ASSESSMENT OF ENVIRONMENTAL IMPACTS NEAR ABANDONED URANIUM MINES WITHIN THE CAVE HILLS AND SLIM BUTTES COMPLEXES, CUSTER NATIONAL FOREST, SOUTH DAKOTA[1]

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Abstract: Prospecting and mining of uraniferous lignite in the Tertiary Fort Union formation occurred from 1954 through 1967 in northwestern South Dakota. Activity was centered on US Forest Service land and abandoned mine sites received limited reclamation. Subsequent erosion and transport of mine waste has resulted in environmental impacts to soil and water resources down gradient of the mine sites. Through US-EPA Region 8 funding, a Joint Venture Agreement between the USDA-Forest Service Northern Region and the South Dakota School of Mines and Technology (SDSM&T) has been established to evaluate environmental impacts from uranium mining to soil, water, and air resources occurring on private lands surrounding the Cave Hills and Slim Buttes complexes within Custer National Forest. Results from this impact study indicate historical mining activities have caused degradation of regional ecological and environmental resources through the transport and deposition of sediments and spoils containing elevated concentrations of arsenic and uranium. Within the watershed downgradient of the North Cave Hills, surface water concentrations of arsenic and uranium exceeded established background concentrations within 27 km of stream length below the abandoned mines. Sediment results suggest secondary arsenic and uranium mineral phases were typically limited to the upper depths of drainage sediments. Results show that 14 watersheds were potentially impacted by sediment transport from previous mining activity. The most impacted area was the Upper Pete's Creek drainage below Bluff B where two U samples were 3× and 4× established background. Groundwater results indicate that metals and radionuclides were natural components of the groundwater systems. Results of the surface dust study indicate the general ubiquity of target analytes in the soils around the North Cave Hills. All metals concentrations in the surface dust were decreasing or below background levels within 15 km from the mine sites.

Additional Key Words: uranium mining impacts, wind dust transport, groundwater, surface water, sediments

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Introduction

Uranium exploration in northwestern South Dakota began in 1954 when the Atomic Energy Commission planned to fly radiologic airborne surveys over the Slim Buttes. Weather conditions precluded flying the Slim Buttes and instead, the survey was flown over the North Cave Hills (NCH) (Curtiss, 1955). As a result, the first claims were staked in the NCH on August 15, 1954 and mining ensued that year. Mine sites were located primarily within an approximately two mile wide north-west trending strip crossing the central NCH, referred to as the hot zone (Fig. 1). Mining was permitted under the General Mining Laws and Public Law 357, which required no form of restoration. Most mines and mining prospects were located on United States Forest Service (USFS) administered land, but at least two actively mined sites and several prospects and exploration cuts and digs were located on private land surrounding the NCH. Most of the uranium mines were abandoned coal strip mines located on relatively flat areas along the top of the numerous buttes that characterize the area. Mining consisted of the removing overburden (up to 80 feet) to reach the ore zone which consisted of uranium-bearing lignite beds. Mining activity increased through the next decade but ceased altogether in 1964.

During mining, most of the overburden and mine spoils were pushed over the edges of the buttes (Fig. 2) where subsequent erosion has spread materials over almost 1,000 acres in the NCH alone. Since mining ceased, additional deposition has occurred down-slope onto private land by water and wind transport. The bluffs and slopes immediately below mine sites often were covered by spoils forming highly over-steepened talus slopes, several of which have failed or are deeply incised. Currently, spoils are mostly devoid of vegetation and their composition has promoted water channeling, gullying, and tunneling. In addition to mined sites, numerous prospecting pits or contour benches were excavated on both USFS and private land and have been mapped and documented (Pioneer, 2005). Characterization of the abandoned mine sites on USFS land has occurred (Pioneer, 2005; Portage, 2005) but no work had characterized off-site impacts on surrounding private land. A Joint Venture agreement between the United States Department of Agriculture-Forest Service and the South Dakota School of Mines and Technology (SDSM&T), including a subcontract with Oglala Lakota College, was established in 2006 to address potential off-site impacts. Funding for this on-going study has been provided through US-EPA Region 8.

