Lectures 24 and 25
Nuclear Waste

Low-Level Radioactive Waste:

- **Fuel rods**: ^{235}U → neutrons which interact with other nuclei to produce radioactive material

- Short term: ^{90}Sr ^{137}Cs ^{60}Co

  $_{t1/2}$ = 29 years =30 years =27 years

  -After 5 half lives : 1/e^5, <1% of the original activity→ background levels.

- Long term presents problems

  - ^{239}Pu, $t_{1/2}$ = 24,400 years→ will exist nominally for 100,000 years.

- Thus, stringent storage requirements are necessary -- at present are all stored on site.

- *West Valley*: Commercial reproduction of fuel (Plutonium).

→ Need possible site for Pu burial

- **Deep Sea burial** (near Hawaii)
- **Columbia River basalt** (Hanford, Washington)
- **Salt Dome/Caverns** (Carlsbad, NM) → WIPP = Waste Isolation Pilot Project.
- **Yucca Mountain** (Nevada).

[Options not considered by US:]

- Above ground pyramid (French)
- Abandoned iron mine (German, less than 1000 m below land surface)
- High U granite (Stripa, Sweden)

  Swedes: for their high level waste repository → have been very open about it - invited a large number of non-Swedish investigators.

→ **<u>Stripa Granite</u>**: good in political and good P.R.

- About 60 miles from Stockholm
- Not very tectonically active, no recent volcanoes.
- 1.7 Ga granite mass of quartz monzonite about 70% Si

  (mostly quartz & k-spar).
- Had intruded into older terrain of high grade gneiss surrounded by a leptite.

- **Formation of a skarn** (Fe-bearing ore, mined by Swedes).

  → Sets up hydrothermal → heat → metals which precipitates when cool to form the skarn.
- Granite: extremely high in U at 40 ppm (8-10 x normal).

  Therefore, this area is already pretty radioactive.
- **Cooling results in fracturing, a large fracture network has developed.**
  - Increased depth, fewer connected fractures.
  - Flow net disrupted by mine- draws large amounts of water into mine!
  - What will it do if not able to control? →Water will pass through this system relatively quickly.

Problems with:

1. **Deep sea bed**:

- Not viable because even though waste is relatively disposed off, will **generate a lot of heat** (in glass, then in stainless steel) → Glass is out of equilibrium- was quenched at 800-1000°C, will attempt to return thermodynamically.
  - How to keep it from returning? → Water tends to help bring species back to equilibrium. Glass, in the presence of water will break down into a clay (or chlorite or epidote).
  - Therefore, may get ^{239}Pu dissolved in water which was heated.
- **Salt water is highly corrosive** to stainless steel, even more so if heated.

  **Therefore, don't want this type of containment vessel in sea water.

  Retrieval? Good luck.

This idea was rejected.

Problems with:

2. **Columbia River Basalts**:
   - The people there wanted the waste. Hmm.... it is Nuclear town, USA
     - But, if buried there, would go to Portland, (recharge, lot of water).
     - **It was too wet an environment**.

This idea was also rejected.

