## Characterisation of gypsum karst aquifers by heat and solute transport simulations

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## ABSTRACT:

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Environmental risks in gypsum karst areas such as geomechanical problems as a result of gypsum dissolution or the vulnerability of the aquifers due to the unretarded transport of pollutants are mainly determined by the karst conduit system. Therefore, in order to provide a reliable basis for risk assessment, an adequate hydrogeological characterisation of the conduit system is required. A newly developed modelling tool is presented, which has been designed to support the characterisation of the conduit system of gypsum karst aquifers by simulating short-term fluctuations of solute concentrations and temperatures of the spring water. Both solute concentration and temperature of the spring water depend on the geometric and hydraulic properties of the conduit system. If only one of these parameters is analysed a unique identification of the structure of the conduit system may not always be obtained. Unsteady-state simulations of both heat and solute transport, however, show that different conduit systems, which are equivalent with respect to spring signals of one parameter, can be distinguished by taking into account a second parameter.

# Key words: Gypsum karst, Karst modelling, Conduit system, Heat transport, Solute transport.

## INTRODUCTION

Research into the hydraulics of karst aquifers is presently mainly focused on carbonate terranes. However, karstification is also common in evaporitic rocks such as gypsum, which are present beneath about 25% of the continental surface (FORD & WILLIAMS 1989). Gypsum karst occurs in many regions of the world. In Europe it is found, for instance, in the Pre-Ural, the Volgo-Kamsky and the Pinego-Severodvinsky region of Russia, in the Western Ukraine, in the south of Poland and on the southern flank of the German Harz Mountains (KLIMCHOUK & *al.* 1996). Environmental risks encounterd in gypsum karst areas are described, e.g., by KLIMCHOUK & ANDREJCHUK (1996). Severe problems such as collapses and subsidence hazards caused by solutionally enlarged voids or the vulnerability of the aquifers as a result of the fast and unretarded transport of pollutants are mainly determined by the conduit system, i.e. the fast flow system of a karst aquifer. Therefore, in order to provide a reliable basis for risk assessment, an adequate knowledge of the structure and the hydraulic properties of the conduit system is required.

ASHTON (1966) suggested to characterise karst drainage systems by analysing spring flow data. His method makes use of the time lag usually observed between the increase in flow and the change of hydrochemical parameters at a spring after a flood. It was applied e.g. by ATKINSON (1977) and SAUTER (1992) to estimate the volumes of conduit water in regional carbonate aquifers. In addition, not only this time lag appears to provide information about the conduit system, but also the amplitude of the chemical variation at the spring. Based on the analysis of concentration data and numerical model simulations GRASSO (1998) proposed a relationship between the geometric properties of conduits and the variations of calcium concentrations measured at carbonate karst springs. Thus, the concept of analysing hydrochemical parameters at karst springs has been shown to be useful in the characterisation of carbonate aquifers. In addition, SAUTER (1992) and BENDERITTER & al. (1993) obtained quantitative information about the properties of carbonate aquifers by analysing spring water temperatures. Moreover, simulations of heat transport processes in karst aquifers by numerical models showed that under unsteady flow conditions temperature signals at a spring can be used to identify conduits of different geometry even if the total conduit volume is identical (Renner 1996 HÜCKINGHAUS & al. 1997, LIEDL & al. 1998, HÜCKINGHAUS 1998). It is therefore concluded that variations of both hydrochemical parameters and water temperature measured at a spring can be used to characterise a karst aquifer.

Although the examples quoted above deal with carbonate aquifers only, it appears reasonable to apply the same methods to gypsum aquifers as well. Variations of water temperature and solute concentration can be found, e.g., at gypsum karst springs in Southern Germany (BUNDSCHUH 1997). At one of these springs, a time lag between minima of temperature and concentration is observed after precipitation events, suggesting that heat and solute transport in gypsum aquifers might be governed by different processes. If this were true temperature and concentration variations would probably contain not the same but complementary information about the karst drainage system. Therefore, a process based numerical modelling tool was developed to simulate both heat and solute transport processes in gypsum karst aquifers, in order to be able to investigate in how far spring water signals can be interpreted in terms of physical parameters and whether an integrated approach can reduce the ambiguity in the interpretation.

