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SULFIDE SCALING OF DEEP-GEOTHERMAL WELL AT KAKKONDA GEOTHERMAL FIELD IN JAPAN

Norio Yanagisawa[1], Koichiro Fujimoto[1] and Yasuyuki Hishi[2]
[1]Geological Survey of Japan, Higashi 1-1-3, Tsukuba, Ibaraki,Japan
[2]Geothermal Engineering Co. Ltd.,Sasamori-72, Ukai, Takizawa, Iwate, Japan

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ABSTRACT

Sulfide rich scales in the Kakkonda geothermal area, Northeastern Japan, are investigated. They are precipitated from the geothermal fluid from the deep reservoir that exists at the boundary between Quaternary Kakkonda granite and Pre-Tertiary formations. The scales are classified into two types based on sulfide mineralogy, which are Pb-Zn rich type and Cu rich type. Pb-Zn rich scale is found in Well-19 located at the marginal part of the Kakkonda granite. It is mainly composed of amorphous silica, galena (PbS), sphalerite (ZnS) and pyrite (FeS_2). Cu-rich scale is found in Well-13, located at the central part of the Kakkonda granite. It is mainly composed of amorphous silica, chalcocite (Cu_2S), bornite (Cu_5FeS_4), chalcopyrite ($CuFeS_2$), galena (PbS), loellingite ($FeAs_2$) and native antimony (Sb). It is also rich in Au, Ag, As, Cr, Ni and Mo compared to Pb-rich scale. The metal contents of the scale in Well-22 show intermediate features between the two types of scales. The existence of both Pb-Zn rich scale and Cu-rich scale is a characteristic feature of the Kakkonda geothermal area. This is probably due to the difference in the parent fluid. The brine in Kakkonda granite may be an important metal source of the scales.

1. INTRODUCTION

The Kakkonda geothermal field is located in the northern part of Honshu Island, where a liquid-dominated geothermal system has been utilized for power generation totaling 80 MW.

A Quaternary granite underlies the Pre-Tertiary sediments at more than 2000 meters depth. The granite influenced the surrounding rock with thermal metamorphism and acts as a heat source of the present geothermal activity. The deep geothermal reservoirs exist near the boundary between Pre-Tertiary formations and the Quaternary granite with a fracture system (Doi *et al. (*1995), Kato and Doi (1993) and Kato and Sato, (1995)).

Because the fluid from the deep reservoir has higher vapor/liquid ratio and higher enthalpy than that of the shallow reservoir, it has been used for the second power generation unit of Kakkonda since 1996. The fluid from the deep reservoir is about pH 4, weakly acid (under atmospheric pressure and room temperature), and shows a difference in composition from neutral shallow reservoir fluid (Yanagiya *et al.*,1996).

The higher enthalpy fluid with much dissolved silica causes the deposition of scale in the pipe-line. The scale commonly causes problems by restricting fluid flow, preventing valves from closing, and scaling turbine blades. Thus scaling can be a serious problem during geothermal exploitation.

On the other hand, the scales give us critical information about the nature of the parent fluid, the origin of the fluid and the evolution process.

In this paper, we investigated the scales of the ground pipe line near the separator of several production wells from a deep reservoir in the Kakkonda geothermal system. We analyze the mineral assemblage and chemical composition of the scales, clarify the difference of the scales between wells and discuss the relationship between the scales and the hydrothermal activity in the Kakkonda area.

2. GEOLOGY AND SAMPLING POINT OF THE SCALE

Figure 1 shows a schematic geological profile of the area along the Kakkonda River (Doi et al., 1995). Among the rocks that have been reached by drilling in this area, pre-Tertiary sedimentary rocks (sandstone and slate) intercalated with acidic tuff are the oldest. The rocks are unconformably covered by Miocene formations (the Kunimitoge, the Takinoue-onsen and the Yamatsuda Formations). These sedimentary rocks are intruded by the 4.9 Ma Torigoeno-taki dacite. A Quaternary-granite pluton intruded the sedimentary and dike rocks, metamorphosing the surrounding rocks. Radiogenic age is estimated to be about 0.2 Ma (Doi et al., 1995). Rocks above -900 m ASL are intensively fractured, forming shallow geothermal reservoirs. Geothermal fluid in these reservoirs is weakly alkaline NaC1 type and has temperatures of 220-260℃. Below -900 m ASL, productive fractures are found at and near the margin of the granitic pluton. Geothermal fluid in these deep reservoirs is slightly acid NaCl type and has temperatures of approximately 300-350℃.

