

The flux of gold and related metals through a volcanic arc, Taupo Volcanic Zone, New Zealand

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ABSTRACT

We determined the concentrations of gold, silver, arsenic, antimony, and mercury in deep hydrothermal solutions (~1 km depth, 200 to >300 °C) from six geothermal systems and calculated their fluxes for a 250-km-long segment of volcanic arc in the Taupo Volcanic Zone, New Zealand. The three orders of variation in the concentrations (<0.1–23 ppb Au; 2.7–2400 ppb Ag) and fluxes of precious metals across the arc indicate that the supply of metals in deep hydrothermal solutions is influenced by the intrusion of andesitic and basaltic magmas, which also govern the convective heat transfer and flux of metals. Two of the geothermal systems, Rotokawa and Mokai, have the highest hydrothermal fluxes of gold and/or silver known, and can supply enough metal in ~50,000 yr or less to match the inventories of the largest hydrothermal ore deposits in the world; although, so far, no ore mineralization has been found in these systems. If the huge amounts of hydrothermal precious metals (minimum values range 80–163 kg Au/yr and 6800–13,850 kg Ag/yr) that flow through the Taupo Volcanic Zone are representative of all arcs throughout time, then focused fluid flow and efficient metal deposition are the most important processes governing the formation of gold and silver ore deposits.

Keywords: gold, silver, hydrothermal, volcanic arcs, metal fluxes.

INTRODUCTION

Precious-metal ore deposits (including epithermal and porphyry styles) are valuable economic products of hydrothermal activity and subduction zone magmatism in modern and ancient volcanic arcs (Hedenquist and Lowenstern, 1994; Simmons et al., 2005), accounting for more than 20% of the global production of gold and silver (Singer, 1995). While magmas may be the source of precious metals (e.g., Hedenquist and Lowenstern, 1994; Ulrich et al., 1999; Sillitoe, 2000a; Sillitoe and Hedenquist, 2003; Heinrich, 2005; Simmons and Brown, 2006), the formation of ore deposits is constrained by the concentrations and fluxes of metals in hydrothermal solutions, yet these are poorly known because of the great difficulty in obtaining samples of deep hot solutions for trace metal analyses. Here we report our results on sampling and analyses of gold, silver, arsenic, antimony, and mercury in deep hydrothermal solutions from the Taupo Volcanic Zone. This is an ideal natural laboratory because precious metals are known to deposit in geothermal systems and volcanoes along the 250 km segment of the arc, while sharing a common geological framework (Weissberg, 1969; Henley, 1985; Brown, 1986; Krupp and Seward, 1987; Hedenquist et al., 1993; Simmons and Browne, 2000; Brown and Simmons, 2003). Wells drilled

to deeper than 1 km into the upflow zone of geothermal systems provide optimal fluid sampling sites. Because precious metals precipitate in wells from the ascending fluids upon boiling and gas loss (Brown, 1986; Krupp and Seward, 1987; Hedenquist et al., 1993; Simmons and Browne, 2000; Reyes et al., 2003), we constructed a titanium down-hole sampler to obtain deep fluids for analysis. Any metals that precipitate in the sampler during its ascent to the surface are incorporated into the liquid sample by rinsing the sampler with strong acid. The analytical results are combined with fluid flows to estimate the metal fluxes for separate geothermal systems and for the whole volcanic arc. These data shed light on the sources of metals in hydrothermal solutions consisting mainly of meteoric water, and they show how metal fluxes limit the duration of hydrothermal ore formation.

TAUPO VOLCANIC ZONE

The Taupo Volcanic Zone is a young (<1.6 Ma) rifted arc that extends ~250 km from Ruapehu to White Island (Fig. 1). Volcanism in the northern and southern segments is dominated by cone-building eruptions of andesite, whereas the central segment is dominated by explosive caldera-forming eruptions of rhyolite (Wilson et al., 1995). The high heat flow (4000–5000 MW) that makes the Zone one of the world's premier thermal belts derives mainly from convective heat transfer associated with widespread geo-

thermal activity, which accounts for ~70% of the total heat flow (Bibby et al., 1995; Hochstein, 1995). This activity results from magmatic intrusion driving deep circulation (>5 km) of meteoric water and produces separate hydrothermal plumes that are distinguished on the basis of their physical and chemical characteristics (Bibby et al., 1995; Giggenbach, 1995). Most of these geothermal systems are concentrated in the central segment of the Taupo Volcanic Zone (Fig. 1), which has the highest rate of rhyolite magma eruption known on Earth (Wilson et al., 1995). The N₂/Ar, CO₂/He, and B/Cl ratios of the hydrothermal fluids, however, suggest that the deep magmas intruding beneath the convection cells are andesitic in the eastern Zone but are basaltic in the western Zone (Giggenbach, 1995; Christenson et al., 2002; Fig. 1).

