



Uranium and Arsenic in the Baseline Environment, Dawson Range, Yukon

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**THE UNIVERSITY
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**LORAX
ENVIRONMENTAL**

Lay Summary

Lay Summary

This report provides a regional assessment of uranium (U) and arsenic (As) sources, distribution, and mobilization processes in the baseline environment in the Dawson Range, Yukon. Both of these chemical elements are naturally occurring, but are present at concentrations exceeding federal water-quality guidelines (*e.g.*, for the protection of aquatic life and for drinking water) in certain Dawson Range groundwaters (U and As) and surface water (U only). A dataset including chemical analyses of thousands of surface water, sediment, and rock samples and hundreds of groundwater samples was compiled for review in this report by pooling results of previous work by the Geological Survey of Canada, the Yukon Geological Survey, and the Coffee and Casino mineral exploration projects. Uranium is released through weathering of granitic and metamorphic rocks in the Dawson Range, and exceeds water-quality guidelines in a number of groundwater and surface-water monitoring locations in the region. Arsenic concentrations in surface water are consistently low, but can exceed water-quality guidelines in groundwater. Arsenic enrichment is more localized, and is associated to ore deposits where As-rich rock and As-rich groundwater is encountered. The presence of geogenic (natural) sources of U and As in the Dawson Range indicates that projects that will expose large amounts of rock to chemical weathering (*e.g.*, mining, quarrying) should evaluate the potential for increased release of these elements to the environment. In addition, site-specific water-quality objectives may have to be developed for projects in the Dawson Range given the frequently elevated U in baseline surface water.

Executive Summary



Executive Summary

Yukon Environment Water Resources Branch has contracted Lorax Environmental Services in partnership with the Department of Earth, Ocean and Atmospheric Sciences of the University of British Columbia to conduct a study on uranium and arsenic in the baseline environment in the Dawson Range, Yukon, in response to observations of concentrations of these metal(loid)s in groundwater and/or surface water at levels that exceed federal water-quality guidelines. The primary objective of this report is to compile available groundwater, surface water, sediment, and rock geochemical data to characterize the geological sources of U and As in the Dawson Range, and identify processes leading to elevated baseline concentrations of these elements in water.

Data considered in this report included baseline environmental monitoring analyses conducted by the Coffee and Casino mine projects and geochemical reconnaissance sampling conducted by the Geological Survey of Canada. Geochemical data comprising thousands of rock, sediment, and surface water samples, and hundreds of groundwater samples collected in the Dawson Range through these industrial and government initiatives were synthesized and analyzed.

Widespread U at levels near or above the average of Earth's upper crust (2.7 µg/g; Rudnick and Gao, 2013) are observed in Dawson Range geologic units. Uranium-rich groundwater is associated with dissolved Ca and alkalinity, which forms uranium-calcium-carbonate aqueous complexes that can hinder sorption and reduction reactions that lead to U removal, thus promoting U solubility in the groundwater system. Uranium loading to surface water bodies in the region is a result of groundwater discharge.

Elevated U concentrations are observed in some instances in high-sulfate groundwaters around mineralized areas, which may indicate that U mobilization is indirectly enhanced by sulfide-mineral oxidation. Sulfide-mineral oxidation promotes Ca and alkalinity release through carbonate buffering reactions, which may increase uranium-calcium-carbonate complexation and thus increase U solubility. However, because groundwater monitoring has been centered around prospective ore deposits that are associated with sulfide minerals, there is relatively limited information on U content in groundwater outside of mineralized areas. Geogenic enrichment of U in the baseline (pre-mining) environment suggests that this parameter constitutes a contaminant of potential environmental concern during weathering of mine waste-rock and/or tailings in the Dawson Range, in particular carbonate-bearing deposits producing alkaline- and Ca-rich drainage.

In contrast to U, As mobilization is more localized and tied to areas of gold mineralization in the Dawson Range; unmineralized country rocks in the Dawson Range otherwise have

low As content. Arsenic is released from the rock through sulfide-mineral oxidation and is sorbed onto Fe-(oxyhydr)oxides. Development of anoxic groundwater conditions leads to reductive dissolution of Fe-(oxyhydr)oxides and release of sorbed As to groundwater at levels often exceeding regulatory guidelines for the protection of aquatic life (5 µg/L). However, unlike U, dissolved As in groundwater is not transported into surface water in the Dawson Range. Mobilization of As occurs most strongly under anoxic conditions where Fe-(oxyhydr)oxides are unavailable for As sorption. The mechanism of As attenuation during groundwater discharge into surface water is probably related to groundwater oxidation and sorption of As onto Fe-(oxyhydr)oxides.

Overall this study shows that elevated U in water is widespread in the Dawson Range through weathering of disseminated U in rocks, a process that is enhanced by elevated Ca and alkalinity in groundwater. Arsenic release is more localized and associated with ore deposits. The potential for release of U and As should be considered for development projects that generate large amounts of waste rock (in particular mining or quarrying) where weathering reactions are enhanced by increasing rock exposure to water and air. In addition, concentrations of U in surface water often exceed water-quality guidelines. Establishment of site-specific U water-quality objectives for industrial projects (*e.g.*, mining) is recommended under these conditions, and these water-quality objectives should consider risks to ecological receptors if higher U concentrations relative to the baseline environment are produced through industrial activities.

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Glossary

Glossary

ARD	Acid-rock drainage
AUC	Average upper crust elemental abundance
Casino	Casino property, Casino Mining Company
Coffee	Coffee Gold Project, Newmont-Goldcorp
CCME	Canadian Council of Ministers of the Environment
GSC	Geological Survey of Canada
ICP-MS	Inductively coupled plasma mass spectrometry
OF-8500	Open File 8500 dataset
NRD	Neutral-rock drainage
QEMSCAN	Quantitative Evaluation of Materials by Scanning Electron Microscopy
RDL	Reported instrumental detection limit
RGS	Regional Geochemical Survey
TDS	Total dissolved solids
μ -XRF	micro X-ray fluorescence
WQO	water-quality objective
XANES	X-ray absorption near-edge spectroscopy
XRD	X-ray diffraction

1. Introduction



1. Introduction

Uranium (U) and arsenic (As) contamination pose an important concern for the protection of the environment. Neither of these elements have any known biological function (Acharya and Apte, 2013; Plant *et al.*, 2014). Arsenic is mutagenic, carcinogenic, and teratogenic (Plant *et al.*, 2014). Uranium toxicity can involve oxidative damage to biological tissues in fish and aquatic invertebrates, and kidney damage to humans (Goulet *et al.*, 2011). Natural U generally has low radioactivity, such that its toxicity arises primarily through chemical (and not radioactive) effects (Goulet *et al.*, 2011).

Both U and As have been observed at concentrations exceeding regulatory guidelines for the protection of aquatic life in groundwater unimpacted by human activity the Dawson Range. The Dawson Range is also host to a number of prospective mineral deposits. An understanding of the geological source of U and As in Dawson Range rocks and processes causing the mobilization of these elements into water under baseline (pre-mining) conditions is critical for two main reasons: (1) improving a mine project's ability to predict water-quality concerns associated to weathering of mine waste rock and tailings; and (2) defining water-quality objectives at industrial sites where elevated U and As (relative to regulatory guidelines) may be present in the baseline environment.

Aside from mining industry, humans have had a presence in the Dawson Range for millenia. First Nations that have traditional and/or asserted territory in the region include Tr'ondëk Hwëch'in, White River, Na-cho Nyäk Dun, and Selkirk First Nations (Yukon Territory, 2013). Human traffic in the area also includes a variety of traditional indigenous and non-traditional activities such as habitation, hunting, trapping, fishing, outdoor recreation, and plant harvesting (Casino Mining Company, 2014; Goldcorp, 2017), creating a possible course of exposure of people to U and As even in the absence of major industrial projects.

The objective of this report is to synthesize the available geochemical data on U and As in the Dawson Range and constrain geological sources and geochemical mobilization pathways for these elements under baseline conditions. Implications for mine-waste management are also discussed.

1.1 Objectives

1. Synthesize the existing rock, sediment, and water-quality data in the Dawson Range and characterize the occurrence and abundance of U and As in the regional baseline environment.
2. Identify the geological sources of U and As in the region

3. Infer mechanisms of U and As mobilization and attenuation.
4. Identify knowledge gaps and implications for water-quality objectives and mine-waste management in the Dawson Range

1.2 Report structure

This report is organized into five sections following the introduction. Section 2 presents an overview of the study area and a review of the environmental geochemistry of U and As. Section 3 summarizes the data sources and data analysis methods. Section 4 presents results of the study. Section 5 integrates results to discuss U and As sources, mobilization pathways, and implications for water-quality objectives and mine waste management. Section 6 provides a closing summary and conclusions.

2. Background



2. Background

2.1 Geological and geomorphological setting

The Dawson Range broadly trends in a NW-SE direction from the Yukon-Alaska border (north of Northway Junction, Alaska) to the mountains surrounding the Mount Nansen Mine in Yukon (MacWilliam, 2018). Its topography is characterized by broad and convex ridges and v-shaped valleys (Bond and Lipovsky, 2011), with elevations generally ranging between approximately 400 and 1,400 metres above sea level. Despite its northern latitude, the region was unglaciated during the Pleistocene (Duk-Rodkin, 1999). Today, semi-discontinuous permafrost is present in the area, with the thickest accumulations located on north aspects and near valley bottoms (Bond and Lipovsky, 2011). Surficial geology includes weathered bedrock mostly present on ridgetops, colluvium, and loess (Bond and Lipovsky, 2011). Thicker alluvium and organic soils are present in valley bottoms (Knight Piésold, 2013; Goldcorp, 2017).

In addition to the Coffee and Casino deposits, there are also numerous other mineral exploration targets in the Dawson Range including Au, Cu, Mo, Ag, Mn, and volcanogenic massive sulfides (MacWilliam, 2018). Also of relevance for the current study are the HASL (MINFILE 115J 092) and Onasick (MINFILE 115J 093) U exploration properties located in the Klondike Plateau, on the north bank of the Yukon River; while no mine projects are currently proposed at these locations, recent exploration found sediment samples containing up to 4,790 µg/g U and localized uraninite mineralization at HASL, and up to 408 µg/g U in soil samples at Onasick. However, a drilling campaign at HASL in 2007 failed to locate economic U (Yukon Geological Survey, 2019).

In the context of the present study, the study area, shown in Figure 2-1, was constrained within:

- All watersheds draining into the White River's eastern flank, beginning at the confluence with the Yukon River, and extending up the White River's stream until the confluence with the Donjek River.
- Watersheds draining into the east bank of the Donjek River upstream until and including Dip Creek, but excluding the Klotassin River upstream of Dip Creek.
- Watersheds draining into the S bank of the Yukon River from the confluence with the White River, going upstream in the Yukon until (and including) Isaac Creek.
- Watersheds draining into the N bank of the Yukon River from the confluence with the White River (Frisco Creek to the N) upstream until (excluding) Cripple Creek to the S/SE (while not strictly part of the Dawson Range, these watersheds were included given the historical U mineral exploration properties around HASL and Onasick).

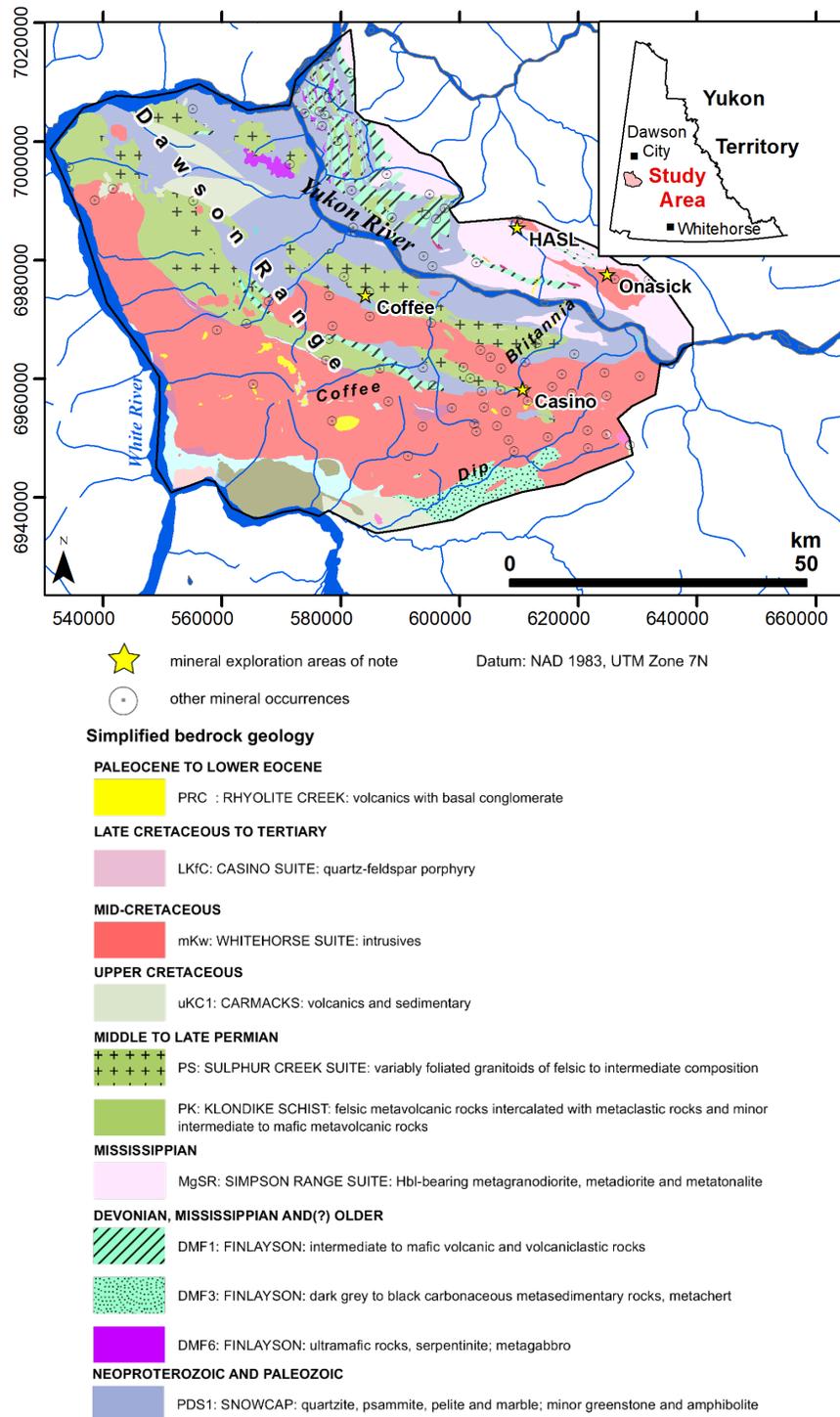


Figure 2-1: Dawson Range geological map with select mineral exploration projects including the Coffee and Casino deposits and the HASL and Onasick U anomalies. The inset map shows the study area location within Yukon, Canada. Data from Geomatics Yukon (<ftp://ftp.geomaticsvukon.ca/GeoYukon/>). Legend only includes a subset of simplified geological units.

This study area encompasses the key geologic units of the Dawson Range and includes the Casino and Coffee Gold properties, which have produced the most detailed baseline environmental characterization data in the region (Figure 2-2 and Figure 2-3).

