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CONTROLS ON DISTRIBUTION OF Hg IN GEOTHERMAL WELLS FROM NGAWHA, NEW ZEALAND AND PUNA, HAWAII

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ABSTRACT

Cuttings from five geothermal wells at Ngawha, New Zealand (Ng3, 18, 20) and Puna, Hawaii (HGP-A and KS1) were analysed for Hg at 30-50m intervals. Results show a primary control by temperature over Hg concentration and a consequent distribution which reflects the form of the Hg present. Adsorbed Hg is stable in rocks up to c. 150°C, Hg in silicates to c. 250°C, Hg in pyrite to c. 300°C and over c. 320°C the Hg values are comparable to unaltered rocks and low. The location of a well within a field relative to zones of permeability is important and higher Hg values in shallower parts may reflect the more permeable location. Temperature, however, must also be considered and higher values throughout a hole may reflect lower temperatures. Apart from any shallower enrichment by adsorbed forms of Hg, downhole Hg enrichment was found to be associated with secondary minerals; those having formed directly from solution by boiling containing higher Hg concentrations than those formed by processes involving steam separation - condensation. Repeat analyses over time show that after periods on the order of one year appreciable Hg can be lost during storage, and after several years most of the Hg introduced to the rock can be lost, at least for lirhologies such as basalt.

IXTRODUCTION

Surface enrichment of Hg is a common feature over many geothermal systems and in some cases mineable deposits of Hg have formed in association with geothermal systems (e.g. White, 1967). Mapping of Hg concentrations in soils is now a commonly applied technique in geothermal exploration and can delineate anomalous zones representing both permeability and convective heat transfer (e.g. Cox, 1981). Mercury analyses of cores and cuttings from geothermal wells have shown features of Hg distribution and form in some systems and assisted in delineating their margins (Christensen et al., 1980; Moore et al., 1982). To understand further the controls involved I have examined the vertical distribution of Hg in relation to measured downhole temperatures in two geothermal systems of different character. Five wells were chosen, two at Puna, Hawaii and three at Ngawha, New Zealand.

The Ngavha field is situated in the northern part of the North Island of Nev Zealand about 400 km from the area of current, subduction related volcanic activity of the Taupo Volcanic Zone. The Ngawha geothermal system is, therefore, isolated and its heat source is considered to be most likely a hot, solidified rhyolitic body (e.g. Heming, 1979). The reservoir of the system is within Permian-Jurassic greywacke basement rocks (e.g. Skinner, 1981) and permeability appears to be provided by faults, and joints (e.g. Browne, 1980). The fluid reservoir is contained by caprocks formed of 500-600m of very low permeability Cretaceous-Tertiary sediments. These sediments are a chaotic megabreccia comprised of a variety of siltstone, sandstone and shales which inhibit water movement.

The greywackes and argillites which form the reservoir (Browne, 1981) are hard, dense, moderate to poorly sorted and contain quartz, albite, K-feldspar, calcite, illite, chlorite, ziron, pyrite, prehnite and pumpellyite. Their hydrothermal alteration is mainly of two types (a) veining, and (b) alteration of primary clay minerals to other clays. Additionally, there are local occurrences of disseminated hydrothermal minerals such as pyrite, calcite and epidote. Veins and lenses of secondary minerals occur in both caprock and reservoir rocks. Minerals comprising the veins are mainly quartz, calcite, chlorite, illite, K-feldspar, pyrite, pyrrhotite and hematite. Overall, calcite is widespread in the caprock, but becomes less abundant with depth. Pyrite has a similar distribution.

Surface features at Ngawha are limited to three parallel permeable zones along which gas discharge occurs. Mercury deposited at less than c. 10m within lacustrine sediments along the central zone has been mined, producing about 85 tonnes of Hg. The average grade of earlier mining was 1.5% Hg (Bell and Clarke, 1909). As a consequence of the low permeability of the caprock, hot springs, as such, have not developed and the hot pools (to 50°C) are formed of surface or shallow groundwater heated by rising hot gases. The reservoir fluid is of NaCl type and temperatures typically 230°-240°C; well dfscharge water is typically, pH = 7.6 (at 160°C), SiO_2 = 460 ppm, SO_4 = 30 ppm, Mg = 0.1 ppm, and Cl = 1470 ppm (Sheppard and Ciggenbach, 1980).