We sampled the deep hydrothermal solutions from three eastern and three western geothermal systems in the central Taupo Volcanic Zone, where there is access to production, monitoring, and exploration wells (Fig. 1). The geology of the drilled parts of these systems is dominated by a layer-cake sequence (1 to >3 km thickness) of rhyolitic to andesitic volcanic rocks, which rest unconformably on Mesozoic metasedimentary rocks, locally disrupted by normal faults. The chemical and isotopic compositions of the hydrothermal waters reflect their meteoric origin and deep circulation, the incorporation of magmatic gases, and the subsequent fluid-mineral

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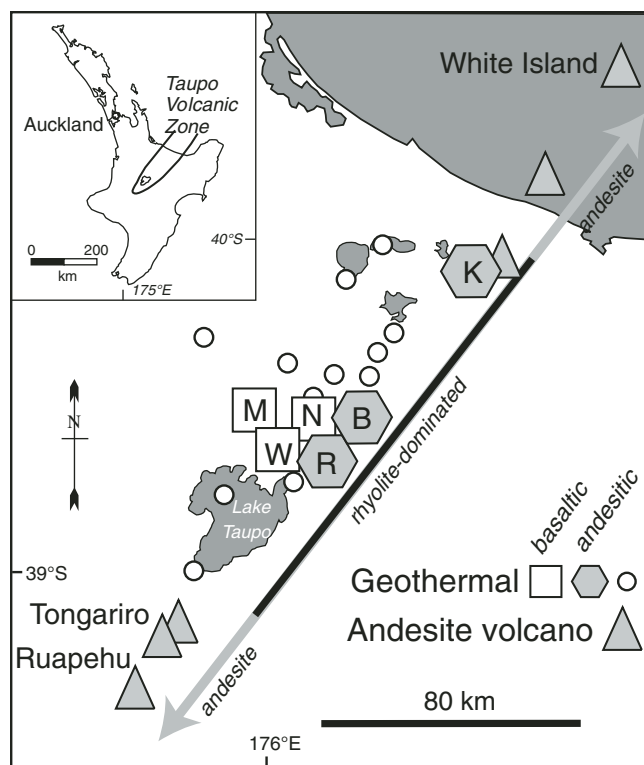


Figure 1. Locations of geothermal systems and andesitic volcanoes in Taupo Volcanic Zone. Eastern geothermal systems with fluid signatures indicating andesitic magmatic intrusions (hexagons) are aligned along a northeast trend, extending the length of the Zone, including Ruapehu and White Island andesite volcanoes. Western geothermal systems with fluid signatures indicating basaltic magmatic intrusions (squares) are restricted to the central rhyolite-dominated part of the Zone. Unsourced geothermal systems (small open circles) are also shown. B—Broadlands-Ohaaki; K—Kawerau; M—Mokai; N—Ngatamariki; R—Rotokawa; W—Wairakei.

interactions (Giggenbach, 1995; 1997; Giggenbach et al., 1993). All of the deep solutions sampled were near-neutral pH and reduced, and they are close to thermodynamic equilibrium with hydrothermal minerals that commonly occur in the deep altered volcanic host rocks, including albite, adularia, quartz, chlorite, illite, calcite, epidote, and pyrite (Giggenbach, 1997). Apart from the ratios of inert gases and deeply derived components (N_2/Ar , CO_2/He , and B/Cl) that distinguish their magmatic sources, the hydrothermal solutions from eastern geothermal systems have higher concentrations of gaseous constituents (CO_2 , H_2S) but lower total dissolved salts (Na, K, Cl) than solutions from western systems (see GSA Data Repository Table DR1¹). In pro-

duction wells, the hot liquid boils as it ascends, discharging a mixture of liquid and gas. The sampling depths (950–1600 m) were therefore chosen to coincide with known liquid-only feed points at the hottest temperatures (195–322 °C) and the deepest levels practical so that we could determine the concentrations of metals in rising solutions before they boiled.

