

^{129}I odine: A New Hydrologic Tracer for Aquifer Recharge Conditions Influenced by River Flow Rate and Evapotranspiration

Kathleen A. Schwehr¹,
Peter H. Santschi¹, and Jean E. Moran²

¹Texas A&M University

²Lawrence Livermore National Laboratory



Acknowledgements



- Allan Jones
- Jan Gerston
- Ric Jensen



- David Elmore
- Pankaj Sharma
- Xiueng Ma



- Greg Woodside
- Adam Hutchinson

Outline

- Objective
- Introduction
- Input function
- Study Site
- Results
- Conclusions



Objective

- To test the potential tracer application of the iodine isotopic ratio $^{129}\text{I}/^{127}\text{I}$ in recent ground waters by analyzing its behavior in a well-characterized aquifer system.



Introduction

Importance of Iodine

- **Largest fraction** of short term and long term dose from nuke releases & fallout
- ^{129}I one of two long lived nuclides with **high mobility** in stored radioactive waste
- **New tracer and geochronological applications**
- Sea atm: **VOI** (greenhouse active & ozone destructive)



Introduction

Background for Iodine

- Biophilic
- ^{127}I 100% abundance
- ^{129}I $t_{1/2} = 15.6 \text{ ma}$

- Natural surface inventory 100 kg
- Bomb testing 150 kg
- Nuclear fuel reprocessing 2600 kg
(Cap de La Hague, Sellafield)
- Chernobyl reactor accident (1986) 1.3 kg

1 Liter drinking water:

10^{-12} Ci

^{226}Ra ~0.2 (Eisenbud & Gesell, 1997)

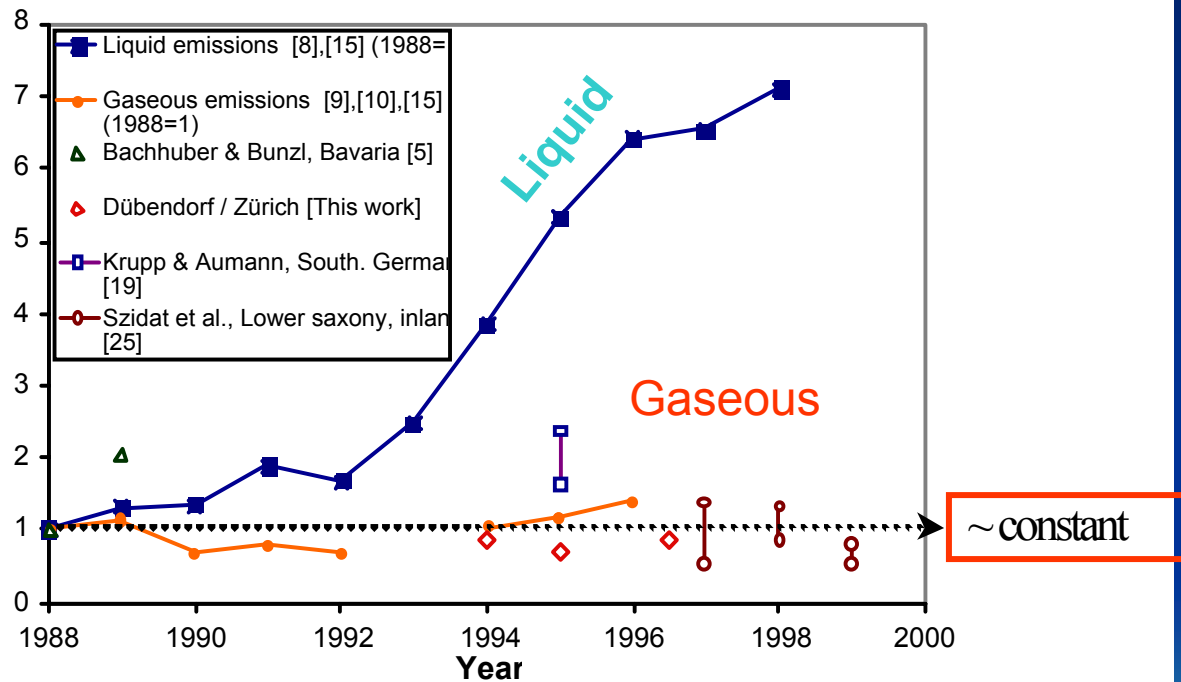
^{129}I 0.0000003



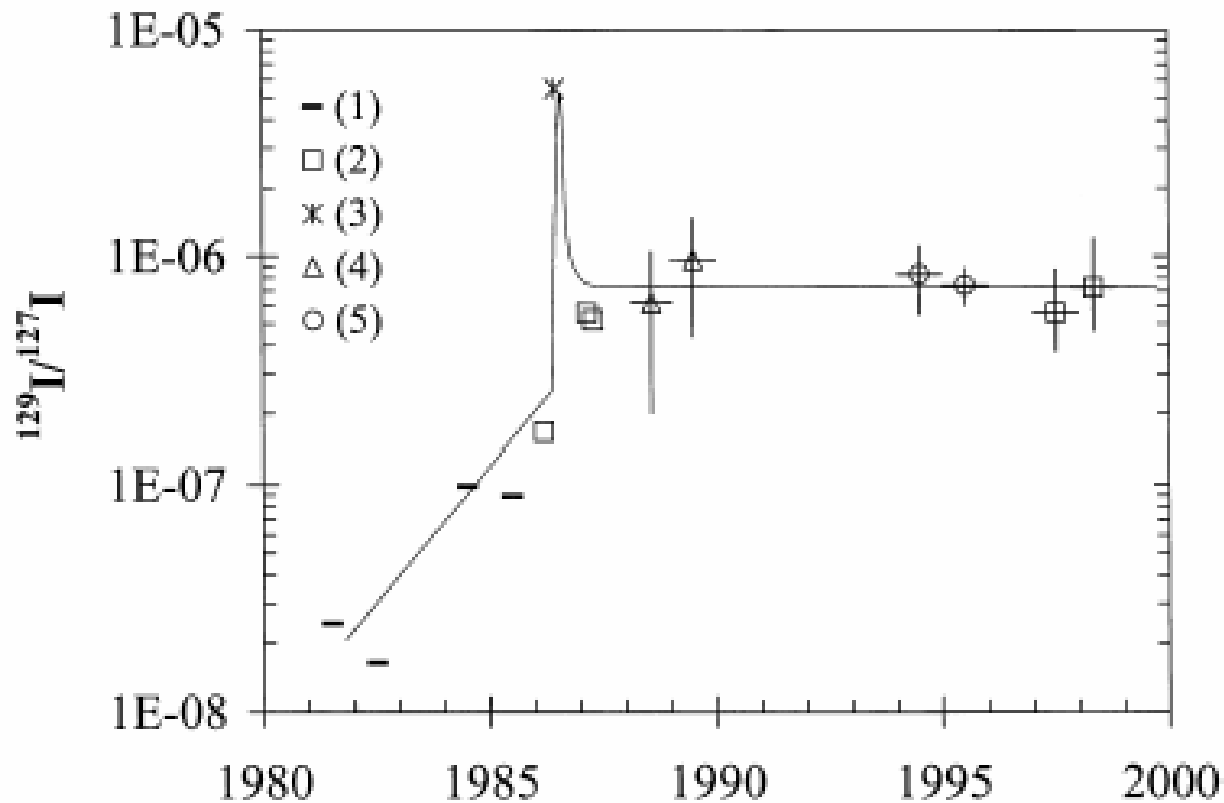
Atmospheric source
input function for ^{129}I
~constant over last decade



