UNM METALS Research Center

Metal Exposure Toxicity Assessment on Tribal Lands in the Southwest

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UNM METALS Monograph 1*

Uranium in Soil, Mine Waste and Spring Water near Abandoned Uranium Mines, Tachee/Blue Gap and Black Mesa Chapters, Navajo Nation, Arizona

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Abstract

Previous investigations identified elevated radiation levels at abandoned uranium mines (AUMs) operated in the 1950s-1960s in the Tachee Wash and Waterfall Canyon areas of Tachee/Blue Gap and Black Mesa Chapters of the Navajo Nation in northeastern Arizona. The occurrence of trace metals in wastes at these sites has not been previously characterized. Community concerns about possible ongoing release of hazardous substances from these sites, and possible contamination of water in a spring used by local families for drinking water, prompted a preliminary field study by the UNM METALS Center. Field gamma radiation spot surveys on the largest waste site and geospatial analysis of the proximity of residences to the mine wastes indicate that the Claim 28 Mine site satisfies the Navajo Nation's screening criteria as a highpriority AUM. ICP-MS analysis of a water sample from a local spring confirmed the presence of uranium in concentrations exceeding the federal and tribal drinking water standard. XPS and XRF analyses of soil and waste samples collected from the largest AUM in the area revealed high levels of uranium, vanadium, arsenic, iron and aluminum, compared with non-impacted local soils. SEM analyses of the wastes found submicron-size U particles, which pose a potential inhalation risk. Further studies are needed to fully characterize the physical and chemical properties of the mine wastes, determine release patterns through runoff, and better understand hydrogeologic relationships between waste sites and natural springs.

*ACKNOWLEDGMENT: We acknowledge Navajo EPA's authorization to collect samples from the Claim 28 Mine complex and dedicate this first report of the UNM METALS Research Center to the memory of **Navajo EPA health physicist Eugene Esplain**, who worked tirelessly for a quarter century helping Navajo communities remediate abandoned uranium mines.

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Background

The region is characterized physiographically by pinon-juniper forests on the mesa tops and deep incised valleys that carry runoff from Black Mesa to points south and west. Uranium deposits were explored and mined from more than a dozen sites in the northern portion of the Tachee/Blue Gap Chapter and the southeastern portion of Black Mesa Chapter in the mid-1950s to the late-1960s (Chenoweth, 1990). The region is known as the Black Mesa East AUM District (USEPA, 2000a). Thirteen different mines produced 16,779 tons of uranium ore and 4,391 tons of vanadium ore during this period (Chenoweth, 1990).

The largest of the Tachee/Blue Gap uranium mines, called "Claim 28" (also identified as Mine #78), located about 5.75 miles north of the Tachee/Blue Gap Chapter House, produced 4,181 tons of uranium ore and about 2,600 tons of vanadium ore (Chenoweth 1990). An estimated 4,995 cubic yards of waste material are buried or exposed at the site (Weston Solutions, 2011) (**Figure 1**). Surface reclamation conducted by the Navajo Nation Abandoned Mine Lands Department in 1991-1992 consisted of burying mine wastes in open pits and trenches on the mesa-top flat land, applying a clean-soil cover and re-contouring the site to mitigate runoff (OSM, 1992). Waste materials are present on a 150-foot slope below the mine, and in a fenced area (identified as Mine #79) at the base of the slope about 1,500 feet north of Route 29 (**Figure 2**). Several residences are located southeast and south of these waste sites. The most recent radiological survey of the Claim 28 "complex" (Mines #78 and #79) by Weston Solutions, Inc., for USEPA Region 9, found maximum gamma radiation rates on the waste slope ranging from 9.1 to 10.3 times higher than the average and lowest recorded background levels, respectively.

Residents and leaders of Tachee/Blue Gap Chapter have expressed concerns about potential uranium contamination from long-abandoned uranium mines for many years (Tachee/Blue Gap Chapter, 1988; Morgan, 2009; Pasternak, 2010). Community members who contacted Southwest Research and Information Center (SRIC), a partner organization in the UNM METALS Center, identified concerns ranging from dust blowing off the slope wastes and into nearby homes (**Figure 2**), runoff carrying potentially contaminated sediments onto grazing lands in the valley, and mine wastes or natural ore deposits possibly contaminating local springs and seeps. They requested assistance in further characterizing mine wastes and sampling Waterfall Spring to ascertain current waste chemistry and water quality conditions.

