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

Kathleen A. Schwehr[1],
Peter H. Santschi[1], and Jean E. Moran[2]

[1]Texas A&M University
[2]Lawrence Livermore National Laboratory

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make every drop count

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It is the mission of Orange County
Since 1933 Orange County Water District A Tradition of Innovation

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Outline

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

Objective

- To test the potential tracer application of the iodine isotopic ratio $^{129}I/^{127}I$ 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 $t_{1/2} = 15.6$ ma

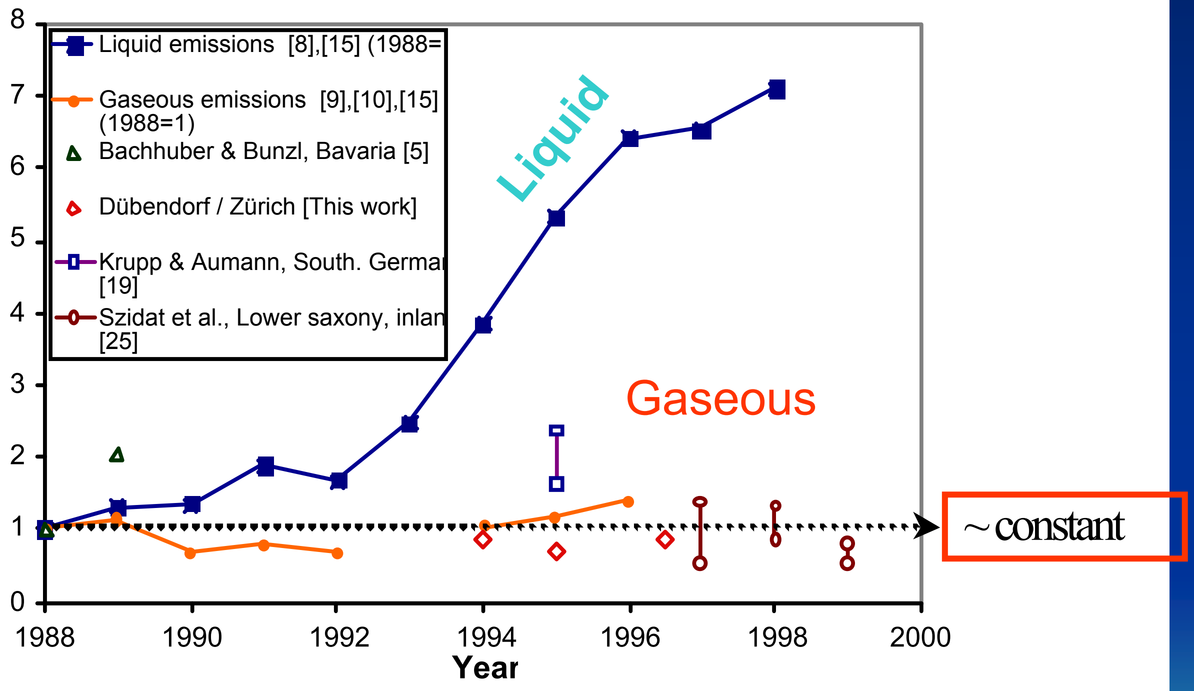
- Natural surface inventory 100 kg
- Bomb testing 150 kg
- Nuclear fuel reprocessing 2600 kg
  (Cap de La Hague, Sellafield)
- Chernobyl reactor accident (1986) 1.3 kg

1Liter drinking water:

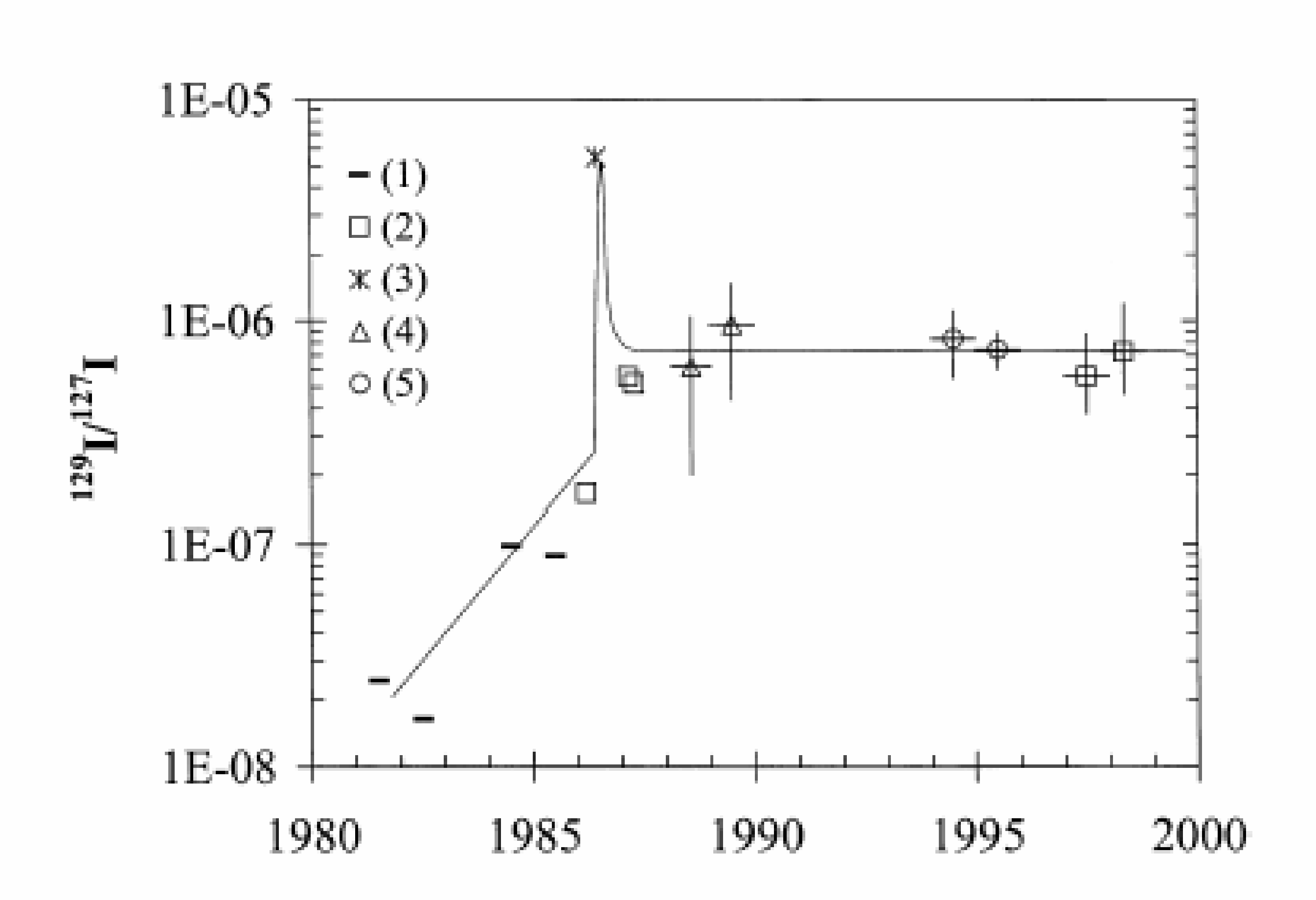
	10^{-12} Ci	
^{226}Ra	~0.2	(Eisenbud & Gesell, 1997)
^{129}I	0.0000003	

