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**MODELLING CHANGES IN SULPHATE CONCENTRATIONS IN DISCHARGE WATER FROM THE ABANDONED IRON MINE OF SAIZERAIS**

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**INTRODUCTION**

The closure of large mines causes a systematic disturbance to the hydrogeological environment. Pollution arises after flooding of the abandoned workings leads to discharge at the surface. As the general character of mine systems is one of extreme yet orderly heterogeneity, in terms of both permeability and dominant flow regime, standard techniques for modelling groundwater flow and quality are inappropriate. Furthermore, spatially distributed models require a large amount of data, generally not available when considering large mining areas. We adopt a global approach to simulate changes in sulphate concentrations in mine discharge water in the Lorraine iron basin, France, and test this method on the iron mine of Saizerais, abandoned 20 years ago.

**SITE DESCRIPTION**

The Saizerais mine is located in the Lorraine iron basin along the eastern edge of the Paris Basin. The Lorraine iron-ore deposit extends from the Luxembourg border and the Moselle area, where it crops out in the north and east (Bajocian cliffs of the Briey and Haye plateaux), to the marshy Woëvre area in the west. The Pont-à-Mousson anticline causes a break in the mineralisation, thus dividing the horizon into two basins of unequal size (Figure 1):

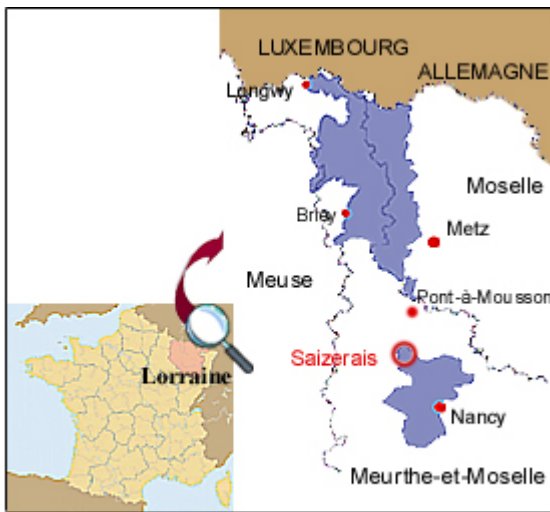


Figure 1 : Geographical location of the LIB

(i) the Longwy-Briey Basin to the north and (ii) the Nancy Basin to the south where the formations are thinner and have different characteristics. The Saizerais mine lies in the northwest of the Nancy Basin. The iron-bearing formation, dated as Aalenian, overlies Early and Middle Toarcian deposits comprising paper shale at the base and then marl containing *Septaria*, capped in places by a very thin layer of Late Lias sandstone that has not been observed with certainty at Saizerais. Regionally, the iron-bearing formation is composed of alternating marly levels (intercalations) and ferruginous limestone (mineralised intervals) forming a regressive sequence. Its thickness varies from 0 to 65 m with an average of 40 m (Montagne *et al.*, 1992). Nine mineralised intervals, distinguished by their colour, are recognised in the Lorraine iron basin (Maubeuge, 1955), although generally no more than two were mined in a single concession. The marly intercalations are rich in carbonates and phyllosilicates, with a low iron content but a notable proportion of pyrite. The iron-bearing formation is overlain by about 5 m of micaceous marl, followed by a Dogger (Bajocian) limestone succession, some

50-60 m thick, that constitutes a major karstic aquifer.

At Saizerais, the Aalenian is relatively thin (0 to 6 m) and shows lacunas. The classic subdivisions established by Bubenicek (1961) in the Briey and Longwy basins cannot be applied here. The deposits are generally characterised by lenticular bodies, particularly around the small Dieulouard basin, and stratigraphic condensation is common.

The iron-ore deposit was discovered at Saizerais in 1916 during water-resource prospecting, but mining only began in 1957 using the room and pillar method. The crisis that hit the iron and steel industry during the 1970's led to closure of the Saizerais mine on 30 June 1980. A total of 21 million tons of ore had been extracted.

**MINE AND GROUNDWATER**

The cutting of mine galleries, associated with the common collapse of the hanging wall, brought the Dogger groundwater into contact with the Aalenian groundwater. The Saizerais mine thus artificially drains the Dogger aquifer.

The water flowing into the mine during exploitation was not diffuse throughout the rock mass, but restricted to specific areas that did not seem related to either preferential surface infiltration zones or to tectonism. The inflow points are probably ‘conduits’ of karstic or anthropic origin that induce preferential flow paths. Tracing tests have revealed an east-west circulation over a distance of more than 8 km. Flow velocities vary between 10 and 20 m/h.

