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Critical parameters controlling pollutant migration from waste disposal sites: Field investigations and influence on aquifer pollution

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Key words: Groundwater, pollutant transport, waste disposal, heterogeneity, barrier, safety, field investigations, modelling

ZUSAMMENFASSUNG

Der vorliegende Artikel betrachtet jene Prozesse, welche für den potentiellen Schadstoffaustrag aus vier verschiedenen Deponietypen in der Schweiz relevant sind. Es werden die folgenden Typen betrachtet: Reaktordeponie für Hausmüll, Reststoffdeponie für Rauchgasreinigungsrückstände, nukleare Endlager für schwach- und mittelaktive Abfälle (SMA) bzw. für hochaktive Abfälle (HAA). Ein Vergleich der Dauer der Schadstofffreisetzung mit der erwarteten Lebensdauer der künstlichen Barrieren zeigt die Wichtigkeit der geologischen Barriere für die langfristige Sicherheit der Deponien auf. Die Technische Verordnung über Abfälle schreibt für die geologische Barriere nur gemittelte hydraulische Parameter vor. Obwohl diese durchschnittlichen Eigenschaften die Stabilität der künstlichen Barrieresysteme beeinflussen können, reichen sie nicht aus, um die langfristigen Umwelteinflüsse einer Deponie zu beurteilen, da der Transport von Schadstoffen in der Geosphäre hauptsächlich durch Heterogenitäten und präferentielle Fliesswege bestimmt ist. Der Artikel beschreibt Feldmethoden zur Charakterisierung solcher Heterogenitäten und zeigt auf, dass nahezu alle typischen geologischen Barrieren in der Schweiz stark heterogen sind und präferenzielle Fliesswege (Klüfte, Störungen, grobkörnige Sandsteinhorizonte) aufweisen. Eine Sensitivitätsstudie mittels einem numerischen Modell und Parametern eines potentiellen Deponiestandortes in der Nordschweiz zeigt, dass eine Vernachlässigung oder falsche Einschätzung solcher präferentieller Fliesswege zu einer um mehrere Grössenordnung falschen Einschätzung der Durchbruchszeiten und Durchbruchskonzentrationen der Schadstoffe führen kann.

ABSTRACT

This paper considers processes which are of relevance to contaminant release from four types of waste disposal sites in Switzerland: disposal sites for municipal solid waste, for solidified filter ashes, for low- to intermediate-level radioactive waste (SMA), and for high-level radioactive waste (HAA). A comparison of contaminant release times and expected lifetimes of engineered barriers underlines the importance of the geological barrier for long-term disposal site safety. With respect to the geosphere, current regulations concerning non-radioactive waste disposal in Switzerland (TVA) relate only to bulk (effective) hydraulic properties. Although bulk properties may influence the stability of engineered barriers, they are inadequate for the evaluation of the disposal site's long-term impact because contaminant migration in the geosphere is controlled primarily by heterogeneity and preferential pathways. Field methods are described which allow such heterogeneities to be detected and characterized, and are illustrated by data which suggest that practically all geological barriers considered in Switzerland include preferential pathways (fractures, faults, coarse-grained layers). A sensitivity analysis using a numerical transport code and parameters from a field investigation for a potential landfill site in Northern Switzerland shows that neglecting the existence of preferential pathways may lead to severe errors when estimating contaminant breakthrough times and peak concentration levels.

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1. Introduction

Waste disposal in Switzerland and in most developed countries employs the multiple barrier system to ensure long-term post-closure safety: the waste is emplaced within a series of engineered barriers (compacted clay layers, synthetic liners, cement, bentonite, drainage systems etc.), which in turn are embedded in a geological barrier (for example a clay aquitard). This general concept holds for both principal Swiss waste disposal types, namely landfills for non-radioactive wastes and underground repositories for radioactive waste. The primary objective of both types of waste disposal is to "concentrate and confine" although dilution of contaminants in the event of release to the biosphere, which cannot be excluded for long time scales, is an important aspect of the corresponding safety analyses.

While safety requirements for a radioactive waste repository are defined in terms of an individual dose unit (10 mrem per year) which must not be exceeded at any time (KSA 1990, guideline R-21), those for non-radioactive waste disposal are defined in terms of elemental leachate and waste concentrations, and in terms of engineered and natural barrier hydraulic conductivity and thickness (TVA 1990). For Swiss radioactive waste, the R-21 guideline requests final disposal with no long-term surveillance. This objective leads to the concept of deep underground disposal in geological formations. In contrast, chemically toxic wastes are in general continuously monitored and the waste deposits are at land surface and only covered with engineered layers of small thickness.

Because the geosphere is a natural and therefore complex system, it may be tempting to overlook its properties and to rely primarily on the integrity of engineered barriers for waste confinement. In this paper we show that, because the duration of contaminant release from waste disposal sites may substantially exceed the lifetime of engineered barrier systems, the geosphere is an essential component for the long-term safety of a waste disposal site and as such it should not be neglected. This point is illustrated by comparing modelled or estimated contaminant release times for various waste types i.e. municipal solid waste, solidified filter ashes (Reststoffe according to TVA 1990), low- to intermediate-level radioactive waste (SMA), and high-level radioactive waste (HAA).

With the importance of the geosphere for long-term waste disposal safety in mind, it becomes apparent that a minimal understanding of the geosphere's complexity is warranted. This paper summarizes which geosphere characteristics (parameters) are the most critical for pollutant migration from a waste disposal site. The major influence of hydrogeologic heterogeneity on long-term migration is emphasized and field methods which can be used to characterize it are presented and illustrated by data measured in real field situations.

Finally, the influence of hydrogeologic heterogeneity on aquifer pollution is illustrated by performing a sensitivity analysis of pollutant migration with a numerical model. Parameters for the analysis are selected based on a field investigation for the characterization of a potential landfill site for solidified filter ashes in Switzerland.