#### MODELLING APPROACH

Flow in karst conduits is simulated by a discrete pipe network model developed by HÜCKINGHAUS (1998). Computation of flow in the pipe network is based on Kirchhoff's law, which states that total inflow and total outflow balance at each node of the network (HORLACHER & LÜDECKE 1992):

$$\sum_{i=1}^{n} Q_i + R = 0$$
 (1)

where  $Q_i$  are flow rates in n pipes connected to a node (m<sup>3</sup> s<sup>-1</sup>), R is direct recharge into the karst conduit system at a node (m<sup>3</sup> s<sup>-1</sup>). Flow in pipes is governed by the Darcy-Weisbach equation

$$\Delta h = \lambda \frac{L}{d} \frac{u \mid u \mid}{2g}$$
(2)

where  $\Delta h$  is head difference along pipe (m),  $\lambda$  is friction factor (-), d is pipe diameter (m), L is length of pipe (m), u is average flow velocity (m s<sup>-1</sup>), and g is gravitational acceleration (m s<sup>-1</sup>). The friction factor is calculated depending on flow conditions, thus distinguishing between laminar and turbulent flow in each individual pipe of the network. In order to deal with the non-linearities occurring for turbulent flow conditions, the iterative Newton-Raphson method is applied to solve the model equations.

Simulation of reactive solute transport in the pipe network is based on the 1D advection equation, which is extended by an additional source term  $S_C$  (mol m<sup>3</sup> s<sup>-1</sup>) accounting for the increase of solute concentrations due to dissolution of gypsum:

$$\frac{\partial C}{\partial t} = -u \frac{\partial C}{\partial z} + S_c \qquad (3)$$

where C is concentration of dissolved gypsum (mol m<sup>-3</sup>), t is time (s), z is co-ordinate in flow direction (m) and u is mean flow velocity in the pipe (m s<sup>-1</sup>). Eq. (3) is solved numerically using an explicit upwind finite-difference scheme. The concentration of the inflow to the pipes is obtained by assuming an instantaneous mixing of water at the nodes of the pipe network.

JAMES & LUPTON (1978) showed experimentally that the dissolution of gypsum in laminar flow through pipes obeys a first-order rate law:

$$S_{\rm C} = h_{\rm M} \frac{4}{d} \left( C_{\rm eq} - C \right) \tag{4}$$

where h<sub>M</sub> is mass transfer coefficient (m s<sup>-1</sup>), d is pipe diameter (m) and is ratio of surface area exposed to water and pipe volume, C is concentration of dissolved gypsum in bulk solution (mol m<sup>-3</sup>), and  $C_{eq}$  is equilibrium concentration with respect to gypsum (mol m<sup>-3</sup>). Since their experiments revealed a dependence of h<sub>M</sub> upon the flow velocity, JAMES & LUPTON concluded that the dissolution process under laminar flow conditions is governed by diffusion of dissociated ions across a boundary layer, which separates the mineral surface from the bulk solution (Textfig. 1). In karst pipes, however, flow conditions are frequently turbulent (FORD & WILLIAMS 1989). Therefore, we have conducted similar experiments circulating water through holes, which were drilled axially into cores of gypsum rock, in order to check the validity of eq. (4) for turbulent flow conditions up to Reynolds numbers of about 10000 (VIERNEISEL 2000). Our results indicate that the first-order rate law is valid under these conditions. More importantly, we were able to calculate the value of the mass transfer coefficient  $h_M$  reasonably well by

$$h_{\rm M} = \mathrm{Sh} \frac{\mathrm{D}}{\mathrm{d}} = \frac{\mathrm{D}}{\varepsilon_{\rm M}} \tag{5}$$

where D is diffusion coefficient (m<sup>2</sup> s<sup>-1</sup>), d is pipe diameter (m), and Sh is the dimensionless Sherwood number, which may be interpreted as ratio of pipe diameter d and thickness of diffusion boundary layer  $\varepsilon_{M}$ . For turbulent pipe flow the Sherwood number is given by empirical mass transfer correlations (BEEK & MUTTZALL 1975)

$$Sh = 0.027 \text{ Re}^{0.8} \text{ Sc}^{1/3}$$
 (6)

$$\operatorname{Re} = \frac{\operatorname{ud}}{\operatorname{v}} \tag{7}$$

$$Sc = \frac{v}{D}$$
 (8)

where Re is Reynolds number, Sc is Schmidt number and v is kinematic viscosity of water  $(m^2 s^{-1})$ .