DISCUSSION

The precious-metal concentrations in deep solutions (Table 1) span three orders of magnitude (<0.1 –23 ppb, Au; 2.7–2400 ppb Ag) and correlate with H_2S concentration and temperature (Fig. DR1), consistent with experiments on gold and silver solubility (Stefánsson and Seward, 2003, 2004). Accordingly, gold concentrations are highest at Rotokawa and lowest at Wairakei and Ngatamariki. Silver concentrations are also highest at Rotokawa, but the lowest values are shared between Broadlands-

Ohaaki, Kawerau, Wairakei, and Ngatamariki. High-grade precious-metal scale deposits have been found in the surface pipes of the eastern geothermal systems at Rotokawa, Broadlands-Ohaaki, and Kawerau (Brown, 1986; Krupp and Seward, 1987; Simmons and Browne, 2000; Reyes et al., 2003), and these systems host fluids with high aqueous concentrations of precious metals. The high silver concentration at Mokai (250 ppb) indicates that significant precious-metal concentrations also occur in hydrothermal solutions from western geothermal systems. The aqueous concentrations of As, Sb, and Hg, which typically occur in precious-metal ores, range only about two orders of magnitude, independent of gold or silver concentrations (Table 1, Fig. 2; Fig. DR1).

Speciation calculations show that deep solutions from all six systems are significantly undersaturated in gold ($\log Q/K < 0$; Table DR3) and, in three systems, are undersaturated in silver sulfide (Ag_2S), indicating that the metal budget, rather than temperature or aqueous H_2S concentration, limits the amount of gold and, to some extent, silver in solution. These results, and the similar stratigraphy of all six geothermal systems, point to a deep local control on the supply of precious metals, exemplified by the sharp differences in fluid compositions and metal concentrations between Rotokawa and Wairakei (Fig. 2), which are only 10 km apart (Fig. 1). For the eastern geothermal systems, intrusions of andesite magma are likely metal sources as indicated by the gold, arsenic, and antimony fluxes in volcanic emissions from White Island volcano (Hedenquist et al., 1993; Wardell et al., 2004; Table 2); transport of gold by a deep, vapor-like phase (Ulrich et al., 1999; Heinrich, 2005) is consistent with the evidence

¹GSA Data Repository item 2007272, methods, Tables DR1–DR4, and Figure DR1, is available online at www.geosociety.org/pubs/ft2007.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

TABLE 1. TRACE METAL CONCENTRATIONS OF THE DEEP GEOTHERMAL SOLUTIONS AND SAMPLING TEMPERATURES

	T (°C)	Au (ppb)	Ag (ppb)	As (ppb)	Sb (ppb)	Hg (ppb)
Broadlands-Ohaaki	290	0.3–1.5*	2.7–8*	1540	360	12.82
Kawerau	260–295	0.7–2.1	14.3–31.5	3120–4860	480–540	44–78
Rotokawa	320	7.8–23	1100–2400	1200–1400	180–230	1.86–5.18
Mokai	322	0.6	250	3900	1200	1.11
Ngatamariki	195	<0.1	3.1–8.4	100–470	13–27	1.21–3.40
Wairakei	255	<0.1	6.0–14.1	2900–3200	41–58	0.68–2.82

Note: Summarized from Table DR2 (see footnote 1).

*Upper estimates of gold and silver concentrations in deep waters from Broadlands-Ohaaki are from Brown (1986).

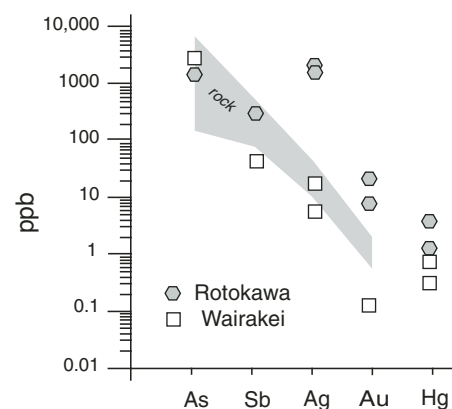


Figure 2. Range of trace metal concentrations in deep thermal solutions from Wairakei (western geothermal system) and Rotokawa (eastern geothermal system) compared to the range of trace metal concentrations in volcanic and metasedimentary rocks (gray shaded area labeled “rock”; Table DR4 [see footnote 1]).

