Surface-Water, Ground-Water, and Sediment Geochemistry of Epizonal and Shear-Hosted Mineral Deposits in the Tintina Gold Province—Arsenic and Antimony Distribution and Mobility

By Seth H. Mueller, Richard J. Goldfarb, Philip L. Verplanck, Thomas P. Trainor, Richard F. Sanzolone, and Monique Adams

Chapter G of

Recent U.S. Geological Survey Studies in the Tintina Gold Province, Alaska, United States, and Yukon, Canada—Results of a 5-Year Project

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Abstract

Epigenetic mineral deposits in the Tintina Gold Province are generally characterized by high concentrations of arsenic and antimony in their mineral assemblage. A total of 347 samples (ground water, surface water, and stream sediment) were collected to investigate the distribution and mobility of arsenic and antimony in the environment near known mineral deposits. Samples were collected from east to west at Keno Hill and Brewery Creek, Yukon, Canada; and Cleary Hill, True North, Scrafford Mine, Fairbanks, Ryan Lode, Stampede Creek, Slate Creek, and Donlin Creek, all in Alaska. Surface- and ground-water samples are all slightly acidic to near-neutral in pH (5–8), have a wide range in specific conductance (surface water 17–2,980 microsiemens per centimeter and ground water 170–2,940 microsiemens per centimeter), and show elevated dissolved arsenic and antimony concentrations (arsenic in surface water is less than 1 to 380 micrograms per liter and in ground water is less than 1 micrograms per liter to 1.5 milligrams per liter; antimony in surface water is less than 2 to 660 micrograms per liter and in ground water is less than 2 to 60 micrograms per liter). Stream sediments downstream from these deposits have high concentrations of arsenic and antimony (arsenic median is 1,670 parts per million, maximum is 10,000 parts per million; antimony median is 192 parts per million, maximum is 7,200 parts per million). The mobility of arsenic and antimony is controlled by the local redox environment, with arsenic being less mobile in oxidized surface waters relative to antimony, and arsenic more mobile in reduced ground water. These factors suggest that both antimony and arsenic may be useful pathfinder elements in water and sediment for targeting similar style deposits elsewhere in the Tintina Gold Province.

Introduction

In recent years, trace elements such as arsenic and antimony have garnered significant attention from the scientific community and the general public due to their toxicity and our initially limited, but rapidly growing understanding of their geochemical behavior in the natural environment (Welch and others, 2000; Filella and others, 2002a,b; Smedley and Kinniburgh, 2002). Arsenic and antimony are strongly associated with both gold and base metals in epigenetic shear-zone-related mineral deposits within the Tintina Gold Province (TGP; Hart and others, 2002). We have carried out numerous investigations on the distribution, speciation, and mobility of arsenic and antimony in ground and surface waters as well as stream sediments in multiple environments (prior to, during, and after mining); and on their efficiency as pathfinder elements in exploration for similar, as of yet undiscovered gold deposits in the TGP. Studies of the speciation and mobility of trace metals, such as arsenic and antimony, are also critical to understanding the natural degradation of both mined and undisturbed mineral deposits and can provide empirical data necessary for the development of environmental geochemical models of epizonal shear- and shear zone-hosted mineral deposits.

Four primary areas within the TGP were studied. As shown in figure G1 (from east to west) our study included (1) Keno Hill and Brewery Creek in the Tombstone district of Yukon, Canada; (2) several deposits in the Fairbanks mining district, including Cleary Hill, True North, Scrafford, and Ryan Lode, as well as undeveloped shear- and fault-hosted zones of mineralization within the vicinity of the city of Fairbanks; (3) the Stampede and Slate Creek deposits in the Kantishna Hills mining district; and (4) the Donlin Creek gold deposit in the Kuskokwim district in southwestern Alaska.