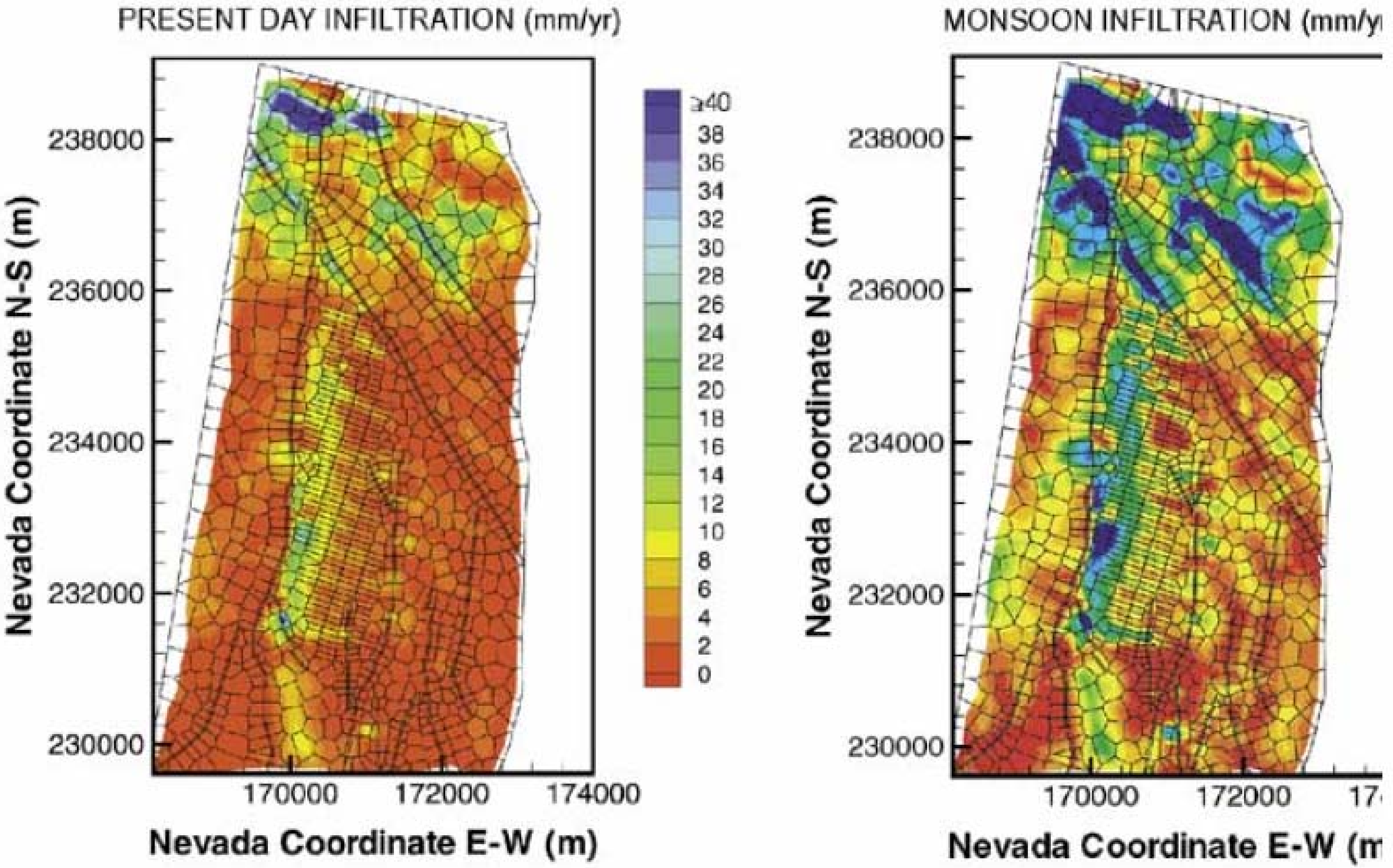
Problems with:

3. **WIPP**:
   - Very low permeability.

     K < 10^{-10} cm/s (lab) → < 1cm/1000 year.
   - But, can get fractures which will carry water much more significantly than porous media flow, because as water flows, it dissolves salts, which opens fractures allowing more water to flow and don't heal
   - *This also rejected.*

Problems with:

4. **Yucca Mountain**:
   - Very dry, less than 10 cm of rainfall/year.
   - Carbonates carry water, and there is Paleozoic carbonate rocks.
   - About 100 miles from Las Vegas, no one uses this water.
   - As opposed to other countries, solutions have taken a definite US orientation.
     - Ie. Sweden ---> in site, open access, formed by a team of international scientists.
     - US went along at a good rate until about 1985 - 1986