TABLE 2. THE THERMAL OUTPUTS (MW), GEOTHERMAL FLOW RATES, AND HYDROTHERMAL METAL FLUXES FOR THE TAUPO VOLCANIC ZONE

	MW	Upflow (kg/sec)	Au (kg/yr)	Ag (kg/yr)	As (kg/yr)	Sb (kg/yr)	Hg (kg/yr)
Broadlands-Ohaaki	125	95	0.9–4.5	8–24	4600	1100	38
Kawerau	100	85	1.9–5.6	38–84	8400–13,000	1300–1450	120–210
Rotokawa	220	150	37–109	5200–11,400	5700–6600	850–1090	9–25
Mokai	275–400	190–275	3.6–5.2	1500–2170	23,400–33,800	7200–10,400	7–10
Ngatamariki	38–55	30–45	<0.14	3–12	95–670	12–38	1–5
Wairakei	350–420	290–350	<1.1	55–156	26,500–35,300	375–640	6–31
Subtotal	1110–1320	840–1000	43–126	6800–13,850	68,700–94,000	10,800–14,700	180–320
White Island	240–800	?	37	?	6200	660	310
Total	1350–2120	840–1000	80–163	6800–13,850	74,900–100,200	11,460–15,360	490–630

Note: Thermal outputs and deep-water upflow (Bibby et al., 1995; Hochstein, 1995) are combined with the metal concentrations of deep solutions to calculate the metal fluxes for geothermal systems. The results are added to metal fluxes of White Island volcano (Hedenquist et al., 1993; Wardell et al., 2004) to determine the minimum overall metal fluxes for the Taupo Volcanic Zone.

for the contributions of magmatic gases into the deep portion of these systems (Giggenbach, 1995). Country rocks represent another possible source of trace metals (Table DR4), since deep fluids are undersaturated in gold, and the Wairakei data resemble the distribution patterns of As, Sb, Ag, and Au in unaltered volcanic and metasedimentary rocks (Fig. 2). Even in this case, the role of magmas remains important as the main source of aqueous chloride and sulfide (Giggenbach, 1995), which provide ligands to mobilize the metals into deep solutions (e.g., Seward and Barnes, 1997).

Metal fluxes (Fig. 3; Table 2) were calculated from the trace-metal concentrations (Table 1) and the rates of thermal fluid upflow (Bibby

et al., 1995; Hochstein, 1995). The combination of high flow rate and precious-metal concentrations makes Rotokawa stand out as having the highest fluxes of gold (37–109 kg/yr) and silver (5200–11,400 kg/yr). The Rotokawa gold flux is substantially greater than that estimated for the Ladolam geothermal system (24 kg/yr; Simmons and Brown, 2006), which hosts one of the largest hydrothermal gold deposits in the world; it is also greater than that for White Island volcano (37 kg/yr; Hedenquist et al., 1993) and overlaps the lower part of the estimated flux range for Mount Etna volcano (80–1200 kg/yr; Andres et al., 1993). The gold flux for Mokai is considerably less than that for Rotokawa, but it is similar to Kawerau and Broadlands-Ohaaki, which both have higher metal concentrations in solutions. Mokai is also notable for its high silver flux, which greatly exceeds that for Kawerau, Broadlands-Ohaaki, Wairakei, and Ngatamariki. The highest arsenic fluxes are at Mokai and Wairakei, the highest antimony flux is at Mokai, and the highest mercury flux is at Kawerau. However, the concentrations and fluxes of these metals correlate poorly with gold and silver (Table 2), suggesting that the controls on their respective inventories are decoupled from those influencing gold and silver.

