



## Geochemical controls of elevated arsenic concentrations in groundwater, Ester Dome, Fairbanks district, Alaska

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### ABSTRACT

Ester Dome, an upland area near Fairbanks, Alaska, was chosen for a detailed hydrogeochemical study because of the previously reported elevated arsenic in groundwater, and the presence of a large set of wells amenable to detailed sampling. Ester Dome lies within the Fairbanks mining district, where gold-bearing quartz veins, typically containing 2–3 vol.% sulfide minerals (arsenopyrite, stibnite, and pyrite), have been mined both underground and in open cuts. Gold-bearing veins on Ester Dome occur in shear zones and the sulfide minerals in these veins have been crushed to fine-grained material by syn- or post-mineralization movement. Groundwater at Ester Dome is circumneutral, Ca–HCO<sub>3</sub> to Ca–SO<sub>4</sub> type, and ranges from dilute (specific conductance of 48 μS/cm) to more concentrated (specific conductance as high as 2070 μS/cm). In general, solute concentrations increase down hydrologic gradient. Redox species indicate that the groundwaters range from oxic to sub-oxic (low dissolved oxygen, Fe(III) reduction, no SO<sub>4</sub> reduction). Waters with the highest Fe concentrations, as high as 10.7 mg/L, are the most anoxic. Dissolved As concentrations range from <1 to 1160 μg/L, with a median value of 146 μg/L. Arsenic concentrations are not correlated with specific conductance or Fe concentrations, suggesting that neither groundwater residence time, nor reductive dissolution of iron oxyhydroxides, control the arsenic chemistry. Furthermore, As concentrations do not covary with other constituents that form anions and oxyanions in solution (e.g., HCO<sub>3</sub>, Mo, F, or U) such that desorption of arsenic from clays or oxides also does not control arsenic mobility. Oxidation of arsenopyrite and dissolution of scorodite, in the near-surface environment appears to be the primary control of dissolved As in this upland area. More specifically, the elevated As concentrations are spatially associated with sulfidized shear zones and localities of gold-bearing quartz veins. Consistent with this interpretation, elevated dissolved Sb concentrations (as high as 59 μg/L), also correlated with occurrences of hypogene sulfide minerals, were measured in samples with high dissolved As concentrations.

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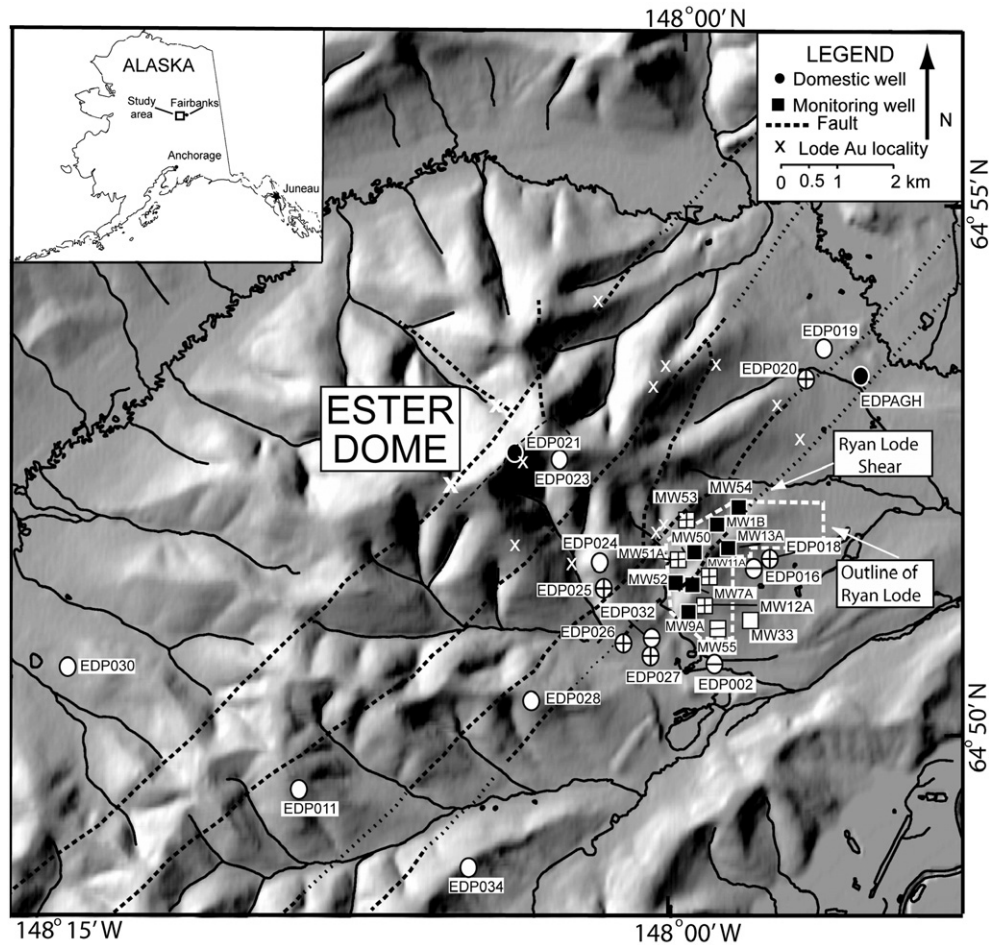
### 1. Introduction