Usage of eq. (4) relies on the assumption of a diffusioncontrolled dissolution process, i.e. diffusion across the boundary layer is believed to be slow compared with the dissociation of gypsum molecules at the mineral surface. Therefore, it is important to compare the mass transfer coefficients  $h_M$  resulting from eqs. (5)-(8) with the rate constant of the dissolution reaction at the mineral surface. The latter was determined experimentally for several varieties of gypsum by LEBEDEV & LEKHOV (1989) and JESCHKE & *al.* (2001), yielding dissolution rate constants in the order of  $10^4$  m s<sup>-1</sup>. This is well above the maximum value calculated for mass transfer coefficients in the model simulations presented in this paper ( $6.5 \cdot 10^{-6}$  m s<sup>-1</sup>), thus justifying the assumption of a diffusion-controlled dissolution process.



Fig. 1. Mass transfer processes in gypsum karst pipes

Heat transfer between pipe wall and water flowing through the pipe is analogous to the diffusion mass transfer model considered above (BEEK & MUTTZALL 1975). Thus, the governing equations of the solute transport model are easily adapted to allow for heat transport simulations. The equation of heat convection along the pipe is expanded by a source term  $S_T$  accounting for heat transfer across the thermal boundary layer between pipe wall and bulk water (Text-fig. 2):

$$\frac{\partial T}{\partial t} = -u \frac{\partial T}{\partial z} + S_{T}$$
(9)

The source term is given by

$$S_{T} = \frac{h_{H}}{c_{w}\rho_{w}} \frac{4}{d}(T_{s} - T)$$
(10)

where T is water temperature (K),  $T_s$  is temperature at rock surface,  $\rho_w$  is density of water (kg m<sup>-3</sup>),  $c_w$  is specific heat of water (J kg<sup>-1</sup> K<sup>-1</sup>) and d is pipe diameter (m). The heat transfer coefficient  $h_H$  (J s<sup>-1</sup> m<sup>-2</sup> K<sup>-1</sup>) is related to thermal conductivity of water  $\lambda_w$  (J s<sup>-1</sup> m<sup>-1</sup> K<sup>-1</sup>) and pipe diameter via the dimensionless Nusselt number Nu, describing the ratio between pipe diameter and thickness of thermal boundary layer  $\varepsilon_H$  (BEEK & MUTTZALL 1975):

$$h_{\rm T} = {\rm Nu} \frac{\lambda_{\rm w}}{d} = \frac{\lambda_{\rm w}}{\varepsilon_{\rm H}}$$
(11)

The Nusselt number is analogues to the Sherwood number in mass transfer analysis, thus for turbulent pipe flow it is given by

$$Nu = 0.027 \text{ Re}^{0.8} \text{ Pr}^{1/3}$$
(12)

$$\Pr = \frac{v}{\alpha_{w}}$$
(13)

where Pr is Prandtl number and  $\alpha_w$  is the thermal diffusivity of water (m<sup>2</sup> s<sup>-1</sup>) given by

$$\alpha_{w} = \frac{\lambda_{w}}{\rho_{w} c_{w}} \tag{14}$$

However, in a karst aquifer an important difference between heat and solute transport exists. On the one hand, the concentration at the rock surface equals the equilibrium concentration and thus is constant provided the dissolution process is diffusion-controlled. On the other hand, heat transfer between rock and turbulently flowing water is controlled by heat conduction in the rock matrix rather than by heat transfer across the thermal boundary layer (Text-fig. 2), i.e. the temperature at the rock surface will change its initial value to approach values close to the bulk water temperature. Therefore, the heat



Fig. 2. Heat transfer processes in karst pipes and surrounding rock

transport model has to solve the equation of heat conduction in cylindrical co-ordinates (CARSLAW & JAEGER 1959) around the conduits in order to calculate the time-dependent temperature at the rock/water interface:

$$\frac{\partial T_r}{\partial t} = \alpha_r \left( \frac{\partial^2 T_r}{\partial r^2} + \frac{1}{r} \frac{\partial T_r}{\partial r} \right)$$
(13)

where  $T_r$  denotes rock temperature,  $\alpha_r$  is thermal diffusivity of rock, and r denotes the radial coordinate. Again, the model equations for heat transport are solved numerically using an explicit finite-difference scheme. The temperature of inflow water to each pipe is calculated by assuming an instantaneous mixing of water at the nodes of the network.

