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GEOCHEMICAL INTERPRETATION OF DATA ON THERMAL FLUIDS FROM THE LAUGARVATN LOW-TEMPERATURE AREA, SW-ICELAND

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ABSTRACT

The Laugarvatn field is one of many low-temperature geothermal fields in the Southern Lowlands of Iceland. The study of the chemistry of the water has been used to suggest movement, processes affecting geothermal water during its up-flow and also possible recharge areas using isotope data (δD and $\delta^{18}O$) and Cl/B ratios. Silica and cation geothermometers are applied to predict possible subsurface temperatures using analytical data, and equilibrium calculations with the chemical speciation programme WATCH. The reliability of the geothermometers is checked by studying the equilibrium state of the thermal waters by means of saturation index log Q/K diagrams, Na-K-Mg triangular diagram and by applying both silica-enthalpy and enthalpy-chloride mixing models. In the end, the possible temperatures are predicted after taking into consideration the equilibrium state of the thermal waters.

The geochemistry of the thermal water and cold groundwater in the Laugarvatn area shows that the use of geothermometry is dependent on many physico-chemical parameters, suggesting subsurface temperatures in the range of 120-180°C, and probably different sources of water in the eastern and western parts of the area. In fact, physical processes like conductive cooling, mixing with cold water and boiling during up-flow may affect water-rock equilibria and this can result in unreliable temperature predictions. So, it is necessary to apply different methods and geothermometers for the studies of the equilibrium state, to understand which processes occur in the area.

1. INTRODUCTION

The Laugarvatn region is in the upper part of Arnessysla in the Southern Lowlands of Iceland, about 60 km east of Reykjavik. It lies in Quaternary volcanic formations, in the vicinity of a group of basaltic volcanoes. The area is located just east of the southwestern active volcanic zone and is within the Southern Lowlands hydrothermal area which is one of the two largest low-temperature areas in Iceland, covering some 700 km². Thermal springs with hot water ranging from 40°C to boiling and with a large flow rate, are abundant in the area. The subsurface temperature indicated by geothermometry is around 140°C at Laugarvatn. Present use of geothermal water in the area is mainly for district heating.

Geochemistry has been used quite extensively since the seventies as a tool for the exploration of geothermal resources and for the subsequent development and exploitation of these resources. The thermal fluids are characterized by their chemical composition, which is to a large extent caused by the interaction of the fluid with the host rock. The application of suitable chemical methods to the interpretation of thermal fluid composition will give a better understanding of the origin of the fluid and the processes that take place during its transport.

The general purpose of this report is a geochemical interpretation of chemical analysis of some thermal fluids where the location of the recharge areas and evaluation of the direction of subsurface fluid flow, prediction of subsurface temperature and evaluation of mixing and boiling processes will be carried out.

2. GEOLOGICAL SETTING AND GEOTHERMAL ACTIVITY IN THE AREA

2.1 Geological setting

Iceland is situated on a constructive plate boundary, the North-Atlantic Ridge, which crosses the country southwest to northeast (Figure 1). The boundary is visible as an active volcanic zone characterized by numerous fissure swarms and volcanic systems, most containing central volcanoes. The volcanic zone is flanked by Quaternary rocks, characterized by sequences of sub-aerial lava flows intercalated by hyaloclastites and morainic horizons corresponding to glacial conditions. Quaternary formations are flanked by Tertiary flood basalt (Saemundsson, 1978).

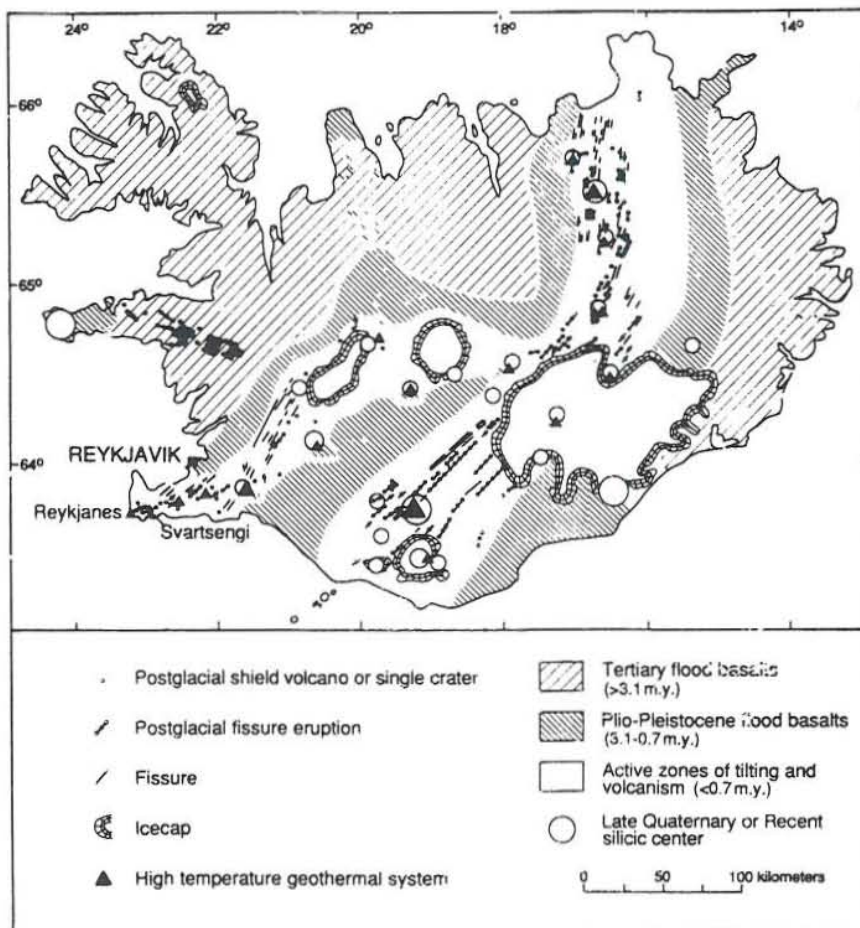


FIGURE 1: The main geological features of Iceland

The area of study is located on the eastern edge of the Reykjanes-Langjokull volcanic zone, i.e. the part termed the Thingvellir-Langjokull zone (TL-zone). As defined, the TL-zone stretches from lake Thingvallavatn in the south to the northern boundaries of the Langjokull glacier (Arnorsson and Olafsson, 1986).

The rocks are upper Quaternary, sub-glacial hyaloclastites and pillow lavas, and inter-glacial basalt lavas. Minor outcrops of acid volcanics occur in the vicinity of the Geysir geothermal field. The geological formations become progressively younger towards the volcanic belt to the west (Figure 2).

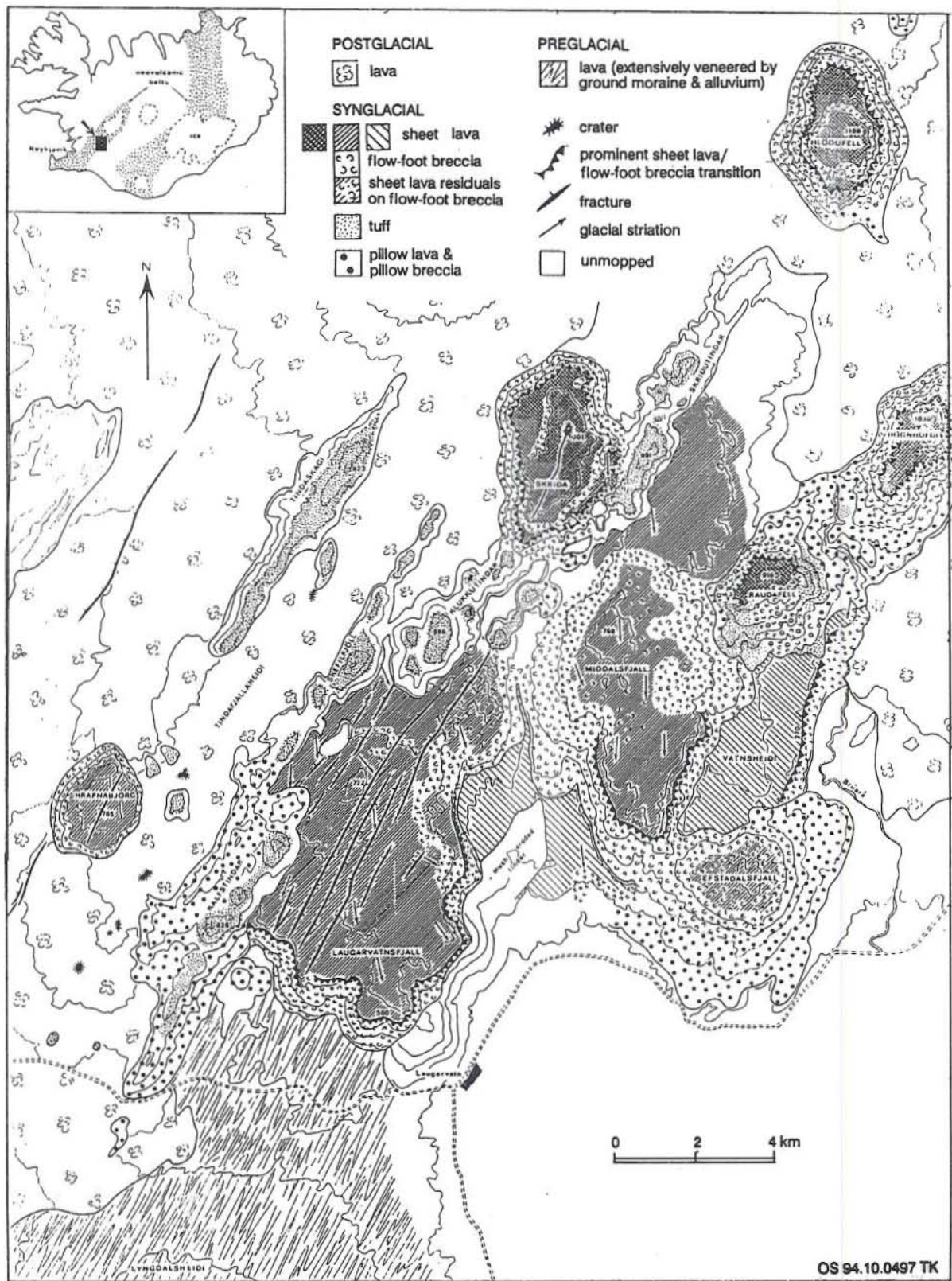


FIGURE 2: Geological map of the Laugarvatn area (Jones, 1969)

The basalt lavas dip a few degrees towards northwest under the active volcanic belt. Towards east, where the rocks are oldest, the hyaloclastite formations are, at least partly, reworked and tend to form sheets of variable thickness between the lavas, but in the western part of the area they form the mountains protruding above the plains. The area is extensively faulted, the NNE-SSW striking faults being most common running approximately parallel to the volcanic belt. Younger fractures striking ENE-WSW and N-S are relatively common in the area (Georgsson et al., 1988). In the southern part of the area recent N-S en echelon fractures occur on an E-W belt coinciding with a major transcurrent fault (Einarsson, 1989). Large earthquakes associated with movements on this fault occur about every one hundred years. At several geothermal sites there is exclusive evidence of movements of fractures in post-glacial times (Georgsson et al., 1988). There are also descriptions, recent and ancient, of changes in the flow and temperature of some hot springs in the area, that occurred after earthquakes.

