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TECHNETIUM-99 AND IODINE-129 IN THE BURIAL GROUND PLUME (U)

by

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TECHNETIUM-99 AND IODINE-129 IN THE BURIAL GROUND PLUME

SUMMARY

As anionic species, both Tc-99 (as TcO_4^-) and I-129 are expected to be mobile in the soils beneath the burial ground. These two isotopes were analyzed in groundwater collected from research wells screened in the tritium plume leaving the burial ground. The maximum observed concentrations of 22 pCi/L of Tc-99 and 12 pCi/L of I-129 in the plume wells are above background levels and confirm the mobility of these species. The earlier measurements included in this report have been reported before.^{1,2} The data indicate that the I-129 concentrations may be increasing with time.

Because of the long time (greater than one year) involved in obtaining results for these ultra low-level analyses, this work included analysis of only a few wells inside the burial ground. This limited selection of wells does not permit a mapping of the groundwater concentration isopleths.

INTRODUCTION

Equilibrium considerations show that Tc-99 should be present as TcO_4^- in the burial ground soil/water system.³ The same considerations for iodine show that it should be present as iodide (I^-) or iodate (IO_3^-), depending on the pH of the water.^{4a} When these data are considered along with the generally low anion exchange capacity of SRP soils, it is expected that these species should be mobile once leached from the waste. The migration which occurs should be along the groundwater flow paths.

Laboratory studies of the soil/water distribution coefficients (K_d 's) confirm that both Tc-99 and I-129 should be mobile in SRP soils. The K_d for Tc-99 as the pertechnetate anion were measured as less than 1 mL/g.³ Similar values measured for iodide on SRP soils show a slightly lower mobility, with K_d 's ranging from 0.6 to 6.6 mL/g.^{4b} To validate the mobility predicted by these laboratory measurements, groundwater samples were collected for analysis from wells intersecting the tritium plume downflow of the burial ground.

WELL LOCATIONS AND SAMPLING

The tritium plume indicates the major flow path of water leaving the SRP low-level waste burial ground. Investigation of Tc-99 and I-129 migration centered on analysis of groundwater samples from wells in the tritium plume. Well PDQ5 is the plume well furthest from the burial ground and is about 600 feet south of the the southwest corner of the burial ground. The well has a 5 foot long screen at the 46 foot depth, which is roughly in the middle of the tritium plume, approximately 30 feet below the surface of the water table.

Wells BGC-2C and BGC-3C are located adjacent to the burial ground fence on the south side of 643-G. They are wells in 2 of the 3 clusters designed to determine the depth and thickness of the tritium plume.⁵ These two particular wells are those in each cluster whose screens are in the middle of the tritium plume, as indicated by the highest tritium concentration of any well in the cluster. By virtue of the location of these wells (see Figure 1) they should be sampling water from different portions of the burial ground, and should not necessarily have the same composition.

Well BG-109 was used as a control well for these studies. It is located to the north and east of the 643-G burial ground, just to the south of the water table divide. The water which is sampled by this well should not have been in contact with any

waste, but should contain any nuclides which might be present due to fallout or rainout. Its location is also shown in Figure 1.

All four of these wells have been sampled using a brass, Kemmerer-type sampler which collects the water at the screen depth. It was washed carefully between uses to avoid cross contamination of the wells. In addition, the control well was sampled first, followed by the other wells in order of increasing likelihood for the nuclides of interest being present.

Wells E-17, G-21, and I-13 are located within the burial ground fence, and are screened at the water table. They represent three of the wells chosen for these analyses, based on their proximity to wastes which may have contained the radionuclides of interest. These wells were sampled using stainless steel bailers dedicated to each well. This technique samples only the groundwater at the surface of the water table, which was adequate for the purpose of this study.

All of the samples were split upon collection to allow a variety of analyses. The samples were poured into clean polyethylene bottles and were spiked with the tracers necessary for chemical yield determinations within 24 hours of collection. Any losses during storage are accounted for by this method.

METHODS OF ANALYSIS

The Tc-99 and I-129 analyses were performed by M. V. Kantelo of ETD. Tc-99 was analyzed by Isotope Dilution Mass Spectrometric methods.⁶ The I-129 analysis was by neutron activation analysis using K reactor as the source of neutrons. This method also gives an analysis of I-127 (stable iodine) on the same sample. Analyses by both techniques were performed in duplicate. The samples were not treated prior to submission to ETD.

