An Alarming Change in the Status of Technetium-99 in the Vadose Zone and Aquifer at INL

Technetium-99, with half life 213,000 years, is a long lived carcinogenic radionuclide created by nuclear fission from nuclear reactors or nuclear weapons testing. Nuclear fuel debris buried underground at INL's Radioactive Waste Management Complex (RWMC) and waste from nuclear fuel reprocessing at INL's Idaho Nuclear Technology Center (INTEC) have released Tc-99 into the ground. Technetium-99 contamination from INTEC operations of Tc-99 has been detected in the aquifer since 1998. So, why have you read so little about this?

Despite the Cleanup News at RWMC, Vadose Zone Tc-99 Levels Are Increasing

Examine recent USGS reports for selected contaminants like USGS 2013-5214 for 2009 through 2011, or USGS 2015-5003 and you'll find various radionuclides included—but sweetly and oh so neatly, Tc-99 is completely absent from the reports.¹ You must search the CERCLA cleanup studies to find information about Tc-99 monitoring.

Technetium-99 has not been found in the aquifer below RWMC where chemical contaminants (prominently carbon tetrachloride, uranium, cesium-137, and others continue to be the largest aquifer contaminants. And success stories of RWMC cleanup are prominent, with successful retrieval of "targeted" waste continuing, even though shipments to WIPP have been interrupted due to the accidents last February 2014. Restart efforts are expected to take several years at WIPP, the New Mexico salt mine for underground disposal of defense-generated wastes.²

In 2006, here's what you might have read about Tc-99 at RWMC above the aquifer: ³

"Technetium-99 is consistently detected at depths of 27 m (88 ft) in two locations. . .at the western end of the [subsurface disposal area]. The concentration associated with Well D06 is increasing. Historically, Tc-99 has not been a priority analyte for vadose zone monitoring; therefore, data are sparse." (No concentration level is given.)

"Few radionuclides are detected in core samples. Most are detected only sporadically and have no associated temporal or spatial trends; however, some radionuclides are consistently detected in RWMC core samples. In order of detection frequency from highest to lowest, these radionuclides are Tc-99, Am-241, Pu-239/240, Sr-90, and Pu-238."

"Detections of Tc-99 in the I-series wells in 1999 were not corroborated by detections in the 2003 core sampling campaign. Some evidence supports the conclusion that Tc-99 is present,

¹ US Geologic Survey, An Update of Hydrologic Conditions and Distribution of Selected Constituents in Water, Eastern Snake River Plan Aquifer and Perched Groundwater Zones, Idaho National Laboratory, Idaho, Emphasis 2009-11, Report 2013-5214 (DOE/ID-22226), 2013 or Water-quality characteristics and trends for selected wells possibly influenced by wastewater disposal at the Idaho National Laboratory, Idaho, 1981–2012, Report 2015–5003 (DOE/ID-22233), 2015.

² Read more at wipp.energy.gov or <u>http://www.abqjournal.com/540901/news/flynn-wipp-leak-fines-have-national-implications.html</u>

³ J. Holdren et al., Idaho Cleanup Project, Department of Energy, *Remedial Investigation and Baseline Risk* Assessment for Operable Unit 7 13/14, DOE/ID-11241, May 2006. http://ar.inel.gov

while some evidence is to the contrary. However, lysimeter data imply Tc-99 transport may be occurring."

So, they are saying, gee, we just don't see any trends and the detections are sporadic. The metamessage seems to be we're not really sure that Tc-99 transport is occurring so there doesn't appear to be anything to worry about.

But along comes the 5 year monitoring report for reporting in 2010 to 2014. ⁴ This report documents detections of concentrations of 15,700 pico curies per liter (pCi/L), well above the derived maximum contaminant level (MCL) of 900 pCi/L. This same location has also yielded concentrations of nitrate, tritium, and total uranium that exceeded respective MCLs. "Tc-99 continues a generally upward trend." ⁵

Technetium-99 is highly soluble and mobile in soil. Contamination in the vadose zone above the aquifer will continue downward to the aquifer. RWMC has an estimated 42.3 curies of Tc-99.⁶

But cleanup is happening at RWMC, so aren't the levels of Tc-99 going to be reduced? No they may argue that once an engineered cap is installed the leaching will slow. But, the Tc-99 is mainly from INL operations and not from Rocky Flats. Only the chemical waste and the transuranic waste from Rocky Flats is targeted for retrieval and shipment to the WIPP.

Over 16 curies of technetium-99 to be disposed of at the proposed Replacement RH-LLW facility to replace RWMC roughly matches estimates of what INTEC released to the aquifer from injection well practices.⁷ And, the RWMC technetium-99 waste is supposedly stored in metal canisters.⁸

Technetium-99 at INTEC

As part of ongoing CERCLA activities at INTEC, in May 2003 Tc-99 was found **in the aquifer** at concentrations between 2,000 and 3,000 pCi/L. This was the first time Tc-99 concentrations exceeded the derived maximum contaminant level of 900 pCi/L. ⁹ Elevated Tc-99 in groundwater at monitor well ICPP-MON-A-230 is thought to be from historical liquid waste releases (leakage) at the INTEC tank farm. As the contamination disperses and spreads in the aquifer, the detected levels are expected to decrease as the contaminant disperses in the aquifer.

