

Chemical Compositions of Fluid Inclusions in Intrusion-Related Gold Systems, Alaska and Yukon, Using PIXE Microanalysis

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Abstract

Proton-induced X-ray emission (PIXE) has been used to characterize the multielement chemistry of the diverse fluid inclusions found in intrusion-related gold systems in the Tintina gold province, Yukon and Alaska. The studied samples are from shallow-level examples that contain coexisting brine (type 3) and carbon dioxide-bearing vapor (type 4) inclusions (e.g., Shotgun, Donlin Creek Dome area, Mike Lake, and Brewery Creek) and deeper level deposits (e.g., Pogo, Dublin Gulch, and Emerald Lake) that contain low-salinity carbon dioxide- (type 1) and/or methane-rich (type 5) inclusions, which locally are overprinted by late secondary type 3 inclusions (e.g., Pogo and Emerald Lake).

Major element ratios, K/Ca and Mn/Fe, of both synore high-salinity (type 3) and low-salinity (types 1, 4, and 5) inclusions are >1 and <0.24 , respectively. The latter is consistent with the reduced conditions in which intrusion-related gold systems form. Late, secondary type 3 inclusions at Emerald Lake and Pogo, however, are chemically distinct, with higher Mn/Fe ratios (>0.24), and at Pogo low K/Ca ratios (<0.2). Chlorine and bromine data have been used to trace the source of salinity. Two distinct groups of Br/Cl mol ratio are recognized. Group 1 includes type 3 inclusions from the Pogo region, Mike Lake, Brewery Creek, and Emerald Lake, which have Br/Cl mol ratios consistent with typical magmatic values, mostly above 0.5×10^{-3} and below 1.54×10^{-3} (seawater). Group 2 comprises type 3 inclusions from Donlin Creek and Shotgun in southwestern Alaska, which have Br/Cl mol ratios from 2.34×10^{-3} to 6.37×10^{-3} , potentially reflecting a halogen contribution from the local sedimentary crust (the Kuskokwim basin) considered to be the primary source of the granite melts.

The data also provide insights into important metal contents of the fluid inclusions, including copper, zinc, lead, tungsten, and arsenic; however, gold, bismuth and antimony were all below the detection limits for these elements by the PIXE technique. The results explain some of the distinct metal associations of shallow and deep intrusion-related gold systems. Fluid inclusions in deposits emplaced at shallow crustal levels are characterized by higher iron, manganese, zinc, and lead contents due to the greater abundance of chlorine. Tungsten is more elevated in the low-salinity, carbon dioxide-bearing fluid inclusions in deposits at deeper levels, consistent with high tungsten in the deposits and likely due to the formation of tungstate rather than chloride complexes. Copper and arsenic have similar concentrations in both low- and high-salinity inclusions, also suggesting that ligands other than chlorine were important for these elements. Experimental and microanalytical studies have shown that copper, arsenic, and gold can complex with sulfur and do not require chlorine, exclusively, for metal transport. This may explain why deposits at both shallow and deep levels contain gold despite the wide variation in salinity and different fluid types present.

Introduction

INTRUSION-RELATED gold deposits associated with tin-tungsten terranes are a recently recognized class of deposit (Fig. 1; Thompson et al., 1999; Lang et al., 2000; Lang and Baker, 2001; Hart et al., 2002; Baker et al., 2005). The Tintina gold province is the best described intrusion-related gold deposit region and contains some of the most significant resources

including Donlin Creek (117 Mt at 3 g/t Au: Ebert, 2003; Goldfarb et al., 2004), Fort Knox (169 Mt at 0.9 g/t Au: Bakke, 1995), Shotgun (~1 Moz: Rombach and Newberry, 2001), and Pogo (10 Mt at 16 g/t Au: Smith et al., 1999; Rhys et al., 2003) in Alaska, and Brewery Creek (13.3 Mt at 1.4 g/t Au: Diment, 1996) and Dublin Gulch (50.3 Mt at 0.9 g/t Au: Maloof et al., 2001) in Yukon. The deposits in the Tintina gold province share many characteristics, including a spatial and temporal association with middle to Late Cretaceous, moderately reduced, I-type felsic intrusions, that typically host gold-bearing quartz veins characterized by a reduced (pyrrhotite-stable

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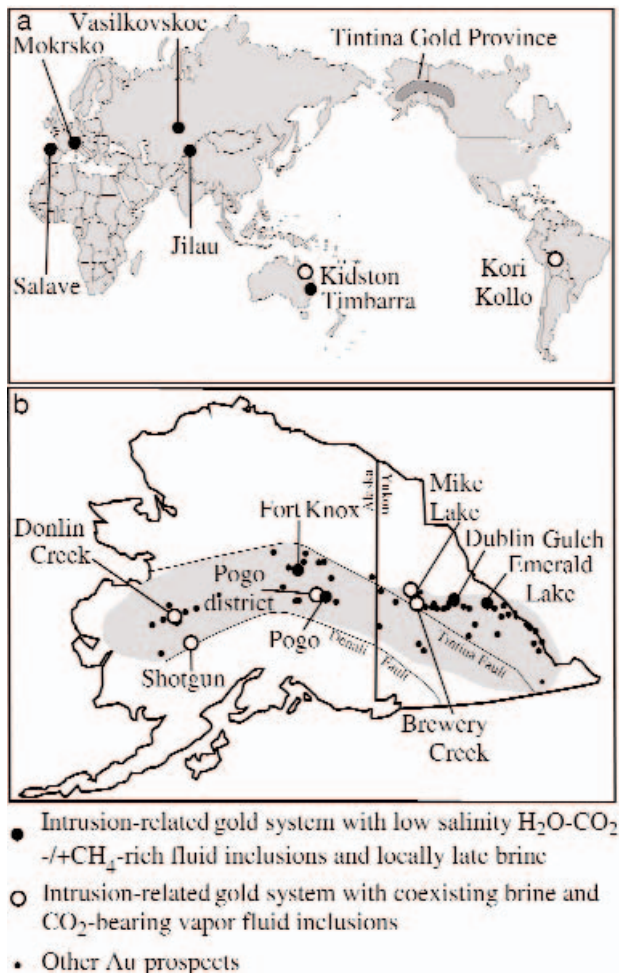


FIG. 1. a. World map showing the locations of major intrusion-related gold systems and their major fluid inclusion characteristics. b. Tintina gold province map showing the locations of deposits and prospects selected for this study. Their major fluid inclusion characteristics are indicated.

with no magnetite or hematite), low sulfide (<5 vol %) ore assemblage. The deposit styles, however, vary greatly and include large flat veins (Pogo), sheeted veins (Fort Knox and Dublin Gulch), breccia and stockwork (Shotgun), and dike-sill-hosted veinlets (Brewery Creek and Donlin Creek). This variation in style has been attributed to depth of emplacement and proximity to intrusions, with systems such as Donlin Creek and Brewery Creek considered to be shallow-level epithermal and/or epizonal deposits (<2 km; Baker, 2002; Ebert, 2003; Goldfarb et al., 2004), Shotgun emplaced at typical porphyry levels (2–5 km; Rombach and Newberry, 2001) and Pogo, Fort Knox, and Dublin Gulch emplaced in deeper plutonic environments (>5 km; McCoy et al., 1997; Smith et al., 1999; Maloof et al., 2001). Shallow-level deposits are characterized by gold associated with arsenic and antimony and locally elevated base metals, whereas deeper systems commonly contain abundant bismuth, tungsten, and arsenic. Similar features have been recognized globally, with deposits such as Kori Kollo considered to be a shallow-level example, Kidston interpreted to have formed at shallow to moderate depths, and systems such as Timbarra, Jilau, Salave, Mokrsko,

and Vasilovskoe thought to be emplaced into the plutonic environment (Fig. 1; Thompson et al., 1999).

There has been significant debate regarding the genesis of intrusion-related gold deposits and, in particular, whether the deposits were formed primarily through focused magmatic-hydrothermal processes (e.g., Thompson et al., 1999; Baker, 2002) or through large-scale metamorphic processes (e.g., Groves et al., 2003; Goldfarb et al., 2004). Much of this controversy centers around the diversity of hydrothermal fluid types, preserved as fluid inclusions, in the deposits. Deposits emplaced at shallow to moderate level (<5 km) commonly contain moderate- to high-temperature (>300°C) immiscible brine and carbon dioxide-bearing vapor inclusions in addition to lower temperature (<300°C) low-salinity, carbon dioxide-bearing aqueous inclusions (Fig. 1; Baker, 2002). Deeper deposits have only localized occurrences of brine inclusions (typically late and secondary; Baker and Lang, 2001) and contain abundant moderate-temperature (200°–400°C) low-salinity, carbon dioxide (and/or methane)-bearing aqueous inclusions. The latter are consistent with fluid types typical of metamorphic environments (Phillips and Powell, 1993), whereas brine and vapor inclusions are commonly found in magmatic-hydrothermal deposits (Bodnar, 1995). However, Baker (2002) argued that the low-salinity carbon dioxide-bearing aqueous inclusions may well be magmatic in origin and cited experimental work by Lowenstern (2000, 2001) and Giggenbach (1997), which showed that carbon dioxide degasses from felsic magmas at much higher pressures than water and chlorine. Consequently, the carbon dioxide contents of deep magmatic-hydrothermal systems are higher and salinities are lower than in shallow environments.

Recent advances in microanalytical techniques such as the laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS; Audetat et al., 1998; Heinrich et al., 1999; Ulrich et al., 1999, 2002) and proton-induced X-ray emission (PIXE; Heinrich et al., 1992; Ryan et al., 1993, 2001; Williams et al., 2001; Baker et al., 2004) techniques have enabled the direct analysis of the compositions of fluid inclusions and provided exciting new insights into the origin and evolution of ore fluids. This paper presents the first multielement chemistry of fluid inclusions from intrusion-related gold systems in the Tintina gold province utilizing the PIXE technique. The data presented include analyses of fluid inclusions from shallow and deep intrusion-related gold systems (Table 1) and build upon previously published fluid inclusion microthermometric studies (Table 2) in order to compare and contrast the chemistry of different brines, carbon dioxide-bearing vapor, and low-salinity, carbon dioxide (and/or methane)-bearing aqueous inclusions, thereby providing additional insights into the origin and evolution of the ore fluids.