Chemical analyses were performed by ion chromatography (nitrate, sulfate, chloride, and fluoride), ICP emission spectroscopy (Si, Na, Ba, Ca, Sr, Mn, Mg, Fe, and P), or atomic emission spectroscopy (K, Mg, and Fe), by ADD. The samples were filtered prior to submission, and those samples for ICP/ES or AES were acidified with ultrapure nitric acid after filtering.

Dissolved oxygen content, pH, eH, and conductivity were measured on unfiltered samples the same day as collection, immediately upon return of the samples to the laboratory.

RESULTS

Technetium

Results of the Tc-99 and chemical analyses are given in Table 1 with the time trend for Tc-99 given in Table 3. The maximum Tc-99 concentration observed was 22 ± 11 pCi/L. This compared to a concentration in the control well of 0.021 pCi/L. In all of the plume well samples analyzed, there was evidence for Tc-99 in the plume water in excess of the background levels measured in BG-109. Presence of the Tc-99 in the plume wells confirms the expected mobility of this radionuclide.

Concentrations of up to 5 pCi/L were observed in the groundwater monitoring wells E-17 and G-21 inside the burial ground fence. Although it does not directly apply to the groundwater beneath the burial ground, the EPA Interim Drinking Water Standards for radionuclides can be used as benchmarks with which to compare these measured values. For Tc-99 this benchmark is 900 pCi/L.

Iodine

Results of the I-127 and I-129 analyses are given in Tables 2 and 4, in the same format as for technetium. The maximum concentration of I-129 observed in any of the plume wells was 12 pCi/L in well PDQ5. In addition, the three samples collected from well PDQ5 between April 1982 and August 1983 show an increasing trend for both I-129 and I-127, from 0.25 ± 0.05 pCi/L to 12 ± 2 pCi/L. No samples were collected after August 1983.

These results agree quite well with the observed concentration range of 0.5 to 5.0 pCi/L from a lysimeter containing iodine on berl saddles.⁷ The results of the lysimeter indicated that the observed concentrations were close to the solubility limit for the silver iodide waste. The silver iodide coated berl saddles have contributed about 12 Ci of I-129 to the burial ground, and are probably the major source of I-129 in the burial ground.⁷

The fact that the I-129 and I-127 are always present in the same samples suggests that they are both from the waste. However, the present results indicate far more I-127 than would be expected from fission product waste alone (assuming typical fission yields). The present work indicates that the mass ratio of I-127 to I-129 ranges from 10^4 to 10^6 . For the lysimeter, where the only waste was the berl saddles this ratio was reported as 400.⁷

Again, one can use the 4 mrem EPA Interim Drinking Water Standard for I-129 as a benchmark for comparison to the measured concentrations, even though the standard does not apply to the groundwater beneath the burial ground. The standard for I-129 of 1 pCi/L is less than the observed maximum for well PDQ5. However, the presence of the stable I-127 in great excess of that expected from fission yields should minimize the radiation dose from I-129, even to a hypothetical user of the groundwater at some point in the distant future. This is because the critical organ for I-129 is the thyroid, and the concentration of iodine by the thyroid is independent of isotope. Therefore, the radiation dose from I-129 in the groundwater beneath the burial ground should be lower than if the stable I-127, in excess of the normal fission yields, was not present.

CONCLUSIONS AND RECOMMENDATIONS

The measurements of Tc-99 and I-129 in the plume wells represent the first attempts to measure non-gamma emitting radionuclides other than tritium in these plume wells. The presence of these two species indicate that they are mobile radionuclides which are migrating from the burial ground toward Four Mile Creek. The Tc-99 in the burial ground is present due to the disposal of mixed fission product waste. Since the observed concentrations are less than the EPA Interim Drinking Water Standard, and the impact of the presence of this radionuclide in the groundwater is expected to be minimal, no changes in waste handling specifically for Tc-99 are recommended.

Although dose projections from I-129 in the groundwater are expected to be minimal and do not justify changes in disposal practice, the I-129 waste is a compact source which is already segregated from other waste. Storing it in a segregated, retrievable form would leave open the option for disposal of this long-lived radionuclide in a future geologic repository. I recommend that steps to implement such storage of I-129 be considered.

