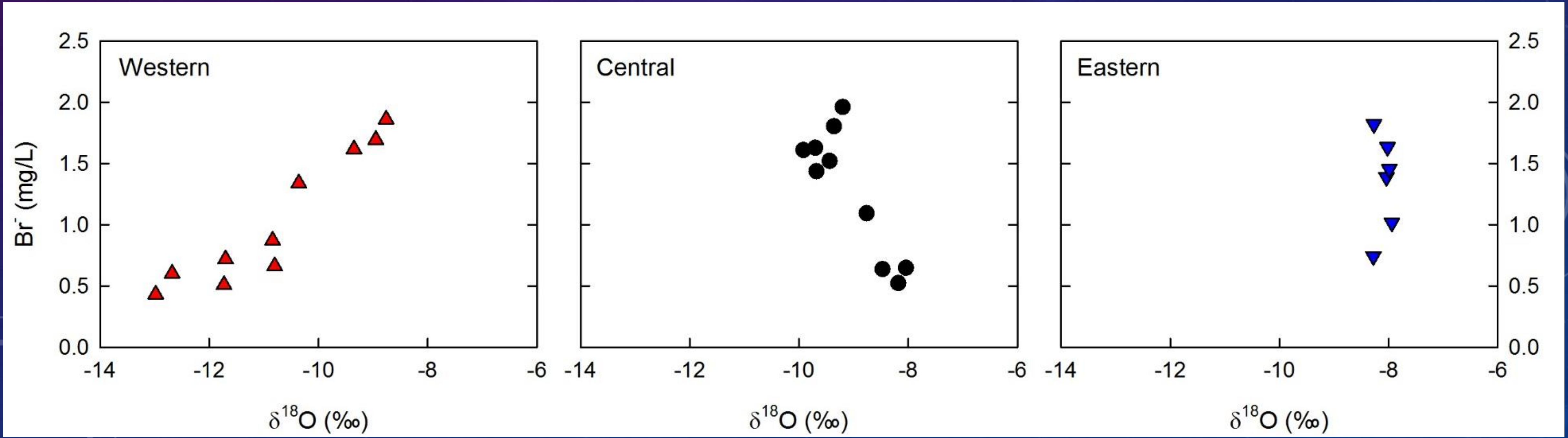
- Regional differences for cations and anions



RELATIONSHIP BETWEEN δ^{18}O AND BROMIDE

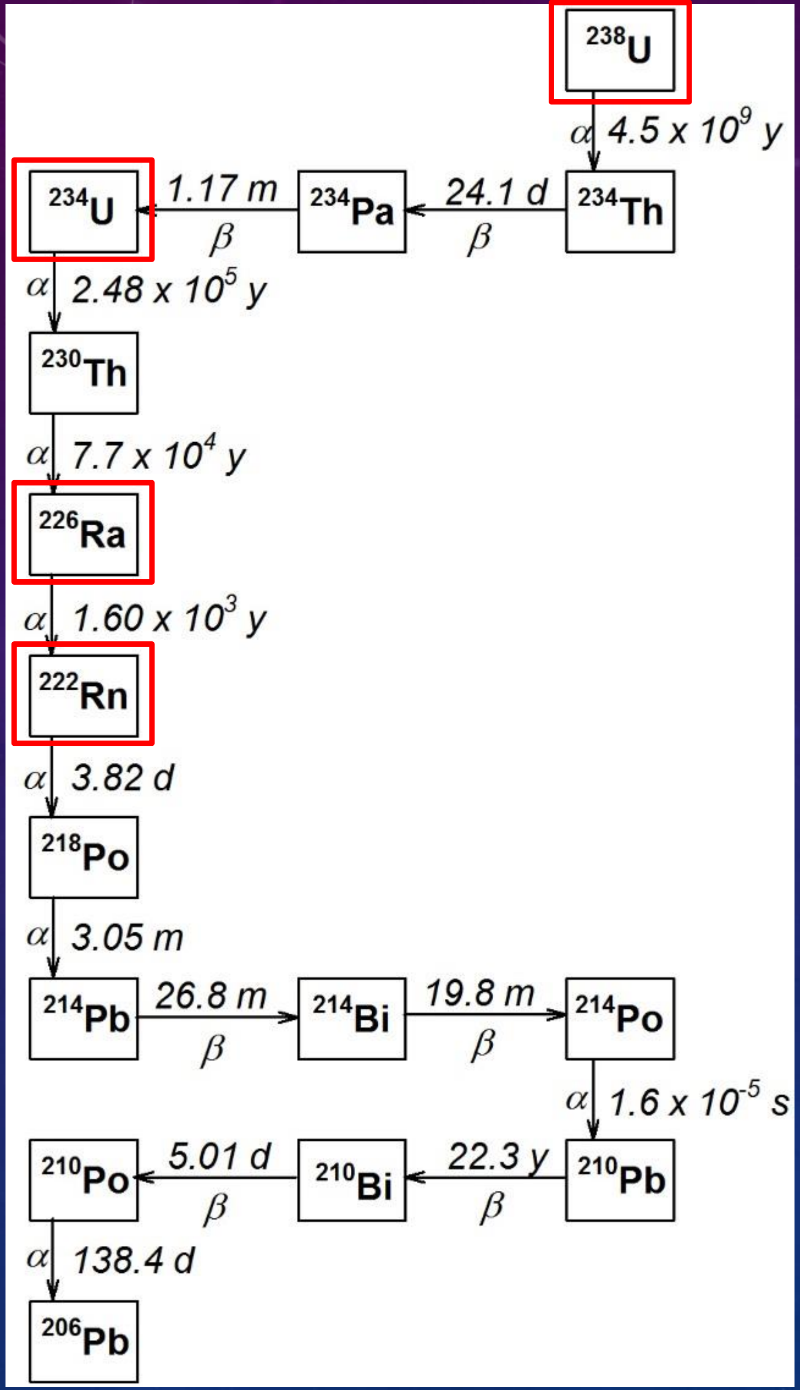
- Positive relationship if simple mixing
- Must be other processes as well

