1	Active geothermal systems with entrained seawater as analogues for low-sulphidation
2	epithermal mineralization
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11 Abstract (207 words)

The paradigm for low-sulphidation (LS) volcanic-arc associated mineralization is the active geothermal systems located along the Taupo Volcanic Zone (e.g. Broadlands). However, this analogue is inapt where fluid salinities are consistently in excess of 3.5 wt % NaCl.

LS mineralization on Milos (Aegean arc) records high paleofluid-salinities. The δD and $\delta^{18}O$ data do not exemplify ¹⁸O-shifted meteoric waters—typical of terrestrial geothermal systems. Nor is a submarine origin indicated—stable isotope data show mixing between meteoric, seawater and volcanic-arc gases. Strontium isotope data are comparable to a nearby active seawater-entrained geothermal system. These are features seen in hydrothermal systems associated with emergent volcanoes.

21 For the Milos LS mineralization, high-salinity fluids show it cannot be explained by a 22 Broadlands-type model. The absence of saliferous sequences and significant intrusive rocks 23 preclude these as salinity sources. The similarities between paleo and active systems in terms of salinity, $\delta D - \delta^{18}O$ and strontium isotope systematics strongly suggest that seawater is the 24 main source for Na and Cl. We suggest geothermal systems, containing seawater, associated 25 26 with emergent volcanoes are an alternative analogue for LS epithermal mineralization. Furthermore, they bridge the gap between submarine, and large-scale terrestrial geothermal 27 28 systems-the modern analogues for VHMS and epithermal mineralisation in the scheme of 29 intrusion-centered hydrothermal mineralization.

30 Keywords

31 Epithermal processes, seawater, Milos, isotopes

32 Introduction

Geothermal systems in convergent plate margin settings are the active equivalents of high-33 34 level (2–3 km) intrusion-centred hydrothermal ore-deposits (e.g. Hedenquist and Lowenstern, 1994). Close to magmatic activity, volcanic emanations are the surface expressions of 35 porphyry-Cu and high-sulphidation epithermal-Au mineralizing processes at depth 36 37 (Hedenquist et al., 1993). Located farther from the magmatic source are low-sulphidation 38 (LS) epithermal deposits, for which the Broadlands-Ohaaki geothermal system is the 39 paradigm (e.g. Simmons and Browne, 2000). Generally, in LS mineralization, fluid inclusions 40 document low-salinity fluids (< 1 wt % dissolved salts + CO₂) and these are equivalent to the 41 low-chorinity (< 1000 ppm) fluids seen in Broadlands-type geothermal systems. However, they also identify saline fluids (up to 15 wt % salts) and, in this case, there are no documented 42 active analogues (Hedenquist and Lowenstern, 1994). Conversely, deep water (> 1000 m) 43 44 geothermal systems, such as TAG (e.g. You and Bickle, 1998) are exemplars for VHMS 45 deposits. However, where boiling submarine geothermal fluids vent into shallow water (< 20046 m) mineralization is epithermal in style (Stoffers et al., 1999), but there are no clearly defined 47 ancient equivalents (Huston, 2000). Thus, in both the submarine and subaerial environments 48 there is a missing link between active geothermal systems and their ancient counterparts.

Below, we provide new strontium isotope data and summarize features of epithermal systems on Milos island. By comparing these with well-characterised modern analogues we suggest that active geothermal systems with entrained seawater, such as those in the Aegean arc (Aegean arc-type), are an alternative to the Broadlands–Ohaaki LS paradigm in the scheme of intrusion-centred hydrothermal systems.

54 The Aegean arc

55 The Aegean arc is a zone of Pliocene to modern volcanism related to active back-arc 56 extension caused by the subduction of the African plate beneath the Aegean micro-plate (e.g. 57 Pe-Piper and Piper, 2002). It is built on continental crust, comprises seven major volcanic 58 centres and is located 120–250 km north of the Hellenic trench (Fig. 1). The volcanic rocks 59 are calc-alkaline with localised high-K variants and range from basalt to rhyolite in composition, with dominant andesites and dacites. Present day hydrothermal activity 60 61 comprises both low-enthalpy (Aegina, Sousaki, Methana) and high-enthalpy systems (Milos, 62 Nisyros).