Closure of the Saizerais mine in 1980 and the decision to end dewatering brought about the progressive flooding of the mine workings until the overflow point at 200 m a.s.l. in Drift 2 was reached in December 1982, and from which the water flows directly into the Moselle River (Figure 2). Flooding of the mine did not significantly affect the piezometric surface outside the mining area. Even a certain degree of coherence is observed between levels in the Bajocian aquifer and those of the mine reservoir, thus suggesting hydraulic continuity. Mine-water reserves were estimated at about 8,000,000 m<sup>3</sup> by the mining companies.

From 1972, the water pumped from the Saizerais mine, of good physico-chemical quality, was used directly for drinking-water supply. In 1980, the Saizerais mine was closed and dewatering stopped. To compensate for this sharp decrease in water resources used to supply Pont-à-Mousson with drinking water, a new 160-m-deep pumping well was drilled to intersect a gallery at depth point 91.95 m a.s.l. (Figure 2). A dewatering overpumping system was installed on 14 September 1987 in Drift 2 so as to increase groundwater recharge.

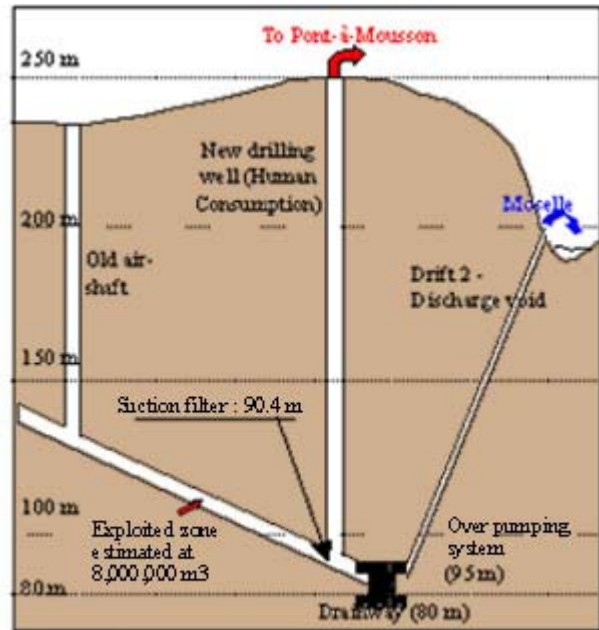


Figure 2: Schematic cross section of the Saizerais mine

Mine flooding was accompanied by a marked decrease in water quality. SO<sub>4</sub> concentrations rose from 42 mg/l before flooding to 1200 mg/l at overflow. The overflow discharge also shows high Na (940 mg/l) and Cl (995 mg/l) concentrations. These values are similar to the quality of water pumped from the Pont-à-Mousson well (550 mg/l of sulphate and 440 mg/l of sodium) at the end of overflow. Based on these results, the Water Board concluded in a 1983 report "that there is a stratification of water quality within the mine, and that the overflow (discharge derived from the deepest parts of the mine via the drift and dewatering pipes) served to drain off a ‘dead storage’ of highly mineralised water". According to an estimate carried out by the former mining company, a pore volume of approximately 500,000 m<sup>3</sup> exists beneath the abstraction level in the Pont-à-Mousson well, which is concreted its entire length and lined at about 90 m a.s.l.). This probably corresponds to a reservoir with different physico-chemical properties to the overlying one (in contact with the footwall and hanging wall of the lower silica-rich layer).

**CONCEPTUAL FRAMEWORK OF GROUNDWATER FLOW**

The Saizerais abandoned mine system can be considered as being composed of two superimposed sub-reservoirs with different chemical and physical properties:

		Upper sub-reservoir	Lower sub-reservoir
<b>Chemical characteristics</b>	Maximum SO <sub>4</sub> (mg/l)	600	1200
	Associated mineralised layer (Hervé, 1980)	Hanging wall of the grey layer	Hanging wall of the red layer
<b>Physical characteristics</b>	Volume (m <sup>3</sup> )	8.35 · 10 <sup>6</sup>	0.5 · 10 <sup>6</sup>
	‘Inflows’ of the system	•% Effective rainfall	•% Effective rainfall •Overflow from the upper layer
	‘Outflows’ of the system	•Drinking water supply •Karst leakage	•Overpumping •Overflow

Groundwater flow within the mine reservoir can be divided into three phases (Figure 3).