Regional Geology

The Tintina gold province is a broad arc-shaped belt that extends for >1,500 km from southwestern Alaska, across central Alaska and Yukon, to the westernmost Northwest Territories (Hart et al., 2002). The Tintina gold province coincides with middle (110–85 Ma) and Late (70–65 Ma) Cretaceous magmatic suites that are spatially and temporally associated with the gold mineralization. The Tintina gold province is underlain by polymetamorphosed Neoproterozoic and Paleozoic

metasedimentary and meta-igneous assemblages of the Yukon-Tanana terrane in west-central Yukon and interior Alaska, dominantly clastic sedimentary rocks of the Neoproterozoic to Carboniferous Selwyn basin in east-central Yukon, and amalgamated basement assemblages of southwestern Alaska, which are overlain by thick, weakly deformed Cretaceous flysch of the Kuskokwim and related basins (Hart et al., 2002). Intruded across these diverse elements is a broad array of middle and Late Cretaceous plutonic belts. In the Yukon and east-central Alaska, several middle Cretaceous plutonic belts were emplaced progressively inboard of the older suites. The youngest (97–91 Ma) and most inboard of these belts comprise the Tombstone and Tungsten plutonic suites in the Yukon and the fault-displaced equivalents in the Fairbanks district of Alaska (Hart et al., 2002). These are the most prolific gold mineralizing suites and are associated with the genesis of the Fort Knox, Brewery Creek, and Dublin Gulch gold deposits, as well as the very large tungsten skarns at Mactung and Cantung. Despite their similarity in age and tectonic setting, the plutonic suites have a wide range of lithological, geochemical, and metallogenic characteristics that range from variably alkalic, metaluminous to peraluminous, and include associated aplite, pegmatite and lamprophyre dikes. Most of these plutons have low magnetic susceptibilities and ferric/ferrous ratios and are dominated by titanite as the major oxide phase. Except for the more oxidized alkalic rocks, these plutons are characterized as ilmenite series (Baker et al., 2005). The intrusions have ϵNd_T values between -7.6 and -15 and initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from 0.709 to 0.720, suggesting a significant crustal component to the magmas (Lang et al., 2000), but locally there are also important contributions from ancient metasomatized mantle.

Geology and Fluid Inclusion Characteristics of Intrusion-Related Gold Systems

Fluid inclusions from eight intrusion-related gold systems were selected for the PIXE study to cover the wide range of geologic environments that host gold mineralization within the Tintina gold province (Table 1). The samples also were selected to be representative of the diverse fluid inclusion types in the intrusion-related gold systems and were well constrained in terms of their relationship to gold (the majority being synore stage) and microthermometric characteristics (Table 2). The following section briefly summarizes the geologic setting of each of the intrusion-related gold systems and their fluid inclusion characteristics. The terminology of Baker and Lang (2001) has been used to classify the fluid inclusions; type 1 inclusions are low-salinity (<10 wt % NaCl equiv) carbon dioxide ($\pm$ methane)-bearing inclusions, type 2 inclusions are low- to moderate-salinity (<20 wt % NaCl equiv) aqueous inclusions, type 3 inclusions are high-salinity, halite ($\pm$ sylvite $\pm$ other daughter phases)-bearing inclusions, type 4 inclusions are carbon dioxide vapor-rich inclusions that coexist with type 3 inclusions, and type 5 inclusions are low-salinity (<10 wt % NaCl equiv) methane ($\pm$ carbon dioxide)-bearing inclusions.

Emerald Lake

The composite Emerald Lake pluton occurs in eastern Yukon (Fig. 1b) where granites of both the Tombstone and Tungsten plutonic suites occur. The Emerald Lake pluton

TABLE 1. Summary of the Geologic and Metallogenic Characteristics of the Selected Intrusion-Related Gold Systems

Deposit/prospect	Gold endowment	Style	Host rock	Metal signature	Pressure (kb), Depth (km)	References
Emerald Lake	Prospect	Sheeted veins	Granite pluton	Au-Bi-Te-As-W	>1.3 , >4.0	Baker and Lang (2001)
Dublin Gulch	50.3 Mt @ 0.9 g/t	Sheeted veins	Granodiorite pluton	Au-Bi-Te-As-W-Sb-Pb-Cu	>1.1 , >3.5	Malloof et al. (2001)
Brewery Creek	13.3 Mt @ 1.4 g/t	Veins/veinlets	Monzonite sills	Au-As-Sb	~ 0.5 , ~ 2.0	Diment (1996), Baker (2002)
Mike Lake	Prospect	Sheeted veins	Syenite stock	Au-Bi-Cu-As-Sb	<0.7 , <2.5	Baker and Lang (2001)
Pogo	10 Mt @ 15 g/t	Flat veins	Gneiss, granite/aplite	Au-Bi-Te-As-Ag-Cu-Pb	>1.7 , >5.0	Smith et al. (1999)
Pogo district	Prospects	Veins	Gneiss, granite/aplite	Au-Bi-Te-As-Ag-Cu-Pb	0.9–1.8, 3.0–6.0	Ebert (2003)
Shotgun	Prospect (~1 Moz)	Breccia, stockwork	Granite stock	Au-Bi-Te-Mo-As-Cu	~ 0.5 , ~ 2.0	Rombach and Newberry (2001)
Donlin Creek	259.6 Mt @ 3 g/t	Veins/veinlets	Rhyolite sills/dikes	Au-Ag-As-Sb-Hg	~ 0.5 , ~ 2.0	Ebert (2003), Goldfarb et al. (2004)

TABLE 2. Summary of the Sample Descriptions and Fluid Inclusion Characteristics of the Selected Intrusion-Related Gold Systems

Deposit/prospect	Sample no.	Sample description	Type	Paragenesis	No. PIXE analyses	Th range (°C)	Salinity (wt % NaCl equiv) ¹	Reference
Emerald Lake	EL96 9, EL9615A, EL9616E	Sheeted quartz veins with py-apy-Au-Bi in quartz-monzonite/ biotite granite; syn-Au	1	P to S	19	208 to 338	0.5 to 10.3	Baker and Lang (2001)
			3	S	7	217 to 284	24 to 45	
Dublin Gulch	DG96 208C 179	Sheeted quartz veins with apy-py-po-sch-Au-Bi in biotite granodiorite; syn-Au	1	P to S	7	205 to 355	0.2 to 9.8	Baker and Lang (2001)
Brewery Creek-Schooner	BC141	Quartz monzonite porphyry with quartz phenocrysts containing fluid inclusions; pre-Au	3	P to S; coexisting with type 4	5	432 to 631	30 to 43	Baker (2002)
			4	P to S; coexisting with type 3	4	418 to 445	2.7 to 11.5	
Mike Lake	ML166, ML167	Sheeted quartz veins within syenite porphyry with apy-py-ccp-tur; syn-Au	3	P; coexisting with type 4	9	277 to 355	27 to 64	Baker and Lang (2001)
			4	P; coexisting with type 3	6	252 to 362	0.9 to 2.8	
Pogo	P158	Quartz vein with apy-py-po-Au-Bi in biotite-quartz-feldspar gneiss	5	P	5	308 to 470	0.5 to 5.0	Smith et al. (1999), Rombach et al. (2002)
			3	S	7	175 to 328	25 to 55	
Pogo district-Spring Grid	00-7710	Quartz vein with apy-py-po-Au-Bi in biotite-quartz-feldspar gneiss; syn-Au (9.65 ppm Au)	1	PS	1	298 to 405	8 to 10	Ebert (2003)
			3	PS	4	290 to 419	33 to 35	
Shotgun	98-15, 80 m	Quartz stockwork with py-ccp-apy-sph in granite porphyry; syn-Au (Au > 700 ppb)	3	P; coexisting with type 4	7	270 to 395	27 to 69	Rombach and Newberry (2001)
			4	P; coexisting with type 3	1	No data	<7	
Donlin Creek-Dome	DC97-392 193.85	Quartz vein with py-ccp-apy in porphyritic rhodocite; synhigh-temperature Au (Au = 90.5 ppm); premain stage Au	3	P; coexisting with type 4	2	265 to 403	up to 74	Ebert (2003)
			4	P; coexisting with type 3	3	320 to 390	Low (<10)	

Abbreviations: apy = arsenopyrite, ccp = chalcopyrite, P = primary, po = pyrrhotite, PS = pseudosecondary, py = pyrite, S = secondary, sch = scheelite, sph = sphalerite, tur = tourmaline
¹ Salinity estimates include ranges from cited references and estimates from this work

belongs to the Tombstone plutonic suite and is zoned from an outer alkalic augite syenite, through subalkalic metaluminous hornblende quartz syenite and hornblende quartz monzonite, to a core of peraluminous biotite granite (Duncan et al., 1998; Duncan, 1999; Coulson et al., 2001, 2002). Ages (U-Pb and Ar-Ar) of intrusive phases and late-stage pegmatitic veins overlap and suggest a duration of magmatic-hydrothermal activity of <100 kyr at ~92.5 Ma (Coulson et al., 2002). Baker and Lang (2001) described the characteristics of fluid inclusions from gold-bismuth-bearing sheeted quartz veins that had a strong spatial and temporal relationship with late-stage aplite dikes, pegmatites, and miarolitic cavities hosted by granite. The veins contain minor sulfide, mostly arsenopyrite and pyrite, with locally coarse bismuthinite, and native gold. Type 1 inclusions are the most abundant and earliest inclusions (Fig. 2a, b) and have homogenization temperatures (T_h) between 208° and 338°C, with salinities between 0.5 and 10.3 wt percent NaCl equiv (Table 2). Carbon dioxide contents are variable (X_{CO_2} = 0.04–1.00), and estimated carbon dioxide densities range from 0.63 to 0.89 g/cm³. Minor methane was detected in some inclusions (X_{CH_4} <0.03) using laser Raman analysis. Type 2 inclusions locally occur with type 1 inclusions but are mostly secondary (T_h = 154°–261°C; salinity = 4.3–16.2 wt % NaCl equiv; Fig. 2b). Spatially separate type 3 (halite-only) inclusions were restricted to secondary trails (T_h = 217°–284°C; salinity = 24–45 wt % NaCl equiv), and type 4 inclusions are absent. Pressure estimates based on halite dissolution temperatures (halite was the last phase to disappear) in type 3 inclusions indicated minimum pressures of 1.3 kbars (Baker and Lang, 2001). Baker and Lang (2001) discussed the difficulties in relating the timing of the different fluid inclusions to the gold event. They suggested an evolving fluid evolution from carbon dioxide-rich type 1 inclusions (type 1A >50% by vol carbon dioxide bubble) through to type 1 inclusions with less carbon dioxide (type 1B <50% by vol carbon dioxide bubble) to late secondary type 2 inclusions. Gold is interpreted to have precipitated as the fluid evolved from carbon dioxide-rich to -poor conditions. The samples that were analyzed by PIXE in this study included 19 inclusions in gold-bearing quartz veins with variable carbon dioxide contents (both 1A and 1B of Baker and Lang, 2001; however, this separation has not been made in the presentation of the PIXE data because of the difficulty in determining gas/liquid ratios through the PIXE optics). The type 1 inclusions are therefore considered to be representative of the key depositional stage for gold. Seven type 3 inclusions from the sheeted veins were also analyzed but are interpreted to have been trapped after deposition of gold (Table 3).