Drillhole data show that permeability decreases in general with the age of the flood basalt pile in Iceland (Sæmundsson and Fridleifsson, 1980). By inference from the results of Walker (1960) in Eastern Iceland

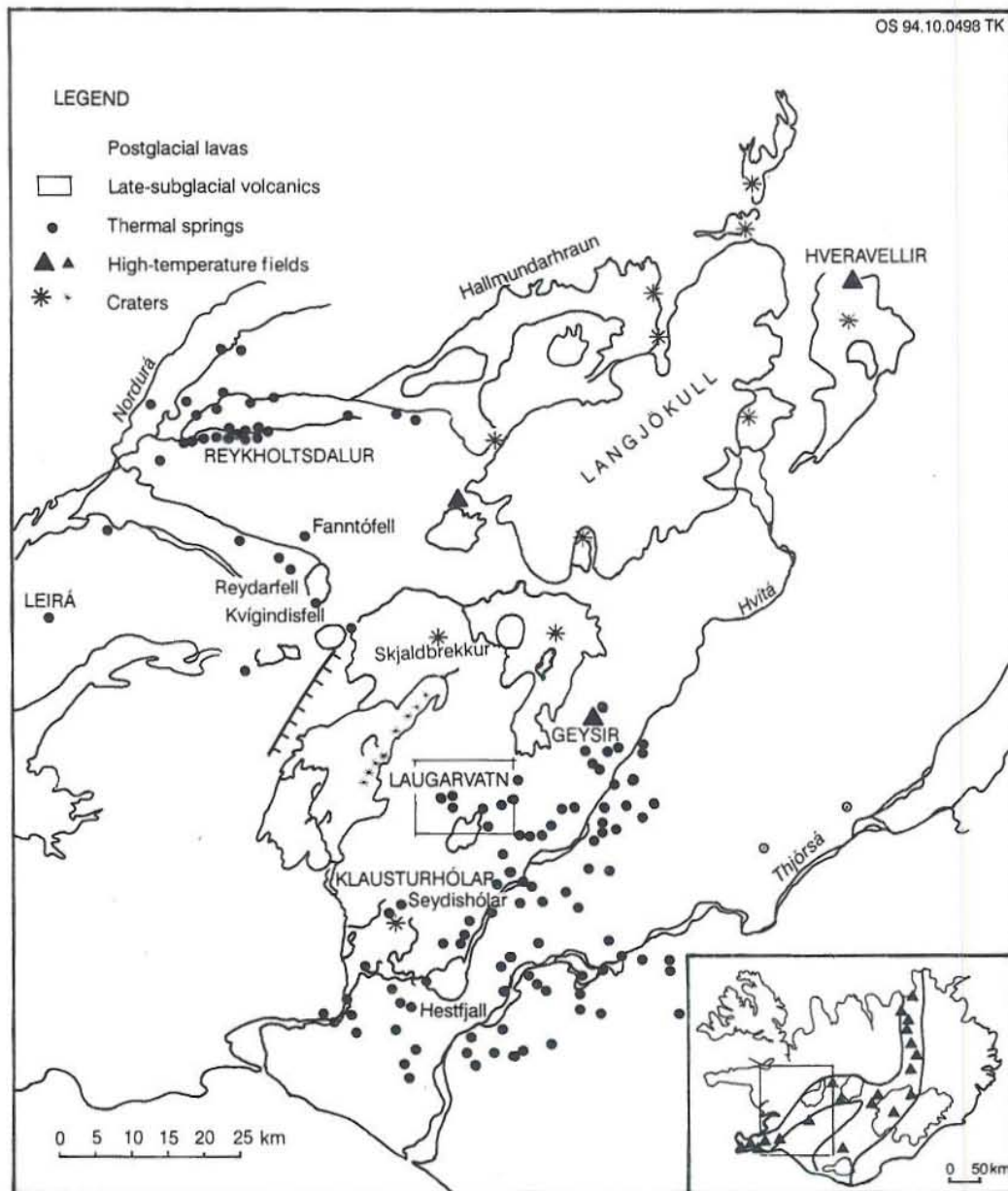


FIGURE 3: Geothermal map of the Southern Lowlands and the Borgarfjörður area (Arnorrsson and Olafsson, 1986)

it is expected that the decreasing permeability with age is due to compaction and increasing abundance of secondary minerals filling pores and cracks. The rocks in the area are Upper Quaternary and would, therefore, be expected to have some primary permeability. The regional thermal gradient is high in the area from about 140°C/km at the edge of the volcanic zone to about 60°C/km 50 km to the east (Palmason et al.; 1979).

2.2. Low-temperature geothermal activity in Iceland

Low-temperature hydrothermal activity is widely distributed in the Tertiary and early Quaternary basalt formations in Iceland (Figure 3). However, some Tertiary areas, particularly Eastern Iceland, are almost devoid of any low-temperature activity. Surface manifestations are isolated springs and spring clusters with temperature ranging from just above the mean annual temperature to boiling. The temperatures are generally below 150°C in the uppermost 1000 m. Most of the springs are located on relatively low ground in valleys and lowlands by the coast. Total flow from thermal springs is estimated to be 1800 l/s and that from springs and drillholes roughly 4600 l/s (Gudmundsson and Palmason, 1981). The flow rate from individual spring clusters and isolated springs is highly variable (upto 180 l/s). Distance between isolated springs may be several tens of kilometres. Generally there are no surface manifestations in the area in between. This scattered low-temperature activity is to be contrasted with the high-temperature hydrothermal activity in Iceland, which is concentrated and intense in relatively small areas of 1 to 100 km² within the volcanic zone.

The heat source for the geothermal systems in upper Arnessysla, is considered to be magmatic. The magma is assumed to have originated under the nearby Thingvellir-Langiokull volcanic zone. Diminishing dilatation in this zone, favours intrusion into the old crust flanking it as witnessed by volcanic edifices of Upper-Quaternary age resting unconformably upon considerably older volcanics. Chemical geothermometry indicates that subsurface temperature in the area may be as high as 200°C. The geothermal systems are considered to be convective. Convection is envisaged to be concentrated in tectonic fractures but downflow could occur additionally through pores of primary permeability. The high H₂S content of the Laugarvatn hot springs is taken to indicate that magma is replenishing the heat source at present (Arnorsson and Olafsson, 1986). Hot springs are relatively abundant in the area.

3. ANALYTICAL DATA

The present geochemical study is based on 20 samples from hot springs, cold springs and cold water from the area. Chemical composition of all the samples is shown in Table 1 and their locations in Figure 4. The composition of geothermal water depends on many factors. Temperature dependent reactions between host rock and water are one of the major ones. Leaching plays an important role when the amount of a particular constituent is too small to achieve equilibrium. However, processes of mixing, boiling and cooling usually have a pronounced influence on the final composition of the geothermal waters. The first four samples are from the same hot spring (Laugarvatn), taken at different times to look at time dependent changes. Silica and chloride concentrations versus time are plotted for comparison in Figure 5 and Figure 6, respectively. The water from this hot spring shows variation in chemical composition with time, in general silica (SiO₂), sulphate (SO₄) and H₂S decrease with time, whereas, the pH and the sodium (Na) increase. The concentration of dissolved solids in geothermal water in Iceland is highly variable. In these samples, the T.D.S. concentration is relatively low, it ranges from about 13 (cold water) to 415 ppm (Laugarvatn) (Table 1). Sulphate tends to be the dominant anion in nearly all the samples, followed by chloride. In dilute water chloride is the dominant anion. Sodium is the most abundant cation. pH ranges from about 7.8 (sample No. 7) to 9.62 (sample No. 12). The Al concentration was determined only in 3 samples (No. 4, 17 and 21) and the iron (Fe) concentration only in sample No.17. Samples 17, 20 and 6, 13 respectively are of similar chemical composition, except for the δD and δ¹⁸O isotope ratio in No. 6. H₂S is relatively high in samples No. 1, 3, and 9.

TABLE 1: Chemical composition of thermal and cold waters from the Laugarvatn area (conc. in ppm)

N°	CO2	H2S	B	SiO2	Na	K	Mg	Ca	F	Cl	SO4	SD	δO18	TDS	T.meas	pH
1	35,5	17,4	0,14	164	70,8	3,5	0,03	3,1	3,3	33,1	76,6			345	99	8,2
2	25,2	15,1	0,14	155,5	75,5	4,02	0,004	3,12	3,02	36,1	44,3			372	99	9,14
3	27,3	13		144,87	76,23	3,78	0,04	3,08	2,939	35,05	33,68			344,7	97,5	9,48
4	37,5	1,43	0,1	144,9	76,9	3,35	0,005	2,96	2,98	34,6	32,1	-66,8	-9,58	371	99	9,42
5	10,1	0		13,89	11,87	0,14	0,07	2,33	0,07	4,24	1,47	-71,67	-10,47	60,5	3,6	10,1
6	13,1	0		15,79	7,24	0,44	0,62	3,91	0,055	4,84	1,53	-65,37	-9,56	60,3	3,6	9,5
7				16,5	11,9	0,3	1,28	4,1	0,188	8,1				66,9		7,8
8	13	5,2	0,14	78	38	1,6	0,15	2,3	1,6	18,1	60,5			224	44	9,2
9	38,1	22,4	0,19	141	78,8	3,5	0,04	5,4	2,8	49	105,6	-71,2	-9,61	400	99	8,4
10	18	3,9	0,31	164	83,3	3,6	0,07	3,5	2,2	43,3	54,8	-71,4		402	86	8,9
11	52,6	0,78	0,06	72,8	52,5	1,2	0,82	6,78	1,059	24,2	20,3		-9,35	200,5	43	9,33
12	25,41	3,38	0,127	107,94	77,14	2,58	0,01	2,48	1,837	48,62	37,97		-9,8	341,5	93,5	9,62
13	12,64	0		13,53	7,04	0,37	0,53	2,68	0,063	3,74	2,69		-10,93	42,5	3,2	8,92
14	16,6	0	0,0035	16,01	10,08	0,35	0,53	5,06	0,79	5,2	3,37	-71,5	-9,3	54,9	4,3	9,34
15	16,1	0													4,3	9,32
17	35,9	2,12	0,15	200,4	86,8	5,06	0,0019	2,02	2,5	46,2	52,6			451	97	9,25
18	45,4	1,89		212	90	4,8	0,02	1,7	2,75	46,7	55,8	-71,9	-9,65	495	88	9,05
19	22,4	1	0,2	206	88	4,7	0,03	2,6	2,6	51,8	53,2	-74			99	8,6
20	37,3	0,82		153,16	79,66	3,19	0,1	3,08	2,153	46	50,68				84,6	9,08
21	61,8	0	0	32,09	15,3	8,01	4,08	13,15	0,166	25,93	4,64				11	

