Application of $^{129}I/^{127}I$ Ratios in Groundwater Studies Conducted at Los Alamos National Laboratory, New Mexico

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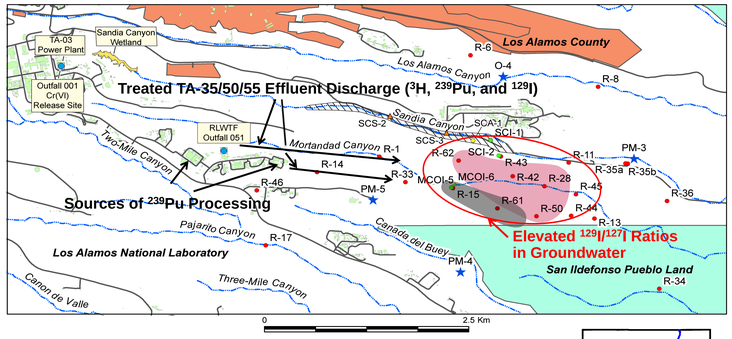
Abstract

Los Alamos National Laboratory (LANL) is an operating nuclear site that has released treated effluents from three plutonium-processing facilities since the mid 1940s. The radioisotope ^{129}I ($T_{1/2}$ = 15.7 Myrs) derived from ^{235}U and ^{239}Pu processing at LANL is locally detected in groundwater above background concentrations. This isotope provides a unique tracer for groundwater investigations conducted at LANL that helps to identify source releases linked to groundwater-flow paths in aquifers subject to binary and ternary mixing of natural- and industrial-derived waters containing chromate and other chemicals. Bromide, chlorate, chloride, nitrate, perchlorate, sulfate, and tritium were associated with multiple outfalls at LANL and, therefore, do not provide unique chemical signatures identifying a specific point of release or source. Natural and anthropogenic ratios of $^{129}I/^{127}I$ measured in groundwater samples collected at LANL were quantified using accelerator mass spectrometry at Purdue Rare Isotope Measurement Laboratory, Purdue University. Anthropogenic ratios of $^{129}I/^{127}I$ range from 1,531 X 10^{-15} to 10,323 X 10^{-15} within perched-intermediate groundwater present in volcanoclastic and basalt aquifers (210 – 216 m depth). Anthropogenic ratios of $^{129}I/^{127}I$ range from 359 X 10^{-15} to 4,350 X 10^{-15} within the regional aquifer (280 m depth) consisting of volcanoclastic sediments of variable hydraulic properties. Local background ratios of $^{129}I/^{127}I$ have a narrow range of 171 X 10^{-15} to 378 X 10^{-15} in the regional aquifer. Dissolved iodide measured in groundwater at LANL is stable dominantly as iodate. Background concentrations of dissolved iodate (0.1 to 33.2 nM) are less variable compared to anthropogenic iodate (8.0 to 246 nM) in groundwater at the site. Variability in concentrations of anthropogenic iodate is controlled by heterogeneous source releases of iodate over time and non-uniform mixing of groundwater in the different aquifers.

Plume Map of Total Dissolved Chromium and Elevated Above Background $^{129}I/^{127}I$ Ratios within the Regional Aquifer, Los Alamos National Laboratory



Natural and Anthropogenic Sources of $^{129}Iodine$

Natural sources of ^{129}I include cosmic spallation of xenon and fission of uranium occurring in the subsurface. Fission of uranium releases ^{129}I to groundwater and the atmosphere from volcanic emissions. Residence times for ^{129}I in the atmosphere and oceans are 2 weeks and 40,000 years, respectively.

Anthropogenic ^{129}I is a fission product of ^{235}U and ^{239}Pu processing at nuclear facilities. Isotope ratios of $^{129}I/^{127}I$ increased in some parts of the world during the 1960's resulting from atmospheric nuclear testing. Atmospheric $^{129}I/^{127}I$ ratios ranged from 10^{-7} to 10^{-4} in the past.