Dublin Gulch

The Dublin Gulch deposit occurs in central Yukon and is hosted by a biotite granodiorite pluton of the Tombstone plutonic suite. The deposit comprises quartz-dominated veins, 1 to 2 cm wide, which contain gold, arsenopyrite, pyrite, pyrrhotite, native bismuth, bismuthinite, and scheelite (Table 1; Hitchins and Orssich, 1995; Baker and Lang, 2001; Maloof et al., 2001). Adjacent to the pluton, within a hornfelsed embayment, lies the Ray Gulch tungsten-skarn deposit (5.4 Mt at 0.82% WO₃; Lenman, 1983). Baker and Lang (2001) reported fluid inclusion characteristics from sheeted veins at

Dublin Gulch. Type 1 inclusions contain variable X_{CO_2} (0.01–0.75; CO₂ density = 0.16–0.9 g/cm³), have low salinity (0.2–9.8 wt % NaCl equiv), and temperatures of homogenization from 205° to 355°C. Methane (X_{CH_4} <0.23) and nitrogen (X_{N_2} <0.12) are locally present, and methane-rich inclusions (type 5) were found in the W skarn (Brown et al., 2001). Type 2 inclusions occur with, and commonly postdate, type 1 inclusions (salinity = 2.1–15.7 wt % NaCl equiv; T_h = 141°–219°C). Rare type 3 inclusions (halite-only) were observed but no microthermometric data was recorded. Minimum pressure estimates of 1.1 kbars were calculated from type 1 inclusions (Baker and Lang, 2001). Baker and Lang (2001) considered that type 1 inclusions were syngold at Dublin Gulch, similar to those described at Emerald Lake. PIXE analysis was carried out on seven type 1 inclusions from the sheeted veins (Table 3).

Brewery Creek

The Brewery Creek gold deposit occurs in west central Yukon and is hosted by alkalic quartz monzonite sills (Lindsay et al., 2000). Gold occurs primarily with arsenopyrite and in arsenic-rich rims in pyrite and locally in quartz-carbonate-pyrite-arsenopyrite filled veinlets. Late stibnite veins postdate the main gold event. Fluid inclusion studies have been carried out in preore quartz-phenocrysts from quartz-monzonite sills (Baker, 2002; Fig. 2c) and from syn- to postore quartz-calcite-pyrite-arsenopyrite and quartz-stibnite veins (unpub. report, Dunne, K., 1995, Brewery Creek: Preliminary fluid inclusion study, Yukon Geology Program and Viceroy Mining Pty.). The phenocrysts contain coexisting type 3 (30–43 wt % NaCl equiv; halite ± unknown daughter phases) and type 4 inclusions (carbon dioxide was identified by low melting temperatures of –57.2° to –58.1°C and rare clathrate formation) with temperatures of homogenization ranging from 432° to 631°C. The inclusions were trapped in the two-phase immiscible field at pressures ~0.5 kbars. Rare fluid inclusions associated with the mineralized veins reported by K. Dunne (1995, unpub. report) are liquid rich (with minor carbon dioxide indicated by crushing) and have low salinity (0–10 wt % NaCl equiv) with temperatures of homogenization ranging from 100° to 350°C. Unfortunately no inclusions of this type were preserved in veins examined in this study, and the PIXE analysis was restricted to type 3 (n = 5) and type 4 (n = 4) inclusions in preore quartz phenocrysts (Table 3).

Mike Lake

The Mike Lake stock is a composite, middle Cretaceous alkalic biotite-hornblende syenite to monzonite porphyry of the Tombstone plutonic suite in west central Yukon (Table 1; Baker and Lang, 2001). Several styles of gold-copper-bismuth-arsenic-antimony mineralization are spatially associated with the Mike Lake pluton and related dikes (Poulsen et al., 1997; Baker and Lang, 2001). Sheeted quartz-arsenopyrite-pyrite-chalcopyrite-gold-tourmaline veins, 1 to 5 cm thick, are found within the main pluton and grade laterally to pegmatitic veins with quartz, K-feldspar, and minor chalcopyrite. Baker and Lang (2001) described fluid inclusions from mineralized, sheeted pegmatitic veins and quartz-arsenopyrite veins from within the intrusion (Table 2). The veins contain primary to secondary type 3 inclusions that commonly host

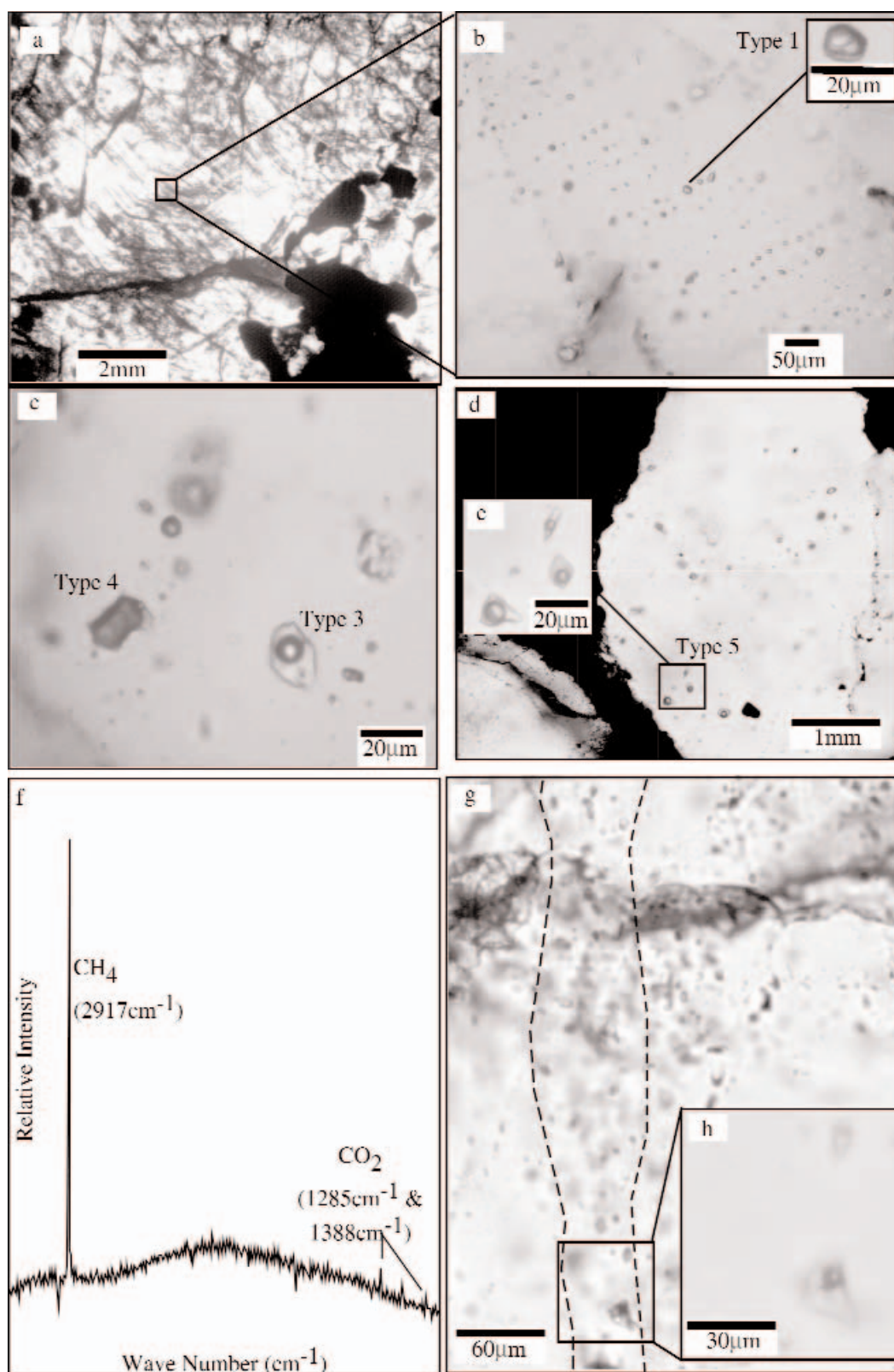


FIG. 2. Photomicrographs of fluid inclusions from various intrusion-related gold systems. a. Primary fluid inclusion populations from Emerald Lake (from Baker and Lang, 2001). b. Type 1 inclusions in primary growth zones; note also type 2 secondary inclusions (from Baker and Lang, 2001). c. Coexisting type 3 and 4 inclusions from quartz phenocrysts at Brewery Creek. d. and e. Primary type 5 inclusions in quartz surrounded by ore-stage sulfides at Pogo. f. Laser Raman spectra from type 5 inclusions at Pogo illustrating their methane-rich nature. g. Secondary trail of late type 3 inclusions (h) from Pogo.

TABLE 3. Results of the PIXE Analysis (ppm unless otherwise stated)