2. Efficiency of engineered and geological barriers

2.1 *Temporal evolution of waste toxicities*

Whereas in the case of radioactive waste, the long-term evolution of the waste's toxicity can be predicted precisely based on the radionuclide half-lives, the long-term evolution of chemical waste can only be estimated by extrapolating into the future observations made over relatively short periods of time, primarily with the help of models and occasionally using natural analogs. Table 1 attempts to compare temporal evolutions of radioactive and non-radioactive wastes. Such a comparison is of course difficult because a uniform set of units concerning toxicity cannot be defined. In Table 1, the Swiss running water quality standard (Swiss Ordinance 1975) is selected as a reference toxicity level for non-radioactive waste whereas for radioactive waste the reference toxicity is the natural radioactivity of the host rock surrounding the repository. The unit for the radioactivity (ARI) is the number of 10 mrem dose equivalents which would result from ingestion of representative rock or waste masses (NAGRA 1985).

According to Belevi & Baccini (1989), of all the contaminants found in municipal solid waste, organic carbon (C_{org}) has the longest modelled release time at significant concentrations. The results of their modelling suggest that the time required for C_{org} in the leachate to drop below the quality standard is in the range of 500 to 1,700 years. In the case of cadmium, a heavy metal present in landfills for solidified filter ashes (Reststoffdeponie), the estimated release time according to IMRA (1991) is much longer (>0.5 million years) and is related to the dissolution of the cement which is mixed with the waste to stabilize it. For low- and intermediate-level radioactive waste (SMA, NAGRA 1985), the modelled radionuclide release time is comparable to the release time of C_{org} from a municipal solid waste site (on the order of 1,000 years), whereas for high-level waste (HAA) the modelled release times are similar to those for solidified filter ashes (on the order of a few million years).

The duration of cement dissolution is generally estimated based on equilibrium carbonate solubility and water flux calculations which do not quantitatively consider alteration due to ageing processes which are very complex to describe. As the modelling analyses which led to the values in Table 1 are mostly based on simplifying assumptions, the results are highly uncertain. This is particularly apparent when comparing the release time estimated in the IMRA study (>0.5 million years), to the cement dissolution time estimated in NAGRA (1985) for an underground repository (10,000 to 100,000 years). Yet one would expect cement to have a far greater lifetime in the case of an underground repository than in a surface landfill where the hydrologic system is much more aggressive.

Parameter	Municipal Solid Waste	Solidified Filter Ashes (Reststoffe)	Low-Level Radioactive Waste (SMA)	High-Level Radioactive Waste (HAA)
Initial Toxicity	750 mg C_{org} /l Leach. [1]	0.06 mg Cd/l Leach.[2]	$3 \cdot 10^{13}$ ARI[3]	$2 \cdot 10^{16}$ ARI[3]
Toxicity after 100 y	600 mg C_{org}/l Leach. [1]	0.06 mg Cd/l Leach.[2]	$3 \cdot 10^{13}$ ARI[3]	$4 \cdot 10^{15}$ ARI[3]
Toxicity after 1'000 y	100 mg C_{org}/l Leach. [1]	0.06 mg Cd/l Leach.[2]	$5 \cdot 10^{12}$ ARI[3]	$6 \cdot 10^{14}$ ARI[3]
Toxicity after 10'000 y	0 mg C_{org}/l Leach. [1]	0.06 mg Cd/l Leach.[2]	$6 \cdot 10^{11}$ ARI[3]	$6 \cdot 10^{13}$ ARI[3]
Toxicity after 100'000 y	0 mg C_{org}/l Leach. [1]	0.06 mg Cd/l Leach.[2]	$1 \cdot 10^{11}$ ARI[3]	$3 \cdot 10^{13}$ ARI[3]
Toxicity after 1'000'000 y	0 mg C_{org}/l Leach. [1]	> 0.06 mg Cd/l Leach.[2]	$1 \cdot 10^{10}$ ARI[3]	$3 \cdot 10^{13}$ ARI[3]
Reference Toxicity of Quality Standard	20 mg C_{org}/l (Running Water)	0.005 mg Cd/l (Running Water)	$3 \cdot 10^{12}$ ARI (Limestone)	$2 \cdot 10^{13}$ ARI (Granite)

Tab. 1. Modelled temporal evolution of waste toxicities for different waste types. [1] Belevi & Baccini (1989), [2] IMRA (1991), [3] NAGRA (1985). ARI is the number of 10 mrem effective dose units.

2.2 *Lifetimes of engineered barriers*

Long-term predictions concerning the behaviour of engineered barrier systems must also rely on model results and are therefore inherently uncertain. In the case of non-radioactive waste disposal sites, estimated barrier lifetimes are often based primarily on "expert opinions" which are sometimes contradictory. For example while some experts believe that the latest synthetic liners have lifetimes on the order of thousands of years, field investigations of landfills after 10 years of operation reveal rents and perforations in the liners (Düllmann & Eisele, 1993). These authors also found that drainage systems had ceased to function due to iron oxide and carbonate precipitates.

Possibly part of the divergence of opinion concerning long-term engineered barrier performance is the result of an underestimation of the detrimental impact which time, and the accompanying ageing process, has on any man-made construction. Due to the complexity of this ageing process, similar simplifying assumptions must be made when modelling barrier performance as for the contaminant release durations. Two commonly made assumptions are those of equilibrium chemistry and steady-state water fluxes. Yet transience can be a major issue for engineered barrier stability. For example water-table fluctuations may cause cyclic wetting and drying of synthetic clay liners and result in dessiccation fractures which lead to a dramatically reduced barrier efficiency.

From a hydrogeological standpoint, a critical factor for the stability of engineered barriers is water circulation within and around the waste disposal site. Bentonite stability, for instance, is dependant on the rate of potassium supply which in turn is a function of the water supply rate. Total water fluxes are primarily dependant on precipitation and on bulk (effective) hydraulic properties of the surrounding rock body. Other critical factors are the hydrochemistry inside and around the barriers, and the mechanical stability of the waste stabilization matrix and the concrete liner (IMRA 1991, NAGRA 1985, 1988).