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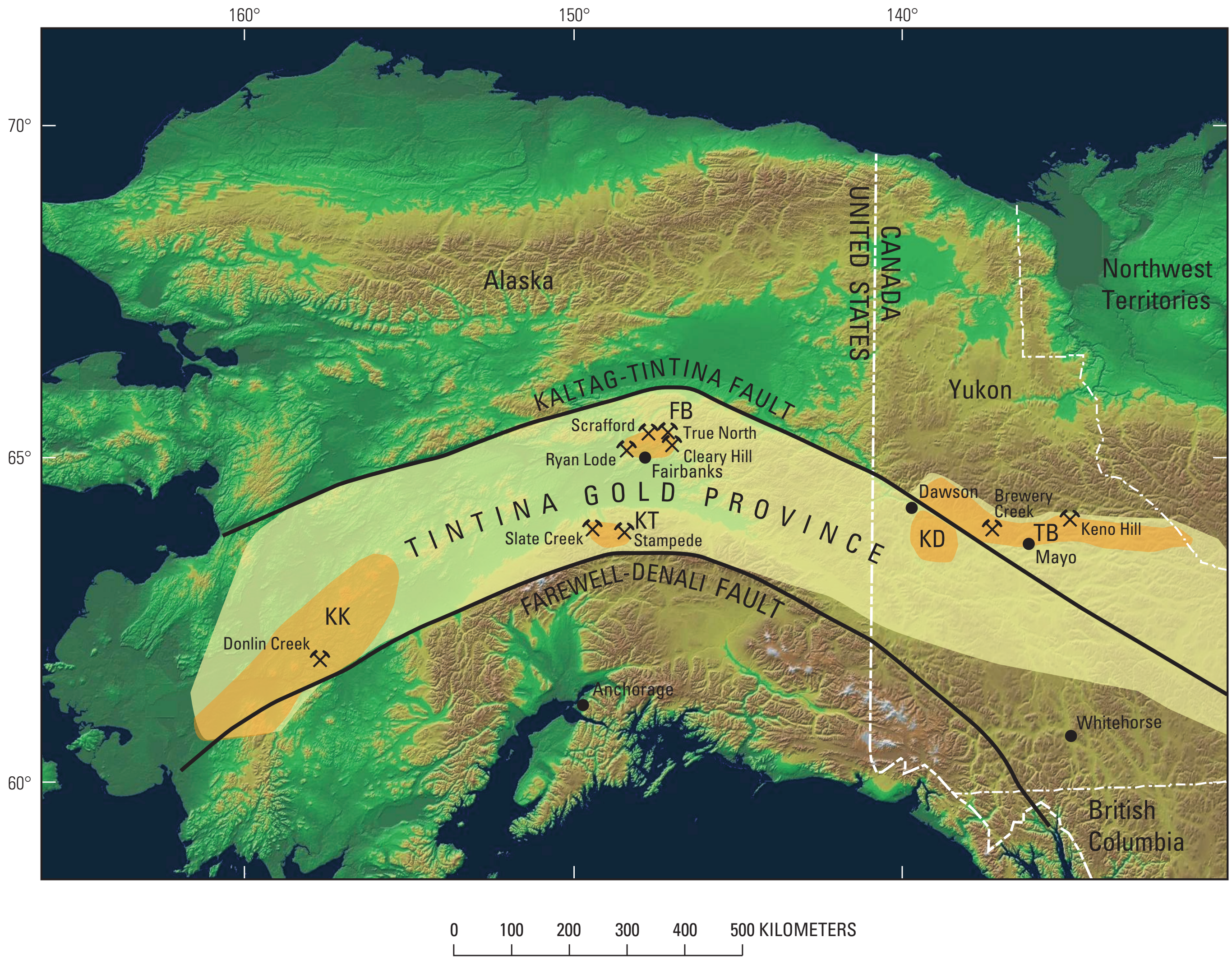

Figure G1. Landsat-based shaded-relief map showing the outline of the Tintina Gold Province, major faults, mining districts (KK, Kuskokwim; KT, Kantishna; FB, Fairbanks; KD, Klondike; TB, Tombstone Gold Belt), and study areas.

Geology of Mineral Deposits

The TGP is an arcuate east-west-trending belt of rocks made up of multiple and diverse geologic terranes generally bounded on the north by the Kaltag-Tintina fault system and on the south by the Farewell-Denali fault system (fig. G1). Three dominant geotectonic regimes lie within the TGP. They are, from east to west, (1) Neoproterozoic to middle Paleozoic turbidites, basinal clastic rocks, and limestones of the Selwyn basin continental margin sequence (Gordey and Anderson, 1993); (2) Neoproterozoic to Paleozoic polymetamorphosed metasedimentary and meta-igneous assemblages of the Yukon-Tanana Upland in east-central Alaska (Foster and others, 1994); and (3) Paleozoic to Mesozoic basement terranes overlapped by Cretaceous flysch sequences of the Kuskokwim basin (Decker and others, 1994).

