Proceedings World Geothermal Congress 2015
Melbourne, Australia, 19-25 April 2015

Hydrochemical Characteristics of Erma Reka Geothermal Reservoir (S. Bulgaria)

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Keywords: geothermal water, hydrogeochemistry, mineral depositions, Bulgaria

ABSTRACT

Erma Reka geothermal reservoir is located in the southern part of the Rhodopes massif (South Bulgaria), close to Greek border, and is a unique combination with a polymetallic Pb-Zn mineralization. The deposit was discovered in 1955 and the exploitation of polymetallic ore continues until now. The water temperature is within the range of 30 to 90°C. The thermal water is accumulated in karst and cavernous marble body, located at a depth of 450 m below the surface and embedded in a gneiss complex. It is pumped to the surface and discharges in a small river, without using its thermal energy. Cavities and caverns, some of which are huge in size (from 400-1300 m) have been crossed during the drilling in the marble body. Thermal water from Erma Reka reservoir discharges through several springs located in the region of Thermes (N. Greece). The obtained TDS values of the water samples vary between 0.6 to 1.3 g/l depending on the depth and special deposition of the reservoir. The TDS is correlating with the ratio change of concentrations of the main species HCO_3^-/SO_4^{2-}, Ca^{2+}/Na^+, and with the H_2SiO_3 content, which reaches up to 250 mg/l at highest mineralization. Contents of other elements are analyzed in thermal water. Varied in composition and texture mineral depositions occur in wellhead where a significant temperature decrease is observed. The values of saturation index of various compounds regarding mineral phases have been determined. It has been established that various carbonate, phosphate and silicate mineral phases could be deposited in the cooling down process. Knowing the process of thermal water scaling is essential for the future use for a space heating in the town of Zlatograd, located at about 12 km to the southeast of Erma Reka geothermal reservoir.

1. INTRODUCTION

The territory of Bulgaria is rich in thermal waters of temperature up to 98°C, Petrov et al. (1970), Shterev, (1964), Bojadgieva et al. (2000, 2010). Erma Reka reservoir is located in Rhodopes massif, Southern Bulgaria, Fig.1. It was discovered during the period 1953-1955 in the process of reconnaissance drilling for lead-zink ores, Bogdanov (1960), Dragiev (1996). Hot water of temperature up to 90°C is associated with an unrevealed on the surface karst and cavernous marble body located at depths below 450 m and imbedded in a gneiss complex, Antonov and Ivanova (1967); Petrov and Andreev (1973), Andreev (1995); Yosifov and Andreev (1996), Andreev et al. (1998), Teneva-Georgieva and Andreev, (2005). About 60 wells of depth up to 1500m have been drilled in the deposit. They crossed huge cavities and cavern up to 400-1300 m without being able to reach their bottom. The high water permeability and temperature of the gneiss complex created high geothermal anomaly which hindered mining activity. This required additional survey in order to drainage the marbles, Antonov et al. (1980), Petrov and Jotov (1991). The heat flow density in Erma Reka reservoir above the marbles is calculated to $440x10^{-3}W/m^2$, Bojadgieva and Gasharov (2001). The reservoir is cross-bordered and is discharged in the springs Thermes, situated in Northern Greece, Minissale et al. (1989).

The exploitation of the majority of the lead-zinc deposit has been gradually terminated since 1990 and currently only 3 sectors are under operation. A Phare Project "Integrated use of Erma Reka –Ilidza geothermal system" was completed in 2011. The project was focused on thermal water use for heating of public buildings in the town of Zlatograd located at a distance of about 12 km in southeast direction, Fig.1. The system construction hasn't still started due to some financial problems and additional survey currently going on of the hilly terrain between the source and consumer.

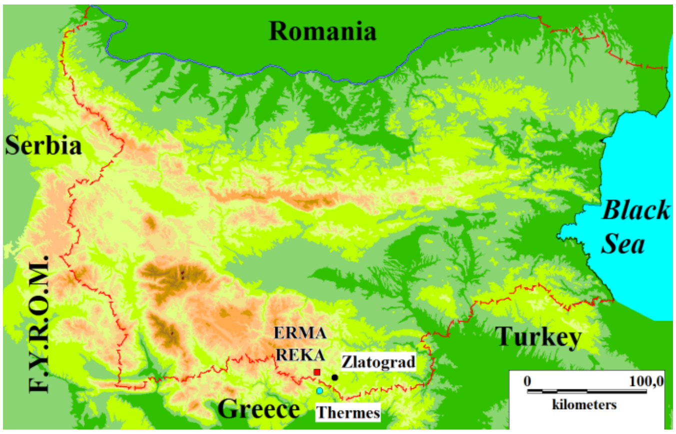

Figure 1: Location map

The future water use requires a extensive knowledge of hydro chemical water composition. In the long-term studies many water samples characterizing the thermal and cold waters in the area, and sources partly influenced by hot water have been analyzed. The aim of the paper is to summaries the existing hydrochemical data, to study the temperature influence on the water chemical composition change and values of saturation index of various compounds regarding mineral phases.