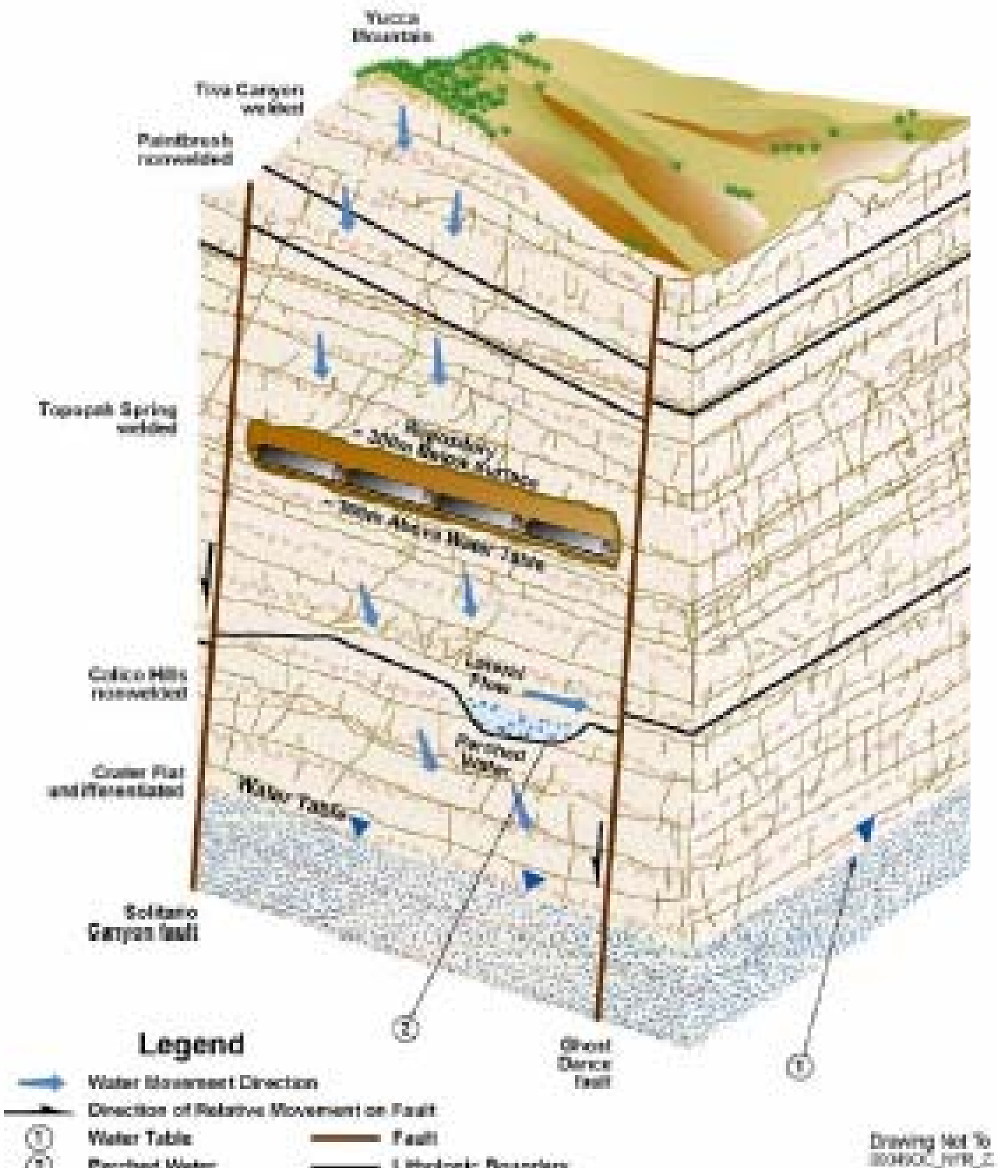
PRESENT DAY INFILTRATION (mm/yr)
Nevada Coordinate N-S (m)
238000
236000
234000
232000
230000
170000
172000
174000
Nevada Coordinate E-W (m)
≥40
38
36
34
32
30
28
26
24
22
20
18
16
14
12
10
8
6
4
2
0
MONSOON INFILTRATION (mm/y
Nevada Coordinate N-S (m)
238000
236000
234000
232000
230000
170000
172000
Nevada Coordinate E-W (m

Problems with **Yucca Mountain**:

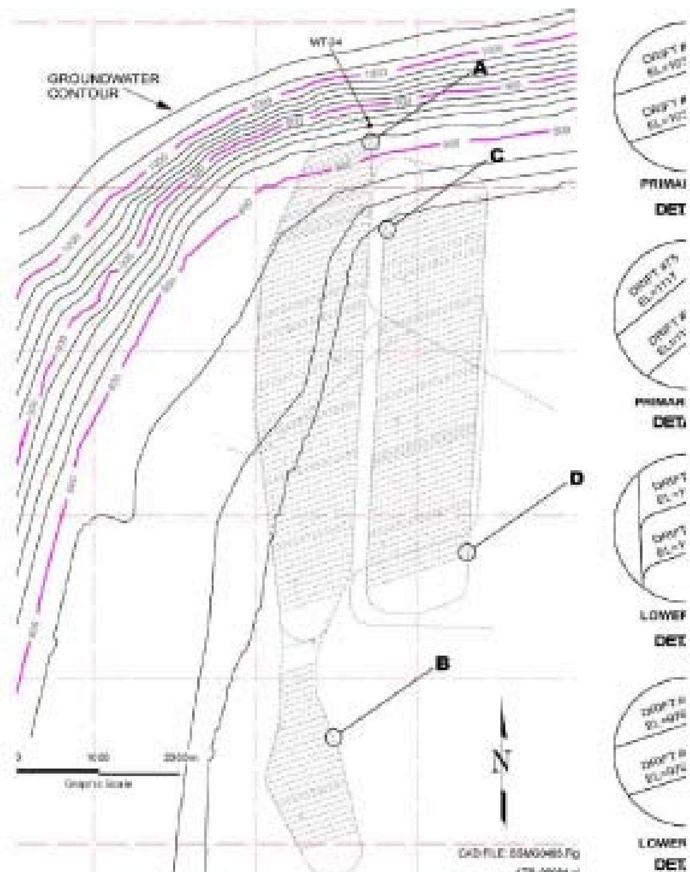
- **Site is a welded tuff** (result of explosive material laid down 10 -15 million years ago).
- *Concern : Can water migrate through and interact with repository surface?*
  - Would say no because **recharge is almost non-existent**.
  - Under current conditions, would say that it would stay reasonably intact.
  - **Jerry Zamansky** gave a public accusation (in NY Times) that the area was subject to periodic groundwater movement, based on the study of carbonate veins
    - **Carbonate veins** had been subject of study.