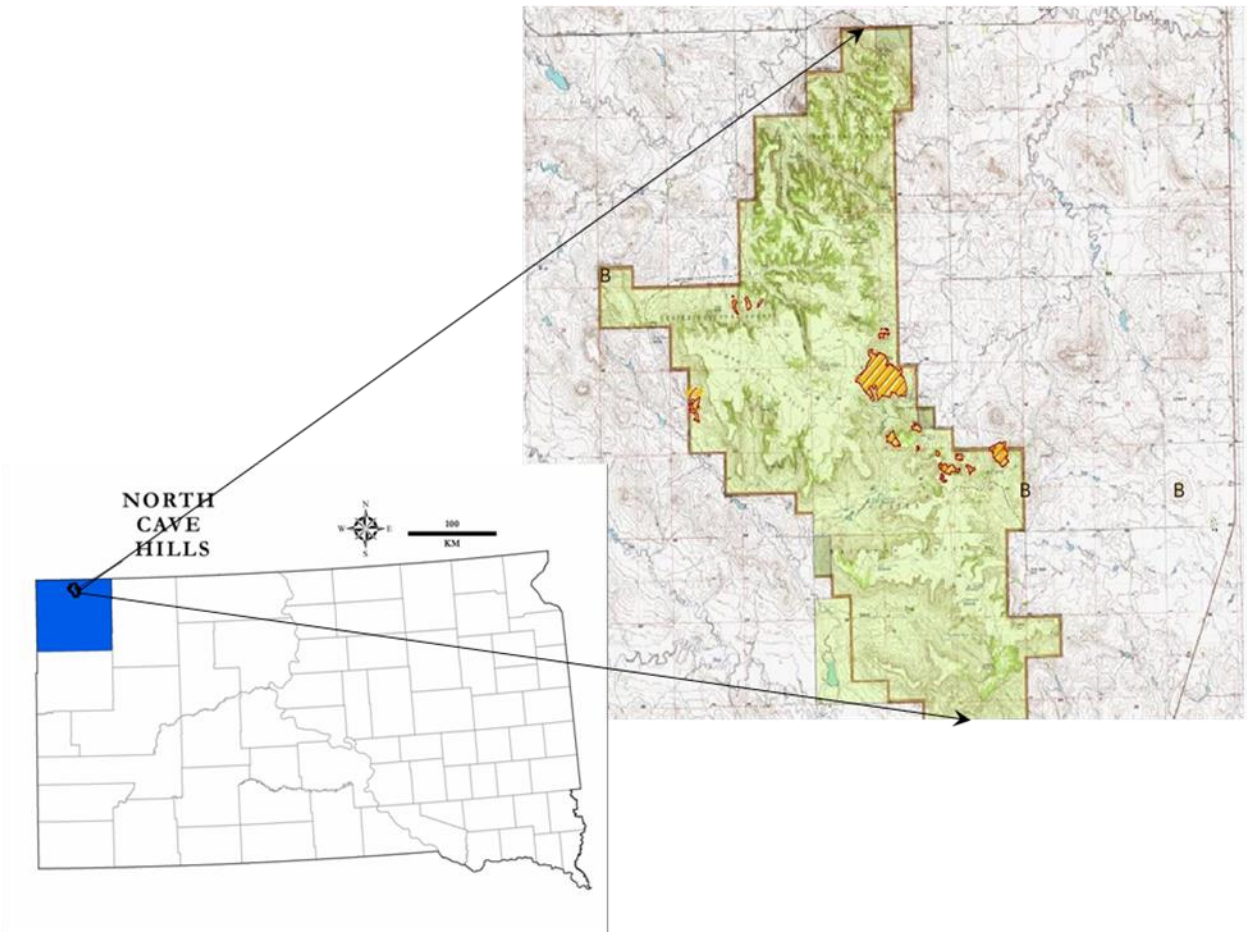

Figure 1: Site location map for the North Cave Hills complex of Custer National Forest, Harding County, South Dakota.

Figure 2: Current overburden and mining subsidence at Bluff B of the North Cave Hills complex

Objectives of the NCH project were to determine whether heavy metal and radionuclide environmental contaminants have been transported from historical mine sites located on USFS land onto surrounding private land. Mechanisms of environmental transport investigated included the following:

- erosion of spoil sediments through surface runoff into adjacent drainages;
- erosion of spoil sediments through slope failures below mined bluffs;
- dissolution of hazardous metals within runoff water that form tributaries to streams;
- erosion/deposition of small particle contaminants by wind transport; and
- infiltration of hazardous metals and radionuclides into local and regional ground water aquifers.

Methods

Target analytes for metals and radionuclides and analytical methods and procedures for determination of analyte concentration were similar to those used in previous investigations (Pioneer, 2005; Portage, 2005). Metals of interest included: As, Se, Cu, Mo, Pb, Th, U, and V. Water samples were further analyzed for gross alpha radiation, Ra^{226}, and U^{235}.

A watershed approach was developed to discern potentially impacted surface waters. It was assumed that all runoff water and eroded sediments (except for wind erosion) would ultimately end up in adjacent drainages and subsequently migrate downstream through established drainage networks. Surface-water sampling events were divided into two separate phases. Initial Phase I sampling evaluated target analyte concentrations of all potentially impacted drainages at the USFS/private land boundary. Phase I data also were used to establish background concentrations based on sampling locations assumed to be unaffected by mining activities. These data were subsequently used to evaluate which drainages were most heavily impacted. After evaluation of the Phase I results, drainages with the highest environmental concern were selected for Phase II sampling which continued downstream to a point where target analyte concentrations were comparable to established background levels. Phase I and II sampling located are shown in Figure 3 and 4, respectively.

Groundwater quality was evaluated by selecting 34 wells that were evenly distributed around the NCH within eight km of the USFS boundary (Figure 5). Approximately half of the wells were domestic supply wells and the others were stock wells. Well depths ranged between 10 and