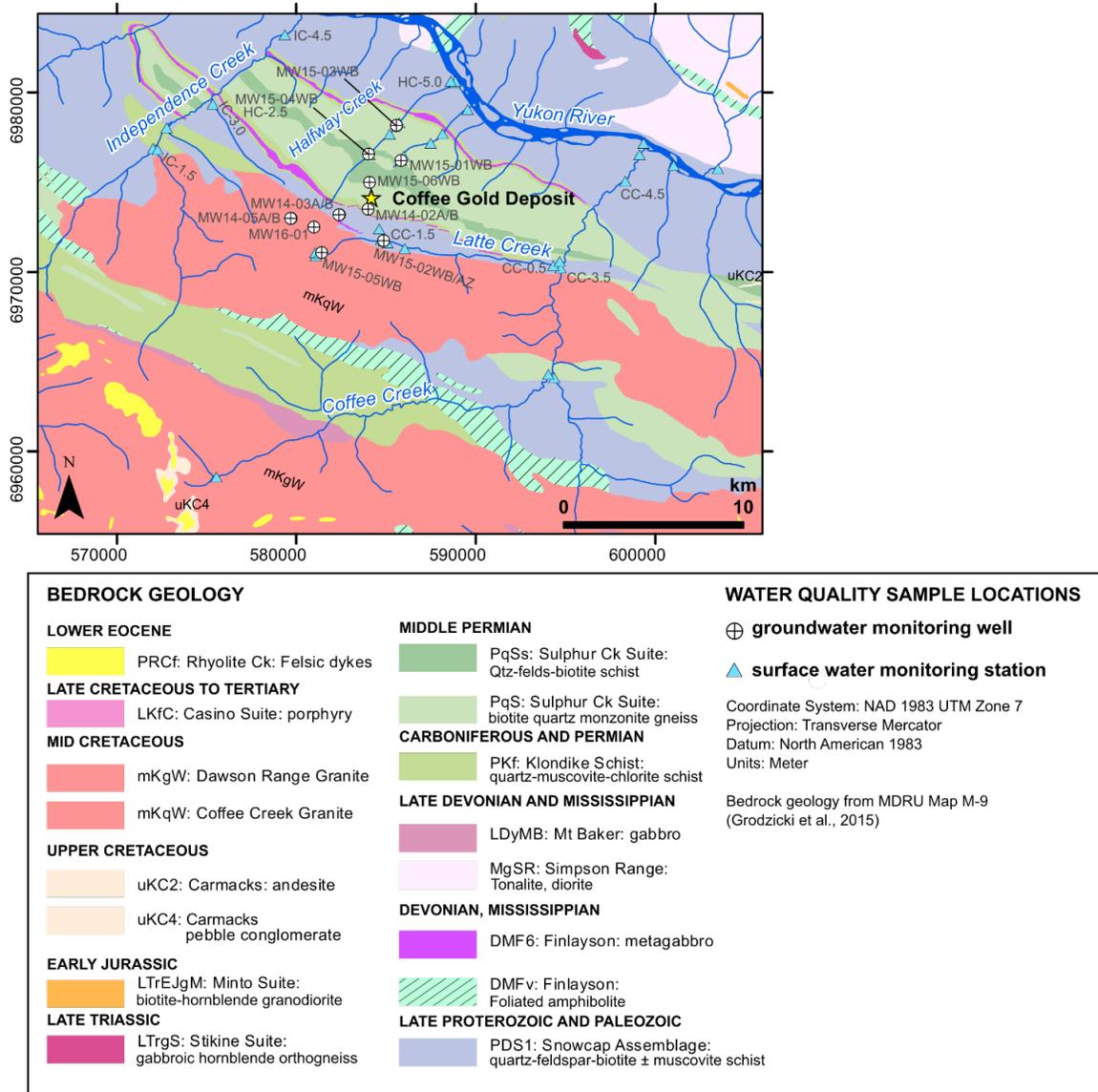


Figure 2-2: Groundwater and surface water monitoring locations around the Coffee Gold property along with underlying bedrock geology. Labels are only shown for a subset of water-quality locations.

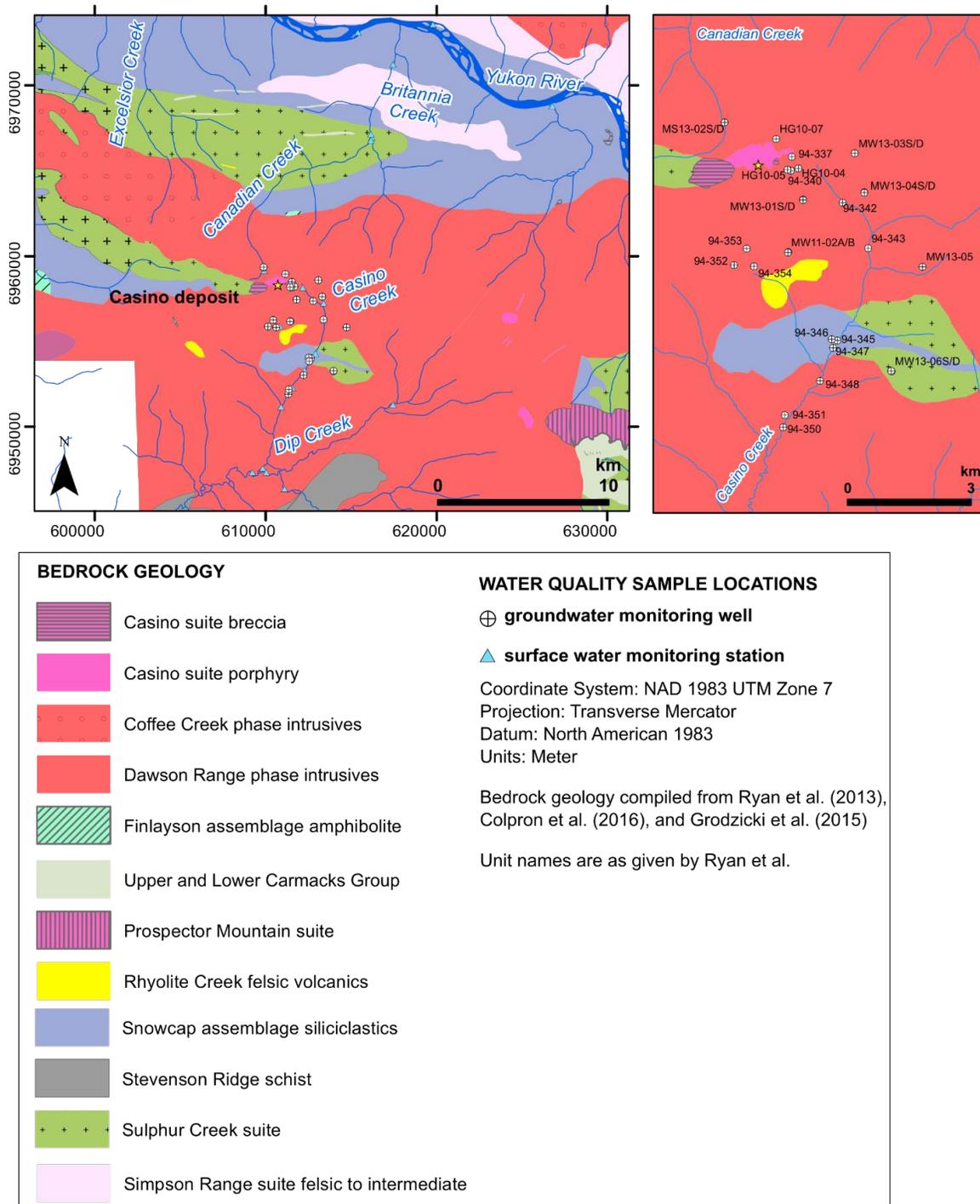


Figure 2-3: Groundwater and surface water monitoring locations around the Casino property along with underlying bedrock geology. Inset at right shows groundwater well IDs.

2.2 Environmental geochemistry of U and As

2.2.1 Uranium

Uranium is a lithophile metallic element that is typically enriched in crustal rocks relative to the mantle due to its incompatible behaviour in magmatic melts (Kyser, 2014). Typical U contents in rock therefore increase from ~ 1 µg/g in mafic rocks up to several tens of µg/g in granitic and rhyolitic rocks (Rudnick and Gao, 2013; Cumberland *et al.*, 2016). The average upper crust U content is estimated at 2.7 µg/g (Rudnick and Gao, 2013).

The mineralogy of U includes both distinct U-bearing phases and U substitutions into various minerals. The most common U minerals are uraninite (UO₂), coffinite (USiO₄), brannerite (U,Ca,Ce)(Ti,Fe)₂O₆, autunite [Ca(UO₂)₂(PO₄)₂•10-12H₂O], and uranothorite (Th,U)SiO₄ (Kyser, 2014). The U⁴⁺ cation can substitute for Th⁴⁺, Zr⁴⁺, Nb⁵⁺, Ta⁵⁺, and rare earth elements in other minerals (Kyser, 2014; Wu *et al.*, 2014).

Uranium solubility in natural waters is tied to sorption and U oxidation state, which are controlled by water pH, redox, and the presence of ligands that can form aqueous complexes with U. Uranium(VI) and U(IV) are the two most common oxidation states of U; U(IV) being generally insoluble at pH > 3, while U(VI) forms the relatively soluble uranyl (UO₂²⁺) aquocation (Cumberland *et al.*, 2016). U(VI) therefore constitutes an important mechanism of U removal in groundwater through precipitation of biogenic or abiotic uraninite [U(IV)O_{2(c)}] (Abdelouas *et al.*, 2000). The importance of this mechanism is driven by observations of U accumulation in organic-rich sediments (Tixier and Beckie, 2001; Cumberland *et al.*, 2016). Uranium(VI) solubility is generally limited by sorption of UO₂²⁺ to mineral surfaces. However, U(VI) sorption is hindered by complexation of UO₂²⁺ with various anionic ligands, the most important of which are carbonate (CO₃²⁻), silicate (SiO₄⁴⁻), phosphate (PO₄³⁻), vanadate (VO₄³⁻), and, at low pH, sulfate (SO₄²⁻) (Cumberland *et al.*, 2016).

In many groundwater settings, UO₂²⁺ complexation with carbonate and calcium are important drivers of U mobilization (Stewart, 2008). This effect is primarily because the ternary aqueous complexes Ca₂UO₂(CO₃)₃ and CaUO₂(CO₃)₃²⁻ outcompete mineral surfaces for UO₂²⁺ binding, thus weakening U sorption (Figure 2-4) (Stewart *et al.*, 2010). In addition, the thermodynamic stability of these ternary complexes is such that they are more resistant to biogenic U(VI) reduction, further enhancing U mobility (Stewart *et al.*, 2011).

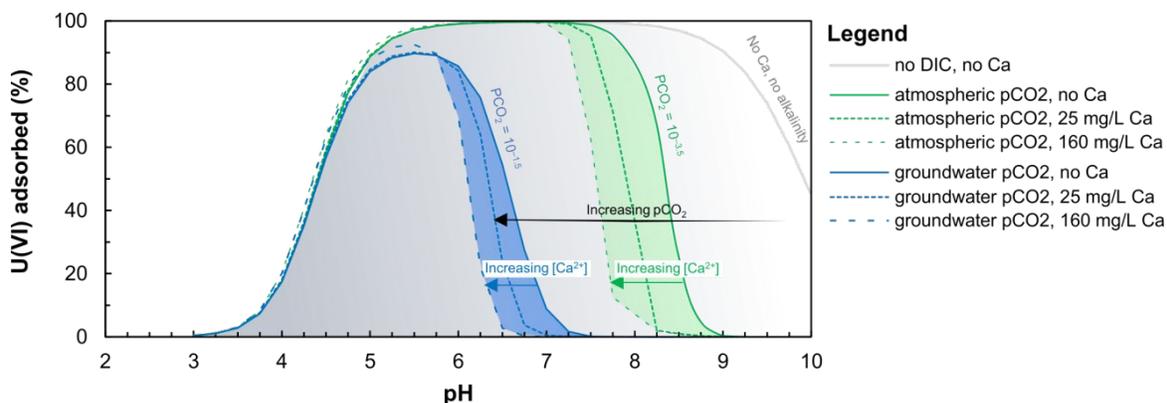


Figure 2-4: Generalized U(VI) sorption behavior on Fe-(oxyhydr)oxide as a function of pH, Ca, and alkalinity in a PHREEQC simulation. Alkalinity is fixed by pCO_2 representative of typical atmospheric ($10^{-3.5}$ atm) and groundwater ($10^{-1.5}$ atm) concentrations. The U sorption envelope decreases from pH ~6.5 to 9 as pCO_2 and $[Ca]$ increase, indicating greater U(VI) solubility.

Excessive U intake can be toxic. Studies of acute U exposure to fish show gill damage, reduced blood flow, asphyxiation, and renal damage (Goulet *et al.*, 2011). Toxic effects from chemical interaction of U with biological tissues is a greater concern in natural environments than is radiation damage due to the long half-life of ^{238}U and its low tissue penetration (Goulet *et al.*, 2011). As a result of U toxicity, regulatory guidelines are in place in Canada. The long-term exposure guideline for the protection of aquatic life is 15 $\mu g/L$ (Canadian Council Of Ministers Of the Environment, 1999b). The proposed maximum allowable concentration in drinking water is 20 $\mu g/L$ (Health Canada, 2017).

A survey of tens of thousands of U analyses in Canadian lakes and streams shows median concentrations of <0.05 and 0.06 $\mu g/L$. However, groundwaters with U concentrations reaching hundreds to thousands of $\mu g/L$ through natural mobilization processes have been reported in several localities, including Finland (Asikainen and Kahlos, 1979; Kurttio *et al.*, 2017), Burundi (Post *et al.*, 2017), the USA (Welch and Lico, 1998; Orloff *et al.*, 2004), Manitoba, Canada (Gascoyne, 2004), British Columbia, Canada (Tixier and Beckie, 2001), China (Wu *et al.*, 2014), and Korea (Lee *et al.*, 2001; Kim *et al.*, 2004). In many instances, U-rich groundwater is associated to granitic or sedimentary basins (Smedley *et al.*, 2006). In addition, hundreds to thousands of $\mu g/L$ U are possible downstream from U mining activities (Goulet *et al.*, 2011).

2.2.2 Arsenic

Arsenic is a metalloid of strong chalcophile character (Plant *et al.*, 2014) which tends to form covalent-type bonding with sulfur. Sulfides therefore constitute an important reservoir of As-bearing minerals such as arsenopyrite (FeAsS), arsenian pyrite Fe(As,S)₂, realgar (As₄S₄), and orpiment (As₂S₃). Arsenic has been used as an indicator of gold mineralization because Au is often associated with arsenopyrite (Leeuwen *et al.*, 1990; Yang and Blum, 1999; Clark and Raven, 2004; Blowes *et al.*, 2014). Arsenates and sulfosalts, often found in evaporitic or post sulfide-oxidation environments, constitute another important pool of As minerals (Rudnick and Gao, 2013; Plant *et al.*, 2014). In contrast, the As content in silicate minerals is usually < 1 µg/g due to its poor compatibility in silicate mineral lattices (Plant *et al.*, 2014). Sedimentary rocks rich in organic matter, Fe-(oxyhydr)oxides, and phosphates can contain hundreds of µg/g As (Plant *et al.*, 2014). The average upper crustal abundance of As is 4.8 µg/g (Rudnick and Gao, 2013).

Like U, As solubility is redox-sensitive. Multiple oxidation states are possible, including As(-III), As(-I), As(0), As(III), and As(V). In oxidizing aqueous environments, As forms the oxyanion arsenate [As(V)O₄³⁻]. Under moderately reducing condition, As is found as arsenite [As(III)O₃³⁻]. Both arsenate and arsenite become increasingly protonated as pH decreases. In more reducing conditions, dissolved As can be reduced to the -III, -I or 0 oxidation states where it is often co-precipitated with secondary sulfide minerals (Plant *et al.*, 2014).

Arsenate and arsenite mobility can be limited by sorption and secondary mineral formation. Sorption is typically the most important control on As transport in natural waters, and occurs on the surface of various phases including organic matter, calcite, clays, and (oxyhydr)oxides of Al, Mn, and especially Fe (Smedley and Kinniburgh, 2002). Arsenic sorption weakens under alkaline-pH conditions, especially for arsenate (Goldberg, 2002). Arsenate sorption is generally stronger than that of arsenite (Grafe *et al.*, 2001; Goldberg, 2002; Grafe *et al.*, 2002; Dixit and Hering, 2003; Helle *et al.*, 2008). Complexation of dissolved As (*e.g.*, with dissolved organic carbon) or competition for the sorption surface sites with other oxyanions (*e.g.*, from phosphate) can weaken As sorption (Manning and Goldberg, 1996; Grafe *et al.*, 2001; Grafe *et al.*, 2002; Bissen *et al.*, 2003). Arsenic solubility can also be limited by precipitation of arsenate-group minerals including scorodite (FeAsO₄) and yukonite [Ca₃Fe(AsO₄)₂(OH)₃ · 5H₂O].

Given the mutagenic and carcinogenic properties of As (Plant *et al.*, 2014), water quality guidelines targeting this element exist in many jurisdictions. The maximum allowable As concentration in drinking water in Canada is 10 µg/L, and the CCME guideline for the protection of aquatic life is 5 µg/L (Canadian Council Of Ministers Of the Environment,

1999a; Health Canada, 2006). In natural aqueous environments, As concentrations are typically < 10 µg/L, but locally much higher concentrations are possible, in particular in groundwater (Smedley and Kinniburgh, 2002). Groundwater As enrichment is associated with either (1) regions of geologically young sediments with slow hydrogeological flow rates; (2) geothermal activity; or (3) ore deposits where sulfide-mineral oxidation has occurred (Smedley and Kinniburgh, 2002). In Canada, uncontaminated groundwaters and surface waters typically have 1 to 5 µg/L As; however, elevated As has been reported in the vicinity of many gold mining and roasting operations (Wang and Mulligan, 2006) as well as natural aquifers intercepting sulfide-mineralized geology (Boyle *et al.*, 1998).

3. Methods

3. Methods

3.1 Data sources and data processing

A search of publicly available geochemical datasets was conducted to compile rock, sediment, and water-quality analyses in the Dawson Range. These datasets were complemented by baseline environmental characterization data provided by Casino Mining Company (CMC) and Goldcorp (Casino Mining Company, 2014; Goldcorp, 2017) (Table 3-1).

