SPATIAL AND TEMPORAL RELATIONSHIPS BETWEEN HYDROTHERMAL ALTERATION ASSEMBLAGES AND INTRUSIONS AT THE PALINPINON GEOTHERMAL FIELD, PHILIPPINES – IMPLICATIONS FOR PORPHYRY AND EPITHERMAL ORE DEPOSITS

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Summary - Biotite, calc-silicate, propylitic, advanced argillic and illite alteration assemblages at Palinpinon are genetically associated with the Nasuji and Puhagan intrusions. Age determinations of hydrothermal biotite ($^{40}Ar/^{39}Ar$ = 0.7-0.6 Ma) and alunite (K/Ar = 0.9-0.8 Ma) demonstrate that biotite and hypogene advanced argillic assemblages formed contemporaneous with the Nasuji Pluton ($^{40}Ar/^{39}Ar$ = 0.7-0.3 Ma). The 'blind' intrusion at Puhagan provides the heat source for present geothermal activity. Magmatic-hydrothermal alteration assemblages (biotite, calc-silicate) have developed above this intrusion with a halo of propylitic alteration. At <2 km, an illite-bearing alteration assemblage is in thermal equilibrium with the present geotheml system. The intimate spatial and temporal relationship between intrusion emplacement and the styles of alteration at Palinpinon are characteristic of deposit styles such as porphyry, skarn, HS and LS epithermal. However, mineralized zones have not been detected at Palinpinon, possibly due to poor fiacture permeability, insufficient fluid flux, or a lack of metals in the hydrothermal fluids.

1. INTRODUCTION

Palinpinon geothermal field, Southem Negros, Philippines, is a high temperature, liquid-dominated geothermal system in an active volcanic arc. Previous workers (Leach and Bogie, 1982; Celenk et al., 1987; Reyes, 1990; Mitchell and Leach, 1991; Corbett and Leach, 1998) have drawn parallels between the types of alteration recognised at Palinpinon and magmatic-hydrothermal ore deposits (specifically, porphyry Cu deposits).

Hydrothermal alteration mineral assemblages associated with active and extinct hydrothermal systems can provide important insights into their fluid chemistry and temperature. The zonal distribution pattern and overprinting relationships of alteration assemblages indicate how fluid chemistry has evolved both spatially and temporally and can be used to understand genetic relationships between mineral deposits within a mineral district. Genetic relationships between any two of the four main styles of hydrothermal ore deposits related to arc-magmatism (e.g., porphyry Cu-Au, high sulfidation {HS) and low sulfidation {LS) epithermal Au, skarn) have been demonstrated (e.g., Arribas, 1995) or inferred by previous workers (e.g., Sillitoe, 1989; Cooke and Bloom, 1990), and such relationships have obvious implications for mineral exploration throughout the circum-Pacific. However, in order to understand the spatial and temporal associations between all four environments it is necessary to investigate their mineralogy, fluid chemistry and timing within a single district.

This paper describes the spatial and temporal distribution of hydrothermal alteration types at Palinpinon (biotite, calc-silicate, propylitic, illite and advanced argillic), together with the associated minor base and precious metal mineral occurrences. Techniques employed include thin section petrography, infrared spectroscopy (PIMA, FTIR), X-ray fluorescence (XRF) analyses, scanning electron microscopy (SEM), fluid inclusion microthermometry, proton-induced X-ray emission analyses (PIXE), and K-Ar and *Ar-Ar* geochronology.

2. GEOLOGY & HYDROLOGY

The Palinpinon geothermal field is hosted by several volcano-sedimentary sequences (Figure 1). The lowermost drilled unit is the andesitic volcaniclastic Lower Puhagan Volcanic Formation (Middle Miocene). The Okoy Formation (Upper Miocene-Early Pliocene) is a marine sequence of calcareous siltstones and sandstones. The uppermost formations are the Southern Negros (Late Pliocene-Pleistocene) and Cuernos Volcanic Formations (Pleistocene-Recent) which both consist of volcanic and volcaniclastic rocks of basaltic andesite to andesitic composition. Diorite to quartz diorite intrusions were emplaced into these successions during Early Pliocene to Recent times. The Puhagan dikes ($^{40}Ar/^{39}Ar$ = 4.2-4.1 Ma) and the Nasuji Pluton ($^{40}Ar/^{39}Ar$ = **0.7-0.3** Ma) intruded the Middle Miocene, Late Miocene and Early-Late Pliocene rock formations (Rae et al., submitted).