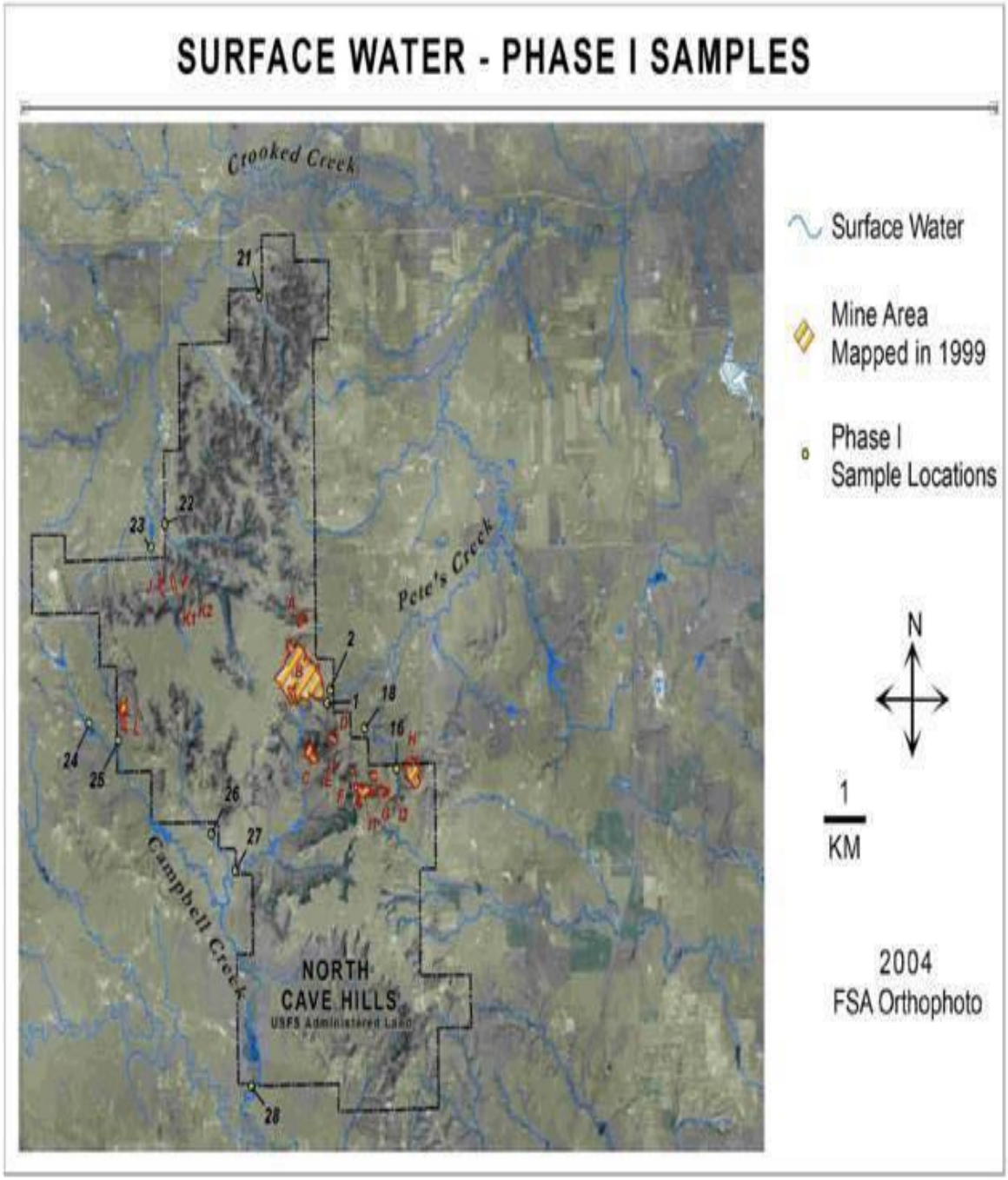

Figure 3: North Cave Hills Phase I surface water sampling locations.