Atmospheric Inputs of ^{129}I in Europe [Schnabel et al., 2001]



$^{129}\text{I}/^{127}\text{I}$ Ratios in Precipitation in Europe [Szidat et al., 2000]

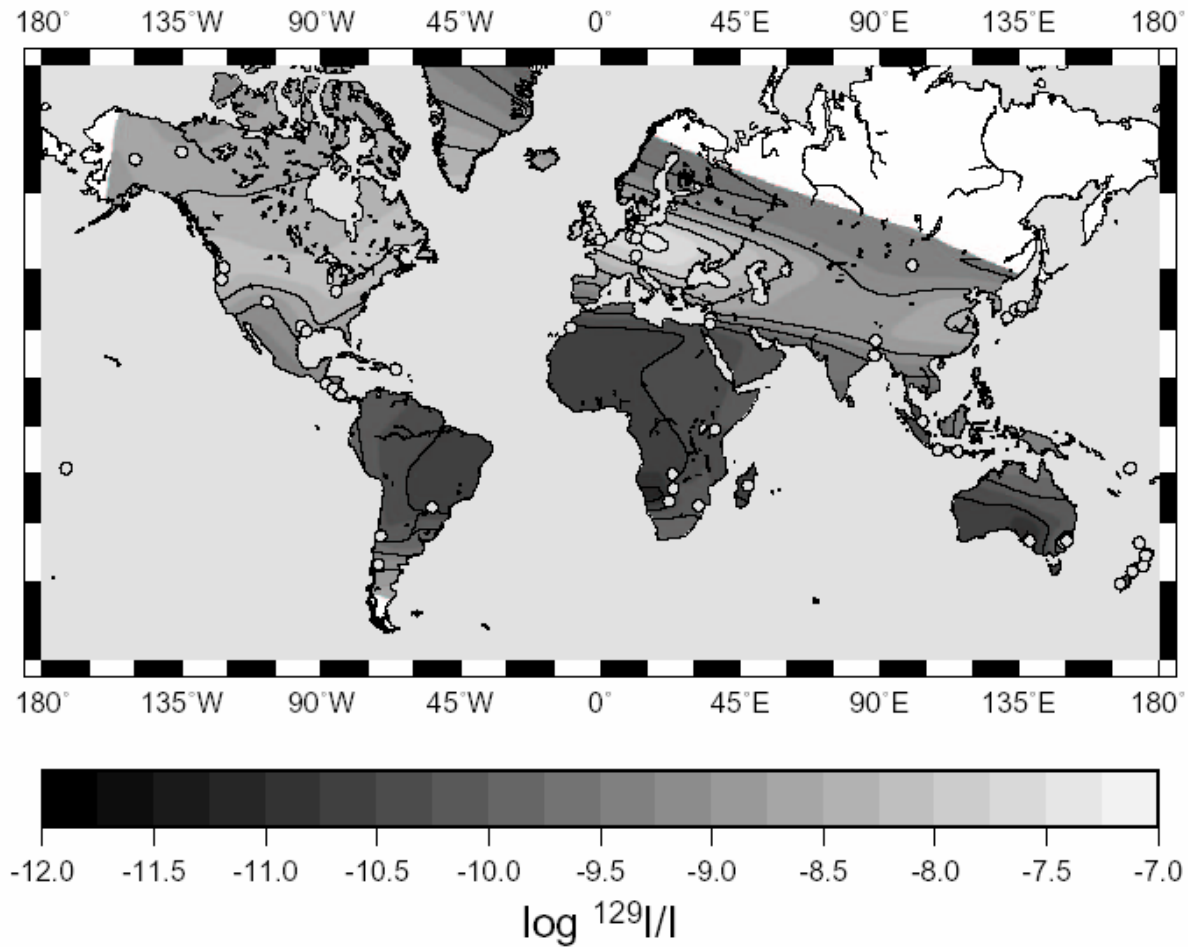


Global atmospheric
transport in 11 to 18 days



Global Distribution of $\log \frac{^{129}\text{I}}{^{127}\text{I}}$

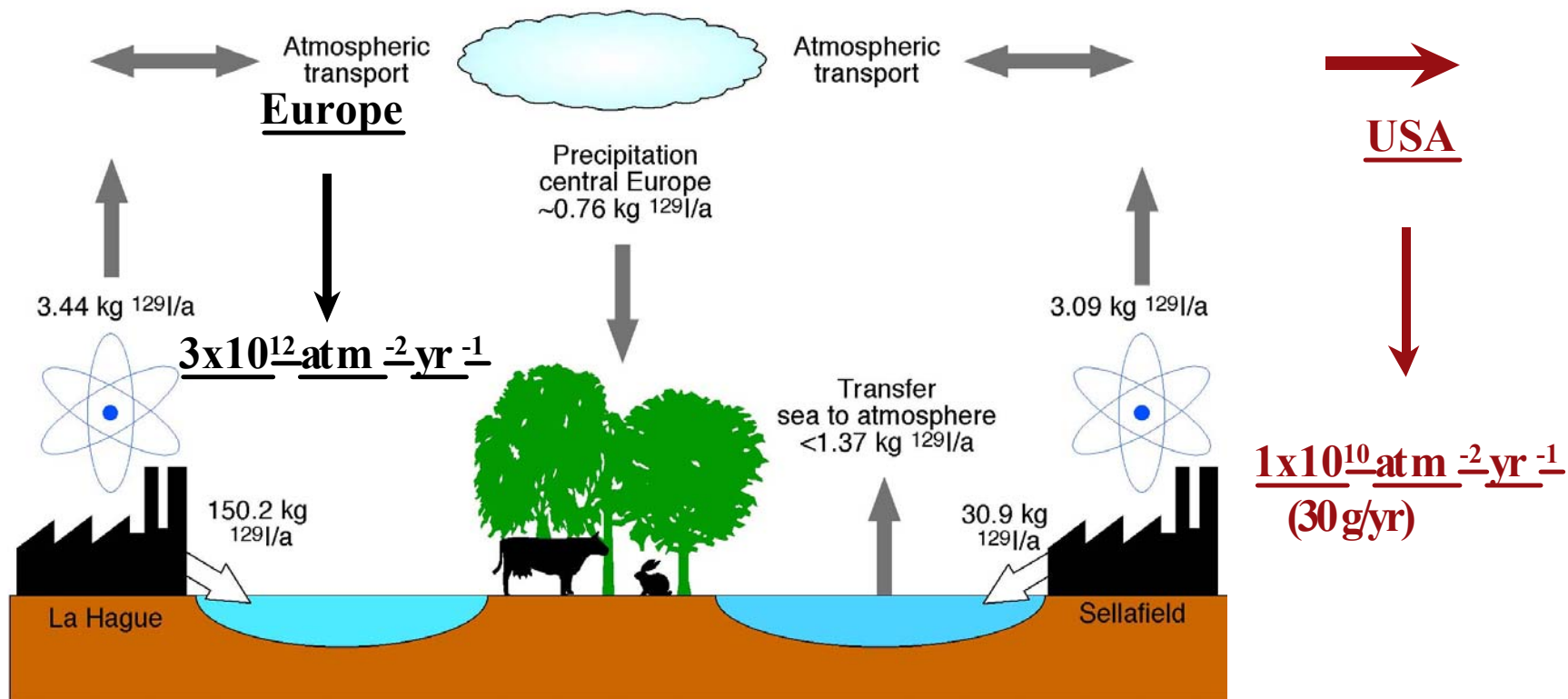
[Snyder and Fehn, 2003]



