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ORIGINAL PAPER

U. Lang · T. Gudera · P. Elsass · G. Wirsing

Numerical modelling of chloride propagation in the quaternary aquifer of the southern Upper Rhine Graben

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Abstract The deep groundwater in the quaternary gravel sequence of the southern Upper Rhine Graben locally contains high chloride concentrations near the river Rhine between Fessenheim (France) in the South and Breisach (Germany) in the North. This historical pollution is mainly due to past infiltration from the former brine storage basins of the French potash mines on the "Fessenheim Island" and—to a lesser extent—from the leaching of the salt dumps of the German potash mines in Buggingen and Heitersheim. The spreading of the salt plume was investigated by means of a groundwater model. The aim of the model was to understand the brine movement, the present distribution of chloride as defined by recent hydrochemical investigations, and to select locations for new reconnaissance boreholes. The geological structure was reproduced by a three layer model, which was calibrated for steady state flow conditions. The hydraulic conductivity of the first layer was determined by comparing measured and calculated heads in the model area. The vertical resolution was refined to simulate the density-dependent salt transport processes. The transport of the salt plumes was simulated over a 40-year period, starting at the beginning of brine storage in the 1950s. The relevant transport parameters have been estimated in a sensitivity analysis, where the simulated breakthrough curves of chloride concentration have been compared with the measured data. The results of the groundwater model indicate that brines containing approximately 1 million tons of chloride are still present at the bottom of the aquifer. These highly concentrated salt brines mix with fresh water from the upper part of the aquifer. This dispersive process leads to the formation of a plume of chloride-rich water extending downstream, where pumping wells for several local water supplies are located.

Keywords Upper Rhine Graben · Groundwater salinisation · Density driven flow and transport · Groundwater model · Rhine water infiltration

U. Lang (✉)
Ingenieurgesellschaft Prof. Kobus und Partner GmbH,
Wilhelmstr. 11, 70182 Stuttgart, Germany
E-mail: lang@kobus-partner.com
Tel.: +49-711-237193600
Fax: +49-711-237193601

T. Gudera
Landesanstalt für Umweltschutz Baden-Württemberg,
Griesbachstr. 1, 76185 Karlsruhe, Germany
E-mail: Thomas.Gudera@lfuka.lfu.bwl.de
Tel.: +49-721-9831363
Fax: +49-721-9831514

P. Elsass
Service Géologique Régional Alsace, BRGM,
BP177 15, rue du Tanin, Lingolsheim,
67834, Tanneries cedex, France
E-mail: p.elsass@brgm.fr
Tel.: +33-3-88774895
Fax: +33-3-88761226

G. Wirsing
Landesamt für Geologie, Rohstoffe und
Bergbau Baden-Württemberg, Albertstraße 5,
79104 Freiburg i. Br., Germany
E-mail: wirsing@lgrb.uni-freiburg.de
Tel.: +49-761-2044397
Fax: +49-761-2044397

Introduction and purpose of study

Beside widespread elevated concentrations of nitrate, groundwater in the Quaternary sediments of the central part of the southern Upper Rhine Graben locally shows high concentrations of chloride. This pollution is mainly a consequence of the mining for potash of Oligocene salt deposits. The area was extensively mined in the Alsace Potash Basin on the French side, but also in the Heitersheim and Buggingen area on the German side. Mining started in 1910 in the so-called Wittelsheim Basin north of Mulhouse (France), and on the German side in 1927 near Buggingen. It ended on the German side in 1976 and was operated on the French side until 2002. The salt processing involved disposal of large quantities of brine with high sodium chloride concentrations, which was piped into the Rhine from 1927 on. In order to handle the brine that accumulated when the Rhine was at low water conditions, storage basins were created in 1957 on the "Fessenheim Island" between the old Rhine and the Canal, and were used until 1976 (Fig. 3). The saturated salt brine which was piped into the basins infiltrated into the deep subsurface. Smaller storage basins were also used on the German bank of the Rhine. Rainwater infiltrating through the mining dumps near Heitersheim and Buggingen dissolves salt and seeps into the subsurface, contributing to a lesser extent to the present day chloride concentrations in the groundwater of the region. Chloride input from dissolution of salt in the underlying Oligocene sediments is not relevant compared to the amount of anthropogenic salt input and was therefore not incorporated into the numerical model (Bauer et al. 2004).

The chloride pollution may have consequences for water management as well as for the extraction of sand and gravel in the area. In order to understand the present day distribution of chloride in groundwater, a numerical groundwater model was set up with the aim of simulating the spreading of the salt plume. The numerical model was also used to define the most suitable positions for new observation wells, which were drilled for the purpose of hydrochemical and isotopic groundwater studies.

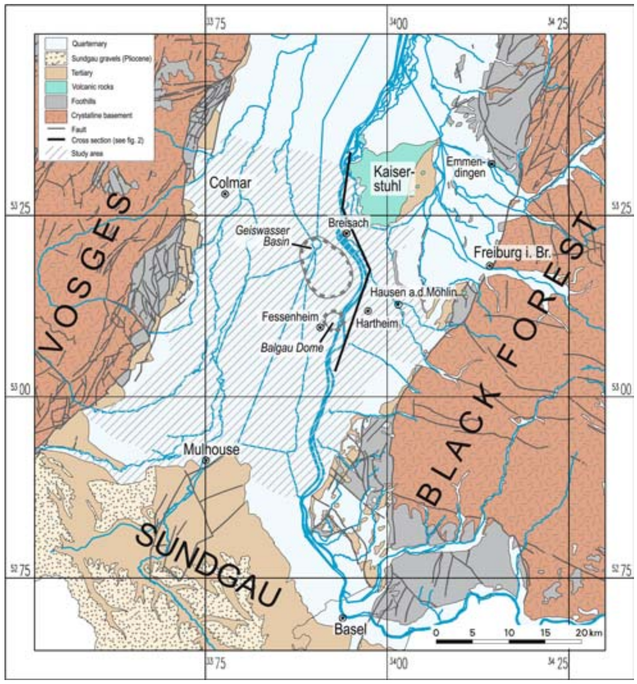


Fig. 1 Location of the study area in the southern Upper Rhine Graben

2–6 m below ground in the North of the study area. The magnitude of mean groundwater flow velocities is 3–5 m/day. The groundwater within the Quaternary sediments is unconfined.

Hydrogeology

The study area is situated in the southern part of the Upper Rhine valley (Fig. 1), occupying the Upper Rhine Graben, which constitutes the central and most important part of a NNE–SSW trending West European rift system (Illies 1977; Pflug 1982; Hüttner 1991). During the Quaternary, this structure was filled in by the Rhine and its tributaries with coarse-grained clastic sediments, derived from the Alps as well as from the Vosges and the Black Forest respectively. Beside some occurrences of highly mineralized water and thermal water in Triassic and Jurassic sediments of the deeper subsurface (Buntsandstein, Upper Muschelkalk, Hauptrogenstein), the groundwater resources are concentrated in this Quaternary sedimentary fill. The Quaternary sediments host one of the largest groundwater occurrences in Central Europe with international importance for the supply of drinking water and water for agricultural and industrial use (HGK 1977; Landesanstalt für Umweltschutz Baden-Württemberg, Région Alsace 1996; HGK 1999).

On the eastern and western boundaries of the Quaternary sediments with the Black Forest and the Vosges respectively, the flow direction is almost perpendicular to the Rhine valley. In the central part of the study area, the groundwater flow direction is more or less parallel to the Rhine river with a northerly direction. The depth of the water table ranges from 8–10 m in the South to

Hydrogeological model

The Quaternary sediments in the study area are subdivided into three hydrogeological units, mainly on the basis of lithological criteria (content of gravel/sand/silt, source of the material, degree of alteration of the gravels and density of the material), from top to bottom: Neuenburg-Formation (Younger Alluvial, Jüngere Schotter), Breisgau-Formation (upper part, Upper Breisgau-Schichten and lower part, Lower Breisgau-Schichten) (see Fig. 2 and Table 1). The base of the aquifer comprises sediments of Plio-Quaternary (Iffezheim-Formation, Fig. 2) and Tertiary (Oligocene) age. Thickness and distribution of the hydrogeological units are influenced by the tectonics that affected the top of the Tertiary sediments as well as locally the Quaternary sediments, and by salt-induced tectonic movements, which were still active during the Quaternary (Lutz and Cleintuar 1999). Isoline contour maps of the base of the hydrogeologic units are given in LGRB (2000).