There are 6 wells that encounter the Quaternary granite. But several wells did not encounter deep reservoir. Therefore, we collected scale for analysis at three production wells: Well-13 is in the central part of the Kakkonda granite, Well-19 is in the marginal part of the Kakkonda granite and Well-22 is between Well-13 and Well-19. Scale was collected at production pipe line before separator on ground and after one-year of precipitation.

3. SAMPLES AND ANALYSIS METHODS

Scales from Well-13 and 19 are layered with a thickness of a few millimeters; polished thin sections were made in the direction parallel to scale growth. Scale from Well-22 is a powdered sample with a diameter of several tens of a micrometer.

Textures and mineral assemblages were observed with a reflection microscope and a scanning electron microscope (SEM).

The bulk sample was powdered for X-ray diffraction and chemical analysis. Sulfide minerals in the scale were picked out, and the ratio of sulfur isotopes was measured.

The methods of the chemical analysis are as follows:.
1) Major metal elements were analyzed by Inductively Coupled Plasma Emission Spectrometry (ICP-ES) method. For analysis, 50 ml hydrochloric acid solutions were made from 100 mg samples.
2) Na and K in the solution made by the above method were analyzed by a compact ion meter after a calibration curve was made with the standard solution of 2000 ppm and 150 ppm.
3) As was analyzed by the Instrumental Neutron Activation Analysis (INAA) method.
4) Au and Ag were analyzed by Atomic Absorption Spectrophotometry (AAS) method with tens of a microgram powder sample.
5) S and C were analyzed by High-frequency burning infrared absorption method.

4. RESULTS

4-1. Textures and Mineral assemblages

The results of X-ray diffraction and SEM observation are shown in Table 1. The scale at Well-13 contains chalcocite (CuS_2), bornite (Cu_5FeS_4) and loellingite ($FeAs_2$) as heavy metal minerals. These minerals are scattered in amorphous silica and considered of detrital origin as shown in figure 2.

The scale at Well-19 is composed of an amorphous silica layer and a galena-sphalerite layer. Galena and sphalerite show dendritic growth as shown in figure 3.

The scale at Well-22 is mainly composed of amorphous silica, barite, and galena. Small amounts of Cu-minerals are also present.

4-2. Chemical composition and sulfur isotope

The results of chemical analysis are shown in Table 2. From this table, the principle components of scales were amorphous silica and sulfide minerals. Total content of metal and sulfur is 10-50%.

Figure 4 shows the relative concentrations of several metal elements normalized to Well-19. Metal elements were separated into two groups. Firstly, base metals such as Mn, Zn and Pb are rich in the scale of Well-19; the Zn content is close to 20%. Secondly, Cu, Au, Ag, As, Cr, Ni, and Mo are rich in the scale of Well-13; the Cu content is close to 15%. Concentrations of Au and Ag in the scale of Well-13 are 19.4 and 550.0 ppm, respectively. The metal contents in the scale of Well-22 show the intermediate value between those of Well-13 and Well-19.

S and metal contents are compared for speciation. For the scale of Well-19 (Pb-Zn rich scale), analyzed S content is equivalent to the calculated one assuming all Zn and Pb form sphalerite and galena. Thus, all S is probably included in the sulfide minerals galena and sphalerite.

As for the scale of Well-22 (intermediate type scale), analyzed S content is nearly equivalent to the calculated one assuming all Ba forms barite and all Cu, Pb and Zn form the sulfide minerals chalcocite, galena and sphalerite. Thus, S containing minerals are both barite and the sulfides.

On the other hand, for the scale of Well-13 (Cu-rich scale), the analyzed S content is much smaller than the calculated one assuming all Cu and Pb are included in chalcocite and galena, both of which are detected by XRD. Such as native antimony and loellingite, not sulfide but metal element or metal composite of Cu or Pb may exist in this scale.

Sulfur isotope ratios and δ^{34} for sulfide minerals in Well-13 and Well-19 are -0.9‰ and +1.9‰, respectively.