^{129}I Flux comparison between
Western Europe and the
Contiguous United States



Recent ^{129}I Emissions in Europe (Schnabel et al., 2001) and USA (Moran et al., 2002)



Santa Ana River Basin



Study Site in Semi-Arid Region



Precipitation

- Upper Basin 46 cm
- Lower Basin 35 cm (ET)
- (Texas ~20 to 160 cm)

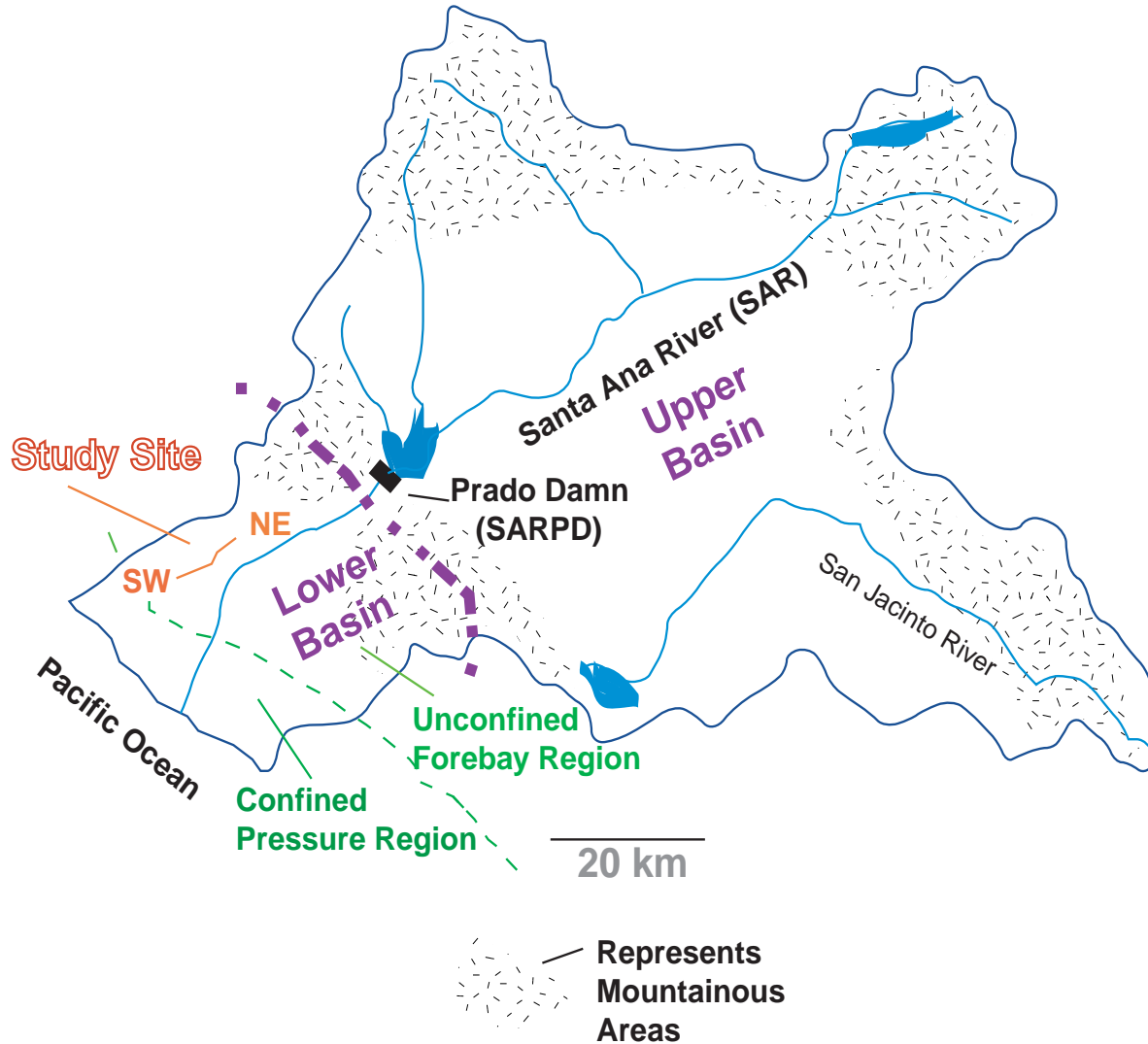
Aquifer Properties

- Artificial recharge
- Linear flow velocity ~5m/d
- Hyd. cond. 200 to 300 m/d

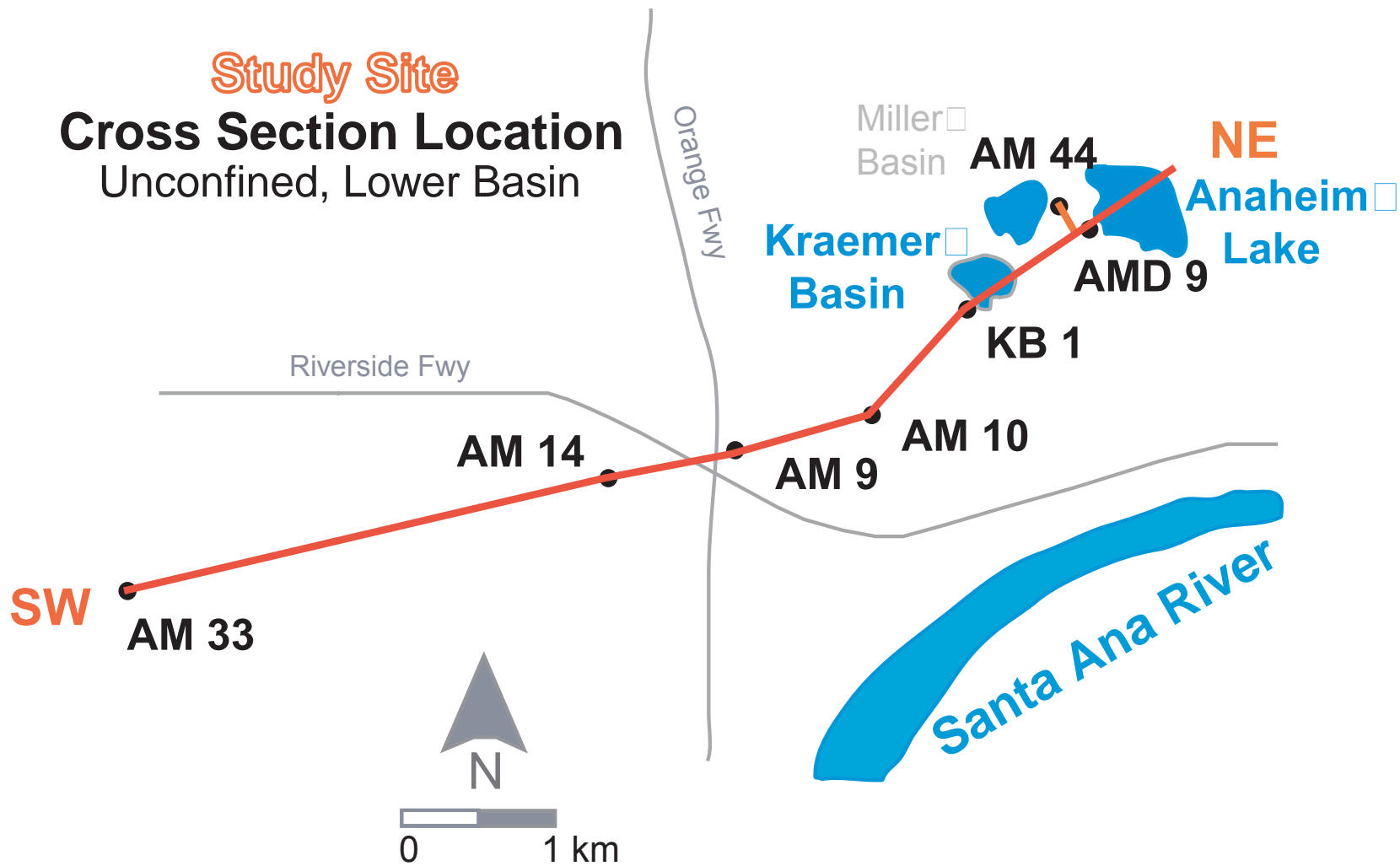
Aquifer System

- Unconfined
- Alluvial fill sands & gravels