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

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

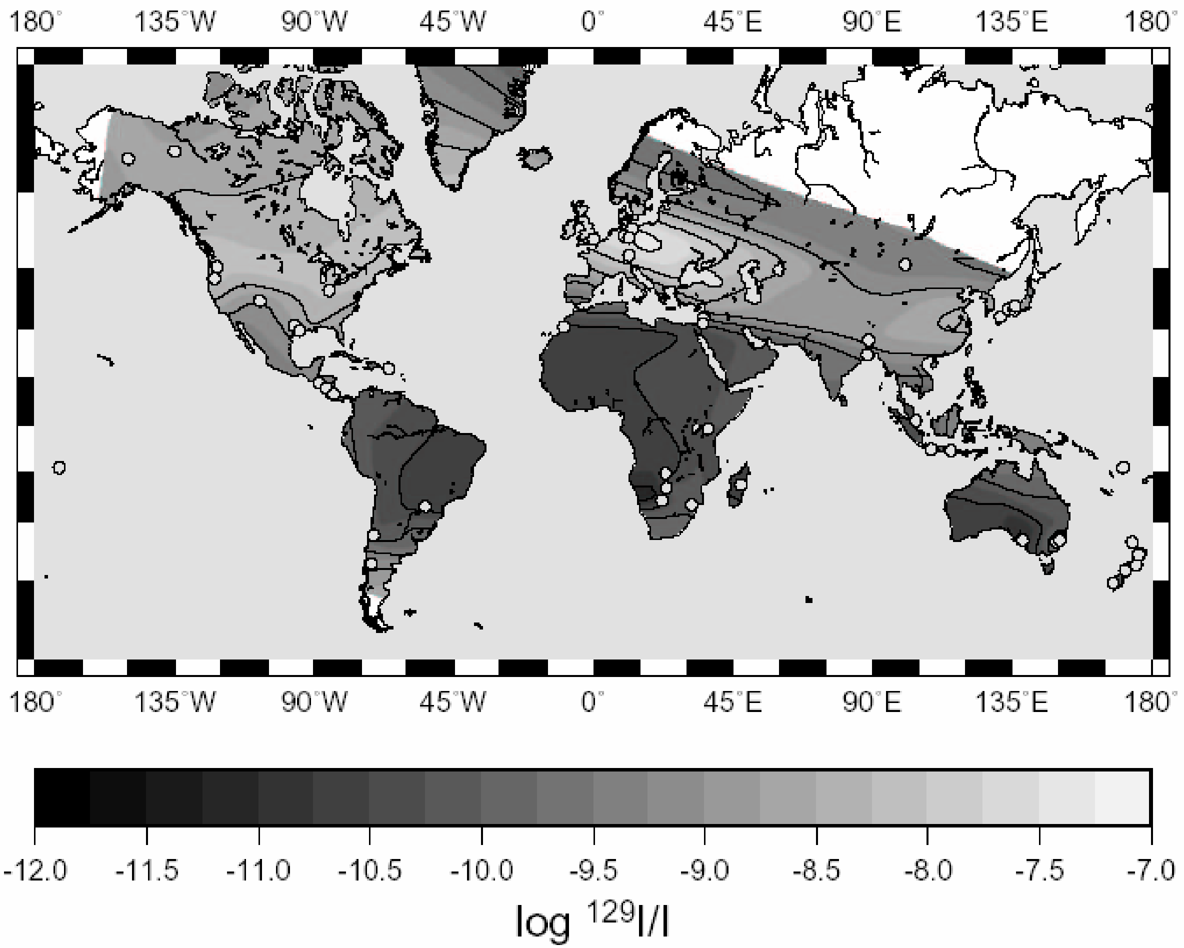


$^{129}I/^{127}I$ Ratios in Precipitation in Europe [Szidat et al., 2000]



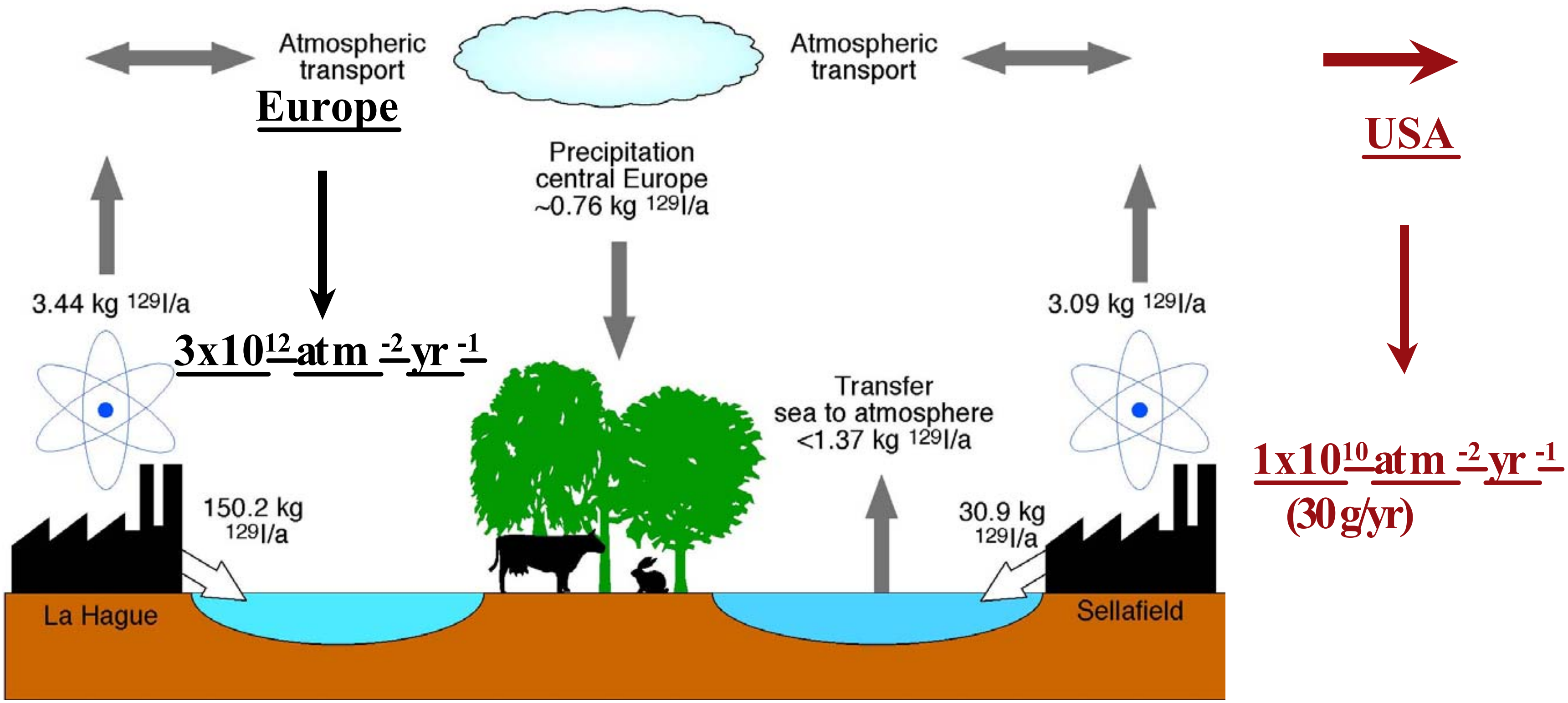
Global atmospheric transport in 11 to 18 days