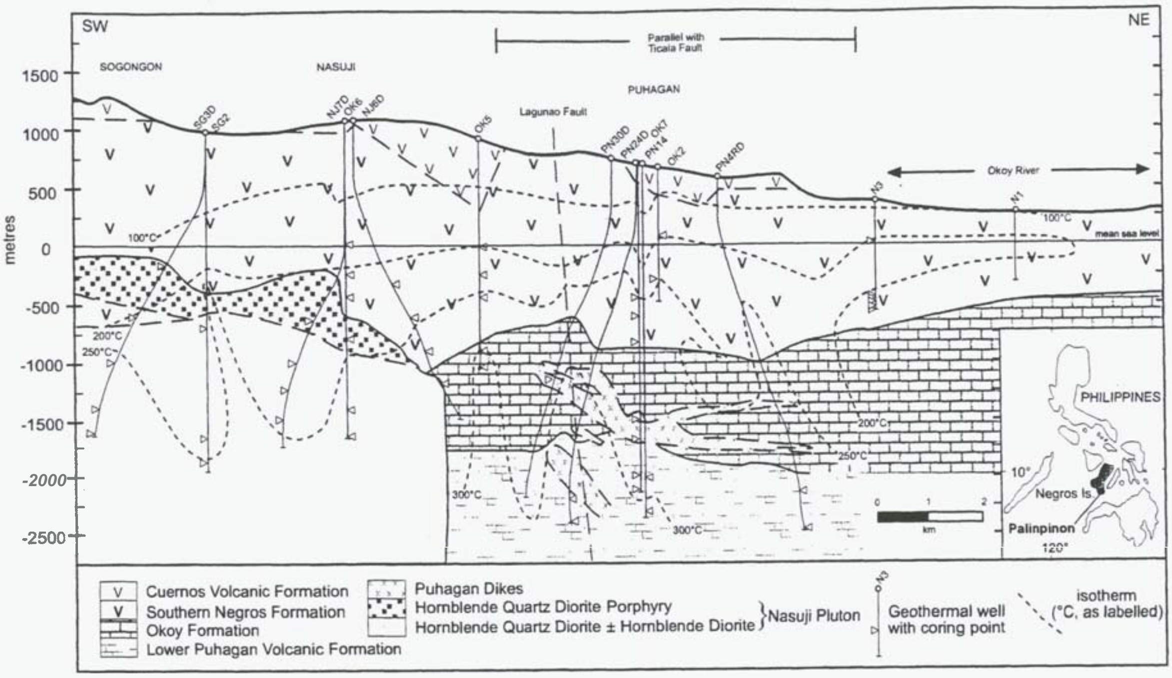

Figure 1. Geological cross-section (SW-NE) through Palinpinon geothermal field with the distribution of isotherms.