- Dip & flow toward coast

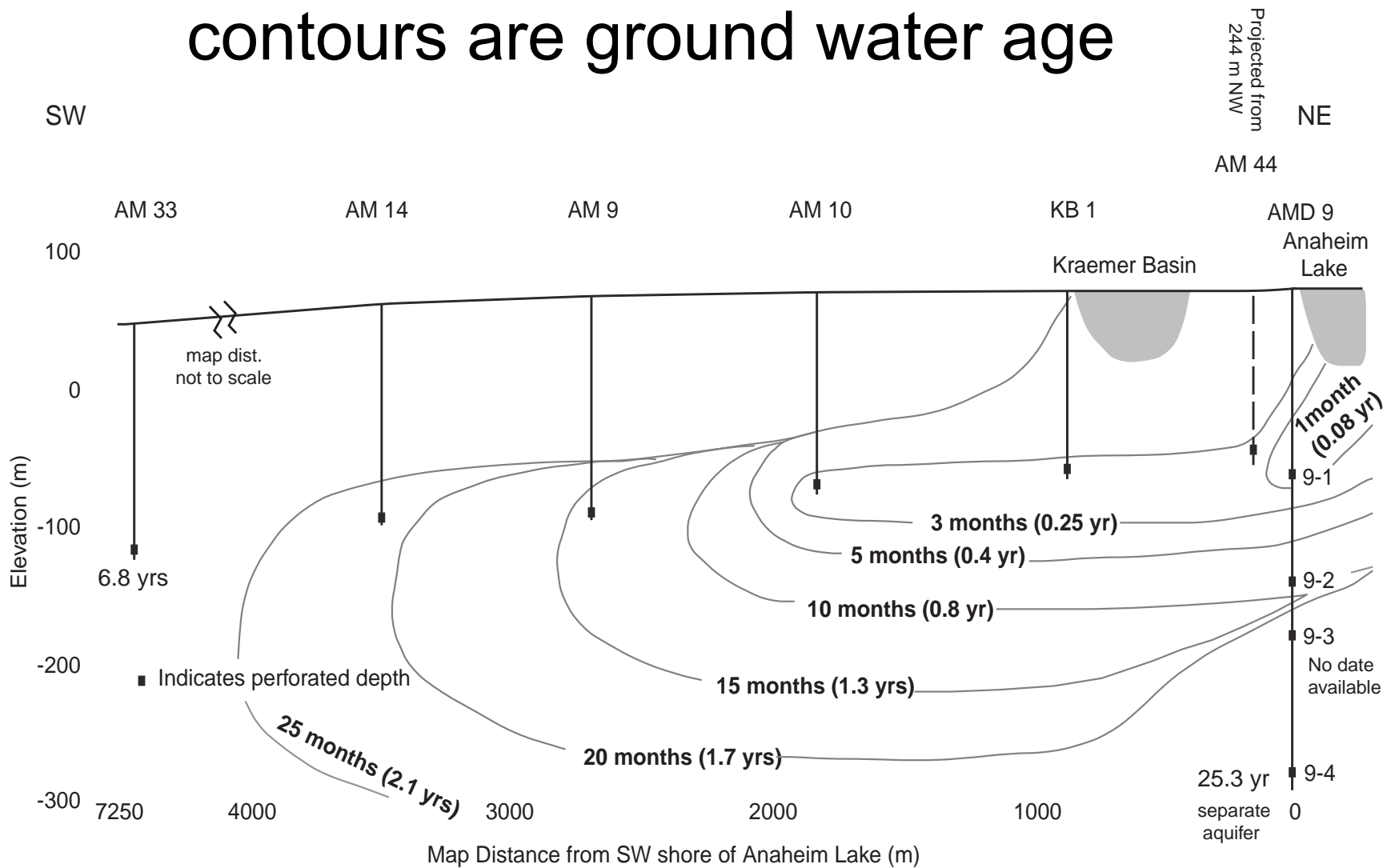
Santa Ana River Basin



Study Site
Cross Section Location
Unconfined, Lower Basin



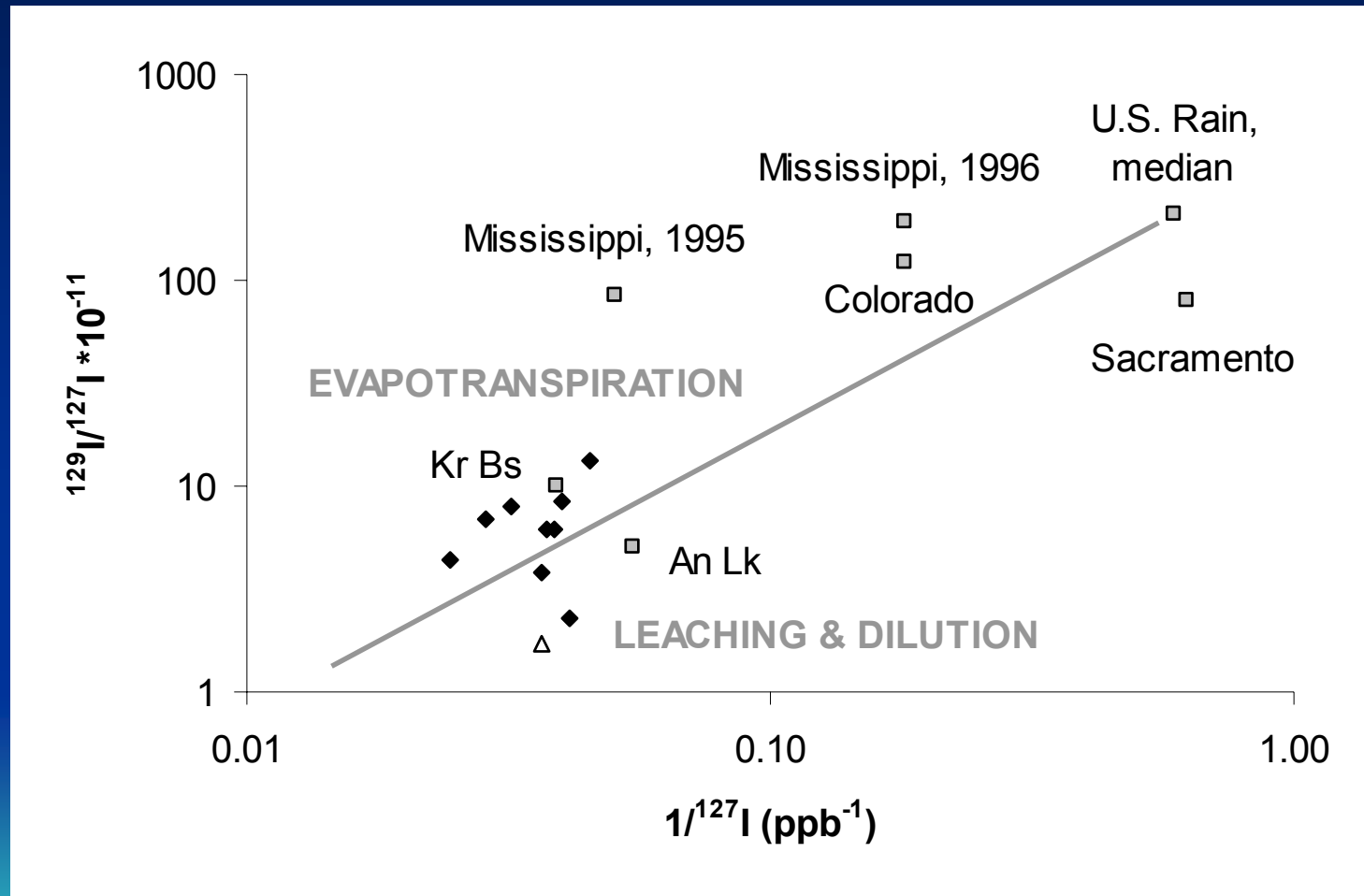
Cross Section contours are ground water age



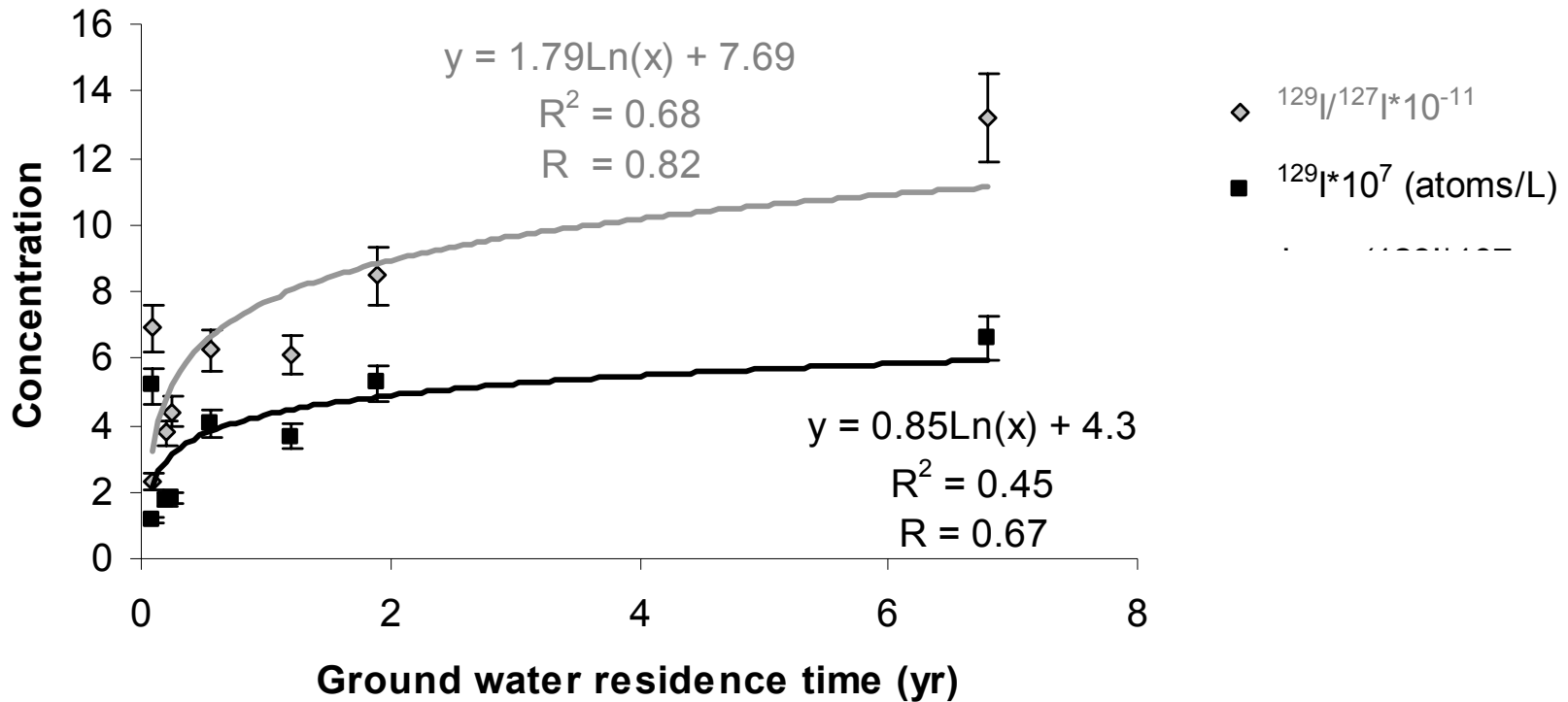
Results



Mixing diagram



Concentration vs. age



TOC & Biophilic Iodine

- **TOC** (Davisson et al., 1998)
 - Conc ↓ 50 to 70 % from surface to gw
 - ↓ in size fraction (from < 1 μm to < 0.2 μm)
- **¹²⁹I** (Santschi et al., 1999; Dissanayake & Chandrajith, 1999; Quiroz et al. 2002)
 - Conc ↓ 50 to 70 % from surface to gw
 - Colloidal fraction 50 to 70 % > dissolved in Miss. River (Oktay et al., 2001)



Removal of macromolecular colloidal material during infiltration



Factors affecting recharge

- Subsurface aqueous geochem. ppt or dissolution---I, Cl: conservative behavior
- Mixing: reclaimed water & imported water (10 to 25%) from Colorado River (COR)
 - COR ^{129}I : $3.2 * 10^7$ atoms/L
 - SARPD ^{129}I : $4.1 * 10^7$ atoms/L
- ET: salts concentrate & ppt during dry cycles; leach & dilute during wet cycles