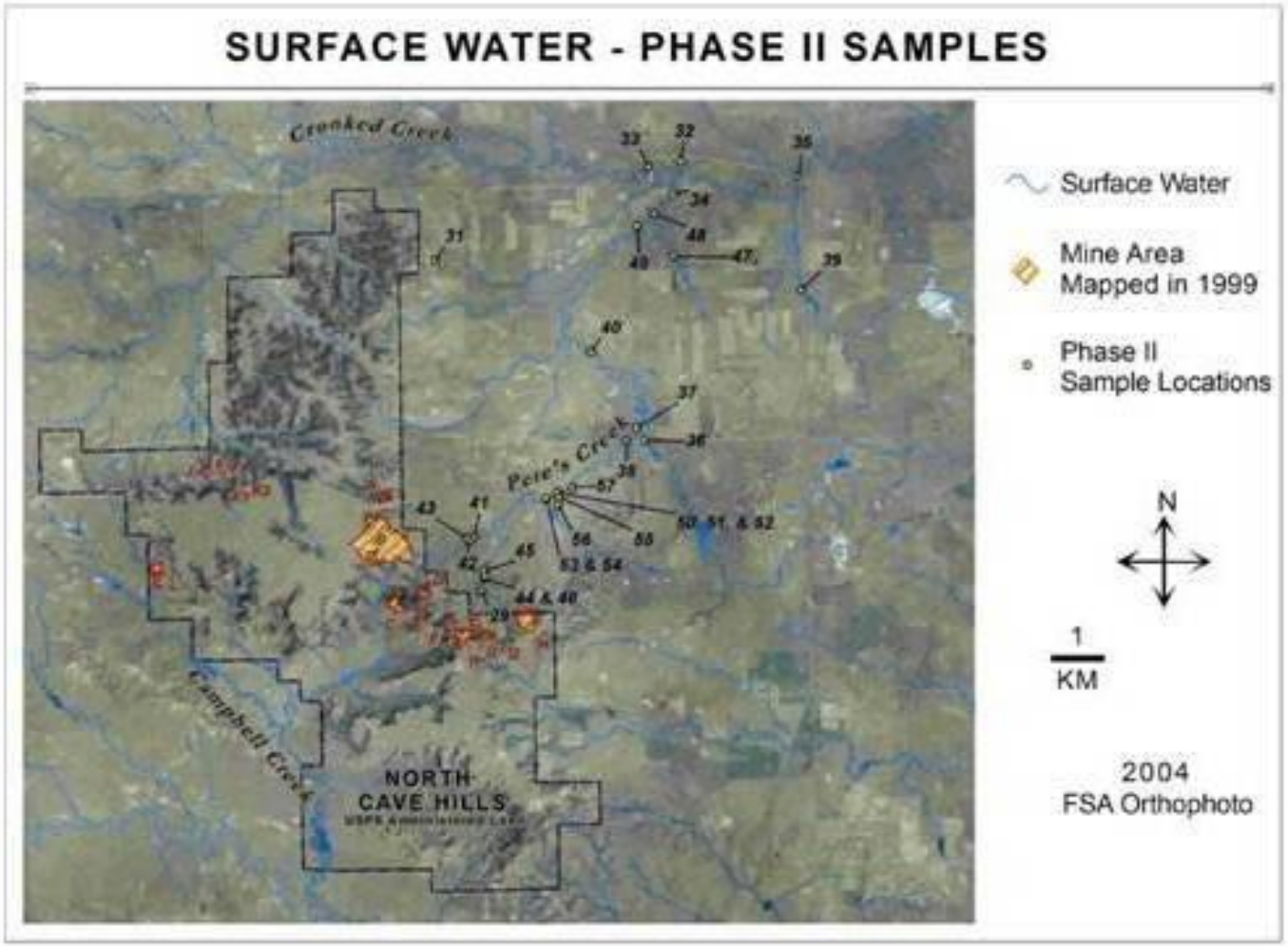

Figure 4: North Cave Hills Phase II surface water sampling locations

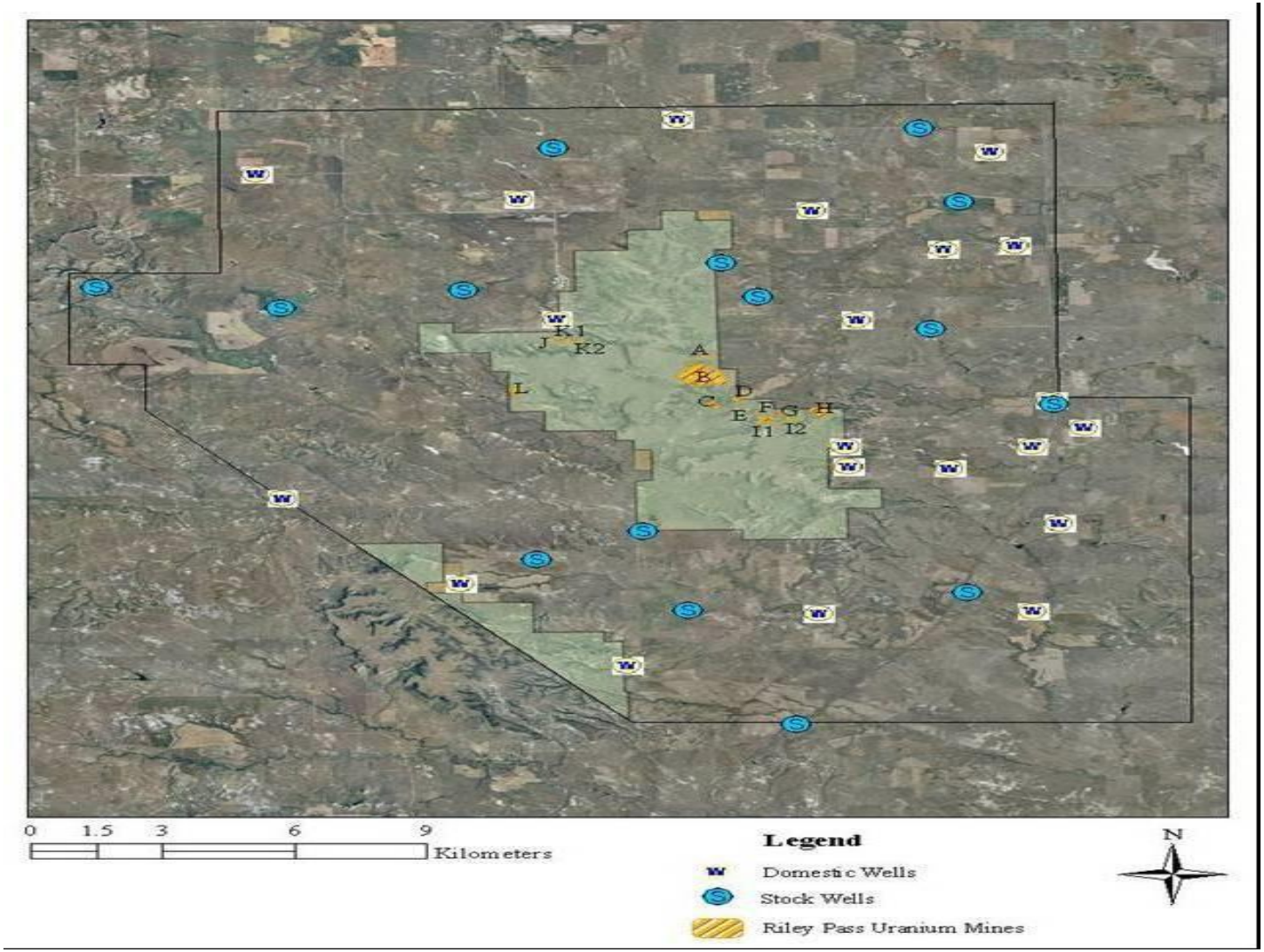

Figure 5: Groundwater sampling locations. Domestic and stock wells are designated by symbol

240 m and represented shallow unconfined alluvial aquifers and deep confined aquifers. Samples were collected after pumping a volume of water equal to or exceeding two times the casing volume.

Airborne dust particulates were collected from 30 locations in two sampling phases. Phase I sampling occurred within the set 8 km radius from the USFS boundary. These data were used to establish background concentrations in regions where impacts from wind were assumed not to be influenced by historical mining activities.. Phase I results were utilized to identify directions and regions to obtain additional samples in Phase II until a reduction in concentrations were observed. Airborne dust particulates were collected using a portable wind tunnel (Stetler, 1999) following these criteria:

- sites were high elevations above stream courses to ensure dust was either deposited directly by air or was derived by soil-forming processes;
- sampling sites were designed in a 'gridded' network around the NCH and were sufficiently distributed in accordance with the prevailing wind directions;
- sites were at locations that had physical indications of the presence of fine-grained surface materials derived from mechanisms excluding fluvial processes;
- locations had surfaces where dry and loose fine-grained materials were readily available to wind processes and could be collected either from the existing surface or generated through preparation of a standard loose surface (Saxton et al., 1998)

Samples collected to represent potential environmental transport by the above listed mechanisms were compared to established background values calculated using:

$$mean + 3x \,\text{standard deviation} \qquad (1)$$

These background values were determined using samples collected from non-impacted drainages and represent natural metals concentrations. For airborne dust particulates, a background value of mean + 2 × standard deviation was used.

Further discussion on sample collection, analysis, and quality assurance and quality control methodology utilized within this study may be found in Stone et al. (2007).

Results

The key metals of interest in this study were As and U, although concentrations for the full suite of analytes listed above were determined from each sample. In the following discussions,

only As and U results will be presented within this paper. Further discussion regarding specific contaminant trends within all environmental mediums studied may be found in Stone et al. (2007). This final project report may be downloaded at http://uranium.sdsmt.edu.

