WATER ISOTOPE GEOCHEMISTRY AS TOOLS FOR THE EXPLORATION OF A LOW-ENTHALPY COMPLEX RESERVOIR: THE CASE OF THE GEOTHERMAL FIELD OF HAINAUT (BELGIUM – NW EUROPE)

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Keywords: geochemistry – isotopes – karstic reservoir – low enthalpy – Belgium

INTRODUCTION

The geothermal reservoir of Hainaut is a mainly carbonated fissured and karstic aquifer, composed by Lower Carboniferous limestone and dolostone. It is located in the Mons region (see Figure 1), near the French border.

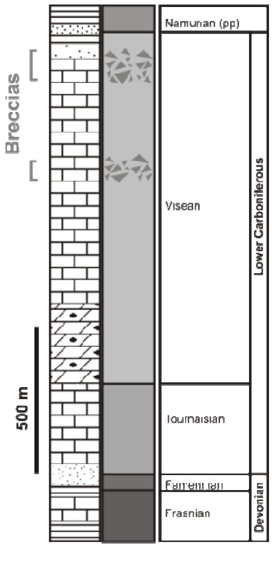
Figure 1 : Localization of the geothermal field of Hainaut

This geothermal resource was first discovered in 1976 thanks to a geological exploration borehole in Saint-Ghislain (Delmer, 1977). It encountered massive layers of anhydrite in the Upper and Middle Visean carbonate, overlying a thick productive layer of karstic breccias at 2.500 m deep. This borehole, that was turned into a production well ten years later, is the starting point of a district heating system. It gives 73°C hot water at an artesian flow rate of 100 m³/h.

Two other wells were drilled short after the Saint-Ghislain well, in Douvrain and Ghlin, a few kilometers North and North-East. The first one is exploited for heating buildings and sanitary water of a nearby hospital. The second one is still inactive. These two wells produce respectively 67°C and 71°C water, at 1.370 m and 1.575 m deep, from Upper Visean breccia. The flow is artesian as well as in Saint-Ghislain, and reaches the same value (100 m³/h). Another geothermal well drilled in 1985 in Condé (France), a few kilometers off the border, produced water at 29°C from 1.240 m deep, far from the expected temperature of 50°C. This heterogeneous temperature distribution demonstrates the necessity of investigating flow patterns in the deep reservoir before any new exploitation prospect.

Presently, the exploitation scheme of the reservoir is extremely simple. The two exploited wells draw water from the depths, use heat in a serie of heating applications, and then discharge cooled water to the hydrographic networks, thanks to its low salinity (less than 2g/l). Recent development programs involve present networks extension and new drilling works. Further investigations have to be led in the aim of avoiding other failure as it occurred in Condé and predicting the limitations of the single-well technique, amongst others.

Figure 2 : Reservoir lithostratigraphy



The reservoir is thought to be recharged from infiltrating meteoric waters in the outcropping area. It has no natural exsurgence, except a few springs showing anomalies in temperature and/or chemical content. In the aim of assessing the influence of deep water on shallow water characteristics an extensive study has been made to gather chemical and isotopic data on both deep and shallow waters of this aquifer.

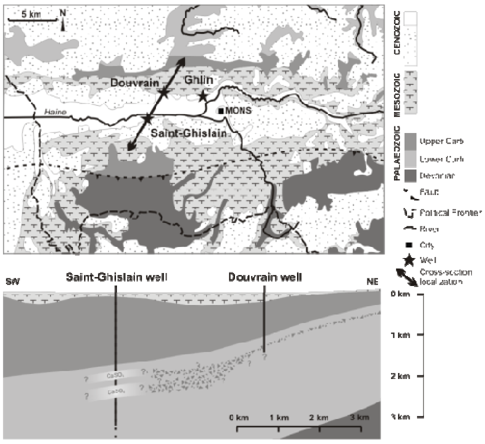
GEOLOGY OF THE GEOTHERMAL RESERVOIR OF HAINAUT

The rocks composing the geothermal aquifer are mainly Lower Carfoniferous carbonates. These strata outcrop at the North of the geothermal exploitation area, and the lithostratigraphical sequence is quite well known. Figure 2 gives a schematic log of the aquifer formations, as they have been described in Doremus and Hennebert (1995), and Hennebert (1999).