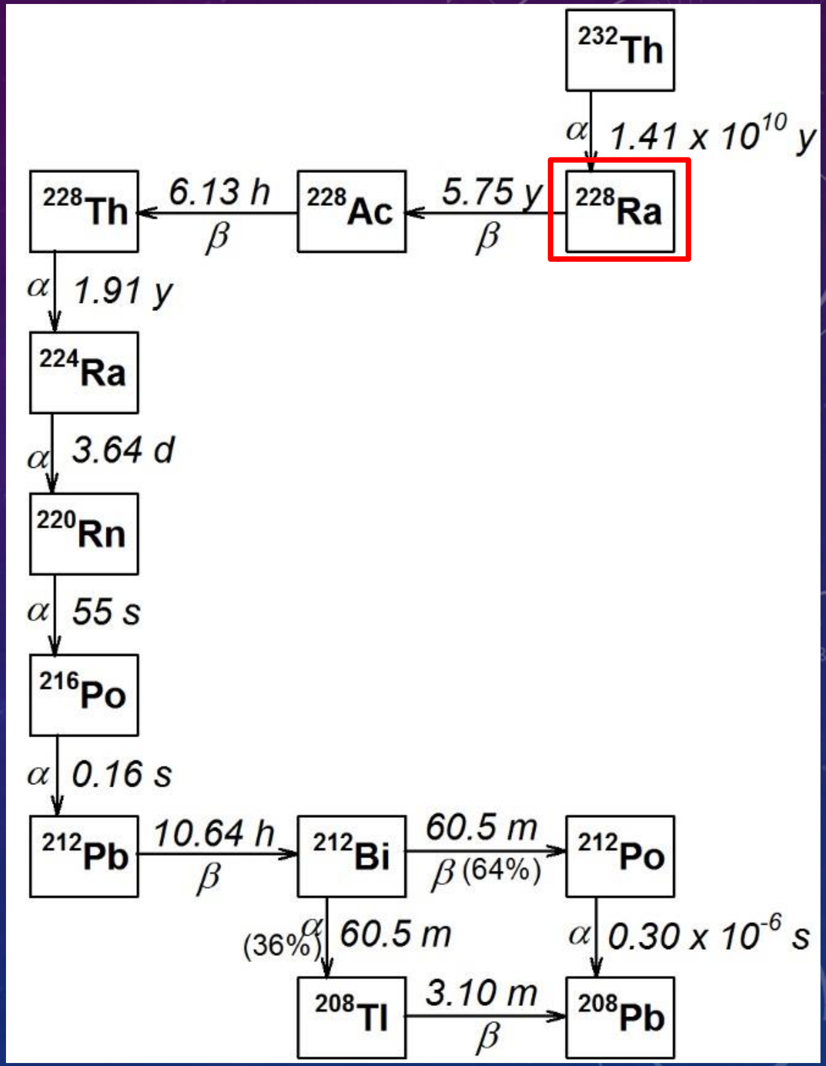




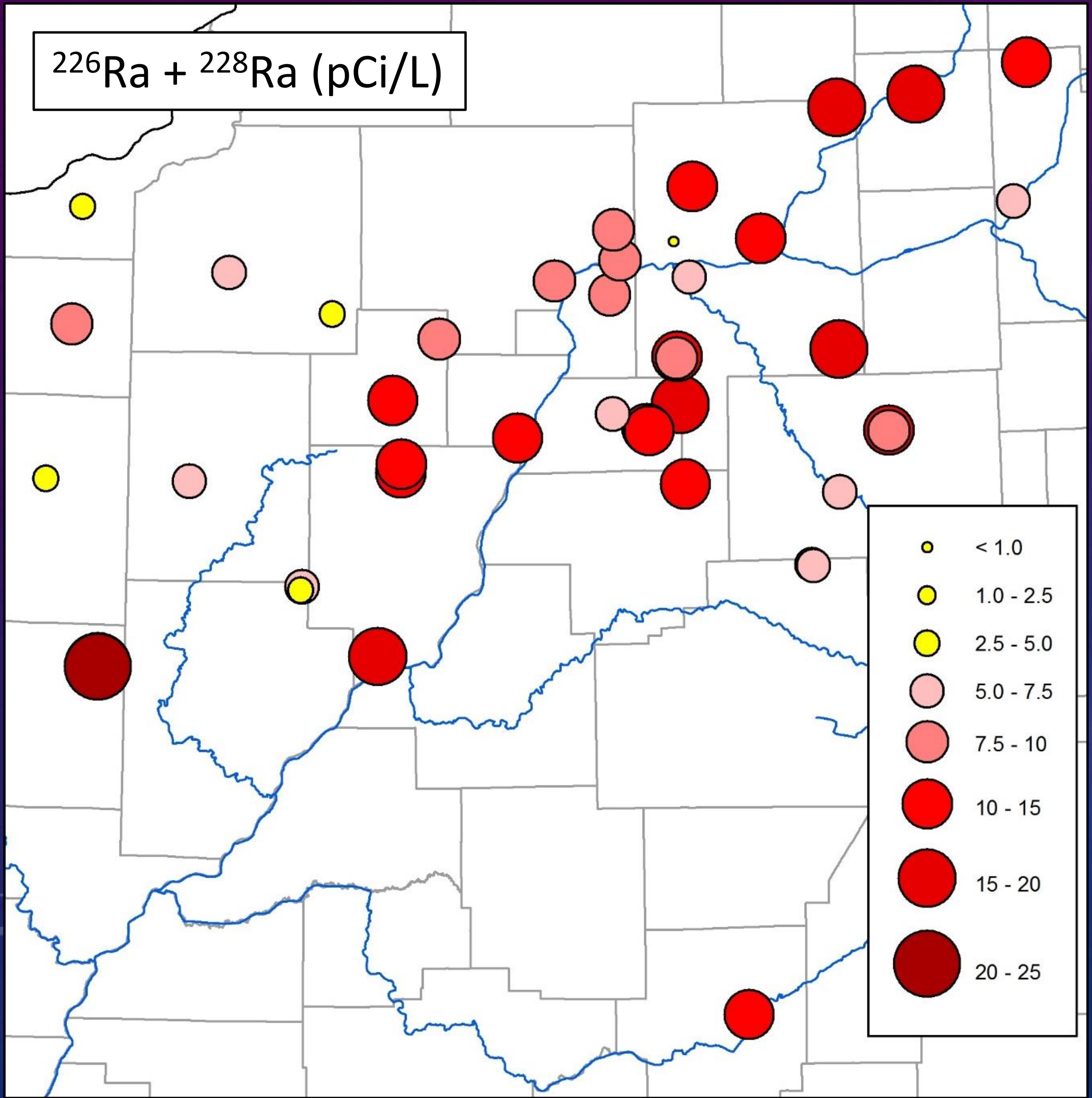
DECAY CHAINS THAT PRODUCE RADIUM

- ^{226}Ra and ^{228}Ra produced by different chains
- Enter solution via mineral dissolution, desorption, recoil
- Different parents may mean different solution processes
- Short half-lives: don't travel far from source
- Once in solution, both ^{226}Ra and ^{228}Ra should behave same