Soils

Soil samples were collected from cores (0.3—8 m deep) drilled in numerous watersheds surrounding the mined and non-mined areas. Two or more composite samples were prepared from most of the deeper cores, with sampling intervals of 0.5 m used for > 2.0 m cores. Shallow cores yielded a single sample. Background concentrations were determined using sites representing pristine environments not affected by mining.

Uranium concentrations in soils were mostly below the background value of 22 mg/kg and had a generally decreasing trend with increasing distance from the source areas (Stone et al., 2007). Higher values were associated with washover deposits and channeling of sediments. Values exceeding 3× the background were located immediately below Bluff B (Fig. 6).

Arsenic concentrations followed a trend similar to U concentrations. Background concentration was 32 mg/kg and was exceeded at several locations at values up to 64 mg/kg. The highest As value (96 mg/kg) was at the site with the highest U concentration.

Results showed that 14 watersheds were potentially impacted by sediment transport from previous mining activity. The most impacted area was in the upper Pete's Creek drainage below Bluff B where two U samples were 3× and 4× of background. All other U concentrations were below 2× background.

Surface Water

Fourteen watersheds were identified within the study area that were potentially impacted by uranium mining. Four pristine watersheds were used to determine background concentrations for all analytes. Sampling of surface water was completed in two phases; Phase I was a large-scale attempt to define contamination regionally, and Phase II sampling isolated impacted areas and delineated contaminant extent. Results indicated there were two impacted areas: the Upper Pete's Creek watershed below and east of Bluff B where surface waters were derived from direct runoff of the spoils piles, and Campbell Creek which received runoff down the western slopes of the North Cave Hills.

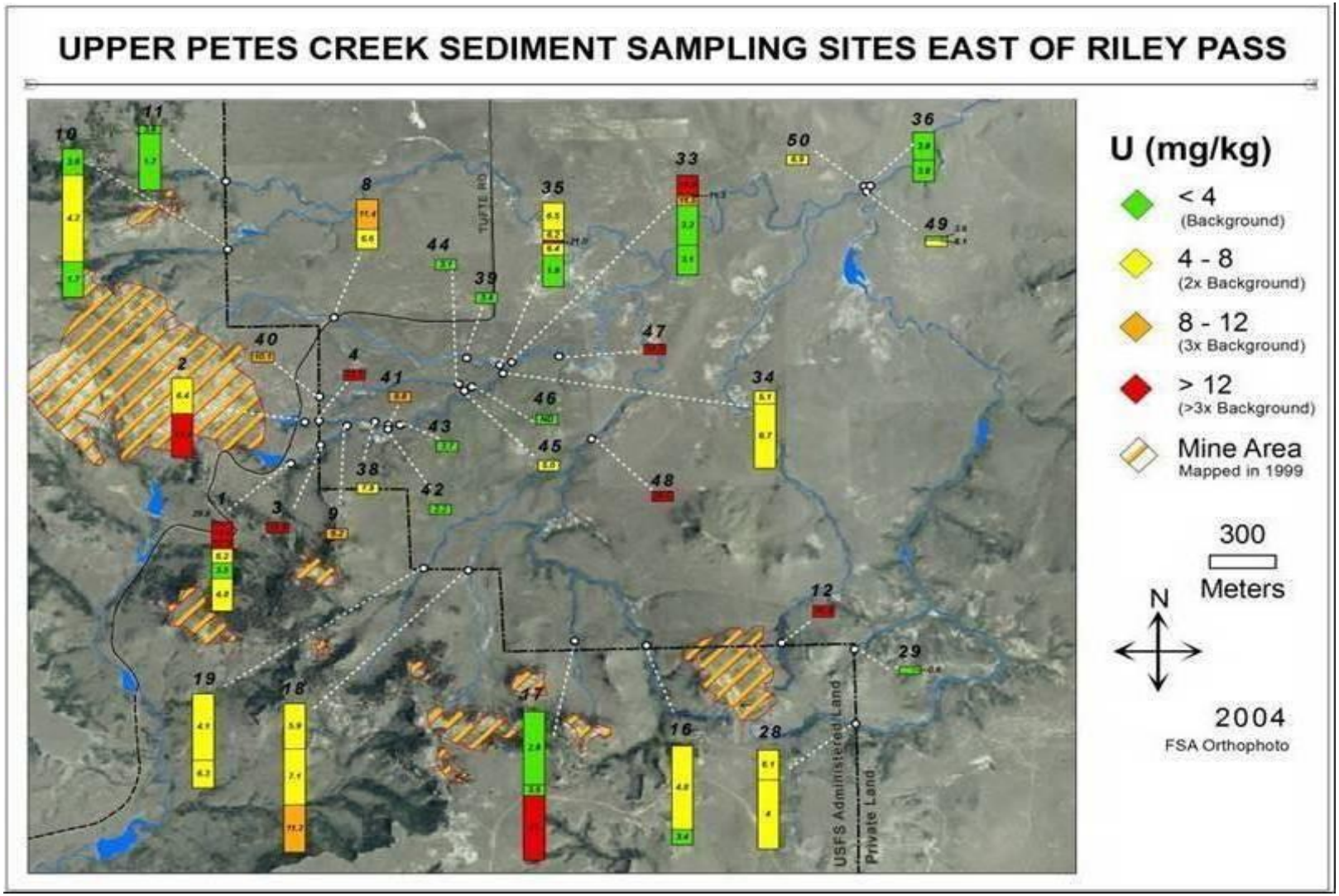

Figure 6: Uranium concentrations of core samples found in upper Pete's Creek. Bluff B is the largest mined area located in the upper left of this figure.

Phase I sampling indicated elevated U and As concentrations in both watersheds. Elevated U concentrations in the Upper Pete's Creek drainage ranged from 2.9× to 5× above the background value of 0.027 mg/L. Elevated As concentrations ranged from 28× to 33× above the background value of 0.020 mg/L. In Campbell Creek Draw, elevated U concentrations were 3.6× background. These results were used to define Phase II sampling to isolate the contamination and identify locations with the greatest contamination.