    ** **Thure Cerling** had already studied this area, was the only one with hard data to present to Congress.

Figure 3-23. Diagrammatic Orthogonal Section of Yucca Mountain Illustrating Hydrologic Features and Processes of the Unsaturated Zone



This figure shows the thick unsaturated zone, the location of the repository horizon, principal



The water table in the vicinity of Yucca Mountain shows a steeper gradient toward the north and northwest. The northernmost emplacement drifts in the primary block area would be closest to the water


FIRE

Problems with Yucca Mountain:

Questions:

- Source of carbonates?
- Source/Age of volcanism?
- Is it possible for groundwater to have migrated up the fault zone?

Possible source of CO_3

- Soil carbonates (pictures don't look like typical soil carbonates).
- precipitation of $CaCO_3$ → looked like it had precipitated from groundwater.
- To estimate conditions, excavated large trenches & found large veins of $CaCO_3$

Problems with Yucca Mountain:

***Trench 14**

What is the source of the carbonate veins?

- Simply soil carbonates.
- Still carbon dioxide generated in photosynthesis in soil zone, dissolved in water, in contact with Ca ion to form $CaCO_3$. Will precipitate to form calcite.
- Amount of soil carbonate depends on type of vegetation.
- Scientific study turned to **isotopes of carbon and oxygen** to determine whether the carbonate was :

  1. Meteoric or 2. Paleozoic

- **$^{13}C/^{12}C$ is a function of CO_2 →** is a function of the source.

Problems with Yucca Mountain:

- Paleozoic $CaCO_3$ hasn't changed than much, therefore, this $CaCO_3$ look very much like a marine carbonate.

$^{13}C/^{12}C \rightarrow \delta^{13}C_{CaCO3} = 0$ -- looks like sea water, but:

$\delta^{18}O$ looks like meteoric water.

$\delta^{13}C$ of most plants is about -15 to –25

$\delta^{13}C$soil carbon = -2 to -12.

** Depends on the type of plant that exists.

Problems with Yucca Mountain:

- **C3** - 3 carbon acid → ribulose phosphate make C3 plants about -25_
    - Includes trees and shrubs.
- **C4** - Starting about 8-9 million years ago, C4 pathway in response to decreasing CO_2 levels in atmosphere.
    - Is a 4 carbon acid, dominating in water-stressed (low water) environments.
    - Includes grasses, corn.
        → Useful for determining when corn was domesticated, can track human dirt based on isotopic content of bones.
    - moderate soils at low elevation, mainly C4 plants (shown by high 13C content.)
    - moderate soils in higher elevation, C3 plants tend to dominate.

General distribution of C_3 and C_4 ecosystems. Note that the northern limit of temperate grasslands in North America and Asia include a significant proportion of C_3 grasses due to the cool growing season.