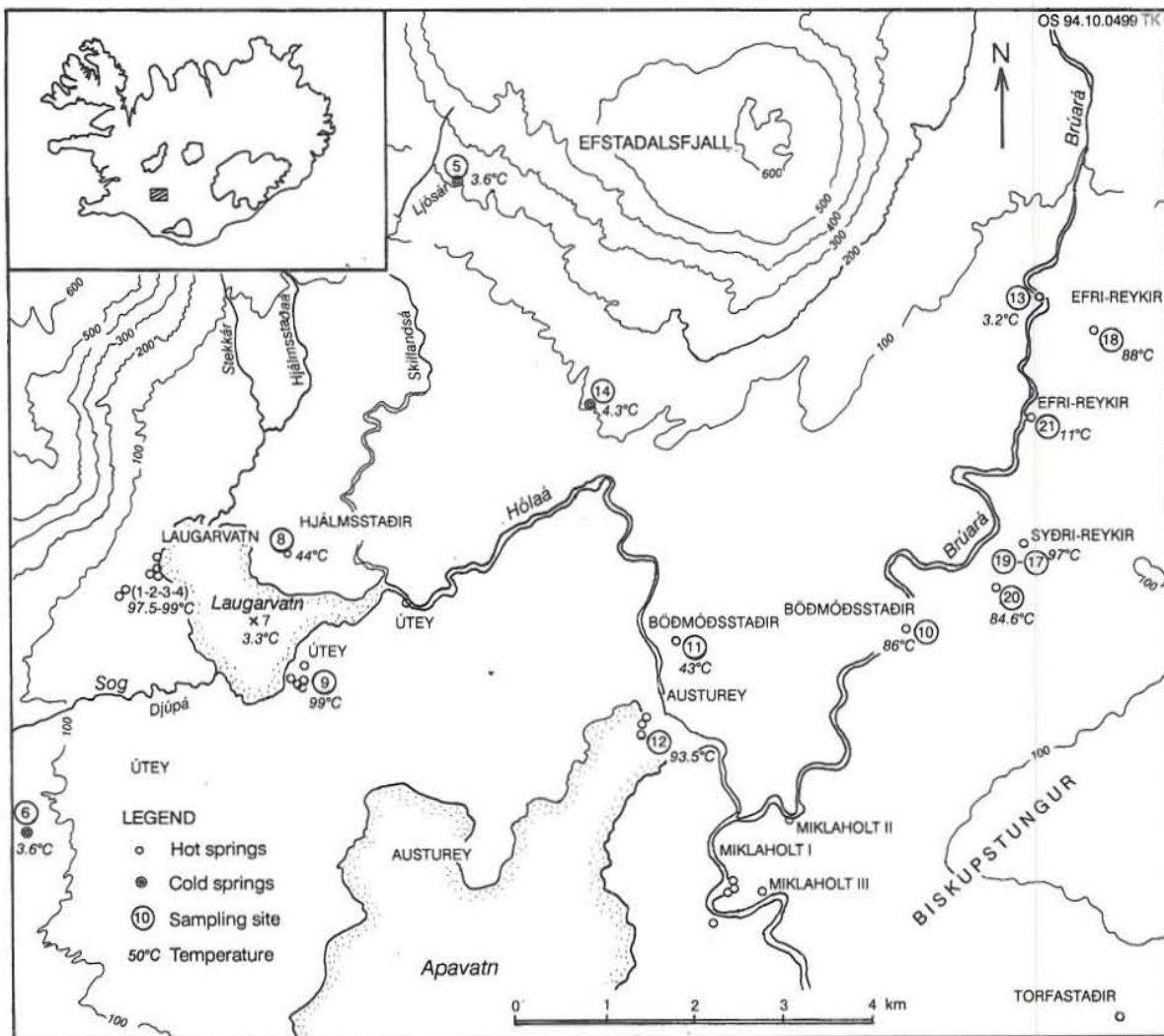


FIGURE 4: The Laugarvatn area and sampling sites

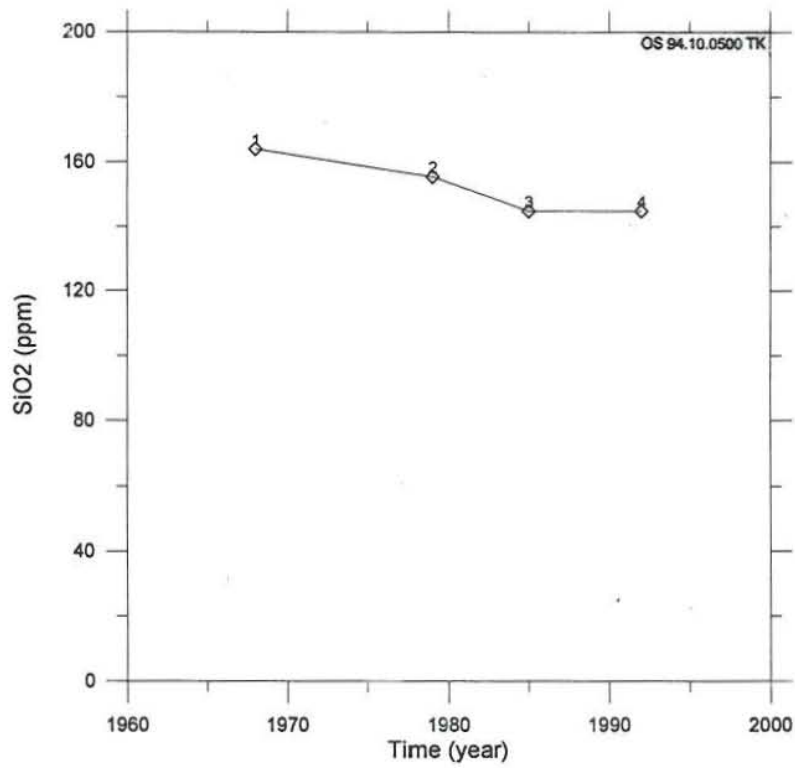


FIGURE 5: Changes in silica concentration at Laugarvatn with time

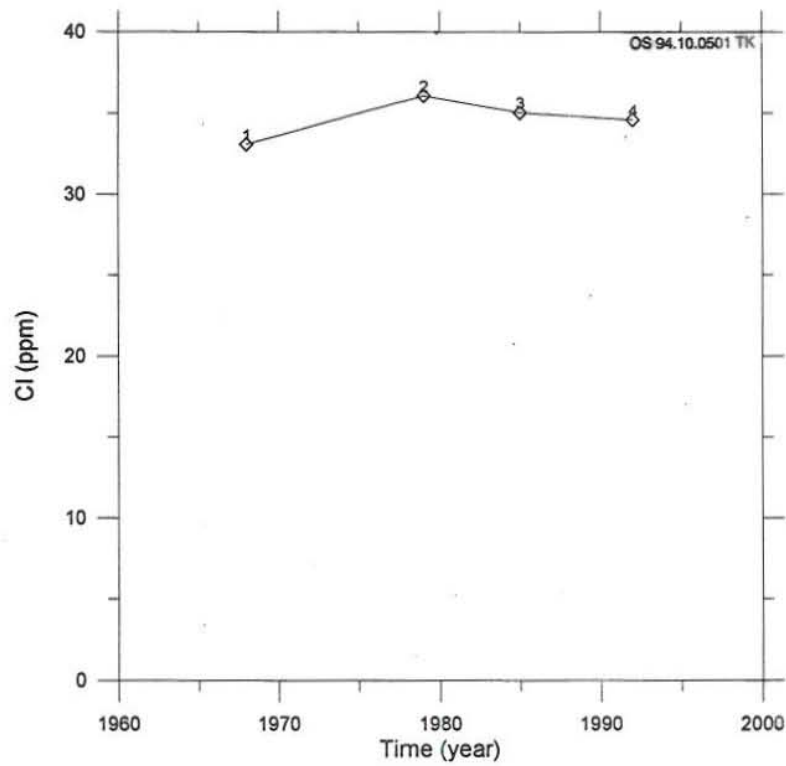


FIGURE 6: Changes in chloride concentration at Laugarvatn with time

4. METHODS

4.1 The computer programme WATCH

The aim of the computer programme WATCH (Arnorsson et al., 1982; Bjarnason, 1994) is to serve as a tool for interpreting the composition of geothermal fluids. This is done by calculating their chemical speciation, for different geothermometer temperatures and is also applicable to other chemical problems. The input to the programme is from a component analysis of each phase (liquid, gas, steam) of the geothermal water at the surface, including the water pH and the temperature at which it was measured, and the reference temperature at which the composition is to be calculated. The concentrations of all the species (67 and H^+ and OH^- , which are treated separately) considered in the programme are expressed in terms of the component concentrations by mass balance equations. The two sets of equations are solved simultaneously by an iterative procedure. This procedure is carried out a few times during each run of the programme. An initial estimate of the ionic strength of the water is obtained using only the concentrations of the major cations. This value of the ionic strength is used to compute activity coefficients, at the temperature of the pH measurement, using the extended Debye-Huckel formula. The chemical equilibria and mass balance equations are now solved simultaneously at this temperature to yield a distribution of the species. From the new distribution of the species, a proper ionic strength is computed. This ionic strength is used to recompute the activity coefficients and species concentrations still at the temperature of the pH measurement.

The pH-calculation, the calculation of the activity coefficients, the mass balance iteration, and the ionic strength calculation are all, in turn, carried out a couple of times. The equilibrium gas pressures are then computed from the final equilibrium species concentrations. Reference temperature can be chosen in several ways, i.e. as the measured temperature of the well or the spring, or a chalcedony, quartz, or sodium - potassium chemical geothermometer value, or in fact by any arbitrary value. The geothermometer temperatures are calculated from the activities of the chemical species. This means that the pH and speciation calculations must be repeated each time the geothermometer temperature is updated. This is iterated until the temperature is consistent to within 0.01°C.

The output from the programme, lists the component and species concentrations at the reference temperature, as well as the activity coefficients. Ionic balance, geothermometer temperatures, partial pressures of gases, and redox potentials are also calculated. Finally, the ion activity products ($\log Q$) and solubility products ($\log K$) of selected minerals (29 geothermal minerals) are computed. From these, the corresponding saturation indices ($\log Q/K$) can be obtained.

The WATCH programme can also be used to compute the resulting activity coefficients for each species and activity and solubility products when the equilibrated water is allowed to cool conductively or by adiabatic boiling from the reference temperature to some lower temperature.

4.2 Characterization of the water according to origin and type

4.2.1 Stable isotope composition

When water evaporates from the ocean under equilibrium conditions, the vapour should be depleted by about 80‰ in deuterium reported as δD and oxygen-18 9‰ reported as $\delta^{18}O$ (Craig 1961) relative to the ocean water. However, this change is also controlled by geographical and time variations. The deuterium concentration of the precipitation in Iceland varies significantly from one place to another. This suggests the possibility of using deuterium as a natural tracer to estimate the recharge areas of groundwater systems and to study their flow patterns. When a sample of groundwater discharging from either a cold or hot spring which has a deuterium value similar to the mean value of the local precipitation, this can indicate a local

origin for the groundwater (Arnason, 1976). The use of the deuterium as a natural tracer in groundwater studies is based on the assumption that the groundwater is derived from precipitation and that the deuterium content of the water does not change in the ground. Furthermore, the method assumes that the deuterium value of precipitation has not changed significantly during the time the water has remained on the ground. Also, Arnason (1976) concluded that δD - $\delta^{18}O$ relation obtained for hot and cold water in Iceland supports the contention that the groundwater is almost entirely of meteoric origin. He also mentioned that the stable isotope content of the precipitation has not changed greatly during the last 8000 years, so deuterium values of groundwater younger than 8,000 years, should be comparable to the values of the deuterium map of Iceland. Comparison of deuterium content of a deep ice core with $\delta^{18}O$ content of a more recent ice core, indicates that precipitation deposited between 10,000 to 60,000 years ago was -50 to -100‰ lower than today. However, the results of deuterium measurements suggest that most groundwaters in Iceland are younger than 8,000 years.

4.2.2 Cl/B ratio

The sources of Cl and the B in geothermal waters are 1) seawater spray/aerosols, 2) seawater that has infiltrated the bedrock and 3) the rock being altered by the waters. Rock dissolution which seems to be stoichiometric with respect to Cl and B causes lowering of aqueous ratios since the Cl/B ratio in Icelandic basalts lies in the range 50-150 but that of seawater spray is the same as the Cl/B ratio of seawater or 4350 (Arnorsson et al., 1989). Thus, a study on this ratio can yield valuable information on the origin of the water.

4.2.3 Cl-SO₄-HCO₃ triangular diagram

Cl is the most common anion in seawater and sea spray but also originates from dissolution of rock. Once in solution it does not take part in water-rock reactions. Sulphate comes to smaller extent from sea water and a large concentration is usually derived from sulphide in volcanic (magmatic) steam. It is less stable than Cl and will take part in reactions and steam heated water often tend to be quite acid. Carbonate is often found at the periphery of high-temperature systems, where there is an excess of it, often from a magmatic source. In cool water carbonate may also originate from biological sources. It is also reactive. Thus relatively high concentrations of sulphate and carbonate often reflect reactive waters whose equilibria with alteration minerals may be disturbed and therefore the basis for geothermometry is shaky. This type of diagram, Cl-SO₄-HCO₃ (Giggenbach, 1991) is therefore often used to weed out waters that are not suitable for geothermometry. In some ageing magmatic systems equilibrium with alteration minerals may be established even though there is an excess of sulphate (and sulphide) in the water. In such cases the waters will not be acid.

The Cl-SO₄-HCO₃ triangular diagram is based on the relative concentrations of the three major anions (Cl, SO₄, HCO₃). The position of a data point in such a diagram is simply obtained by first forming the sums of the concentrations C_i in (mg/kg) of all the three constituents involved:

$$S = C_{Cl} + C_{SO_4} + C_{HCO_3} \quad (1)$$

and then working out the percentage concentrations for each of the three constituents.

4.3 Geothermometry

4.3.1 General

The determination of the subsurface equilibrium temperature of the geothermal fluid, is one of the important aims of geothermal investigations. Both quantitative and qualitative techniques are used to predict the

temperature. The quantitative techniques currently available require chemical analyses of thermal waters from springs and wells. In contrast, qualitative techniques may be used to look for anomalous concentrations of various "indicator" elements in a large variety of hosts, including soil, soil gases, fumaroles, hot and cold springs, and steam.

