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GEOSPHERE MIGRATION STUDIES AS SUPPORT FOR THE COMPARISON OF CANDIDATE SITES FOR DISPOSAL OF RADIOACTIVE WASTE IN ROCK- SALT

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The Dutch research program on the geological disposal of radioactive waste was designed to supply a basis for the selection of combinations of three factors, i.e., type of rock-salt formation, site, and disposal technique, satisfying radiological standards and other criteria for final disposal. For the present phase of the research program, no field work is foreseen. Due to the large number of potential sites and the lack of site-specific data, a complete safety assessment for each location cannot be completed within the time scheduled for the project. Therefore, the potential sites have been grouped according to the type of rock-salt formation (e.g. bedded salt and salt domes) and two classes of depth below the surface of the ground. Values for geohydrological parameters

were obtained by extrapolation of data from existing boreholes and analysis of the sedimentary environment. Experiments were carried out in soil samples from formations in Belgium and Germany, which are comparable to those under study in The Netherlands, to obtain retardation data for radionuclides. A three-dimensional model of groundwater flow and contaminant transport, called METROPOL, has been developed. A particle-tracking facility provides a quick impression of the main flow direction and travel times. Because any release pathway passes through zones differing in salinity, temperature, pressure, and lithology, the model was designed to take these factors into account.

To investigate the effect of high salinity on nuclide transport properly, a theoretical experimental study was carried out. Use of a thermodynamic approach showed that terms related to salt mass fraction have to be added to Darcy's and Fick's laws. These terms give rise to higher rates of nuclide mass flux if they exceed certain critical values. Thus, neglect of these terms may lead to underestimation of nuclide mobility. An experimental study to investigate effects of these modifications is in progress.

INTRODUCTION

The Dutch research program for geological disposal is intended to supply a basis for assessment of the feasibility of disposal of radioactive waste in rock-salt. This research program was designed by the Onshore Disposal Study Committee (OPLA)¹, and is divided into three stages:

1. inventory and comparison of potential disposal concepts and sites;
2. preliminary field research in the area of selected sites; and
3. above-ground field research of a chosen site.

Specific items for research in the first stage include the disposal techniques, the types of rock-salt formation, and the local geological and geohydrological situations. The findings were expected to facilitate the selection of an optimal combination of formation type, location, and disposal technique in terms of radiological impact. However, during the course of the research it has become evident that an absolute selection and ranking of sites cannot be achieved. This is due partly to the lack of data, but especially to the uncertainty associated with the prediction of the future behaviour of a disposal site. The migration of

radionuclides can be expected to lead to overlapping ranges of radiation doses. The main item in the research program is, of course, the safety evaluation. A number of studies have been designed to support the safety evaluation. Besides the research now being performed in The Netherlands by several research institutes, universities and consulting companies, active participation in international programs has been sought.

For almost all disposal concepts, the primary accident scenario comprises flooding of the repository followed by squeezing of the contaminated brine out of the salt formation. Because geosphere migration is considered to be the indispensable link between near-field release studies and environmental pathway studies, special attention was given to this subject.

The present paper reviews the relevant Dutch research work in the field of geosphere migration. It deals with the collection of geohydrological as well as geochemical data, model development, and model validation.

THE GEOHYDROLOGICAL INVENTORY

In view of the purpose of the first stage of the Dutch research program for underground disposal, no field work was carried out. To obtain as much data for the safety assessment study as possible, an inventory of all accessible geohydrological data was made. The database stored in a computer permits easy handling of data needed for model calculations of geosphere transport. The database can be used to construct geohydrological cross-sections. The database can also be used to obtain insight into the variability of a parameter. Figure 1, for instance, gives the distribution of the permeability of one of the aquifers in the Lower Tertiary, which overlies several salt structures. This information will be of use for the eventual calculation of the effects of parameter variation considered in the safety assessment. So far the database has been limited to the Tertiary. The available data concerning older geological formations are not sufficiently numerous or detailed to be suitable for inclusion in the database. The inventory of Upper-Paleozoic and Mesozoic data made it very clear that additional techniques are