Site Access and Permissions

Local residents who use Waterfall Spring (location shown in **Figure 2**) gave permission to staff of SRIC (Chris Shuey and Paul Robinson) and the University of New Mexico (UNM) scientists (Adrian Brearley and Steve Cabaniss) to collect a water sample from Waterfall Spring on November 11, 2013. Navajo Nation Environmental Protection Agency (NNEPA) staff granted permission for Mr. Shuey and Mr. Robinson to collect soil and waste samples from the Claim 28 complex on January 29, 2014. In both field trips, UNM METALS Center personnel were occupied by resident Sadie Bill, a member of the Tachee Uranium Concerns Committee (TUCC) and resident of Tachee/Blue Gap Chapter.

Field Methods

Water Sampling. The site of Waterfall Spring (36.29468N, -109.83154W) was accessed by foot from the mesa top into a incised canyon located about 5 miles north of Indian Route 29. A sample was collected in a plastic drinking bottle from a pool at the base of the cliff by Mr. Shuey, witnessed by Mr. Robinson and Ms. Bill. Water from the pool was used to rinse out the sample container. A sample of approximately 300 ml was collected by dipping the bottle into the pool, taking care not to disturb sediments at the bottom of the pool. The container was marked with the sample time and date and handed over to Dr. Brearley. The sample was driven to the UNM Earth and Planetary Sciences (E&PS) laboratory for chemical analysis.

Soil and Mine Waste Sampling. Soil and soil-waste mixture samples were collected by Mr. Shuey, Mr. Robinson and Ms. Bill from three locations on Mine #79 and Mine #78, which collectively are referred to as the "Claim 28 Mine Complex." Gamma radiation rates were recorded for each sample location using a recently calibrated Ludlum-19 MicroR Meter, owned by SRIC. The sample procedures, sample identification numbers, latitude-longitude coordinates, and gamma radiation rates recorded at each sample location are described in detail in a February 4, 2014 (Revised March 10), memorandum from Mr. Shuey and Mr. Robinson to the UNM METALS Center Working Group. The memorandum, which contains photos of the sampling locations, is attached to this reported as Appendix A. A soil sample was collected near the intersection of Indian Route 29 and Waterfall Road, a location believed to be unaffected by uranium mine wastes. This sample is referred to as "non-impacted" soil and is used as a background or reference sample for comparison with soil-waste samples collected from the abandoned mine waste sites. **Table 1**, compiled from the SRIC February 4/March 10 memorandum, summarizes all field data for the soil-waste specimens.

Laboratory Methods

Water Sample. The Waterfall Spring sample was analyzed by ICP-MS (Inductively Coupled Plasma Mass Spectrometry) consistent with USEPA standard methods at the UNM-E&PS laboratory under the direction of chemist Abdul-Medhi Ali, Ph.D. Twenty-seven analytes, including trace metals, major ions and TDS (total dissolved solids), were analyzed. The results are shown in **Table 2**.

Soil and Soil-Waste Samples. Metal content of soil and waste samples were determined by X-ray photoelectron spectroscopy (XPS) and X-ray fluorescence (XRF) by Dr. Abdul-Mehdi Ali, Dr. Kateryna Artyushkova, Research Associate Professor in the UNM Chemical Engineering, Sumant Avasarala, Ph.D. student in UNM Civil Engineering Department, and Dr. Jose Cerrato, Assistant Professor of Environmental Engineering in the UNM Civil Engineering Department. Results of these analyses are shown in **Tables 3, 4 and 5.** A scanning electron microscope (SEM) was used by Dr. Brearley to examine the distribution, sizes and composition of metal-rich particles within the soil and waste samples.

Results and Discussion

Distances from Residences to Mine Wastes. Using the distance function in Google Earth, we calculated a distance of 0.25 miles from two occupied residences to the closest point on the Claim 28 Mine waste dump (**Figure 1**). Similarly, another occupied residence is located within 0.25 miles of the fenced waste repository site (Mine #79). Additionally, storm water runoff from the Claim 28 Mine flows toward these homes located down gradient of the mine wastes in well-defined channels at the base of the mesa slope.

Gamma radiation rates. As shown in **Table 1**, gamma radiation rates ranged from 12-13 microRoentgens per hour (μ R/hr) at the background site (Sample #3) to 290-320 μ R/hr at the Sample #2A site, which was located in a deep gulley in the mine wastes covering the steep slope of the Mine #78 site (**Figure 2**). Gamma rates ranged from 120-230 μ R/hr at the Sample #1 (Mine #79) site, which NNAML staff indicated is a repository for wastes removed from a haul road that provided access to the mesa top during reclamation of the Claim 28 Mine in 1991-1992. The gamma rate at the site of Sample #2B, on top of the mesa in the area where reclamation was performed, ranged from 160-180 uR/hr. Using the lower end of the gamma rates measured at the non-impacted background location (i.e., 12 uR/hr) as a reference point for comparison, all gamma rates measured at the soil-waste sample sites were at least 10 times higher than the background rate, and ranged up to nearly 27 times higher (maximum value at Sample Site #2A).