The majority of the mineral deposits are shear zone- and (or) fault-hosted epizonal gold-bearing ores, typically containing as much as several volume percent arsenopyrite, pyrite, and (or) stibnite. The Keno Hill silver-lead-zinc deposit in the Tombstone district (fig. G1), although having a different metallogenic signature, was included for study because it is a similar style of epigenetic vein-type mineralization, and appears to have a spatial and temporal relationship with gold-bearing deposits in the eastern part of the TGP. Other gold deposits in the TGP with significantly lower sulfide volumes (for example, Fort Knox in Alaska and Scheelite Dome in Yukon), commonly termed intrusion-related gold systems (Hart and others, 2002), were not examined in these studies because arsenic and antimony mobility is less of a concern surrounding this deposit type. The following are brief summaries of the local geology of each study location from east to west across the TGP. For more detailed descriptions of the characteristics of these deposits, see Goldfarb and others (this volume, chap. A).

Keno Hill

The Keno Hill silver-lead-zinc deposits are located 270 kilometers (km) east of the Alaska-Yukon border, approximately 60 km northeast of the Brewery Creek deposit (fig. G1). This district is composed of about 70 subparallel veins (or vein systems) comprising a composite 6-km-wide by 30-km-long, east-west-trending mineralized zone. The vein deposits are hosted in the Carboniferous quartzite, carbonaceous phyllite, and calcareous quartzite of the informally named Keno Hill quartzite, and Middle to Late Devonian quartz-sericite-chlorite phyllite, carbonaceous phyllite, siliceous carbonaceous metasiltstone, and crystalline limestone of the Earn Group. These rocks are intruded by numerous granitic dikes and sills dated at about 90 million years before present (Ma) (Sinclair and Tessari, 1981; Franzen, 1986). Stibnite and arsenopyrite are associated with the silver-lead-zinc mineralization. Alteration and gangue minerals include quartz, calcite, and siderite. The majority of the historical mines in the Keno Hill district have been abandoned without reclamation.

Brewery Creek

The Brewery Creek deposit is located 60 km east of Dawson in east-central Yukon, in the hanging wall of the Jurassic to Cretaceous Robert Service thrust fault (Diment, 1996; Lindsay and others, 2000). The area is underlain by graptolitic shale, chert, mudstones, calcareous andesitic flows, tuffs and breccias, and siltstone of the Cambrian to Upper Devonian Road River Group, and argillite, silty shale, sandstone, graywacke, and limestone of the Earn Group (Diment and Craig, 1999; Lindsay and others, 2000). Granitic dikes and sills intrude these units and are the dominant ore host rocks. The deposit is characterized by an epizonal style of mineralization and is dated at 91 Ma (Lindsay and others, 2000). The ore-bearing quartz veinlets and disseminated sulfide zones are continuous along strike for 12 km. The ore contains a few percent arsenopyrite and pyrite, with gold-enriched rims. Stibnite postdates the main phase of gold mineralization (Diment and Craig, 1999; Lindsay and others, 2000). The mineralogical characteristics result in a gold-arsenic-antimony signature. Strong carbonate alteration occurs along the mineralized zone. The deposit is presently being reclaimed.

Fairbanks District

The deposits within the Fairbanks mining district included in this study are the Cleary Hill, True North, Scrafford, and Ryan Lode deposits. In addition, ground- and surface-water samples were collected throughout the city of Fairbanks, Alaska, area to determine the extent and environmental aspects of undeveloped fault- and shear-hosted disseminated sulfide zones concealed under Quaternary surficial materials. The mineral deposits of the Fairbanks mining district are hosted in a heterogeneous mixture of metasedimentary rocks that include quartz-muscovite schist, quartzite, chlorite quartzose schist, amphibolite schist, biotite schist, and marble of the Fairbanks Schist; slate, phyllite tuff, quartzite, calcareous schist, and marble of the Birch Hill sequence; and, in the case of True North, eclogite-bearing amphibolite and quartzite of the Chatinika terrane.

The Cleary Summit area is located 30 km northeast of the city of Fairbanks and contains about 30 small, high-grade (about 10 grams/ton gold) historical lode mining operations, including the Hi-Yu, Chatham Creek, and McCarty deposits. The deposits of Cleary Hill are best described as shear-hosted orebodies, with veins occurring as open-space fractures dominated by massive white quartz with variable sulfide content (Metz, 1991; McCoy and others, 1997). Alteration minerals include quartz, sericite, calcite, and ankerite.