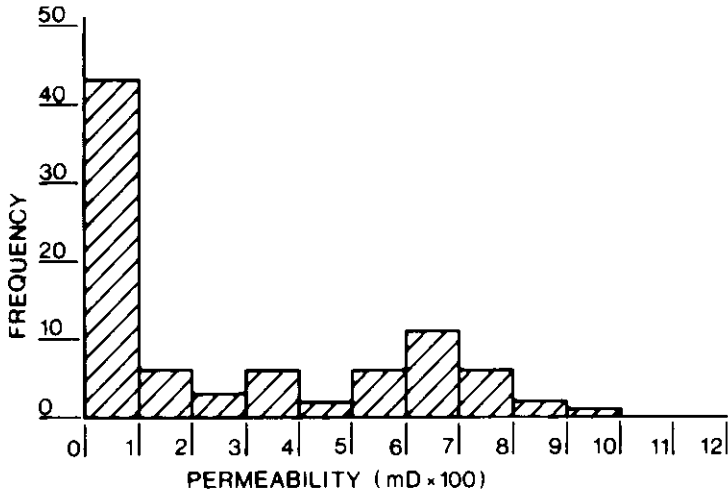


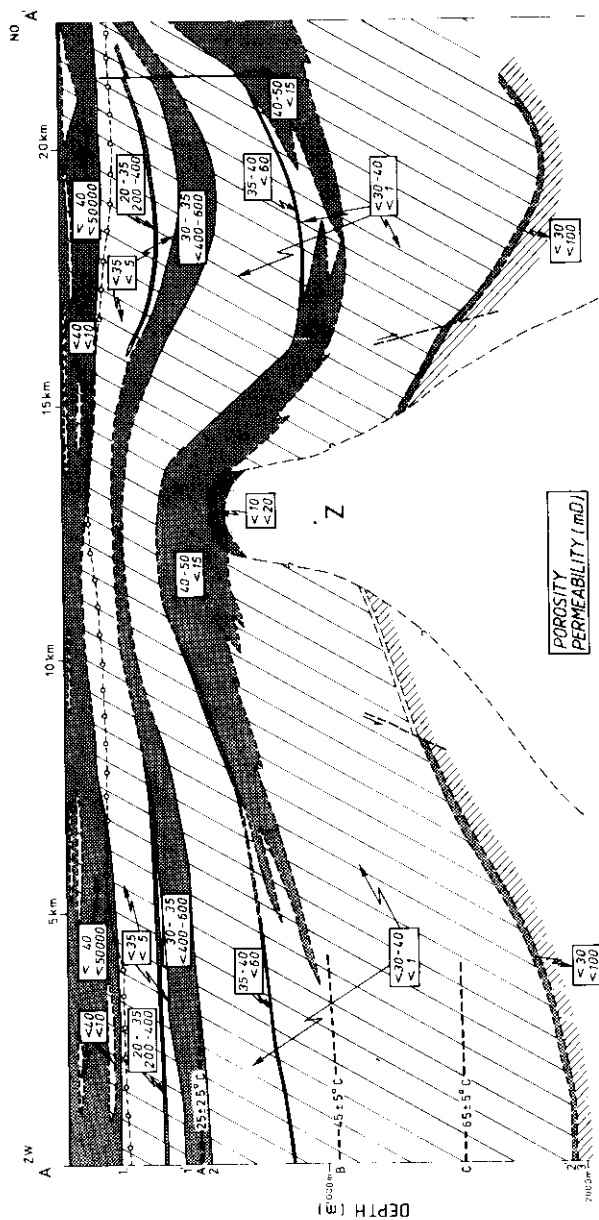
FIGURE 1. Example of distribution of available permeability data

required to quantify input data for model calculations. Because interpolation and extrapolation methods could not be applied due to lack of data on these formations as well as the dubious reliability and wide spread of the few available data, we used the method of reservoir analysis. This method which recognizes the importance of the sedimentary environment is based on study of genesis, geological history, hydrochemistry of areas for which data are scarce, and yields a spatial distribution of their hydrogeological characteristics. The results obtained with this approach are presented here in the form of a hypothetical cross-section over a salt-dome (Figure 2).