2. GEOLOGY AND HYDROGEOLOGY BACKGROUND

Erma geothermal field is located in the Rhodopes massif. According to the geological map of Bulgaria in the scale 1:100000, Kozhuharov et al. (1989) the region is composed of amphibolites, amphibolite gneisses, leptinites, schists, gneissic shists, and in the high parts of the relief – by graniteschists. Tectonically it is a southern part of an anticline where a dome-like structure was formed. The discovered large deep seated marble body during the well logging is shown in Fig.2. It is located at a depth of over 450 m and has a thickness of more than 1000 m. Cavities and caverns, some of which are huge in size (from tens of centimetres to more than 10-15 meters) have been crossed during the drilling. For example, one well passed through an open cavity at a depth of 800 m to 1250 m and another from 670 to 2020 m without reaching their bottom. The marbles are bordered to the west and north by faults and to the east and south they tin out and cover an area of about 25-30 km^2. The marble body is also crossed by many faults and fractures, which are essential for the hydrothermal system. The faults are characterised by three main directions - NW (275 - 305^o), SE (60- 80°), NE (40 - 70^o) and E (90 - 100^o). A great fault dislocation oriented northeast-southwest direction separates the body in two blocks, which are hydraulically connected. The approximate water quantity is estimated to 200x10^6 m^3.

Hot water upwards through the faults and fractures in the gneisses to the piezometric surface of the absolute elevation +470 to +490 m. The faults create also conditions for cold waters penetration in depth. A hydraulic link exists between cold and warmed fractured unconfined waters formed in gneisses (zone of regional fracturing) and confined waters containing in the upper zone of the marble body. The rainfall influences on the variation of water level in the whole massif. When cold waters penetrate in depth the thermal water piezometric level becomes deeper as a result of cooling.

Pumping tests have been conducted in many of the wells in the studied area. The hydrogeological parameters of the aquifers vary widely: transitivity **(T)** is from 60 to more than 2 000 m^2/d, hydraulic conductivity is from less than 5 to more than 100 m/d. Cavernous quartz sediments are formed as a result of complicated interactions between thermal and cold waters, temperature barrier existing on the sectors of border zone between warmed marbles and cooler gneisses and along some faults dislocations. Simultaneously, in the scope of the hydrothermal reservoir are formed metasomatic polymetallic ores, which are subject to underground mining.

Numerous mining galleries, exploitation horizons, mine and ventilation shafts, including the main shaft to a depth of 650 m (the deepest in the Rhodopes massif) were built. Their construction was done under extreme high temperatures up to 70^{o}C, hazardous geological, mining and hydrogeological conditions. Before ore exploitation a lowering of water level in the heated marble body was done. This process reflected on the decreasing of water quantity of the Thermos spring, located in Greece in the valley of Ilidza river. The natural flow of this spring is about 50-60 l/s. The link between them is realized through a fault.

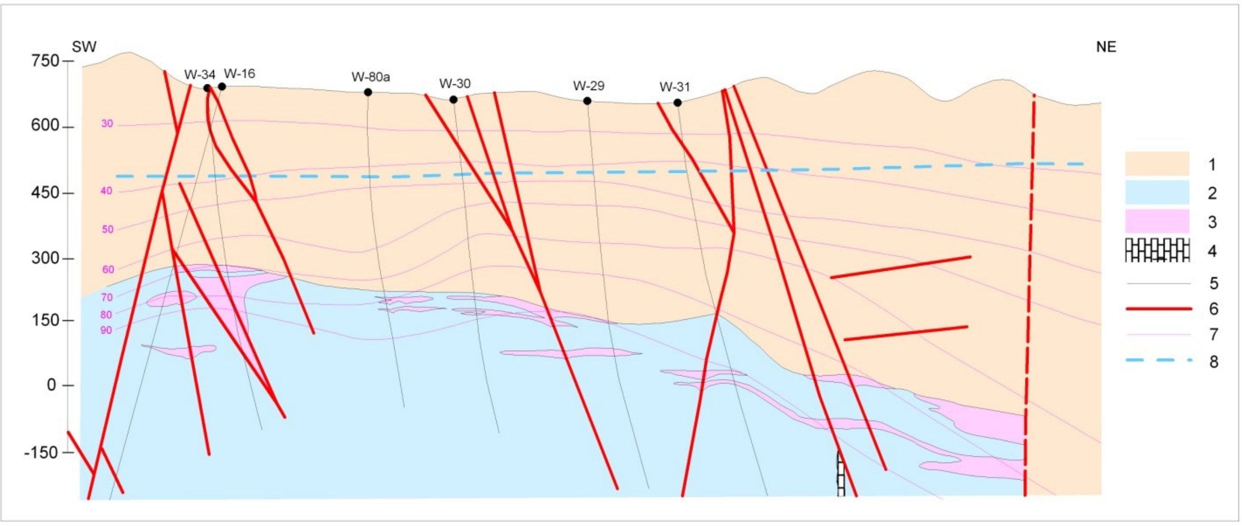

Figure 2: Hydrogeological and geothermal profile along Erma Reka hydrothermal reservoir