63 Milos geology and LS epithermal Au-Ag mineralization

64 Upper Pliocene (2.66 ± 0.07 Ma; Stewart and McPhie, 2003) submarine and Late Pleistocene to present (1.9-0.1 Ma) subaerial volcanic rocks overlay Mesozoic metamorphic basement 65 66 and Upper Miocene-Lower Pliocene marine sediments, and record a transition from a shallow 67 submarine (< 200 m) to subaerial volcanic setting (Fytikas et al., 1986; Rinaldi and Venuti, 68 2003; Stewart and McPhie, 2003). Emergence probably occurred at 1.44 ± 0.08 Ma (Stewart 69 and McPhie, 2003). Plutonic rocks are not known on Milos and have only been reported as 70 ignimbrite-hosted granitic xenoliths from the nearby islet of Kimolos (Pe-Piper and Piper, 71 2002).

72 The oldest submarine volcanic rocks occur on western Milos and host LS Au-Ag 73 mineralization (the Profitis Ilias [PI]-Chondro Vouno [CV] epithermal system), which extends over a 20 km² area (this study; Kilias et al., 2001). Fluid inclusion data show the 74 75 hydrothermal fluids underwent extreme boiling and vaporisation. Importantly, final ice-76 melting (T_{mice}) data show that 70 % of the fluid inclusions have net salinities in excess of 77 seawater (Tab. 1), showing that throughout its lifespan, fluids in the system were saline. The 78 tops of the paleosystem (now ~600 masl) show crustiform/colloform quartz-barite±alunite 79 veins and quartz-cemented breccias, with locally high gold (PI: 56 ppm; CV: 250 ppm) and 80 silver (PI: 197 ppm; CV: 90 ppm). Deeper in the system (now ~300 masl), the mineralization 81 is dominated by a base-metal-bearing stockwork. Elevated gold values at PI are concentrated 82 above the base metal zone and are spatially related to boiling (Kilias et al., 2001).

83 Active geothermal system

84 In the active geothermal system (Liakopoulos, 1987; Pflumio et al., 1991), data indicate a 85 two-component reservoir: (1) A high enthalpy system with deep seawater recharge located 1– 86 2 km below sea-level. Reservoir temperatures range 250-350 °C and salinities can be 87 significantly higher than seawater (up to 9 wt % salts). This results from Rayleigh distillation 88 as seawater percolates, through progressively hotter rocks, into the reservoir. Due to its high 89 salinity, venting of the deep geothermal fluid is accompanied by boiling close to the top of the 90 reservoir and in some cases the reservoir may be two-phase. (2) A shallow reservoir (100-175 91 °C) overlies the high-enthalpy system. It is located close to sea-level, recharged by meteoric 92 water and seawater intrusion, is commonly saline (up to 5 wt % salts) and heated by gas 93 escapes from the underlying deep reservoir. Seawater, as a major component of both the deep

- 94 and shallow reservoirs, is documented on the basis of 87 Sr/ 86 Sr and δ D– δ 18 O–Cl systematics
- 95 (Pflumio et al., 1991). In the shallow (<100 m) submarine environment, venting geothermal
- 96 fluids contain suspended particulate matter strongly enriched in Fe, Mn, Si and Ba and locally
- 97 deposit APS minerals, pyrite, marcasite, barite, gypsum, and calcite (Baltatzis et al., 2001;
- 98 Varnavas et al., 2000). In addition, the deep reservoir is metalliferous (Pb: 180 ppb; Zn: 1458
- 99 ppb) (Christanis and Seymour, 1995) and has gold concentrations in the region of 0.3 ppb
- 100 (Liakopoulos, 1987).

101 Strontium isotopes

Sr-isotope data show that the least altered igneous rocks have low ⁸⁷Sr/⁸⁶Sr, whereas their hydrothermally altered counterparts are closer to a seawater signature (Fig. 2). The basement rocks have variable strontium ratios (0.7033–0.7136)) (Fig. 2). In the modern system, ⁸⁷Sr/⁸⁶Sr in the fluids vary from 0.7092–0.7102 (Pflumio et al., 1991). Barites from the Profitis Ilias mineralization display similar ⁸⁷Sr/⁸⁶Sr to the modern system (Fig. 2). All values for geothermal water and hydrothermal minerals are slightly more radiogenic than seawater but relatively constant.