Phase II sampling indicated severe contamination in the Upper Pete's Creek watershed below and to the east of Bluff B where the highest U and As concentrations were 23× and 89× background, respectively (Fig. 7 and 8). Sampling in the downstream direction showed that background concentrations were achieved within a distance of ~5 km below Bluff B, and were continuously observed for all analytes at the confluence of Pete's Creek and Crooked Creek, ~ 15 km northeast of Bluff B.

Groundwater

Contaminant occurrence was ubiquitous in groundwater samples at all depths of wells tested and was distributed within the study area both up and down gradient. The exceptions were Th, As, and V, which were not detected in any samples. Trends of occurrences suggest that metals detected were regional, naturally occurring contaminants and not directly attributable to uranium mining.

Water quality was compared against maximum contaminant levels (MCLs) established by the US EPA (US EPA, 2006). Uranium was detected in seven wells and ranged from 0.001 to 0.064 mg/L (Fig. 9). One well was 0.064 mg/L and was the only sample that exceeded the MCL of 0.03 mg/L. This well was 18 m deep and was fed by shallow alluvial gravels with hydraulic connectivity to uraniferous lignites updip from the well. A second well contained a U concentration of 0.027 mg/L and was also a shallow well in alluvium.

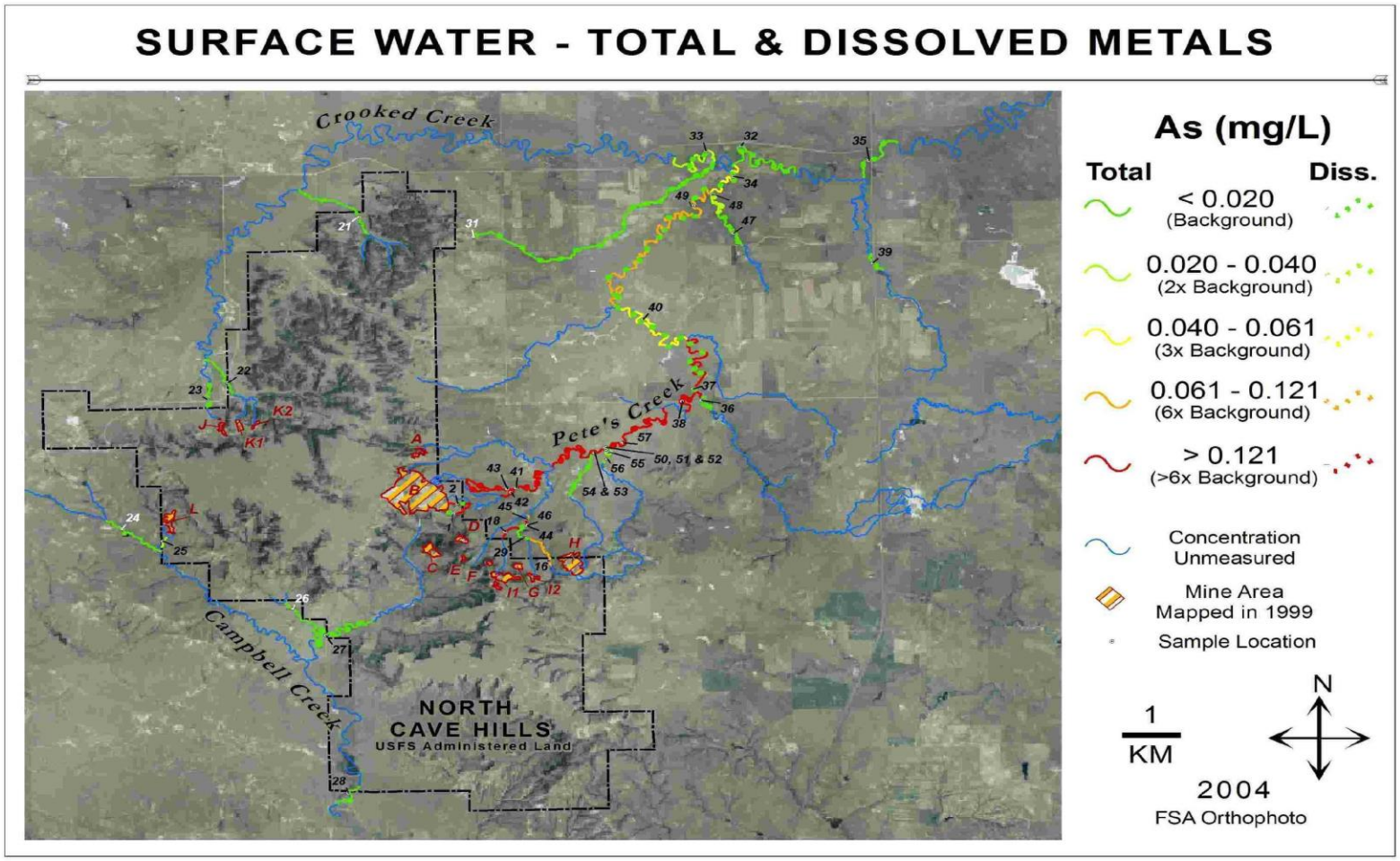

Figure 7: North Cave Hills surface water results for total and dissolved arsenic..