The aquifer includes Famennian and Upper Frasnian permeable strata. These formations show different lithologies, including sandstones and thin shale layers. Tournaisian rocks are mainly limestones. Visean strata often show dolostones in the lower part, then limestones containing several breccia layers. Phtanites are the uppermost rocks considered that are part of the aquifer.

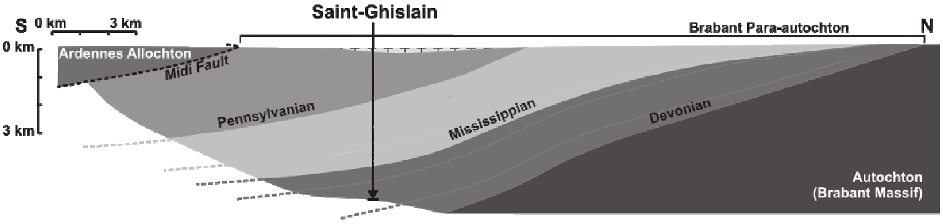
In the Mons region, the aquifer strata have an E-W direction and show a little dip of 10 to 15 degrees Southward. They can be found under Meso-Cenozoïc formations of the structure called “Mons Basin” and Pennsylvanian coal-bearing rocks that were intensively mined during the last two centuries. Figure 3 illustrates the geological context of the geothermal reservoir.

Figure 3 : Geological map and schematic cross-section of the exploited area



Palaeozoïc formations are shaped by Hercynian orogeny. The results of this influence are represented on figure 4. Palaeozoïc strata are divided in several tectonic units. The Ardennes Allochton overlays the Para-autochton unit, composed by Carboniferous and Devonian formations. The Para-autochton lies on the Cambro-Silurian autochton Brabant Massif.

Figure 4 : Schematic North-South cross-section



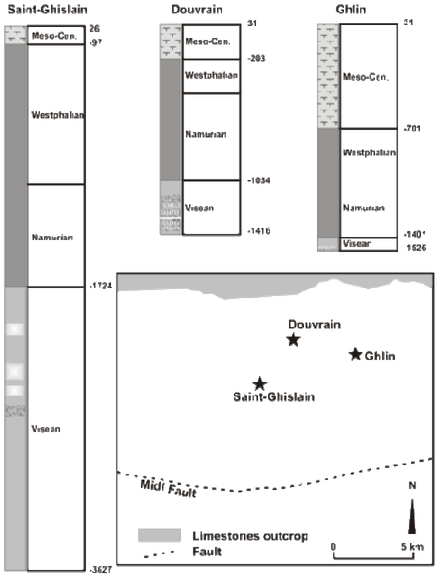
Hercynian tectonic only left rare traces on the reservoir strata, which show few faults and folds, but the overlying coal-bearing formations of Upper Carboniferous are heavily affected by thrust-faults. The Midi fault is the last of these numerous faults, and separates the Allochton from the Para-autochton unit.

The opening of the reservoir to meteoric waters is supposed to have taken place in Late Jurassic/Early Cretaceous times and to be related to fracturation of the carbonates under extensional tectonic regime (Quinif *et al.*, 1997). Penetration of fresh waters in the deep massif has induced anhydrite dissolution, which is one of the supposed origins of the basin-shaped top Palaeozoïc surface, where Cretaceous sediments have been conserved in the present Mons Basin (Dupuis and Vandycke, 1989).

The Saint-Ghislain well revealed thick anhydrite layers in Middle and Late Visean series, as described in Groessens *et al.* (1979) (see Figure 5). These anhydrites seem to correspond to breccia layers that appear at the outcrop and in the others geothermal wells of Douvrain and Ghlin. Anhydrite and gypsum pseudomorphs were found in most of these breccias, and indicate the contribution of anhydrite removal to their origin, though they may not be pure collapse breccias and be the result of several phenomena (Rouchy *et al.*, 1993; De Putter *et al.*, 1994).