Since the uncertainty of the data increases rapidly with depth, parameters are given in the form of ranges of values derived by reservoir analysis². The data presented here concerning the disturbed zone surrounding salt domes are inaccurate.

GEOHYDROCHEMISTRY

For the prediction of radionuclide migration patterns in the geosphere, geochemical data are needed. These data can be partially obtained from



laboratory experiments under conditions expected to occur in Dutch aquifers. The findings are expressed in terms of so-called K_d values. Four radionuclides were studied: Am, Pu, Np, and Tc. The impact of several parameters, such as pH, redox potential, salt concentration, colloidal and organic matter content, iron-containing particles and microbiological activity was assessed. The geological materials used for this purpose were glauconite-containing sand from Belgium and soil material from boreholes above the Gorleben salt-dome (FRG). The main laboratory techniques were column and batch experiments. Batch experiments provided a rather easily obtainable description of the impact of certain parameters, and the more laborious column experiments permitted more reliable and detailed description of certain cases³. For special purposes, additional experiments were performed as well.

A very strong impact on radionuclide mobility was found to be exerted by the organic matter and the iron-containing particles in the soil. The organic matter in the Gorleben soils proved to lead to very similar behaviour of all nuclides when the redox potential is sufficiently low (about -300 mV)⁴. Adsorption on the glauconite-containing soil, which was free of organic matter, seems to be strongly influenced by iron-containing particles (Figures 3 and 4). The migration of these iron-containing fractions cannot be described by equilibrium chromatography and it is impossible to describe co-migration of nuclides with these fractions in terms of K_d values, because the K_d concept is based on chromatographical equilibrium adsorption/migration behaviour. The tendency of the nuclides to migrate in several fractions each with its own mobility makes it necessary to describe the migration of one radionuclide with more than one K_d value. It is believed, however, that on a geological time scale conversions between these fractions will lead to a single average K_d value per nuclide. The K_d values derived from experiments are given in table I. These values are assumed to be representative for the sandy aquifers near salt formations in The Netherlands.

Because these data were restricted to samples of sandy formations from other countries, additional data on retardation are currently being collected from the international sorption information retrieval system (ISIRS) of the Nuclear Energy Agency.