**Table 3-1:
Overview of geochemical data compiled for use in this study**

	Total sample count	Sample count within study area	Notes
<i>Rock geochemistry</i>			
GSC Open File 8500	811	151	whole-rock analyses
Coffee Gold	460	460	aqua-regia with ICP-MS
Casino	464	464	aqua-regia with ICP-MS
<i>Sediment geochemistry</i>			
RGS Re-analysis 2016	20,411	365	aqua-regia with ICP-MS
<i>Surface-water quality</i>			
RGS 2003	28,721	457	no As data
Casino baseline	525	525	38 locations, data from 2008 to 2014
Coffee baseline	2,358	2,185	43 locations, data from 2010 to 2018
<i>Groundwater quality</i>			
Casino baseline	168	168	28 monitoring wells, data from 1994 to 2014
Coffee baseline	215	215	18 monitoring wells, from 2014 to 2018

Bedrock geological maps were compiled from Colpron *et al.* (2016), Godzicki *et al.* (2015), and Ryan *et al.* (2013). Several hundred whole-rock major and trace element analyses for the various Dawson Range lithologies were recently published by the Geological Survey of Canada (Ryan *et al.*, 2018). For the purpose of this study, geochemical data covering the geological units that outcrop within the study area were extracted from the dataset of Ryan *et al.* (2018) to define U and As abundance in local rocks. This dataset was complemented with geochemical data produced by the Coffee and Casino projects, which included >900 trace-element analyses by inductively coupled plasma mass spectrometry (ICP-MS) after aqua-regia digestion. Aqua regia digestion is a partial extraction that does not completely break down silicate minerals and other refractory phases, but can inform on the mobilization behavior of various elements under aggressive acidic and oxidizing

conditions. Mineralogy of the local rocks was assessed in the baseline characterization work at the Coffee and Casino projects (Casino Mining Company, 2014; Goldcorp, 2017). More detailed understanding of solid-phase U and As speciation is available through X-ray absorption near-edge spectroscopy (XANES), micro-X-ray fluorescence (μ -XRF), and Quantitative Evaluation of Materials by Scanning Electron Microscopy (QEMSCAN) analyses on a subset of samples from the Coffee site.

The Regional Geochemical Survey (RGS) provides a large number of stream sediment (>20,000) and surface water (>28,000) analyses for samples collected across Yukon by the Geological Survey of Canada between 1976 and 2006 as part of its National Geochemical Reconnaissance Program (Héon, 2003). Archived sediments were re-analyzed by ICP-MS after aqua-regia digestion between 2011 and 2016 (Yukon Geological Survey, 2016). Stream water chemistry in the RGS dataset includes U measurements by laser-induced fluorimetry on unfiltered water samples. RGS surface water analyses were merged with the more recent dissolved trace-element analyses from the Coffee and Casino projects that were conducted by inductively-coupled-plasma mass-spectrometry (ICP-MS) to provide a regional assessment of U and As. Comparison of total (unfiltered) and dissolved (filtered) U concentration data from the Casino and Coffee projects suggests these are generally comparable, justifying the use of unfiltered RGS U concentration data.

Groundwater analyses of samples collected between 1994 and 2014 from 28 monitoring wells at the Casino site (Figure 2-3) and between 2014 and 2018 from 18 monitoring wells at the Coffee site (Figure 2-2) were considered in this study. Groundwaters were analyzed for field parameters (*e.g.*, pH, electrical conductivity) and a suite of laboratory parameters including alkalinity, major anions, and trace elements by ICP-MS. Groundwater analyses that had > 5 % charge balance errors (as calculated in the geochemical modelling program PHREEQC) were discarded.

Data processing involved the construction of a database compiling all water-quality analyses from the various data sources. For the purpose of statistical and geospatial analyses, all replicate sample analyses were removed and values that were below reported detection limits (RDL) were converted to half of the RDL. Geospatial analysis of surface water and sediment chemistry was conducted to identify hotspots of U and As enrichment through interpolation using ordinary spherical kriging with ArcMap software (version 10.6, ESRI, Redlands CA). Because U concentrations are seasonally sensitive to dilution, the maximum U concentration at each sampling location was used as input in the kriging computation.

Calculation of mineral saturation indices and uranium speciation was completed using PHREEQC with the wateq4f.dat database (Parkhurst and Appelo, 2013). The association constants for the species $\text{Ca}_2(\text{UO}_2)(\text{CO}_3)_3$ and $\text{Ca}(\text{UO}_2)(\text{CO}_3)_3^{2-}$ were added to the database prior to simulations (Stewart *et al.*, 2010).

4. Results

4. Results

4.1 Geological occurrence of U and As in the Dawson Range

4.1.1 Uranium in rock

Bedrock geochemical analyses indicate that Dawson Range rocks have U content that is close to the average upper crustal abundance (AUC) value of 2.7 µg/g (Table 4-1). Whole-rock analyses in Open File (OF) 8500 data have a median U content of 2.1 µg/g and a maximum value of 19 µg/g. In contrast, aqua-regia digestions of rock samples from the Coffee and Casino projects released higher U (medians 3.5 and 3.7 µg/g, respectively; maximums 53 and 75 µg/g, respectively) (Figure 4-1). Within the geological units present in the Dawson Range, OF 8500 data indicate that four have a median U content above the AUC: Rhyolite Creek complex; Casino suite; Whitehorse suite; and Sulphur Creek suite. These units are all composed of igneous rocks or igneous protoliths and dominated by felsic to intermediate compositions. The Whitehorse and Sulphur Creek suites also account for 43 % of bedrock geology in the study area (Figure 4-2). Spatially, rock samples with U > AUC are consistently observed along Whitehorse and Sulphur Creek occurrences throughout the region, with the many U-rich samples concentrated in units correlating to Sulphur Creek Suite, Whitehorse Suite, and Klondike Schist lithologies bordering the Yukon River (Figure 4-3).

Uranium enrichment is strongest in Dawson Range rocks of felsic and intermediate composition, while mafic and ultramafic rocks have lower U, consistent with the incompatible behavior of U in magmas. Magmatic U enrichment is further shown by increasing U content with Zr/TiO₂ and (Na+K)/Al (Figure 4-4). Zr⁴⁺ is an incompatible element relative to Ti⁴⁺, and (Na+K)/Al indicates the degree of silicate-chain polymerization in melts; at high (Na+K)/Al there is less silicate polymerization, which enhances the ability of U to be incorporated into the solid phase during magmatic cooling (Kyser, 2014).

Table 4-1:
Summary statistics of U and As content in Dawson Range rocks

Dataset	U (µg/g)						As (µg/g)					
	Upper continental crust* = 2.7 µg/g						Upper continental crust* = 4.8 µg/g					
	5 th percentile	95 th percentile	max	median	mean	n	5 th percentile	95 th percentile	max	median	mean	n
Open File 8500 [†]	0.1	6.8	19	2.1	3	651	0.3	7.2	1,236	1.5	5	485
Coffee	0.7	18	53	3.5	5	459	3.2	2,091	>10,000	39	393	459
Casino	0.6	10	75	3.7	4	464	1.7	56	389	8.0	16	464

Notes:

*Upper continental crust estimate from Rudnick and Gao (2014)

[†]Includes only Open File 8500 data for geologic units that are present within the study area

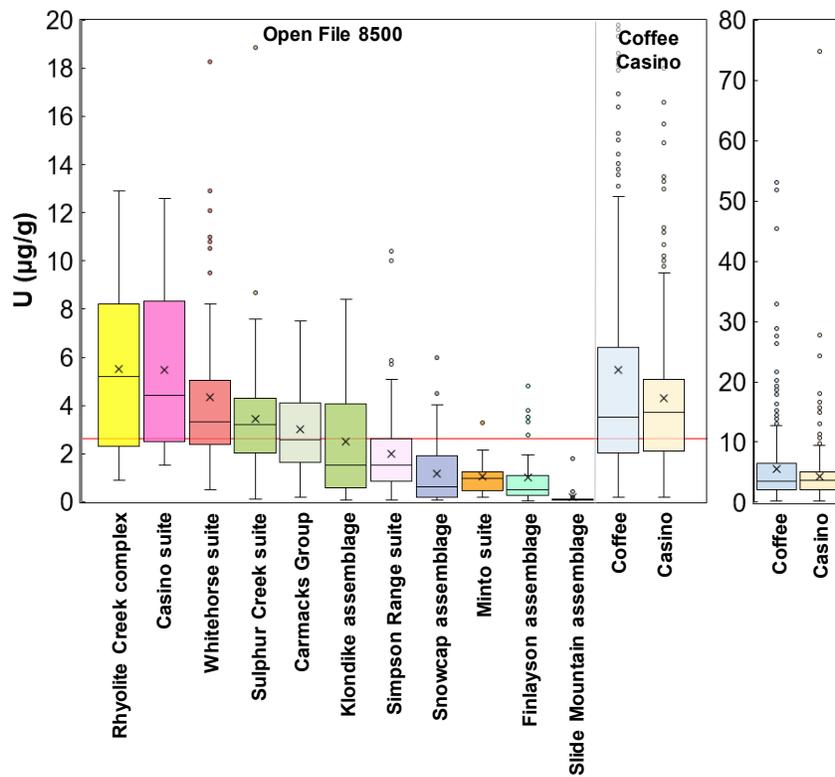


Figure 4-1: Uranium content for the different geological units present in the Dawson Range and in Coffee and Casino static testing databases. The full extent of data from Coffee and Casino is shown in the right-hand panel, which has an extended y-axis range. Data for geological units with less than 10 sample analyses are not shown. The red horizontal line in the left panel indicates average crustal abundance (2.7 µg/g).

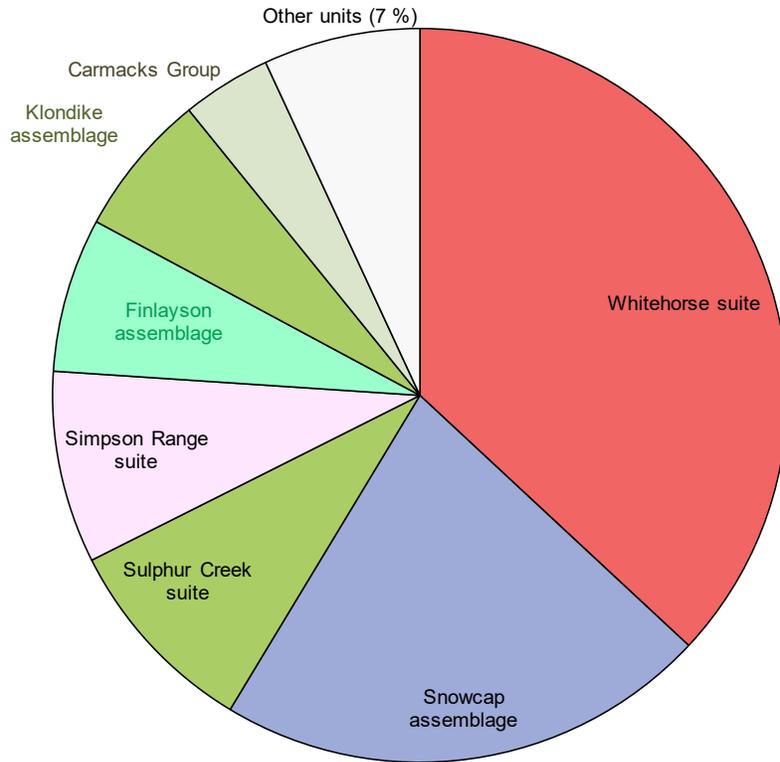


Figure 4-2: Relative distribution of geological units within the study area (as defined in Section 2.1.)

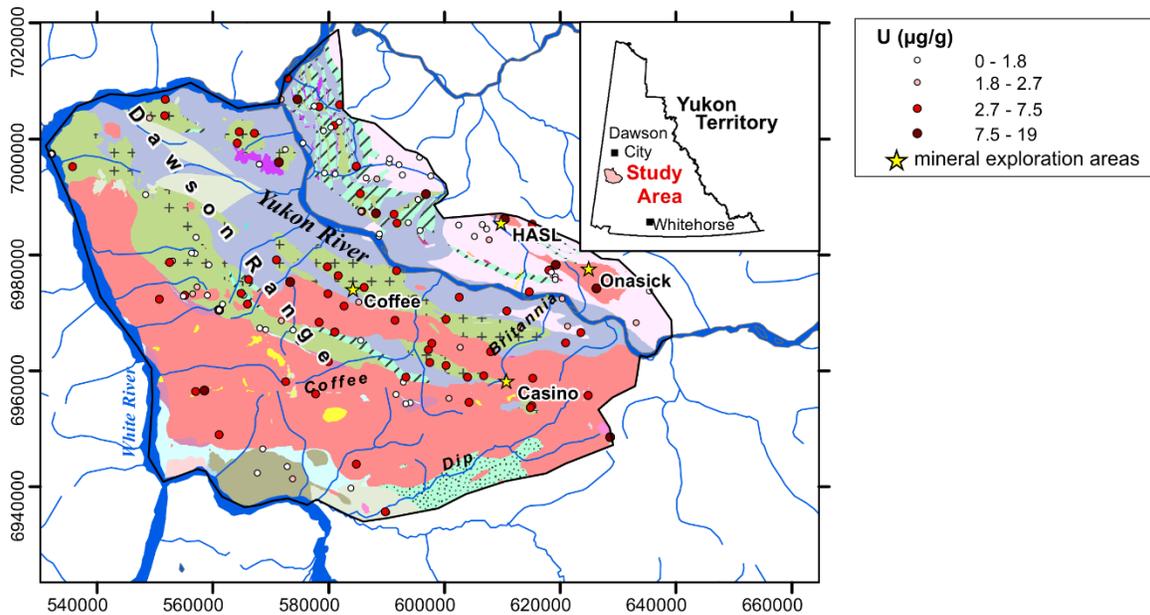


Figure 4-3: Spatial distribution of U in rocks with bedrock geology in the background. Data from GSC Open File 8500.

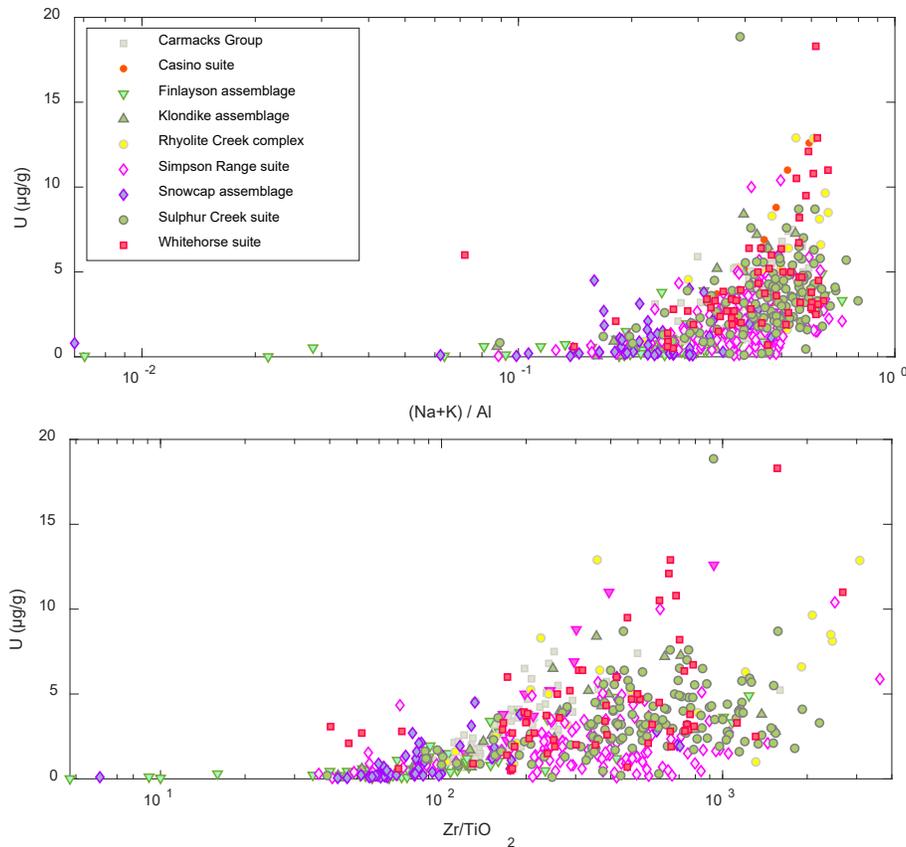


Figure 4-4: Enrichment of U through magmatic processes in Dawson Range rocks, as seen through positive correlation with Zr/TiO_2 and $(Na+K)/Al$. Data from GSC Open File 8500.

Precise identification of U mineralogy in Dawson Range rocks is hindered by the low U content relative to mineralogical instrumental detection limits. Available data indicates that U is present through element substitution in accessory minerals, and may also be present in U-forming minerals. Electron microprobe analyses of four samples from the Coffee project showed up to 10,000 $\mu\text{g/g}$ in zircon (Goldcorp, 2017). However, this pool of U is not expected to be mobile given the high resistance of zircon to chemical weathering. Apatite, another common U-bearing mineral, did not contain U above detection limits in these samples. Rutile and anatase, two polymorphs of TiO_2 , were frequently observed at low levels by X-ray diffraction (XRD) (Goldcorp, 2017) and these phases can also incorporate U through cation substitution (Luvizotto *et al.*, 2009), although no U content data are available for rutile in Dawson Range samples. No distinct U minerals could be directly identified by powder X-ray diffraction and synchrotron-based powder XRD. However, μ -XRF analyses were used to identify elemental associations of U: spatial overlap of U with Fe, Ca, and Cu were observed in a majority of samples analyzed by μ -XRF, and overlap with Th, Ti, and As was observed in select samples (Goldcorp, 2017).