The True North epizonal gold deposit is located 8 km north of the Cleary Hill mines. The ore zones are shallowly dipping and variably brecciated within the eclogitic rocks of the Chatinika terrane. Strong structural control is indicated by coincidence of ore zones with local thrust faults (Bakke and others, 2000). Gold occurs with fine-grained pyrite, arsenopyrite, and stibnite. In the developed oxidized zone, the pyrite and arsenopyrite have weathered to goethite and scorodite, respectively. Alteration minerals include quartz, manganese oxides, ankerite, mariposite, sericite, and graphite. The deposit is now abandoned and there is no reclamation.

The Scrafford deposit, located 18 km to the north of Fairbanks, was mined from 1915 to 1918 and was the second largest producing antimony mine in Alaska (Robinson and Bundtzen, 1982). Massive, fibrous stibnite occurs in blocks (about 3 m wide) within shear zones and in quartz stockwork veinlets (Robinson and Bundtzen, 1982). Minor arsenopyrite, gold, and galena are present within the lode. The shafts have since collapsed, leaving tailings piles and an open trench.

The Ryan Lode deposit is 13 km west of Fairbanks. The ore zone consists of northeast-striking, subparallel shear zones along the margin of a 90 Ma granodiorite plug (McCoy and others, 1997). The ore is mixed oxide-sulfide and contains several percent arsenopyrite and stibnite. Alteration minerals include albite, sericite, ankerite, and calcite.

Kantishna Hills

The Kantishna Hills mining district is located within the northwestern part of Denali National Park and Preserve. This district contains a number of gold- and antimony-bearing lode deposits, including Alaska's largest past antimony producer, the Stampede mine, as well as occurrences along Slate Creek. The host rocks consist of Precambrian to Paleozoic chlorite and graphite schists, marble, and metavolcanic rocks of the Spruce Creek sequence and calcareous schist, marble, slate, phyllite, tuff, and quartzite of the Birch Creek schist (Bundtzen, 1981). The district is defined by a number of vein deposits that trend toward the northeast for greater than 60 km, from Slate Creek in the southwest to Stampede in the northeast. Individual veins that make up the bulk of the orebodies in the area are structurally controlled along faults and range in width from 8 cm to greater than 9 m and range in length from 30 m to greater than 500 m. The orebodies at both Stampede and Slate Creek are composed predominately of massive stibnite.

Donlin Creek

The Donlin Creek deposit is located 450 km northwest of Anchorage, in the Kuskokwim Mountains. The approximately 70 Ma deposit consists of a hypabyssal dike-sill complex that intrudes the Kuskokwim Group sedimentary rocks (Miller and Bundtzen, 1994; Bundtzen and Miller, 1997). The gold occurs in north- to northeast-striking, steeply dipping quartz veins and veinlets with lesser dolomite that fill brittle extensional fractures in the rhyolite-rhyodacite porphyry dikes and sills. The gold is contained in arsenopyrite and rarely pyrite, and is refractory in nature (Szumigala and others, 2000). Late-stage stibnite, orpiment, realgar, cinnabar, and native arsenic are also present in small quantities (Goldfarb and others, 2004). Sericite and carbonate are common alteration minerals.

Sample Collection and Methods

A total of 196 surface-water samples, 65 ground-water samples, and 86 stream-sediment samples were collected from sites surrounding active mines, exploration targets, and unreclaimed and partially reclaimed sites, and areas in the vicinity of Fairbanks previously identified as having elevated ground-water arsenic concentrations (table G1, fig. G2). Detailed information regarding sample collection, analysis, quality assurance, and quality control can be found in Mueller (2002), Mueller and others (2002, 2003, 2004), and Verplanck and others (2003).

Ground-Water Samples

Three types of ground-water sites were sampled: individual domestic supply wells, ground-water monitoring wells, and drilling water-supply wells. In the case of domestic supply wells, a flow-through cell was connected to the plumbing system upstream of any household filtration or treatment systems. Water pH, specific electrical conductance, and temperature were monitored until all three readings stabilized, then a composite sample was collected in a sterilized 5-gallon bucket, from which a subset of samples was transferred into polypropylene bottles. The subset included the following samples: unfiltered-acidified (ultrapure nitric), filtered-acidified (0.45-micrometer (μm) disposable capsule filter and ultrapure nitric acid), filtered-unacidified (0.45-μm disposable capsule filter), and filtered-acidified (0.45-μm disposable capsule filter and ultrapure hydrochloric acid). All samples were refrigerated until analysis.