	Analysis	Cl	K	Ca	Mn	Fe	Cu	Zn	As	Br	W	Pb	K/Ca	Mn/Fe	Br/Cl ¹	Zn/Pb
Emerald Lake																
Type 1	el969a	62,000	6,600	683	1,130	811	304	1,180	905	(92)	(269)	(397)	9.66	1.39		
	el969c	40,000 ²	4,850	920	76	468	(68)	158	(13)	(45)	(140)	(100)	5.27			
	el969d	42,100	5,940	1,310	(34)	315	(146)	889	(111)	224	68	(22)	4.53		2.33	
	el969f	47,000	5,600	1,650	589	528	(238)	377	na	(206)	898	na	3.39	1.12		
	el969g	54,800	7,300	841	2,780	9,130	206	983	976	(71)	(100)	(393)	8.68	0.30		
	el16ed	40,000 ²	4,800	1,440	(101)	257	178	181	(58)	na	681	(53)	3.33			
	el16ee	47,100	7,180	2,140	(138)	356	336	515	(137)	(158)	856	(53)	3.36			
	el16eb	60,200	12,700	2,880	(11)	639	149	721	(75)	(2)	(207)	(17)	4.41			
	el969q1	12,100	2,250	824	187	250	150	191	na	(24)	577	na	2.73	0.75		
	el15ah	44,700	4,710	6,500	(3)	1,300	315	362	(72)	na	526	(121)	0.72			
	el15al	49,700	5,260	1,070	(57)	773	222	326	(88)	(12)	418	(28)	4.92			
	el15aj	48,100	9,050	5,750	(31)	1,180	587	2,070	529	(86)	1,840	(398)	1.57			
	el15ai	36,700	5,140	1,310	(170)	387	243	209	(13)	na	626	(120)	3.92			
	el15ag	39,900	6,350	5,840	(147)	662	372	1,090	(247)	(137)	1,380	(175)	1.09			
	el954a	9,530	781	261	(14)	96	45	91	(19)	(35)	196	(36)	2.99			
	el954d	10,200	1,640	1,270	(193)	497	293	435	(58)	(144)	550	na	1.29			
	el954c	3,290	551	(118)	(27)	182	107	157	na	na	294	(167)				
	el16eg	40,000 ²	18,400	2,940	405	2,930	198	605	486	(264)	(549)	(385)	6.26	0.14		
	el16eh	29,500	3,210	(871)	(163)	1,420	367	357	793	na	1,030	(764)				
Type 3	el16ef	272,000	35,100	1,690	na	1,070	792	553	(194)	805	1,390	(22)	20.77		1.29	
	TB-elA	210,000 ²	5,980	3,939	(155)	251	(157)	(146)	(121)	244	na	642	1.52		0.05	
	TB-elB	20,500	18,400	10,069	3,390	8,600	(88)	1,780	176	372	na	1,810	1.83	0.39	7.94	0.98
	TB-elC	188,000	23,700	11,209	2,690	7,480	(142)	1,420	264	381	na	1,632	2.11	0.36	0.89	0.87
	TB-elD	265,000	12,600	3,501	518	1,360	39	809	192	146	na	1,597	3.60	0.38	0.24	0.51
	TB-elI	210,000 ²	163,000	31,927	5,220	7,970	(121)	3,510	108	771	na	4,244	5.11	0.65	0.16	0.83
	TB-elN	210,000 ²	245,000	61,471	9,180	24,500	(194)	3,170	184	746	na	3,647	3.99	0.37	0.16	0.87
	TB-elO	147,000	25,100	7,552	4,370	12,600	(74)	1,850	120	331	na	2,094	3.32	0.35	0.99	0.88
Dublin Gulch																
Type 1	ez179r	40,000 ²	22,500	20,600	3,640	26,200	434	1,920	631	(65)	(357)	(62)	1.09	0.14		
	ez179p	41,000	7,230	1,990	325	2,460	84	491	157	(36)	96	(81)	3.63	0.13		
	ez179q	43,800	5,680	1,240	39	1,050	86	427	160	141	210	(111)	4.58		1.41	
	ez179n	40,500	7,230	3,450	438	3,230	194	551	(117)	(145)	476	(317)	2.10	0.14		
	ez179o	40,000 ²	2,200	812	(40)	138	71	115	na	(25)	168	(16)	2.71			
	ez179m	37,900	6,890	1,450	306	2,130	(66)	277	157	217	344	na	4.75	0.14	2.50	
	ez179h	40,000 ²	29,900	52,000	589	7,290	298	454	(69)	(42)	470	44	0.58	0.08		10.32
Brewery Creek																
Type 3	bc1	210,000 ²	23,600	3,921	2,260	10,800	(56)	780	100	(211)	(119)	741	6.02	0.21		1.05
	bc13	265,000	46,500	17,324	6,026	64,800	278	6,860	276	541	(264)	3,530	2.68	0.09	0.89	1.94
	bc14	241,000	39,700	24,130	4,780	24,700	114	2,700	(61)	480	(164)	2,099	1.65	0.19	0.87	1.29
	bc15	210,000 ²	5,320	2,320	1,670	7,440	(99)	1,970	107	(398)	(219)	1,406	2.29	0.22		1.40
	bc2	237,000	54,500	6,584	2,290	11,700	134	857	93	(399)	(215)	773	8.28	0.20		1.11
Type 4	bc4	46,800	17,700	4,138	726	3,710	(260)	(530)	(207)	(1095)	(564)	(1905)	4.28	0.20		
	bc5	16,200	16,000	2,283	(367)	1,950	(396)	600	891	(1736)	934	(2930)	7.01			
	bc8	69,400	22,100	3,778	1,300	11,900	(171)	621	(128)	(701)	(366)	676	5.85	0.11		0.92
	bc9	18,000	4,900	(651)	(215)	312	(189)	(166)	226	(815)	(385)	(1565)				
Mike Lake																
Type 3	ml167a	305,000 ²	27,300	23,000	4,700	28,400	240	2,260	486	381	(103)	1,290	1.19	0.17	0.05	1.75
	ml167b	305,000 ²	16,300	17,700	3,020	18,900	181	1,460	187	280	(79)	832	0.92	0.16	0.04	1.75
	ml167j	305,000 ²	24,500	22,800	4,550	30,900	257	2,670	410	487	(95)	1,420	1.07	0.15	0.07	1.88
	ml166n	215,000	37,500	7,990	4,750	57,100	785	2,470	660	663	286	1,230	4.69	0.08	1.35	2.01
	ml166k	257,000	24,100	13,000	3,640	36,500	402	2,990	455	655	(194)	2,210	1.85	0.10	1.12	1.35
	ml166l	305,000 ²	28,300	9,080	2,520	32,400	787	1,970	704	544	(142)	1,040	3.12	0.08	0.08	1.89
	ml166j	305,000 ²	36,100	10,900	4,640	49,900	561	3,450	862	560	324	1,930	3.31	0.09	0.08	1.79
	ml166g	386,000	30,500	12,600	10,700	125,000	1,160	7,660	1,760	2,300	(484)	5,000	2.42	0.09	2.61	1.53
	ml166i	163,000	21,500	9,480	2,650	43,200	2,200	3,110	1,190	(226)	1,530	4,300	2.27	0.06		0.72
	ml167e	9,000 ²	2,550	639	129	1,120	623	214	103	(52)	232	na	3.99	0.12		
Type 4	ml167h	9,020	2,530	1,230	151	1,350	619	341	(75)	na	(209)	na	2.06	0.11		
	ml167i	7,090	2,470	1,560	206	1,980	751	350	155	(23)	119	na	1.58	0.10		
	ml166q	9,000 ²	2,540	1,200	726	5,880	229	661	138	170	(128)	322	2.12	0.12	0.83	2.05
	ml167qb	9,000 ²	1,520	420	90	363	na	73	(14)	na	99	(23)	3.62	0.25		
	ml166qc2	2,250	497	169	46	241	na	39	(9)	(3)	(58)	(34)	2.94	0.19		
Pogo																
Type 3	pogo1	157,000	9,780	86,616	2,830	1,390	(48)	508	(39)	262	(107)	477	0.11	2.04	0.73	1.06
	pogo2	210,000 ²	10,600	68,935	28,120	1,560	192	526	(136)	(646)	(385)	(1340)	0.15	18.03		

TABLE 3. (Cont.)

	Analysis	Cl	K	Ca	Mn	Fe	Cu	Zn	As	Br	W	Pb	K/Ca	Mn/Fe	Br/Cl ¹	Zn/Pb
Type 5	pogo4	331,000	14,100	147,027	5,150	2,760	(237)	800	(171)	(909)	(515)	(2020)	0.10	1.87		
	pogo5	140,000	7,710	43,875	1,760	1,030	(117)	350	90	(432)	(244)	(1010)	0.18	1.71		
	pogo6	210,000 ²	7,540	41,926	3,490	2,700	(122)	856	(82)	(446)	(261)	1,090	0.18	1.29		0.79
	pogo7	210,000 ²	4,060	54,019	1,590	985	(80)	407	(49)	(242)	(135)	(513)	0.08	1.61		
	pogo9	120,000	8,220	48,859	911	541	(47)	205	(36)	(169)	(102)	401	0.17	1.68		0.51
	pogo10	30,000 ²	13,000	10,426	432	2,580	(409)	(379)	(346)	(1560)	(877)	(3130)	1.25	0.17		
	pogo11	30,000 ²	8,390	2,873	(360)	1,810	(376)	(352)	(230)	(1390)	(804)	(2810)	2.92			
	TB-PH	27,000	2,560	1,853	(120)	2,180	272	(113)	(279)	187	na	(214)	1.38		3.03	
	TB-PG	25,000	4,850	5,492	(336)	7,150	2,660	(321)	1,560	503	na	(554)	0.88		8.80	
	TB-PJ	3,400	1,280	1,334	(77)	3,060	2,400	98	106	(107)	na	(119)	0.96			
Pogo district																
Type 1	pogo_r4	51,000	19,400	13,728	1,960	21,500	(904)	2,490	(620)	(3316)	(1957)	(6800)	1.41	0.09		
Type 3	pogo_r1	210,000 ²	na	15,162	1,570	10,400	104	2,790	74	(153)	(105)	1,530		0.15		1.82
	pogo_r3	210,000 ²	31,000	19,387	2,350	14,200	(104)	3,850	313	371	(227)	2,340	1.60	0.17	0.08	1.65
	pogo_r8	210,000 ²	19,400	53,291	4,190	27,800	129	6,260	131	477	(190)	3,520	0.36	0.15	0.10	1.78
	pogo_r11	210,000 ²	14,000	11,624	3,340	25,900	138	2,620	70	187	(70)	1,500	1.20	0.13	0.04	1.75
Shotgun																
Type 3	sg1	395,000 ²	70,000	10,961	29,500	142,000	572	6,230	(119)	3,710	(354)	1,450	6.39	0.21	0.41	4.30
	sg2	296,000	42,300	3,562	11,100	60,100	319	3,460	(61)	1,780	(187)	738	11.88	0.18	2.63	4.69
	sg3	279,000	54,700	6,620	20,400	112,000	516	5,780	144	3,120	(301)	953	8.26	0.18	4.89	6.07
	sg11	251,000	33,700	6,207	1,690	110,000	438	5,790	(80)	3,300	(262)	1,350	5.43	0.02	5.75	4.29
	sg12	322,000	56,700	10,024	23,500	149,000	696	7,320	(121)	4,310	(400)	2,180	5.66	0.16	5.86	3.36
	sg9	166,000	34,100	4,771	6,320	43,200	260	2,790	(114)	1,360	(307)	(947)	7.15	0.15	3.58	
	sg7	323,000	56,100	10,705	24,700	158,000	523	8,690	(139)	4,700	(443)	1,350	5.24	0.16	6.37	6.44
Type 4	sg5	19,900	29,500	(1070)	338	4,180	1,830	(409)	(263)	(1620)	(949)	(3500)		0.08		
Donlin Creek																
Type 3	dc1	275,000	85,500	18,424	11,400	187,000	571	16,500	(118)	2,380	(354)	2,180	4.64	0.06	3.79	7.57
	dc3	449,000	142,000	22,275	13,300	164,000	644	16,800	(161)	2,400	(453)	2,090	6.37	0.08	2.34	8.04
	dc2 ³	131,000	65,600	23,442	3,710	60,000	10,500	3,730	1,950	(4100)	21,732	(7130)	2.80	0.06		
	dc7 ³	203,000	19,000	6,402	177	2,370	638	292	(55)	(314)	(170)	(546)	2.97	0.07		
	dc5	51,200	9,860	3,773	436	6,970	136	687	(77)	(385)	(205)	(677)	2.61	0.06		

() = Maximum estimate based on minimum detection limit; na = not analyzed

¹($\times 1000$) mole ratios²Inclusion too deep for accurate PIXE Cl estimate; microthermometry value used instead³High Cl estimate for low salinity inclusion (possible heterogeneous inclusion)

opaque and locally sylvite daughter phases, in addition to coexisting type 4 ($X_{\text{CO}_2} = 0.7\text{--}0.78$), low-density ($0.08\text{--}0.19\text{ g/cm}^3$) vapor inclusions. The two inclusion types have identical ranges of homogenization temperatures, mostly between 250° and 350°C and were trapped at pressures of <0.7 kbars. The coexisting brine and vapor inclusions were interpreted by Baker and Lang (2001) to have been trapped at the time of gold deposition due to the intergrown nature of the quartz and gold-bearing sulfide and localized occurrence of opaque phases within the fluid inclusions. PIXE analysis was carried out on nine type 3 and six type 4 inclusions (Table 3).