Table 2 summarizes modelled/estimated lifetimes of engineered barrier systems in the case of non-radioactive and radioactive waste types. The results presented for radioactive

Parameter	Municipal Solid Waste	Solidified Filter Ashes (Reststoffe)	Low-Level Radioactive Waste (SMA)	High-Level Radioactive Waste (HAA)
Hazardous Contaminant Release Time (y)	500-1700 [1]	> 500'000 [2]	2'000 [3]	2'000'000 [3]
Waste Stabilization Matrix: Assumed Total Dissolution Time (y)	No Stabilization	contradictory (cement)	10'000-100'000 [3] (cement)	150'000 [3,4] (glass)
Factors Controlling Integrity of Stabilization Matrix		Water Supply Rate Hydrochemistry	Water Supply Rate Hydrochemistry Mechanical Stability	Temperature Hydrochemistry Mechanical Stability
Engineered Barriers: Assumed Total Failure Time (y)	10-1000 (?) [5,6] (mineral liner)	10-1000 (?) [5,6] (mineral liner, geomembrane)	10'000-100'000 [3] (cement)	> 1'000'000 [3,4] (bentonite)
Factors Controlling Engineered Barrier Stability	Water circulation Hydrochemistry (Waste) Settling (Compaction)	Water circulation Hydrochemistry (Waste) Settling (Compaction)	Water circulation Hydrochemistry Mechanical Stability	Water circulation Hydrochemistry

Tab. 2. Modelled/estimated lifetimes of engineered barrier systems for different waste types. [1] Belevi & Baccini (1989), [2] IMRA (1991), [3] NAGRA (1985), [4] NAGRA (1988), [5] Resele et al. (1993), [6] Düllmann & Eisele (1993)

waste repositories are based on substantially more comprehensive and sophisticated investigations than for non-radioactive waste landfills. On the other hand, the relatively long lifetimes of HAA waste barriers assume in part geosphere properties (e.g. bulk hydraulic conductivities of 10^{-11} m/s) which have not yet been proven to exist at suitable repository locations. The values in Table 2 suggest that, in the case of non-radioactive waste, estimated lifetimes of engineered barriers are shorter than the contaminant release times. For radioactive wastes stored under suitable conditions, the lifetimes are expected to be similar to or greater than the contaminant release times.

2.3 *Critical Geosphere characteristics*

Although bulk hydraulic properties of the geosphere may influence the stability of engineered barriers, once these barriers have failed it is no longer bulk properties which are of relevance for the disposal site's environmental impact. Contaminant transport in the geosphere is strongly influenced by the spatial distribution of hydrogeologic characteristics and relatively small-scale heterogeneities in the subsurface, such as fractures or faults, lead to the development of preferential flow and transport pathways, within which most of the water flow occurs.

Therefore, transport in fractured rock is in general not compatible with the concept of a representative elementary volume (REV) and a corresponding continuum approach (e.g. Schwartz & Smith 1987). For a network permeability REV to exist in such a rock the individual pathways need to be fully interconnected and smaller than the dimension of the REV (e.g. Witherspoon 1986). The individual pathways might show different scales (e.g. fracturing), such that the averaged parameters vary continuously, or be so large that there is no average at the scale of interest. In all these cases, transport is normally modelled in terms of discrete networks (e.g. Schwartz & Smith 1987).

Such heterogeneity affects not only advective transport, but also processes such as degradation and retardation which depend on local hydrochemical and biological conditions. If transport is confined to specific preferential pathways, the total amount of sites available for sorption is substantially reduced, and saturation of sites will be reached much faster than if the contaminants were more evenly distributed. Also, radioactive decay and biodegradation can be expected to be much less effective for pollutant removal in the case of transport in preferential pathways because the contaminant residence time during which degradation occurs is decreased.

As migration proceeds, the contaminants may reach an aquifer in which water fluxes are much greater than in the geological barrier. In this case dilution of leachate with uncontaminated groundwater is an essential process affecting the contaminant plume's impact. In the case of contaminants which are not affected by a degradation or decay process, dilution is essentially the only mechanism which leads to a reduction of contaminant concentrations. The aquifer's ability to dilute the leachate is also strongly influenced by its hydrogeologic heterogeneity.

Table 3 presents a summary of the critical factors and processes which are believed to be particularly relevant to the long-term safety of a waste disposal site. Considering the importance of heterogeneity and preferential pathways for contaminant migration in the geosphere, the localization and characterization of this heterogeneity becomes essential for a realistic evaluation of a waste disposal site's long-term environmental impact.

Relevant Process for Safety	Controlling Hydrogeologic Factors/Parameters
Dissolution of Waste Stabilization Matrix	- Infiltration - Waste hydrochemistry - Hydraulic conductivity of technical barriers - Hydraulic gradient across technical barriers - Mechanical characteristics of waste
Failure of Liner / Engineered Barrier	- Waste leachate chemistry - Groundwater chemistry - Effective (mean) hydraulic conductivity of natural barriers - Hydraulic gradient across natural barriers - Mechanical Characteristics of engineered barriers
Contaminant Transport through the Geological Barrier (Aquitard) and in the Aquifer	- Transport velocities: distribution of preferential pathways - Dilution in the aquifer: as above - Retardation: foc, clay content, Eh, pH, K_d, species in solution etc. - Degradation: bacteria, oxygen/nutrient supply

Tab. 3. Summary of critical factors controlling the processes relevant to safety

3. Localization and characterization of preferential transport pathways

3.1 Field methods

As illustrated below, for most potential geological barriers in Switzerland, the existence of preferential pathways is the rule rather than the exception. The geometry of these preferential transport pathways is generally 3-dimensional and their characterization is far from trivial as there exists no single method which can answer all questions.