Global Distribution of log $^{129}I/^{127}I$ [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)

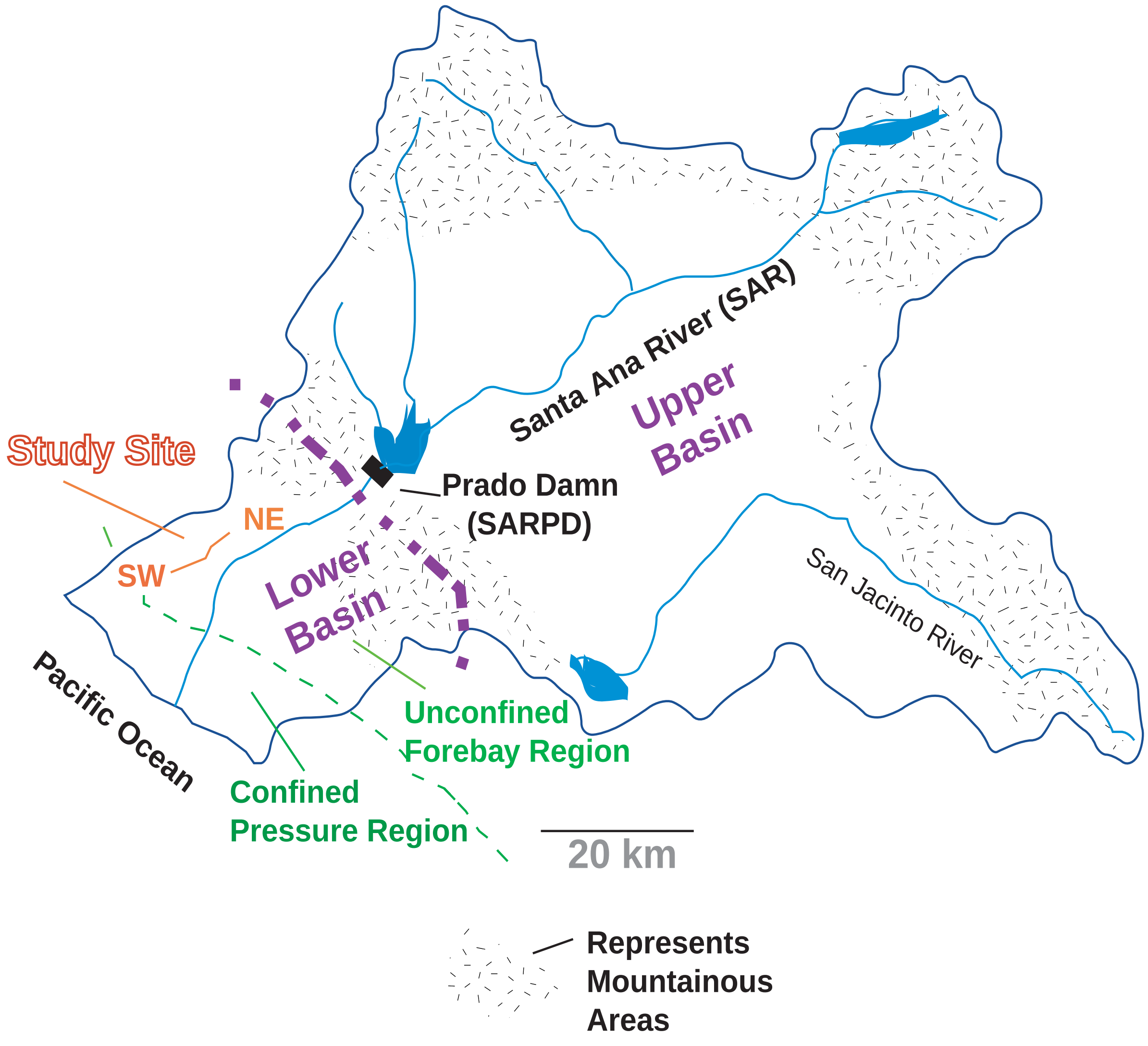
Aquifer Properties

- 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