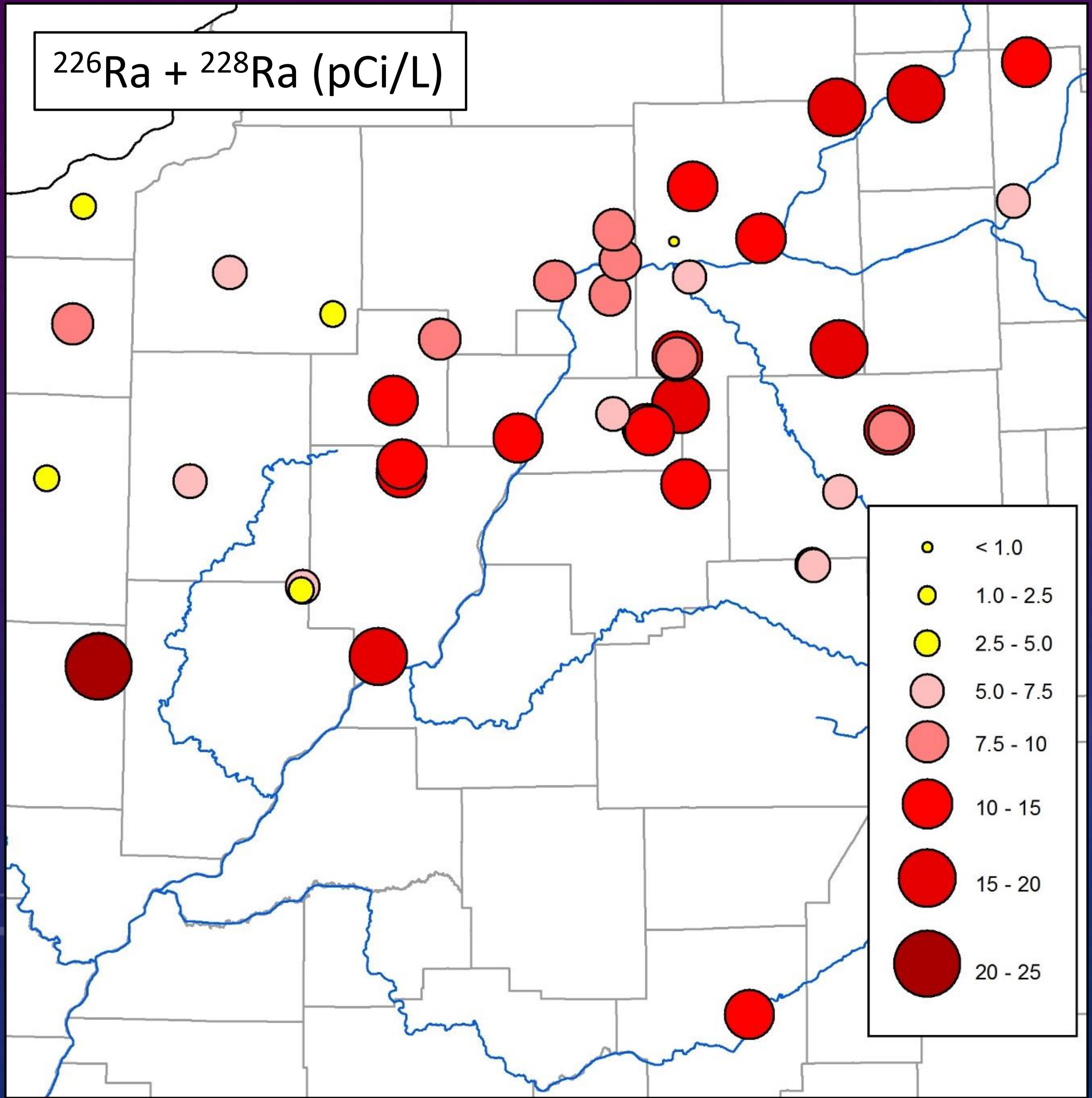


RADIUM

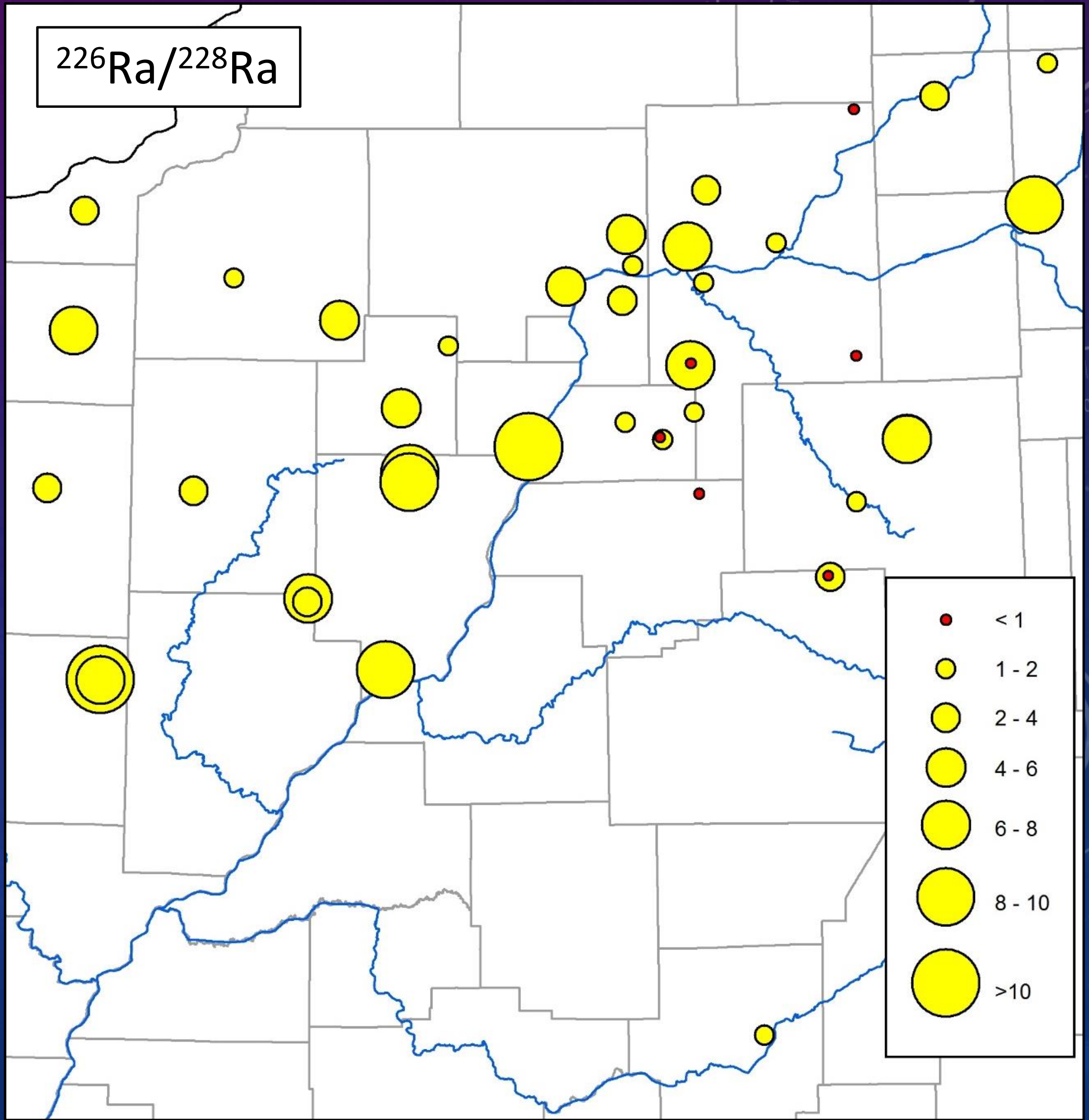


- Elevated throughout region, including recharge zone
- Brines or leakage not main source of Ra
- Ubiquitous sources

RADIUM



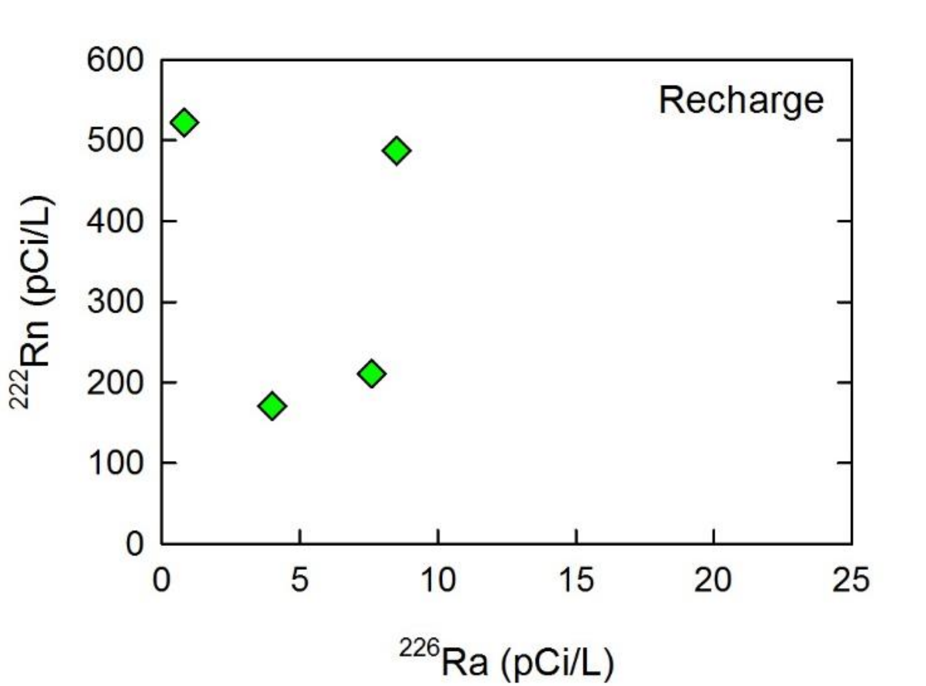
$^{226}Ra + ^{228}Ra$ (pCi/L)
< 1.0
1.0 - 2.5
2.5 - 5.0
5.0 - 7.5
7.5 - 10
10 - 15
15 - 20
20 - 25