⁴ Department of Energy, "Operable Unit 7-13/14 Five-Year Monitoring Report for Fiscal Years 2010-2014, DOE/ID-11507, August 2014. p. 31-32, at Zone 1, R2004. <u>https://ar.inl.gov/images/pdf/201409/2014091800949BRU.pdf</u>

⁵ DOE/ID-11507, August 2014. p. 31-32, at Zone 1, R2004. https://ar.inl.gov/images/pdf/201409/2014091800949BRU.pdf

⁶ ibid. RI/BRA DOE/ID-11241Table 4-2.

⁷ Environmental Defense Institute report: Unwarranted Confidence in DOE's Low Level Waste Facility's Performance Assessment.

⁸ ibid. RI/BRA DOE/ID-11241. p 7-44.

⁹ Idaho Completion Project, Bechtel BWXT Idaho LLC, "Evaluation of Tc-99 in Groundwater at INTEC: Summary of Phase 1 Results," ICP/EXT-04-00244, September 2004. p. 2-2.

The former INTEC injection well, used from the 1953 until 1986 and plugged in 1989, likely constituted an earlier source of Tc-99 to the aquifer, but the resulting groundwater Tc-99 concentrations did not exceed the MCL. The injection well flushed high volumes of water, creating a more dilute Tc-99 plume that extends south and down gradient of INTEC.

The report of detections of Tc-99 at INTEC in 2003 cites a USGS report of Tc-99 detection in the aquifer. ¹⁰ This came as quite a surprise to me, as I had been searching for USGS reports for aquifer contamination using the UGSG bibliography for INL but found none. This 1998 report for Tc-99 and other contaminants at INL was only printed in a journal. It is not in the INL bibliography, nor is any report of Tc-99 included in the bibliography nor is it included as a "selected" radionuclide in reports I have reviewed.

In this 1998 journal article by T. M Beasley,¹¹ Tc-99 was found in groundwater over a large area extending south from the INTEC to the area of the Big Southern Butte. The plume was similar in size and shape to the known tritium plume south of INTEC. An estimated 15 Ci of Tc-99 was disposed of in the aquifer attributed to the former INTEC injection well which operated from 1953 to 1986. The INL and Department of Energy had made no documented measurements (or estimates) of the fuel reprocessing contaminant, Tc-99, released by INTEC.

Technetium-99 Health Summary

Technetium-99 human health risk is primarily through groundwater ingestion and irrigation of crops with contaminated groundwater. The Environmental Protection Agency (EPA) has established a Maximum Contaminant Level (MCL) of 4 millirem per year for beta particle and photon radioactivity from man-made radionuclides in drinking water. The average concentration of technetium-99 to yield 4 millirem per year is a derived value of 900 pCi/l. It is crucial to remember the additive effects of other radionuclides present in addition to technetium-99. The annual dose from all the beta emitting radionuclides combined should not exceed 4 millirem/yr.¹²

Once in the human body, Tc-99 concentrates in the thyroid gland and the gastrointestinal tract. The body, however, constantly excretes Tc-99 once it is ingested. As with any other radioactive material, there is an increased chance that cancer or other adverse health effects can result from exposure to radiation.

At radioactively contaminated sites with Tc-99 contamination, the primary routes of exposure to an individual are from the potential use of contaminated drinking water and ingestion of contaminated plants. The contamination at INTEC has been generously spread into the aquifer and spread downstream. The leaching of Tc-99 at RWMC has only just begun and will continue for thousands of years. The leaching of Tc-99 from the new Replacement Remote-Handled Low-

¹⁰ ibid. ICP/EXT-04-00244. p. 2-2, 5-1.

¹¹ T. M. Beasley, P. R. Dixon, and L. J. Mann, "⁹⁹Tc, ²³⁶U, and ²³⁷Np in the Snake River Plain Aquifer at the Idaho National Engineering and Environmental Laboratory," *Environmental Science & Technology*, 32:3875-3881, 1998.

¹² EPA Facts about Technetium. <u>http://www.epa.gov/superfund/health/contaminants/radiation/pdfs/technetium.pdf</u>

Level Waste (RH-LLW) at INL will add to the legacy of dumping radioactive waste into the Snake River aquifer. See Table 1 for a summary of Tc-99 in or headed for the aquifer.

Table 1. Estimated inventory of technetium-99 in disposed waste, vadose, or aquifer at INL.

				Percent of
			Highest	900 pCi/L
			measured	Maximum
	Technetium-		concentration,	Contaminant
Location	99, curies	INL Facility ^a	pCi/L	Level ^b
RWMC buried	42.3 curies	MFC, INTEC, ATRC,	15,700 pCi/L	1744
waste		NRF	(2010-2014)	1/44
INTEC disposal	15 curies	Government-owned	518 pCi/L	57
well		nuclear fuel reprocessing	(1994)	57
INTEC tank farm	less than 4	Government-owned	3000 pCi/L	333
leakage	curies	nuclear fuel reprocessing	(2003)	333

Information source: DOE/ID-11241, ICP/EXT-04-00244 Notes:

a. Facilities: Advanced Test Reactor Complex (ATRC); Idaho Nuclear Technology Center (INTEC); Materials and Fuels Complex (MFC), formerly ANL-W; Naval Reactors Facility (NRF).

b. The maximum contaminant level in pico curies per liter (pCi/L) for technetium-99 is derived from the EPA drinking water regulation, 40 CFR 141.66, of 4-mrem/yr for a beta-emitting radionuclide. The presence of more than one radionuclide reduces the MCL for each so the total dose does not exceed 4 mrem/yr to the critical organ. Beta emitters cesium-137, iodine-129 and technetium are summed for the 4 mrem/yr limit; however, strontium-90 and tritium have their own MCL.