5. DISCUSSION

Two types of sulfide rich scale, Pb-Zn rich scale and Cu rich scale, are distinguished in the Kakkonda geothermal area. Shikazono and Shimizu (1992) classified Japanese vein type deposits into two types. One is Au-Ag type that concentrates the elements such as Cu, Au, Ag, As and Sb and the other is base-metal type that concentrates the elements such as Pb, Zn, and Mn. After their classification, the Cu rich scale and Pb-Zn rich scale is similar to Au-Ag deposit and base-metal deposit, respectively. The scale of Well-22 is an intermediate type.

Sulfide scales are reported at several geothermal areas. Scales of the Salton Sea geothermal area in southern California consist of bornite (Cu5FeS4), chalcopyrite, dignite (Cu9S5), pyrite, stromeyerite (AgCuS) and arsenopyrite(FeAsS) (Skinner et al. (1967) and McKibben and Williams (1985)). Scales of the Broadland geothermal area in New Zealand consist mainly of chalcopyrite (CuFeS2) with lesser amount of galena, sphalerite, pyrite and electrum (Brown, 1986). Scales of the Yamagawa geothermal area in Japan consist mainly of galena and sphalerite, with lesser amount of pyrrhotite (FeS) (Akaku, 1988). Scales of the Oku-Aizu geothermal area in Japan consist of tetrahedrite ([Cu,Fe]12Sb4S3) with fine-grained galena and sphalerite in Well-87N-15T (Nitta et al., 1991) and pyrrhotite, pyrite, alabandite (MnS), sphalerite, wurtzite, galena, and cubanite in the Well-84N-2t (Imai et al., 1988).

Scales from these geothermal areas commonly include galena, sphalerite and copper sulfides. Figure 5 shows a Pb-Cu-Zn diagram of the scales of these area. The scales of the Kakkonda area have the widest variation in base metal ratio compared to the other areas. The scale of Well-19 is Pb-Zn rich and that of Well-13 is Cu rich. Scale of the Broadland and Salton Sea geothermal areas is close to that of Well-13 on the diagram, whereas the Cu minerals are different from Kakkonda. The Scales of Oku-Aizu and Yamagawa geothermal areas are intermediate between that of Well-13 and Well-19.

The salinity of the deep reservoir fluids in the Kakkonda area is about 1,000 mg/kg (Yanagiya et al., 1996). That is close to Broadland (Brown, 1986) and lower than the Yamagawa, Oku-Aizuthat and Salton Sea which, exceeds 10,000 mg/kg (Skinner et al., (1967), Akaku (1988) and Imai et al.(1988)). This low salinity arises from the meteoric origin of the Kakkonda geothermal fluid and lack of composite minerals in the rocks.

Among the three wells we investigated in the Kakkonda area, the contents of SiO_2, Cl and Na show similar values, while the ratio of B/Cl and As in fluid of Well-13 are much higher than the other well fluids. This suggests that a vapor rich region may exist around Well-13 (Yanagiya et al., 1996). This is supported by the occurrence of pneumatolytic minerals such as tourmaline, molybdenite and axenite around Well-13 (Kato and Sato, 1995).

On the other hand, host rocks around the feed points are Quaternary granite. Quartz diorite exists around Well-13 and tonalite exists around Well-19 (Kanisawa et al.,1996) as shown in Fig.1. Figure 6 shows that the relative concentration of Pb, Cu and Zn of scales and host rocks including granite rocks are Pre-Tertiary formations. The relative concentration of Pb, Cu and Zn of scales do not correspond with that of host rocks.

Figure 6 shows that the relative concentration of Pb, Cu and Zn in brine with over 10% salinity collected at a depth of 3600 m in granite of WD-1a by NEDO (Kasai et al.,1996), the ratio of those in fluid inclusion observed with the Kakkonda granite (Sasaki et al.,1998), and the ratio in fluid from WD-1b by NEDO around Well-19 (NEDO, in press).

From Figure 6, the relative concentration of Pb, Cu and Zn in Pb-Zn rich scale is similar to that of the liquid rich inclusion, brine of WD-1a with high salinity in kakkonda granite and fluid of WD-1b. This observation suggests that the meteoritic water diluted the brine in the Kakkonda granite and deposits the Pb-Zn rich scale.

On the contrary, the origin of the Cu in Cu rich scale is not clear as the base metal contents of the geothermal fluid from Well-13 has not been analyzed. Sasaki et al.(1998) mentioned that the vapor rich inclusion is slightly rich in Cu relative to polyphase inclusion in the granite in WD-1a. Considering that the geothermal fluid from Well-13 is originated from vapor rich environment.