Legend: 1. amphybolites, gneisses; 2. marbles; 3.quartz zones and ore zones; 4. caverns; 5. geological boundaries; 6. faults;. 7. temperature isolines, ^{o}C; 8. piezometric water level (m); W-29 wells

3. HYDROCHEMICAL CHARACTERISTICS

Since 1955 till present many water samples have been taken from different sources for chemical analysis: surface water, mine water and water from well intersecting cold and hot water horizons. Some of the sources have been analyzed only once and others several times. Major problem for hydrochemical data interpretation was that temperature and some of the main components characterizing the chemical composition have not been determined in the past. For this reason are processed and analyzed the data obtained for 110 water samples, including a sample taken on 22.8.2002 from Thermos spring, Greece. Its water temperature is 48^{o}C and TDS - 1378 mg/l.

Water temperature and TDS vary within Erma Reka reservoir, respectively in the range of 10 to 89°C and 79 to more than 1646 mg/l depending on the geological location and the type of water source, Fig.3.

Surface waters and waters accumulated in the gneisses overlying marbles and not influenced by them are characterized by low temperature and mineralization. TDS measured for thermal water samples taken from wells vary between 700 and 1646 mg/l. For all of them the temperature is higher than 40°C. The water temperature of the rocks overlying the marbles and those from the zones where cold and hot water are mixing is lower than 40°C and TDS is reduced.

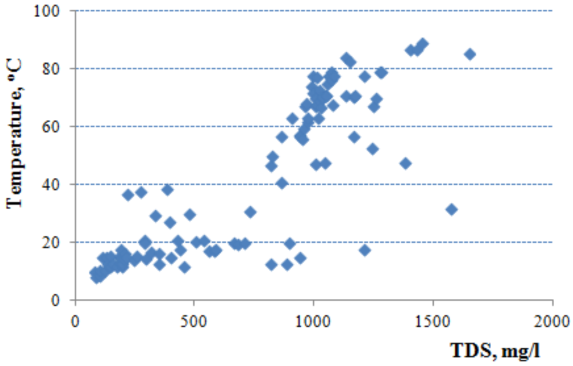

Figure 3: Relationship between temperature and TDS

The prevailing ions are related to the type of water source and depend on the temperature and TDS values, Fig.4. Some ions, like HCO_3^-, Ca^{2+}, Na^+ are typical for cold water, while HCO_3^-, SO_4^{2+}, Na^+ - for thermal water. Similar results are obtained for the Thermes thermal spring in Greece. With few exceptions, mixed and mine water are characterized by intermediate values. The content of chloride and magnesium is less representative. Higher magnesium content is registered only in single samples taken from surface sources. Two groups are formed based on the analysis of main anions HCO_3^-/ SO_4^{2-} (Fig. 5) and cations Ca^{2+}/ Na^+ ratio (Fig. 6).

