

Chemical hierarchy of hydrothermal signals at surface and hierarchy of eventual geophysical investigations in geothermal exploration

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ABSTRACT

In this paper a strategy is presented for geothermal prospecting in developing countries that brings to locate the first exploratory geothermal well by using only geochemical parameters in fluid phase in natural thermal emergences. The paper suggests a hierarchy of natural thermal manifestations, briefly describing how to interpret the most relevant chemical parameters.

Once the selection of the most promising area on a country, or a regional scale has been done, local investigation of the aquifer(s), especially in flat areas, in terms of: temperature and *pH*, and eventual tele-thermal parameters that can be analyzed in the field (e.g. NH_4 , SiO_2 , Hg) should bring to the delimitations of closed anomalies. If more parameters will converge to the same area(s), it is very likely that that place is the top of a buried thermal fluid convective high and likely the best place to drill the first exploratory well.

1. INTRODUCTION

There are several reasons why geothermal energy (n.b.: conventional, i.e. hydrothermal), on a worldwide scale, is not still well developed; some are economic (and/or political) others are technical.

From a technical point of view the main reason has to be sought in the too many disciplines involved during the exploration phases and the difficulty to prioritize the single aspects. From the political point of view, the main problem is in the communication, during the decisional phases, between technical experts and decision makers (or general managers), with their different background, when investments have to be decided.

The two opposite situation, the best and the worst, are in Switzerland and Indonesia, respectively. In Switzerland there is a lot of propaganda but bad results (expensive deep drilling and induced seismicity); in Indonesia there is not much communication to the scientific community, but more than 1,200 installed MW, with the highest installing rate among geothermal countries in the last decade.

Although it is impossible to demonstrated as true everywhere, there is a common sensation in the geothermal community that the best places on the Earth for the development of geothermal energy must have: *i)* active volcanoes and *ii)* a good meteoric recharge. The meteoric recharge of geothermal systems from nearby local meteoric water was as early demonstrated in the 60's through the isotopic composition of the steam produced in active geothermal wells (**Craig, 1963**). This combination of presence of a lot of volcanoes in a region and a huge meteoric recharge is, as a matter of fact, the intimate success of the recent geothermal expansion in Indonesia in the past decade, and the Philippines in the past two decades.

Under the prospected light, the African rift, especially in those countries located along the equator, i.e.: Kenya, Uganda, Democratic Republic of Congo and Ruanda, with their many active volcanoes and very relevant rainfalls, have the same potentiality of expansion as Indonesia, and they should definitely increase their geothermal energy faster than at present.

In the present paper the strategy to be adopted in these developing countries, to accelerate the exploration phase is presented. It is largely based on the use of geochemical prospecting tools applied to natural spring and gas emergences and relatively inexpensive field campaigns to delimited thermal anomalies. It is also largely based on concepts developed in the 60s by the Italian geothermist Franco Tonani, that unfortunately passed over last December to which the paper is dedicated.

2. GEOTHERMAL BACKGROUND

One of the most important conceptual model introduced in Geothermics, is the “*self sealing geothermal field*” (**Facca and Tonani, 1967**). This type of field overcomes the previous one(s) where the reservoir was supposed to have the necessity to be separated, from the surface, by an impermeable geological cap rock, in terms of lithology, such as clay-rich formations or volcanic lahars, or others. As a consequence, the concept of real reservoir in opposition with the potential reservoir (typically the Mesozoic limestone reservoir of the Italian fields) was derived, that not necessarily has to coincide with a specific geological horizon. As a matter of fact, at the Geysers in California, the reservoir and the cap rock are in the same “*Franciscan Sequence*”, a fractured tectonic melange of graywackes and igneous mafic rocks (**Bailey et al., 1964**), sealed in the top parts by silica precipitation.

From an opposite point of view, although the concept of geothermal reservoir was introduced in Italy, at Larderello, paradoxically, the steam escapes to the atmosphere directly from the southern reservoir outcropping areas (**Minissale, 1991**).

One important factor that strongly affects the possibility of discovering new geothermal systems from the surface, is the apparent decrease of the shallow thermal heat-flows by the presence of powerful, very dynamic shallow aquifers(s), that might be present above the *self-sealing* zone. Therefore, a perfect and well sealed (at the top) geothermal system, possibly located in a flat sedimentary area, where rainfalls are abundant, has no possibility to be discovered; and it is possible that on the Earth there are several of this type..., still undiscovered. Fortunately, geothermal systems are prevalently located in active areas, mostly around volcanoes, where several active faults and fractures are present, that allows, in convective areas, the rising and emergence of more or less contaminated thermal fluids, such as: fumaroles, thermal springs and CO₂-rich gas emissions.