The geothermal system at Puna is within the lower east rift zone of Kilauea volcano, 25km from the summit. Drillholes and surface surfaces indicate that the reservoir is horizontally rift-contained by impoundment by the numerous dykes within the rift, and vertically by reduction of permeability by secondary mineral deposition. The east rift is volcanically active, the last eruption in the Puna area being in 1960. Because of the spatial relation to the summit, the heat source to the geothermal system is most likely to be hot basalt intrusions within the rift. Reservoir fluids are NaCl type of 10-15% seavater, temperatures are 320-330°C and the deep pH around 3.5. Discharge chemistry is typically pH 6-7 (190°C), SiO_2 = 875 ppm, SO_4 = 170 ppm, Mg = 0.2 ppm, Cl = 2920. At full discharge HCP-A can produce 45.5 t/hr of fluid with a steam fraction of .48 (Thomas, 1982). The reservoir is within a sequence of thin flows and pillow lavas of tholeiitic basalt, and permeability is usually due to fracturing and zones between flows rather than vesicularity (Stone, 1977). Beneath a shallow zone of unaltered lavas, three zones of characteristic hydrothermal alteration occur (Stone, 1977): 675-1350m, smectlte (montmorillonite) with minor calcite, quartz, zeolite and chlorite; 1350-1894m, chlorite with quartz, actinolite and smectite; from 1835m actinolite begins to dominate becoming abundant at 1959m, with accessory chlorite, quartz and opaque grains (mainly hematite and pyrite).

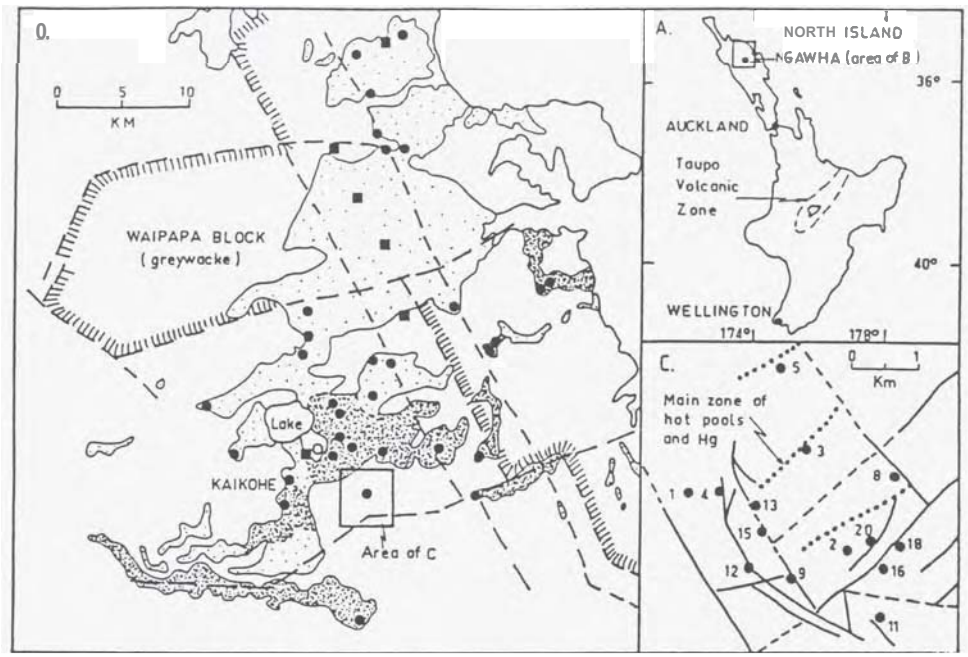

Fig 1: A. Location of Ngawha area. B. Simplified geology of Ngawha area showing mapped faults, uplifted greywacke basement and Quaternary basalt flows (older Horeke, light stipple; younger Taheke, dense stipple). Solid circles are basalt volcanic centres; solid squares, rhyolites locations; open circle, andesite centre. C. Ngawha drill field showing wells, simplified faults (after High, 1982) and zones of surface activity.