The spatial overlap of U and Fe may indicate association of U with Fe-oxides through UO_2^{2+} sorption, while associations of U with Ti and Th in gneiss samples may indicate the presence of brannerite and thorite. The oxidation state of U in gneiss and schist samples from the Coffee project includes both U(VI) and U(IV), although samples from the oxidized portion of the deposit were depleted in U(IV) (Figure 4-5). The presence of U(VI) in all rock samples suggests U mobilization does not require oxidation of the rock but may simply arise through water-rock contact.

The distinct weathering facies at the Coffee and Casino deposits translate into different U mobilization behaviors. The oxide leach cap at Casino is relatively depleted in U, carbonates, and sulfides relative to subjacent supergene and hypogene zones (Figure 4-6). In contrast, there is no observable U depletion in the oxidized facies at the Coffee site, despite relatively lower sulfide and carbonate content in comparison to the transition and sulfide facies at Coffee (Figure 4-6).

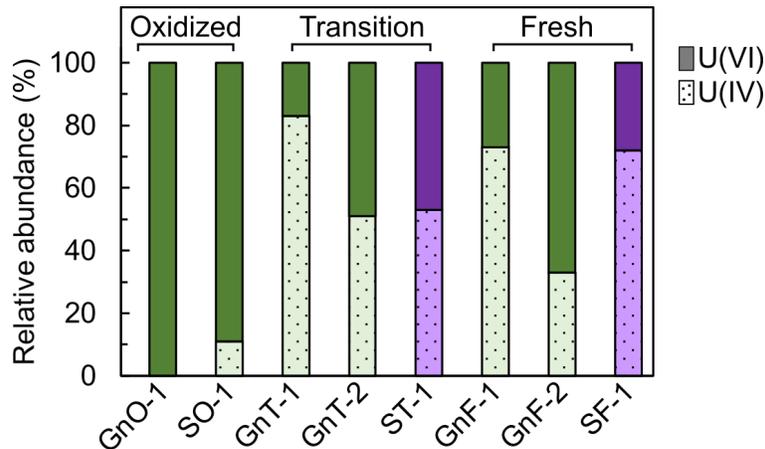


Figure 4-5: Uranium oxidation state in rock samples from the different weathering facies at the Coffee project determined by XANES. Green colors = gneiss; purple colors = schist.

4.1.2 Uranium in sediment

Sediment samples in the region frequently have elevated U, with 36 % of the 364 samples having $U > AUC$. Sediments often have higher trace-metal content than underlying bedrock because of accumulation of U through sorption onto fines or U accumulation in organic-rich sediments, making sediment data useful vectors of U source (Yao *et al.*, 2015). Consistent with whole-rock analyses, the most U-rich sediment is found near the Yukon River, with a noteworthy hotspot around an outcrop of Whitehorse suite granite southeast of the HASL property. Localized elevated U in sediment is also observed near the Coffee and Casino properties, along with the intervening drainages (Excelsior and Casino Creeks) (Figure 4-7).

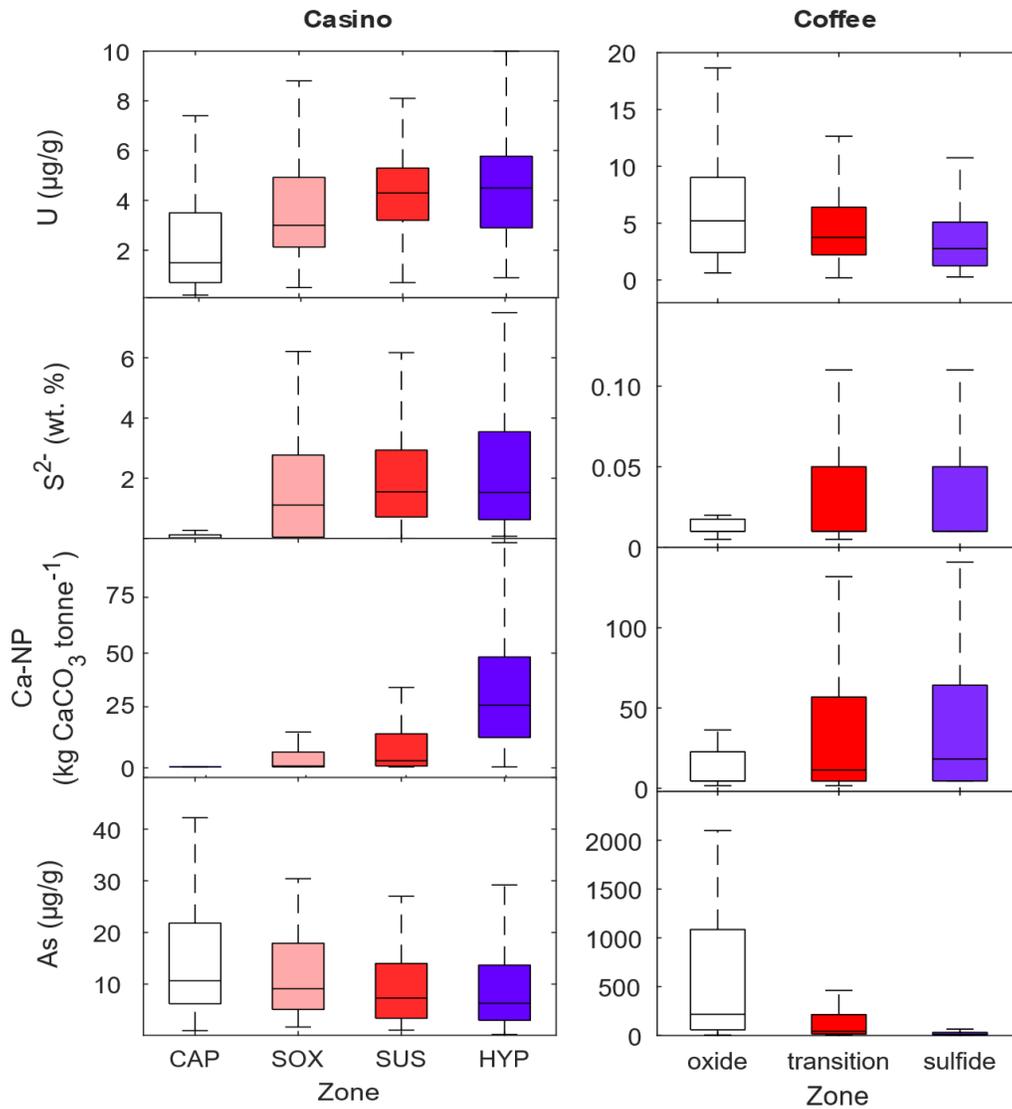


Figure 4-6: Uranium, sulfide, carbonate neutralization potential (Ca-NP) and arsenic content in Casino and Coffee drill-core samples by weathering facies. At Casino, CAP = oxide cap, SOX = supergene oxide, SUS = supergene sulfide, HYP = hypogene; at Coffee: weathering zones are defined as “oxide”, “transition”, or “sulfide”. Note that y-axis scales are different between left and right panels.

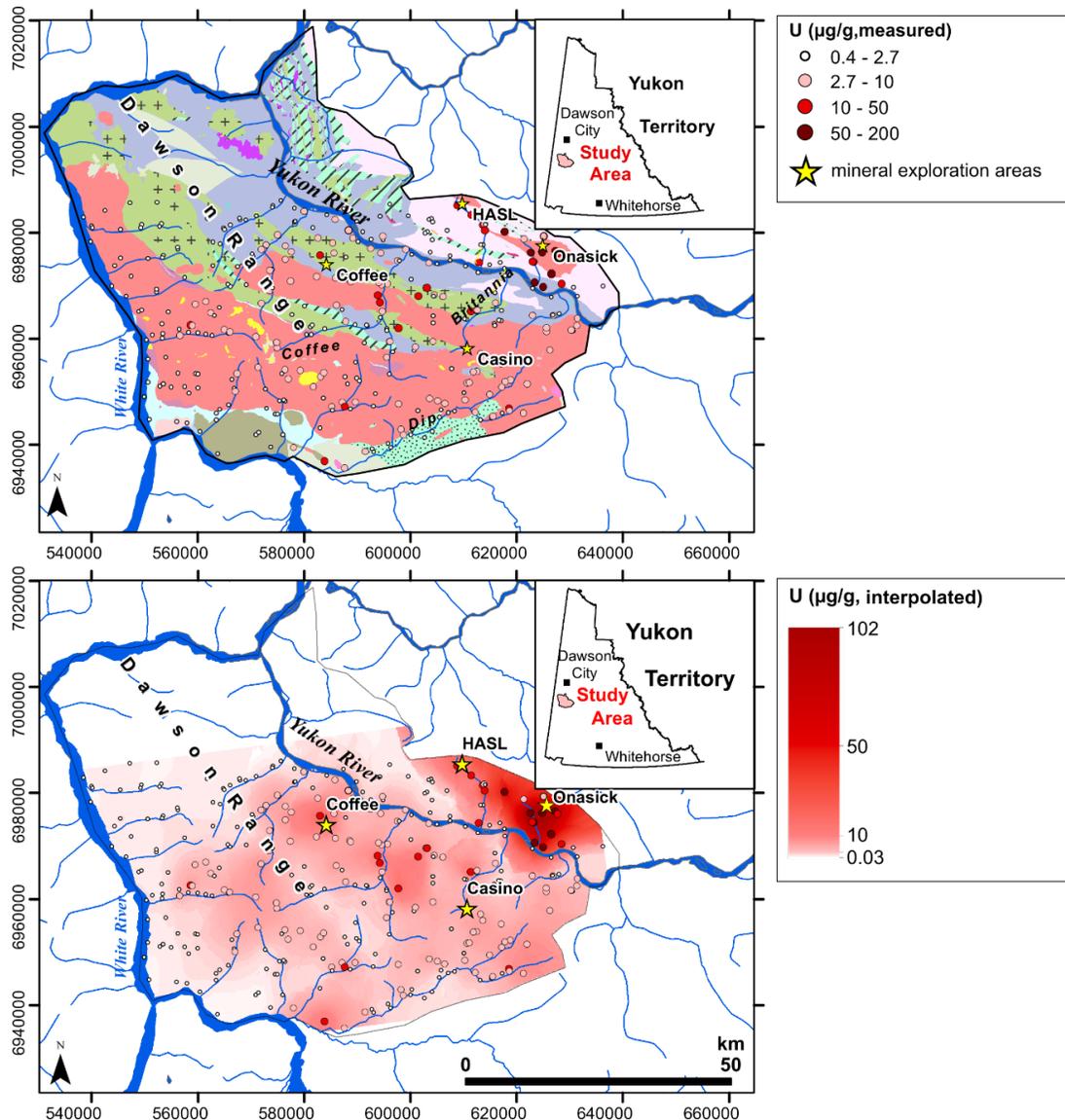


Figure 4-7: Spatial distribution of U in sediment samples, showing measured U content and bedrock geology with measured and interpolated U.

4.1.3 Arsenic in rock

Geochemical analyses of As abundance in Dawson Range show notable differences between regional whole-rock analyses in Open File 8500 in comparison to rock samples collected at the Coffee and Casino sites (Figure 4-9). Samples from Open File 8500 have a median As content of 1.5 µg/g, which is below the AUC value of 4.8 µg/g (Table 4-1). In contrast, As content is considerably higher in rock at the Casino (median 8.0 µg/g, maximum 389 µg/g) and highest at Coffee (median 38 µg/g, maximum 7,940 µg/g).

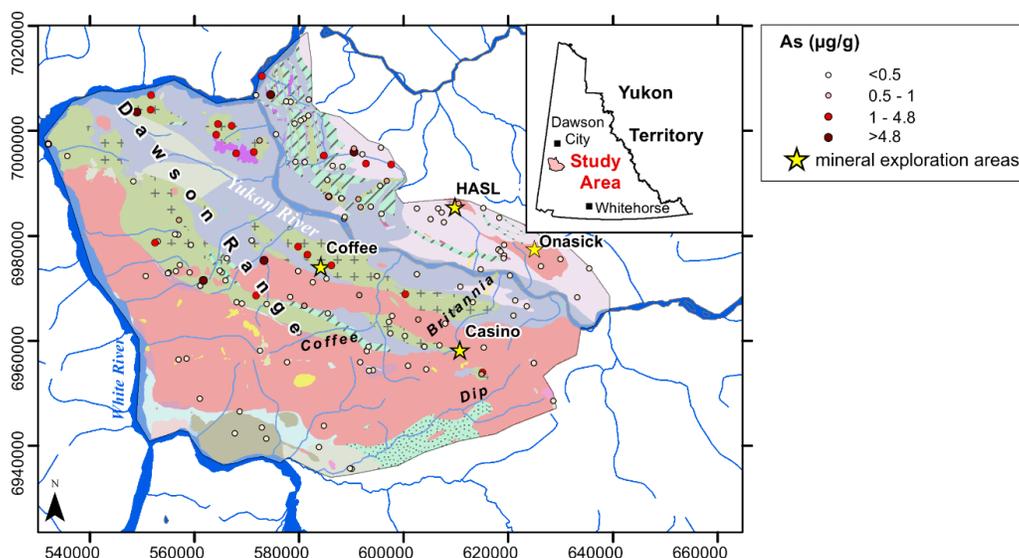


Figure 4-8: Spatial distribution of As in whole-rock with bedrock geology in the background.

Open File 8500 samples collected within the study area show localized As-rich samples spatially overlapping with the Sulphur Creek gneiss (Figure 4-8); however, the Sulphur Creek gneiss overall has relatively low As content suggesting heterogeneity in the As distribution in this geological unit (Figure 4-9). Carmacks Group volcanic rocks are the only geologic unit containing a median As > AUC. Localized samples with extremely high As content (up to 1,236 µg/g) in the Open File 8500 dataset are sporadically distributed and without clear relationship to bedrock geology.

Arsenic occurrence is strongly tied to the presence of sulfide and Fe-oxide minerals (derived from sulfide-mineral oxidation) in the rock. Distinct As-bearing sulfides including arsenopyrite and arsenian pyrite are present at Coffee (Goldcorp, 2017). Arsenian pyrite is commonly observed at Coffee and Casino; and electron microprobe analyses at Coffee show that this phase can contain up to 76,000 µg/g (Table 4-2). Oxidation of As-bearing sulfides leads to association of As to oxides and Fe-(oxyhydr)oxides (Figure 4-10). Scorodite (FeAsO₄) was observed in select rock samples from the oxidized and transition facies at Coffee (Goldcorp, 2017). Evidence for oxidative dissolution of As-bearing sulfides is observed in the redox speciation of As in rock samples from Coffee, where rock collected in the fresh weathering facies contains As(III) and As(-I) while rock collected in the oxidized and transition facies are dominated by As(V) (Figure 4-11). In contrast to U, depletion of As from the oxidized weathering facies at Coffee and Casino is not observed; in fact, median As contents are highest in more oxidized portions of these deposits (Figure 4-6).

Table 4-2:
Median and maximum As content in (arseno)pyrite grains from rock samples at the Coffee site determined by electron microprobe analysis.

	median	max	number of	lithology
Sample ID	µg/g	µg/g	spots	
GT	3,967	76,300	25	gneiss
ST	68	4,135	15	schist
SS	31	17,500	21	schist

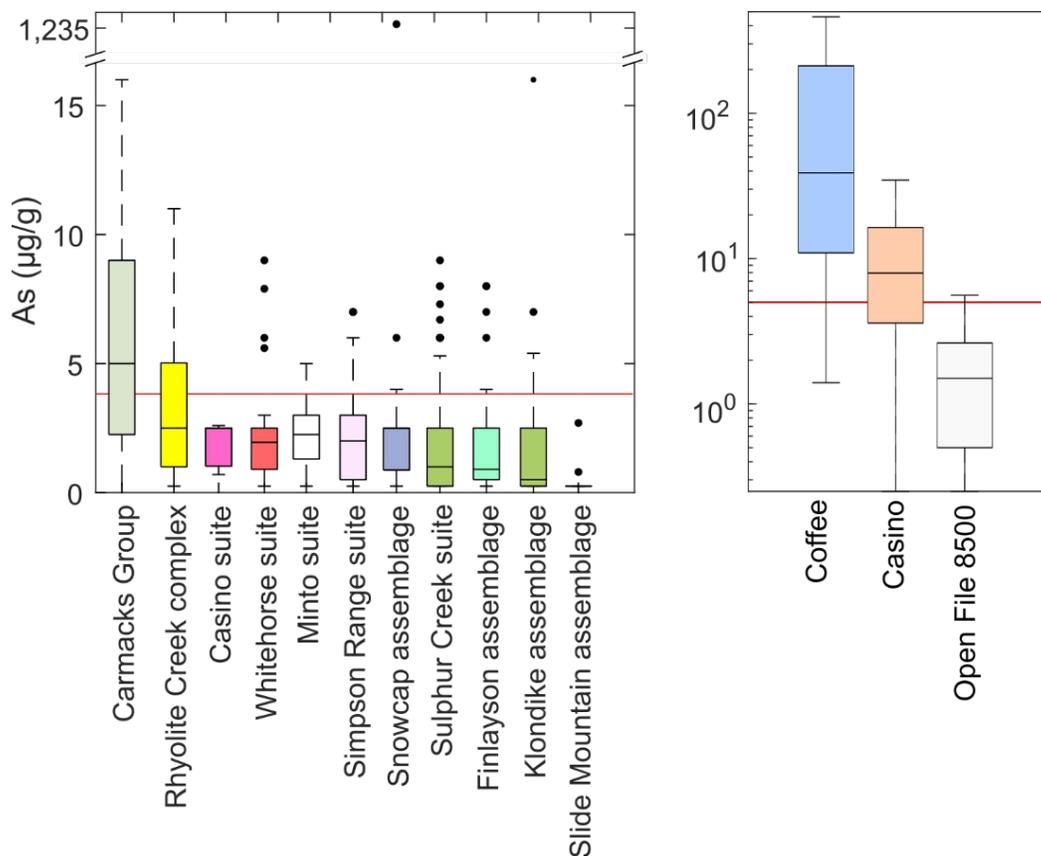


Figure 4-9: Boxplot of As content by selected geological units in Open File 8500, and As content in Coffee and Casino drill-core. Horizontal red line indicates average upper continental crust value. Note break in y-axis scale in left panel and difference in y-axis scales between the two panels.