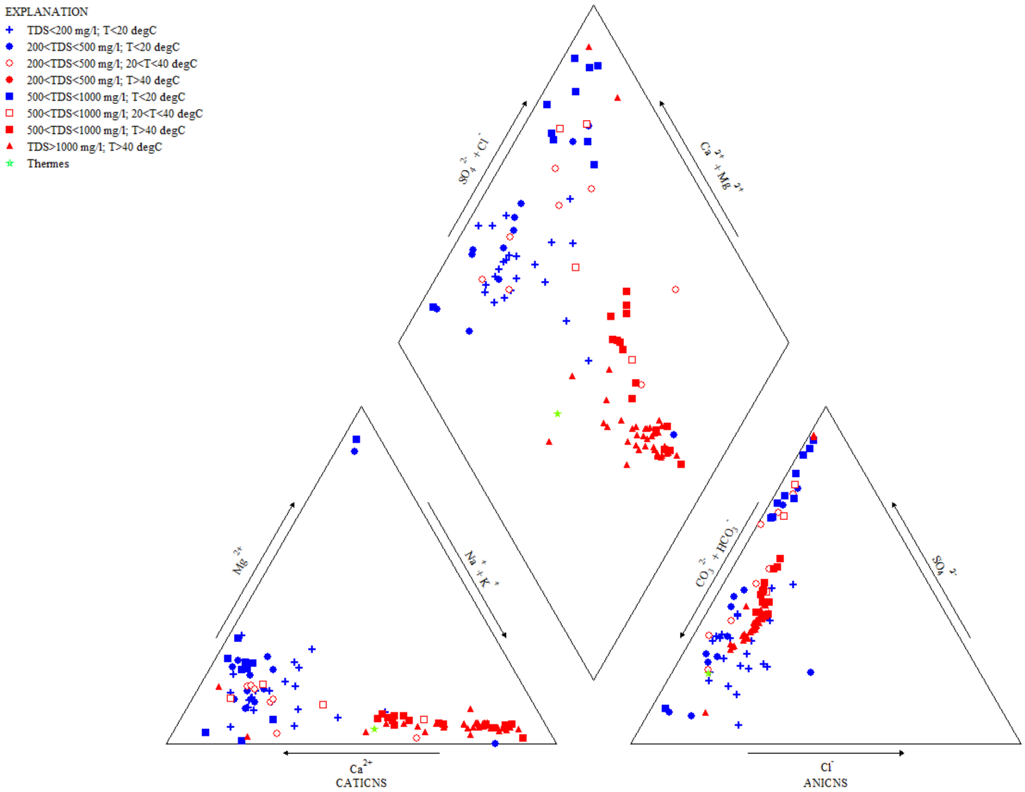

Figure 4: Piper diagram for Erma Reka reservoir

Regardless of the elevated TDS for thermal waters of temperature above 40°C sodium preserves its leading role, while SO_4^{2-} and HCO_3^- ions mark a slight increase.

The ratio of main ions defined for waters of temperature less than 40°C – surface, cold ground waters, and cooled mine water in the shafts are characterized by great variety.

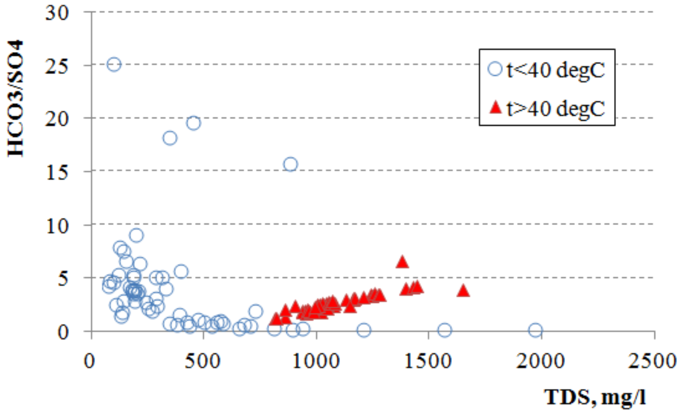

Figure 5: Relationship between HCO_3^-/ SO_4^{2-} and TDS

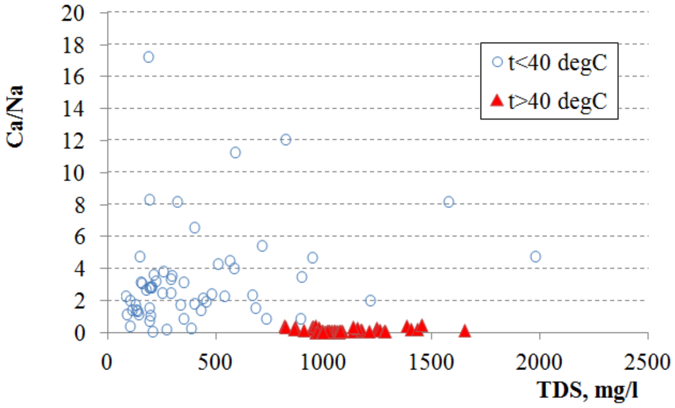

Figure 6: Relationship between Ca^{2+}/Na^+ and TDS

Similar groups are formed for one of the main thermal water component - H_2SiO_3 (Fig.7). A trend of growing H_2SiO_3 content proportional of TDS increasing is observed for thermal water. The concentration of the same component is changing in a wide interval for cold, mixed and cooled thermal water not exceeding 50 mg/l. High H_2SiO_3 concentration is prerequisite for the existence of quartz deposition in the reservoir.