Surface-Water Samples

At surface-water sites (springs, seeps, and streams), pH, specific conductance, and temperature measurements were taken after the readings stabilized. Then, a representative sample was collected and a subset of samples transferred to polypropylene bottles and refrigerated until analysis (see above).

Table G1. Sampling locations, sample types, and number of samples collected for each location in this study.

Mining district	Sample location	Number of samples		
		Surface water	Ground water	Stream sediment
Tombstone	Keno Hill	14		14
	Brewery Creek	13		7
Fairbanks	Fairbanks (city of)	27	[1]40	
	Cleary Hill	9		7
	True North	6	[2]10	
	Scrafford Mine	6		6
	Ryan Lode		132	
Kantishna	Slate Creek	22		22
	Stampede	18		[3]18
Kuskokwim	Donlin Creek	81	[4]2	12
	Total	196	65	68

[1] Ground-water samples not associated with known mineralization.

[2] Monitoring wells.

[3] Analyses have not been completed on these samples as of this publication.

[4] Drilling supply wells.

Figure G2. Photographs of geologists performing data collection at various locations. *A*, Sampling the reclaimed area of Slate Creek, Kantishna Hills mining district, Denali National Park and Preserve. *B*, Sampling streams near Keno Hill, Yukon. *C*, Monitoring well sampling setup, Ryan Lode mine, Fairbanks, Alaska. *D*, Examining the Ryan Lode shear zone, Fairbanks, Alaska.

Sediment Samples

Bedload stream-sediment samples (less than 63-µm wet sieved onsite) were collected using a variation of the method described in Shelton and Capel (1994). This size fraction includes high surface area, reactive materials such as clays and iron and aluminum hydroxides that are likely associated with downstream transport of adsorbed or co-precipitated arsenic and antimony. Samples were split, with one portion air dried at room temperature for inductively coupled plasma–mass spectrometry (ICP-MS) analysis.

Results and Discussion

The information described herein is partially drawn and summarized from Mueller (2002), Mueller and others (2002, 2003, 2004, 2005), and Verplanck and others (2003).

Water Geochemistry

Generally, both ground and surface water can be classified as $Ca\text{-}Mg\text{-}HCO_3^-\text{-}SO_4^{2+}$ dominated. Specific conductivity values show wide variation across the TGP (surface water 17–2,980 microsiemens per centimeter (µS/cm), ground water 170–2,940 µS/cm). Regardless of lithology, and absence or presence of permafrost, ground and surface waters associated with epizonal vein mineralization in the TGP have slightly acidic to slightly basic pH (5–8). Acid-rock drainage is generally not an issue with these types of deposits in the TGP because the metasedimentary host rocks are often calcareous; both gangue minerals and alteration products include calcite, ankerite, and dolomite, all of which contribute to a natural acid-neutralizing capacity within the vicinity of the deposits.

Sediment Geochemistry

Elemental concentrations in stream sediments collected downstream from the gold deposits vary greatly. Arsenic, antimony, iron, and aluminum occur in relatively high concentrations (arsenic median=1670 ppm, maximum=10,000 ppm; antimony median=192 ppm, maximum=7200 ppm; iron median=5.9 percent, maximum=43 percent; aluminum median=8 percent, maximum=15 percent). Median concentrations for arsenic and antimony downstream from known lodes are at least an order of magnitude greater than in sediments from streams draining unmineralized areas.

Arsenic and Antimony Distribution and Mobility

Arsenic concentrations in surface-water samples from different deposits in the TGP range from below detection (less than 1 microgram per liter, µg/L) up to 380 µg/L (fig. G3*A*). The sampling locations with the highest surface-water arsenic concentrations were downstream from deposits at Slate Creek, undeveloped sulfidized shear zones in the Fairbanks area, Scrafford, Cleary Hill, and Brewery Creek. Surface-water antimony concentrations ranged from below detection (less than 2 µg/L) to 660 µg/L (fig. G3*A*). Antimony concentrations were significantly higher in streams draining the Slate