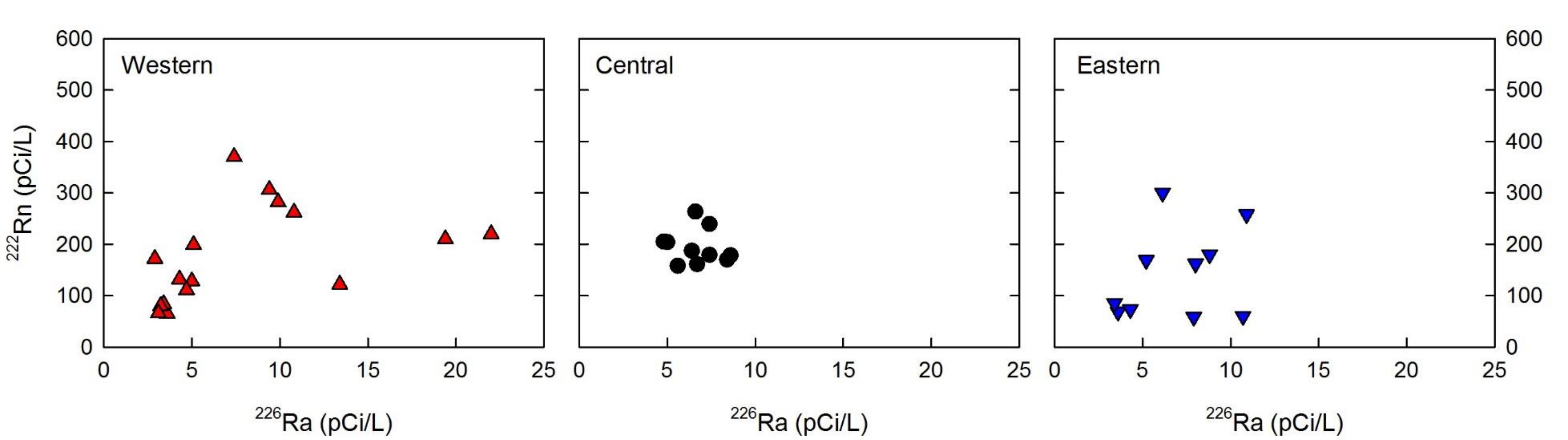


$^{226}Ra/^{228}Ra$
< 1
1 - 2
2 - 4
4 - 6
6 - 8
8 - 10
>10

RADON-222 VS. RADIUM-226

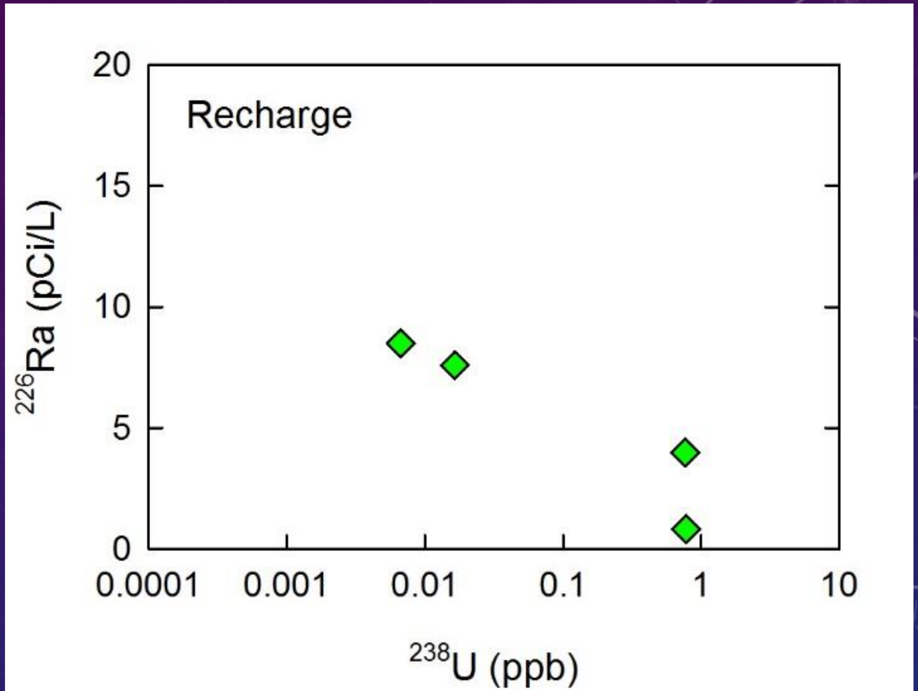
- ^{222}Rn very short half-life
- ^{222}Rn >10 times ^{226}Ra activity; ^{226}Ra mainly associated with solid phase

