Use of Isotopic Tracers at the Hanford Site

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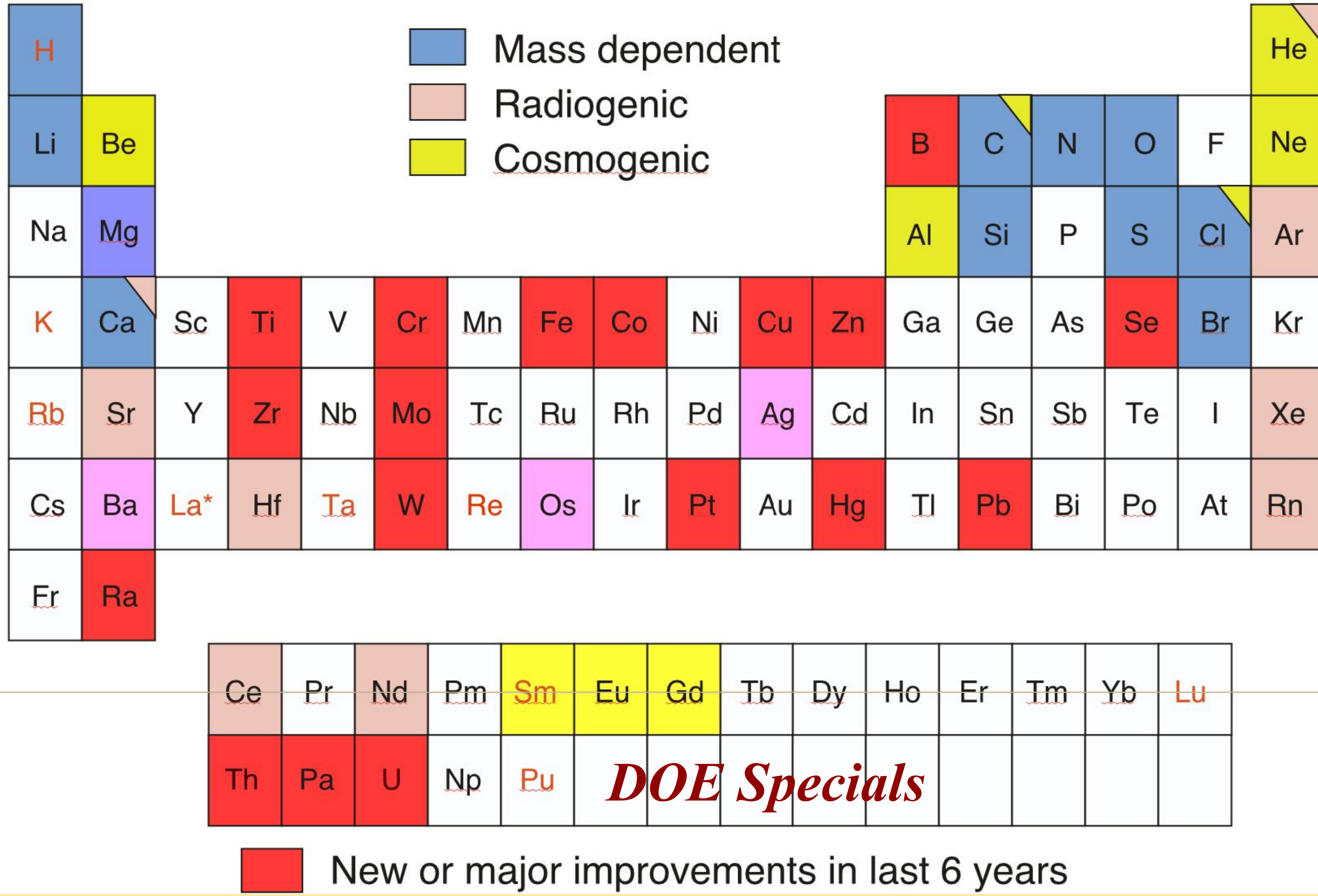
What can we do with isotopes?

- Tracers of sources and processes (natural and otherwise)
- Vadose zone infiltration and seepage rates
- Natural versus industrial sources of waters and contaminants
- Mineral-fluid reaction rates; fixation and retardation of contaminants
- Connecting vadose zone contamination with groundwater plumes (Cribs vs. trenches, spills vs. leaks, old vs. recent)
- Identifying sources of water as well as sources of contaminants (process water, Columbia R. water, natural water, evap. Ponds)
- Connecting groundwater plumes with Columbia River contaminants

ELEMENTS WITH NATURAL ISOTOPIC VARIATIONS



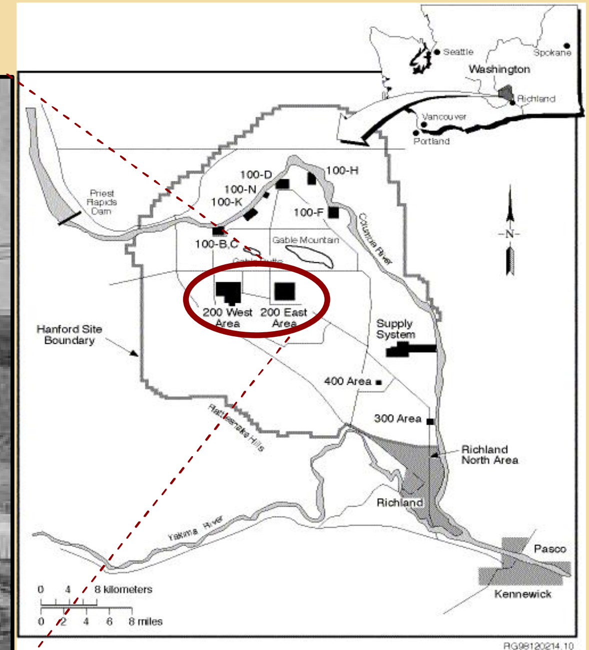
Research questions.....

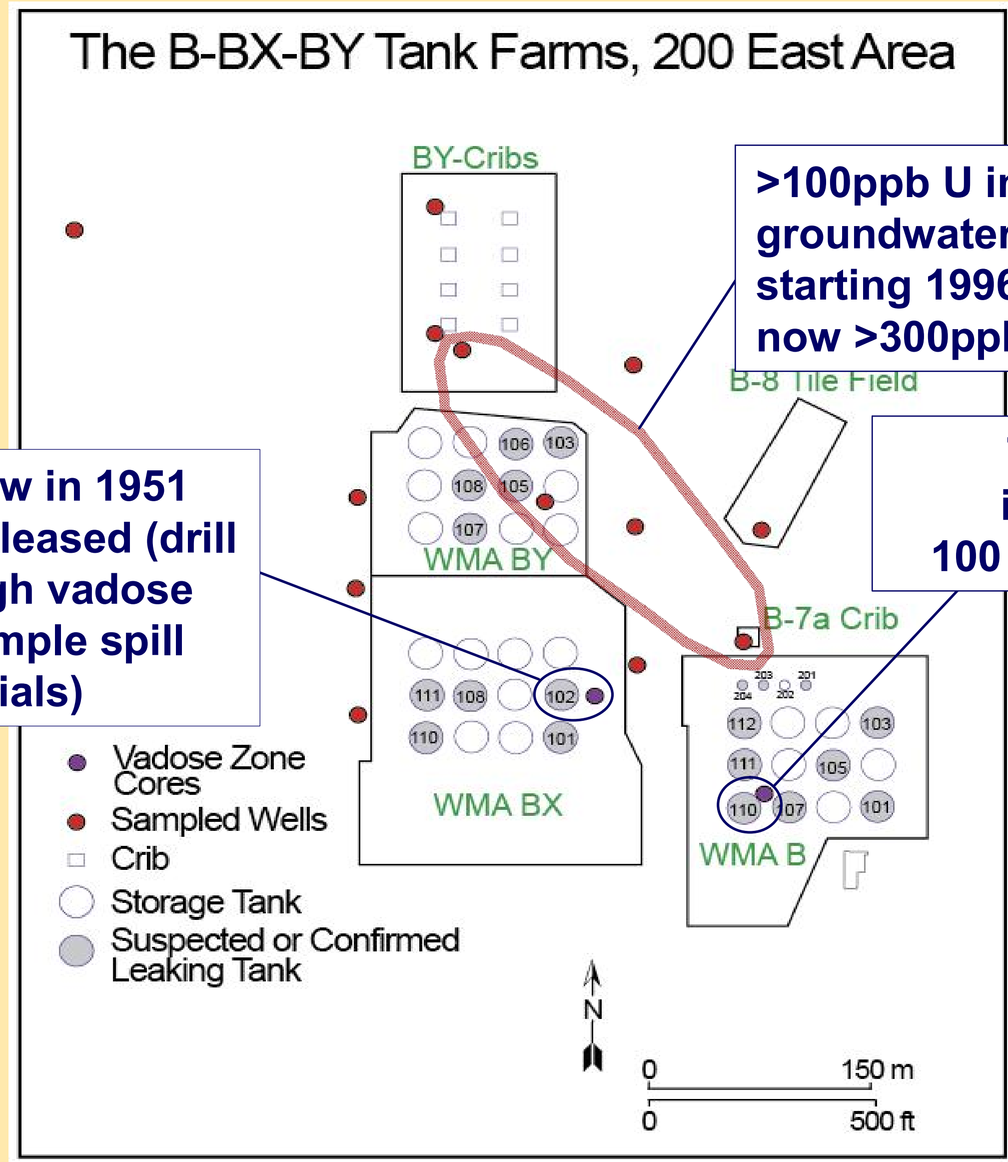
- **Are isotopic variations actually diagnostic in real life field situations?**
- **Can materials be sampled appropriately for isotopic characterization? Can studies piggy-back on standard characterization activities?**
- **Are background effects (natural variations) separable from contamination or other industrial modifications of the environment?**
- **What conceptual and mathematical models are necessary to properly interpret isotopic data?**
- **Which elements/isotopes show the most promise?**

Topics for this presentation

- **Contaminant tracing**
  - ***U isotopes, B-BX-BY tanks and groundwater***
  - ***N, O isotopes; T-TX tanks and groundwater***
  - ***U isotopes in Columbia River***
- **Vadose zone infiltration**
  - ***Sr-U isotope fluxmeter***
  - ***Distinctive O-H isotopes in VZ porewater***
- **Groundwater:**
  - ***Sr isotopes, weathering rates, VZ flushing***