In the following paragraphs a hierarchy of natural fluid emissions will be presented in relation with their chemical and isotopic composition. The chemical aspects are strongly related with the outstanding work done by Werner Giggenbach in the 70s and 80s (**Giggenbach 1980; 1987**) that clearly demonstrated that, in geothermal systems, the composition of geothermal gases (the most useful tool to forecast deep equilibration temperatures using geothermometers) is strongly buffered by the FeO/FO_{1.5} ratio at a fixed H₂/H₂O ratio of 2.8, and that this buffer works worldwide. The main consequence of this extremely important factor is that: the interpretation of fluid composition(s) cannot be ambiguous...if they reach the surface....uncontaminated.

3. HIERARCHY OF NATURAL FLUID EMISSION TO THE SURFACE

For the present purposes of the paper, the following list of fluid emissions, with decreasing temperature, can be proposed.

- 1) Supercritical low-pressure fumaroles (T>371 °C, up to 1150 °C).
- 2) Superheated fumaroles (low pressure, T<371 °C).
- 3) Steam-saturated fumaroles (often about 160 °C): “*sofatarata*” type.
- 4) Boiling fumaroles at atmospheric pressure (85-99 °C according to elevation).
- 5) Boiling water/mud pool/mud/pot (90-100 °C), sometimes very acidic.
- 6) Steaming ground (diffuse emission) 30 °C<T< 99°C.
- 7) Springs at near boiling conditions (90-95 °C), with high flow rate, N₂-and-He rich.
- 8) Thermal springs (20-90 °C) with associated CO₂.
- 9) Cold gas (CO₂, H₂S) bubbling pool (stagnant water, very acidic if with H₂S).
- 10) Dry CO₂ emissions.

They are ordered in terms of potential significance in geothermal prospecting.

Fumaroles (n. 1 and 2) clearly suggest the presence of magmatic gases. They have SO₂, HCl and HF that clearly points to the absence of steam condensation in the feeding conduit. Sometimes these very acidic fumaroles, with free SO₂, may persist event at very low temperature (water boiling conditions at atmospheric pressure, or even lower temperature), simulating the steaming ground type emission (n. 5), suggesting evaporation of local water, unable anyhow to remove the acidic components from the gas phase.

Saturated fumaroles (n. 3) of the “*sofatarata*” type, at about 160 °C, is the best indication of the presence of shallow steam-dominated geothermal systems underground, at the maximum enthalpy point for water (about 230/240 °C and 30/35 atm.). The expansion of such deep steam to atmospheric conditions, in fact, produces a stable 160 °C fumarolic activity. Such fumaroles are present in the Phlaegrean Fields in the Neapolitan area (**Cioni et al., 1984**) where the “*sofatarata activity*” was defined, and e.g.: at Sufrière in the St. Lucia Island in the West Indies (**Lindsay 2001 in Joseph et al. 2013**). Their presence can also be envisaged in the Italian geothermal field of Larderello, where some peripheral geothermal wells in the south-east sector of the field, likely drain a similar steam (in terms of temperature) even if at a slightly higher pressure than atmospheric (**Minissale, 1991**).

Fumaroles at boiling temperature for the elevation (n. 4) and/or boiling water/mud pools...etc. (n. 5) type emissions are quite diffuse natural manifestations in both volcanic and geothermal areas all over the World. Their chemical composition is extremely variable: from extremely acidic and extremely saline in most of active volcanic environments and craters, because of the condensation of acidic magmatic components (prevalently H₂S and SO₂), to alkaline and very low in salinity in the NH₃-rich steam condensates of Larderello in Italy (**Minissale et al., 1992**).

The last “steam” activity at the surface, in both geothermal and volcanic areas, sometimes wrongly referred as fumarolic activity, is what is called, by volcanologists: steaming ground or diffuse steam emission (n. 6). There is a large variety of steaming ground(s) covering: from steam at near boiling temperature at atmospheric pressure to a recent reported value of the soil around the Baekdu volcano in Nord Corea of 6 °C in winter, when the outside temperature was -25 °C (**Kim Hyok pers. commun.**), clearly suggesting anyhow local hot fluid emission. In the middle between these two extremes, there is steaming in temperate or equatorial regions, especially after rainfalls, and hot air emission without any steam emission in arid regions. If manifestation from 1-to-5 can be considered very good indicators of high temperature underground, the steaming ground, especially far from active volcanic areas, may be very misleading.