The productive layer was crossed in Saint-Ghislain at a depth of 2.500 m, under the massive anhydrite strata, in a highly conductive

Figure 5 : Geological succession in the geothermal wells



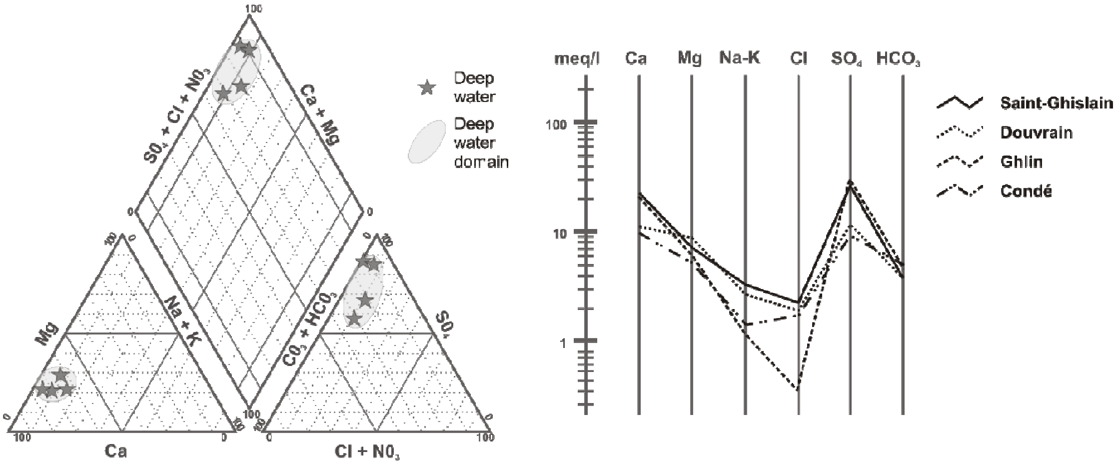
and probably karstified breccia. In Douvrain and Ghlin, large fissures in Upper Visean breccia give most of the hot water flow. Permeability and breccia layers related to anhydrite dissolution therefore seem strongly linked. On the other hand, natural springs of the Southern margin of the shallow aquifer, than issuing from to Upper and Middle Visean, tend to show temperature exceeding normal (10 – 12 °C), sometimes associated with high sulfate content.

The hypothesis has already been expressed in Delmer *et al.* (1982) that these springs can be influenced by deep water upflow. The investigations described below notably aim in confirming this hypothesis.

DEEP AND SHALLOW WATER GEOCHEMISTRY

Available data on Lower Carboniferous aquifer waters come from two main origins: shallow waters from springs, water catchment and quarries pumping on one hand, and, deep waters from the geothermal wells on the other hand.

Figure 6 : Piper and Schöeller-Berkaloff diagrams for geothermal water

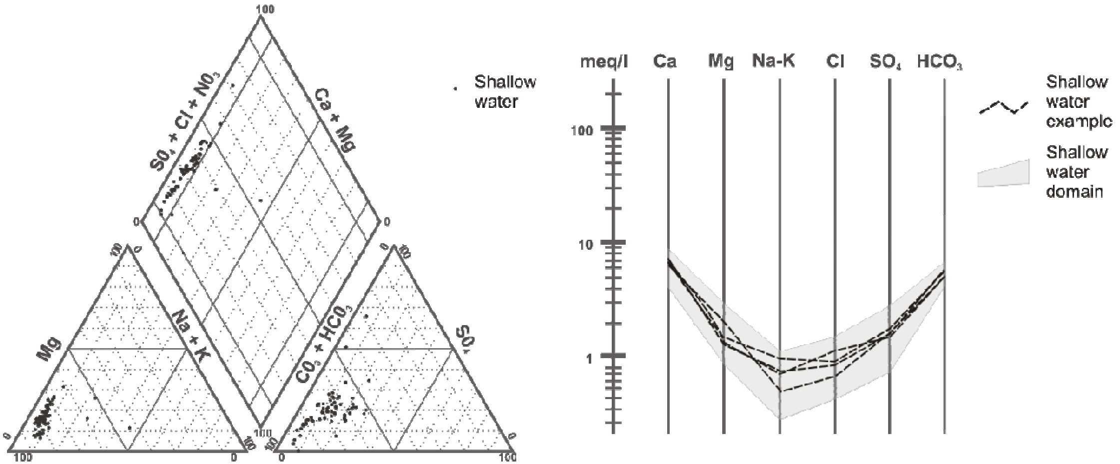


The two active Hainaut geothermal wells waters are regularly sampled and analyzed since they were set in use. The third well has recently been opened for a several month flow test, and the water chemical content has then been analyzed. For the Condé well, data come from several unpublished reports.