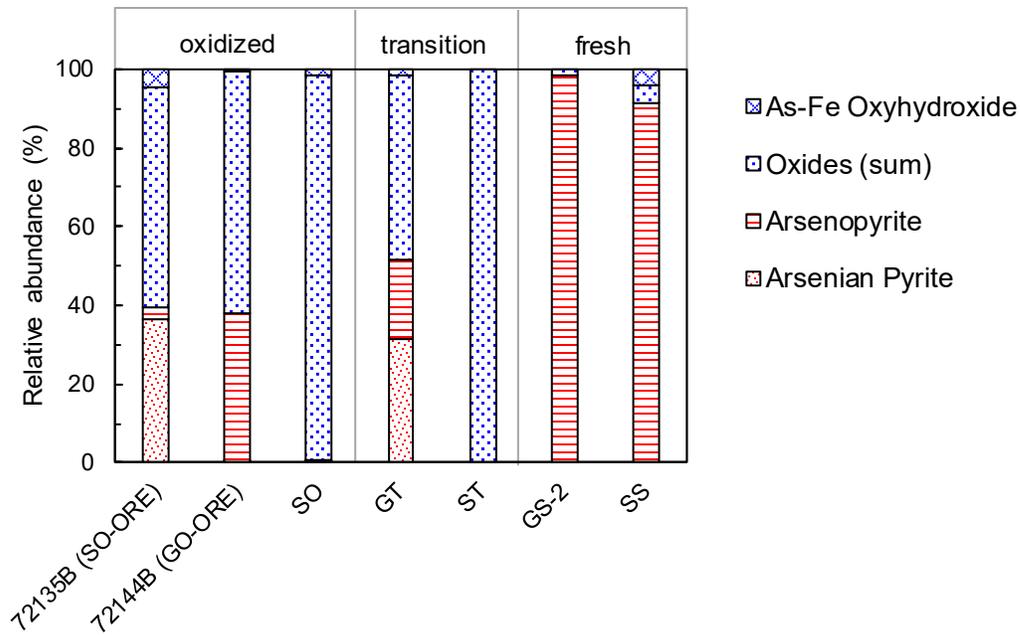


Figure 4-10: Mineralogical distribution of As in rock samples from the Coffee site determined by quantitative evaluation of materials by scanning electron microscopy (QEMSCAN) in samples from the Coffee site. Red colors = primary sulfides; blue colors = secondary oxides.

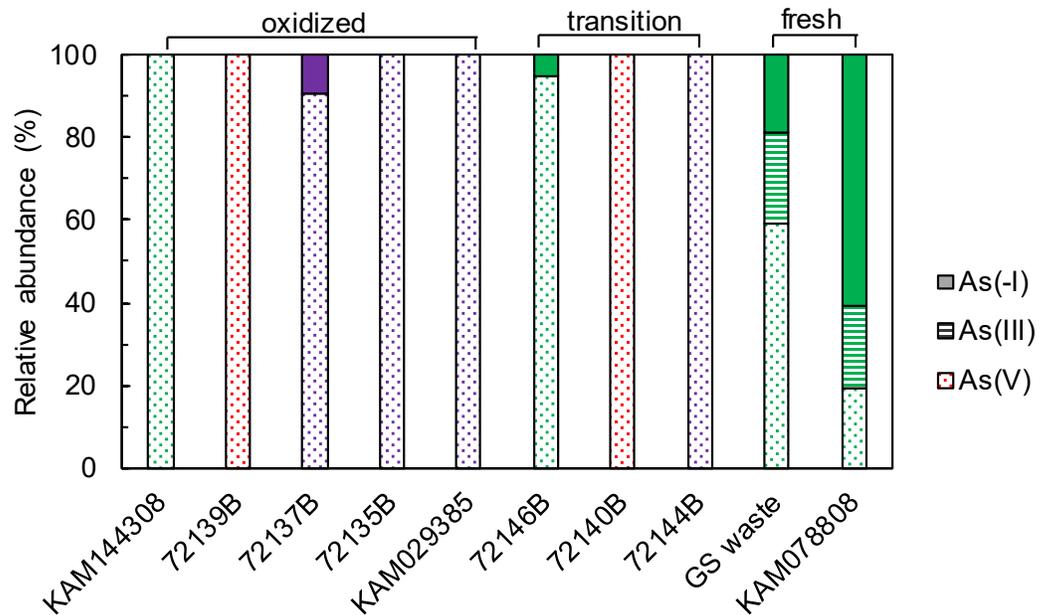


Figure 4-11: Arsenic redox speciation determined by XANES in rock samples from the Coffee project. Green = gneiss; red = granite; purple = schist.

4.1.4 Arsenic in sediment

The median As content of sediment samples within the study area is 5.2 µg/g, with a maximum value of 157 µg/g. Arsenic distribution in sediment generally follows a NW-SE trend, with lower As in sediment overlying the Whitehorse suite intrusives in the SW of the region and Simpson Range and Whitehorse suite rocks on the NE bank of the Yukon river, while sediment with more elevated As is concentrated around the Coffee and Casino deposits (Figure 4-12). Sediments with As content > 10 µg/g are frequently found in creek drainages surrounding these mineralized areas, including Independence, Halfway, Coffee, Excelsior, Britannia, and Casino Creeks (Figure 4-12).

4.2 Natural occurrence of uranium and arsenic in water

4.2.1 Uranium is naturally present at concentrations above CCME guidelines in surface water

Territorial-scale mapping of the >28,000 surface-water analyses conducted during the RGS shows that the Dawson Range is one of a handful of regions in Yukon where U approaches and exceed CCME guidelines for the protection of aquatic life (15 µg/L) (Figure 4-13). Within the Study Area, U enrichment in surface water appears concentrated in a NW-SE trending region along the Yukon River that encompasses the Coffee, Casino, HASL, and Onasick properties (Figure 4-14). The median U concentration calculated at each of the 535 surface-water sampling locations available in the dataset within the Study Area exceeds 15 µg/L in 5.4 % of locations. In comparison, only 0.4 % of the 28,721 water samples analyzed for U across Yukon in the RGS have U > 15 µg/L. While many U-rich (>15 µg/L) surface waters are found in proximity to the Coffee and Casino deposits, high-U water is also found in other nearby drainages, such as Independence Creek, Excelsior Creek, Pedlar Creek, and an unnamed drainage E of Pedlar Creek (Figure 4-12).

To identify relationships of U-rich surface water with other aqueous geochemistry parameters, the proportion of samples having U concentrations above the 15 µg/L CCME guideline for the protection of aquatic life was calculated over different intervals of other parameters (a method proposed by Riedel and Kübeck, 2018). Calculations show surface-water U relationships with alkalinity, electrical conductivity, dissolved Ca, and pH (Figure 4-15). In samples with alkalinity concentrations exceeding 150 mg/L (as CaCO₃) and Ca concentrations exceeding 80 mg/L, U concentrations are above 15 µg/L in 80 % of analyses. More than half of surface water samples with electrical conductivity above 600 µS/cm also have U concentrations greater than the CCME guideline. The parabolic relationship of U and pH shows that U concentrations are maximal in circumneutral to alkaline pH and highly acidic surface waters, while U concentrations are lowest in the pH 4.5 to 6.5 range (Figure 4-15).

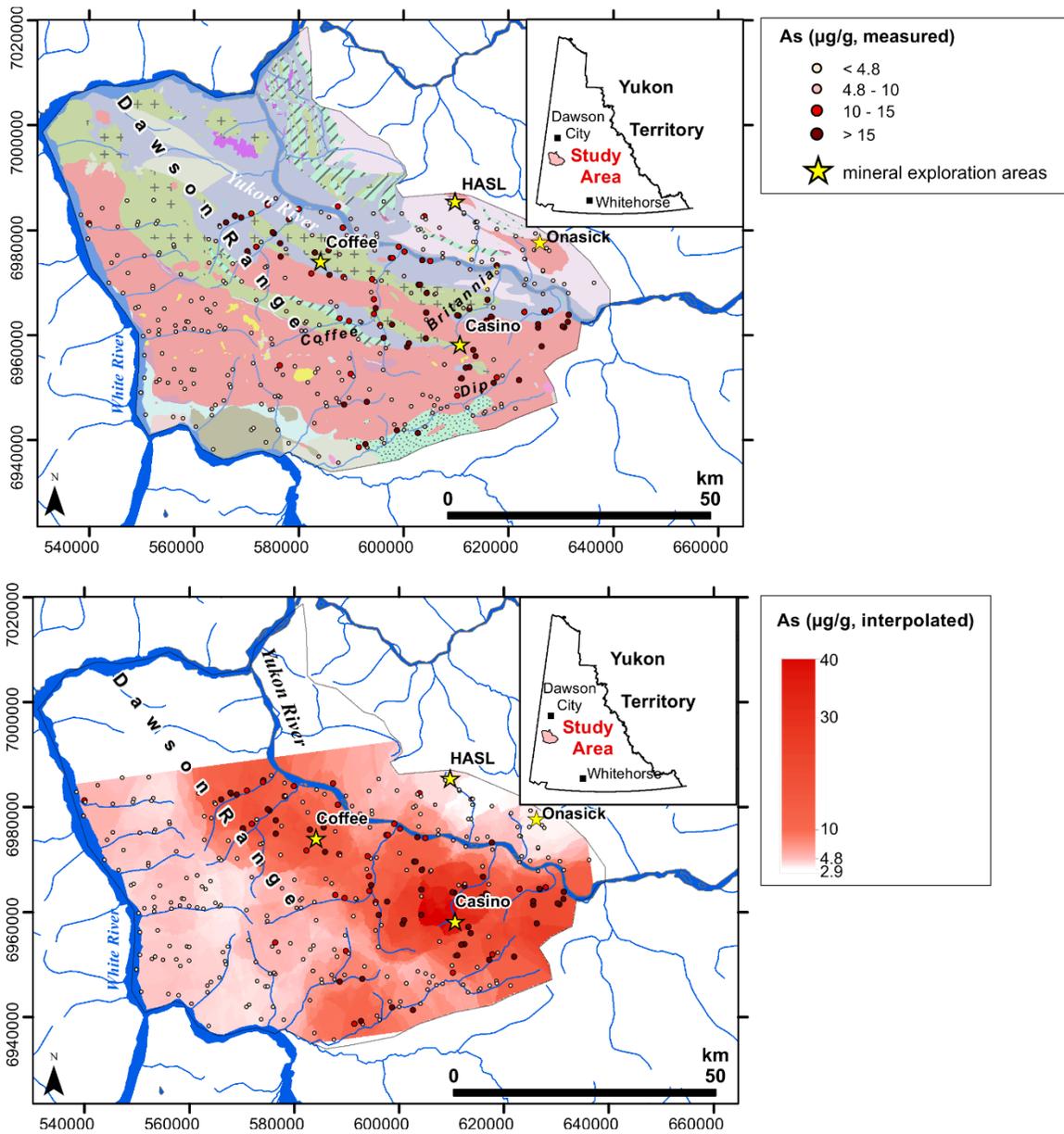


Figure 4-12: Spatial distribution of As in sediment samples, showing measured As content and bedrock geology (above) and measured and interpolated As (below).

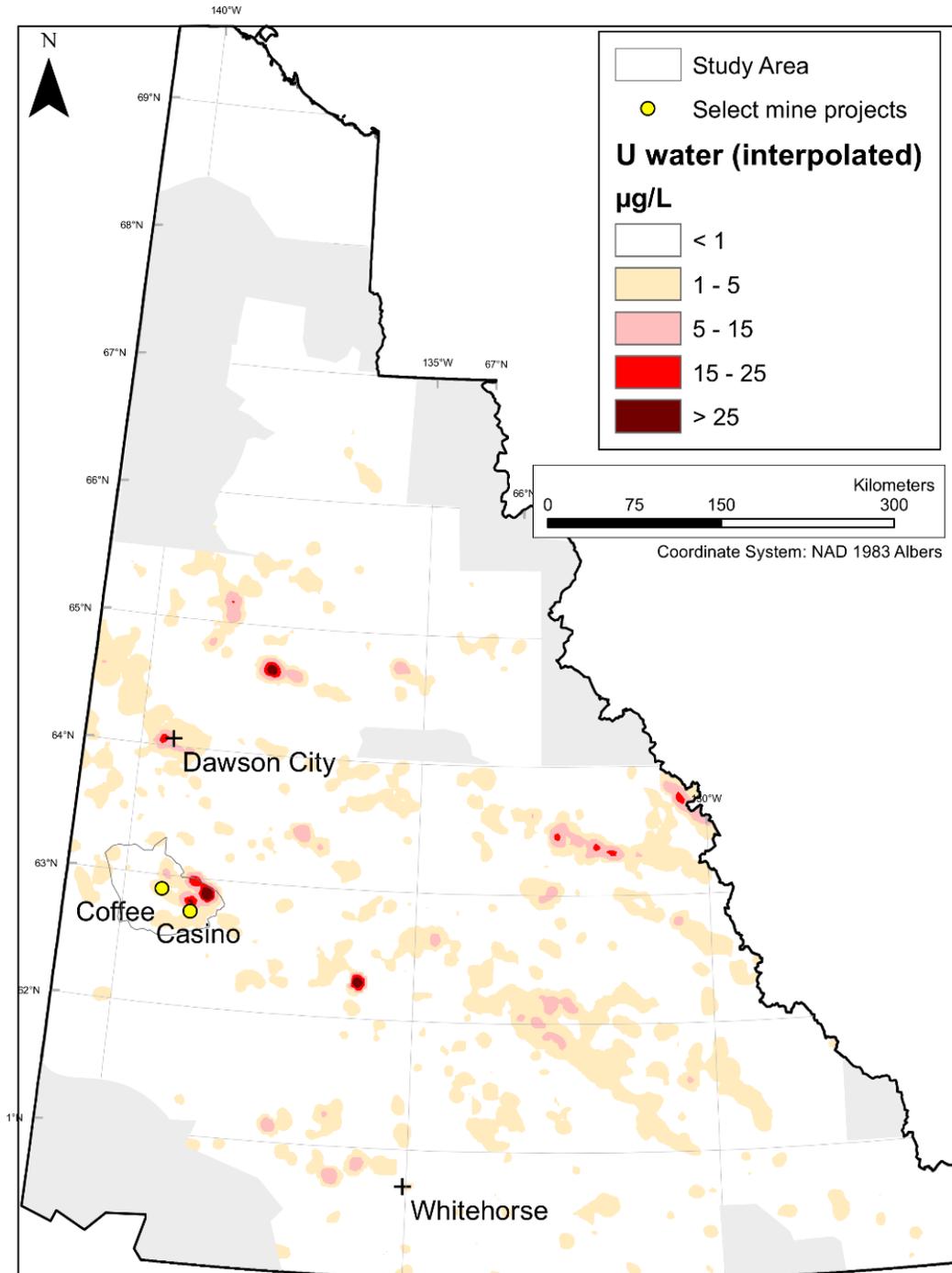


Figure 4-13: Uranium concentrations surface water at the territorial scale, interpolated from the RGS dataset. Areas with gray shading do not have samples available in that dataset.