Cu rich brine might exist around Well-13 and it may be the source of Cu. As mentioned before, Well-13 is located at the central part of the Kakkonda granite while Well-19 is located at the marginal part. The brine around Well-19 is Pb-Zn rich. If the brine around Well-13 is Cu rich, the zonation is similar to that found in Porphyry Copper deposit.

6. CONCLUSIONS

We characterized the scales from the Kakkonda deep reservoir and discussed the origin of the scale.

(1) The principal component of the scale of deep wells Well-13, 19, and 22 was amorphous silica and sulfide minerals with a metal element and 10-40%.

(2) There are two extreme types of sulfide deposition based on sulfide mineralogy: 1) Pb-Zn rich scales including galena (PbS), sphalerite (ZnS), and pyrite (FeS_2) are found in Well-19, at the margin of the Kakkonda granite. 2) Cu rich scales including chalcocite (Cu_2S), bornite (Cu_5FeS_4), chalcopyrite ($CuFeS_2$), galena (PbS), loellingite ($FeAs_2$), and native antimony (Sb) are found in Well-13, near the top of the Kakkonda granite. Well-19 is rich in Zn and Mn and Well-13 is rich in Cu; Well-22 has an intermediate composition.

(3) The relative concentration of Cu, Pb and Zn in the scales at the Kakkonda area have a wide variation compared to the other active geothermal areas.

(4) The relative concentration of Pb, Cu and Zn in the scale of Well-19 is similar to liquid rich inclusions and brine of WD-1a in Kakkonda granite.

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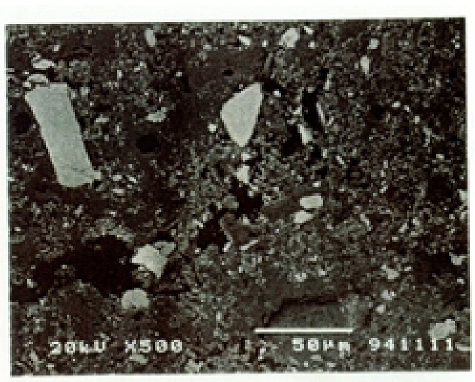

Figure 2: SEM image of the scale at well-13: Bright particles are bornite (Cu2S) and dark part are amorphous silica.

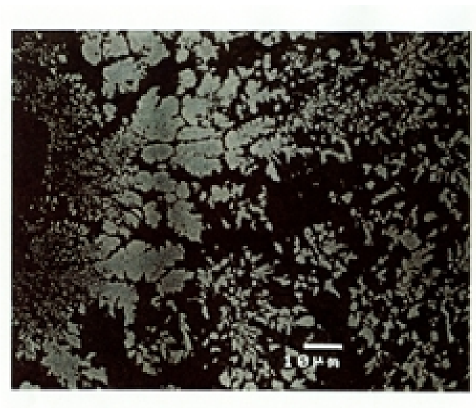

Figure 3: SEM image of the scale at well-19: Bright part is dendritic growth pattern of galena and sphrelite