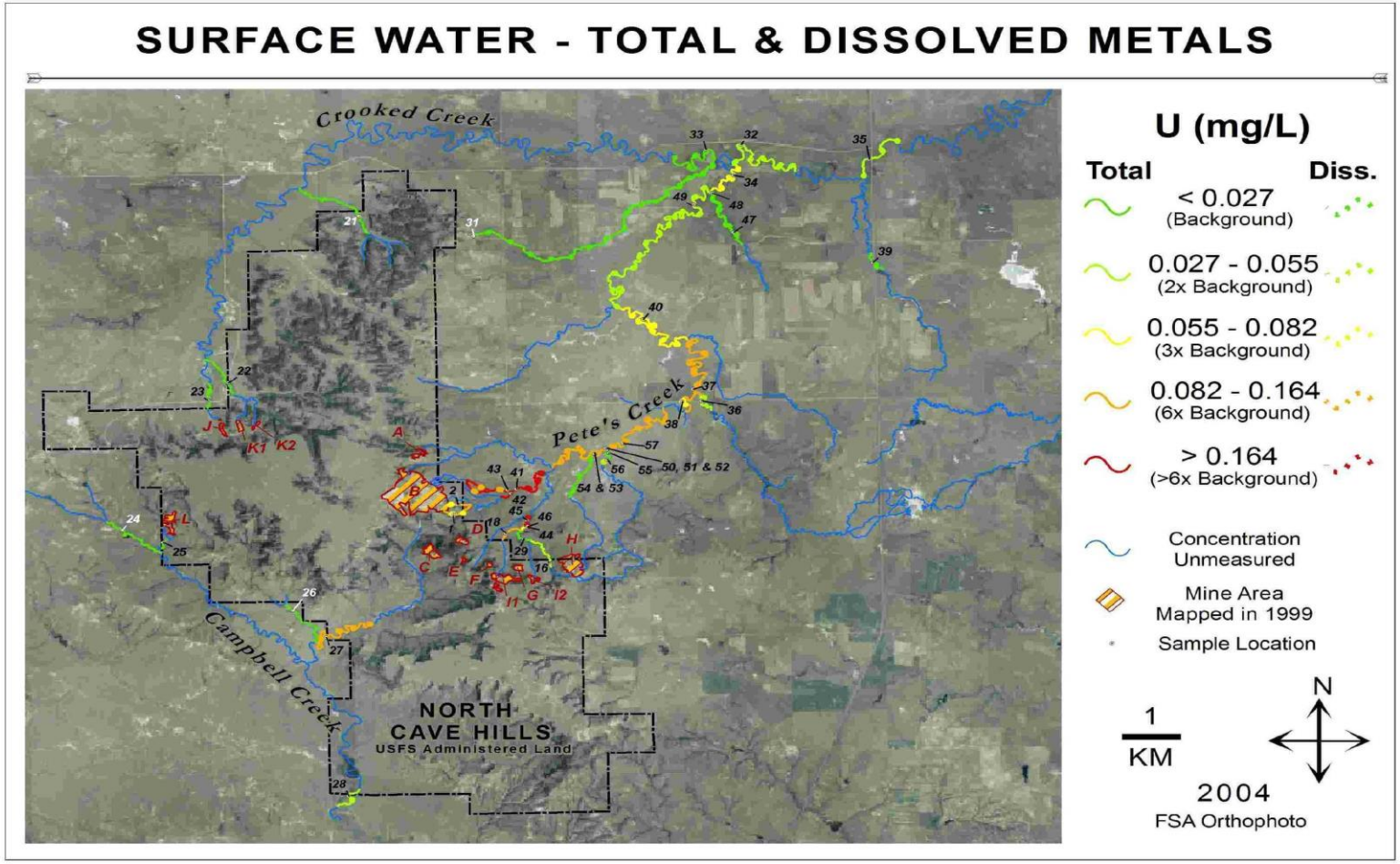

Figure 8: North Cave Hills surface water results for total and dissolved uranium.

Many elements in the U decay series can contribute radionuclide counts in a sample. Analysis to determine which element contributed what amount of radioactivity is expensive and time consuming. Thus, gross alpha counts were used to assess the radioactivity of the sample. The EPA has established a MCL of 15 pCi/L for gross alpha. Twenty-six of the 34 wells contained gross alpha radiation ranging from 1.8 to 44.4 pCi/L. Distribution of these wells were up and down gradient of the abandoned mines and at depth from 18 to over 240 m. Three wells exceeded the MCL for uranium. The two highest concentrations were the shallow wells discussed above and the third highest concentration was a 122 m deep well completed into the Fox Hills sandstone located northeast of Bluff B.

In addition to U concentrations and gross alpha contents, three wells contained Ra^{226} in concentrations ranging from 0.5 to 0.7 pCi/L and were all well below the MCL of 5 pCi/L. Two of the wells had concentrations of 0.7 pCi/L and were the same wells as those with the highest gross alpha contents discussed above. U^{235} was detected in five wells at concentrations between 0.3 and 1 pCi/L, up and down gradient of the mine sites. There is no MCL for U^{235} data collected and analyzed during the groundwater study indicate that metals and radionuclides were natural components of the groundwater systems. Further, the distribution of the contaminants shows metals were dissolved between the recharge areas to the west and the North Cave Hills. It is not clear if the abandoned uranium mines in the North Cave Hills contribute directly to the metals content of the ground water. Most likely the chemistry of surface water and local springs were affected by the mines but the deep aquifers should not be affected directly. Shale confining layers above the deep sources theoretically protect infiltrating waters from reaching these aquifers. The exception would be the presence of deep fracture systems allowing local infiltration to reach the water table, i.e., a leaky confining layer. These conditions will be evaluated in future studies.

Surface Dust

Results of the surface dusts indicate the general ubiquity of target analytes in the soils around the North Cave Hills. Uranium was present in all but two samples and arsenic was present in all samples. Vanadium, Cu, and Th also were present in all samples and Pb occurred in all but one sample. Molybdenum and Se had the least occurrences.