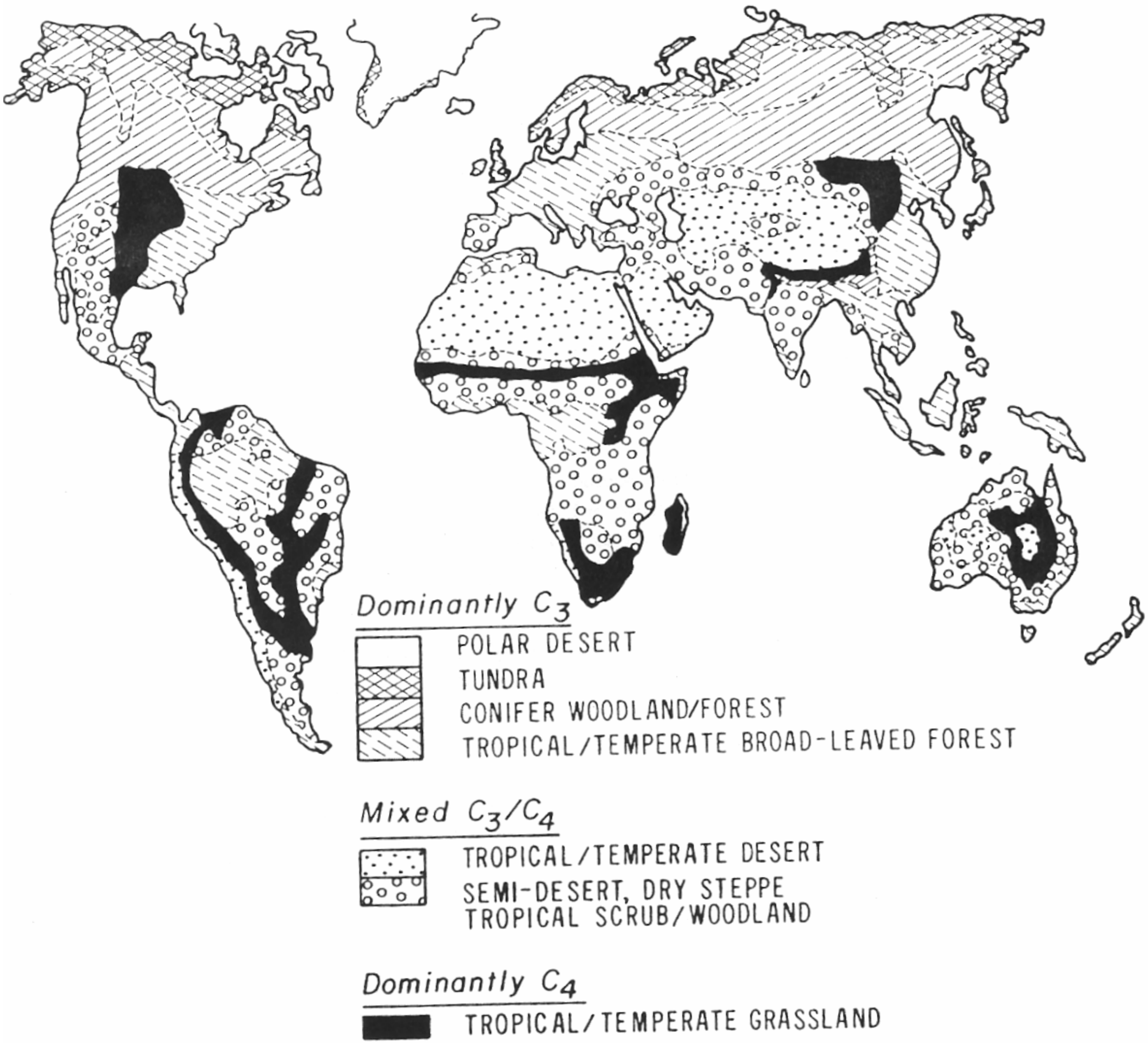
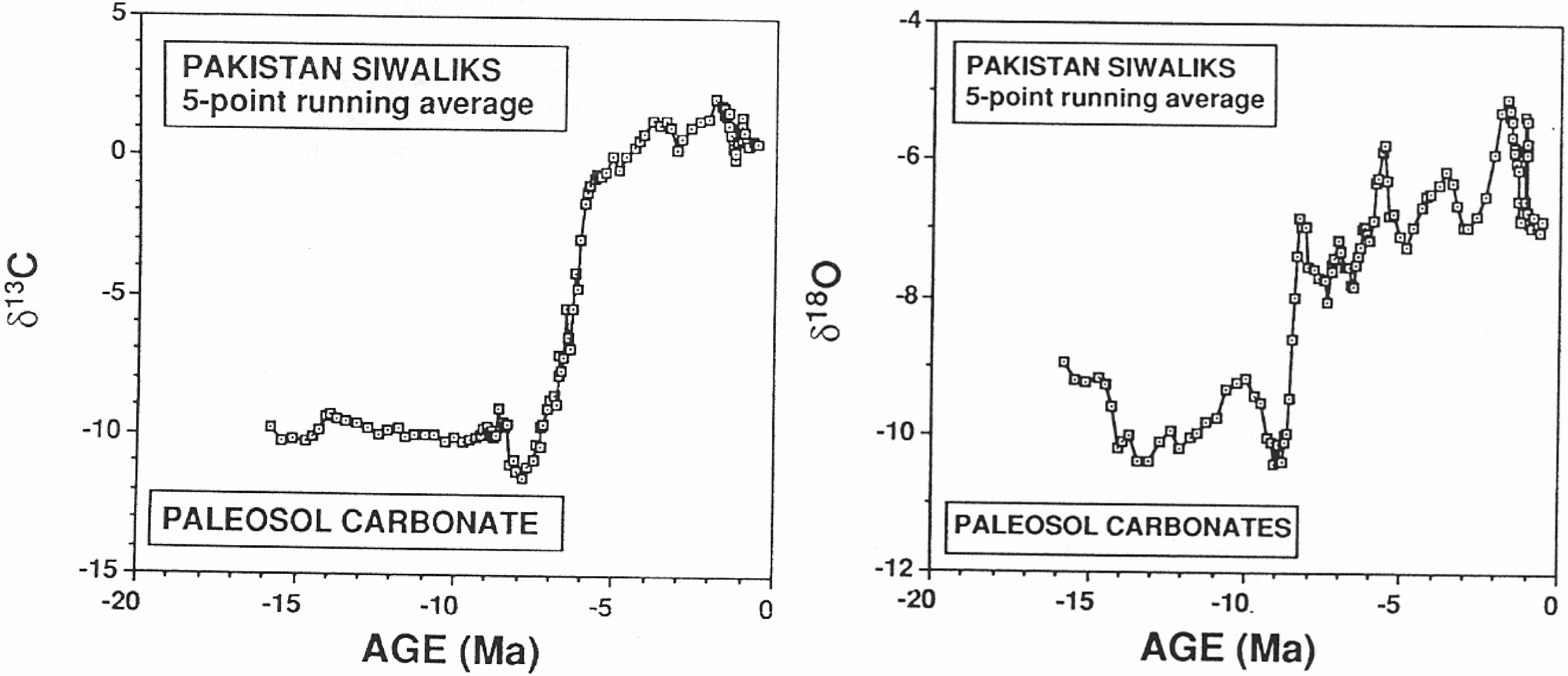