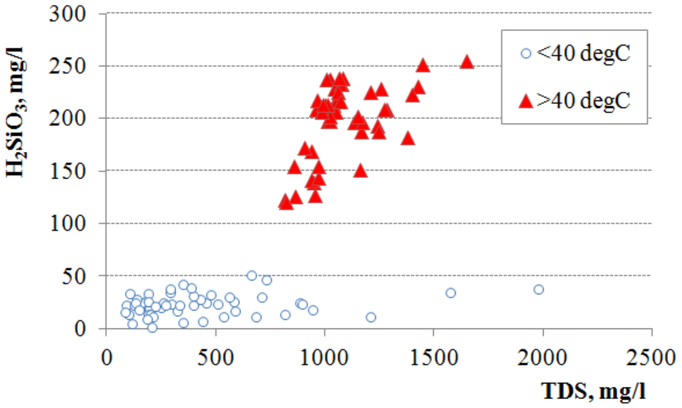

Figure 7: Relationship between H_2SiO_3 and TDS

The variation of intervals characterizing some hydrochemical indicators for thermal water containing in the marble body are studied. Between 5 and 10 water samples are taken from most of the wells during long lasting pump tests. The obtained results show that the concentration changes are in comparatively small interval for most of the them, Fig.8. More significant variations are observed only in the zones where cold waters penetrate the marble body.

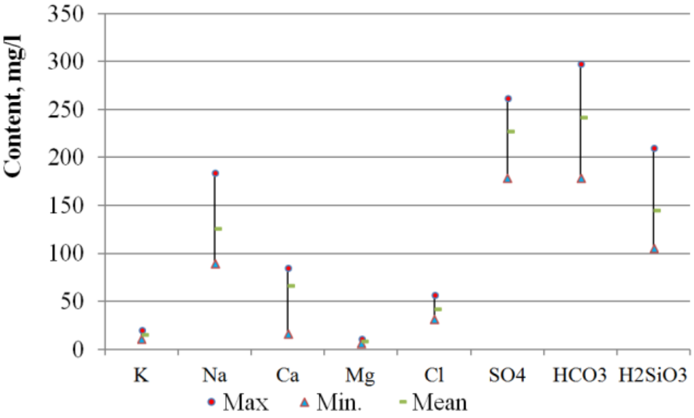

Figure 8: Intervals of main ions variation in well, discovered thermal water

The concentration of micro components in water samples forming the two groups (temperature above and below 40°C) differs (Table 1). Some of them Ga, Ge, Li, Ti, Mo, V increase their concentration in hot water, which is associated with ascending water from the deeper part of the reservoir. Other components like Mn, Ni, Pb, Sr, Zn, Cr, Zr, Cd exhibit higher concentrations in cold, mixed and cooled thermal waters. They are probably coming as a result of interaction with the host rocks or from waters passed through ore mineralization. No significant change in Ag, Al, As, Ba, Cu, Fe, Co concentrations in terms of water temperature is established. Enhanced F concentration up to 8 mg/l is observed in single samples taken from hot water sources.

The incoming H_2S and CO_2 from depth play an essential role for the formation of gas composition. The water-rock interaction and the process of mixing waters of different temperature and composition also increase gas content. The quantity of CO_2 reaches about 140-150 mg/l in thermal water sources. Due to the high hydrostatic pressure in depth CO_2 is expected to be in liquid phase.

4. MINERALIZATION

Complex hydrogeological conditions, the presence of geological and thermal barriers, mixing waters of different genesis and anthropogenic impact create prerequisites for various physicochemical processes, including elimination of some substances dissolved in water and deposition of others. Such processes have been observed in the area of Thermes spring (Greece) where thermal water is discharged, Fig.9.

Figure 9: Mineral depositions in the region of Thermes spring, Greece

Quartz-cavernous areas in Bulgarian territory are formed at the contact of the heated marbles and cold gneisses, amphibolites and other metamorphic rocks, chilled by cold water infiltration. The polymetallic ores exploitation has led to significant anthropogenic interference associated with thermal waters drainage.

Well W-29 is an example of a rapid sedimentation process led to formation of a thick no homogeneous depositions along the casing as a result of water table lowering and pump tests, Fig.2.

Erma Reka travertine depositions are composed of almost pure $CaCO_3$, which occur as calcite or aragonite. They occur as encrustations or incrustations from thermal waters of W-29. The concentric incrustations from a drill pipe was examined in order to represent existing sequences.

The incrustations are composed by several concentric layers with size from 1-2 mm to ~1.5 cm in width. They are formed of alternations of porous, earthy accumulations and crystalline crust. In general the earthy layers are in the outer part of the sequence while the crystalline formations are developed mainly in the inner part. The XRD analyses showed that mineralogical composition of the crystalline crust is pure calcite while porous masses are aragonite (~82%) + calcite (18%).

The dense porous sections represent bundles of small elongated aragonite crystals (Fig.10 a, d). The crystalline crust is formed by spherulitic aggregates which are shaped by larger terminations. SEM observations show that spherulites 0are composed by distorted and skeletally developed rhombohedral crystals (Fig. 10). On places, side faces of some individuals are striated or equivalent faces in other are affected by dissolution (Fig. 10 c). On the boundaries between calcite crust and aragonite areas are observed complex intergrowths of non regular developed calcite individuals, aragonite needles as well rhombohedral single crystals.