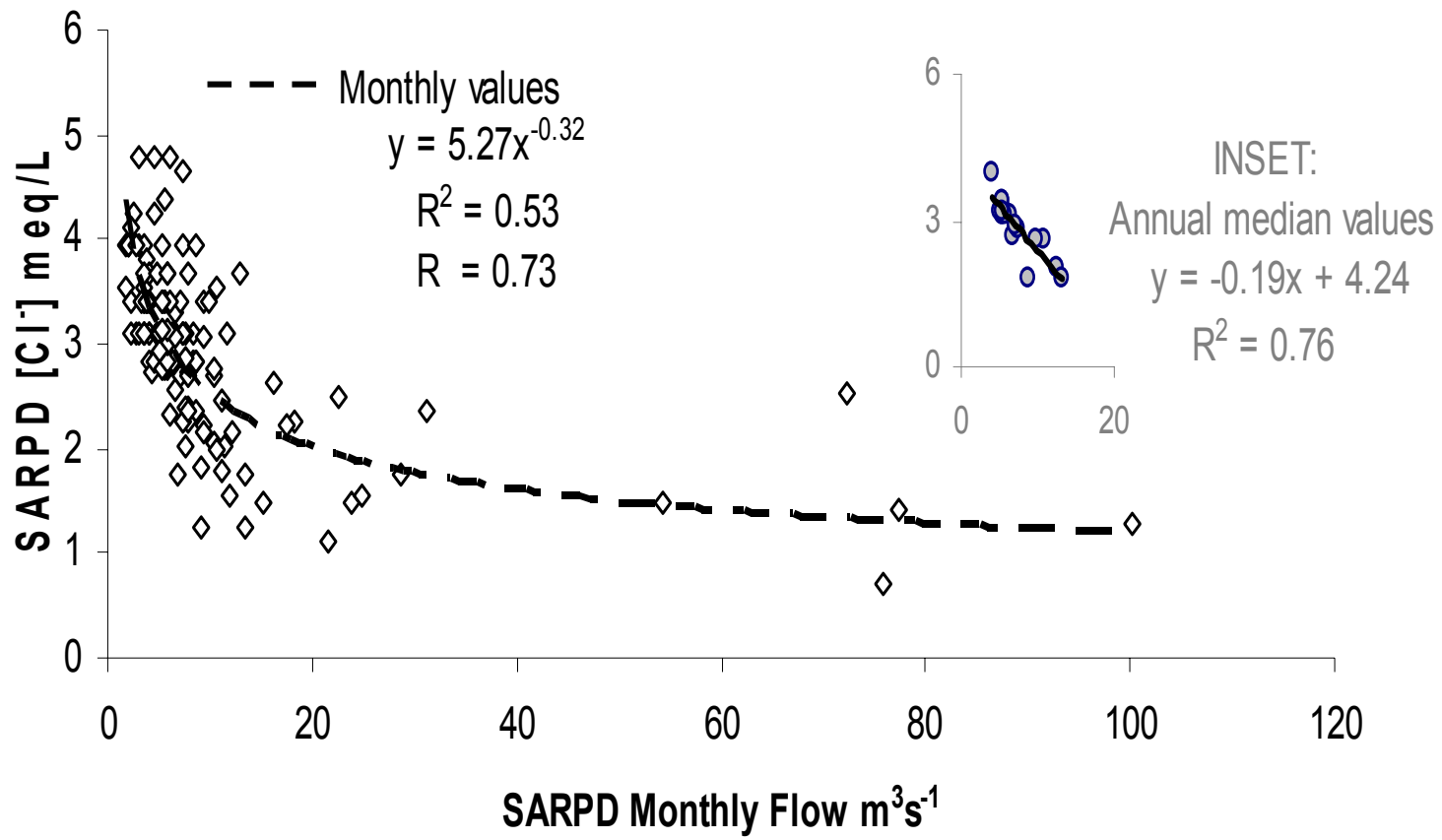


Evapotranspiration (ET)

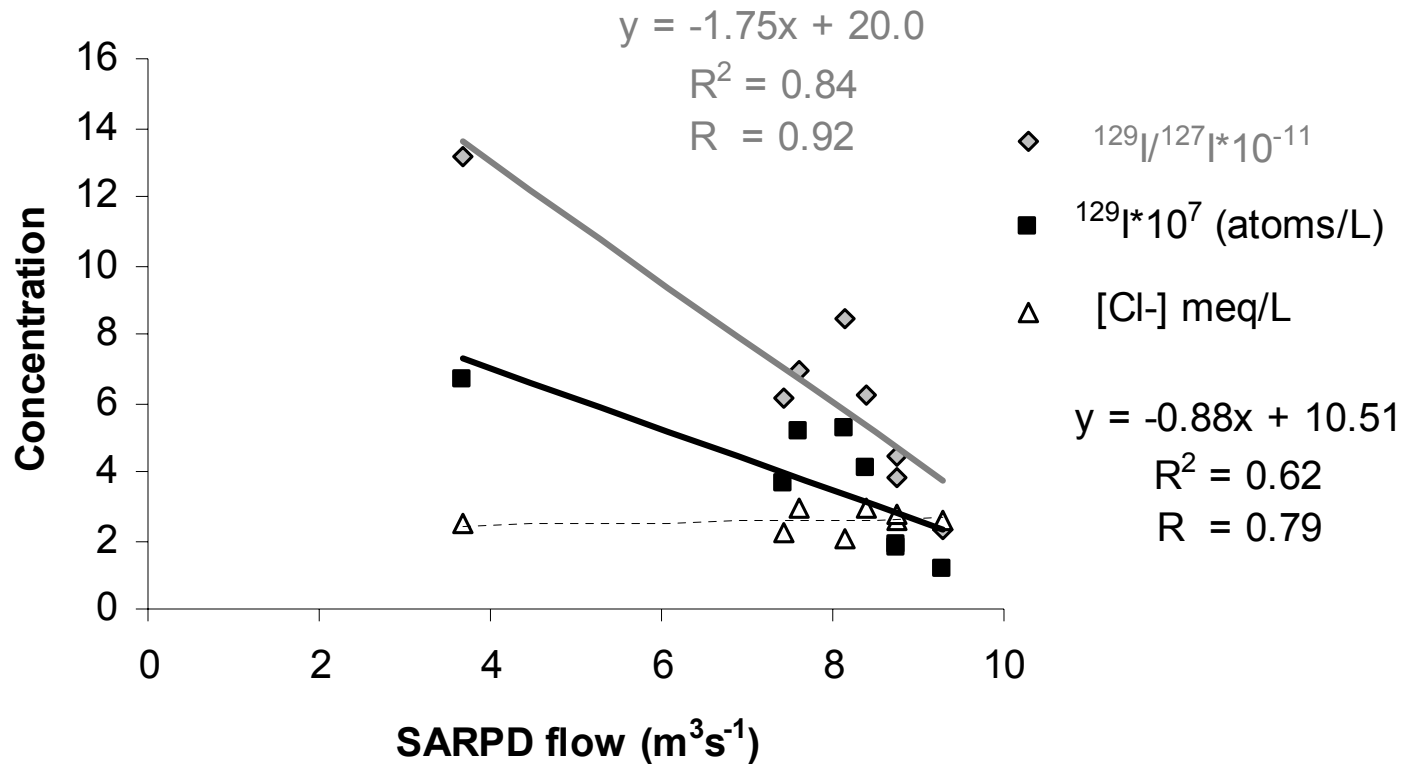
- Catchment behavior through analogy to chloride
- Long term database