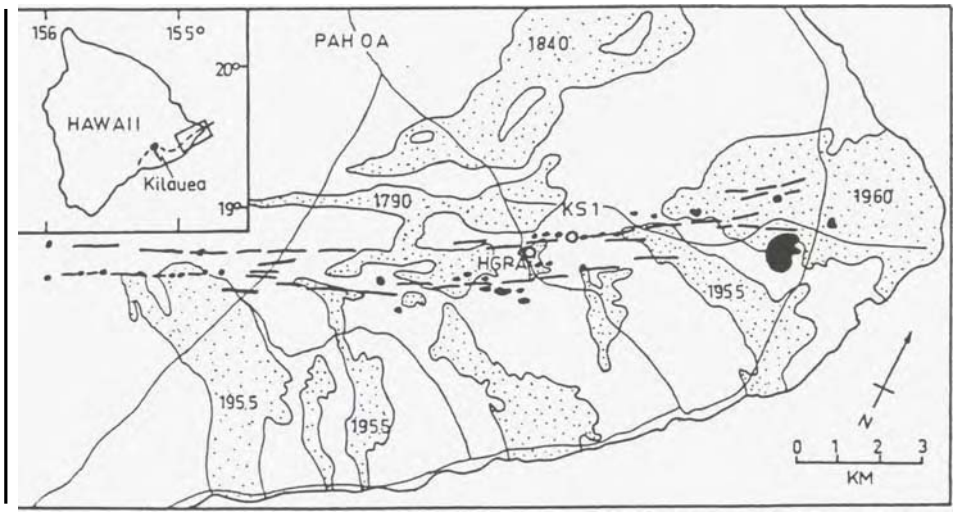

Fig 2: Puna area, lower east rift zone of Kilauea volcano, island of Hawaii. Trace of tift zone is shown by cinder cones and fissures; historic lava flows are shown (stippled). Locations of wells HGP-A and KS1 are shown by open circles.

TABLE 1: Average Contents of Hg in Geothermal Fluids (in ug/l)

Sample	Water	Steam	Total Discharge	Ref.
NGAWHA				
Main surface pools	27.3	129		a
	(temp-dependent)			
Ng2 discharge	5.2	55.3		b
Ng4 "	1.2	168.3	40.7	b
Ng9 "	3.1	182.5	50.1	b

Sample		Water	Steam	Total Discharge	Ref.
PUNA			Approx temp		
Downhole	305m	35.4	78°C		
	692m	3.0	245°C		c
"	1310m	3.5	318°C		c
"	1769m	3.2	308°C		c
"	1920m	1.6	350°C		c
Discharge		0.4			d

a. Davey and Van Moort, 1974
b. Giggenbach and Sheppard, 1980
c. S Siegel, pers. comm. 1978.

Mercury has been analysed on powdered cuttings from intervals of 30-50m. Approximately 5g of material was selected with around 0.5g of homogenised powder analysed using a Jerome gold film mercury detector. Samples were heated to over 800°C and the concentration of Hg released determined from changes in resistivity of a gold film due to Hg adsorption by comparison to injected standards of Hg vapour and sample weight. The limit of detection is 2 ppb.

Wells analysed

Three Ngawha wells were selected (Ng3, Ng18, Ng20) to provide variation in Location and characteristics; these were drilled in 03.81, 06.82 and 07.82, respectively, by NZ Ministry of Works and Development (MUD). Cuttings from two Puna wells were analysed, HGP-A, a research drillhole (University of Hawaii; State of Hawaii; US Department of Energy) which was completed in mid 1976, and KS1 (Amfac; Dillingham; Thermal Power Company) completed 10.81. KS1 is about 800m north of HGP-A. The most representative downhole temperature profiles are shown in Figures 3 and 4; as KS1 is a commercial venture, data from that well are proprietary and unavailable, consequently, some approximations are made using HCP-A downhole data.