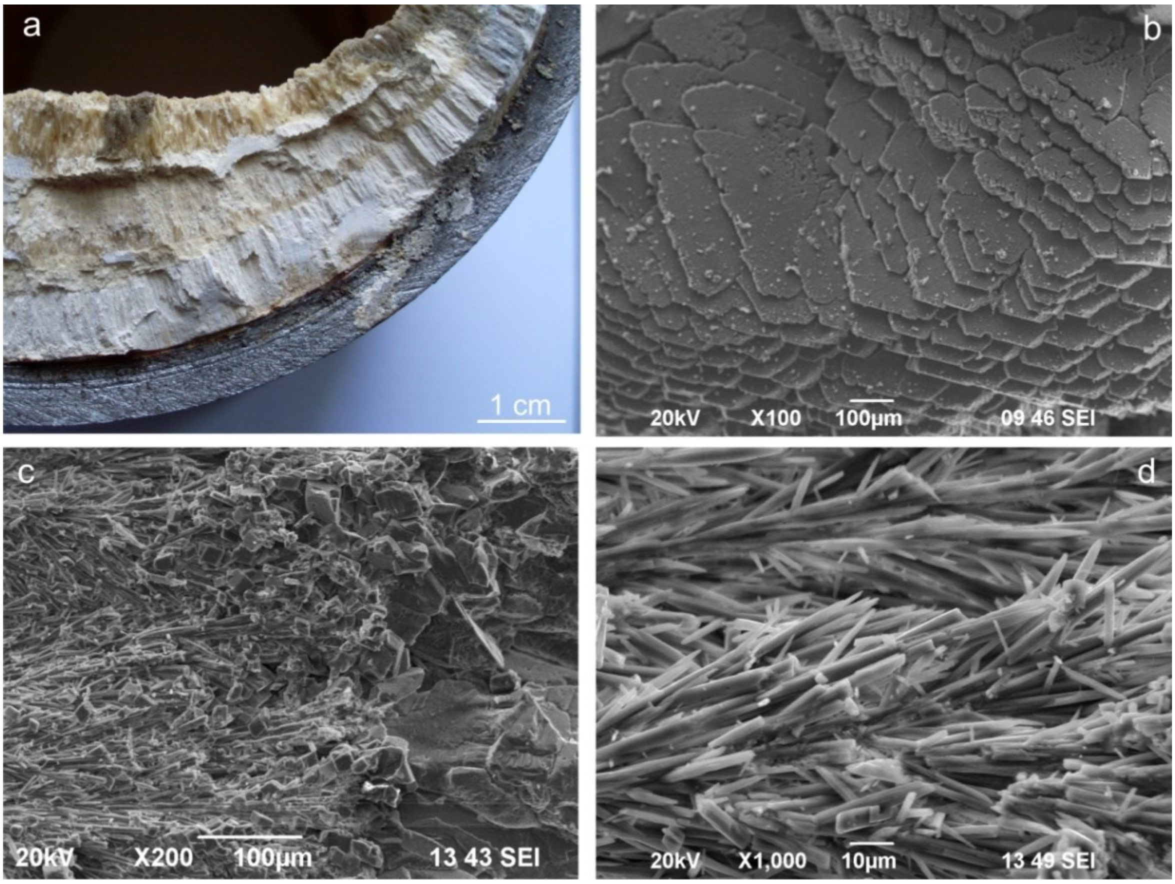

Figure 10: a) cross-section view of radial incrustations of a pipe; b) outer terminations of calcite aggregates; c) alternation of fibrous aragonite and distorted calcite crystals and sporadic rhombohedral crystals; d) close view of aragonite elongate needles, b-d) SEM

SEM-EDS determinations revealed small amounts of Mg (up to 0.80 wt.%), Na (up to 0.60 wt.%), and Fe (up to 1.40 wt.%) as impurities in carbonates.

5. DISSCUSION

Travertines occur as calcite or aragonite depending on numerous controlling factors as temperature, Fouke et al. (2000), chemical composition (Mg/Ca ratio) of thermal water, Folk (1994), saturation rate, Buczynski and Chafetz (1991), pCO_2 and rate of CO_2 degassing, Kele et al. (2008) and others. Friedman (1970) pointed out that aragonite usually occurs around thermal springs, where the temperature ranges from 30°C to 60°C, whereas Folk (1994) and Fouke et al. (2000) noted that aragonite would precipitate, if the water temperature is above 40°C, and calcite would form, if the water is rich in Ca and cooler than 30°C. Calcite can, however, precipitate directly from waters with temperatures >90°C in Kenya (Jones and Renaut, 1995) and in New Zealand, Jones and Renaut (1996). The Mg/Ca ratio of thermal water has an effect on the formation of aragonite, Folk (1994) and according to Fischbeck and Müller (1971) the aragonite precipitation becomes significant when the Mg/Ca ratio is ~2.9.