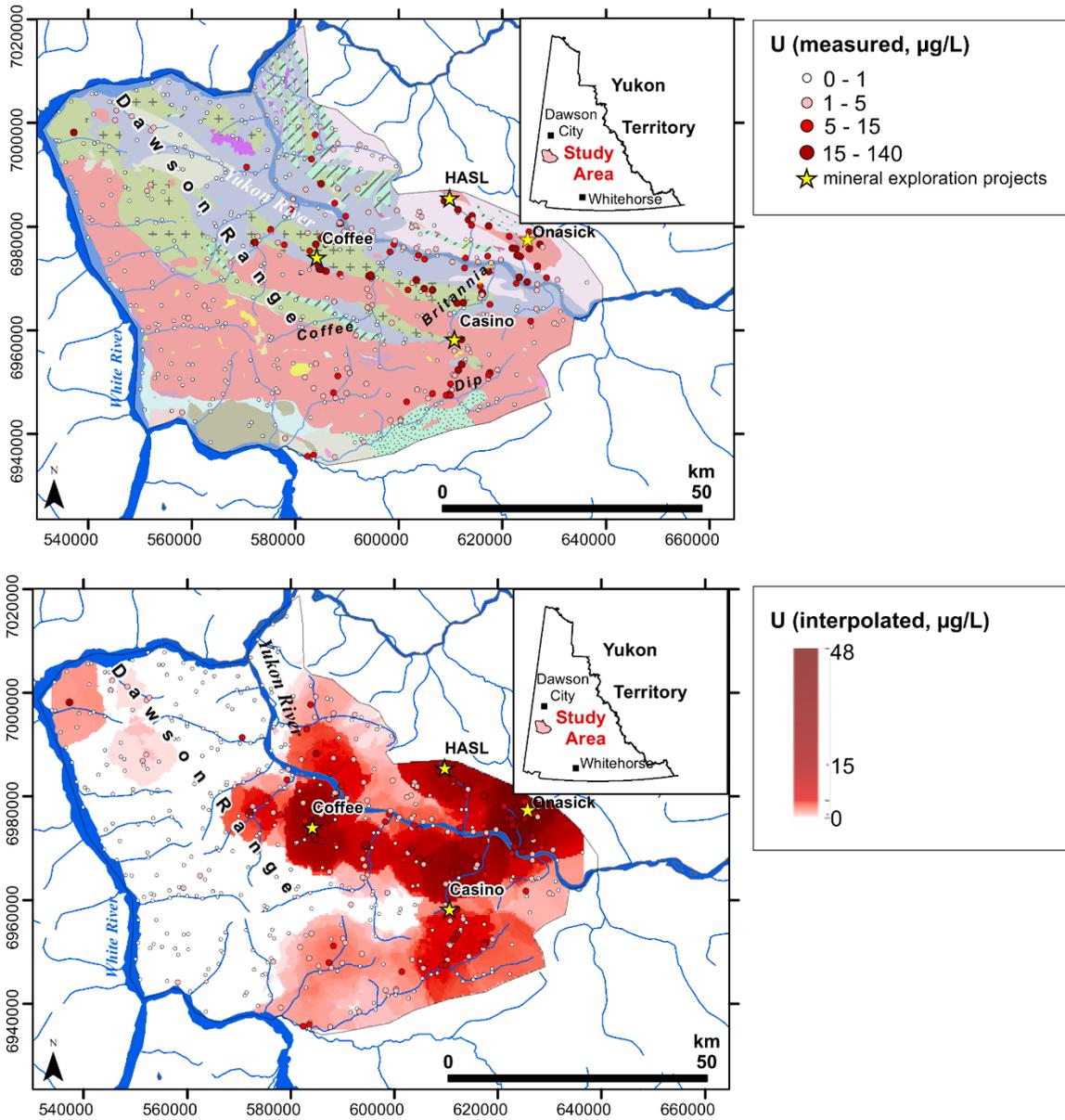


Figure 4-14: Spatial distribution of U in surface water samples, showing measured U concentrations and bedrock geology (above) and measured and interpolated U (below). For sampling locations with multiple samples, the maximum U concentration is shown and was used in interpolations to emphasize U hotspots.

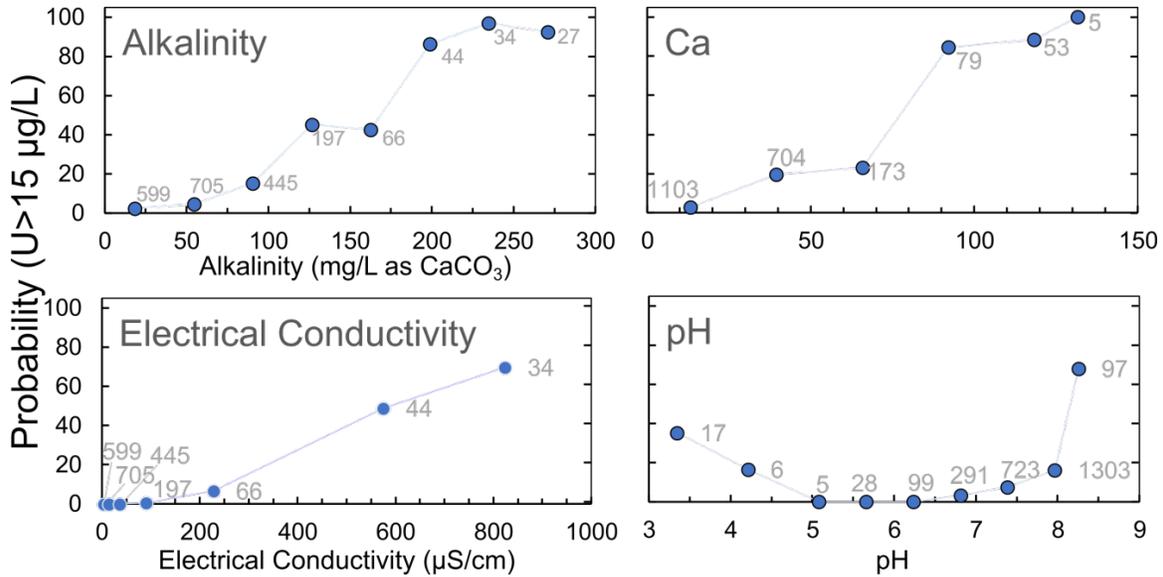


Figure 4-15: Probability of having U concentrations above the CCME guideline in surface water calculated over different ranges in alkalinity, Ca, electrical conductivity, and pH. Data labels in grey font correspond to the number of water-sample analyses used to calculate each probability value.

There is a strong inverse relationship of U and flow at surface-water monitoring locations around the Coffee site where regular flow monitoring has occurred and where U concentrations frequently exceed 15 µg/L (Figure 4-16). These relationships are most visible in upper Latte, Coffee, Halfway, and Independence Creeks, and also correspond to water-quality monitoring locations in creeks draining rocks from the Whitehorse Suite intrusive or Sulphur Creek metamorphic geologic units. In contrast, monitoring stations in Independence Creek which are dominated by Klondike or Snowcap assemblage rocks (*i.e.*, IC-0.5 and IC-3.0) have lower U concentrations over a wide range in flows (Figure 4-16). Downstream of U-rich surface water locations, the U-flow relationship also breaks down, suggesting that there are localized areas of U-rich groundwater discharge into creeks but that creek U concentrations then decrease through dilution by runoff and U-poor groundwater discharge along the stream course. While flow data are not available at the Casino site, the U concentrations there are also typically higher during winter baseflow conditions (Figure 4-16), further suggesting groundwater discharge as the source of U loading to Dawson Range creeks.

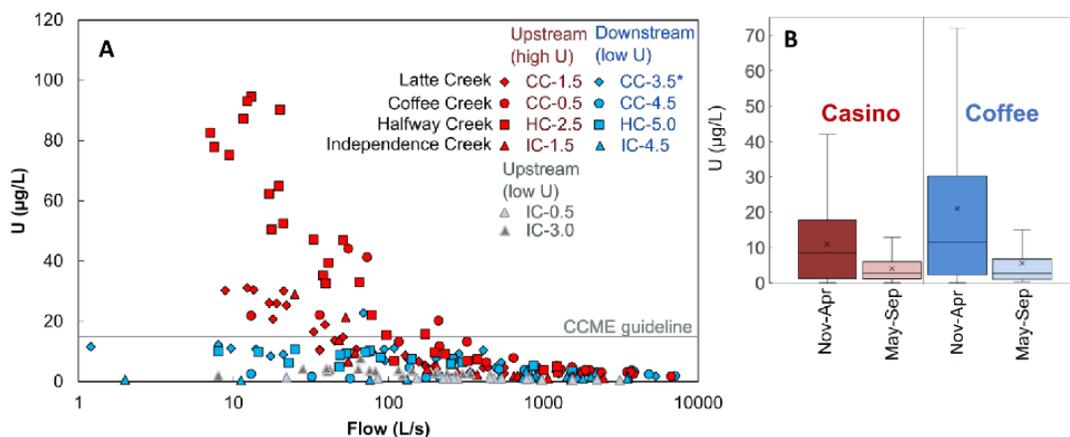


Figure 4-16: Relationship between U concentrations and spot measurements of flow in select drainages around the Coffee site. (A) A subset of upstream locations with high U concentrations also show strong inverse relationships between U and flow, while downstream locations typically have lower U concentrations that are independent of flow. (B) Comparison of U concentrations in water baseflow (November-April) vs. snow-free (May-September) conditions in creeks at the Casino and Coffee sites showing high U concentrations under low flow regimes.

4.2.2 Groundwater uranium concentrations frequently exceed CCME guidelines

Groundwater U enrichment is even greater than in surface water (Figure 4-17): of the 46 monitoring wells at Coffee and Casino, 39 % have median U concentrations > 15 µg/L. The median U concentration over the entire dataset is 17 µg/L. To account for wells that have been sampled multiple times, a median groundwater concentration was determined at each well. The overall median of this dataset (*i.e.*, of median U concentrations by well location) is 10 µg/L, with a tendency towards higher U in wells near the Coffee site (median 29 µg/L, maximum 589 µg/L) in comparison to those at Casino (median 7 µg/L, maximum 101 µg/L).

Uranium-rich groundwater is associated with elevated total dissolved solids (TDS), alkalinity, Ca, and sulfate (Figure 4-20). Accordingly, more widespread U-rich groundwater is found at the relatively more carbonate-rich Coffee property, where 11 out of 15 groundwater monitoring wells have median U concentrations > 15 µg/L (Figure 4-18) and where circumneutral to basic pH (median pH 6.9 to 8.0) is encountered in groundwater. As a result of the presence of alkalinity and dissolved Ca, U aqueous speciation at the Coffee site is therefore dominated by $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3$ and $\text{Ca}_2\text{UO}_2(\text{CO}_3)_2^{2-}$; species which decrease the ability U(VI)O_2^{2+} to adsorb and to undergo reduction to insoluble U(IV) (Stewart *et al.*, 2010; Stewart *et al.*, 2011).

Bedrock lithology and redox conditions influence U occurrence in groundwater at Coffee. Of the 8 monitoring wells screened in schist or gneiss, only one has a median U concentration $< 15 \mu\text{g/L}$ (MW15-03 WB). In contrast, three out of four wells screened in granite at Coffee have a median U $< 15 \mu\text{g/L}$. The lower-U granitic wells are also characterized by FeS-saturated or supersaturated conditions suggesting Fe(III) and sulfate-reducing conditions. Similarly, MW15-06WB, screened in gneiss, holds relatively low-U relative compared to other gneiss wells at the site and also has FeS-supersaturated water, while the majority of U-rich monitoring wells at Coffee have FeS-undersaturated conditions (Figure 4-18). As biogenic U(VI) reduction can be driven by iron- and sulfate-reducing bacteria (Abdelouas *et al.*, 2000), Fe(III) and sulfate-reducing conditions in these waters may indicate conditions favoring U(VI) reduction.

At the Casino site, there is proportionally less U-rich groundwater in comparison to Coffee: only 7 out of 28 monitoring wells at Casino have median U concentrations above the CCME guideline (Figure 4-19). Accordingly, the majority of groundwater wells at Casino are screened in granitic rock, with only two wells being screened in gneiss (94-346 and 34-345, with median U concentrations of $45 \mu\text{g/L}$ and $12 \mu\text{g/L}$, respectively).

Uranium speciation in the majority of groundwater at Casino is overwhelmingly dominated by Ca-UO₂-CO₃ complexes (Figure 4-18), similar to Coffee groundwater. However, a distinct U speciation is present in the two most U-poor wells (MW13-02D and 94-337, Figure 4-18). These wells have median U concentrations $\leq 1 \mu\text{g/L}$ and are generally characterized by low Ca ($< 30 \text{ mg/L}$) and low alkalinity ($< 30 \text{ mg/L}$). Well 94-337 preserves a unique signature dominated by sulfide oxidation as indicated by its Ca-SO₄ water type and relatively low pH (median pH 5.7), which promotes the presence of other U complexes including UO₂²⁺, UO₂F⁺ and UO₂F₂, UO₂SO₄, and UO₂OH⁺ that are otherwise absent in higher-pH Dawson Range groundwater.

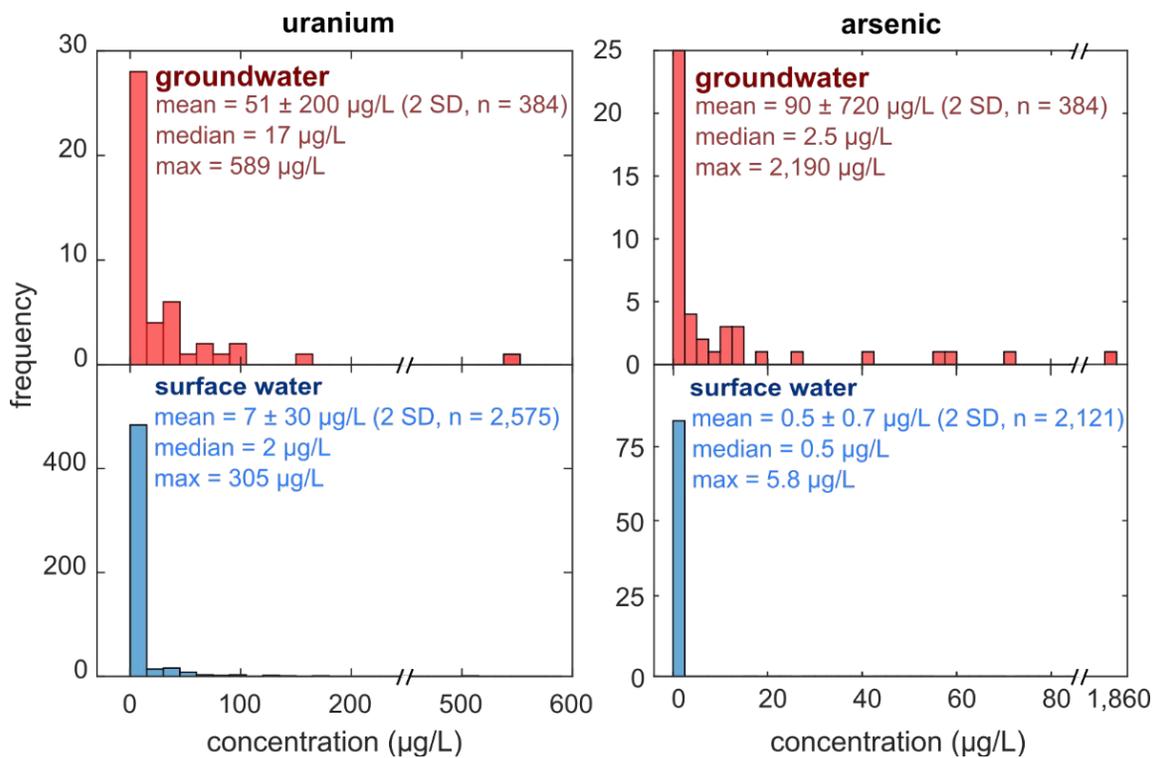


Figure 4-17: Uranium and arsenic frequency-distribution diagrams for groundwater and surface water data. Histograms show the frequency distribution of median U (above left) and As (above right) concentrations for each groundwater sampling location and maximum U (below left) and As (below right) concentrations for each surface water sampling location. Maximum concentrations are used for surface water to highlight U enrichment given the inverse relationship of U and flow in creeks.