Pogo deposit

The Pogo gold deposit, east central Alaska, has been interpreted to be part of the intrusion-related gold deposits class by Smith et al. (1999) but is quite different in style from many of the deposits in the Tintina gold province. Similarities to intrusion-related gold deposits include a strong gold-bismuth correlation, a reduced, low sulfide (<3 vol %) ore mineral assemblage (pyrrhotite, pyrite, and arsenopyrite), and the occurrence of pegmatite and aplite within the deposit.

However, the veins are hosted by gneiss and are located distally (~ 1.5 km) from a major Cretaceous intrusive body rather than within a pluton. The quartz sulfide (pyrrhotite, loellinite, chalcopyrite, bismuthinite, arsenopyrite, and pyrite) veins that host the gold contain primary to secondary type 5 inclusions (Fig. 2d-f) with homogenization temperatures from 308° to 470°C and trapping pressures estimated to be 1.7 to 2 kbars (Smith et al., 1999; Rombach et al., 2002). Late, type 3 inclusions (halite-only) occur in secondary trails and have estimated salinities from 25 to 55 wt percent NaCl equiv and temperatures of homogenization between 175° and 328°C (Fig. 2h, g; Rombach et al., 2002). Type 5 inclusions are considered to be syngold, and type 3 inclusions postgold, based on their respective primary and secondary origins and their relationship to sulfide phases. PIXE analysis was carried out on five type 5 inclusions and seven type 3 inclusions (Table 3).

Pogo district

Ebert (2003) reported on the fluid inclusion characteristics of mineralized quartz veins hosted by gneiss and granite in

the Pogo district. Sample 00-7710 is typical of mineralized veins in the region with gold (9.65 ppm) and anomalous bismuth, arsenic, tungsten, and tellurium (Table 2). It contains abundant type 1 and 3 inclusions (halite $\pm$ sylvite $\pm$ carbonate) that commonly occur in the same quartz zones. Type 5 inclusions are rare, whereas type 2 inclusions are commonly late and secondary. Type 1 and 3 inclusions homogenize over the same temperature range (T_h $\sim$ 300°–400°C) and have salinities of 8 to 10 and 33 to 35 wt percent NaCl equiv, respectively. Pressure estimates range from 0.9 to 1.8 kbars. Ebert (2003) considered that the intimate association of type 1 and 3 inclusions within gold-bearing quartz-arsenopyrite-pyrite-pyrrhotite veins indicated that they were trapped at the time of gold deposition. PIXE analysis was carried out on four type 3 and one type 1 inclusions (Table 3).

Shotgun

The Shotgun deposit in southwest Alaska is hosted in a Late Cretaceous (70 Ma) ilmenite series, I-type granite porphyry stock which has been subjected to intense quartz stockwork veining and brecciation within pervasive albite-sericite-quartz-carbonate alteration (Rombach and Newberry, 2001). Gold occurs within the veins and breccias and is associated with arsenopyrite, pyrite, pyrrhotite, molybdenite, and chalcopyrite and correlates strongly with bismuth, tellurium, molybdenum, and silver ($r^2 > 0.6$). Rombach and Newberry (2001) reported characteristics of ore-stage fluid inclusions and described coexisting type 3 (27 to >60 wt % NaCl equiv; halite $\pm$ sylvite $\pm$ unknown daughter phases) and type 4 inclusions (<7 wt % NaCl equiv; $\pm$ methane) that homogenized at temperatures of 280° to $>600^\circ\text{C}$ (limit of the heating-freezing stage). Pressures of ~ 0.5 kbars were estimated for the entrapment of the immiscible phases. Seven type 3 and one type 4 inclusions were analyzed by PIXE (Table 3).

Donlin Creek

The Donlin Creek deposit in southwest Alaska is hosted by rhyolite sills and dikes (Ebert, 2003; Goldfarb et al., 2004). There are several mineralized zones including the high-temperature porphyry-style mineralization at the Dome and the epithermal- and/or epizonal-style mineralization in the ACMA-Lewis area (the largest part of the resource; Ebert, 2003; Goldfarb et al., 2004). The Dome mineralization is characterized by gold, arsenic, silver, bismuth, copper, zinc $\pm$ tellurium in high-temperature quartz veins containing arsenopyrite, pyrrhotite, pyrite, and iron-rich sphalerite. Fluid inclusions in the Dome area comprise immiscible type 4 ($\pm$ methane $\pm$ nitrogen) and type 3 inclusions (up to 74 wt % NaCl equiv), which were trapped between $\sim 300^\circ$ and 400°C and <0.5 kbars. However, the majority of gold occurs in narrow veinlets in low-temperature illite-kaolinite alteration and is associated with anomalous arsenic, antimony, and mercury (ACMA-Lewis area). The gold-bearing quartz-carbonate-arsenopyrite-stibnite veins contain type 1 (2–6 wt % CO_2) fluid inclusions that have temperatures of homogenization from 140° to 220°C and low salinities (<5 wt % NaCl equiv; Ebert, 2003; Goldfarb et al., 2004). Only type 3 ($n = 2$) and type 4 ($n = 3$) inclusions from the Dome area were available for PIXE analysis in this study (Table 3) and are considered to belong to the ore stage in the Dome zone of Donlin Creek but are

unrelated to the main ore-stage fluid in the main part of the deposit (the ACMA-Lewis area).

PIXE Analysis

Methodology

PIXE analysis was carried out on the CSIRO-GEMOC nuclear microprobe, North Ryde, Australia. Details of the technique are reported in Ryan et al. (1993, 1995, 2001). Analyses for this study were performed on carbon-coated doubly polished fluid inclusion sections and individual fluid inclusions were analyzed for 10 to 15 minutes, using a 3 MeV proton beam with a spatial resolution of 1 to $2\ \mu\text{m}$. The average concentration of elements $Z > 17$ in the trapped fluid is calculated by the method of Ryan et al. (1993, 2001). Reported errors of ~ 30 percent include analytical error of 10 to 15 percent and geometry-related errors, such as estimates of depth, thickness, and irregularity of natural inclusion shapes from the modeled ellipsoid (cf. Heinrich et al., 1992; Ryan et al., 2001). Interelement ratios, particularly for elements with similar atomic numbers, are more accurate than estimates of actual concentrations because the main sources of error have similar effects on all elements. Errors for lighter elements, particularly chlorine, are larger (up to 50%) due to the attenuation of X-rays by the host mineral, particularly in inclusions deeper than $10\ \mu\text{m}$. In this study, analyses with chlorine contents that were initially found to deviate significantly from the salinities determined by microthermometry have been corrected, using a procedure similar to that which has been applied for the internal standardization of LA-ICP-MS fluid inclusion analyses (Heinrich et al., 2003; Table 3).

Important elements such as sodium and sulfur cannot be measured quantitatively ($Z < 17$), however, monitoring of gamma-ray emissions (PIGE) can provide an indication of sodium distribution within the fluid inclusion. Limits of detection are further dependent on the depth of the inclusions in the sample and for elements with $Z > 25$ are typically 30 to 300 ppm. Ore-related elements of interest for intrusion-related gold systems include gold, bismuth, silver, and antimony, all of which are likely to be present at concentrations ranging from parts per billion to a few parts per million (tens of ppm at most). Consequently, none of these elements were detected using the PIXE technique and may be better determined by using LA-ICP-MS (cf. Ulrich et al., 1999). Metals such as copper, zinc, lead, arsenic, and tungsten were commonly detected (Table 3).

A distinct advantage of the PIXE technique over LA-ICP-MS is the ability to image the distribution of elements in the inclusions and the ability to exclude solid phases in the quartz host that could otherwise contaminate the analysis. However, not all phases containing detectable elements are resolved in images. Quantitative assessment is required on the actual X-ray spectra. Figure 3 shows examples of the images generated from a low-salinity, type 5 inclusion (Fig. 3a), a type 3 brine inclusion (Fig. 3b), a type 4 vapor-rich inclusion (Fig. 3c), and a late secondary type 3 inclusion (Fig. 3d). The PIXE image of sample TB_PG from Pogo shows the inclusion area being defined by chlorine. A solid copper phase can be distinguished in the image which coincides with elevated arsenic. It is not clear whether this occurs within the inclusion or directly adjacent to and/or below it within the quartz host, but sample TB_PJ shows a similar overlap suggesting that the