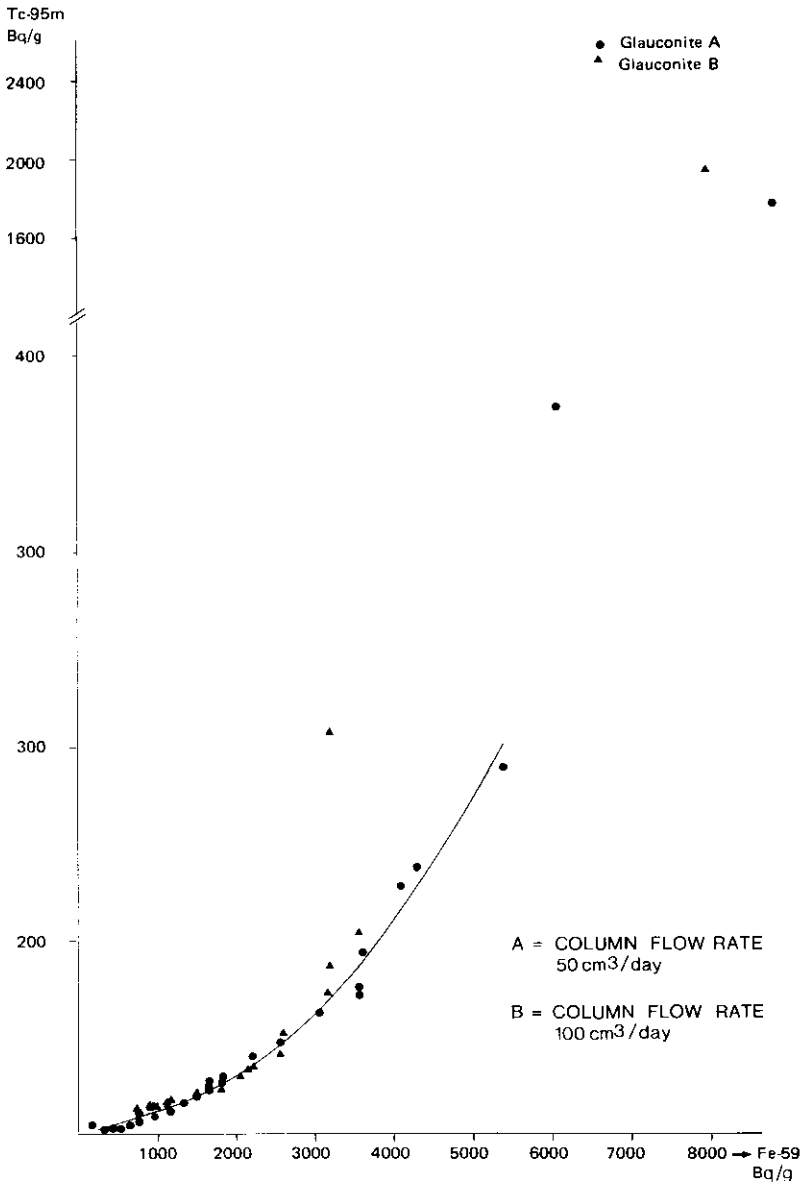


FIGURE 3. Adsorption of Tc as function of Fe content

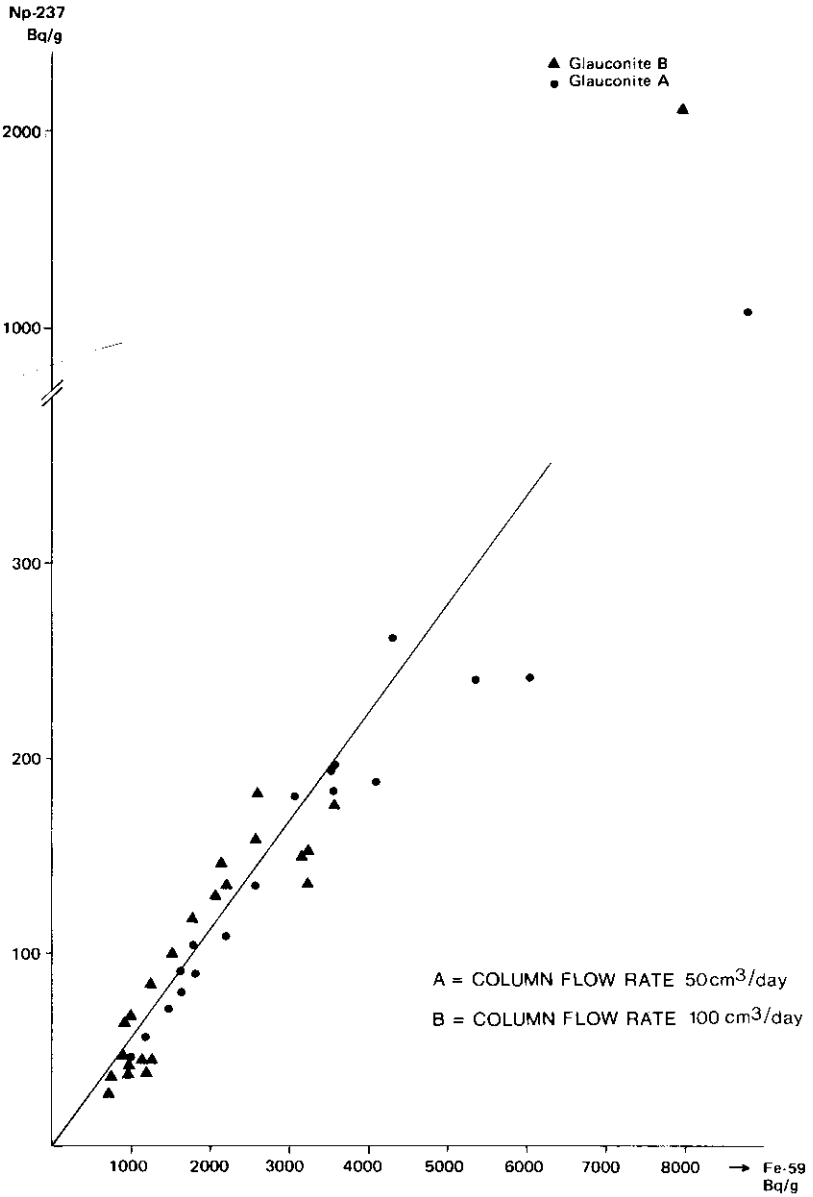


TABLE I
 K_d values found for porous sediments resembling those associated with Dutch aquifers