The gamma rates recorded at the sample locations were spot surveys that were not intended to represent a systematic assessment of gamma rates across the mine sites. The Weston Solutions Site Screen for USEPA Region 9 in August 2011 was a more extensive survey resulting in nearly 2,800 data points at Mine #78 (i.e., Claim 28) and about 600 data points at Mine #79 (waste repository). The range of gamma counts in the Weston survey was 12,843 cpm (counts per minute) to 132,382 cpm at Mine #78, or more than a tenfold spread. Weston calculated an average background of 14,688 cpm at Mine #78, which is about a nine-

fold difference from the maximum site reading. At the fenced repository site (Mine #79), the highest gamma radiation measurement (36,695 cpm) was about 2.7 times greater than the average background for the site (i.e., 13,355 cpm).

Notwithstanding the differences in the ranges of minimum to maximum gamma rates between the UNM METALS spot surveys and the USEPA/Weston site screening survey, both assessments confirmed the presence of elevated radiation levels on mine wastes compared with local background rates. In many cases, maximum gamma rates were equal to or greater than 10 times than the lowest background rate.

Water Samples. Results of the ICP-MS tests run by Dr. Ali compared favorably with results of the only previously known sample from Waterfall Spring, collected by U.S. Army Corps of Engineers (USACE) personnel in November 1998 (USEPA, 2000a). In particular, the U concentration in the November 2013 sample of 0.0717 milligrams per liter (mg/l), or 71.7 micrograms per liter (μ g/l), is quite close to the result reported by USACE from the sample 15 years before (**Table 2**). The USACE result of 61.7 picoCuries per liter total U activity can be converted to mass by multiplying by 1.5 (or dividing by 0.67 pCi/ μ g), which is a rule of thumb used by USEPA to convert concentrations in radioactivity to concentrations in mass when the exact ratios among the uranium isotopes are not known (USEPA, 2000b; p. 76712). The November 1998 result converts to 92.6 μ g/l, or parts per billion. Either way, both concentrations represent a 2.5- to 3-fold exceedance of the current USEPA drinking water standard for uranium of 30 μ g/l (**Table 2**).

The Total Dissolved Solids (TDS) concentration of 1,590 mg/l reported in the UNM analysis indicates water from the spring is slightly brackish. No other contaminant exceeded its primary (MCL) or secondary (SDWS) drinking water standard in the UNM analysis. However, the USACE sample also had concentrations of gross alpha radioactivity (52 pCi/l) and total radium (6.31 pCi/l) that exceeded their respective MCLs (15 pCi/l and 5 pCi/l). Radium is a decay product of uranium and a known human carcinogen associated with bone and blood cancers (ACS, 2014). This is cause for concern because an extended family that lives at the mouth of Waterfall Canyon has been using Waterfall Spring for its drinking water supply for most of the time since the USACE test in 1998. While the family was recently connected to the Navajo Tribal Utility Authority (NTUA) public water supply system, they still depend on the spring for watering their livestock.

Soil and Waste Samples. Results of the XPS analyses are shown in **Tables 3** and **4**. It should be noted that XPS spectra represent the signal obtained from the "near surface region" (top 5 nm) of the solid sample analyzed. The mine wastes had high concentrations of uranium (0.01% or 100 parts per million [ppm]), vanadium (1.03%, or 1,030 ppm) and arsenic (0.49%, or 490 ppm), compared with concentrations in non-impacted soils that were below detection limits of the assays for each of these elements. The arsenic concentration is more than 320 times higher than the crustal average concentration of 1.5 ppm (IGRAC, 2014).

The spectra scan plots in **Table 4** show identifiable peaks for U(VI) and As(0), compared with the plots for non-impacted soils, which show little evidence of U and As contamination. U(VI) is more mobile in the environment than its reduced counterpart, U(IV). The most commonly occurring oxidation states of As are As(III) and As(V). On the other hand, As(0) is rarely observed in nature and is the least mobile naturally occurring oxidation state of As. Future efforts at UNM will focus on performing laboratory experiments to better understand the mobility of U and As present in sediment samples when exposed to water solutions representative of field conditions.