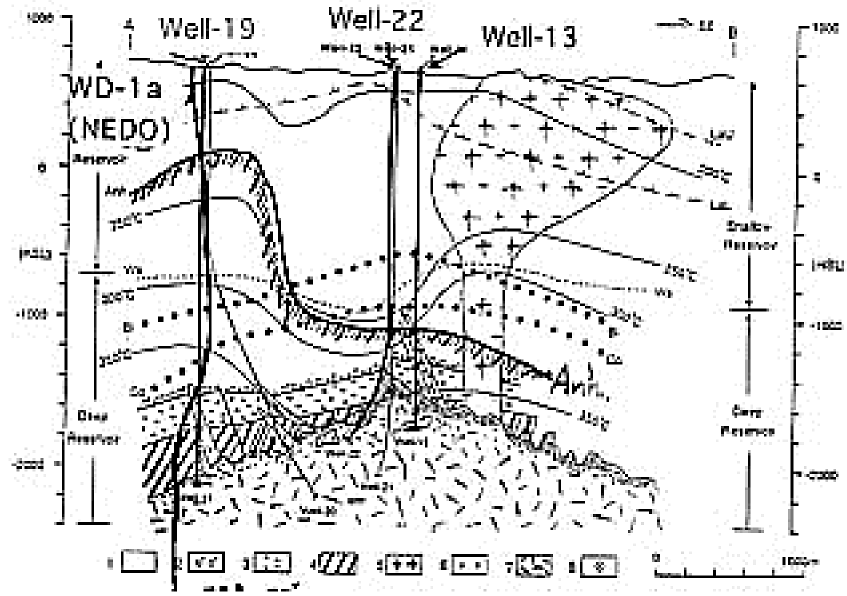

Figure. 1. Schematic geothermal cross-section of the Kakkonda geothermal system along the Kakkonda river in a NE-SE direction (Kato and Doi , 1993).
l : Tertiary formations; 2: basal conglomerate in Tertiary formation; 3,4 : pre-Tertiary formations; 5: 4.9 Ma dacite dike (Torigoeno-taki dacite); 6: old tonalite dike; 7: neo-granitic pluton: 8:sampling point: LaU: upper limit of laurnontite; LaL Lower limit of laumontite; Anh: upper limit of anhydnte; Wa: Iower limit of wairakite; Bi: biotite isograd; Co: cordierite isogra

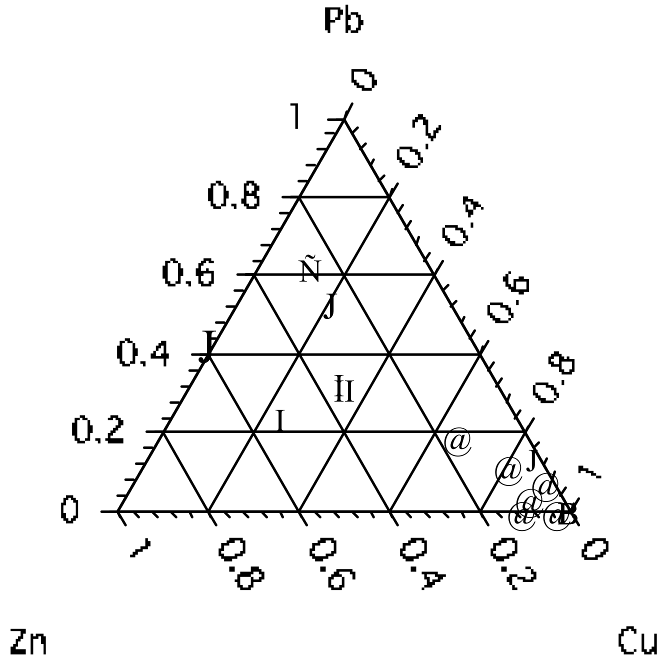

Figure 5: Pb-Zn-Cu ratio (ppm) of scale samples at Kakkonda, Yamagawa, Oku-Aizu , Broadland and Salton Sea. Closed circle: scale samples at Well-19,22and 13 from left to right, Closs (X) : scale at Oku-Aizu, open square : scales at Yamakawa, sharp (#) : scales at Broadland, close squre: scale at Salton Sea

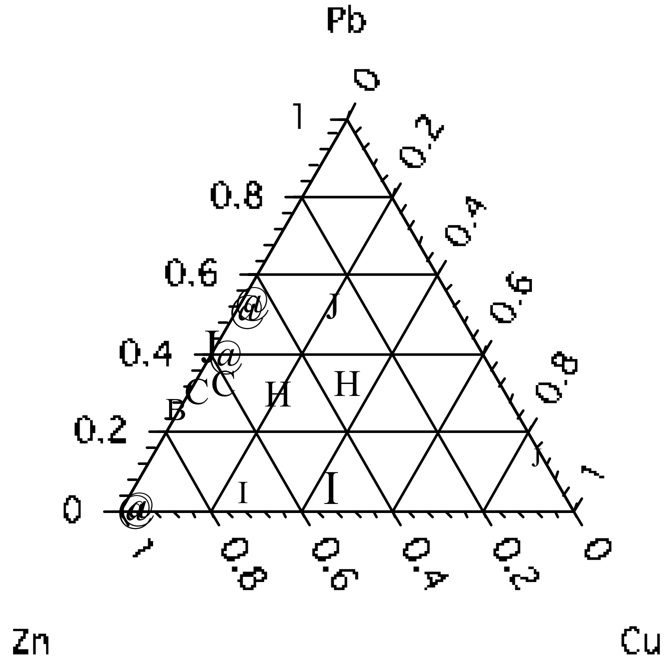

Figure 6: Pb-Zn-Cu ratio (ppm) of scale samples, rock sample and inclusion fluids. Closed circle: scale samples at Well-19,22and 13 from left to right, closed aquare: brine collected at the bottom of the well WD-1a (Kasai,1996), closed triangle and open triangle: inclusion fluids inpolyphase and vapor-rich inclusions from quartz vein in a granitinc rock, respectivity (Sasaki et al.,1998), closs (x): Granite at Well-13 and 19 from left to right,, sharp (#): Fluids from WD-1b (NEDO,inpress)