radionuclide	K_d (cm ³ /g)
Am	1000
Pu	4000
Np	60
Tc	30

PHYSICS OF FLOW AND TRANSPORT; THEORETICAL BACKGROUND AND EXPERIMENTAL VERIFICATION

A vital part of any mathematical model is the physical description of the problem(s) to be solved. This includes the description of the system being considered and physical processes occurring in that system. Physical processes occurring in geological media in the vicinity of salt formations are complex. Often, coupled processes take place and extreme conditions prevail. An important factor in this respect is the presence of high salt concentrations, which is bound to affect the diffusion, dispersion, and adsorption of radionuclides moving away from a salt formation.

It is known that both the density and viscosity of water increase with increasing salt concentration. Both of these effects tend to slow down the upward flow of groundwater. This is evident from Darcy's law, which can be written as

$$\mathbf{q} = -\frac{k}{\mu}(\nabla p - \rho \mathbf{g}),$$

where \mathbf{q} is the Darcy velocity,

k is the intrinsic permeability of the medium,

μ is the dynamic viscosity of brine,

p is the pressure,

ρ is the mass density of brine, and

\mathbf{g} is the gravity vector pointing downward.

Furthermore, the question arises whether the basic equations (in

particular Darcy's law and Fick's law) hold under such extreme conditions. To answer this question, we applied a thermodynamic approach to derive basic governing equations for the flow of high-concentration brine in soil⁵. This approach, combined with principles of conservation of mass and momentum as well as the first and the second laws of thermodynamics, yielded extended versions of Darcy's and Fick's laws. It is concluded that if there is a high salt concentration gradient, one may register a total mass flux for brine even under a hydrostatic pressure distribution. This extraneous mass flux is due to the movement of salt under its own concentration gradient. To account for this effect, not only the usual pressure-gradient term but also a term proportional to the salt mass fraction is introduced into Darcy's equation, which gives:

$$\mathbf{q} = -\frac{k}{\mu}(\nabla p - \rho \mathbf{g}) - D^f \nabla \omega^s, \quad (2)$$

where ω^s is the mass fraction of salt and D^f is the coefficient of density flow.

Similarly, a term depending on the gradient of the salt mass fraction is added to Fick's law for radionuclides. This term accounts for the fact that the salt carries the radionuclides along as it moves through a medium. For assessment of the potential effects of these modifications of basic equations, a number of numerical "experiments" were performed, using a one-dimensional model called TORISM. As Figure 5 shows, beyond a critical value of a dimensionless combination of the system properties, the effect of additional terms becomes pronounced⁶. In this figure, ω_0^s and ω_0^r are mass fraction of salt and radionuclide, respectively, of the inflow fluid.

In such cases, the advective and dispersive mass flux of radionuclides increase due to the enhanced mass flux of the brine. Determination of the validity of these modifications for any real situation requires laboratory and/or field experiments. A physical model was therefore designed and set up to investigate such effects experimentally. Figure 6 gives a schematic representation of the model. As indicated there, flow in the model is upward. For these experiments the model will be filled with fresh or brackish water, which in due course will be displaced by highly concentrated brine. These experiments will provide an assessment of the validity of Darcy's and Fick's laws under high-