Single-Shell Tank Farm Construction (1944)

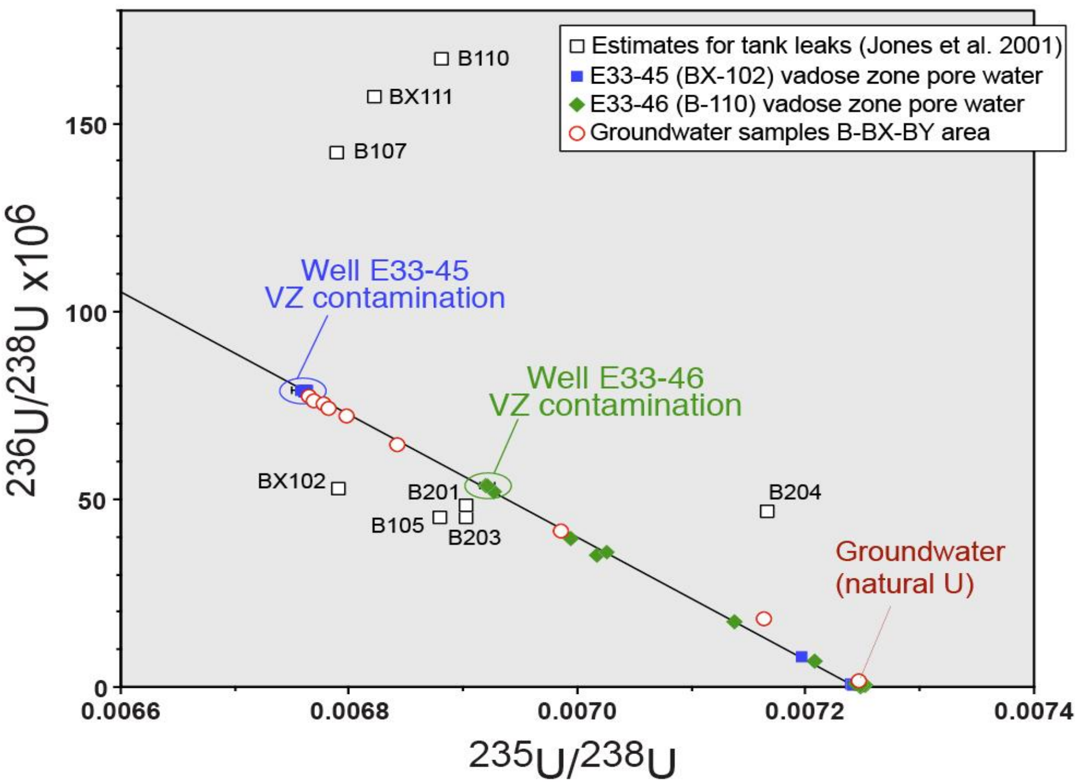


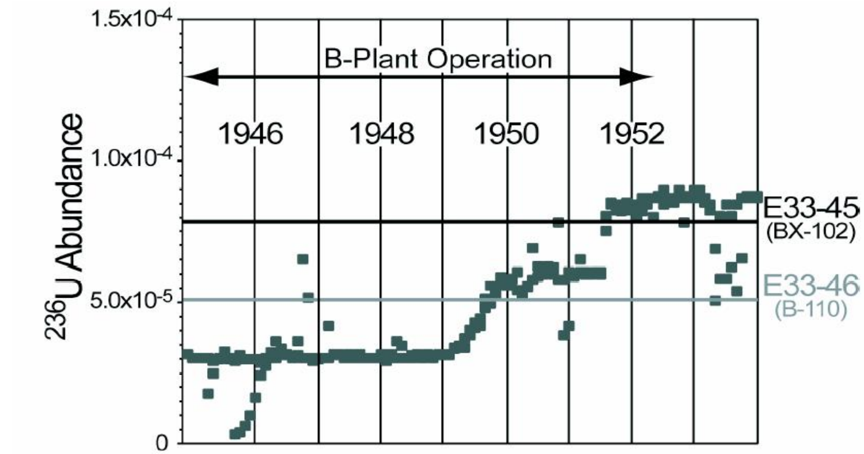


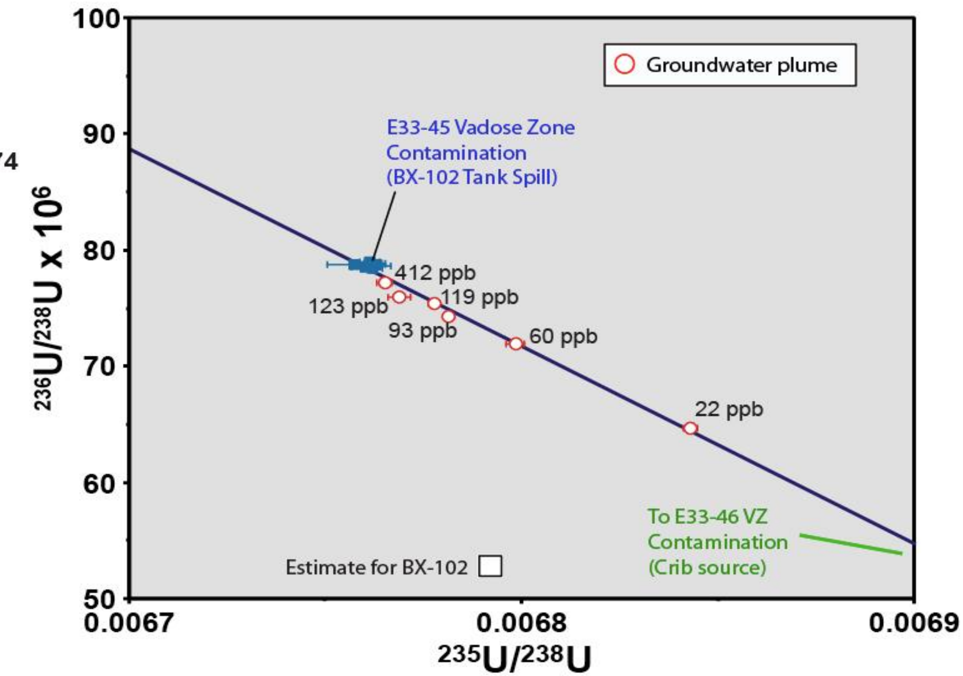
>100ppb U in groundwater starting 1996: now >300ppb

Big overflow in 1951
9 tons of U released (drill core through vadose zone to sample spill materials)