## MODEL SCENARIOS AND RESULTS

As a first scenario we compare temperatures and concentrations of spring water emerging from a single conduit of 1200 length with spring signals resulting from heat and solute transport in a pipe network (Textfig. 3). Both conduit systems consist of 24 pipes, each of a diameter of 0.2 m and a length of 50 m. Thus, the volume of the conduit system and the area of rock exposed to conduit water is equal in both cases. Moreover, the pipe network is arranged such that the distance between the individual injection points and the outlet is always identical. Therefore, the residence time of water is the same in both systems provided the total recharge, which is applied to only one point of the single conduit, is distributed equally on the eight inlets of the pipe network.

The water initially in the pipes is in equilibrium of concentration with respect to gypsum (15 mol m<sup>-3</sup>) and of temperature with respect to rock temperature (281.15 K). Then recharge with both lower concentration (0 mol m<sup>-3</sup>) and lower temperature (279.15 K) is



Fig. 3. Conduit systems of the first model scenario

injected at the nodes marked in Text-fig. 3. The total recharge amounts to  $0.012 \text{ m}^3 \text{ s}^{-1}$  during the first six hours (21600 s) and is reduced then to  $0.006 \text{ m}^3 \text{ s}^{-1}$ , i.e. each inlet of the pipe network is supplied with 0.0015 m<sup>3</sup> s<sup>-1</sup> and 0.00075 m<sup>3</sup> s<sup>-1</sup>, respectively. Under these conditions flow in the pipes is always turbulent.

The resulting solute concentrations and temperatures at the spring were normalised using the following equation:

$$X_{norm} = \frac{X - X_0}{X_r - X_0}$$
(14)

where X denotes solute concentration or water temperature at the spring,  $X_0$  is concentration or temperature of the inflow, and  $X_r$  is equilibrium concentration or initial temperature of rock, respectively. Text-fig. 4 shows that the normalised water temperature at the spring obtained by the heat transport simulation is virtually the same for both the single conduit and the pipe network. At a flow rate of 0.012 m<sup>3</sup> s<sup>-1</sup> the water takes about 52 minutes (3142 s) to flow from the inlet to the spring. Thus, after that time the water temperature falls below its initial value. Since the rock is permanently cooled by recharge water, the temperature keeps falling as long as the flow rate remains constant. When the flow rate is decreased to 0.006 m<sup>3</sup> s<sup>-1</sup> the residence time of water in the conduit system increases. Due to the lengthened contact time of water and rock surface, the water temperature rises. At first, however, water emerging at the spring has been mainly flowing with the higher flow rate through the conduit system. Therefore, water temperatures increase as long as emerging spring water has been flowing through the whole conduit system with the lower flow rate. After this transition period, the cooling of the rock matrix and the thereby decreasing heat flux from the rock



Fig. 4. Normalised temperatures and concentrations at the outlets of the conduit systems of the first model scenario

eventually causes the water temperature to decrease again. Since the temperature signals of single conduit and pipe network are more or less identical during the whole simulation period, it is not possible in this case to distinguish between the different conduit structures by analysing spring water temperatures only. We will look therefore at the second parameter, i.e. the solute concentration of water emerging at the spring.

Although the solute concentration shows basically a similar behaviour in either case, the values obtained for the single conduit are higher than those of the pipe network (Text-fig. 4). When recharge water appears at the spring, the solute concentration drops below saturation reaching temporarily a constant normalised concentration of 0.33 at the outlet of the single conduit and a value of 0.14 at the outlet of the pipe network. These values may also be obtained by solving the advection equation (3) analytically for steady-state conditions (i.e.  $\partial C/\partial t = 0$ ). Inserting eq. (4) in eq. (3) and replacing the mass transfer coefficient by the Sherwood number for turbulent flow conditions as given by eqs. (5-8), the resulting equation can be integrating along a pipe of length L yielding the concentration at the pipe outlet

$$C_{out} = C_{eq} - (C_{eq} - C_0) e^{-0.10v^{-7/5}D^{2/3}Ld^{-4/5}Q^{-1/5}}$$
(15)

where flow velocity has been replaced by flow rate Q, and  $C_0$  denotes concentration of inflow to the pipe. This equation can be directly applied to the single conduit. In the pipe network steady-state concentrations can be calculated in downstream direction using the

average of the outlet concentrations of upstream pipes as inflow concentration for the downstream pipe.