QUALITY ASSURANCE

All of the data contained in this report is recorded in laboratory notebooks DPSTN-3397 and DPSTN-4154 in accordance with the SRL Procedures Manual and the DWPS QA Documents. The analyses were performed by ADD or by M. V. Kantelo of ETD. No

standards were submitted, but standards are routinely analyzed on all of the instruments used. In general, all of the analyses were run in duplicate. Sampling devices used for collection of the groundwaters were carefully rinsed between samplings and wells were sampled in the order of least to greatest potential for Tc-99 or I-129 contamination.

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TABLE 1. Tc-99 AND CHEMICAL ANALYSES OF GROUNDWATER

Samples collected September 22, 1982

Well ID		<u>PDQ5</u>	<u>BG-109</u>	<u>E-17</u>	<u>G-21</u>
Tc-99	pCi/L	1.47	<0.6	4.4	2.7
F	mg/L	0.09	0.03	1.74	<0.1
Cl	mg/L	2.2	2.8	7.1	34.2
Nitrate	mg/L	<0.5	9.5	88.2	<0.5
Sulfate	mg/L	0.3	72.0	133	6.3
K	mg/L	0.62	0.35	10.5	0.68
Mg	mg/L	1.17	0.27	2.2	10.3
Fe	mg/L	1.42	<0.03	0.21	156
Si	mg/L	3.2	3.6	11.1	1.7
Na	mg/L	7.9	5.1	35.7	20.2
P	mg/L	0.05	<0.03	0.1	0.4
Ca	mg/L	2.1	1.4	98	7.6
Ba	mg/L	0.3	0.02	0.02	0.08
Sr	mg/L	0.01	0.01	0.4	0.07
Mn	mg/L	3.0	0.03	<0.01	0.5
Dissolved					
Oxygen	mg/L	3.5	6.6	6.1	2.7
pH		5.9	5.3	7.3	5.7
eH		105	520	435	219
Conductivity		49	40	270	250
	umho/cm				

Samples for anions and Tc-99 filtered prior to analysis.

Samples for cations filtered and acidified prior to analysis.

All other measurements run on unfiltered samples.

TABLE 2. I-129 AND CHEMICAL ANALYSES OF GROUNDWATER

Samples collected August 15, 1983

Well ID		PDQ5	BG-109	BGC-2C	BGC-3C	I-13
I-129	pCi/L	12.0	0.01	0.033	0.92	0.006
I-127	ug/L	1190	<14	99	166	49
Tritium	uCi/L	61.3	0.04	3.2	63.2	0.05
F	mg/L	--	<0.1	<0.1	0.4	<0.1
Cl	mg/L	15.5	1.7	4.6	3.2	133
Nitrate	mg/L	<0.5	8.8	5.4	0.9	4.9
Sulfate	mg/L	<0.5	1.5	2.3	3.3	5.5
K	mg/L	0.68	0.19	5.2	2.3	73.1
Mg	mg/L	1.24	0.26	<0.01	<0.01	3.35
Fe	mg/L	6.07	<0.01	<0.01	<0.01	<0.01
Si	mg/L	3.6	2.9	1.8	0.58	2.2
Na	mg/L	10.4	3.6	4.9	8.8	33.7
P	mg/L	0.15	0.11	0.09	0.05	0.08
Ca	mg/L	2.3	1.0	22.1	393	16.9
Ba	mg/L	0.9	0.02	0.01	0.07	3.0
Sr	mg/L	0.01	0.01	0.15	0.48	0.51
Mn	mg/L	4.3	0.02	<0.01	<0.01	0.06
Dissolved						
Oxygen	mg/L	3.3	7.5	3.9	2.6	7.8
pH		5.8	8.8	10.6	11.9	6.2
eH		212	600	392	210	610
Conductivity		255	35	250	3820	600
umho/cm						

Samples for anions and Tc-99 filtered prior to analysis.
 Samples for cations filtered and acidified prior to analysis.
 All other measurements run on unfiltered samples.

TABLE 3. Tc-99 CONCENTRATIONS AS A FUNCTION OF TIME

Date of Sample	Tc-99 pCi/L	
	PDQ5	BG-109
April 82	22	0.021
September 82	1.5	<0.6

TABLE 4. IODINE CONCENTRATIONS AS A FUNCTION OF TIME

Date of Sample	I-129 pCi/L		I-127 ug/L	
	PDQ5	BG-109	PDQ5	BG-109
April 82	0.25	--	1.52	--
December 82	1.5	0.005	200	35
August 83	12.0	0.04	1190	<14