From n. 7 to n. 10 we are in the more common occurrence of emerging hot fluids from the Earth, i.e. thermal springs and gas emissions (CO_2 or N_2 -rich, and/or mixed between the two). Quite often these manifestations are the only emissions around active volcanoes. There are many types of thermal springs but, their hierarchy in terms of geothermal prospecting significance, can be placed between these two end-members:

- 1) low salinity, low flow-rate, low pH , HCO_3 -type, low He content, high $^3\text{He}/^4\text{He}$ ratio, CO_2 -rich associated gas,
- 2) high salinity, high flow rate, high pH , Cl-type, high He content, low $^3\text{He}/^4\text{He}$ ratio, N_2 -rich associated gas,

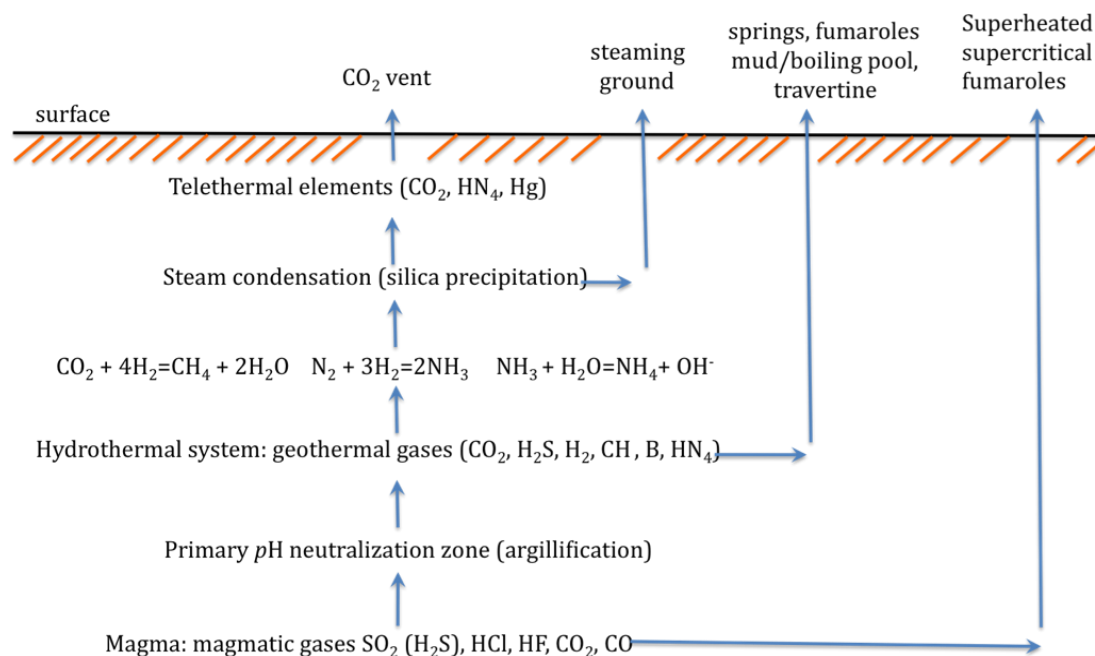
both generally having, in terms of $\delta^{18}\text{O}$ and δD , an isotopic meteoric signature.

The first end member (1) is the typical hot spring of geothermal areas (such as Larderello in Italy; **Duchi et al. 1986**), where the heat-flow is so high that even shallow circulating waters in the cap-rock formations can get hot, although maintaining the HCO_3 composition of groundwater (either Ca or Na according if the circuit is in carbonate or silicic formations). The second member (2), well defined in the India subcontinent (**Minissale et al., 2002**), is the typical long deeply convective circulating water, with high salinity, Na-Cl composition, that gets hot through normal gradients after deep circulation.