Eq. (15) demonstrates that spring water concentrations increase with decreasing flow rate. Therefore, the change in flow rates after six hours (21600 s) initiates a transition period, in which the concentration of the spring water increases. After that period water emerging at the spring has been flowing with a constant flow rate through the whole conduit system. Therefore, the normalised concentration again reaches constant values of 0.37 at the outlet of the single conduit and 0.15 at the outlet of the pipe network. Thus, the solute concentration of the spring water emerging from the single conduit is clearly different from the concentration at the outlet of the pipe network even under steady-state flow conditions, i.e. it is possible to distinguish both conduit systems by analysing the spring water concentrations. This result reflects the different controlling processes of heat and solute transport in a gypsum aquifer. Heat transfer from the rock to the turbulently flowing water is not limited by heat transfer across the thermal boundary layer between rock surface and bulk water, but by heat conduction in the rock matrix. Since the latter does not depend upon the flow conditions in the pipe, conduit systems with identical pipe volume, identical surface area and the same residence time of water in the system will show the identical water temperatures at the spring even if flow velocities differ. Gypsum dissolution, however, is controlled by mass trans-



Fig. 5. Conduit systems of the second model scenario

fer across the diffusion boundary layer between pipe wall and bulk water. The thickness of this boundary layer, and thus mass transfer across it, is influenced by the flow velocity in the pipe. Therefore, if flow conditions are not the same in two conduit systems, which are equal with respect to all other properties, solute concentrations will be different at the outlets of the systems. Within the branches of the pipe network shown in Text-fig. 3 flow rates are obviously smaller than in the pipes of the single conduit. Thus, the diffusion boundary layer is thinner in the single conduit, i.e. the diffusion process is faster and the solute concentration of the spring water is larger as compared to the pipe network.

From the above discussion the question arises, whether a pipe network exists, which shows the same solute concentration at the outlet as the single conduit considered before. In fact, eq. (15) reveals, that it is possible to compensate for different flow rates by changing pipe length and pipe diameter. However, when changing these parameters the total volumes of the two conduit systems still have to be equal, since otherwise the residence times of water would be different in the two systems. Text-fig. 5 shows an appropriate pipe network and the corresponding single conduit, which is the same as in scenario one.

The resulting solute concentrations of the spring water (Text-fig. 6) are now the same for the two conduit systems. However, the temperatures of water emerging at the outlet of the pipe network are larger than those of the single conduit. In order to make the



Fig. 6. Normalised temperatures and concentrations at the outlets of the conduit systems of the second model scenario

pipe network equivalent to the single conduit with respect to solute concentration, the area of rock exposed to water had to be increased as compared to the single conduit. Under these conditions heat transfer between rock and flowing water, which is limited by heat conduction in the rock rather than by velocitydependent heat transfer across the thermal boundary layer, is faster in the pipe network than in the single conduit, thus accounting for larger temperatures of water emerging from the pipe network.

## CONCLUSIONS

The model simulations demonstrate that spring water emerging from conduit systems of different geometry can show either the same concentration or the same temperature. Thus, a unique identification of the structure of the conduit system is not possible by analysing only one of the two parameters. However, conduit structures, which are equivalent with respect to one parameter (e.g. temperature), can be distinguished by taking into account the second parameter (e.g. concentration). We therefore state that a combined analysis of concentrations and temperatures at gypsum karst springs reduces the ambiguity contained in the information about the structure of the conduit system. This conclusion is based on the assumption that heat and solute transport in gypsum aquifers are controlled by two different processes, i.e. heat conduction in the rock and mass diffusion across the boundary layer between pipe wall and turbulent flow. While heat conduction does not depend upon the flow conditions in the pipe, mass diffusion does. Under conditions in which limestone dissolution is diffusion-controlled (compare LIU & DREYBRODT 1997, DREYBRODT & BUHMANN 1991) a similar result, therefore, should be obtained for carbonate karst aquifers.

Further investigation will have to address the question of how far heat and solute transport in karst aquifers is influenced by the dualistic behaviour of the flow system. Flow in karst aquifers may be divided into two flow components, i.e. slow diffuse flow in the fissured system of the rock and rapid localised flow in solutionally enlarged conduits (ATKINSON 1977). The two flow components are coupled, for instance, in the numerical karst modelling tool CAVE (CLEMENS & *al.* 1996), which has been recently expanded for the simulation of gypsum dissolution and transport in karst conduits (BIRK & *al.* 2000). Since the fissured system of the rock contains the major part of groundwater in a karst aquifer, such a coupled model might be better suited for a realistic simulation of karst spring signals than the pure pipe flow model employed in the present study. In addition, future work will have to examine whether the approach of a combined analysis of temperature and concentration data at karst springs is applicable under field conditions.

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