Mercury in Fluids

Mercury in Ngawha fluids is much higher than in other New Zealand geothermal fields, and as for many wells, especially those with a higher gas content, concentrations in separated steam are appreciably higher than in discharge water (Weissberg and Rhode, 1978). Concentrations for a variety of fluids are shown in Table 1; in summary, average values from the Ngawha wells Ng2, 4 and 9 were 2.1 ug/l in discharge water, 175 ug/l in separated steam and about 48.8 ug/l for reservoir fluid (T.D.) (Ciggenbach and Sheppard, 1980). In the central active zone at Ngawha, all gases and most surface waters are saturated with Hg at their temperature; gases at 20°C contained 13 ug/l Hg and at 60°C, 245 ug/l (Davey, 1974; Davey and Van Moort, 1974).

Wellhead water samples from the 1976 production tests of HGP-A are reported to have Hg concentrations of several hundred ugll at the beginning of tests to less than 50ug/l at the end (Kroopnick et al., 1978). Hg in downhole samples (Table 1) shows that a reservoir concentration of c. 3 ugll Hg is likely; this is in contrast to c. 45 ugll calculated for the Ngawha reservoir fluid. These data show a higher Hg concentration in shallow waters and suggest an inverse correlation with temperature. No Hg analyses for separated steam at the wellhead are available.

RESULTS

Hg in Secondary Minerals

Various secondary minerals were separated and identified by XRD. Pyrite grains separated from Ngawha drill cores at various depth contained Hg concentrations of 600 - 3700 ppb; pyrrhotite grains consistently contained lower Hg, commonly 50-60 ppb. Near surface marcasite in the main zone of activity is reported to have Hg contents of 1000 ppm to 4.4% (Davey and Van Moort, 1974). Other secondary materials were also low in Hg: quartz veins, 40-185 ppb and calcite veins 20-70 ppb. Calcite scraped from the casing of Ng9 (400-750m) contained 42 ppb. Amorphous silica deposited as scale on Ng9 silencer tower (presumably from a condensate) contained 19 ppb; amorphous silica scale in a temporary overflow drain from Ng9 weir box contained 110 ppb, and amorphous silica scale in the header tank to which the drain discharged contained 493 ppb. No cinnabar was identified in any of these samples; Hg in the surface forming samples is presumed to be Hg° within their structure. Davey (1974) determined that cinnabar deposition at Ngawha resulted from oxidation of Hg" vapour in acid water during rain, and subsequent reaction with sulphide dissolved in the surface waters.

Pyrite grains from KS1 cuttings (450m) contained 4200 ppb Hg. Secondary materials scraped from HGP-A casing (approximate depths only) contained: calcite, above 990m = 1254 ppb; pyrrhotite, 1200m = 25 ppb; pyrrhotite, 1750m = 20 ppb and anhydrite, 1780m = 14 ppb. Amorphous silica rapidly deposited from HCP-A discharge water by addition of NaOH contained 77 ppb Hg.

In summary, pyrite contains appreciably more (over 10x) Hg than pyrrhotite and quartz usually contains more Hg than calcite. The initial interpretation of these data is that secondary minerals deposited as a result of steam separation and condensation will have low Hg contents and those deposited from an aqueous phase (resulting from boiling and saturation) will have higher Hg contents. Calcite from above 990m in HGP-A was discharged from the well after a sealing packer was installed (10.79) at 993m. It corresponds to a zone of calcite filling vesicles in cores (Stone, 1977) and may result from saturation of fluid with respect to $CaCO_3$ by rapid temperature increase and not condensation.

Hg Loss from Stored Samples

As seen in Figure 3, downhole Hg values in HCP-A cuttings (and cores) are low, commonly 10-20 ppb. Consistent results for these samples were obtained by three separate analyses and were mostly within analytical uncertainty (±10%). These analyses were carried out 3, 5 and 6½ years after the well vas drilled and the cuttings stored in plastic bags at room temperature (25°C). The low values appeared anomalous considering other soil-rock analyses from Hawaii (e.g. McMurtry et al., 1979; Cox, 1981) and a report of concentrations of 30-240 ppb (depths not reported) for HGP-A cores analysed about (?) six months after drilling (S. Siegel, pers. comm., 1978). These aspects and the subsequent analyses of KS1 cuttings, indicated that Hg was lost from the HCP-A cuttings during storage.