Extrapolating the metal flux data (Fig. 3; Table 2) over the last 100,000 yr, a typical minimum lifetime for Taupo Volcanic Zone geothermal systems (Hochstein and Browne, 2000), the total throughputs for the 250-km-long segment of volcanic arc due to hydrothermal activity range from 8000–16,300 tonnes of gold and from 680,000–1,385,000 tonnes of silver, assuming uniform metal concentrations and flow rates over the period. These are conservative estimates because the systems studied only represent ~25% of the total heat and mass transfer due to geothermal activity and volcanic degassing; an additional ~1800 tonnes of gold erupted to the surface with rhyolite and andesite magmas, based on an average gold concentration of ~1 ppb in

fresh volcanic rock (Table DR4) and the magma eruption rate of 0.28 m³/s (Wilson et al., 1995). The hydrothermal metal flux estimates are huge considering the largest magmatic-hydrothermal ore deposits in volcano-plutonic arcs contain ~1000–2000 tonnes of gold (Sillitoe, 2000b) and up to 100,000 tonnes of silver (Simmons et al., 2005). The metal fluxes for Rotokawa, alone, produce such ore deposit inventories in <20,000–55,000 yr for gold and <10,000–20,000 yr for silver, and Mokai could produce a silver inventory of 100,000 tonnes in 45,000–75,000 yr. Unlike the active hydrothermal gold deposit at Ladolam (Simmons and Brown, 2006), however, ore bodies have so far not been discovered in Zone geothermal systems (e.g., Simmons and Browne, 2000). Does this mean that metal concentrations in deep solutions fluctuated over time and/or that metal-depositing conditions required to form ores have not developed in the Zone geothermal systems? Answers to these questions are hard to obtain because: (1) data on the compositions of deep geothermal fluids are only available for the past 50 yr, and this period of time is too short to know if metal concentrations might have fluctuated over a much longer time scale; and (2) information on the occurrence of subsurface mineralization is limited by the relatively wide spacing of geothermal wells of several tens to several hundred meters and the irregular coring intervals, particularly at shallow levels (<500 m depth), where epithermal deposits form (e.g., Simmons and Browne, 2000). Nonetheless, if the metal fluxes in the Taupo Volcanic Zone are indicative of fluxes in other volcanic arcs, and throughout time, then the efficiencies of processes producing precious-metal ores, especially in regard to metal deposition, appear extremely low, as suggested by the rarity of giant precious-metal ore deposits (Sillitoe, 2000b).

In summary, our results show there is a very strong hydrothermal flux of precious and related metals within a volcanic arc. These metal fluxes are regulated by intrusion of andesitic and

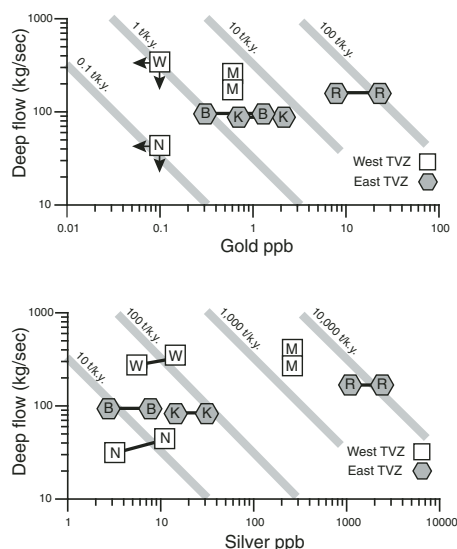


Figure 3. Precious metal concentrations of deep thermal solutions versus deep upflow rates (Tables 1 and 2), showing the metal flux rates (tonnes per 1000 yr, t/k.y.) in Taupo Volcanic Zone (TVZ) geothermal systems. B—Broadlands-Ohaaki; K—Kawerau; M—Mokai; N—Ngatamariki; R—Rotokawa; W—Wairakei.

basaltic magmas: (1) through input of metal-bearing fluids or supply of metal-transporting ligands, which affect the concentrations of metals in solutions; and (2) the heat flow, which dictates the convective mass transfer. The apparent absence of naturally formed precious-metal ore bodies in the Taupo Volcanic Zone geothermal systems, but with spectacular gold-silver scale deposits in wells (Brown, 1986; Simmons and Browne, 2000), emphasizes the importance of focused fluid flow and efficient metal precipitation (e.g., boiling) as ore-forming processes. After all, without them, no matter how much metal might be fluxing through a geothermal system, ore bodies will not form.

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