Most qualitative geothermometers are based on the distribution and relative concentration of volatile elements in waters and soils or variation in soil-gas composition. Anomalously high concentrations of volatile elements may accumulate above or around relatively shallow heat sources especially if subsurface boiling occurs. White (1970) reviewed the qualitative geothermometers that had been suggested up to then. Tonani (1970) suggested that enrichment of B, NH₄, HCO₃, Hg and H₂S in water might result from boiling at depth: High concentrations of volatile components often accumulate above or around a shallow heat source. An increase of H₂S concentration in water may be an indication of a higher temperature at depth than at the surface. Decreasing and very low concentration of Mg is an indicator of thermal water.

The quantitative geothermometers can be divided into two groups, chemical and isotopic geothermometers. The latter group is mostly used for high-temperature waters. Chemical geothermometers are sustained by the evidence of chemical equilibria between deep solutions and types of mineral phases respectively, the silica minerals and alkali feldspars.

Various chemical geothermometers have been developed to predict reservoir temperatures in geothermal systems, silica geothermometers (e.g. Fournier and Rowe, 1966; Arnorsson 1975; Fournier and Potter, 1982; Arnorsson et al., 1983; Arnorsson, 1985), Na-K geothermometers (e.g. Fournier, 1979; Arnorsson et al. 1983b; Arnorsson, 1985; Giggenbach et al., 1983;), Na-K-Ca geothermometers (Fournier and Truesdell, 1973; Arnorsson, 1985). These geothermometers are all based on the assumption that specific temperature dependent mineral solute equilibria are attained in the geothermal reservoir. When applied to the same fluids, the various geothermometers frequently yield appreciably different values for reservoir temperatures. This may be due to lack of equilibrium between the respective species and hydrothermal minerals or due to reactions or mixing with cold water during the up-flow, both of which modify the hot water composition. It is clearly desirable to have an approach that maximizes the use of the available analytical data on the water and allows the validity of the assumptions of specific solution/mineral equilibria to be tested.

4.3.2 The silica geothermometers

The silica geothermometers used at present to predict subsurface temperature of reservoirs, are based on experimentally determined solubilities of chalcedony and quartz. Generally speaking, the quartz geothermometer is applied to high-temperature reservoirs but the chalcedony geothermometer to low-temperature reservoirs. In Iceland, rocks are young, but the chalcedony geothermometer temperature is generally thought to apply up to 180°C and the quartz geothermometer at higher temperatures. Where older rocks prevail, quartz equilibrium seems to be attained at lower temperatures, in some cases down to 90-100°C or even lower. An important fact to be considered when applying the silica geothermometer, is that equilibrium between mineral and solution involves only the unionized silica in the solution. Since 1960 several formulae for the estimation of deep temperatures, based on concentrations of silica in thermal water, have been presented. These formulae give more or less the same temperature results, the only difference being that the quartz geothermometer yields higher temperatures than the chalcedony geothermometer for the same concentration of silica due to their different equilibration solubilities. Fournier (1977) presented the chalcedony geothermometer as the silica concentration, *C*, is in mg/kg:

$$t^{\circ}\text{C} = \frac{1032}{4.69 - \log C} - 273.15 \quad (2)$$

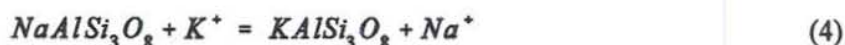
The expression for the quartz geothermometer at the condition of no steam loss presented by Fournier and Potter (1982) is:

$$t^{\circ}\text{C} = -\alpha + \beta C - \gamma C^2 + \delta C^3 + \epsilon C \log C \quad (3)$$

where $\alpha = 42.198$; $\beta = 0.28831$; $\gamma = 3.6686 \times 10^{-4}$; $\delta = 3.1665 \times 10^{-7}$; $\epsilon = 77.034$.

4.3.3 Cation geothermometers

The Na-K geothermometer is based on ion exchange reactions whose equilibrium constants are temperature dependent. An example is the exchange of Na^+ and K^+ between coexisting alkali feldspars :



The equilibrium constant, K_{eq} , for Equation 4 is:

$$K_{eq} = \frac{[\text{KAlSi}_3\text{O}_8] [\text{Na}^+]}{[\text{NaAlSi}_3\text{O}_8] [\text{K}^+]} \quad (5)$$

If the activities of the solid reactants are assumed to be in unity and the activities of the dissolved species are about equal to their molal concentrations, Equation 5 reduces to:

$$K_{eq} = \frac{[\text{Na}^+]}{[\text{K}^+]} \quad (6)$$

Several empirical Na/K geothermometers suggested by various authors, have been presented, but there is a large discrepancy between temperatures, resulting from the different equations at low-temperature. The following formulae presented by Arnorsson et al. (1983) and Giggenbach (1988), are used in this report (the concentrations of Na and K are in mg/kg):

Na/K temperature by Arnorsson et al. (1983), valid in the range of 25-250°C:

$$t^{\circ}\text{C} = \frac{933}{0.993 + \log(\text{Na}/\text{K})} - 273.15 \quad (7)$$

Na/K temperature by Giggenbach (1988)

$$t^{\circ}\text{C} = \frac{1390}{1.750 + \log(\text{Na}/\text{K})} - 273.15 \quad (8)$$

The Na-K-Ca geothermometer (Fournier and Truesdell, 1973) takes account of reactions involving exchange of Na^+ , K^+ , and Ca^{2+} with mineral solid solutions. This minimizes but does not eliminate effects of disregarding the activity coefficients of solids. The geothermometer is entirely empirical and assumes one type of base exchange reaction at temperatures above about 100°C (concentrations are in mg/kg):

$$t^{\circ}\text{C} = \frac{1647}{\log(\text{Na}/\text{K}) + \beta (\log(\sqrt{\text{Ca}/\text{Na}}) + 2.06) + 2.47} - 273.15 \quad (9)$$

where $\beta = 4/3$ for $t < 100^\circ\text{C}$; and $\beta = 1/3$ for $t > 100^\circ\text{C}$.

The Na-K-Mg triangular diagram. Giggenbach (1988) suggested that a triangular diagram with Na/1000, K/100, and Mg at the apices could be used to classify waters according to their state of equilibrium at given temperatures. He adopted the Na/K and slightly modified K/Mg geothermometer equations given by Giggenbach et al. (1983). The full equilibrium curve is for reservoir water composition corrected for loss of steam owing to decompressional boiling. Uncorrected boiled water will plot slightly above the full equilibrium line. This overcomes some of the disadvantages of the direct use of Na-K and Na-K-Ca geothermometers where e.g. mixing with an immature water or incomplete equilibration interferes with their use but the magnesium concentration (low in equilibrated geothermal waters) can help in weeding out unsuitable samples. The magnesium correction suggested by Fournier and Potter, (1979) has the same aim but is more cumbersome and less successful.

The use of the triangular diagram is based on the temperature dependence of the three reactions:



They involve minerals of the full equilibrium assembling after isochemical re-crystallization of an average crustal rock under conditions of geothermal interest. Na, K and Mg concentrations of waters in equilibrium with this assemblage are accessible to rigorous evaluation. The coordinates of a point on the diagram are calculated by

$$S = C_{\text{Na}}/1000 + C_{\text{K}}/100 + \sqrt{C_{\text{Mg}}} \quad (13)$$

$$\% \text{-Na} = C_{\text{Na}}/10S \quad (14)$$

$$\% \text{-K} = C_{\text{K}}/S \quad (15)$$

$$\% \text{-Mg} = 100\sqrt{C_{\text{Mg}}}/S \quad (16)$$

4.3.4 Mineral equilibrium geothermometry

Reed and Spycher (1984) have proposed that the best estimate of reservoir temperature can be attained by considering simultaneously, the state of equilibrium between a specific water and many hydrothermal minerals as a function of temperature. Equilibrium constants are both temperature and pressure dependent. However, pressure in the range occurring in geothermal systems (0-200 bar) has a very limited effect. On the other hand equilibrium constants for mineral dissolution often vary strongly with temperature. Therefore, if a group of minerals converges to equilibrium at a particular temperature, this temperature corresponds to

the most likely reservoir temperature, or at least the temperature of the source aquifer, for the particular water considered. Reed and Spycher (1984) consider their procedure capable of distinguishing between equilibrated geothermal waters and waters that have departed from equilibrium due to boiling or mixing with shallow colder waters.

For the dissolution of a mineral, m , the activity quotient, Q_m , can be written

$$Q_m = \prod \frac{a_{i,m}^{v_{i,m}}}{a_m} \quad (17)$$

Where $a_{i,m}$ is the activity and $v_{i,m}$ is the stoichiometric coefficient of species in the mineral m , written with the mineral on the left hand side of the reaction equation, and the aqueous components on the right hand side, the activity of mineral m , a_m , being equal to one for pure minerals. The free energy, ΔG , of any chemical reaction including mineral dissolution is given by

$$\Delta G_m = +\Delta G_{m,r}^\circ + RT \ln Q_m \quad (18)$$

and the stoichiometric coefficients $v_{i,m}$ are positive for those species that appear on the right and negative for those on the left side of the equilibrium expression.

At equilibrium $\Delta G = 0$. The relationship between the standard partial molal free energy of reaction, $\Delta G_{m,r}^\circ$ and the equilibrium constant, K_m , is

$$\ln K_m = \frac{\Delta G_{m,r}^\circ}{RT} \quad (19)$$

Therefore, $Q_m = K_m$, and $\log (Q_m/K_m) = 0$, at equilibrium. In a plot of $\log (Q_m/K_m)$ vs. temperature, the values for all minerals that are in contact with the solution will converge to a value of 0 at the same temperature.

When geothermal fluids at equilibrium with hydrothermal minerals mix with solutions of different composition, the equilibrium is disturbed causing, a) a shift in the position in which the minerals are apparently at equilibrium to a lower $\log (Q_m/K_m)$ values if the solutions that mix with the geothermal fluid are very dilute; or b) a complete lack of any identifiable equilibrium temperature if the solution mixing with the geothermal fluid contains solutes in proportions very different from the geothermal water. Furthermore there will be a dispersion of the $\log (Q_m/K_m)$ plots, if the geothermal fluid has boiled before it is sampled, since the formation of the vapour and the loss of gases will lead to, a) a simple concentration of none-volatile components in the residual aqueous solution which would tend to give positive $\log (Q_m/K_m)$ values; and b) a change in the gas content of the aqueous solution and a simultaneous increase in pH, both of which tend to cause supersaturation of certain mineral phases (e.g. calcite and pyrite). These factors will lead to complex changes in the various apparent equilibrium mineral/solution temperature.

4.3.5 Mixing models

The geothermal water may cool during up-flow from the reservoirs before it emerges at the surface in hot springs. It may cool by conduction, boiling or mixing with shallow cold water, or by any combination of these

three processes. Therefore, the chemical composition of the thermal waters may be changed. Water formed by mixing of geothermal water and cold ground water has characteristics which serve to distinguish it from unmixed water (Amorsson, 1985). Unmixed geothermal waters have a characteristic chemical composition which is controlled by equilibrium conditions between certain dissolved constituents of the water and minerals in the rock. However, the kinetics of the leaching processes control the changes in composition of cold groundwater or surface water. The chemical composition of the mixed water may be used for temperature estimation by applying mixing models (Fournier, 1977). Mixing models should never be applied unless there is independent evidence for mixing. Some indications of mixing are as follows:

1. Variations in chloride content of boiling springs too great to be explained by steam loss;
2. Variations in ratios of conservative elements, such as B/Cl;
3. Variation in oxygen and hydrogen isotopes;
4. Cool springs with large flow rate and much higher geothermometer temperatures (greater than 50°C) than measured temperatures;
5. Systematic variations of spring compositions and measured temperatures, generally the colder water will be more dilute than the hotter water.