(i) At the beginning of flooding, the mine reservoir is empty. Governed by the effects of gravity, water percolates through the Dogger limestone and recharges the lower sub-reservoir, either directly or via the upper sub-reservoir. As transfer times through the upper sub-reservoir are very short, we consider that the water has a chemical composition similar to that of the Dogger fluids (SO<sub>4</sub> ≅ 50 mg/l), regardless of the actual flow path adopted. The underlying Toarcian marls constitute an impermeable barrier.

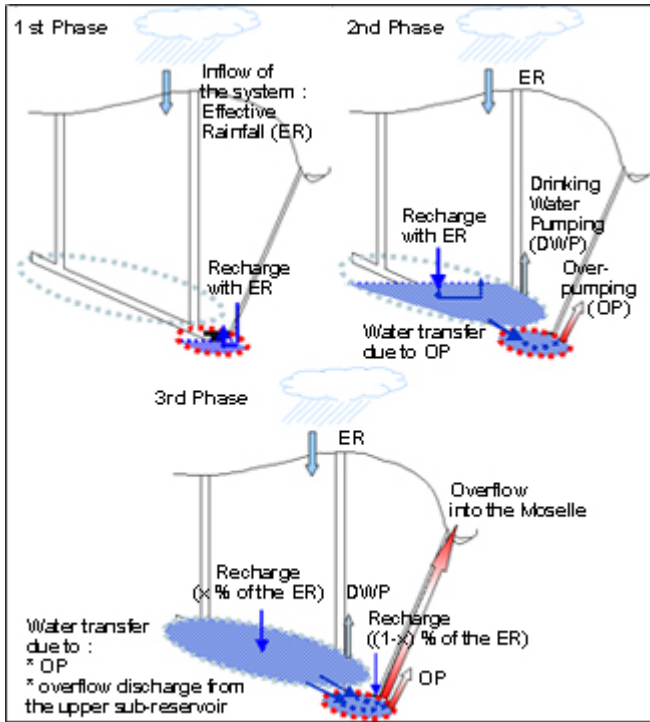


Figure 3: Conceptual model of the Saizerais mine.

(ii) When the lower sub-reservoir is saturated, rainfall no longer recharges the lower sub-reservoir, but now supplies the upper sub-reservoir. Nevertheless, during overpumping, the water extracted from the lower sub-reservoir is immediately replaced by that of the upper sub-reservoir (again governed by gravity), thus generating an ‘induced’ circulation between the upper and lower sub-reservoir.

(iii) When the upper sub-reservoir is saturated, the mine reservoir overflows from Drift 2, which is directly linked to the lower sub-reservoir. We assume that both sub-reservoirs are recharged by water having the same concentration as the Dogger groundwater, and in proportions related to their respective volumes. The lower sub-reservoir is also recharged by overflow from the upper sub-reservoir (adding to the effect of overpumping), and in turn overflows from Drift 2.

### MODEL CONSTRUCTION

Based on the current state of knowledge concerning groundwater flow and the process of sulphate acquisition by water, the hydrochemical behaviour of the Saizerais iron mine has been modelled using an approach of ‘mixing cells in series’ (Jauzein *et al.*, 1989; Villiermaux, 1993), also known as NCR or Networks of Chemical Reactors (Altmann, 1995; Baranger & Kervevan 1998; Adams and Younger, 1999; Baranger *et al.*, 1999a, 1999b).

As shown on Figure 4, the mine reservoir is composed of three units representing the Dogger karst limestone on the one hand, and two chemical reactors corresponding to the two sub-reservoirs on the other.