Figure 2. Cerling and Quade, 1993.

Carbon and isotopic compositions of pedogenic carbonates from the Siwaliks in Pakistan.



Carbon isotopes show a shift from a C_3 dominated to a C_4 dominated ecosystem between 7.3 and 6 million years ago, whereas oxygen isotopes record an important shift at about 8 million years ago as well as a few smaller shifts at other times.

Figure 17. Cerling and Quade, 1993

Isotopic composition of Holocene pedogenic carbonate from two soil profiles from Midwestern North America. The δ^{18} O and δ^{13} C values are essentially constant at depth for pedogenic carbonate. There is more variation in the isotopic composition of soil organic matter, specially near the soil surface, which may be due to differences in rooting depths of C_3 and C_4 plants.

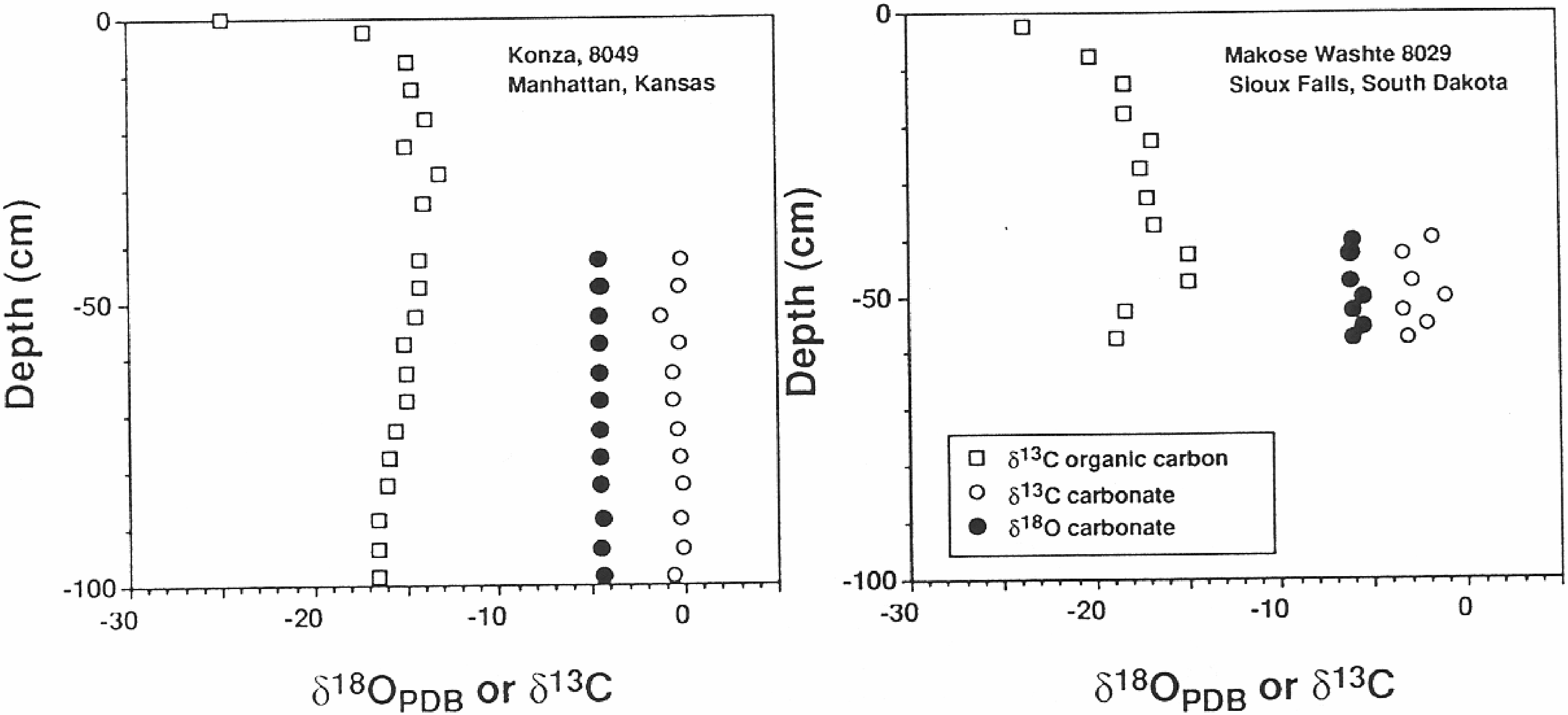


Figure 3. Cerling and Quade, 1993.

Problems with Yucca Mountain:

* **At trench 14, dominant C3 variety.**

* **Isotopic composition of O reflects composition of water for ^{18}O**

  • As go to higher elevations, lower $^{18}O/^{16}O$ ratio.

δ^{13}Csoil carbonate (acts as a marker) -- allow people to examine paleoecology.

** There is no process to get between the Paloezoic aquifer and Trench 14

→ Therefore, there is NO SIMILARITY between the soil carbonate and the Paleozoic aquifer.

Molten lava/ Lanthrop Wells Cinder cone.

– Only about 5,000 years old? Based on morphology

– Volcanic activity more recent than 10,000 years, so an acceptable repository
  → still a tectonically active area.

  [If cone is young & see original flow surfaces, can tell the true age of the last eruption.]

  - **Cosmic rays bombard surface,** penetrate crystal nuclei (called spallation) and cut off either a ^{3}H or a ^{3}He atom about 15% of the time.