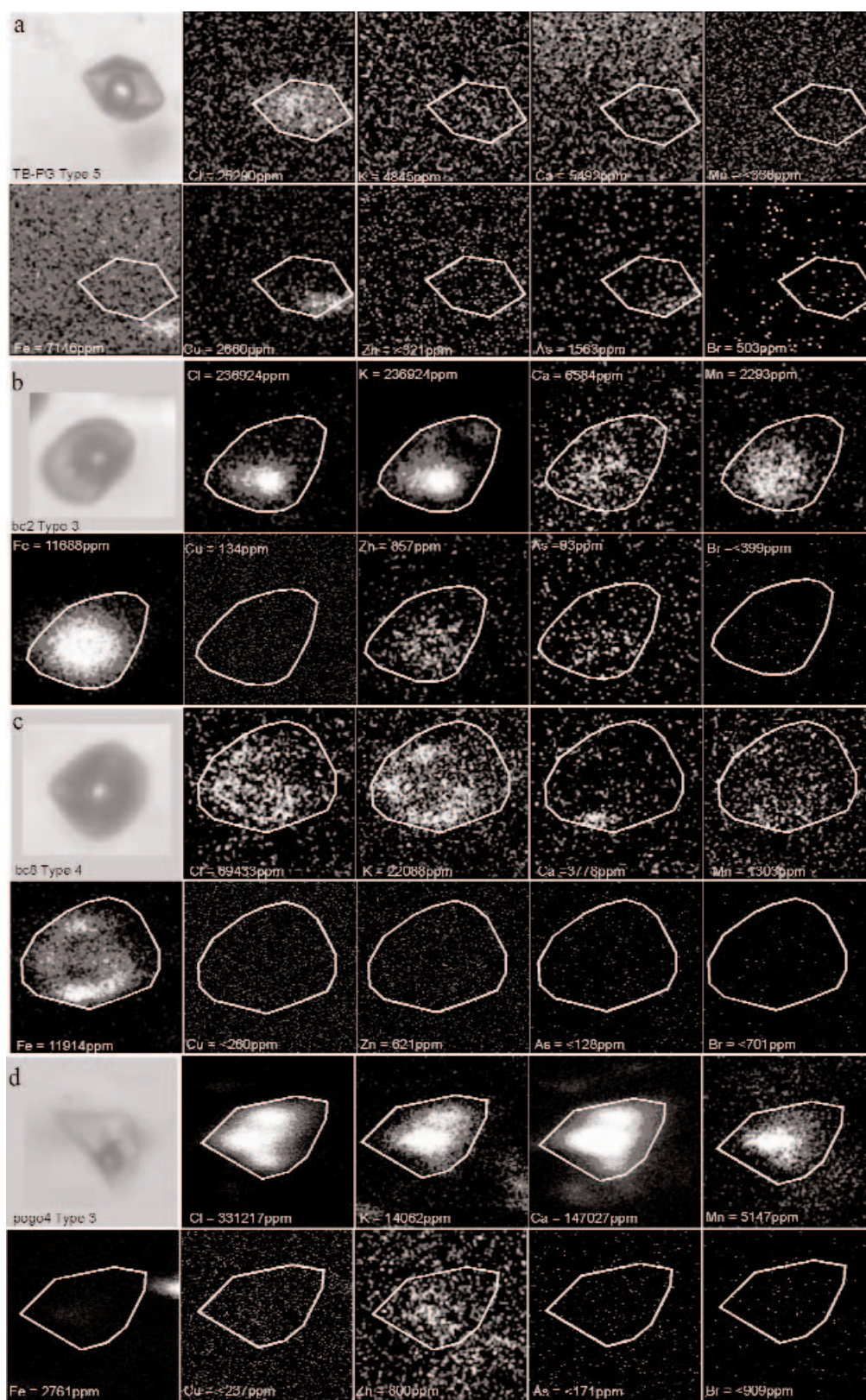


FIG. 3. PIXE images (each box = $25 \times 25 \mu\text{m}$ for scale) showing the distribution of Cl, K, Ca, Mn, Fe, Cu, Zn, As, and Br in a variety of fluid inclusion types including: a. Type 5 inclusion from Pogo (note Cu-As-Fe solid phase). b. Type 3 inclusion from Brewery Creek (note high Cl coincident with halite and sylvite (high K) signal). c. Type 4 inclusion from Brewery Creek (note high solute content of liquid rim around the edge of the vapor phase). d. Secondary type 3 inclusion from Pogo (note the high Ca content compared to type 3 inclusion from Brewery Creek).

solid phase is common to both inclusions (Table 3). Petrographic reexamination of the fluid inclusions could not distinguish any opaque phase, either within the inclusion or in the quartz host, however, the inclusions measured 7 and 13 μm in their maximum length direction, respectively, and consequently the $\sim 1\text{-}\mu\text{m}$ -diameter solids (estimated from the PIXE images) could have been masked by the dark edges of the inclusions. The inclusions occur in primary clusters in quartz surrounded by pyrrhotite, loellingite, chalcopyrite, arsenopyrite, and pyrite, suggesting that they were directly related to the ore stage (Fig. 2d-e). Other inclusions in the same cluster do not show this high concentration of copper, arsenic, and iron (Table 3). Therefore, if the solid phases do occur within the inclusions they may represent accidentally trapped solid sulfide phases that precipitated during ore deposition. Sample bc2 from Brewery Creek illustrates the high chlorine con-

centrations associated with halite and sylvite in type 3 inclusions, whereas all other elements appear to be within the liquid phase (Fig. 3b). High concentrations of solutes are observed in the liquid rim around the large vapor bubble in the type 4 inclusion from Brewery Creek (Fig. 3c). The type 3 inclusion from Brewery Creek contains high concentrations of calcium compared to potassium and has much lower K/Ca ratios than the type 3 inclusion from Brewery Creek (see below).

Major elements

Major components of all fluid inclusion types analyzed by PIXE include potassium, calcium, iron, and manganese (Table 3). Two broad groups of fluid inclusions, with some overlap, can be distinguished based on these major elements; group one comprises the high-salinity, halite-bearing type 3 inclusions (Fig. 4a-b) and group two consists of all low-salinity

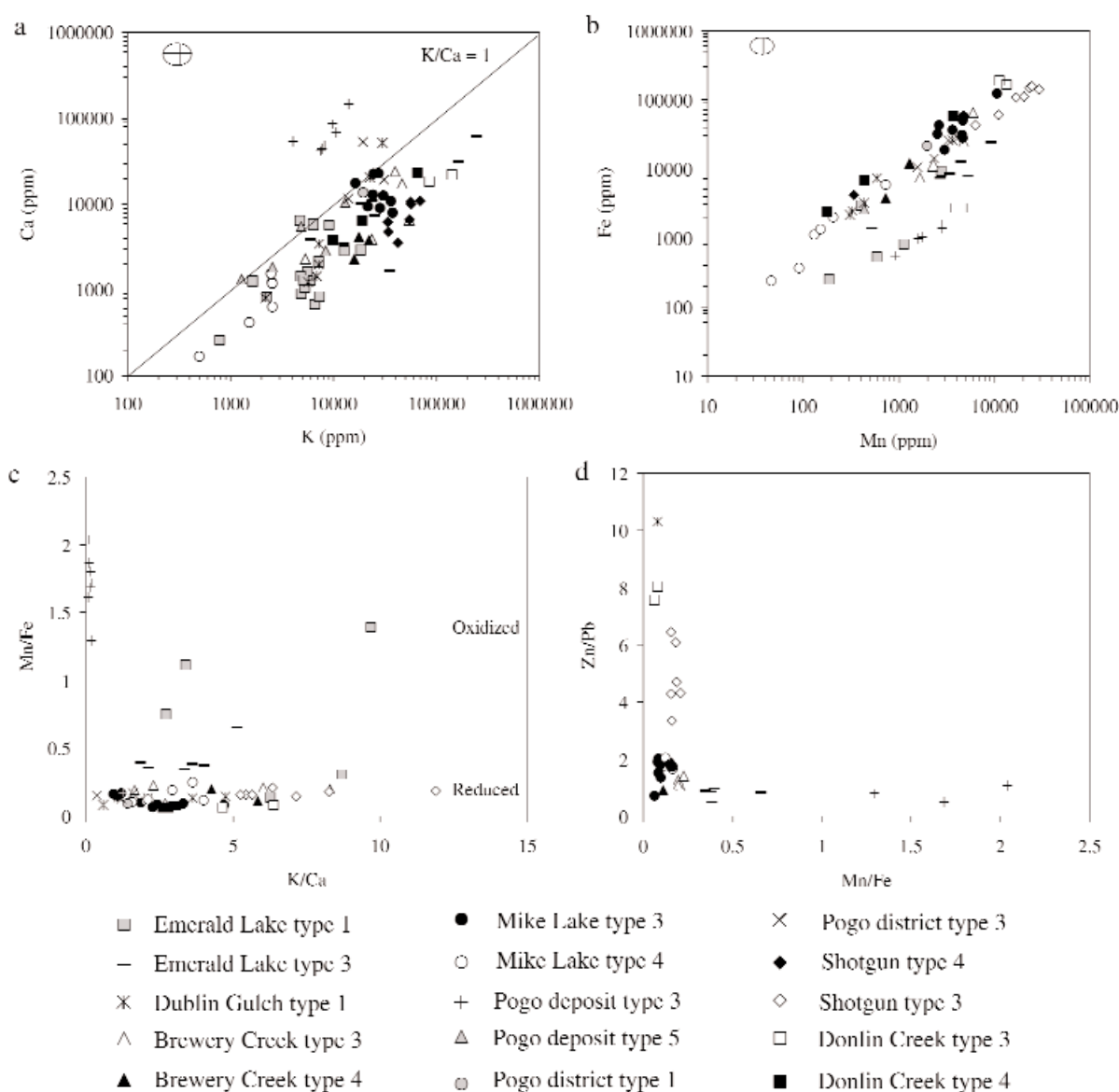


FIG. 4. Plots of the concentrations of major cations in fluid inclusions from Yukon and Alaska intrusion-related gold systems. a. K and Ca. b. Fe and Mn. c. K/Ca and Mn/Fe ratios (Mn/Fe ratio redox estimates from Bottrell and Yardley, 1991). d. Mn/Fe and Zn/Pb ratios. Error bars = 30 percent.

inclusion types (1, 3, and 5; Fig. 4a-b). In general type 3 inclusions contain between 10,000 and 100,000 ppm potassium and calcium, whereas the low-salinity inclusions have concentrations ranging mostly from 1,000 to 10,000 ppm. Both groups have K/Ca ratios of ~ 1 (wt ratios have been used throughout the paper unless stated otherwise), excluding secondary type 3 inclusions from the Pogo deposit (K/Ca = 0.08–0.18), one type 3 inclusion from the Pogo district, and one type 1 inclusion from Dublin Gulch (Fig. 4a). Type 3 inclusions from the two southwestern Alaska deposits, Donlin Creek and Shotgun, have some of the highest K/Ca ratios (4.64–11.87), whereas type 3 inclusions from Brewery Creek (K/Ca = 1.64–8.28) and Mike Lake (K/Ca = 0.92–4.69) span a lower range. Secondary type 3 inclusions at Emerald Lake (K/Ca = 1.52–5.10; excluding one analysis of 20.77) have much higher ratios than similar secondary type 3 inclusions from Pogo. Low-salinity inclusions mostly have K/Ca ratios between 1 and 5.

The majority of type 3 inclusions have manganese and iron concentrations between 1,000 and 10,000 ppm and 10,000 and 100,000 ppm, respectively. Shotgun (6,320–29,500 ppm Mn and 43,210–158,000 ppm Fe) and Donlin Creek (11,400–13,300 ppm Mn and 187,000–164,000 ppm Fe) have the highest concentrations. Mn/Fe ratios for both the high- and low-salinity populations are very narrow (0.06–0.22), excluding inclusions from Emerald Lake and type 3 inclusions from Pogo (Fig. 4b-c). The latter are again anomalous, with very high Mn/Fe ratios (1.29–2.04). Mn/Fe ratios (0.35–0.65) in type 3 inclusions from Emerald Lake are lower than in type 3 inclusions from Pogo but higher than inclusions from other intrusion-related gold systems. Type 1 inclusions from Emerald Lake have a wide range of Mn/Fe ratios, from 0.14 to 1.39, but two thirds of the manganese analyses are below detection limits, indicating that the majority of type 1 inclusions from Emerald Lake have very low Mn/Fe ratios (Table 3).