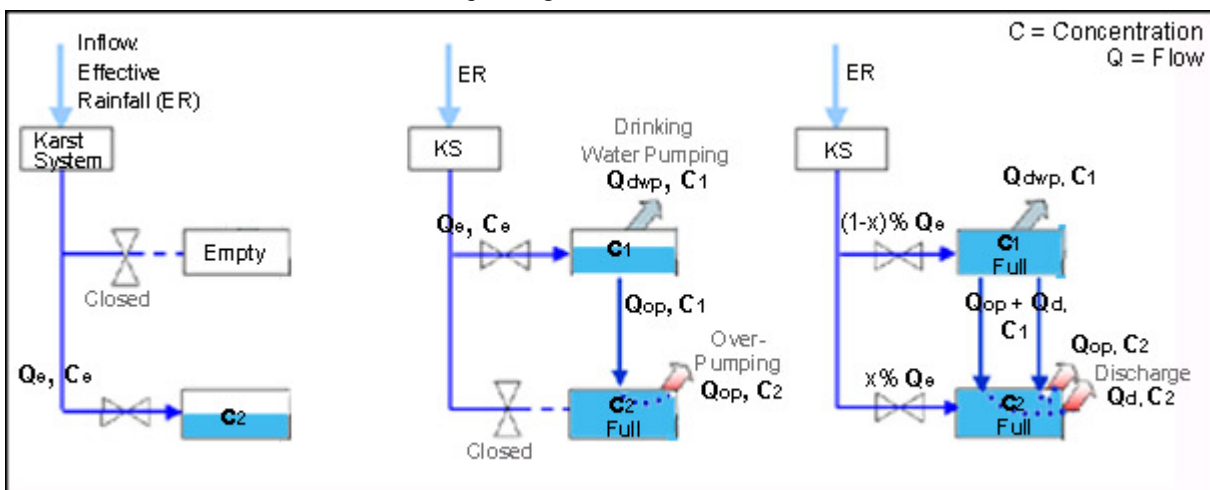


Figure 4: NCR simulators of the Saizerais mine reservoir

The karst limestone is the recharge area of the aquifer. Recharge rates vary depending on seasonal variations, but sulphate concentrations are constant at 50 mg/l, i.e.  $5.2 \cdot 10^{-4}$  mole/l (average value for groundwater circulation in the Dogger limestone). The three phases of groundwater flow in the mine reservoir mentioned in the description of the conceptual framework are modelled using a set of valves linking two chemical reactors that represent the two sub-reservoirs.

In order to model groundwater flow within the two sub-reservoirs, we adopted the classic chemical engineering approach, *i.e.* the ideal mixing chemical reactor. This approach, previously applied to transport modelling in porous media (Jauzein *et al.*, 1989), allows us to overcome a lack of data by considering the system in its entirety: each sub-reservoir is characterised by an inflow  $Q_i$ , an outflow  $Q_o$  (sum of discharge due to overflow, overpumping, abstraction and/or leakage), and a volume  $V$ , variable so as to represent fluctuations in the piezometric level. Groundwater circulating in a sub-reservoir encounters sulphate in the solid state, which it can dissolve and transport away from the

sub-reservoir. The chemical reactor is described by the following mass-conservation equation (Villiermaux, 1993):

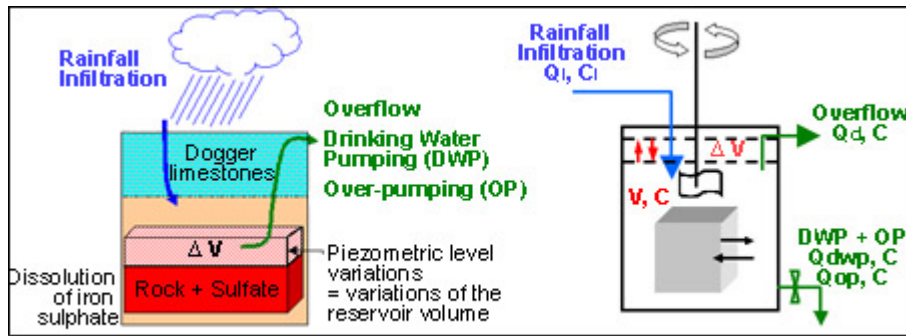


Figure 5 : Principle of the chemical reactor used to simulate a "sub-reservoir"

$$(Q_{\text{over}} + Q_{\text{leak}} + Q_{\text{abstr.}})C_o = Q_i C_i + \text{Vol} (d\chi/dt) - d(\text{Vol} C_o)/dt$$

$Q_{\text{over}}$  = overflow discharge  
 $Q_{\text{leak}}$  = leakage discharge  
 $Q_{\text{abstr.}}$  = abstraction discharge  
 $Q_i$  = reactor inflow  
 $C_o$  = outflow concentration  
 $C_i$  = inflow concentration  
 $\text{Vol}$  = volume of water in the reactor  
 $dC/dt$  = accumulation of the chemical compound in the reactor water  
 $d\chi/dt$  = rate of dissolution or precipitation

A numerical simulator of this chemical reactor was built using the software packages Simulink® and Matlab® (Becquette, 1998). To carry out calculations using this simulator, however, the following data are required: (i) a record of aquifer inflow (recharge flowrate), (ii) volume of water within the aquifer, (iii) initial amount of available solid sulphate, (iv) value of the equilibrium and kinetic constants of the sulphate dissolution reaction. These data can only be obtained after two other models have been built and calibrated: a hydrogeological model and a chemical model.