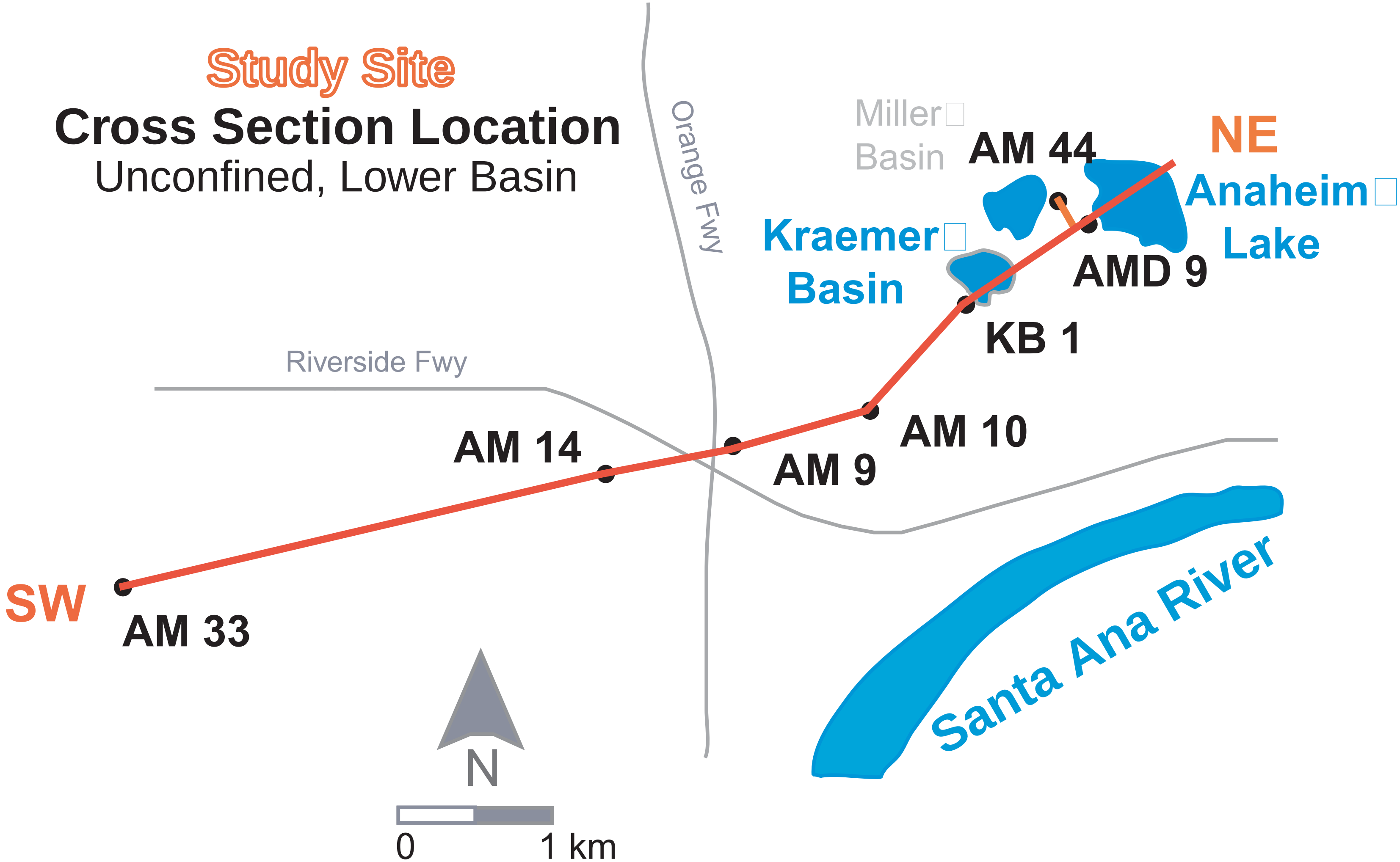
Santa Ana River Basin



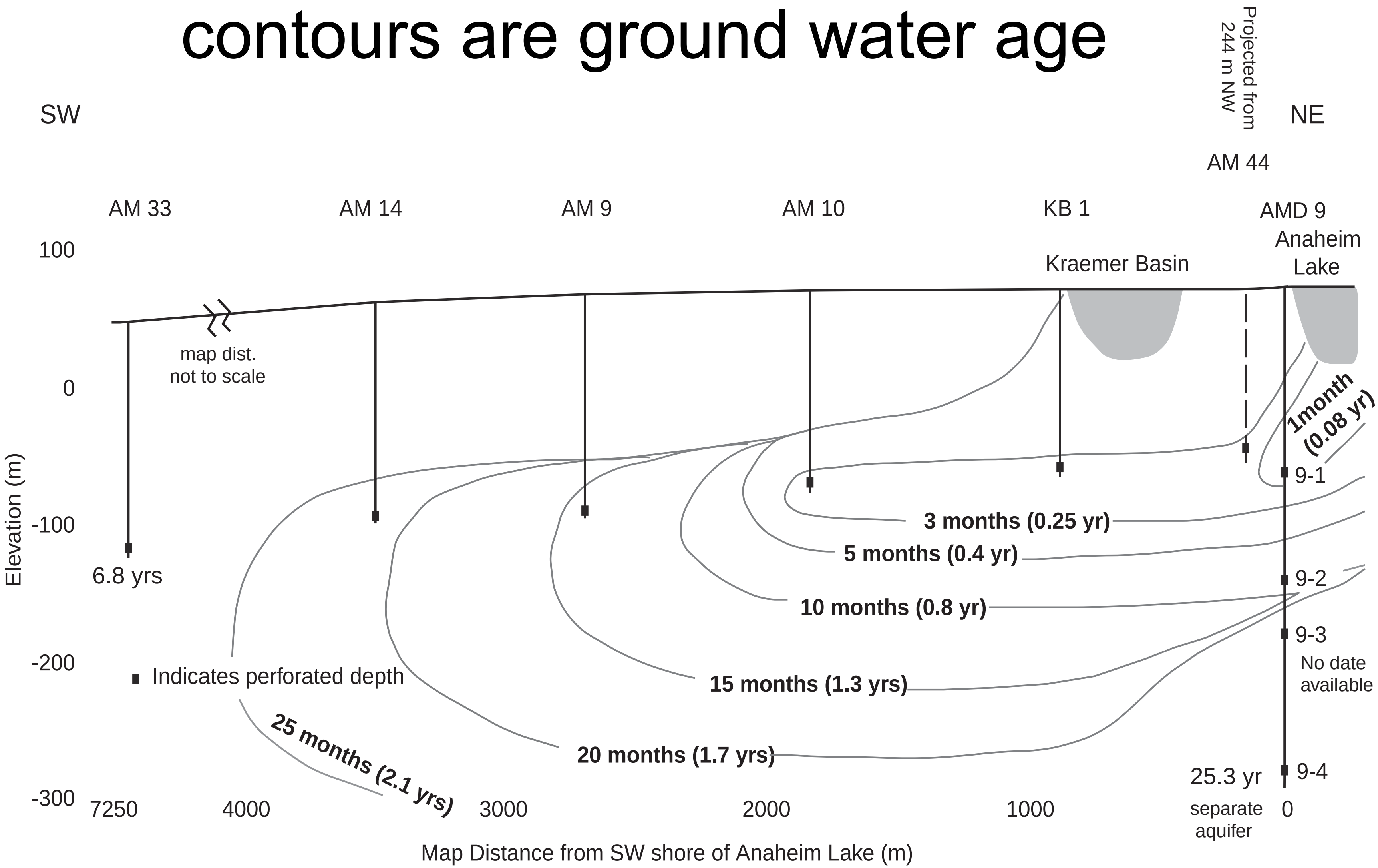
Study Site

Cross Section Location

Unconfined, Lower Basin

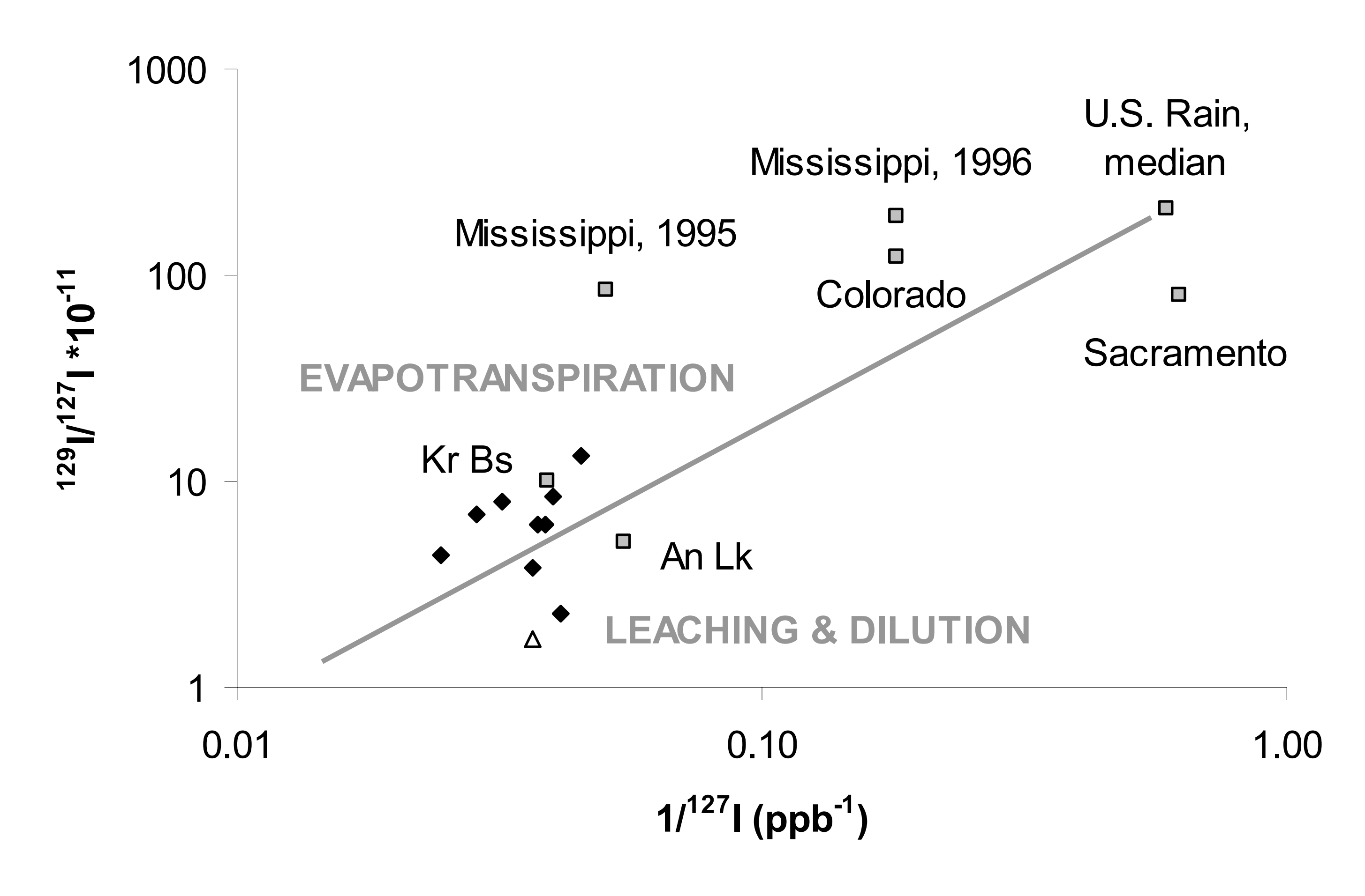


Cross Section
contours are ground water age