Deep water, as illustrated by Figure 6, have a moderate chemical content, which does not exceed 2 g/l. Anhydrite leaching influence is obvious, with a strong sulfate content.

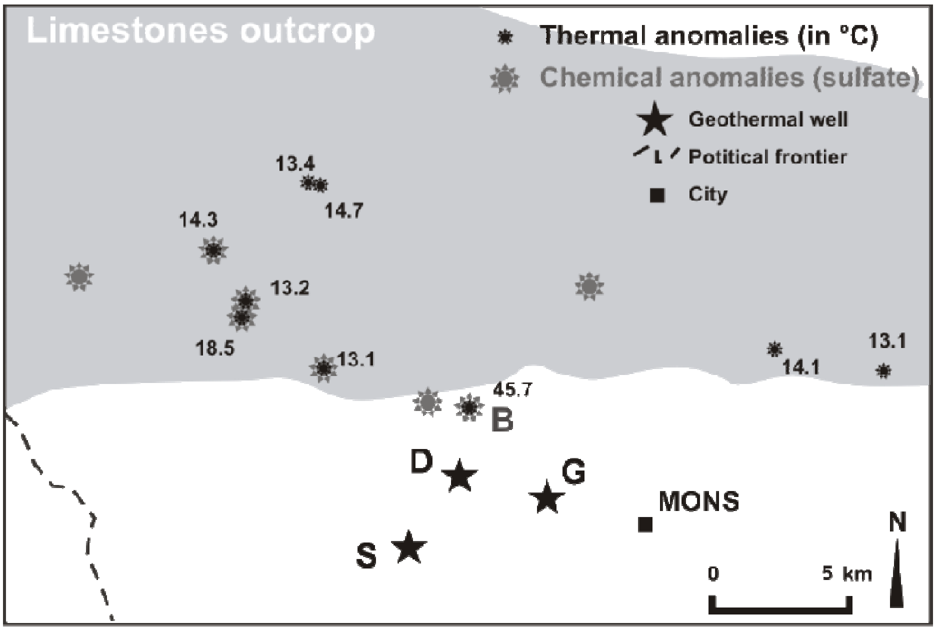
Shallow water can come from different lithologies: Limestone, dolostone, shaly limestone, sandstone… The calcium and carbonate influence is often well expressed (see Figure 7), with variations in magnesium content depending of the localization of the sampling point in relation with geology. Sulfate content can be quite heterogeneous, from 20 to 120 mg/l, but remains most of the time between 70 and 90 mg/l, which is normal for water issuing from these quite pyrite-rich formations.

Figure 7 : Piper and Schöeller-Berkaloff diagrams for shallow water



The normal temperature of shallow waters never exceeds 12°C. However, several natural spring mainly located at the Southern margin of the outcropping area show anomalies in temperature (over 13 °C), and/or in sulfate content (over 100 mg/l). These springs are represented on figure 8.