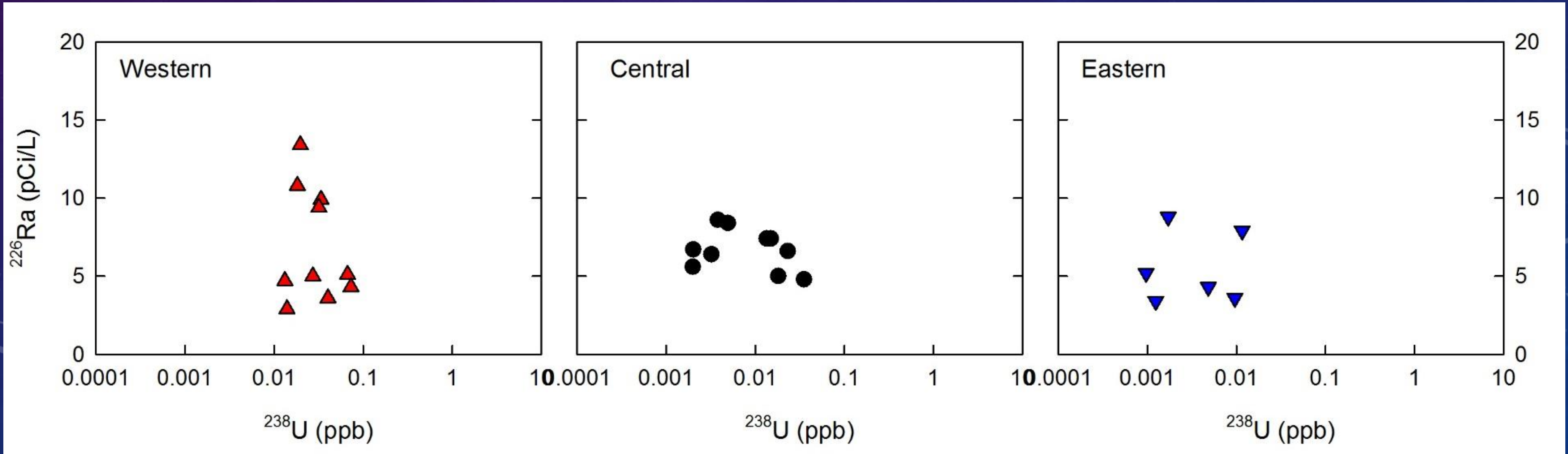




RADIUM-226 VS. URANIUM-238

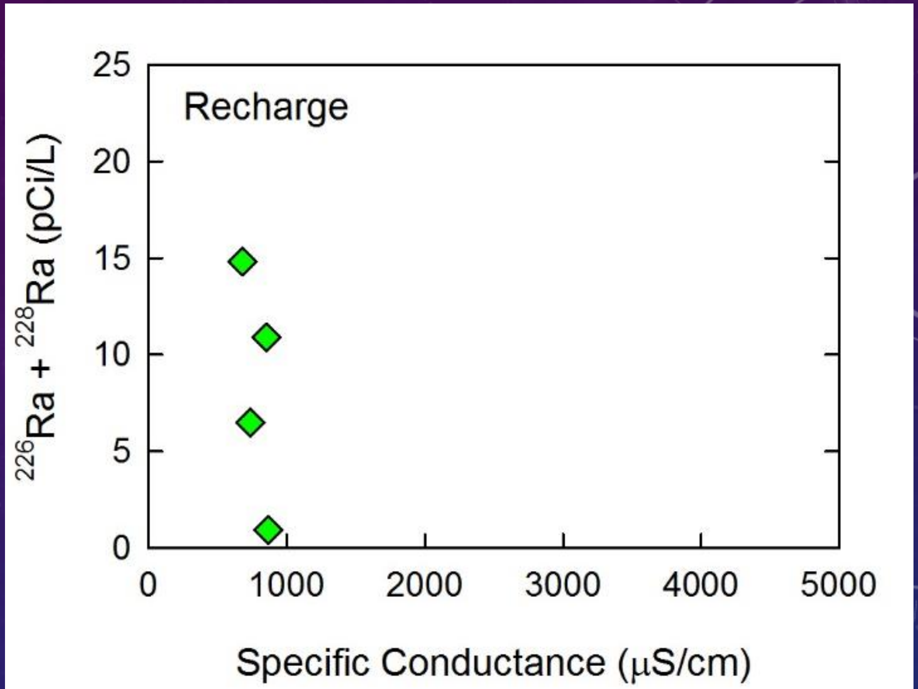
- Lack of correlation suggests ^{238}U also primarily associated with solid phase

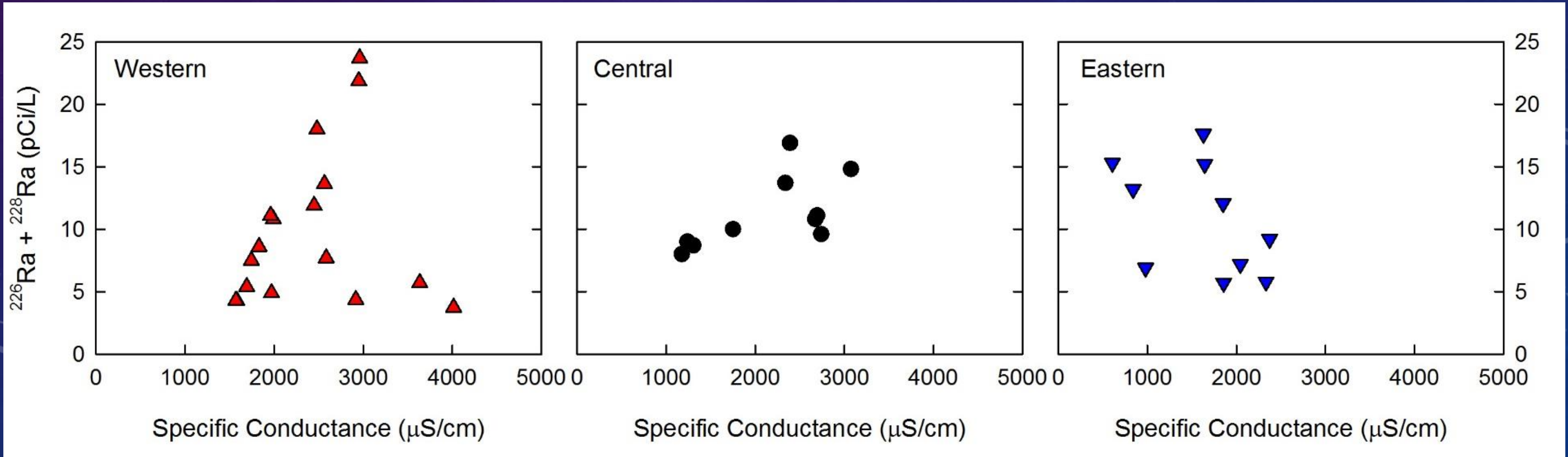




RADIUM-226 + RADIUM-228 VS. SPECIFIC CONDUCTANCE

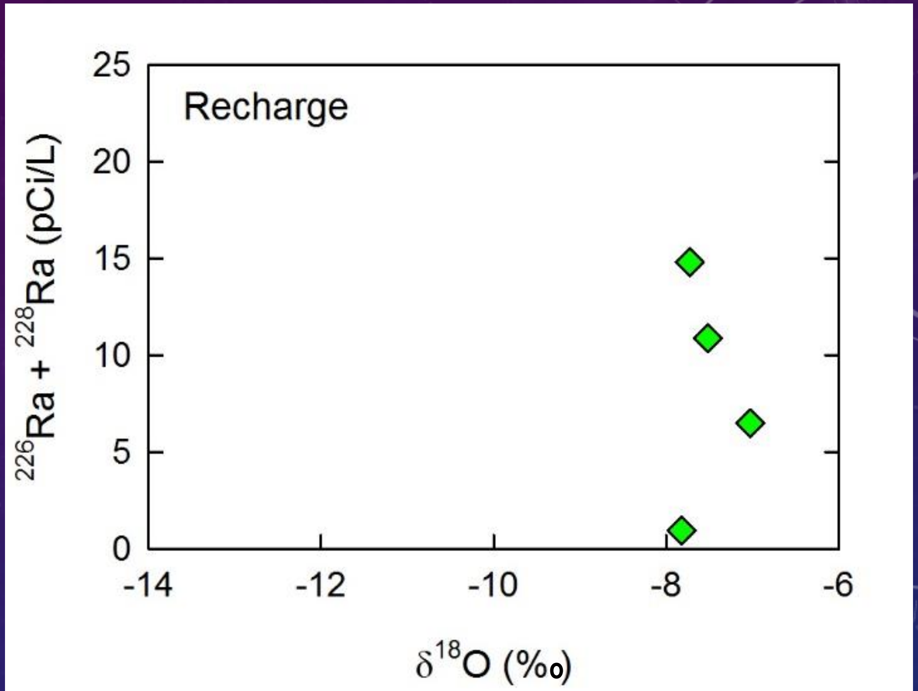
- Higher TDS may lead to increased Ra in solution due to increased exchange off surfaces