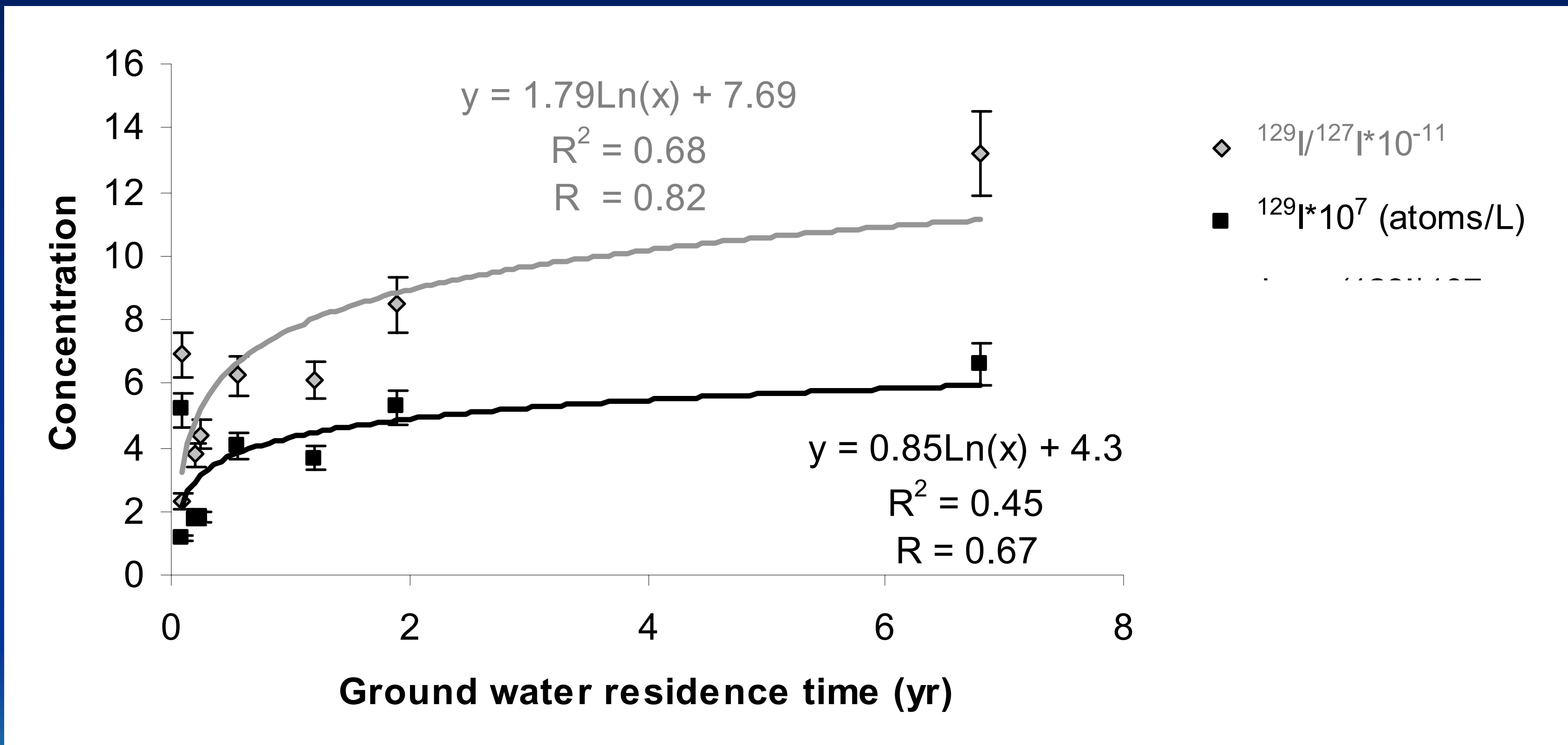


Results

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)

- **^{129}I** (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

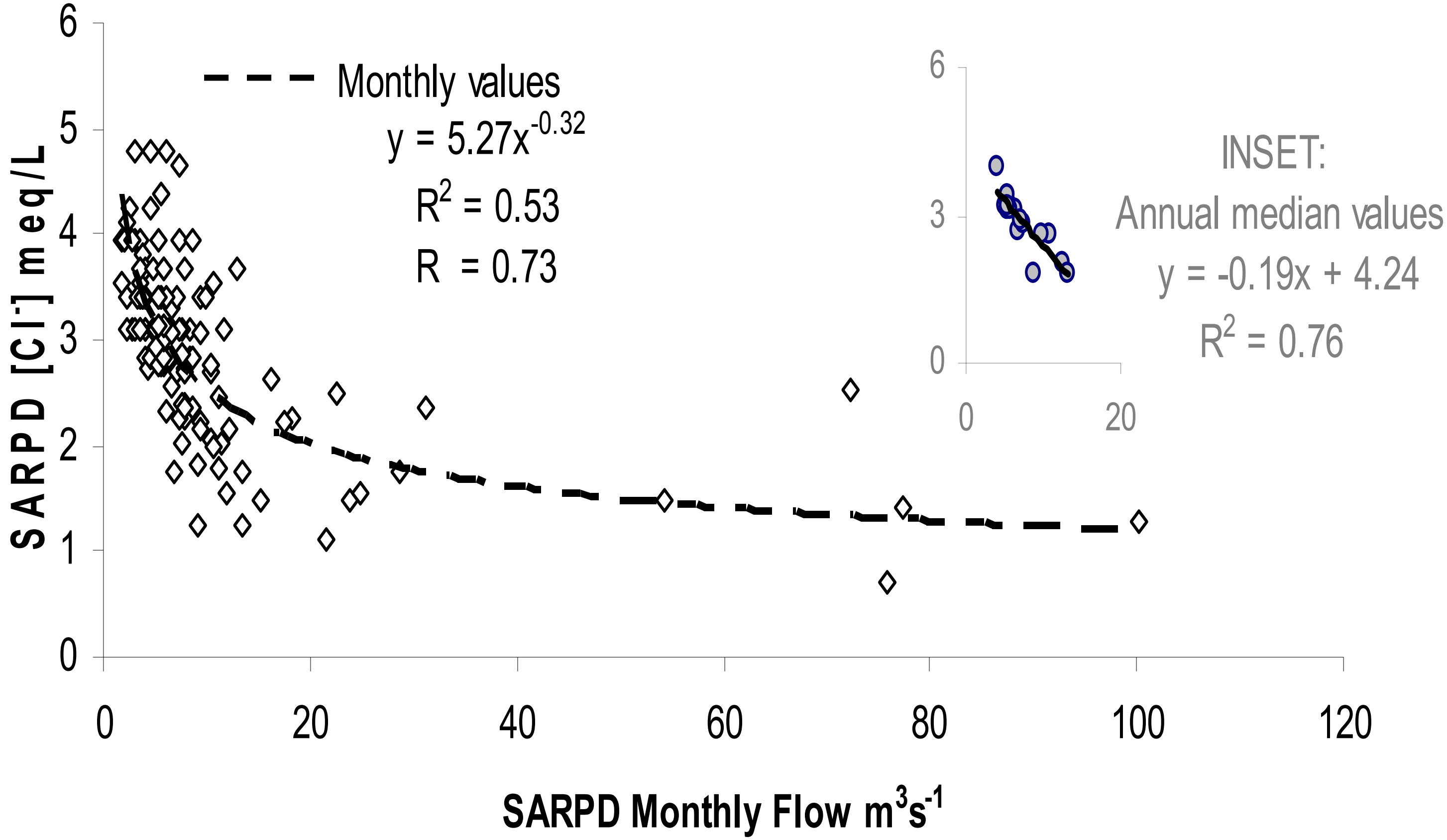
- Mixing: reclaimed water & imported water (10 to 25%) from Colorado River (COR)
  - COR ^{129}I: 3.2 * 10^7 atoms/L
  - SARPD ^{129}I: 4.1 * 10^7 atoms/L

- ET: salts concentrate & ppt during dry cycles; leach & dilute during wet cycles