Figure 8 : Localization of Southern margin springs

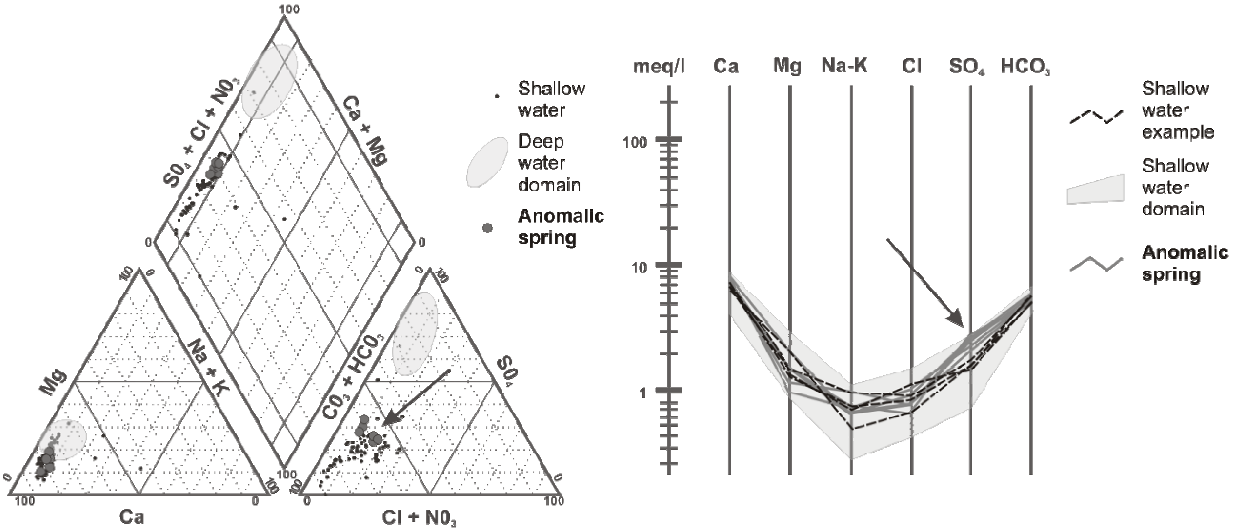


One of these spots show a temperature of more than 45°C, North from Douvrain well (marked “B” on figure 8). It represents the Baudour galleries, drilled at the beginning of the twentieth century by miners to reach coal seams without having to deal with Cretaceous aquifer, located above the coal basin. These galleries had to be abandoned because of hot water coming from fractures in Namurian sandstones. The initial temperature of the water was 53°C, at 300 m deep. Presently, water level is equilibrated with shallow aquifer piezometric level, and its temperature still reaches 45°C.

The springs represented on figure 8 have been sampled in the aim of confirming their chemical content (see Figure 9). Their sulfate content appears to be high, compared with the average content of shallow waters. The origin of this sulfate can be found in several origins. The two main suspected sulfate sources are anhydrite leaching and pyrite oxidation.

Temperature anomalies seem to indicate deep origin for some of these spring waters, and isotope analyses can validate or invalidate this hypothesis. Confirmation would implicate convection loops bringing hot and sulfated water in contact with fresh water at the Southern margin of the reservoir, along Upper Carboniferous impervious sediments, and sometimes reaching surface through them thanks to fractures, as in the Baudour galleries.

Figure 9 : Piper and Schöeller-Berkaloff diagrams for anomalic springs



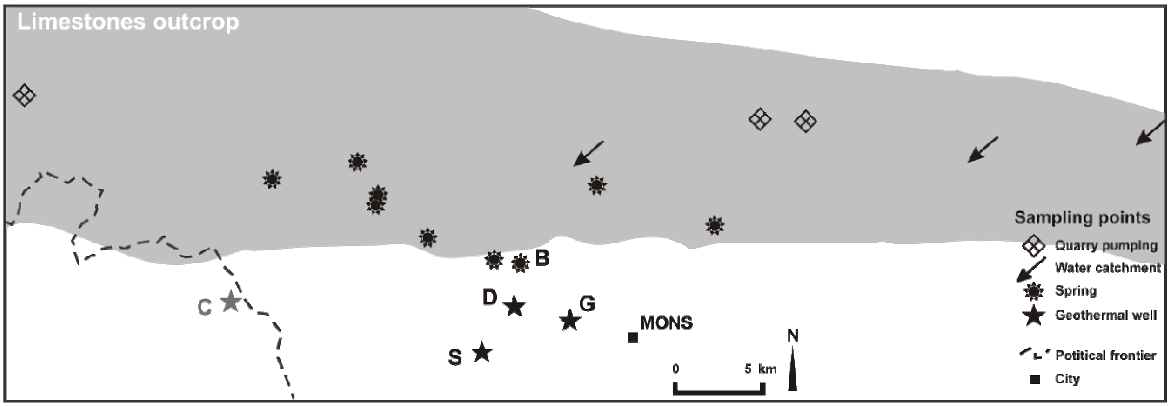
ISOTOPE INVESTIGATIONS IN LOWER CARBONIFEROUS RESERVOIR

Sampling points for isotope characterization of both shallow and deep Lower Carboniferous reservoir have been chosen as illustrated on Figure 10. Three main groups are represented :

1- quarry and water catchment pumpings, as reference of regular shallow water,
2- geothermal wells,
3- anomalic springs already mentioned.

The Condé geothermal well is located on this figure in a different grey tone because isotopical data that will be used below come from literature and not from recent sampling and analyses.