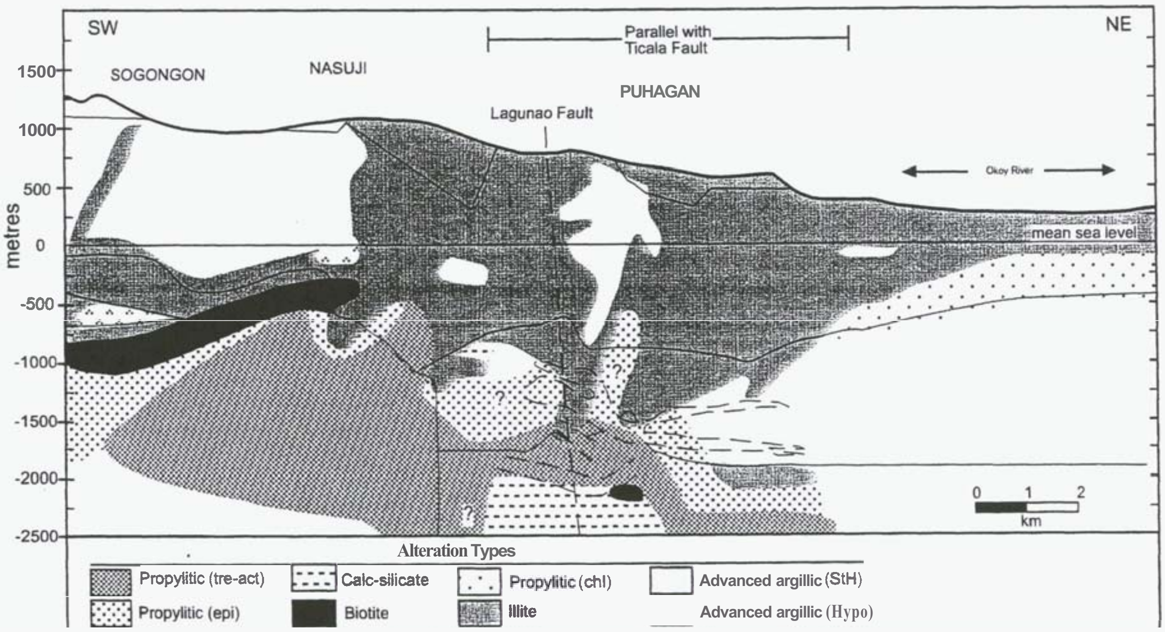

Figure 2. Cross-section (SW-NE), showing the distribution of alteration types relative to the geology. Thin lines represent the geological contacts shown in Figure 1. Alteration type abbreviations: chl = chlorite; epi = epidote; tre-act = tremolite-actinolite; Hypo = hypogene; StH = steam-heated.

Permeable zones related to faults and lithological contacts are the dominant influence on the modern geothermal hydrology at Palinpinon (Rae, 2002). The upflow zone is situated at a fault intersection (Ticala and Lagunao Faults) and the main northeasterly outflow zone is parallel to NE-striking faults (e.g., Ticala Fault) beneath the Okoy River (Figure 1). The reservoir currently consists of neutral to slightly alkaline chloride waters that have been affected by boiling, mixing and conductive cooling. Boiling occurs close to the region of upflow and is mainly restricted to narrow permeable zones. Mixing with steam-heated sulfate and meteoric waters occurs peripherally to the upflow zone.

3. WALLROCK ALTERATION

3.1. Biotite Alteration

Biotite is the diagnostic mineral of this assemblage, which occurs with magnetite, quartz, anhydrite, orthoclase, albite, tremolite-actinolite, hematite, pyrite and/or chalcopyrite. Zones of biotite alteration occur in the upper regions of the Nasuji Pluton, and in the Puhagan sector in the upflow zone beneath the Puhagan dikes (Figure 2).

3.2. Calc-Silicate Alteration

This alteration type is defined by the presence of garnet, clinopyroxene andor scapolite. Other minerals that can occur include tremolite-actinolite, epidote, wairakite, prehnite and albite. The most intense development of calc-silicate alteration occurs in the deepest parts of the geothermal field in the upflow zone beneath the Puhagan dikes (Figure **2).** In Nasuji-Sogongon, calc-silicate alteration occurs adjacent to the Nasuji Pluton.

3.3. Propylitic Alteration

Propylitic alteration assemblages are recognised by the presence of tremolite-actinolite, epidote and chlorite. These minerals occur with albite, orthoclase, **quartz,** pyrite ± chalcopyrite ± bornite ± titanite ± leucoxene. This assemblage has three subzones. The **tremolite-actinolite** subzone occurs in the deepest parts of the geothermal field (Figure **2).** In Puhagan, it surrounds biotite and calc-silicate alteration zones. In Nasuji-Sogongon, it has affected rocks in the deepest drilled portions of the Nasuji Pluton. The **epidote** subzone occurs above the tremolite-actinolite subzone (Figure **2)** and propylitic (epidote)-altered rocks crop out in Nasuji-Sogongon. The **chlorite** subzone occurs above the epidote subzone and in the outflow zone (Figure **2).**