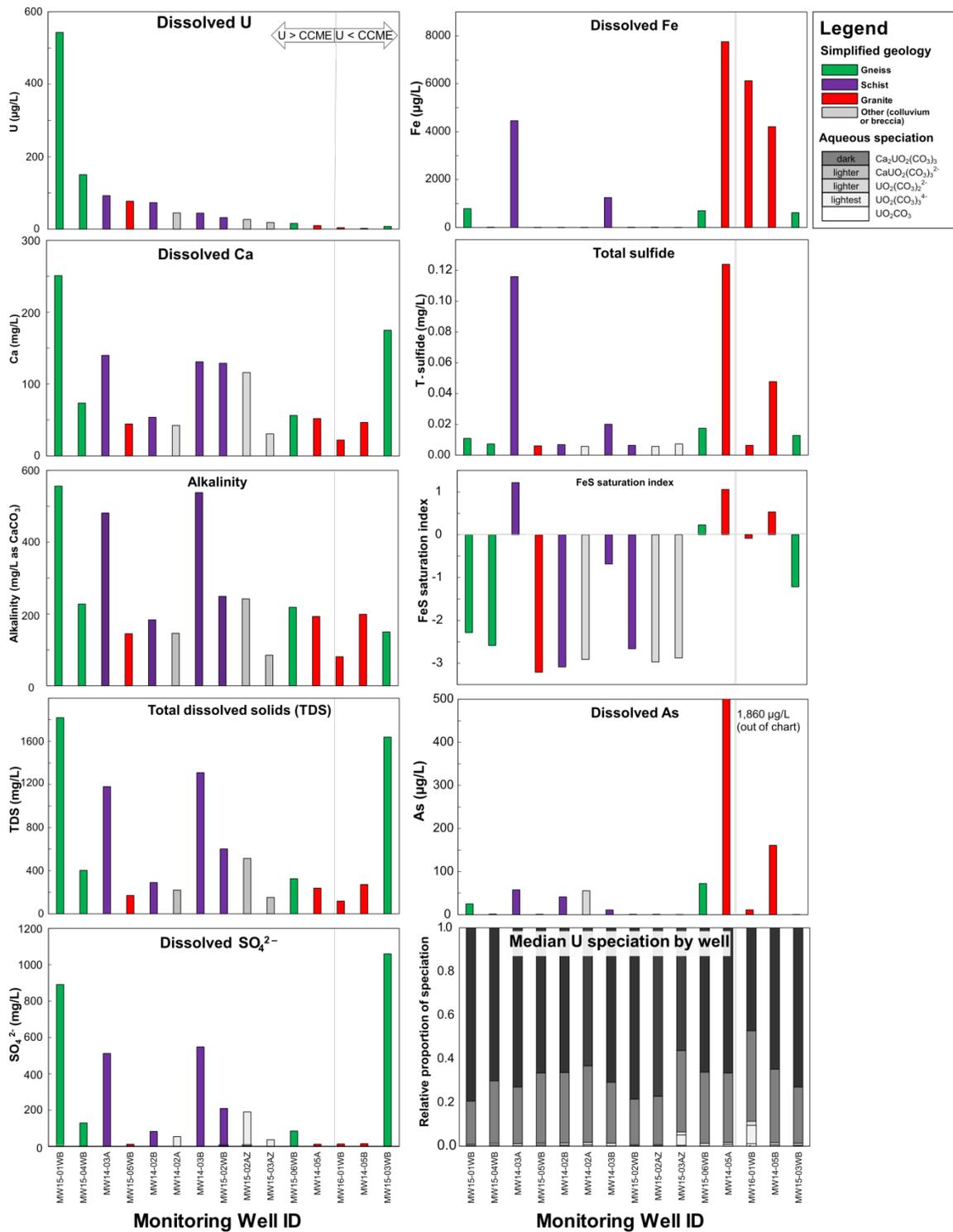


Figure 4-18: Median concentrations (SO₄, Alkalinity, TDS, Ca, U, Fe, As, total sulfide), FeS saturation index, and U speciation in groundwater wells at the Coffee site. Color reflects simplified lithology of screened well intervals. In the speciation plot, the shading distinguishes the aqueous species of U present. The vertical grey line delineates wells with median U concentrations above the CCME guideline value (15 µg/L).

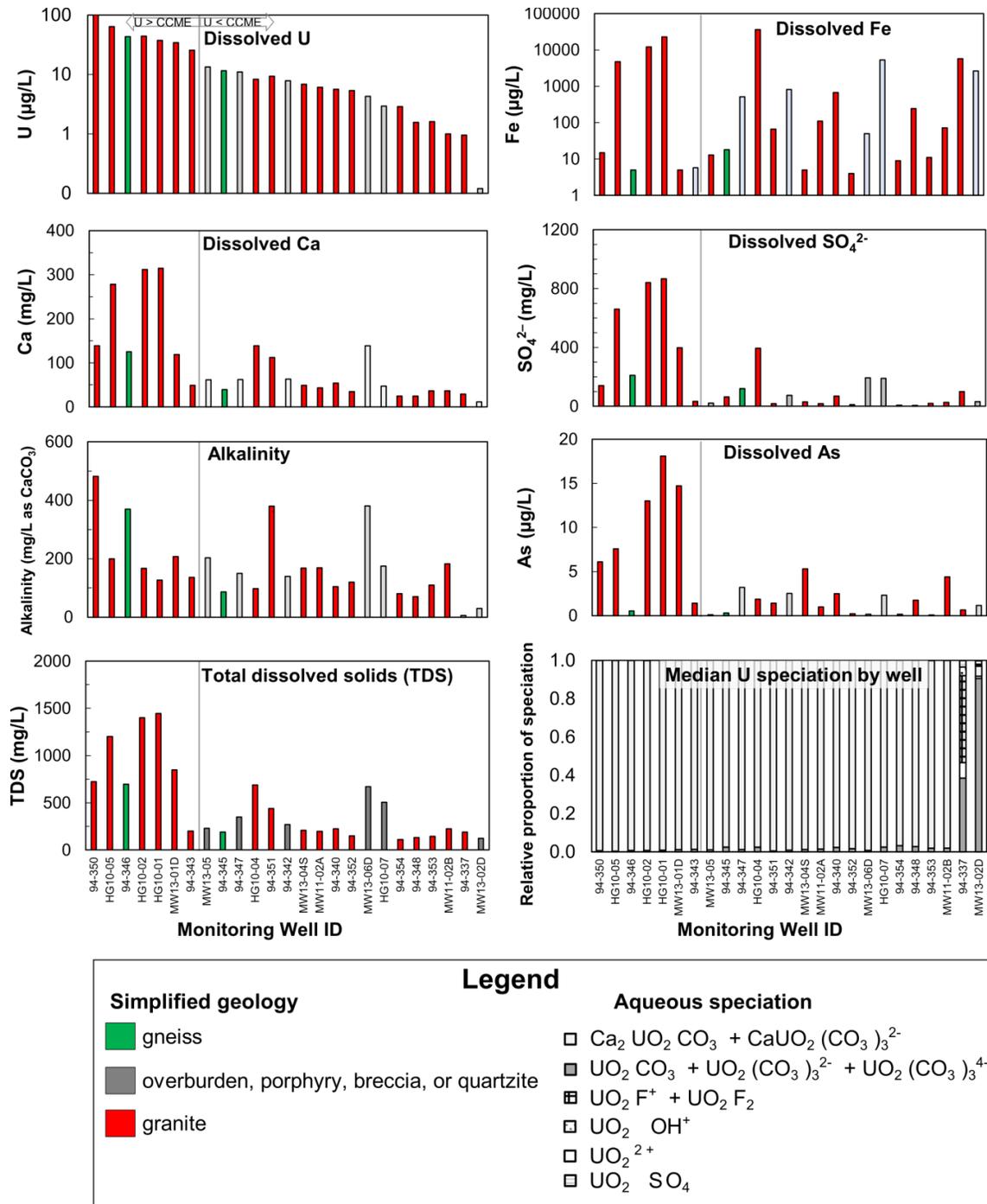


Figure 4-19: Median concentrations (SO_4 , Alkalinity, TDS, Ca, U, Fe and As) and U speciation in groundwater wells at the Casino site. Color reflects simplified lithology of screened well intervals. The vertical grey line delineates wells with median U concentrations above the CCME guideline value ($15 \mu g/L$).

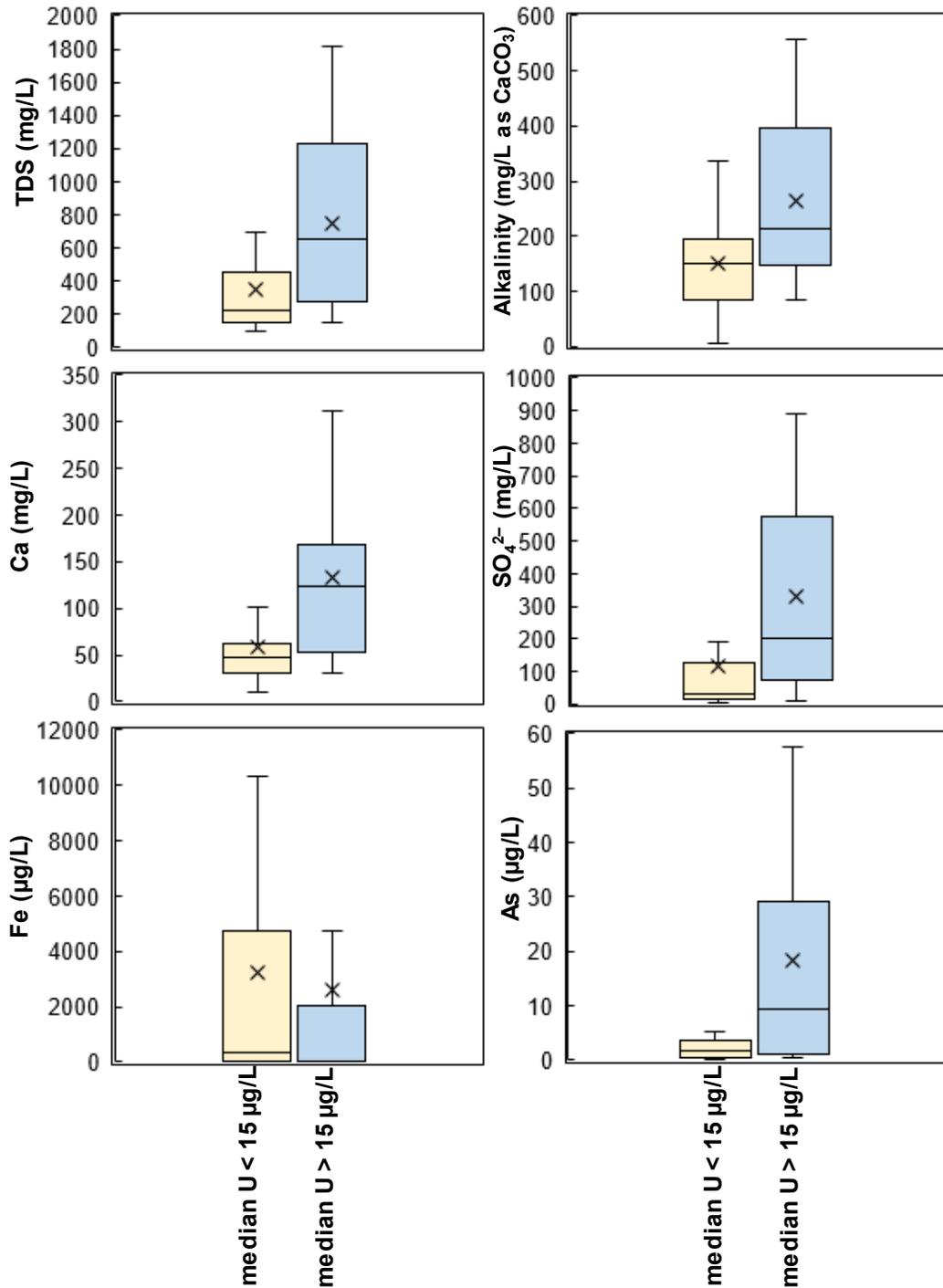


Figure 4-20: Boxplots comparing TDS, alkalinity, Ca, sulfate, Fe, and As for groundwater monitoring wells in the Dawson Range with median U concentrations above vs. below the 15 µg/L CCME guideline value.

4.2.3 Arsenic concentrations are low in surface water but high in groundwater

Arsenic concentrations in surface water are consistently low across the Dawson Range. The median and mean As concentrations of the 2,121 surface water samples that are available are both 0.5 µg/L, with a standard deviation of 0.7 µg/L (Figure 4-17). Only two As concentrations above the CCME guideline were observed in surface water in the entire dataset, both located in Barker Creek and both at concentrations of < 6 µg/L.

Groundwater As concentrations are substantially higher than those of surface water. In 17 of the 46 groundwater wells, median As concentrations exceed the CCME guideline, showing that high As is prevalent in groundwater around metal ore deposits (Figure 4-17 and Figure 4-16). Groundwater at the Coffee site generally has higher As concentrations than at Casino, consistent with a much lower As content in rock at the latter location (Figure 4-9, Figure 4-18, and Figure 4-19). The most As-rich groundwater is found in the MW14-05A/B well cluster screened into granite bedrock below area of gold mineralization at the Coffee site; these wells have median As concentrations of 1,860 µg/L and 161 µg/L, respectively. They are also characterized by reducing conditions, as indicated by elevated Fe(II) and sulfide concentrations (Figure 4-18).

5. Discussion

5. Discussion

5.1 Uranium

5.1.1 Uranium is mobilized in alkaline and calcium-rich groundwater

A combined analysis of whole-rock, sediment, surface water, and groundwater data suggests that there is a general regional enrichment of U in the Dawson Range. Uranium concentrations in surface water exceeding the CCME guideline for the protection of aquatic life are not uncommon. It is important to note that, with the exception of baseline environmental monitoring programs at the Coffee and Casino sites, dissolved U concentration data for surface waters consist of single-event sampling conducted by the Geological Survey of Canada (GSC) during the National Geochemical Reconnaissance Program between 1976 and 2006 which do not account for seasonal variation in surface water chemistry. Although these data show frequently elevated U concentrations in Dawson Range creeks, they are constrained by the strong seasonality U concentrations and flow in surface water. Assuming that the majority of GSC sample was conducted during summer months when groundwater discharge signatures can be diluted in surface waters due to runoff, it is possible that a sampling effort conducted under baseflow conditions might reveal additional U-rich creeks.

Key lithological hosts for U are the Whitehorse and Sulphur Creek suites, because they contain U above the average upper crustal abundance (Figure 4-1) and also comprise a major proportion of Dawson Range surface area (Figure 4-2). Other U-rich units such as the Rhyolite Creek complex and the Casino suite are also relatively enriched in U, although their expression is geographically localized in the Dawson Range.

However, geology alone is unlikely to account for U enrichment in groundwater and surface water: for example, the Casino site is located in a predominantly granitic area that includes the U-rich Whitehorse suite and Casino suite rocks, but has lower aqueous U concentration in comparison to the Coffee site where bedrock geology includes a major proportion of Sulphur Creek, Klondike, and Snowcap schists and gneisses that have lower U content than Whitehorse batholith rocks (Figure 4-2). Groundwater geochemical conditions therefore play a significant role in mobilizing geogenic U, and may play a more significant role in U occurrence in surface and groundwater than whole-rock U content. In the Dawson Range, gneisses and schists of the Sulphur Creek and Klondike assemblages contain more abundant carbonate minerals, the dissolution of which provides alkalinity and Ca that can enhance U mobility through complexation reactions. In contrast, the Whitehorse suite intrusives are generally poor in Ca-bearing carbonates, and so their U is

mobilized less effectively. Therefore, while there is widespread U at comparable levels (*i.e.*, a few $\mu\text{g/g}$) in many Dawson Range geological units, the most substantial U mobilization in groundwater coincides with rocks such as the Sulphur Creek assemblage that generate Ca-rich and high-alkalinity groundwater in combination with U-rich rock.

The relationship between alkalinity, Ca, and U mobilization in groundwater is highlighted in (Figure 5-1), where the most U-rich waters are encountered at higher Ca and alkalinity concentrations. Carbonate mineral dissolution occurs through acid attack, releasing Ca and carbonate into solution that complex with U(VI):

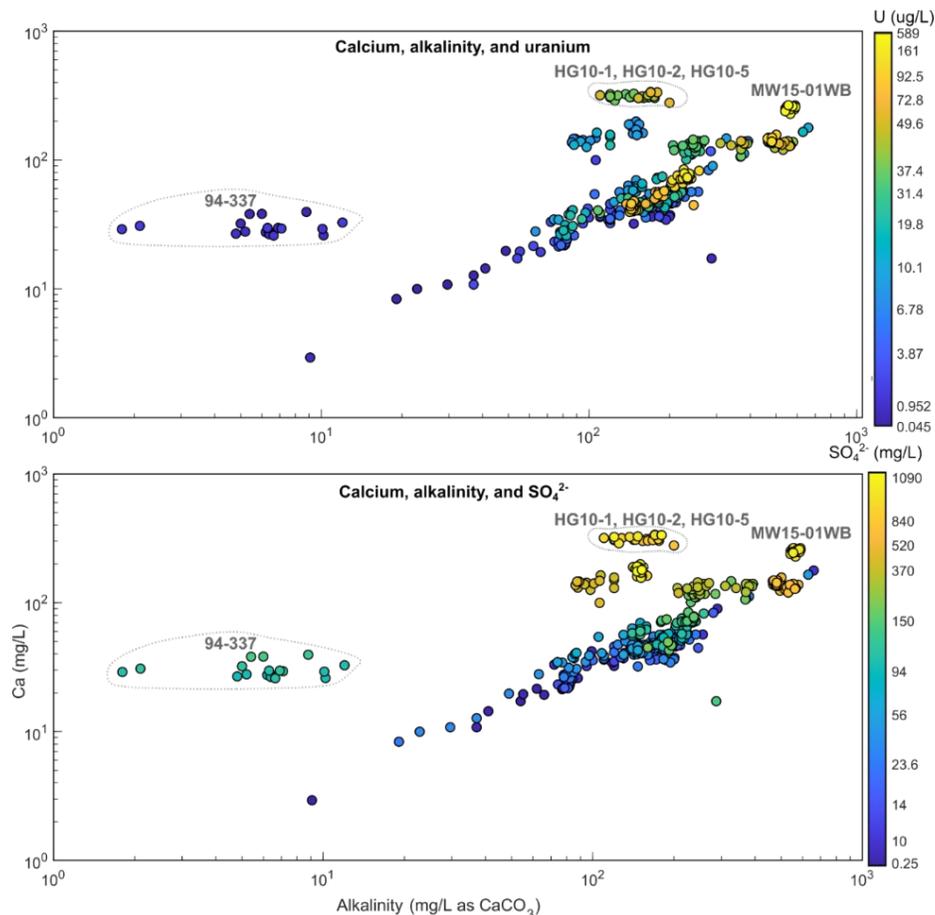
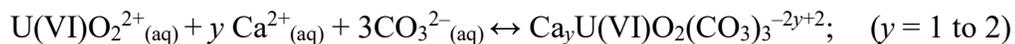
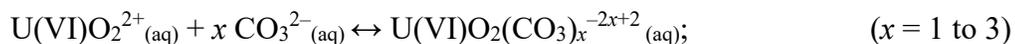
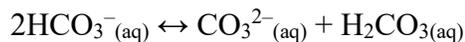
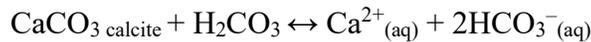


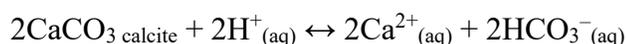
Figure 5-1: Relationship of Ca, alkalinity, sulfate, and uranium concentrations. Uranium (upper panel) and sulfate (lower panel) concentrations are represented by colors of symbols. Data points from select monitoring wells are also highlighted.