The uppermost unit (Neuenburg-Formation) consists of gravel with a variable sand content (mainly medium-grained sand) and a low silt content. Going from the Vosges and the Black Forest respectively towards the Rhine, the proportion of crystalline components within the Neuenburg-Formation as well as the sorting of the

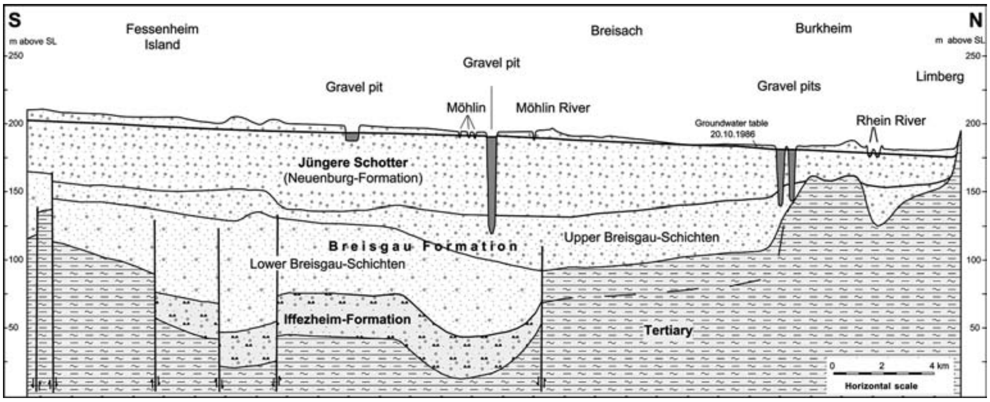


Fig. 2 Schematic geologic cross section (*N–S*) through the study area (see also Fig. 1)

constituents decreases, and gravels of Alpine origin prevail. Based on the ratio of Alpine versus Black Forest/Vosges origin of the components, the Neuenburg-Formation was subdivided into three lithofacies types: in the central part Alpine material prevails with hydraulic conductivities $>10^{-2}$ m/s. The transition zone towards the graben flanks is characterized by a varying content of material derived from the Alps and the Black Forest/Vosges. Hydraulic conductivities range from $1*10^{-3}$ m/s to $1*10^{-2}$ m/s. The outer zone is dominated by material derived from the Black Forest and the Vosges, respectively. The content of sand and silt increases towards the mountains. Hydraulic conductivity of these poorly sorted sediments is between 10^{-4} m/s and 10^{-2} m/s. The Neuenburg-Formation reaches its maximum thickness of about 70 m on the eastern side of the river Rhine near Hausen on the Möhlin, where the water supply wells of the city of Freiburg are located. In places where the Tertiary basement comes close to the surface (e.g. southeast of Bremgarten) the thickness of the Younger Alluvium is reduced to about 10 m.

The unit beneath the Neuenburg-Formation is the Breisgau-Formation. It consists of sandy to silty gravels that are locally densely packed. The gravels are fresh to completely altered. In general, the degree of alteration as well as the packing of the components increases with depth. Due to its lithology, the Breisgau-Formation is subdivided into an upper subunit with a hydraulic conductivity of $1*10^{-4}$–$1*10^{-2}$ m/s (Upper Breisgau-Schichten) and a lower, more densely packed sequence with a hydraulic conductivity of $1*10^{-7}$–$1*10^{-4}$ m/s (Lower Breisgau-Schichten). Similar to the Neuenburg-Formation, the Breisgau-Formation is laterally differentiated into several lithofacies types (LGRB 2000). The Breisgau-Formation occurs throughout the investigation area and reaches its maximum thickness of about 200 m southwest of Breisach (Geiswasser Basin). The base of the aquifer is made up of the Iffezheim-Formation (Plio-Quaternary) and of the Tertiary basement. The Iffezheim-Formation corresponds partly to the ''Altquartär'' of Bartz et al. (1982) and to the *''Pliocène final''* of French literature, and consists of densely packed clay, silt and silty sand. The distribution of the Iffezheim-Formation is controlled to a great extent by faults within the Tertiary basement. The Tertiary basement consists of marl-, silt-, and claystones. The hydraulic conductivity of the Iffezheim-Formation and of the Tertiary basement ranges between 10^{-9} m/s and 10^{-5} m/s.

Base of the aquifer

Due to the density-driven transport of the highly concentrated chloride brines, the morphology of the base of the aquifer is of crucial importance for the present-day distribution and the future propagation of the salt brines. Therefore, this boundary was investigated in detail in the vicinity of Fessenheim Island by means of seismic reflection measurements and additional drilling (Région Alsace 2000; Regierungspräsidium Freiburg 2002). These investigations resulted in the discovery of a dome-like structure North of the Fessenheim Island (the Balgau Dome) which is part of a larger structure known as the ''Diapir of Weinstetten'' (Eßlinger 1968), and of a graben on its eastern flank. Whereas at the Balgau Dome the Tertiary basement comes up to about 120 m a.s.l, it is

Table 1 Maximum thickness and hydraulic conductivities of the hydrogeologic units

Hydrogeologic unit	Hydraulic conductivity (m/s)	Maximum thickness (m)
Neuenburg-Formation	$>10^{-2}$–10^{-3} (central part of the graben)	70
	10^{-2}–10^{-4} (outer zone of the graben)	30
Upper Breisgau-Schichten	10^{-2}–10^{-4}	50
Lower Breisgau-Schichten	10^{-4}–10^{-7}	150
Iffezheim-Formation (base of the aquifer)	10^{-5}–10^{-9}	30

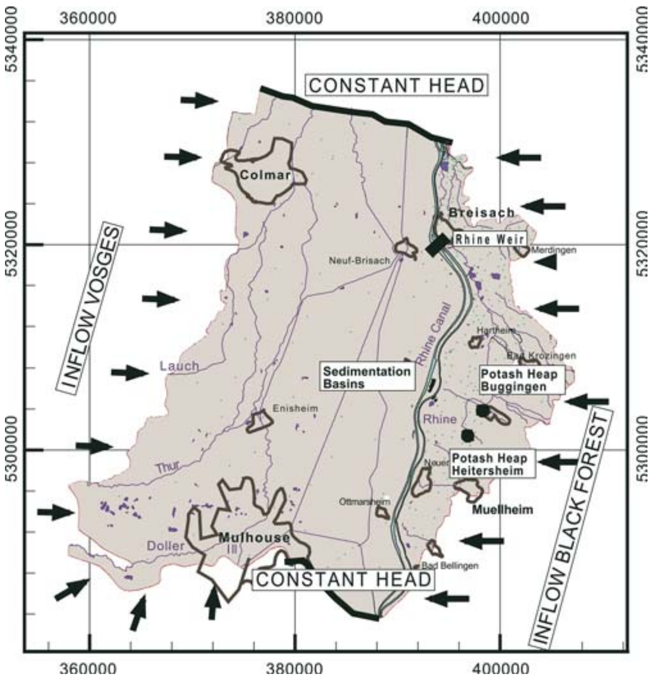


Fig. 3 Modelling area and flow boundary conditions

situated in the graben at 30–40 m altitude. In the North–West direction of the graben structure, the base of the aquifer dips towards the so called Geiswasser Basin, where the Quaternary sediments of the southern Upper Rhine Graben reach a maximum thickness of 240 m.

Numerical groundwater model

Model configuration

A numerical model was set up to investigate the movement of the salt plumes from the sedimentation basins of the Fessenheim island and from the salt heaps of Heitersheim and Buggingen. The finite element program SPRING (GKW 1999) was used for the modelling. SPRING is a three dimensional model for the simulation of flow and transport processes in groundwater with the capability to simulate density-driven effects. The finite element mesh can be generated with triangles and rectangles, which has the advantage that fewer elements and nodes are necessary for the same geometric approximation than with finite element models that only use triangles. The finite element mesh of the groundwater model was generated under consideration of all relevant geometries: rivers, lakes, wells and natural boundaries. The mesh was refined in the area of the expected salt plume, to obtain a better accuracy for the transport modelling. The resulting mesh consists of 52,776 nodes and 61,983 elements in one horizontal layer.