3.4. Illite Alteration

The occurrence of either illite or smectite defines the illite alteration assemblage. Other minerals *that* can be present include **quartz,** leucoxene, anhydrite, calcite, pyrite, chalcopyrite, sphalerite and galena. This is the most extensively developed alteration type and occurs across the field from the Surface to depths greater than 500 m below sea level (bsl; Figure **2).**

3.5. Advanced Argillic Alteration

A hypogene subzone is characterised by tabular alunite, zunyite, **quartz,** aluminium-phosphate-sulfate *(APS)* minerals, pyrophyllite, diaspore, dickite, ± andalusite ± pyrite ± marcasite ± covellite. **A steam-heated** subzone is recognized by **quartz,** or amorphous silica, kaolinite, pseudocubic alunite, APS minerals, ± native sulfur ± diaspore ± pyrophyllite ± dickite ± pyrite ± marcasite. Advanced argillic altered rocks crop out at the surface as a series of northeast-trending ridges along the Okoy Valley. The southwestern ridges are altered to the hypogene assemblage, and the northeastern ridges have a steam-heated assemblage. Below surface in Nasuji-Sogongon, a thick (<1200 m) zone of advanced argillic alteration extends fkom the surface to approximately the top of the Nasuji Pluton (Figure **2).** In Puhagan a zone of advanced argillic alteration extends to approximately 1000 m bsl (Figure **2).** There are sporadic occurrences of andalusite, but most of the subsurface advanced argillic alteration consists of alunite, pyrophyllite, diaspore and dickite.

4. RADIOGENIC ($^{40}Ar/^{39}Ar$, K/Ar) DATING

Two samples of hydrothermal biotite and alunite were radiogenically dated by the Precise Radiogenic Isotope Services (PRISE) group, Australian National University (ANU), Canberra. The samples are from the biotite and hypogene advanced argillic alteration zones spatially associated with the Nasuji Pluton. Duplicate analyses of the alunite **were** within the calculated analytical uncertainties, with an average age of 0.85 ± 0.02 Ma. The results of the biotite analysis yielded a flat spectrum with a plateau age of 0.65 ± 0.01 **Ma.**

5. FLUID INCLUSIONS

5.1. Fluid Inclusion Types and Populations

Only primary and secondary fluid inclusions were identified in the sample suite (Roedder, 1984). Based on observations made at room temperature, six fluid inclusion types are recognised (Table 1). According to the associated types, the fluid inclusions are grouped into three populations (Table **2). Population A** occurs in **quartz** veins, associated with biotite alteration and primary **quartz** from the Nasuji Pluton. **Population B** in **quartz** veins associated with illite and propylitic (epidote + tremolite-actinolite) alteration assemblages. **Population C** occurs in **quartz** veins associated with illite and propylitic (chlorite) alteration zones.

Table 1. Fluid inclusion types with behaviour upon homogenisation.

Fluid Inclusions		Phases (at 25°C)			Homogenisation
Type	Subtype	Number	Dominant	Types	Behaviour
1a		2	liquid	liquid + vapor	vapor disappears
1b		3-4	liquid	liquid + vapor + (± opaque ± unknown)	vapor disappears
2		2	vapor	vapor + liquid	no response on heating
3a	3al	3	liquid	liquid + vapor + halite	vapor disappears
	3ad	3	liquid	liquid + vapor + halite	salt dissolution
3b	3bl	3-5	liquid	liquid + vapor + halite + (± opaque ± unknown)	vapor disappears
	3bd	3-6	liquid	liquid + vapor + halite + (± opaque ± unknown)	salt dissolution
4		4-7	daughter minerals	liquid + vapor + halite + sylvite + (± opaque ± unknown)	salt dissolution

Table 2. Fluid inclusion populations with associated vein mineralogies, alteration and inclusion types and the range of homogenisation temperatures and salinities (eq.wt.% NaCl, KCl & $CaCl_2$). Population A inclusions also occur as secondary fluid inclusions in primary quartz in the Nasuji Pluton.