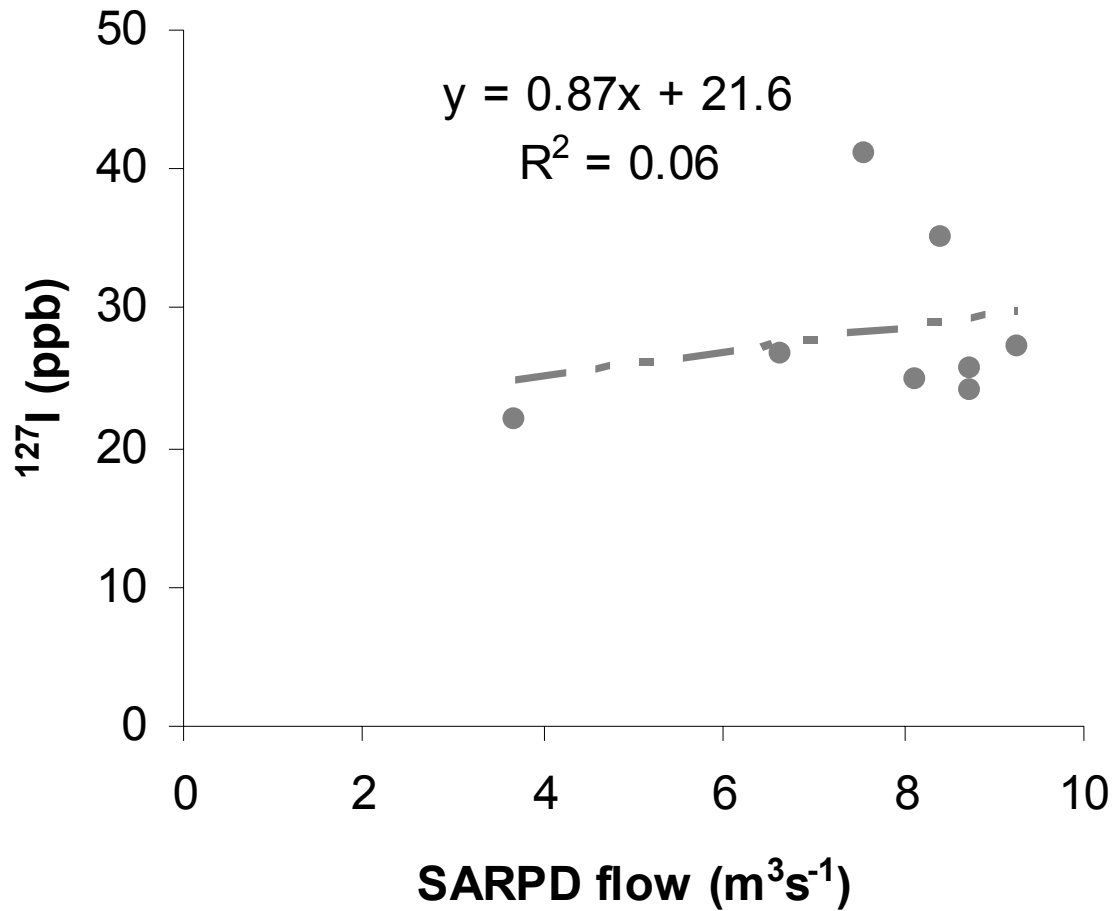
Chloride: Analogy for Iodide



Concentration vs. Flow



^{127}I vs. River Flow



Cl, ^{127}I exhibit different
mobilities than ^{129}I



Conclusions

- $^{129}\text{I}/^{127}\text{I}$ & ^{129}I increase with aquifer residence time
 - Contrasts with constant source function
 - Attributed to river flow rate,
base flow: ET
storm flow: dilution
- ^{129}I exhibits different mobility than ^{127}I , Cl
 - In different chemical form or not equilibrated
(^{127}I : $\tau \sim 1000$ yr)
- Potential for $^{129}\text{I}/^{127}\text{I}$ & ^{129}I as geochronometer for TOC:
? better than ^{14}C for TOC < 50 yrs

Questions ?



References

- Davisson, M.L., Vengosh, A., & Bullen, T. (1999a). Tracing waste-water in river and ground water of Orange County using boron isotopes and general geochemistry, *In* Lawrence Livermore National Laboratory UCRL-ID-133529 (pp. 44).
- Davisson, M.L., Hudson, G.B., Herndon, R., & Woodside, G. (1999b). Report on isotope tracer investigations in the Forebay of the Orange County Groundwater Basin: Fiscal years 1996 and 1997, *In* Lawrence Livermore National Laboratory UCRL-ID-133531 (pp. 44).
- Dissanayake, C.B. & Chandrajith, R. (1999). Medical geochemistry of tropical environments. *Earth-Science Reviews* 47, 219-258.
- Drever, J.I. (1997). Evaporation and saline waters; cyclic wetting and drying. *In* Geochemistry of Natural Waters: Surface and Groundwater Environments (pp. 335-336). New Jersey: Prentice-Hall.
- Eisenbud, M. & Gesell, T. (1997). *In* Environmental Radioactivity, Boston: Academic Press, p.656.
- Fehn, U. & Snyder, G. (2003). Global distribution of ¹²⁹I in rivers and lakes: implications for cycling in surface reservoirs. *Submitted*.
- Kocher, D.C. (1982). On the long-term behaviour of iodine-129 in the terrestrial environment. *In* Environmental Migration of Long-lived Radionuclides (pp. 669-679), Vienna: IAEA.
- OCWD (1999). Orange County Water District, Master Plan Report for 2020, [http://OCWD 2020 rpt\OCWD Online - Year 2020 Master Plan Study.htm](http://OCWD%2020%20rpt\OCWD%20Online%20-%20Year%202020%20Master%20Plan%20Study.htm) .
- McBride, M.B. (2000). Chemisorption and precipitation reactions, *In* M.E. Sumner, *Soils*, (pp. B-265-302). Boca Raton: CRC Press.



References

- Meijer, A., (2002). Conceptual model of the controls on natural water chemistry at Yucca Mountain, Nevada, *Applied Geochemistry*, 17, 793-805.
- Moran, J.E., Oktay, S., Santschi, P.H., & Schink, D.R. (1997). Surface ¹²⁹Iodine/¹²⁷Iodine ratios: Marine vs. terrestrial, Applications of Accelerators, In J.L. Duggan & I.L. Morgan, *Research and Industry*, (pp. 807-810). New York: AIP Press.
- Moran, J.E., Oktay, S.D., Santschi, P.H., & Schink, D.R. (1999a). Atmospheric dispersal of ¹²⁹Iodine from European nuclear fuel reprocessing facilities. *Environ. Sci. Technol.* 33 (15), 2536-2542.
- Moran, J.E., Oktay, S., Santschi, P.H., Schink, D.R., Fehn, U., & Snyder, G. (1999b). World-wide redistribution of ¹²⁹Iodine from nuclear fuel reprocessing facilities: Results from meteoric, river, and seawater tracer studies. IAEA-SM-354/101.
- Moran, J.E., Oktay, S.D., and Santschi, P.H. (2002). Sources of Iodine and ¹²⁹Iodine in Rivers. *Wat. Res. Res.*, 38(8), 24-1 to 24-10.
- Oktay, S.D., Santschi, P.H., Moran, J.E., & Sharma, P. (2001). ¹²⁹I and ¹²⁷I transport in the Mississippi River. *Environ. Sci. and Technol.*, 35, 4470-4476.
- Quiroz, N.G.A., Kotzer, T.G., Milton, G.W., Clark, I.D., & Bottomley, D. (2002). Partitioning of ¹²⁷I and ¹²⁹I in an unconfined glaciofluvial aquifer on the Canadian Shield. *Radiochim. Acta* 90, 1-10.
- Rahn K.A., Borys, R.D., & Duce, R.A. (1976). Tropospheric halogen gases –inorganic and organic components. *Science* 192, 549-550.
- Schnabel, C., Lopez-Gutierrez, J.M., Szidat, S., Sprenger, M., Wernli, H., Beer, J., and Synal, H.A. 2001. On the origin of ¹²⁹I in rain water near Zurich. *Radiochimica Acta*, 89, 815-822.
- Szidat, S., Schmidt, A., Handl, J., Jakob, D., Botsch, W., Michel, R., Synal, H.-A., Schnabel, C., Suter, M., Lopez-Gutierrez, J.M., and Staede, W. 2000. Iodine-129: Sample preparation, quality control and analyses of pre-nuclear materials and of natural waters from Lower Saxony, Germany. *Nucl. Instr. And Methods in Physics Res. B*, 172, 699-710.