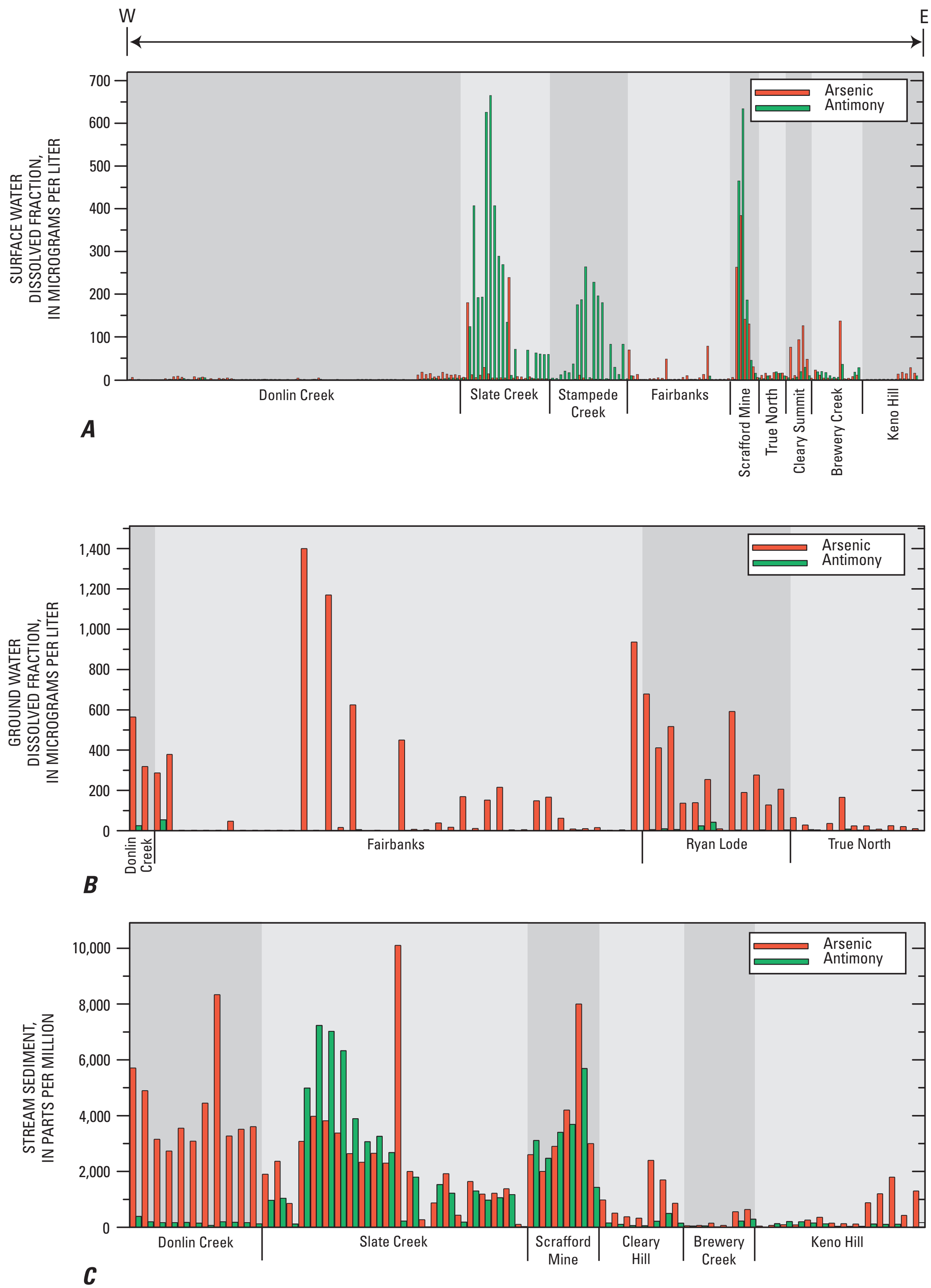

Figure G3. Graphs showing arsenic and antimony concentrations. *A*, Dissolved (less than 0.45-micrometer (µm) fraction) arsenic and antimony concentrations in surface water from west to east across the Tintina Gold Province (TGP). *B*, Dissolved (less than 0.45-µm fraction) arsenic and antimony in ground water from west to east across the TGP. *C*, Arsenic and antimony concentrations in the less than 63 µm fraction of stream-sediment samples from west to east across the TGP.