Inclusion Population	Vein Mineralogy	Alteration Type	Inclusion Types	Homog. Temp. (°C)	NaCl (eq.wt.%)
A	qtz ± anh ± bio ± mt ± tr/at ± cp	biotite	3a + 3b + 2 + 1a + 1b ± 4	267' to >600°C	34.3 to 61.6 (KCl : 11.8 to 19.9)
B	qtz ± epi ± tr/at ± anh ± cc ± wai ± adu ± py ± sph ± ga ± cp	illite, propylitic (epidote+ tremolite-actinolite)	1a + 2	214" to 356°C	0.0 to 10.5 ($CaCl_2$: 3.8)
C	qtz ± anh ± cc ± wai ± adu ± py ± cp	illite, propylitic (chlorite)	1a	189" to 345°C	0.0 to 4.5

Abbreviations: adu = adularia; anh = anhydrite; bio = biotite; cc = calcite; cp = chalcopyrite; epi = epidote; ga = galena; mt = magnetite; py = pyrite; qtz = quartz; sph = sphalerite; tr/at = tremolite-actinolite; wai = wairakite. Homog. Temp. = homogenisation temperature.

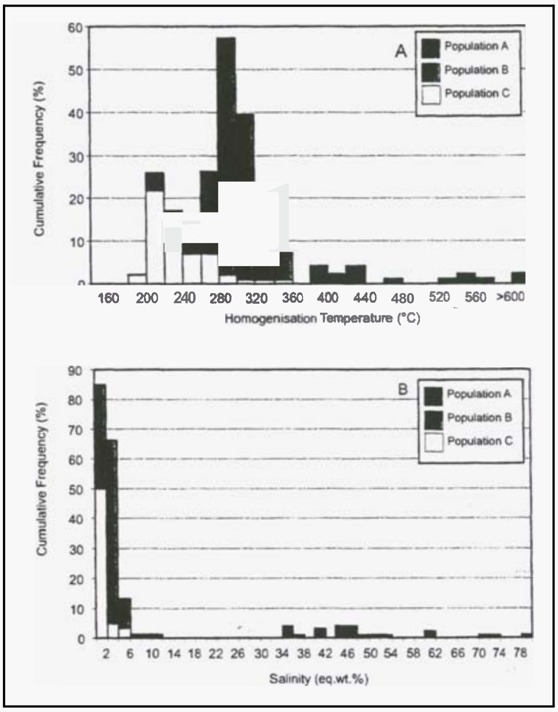

Figure 3. Histograms summarising homogenisation temperature (A) and calculated salinities (B; eq.wt.% NaCl, type 4 fluid inclusions are eq.wt.% NaCl + KCl).

5.2. Microthermometry Analytical Methods

Fluid inclusion heating and freezing measurements on 239 inclusions were made on quartz in veins from zones of illite, propylitic, and biotite alteration. Also included were primary quartz from porphyritic and equigranular Nasuji Pluton hornblende quartz diorite. Measurements were carried out on USGS-type and Linkam MDS600 heating/freezing stages. The precision of measured temperatures is ±2.0°C for heating and ±0.3°C for freezing.

5.3. Microthermometric Results

Homogenisation temperatures for **population** A inclusions range from 267°C to above 600°C (Figure **3A). Type 3** and **4** inclusions have salt dissolution temperatures that correspond to salinities between 34.3 to 61.6 equivalent weight percent (eq.wt.%) NaCl and **11.8** to **19.9** eq.wt.% KCl (Figure **3B). Population B** inclusions have homogenisation temperatures between 214" and 356°C (Figure **3A)** and apparent salinities between 0.0 and 10.5 eq.wt.% NaCl (Figure 3B). However, observed first melting temperatures indicate that some inclusions contain H_2O-NaCl-$CaCl_2$ solutions. In such cases, the observed melting sequence indicates solutions with 1.6 eq.wt.% NaCl and 3.8 eq.wt.% $CaCl_2$ compositions. **Population C** inclusions have homogenisation temperatures between 189" and **345°C** and salinities between 0.0 to **4.5** eq.wt.% NaCl (Figures **3A & 3B).**

5.4. PIXE Results

Fluid inclusions from one sample were analysed by PIXE using the CSIRO-GEMOC Nuclear Microprobe, Sydney. Those analysed were five **population** A inclusions (four type 3b; one type 2) in primary quartz fiom the Nasuji Pluton. These have appreciable metal contents. The four type 3b inclusions have average concentrations of 6.0 weight percent (wt.%) Fe, 0.7 wt.% Zn, 0.3 wt.% Pb and 930 ppm Cu. However there is a wide range of Cu concentrations, with one inclusion containing as much as 0.2 wt.% Cu. The type 2 inclusion also contains very high concentrations of Fe **(3.4 wt.%), Zn** (0.22 **wt.%)** and Cu **(654** ppm).