While geophysical methods (for example seismics and ground-penetrating radar) may provide a 2- or 3-dimensional picture of petrophysical heterogeneities, they do not yield estimates of the hydrogeologic characteristics (in particular the permeability) of these heterogeneities. Even when rock structures are directly accessible to visual inspection (e.g. from core analysis), direct hydraulic interpretation is often erroneous because only a small proportion of observed structures may actually be hydraulically active. Therefore preferential pathways for flow and transport can only be clearly identified and characterized by in-situ tests which measure water flux in the geosphere.

Standard hydraulic tests (e.g. pump tests, slug tests) performed in open holes are not well suited for the description of the geosphere's heterogeneity because they yield hydraulic parameters which are averaged over the entire open borehole section. Also, tests which do not include measurements in observation boreholes (single hole tests) are not directional and yield hydraulic parameters which are generally only representative of the volume of rock in the immediate vicinity of the test borehole. Two relatively new testing concepts can partially lift these difficulties: hydraulic pressure and tracer cross-hole testing and dynamic fluid logging.

Hydraulic crosshole pressure testing requires a network of active/observation boreholes equipped with multiple packer/transducer systems (see for example Guyonnet et al. 1993). Based on the spatial distribution on the pressure response in individual observation intervals, highly conductive features can be identified. Because the testing procedure is relatively cost and equipment intensive, it has been applied primarily in the context of radioactive waste disposal. However, COLENCO has recently applied it for the characterization of a potential filter ash waste disposal site in Switzerland.

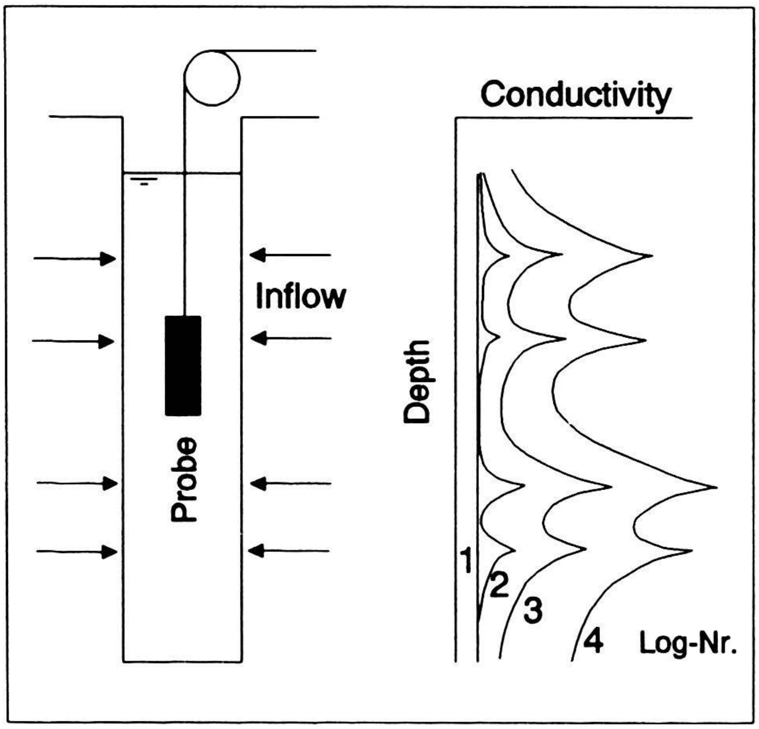

Fig. 1. Schematic representation of the fluid logging method

The dynamic fluid logging method (Tsang et al. 1990) involves first washing out the borehole with water of a different salinity than the formation water, and then measuring the fluid electrical conductivity profile in the borehole at different times while pumping at a low flow rate to stimulate flow from the formation to the borehole. Locations of formation fluid inflow result in electrical conductivity peaks observed in the measured profiles (Fig. 1). The method can be used not only to precisely locate hydraulically active features, but also to quantitatively characterize these features in terms of transmissivities, formation hydraulic heads and formation water salinities. The quantitative analysis methods are based either on numerical modelling (Tsang et al. 1990, Löw et al. 1993) or on direct analytical methods (Löw et al. 1994).

3.2 Preferential transport pathways from fluid logging experiments

Since 1987, dynamic fluid logging tests have been conducted in Switzerland in a wide variety of geologic environments, both for landfill site characterization (shallow boreholes) and for potential underground repository investigations (deep boreholes). Most of these fluid logging tests were performed for NAGRA, the Swiss National Cooperative for the Disposal of Radioactive Waste. Measurements were collected by GEOTEST AG while COLENCO was responsible for test design and interpretation. The rock types investigated include all the geological barriers considered in the context of waste disposal in Switzerland, namely crystalline rocks (granites, gneiss, diorites), fluvial sediments (molasse), marine sediments (marls, mudrocks, evaporites) and lacustrine sediments (clays). In nearly all these environments, strongly heterogeneous flow conditions were observed. Some typical examples are illustrated below.