### HYDROGEOLOGICAL MODEL

The various groundwater flows in the mine reservoir are estimated on the basis of the available data records: rainfall, potential evapotranspiration and discharge (rivers, dewatering, overflow). The computer code TEMPO, based on signal-processing methods (Pinault *et al.*, 1999; Pinault *et al.*, 2001), was used to process the data records.

This technique was first applied to modelling the flowrate of the Esche River, located a few kilometres from the Saizerais mine, using rainfall records. These results were then used to establish an effective rainfall record.

Effective rainfall is used in two sub-models derived through direct deconvolution by identifying impulse responses relating effective rainfall and records of dewatering discharge (1976 and 1980 data) or piezometric levels (1988 to 1991 data). These impulse responses are then compared to those periods when no outflow data are known for the model: based on the calculated effective rainfall records from 1981 to 2000, the potential dewatering discharge can be calculated, and thus the recharge flowrate of the mine over this period. Similarly, fluctuations in the piezometric level between 1981 and 2001 are determined, which helps overcome the lack of field measurements (no data from 1985 to 1987). By integrating the inflow of the mine from the end of dewatering until the beginning of overflow, from which the abstraction discharge is subtracted, we estimate the total volume of the mine reservoir to be  $8.85 \cdot 10^6 \text{ m}^3$ . This result not only confirms the validity of the mining company's measurements, but also guarantees to a certain extent the validity of our model.

### CHEMICAL MODEL

The main hydrochemical processes involved in water-rock interactions constitute a current GISOS research topic. The aim here is to test the strategy adopted, with only sulphate dissolution being taken into account. A simple chemical kinetic model was selected that only simulates sulphate dissolution/precipitation processes. Leaching experiments carried out on marl in the southern and northern parts of the Briey Basin, respectively by Hervé (1980) and by our team, suggest that sulphate is produced through gypsum dissolution. The gypsum itself would be the product of oxidation of pyrite contained in the marly intercalations during mine cutting. We consider a global reaction and use apparent kinetic constants in our calculations. These implicitly incorporate elementary kinetic mechanisms. The considered reaction is expressed:  $XSO_4 = X^{2+} + SO_4^{2-}$

The total rate of this reaction is equal to the difference between the dissolution rate and the precipitation rate. The formulae retained for the dissolution and precipitation rates are:

$$v_{\text{dissolution}} = k_{\text{diss}} M_{\text{rct}}$$

$$v_{\text{precipitation}} = k_{\text{pcp}} (SO_4^{2-})(X^{2+}) M_{\text{rct}} = k_p (SO_4^{2-})^2 M_{\text{rct}}$$

$$k_{\text{eq}} = k_{\text{diss}} / k_{\text{pcp}}$$

Where :  $k_{\text{eq}}$  = apparent equilibrium constant

$k_{\text{diss}}$  = apparent dissolution rate constant

$k_{pcp}$  = apparent precipitation rate constant

$M_{ret}$  = amount of available sulphate

The term  $M_{ret}$  (amount of available sulphate) is used to relate kinetics to the reactive mineral mass. The greater the solid sulphate mass, the higher the dissolution rate, because the exchange surface between solid sulphate and water increases. By default, in the absence of more complete data concerning the ion “X”, the precipitation rate is assumed to be of second order with respect to  $SO_4^{2-}$ . At equilibrium, the reaction rate is nil and the equilibrium constant ( $k_{eq}$ ) is equal to the square of the  $SO_4^{2-}$  concentration of the fluid.

Values of the apparent equilibrium constants are extracted by comparing the results of studies undertaken on rocks of the Briey Basin (Hervé, 1980) with the maximum concentrations observed during overflow at Saizerais. In the absence of experimental data (particularly leaching tests) for rocks of the Saizerais mine, the precipitation rate constant ( $k_{pcp}$ ) and the amount of available sulphate ( $M_{ret}$ ) are estimated by calibration with field measurements carried out during the first two years at Saizerais.

**MODELLING SULPHATE CONCENTRATION CHANGES IN SAIZERAIS MINE WATER**

After calibration of the chemical parameters over the first two years of overflow, and based on the experimental results, we obtain the following set of parameters (Table 2):

Table 2 – Values of the constants of the chemical model after calibration with the first two years of overflow.