The model configuration was developed from the hydrogeological model with the aquifer subdivided into three layers and on the basis of two existing numerical models (BRGM 1998 and Landesanstalt für Umweltsschutz Baden-Württemberg Région Alsace 1996) . The horizontal extension of the model is similar to that of the two existing regional groundwater models. The modelling area covers the entire width of the fluvial aquifer in the Upper Rhine Graben between Mulhouse and Colmar (Fig. 3). The eastern and western boundaries of the model area are the boundaries of the Black Forest and the Vosges. The inflow of groundwater from these boundaries was modified during the calibration process. The total inflow from the Black Forest was set to 1.037 m^3/s. The inflow rate from the western boundary (Vosges) was set to 1.497 m^3/s. The inflow rates can be justified by the surface catchment of the inflow boundary and the estimation of 2 l/s km^2 recharge rate in the catchment. This assumption leads to inflow rates of 1 m^3/s from the Black Forest and 1.5 m^3/s from the Vosges. Constant head boundaries were established in the northern and southern part to simulate the groundwater exchange with the regional porous aquifer of the Rhine Graben. The values of the constant head boundaries were derived from measured heads. The northern and southern boundaries are located at the head isoline of 180 m and 245 m a.s.l, respectively.

The aquifer is in contact with a fine mesh of streams and with river Rhine. River Rhine is divided into a canal and a branch, which formed the original river bed. The flow is controlled by two weirs. The exchange between groundwater, streams and rivers is simulated by a leakage boundary condition. The water level of the Rhine was interpolated from several measurement points. The water level of the streams has been derived from the existing models. One hundred and twenty-seven water filled gravel pits are located in the investigation area, which are in direct contact with the groundwater. An extremely high conductivity was assumed for the first layer, to simulate a nearly horizontal water table in the lakes. This results in an equivalent inflow and outflow for the lakes. A specific yield of 1 was attributed to the elements inside the lakes. The potash dumps were assumed to have no influence on the inflow. Only the natural groundwater recharge was assigned to the horizontal area of the potash heaps.

Six hundred and twenty-six water wells for drinking water supply, industrial use and irrigation are located in the model area. The pumping rates there were assigned to the corresponding model layer as sink conditions. Areal drainage systems to drain agricultural areas are present in the northwestern part of the model area. These drainage systems have been simulated as surface leakage boundary conditions.

An areal recharge due to precipitation has been derived from the existing models. The distribution of the recharge varies between 0.3 l/s km^2 and 17 l/s km^2. The highest recharge values are found in the eastern part of the modelling area near the Black Forest. Low recharge rates are estimated for the center of the Rhine valley.

Steady state flow calibration

The numerical model was calibrated for steady state flow conditions. The piezometric heads from 360 observation wells were available for the calibration. The calibration was performed for medium flow conditions. The measurements of the reference day (10/20/1986) were chosen for comparison between measured and calculated heads. Hydraulic conductivity, the leakage coefficients of the rivers and streams, and the boundary inflow were modified during the calibration process.

The most important result of the steady state flow calibration is the distribution of hydraulic conductivity, which could be well determined for the first layer ("Jüngere Schotter" or Neuenburg-Formation) because most of the observation wells are located in this layer. It is known from borehole material and pumping tests that the conductivity decreases with depth. Therefore, the conductivity in the second and third layers was assumed to be lower than in the first layer by a factor of 0.1 and 0.01 respectively. The calibrated distribution of hydraulic conductivity in the first layer is presented in Fig. 4. The highest conductivities ($k_f = 10^{-3}$ –10^{-2} m/s) are found in the Neuenburg-Formation near river Rhine. Low conductivities have been determined for the regions with high head gradients at the borders of the Rhine valley. The calibrated distribution matches well with the geological differentiation by lithofacies types and the observed conductivities with highest values in the central part and decreasing conductivities towards the graben flanks.

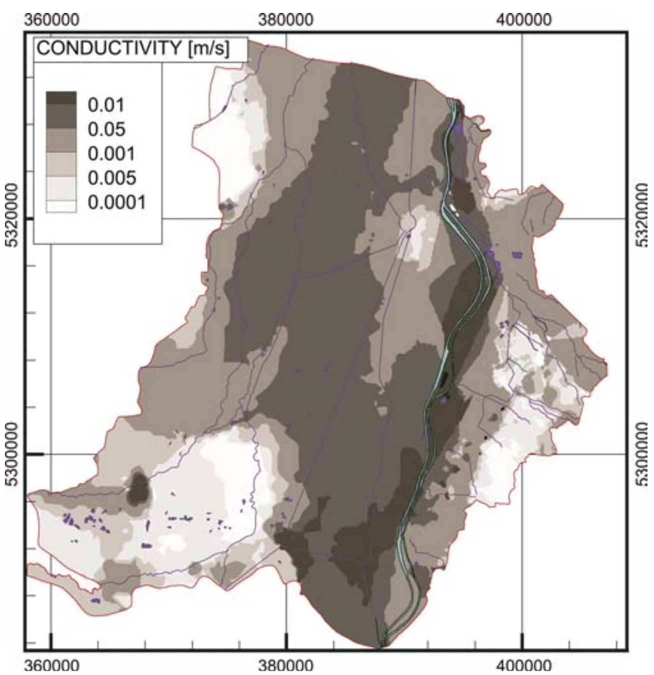


Fig. 4 Calibrated distribution of the hydraulic conductivity in the first model layer (Neuenburg-Formation)

The leakage coefficient between streams and groundwater was an additional calibration parameter for the steady state flow calibration. The leakage coefficient for streams is defined as follows:

$$C = \frac{k_f}{d} \cdot B \qquad (1)$$

where B is the width of the stream, k_f is the hydraulic conductivity of the stream bed layer and d is the thickness of the stream bed layer. The calibrated leakage coefficients vary between 10^{-8} m/s and 10^{-4} m/s.

The result of the calibration is shown in Fig. 5 as calculated versus measured heads. There is no significant trend or shift to high or to low calculated heads. The average difference between calculated and measured heads is 0.78 m for the total model area, which is less than 1% of the maximum head difference in the model area.

The resulting distribution of heads is shown in Fig. 6. The general flow field is parallel to the Upper Rhine valley from South to North. The flow direction in the western and eastern part of the investigation area, which is mostly perpendicular to the Rhine valley, is caused by the inflow from the graben boundaries. The highest heads can be found in the border area of the Upper Rhine Graben near Müllheim and Mulhouse due to the rising aquifer base. The head gradient in this regions is 1–5% and about 0.1% in the center of the Rhine valley.

Additional particle tracking investigations show the pathlines (Fig. 6) from the present salt infiltration areas (potash dumps Buggingen and Heitersheim, sedimentation basins of Fessenheim) to the Rhine and a parallel flow according to the line of the talweg of the head distribution. This indicates the exfiltration of groundwater into river Rhine as the main receiving stream. The backwater from the weir of Breisach