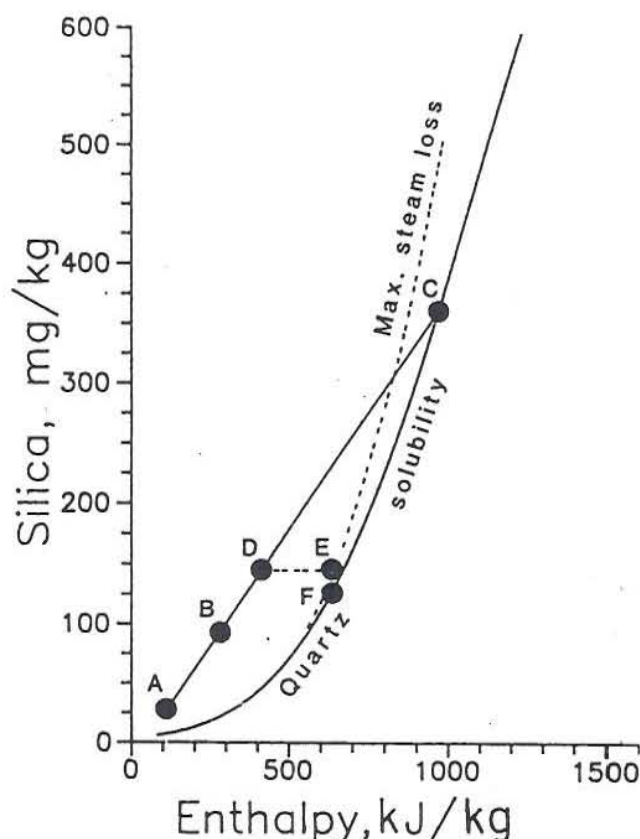


FIGURE 7: The silica-enthalpy graph illustrating calculation of silica mixing model temperature

The silica-enthalpy mixing model (Fournier, 1977). In order to determine the temperature of the hot water component the dissolved silica concentration of mixed water and a silica-enthalpy diagram may be used (Figure 7). A straight line drawn from a point representing the non-thermal component of the mixed water (point A) through point B representing warm mixed-water, to the intersection with the quartz solubility curve gives the initial silica concentration and the enthalpy of the hot water component (point C). This procedure assumes that any steam that formed adiabatically (as the hot-water component moved up to a more shallow environment) did not separate from the residual liquid water before mixing with the cold-water component. D represents the boiling point, to account for separation by boiling. A line is horizontally extended from D, the point at which the line A-B intersects the curve representing the boiling point enthalpy at atmospheric pressure, until it intersects the curve representing the SiO₂ concentration after maximum steam loss at point E which represents the original enthalpy of the geothermal component. The concentration of silica may be read directly from the graph as well as the predicted temperature using enthalpy and steam tables.

The enthalpy-chloride mixing model (Fournier, 1977) has been used to predict underground temperatures, salinities, boiling, and mixing relations. Referring to Figure 8, waters that result from the mixing of a low-enthalpy (low-temperature) water, such as G, with high-enthalpy waters such as P, B, or C, lie along lines radial to G. For example, waters M, N, and R could result from the mixing of waters G and P in different proportions. Similarly, waters D and L could result from the mixing of G and C in different proportions. The

points B, C, and E can be related to P by boiling along curve 1. Point K can be related to N by boiling along curve 3. The water represented by the point L is well below boiling after mixing; the water represented by D has a temperature well above 100°C after mixing, and water-rock chemical re-equilibration may occur in hours or days owing to the higher temperature. Point A is drawn to illustrate that a deep parent water, P, can be related to a water such as A by various mechanisms such as: 1) water P can first boil to produce water B, and then B can mix with G to give A; 2) water P can first mix with G to give water M, and M can then boil to produce A, or it can cool directly to A by conductive cooling.

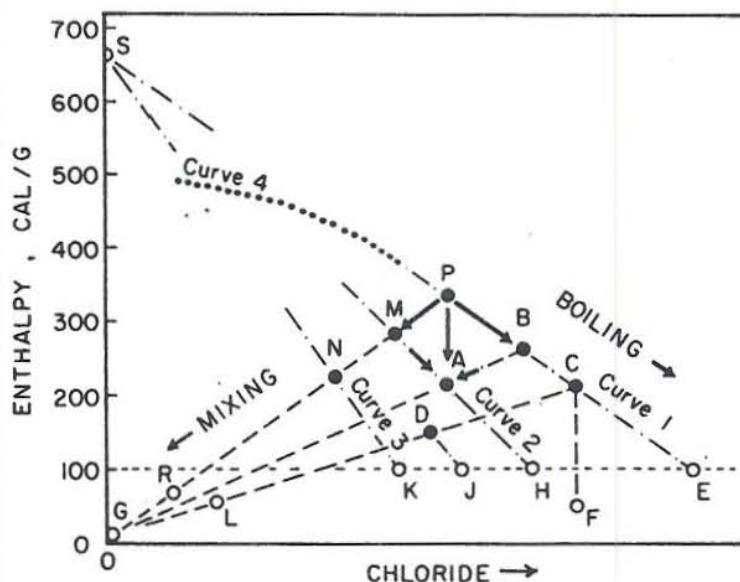


FIGURE 8: The enthalpy-chloride graph illustrating calculation of enthalpy-chloride mixing model temperature

The silica-carbonate mixing model. Arnorsson et al. (1983) found that the concentrations of carbon dioxide in waters in geothermal reservoirs were only dependent on temperature. They concluded that this was the result of overall solute/mineral equilibration in these reservoirs. At temperatures above about 200°C, most of the dissolved total carbonate is in the form of carbon dioxide, so it is a satisfactory approximation to take analyzed carbonate to represent carbon dioxide. It is well known that silica levels in high-temperature waters are determined by quartz solubility. It follows, therefore, that it is a satisfactory approximation to assume a fixed relation between silica and total carbonate in high-temperature geothermal reservoir waters. Boiling of such waters will lead to a drastic reduction in its carbonate content but mixing without boiling will follow the silica/carbonate relationship. The silica-carbonate diagram may be used in two ways to aid geothermometry interpretation. First, it serves to distinguish boiled waters from conductively cooled waters and mixed waters which contain an undegassed (and therefore unboiled) hot water component, assuming that the boiling occurs between the points of last equilibrium with quartz and sampling. Second, if there are sufficient data on warm waters containing an unboiled water component, the diagram may be used to evaluate the temperature of the hot water component. The diagram is useful as a supplement to interpretation of the silica-enthalpy warm spring mixing model because a choice often needs to be made as to whether it should be assumed that boiling occurs before mixing or not. Little steam formation suffices to deplete the original hot water almost quantitatively in total carbonate. If it has boiled, the points fall above the $\text{SiO}_2\text{-CO}_2$ curve.

If there is no boiling the model can be used for estimation of underground temperatures by model extrapolation of a line through the data points for mixed and undegassed warm waters and determination of the point of intersection with the silica-carbonate curve for equilibrated waters. From the silica concentration corresponding to this point, the temperature may be derived using the quartz geothermometer.

5. INTERPRETATION OF THE RESULT

5.1 Origin and type of water

Stable isotope data: The low-temperature geothermal water in Iceland is of meteoric origin. This has been confirmed by deuterium isotope measurements (Arnason, 1976). On the basis of drillhole data on

permeability and temperature Bjornsson (1980) showed that, some of the low-temperature systems were convection systems. Bodvarsson (1983) presented a conceptual model of the low-temperature fields. According to his model the circulating groundwater is heated in narrow fissures, which continuously migrate downwards during the heat mining process. Theoretical considerations of the heat process suggested by Bodvarsson (1983) show that it explains the power of even the largest low-temperature areas in Iceland. In addition, these considerations show that the power of such low-temperature systems is controlled by the temperature conditions in the crust and in particular the local stress field. Given the thermal conditions in the crust of Iceland it appears therefore that the regional tectonics and the resulting local stress field control the low-temperature activity.

Arnason (1976) mapped the deuterium content of rain water in Iceland and presented as δD values. It varies between -50 and -100‰ and decreases with distance from the coast. The deuterium content of water from the Laugarvatn spring, (samples 1-4) and Sog Laugardalur (sample 6) suggest the same origin of water, because they have a similar δD value. According to these δD values, the recharge area of the water is probably somewhere in the mountainous area south of the Langjokull glacier. All these samples are located in the western part of the area. The δD values of samples 5, 9, 14, 18, and 19 are similar, with an average of -71.4‰ and they are located in the eastern part of the area. According to this, the likely recharge area for this thermal water should lie just south of the Langjokull glacier (Figure 3).

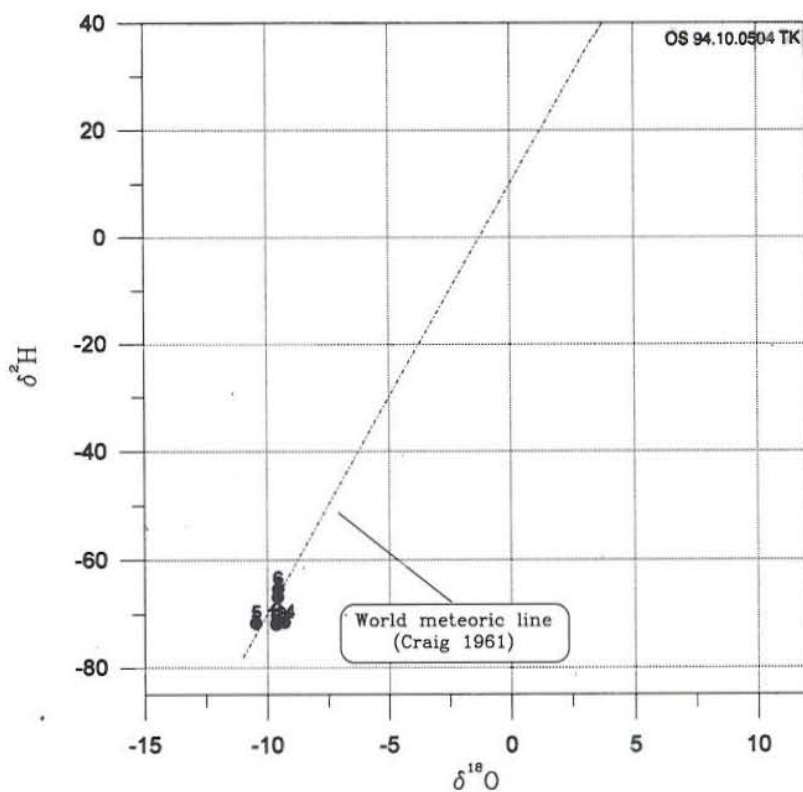


FIGURE 9: δD (δ^2H) vs. $\delta^{18}O$ for the Laugarvatn waters

The deuterium content of the geothermal systems in the Southern Lowlands is consistent with an origin in the interior highlands (Arnason, 1976). Deep flow of groundwater from the recharge area driven by the general hydraulic gradient is assumed to feed the geothermal systems. Figure 9 shows that all the data points plot approximately on the world meteoric line.

Cl/B ratios: On the basis of Cl/B ratios Arnorsson et al. (1993) classified the Southern Lowlands of Iceland into three geographically distinct systems, the Hreppar, Biskupstungur including the Laugarvatn area, and Selfoss systems. Geothermal waters not containing a component of infiltrated seawater and with temperatures near and

above 100°C have generally reacted sufficiently with the rock to obtain a Cl/B ratio similar to that of basalt. In the Biskupstungur system the Cl/B ratios are in the range of 100-400, i.e. similar to, or somewhat higher than, such ratios in tholeiitic basalts. The Cl/B ratios of samples 8 and 10 are similar and they may be associated with alkali basalts. Samples 1, 2, 9, and 19 range between 236-259, so they seem to be similar to such ratios in tholeiitic basalts. The Cl/B ratios of samples 11, 12 and 17 range between 300-400 and also fall in the same range of the tholeiitic basalts. This means that the Cl/B ratios only suggest a slightly different origin of the thermal waters within the area.

The Cl-SO₄-HCO₃ triangular diagram: Figure 10 shows that most of the samples plot either in the volcanic or steam heated water regions (samples 1, 2, 8, 9, 10, 17 and 19), which are sulphate, volcanic and steam heated waters and are generally encountered in the elevated parts of a field. They are, however, not ordinary steam heated waters, because their pH is quite high. The sulphate is probably formed by the oxidation of the H₂S during up-flow. Arnorsson and Olafsson (1986) argued that this is an old high-temperature system that is cooling down but is still receiving sulphide from a magmatic source. This is borne out by the fact that the pH is quite high and therefore these can not be regarded as ordinary steam heated waters. Samples 5, 6, 13, 14 and 21, are located in the HCO₃ corner and represent cold groundwater. Samples 3, 18, and 20, are close to chloride and sulphate waters, but are still in bicarbonate water range which suggests some degree of mixing. Only one sample, 12, plots in the chloride side.