Figure 10 : Localization and type of sampling points for isotope analyses



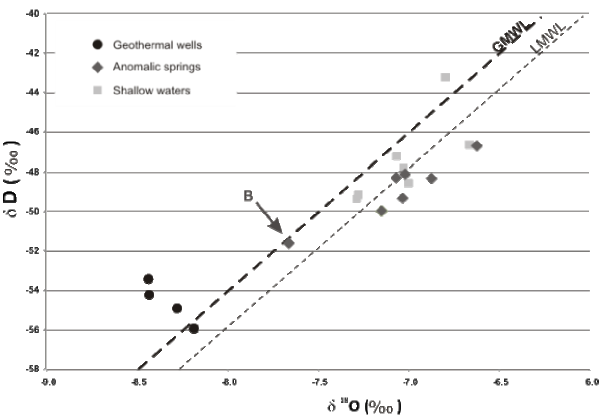
Stable isotopic ratios have been measured for deuterium and ^{18}O in water, ^{13}C of DIC in bicarbonate, and ^{34}S and ^{18}O of sulfates (except for ghlin well and Baudour galleries). The results are given in table 1. ^{14}C content has been analyzed for Saint-Ghislain and Douvrain geothermal waters.

Table 1: Isotopic content of deep and shallow water of the Lower Carboniferous reservoir

Locality	‰	H_2O $\delta^{18}O$ ± 0.20	δD ± 2	HCO_3^- $\delta^{13}C$ ± 0.20	HCO_3^- mg/l	$SO_4^=$ $\delta^{34}S$ ± 0.20	$\delta^{18}O$ ± 0.20	$SO_4^=$ mg/l	t° ℃
Saint Ghislain	**PG - 01**	-8.44	-55.35	-3.11	188.3	17.10	16.70	1316.0	65.5
Douvrain	**PG - 02**	-8.20	-57.47	-4.85	209.9	17.90	16.30	575.9	55.4
Ghlin	**PG - 03**	-8.29	-56.20	-3.60	274.7			1402.1	710
Condé	**PG - 04**	-8.50	-54.60	-7.60	303.0	14.80		440.0	27.9
Baudour	**B - 01**	-7.67	-52.05	-6.90	223.5			660.0	45.7
Stambruges	**SAG - 01**	-7.04	-49.13	-11.43	289.8	-0.20	9.90	129.8	18.5
Stambruges	**SAG - 02**	-7.08	-47.81	-12.18	305.0	-5.80	7.50	119.2	13.3
Wadelincourt	**SAG - 03**	-6.63	-45.81	-13.88	347.7	-4.20	6.10	120.7	11.1
Sirault	**SAG - 04**	-6.88	-47.86	-13.62	305.0	120	8.90	94.3	13.1
Beloeil	**SAG - 05**	-7.16	-49.93	-12.41	329.4	-6.80	7.20	113.7	14.3
Jurbise	**SAG - 06**	-7.03	-47.52	-14.11	311.1	-7.10	6.10	103.3	10.9
Feluy	**PCCN - 01**	-7.03	-47.19	-11.51		-5.10	5.00	718	
Lens	**PCCN - 02**	-7.07	-46.50	-13.35	340.3	-6.00	6.50	75.1	12.3
Buzet	**PCCN - 03**	-7.30	-48.96	-13.39	254.1	-3.60	170	77.6	10.6
Spy	**PCCN - 04**	-7.29	-49.01	-12.09	347.0	-3.90	6.80	77.1	12.5
Antoing	**PCCN - 05**	-6.80	-4142	-13.76	355.0	-4.40	4.60	183.7	
Soignies	**PCCN - 06**	-6.67	-45.74	-9.35	327.6	-12.10	-120	595.6	
Soignies	**PCCN - 07**	-7.02	-48.11	-11.60	342.8	-7.00	170	85.5	12.1

Deuterium and ^{18}O content in sampled water are represented on Figure 11. The Global Meteoric Water Line (GMWL) according to Craig (1961) and Local Meteoric Water Line (LMWL) are illustrated as well. The d-excess of LMWL has been chosen to fit with experimental results, and is consistent with GNIP data (IAEA, 2001), which give a d-excess value of 8 to 9 in the area.