Saturation indexes of the waters are calculated using the Visual MINTEQ 3.0 software and databases. Visual MINTEQ 3.1 is used for the SI calculations. Visual MINTEQ is a freeware chemical equilibrium model developed by Jon Petter Gustafsson at KTH, Sweden (*http://www2.lwr.kth.se/English/Oursoftware/vminteq/index.html*). The following database files which are part of Visual MINTEQ software are used: comp_2008.vdb – for components; thermo.vdb - constants, enthalpies etc. for all aqueous, diffuse-layer, solid, redox and gas species; type6.vdb - enthalpies etc. for the solid phase species. The real temperature conditions, pH and concentrations for the each water sample are imputed.

The saturation index (SI) is calculated by comparing the chemical activities of the dissolved ions of the mineral (ion activity product, IAP) with their solubility product (Ksp). In equation form, SI = log(IAP/Ksp). The most important results of speciation calculations are saturation indices for minerals, which indicate the saturation state of each mineral relative to the water.

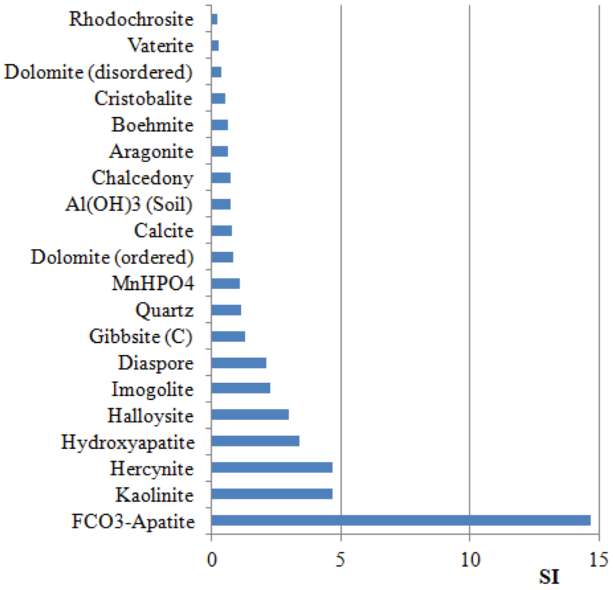

Figure 11: Mineral phases with positive saturation index values (SI) for Thermes spring, Greece

Positive values of saturation index are obtained for most of the mineral phases in the zone of natural draining in Thermes, Greece, Fig. 11. Carbonate sediments are result of CO_2 transition from gas to liquid phase at surface conditions. Carbonate depositions in well W-29 have been formed after three pump tests in the upper part of the water saturated marble in three consecutive years. Different TDS values are measured for samples taken during the three pump tests. They are mostly between 700 and 800 mg/l for the first and third pump test and higher – between 1000 and 1100 mg/l – for the second test. The calculated equilibrium index for waters of different TDS shows mostly similarity and small differences, Fig.12 .

Although the equilibrium index calculated for W-29 is positive for more than 20 mineral phases, most appropriate conditions for deposition are for carbonate minerals. This is due to airlift technology used during the pump test.

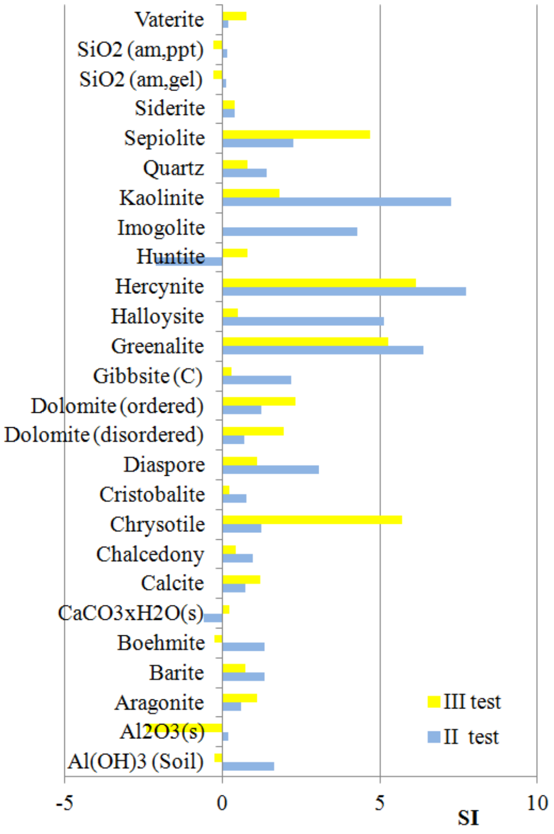

Figure 12: Mineral phases with positive values of saturation index (SI) for well W-29