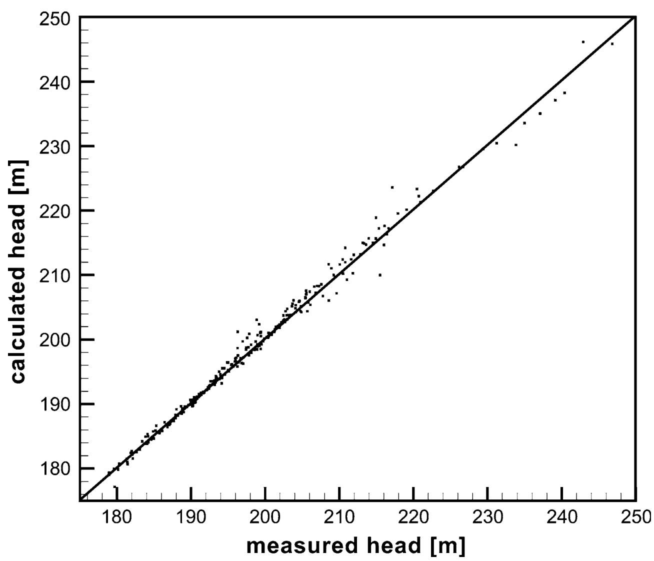


Fig. 5 Calculated versus measured heads

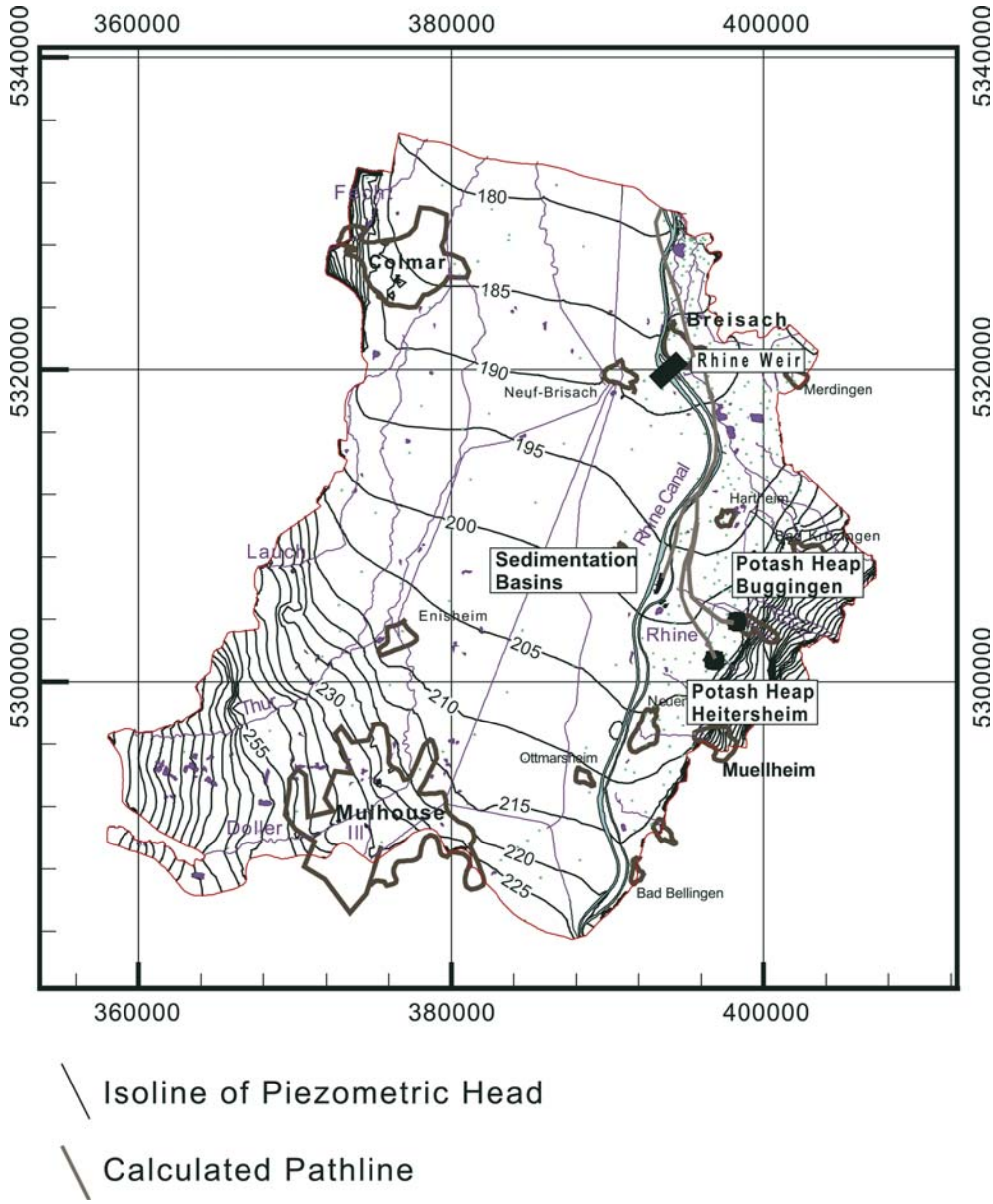


Fig. 6 Calibrated isolines from piezometric heads and pathlines from salt intrusion areas

causes an infiltration of Rhine water into the aquifer. Therefore the pathlines shift from the Rhine towards Breisach.

The overall groundwater budget was determined on the basis of the modelling results. The budget is listed in Table 2. The recharge due to precipitation is the main inflow component. The fine mesh of streams produces an inflow of 4 m^3/s at the border area of the model. The main outflow components are the withdrawals by wells and the outflow into river Rhine. The total sum of inflow or outflow is 18.6 m^3/s.

Determination of specific yield

The steady state simulation during the calibration process is only a rough approximation of the transient flow system due to seasonal changes of the recharge and to the dynamic movement of the water levels in the rivers and streams. However, it was assumed that this

Table 2 Water budget of the steady state groundwater model in cubicmeter per second

	Inflow	Outflow
Constant head	2.397	2.733
Recharge	8.013	
Withdrawal		6.498
Boundary inflow	2.534	
Leakage by drains		3.805
Exchange Rhine	0.811	4.243
Exchange Rhine Canal	0.765	0.017
Exchange stream mesh	4.075	1.299
Total	18.595	18.595

approach is sufficient as a base for the long term investigations of the salt movement. Nevertheless the existing model was enhanced to transient flow conditions for a 1-year period between November 1986 and October 1987, to estimate the storage coefficient. The unconfined storage coefficient (specific yield) is needed for the transient density driven flow and salt transport model investigation. Further on, it was assumed that the porosity is equal to or lower than the specific yield.

The transient simulation period includes a flood event of the river Rhine. During the flood event the water table of the river Rhine rose more than 4 m above medium conditions. This significant impulse was used to examine the behaviour of the introduced wave in the groundwater and to estimate the specific yield. On the basis of the existing models (BRGM and Life) the specific yield was determined to vary in three different zones with the higher values of 0.15 and 0.10 in most parts of the model area and lower values of 0.06 near the river Rhine. These lower values of specific yields correspond with the local zones of highest conductivities around the Rhine. Coarse gravel sediment can be found around the central part of the Rhine graben (see Table 1) with a smaller specific yield than in the outer zone of the graben.

This estimated distribution of the specific yield leads to transient movement of the piezometric heads as shown in Fig. 7. The comparison of measured and calculated heads for a selected well demonstrates that the numerical model reproduces the measured damping of the wave during its propagation in the groundwater.

Model validation by simulation of the propagation of infiltrated Rhine water

The calibrated groundwater model was validated with additional information on the movement of Rhine water derived from isotopic data. An interpretation of the extension of Rhine water into the aquifer was developed from isotope analysis (Regierungspräsidium Freiburg 2002). δ^{18}O-values typical for Rhine water were found in the area of Breisach more than 2 km away from the river Rhine. The Rhine water distribution shows the influence of the weir at Breisach. The backwater of the weir leads to an inflow of Rhine water into the aquifer. Rhine water will be transported parallel to the Rhine in the northern direction. This hydraulic effect was validated by the isotopic data.

A transport model was developed on the basis of the steady state flow field. Water infiltrating from the Rhine was marked with a concentration of 1.0 and the transport of this water was modelled over a period of 16 years. The existing model was refined in the vertical direction to get an appropriate approximation of the vertical transport processes. The Neuenburg-Formation was divided into four model layers. The upper part of the Breisgau-Formation (Upper Breisgau-Schichten) is represented by two model layers and the lower part of the Breisgau-Formation (Lower Breisgau Schichten) was divided into three additional layers.

The results of this transport investigation are shown in Fig. 8. Infiltrated Rhine water flows mostly near the Rhine in the southern model region. Only the meander of the Fessenheim Island leads to an underflow of the river Rhine. Therefore, groundwater infiltrated by river Rhine flows upto a distance of 100–500 m beside the Rhine. Due to the weir of Breisach the Rhine infiltrates into the groundwater. This Rhine water flows according to the measured data in the direction of Breisach.

A Rhine water component has been determined by the isotopic data in the deeper part of the aquifer from

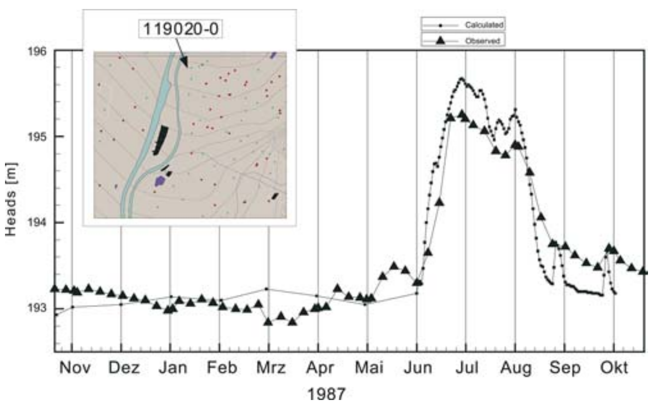


Fig. 7 Calculated and observed heads during flood event 1987 for a selected well north of the sedimentation basins