Figure 11 : δD and $\delta^{18}O$ vs. SMOW



Meteoric origin of deep waters is confirmed. Heavy isotope depletion of deep water compared to that of shallow water suggests infiltration under cold climate.

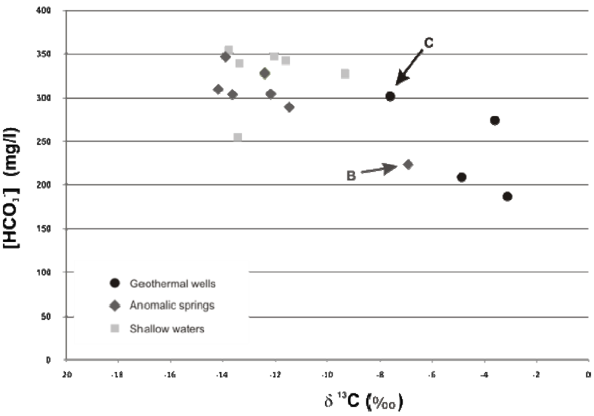
The "B" marker represents the Baudour galleries. Its intermediate position between geothermal water and shallow water confirms a very strong contribution of deep water. Figure 11 shows that the other springs isotopic contents are too close to that of regular shallow water, forbidding any distinction between them.

$\delta^{13}C$ in bicarbonates is represented on Figure 12, compared to HCO_3^- concentration. DIC in water can come from several origins. Carbon resulting from dissolution of the aquifer carbonated matrix ($\delta^{13}C$ between 0 and +2 ‰) would show isotopic ratios between -0.7 and -2.7 ‰. Atmospheric CO_2 dissolved in water would be characterized by ratios between -15 and -17 ‰, depending on temperature (Mook, 2000).

Geothermal water appears to be in equilibrium with carbonated matrix, except for Condé water ("C" marker), of which low ratio suggests mixing with fresh waters. The group of shallow waters shows ratios between -11 and -14 ‰, in good coherence with a situation where water is in contact with both atmosphere and carbonated rocks. Here again, the Baudour water ("B" marker) tends to join geothermal water area, and reveals a strong influence of carbonated aquifer.

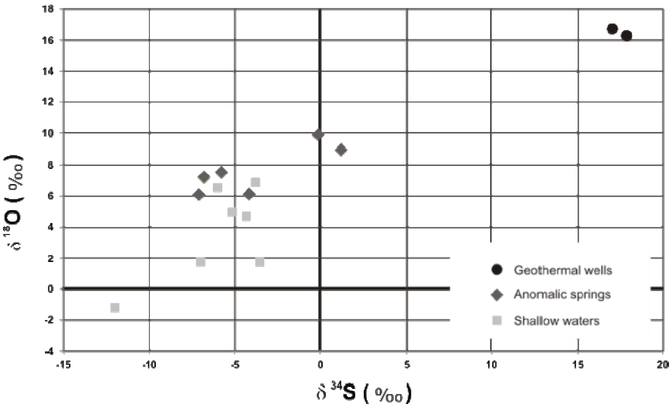
^{14}C content has been measured on HCO_3^- in Saint-Ghislain and Douvrain deep waters. The residence time resulting from different interpretations ranges from 35.000 to 21.000 years

Figure 12 : $\delta^{13}C$ vs PDB and HCO_3^- concentration



for Saint-Ghislain and from 20.000 to 8.000 for Douvrain (Ingerson and Pearson, 1964; Vogel, 1970; Tamers, 1975). This residence time is consistent with geothermal water depletion in ^{18}O and D, as they date infiltration back to the last ice age.

Figure 13 : $\delta^{18}O_{SO4}$ vs SMOW and $\delta^{34}S_{SO4}$ vs CDT



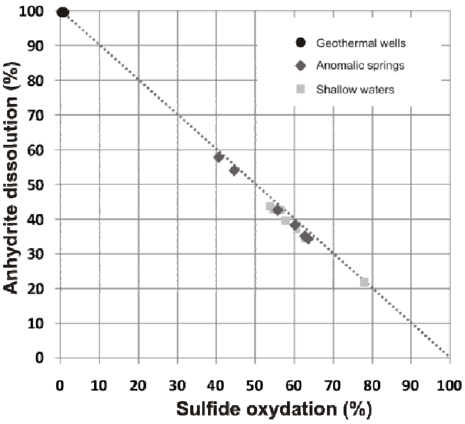
The two main origins of sulfate ion in Lower Carboniferous reservoir water are supposed to be sulfide oxidation and anhydrite leaching, which give totally different isotopic ratios. Sulfate from anhydrite would show high values for δ^{34}S as well as for δ^{18}O (more than +15 ‰), reflecting that of anhydrite itself, as measured by Langguth and Nielsen (1980), and Pierre (1986). Sulfate coming from sulfide oxidation would present typical negative values (Krouse, 1980).