As population grows, groundwater resources are being increasingly exploited for drinking water. Arsenic commonly occurs in groundwater at concentrations greater than the World Health Organization (WHO) guideline and the United States Environmental Protection Agency's (USEPA) drinking water standard of 10 μg/L. High arsenic concentrations are found in a variety of natural environments, including geothermal systems (Ballantyne and Moore, 1988; Webster and Nordstrom, 2002), sedimentary basins (Smedley and Kinniburgh, 2002; Bhattacharya et al., 2002; Welch and Stollenwerk, 2002), and metallic mineral deposits (Boyle et al., 1998; Groves et al., 1998; McCuaig and Kerrich, 1998; Camm et al., 2004; Smedley et al., 2007). Arsenic contamination of drinking water supplies has been found in

most countries, resulting in more research regarding the hydrogeochemical parameters that control arsenic mobility in groundwater (Smedley and Kinniburgh, 2002; Nordstrom, 2002). Some of the highest groundwater arsenic concentrations in the United States are located in east-central Alaska. This paper reports on the hydrogeochemistry of groundwater in Ester Dome, Alaska and evaluates the factors leading to high arsenic concentrations.

Ester Dome, located 10 km west of Fairbanks, Alaska (Fig. 1), lies within the Fairbanks mining district and is known to contain groundwater with high dissolved arsenic concentrations (Wilson and Hawkins, 1978; Hawkins et al., 1982). In the early 1970s, As concentrations in excess of 1 mg/L in domestic wells were first reported (Harrington et al., 1978). The Fairbanks region was once a center of placer gold mining, and related small lode mines have been worked on Ester Dome intermittently since the early 1900s. Within the gold-bearing quartz veins, sulfide mineral abundances typically are 2–3 vol.% and are dominated by arsenopyrite, stibnite (Sb<sub>2</sub>S<sub>3</sub>), and pyrite (Newberry et al., 1996; McCoy et al., 1997). Although arsenopyrite is

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**Fig. 1.** Shaded relief map of Ester Dome, Alaska displaying sampling site locations coded by dissolved arsenic concentration (open symbol  $<10 \mu\text{g/L}$ , single divided symbol  $10\text{--}49 \mu\text{g/L}$ , double divided symbol  $50\text{--}200 \mu\text{g/L}$ , and closed symbol  $>200 \mu\text{g/L}$ ). Fault locations and lode Au localities from Newberry et al. (1996) and Cameron (2000).

likely the primary source of most As, weathering has redistributed the arsenic such that it is also associated with secondary phases and oxide crusts.

In many gold-bearing deposits, arsenic may occur as a primary constituent in sulfide minerals such as arsenopyrite and enargite, or as a minor or trace constituent in minerals such as pyrite. Examples of gold occurrences with substantial amounts of arsenopyrite include many deposits throughout the Fairbanks region and elsewhere in the Tintina gold province of east-central Alaska (Flanigan et al., 2000; Hart et al., 2002; Goldfarb et al., 2005), the Macraes gold deposits on the South Island of New Zealand (Craw et al., 2004), and the Osilo area of Sardinia, Italy (Cidu et al., 1995). In fact, the Salsigne gold deposit in the French Alps was the world's largest arsenic producer for many years (Le Guen et al., 1992; Demange et al., 2006).

Oxidation of sulfides, mediated by bacteria, releases the arsenic (Nordstrom and Southam, 1997). Depending on the geochemical conditions, such as redox, pH, temperature, bacterial ecology, and aqueous chemistry, As mobility can be retarded by the formation of secondary phases such as sulfosalts and scorodite, a hydrated iron arsenate that forms under acidic, oxidizing conditions. Arsenic mobility can also be retarded by sorption to Fe, Al, and Mn precipitates and crusts, as well as to clays (Smedley and Kinniburgh, 2002). Arsenic can be remobilized from these secondary sources as hydrologic and geochemical conditions change. In circumneutral, sub-oxic groundwaters, such as Ester Dome, scorodite is unstable and tends to dissolve incongruently, releasing As into solution (Robins, 1987; Vink, 1996). Release of As from Fe oxides has been studied in detail because Fe oxides tend to be in higher concentration than other oxides (Smedley

and Kinniburgh, 2002). A change in redox conditions from oxidizing to reducing can lead to the reductive dissolution of Fe and the release of As (Smedley and Kinniburgh, 2002). Arsenic transport in groundwater is complex because as pH and redox conditions change along the groundwater flowpath, different processes can control its distribution between aqueous and solid phases in different parts of an aquifer.

Since the discovery of elevated arsenic concentrations in domestic wells in the Fairbanks area in the mid-1970s, geochemical studies have documented the range of As concentrations and have tried to understand the hydrogeochemical processes responsible for the anomalies. Most of these studies either focused on As and just a few other constituents (Wilson and Hawkins, 1978; Johnson et al., 1978; Hawkins et al., 1982) or looked at the entire Fairbanks area (Goldfarb et al., 1997; Farmer et al., 1998; Mueller, 2002). Most authors have noted the widespread, but heterogeneous, occurrence of arsenopyrite and scorodite within the bedrock and concluded that these minerals are the primary source of dissolved As in the groundwater. In the Fairbanks area, Hawkins et al. (1982), Farmer et al. (1998), and Mueller (2002) found that As in groundwater with the highest concentrations occurred primarily as As (III) and concluded that wells with chemically reducing groundwater have the greatest potential for elevated arsenic concentrations.

The objective of this hydrogeochemical investigation was to determine baseline conditions and identify processes controlling the variability of As specifically in one local part of the geologically and hydrologically complex Fairbanks region. Ester Dome was chosen for this detailed study because previous investigations documented consistently elevated As concentrations in groundwater within an

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