KS1 Hg results in Figure 3 (solid lines) are analyses made six weeks after drilling; solid circles show partial repeat analyses of the same cuttings 16 months after drilling. From these results it is apparent that Hg loss can occur during storage, at least for basalts. The form of Hg in the sample is also important. For the KS1 samples the percentage loss is highest in shallow samples (70-90%); loss was 40-70% at intermediate depths, but in the lower part of the hole there was no apparent loss. This is interpreted as indicating differing degrees of stability of the form of the Hg present. Although not exhaustively tested for Ngawha samples, work to date suggests that such storage loss from the lithologies at Ngawha had not occurred by the time of sampling, and would be much less over longer periods.

Vertical Distribution of Hg-Puna

Due to Hg loss during storage HGP-A results cannot be interpreted, and the original KS1 concentrations are assessed using the HGP-A temperature profile as an approximation- For HGP-A cuttings Stone (1977) first observed pyrite at 485m and a general increase in pyrite with depth. Euhedral pyrite grains were described as occurring with quartz and clay minerals in voids in the lower 500m of HGP-A. A qualitative comparison between KSL and HGP-A cuttings shows that KS1 cuttings contain significantly more pyrite, mostly as disseminated grains, from a depth of c. 450m. The location of these wells is important, HCP-A being between two linear fissure zones (marked by cinder cones) In an apparent offset; KS1 is sited within the fissure zone from which the 1960 lavas erupted (Figure 2). It is considered likely, therefore, that KS1 occurs within a more permeable zone which may result in a higher concentration of volatile elements within fluids there; this is in turn reflected in the greater abundance of pyrite. This suggestion is also supported by surface Hg surveys (Cox, 1981) in which soil Hg values were 95-178 ppb in the area of the vells arid KS1 (drtlled after these surveys) Is located within a linear anomaly along the fissure zone with Hg