Tank Leak in 1965-69; 100 kg U released



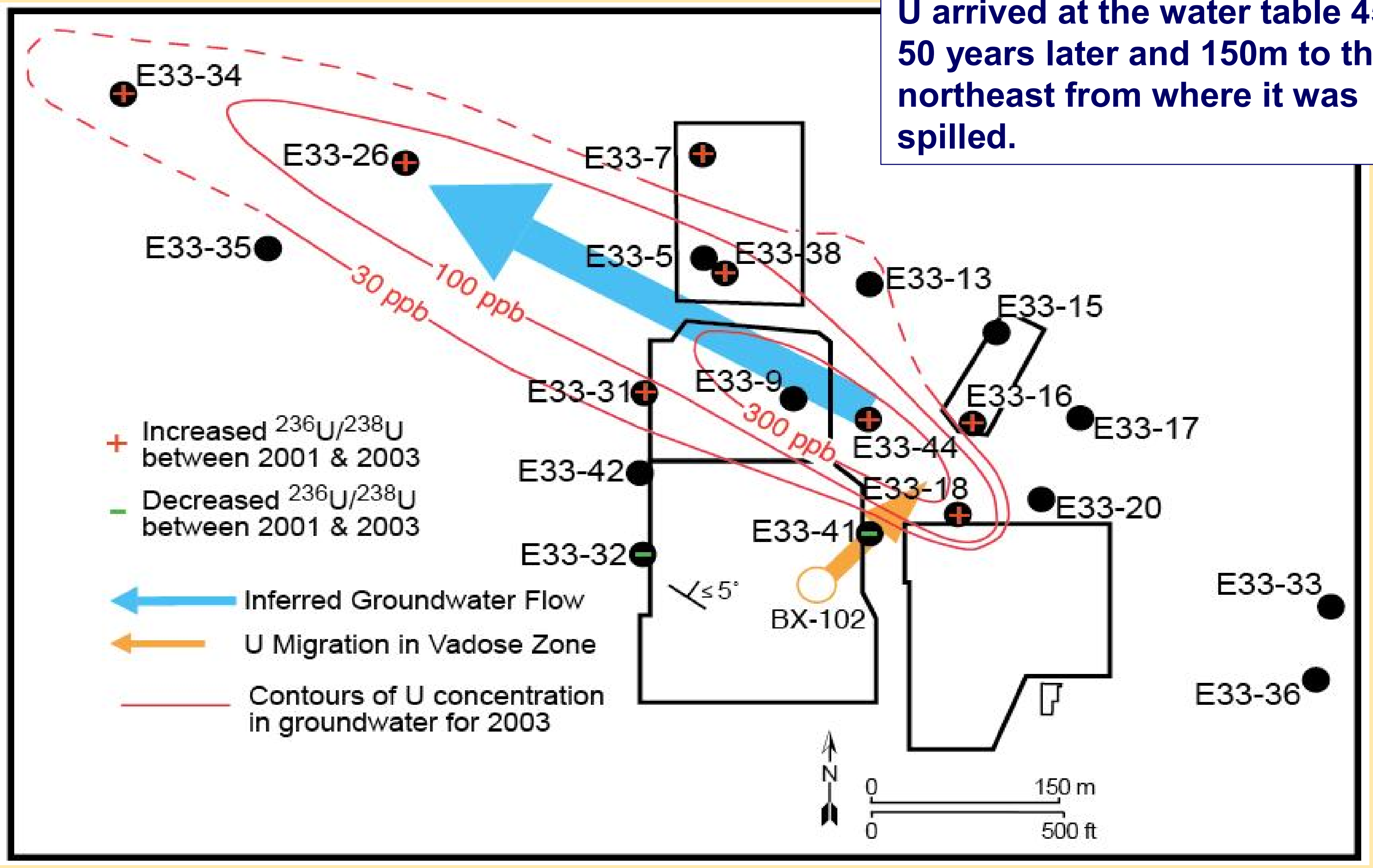




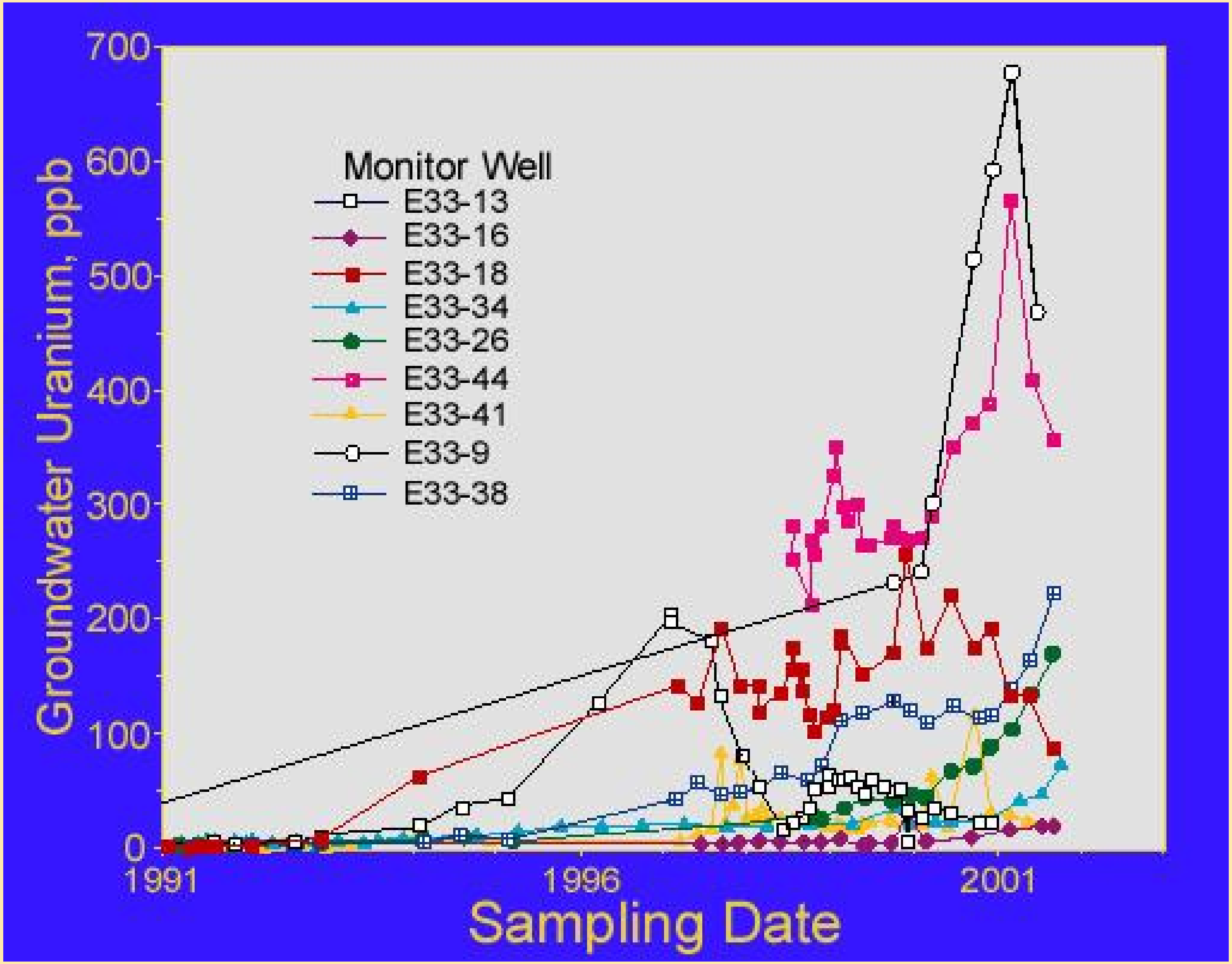
U isotopes in the groundwater plume exactly match those in the vadose zone under the BX-102 spill location, and are consistent with the U isotopes in waste from the B plant at the time of the spill.

Christensen et al., Env. Sci.& Tech (2004)

U arrived at the water table 45-50 years later and 150m to the northeast from where it was spilled.



Delivery of the U to groundwater is complex....



Use of N and O Stable Isotopes for Sources of Nitrate

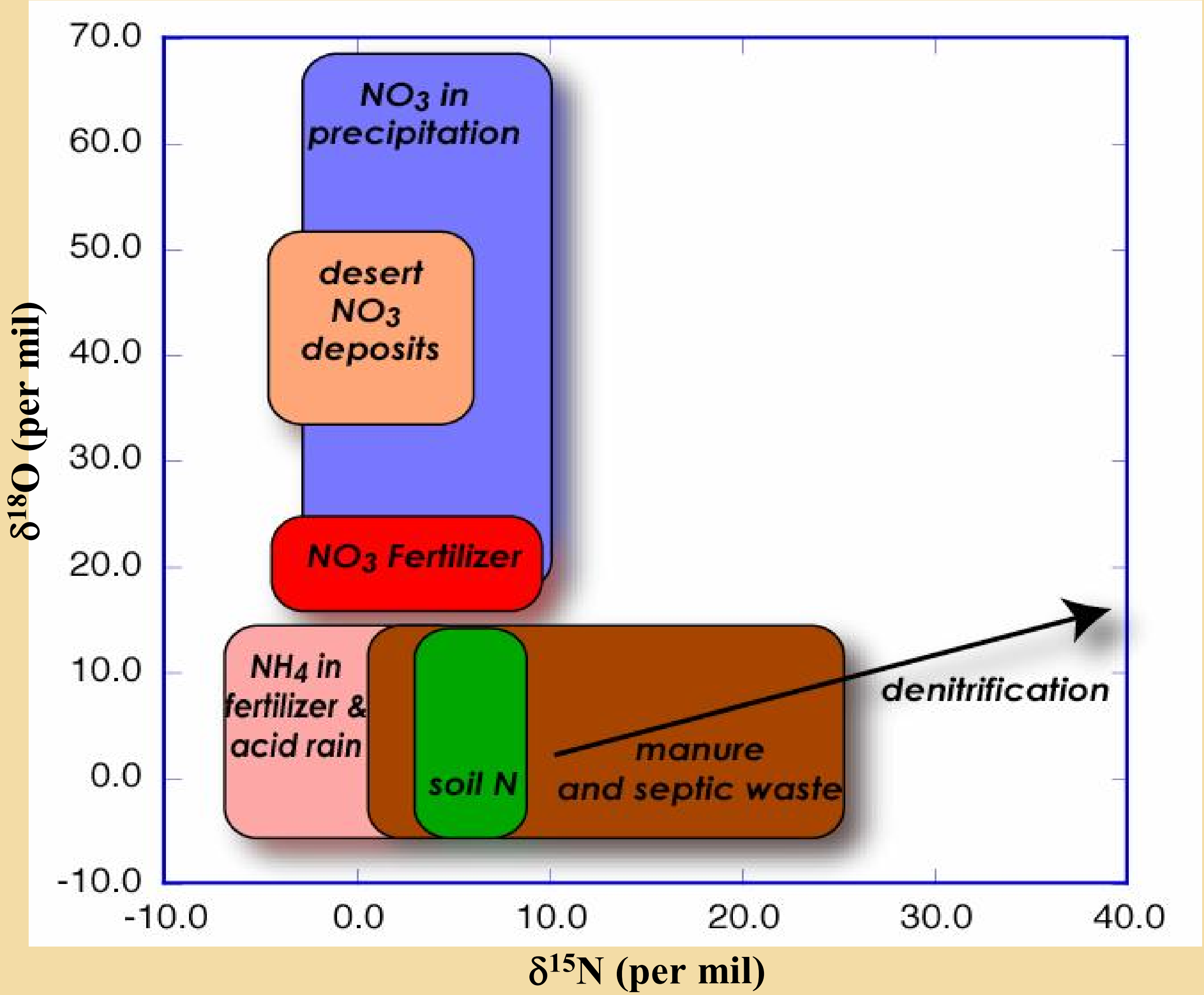
Nitrate is a common contaminant, but……

•High natural backgrounds, especially in arid and semi-arid environments.

•Widespread use as fertilizer.

•Byproduct of animal waste.

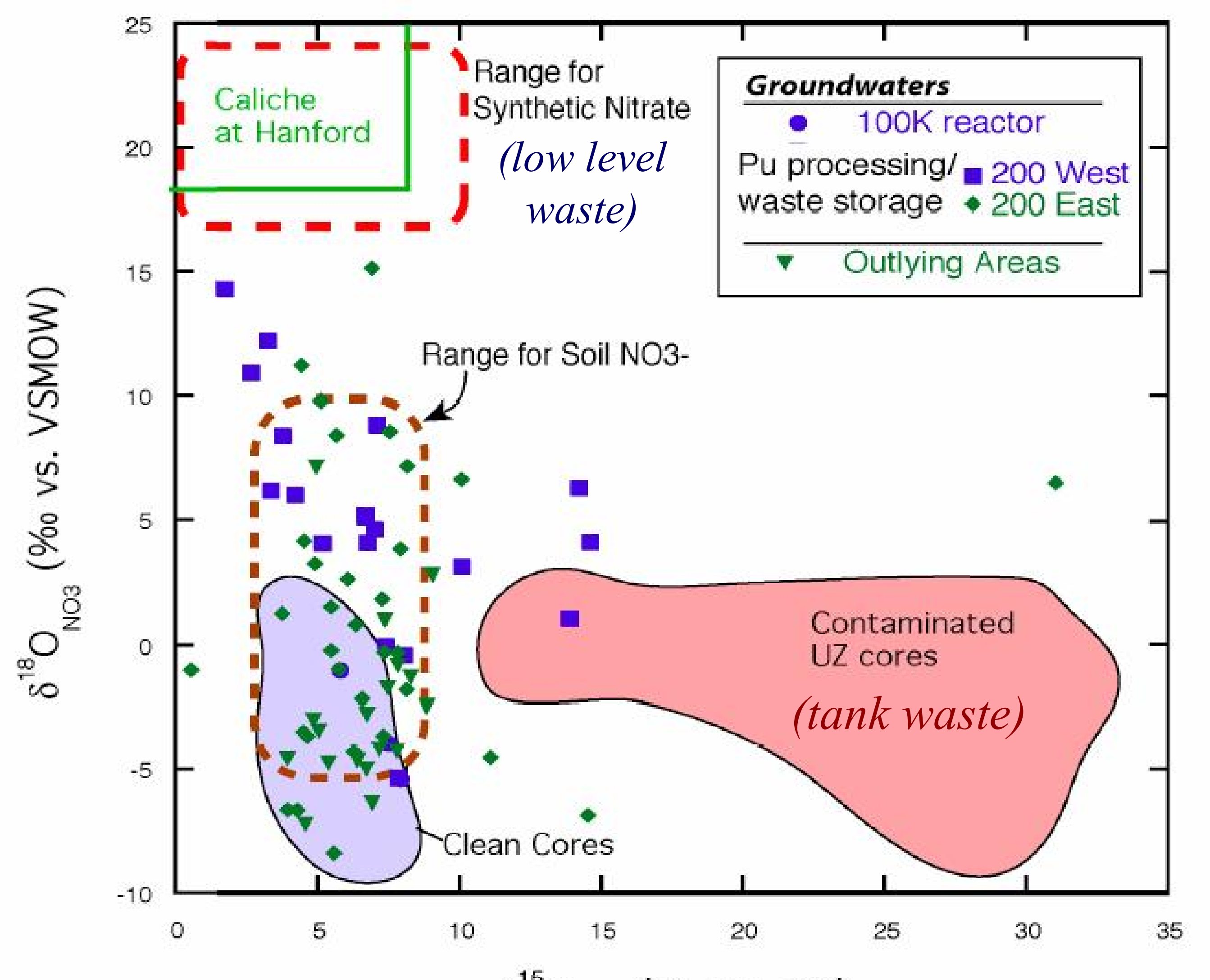
•Common constituent of chemical processing