Chemical model constants	Log $K_{eq}$	Log $k_{pcp}$	Amount of available sulphate
Hanging wall of the lower layer	-3.43	-3.8	1872 mg/l
Hanging wall of the upper layer	-4.35	-3.1	3744 mg/l

A simulation was then carried out until the end of 1994. Concentrations were considered at two points: the overflow point in Drift 2, also the site of overpumping, and the Pont-à-Mousson drinking-water abstraction point.

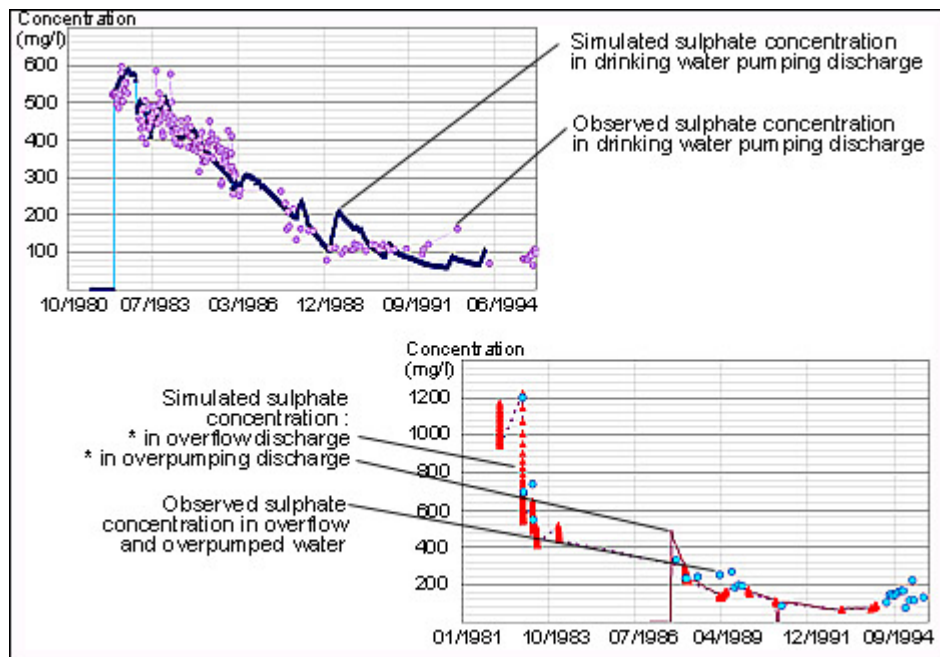


Figure 6 : Simulated and observed sulphate concentrations in Saizerais

Sulphate concentrations at the overflow point reach 1200 mg/l when the water first overflows after 521 days. Concentrations then fall sharply and when the second overflow occurs, in April 1983, concentrations are similar to those observed in the abstraction discharge. On the whole, the simulated concentrations are similar to the measured concentrations.

Maximum sulphate concentrations in the abstraction discharge are some 600 mg/l when the water first overflows. Concentrations then fall gradually until 1988 when values are around 100 mg/l. The curve is characterised by the marked influence of rainfall intensity on dilution, giving a ‘sawtooth’ impression already observed for the measured values. The overflows are identified on the curve by a drop in concentration marking the mixing between fluids of the two ‘sub-reservoirs’ and the effect of dilution. Once again, the values calculated using the model are similar to the measured concentrations.

When flowrate data are reliable, and after calibration of the chemical parameters over the first two years of overflow, it is possible to simulate changes in sulphate concentrations of the mine water at Saizerais.

## CONCLUSION

The iron mines of Lorraine have an impact on water resources with associated major environmental consequences. This issue is currently being studied in the aim of characterising the situation and establishing a water-management model. The lack of data in terms of hydrology (only a dozen or so observation wells exist over the entire Lorraine iron basin), geology (rocks of different type and chemical composition) and, to a greater extent, mapping of the mines, means that it is difficult to apply traditional modelling techniques.

Global modelling based on an approach of signal processing and a Network of Chemical Reactors offers a promising alternative for simulating changes in sulphate concentrations in waters of the Lorraine iron basin. The application of this method to the iron mine of Saizerais, flooded for the past 20 years, has produced correct simulations of the concentrations measured at different overflow and abstraction points. Laboratory experiments on various rock samples are currently underway in order to complement and refine the chemical data. The results will enable us to integrate more complete chemical mechanisms, and combine the study of changes in  $\text{SO}_4^{2-}$  concentrations with that of other major ions observed in solution, such as  $\text{Na}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{HCO}_3^-$ .

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