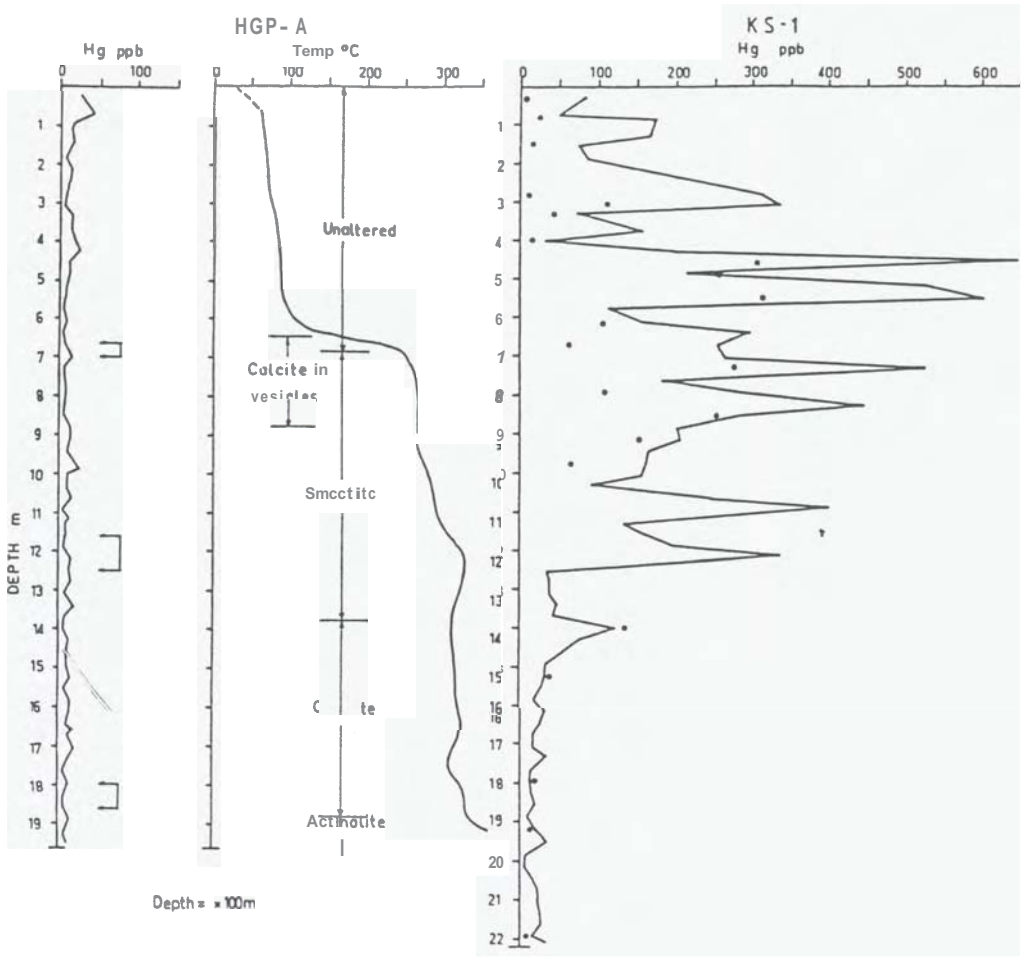

Fig 3: Downhole Hg analyses in ppb for wells HGP-A and KS1. Temperature profile of HGP-A is from Kroopnick et al., 1978; zones of alteration minerals, Stone, 1977 ; production zones (arrows) from Thomas, 1982 ·

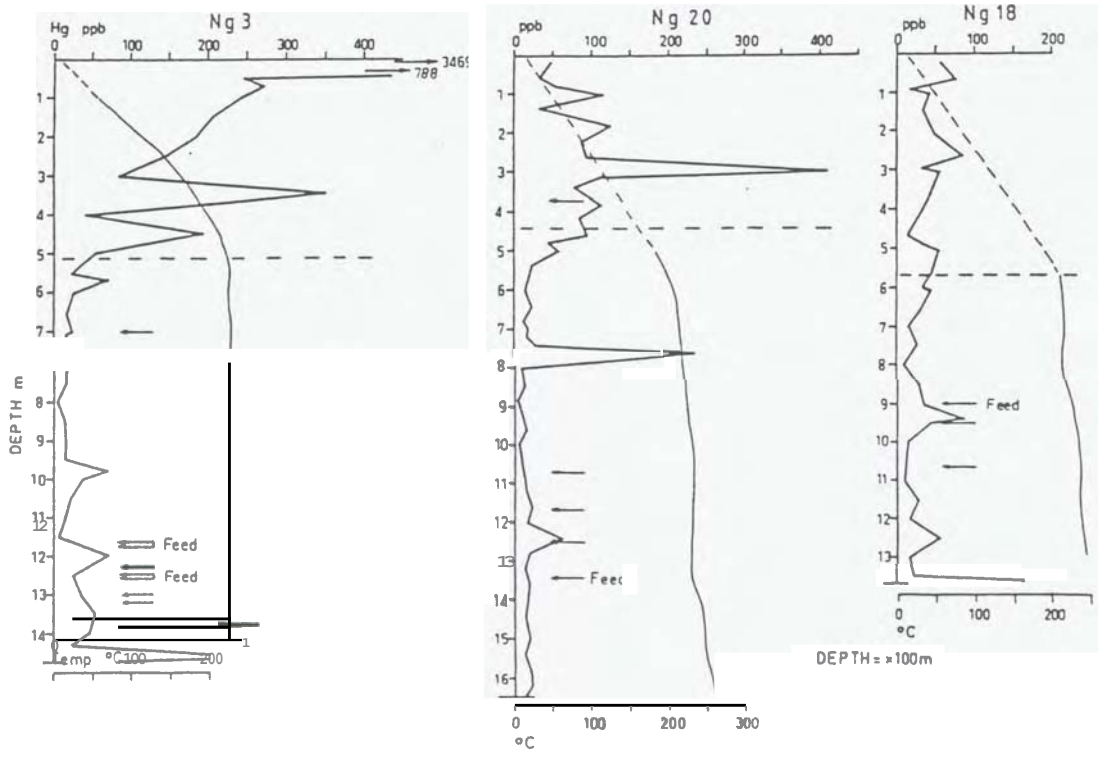

Fig 4: Downhole Hg analyses of cuttings from Ngawha wells Ng3, 18, 20. Temperature profiles and permeable zones (arrows) from unpub. MVD reports. caprock-reservoir greywacke contact (broken Line) from unpub. reports

The Hg profile of KS1 cuttings suggests three zones: shallov, 0-450m of 100–300 ppb; intermediate, 450-1250m of 150-500 ppb; and deep, below 1250m of mostly less than 50 ppb. Due to the uniform lithology in this well, other parameters are important in influencing Hg concentration - mainly temperature and secondary mineralisation.