(modified from Kendall, 1998)

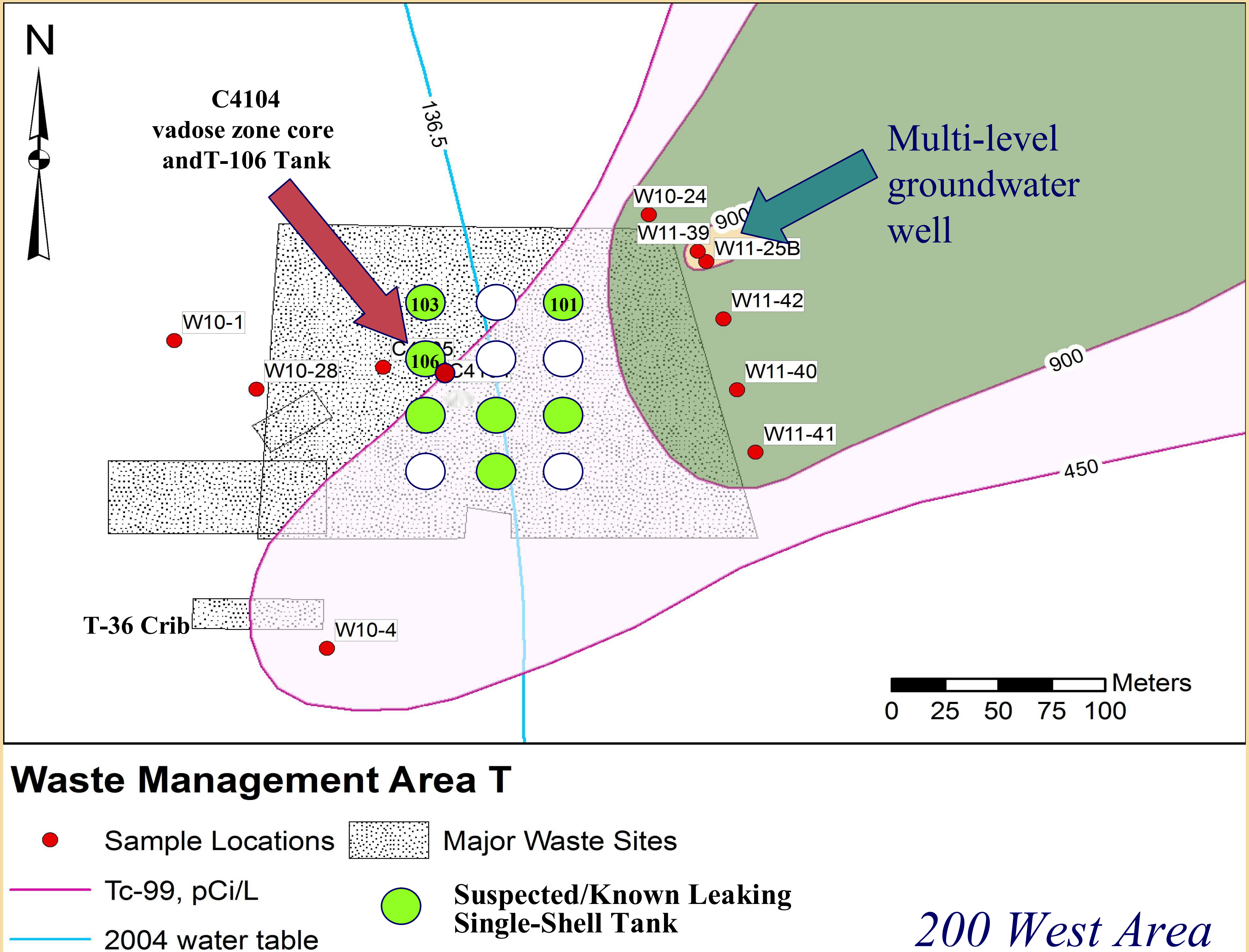
At Hanford:

Nitrate in high level waste from tanks is distinguishable from soil nitrate, low level waste nitrate, and natural vadose zone nitrate



Singleton et al., ES&T, 2005

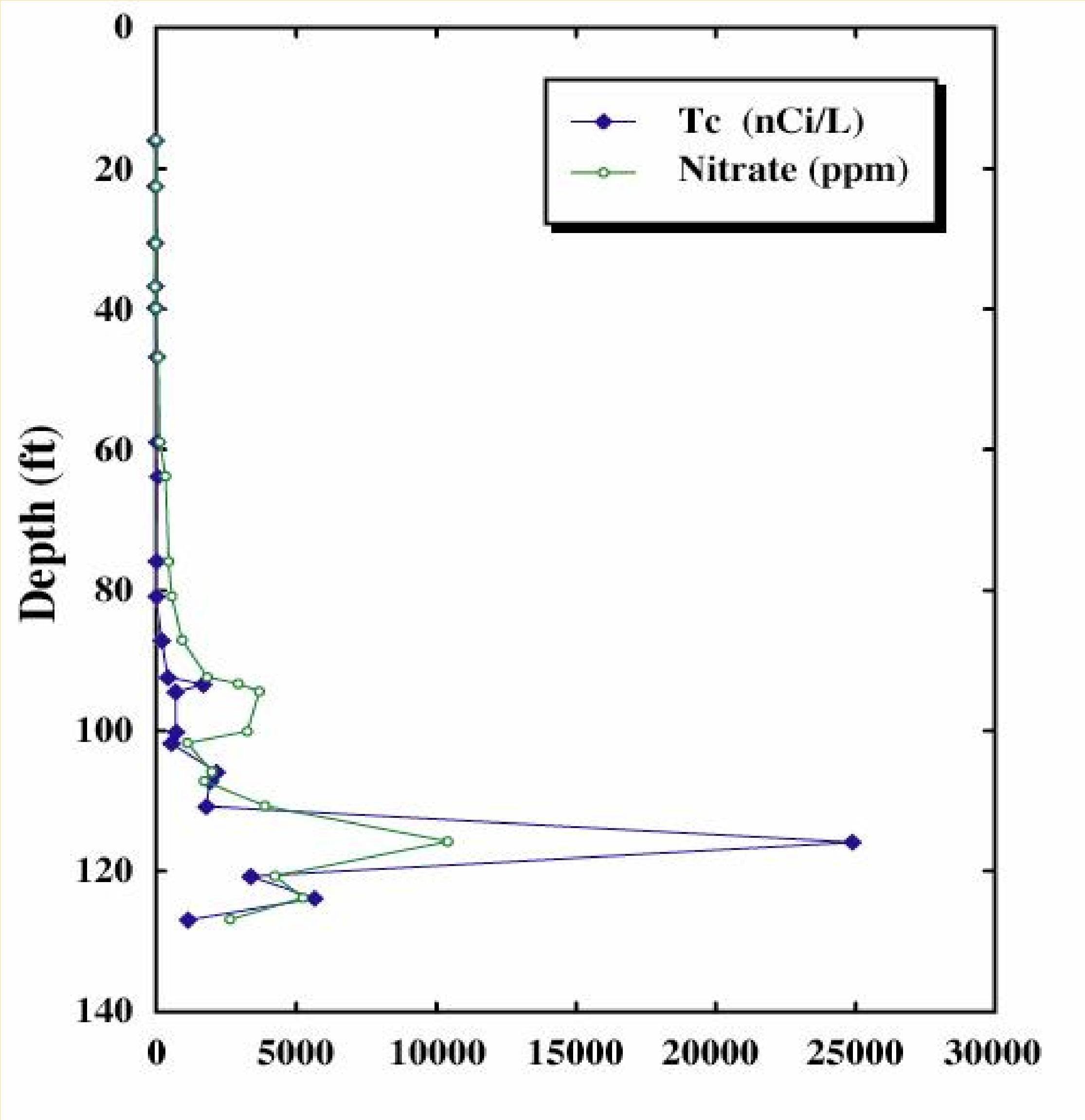
Nitrate is involved in the ^{99}Tc Plume Near the T- Tank Farm