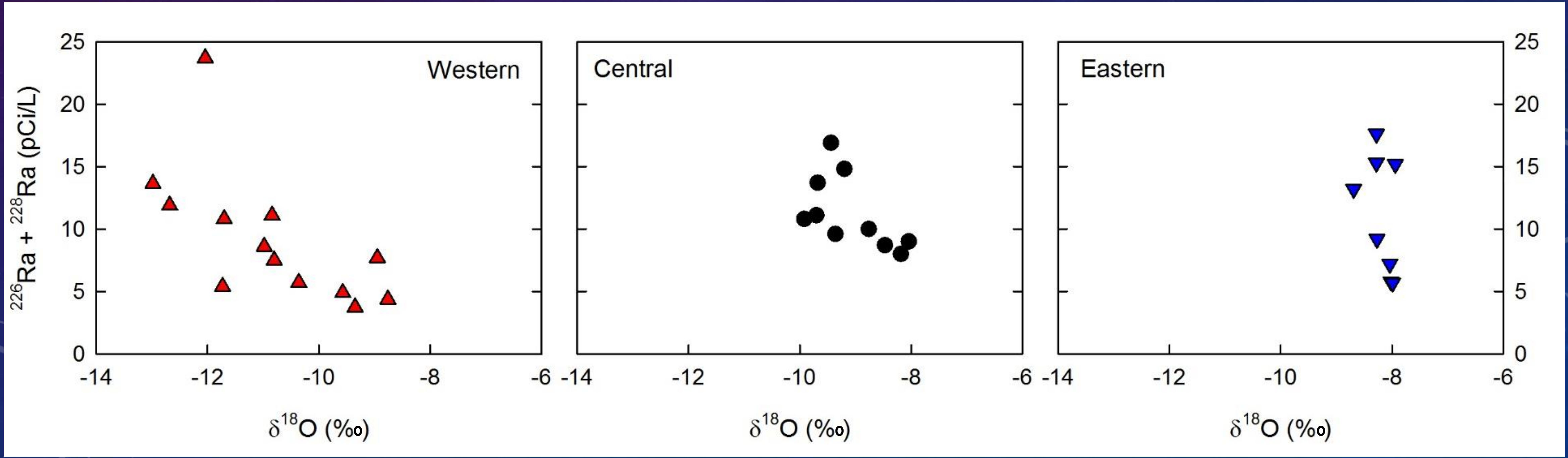




RADIUM-226 + RADIUM-228 VS. $\delta^{18}O$

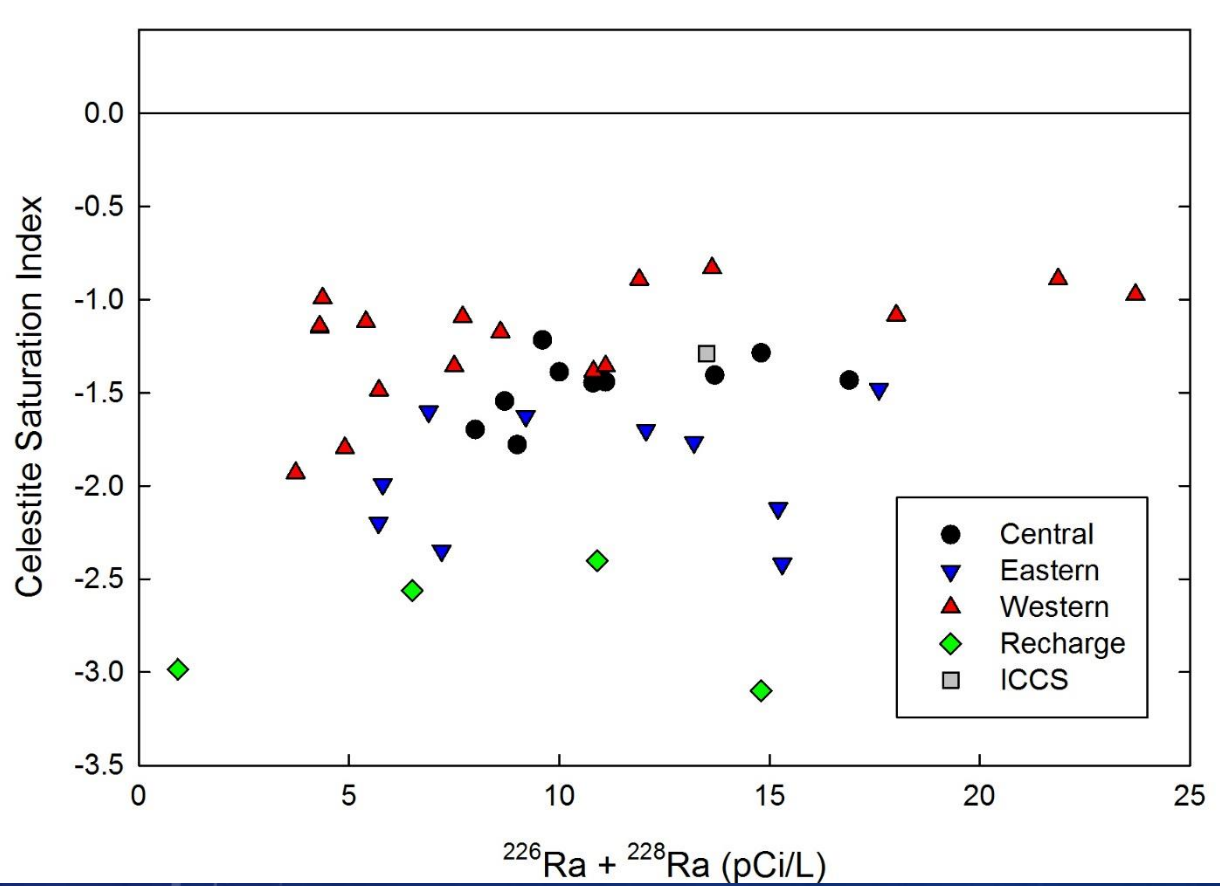
- Negative relationship suggests greater Ra where there is more meltwater recharge
- In situ brines not main source of Ra