The injection of compressed air into the aquifer results in release of dissolved CO_2 from thermal water and creates a prerequisite for mineral deposition. The saturation index for samples taken from water level lowering system is calculated. Most of the values are close to those obtained for W-29 and also some positive index values are defined for some phosphorus containing mineral phases - $Ca_4H(PO_4)_3{:}3H_2O$, chloropyromorphite and cuprite. The water level lowering system is based on a shaft at 300 m level and drains the zone around large karst caverns where the hottest (about 90°C) thermal water is accumulated.

Different factors control the deposition of oversaturated water with respect to corresponding mineral phases. Simulated is the influence of the temperature decreasing on the saturation indices. One of them is a temperature change taking place in the upper part of the geothermal zone. The hot water becomes unsaturated at temperature below 50-60°C as regards aragonite and calcite and is permanent saturated as regards quartz, Fig.13.

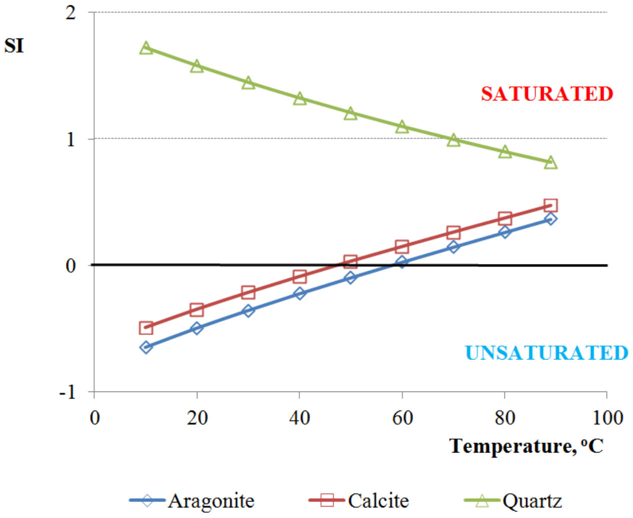

Figure 13: Temperature impact on saturation index

The values of saturation index of various compounds regarding mineral phases show that most significant risk is associated with the deposition of carbonate materials, particularly in the areas of abrupt temperature change and water contact with atmospheric air. It has been established that various carbonate, phosphate and silicate mineral phases could be deposited in the process of cooling down.

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Table 1: Micro components for different types of water, mg/l

Elements	Water of temperature below 40^{o}C (58 samples)				Water of temperature above 40^{o}C (52 samples)			
	Typical values			Number of samples	Typical values			Number of samples
	Average	Min	Max		Average	Min	Max	
Elements of low concentration variation								
Ag	0.0003	0.0001	0.0017	18	0.0010	0.0008	0.0018	13
Al	0.787	0.019	4.795	58	0.426	0.012	2.787	52
As	0.027	0.007	0.070	18	0.084	0.080	0.088	2
Ba	0.341	0.007	3.470	58	0.250	0.077	0.540	50
Cu	0.0041	0.0003	0.0270	57	0.0052	0.0022	0.0240	36
Fe	0.104	0.003	0.565	58	0.122	0.012	1.500	52
Elements, decreasing their concentrations in warm waters								
Mn	0.9847	0.0003	10.0980	58	0.0353	0.0024	0.4100	52
Ni	0.0032	0.0001	0.0650	39	0.0016	0.0008	0.0030	35
Pb	0.0253	0.0003	0.8960	54	0.0043	0.0005	0.0200	32
Sr	1.093	0.005	19.490	57	0.416	0.056	1.660	52
Zn	0.655	0.003	8.340	31	0.147	0.024	1.031	21
Cr	0.0080	0.0009	0.0210	3	0.0020	0.0020	0.0020	1
Zr	0.0017	0.0003	0.0080	15	0	0	0	0
Cd	0.037	0.001	0.093	8	0	0	0	0
Co	0.031	0.004	0.065	5	0	0	0	0
Elements, increasing their concentration in warm waters								
Ga	0.0007	0.0003	0.0021	12	0.0143	0.0008	0.0550	4
Ge	0.0001	0.0001	0.0001	1	0.0307	0.0010	0.0640	49
Li	0.819	0.076	6.560	11	7.186	1.030	10.000	44
Mo	0.0039	0.0001	0.0760	39	0.0200	0.0002	0.2640	50
Ti	0.133	0.003	1.683	58	0.305	0.025	1.069	48
V	0.0029	0.0003	0.0190	43	0.0319	0.0024	0.0870	48