Supplementary Information



Aquifer parameters

- Linear flow velocity

$$v = d/t$$

where d = distance (m), t is age (days)

- Hydraulic conductivity (Darcy's Law)

$$v = (K/p) \Delta h$$

where v = linear flow rate (m/day)

K is the average hydraulic conductivity (m/d)

p is the effective porosity the aquifer

(m^3 water/ m^3 soil and water)

Δh is the gradient in groundwater elevation between the endpoints (m/m),



Comparison & Units

- ^{129}I : $5.6 \text{ kg} = 1 \text{ Ci} = 3.7 \cdot 10^{10} \text{ dps or Bq}$
 $= \sim 3 \cdot 10^{25} \text{ atoms}$

$^{129}\text{I}/^{127}\text{I}$

- Natural 10^{-13} to 10^{-12}
- USA (this study) 10^{-11}
- Europe 10^{-10} to 10^{-8}
- Peak near nuclear facility 10^{-7}
- ^{129}I (this study) 10^7 atoms/L
- ^{127}I : $1 \text{ ppb} \sim 5 \cdot 10^3 \text{ atoms/L}$



Table 1. Iodine and water age data for the Orange County study wells.

| | AM 33 | AM 14 | AM 9 | AM 10 | KB 1 | Kraemer Basin | AM 44 | Anaheim Lake | AMD 9-1 | AMD 9-2 | AMD 9-3 | AMD 9-4 |
|---|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|----------------------------|--------------|---------------------------|----------------------------|-------------------|-------------------|
| $^{129}\text{I}/^{127}\text{I} * 10^{11}$ | 13.2 | 8.47 | 6.11 | 3.78 | 2.29 | 9.88 | 4.40 | 5.04 | 6.89 | 6.25 | 8.02 | 1.74 |
| ^{129}I (10^7 atoms/L) | 6.63 | 5.25 | 3.67 | 1.77 | 1.16 | 4.77 | 1.83 | 3.50 | 5.17 | 4.07 | 4.38 | 0.91 |
| ^{127}I (ppb) | 22.0 ^a | 25.0 ^a | 26.8 ^a | 25.7 ^a | 27.2 ^a | 18.3 ^a | 24.2 ^b | 26 | 41.1 | 35.1 | 31.2 | 27.3 |
| Ground water age (years) | 6.8 ^c | 1.9 ^d | 1.2 ^d | 0.2 ^d | 0.1 ^d | | 0.25 ^e [20%] | | 0.08 ^e [0%] | 0.55 ^e [40%] | n.d. ^f | 25.3 ^d |
| Sample collection date | Aug 99 | Aug 99 | Aug 99 | Sep 99 | Aug 99 | Aug 99 | Aug 99 | Sep 99 | Sep 99 | Sep 99 | Sep 99 | Sep 99 |
| Date incl. τ_w ^g | Aug 92 | Jun 97 | Aug 98 | Feb 99 | Jan 99 | May 99 | Feb 99 | Jun 99 | May 99 | Dec 98 | n.d. | May 73 |
| TOC (mg/L) ^h | 0.89 | 1.13 | 0.9 ⁱ | 0.89 | 1.88 | 3.74 | 1.19 | 4.53 | 2.44 | 1.76 | 1.52 | 1.37 |
| Cl (mg/L) ^h | 86.9 | 73.8 | 78.8 | 89 | 91.6 | 87.9 | 103 | 89.2 | 105 | 98.9 | 86.9 | 121 |



Table 2. Comparison of ^{129}I , ^{127}I , Cl concentrations and flux for regional rivers and the recharge ponds.

| River or Pond | Date | Discharge (m ³ /s) | Discharge (*10 ¹² L/yr) | Drainage (*10 ¹⁰ m ²) | ^{129}I (10 ⁷ atoms/L) | $^{129}\text{I}/^{127}\text{I} * 10^1$ | ^{127}I ppb | Cl (mg/L) | Flux $^{129}\text{I} * 10^{18}$ (atoms/yr) |
|----------------------------|-------|-------------------------------|------------------------------------|--|--|--|----------------------|-------------------|--|
| Mississippi ^a | 05/95 | 16400 | 517.00 | 327.00 | 8.00 | 85.0 | 19.8 | n.d. ^d | 41400 |
| Mississippi ^a | 06/96 | n.d. ^d | | | 5.10 | 194.0 | 5.5 | n.d. ^d | |
| Sacramento ^a | 12/95 | 845 | 26.60 | 5.00 | 0.60 | 80.5 | 1.6 | 1.7 | 160 |
| Colorado ^a | 08/96 | 111 | 3.50 | 32.00 | 3.20 | 123.2 | 5.5 | 57.0 | 112 |
| SARPD ^b | | 6.6 ^e | 0.21 | 0.58 | 4.14 ^f | | | | 8.62 ^g |
| Anaheim Lake ^c | 09/99 | | | | 3.50 | 5.04 | 26.0 | 89.2 | |
| Kraemer Basin ^c | 08/99 | | | | 4.77 | 9.88 | 18.3 | 87.9 | |

Table 3. Statistics for TOC (mg/L) from May 1990 through April 2001.
Data provided by G. Woodside, OCWD.

| Site ID | Median | Minimum | Maximum | Number of samples |
|-----------------------|--------|---------|---------|-------------------|
| <i>Surface Waters</i> | | | | |
| Anaheim Lake | 5.13 | 2.82 | 12.20 | 118 |
| Kraemer Basin | 4.32 | 2.75 | 6.56 | 17 |
| | 4.72 | | | |
| <i>Ground Waters</i> | | | | |
| AM 33 | 0.78 | 0.49 | 1.01 | 8 |
| AM 14 | 0.80 | 0.65 | 1.13 | 16 |
| AM 9 | 1.01 | 0.70 | 2.67 | 67 |
| AM 10 | 1.09 | 0.73 | 4.03 | 55 |
| KB 1 | 2.27 | 1.29 | 5.07 | 77 |
| AM 44 | 1.64 | 1.08 | 2.48 | 67 |
| AMD 9-1 | 2.46 | 1.55 | 4.32 | 110 |
| AMD 9-2 | 1.39 | 0.80 | 2.89 | 98 |
| | 1.24 | | | |