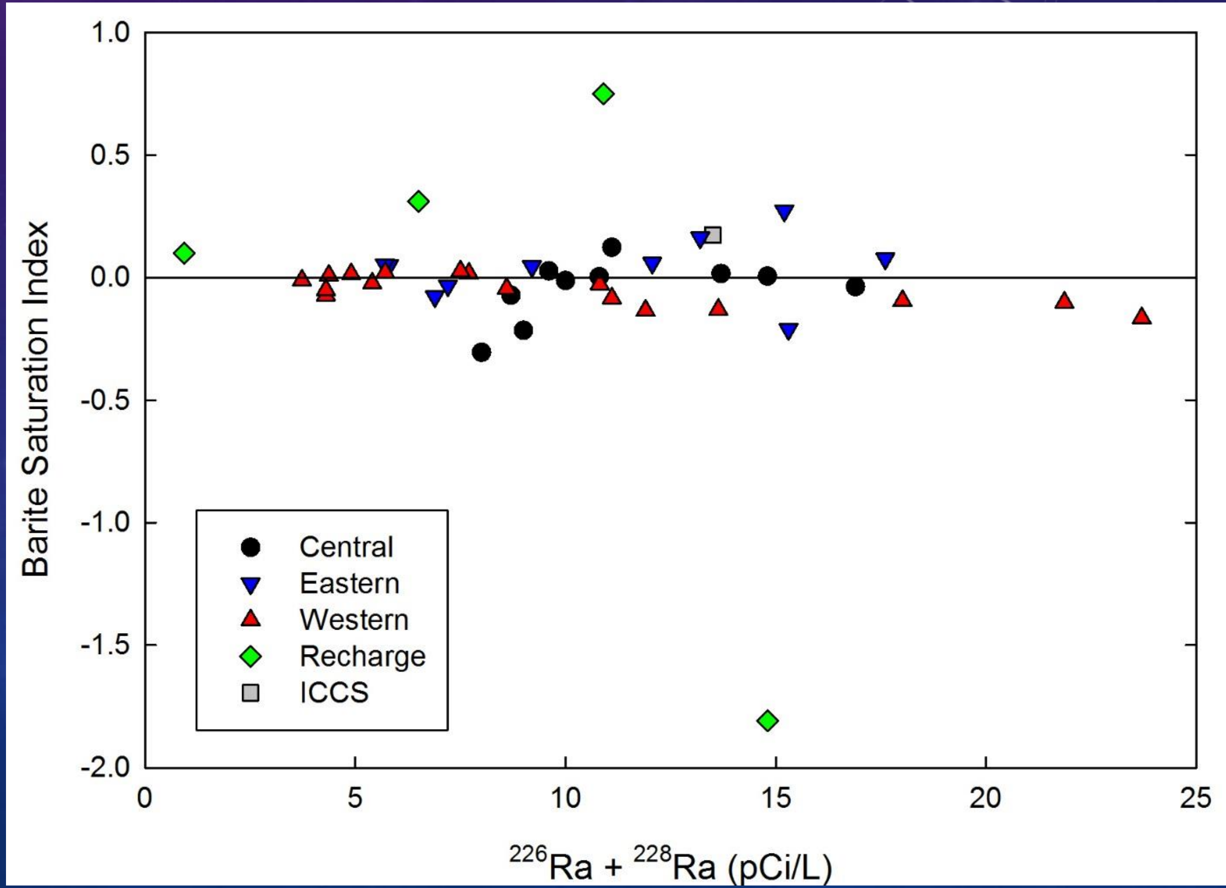




MINERAL SOLUBILITY CONTROL?

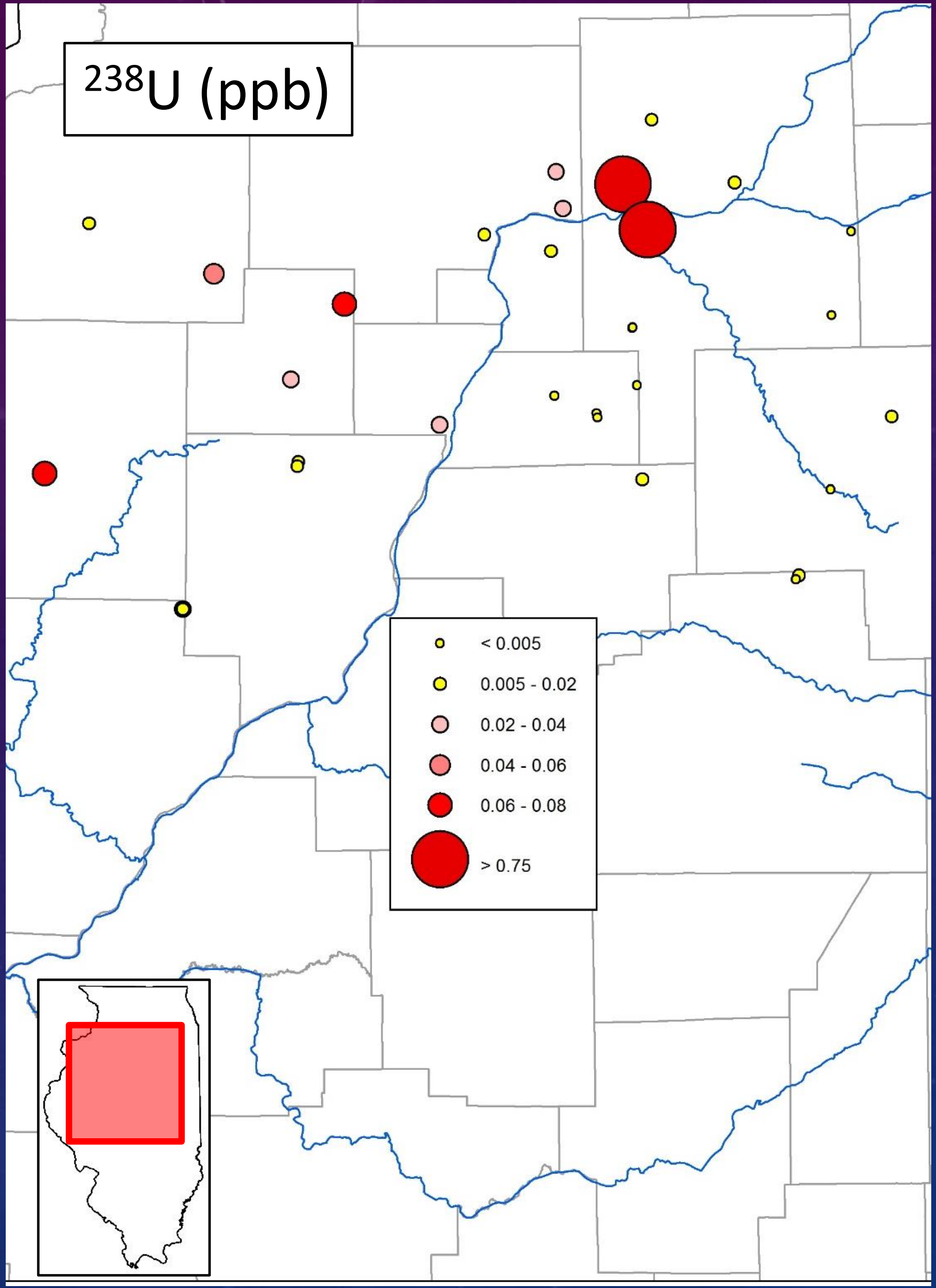
- Lots of sulfate in the St. Peter
- Some studies suggest barite ($BaSO_4$) or celestite ($SrSO_4$) can control Ra
- Adsorption likely more important