Vertical Distribution of Hg-Ngawha

For assessment, the location of the Ngawha wells within the drillfield should be considererd (Figure 1,c). Ng3 is near the centre of the drillfield closeby the main zone of surface activity (and main zone of natural Hg deposition); MWD drilling/ completion test reports shov it to be a small producer (maximum flow 107 t/hr). Ng20, a poor producer (maximum flow c. 80 t/hr) but with temperatures to 260°C is 40m south of the southern zone of surface features (acid lakes and gas seeps); Ng18 is located towards the SE margin of the field and is separated from Ng20 by a fault with a vertical displacement of 130m. It is a small producer (flov 103 t/hr).

Vertical profiles of Hg are shown for the wells (Figure 4) with temperature profiles (MWD reports) and caprock–basement contact (from NZCS logs and P R L Browne unpub. petrology reports). Hg in all wells shows an overall decrease within the greywacke reservoir to similar baseline values of 10–30 ppb, but with peaks of higher values. Reservoir Hg concentrations are lowest in Ng20, and the difference between caprock and reservoir values is lowest in Ng18. Within the caprock the highest concentrations occur in Ng3, overall increasing towards the surface vith 3469 ppb at 10m depth. Caprock Hg in Ng20 is mostly 50–100 ppb with a peak of 409 ppb at 300m. Ng18 caprock concentrations are the lowest and least variable, between 20–80 ppb. Neither Ng20 or Ng18 have the very high Hg concentrations at shallow depth. Virtually all the high peaks of Hg can be correlated with the presence of pyrite in cuttings, usually in veins associated with calcite and quartz, but also as disseminated fine grain pyrite.

Discussion of Hg Distribution

Figures 3 and 4 show the importance of temperature in controlling the overall pattern of Hg distribution. In the Ngawha wells studied measured reservoir temperatures were commonly 230-240°C with a maximum of 260°C in Ng20, whereas at Puna (HCP–A) the reservoir temperature is 300–320°C with a maximum of 358°C. The high Hg values in the almost impermeable caprock at Ngawha indicate that Hg is being transported through that material as vapour. Vapour transport was also concluded by Davey and Van Moort (1974) to have formed the shallow Hg deposits at Ngawha.

Although the Puna area is diffetent from Ngawha hydrologically the reservoir is "sealed" and there is no surface water discharge. Rising separated vapour passes through cool shallow groundwater to the surface and the highest soil Hg concentration measured along this part of the rift was 1250 ppb (Cox, 1981) which suggests a lower concentration of Hg in gas than at Ngawha.

The association of higher Hg values with pyrite has been noted, plus higher values at shallow depth (Ng3) which are not associated with pyrite. Also noted are the lowest values in the deeper, hotter parts of all wells. Aspects of temperature have been considered elsewhere, experimentally and in field studies. Dickson (1964) determined that with an increase in temperature to c. 250°C HgS begins to become unstable. Heating experiments (Landa, 1978; Christensen et al., 1980) show that adsorbed Hg is largely mobllised over 100°-200°C (major losses from 100°-150°C) and that loss of normally more stable Hg sulphide begins at 210°-270°C with complete liberation by 340°C.

For samples from The Geysers, Moore et al. (1982) consider Hg loss beginning at 150°C represents release of Hg within silicate minerals, the major form of Hg occurrence in the rocks. They considered that loss at c.250°C may represent Hg as cinnabar (although not recognised) and Hg held to 400°C vas probably as a trace constituent of pyrite. No cinnabar vas recognlsed or Identified by XRD in any of the Ngawha or Puna samples.