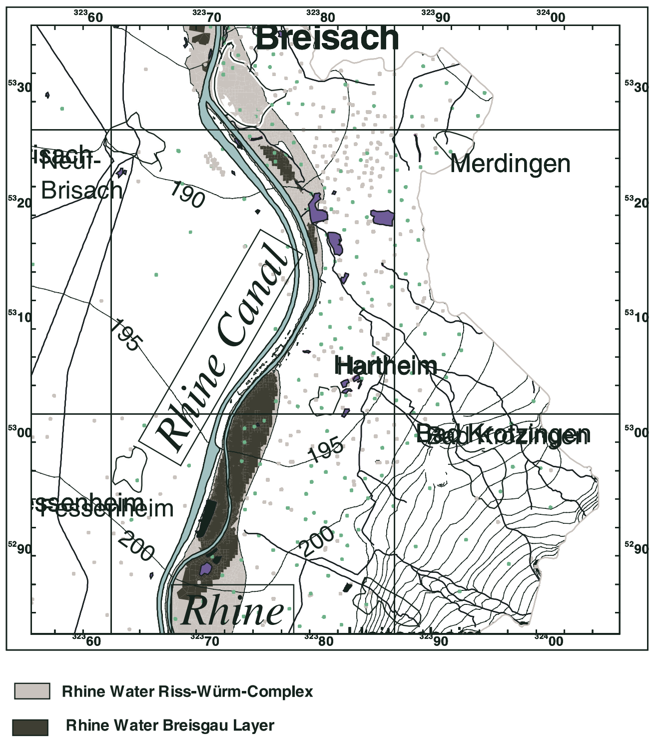


Fig. 8 Calculated groundwater regions with Rhine water

70 m to 100 m deep boreholes. The simulated Rhine water plume also showed the existence of Rhine water in the deeper part of the aquifer. The analysis of the simulated data demonstrates that the vertical flow gradient causes vertical transport of Rhine water to depths of 100 m within 10 years. This is in good accordance with the results of the isotope studies carried out in this region (Regierungspräsidium Freiburg 2002).

The density problem caused by salt intrusion

Density driven salt problems have been variously investigated and documented in the literature. An overview of salt problems and their numerical solutions is given in Diersch and Kolditz (2002) and Ackerer et al. (1999). Three typical salt problems have been described, which may often occur in natural groundwater systems (Fig. 9). These salt problems were mainly used to validate the program codes of density dependent flow and transport models.

The flow and transport processes in porous media in case of variable fluid density can be described by the fluid and solute mass balances, generalised Darcy‘s law and the equations of state for density and viscosity. The Darcy’s law is defined as (Bear 1979):

$$v = -\frac{k}{\varepsilon\mu} \cdot (\nabla P + \rho g \nabla z) \quad (2)$$

The fluid mass balance and solute mass balance can be described as follows:

$$\rho S \frac{\partial P}{\partial t} + \varepsilon \frac{\partial \rho}{\partial C_{\mathrm{m}}} \frac{\partial C_{\mathrm{m}}}{\partial t} + \nabla \cdot (\varepsilon \rho v) = 0 \quad (3)$$

$$\varepsilon \rho \frac{\partial C_{\mathrm{m}}}{\partial t} + \varepsilon \rho v \cdot \nabla C_{\mathrm{m}} = \nabla \cdot (\varepsilon \rho D \cdot \nabla C_{\mathrm{m}}) \quad (4)$$

The dispersion tensor D is defined as:

$$D = |v| \cdot \alpha_T I + \frac{\alpha_L - \alpha_T}{|v|} vv + D_m I \quad (5)$$

where:

P Fluid pressure (M/LT^2)
v Fluid velocity (L/T)
S Specific pressure storativity for a rigid matrix $[(M/LT^2)^{-1}]$
ϵ Porosity (-)
k Permeability tensor (L^2)
g Gravity acceleration (L/T^2)
ρ Fluid density (M/L^3)
μ Fluid dynamic viscosity (M/LT)
C_m Solute mass fraction (-)
α_L, α_T Longitudinal and transversal dispersivity (L^2/T)
D_m Diffusion coefficient (L^2/T)
I Unit matrix

The fluid viscosity was assumed to be constant for the density driven salt transport modelling in the Upper Rhine Graben. A linear function was applied for the dependence between fluid density and solute mass fraction. A density dependence on solute temperature was neglected.

Henry problem

The Henry problem describes propagation of a saltwater front in a confined aquifer, which was initially saturated with freshwater. This is a typical situation at coastal regions where seawater propagates at the bottom of the aquifer into the freshwater aquifer. The modelling area is two dimensional in a vertical cross section. The top and the bottom are impermeable. An inflow of freshwater is assumed on the left side. The intrusion of salt water is simulated by a constant concentration boundary condition and the assumption of hydrostatic pressure at the sea side.

Henry (1964) developed a semi-analytical solution for the intrusion problem. Based on the Oberbeck-Boussinesq approximation he derived analytical

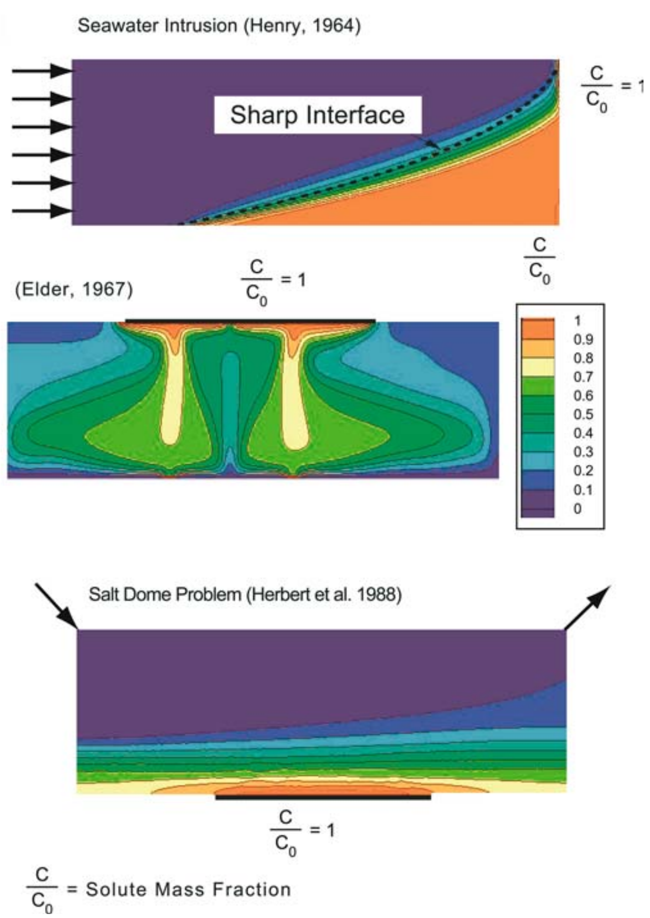


Fig. 9 Simulated salinity distribution of typical salt intrusion problems: seawater intrusion, Elder and salt dome problem

expressions for the salt concentration in form of Fourier series. Segol (1994) showed that the differences between calculated distributions and analytical solutions can be eliminated by the consideration of higher order terms of the analytical solution which were neglected in previous studies.

Elder problem

The typical distribution of water with a high density on top of water with a lower density was investigated by Elder (1967a). Elder (1967b) set up a small size laboratory experiment with water between two vertical plates. Heat was introduced at the lower boundary, which led to an upwelling of hot water in different fingers. Voss and Souza (1987) transformed the thermal Elder problem into a solute-analogous convection problem, where heavy salt water is placed on top and the investigation area is extended to 150 m depth and 600 m length. The unstable flow situation leads to a gravity-driven flow of the water on top with a higher density down into the fresh water. Simulations with different mesh sizes showed totally different simulation results with different concentration distributions especially for the central part, where upwelling and also downwelling occurs (Kolditz et al. 1998).

Frolkovic and De Schepper (2001) performed a sensitivity study with consecutive mesh refinement. They defined a mesh level l, which characterizes the discretisation. The dependence between mesh level l, number of elements (NE) and number or nodes (NN) can be described for a uniform discretisation as followed:

$$\begin{aligned} \mathrm{NE} &= 2 \cdot 4^{l} \\ \mathrm{NN} &= 2 \cdot (4^{l/2}+1)^2 - (4^{l/2}+1) \end{aligned} \tag{6}$$

They showed that mesh levels larger than six leads to the same distribution of salt fingers with downwelling in the center of the investigation area. Lower mesh levels show both downwelling and upwelling.

Ackerer et al. (1999) and Johannsen (2003) demonstrated that accurate results will also be obtained with coarser meshes if the convergence criteria for the transport solution are small enough.

Salt dome and salt pool problem

The salt dome problem describes the proceeding of fresh water over a salt dome. This benchmark was proposed by the participants of the international HYDROCOIN project for the verification of groundwater models (Swedish Nuclear Power Inspectorate 1988). The modelling area is a vertical cross section of a natural aquifer with a salt dome at the bottom. Fresh water is introduced at the top of the aquifer by a fixed pressure boundary condition and an inflow concentration of 0. The salt dome problem was investigated by several authors (Herbert et al. 1988; Konikow et al. 1997). A freshwater region with higher velocities is observed in the upper part and a brine pool is developing along the bottom, where flow with small velocities recirculates in two convection zones. Oldenburg and Pruess (1995) presented different results with only one convection zone. The comparison demonstrated that the development of the convection zone depends on the numerical representation of boundary conditions (either salt concentration or salt mass flux) and the chosen diffusion coefficients.