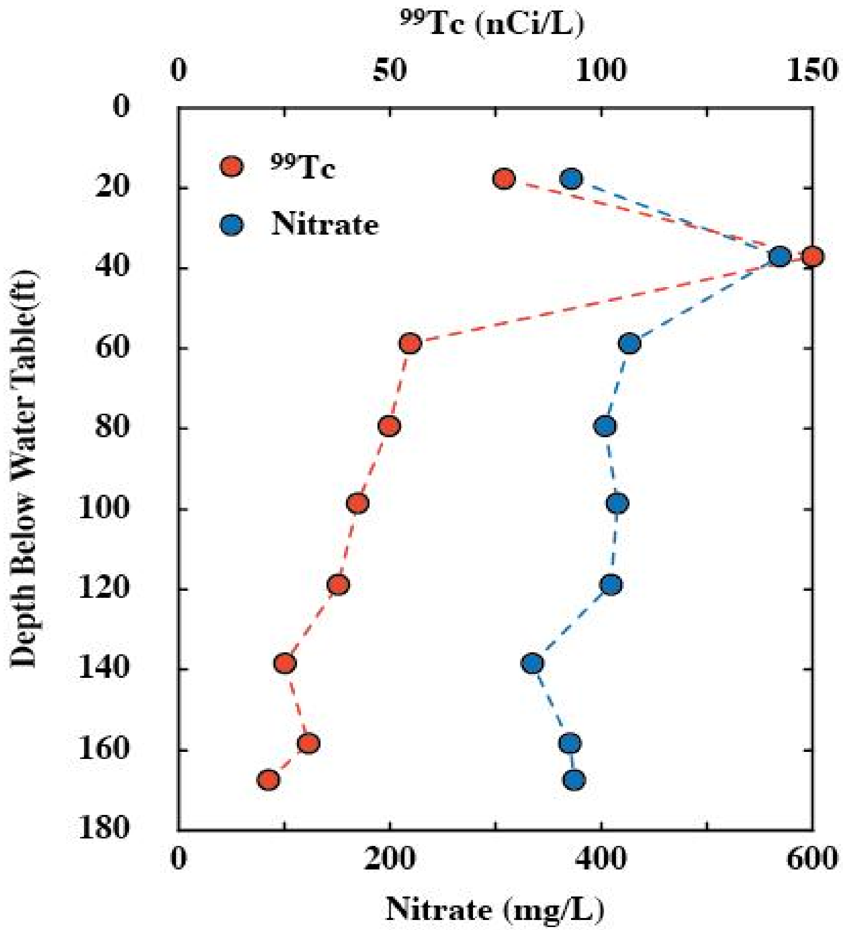
^{99}Tc and Nitrate in C4104 Pore Water (near T-106 Tank Leak)

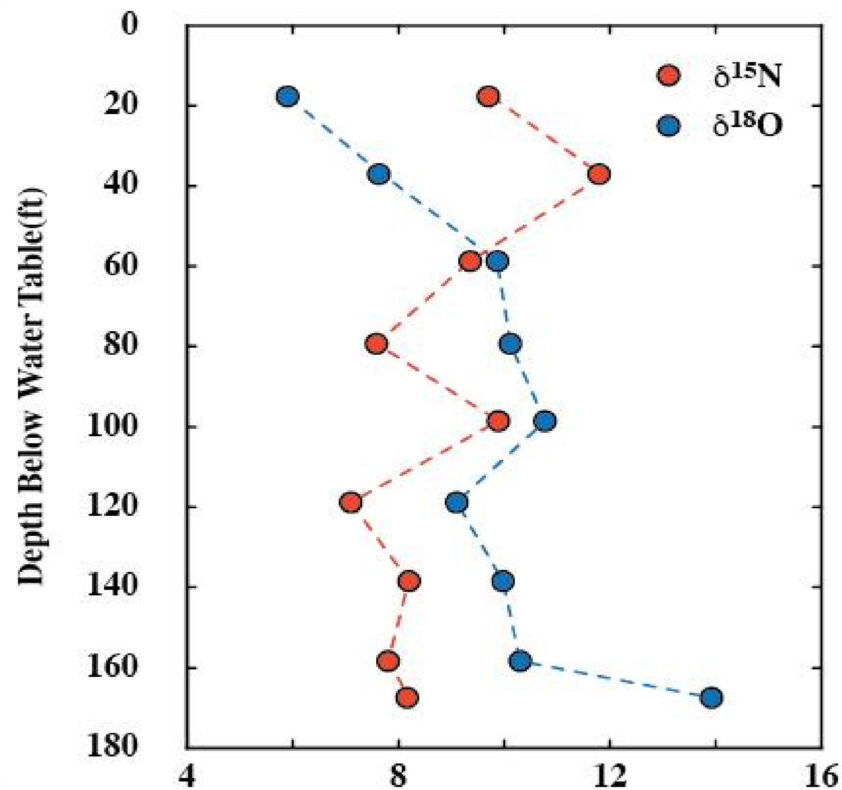
Nitrate is clearly associated with ^{99}Tc in vadose zone core.

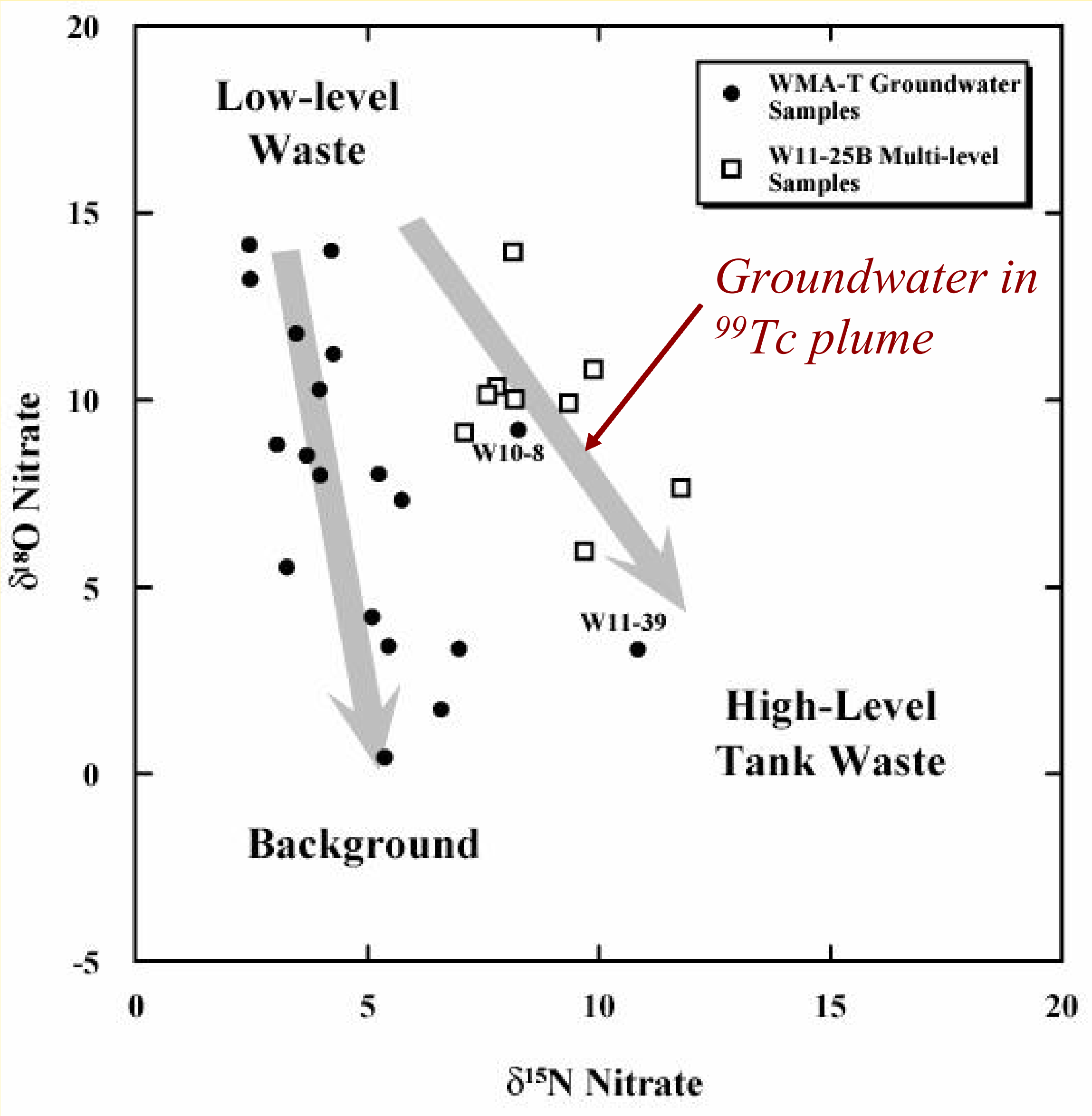
Nitrate has high δ^{15}N (tank waste signature)