6. DISCUSSION AND CONCLUSIONS

6.1. Evolution of the Palinpinon Hydrothermal System

The evolution of hydrothermal activity at Palinpinon, conspicuous by the variety of its alteration types is interpreted to have been driven by two intrusive events: the Nasuji Pluton, and an inferred 'blind' intrusion below drilled depths in the Puhagan area.

The most extensive areas of biotite and hypogene advanced argillic alteration are in the Nasuji-Sogongon region, associated with the intrusion of the Nasuji Pluton. Also present in this region are calc-silicate, propylitic, illite and steam-heated advanced argillic alteration assemblages. Ar/Ar dating of primary hornblende from the Nasuji Pluton has established its emplacement between 0.7-0.3 Ma (Rae et al., submitted). This age is close to the formation of hydrothermal biotite (0.7-0.6 Ma) and alunite (0.9-0.8 Ma) from proximal zones of biotite and hypogene advanced argillic assemblages. This implies that a hydrothermal alteration imprint was established during intrusion of the Nasuji Pluton. However, the older advanced argillic alteration is probably the better estimate for the timing of the intrusion emplacement and the establishment of the associated magmatic-hydrothermal system with the deeper samples taking longer to cool below their argon closure temperatures.

Based on the fluid inclusion microthermometry by Rae et al. (submitted), during the latter stages of magma crystallisation, a two phase, aqueous saline fluid exsolved from the siliceous melt under lithostatic pressures at temperatures in excess of 600°C and minimum depths of 2-2.5 km. This two phase fluid consisted of a low density vapor and a high density saline (60-80 wt.% NaCl) liquid. Under lithostatic pressures and ductile deformation, these aqueous fluids escaped into the surrounding rocks to form high temperature (~550°C) biotite alteration assemblages at approximately 1.5 km depth (Rae, 2002).

With cooling to approximately 400°C, there was a change in pressure conditions due to hydrostatic pressures exceeding lithostatic load. This caused the vestiges of liquid magma to quench and form the porphyritic quartz diorite of the Nasuji Pluton, but also hydrofracturing and brecciation of solidified parts of the host intrusion and proximal country rocks. Hydrofracturing caused enough permeability to develop breccia zones and quartz veins, both associated with the biotite alteration assemblages. PIXE analyses show that these fluids were endowed with base metals (e.g., up to 0.2 wt.% Cu), but the lack of sulfide accumulations at Palinpinon implies either low fluid volumes andor lack of an effective trap for sulfide deposition.

Coeval with the biotite alteration assemblage, a zone of hypogene advanced argillic alteration formed at shallow levels above the Nasuji Pluton. Magmatic volatiles (e.g., SO_2, HCl) escaped fiom the crystallising magma, and ascended to shallower levels along near-vertical zones of permeability (faults and/or joints). Near the surface these gases *mixed* with groundwaters, to form acidic aquifers (i.e., pH < 2; Hedenquist, 1986) that developed the hypogene advanced argillic alteration assemblages. The depth of formation of this alteration zone remains unknown, however the occurrence of high temperature (>250°C) minerals associated with propylitic (epidote) and advanced argillic (zunyite) alteration assemblages in surface outcrops implies at least 450 m of erosion (Reyes, 1990) over the past 0.9-0.8 Ma.

As the Nasuji Pluton cooled the proportion of fluid sourced from the intrusion was reduced and hydrothermal circulation became dominated by dilute, meteoric-derived groundwaters (Rae, 2002). These moved into the pluton where they were heated and caused propylitic (epidote + tremolite-actinolite) alteration of the intrusive rocks, overprinting earlier-formed biotite alteration assemblages. At shallower levels, hydrothermal circulation of meteoric groundwaters probably resulted in the formation of illite and propylitic (chlorite) alteration mineral assemblages in Nasuji-Sogongon and extending eastwards into Puhagan. In regions above the paleo-water table, gas dissolved into perched aquifers and may have caused the foxmation of steam-heated advanced argillic alteration assemblages.