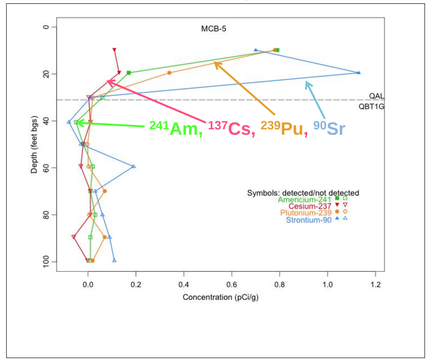
$^{129}I/^{127}I$ Ratios, ^{129}I, and IO_3^- Analytical Results

Name	$^{129}I/^{127}I$ (10^{-15})	Uncertainty (10^{-15})	Atoms of ^{129}I/g of Sample	Uncertainty	IO_3 (ug/L)
Background					
LAM Spring-12-17-13	301	91	87681	2153	6.90
LAOI(a)-1.1-8-15-13	303	89	87559	529	1.60
PM-4-7-10-13	220	261	64430	643	4.50
PM-5-9-18-13	378	135	109950	595	5.20
R-24	171	27	48209	849	1.23
Perched Intermediate Zones					
LAOI-3.2-8-13-13	190	35	55256	13257	3.45
LAOI-3.2a-8-14-13	332	112	97772	11857	3.20
LAOI-7-8-8-13	163	31	47094	13918	4.60
MCOI-5-11-8-13	1924	289	565618	21112	2.90
MCOI-6-1-17-14	10172	879	2976228	7422	1.20
MCOI-6-5-8-13	15431	394	4681291	42393	1.20
POI-4-6-3-13	389	53	118040	2652	11.00
R-6i-8-12-13	264	67	76543	2621	7.30
SCI-2-11-14-13	10323	632	3047588	6125	2.10
TA-53i-8-9-13	274	37	79459	17751	3.55
TW-2Ar-6-6-13	265	28	77676	3747	43.00
Regional Aquifer					
R-4-6-10-13	289	202	86243	1965	5.80
R-11-1-9-14	209	94	61113	1176	2.60
R-11-11-5-13	424	105	123392	1368	2.60
R-28-5-6-13	4350	157	1291933	5078	1.20
R-35a-1-16-14	497	127	146497	694	5.60
R-42-11-7-13	3201	417	925831	6087	0.92
R-43 S1-1-21-14	534	123	154879	4000	1.50
R-43 S1-11-19-13	359	94	105853	1579	1.50
R-44 S1-1-13-14	470	137	136196	518	3.50
R-45-S1-1-14-14	674	191	195613	746	2.80
R-50-S1-1-15-14	559	123	164640	994	1.90
R-50-S1-11-12-13	466	112	135424	1272	1.90
R-62-11-12-13	779	187	229680	16117	1.70
R-62-7-19-13	898	225	254214	1389	1.90
Blanks					
Cblk-3354-4	6	2			
Cblk-3354-3	10	2			
Cblk-3354-2	6	1			
Cblk-3354-1	5	1			

$^{129}Iodine$ Yield, Percent Per Fission (http://www-nds.iaea.org/sgnucdat/c3.htm)

Isotope	Thermal	Fast
^{232}Th	not fissile	0.431 ± 0.089
^{233}U	1.63 ± 0.26	1.73 ± 0.24
^{235}U	0.706 ± 0.032	1.03 ± 0.26
^{238}U	not fissile	0.622 ± 0.034
^{239}Pu	1.407 ± 0.086	1.31 ± 0.13
^{241}Pu	1.428 ± 0.36	1.67 ± 0.36