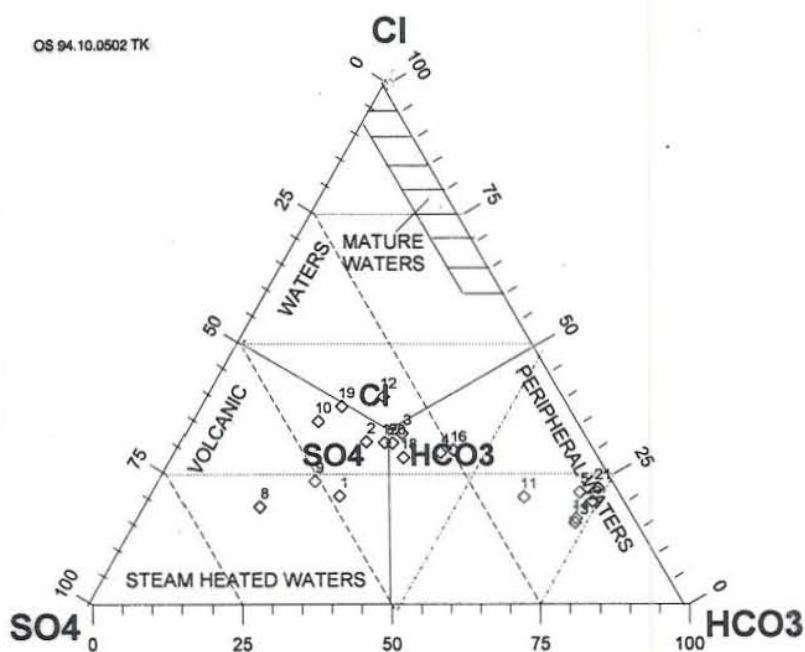


FIGURE 10: The Cl-SO₄-HCO₃ diagram for the Laugarvatn waters

5.2 Subsurface temperatures

5.2.1 Solute geothermometers

In this report chemical geothermometers are used on hot spring waters to predict subsurface temperatures. For various reasons different geothermometers may give different results. Table 2 shows the results for different solute geothermometers. The quartz geothermometer gives about 25°C higher temperature for all the samples than the chalcedony geothermometer. For samples 1-4 there is a clear decrease in temperature with time for both the quartz and chalcedony geothermometers. In fact, all the cooling seems to have taken place from 1986 to 1985. The highest temperatures are recorded for samples 17-19 (in the east part of the area). The silica geothermometer temperature ranges from 105 to 176°C and the chalcedony temperature one from 75 to 144°C. There is a large difference between these results and those of Giggenbach's (1988) geothermometer and Na-K geothermometer, or about 50°C, the latter yielding higher results for all the samples (Arnorsson et al, 1983). The results for the Na-K (Arnorsson et al. 1983) geothermometer are similar to those for the chalcedony geothermometer, the difference being only a few degrees. All samples range between 80-148°C for Na-K (Arnorsson et al., 1983) and 136-192°C for Na-K (Giggenbach, 1988). The highest temperatures are found in samples 17-19.

The Na-K-Ca (Fournier and Truesdell, 1973) geothermometer, using $\beta = 1/3$, yields reasonable results similar to those for the chalcedony and Na-K geothermometers (Arnorsson et al. 1983), in the range of 114-155°C and the highest temperatures are found for samples 17-19. The chalcedony, Na-K (Arnorsson et al. 1983) and Na-K-Ca geothermometers give similar results and seem to be most suitable in this area.

TABLE 2: Results of different geothermometers for the Laugarvatn waters

No.	Sample no.	T _{mea} (°C)	T _q (°C)	T _{chal} (°C)	T _{NaK} (°C)	T _{NaK-Gigg} (°C)	T _{NaKCa-L} (°C)	T _{NaKCa-H} (°C)
1	68-3244	99	166	142	133	182	107	148
2	79-3002	99	154	129	140	187	110	151
3	85-0237	97.5	142	116	134	182	107	148
4	92-0357	99	144	118	124	174	102	142
8	68-3246	44	106	76	121	172	101	140
9	68-3245	99	142	115	115	175	103	143
10	68-3235	86	134	106	124	173	102	141
11	90-0131	43	108	79	80	137	79	114
12	85-0184	93.5	123	94	104	158	93	130
17	79-3003	97	168	145	148	193	114	156
18	71-0117	88	176	154	140	187	110	151
19	68-3234	99	157	132	141	187	110	151
20	85-0238	84.6	154	129	117	169	99	138

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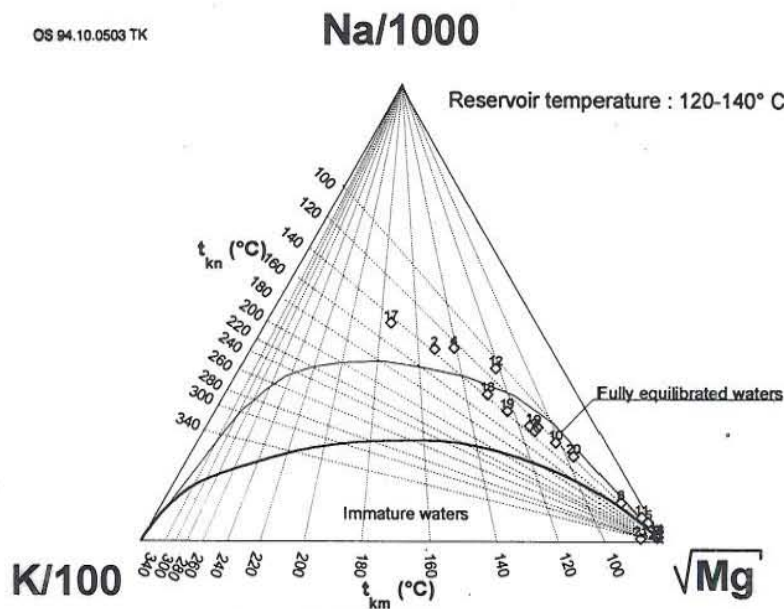


FIGURE 11: The Na-K-Mg diagram for the Laugarvatn waters adiabatic boiling. The Na-K-Mg triangular diagram yields reservoir temperature ranging between 120-140°C for all the geothermal samples.

The Na-K-Mg triangular diagram (Giggenbach, 1988) is shown in Figure 11. The data points on the diagram mainly fall in three groups: 1) The cold groundwater samples (4, 6, 14 and 15) plot near the Mg corner; 2) samples No. 1, 3, 9, 10, 18, 19 and 20 are slightly below the equilibrium curve of Arnorsson et al. (1983) and are partially or nearly equilibrated waters; this is probably due to mixing of the thermal water with a small amount of cold groundwater of meteoric origin during its up-flow; 3) samples 2, 4, 12 and 17 are located a little above the equilibrium curve and are possibly to a small extent affected by

5.2.2 Log Q/K diagrams (SI versus Temperature)

Figure 12 is composed of four graphs representing respectively samples 1, 2, 3, and 4 showing SI (saturation index) from 20-160°C for the Laugarvatn samples. It is the temperature at which the SI curves intersect the zero line that determines the equilibrium temperature for each mineral. In the four graphs, there is no clear temperature-rock equilibrium reached between all the minerals and the thermal fluid, but in some cases two or three minerals attain equilibrium at the same temperature. There are basically two interpretations that can

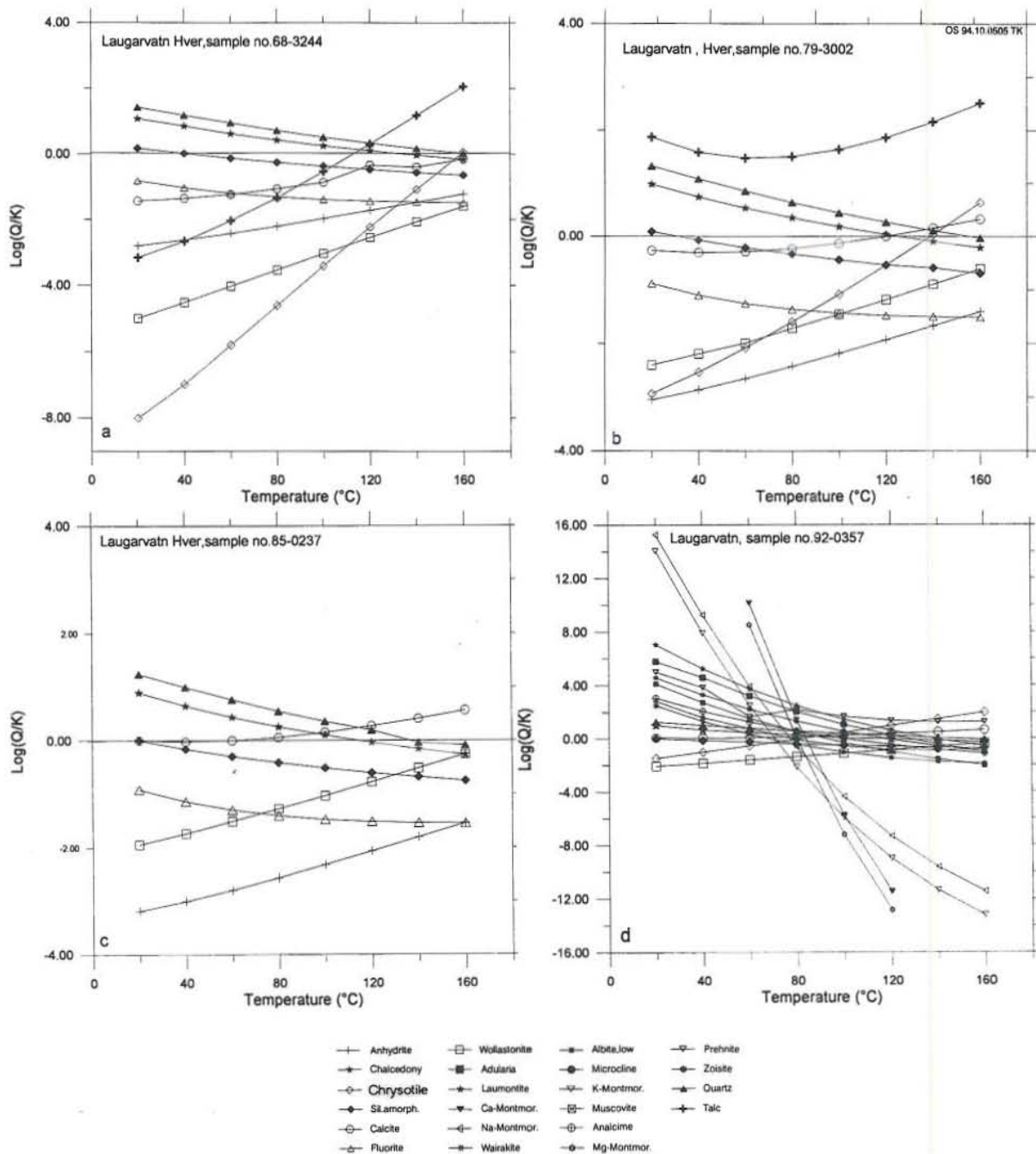


FIGURE 12: Saturation index (log Q/K) vs. temperature calculated with WATCH for the Laugarvatn waters

be given to Figure 12a. If the fluid is in equilibrium with chalcodony we get two intersections in the region 120-135°C, i.e. chalcodony and talc. On the other hand if we assume that the fluid is in equilibrium with quartz we have two intersections at 160°C, i.e. quartz and calcite. In Figure 12d on the other hand there are several intersections in the region 80-100°C which could be interpreted as showing that subsurface temperatures are not likely to be higher than the measured ones. Generally, the calcite, quartz and chalcodony reach saturation in equilibrium in the temperature range studied. The anhydrite, chrysotile, fluorite and wollastonite are under-saturated while talc is always super-saturated at these temperatures. Similar results are obtained for Hjalmsadalauagar, Utey, Austurey, Bodmodsstadir, and Efri Reykir (Figure 13 and 14). Most minerals intersect in the range 90-130°C. The sample from Sydri Reykir clusters around 110°C (Figure 14c). The interpretation of all these figures suggests some mixing of cold ground water with thermal fluid during its up-flow.