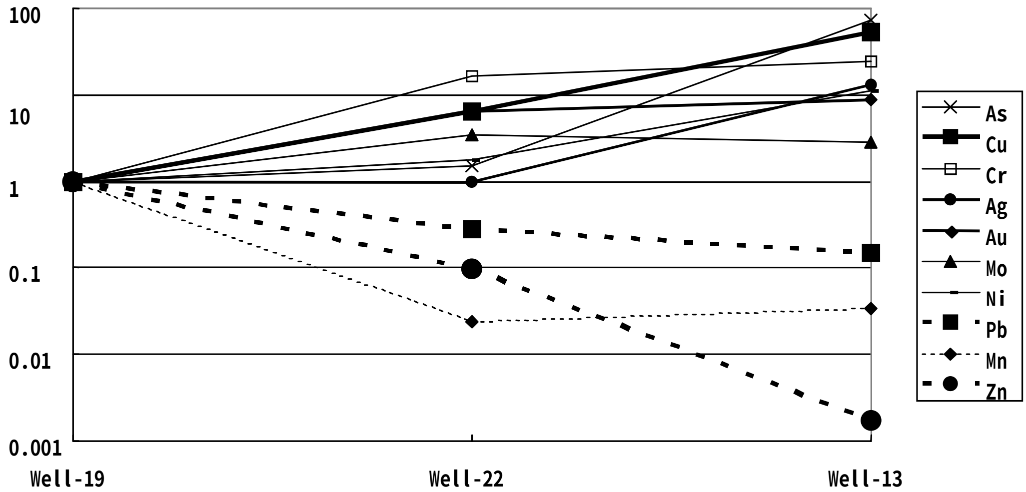

Figure 4: Chemical composition of the major metal elements standardized with the contents of Well-19

Table 1:The results of X-ray diffraction and SEM observation of the the scale at well-19,22 and 13.

Well-name	Appearance	X-ray diffraction	SEM observation
Well-19	Layer of black and lustcrons: 3mm thickness	Amorphous silica, galena, sphalerite	Alternative layer was shown with amorphous silica and sulfide minerals. Sulfides consist of galena and sphalerite, with **authigenic** growt
Well-22	Grey powder	Amorphous silica, barite, galena	Barite, quartz , galena and copper minerals were observed as particles of several tens micron meter.
Well-13	Black block and 1.5mm thickness.	Amorphous silica, galena, chalcocite	Sulfide rich layer was precipitated at the initial stage. This layer consists of **authigenic** loellingite, native antimony, bornite and galena. Most part of the sample consists of amorphous silica with **detrital** chalcocite and loellingite.

Table 2: Chemical composition of the the scale at well-19,22 and 13.

1. Major elements (%)

	SiO2	Al2O3	Fe2O3	MnO	MgO	CaO	Na2O	K2O	TiO2	P2O5
Well-19	25	2.07	17.47	2.92	0.01	0.06	0.2	0.31	0	0.01
Well-22	60	10.37	3.71	0.07	0.25	0.41	0.79	1.05	0.1	0.05
Well-13	47	2.47	18.21	0.1	0.03	0.05	0.33	0.48	0.03	0

2. Metal elements (ppm, but Fe,S and C in %)

	Au	Co	Cr	Mo	Sb	As	Cu	Zn	Fe(%)
Well-19	2.18	29	9	170	2535	620	2712	199219	12.22
Well-22	13.90	8	150	585	1866	940	17458	19525	2.59
Well-13	19.40	81	216	490	5030	45000	142747	351	12.74

	Ag	Ni	Cd	Ba	Sr	Pb	Mn	S(%)	C(%)
Well-19	42.0	30	230	984	14	147343	22600	12.86	0.30
Well-22	41.5	53	269	5461	51	41359	540	2.10	<0.2
well-13	550.0	332	221	46	4	22337	780	2.70	5.80