Results of the XRF tests (**Table 5**) showed substantial enrichment of U, V and Al in the mine wastes compared with non-impacted soils. The concentrations are expressed in micrograms per gram (dry weight), which is equivalent to 1 ppm. The U concentration of 2,248 ppm is more than 1,700 times the crustal average U concentration of 1.3 ppm (WNA, 2014). The V concentration of 15,814 ppm is 158 times the natural concentration of about 100 ppm (ATSDR, 2010). Aluminum was also elevated relative to the Al concentration in the non-impacted soils.

Dr. Brearley has reported (personal communication, March 15, 2014) scanning electron microscope observations from mine waste samples from both the fenced waste repository site and the waste dump on the side of the mesa. Both of these samples show the presence of particles of uranyl vanadates (compounds of uranium and vanadium) with grain sizes ranging from a few microns down to at least 300 nm dispersed throughout the two samples studied. While detailed size distribution data are not yet available, a significant proportion of these particles is submicron in size and can be breathed into and retained in the deepest parts of the lung. No uranyl vanadates were found in the sample of uncontaminated soil studied by scanning electron microscopy. No association of Fe was observed with U from the microscopy analyses. Future microscopy analyses will focus on better understanding U- and As-bearing phases. These tests are preliminary and additional results will be reported at a later date.

Conclusions

Recent field investigations by the UNM METALS Research Center in Tachee/Blue Gap Chapter found that these abandoned uranium mines associated with the Claim 28 complex — Mines #78 and #79 — meet the Navajo Nation's criteria as priority sites. The closest points on the mines are 0.25 miles from the nearest occupied residences, and gamma radiation rates equal to or greater than 10 times local background rates are prevalent on these mine sites. Furthermore, XPS and XRF analyses at UNM show the presence of hazardous substances — U, V and As — in mine wastes in concentrations far exceeding both their respective crustal averages and local background in non-impacted soils. The mine wastes also appear to be enriched in Fe and Al compared with non-impacted soils. Preliminary SEM analyses indicate the wastes contain uranium-vanadium compounds on fine-grained particles that are vulnerable to re-suspension in windy conditions, posing a potential inhalation risk. A water sample from Waterfall Spring had a uranium concentration 2.3 times higher than the federal and tribal drinking water standard, confirming the result of a 1998 test by the U.S. Army Corps of Engineers. Additional tests are needed to determine if radium is still present in Waterfall Spring at a level exceeding its drinking water standard. Local residents have been advised not to drink from Waterfall Spring and to avoid crossing or playing on the Claim 28 mine sites and other abandoned mines in the area. Further studies are needed to fully characterize particle-size distributions and chemistry of the mine wastes, investigate hydrogeological relationships between the orebearing Toreva Sandstone and discharge points for springs and seeps like Waterfall Spring, and assess the extent of release of hazardous substances in runoff emanating from the Claim 28 slope wastes. This report will be shared with community members, chapter officials and Navajo Nation regulatory agencies.

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Figures and Tables





Sample #	DDLat	DDLong	Site Description	Gamma rate (µR/hr) at sample location	Depth of sample(s)	Sample weight			
1	36.24352	-109.89422	Mine #79, fenced area (~11,600 ft ²)	120 at surface contact 230 at bottom of sample hole	~6-7 inches (15.2- 17.8 cm)	1.5 lbs (0.68 kg)			
2 – Compos	site of 2A and	d 2B							
2A	36.24488	-109.89213	Mine #78; mine- waste slope under dirt cover	290-320	sample taken from exposed rock ~6 ft below soil cover on waste slope	1.0 lbs (0.454 kg)			
2B	36.24540	-109.89229	Mine #78; mine waste on west side of mesa-top mine site	160-180	~2 inches (5.8 cm)				
3 non- impacted	36.23699	-109.88909	Undisturbed soils near intersection of NR 29 and Waterfall Road	12-13	~5 inches (12.7 cm)	15 oz (0.43 kg)			

 Table 2. Historic and current water quality data for Waterfall Spring, Black Mesa Chapter, Navajo

 Nation (Arizona). Location Coordinates: 36.29468N, -109.83154W. (Blank cells indicate no analysis.)