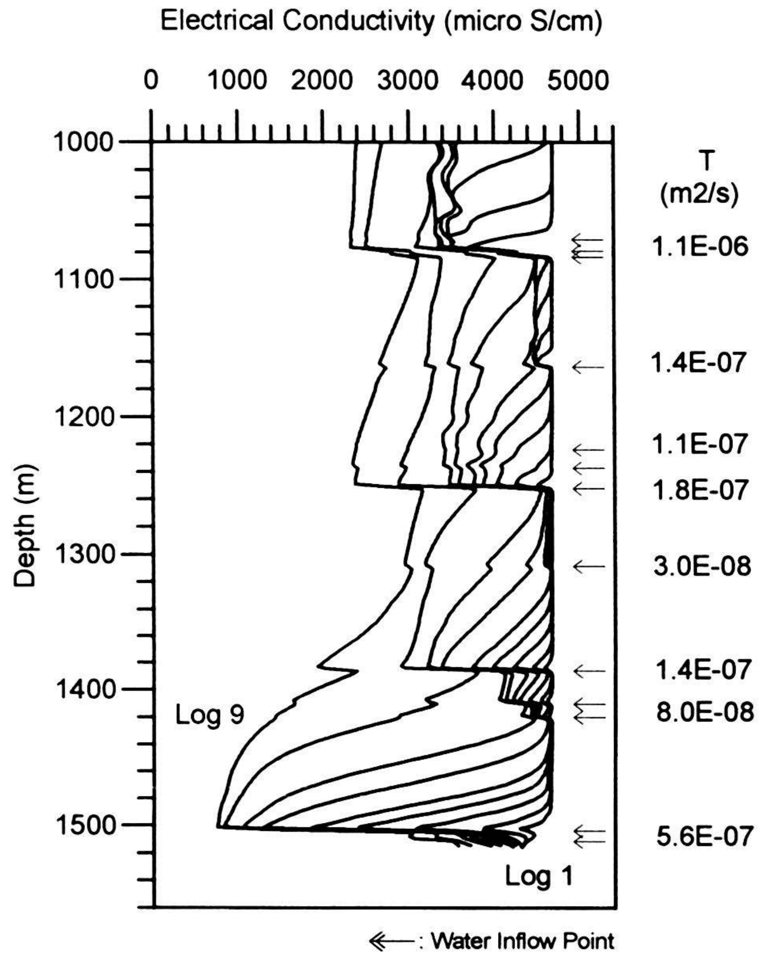

Fig. 2. Electrical conductivity logs measured in the Siblingen borehole. Logging times are between 1 and 37 hours after the start of pumping (Kelley et al. 1991).

Figure 2 presents electrical conductivity logs measured in NAGRA's Siblingen deep borehole which intersects the crystalline basement of Northern Switzerland. Details concerning the borehole geology, test conditions and the test analysis are found in Kelley et al. (1991). In the granite section between 1,000 and 1,500 m, 13 preferential water inflow locations could be identified with transmissivities ranging between 3E-08 and 1E-6 m^2/s. These inflow points correspond primarily to fractures and brittle fault zones.

Conductivity logs measured in a geothermal exploration borehole near Burgdorf are presented in Figure 3. This borehole intersects a strongly heterogeneous sequence of marls and sandstones from the Lower Freshwater Molasse. Details concerning the borehole geology, the test conditions and the analysis are documented in Ammann et al. (1993). In the section between 50 and 250 m, 2 preferential water inflow locations are identified with transmissivities of 1.6E-5 and 1.5E-7 m^2/s. Additional logs measured after those presented in Figure 3 and under different drawdown conditions enabled the identification of several other inflows. The inflows correspond to thin intervals located within coarse grained sandstone layers.

Figure 4 illustrates the only fluid logging experiment performed to-date where the tested geological barrier showed no evidence of preferential pathways within the method's range of resolution (in this example the lower transmissivity detection limit is estimated as $\approx$ 1E-9 m^2/s). This test was performed in the Opalinus Clay and the Lias For-

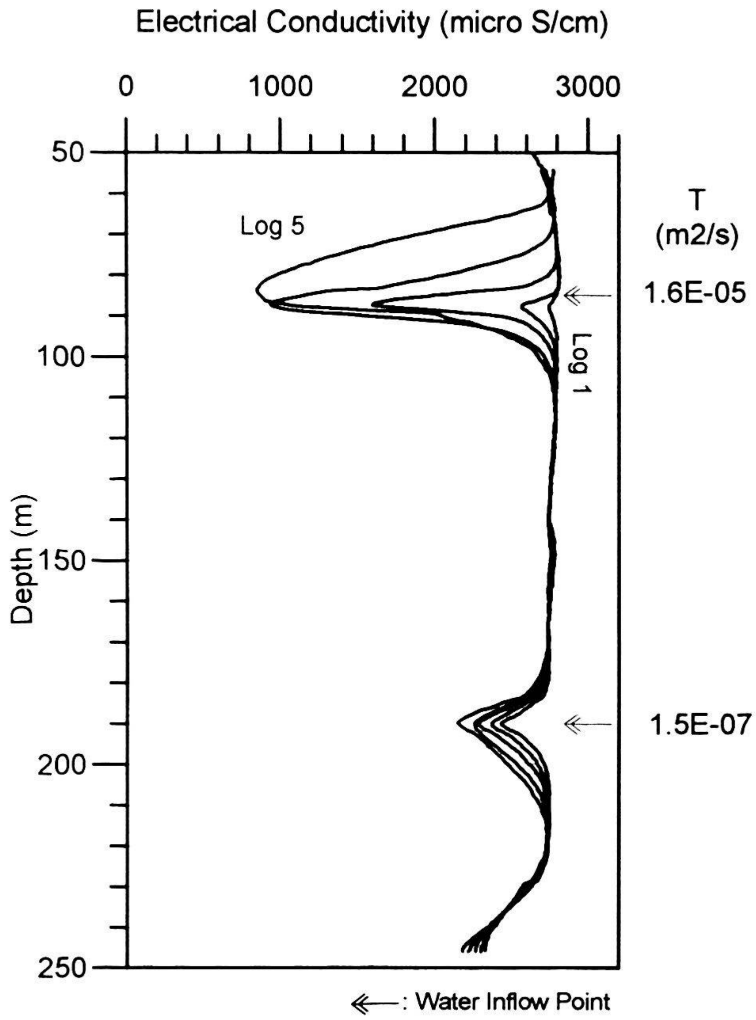

Fig. 3. Electrical conductivity logs measured at Burgdorf. Logging times are between 6 and 13 hours after flushing the borehole with brine (Ammann et al. 1993).

mation of the Folded Swiss Jura. In the tested borehole the geologic barrier (Opalinus Clay) is intersected between 14 and 50 m. No conductivity peaks appear in Figure 4 in this section. On the other hand in the Lias, a heterogeneous sequence of clayey and calcareous marls, an important inflow appears at 57 m depth with a transmissivity of 2E-6 m^2/s.