Trace metals

Zinc concentrations correlate with salinity (Fig. 5a); most type 3 inclusions have >1,000 ppm Zn, whereas concentrations in the low-salinity inclusions range from below detection to $\sim 1,000$ ppm Zn. Secondary type 3 inclusions from Emerald Lake and, in particular, Pogo form a distinct group with relatively low zinc compared to other type 3 inclusions. Lead concentrations likely have a similar relationship to salinity, however, detection limits for lead are typically higher than for zinc using the PIXE method, and therefore the data from the lower salinity inclusions are more variable (Table 3). Secondary type 3 inclusions from Emerald Lake and Pogo again are distinctly different from other inclusion types when ratios are evaluated (Fig. 4d). The inclusions have very low Zn/Pb ratios (0.51–0.98 and 0.51–1.06, respectively), reflecting their anomalously low zinc contents and high Mn/Fe ratios. Type 3 inclusions from Mike Lake (Zn/Pb = 0.72–2.01), Brewery Creek (Zn/Pb = 0.92–1.94), and the Pogo district (Zn/Pb = 1.65–1.82) have low Mn/Fe and low Zn/Pb ratios, whereas type 3 inclusions from Shotgun (Zn/Pb = 3.35–6.45) and Donlin Creek (Zn/Pb = 7.59 and 8.05) have high Zn/Pb and low Mn/Fe ratios.

Copper and arsenic concentrations are below 1,000 ppm in the majority of inclusions and show no systematic variation

with salinity (Fig. 5b-c). Heinrich et al. (1999) have shown that both copper and arsenic partition preferentially into the vapor phase rather than the brine in porphyry copper environments. Based on the data presented here, from intrusion-related gold systems that contain coexisting type 3 and type 4 inclusions, it is difficult to assess whether there is any significant enrichment of copper or arsenic in the vapor inclusions. Concentrations of copper in type 3 inclusions at Brewery Creek are low to below detection and average 175 ppm Cu for values above detection. Arsenic concentrations are similar, with an average concentration of 144 ppm. Concentrations of copper in coexisting type 4 inclusions at Brewery Creek are all below detection; two analyses of arsenic were above the detection limit (891 and 226 ppm). Type 3 inclusions at Mike Lake all contain detectable copper and arsenic with similar concentrations that average 730 and 746 ppm, respectively. Type 4 inclusions average 556 ppm Cu and 132 ppm As. Type 3 inclusions contain on average 475 ppm Cu and 144 ppm As; however, only one vapor inclusion was analyzed, which had very high copper (1,831 ppm) but arsenic concentrations below detection. Type 3 inclusions in the Dome area of Donlin Creek contain an average of 608 ppm Cu, but arsenic concentrations are below detection. The high chlorine contents of two of the three type 4 inclusions analyzed suggest that they trapped mixed brine and vapor, and therefore it is uncertain whether the high copper and arsenic in these inclusions were concentrated in the brine or vapor phases. The one lower salinity vapor inclusion contained 136 ppm Cu, with arsenic concentrations below detection.

Copper concentrations in three type 5 inclusions from Pogo are below detection. However, two inclusions contained significant copper (2660 and 2410 ppm), likely trapped as solid phases (see above). Copper concentrations in late, secondary type 3 inclusions from Pogo range from below detection to up 138 ppm, and a single type 1 inclusion from the same sample contained no copper. Type 1 inclusions from Emerald Lake and Dublin Gulch contain tens to a few hundred parts per million copper (up to 587 ppm). Copper in late secondary, type 3 inclusions from both Emerald Lake and Pogo was detectable in only two inclusions each (39 and 792 and 80 and 192 ppm, respectively). Arsenic concentrations in type 1 inclusions from Emerald Lake (<13–905 ppm) and Dublin Gulch (<69–631 ppm) vary from below detection to several hundred parts per million. Type 5 inclusions from Pogo contain variable arsenic, similar to copper described above. Late, secondary, type 3 inclusions from Emerald Lake contain <108 to 264 ppm As, whereas arsenic concentrations in late type 3 inclusions from Pogo are all below detection, excluding one value of 90 ppm. Elevated tungsten only occurs in inclusions from the Yukon (excluding one highly anomalous value from Donlin Creek; Fig. 5d). In particular, tungsten is enriched in type 1 inclusions from Dublin Gulch (96–476 ppm) and Emerald Lake (68–1840 ppm), and type 3 (<79–1530 ppm) and type 4 (<58–232 ppm) inclusions from Mike Lake.

Halogens

Bromine and chlorine have been used widely to evaluate potential sources of salinity in fluids (Fig. 6). The PIXE method is able to measure the concentrations of both these elements, however, careful analysis of the data is required due

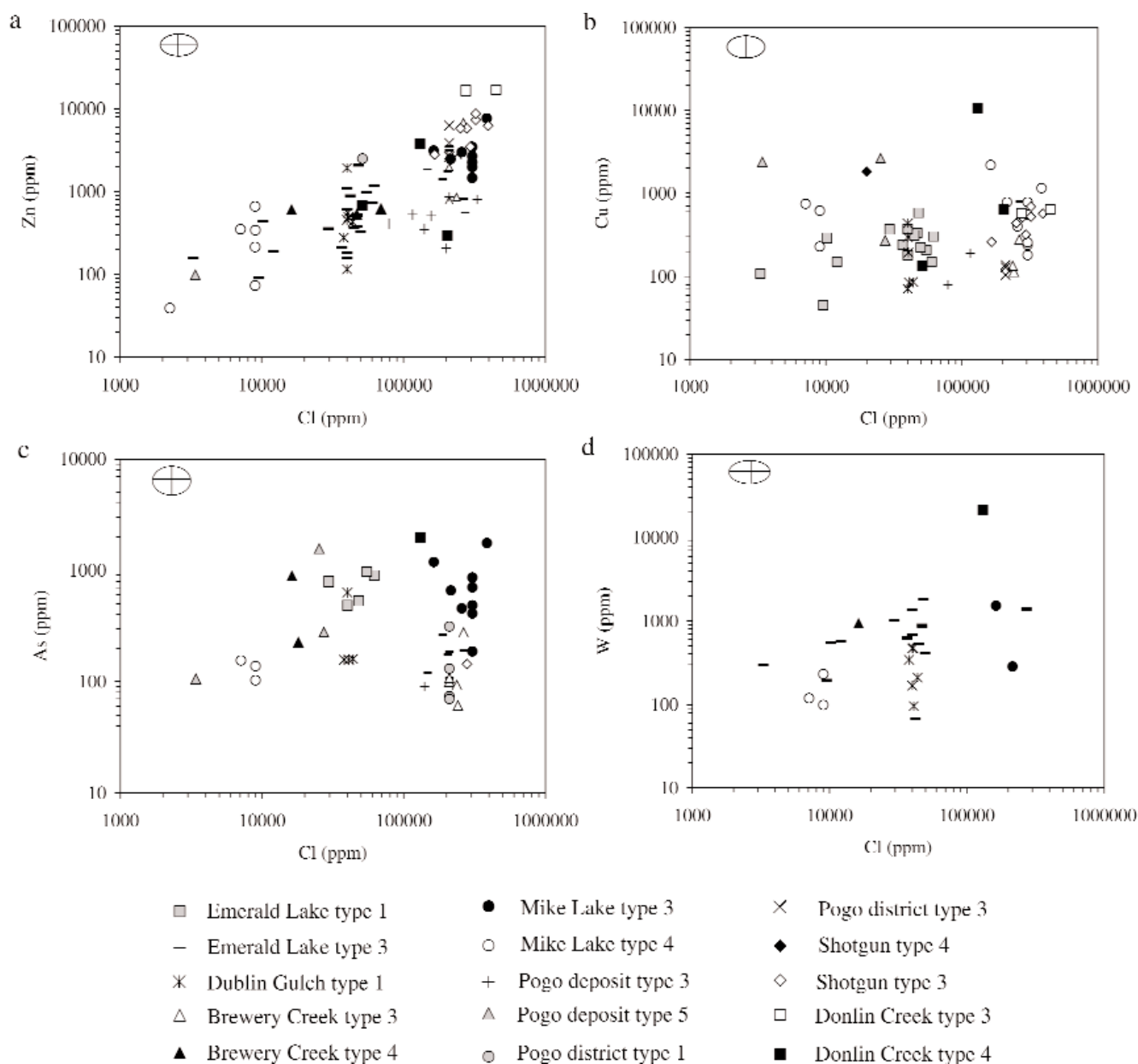


FIG. 5. Plots of the concentrations of chlorine and metals in fluid inclusions from Yukon and Alaska intrusion-related gold systems. a. Zn. b. Cu. c. As. d. W. Error bars = 30 percent.

to the problems of absorption of X-ray from chlorine in deeper inclusions and the commonly low concentration of bromine in many crustal fluids, particularly in low-salinity fluids. Only six low-salinity inclusions had detectable bromine, but all were within analytical error of their detection limits and are therefore not considered further. Halogen data from high-salinity inclusions are more reliable due to the higher concentrations of these elements. Two distinct populations can be discerned; type 3 inclusions from Shotgun ($\text{Br/Cl} = 2.63 \times 10^{-3}$ – 6.37×10^{-3}) and Donlin Creek ($\text{Br/Cl} = 2.34 \times 10^{-3}$ and 3.79×10^{-3}) have high Br/Cl mol ratios whereas Pogo district ($\text{Br/Cl} = 0.39 \times 10^{-3}$ – 0.77×10^{-3}), Mike Lake ($\text{Br/Cl} = 0.4 \times 10^{-3}$ – 1.35×10^{-3} excluding one value of 2.61×10^{-3}) and Brewery Creek ($\text{Br/Cl} = 0.87 \times 10^{-3}$ and 0.90×10^{-3})

have lower ratios. Interestingly, secondary type 3 inclusions from Emerald Lake ($\text{Br/Cl} = 0.24 \times 10^{-3}$ – 1.61×10^{-3}) and Pogo ($\text{Br/Cl} = 0.73 \times 10^{-3}$) also fall within this group.