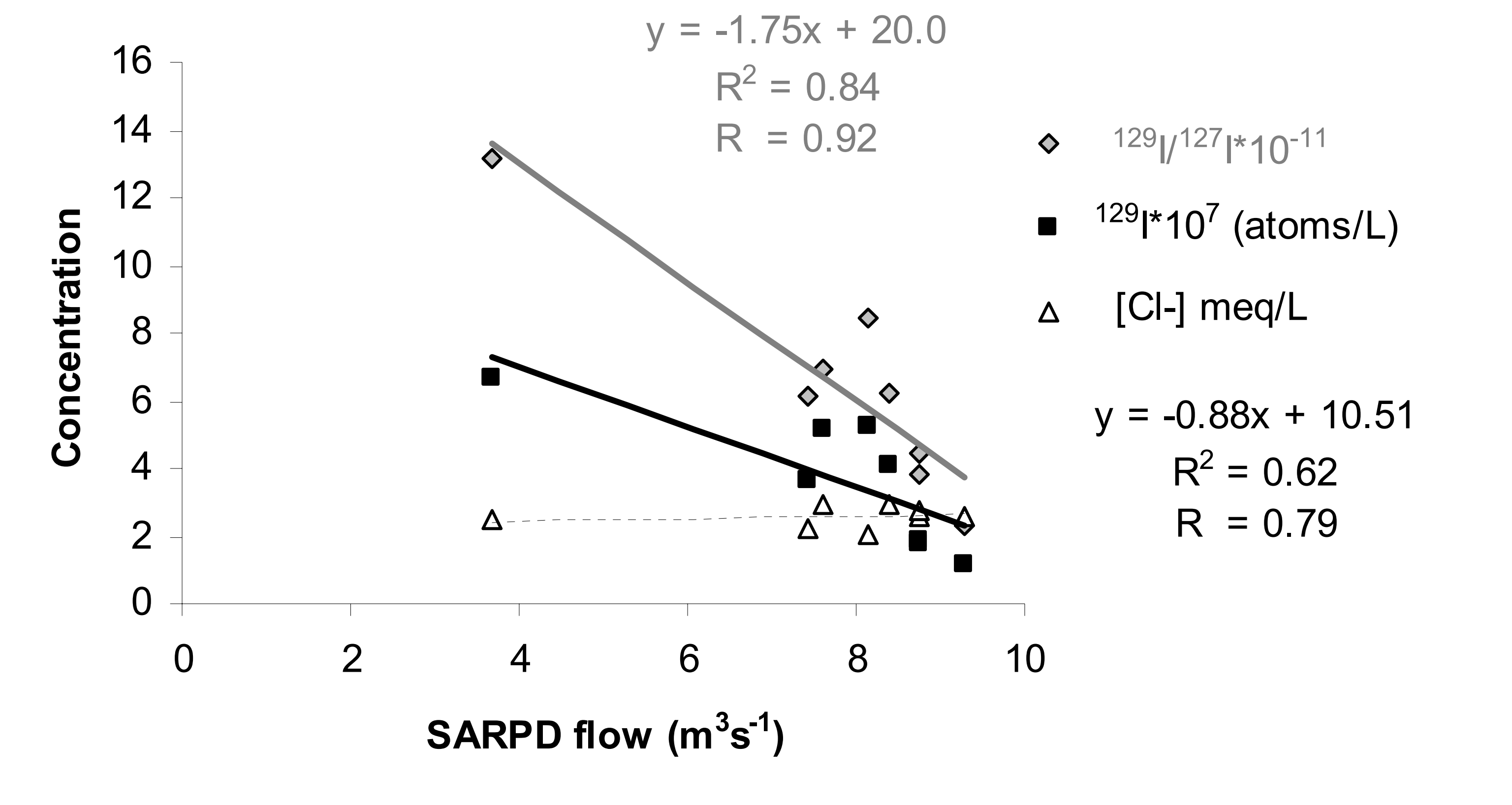
Evapotranspiration (ET)