Geothermal water, as Figure 13 shows, is the perfect example of anhydritic sulfate. On the opposite corner of this figure, one of the quarry pumping water, known for issuing from a pyrite-rich alteration zone in the Tournaisian limestone, shows quite negative values for δ^{34}S.

On Figure 13, two anomalic springs clearly stand out from the shallow waters group and tend to show more positive ratios, indicating a significant anhydrite leaching influence. This effect is not obvious for the other springs, which stay close to the group of shallow waters.

Shallow waters isotopic ratios however show values that can't be the result of sulfide oxidation alone. Sulfate concentration in them indicates another participation that can be found in deep sulfated water. Mass balance for sulfate and resulting isotopic ratios have been calculated using three possible origins : anhydrite, sulfide and in a much lesser proportion, sulfate from meteoric water. Figure 14 gives the result of this treatment. With a meteoric sulfate having a +5 ‰ δ^{34}S for an average concentration of 2 mg/l (Mook, 2000), the shallow water markers spread along a straight line joining the 100 % sulfide corner to the opposite 100 % anhydrite corner. The gap to this line illustrates the influence of meteoric sulfate, which appears to be very limited, as expected.

Figure 14 : Origins of sulfate in the Lower Carboniferous aquifer water



This figure confirms that two of the anomalic springs are clearly more influenced by deep water than the others. But it also indicates that even in regular shallow water, where sulfide oxidation remains the main sulfate provider, anhydritic sulfate enters as a part of the total sulfate content for 30 to 40 %. This proportion can finally be seen as an image of the mixing between shallow fresh water and deep warm and sulfated water in the shallow Lower Carboniferous aquifer.

Considering these results, the combined role of outcropping areas as both recharge and discharge zones is now proved. Further studies are presently going on to locate regions where recharge dominates and where deep water temperature could therefore be lower than in the presently exploited wells, as in Condé.

CONCLUSIONS

The Lower Carboniferous aquifer of Hainaut has been characterized in terms of water geochemistry and isotopic geochemistry, in both deep and shallow part of the reservoir. Deep geothermal water show strong sulfate content related to the leaching of Visean anhydrites still present in the deep strata. Oxygen and hydrogen isotopes analyses indicate its meteoric origin, and depletion in heavy isotopes suggests infiltration during or at the end of the last ice age, which is consistent with estimation of residence times based on ^{14}C. Stable carbon isotope ratios in bicarbonate confirm that geothermal water is in equilibrium with the carbonated rock matrix. Sulfur and oxygen stable isotopes on sulfate ion show the signature of Visean anhydrite dissolution.

Shallow water is not surprisingly a bicarbonate and calcium-rich water. Sulfate can appear in quite important quantities, and several natural springs, particularly on the South margin of the aquifer which corresponds to the top of the reservoir, also show temperature anomalies that lead to suspect a deep contribution to the shallow water chemical content. The Lower Carboniferous carbonates contain sulfides, especially pyrite, that is locally abundant (e.g. : in karstic altered fillings). In the aim of estimating the relative influence of each sulfur potential origin, δ^{34}S and δ^{18}O of sulfate ion has been measured in anomalic spring water and in several quarry and water catchment pumping as a reference of regular shallow aquifer water. The results confirm mixed contribution of anhydritic sulfate and oxidized sulfite in shallow water, with a mixing rate that exceeds 50 % of deep sulfates in several cases.

These investigations confirm the double role of the shallow part of the aquifer, both as recharge and discharge area for the deeper part, presently exploited for geothermal heating. Flow patterns in the reservoir are not homogeneous, as temperature repartition tends to show, and further careful investigations have to be led before setting new exploitations to avoid regions where recharge dominates, and where deep temperature might be lower than expected.

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