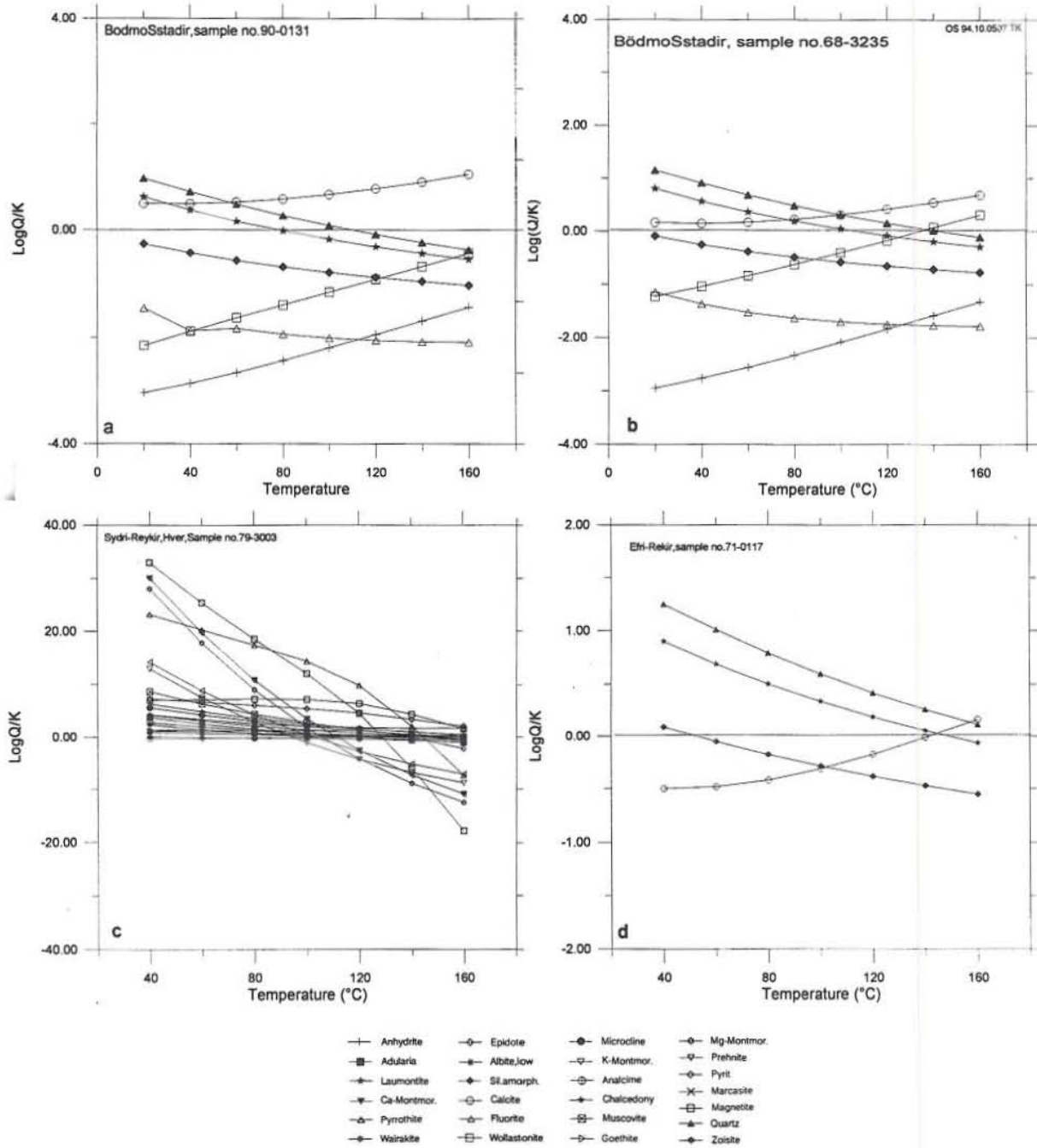


FIGURE 13: Saturation index (log Q/K) vs. temperature calculated with WATCH for waters from some hot springs in the eastern part of the Laugarvatn area

5.2.3 Mixing models

Evidence of mixing: Mixing has been inferred from more than one piece of evidence through the field. First, relatively low chloride and total dissolved salt concentrations in some samples have been recognized. Second, large flow rates in some of the springs and large differences between measured and geothermometer temperatures in some cases. Third, Arnorsson (1985) suggested a linear relation between Cl and most of the constituents as an evidence of mixing which can be demonstrated by Figures 15, 16, 17, and 18 for samples from the Laugarvatn area.

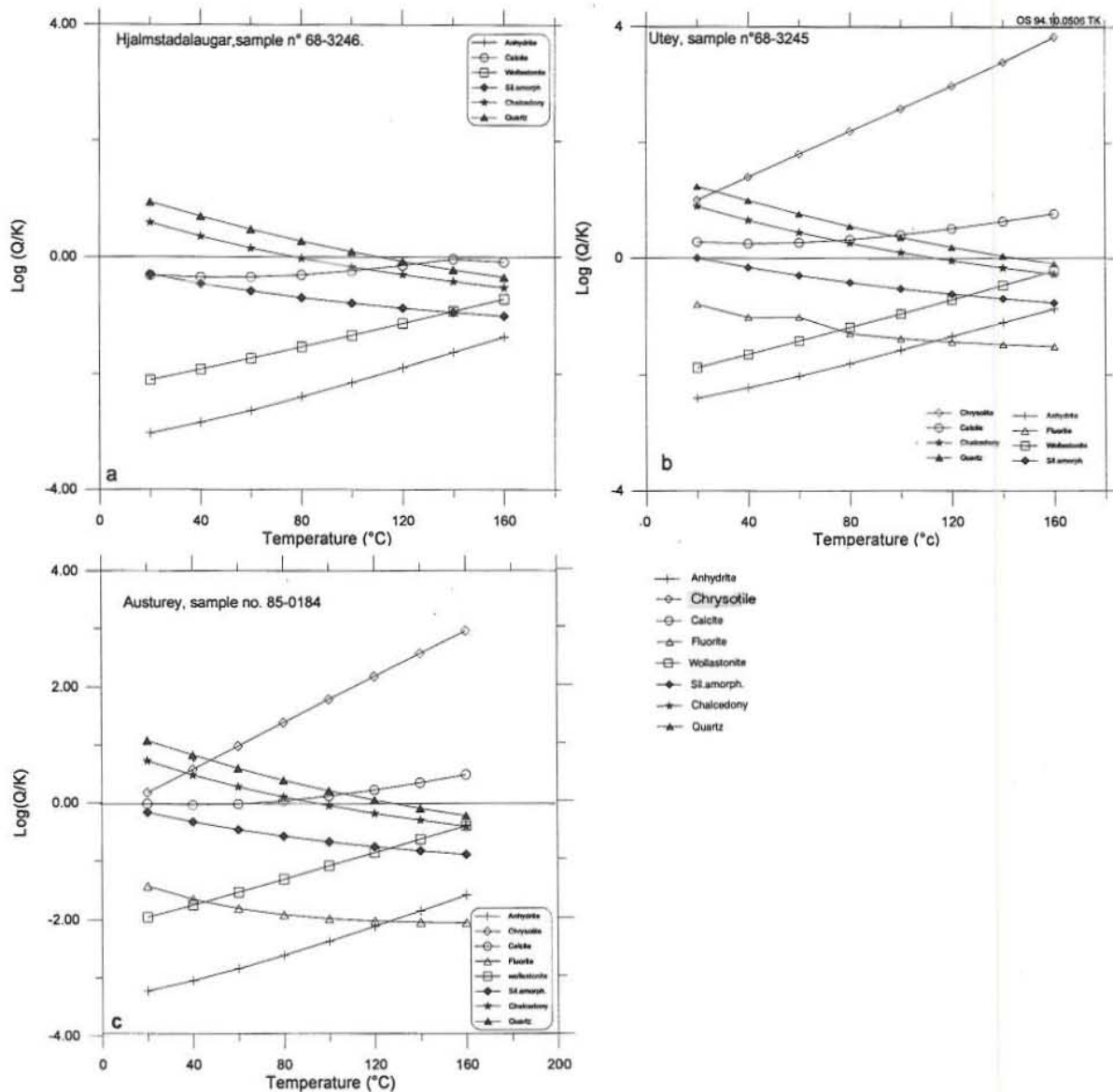


FIGURE 14: Saturation index (log Q/K) vs. temperature calculated with WATCH for waters from some hot springs in the vicinity of Laugarvatn

Silica-Enthalpy mixing model: Figure 19 shows a silica-enthalpy mixing model where mainly two groups of waters are represented. The line a, connects cold water samples 5, 6, 13, 14, and 21 to samples 11 and 12 and intersects at the chalcedony curve (Arnorsson 1983) at the enthalpy value of about 620 kJ/kg which corresponds to 147°C reservoir temperature. The second line, b, connects 5, 6, 13, 14, 21, and 8 with samples 1-4 and intersects with the chalcedony curve at an enthalpy value of about 760 kJ/kg which corresponds to 180°C reservoir temperature which is higher than expected. So the silica temperature is mainly controlled by the chalcedony solubility as it falls in the range suggested by all the authors for silica solubility. The silica concentrations of samples 10, 17, 18, 19 and 20 are higher probably, due to boiling.