109 **Discussion**

110 Seawater as a fluid component on Milos

 T_{mice} in fluid inclusions provides information on salinity. However, above -1.5 °C, it is unable 111 112 to distinguish between dissolved salt (< 2.5 wt % NaCl eq.) and gas (< 4.4 wt % CO_2 eq.). 113 This permits gas-charged low-chlorinity terrestrial geothermal systems to be the paradigm for 114 LS epithermal mineralization, as excess chlorinity can be assigned to dissolved gas 115 (Hedenquist and Henley, 1985). Moreover, in Broadlands-Ohaaki-type geothermal systems 116 derivation of chlorinities in excess of 5000 ppm through fluid-rock interaction is extremely 117 difficult and where high salinities are recorded they are attributed to boiling to dryness and 118 have a localised effect (Simmons and Browne, 1997). Hence, when comparing ancient 119 systems with Broadlands–Ohaaki-type equivalents, when the apparent salinity is up to 3–4 wt 120 % NaCl eq there is an implicit requirement to assign freezing point depressions to dissolved gas rather than chlorinity. However, in our case > 70 % of the fluid inclusions have net 121 salinities in excess of 3.5 wt % NaCl eq (Tab. 1). Thus, we cannot assign excess T_{mice} to 122 123 dissolved gas. Nonetheless, our high salinities have to be explained. There are three main 124 sodium and chlorine reservoirs available to large-scale geothermal systems: (1) evaporites and

125 evaporitic sediments (2) magmatic brines and (3) seawater. An evaporitic origin of salinity is 126 considered highly improbable, as there is no record of saliferous rocks within the Milos sediments. Concerning a magmatic brine, it is estimated that during its lifetime a geothermal 127 systems turns over between $10^{6\pm1}$ km³ of water (Barnes and Seward, 1997). If it is assumed 128 that and esitic magmas contain about 1000 ppm chlorine, then approximately $10^{7\pm1}$ km³ of 129 130 magma would be required maintain salinities in the geothermal system at 9 wt % NaCl. This 131 is not impossible, but on Milos, geological and geophysical evidence is lacking (Pe-Piper and 132 Piper 2002). Thus, a magmatic origin of salinity is also considered unlikely. This leaves a 133 seawater origin for Na and Cl. We consider that the chemical similarities between the ancient 134 and modern systems on Milos (see above) support a seawater origin for the PI-CV epithermal 135 fluids.

Comparison of $\delta D - \delta^{18} O$ of inclusion fluids from the Profitis Ilias epithermal mineralization 136 137 and several active geothermal systems associated with emergent volcanoes reveals remarkable 138 similarities (Fig. 3). In the active systems, the geothermal fluids have a three component 139 source (sea, meteoric and magmatic) and the waters fall in a zone projecting from the 140 meteoric water line to values intermediate to seawater and volcanic-arc gases (Fig. 3). The 141 fluid inclusion data for the Profitis Ilias LS mineralization show an analogous trend and lie in 142 a similar zone; this is in sharp contrast to typical LS mineralization where stable isotope data show ¹⁸O-shifted fluids at constant δD (see Broadlands field in Fig. 3). 143

In terms of strontium isotope data (Fig. 2, the basement metamorphic sequence has ⁸⁷Sr/⁸⁶Sr 144 145 (0.7033–0.7137) encompassing the entire range, permitting a variety of fluid–rock interaction interpretations. However, we think the clustering of measured ⁸⁷Sr/⁸⁶Sr for the modern 146 geothermal fluids (0.7092–0.7100) and mineralization (0.7096–0.7100 [epithermal Au–Ag]; 147 0.7092–0.7098 [Mn–Ba] close to the value for late-Pliocene seawater (0.7090–70906), also 148 indicates of a seawater source. In addition, as ⁸⁷Sr/⁸⁶Sr for the modern and ancient systems are 149 significantly different from most of the unaltered igneous rocks (0.7050-0.7080) mitigates 150 further against a magmatic fluid source. Indeed, most of the ⁸⁷Sr/⁸⁶Sr (0.7082–0.7098) for 151 152 hydrothermally altered igneous rocks cluster within or close to the range recorded by the 153 modern geothermal fluids and seawater.

Taken together, the above lines of evidence show that in addition to being a fundamental component in the active system, seawater has played a key role in LS mineralization.