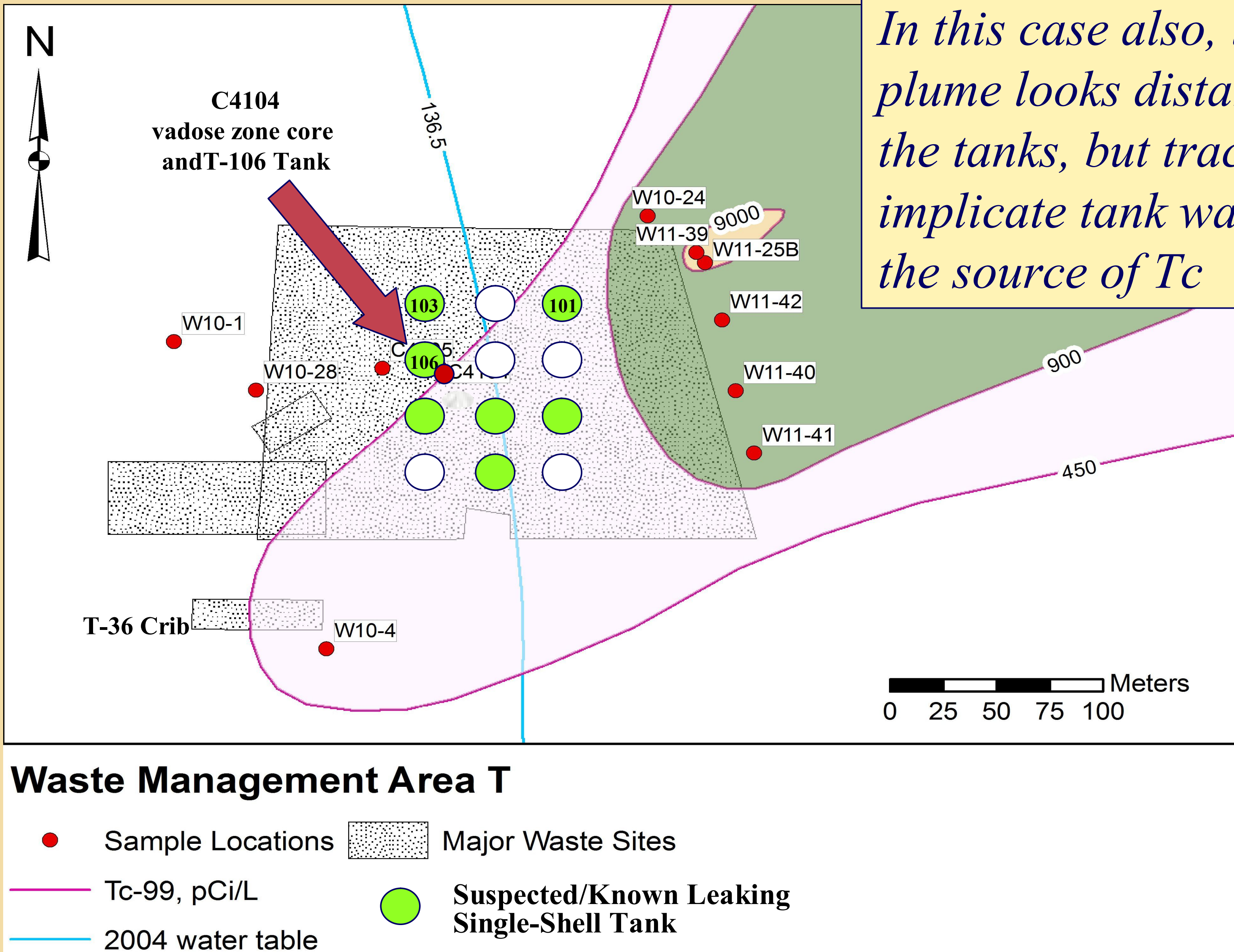
In groundwater plume, nitrate is also associated with ^{99}Tc, nitrate has high $\delta^{15}N$ + low $\delta^{18}O$ (tank waste signature)





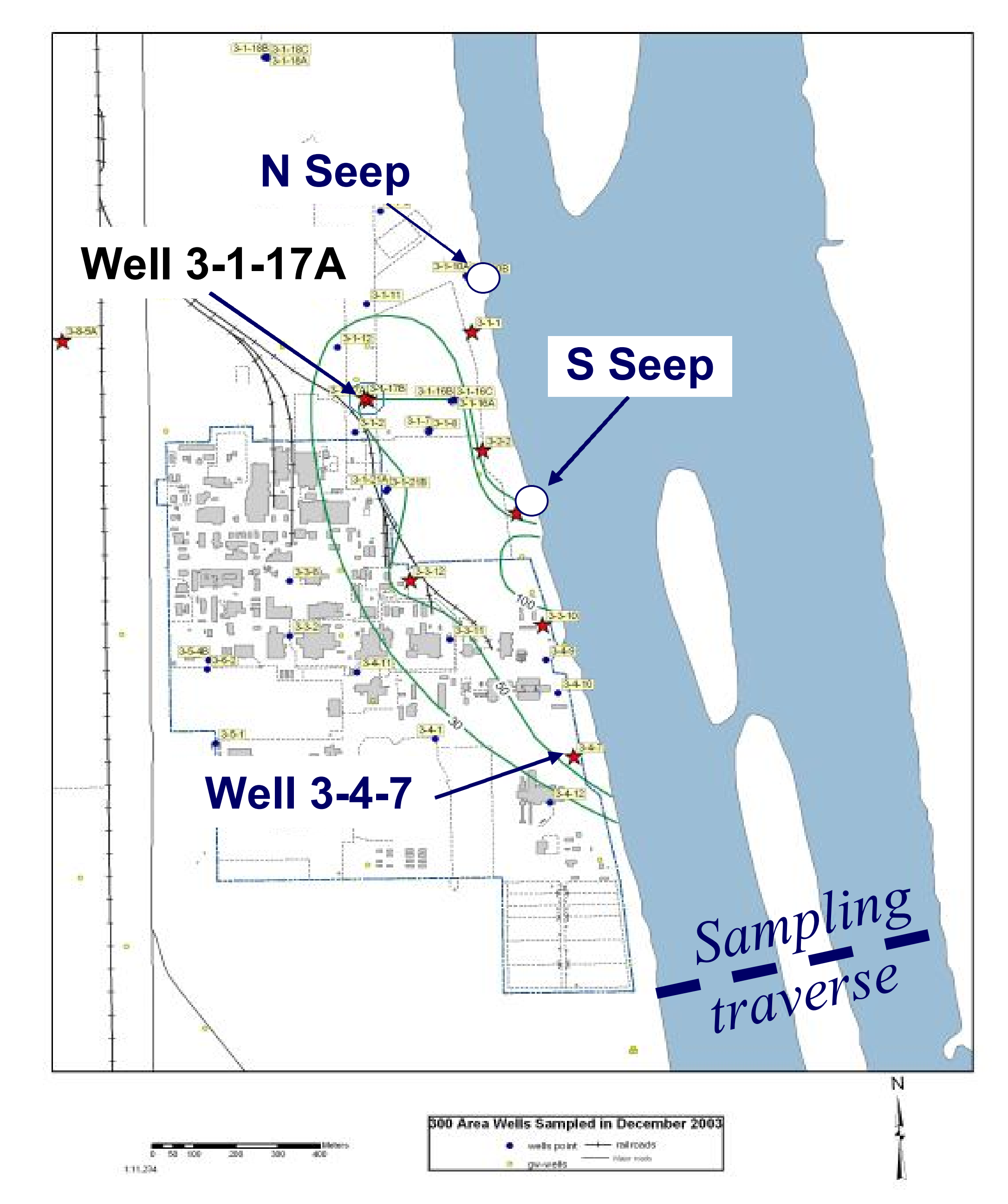


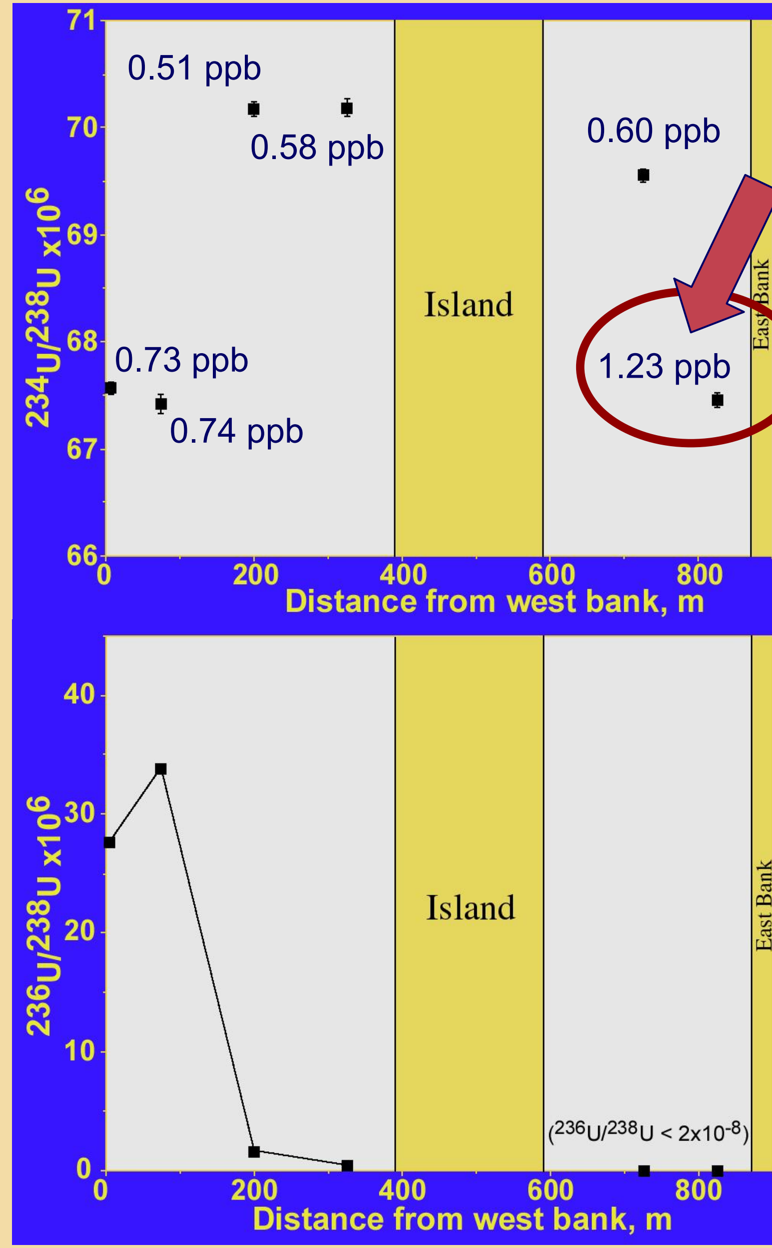
Groundwater in ^{99}Tc plume has a large tank waste nitrate component; but other groundwater samples in the T-TX tank farm area have nitrate from low-level waste and natural sources



In this case also, the plume looks distant from the tanks, but tracers implicate tank waste as the source of Tc