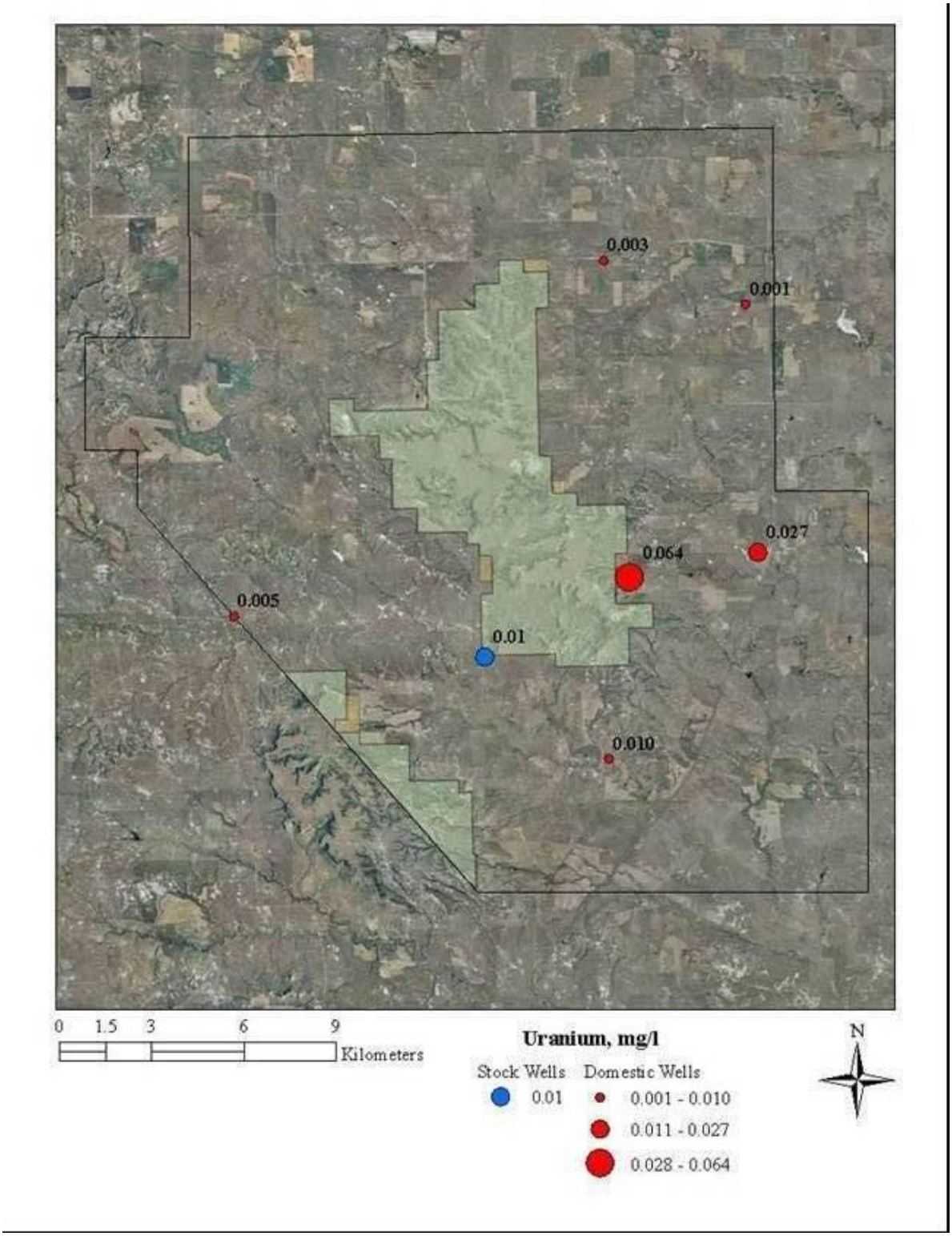

Figure 9: Uranium distribution and occurrence in the groundwater system. One well exceeded the MCL of 0.03 mg/L

Thirteen samples contained U concentrations in excess of the calculated background value of 0.74 mg/kg. Distribution of these sites extended across the entire sampling area and they were classified into three distinct domains:

1. Eight locations, including the three with the highest uranium concentrations (1.96, 1.66, and 1.6 mg/kg), occurred in a northwest to southeast trend cutting across the center of the North Cave Hills and containing the largest abandoned uranium mine areas. The long axis of this high dust concentration area also correlated to the predominant wind direction, indicating a probable wind influence on the observed distribution. The greatest uranium concentration (1.96 mg/kg) unexpectedly occurred on the western side of the North Cave Hills upwind (predominant wind direction) of all of the abandoned mine sites;
2. Two locations north of the North Cave Hills had uranium concentrations greater than 1.0 mg/kg and both were in a direction of minimal above-threshold wind occurrences;
3. Two locations in a topographic low between the North and South Cave Hills (Bull Creek drainage) had uranium exceedances.

Uranium concentrations were below background levels in all areas away from these three identified areas of high concentrations.

Arsenic was detected in all samples and five contained concentrations above the calculated background value of 11.93 mg/kg. All exceedance concentrations were east and south of the North Cave Hills. However, concentrations below the background level that were between 9.8 and 11.6 mg/kg were located near the exceedance locations and appeared to form a similar northwest to southeast pattern across the center of the North Cave Hills as was noted for uranium Domain 1 listed above. Two locations in the north were close in value to the As background level that corresponded to uranium domain 2. Thus, there is some evidence for wind distribution of fine particles that contained these metals.

Discussion

Significant environmental degradation has occurred from transport of heavy metals and radionuclides downstream of abandoned uranium mines in the North Cave Hills. The most impacted regions occurred down gradient of Bluff B and in Campbell Creek, SW of the study area. Surface water contained U and As up to 90× background values and sediment had concentrations 4× background levels. Metals in sediments and surface water were naturally

attenuated within ~15 km below the mine sites, although large deposits of contaminated soils occurred on private lands

Ground water contained many metals and radionuclides in both the up and down gradient directions and at shallow to deep aquifer depths. This indicated that metals contamination was regional in extent and most likely were not affected by U mining

Surface dust had significant metals concentrations in nearly all locations sampled. Uranium was ubiquitous and contained at least one plume of high concentrations that correlated to the predominant wind direction. All metals concentrations in aerosols were decreasing or below background levels within about 15 km from the mine sites. Although U, As, and other metals were detected in surface dusts, their concentrations were on average seven times less than metals concentrations in soils, indicating wind-driven transport of metals remains low.

Acknowledgements

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