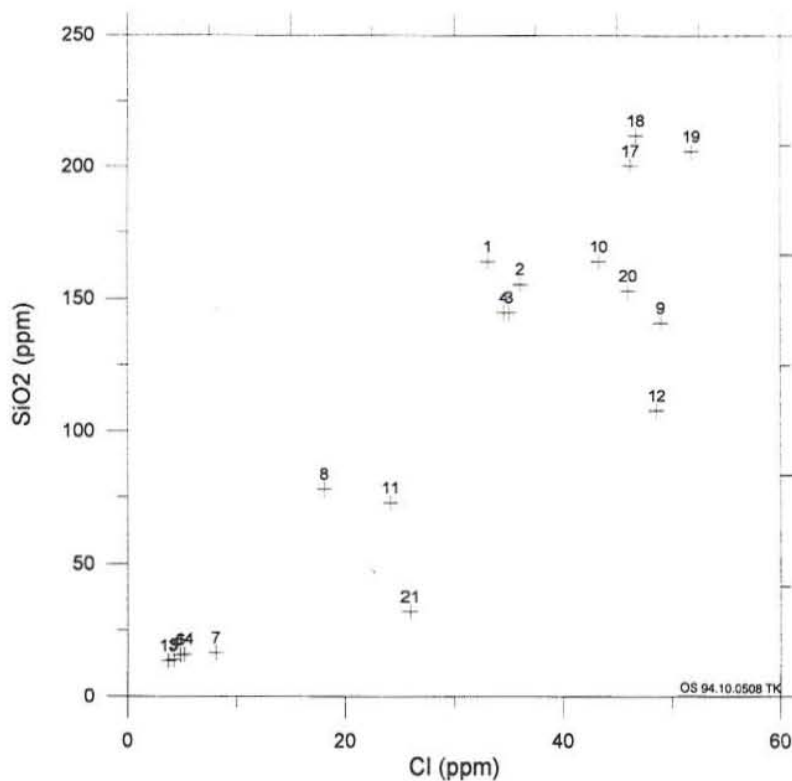


FIGURE 15: Cl vs. SiO₂ for the waters from the Laugarvatn area

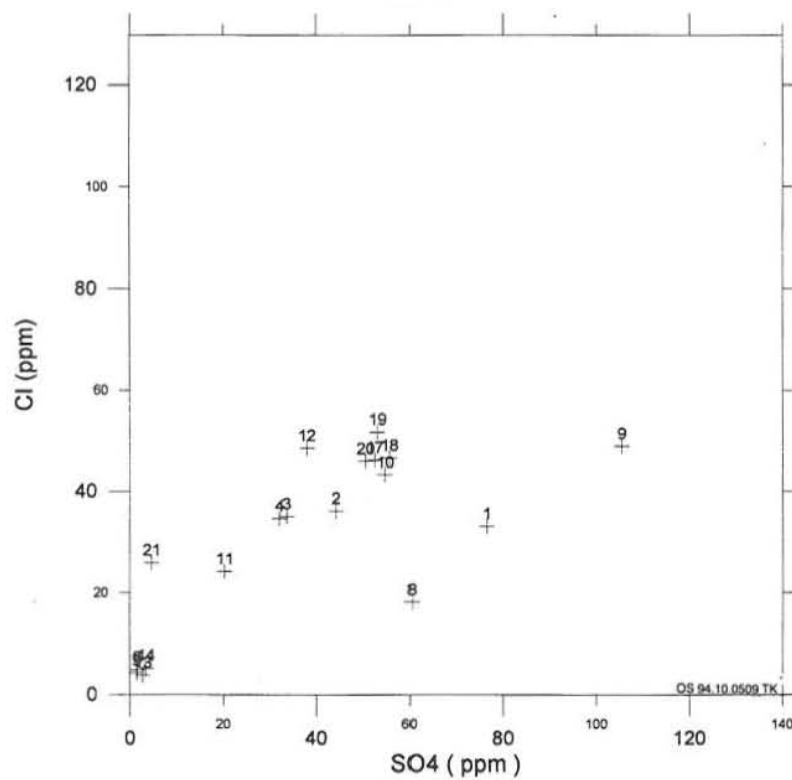


FIGURE 16: SO₄ vs. Cl for the waters from the Laugarvatn area

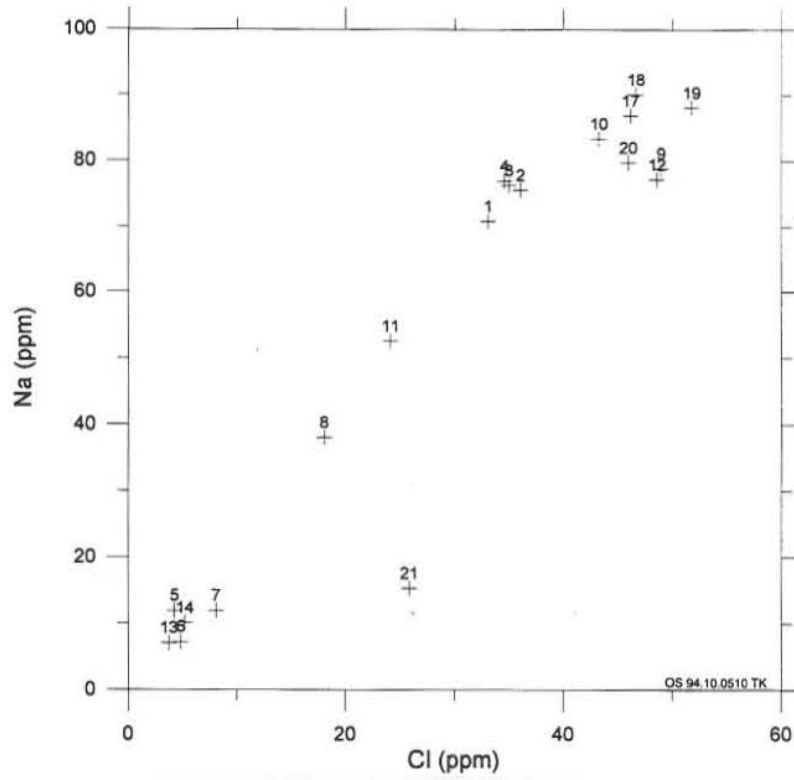


FIGURE 17: Cl vs. Na for the waters from the Laugarvatn area

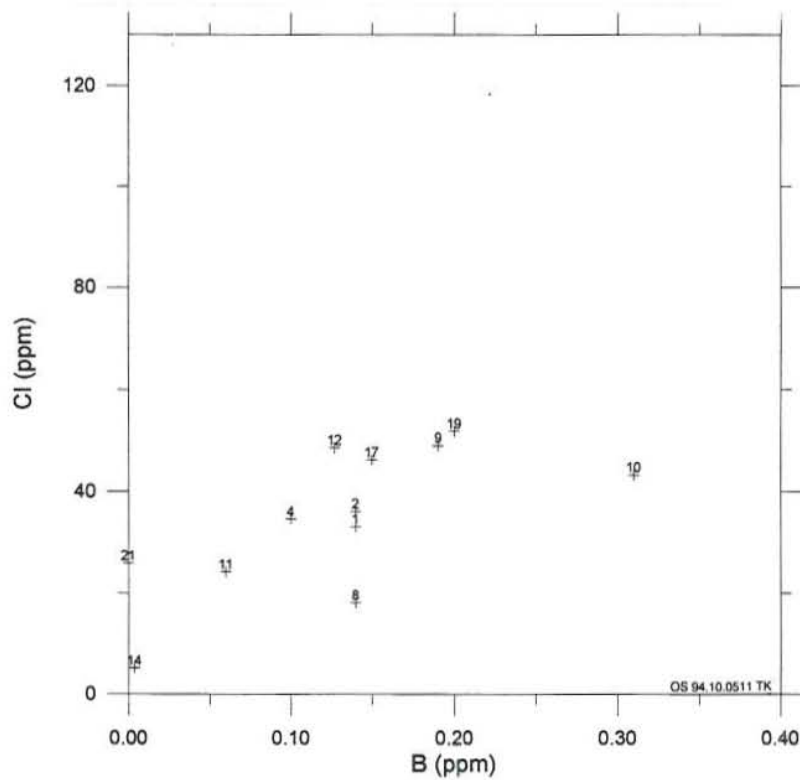


FIGURE 18: B vs. Cl for the waters from the Laugarvatn area

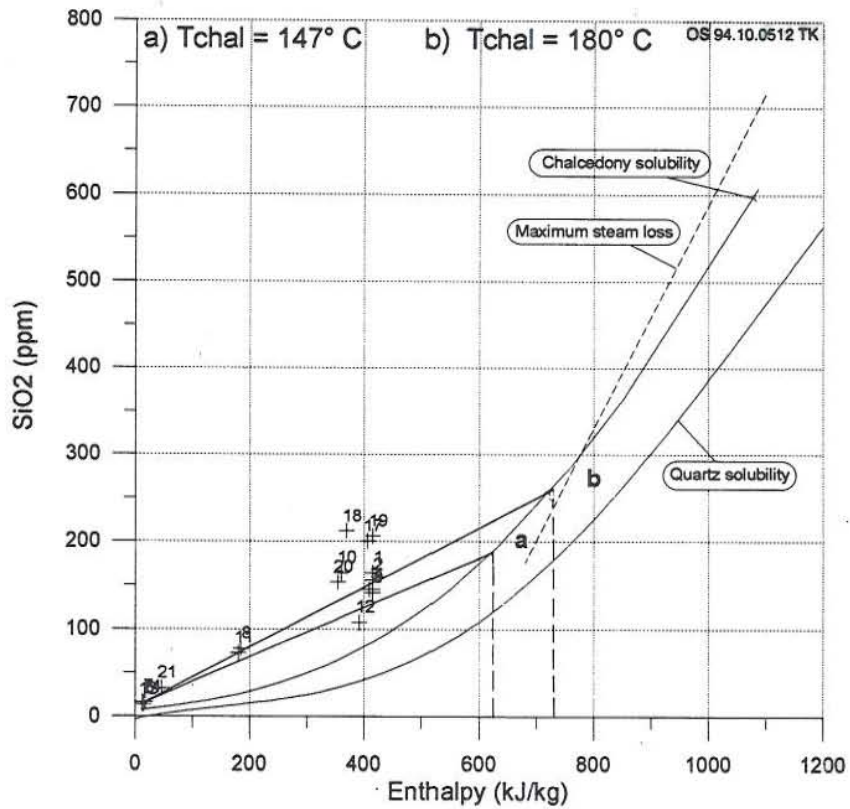
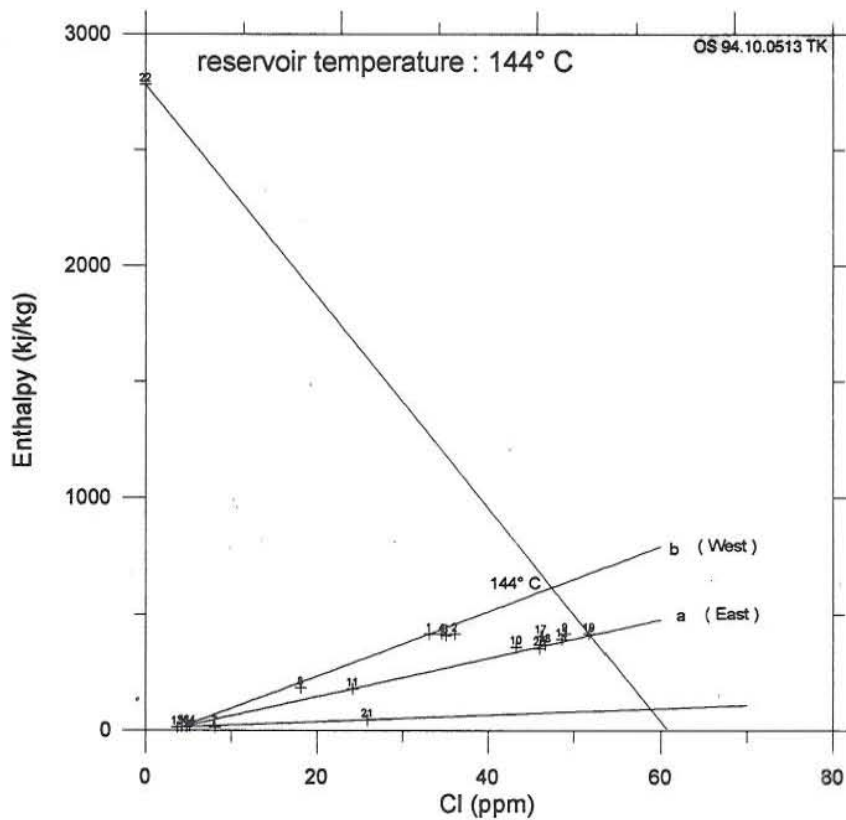


FIGURE 19: The silica-enthalpy mixing model for the geothermal waters from the Laugarvatn area



Enthalpy-chloride mixing model for the Laugarvatn waters is shown in Figure 20. It shows clearly two groups of waters:

Line a connects samples 5, 6, 14, 15, 11 and 9, with 10, 12, 17, 18, 19 and 20, which are affected by cooling probably by two processes, boiling and mixing with cold water and possibly conductive cooling. All these samples are located in the eastern part of the area.

Line b, connects samples 5, 6, 14, 15, and 8 with 1-4 in the west of the area, where the dominant process seems to be mixing with cold water rather than boiling. The enthalpy-chloride mixing model gives a reservoir temperature about 144°C which is similar to the temperatures given by the Na-

FIGURE 20: The enthalpy chloride mixing model for the waters from the Laugarvatn area showing two different groups of waters

K-Mg triangular diagram and the silica-enthalpy mixing model.

In the west of the area cooling is probably due to mixing with cold groundwater rather than boiling, in the east the boiling process is more dominant than mixing with cold water. In the west the high difference in hydraulic pressure increases the possibility of mixing.

6. CONCLUSIONS

1. The reservoir temperature of geothermal water in the Laugarvatn area, seems to be reflected by the chalcedony solubility and is estimated to be in the range of 120-140°C. Deeper down (or in the past) the temperature is expected to be (or having been) higher, possibly as high as 180°C.
2. The Laugarvatn hot spring has cooled with time. This can be seen from the decrease of calculated geothermometer temperatures with time, and can also be confirmed by Cl and SiO₂ calculations. At the same time the flow rate has increased suggesting mixing with cold water at shallow depths.
3. The heat source is probably more powerful in the eastern part of the area (Sydri Reykir, Efri Reykir) as geothermometer temperatures increase towards east.
4. The δD values of the thermal water increase towards the volcanic zone. On the basis of the isotopic data two groups of waters can be suggested. The water in the western part of the Laugarvatn area (Laugarvatn, Sog, Ljosar) is of local origin, while in the east (Sydri Reykir, Efri Reykir, Bodmodsstadir) the δD of the water is lower suggesting a source further inland.

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