300 Area plume and U in the Columbia River

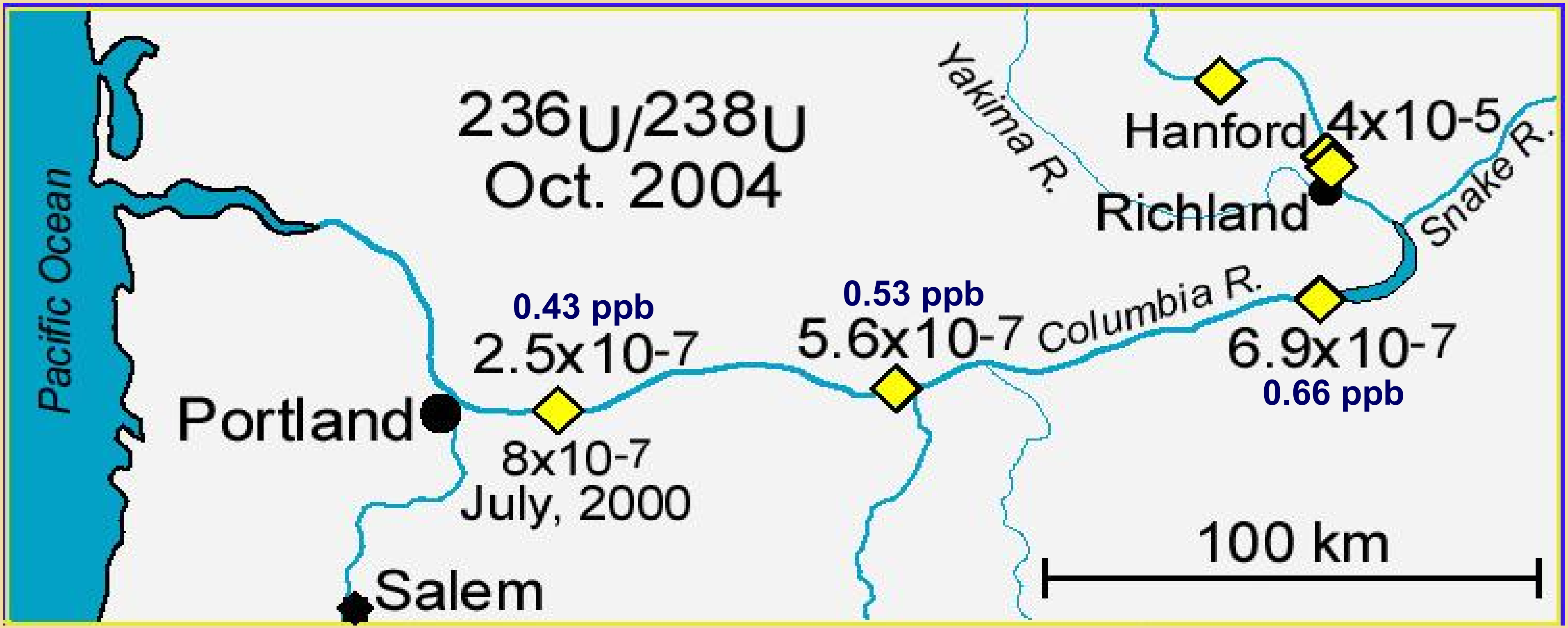




Flux of Contaminant U to the Columbia River varies seasonally (based on ^{236}U)

Fall 2003	Spring 2004
~3 kg/day	~0.5 kg/day
~4% of total river U flux	~0.6% of total river U flux
~30 kL/min of contaminated groundwater	~10 kL/min of contaminated groundwater
0.03% of river flow	0.009% of river flow

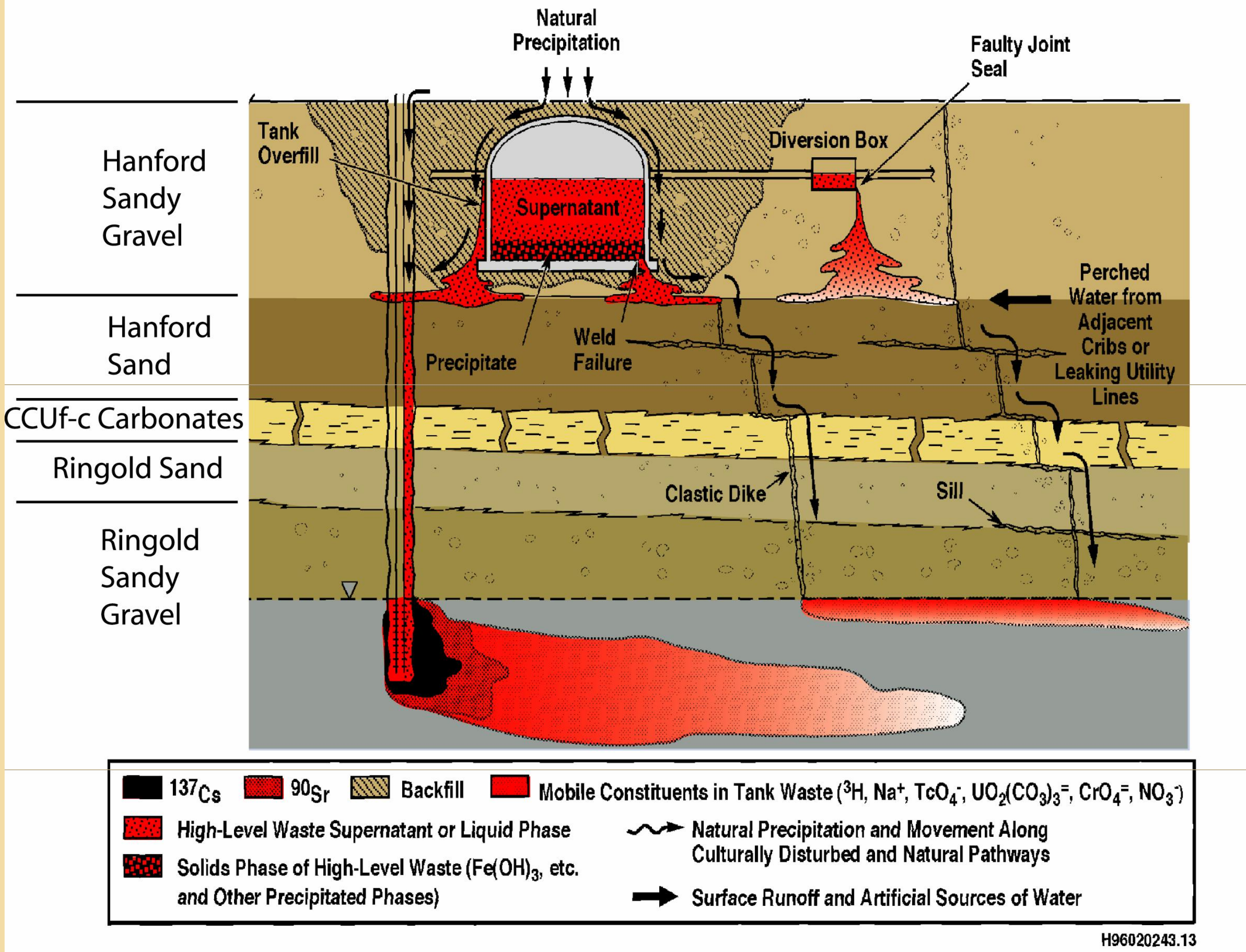
Although enhancement to U concentrations are small; Hanford U can be detected in river water 250 km downstream (and traced to 300-Area plume)



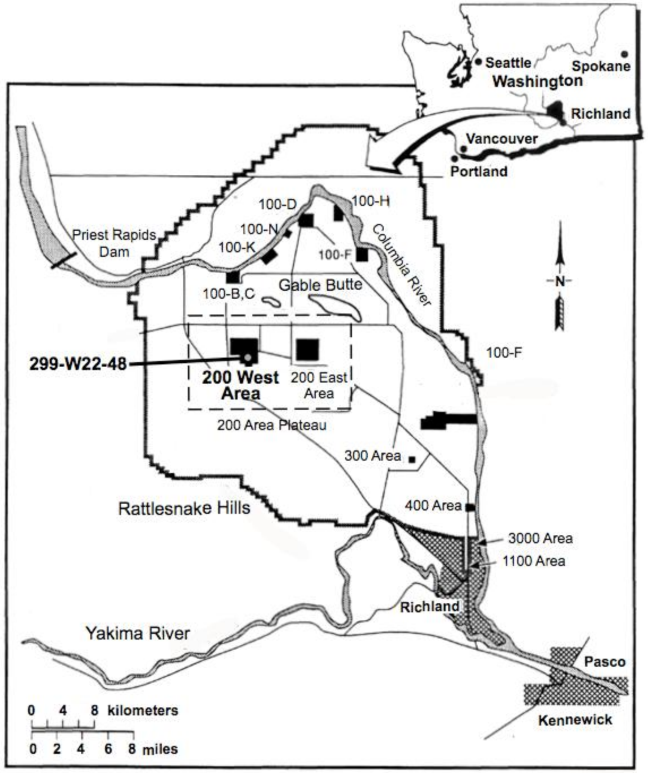
The point is that the Hanford contribution can be precisely measured (and shown to be minor at present)

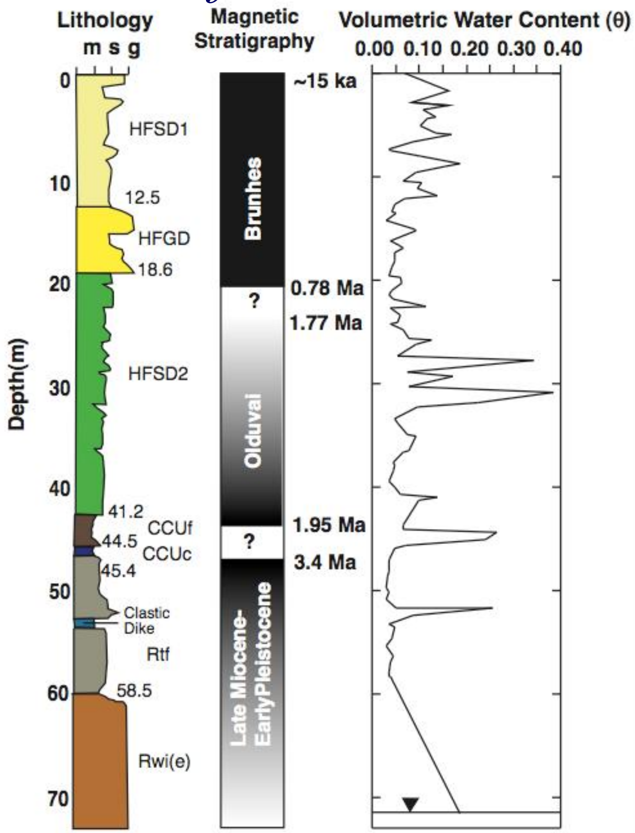
Applications of isotopes to Hanford hydrology: Vadose zone fluid fluxes from isotope measurements