4. Influence of preferential transport pathways on aquifer pollution

4.1 Conceptual and numerical models

In the preceding sections the critical influence of geosphere heterogeneity on contaminant subsurface migration was introduced and illustrated with field examples of hydraulic tests where transport was seen to be controlled by discrete preferential pathways. The purpose of this section is to illustrate the influence of such pathways on aquifer pollution in a semi-quantitative fashion based on model simulations of pollutant migration from a generic waste disposal facility.

The conceptual models for the simulations are shown in Figure 5a and 5b. A waste disposal site of known dimensions with a known mass of contaminants overlies a clay

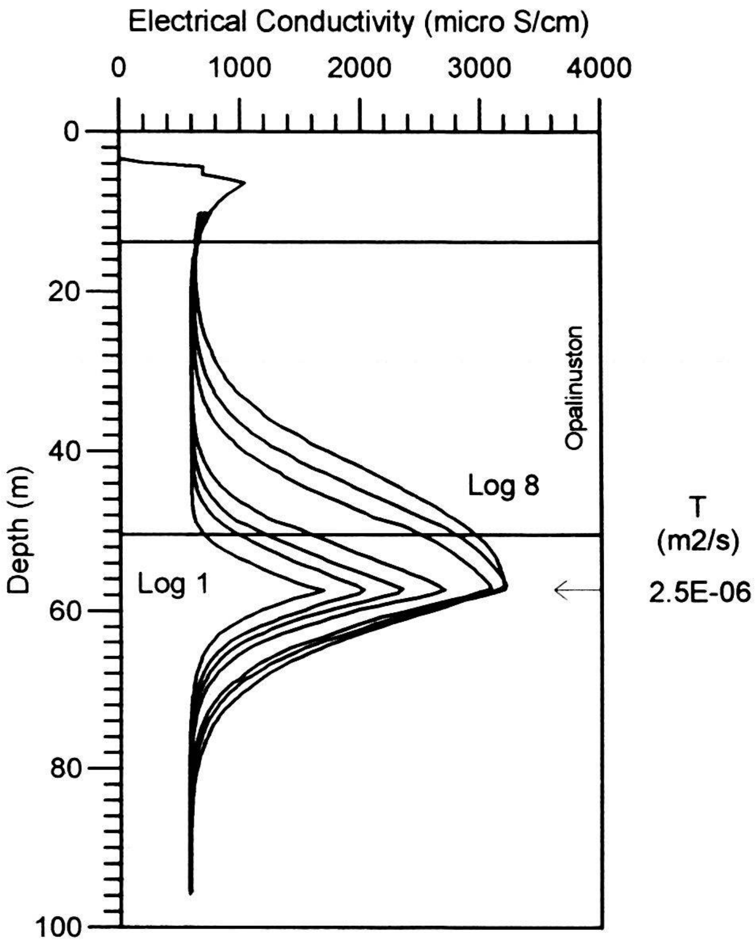

Fig. 4. Electrical conductivity logs measured in the Jura Mountains. Logging times are between 0.5 and 7.3 hours after the start of pumping.

aquitard and a sandy aquifer. Because the simulations aim at addressing the performance of the geosphere with respect to contaminant migration, the drainage system and the synthetic liner shown in Figure 5 are not included in the simulations. The contaminant breakthrough times presented below can therefore be considered as times after failure of the engineered barriers. As stated in the introduction, the long-term efficiency of engineered barriers is questionable when one considers the time scales during which the stored pollutants remain toxic and must be isolated from the biosphere.

Critical parameters (thicknesses, porosities, hydraulic conductivities, hydraulic gradients) for the geological components in Figure 5 were selected based on field investigations performed by COLENCO at a potential solidified filter ash waste disposal site in Switzerland. The potential site is a clay pit with a clay thickness of approximately 10 m. Field and laboratory hydraulic tests suggest that the conductivity of the intact (unfractured) clay is low and on the order of 10^{-9} m/s. The clay is underlain by an important aquifer system. The aquifer has a thickness on the order of 30 m and a hydraulic conductivity of 10^{-3} m/s.

For the simulations, the dimensions of the disposal site were taken as 200 by 300 by 10 m. The specific contaminant selected for the calculations is the organic solvent trichloroethylene (TCE, C_2HCl_3), a chemical compound which is used in massive amounts for the degreasing of metal components. According to TVA (1990) regulations for solidified