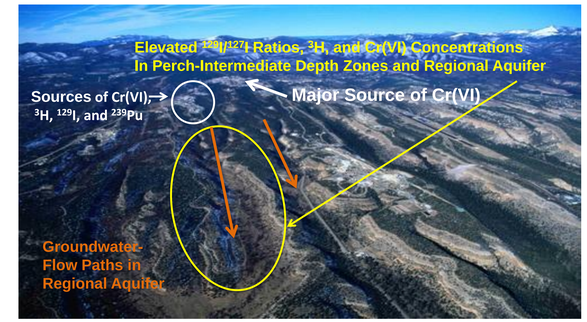
Distributions of Radionuclides in Core Hole MCB-5, Mortandad Canyon, New Mexico



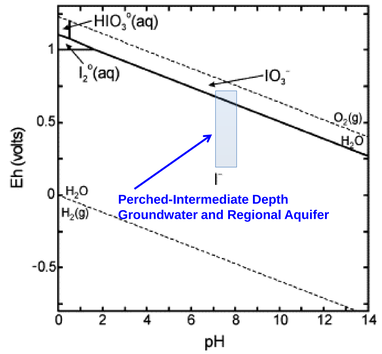
Analytical Methods

^{129}I and ^{36}Cl: accelerator mass spectrometry
Oxyanions: liquid chromatography/mass spectrometry-mass spectrometry
Actinides: alpha spectrometry; ^{90}Sr and ^{137}Cs: gamma spectrometry
Tritium: electrolytic enrichment, direct counting, and liquid scintillation
Anions: ion chromatography
Metals: high resolution-inductively coupled plasma-mass spectrometry

Jemez Mountains and Pajarito Plateau, New Mexico (view to the west showing dominant industrial discharges)

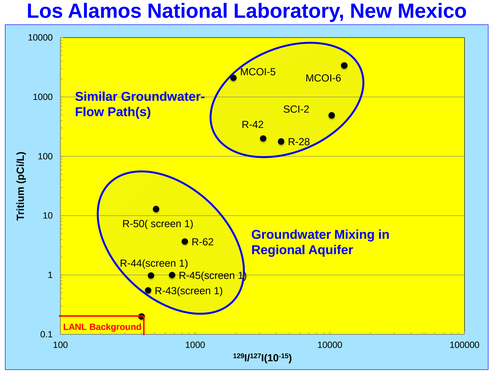


Eh-pH Diagram for Iodine at 25°C and 1 Bar



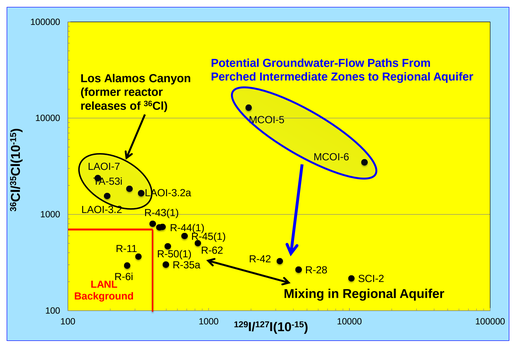
Eh-pH diagram showing the dominant aqueous complexes of iodine at 25°C, 1 bar, and a concentration of 10^{-8} mol/L total dissolved I. Source: Um et al, 2004.

$^{129}I/^{127}I$ Ratios Versus Tritium in Groundwater, Los Alamos National Laboratory, New Mexico



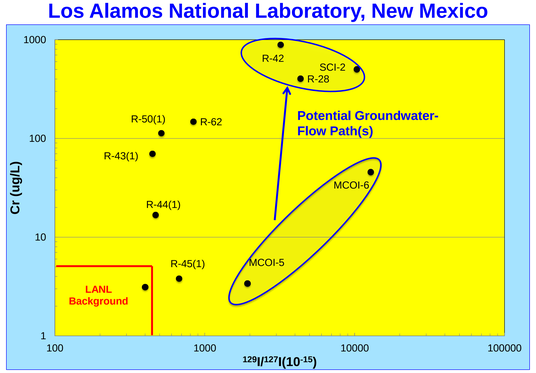
This figure shows $^{129}I/^{127}I$ ratios versus tritium activities in perched-intermediate depth groundwater (monitoring wells MCOI-5, MCOI-6, and SCI-2) and in the regional aquifer (monitoring wells R-28, R-43(1), R-44(1), R-45(1), R-50(1), and R-62). The two groups show that tritium and $^{129}I/^{127}I$ ratios in groundwater samples collected from monitoring wells MCOI-5, MCOI-6, and SCI-2 are associated with groundwater-flow paths originating from the Mortandad watershed, where voluminous treated effluent containing ^3H, ^{239}Pu, and other species were discharged since the 1950s.