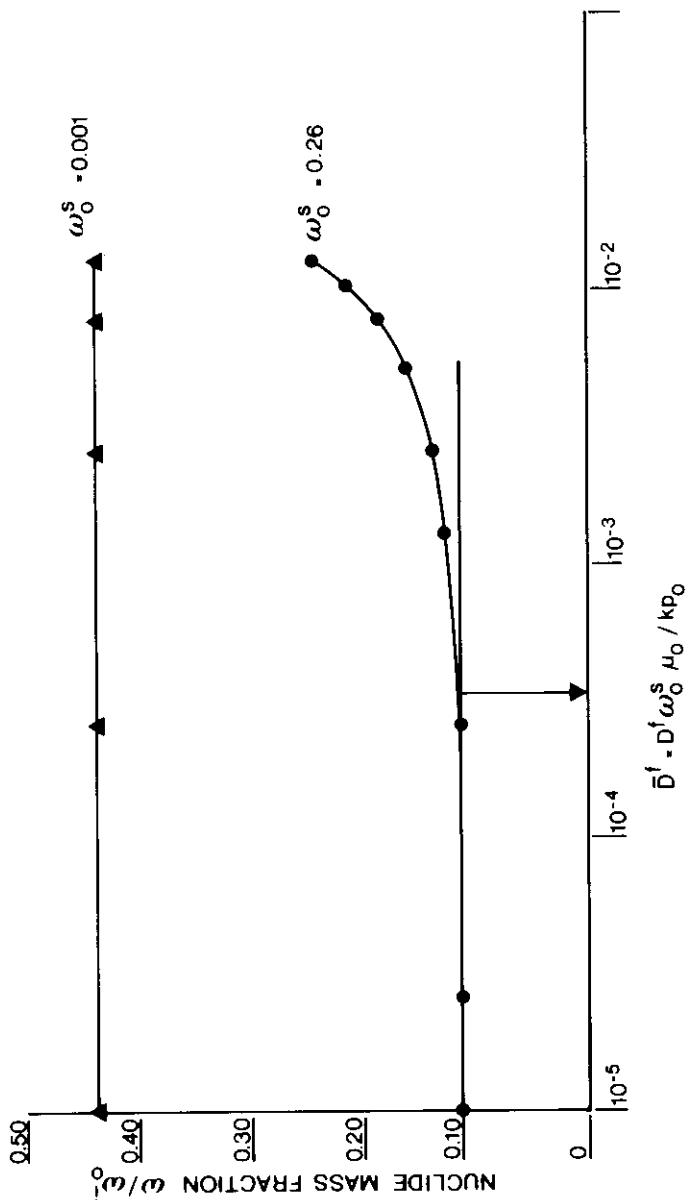


FIGURE 5. Influence of variation of the extra term D^f in Darcy's law on nuclide peak value