The presence of uranyl complexes, in particular $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3$ and $\text{CaUO}_2(\text{CO}_3)_3^{2-}$, governs aqueous U speciation in Dawson Range groundwater, enhancing U solubility (Figure 4-18 and Figure 4-19) by limiting the ability of $\text{U(VI)O}_2^{2+}_{(\text{aq})}$ to be attenuated via sorption or reduction to insoluble U(IV) (Stewart *et al.*, 2010; Stewart *et al.*, 2011).

5.1.2 Mobilization of U may be increased by sulfide-mineral oxidation

Overall, these results suggest that U mobilization in the Dawson Range does not necessarily require a sulfide-mineral oxidation environment, and U leaching may therefore be more regionally pervasive than that of As, which is limited to weathered orebodies. However, the presence of sulfide mineral orebodies may further enhance U mobility relative to non-sulfide bearing geologic environments through two mechanisms: (1) enhanced Ca-U-carbonate complexation through sulfide-mineral oxidation and carbonate buffering reactions, and (2) enhanced leachability of U in country rock caused by the ore-genesis process.

At the Casino site, the most U-rich groundwaters are typically found in proximity to the sulfide ore body where SO_4^{2-} concentrations are elevated as a result of sulfide-mineral oxidation. This process can enhance the release of Ca and alkalinity to solution through carbonate buffering reactions:



So long as there remains sufficient alkalinity to form uranyl-carbonate-calcium complexes, this process might enhance U mobilization both through increasing aqueous complexation and also through decreased UO_2^{2+} adsorption capacity as a result of ion competition from Ca^{2+} and other solutes released during acid-buffering reactions. This setting describes the majority of groundwater geochemical conditions below the oxidized cap and supergene zone at the Casino site, where groundwater in U-rich monitoring wells MW13-01D, HG10-01, HG10-02, and HG10-05 contains ≥ 400 mg/L SO_4^{2-} (released through sulfide-mineral oxidation) but is still well buffered through sufficient carbonate availability (pH 7.1 to 7.7, alkalinity 126 to 145 mg/L as CaCO_3). Uranium removal from weathered bedrock at Casino is reflected by the depletion of U in the oxide leach cap rocks relative to less highly weathered facies (Figure 4-6). Similar high-U, high-alkalinity, and high-sulfate groundwater is also widespread around the Coffee site. The most U-rich concentrations may therefore occur under neutral-rock drainage (NRD) conditions, where sulfide-mineral oxidation leads to carbonate and silicate mineral dissolution, Ca release, and concomitant U complexation and mobilization. As a result, U may constitute a contaminant of potential concern in mining environments in the Dawson Range that have a higher carbonate

buffering capacity relative to acid-generation potential (from sulfide-mineral oxidation) and that generate NRD.

5.1.3 Uranium is less mobile under moderately acidic conditions

In mine waste environments where acid-generation capacity exceeds carbonate mineral buffering (*i.e.*, acid-rock drainage), U mobility will be controlled by pH-dependent U sorption behavior. In the moderately acidic pH range of ~4.5 to ~7, the probability of finding U concentrations greater than the CCME guideline value is minimal in Dawson Range surface water (Figure 4-15), which is likely driven by an absence of uranyl complexation with dissolved calcium and carbonate and an increase in UO_2^{2+} sorption. The single groundwater monitoring location representative of an acid-rock drainage (ARD) environment in the dataset, well 94-337, also shows low U concentrations and relatively elevated sulfate and Ca, consistent with a moderately acidic geochemical environment where U sorption is strongest (Figure 5-1).

5.1.4 Uranium mobility increases in strongly acidic conditions

At even more advanced stages of acid-rock drainage (ARD) (pH ~3 or lower), UO_2^{2+} mobility once again increases because sorption sites become saturated with H^+ and Fe-(oxyhydr)oxide dissolution is favored by Fe(II) solubility in low pH conditions. A natural analogue for this ARD condition is present at the Casino site, where, continued sulfide oxidation has depleted the availability of carbonate minerals. The resulting water chemistry is captured by water quality in Proctor Gulch, a small tributary draining the mineralized area at Casino that is characterized by acidic conditions (median of pH 3.30), no detectable alkalinity, and high electric conductivity (1,020 $\mu\text{S}/\text{cm}$), dissolved Ca (median 104 mg/L), dissolved sulfate (median 439 mg/L), and dissolved iron (median 23,950 $\mu\text{g}/\text{L}$). Under these natural ARD conditions, U is mobile, with a median U concentration of 12 $\mu\text{g}/\text{L}$ and U speciation governed by UO_2OH^+ and UO_2SO_4 .

5.1.5 Uranium attenuation in groundwater by U(VI) reduction

In addition to sorption, reduction of U(VI) to insoluble U(IV) is a common control on U mobility in the environment. The ubiquity of U-rich groundwater in the Dawson Range suggests U(VI) reduction is limited. However, a few groundwaters at the Coffee site may have encountered U-reducing conditions. In particular, granitic wells MW14-05A and MW14-05B are drilled directly below a mineralized zone (the proposed “Kona Pit”). The low sulfate (median < 16 mg/L) and elevated Fe and As concentrations in these wells (Figure 4-18) may indicate that reduction of Fe- and As-bearing oxides has occurred. Biogenic U(VI) reduction to insoluble U(IV)O_2 uraninite can be completed by sulfate-

reducing and iron-reducing bacteria, which may thrive in Fe(II) and SO_4^- -bearing groundwater (Abdelouas *et al.*, 1998; Abdelouas *et al.*, 2000), as is present in many groundwaters around the Coffee and Casino sites. However, co-existence of elevated U and dissolved Fe in the mg/L range is observed in Casino groundwater (*e.g.*, HG10-05, HG10-02, and HG10-01) (Figure 4-19). These wells are also enriched in As, with median concentrations in the range of 8 to 18 $\mu\text{g/L}$, which is indicative of reduction of Fe- and As-bearing oxides. It is unclear whether a portion of the U in these waters has undergone reduction. Dissolved sulfide data are not available in the Casino database, limiting the ability to constrain groundwater redox conditions.

5.2 Arsenic release is promoted by reducing and alkaline groundwater

Arsenic mobilization in the Dawson Range is intrinsically tied to the presence of As-bearing minerals associated to sulfide ores and their oxidized derivatives. Regionally, unmineralized country rock has a low As content (typically a few $\mu\text{g/g}$), as shown in Open File 8500 rock samples. In contrast, ore deposits are enriched in As, which is found in phases such as arsenopyrite and arsenian pyrite and is consistent with the chalcophile behavior of this element (Plant *et al.*, 2014). There is also a correlation of As content in mineral deposits and that of groundwater: the Coffee deposit contains substantially more As than that at Casino, which translates to greater As concentrations in groundwater around the Coffee site (Figure 4-14 and Figure 4-18).

Arsenic mobilization is strongest under reducing groundwater conditions, where there is decreased availability of Fe-(oxyhydr)oxides to act as sorption surfaces. Reducing conditions may also promote conversion of As(V) to As(III), which has lower affinity for sorption sites in circumneutral pH waters (Plant *et al.*, 2014). Weaker retention of As under reducing groundwater conditions is particularly evident at the Coffee site, where groundwater wells with median Fe > 100 $\mu\text{g/L}$ (indicating Fe-reducing conditions) tend to have higher As than Fe-poor wells (Figure 5-2). In contrast, natural attenuation of As occurs under oxidizing conditions through sorption to Fe-(oxyhydr)oxides. This process is evidenced through the association of As to Fe-(oxyhydr)oxides and scorodite, both common sinks of As in oxidized sulfide deposits (Paktunc and Bruggeman, 2010; Blowes *et al.*, 2014). Furthermore, this process likely prohibits As transport from groundwater to surface water through oxidation of dissolved Fe(II) and precipitation as As-bearing Fe(III)-oxyhydr(oxides). As a result, little As is present in surface water (Figure 4-17).

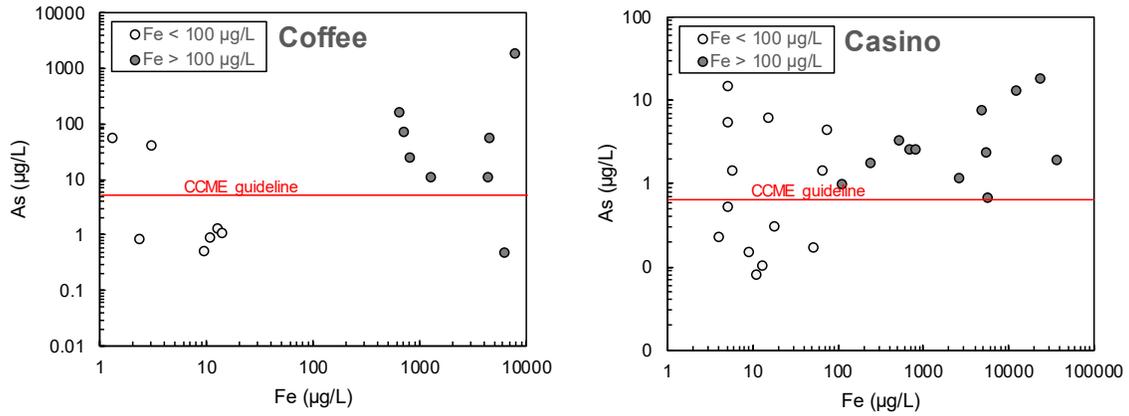


Figure 5-2: Median dissolved As and Fe concentrations calculated at each groundwater well. Coffee data at left, Casino data right. Filled symbols are wells with median Fe concentrations above > 100 µg/L.

***6. Key findings and implications
for mine waste management
in the Dawson Range***

6. Key findings and implications for mine waste management in the Dawson Range

6.1 Uranium in the baseline environment

- Uranium is commonly enriched above average crustal abundance in Dawson Range rocks. The geological units of greatest interest with respect to U mobilization are Whitehorse suite intrusives and Sulphur Creek orthogneiss due to their broad spatial extent and high U content.
- Elevated uranium concentrations in surface water and groundwater are associated with Ca and alkalinity, and uranium speciation calculations in groundwater show a majority of U is mobilized through complexation with aqueous Ca and carbonate ions.
- Uranium is present at concentrations exceeding the CCME guideline for the protection of aquatic life in many Dawson Range creeks, especially under baseflow conditions. Available datasets may underestimate peak U concentrations due to the strong inverse relationship of U concentrations and flowrates and the fact that surface-water sampling in creeks conducted during the RGS consists only of singular sampling events.

6.2 Arsenic in the baseline environment

- In contrast to U, mobilization of As in the Dawson Range appears localized and directly associated to the presence of mineral deposits where As is found in sulfides such as arsenopyrite and arsenian pyrite, or in oxidized derivatives of these minerals. While the As content of unmineralized Dawson Range rocks is generally low relative to average Earth crust, it can be orders of magnitude higher in ore deposits.
- Mobilization of As from rock to groundwater in the baseline environment occurs around the Coffee and Casino sites. Under oxic conditions, oxidative dissolution of As-bearing sulfides leads to As association to oxides and (oxyhydr)oxides. This reservoir of As can be mobilized under anoxic conditions, leading to As concentrations above CCME guidelines in reducing groundwaters.
- Arsenic in groundwater is attenuated prior to groundwater discharge, likely through sorption processes. As a result, As concentrations in creeks remain consistently well below CCME guidelines for the protection of aquatic life (typically < 1 µg/L in surface water).

6.3 Implications for waste rock and tailings management

- Baseline geogenic U enrichment occurs in Dawson Range water through weathering of local lithological units (especially the Sulphur Creek and Whitehorse Suite lithologies). Therefore, projects that include these rock types in their waste stream (*e.g.*, mining, quarrying) should consider the potential for enhanced U mobilization. Mine waste rock and tailings have higher surface area relative to undisturbed bedrock, which can promote metal(loid) mobilization (Blowes *et al.*, 2014; Lindsay *et al.*, 2015). Geochemical assessment of local bedrock sources can help evaluate the likelihood that a project will increase U relative to baseline conditions.
- Uranium mobilization in mine wastes is likely to be greatest under either circumneutral-to-alkaline (pH > 7) conditions with elevated Ca and alkalinity (*e.g.*, neutral-rock drainage), or very-acidic pH (pH < ~4.5) conditions.
- Because baseline U concentrations in surface water (and groundwater) are frequently above CCME guidelines in the Dawson Range area, applying the generic guideline of 15 µg/L to regulate industrial projects will not be practical in some instances. Site-specific or regional water-quality objectives should be considered for projects in the Dawson Range
- The association of As with ore deposits in the Dawson Range means that evaluation of As mobilization potential in mine wastes is required. In particular, monitoring data shows that As mobilization is enhanced by the reducing conditions observed in some groundwater monitoring wells. The potential for As mobilization from any subaqueously stored mine waste should be carefully examined.
- Because As concentrations in surface water are consistently low, special regional water-quality objectives for As are not needed.

6.4 Recommendations for future work

6.4.1 Hydrogeochemistry, mineralogy, and ecotoxicology

- Further research is required to constrain the mineralogical source of U in Dawson Range, which is challenging to assess due to its present at µg/g levels. Identifying the mineralogy of U in the various rock types and geological settings in the Dawson Range will help constrain under which geochemical conditions it is more mobile.
- Uranium occurrence in Dawson Range surface water is widespread but significant variation exists between individual watersheds. While U mobilization can be explained on a regional scale using characteristics of prevalent geologies and first

principles of U geochemistry, the cause of variation among adjacent watersheds is unclear. Local variations in hydrology, bedrock geology, and surface water and groundwater geochemistry will impact U occurrence, mobility and concentrations. Further investigation of individual watersheds at a higher level of detail than presented in this report can refine hydrological and geological conditions under which elevated U concentrations occur.

- Regional surface water chemistry is largely available through single-event reconnaissance sampling conducted by the GSC. Confirmatory sampling of these results under varying hydrological conditions (*e.g.*, freshet, baseflow) with modern techniques (*e.g.*, ICP-MS) can help refine baseline U concentrations in the Dawson Range and address the uncertainty in the GSC dataset that is inherent from the lack of repeated sampling in given monitoring locations at different flow stages.
- This study did not evaluate whether ecological receptors (*e.g.*, fish, benthic macro-invertebrates) are affected in Dawson Range creeks where U concentrations exceed the CCME guideline for the protection of aquatic life. Further ecotoxicological assessment is required to determine whether there are any adverse effects on ecological receptors in the baseline environment, and to quantify at which U concentrations adverse effects are likely.

6.4.2 Regulatory

- This study showed that U exceeds CCME guidelines for the protection of aquatic life in several Dawson Range creeks. This guideline may therefore not constitute a practical water-quality objective (WQO) in parts of the Dawson Range. As such, a specific regional water-quality objective for U could be established. It would be necessary to define a spatial area (*e.g.*, specific watersheds) where this WQO would be applicable. This work should take into consideration the strong seasonal and spatial variation in U concentrations in surface water bodies in the Dawson Range. This work would also benefit from ecotoxicological assessments to quantify the sensitivity of local ecological receptors to elevated U exposure.
- Uranium concentrations in Dawson Range bedrock are only marginally enriched compared to AUC values. Nonetheless, U in surface and groundwater is commonly observed at concentrations significantly greater than CCME guidelines. This result shows that metal enrichment in rock alone is a poor indicator of metal leaching potential, and metal leaching tests are needed to adequately characterize U leaching potential in this environment.

- Monitoring data shows that As mobilization is enhanced by the reducing conditions observed in some groundwater monitoring wells. This type of metal leaching behavior would not be captured in aerobic metal leaching tests (*e.g.*, humidity cells) designed to promote sulfide oxidation. Analytical techniques tests specifically designed to characterize metal release under reducing conditions (*e.g.*, water-saturated columns) should be conducted in support of projects in the Dawson Range which consider sub-aqueous disposal of significant volumes of As-rich geologic material.

7. Acknowledgements

7. Acknowledgements

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8. Closure

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We trust that this Report meets your present requirements. Please let us know if you have any questions or comments.

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