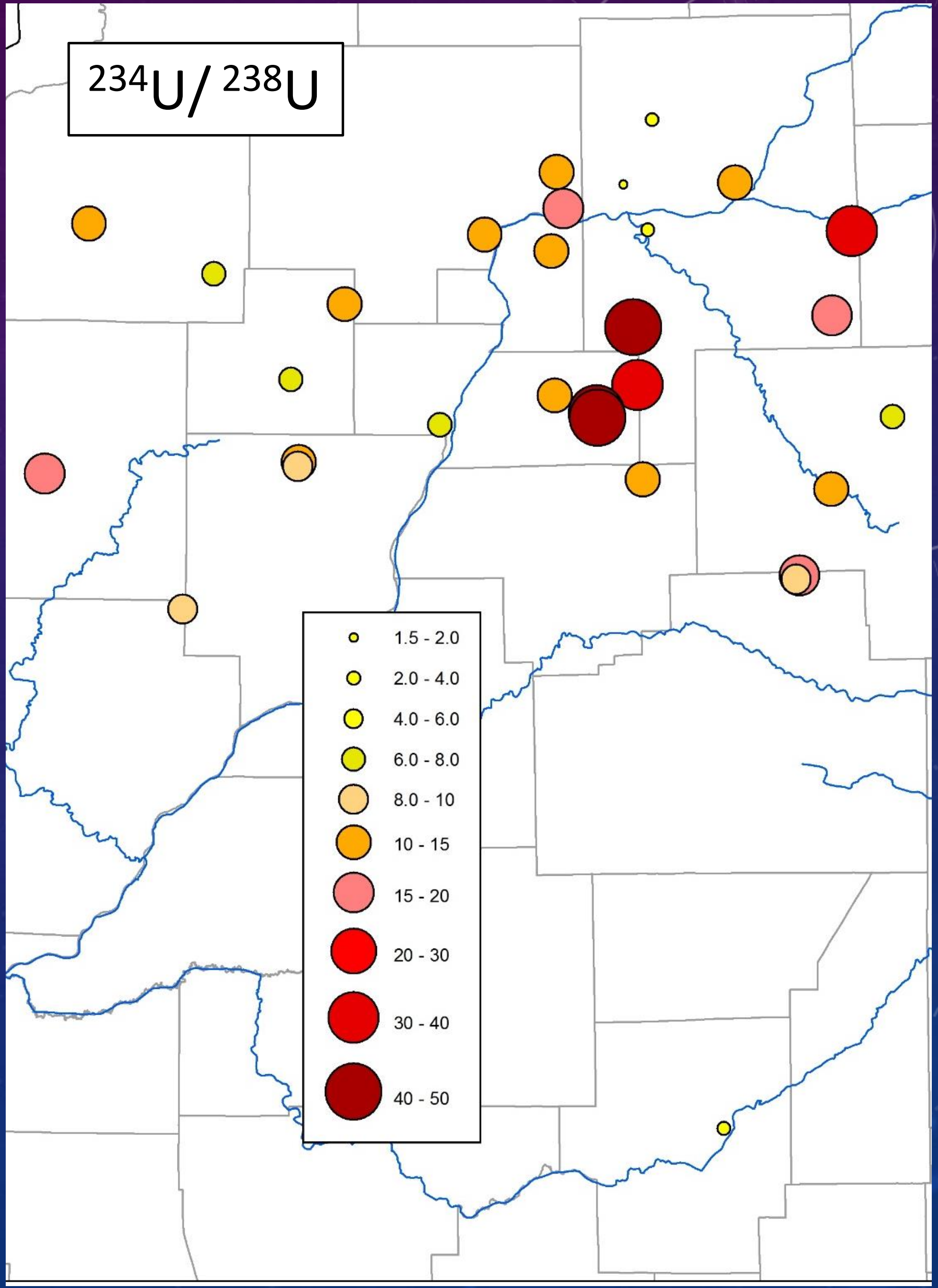




URANIUM

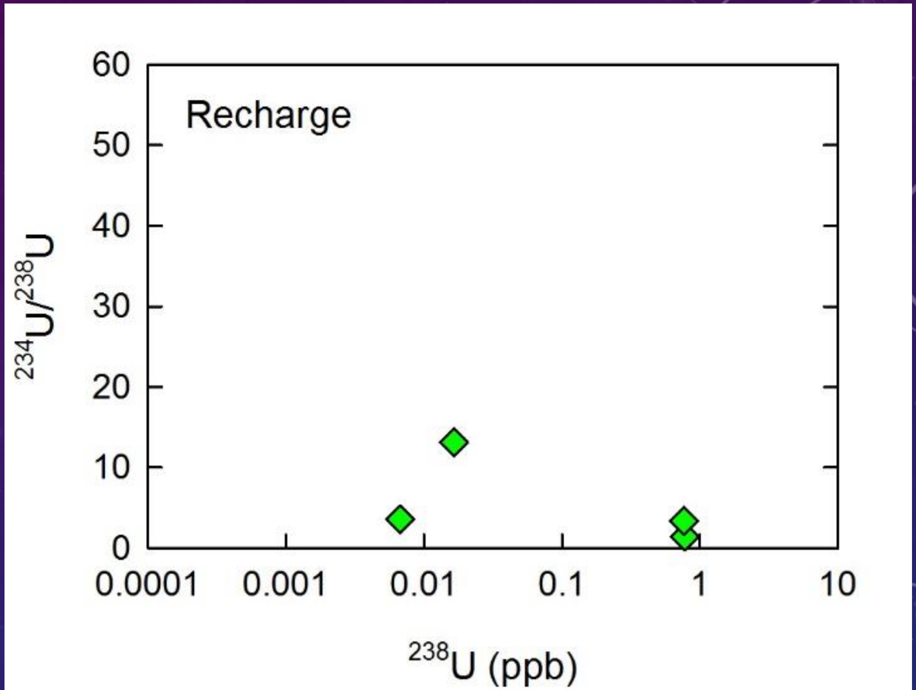
- Most < 0.08 ppb
- High concs. in recharge zone: oxidizing
- Extreme U isotope disequilibrium in central region

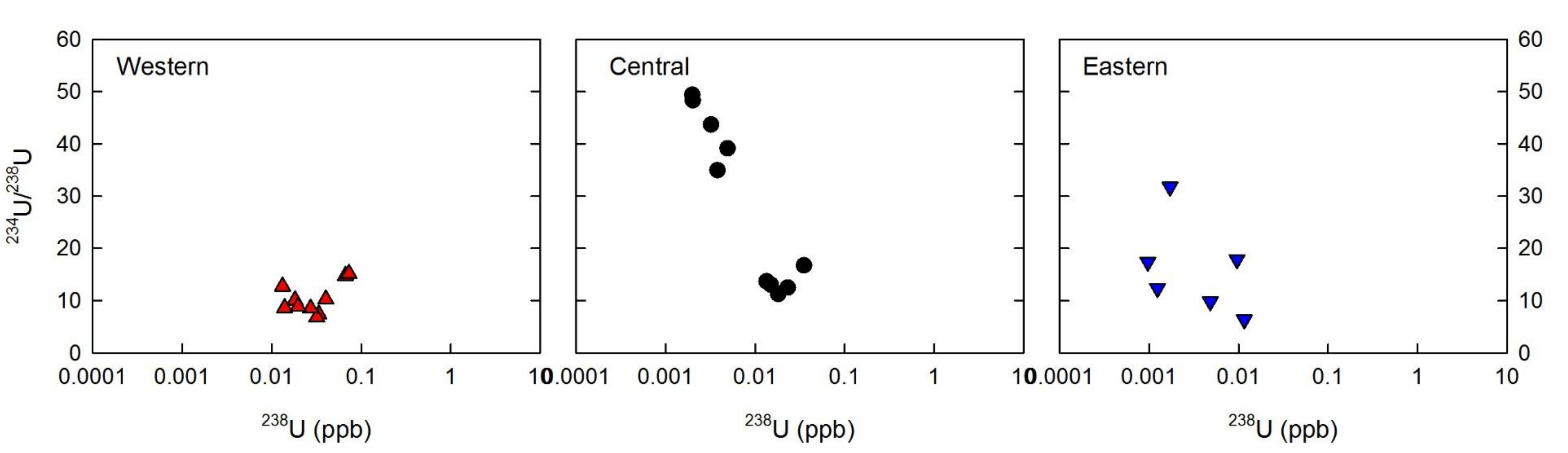




$^{234}U/^{238}U$ VS. ^{238}U

- Negative correlation Central and Eastern
- Disequilibrium must be relatively recent, otherwise decay toward equilibrium would have occurred
- Pulverized shales may be source of U
- Changing redox conditions may lead to enrichment in ^{234}U





SUMMARY

- Hydrogeological and geochemical conditions in the St. Peter Sandstone in central Illinois strongly affected by Pleistocene glaciation
- Structural features exert important control on flow paths
- Simple mixing of glacial meltwater and in situ brines cannot explain aqueous geochemistry
- Elevated radium found throughout the region
- Radium and parents (U, Th) primarily associated with surfaces/solid phases; continuous production of Ra and Rn to solution
- Influx of meltwater/fine-grained sediments may have brought U and Th into the St. Peter, and changing redox conditions may contribute to U disequilibrium