156 Hybrid epithermal systems and modern analogues

157 It is clear that Broadlands-Ohaaki-type meteoric geothermal systems are not a valid analogue 158 for the Milos Au-Ag mineralization and for moderately saline (> 15 000 ppm NaCl eq.) LS 159 epithermal mineralization in general. Furthermore, there is a missing link between saline epithermal LS systems and their modern counterparts. We suggest that the best candidates, 160 which recognize the key parameters of consistently high fluid inclusion salinities, $\delta D - \delta^{18}O$ 161 162 systematics indicating seawater and a seawater Sr isotope signature, are geothermal systems 163 with entrained seawater. Typical examples are the active systems on Milos and Nisyros (Fig. 164 3). These analogues are hybrids, containing elements of both submarine and terrestrial 165 geothermal systems. Indeed, the occurrence of fossil hybrid systems is predicted (Huston, 166 2000) though, to date, no ancient equivalents have been clearly identified. Here, in the 167 emergent environment, circulating sea and meteoric water are the main fluid components. The 168 fluids boil and result in auriferous quartz veins with epithermal textures and proximal quartz-169 adularia, intermediate quartz-sericite-pyrite and distal propylitic/quartz-albite alteration 170 halos—features that are comparable to the Milos epithermal mineralization. Thus, we suggest 171 that epithermal mineralization where the involvement of seawater can be clearly demonstrated 172 (e.g. Milos) are good candidates for fossil hybrid epithermal systems, and active geothermal 173 systems with entrained seawater such as the Aegean-arc type, are their modern analogues.

Features of hybrid epithermal systems can be reconstructed by putting geothermal systems associated with emergent volcanoes into a conceptual framework. Fig. 4 illustrates the model—gold-bearing epithermal veins are located between a shallow low-temperature (100– 177 175 °C) steam-heated zone recharged by meteoric water and seawater intrusion, and a deep, seawater recharged, higher-temperature (250–350 °C) base-metal bearing reservoir.

179 **Concluding remarks**

Broadlands-type models are not appropriate for LS epithermal systems with elevated salinities that cannot be reasonably explained by dissolved gas or localised boiling to dryness. Moreover, where high salinities of this nature are encountered, an explanation for them must be sought. One possibility is a seawater origin for the hydrothermal fluids, though to use this explanation, other parameters have to be consistent. In the case of Milos, the epithermal mineralization can be explained by analogy to seawater-entrained geothermal systems associated with emergent volcanoes (e.g. Nisyros, Milos) and the mineralization data

- 187 (geologic, isotope and fluid inclusion) are in accord with this model. We suggest that our
- 188 Aegean-arc model should be considered as an additional paradigm in the scheme of intrusion-
- 189 centred metallogenesis. Moreover, it may provide a link between submarine and terrestrial
- 190 mineralization processes. Appropriate indicators for its use are fluid inclusion data showing
- 191 consistently elevated salinities (> 3.5 wt % NaCl eq.) and mineralization hosted in submarine
- 192 or transitional to subaerial volcanic rocks in an island arc tectonic setting. However, it must
- be stressed that these are not definitive and other corroborating data must be sought, in our
- 194 case, strontium isotope, δD and $\delta^{18}O$ analyses.

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281	You, C.F., and Bickle, M.J., 1998, Evolution of an active sea-floor massive sulphide deposit:
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283 284 Fig. 1. Maps showing **A**) the main geotectonic elements of the eastern Mediterranean along with volcanic centres and regions of geothermal activity; **B**) the main geological features of Milos island plus the location of LS epithermal mineralisation and the main surface manifestations of the geothermal system (Milos map adapted from Fytikas et al., 1986; LPI: Lower Pleistocene, LPo: Lower Pliocene; LMi: Lower Miocene, M: Mesozoic)

Fig. 2. 'S'-curve of measured strontium isotope data from mineralisation on Milos, showing ⁸⁷Sr/⁸⁶Sr for barite, fresh and altered igneous rocks, marine platform sediments basement rocks and geothermal waters (data: this study; Briqueu et al., 1986; Farrell et al., 1995; Hein et al., 2000; Pflumio et al., 1991)

Fig. 3. Fluid-inclusion $\delta D - \delta^{18}O$ data for Profitis Ilias, comparing the epithermal mineralisation with active systems on the Aegean arc with reference points for eastern Mediterranean seawater (crossed squares labelled S) and estimated present day geothermal liquids (filled triangles labelled M [Milos] and N [Nisyros]). (Milos epithermal mineralisation data: Naden et al., 2003; geothermal data: Brombach et al., 2003; Kavouridis et al., 1999; Liakopoulos, 1987; fields for volcanic arc gases and Broadlands derived from Giggenbach, 1992 and Field and Fifarek, 1985 respectively)

Fig. 4. Conceptual model of hybrid Aegean-arc-type epithermal systems (adapted fromKavouridis et al., 1999; Liakopoulos, 1987)

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Naden et al: Geology, Fig. 1, actual size



Naden et al: Geology, Fig. 2, actual size



Naden et al: Geology, Fig. 3, Actual size



Naden et al: Geology, Fig. 4, actual size