In the shallow parts of the vells, at temperatures to c.150°C, Hg is indicated to have deposited in rocks largely as uncomplexed adsorbed Hg^0 resulting from the temperature decrease, but also associated wlth secondary minerals (pyrite, quartz, calcite) producing individual peaks. In KS1 this appears the case above 600m and c.100°C. Over c.150°C adsorbed Hg is no longer stable and most Hg within the rock is associated with pyrite (and to a lesser extent quartz) to temperatures of c.250°C for quartz and 300°-320°C for pyrite. In the Ngawha wells studied this is basically the condition vithin the reservoir, where temperatures of 300°C are not reached, but Hg peaks associated with pyrite occur. In KS1 this appears to be the condition from c.600-1250m. Over c.300°C Hg is unable to form a stable association and remains mobile, as in the lower part of KS1.

Downhole Hg analyses at Roosevelt Hot Springs geothermal area, Utah (Christensen et al., 1980) shoved Hg vas distributed largely within the outer parts of the system at less than c.225°C. It vas concluded that Hg is progressively deposited by outward flowing, cooling, thermal waters, and its distribution largely reflects the present system's thermal configuration. Mercury concentrations in that system are lover, with maximum values of 100–150 ppb and baseline values of 5–20 ppb. Those authors concluded the Hg vas present as adsorbed Hg, rather than associated with sulphide minerals.

The chemistry of fluids associated with Hg deposits summarised by White (1967) shows that most large deposits are related to discharging alkaline springs. He also notes that where Hg^0 is associated with S vapour and acid environments, Hg deposits are typically small and low grade. Under acid conditions and with the presence of high dissolved S, the total dissolved Hg in fluids will be low (Schwarzenbach and Widmer, 1963). Hg and S are most likely to remain dissociated under these conditions of high temperature and low pH.

Both Puna and Ngawha are high gas systems (discharges are around 48% and *30%* vapour, respectively). Reservoir conditions indicate that the Hg is derived from degassing intrusive masses, the age and size also determining the amount of S being released. Briefly, Ngauha could be considered a low S system (discharge water = 35 ppm SO_4; and H_2S is c. 1%of gases); in comparison Puna is a high S system (SO_4 = 175 ppm; H_2S is c. 39% of gases). It would appear that under natural conditions Hg discharge to the surface at Puna is Low and the geothermal system itself is low in Hg. This is possibly both a function of the relatively small site of the heat source below Puna, its basaltic composition and the high S content of the fluids. The abundant S may react with Fe oxides and silicates to form Fe sulphides, principally pyrite, and the O_2 released may form *SO4* with the available S. The high S, acid character of the fluid apparently inhibits its Hg carrying capacity, and at temperatures below c.300°C allows incorporation of Hg into the structure of pyrite being formed.

At Ngawha the likely silicic heat source and the low S character of the system appear to favour a higher Hg input, and thus content of reservotr fluids. Where conditions enable formation of seconddry minerals directly from solution (e.g. pyrite and quartz in joints) Hg becomes incorporated, however, most Hg remains mobile as uncomplexed Hg vapour.

CONCLUSIONS

This brief study indicates that temperature is the primary control on subsurface distribution of Hg, showing broad zonation of: adsorbed Hg to c.150°C, Hg in quartz to c.250°C, Hg in pyrite to c.300°C, and over 320°C low Hg values comparable to unaltered rocks. It is obvious that chemical characteristics of the fluid within a system, and the type of the system (e.g. heat source) also affect the relative abundance of Hg, notably S content.

There is a generalised increase of Hg in reservoir rocks around permeable and production zones, presumably related to secondary mineral formation, largely pyrite and quartz. Mercury is characteristically different in the main sulphide minerals, higher in pyrite and low in pyrrhotite, related to the conditions of formation. Broadly, secondary minerals deposited directly from aqueous solution by boiling processes (e.g. pyrite and quartz) contain higher amounts of Hg; minerals formed by boiling-steam separation-condensation processes (e.g. pyrrhotite and calcite) are low in Hg with values slightly above background values far unaltered rocks. Marcasite, which is very high in Rg, forms only at the surface along the main zone of activity at Ngawha.

For downhole samples of Lithologies such as basalt, with a low retention ability for Hg, it is important to analyse for Hg as soon as possible after their collection, or to store samples under refrigeration.

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