Analyte	Units	MCL	SDWS	11/4/1998*	11/11/2013**
Alpha activity, gross	pC/I	15		52	
Aluminum	mg/l		0.05-0.2	0.16	0.05
Antimony	mg/l	0.006		ND	
Arsenic	mg/l	0.01		ND	ND
Barium	mg/l	2		0.045	0.018
Beryllium	mg/l	0.004		0.0006	ND
Beta activity	pCi/l	50***		25.1	
Boron	mg/l			ND	
Bromide	mg/l				0.401
Cadmium	mg/l	0.005		ND	ND
Calcium	mg/l				288
Chloride	mg/l		250		56.17
Cobalt	mg/l			ND	
Copper	mg/l	1.3		ND	ND
Fluoride	mg/l	4.0	2.0		0.73
Iron	mg/l		0.3	ND	ND
Lead	mg/l	0.015		ND	ND
Lead-210	pCi/l			2.26	
Lithium	mg/l			ND	
Magnesium	mg/l				86.6
Manganese	mg/l		0.05	0.0027	ND
Mercury	mg/l	0.002		ND	
Molybdenum	mg/l			ND	
Nickel	mg/l			0.0023	ND
Nitrate	mg/l	10			6.34
Nitrite	mg/l	1			0.207
Phosphate	mg/l			no data	
Potassium	mg/l				15.1
Radium-226	pCi/l	5		1.63	
Radium-228	pCi/l	5		4.68	
Radium-total (sum of isotopes)	pCi/l	5		6.31	
Selenium	mg/l	0.05		0.023	ND
Silicon	mg/l				6.34
Silver	mg/l		0.1	ND	
Sodium	mg/l				33.8
Strontium	mg/l				1.86
Sulfate	mg/l		250		153.1
Thorium	mg/l			ND	
Thorium-228	pCi/l			0.133	
Thorium-230	pCi/l			0.007	
Total Dissolved Solids	mg/l		500		1,590
Uranium (mass)	mg/l	0.03		0.0926****	0.0717
Uranium-234	pCi/l	20		31.9	
Uranium-235	pCi/l	20		0.937	
Uranium-238	pCi/l	20		28.9	
Uranium (total isotopes)	pCi/l	20		61.737	
Vanadium	mg/l			0.0194	0.015
Zinc	mg/l		5	0.031	ND

Abbreviations and Notes to Table 2:

MCL = Maximum Contaminant Level; SDWS = Secondary Drinking Water Standard; mg/l = milligrams per liter of water; pCi/l = picoCuries per liter of water. **Red highlighted data** indicate concentrations exceeding MCLs (for gross alpha radioactivity, total radium, and uranium mass and uranium total isotopic); **black boldface** type indicates concentrations exceeding SDWSs.

*Sample collected 11/4/98 by U.S. Army Corps of Engineers personnel; results reported in USEPA, "Abandoned Uranium Mines Project, Arizona, New Mexico, Utah -- Navajo Lands, 1994-2000, Project Atlas" (metals, p. 4-10; radionuclides, p. 4-12).

**Sample collected 11/11/13 by Southwest Research and Information Center; analyses conducted at UNM Earth and Planetary Science Department laboratory, reported 12/5/13, 12/6/13, 1/28/14 and 1/29/14.

***At the time the USEPA Project Atlas was published, 50 pCi/l beta activity was a first-order screen, leading to more elaborated assays; current USEPA MCL is a concentration that gives a dose of 4 millirems per year.

****The total uranium activity in the 1998 sample of 61.737 pCi/l is converted to an equivalent concentration in mass, expressed as milligrams per liter, using EPA's standard conversion factor of 1.5.

	% Content by weight									
	0 1s	C 1s	Al 2p	Si 2p	K 2s	Fe 2p	Mg 2p	U 4f	V 2p	As 3d
Undisturbed Soil	46.6	26.8	6.1	12.5	1.2	0.8	4.0	BDL [*]	BDL [*]	BDL [*]
Mine waste collected under dirt cover	50.8	24.0	8.3	12.1	1.1	1.4	0.2	0.01	1.03	0.49

Table 3. Results for XPS survey scan of selected atomic orbitals obtained for field samples obtained from Tachee Claim 28 site.

* Detection limit of the instrument is 0.01% by weight. BDL = Below Detection Limit.

Table 4. XPS narrow scan spectra of U 4f, As 3d, and Fe 3p obtained photo-peaks for undisturbed soil and mine waste collected under dirt cover from Tachee Claim 28 site.



Table 5. Content for selected elements (ug g⁻¹) from X-ray fluorescence for field samples obtained from Tachee Claim 28 site.

	Elemental Content, ug g ⁻¹								
	Si	S	AI	Fe	Mg	U	V	Ca	
Undisturbed Soil	241,950	1,339	52,129	26,739	3,068	BDL [*]	BDL [*]	16,441	
Mine waste collected under dirt cover	235,563	223	69,533	15,259	181	2,248	15,814	855	

* Detection limit of the instrument is 60 ug g^{-1} for U and 205 ug g^{-1} for V. BDL = Below Detection Limit.