Ex. <u>Shoreline</u> Lake Bonneville Tabernacle Hills

- We know age of surface basalt because we know age of shoreline (14C age of shoreline → therefore, the age of when the basalt erupted).
- Olivines are good phenocrysts- picked out of basalts to determine age.

15,000,000 atoms of ^{3}He → 60,000 years.

Compare to $^{21}Neon$ → ^{23}Na → ^{24}Mg 50 atoms/year

Cosmogenic Isotopes

^{36}CL ^{3}He ^{21}Ne

- Method based on use of a surface.

→ Has there been more than 1 eruption? No, only one in the past 60,000 years.

- ^{3}H has a 1/2 life of 12.4 years (relatively short)
- ^{14}C (important for global research) has a t1/2 = 5730 years (useful for about 30,000 years).
- ^{14}N (n,p) ^{14}C—β→ ^{14}N
  - ^{14}C → relatively neutron rich, 6p, 8n; unstable decays to 14N (7p,7n) by beta decay.
  - ^{14}C → done by counting decays (1st = Libby in 1940's).

Recent Advances:

- Acceleration Mass Spectrometry (AMS)
  - Much easier to measure this than to count the decay.
  - Allows much smaller samples - need only mg rather than grams.

  - $^{14}C \rightarrow {}^{14}CO_2$ (atm) and is incorporated in general C-cycle.
  - $(^{14}C/^{12}C)$atm → constant in atm
  - About 14 dpm/g C or .22 Bq/gC
  - Once decay starts → 7 dpm > 1/2 Ro → 5730 years.

First worry:

1. What is the initial $^{14}C/^{12}C$? Is this really a constant in the atmosphere with time? Not necessarily.
2. Always worry about the exchange of C
   - Relatively inert ---> tooth, protein in bones (collagen), pollen.
   - If intake---> therefore minimum C exchange ---> stable.

$CaCO_3$ ---> C is exchangeable as CO_3.

$$CO_3^{=} (aq) \longleftrightarrow CaCO_3 (s).$$

→ This happens any time with water flowing past aquifer, therefore ^{14}C doesn't work too well with Yucca Mtn.