Apart from the main chemistry in liquid phase, it is actually the gas phase (if this is CO_2) that characterize type 1 springs, pointing to the presence of active hydrothermal systems producing CO_2 . In fact, the strong production of CO_2 , either through silicate or carbonate dissolution and/or water/rock interaction, and/or through the rising from the mantle, or even gas-gas reaction, is able to dilute the ^4He formed in the crust through radioactive decay, and this is the reason for the very low concentration of total He. Such dilution may not affect much the $^3\text{He}/^4\text{He}$ ratio, if the thermal anomaly is triggered by the presence of a mantle magma stocked in the crust.

In the second end member nitrogen, often bubbling from the water in a gently, intermittent way, clearly reflects the atmospheric source of the recharge water, whereas the high content of total helium (up to 6.7 % of the total gas; **Minissale et al. 2002**) suggests long residence time in the shallow crust.

Although is not the intention of this paper to go deeply into this chemical issues, the following block-diagram briefly summarizes what happens underground in volcanic systems, and how fluids, in terms of phases are stratified and rise, eventually, to the surface.



4. STRATEGY TO BE ADOPTED AFTER THE RECONNAISSANCE OF FLUID HIERARCHY

Having always in mind the self-sealing model of a geothermal field, and therefore the possibility that an active hydrothermal system may not leak hot fluids to the surface, what to do when you have mapped in a region all the thermal emergences there present? As usual: sampling and analysis of all of them and, for comparison, eventually analysis of some cold emissions as well. Again, once on a country scale you have been able to select the most promising area, let's say: because there are some $^3\text{He}/^4\text{He}$ anomalies in the gas phase, or some anomalous hydrogen contents (from a Fisher-Throps reaction typical in hydrothermal system), or the presence of some type 1 thermal springs (the best)...etc., how to go further?

Italian scientist Franco Tonani at the 1st World Workshop on Geothermal Energy that was held in Pisa in 1970 (Tonani, 1970) showed that: any hydrothermal system, even in the presence of multiple condensation horizons underground and therefore able to precipitate most hydrothermal components in them, is not able to completely block both NH₃ and CO₂, or mercury....or radon, that continually diffuse to the atmosphere. If these elements spread into a shallow aquifer, together with steam and/or water from the main condensation zone (or even a secondary condensation zone, if the dynamic of the shallow aquifer(s) is not too fast (e.g. in flat morphological areas), the buried hot reservoir top(s) must be revealed with some closed thermal and chemical anomalies inside the shallow(er) aquifer(s). This strategy was adopted by Tonani to discover some geothermal fields in central America and California in the 70s, and more recently by the author in Yemen in the rhyolitic Quaternary volcanic region of Dhamar (Minissale et al., 2011) to delimitate thermal anomalies around the Al-Lisi and Isbil volcanoes (Fig. 1)