The salt dome problem is a synthetic example without any natural data. Therefore Oswald (1998) and Johannsen et al. (2003) investigated the salt distribution in a 40×40×40 cm laboratory experiment, where the lower part of the cube was filled with salt water and the upper part with fresh water. A sink and a source were installed at two upper corners. The laboratory experiment was performed with two different density ratios between freshwater and salt brine. (1) low density case (1% salt mass fraction) and (2) high density case (10% salt mass fraction). In the experiment, salinity breakthrough curves at the outflow opening were measured.

Several numerical models were tested to reproduce this simple laboratory experiment. A sufficient agreement between calculated and measured breakthrough curves was achieved with the models for the low density case. But the numerical models failed, when the density ratio between salt and fresh water was set equal to the high density case, as the saltwater mixing concentration at the outlet was significantly overestimated. This originates in the small transverse dispersivities. The best agreements with the measurements have been achieved by Johannsen et al. (2002). They had to adjust the system parameters as permeability, porosity and transverse dispersivity and used an extremely fine mesh with mesh level $l = 8$.

The salt problem in the Upper Rhine Graben between Fessenheim and Breisach involves the Elder, the salt dome and salt pool problem as follows: highly concentrated salt water is introduced by the potash heaps or sedimentation basins at the top of the aquifer. This leads to an intrusion situation similar to the Elder problem. However, the three-dimensional effects have to be taken into account (Dirsch and Kolditz 1998). The introduced salt water falls due to gravity effects to the bottom of the aquifer and settles in large salt pools. These salt pools are overflown by freshwater, and dispersive processes lead to a downstream plume of low concentration salt brine in the same way as in the salt dome problem. In addition to these transport processes the vertical movement of the salt plume is affected by a layered aquifer system with different hydraulic conductivities. The salt plume at the bottom moves due to the slope of the base of the aquifer and the regional flow field.

Density driven salt transport model

The aim of the transport model was to investigate the salt propagation starting in the 1960s with medium hydrological boundary conditions. The distribution of the current chloride concentration in the Upper Rhine Graben between Fessenheim and Breisach was estimated by a sensitivity analysis of the relevant transport parameters. A detailed calibration of the transport parameters was not undertaken. The numerical interpretation of the chloride plume was the basis for the positioning of two new boreholes in the downstream region of the salt plume (locations are shown in Fig. 14). These locations have been determined according to the numerical results.

The calibrated flow model was enhanced to simulate the density dependent transport processes caused by salt brine intrusion from the potash basins of Fessenheim and the salt heaps from Buggingen and Heitersheim. The flow boundary conditions from the steady state calibration were used for the transport modelling and were assumed to be constant during the simulation period of 40 years.

The intrusion of salt occurs due to rainfall on the salt heaps of Heitersheim and Buggingen. Salt intrusion from the sedimentation basins of Fessenheim are caused by leakage of the sedimentation basins. The salt intrusion from the French salt heaps in the potash basin was not included into the model. Equivalent salt simulations there have been established by BRGM (1998).

Table 3 shows the assumed boundary conditions for the salt transport model. The inflows from the potash heaps are assumed to be very small compared to the inflow from the sedimentation basins.

The inflow concentration of chloride under the potash dumps was assumed to be in the order of the maximum solubility of NaCl in water. The inflow regime from the sedimentation basins is unknown. It is supposed that the inflow concentration was also the maximum solubility. The intrusion of salt was assumed to be constant, although the filling of the sedimentation basins varied through the year, as the basins were used to store the salt brine when the Rhine was at ebb, before discharging it into the river.

Chloride measurements

Chloride measurements are necessary to verify the model results. The investigation of the salt plumes started in the sixties of the past century with 11 deep boreholes. Long term chloride measurements are available from these 11 boreholes (Geologisches Landesamt Baden-Württemberg 1997). Vertical profiles of the electric conductivity (conductivity logs) had been measured annually. The logs were measured in boreholes with deep screens and without pumping.

Breakthrough curves (see Fig. 10) have been derived from the conductivity measurements. The highest chloride concentrations have been found in the direct downstream region of the sedimentation basins at SA VII and IX at the bottom of the aquifer. The chloride concentrations of SA III and VI demonstrate that the plume did not reach this region yet. The boreholes of SA I, II, and IV, which do not reach the bottom of the aquifer, have low concentrations in the upper part of the aquifer. It is inferred that the salt plume moves underneath these boreholes.

Table 3 Boundary conditions of the salt intrusion areas: potash heaps Buggingen and Heitersheim, sedimentation basins of Fessenheim Island

	Inflow	Duration	
	(l/s)	From	To
Potash heap Buggingen	0.179	1957	End of simulation
Potash heap Heitersheim	0.209	1959	End of simulation
Sedimentation basins Fessenheim Island	25-44	1957	1976

Sensitivity analysis for vertical discretisation

The simulation of the vertical transport of the salt brine requires a sufficiently fine discretisation in the vertical direction. On the other hand a vertical refinement of the numerical mesh leads to an increased computer runtime. Therefore a sensitivity analysis has been performed in order to investigate the influence of the vertical discretisation on the simulation results. A model with nine layers and a model with 15 layers have been used for the sensitivity study. The nine layer model shows a discretisation similar to the Rhine transport simulation. The upper three layers of the nine layer model have been divided into six layers and the lower three layers also have been divided into six additional layers so that the model consists of 15 layers.

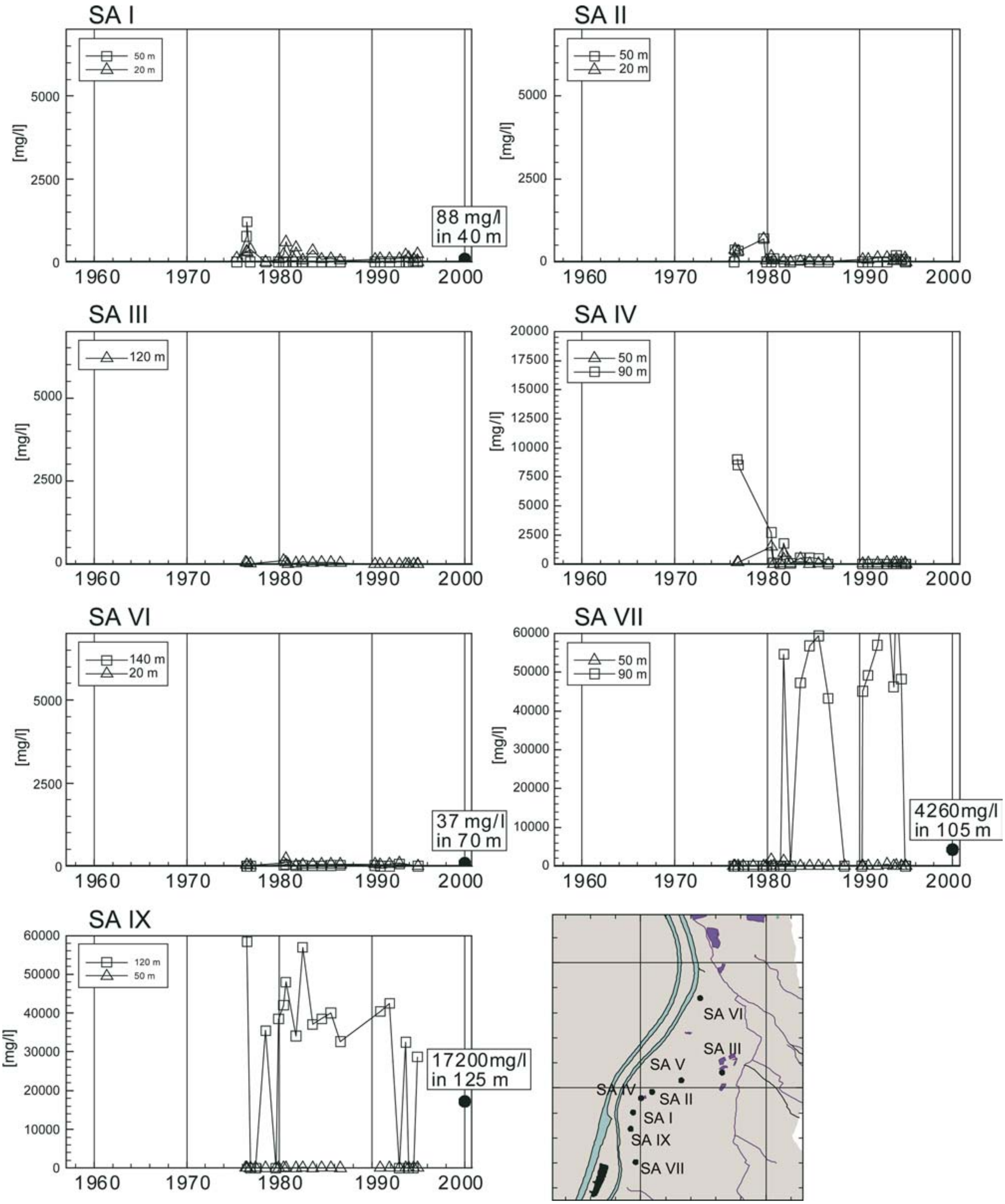
The different simulation results of the two models are shown in Fig. 11 as chloride breakthrough curves at two different depths in borehole SA VII, which is in the downstream region of the sedimentation basins. The salt concentrations of the 15 layer model are 5–10% higher during the infiltration process of the sedimentation basins. This was expected because of the higher vertical resolution, which gives a better approximation of gravity dominated transport processes in the intrusion region. Nevertheless the numerical error in applying the nine layer model was assumed to be small against the uncertainties of the available field data. Therefore the nine layer model was used for the modelling of the density driven salt transport in the Upper Rhine valley.