Discussion

Fluid types, metals, and metal transport in intrusion-related gold systems

The data presented in this study indicate that diverse fluid types were present in intrusion-related gold systems of Alaska and Yukon. Deposits emplaced at shallow crustal levels (<3 km) such as Shotgun, Donlin Creek Dome area, Mike Lake, and Brewery Creek contain coexisting type 3 and 4 inclusions that were trapped at moderate-high temperature ($\sim 300^\circ$ –

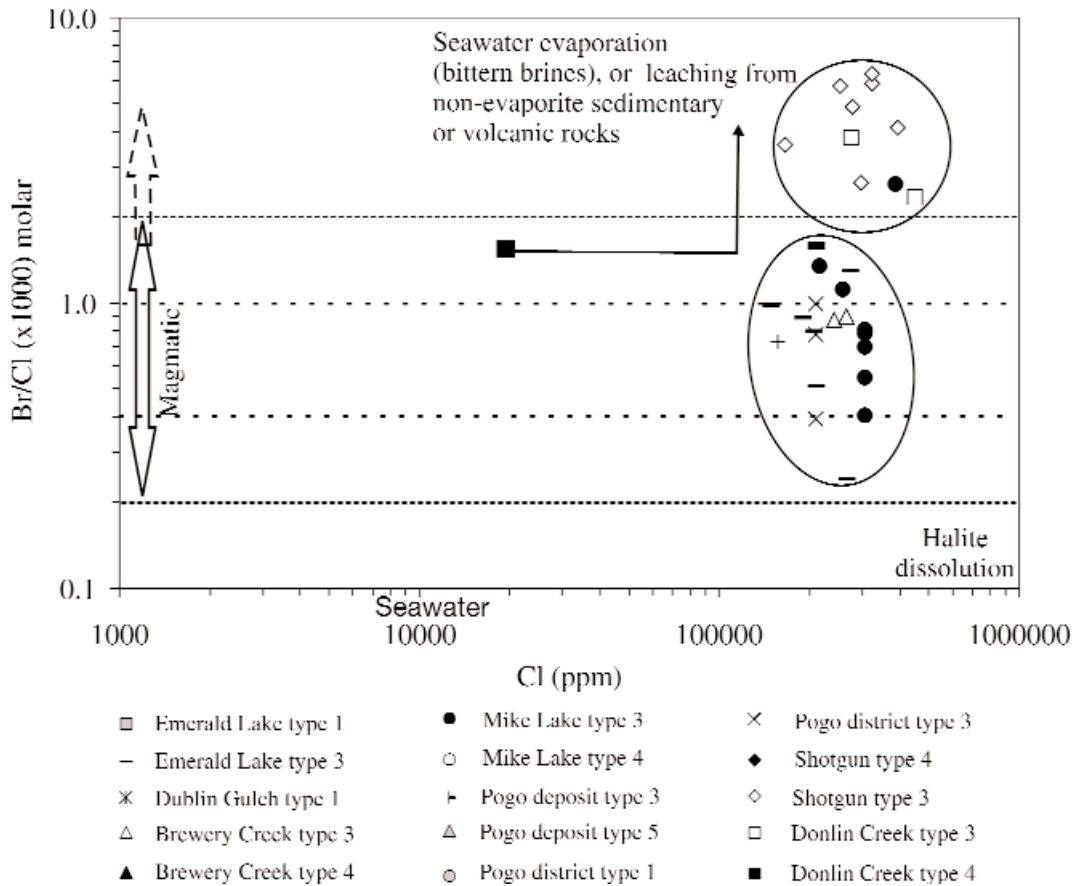


FIG. 6. Plot of Cl vs. Br/Cl ($\times 1,000$) mol ratios in fluid inclusions from Yukon and Alaska intrusion-related gold systems. Fields of different fluid reservoirs are from Bohlke and Irwin (1992), and the seawater evaporation trajectory is from Hermann et al. (1973).

500°C). At Shotgun and Mike Lake, these inclusions are considered to have been trapped at the time of gold deposition. In the Dome area of Donlin Creek the inclusions are interpreted to have been part of a different gold event than the main gold mineralization in the ACMA-Lewis zone. At Brewery Creek the inclusions with immiscible fluid were trapped prior to the main gold stage. Gold-bearing veins in the Pogo district are also characterized by both low-salinity, carbon dioxide-bearing type 1 inclusions and high-salinity type 3 inclusions but were likely formed at moderate to deep levels (Ebert, 2003; Table 1). Deeper systems (>3 km) such as Emerald Lake, Dublin Gulch, and Pogo contain low-salinity, carbon dioxide- and methane-rich type 1 and 5 inclusions that were trapped at moderate temperature at the time of gold mineralization (Smith et al., 1999; Baker and Lang, 2001; Rombach et al., 2002). Both Pogo and Emerald Lake (and locally at Dublin Gulch; Baker and Lang, 2001) also contain a compositionally distinct set of late secondary type 3 inclusions that postdate gold mineralization.

The diversity of fluid types and conditions of mineralization imply a range of transport mechanisms for metals. In particular, there is a wide variation in chloride concentration both within individual deposits and across the spectrum of shallow and deep intrusion-related gold systems. The range of salinities found here is consistent with findings of previous studies

(Smith et al., 1999; Baker and Lang, 2001; Rombach and Newberry, 2001; Baker, 2002; Rombach et al., 2002; Ebert, 2003). In the shallow systems the variation is mostly due to phase separation. In the deeper systems, type 3 high-salinity brines at Emerald Lake and Pogo postdate gold mineralization and are likely related to a separate fluid event (see below). The low-salinity type 1 and 5 fluid inclusions in the deeper systems range from ~0.5 to 10 wt percent NaCl equiv (~3,000–60,000 ppm chlorine), a common range for many gold-only deposits in orogenic settings (Ridley and Diamond, 2000).

The presence of immiscible brine and vapor inclusions in shallow-level intrusion-related gold systems is similar to that of porphyry copper deposits where phase separation has been widely documented as an important process in concentrating and transporting copper and gold (e.g., Heinrich et al., 1999; Ulrich et al., 1999, 2002). These fluids commonly contain in excess of 1,000 ppm Cu to in some cases percent levels and in many cases show greater enrichment of copper, gold, and arsenic in the vapor phase. Copper contents of ores in shallow intrusion-related gold systems are low compared to porphyry copper deposits (mostly <1,000 ppm), which implies that intrusion-related gold systems are formed from copper-poor fluids and should not be considered variants of porphyry copper deposits (e.g., Rowins, 2000). Because copper concentration in the brine inclusions are not consistently higher than in

the vapor inclusions, the concentration of chlorine is apparently not the chief control on copper concentrations. This is further supported by the copper and arsenic concentrations in low-salinity type 1 inclusions in the deeper intrusion-related gold systems. In contrast to copper and arsenic, the concentrations of iron, manganese, zinc, and lead are strongly dependent on the salinity of the fluid (e.g., Fig. 5a). Tungsten on the other hand is anomalous in its absence from the majority of high-salinity inclusions; the highest concentrations were found in low-salinity, carbon dioxide-bearing type 1 inclusions from Emerald Lake and Dublin Gulch in the Yukon (Fig. 5d).

Wood and Samson (2000), in their review of the hydrothermal chemistry of tungsten deposits, suggested that chloride complexes were unimportant for tungsten transport, with tungstate (WO_4^{2-}) being the main solute species. Seward and Barnes (1997) also suggested that reduced sulfur-bearing fluids may form thio complexes with tungstate ($\text{WO}_{4-n}\text{S}_n^{2-n}$). Reduced sulfur has also been implicated in the transport of arsenic (e.g., thioarsenite; $\text{As}_3\text{S}_6^{3-}$), copper as $\text{Cu}(\text{HS})_2^-$, and gold as $\text{Au}(\text{HS})_2^-$ and AuHS^0 (Seward and Barnes, 1997). There are several lines of evidence that support a reduced environment in intrusion-related gold systems: (1) the granite host rocks are reduced (Baker et al., 2005), (2) the ore is characterized by reduced sulfide assemblages, (3) methane is commonly present in ore-related fluid inclusions, and (4) H_2S has been detected in some inclusions using laser Raman techniques (Baker and Lang, 2001). Low Mn/Fe ratios are also indicative of reduced conditions in chlorite-altered host rocks (Bottrell and Yardley, 1991). Chlorite is found in alteration envelopes around gold-bearing veins in intrusion-related gold systems where it commonly replaces igneous biotite and amphibole. All of the inclusions in this study, except for the late secondary type 3 inclusions from Pogo and Emerald Lake, have Mn/Fe ratios of <0.24 consistent with reduced conditions. It is therefore possible that reduced sulfur complexes were important for the transport of tungsten, arsenic, copper, and gold in the fluids studied here.

Fluid sources

Br/Cl mol ratios presented in this study define two populations of high-salinity inclusions that reflect different geographic locations (Fig. 6). The highest Br/Cl mol ratios occur in type 3 inclusions from the southwestern Alaska deposits, Shotgun and Donlin Creek, and the lowest Br/Cl ratios occur in deposits located in Yukon and east central Alaska (Pogo district). Goldfarb et al. (2004) have shown that felsic intrusions in the Donlin Creek area were primarily derived from fine-grained sedimentary rocks of the Kuskokwim (nonevaporite-bearing), flysch basin. Halogens derived from such an environment would likely have elevated Br/Cl ratios, whether they were leached from the surrounding host rock or exsolved into magmatic volatiles following crustal melting (Bohlke and Irwin, 1992). The Shotgun prospect is also hosted by the Kuskokwim group and type 3 inclusions may have similarly derived bromine and chlorine from the sedimentary rocks. However, there is no radiogenic isotope data available to evaluate the origins of granites at Shotgun. The Br/Cl ratios of type 3 inclusions from the Pogo district, Mike Lake (excluding one value that overlaps with the southwestern Alaskan systems) and Brewery Creek are consistent with a magmatic source

(Bohlke and Irwin, 1992). Radiogenic isotopes indicate a significant sedimentary crustal component to these intrusions (Lang et al., 2000), but any halogen component from this source appears to be distinct from that of southwestern Alaska.

The late, secondary type 3 inclusions at Emerald Lake and Pogo have Br/Cl ratios similar to type 3 inclusions in the Pogo district and at Mike Lake and Brewery Creek. However, they are chemically distinct in other ways, including higher Mn/Fe ratios and lower Zn/Pb ratios (Fig. 4c-d). Type 3 inclusions from the Pogo deposit also have very low K/Ca ratios. The high Mn/Fe ratios are indicative of more oxidized conditions and support the suggestion from the paragenesis that these fluids are unrelated to the main ore stage at Emerald Lake and Pogo. The low-temperature, oxidized nature of the fluids and low Zn/Pb and K/Ca ratios (at Pogo) are more consistent with nonmagmatic, basinal-type fluids (Yardley et al., 2000), although this is inconsistent with the halogen ratios. Alternatively, these fluid inclusions may have been derived from distal (and/or deep seated) magmas that cooled and equilibrated with granites (cf. Whitney et al., 1985) and/or country rocks before infiltrating along fractures and veins to be trapped in secondary planes. All other type 3 inclusions have high K/Ca ratios (>1) as do the three low-salinity inclusion types. Both groups also have similar Mn/Fe ratios, consistent with granite-derived or equilibrated fluids. Shotgun and Donlin Creek type 3 inclusions have the highest K/Ca and Zn/Pb, most likely reflecting the very saline nature and high temperature of this particular group of inclusions.

Conclusions

PIXE analyses reveal higher concentrations of iron, manganese, zinc, and lead in the shallow intrusion-related gold systems due to the greater abundance of high-salinity fluids than in the deeper systems. Tungsten, however, is mostly deficient in the high-salinity inclusions but abundant in low-salinity, carbon dioxide-bearing fluid inclusions in deeper level deposits. This is consistent with the high tungsten contents of these systems and may reflect tungsten's preference to form tungstate complexes rather than chloride complexes. Copper and arsenic have similar concentrations in both low- and high-salinity inclusions suggesting other complexes were important. Sulfur complexing may explain why both shallow and deep deposits contain gold despite the wide variation in salinity and different fluid types present. Bromine and chlorine ratios for high-salinity inclusions support a magmatic origin for the salinity, whereas saline fluids at Shotgun and Donlin Creek in southwestern Alaska have distinctly higher Br/Cl mol ratios that may reflect the local sedimentary crustal source of the granite melts (Goldfarb et al., 2004).

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