- Catchment behavior through analogy to chloride
- Long term database

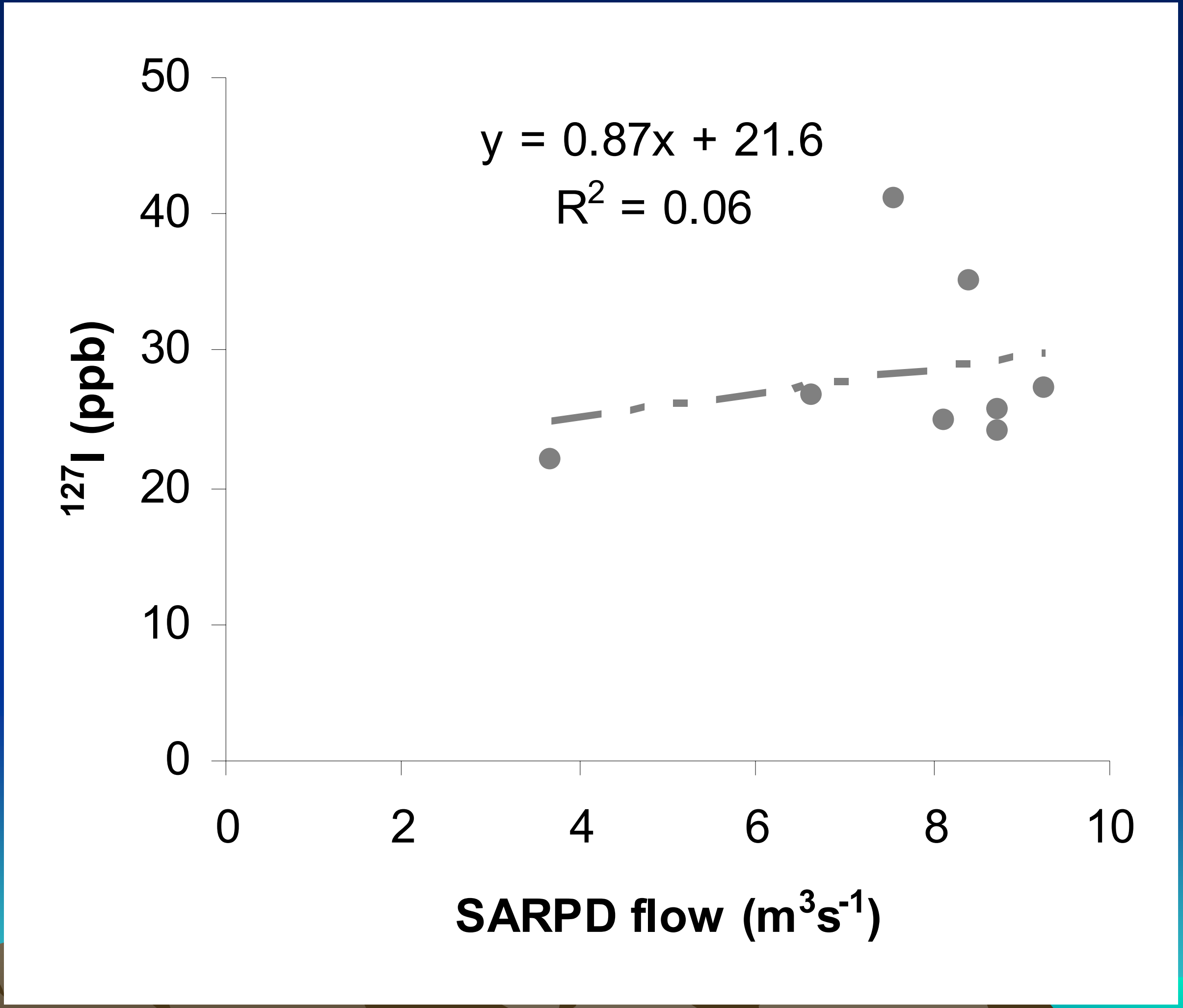
Chloride: Analogy for Iodide



Concentration vs. Flow



^{127}I vs. River Flow



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

Conclusions

- $^{129}I/^{127}I$ & ^{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 $^{129}I/^{127}I$ & ^{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 .
- 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 kg = 1 Ci = $3.7*10^{10}$ dps or Bq
  = ~ $3 * 10^{25}$ atoms

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

• 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 ppb ~ $5 * 10^{3}$ atoms/L

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

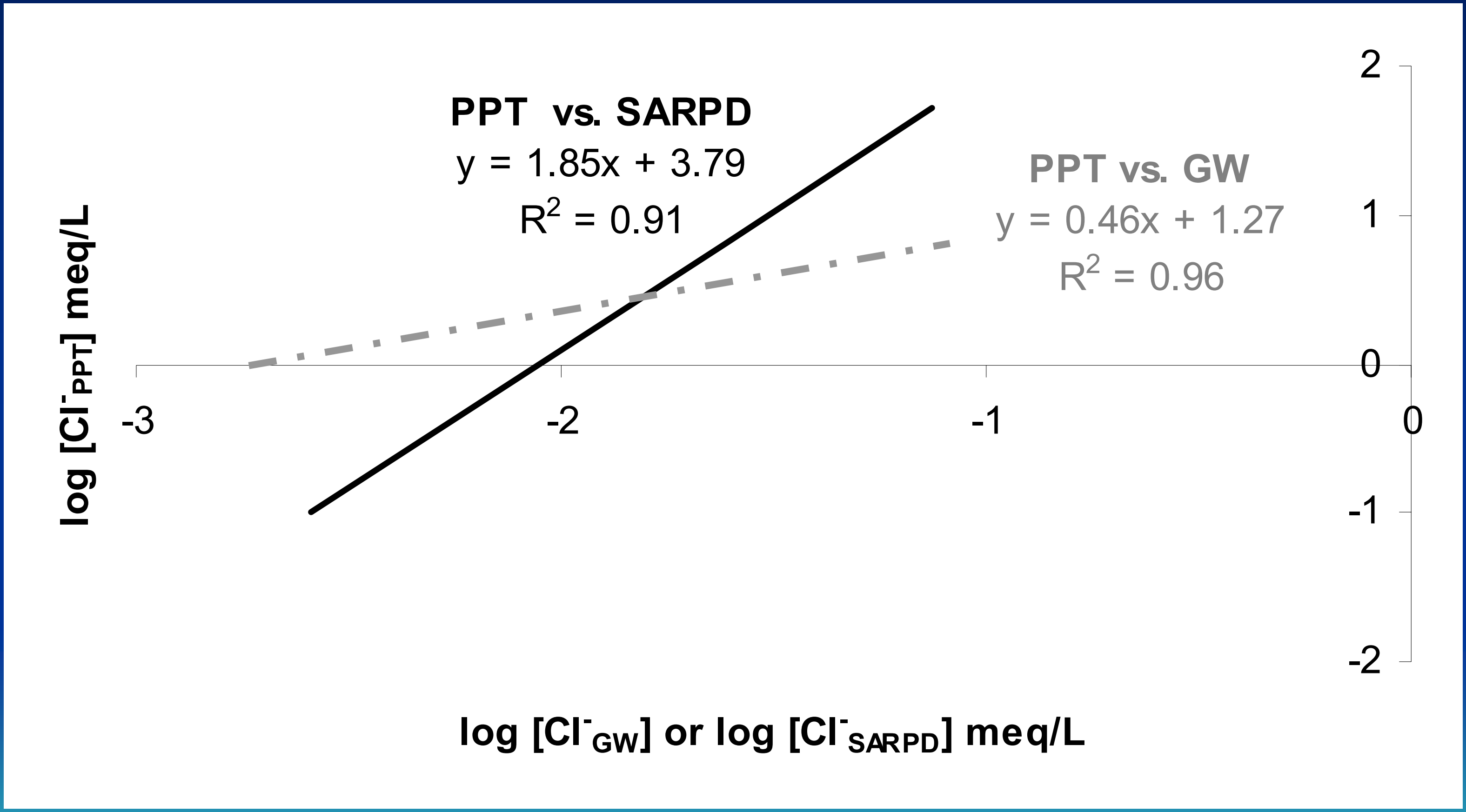
	AM 33	AM 14	AM 9	AM 10	KB 1	Kraemer Basin	AM 44	Anaheim Lake	AMD 9-1	AMD 9-2	AMD 9-3	AMD 9-4
$^{129}I/^{127}I * 10^{11}$	13.2	8.47	6.11	3.78	2.29	9.88	4.40	5.04	6.89	6.25	8.02	1.74
^{129}I (10^{7} atoms/L)	6.63	5.25	3.67	1.77	1.16	4.77	1.83	3.50	5.17	4.07	4.38	0.91
^{127}I (ppb)	22.0 [a]	25.0 [a]	26.8 [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 (10^7atoms/L)	$^{129}I/^{127}I*10^{11}$	^{127}I ppb	Cl (mg/L)	Flux $^{129}I*10^{18}$ (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
Surface Waters				
Anaheim Lake	5.13	2.82	12.20	118
Kraemer Basin	4.32	2.75	6.56	17
	4.72			
Ground Waters				
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			


PPT vs. SARPD
y = 1.85x + 3.79
R2 = 0.91
PPT vs. GW
y = 0.46x + 1.27
R2 = 0.96
log [Cl-PPT] meq/L
log [Cl-GW] or log [Cl-SARPD] meq/L
2
1
0
-1
-2
-3
-2
-1
0