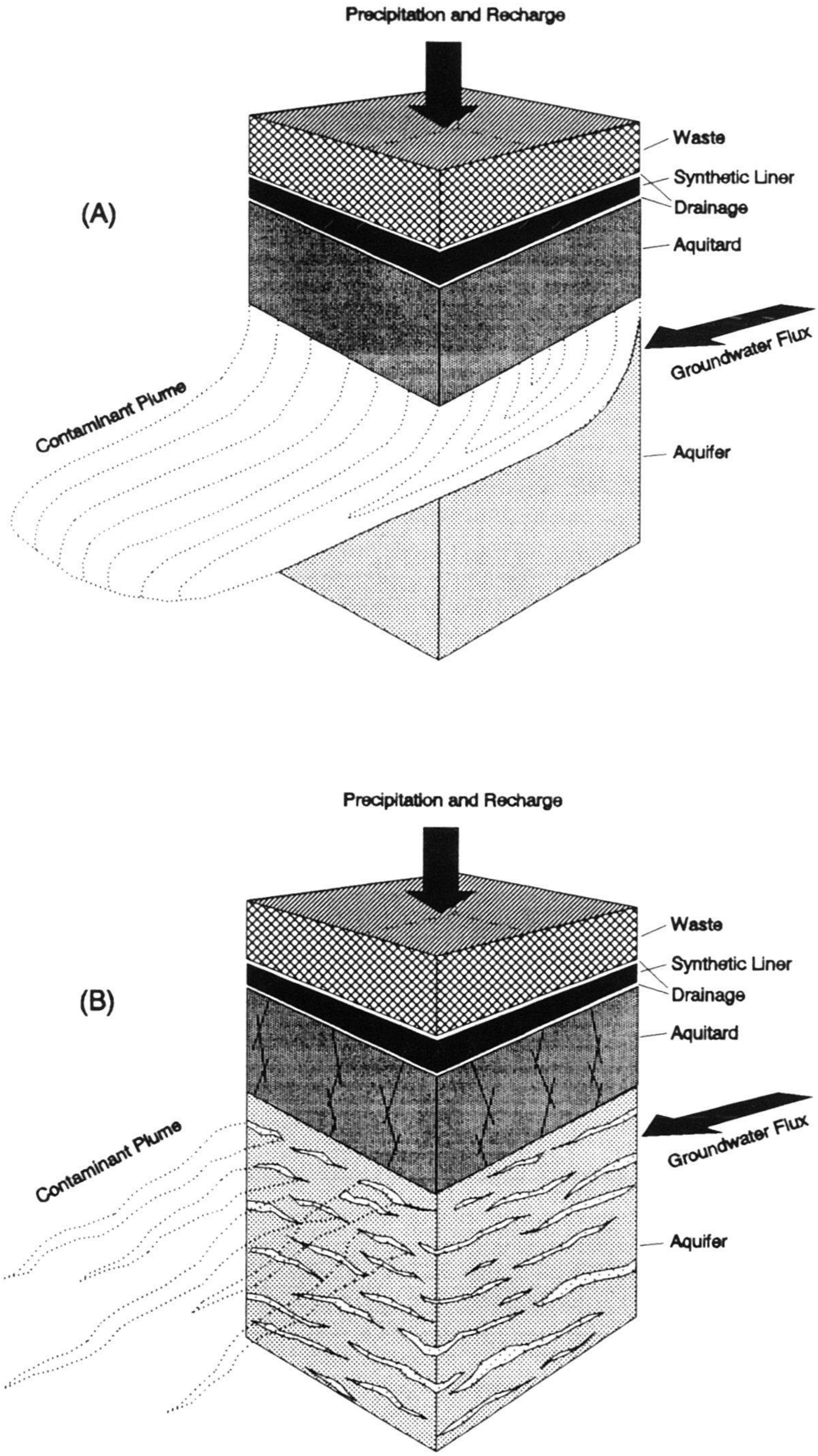

Fig. 5. Conceptual models for the sensitivity analysis.

filter ash waste (Reststoffe), the concentration of hydrophobic halogenated organic compounds (such as TCE) should not exceed 10 mg per kg of dry waste. For the simulations it was assumed that TCE ist the only hydrophobic halogenated organic solvent present, and that the total mass of TCE stored in the disposal site is 1,000 kg. The TVA regulations also state that the maximum admissible concentration of chlorinated solvents in the leachate is 100 µg Cl/l which corresponds to approximately 120 µg/l TCE (considering the molecular weight of TCE). Although such a rule is very difficult to apply in practice, this maximum concentration was nevertheless assumed for the leachate concentration in all the simulations.

As stated previously, an important aspect of toxic waste disposal in Switzerland is the stabilization of the waste by mixing it with cement in various proportions (on the order of 20% cement). Waste stabilization results in a *retardation* of the contaminant transfer into the geosphere and not to an immobilization or to a reduction of the contamination. As stated in section 2.3, for a compound which is poorly affected by processes such as biodegradation and volatilization which act as "sinks" to remove the contaminant from the groundwater (in fact volatilization simply transfers the problem to the air phase), the primary mechanism leading to the reduction of concentration levels is dilution.

The illustration simulations were performed using a 1-D finite element model which accounts for heterogeneity and sources or sinks. The model solves the following transport equation:

$$OR\frac{\delta c}{\delta t} = OD\frac{\delta^2 c}{\delta x^2} - Q\frac{\delta c}{\delta x} - O\lambda c + S \qquad (1)$$

where:

- c is concentration (M/L^3)
- x is distance (L)
- t is time (T)
- Q is Darcy Flux (L^3/TL^{-2})
- D is the dispersion coefficient (L^2/T)
- O is effective porosity ()
- R is the retardation coefficient ()
- λ is the 1st order degradation or radioactive decay constant (T^{-1})
- S is the source/sink term (M/TL^{-3})

The left hand term represents the concentration change as a function of time. The first term on the right hand side expresses concentration changes due to transport by dispersion, the second advection, the third 1st order degradation or radioactive decay, and the fourth sources or sinks.

The initial condition is: $c(x,0) = c_i$

The outflow boundary is of the 2nd type (Neuman): $\frac{\delta c}{\delta x}(L, t) = 0$

The inflow boundary condition is of the 1st type (Dirichlet):

$$c(0,t) = c_0 \ \ if \ \ 0<t<t_0 \ , \ \ c(0,t) = 0 \ \ if \ \ t>t_0 \qquad (2)$$

where:

c $(0,t)$ is the inflow concentration (M/L^3)
c_i is the initial concentration in the groundwater (M/L^3)
c_0 is the concentration in the landfill (M/L^3)
t_0 is release duration of concentration c_0 (T)
L is the length of the simulation domain (m)

The dispersion coefficient D is: $D = \alpha \frac{Q}{O} + D_o \tau$

where:

α is the longitudinal dispersion length (L)
D_o is the free solution diffusion coefficient (L^2/T)
τ is the tortuosity ( )

Dispersion lengths were taken as one tenth of the travel distances, tortuosities were taken from Gillham et al. (1984) and the free-solution diffusion coefficient for TCE was computed using the method of Wilke & Chang (1955). For a detailed description of the parameters presented above, the reader is referred to standard textbooks such as for example Freeze & Cherry (1979). Equation (1) was solved using the Galerkin method. The resulting numerical code has been verified by comparing its results to those of analytical solutions and other numerical codes which allow for multiple sources and sinks (Guyonnet, 1993). Computation accuracy was controlled by performing mass-balance calculations. For all the simulations presented below, mass balances were lower than 10^{-4}%.