Creek, Stampede Creek, and Scrafford deposits. Arsenic concentrations in stream sediments are relatively elevated in stream channels downstream from the Donlin Creek, Slate Creek and Scrafford deposits (fig. G3*C*). Antimony is significantly enriched in sediment samples near the Slate Creek and Scrafford deposits (fig. G3*C*). Proximity to the sulfide source has the greatest effect on the relative concentration of arsenic and antimony. Generally, areas with exposed ore and (or) tailings at or near the surface (for example, Slate Creek and Scrafford Mine) have higher surface and stream-sediment concentrations of arsenic and antimony than do areas that have been revegetated naturally or by reclamation efforts (for example, Cleary Hill, Brewery Creek, and Keno Hill).

The absolute concentrations of arsenic and antimony in sediments are relatively similar from a given mineral deposit area (fig. G3*C*). In contrast, corresponding surface-water samples contain significantly less arsenic than antimony (fig. G3*A*, *C*). Surface-water arsenic concentrations are not as high as surface-water antimony concentrations due to the fact that in these near-neutral-pH oxidizing environments, arsenic undergoes oxidation from the more mobile arsenite (As(III)) to arsenate (As(V)) and adsorbs effectively onto iron hydroxides (Smedley and Kinniburgh, 2002). In contrast, antimony, even as oxidized Sb(V), has an apparent lower affinity for adsorption to iron or aluminum hydroxides and, therefore, remains mobile in solution. Surface-water arsenic concentrations commonly decrease significantly (to below detection limits) within less than 1 km from the source, whereas antimony concentrations can remain significantly above detection limit for as far as 8 km from the source.

Ground-water arsenic concentrations range from below detection (less than 1 µg/L) to 1.4 mg/L, with the highest concentrations being recorded in domestic water supply wells in the Fairbanks area (fig. G3*B*). Ground-water antimony concentrations are significantly lower than arsenic from all areas in the TGP, with concentrations ranging from below detection limit (less than 2 µg/L) to 60 µg/L. The highest concentrations of antimony in the Fairbanks area are in domestic supply wells and monitoring wells at the Ryan Lode deposit. The sources of the arsenic and antimony in the ground water are arsenopyrite and stibnite associated with subsurface shear- and fault-hosted sulfide zones. Both As(III) and As(V) are present in many of the ground-water samples. The presence of mixed arsenic species, along with the absence of acidic ground water, suggests that the source of arsenic in the ground waters is not the direct oxidation of arsenopyrite but rather the reductive dissolution of secondary oxide phases, such as arsenic adsorbed to iron hydroxides (see above discussion).

Ground-water arsenic concentrations in the Fairbanks area have been observed to vary by orders of magnitude over short distances (less than 500 m). The distribution of elevated arsenic concentrations in ground water in the Fairbanks area suggests that developed mineral deposits are not a major source of the arsenic; rather, it appears that undeveloped fault- and shear-hosted disseminated sulfides may extend in a northeast-striking trend several kilometers away from known lode deposits and may be a major contributor to the anomalous arsenic.

Exploration

This study not only provides valuable information on the distribution and mobility of arsenic and antimony but also provides information that can be used in exploration for disseminated and shear-hosted mineralization styles of epigenetic gold deposits in the TGP. We have demonstrated that regardless of the state of development of a given deposit (historical, exploration target, current production), both arsenic and antimony are relatively enriched in stream sediments within and downstream from mineralized areas when compared to background stream-water and sediment samples. Antimony may be detectable in surface waters several kilometers downstream of a mineralized source, whereas arsenic in surface waters will increase closer to the mineralized source.

Conclusions

Arsenic and antimony are typically highly anomalous in the Cretaceous epizonal and shear-zone-related gold deposits within the TGP. The geochemistry of surface-water and stream-sediment samples suggests that arsenic mobility is restricted by oxidation and adsorption processes. Antimony appears to be more mobile, remaining in solution at elevated concentrations for several kilometers downstream from a known source. The exposure and proximity of the mineralization to the surface environment directly affects the concentrations in the stream sediments and waters. The concentration of arsenic in ground water, particularly in the Fairbanks, Alaska, area, reflects proximity to numerous sulfide-rich fault and shear zones. Arsenic concentrations in ground waters are also controlled by the reduction-oxidation reaction (redox) state of the ground water, with the more strongly reduced ground waters having higher arsenic concentrations.

All of these aspects of arsenic and antimony geochemistry in the subsurface- and near-surface environment show that antimony and arsenic are useful pathfinder elements in exploration for epizonal and shear-related gold deposits within the TGP. Owing to the greater mobility of antimony, it could serve to more broadly target a mineralized zone, whereas arsenic could then conceivably be used to focus and target mineralized zones.

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