$^{129}I/^{127}I$ Ratios Versus $^{36}Cl/^{35}Cl$ Ratios in Groundwater, Los Alamos National Laboratory, New Mexico



This figure shows $^{129}I/^{127}I$ ratios versus $^{36}Cl/^{35}Cl$ ratios in perched-intermediate depth groundwater (monitoring wells LAOI-7, LAOI-3.2, LAOI-3.2a, MCOI-5, MCOI-6, SCI-2, R-6i and TA-53i) and in the regional aquifer (monitoring wells R-11, R-28, R-35a, R-43, R-44, R-45, R-50, and R-62). The perched-intermediate depth groundwater (MCOI-5 and MCOI-6) provides potential recharge to the regional aquifer in the vicinity of R-42 and R-28 in Mortandad Canyon. Mixing of regional aquifer groundwater occurs at R-42 and R-28 continuing east-southeast to R-50, R-44, and R-45. Monitoring wells R-11, R-35a, and R-43 represent other separate groundwater-flow paths in Sandia Canyon. Los Alamos Canyon is located north of Sandia and Mortandad Canyons.

$^{129}I/^{127}I$ Ratios Versus Cr Concentrations in Groundwater, Los Alamos National Laboratory, New Mexico



This figure shows $^{129}I/^{127}I$ ratios versus dissolved Cr concentrations in perched-intermediate depth groundwater (monitoring wells MCOI-5, MCOI-6, and SCI-2) and in the regional aquifer (monitoring wells R-28, R-43, R-44, R-45, R-50, and R-62). Monitoring wells R-28, R-42, and SCI-2 contain high concentrations of dissolved Cr (stable as CrO_4^{2-}) and high $^{129}I/^{127}I$ ratios. These three monitoring wells are separated from the other monitoring wells with respect to $^{129}I/^{127}I$ ratios and dissolved Cr concentrations. Most of the Cr(VI) released from LANL outfalls (TA-02, TA-03, and TA-48) has migrated through the vadose zone (280 m thickness) and has mixed with regional aquifer groundwater within Sandia and Mortandad watersheds.

Summary and Conclusions

- The radioisotope ^{129}I ($T_{1/2}$ = 15.7 Myrs) derived from ^{235}U and ^{239}Pu processing at Los Alamos National Laboratory is locally detected in groundwater above background activities.
- This isotope provides a unique tracer for groundwater investigations conducted at LANL that helps to identify source releases linked to groundwater-flow paths in aquifers. Aquifer systems are subject to binary and ternary mixing of natural- and industrial-derived waters containing iodate, chromate, and other chemicals.
- Local background ratios of $^{129}I/^{127}I$ vary from 171 X 10^{-15} to 378 X 10^{-15} in the regional aquifer.
- Anthropogenic ratios of $^{129}I/^{127}I$ range from 1,531 X 10^{-15} to 10,323 X 10^{-15} within perched-intermediate depth groundwater in Mortandad Canyon.
- Anthropogenic ratios of $^{129}I/^{127}I$ range from 359 X 10^{-15} to 4,350 X 10^{-15} within the regional aquifer in Mortandad Canyon.
- Variability in $^{129}I/^{127}I$ x 10^{-15} ratios and concentrations of anthropogenic iodate is controlled by heterogeneous source releases of this isotope and iodate over time and non-uniform mixing of groundwater in different aquifers.

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