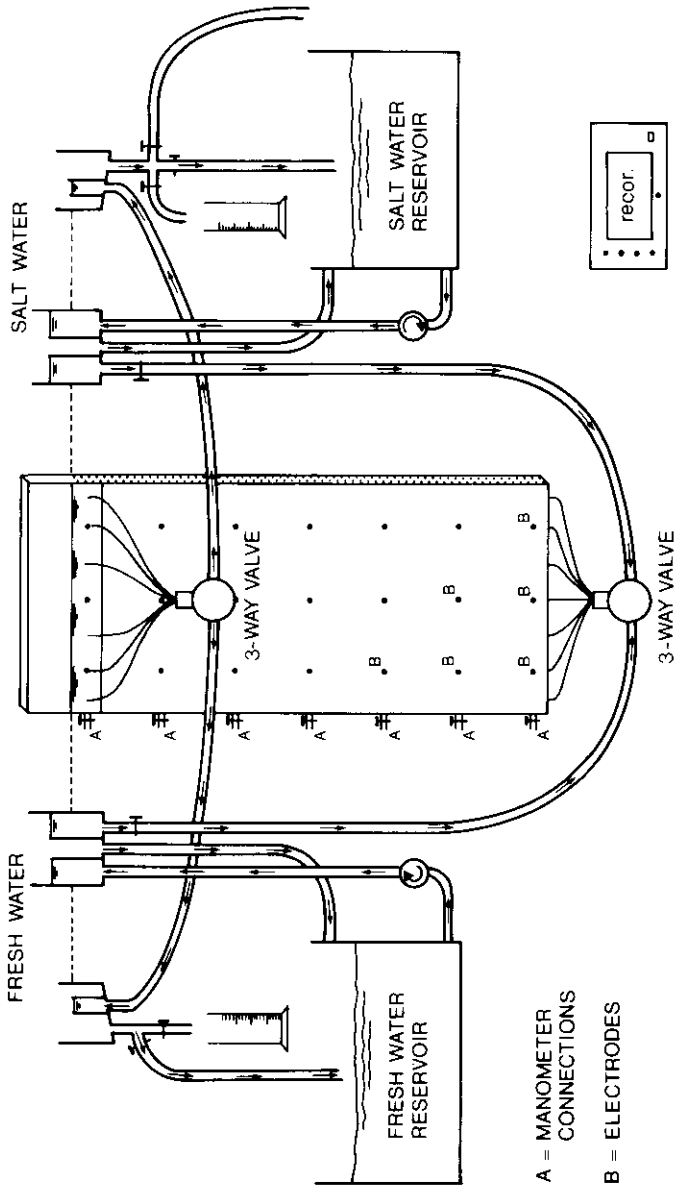


FIGURE 6. A scheme of the laboratory experimental set-up for measuring flow under varying density

concentration conditions. These experiments are in progress, and preliminary results are expected by the end of 1987.

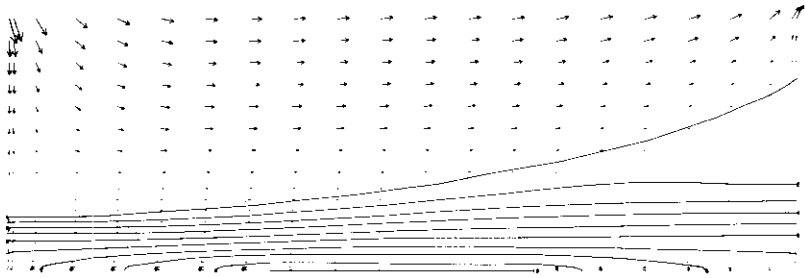
A NUMERICAL MODEL: METROPOL

For the investigation of subterranean radionuclide migration, a numerical model simulating the flow processes in the geosphere is essential. Such a model should have the ability to handle several dissolved species and to take into consideration the interaction between nuclide migration, salinity, and sorption. These demands could not easily be met by existing models, and therefore a new three-dimensional model of groundwater flow was developed at RIVM. This package is called METROPOL, which stands for simulation METHod for the TRansport of POLLutants^{7,8}. At present, METROPOL package comprises of several subpackages developed for mesh generation, computation of steady-state groundwater flow, simulation of transient flow, and the solution of coupled flow and transport equations. The METROPOL package is a fully 3D finite element model. The flow domain is subdivided into a number of small elements, each having the shape of a "deformed" cube. This provides a rather flexible method for modelling a complex geological structure combined with a simple structure for the element grid.

Some recently developed numerical methods promote the numerical solution of the problem, and a version adapted for running on a very fast vector computer (CRAY-XMP) is now available.

Special care has been taken to model correctly the flow of groundwater containing a high concentration solute (e.g. salt in aquifers near salt formations). The numerical model solves two nonlinear coupled equations for p and ω . A mathematical transformation has been introduced to decrease the influence of the non-linear terms in the flow and transport equation, and an iteration between the two equations is performed. Several postprocessing packages (mass balance computations, particle tracking, plotting of computed results) are also available.

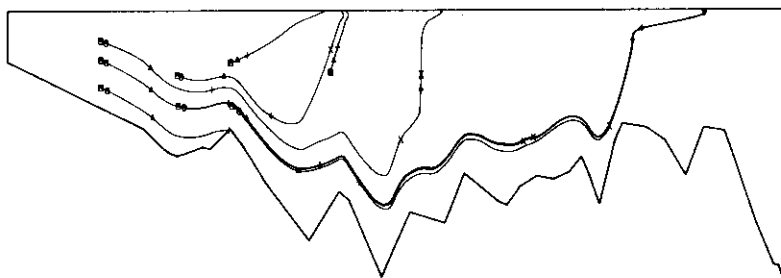
A few examples of the possibilities of METROPOL are given in Figures 7 and 8. The first example concerns a cross-section through a domain in which salt is transported away from part of the bottom