Estimation of parameters relevant for the regional transport of chloride

After the flow calibration the determination of additional aquifer parameters is necessary to simulate the transport processes of the salt brine. Therefore a sensitivity analysis or coarse calibration has been performed to estimate these parameters by comparing the observed chloride concentrations at the boreholes with the calculated values. The relevant parameters were changed during this calibration process, and the result of the analysis can be summarised as follows:

- Porosity for the three hydrogeological units: the porosities were assumed to be constant in each hydrogeological unit. The best results were achieved with 0.15 for the Neuenburg Formation, 0.1 for the upper part of the Breisgau Formation, and 0.08 for the lower Breisgau Formation.

Fig. 10 Observed chloride concentrations



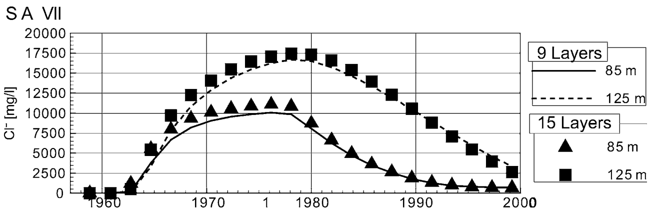


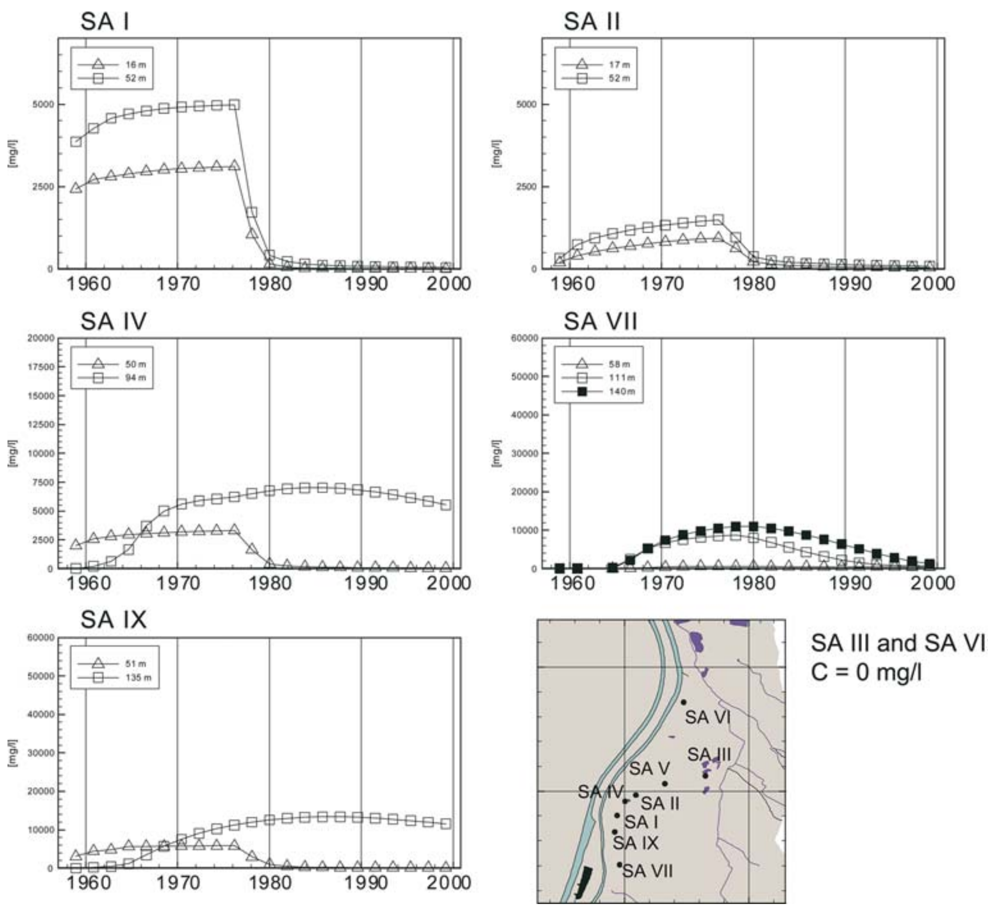
Fig. 11 Simulated chloride breakthrough curves with 9 and 15 layer model

- Dispersivities: it was assumed that the dispersivities are constant for all units and that the ratio between longitudinal and transverse dispersivity is 10. The analysis showed that longitudinal dispersivities higher than 40 m produce a too large spreading of the salt plume at the bottom of the aquifer. A value of 40 m seemed to be appropriate.
- Hydraulic conductivities for the Breisgau Formation: no unique value distribution could be determined from the flow calibration. The resulting conductivities are in the range of two orders of magnitude. The best fit between calculated and measured chloride breakthrough curves was achieved by multiplying the values from the flow calibration by 1/3.
- Anisotropy for the three hydrogeological units: the vertical anisotropy of the hydraulic conductivity was assumed to be constant in each hydrogeological unit. The anisotropy factor (vertical/horizontal conductivity) affects the vertical velocities and therefore leads to different chloride concentrations at the beginning of the intrusion. Since no data is available for that period, only a range of anisotropy factors from 0.03 to 0.1 in the Neuenburg-Formation and 0.03 to 1.0 in the Breisgau-Formation could be determined. An anisotropy factor of 0.1 was applied to the model.
- Leakage rate of the sedimentation basins at Fessenheim: the real leakage rates from the sedimentation basins are unknown, but this inflow boundary condition strongly affects the distribution of chloride in the numerical model. The variation of the leakage rate showed that the average inflow must be between 30 l/s and 44 l/s.

Comparison of observed and calculated chloride concentrations

Figure 12 shows the calculated chloride concentration curves which have to be compared to the chloride data in Fig. 10. The calculated and measured data do not match very well. Only the general trend seems to be similar. There may be two reasons for this: (1) The numerical model is only calibrated in detail for the

Fig. 12 Calculated chloride concentrations



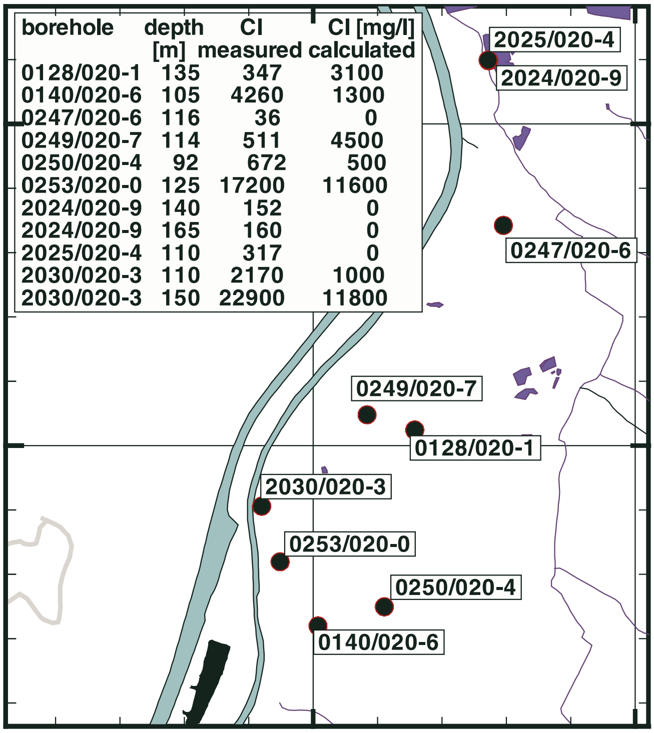


Fig. 13 Calculated and measured chloride concentration at the bottom of the aquifer for the year 2000

steady state flow field. The transport parameters are assumed to be homogeneously distributed and have been estimated by a sensitivity study. A detailed calibration with a differentiation of the parameters was not accomplished. (2) The measured breakthrough curves have uncertainties due to the fact that the chloride concentrations have been derived from measured electric conductivity by vertical profiles of the boreholes.