U- Series:

$^{238}U \rightarrow$ $^{234}Th \rightarrow$ $^{234}U \rightarrow$ $^{230}Th \rightarrow$ ^{226}Ra

$t_{1/2}$ 4.5 Ba 24days 244,000 yrs 77,000 yrs

Activity ratio: should be in secular equilibrium.

- 234A/238A should = 1, but doesn't exactly hold.

- U may move out of grain and become mobile.
- In water (river) U activity >> 1 because ^{234}U has been released preferentially.
- Seawater 234A/238A = 1.15.

What happens if some of the ^{234}U → coral → ^{230}Th stays put?

- U/Th can give an age → $^{230}A/^{238}A$ will give an age.

- Over past 20,000 years, the ocean level has increased and corals had to keep up to rising.
  - ^{14}C and U/Th ages differed by about 3000 years
  - Therefore assumption that 14C is constant is wrong.

    1. Mixing C reservoirs since last glacial.
    2. ^{14}C production.
       - Changes in earth's magnetic field, acts as a giant mass filter which bends cosmic rays away.

Now, have to look a 3 different ages.

^{14}C, $t_{1/2} = 5730$.

^{14}C, $t_{1/2} = 5570$ (radio carbon)

^{14}C corrected based on U-Th timescale.

Yucca Mtn.

Aquifer → High activity ratio

Trench 14 → Low activity ratio

Can do the same thing with Sr isotopes.

- $^{87}Sr/^{86}Sr$ (no production)
- ^{87}Rb --- → ^{87}Sr. Half life 50Ga
- High Rb will result in high $^{87}Sr/^{86}Sr$ ratios.

ie. Granite → high $^{87}Sr/^{86}Sr$ → Used in age dating.

Vein carbonates → low ratio.

Carbonate aquifer: Sr represents the Sr there at the time of formation.

- $(^{87}Sr/^{86}Sr)CaCO_3 = (^{87}Sr/^{86}Sr)$seawater

Can tell what seawater has been doing over the past 600 Ma by measuring Sr.

Controlled by erosion (.712)

- – Input increases $^{87}Sr/^{86}Sr$ from crust and hydrothermal activity (exchanges seawater Sr for basalt (at .703)).
- – Latest peak due to erosion of Himalayas during uplift.

Also has trace quantities of ^{129}I and ^{36}Cl.

- Typically only in atmosphere by interaction of cosmic rays.
- Also produce with very high Cl values

^{4}He ^{129}I ^{36}Cl (300,000 Years)

$t_{1/2} = {}^{16}$Ma

Better potential tracers for processes of 100,000 years.

- ^{35}Cl(n,γ)^{36}Cl → doesn't really work; can be produced by the upper atmosphere.
- ^{129}I → can act from activation of other stable compounds.
- "Dead" carbon → ^{14}C.

^{137}Cs, ^{90}Sr, ^{60}Co → short $t_{1/2}$;

- Had high ^{4}He, ^{129}I, ^{36}Cl and no ^{14}C.
  - **Tended to indicate that the water flowing into the mine was relatively old (> 10,000 years).**
- Gabon, Africa → Natural nuclear reactor. 2.06Ba (very U rich)
  - Had sufficient ^{235}U to produce a melt down!
  - Watch migration → low migration; relatively self contained.

Ward Valley, California:

- Location: a little South of Death Valley, near the CA-NV border
- Significance: was the site for low-level radioactive waste dump

  → for the low-level waste, assurance of about 500 years containment is needed
- Geology of the valley: valley filled with sediments, with underlying faults
- True desert environment: < 100 cm/yr rain ; ≈ 100 cm/yr evapo-transpiration
- Initial plan: apply rigorous (though probably unnecessary) North Eastern radioactive landfill techniques (i.e. liners, etc.)

Ward Valley, California:

PROBLEM:

The Colorado River is only 10 miles away, which led to speculation of how long would it take for the waste to go through the unsaturated zone to the water table about 300 meters beneath?

– Can't use tracers like $^{3}He/^{3}H$ in an unsaturated zone, so the researchers looked at ^{3}H alone. Why did they do this? If ^{3}H existed at a depth, it would indicate fast seepage of water down to the water table (using the 1960's tritium spikes)

- They took water from areas of lower than 100% saturated vadose zone and pumped hard to suck air and available water out with a vacuum pump.

- Water was trapped in a liquid nitrogen trap (-195C), BUT leaks in the pumping system may have allowed moisture from the desert air (which, since it was in an area of high nuclear testing, had about 30-40TU level) into the trap. Because there wasn't a lot of water was collected, small leaks could contribute significantly to the measured tritium.

- Results of the tests: 1TU ± 1TU. → Well, is it 0 or not?

  → more tests are required to give any kind of definitive answer