General parameters	contours
Date 24-JUN-87	number value
Scaling factors	1 1.000 10 ⁻⁰¹
scale in x-direction	2 2.000 10 ⁻⁰¹
1 30.00	3 3.000 10 ⁻⁰¹
scale in y-direction	4 4.000 10 ⁻⁰¹
1 30.00	5 5.000 10 ⁻⁰¹
Corners of the mesh	6 6.000 10 ⁻⁰¹
lowerleft .1	7 7.000 10 ⁻⁰¹
lowerright .23	8 8.000 10 ⁻⁰¹
upperright .1265	9 9.000 10 ⁻⁰¹
upperleft .1243	10 1.000 10 ⁺⁰⁰
Velocity in plane	
→ (1 cm) = 2.16 10 ⁻⁰⁷	
Max. V = 2.162 10 ⁻⁰⁷	
time for contouring	
T = 6.15543610 +10	
cut-off value for	
contour = 0.00 10 ⁺⁰⁰	
contourlines for	
concentrations	

FIGURE 7. Concentration contours in a hypothetical cross-section above a salt dome, calculated with the METROPOL computer code

surface of the domain. The steady-state salt contours and the velocity field are shown in Figure 7. This example was one of the test cases for the international hydrological model intercomparison study HYDRO-COIN, in which RIVM participated with the METROPOL model. Further study of this case is needed because the geometry of the domain



General parameters	markers trajectory
Date : 15-SEP-87	marker time
Scaling factors scale in x-direction 1 : 5685.00 scale in y-direction 1 : 360.00	5 1.902-10 ⁰³
	6 1.246-10 ⁰⁴
Corners of the mesh lowerleft : 43 lowerright : 84 upperright : 1260 upperleft : 1219	▲ 1.128-10 ⁰⁶
	+ 2.488-10 ⁰⁸
	X 1.150-10 ⁰⁸
	◆ 1.873-10 ⁰⁸
	+ 2.194-10 ⁰⁸
	X 2.282-10 ⁰⁸
	Z 4.533-10 ⁰⁸

FIGURE 8. Flow traces and travel times of particles after release from salt formations at different depths and locations, calculated with the METROPOL computer code

and the boundary conditions will probably have especially strong influence on the flow pattern.

Another example concerns a cross section through part of The Netherlands. This case was calculated for fresh water only. Figure 8 shows the calculated flow traces of a few particles followed from release points on different depths and locations within the domain. Travel times depend strongly on the local groundwater pattern in the upper layers. Several cases are currently taken into consideration for modeling of these upper layers. Furthermore, within the framework of the PAGIS project of the European Communities, model calculations for groundwater flow and travel times for a schematic situation derived from the German Gorleben site, have almost been completed.

Further research is now focussed on the incorporation of retardation and decay processes into the code. The shape of the flow as well as transport equations and the type of boundary condition in cases with high density flow are also subjects under investigation.

CONCLUSIONS

In the field of geosphere migration a number of projects are in progress. They serve to provide support for the safety evaluation of final disposal of radioactive waste in salt formations. A database of geophysical parameter values has been entered in a computer, which facilitates handling of data to be used as input for migration modelling. With additional information obtained by reservoir analysis, hypothetical cross-sections through all of the salt structures under study have been constructed. Indications of ranges of parameter values are given. These profiles proved to be very useful in the design of migration models.

Geochemical experiments concerned the effects of variations in e.g. salinity, iron content, organic compounds, and Eh. Presence of iron severely complicates the description of migration processes. The fact that a radionuclide can migrate in different forms makes it necessary to use multiple K_d values.

Because coupled processes in domains where high densities occur might affect the diffusion, dispersion, and adsorption of radionuclides, careful consideration must be given to the basic equations, such as Darcy's and Fick's laws. Extended versions of both laws have been derived, and an experimental validation of the findings is being carried out.

For the predictive calculations on the migration of radionuclides from the near-field domain to the biosphere, a new three-dimensional model for transport of contaminants, called METROPOL, was developed. Because model validation is essential to confirm the reliability of the calculations, several supporting projects have been started. Active participation in the international code intercomparison study HYDRO-COIN was the first step in this direction. The physical value of the theoretical extensions of the basic equations governing the transport code is being established by laboratory experiments on saturated brine flow.

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