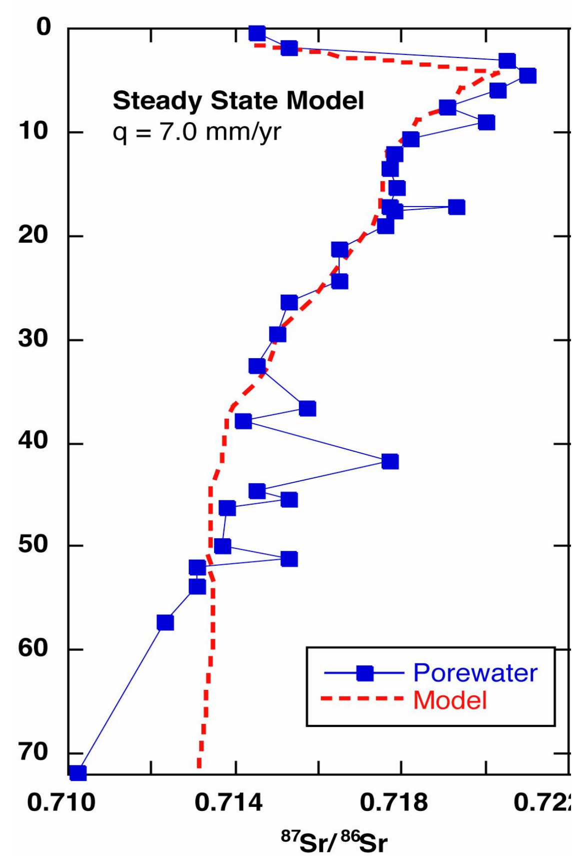
- Transport to groundwater depends on fluid flux
- Isotopes can be used to measure water flux through the vadose zone
- Can obtain 100 - 1000 yr averages needed to predict future migration of contaminants



With Sr isotopes we deduce long-term infiltration flux of 7 ± 3 mm/yr in 200W area for common soil types

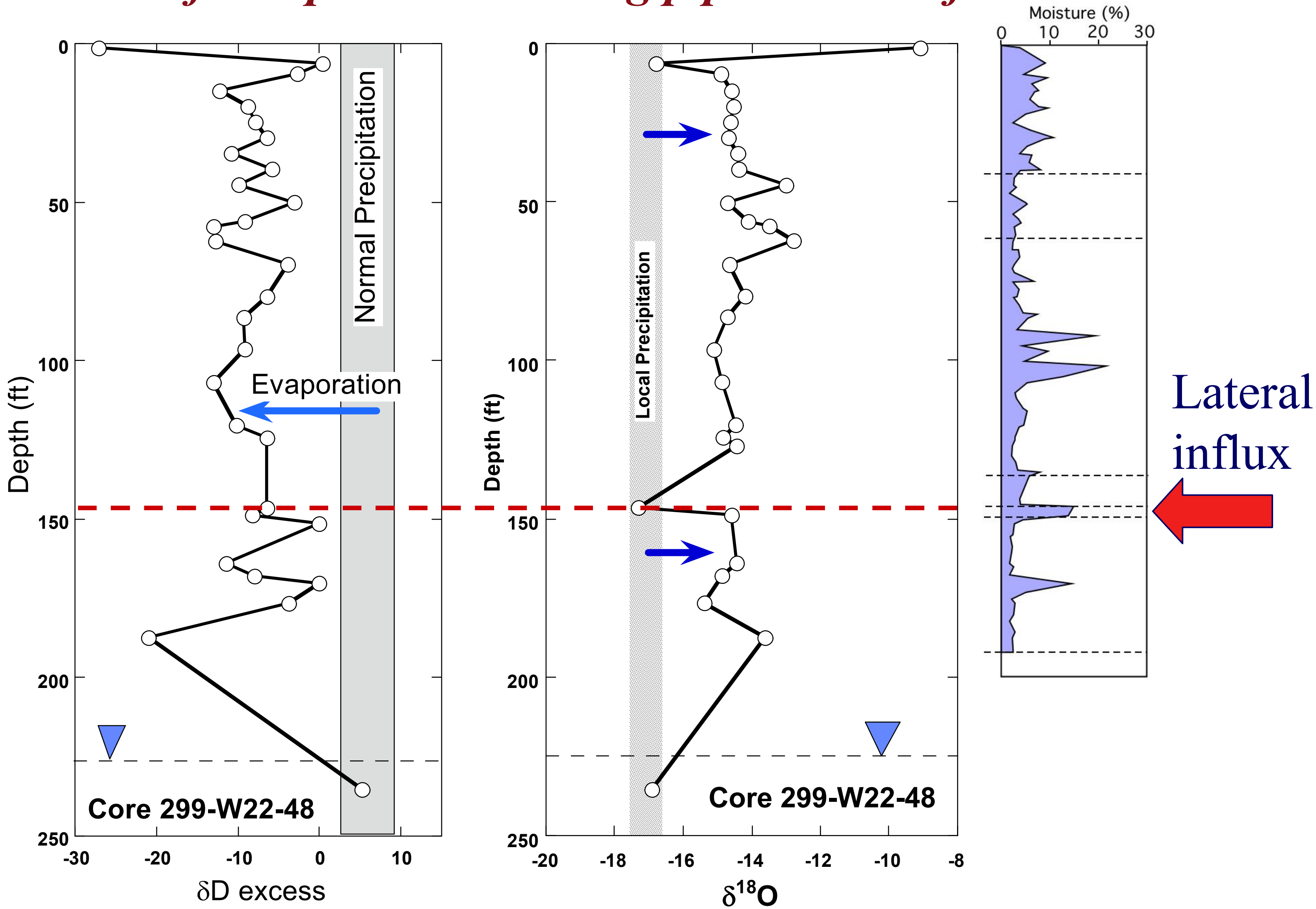


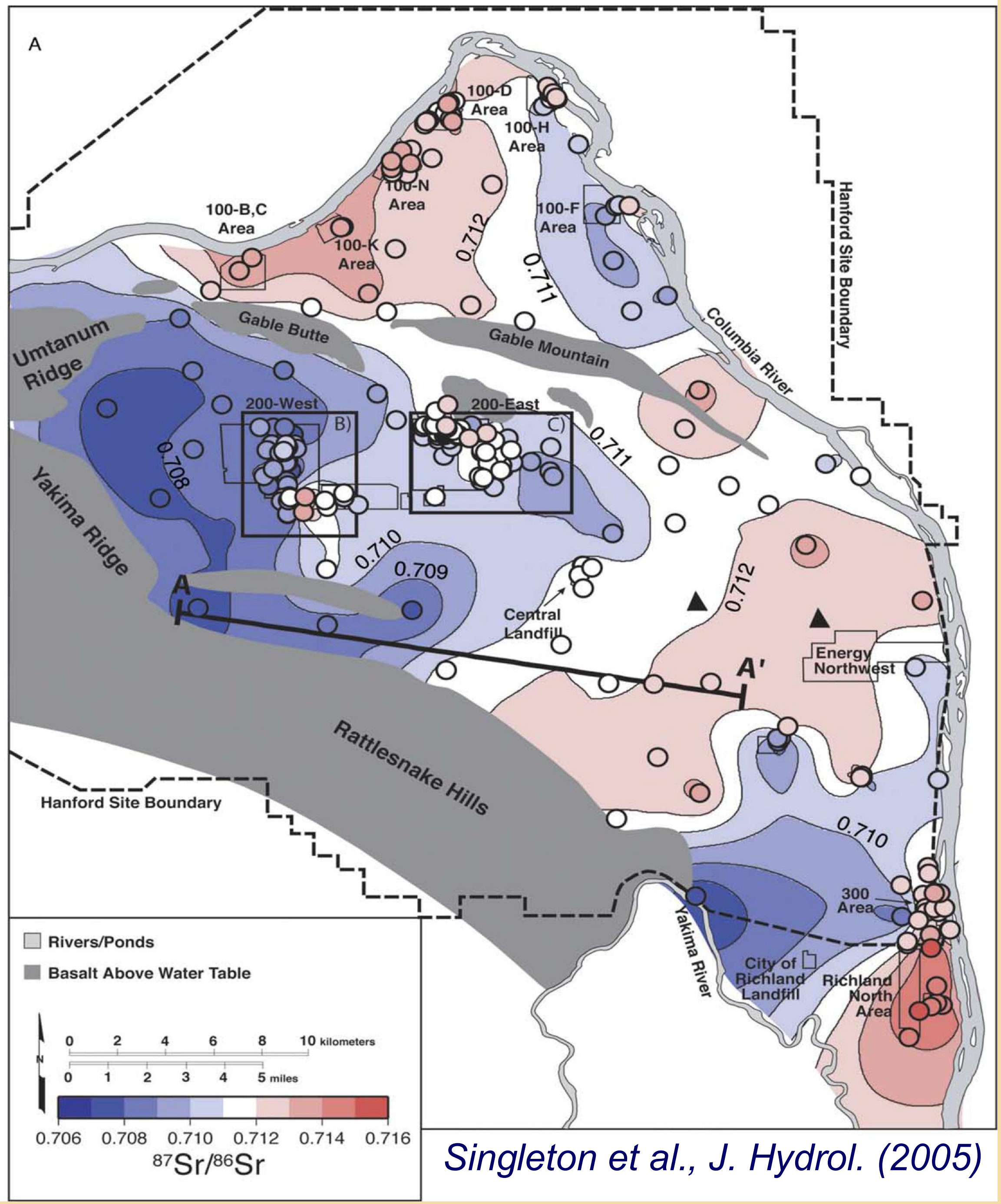




Adding U isotopes removes some ambiguity and slightly modifies result to ca. 4 mm/yr

O and H isotope ratios label VZ water: Water from surface spills and leaking pipes is identifiable





Singleton et al., J. Hydrol. (2005)

Sr isotope map of Hanford groundwater

Low $^{87}Sr/^{86}Sr$ comes from

- *reaction w/basalt (recharge & upwelling)*
- *Yakima River infiltration*

High $^{87}Sr/^{86}Sr$ from

- *reaction w/sediments*
- *VZ flushing &infiltration*
- *Columbia R. infiltration*

Conclusions

- *Isotopic field studies are an essential component of characterization for contaminated watersheds*
- *Major impact on conceptual models*
- *Quantitative estimates of contaminant fluxes and sources - which (should) impact remediation decisions and help to define essential basic research issues*
- *Also useful for monitoring natural and engineered (bio) remediation, and for long-term stewardship*
- *Isotope approaches become more useful as we learn the capabilities and how to apply them*

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