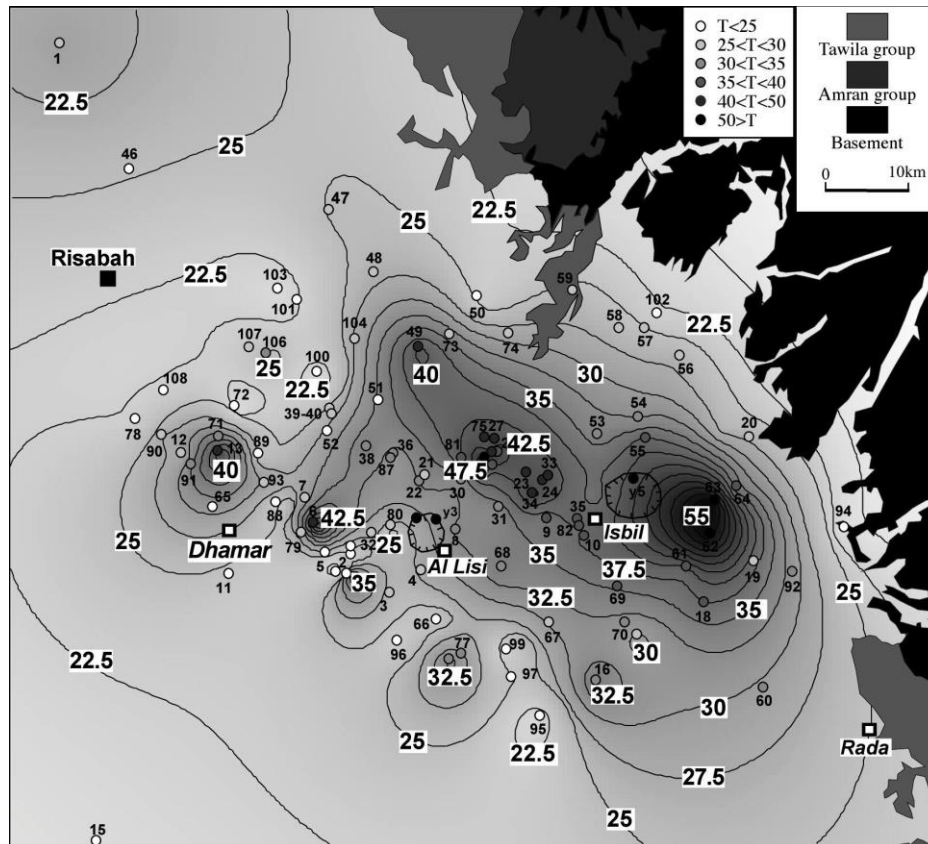


Fig. 1 Delimitation of closed thermal anomaly in the shallow aquifer hosted in the Dhamar intramontane basin characterized by the presence of the Al Lisi and Isbil rhyolitic volcanoes (after Minissale et al., 2011)

In developing countries along the African rift this method can be used to check the temperature distribution in the shallow aquifer, e.g. all around a volcano and, if the hotter parts of the aquifer surrounding the volcano also shows chemical anomalies in the tele-thermal elements (NH₃, CO₂), there is high probability that that is the place where the thermal flux is the highest. Of course this is not always true everywhere but, coupling with usual and inexpensive hydrogeological investigations, it is possible to understand and quantify if there is cooling from shallow running water.

After this preliminary phase of delimitation of thermal anomalies in the shallow aquifer, coupled possibly with some chemical parameters, such as CO₂, from a theoretical point of view, it would be good to confirm the anomalies discovered with some geophysical investigations. Typically, the most used geophysical prospecting methods used in geothermal exploration are geoelectrical profiles and/or Magneto-Telluric surveys. Both methods are strongly influenced by external factors (noise) and, in general, they give contrasting results. But, if the discovered anomalies (if any) in depth coincide with the thermal anomaly in the aquifer at near surface, than, there is high probability that the following phase of drilling an exploratory well will be positive.

A more expensive, but very efficient method to measure real geothermal gradients is through the drilling of 200/300 meters deep wells in impermeable formations. This method was as early applied in Italy in the 60s and led to the discovery of the Mt. Amiata geothermal system (Mouton, 1969). Together with the measuring of the temperature in shallow aquifers (such as shown in Fig. 1) this is the only other direct method to envisage the pattern of the temperature distribution in depth.

The geochemical and geophysical investigation summarized so far, at their best, can only suggest: 1) possible deep equilibration temperature (e.g. through geothermometry in the gas phase), 2) thermal gradients. The existence of a real reservoir,

with a hot fluid inside where to located the calculated temperature(s) remains a guess, and this is the real limit to the expansion of geothermal energy worldwide in both developed and developing countries.

5. DISCUSSION AND CONCLUSIONS

Because the existence of a geothermal system must be verified through the drilling of a well, which is an expensive activity, at least in developed countries, generally, a final seismic geophysical prospecting is attempted to give light to the main geological structures underground. In fact, structural highs (or tops) of any recognized or supposed fractured horizon (either formation or sequence) is the best target for deep, hot, convective fluids, and therefore the best target in geothermal exploration.

By summing up the costs of all geophysical prospecting activities, that are justified for the expensive drilling activities in developed countries, may not be necessary for drillings in developing countries, since that may even equal the cost of a 1,000 m well.

With the proposed chemical methodology to delimitate thermal anomalies in areas where chemical signals suggest the presence of a buried hydrothermal system, with the hierarchy proposed in section 3, and the briefly summarized important chemical parameters (e.g. high $^3\text{He}/^4\text{He}$ ratios, $\delta^{18}\text{O}$ shifts of thermal waters....etc.) any geophysical prospection could be avoided.

Because many countries along the African Rift already have chemical data in most of their hydrothermal areas, but not many wells have been drilled yet, what proposed in the present paper could be tried. This seems particularly easy in those countries that certainly have good geothermal resources and abundant meteoric recharge, like the Democratic Republic of Congo, Tanzania, Uganda and Ruanda, where an integration of relevant chemical data (good gas analyses, $^3\text{He}/^4\text{He}$ ratios...etc.) on the thermal manifestations could help the selection of the best place to try the method.

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