The erosion of at least 450 m of rock in the Nasuji-Sogongon area sometime within the last 0.8 Ma, exposed the Nasuji-Sogongon zone of 'HS-style' advanced argillic alteration as a series of northeasterly striking alunite ridges along the Okoy Valley. During this period, an intrusion was emplaced beneath the Puhagan dikes (>2.5 km depth) at the intersection of the Ticala and Lagunao Faults. This intrusion shifted the focus of hypogene alteration (calc-silicate and biotite) eastwards and caused the development of the hydrothermal convection cell that defines the present-day geothermal system. Since the calc-silicate assemblage formed, this region has cooled by at least 70°-100°C, while parts of the biotite alteration assemblage have remained in thermal equilibrium with present-day fluids.

With convection, zones of propylitic alteration developed as halos to the Puhagan hypogene, calc-silicate and biotite alteration assemblages. These propylitic zones are in thermal equilibrium with the present-day system. The tremolite-actinolite assemblage is restricted to the upflow zone, proximal to the hypogene alteration assemblages. Away from this center, the subzones vary from epidote to chlorite assemblages. Such a systematic variation away fiom a core of biotite alteration has been recognised in the Tintic porphyry Cu system (Norman et al., 1991).

Above the zones of propylitic alteration, illite alteration assemblages overprint biotite, propylitic and advanced argillic alteration zones associated with the Nasuji-Sogongon hydrothermal system. Dissolved gases (CO_2, H_2S) that separated from boiling zones, and redissolved into perched aquifers, have formed steam-heated acid sulfate waters with associated advanced argillic alteration zones. Descent of acid sulfate waters down near-vertical permeable structures (i.e., faults), results in steeply dipping zones of advanced argillic alteration (e.g., in the Puhagan area).

6.2. Implications for Mineral Exploration

The hydrothermal alteration assemblages at Palinpinon are interpreted to record a long-lived (0.8 ± 0.1 Ma) hydrothermal system that evolved with respect to: temperature (600" to 300°C); fluid sources (dominantly magmatic to dominantly meteoric); acidity (acid to near neutral); and confining pressures (0.3-0.4 kb lithostatic to 0.1-0.2 kb hydrostatic). There is an intimate spatial and temporal relationship between the intrusion emplacement and the styles of alteration that are characteristic of deposit styles such as porphyry, skarn, HS and LS epithermal. This work shows that all four alteration system can occur in a single mineral district and may form as a protracted, evolving hydrothermal system with LS epithermal assemblage postdating the hypogene porphyry-HS epithermal alteration assemblages.

Despite the parallels that can be drawn between Palinpinon and mineralized porphyry and epithermal ore deposits, assay results have failed to detect any significant mineralized zones of copper, zinc, lead, gold and silver at Palinpinon (Rae et al., submitted). It may be that wells drilled to date have failed to intersect ore zones, or that such zones have since eroded. Until any evidence to the contrary is obtained Palinpinon is interpreted to be a barren hydrothermal system. Reasons for could be: (1) poor permeability necessary to focus fluid flow and ore deposition; (2) insufficient fluid flux to accumulate large amounts of base and/or precious metals; andor (3) a lack of enough metals. Important features that characterise porphyry, HS and LS deposits are apparently absent at Palinpinon. These include, high density quartz stockwork veining (porphyry), a core zone of vuggy silica (HS epithermal) and vein swarms, massive crustiform veins or mineralized breccia complexes (LS epithermal). All of these features intimate that high grade ores at these deposit types have been promoted by large volumes of fluid and highly permeable wallrocks. The absence of these features at Palinpinon indicates that permeability and/or fluid flux have been insufficient and that a surge in magmatic activity, faulting, or catastrophic decompression and breccia formation are required to promote ore deposition.

7. ACKNOWLEDGEMENTS

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