An additional direct comparison of calculated and measured chloride concentrations is given in Fig. 13, with chloride measurements from the year 2000 at additional boreholes in a depth lower than 90 m. The numerical model reproduces the general distribution with chloride concentrations higher than 1,000 mg/l in the local downstream region of the sedimentation basins. The calculated values differ on the order of 2 from the observed concentrations. The downstream region around boreholes 0249/020-7 and 0128/020-1 shows too high concentrations in the model. This demonstrates that the movement of the highly concentrated brine front is overestimated by the model in the present state. However, no better agreement was achievable with a homogeneous distribution of the porosity and without a detailed calibration of the parameters (conductivity, porosity, dispersivity) in the lower layers.

Chloride concentrations of zero were calculated at observation point 0247/020-6, which corresponds to actual measurements. The increased concentrations in borehole 2025/02-4 and 2024/020-9 on the bottom of the aquifer is not reproduced in the model. The model shows increased concentrations of chloride in the medium depth of the aquifer. It is assumed that a higher vertical dispersivity in the model would result in an increased vertical mixing and would therefore lead to higher concentrations at the bottom. The variation of the transverse dispersivity was not subject of the parameter estimation process.

Present day distribution of chloride

The present day distribution of chloride was determined by the numerical model and is presented in Fig. 14 for two different cases. Case 1: inflow from the sedimentation basins is 30 l/s. Case 2: inflow from the sedimentation basins is 44 l/s . The main mass of chloride can be found at the bottom of the aquifer downstream of the sedimentation basins. This indicates a slow movement of the salt pool in the last 20–40 years. Two new boreholes were placed on the basis of the simulation results at the boundary of the expected salt plume on both sides of the river Rhine. It is expected that in the future, increasing chloride concentrations will be found in the new boreholes.

The distribution at the bottom of the Neuenburg Formation (Fig. 14) shows low chloride concentrations (100–1,000 mg/l) with a plume moving in the direction of Breisach where two drinking water wells are located. The water pumped from these wells currently shows increasing chloride concentrations, which can be explained by the simulation results. The plume of low chloride concentrations is caused by a dispersive exchange process between fresh water and the salt brine pool, which is induced by the natural groundwater flow field.

The numerical model was used to determine the present total mass of chloride in the groundwater. The calculated total mass of chloride is nearly 1 million tons of chloride. Eighty percent of the chloride is stored in the lower part of the Breisgau Formation. The rest is dissolved in the low concentration downstream plume.

Summary and outlook

Salt heaps and sedimentation basins were installed in the middle of the last century by the potash mining companies of Germany and France in the southern Rhine valley. Due to salt water infiltration from the salt heaps and leakage in the sedimentation basins, chloride was introduced into the aquifer of the southern Upper Rhine Graben. A numerical model was set up to investigate the propagation of the chloride plumes. The basis of the numerical model is the conceptual model derived from detailed hydrogeological mapping of the investigation area. Three major hydrogeological units of relevance for the flow and transport processes have been identified for the porous aquifers. The numerical model was calibrated for steady state flow conditions and validated with additional data from isotopic studies on Rhine water infiltration and from transient flow conditions during

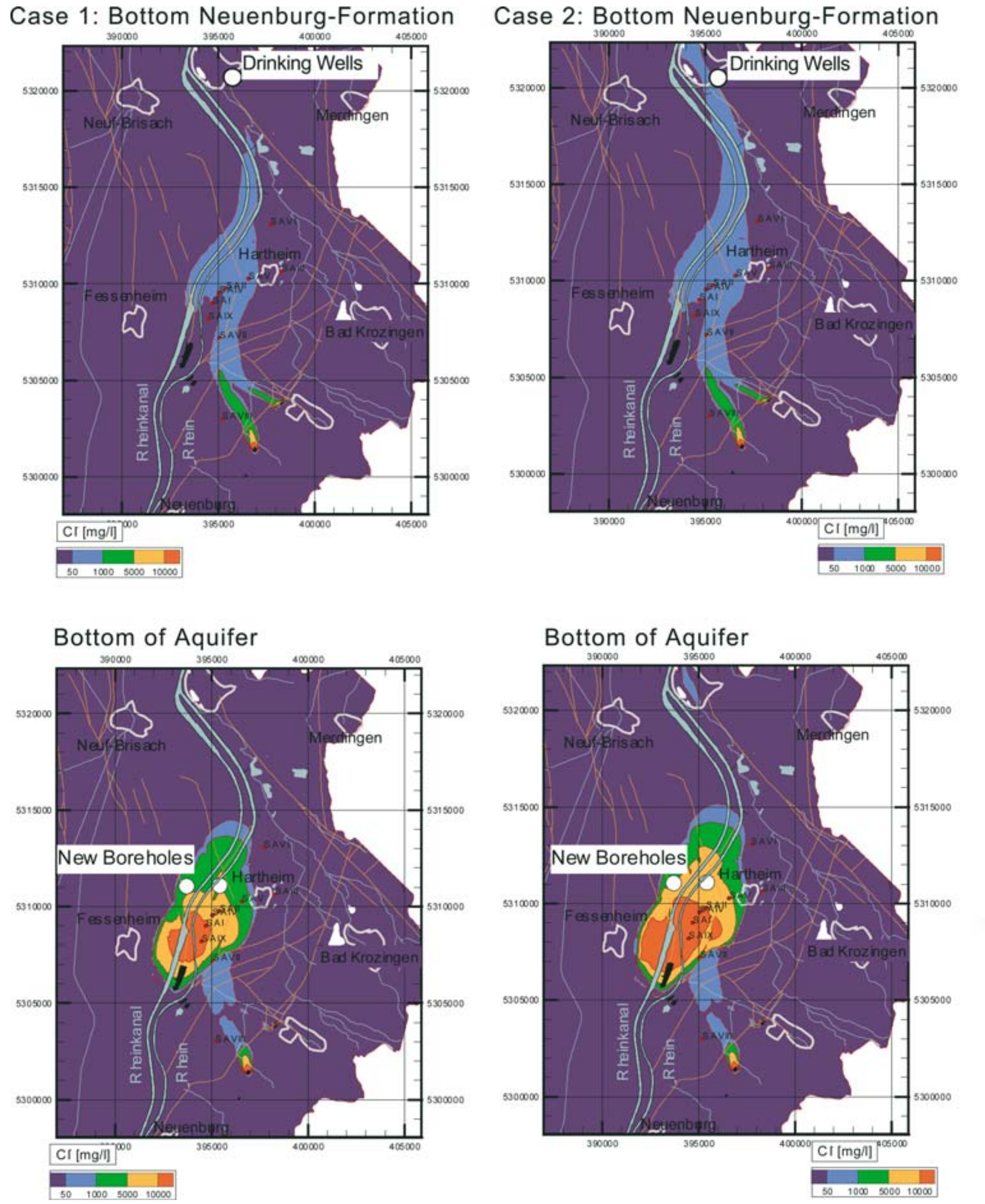


Fig. 14 Calculated chloride concentrations at the bottom of the aquifer and the bottom of the Neuenburg-Formation

the flood event of 1987 in the river Rhine. A rough distribution of the transport parameters has been determined by comparing the observed and calculated chloride breakthrough curves, which have been derived from annual logging of electric conductivity in 11 deep boreholes. The historical breakthrough curves could be simulated only qualitatively. A better agreement between calculated and observed chloride concentrations has been achieved with actual measurements from the year 2000. The first estimation of the current chloride distribution was used to define the position of two new boreholes for further investigations. It is planned to improve the existing numerical model with the hydrogeological data from the additional boreholes and to enhance the flow model with a long term flow calibration. The detailed distribution of the relevant transport parameters will be achieved by an additional calibration of the chloride transport. It is assumed that the prediction of the salt plume movement will be possible on the basis of these future model enhancements.

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