4.2 Scenarios and parameter values

Several scenarios are simulated to illustrate the influence of the critical transport parameters. The total volume of water flowing from the disposal site into the geosphere was assumed to be the same for all scenarios. What changes from one scenario to another is the speed at which transport occurs (due primarily to fractures in the clay) or the extent of dilution in the aquifer (due to the presence of heterogeneities which constitute preferential transport pathways).

Scenario 1 (base-case): the aquitard and the aquifer are homogeneous (Fig. 5a) and the contaminant is not affected by retardation or degradation. The parameter values for the base-case simulation are summarized in Table 4. The total volume of water which flows from the waste down into the aquifer is controlled by the clay aquitard hydraulic parameters. From the values in Table 4 and the dimensions of the disposal site it follows that the total volume of water flowing from the waste into the aquifer is 2.59 m^3/day. This water is charged with a concentration of 120 µg/l but is diluted in the aquifer by a volume of groundwater determined by the aquifer parameters, with zero concentration.

Scenario 2: the aquitard and the aquifer are homogeneous (Fig. 5a) and the contaminant is affected by retardation and degradation. The retardation coefficients for TCE in the clay aquitard was computed using the method of Schwarzenbach & Westall (1983) and assuming an organic carbon fraction (foc) of one percent (see Tab. 5). For retardation in the aquifer, the value was selected based on measurements reported in the literature (see for example Schwarzenbach et al., 1983). TCE is known to be only weakly

PARAMETERS	BASE-CASE VALUES	
	In Clay Aquitard	In Sand Aquifer
Thickness (m)	10	30
K (m/s)	10^{-9}	10^{-3}
Gradient	0.5	0.001
Porosity ( )	0.37	0.2
Tortuosity ( )	0.8	0.67
Dispersivity (m)	1	10
Diffusion Coefficient (m^2/s)	10^{-9}	10^{-9}
Retardation Coefficient ( )	1	1
Half-Life for Degradation (years)	none	none

Tab. 4. Summary of base-case parameter values (scenario 1)

PARAMETERS	BASE-CASE VALUES	
	In Clay Aquitard	In Sand Aquifer
Number of Fractures in Clay	30	-
Fracture Apertures (μm)	23	-
Lateral Extent of Fractures (m)	200	-
Transport Path Thickness over Total Thickness	-	1:10
Transport Path Width over Total Width	-	1:5
Retardation Coefficient	8	1.5
Half-Life for Degradation (years)	200	200

Tab. 5. Summary of parameters relevant to scenarios 2 to 5

affected by degradation under aerobic conditions and to persist in the subsurface for long periods of time (see for example Lerner et al., 1993). To illustrate the effect of degradation relative to the base-case simulation TCE was assumed to decay following a first order rate with a half-life of 200 years. Note that degradation was assumed to occur in the sediment but not to affect the source term.

Scenario 3: the aquitard is fractured (as in Fig. 5b) but the aquifer is homogeneous. The contaminant is affected by retardation and degradation. Calculations based on the cubic law for flow in a parallel plate model (see for example Snow, 1969) show that 30 vertical fractures with apertures of 23 microns and lateral extents equal to the width of the disposal site (200 m) will result in the same flow rate as for scenarios 1 and 2 (2.59 m^3/day). Diffusion of contaminants into the fracture walls is neglected.

Scenario 4: the aquitard is homogeneous but the aquifer is heterogeneous (as in Fig. 5b). The contaminant is affected by retardation and degradation. The type of aquifer heterogeneity represented in Figure 5b is characteristic of for example a fluvio-glacial sedimentary environment. Hydraulic and tracer tests performed at the site used as an example for the simulations suggest that, in the vertical direction, approximately one tenth of the aquifer is actually participating in the transport process. The ratio in the lateral direction could not be evaluated but can be expected to be larger due to geometric anisotropy. A ratio of 1:5 was assumed. These ratios result in a smaller area available for transport and therefore dilution with uncontaminated aquifer groundwater is reduced.

Scenario 5: the aquitard is fractured and the aquifer is heterogeneous (Fig. 5b). The contaminant is affected by retardation and degradation. This scenario combines the effects of heterogeneity of scenarios 3 and 4.

4.3 Results and discussion

The results of the simulations for scenarios 1 to 5 are presented in Figure 6. Concentrations of TCE at a point located in the aquifer 100 meters downstream from the disposal site are plotted versus time (transport length in the aquifer = 100 m). This imaginary point could be thought of as a control piezometer. Note that as the flux through the waste is the same for all scenarios (2.59 m^3/day), and the leachate concentration is also constant (120 μg/l), the time required to deplete the total mass of TCE (1,000 kg) is the same for all scenarios (8,815 years). This depletion time is included in the model by Equation (2).

Compared to the base-case scenario, scenario 2 illustrates the effect of retardation and degradation which result in Figure 6 in a lower concentration than for scenario 1, and a larger time of first contaminant breakthrough at the piezometer.

Although degradation is also included in scenario 3 (fractured aquitard), it has no effect on the late-time (peak) concentration because transport through the fractured clay is so rapid that degradation has virtually no influence: contaminants appear much earlier than for scenario 1 but at the same peak concentration. It should be reminded that in the cases of scenarios 3 and 5, contaminant breakthrough times are grossly underestimated since it is assumed that the engineered barriers (synthetic liner and drainage system) have failed.

In scenario 4 the clay aquitard plays its role as a retarding barrier, but the aquifer is not as efficient at diluting the leachate. The contamination is seen to appear later but at a higher concentration than for scenario 1. Note that the contamination appears earlier than for scenario 2 although clay parameters are the same. This effect is due to the larger concentration gradients in scenario 4 which lead to increased transport by dispersion.

Scenario 5 combines the effects of heterogeneities in both the aquitard and the aquifer. The clay barrier does not fulfil its role for retardation and there is less leachate dilution in the aquifer relative to the base-case. As for scenario 3, due to very rapid transport, degradation has virtually no effect on the peak concentration. This concentration is therefore greater than for scenario 4.

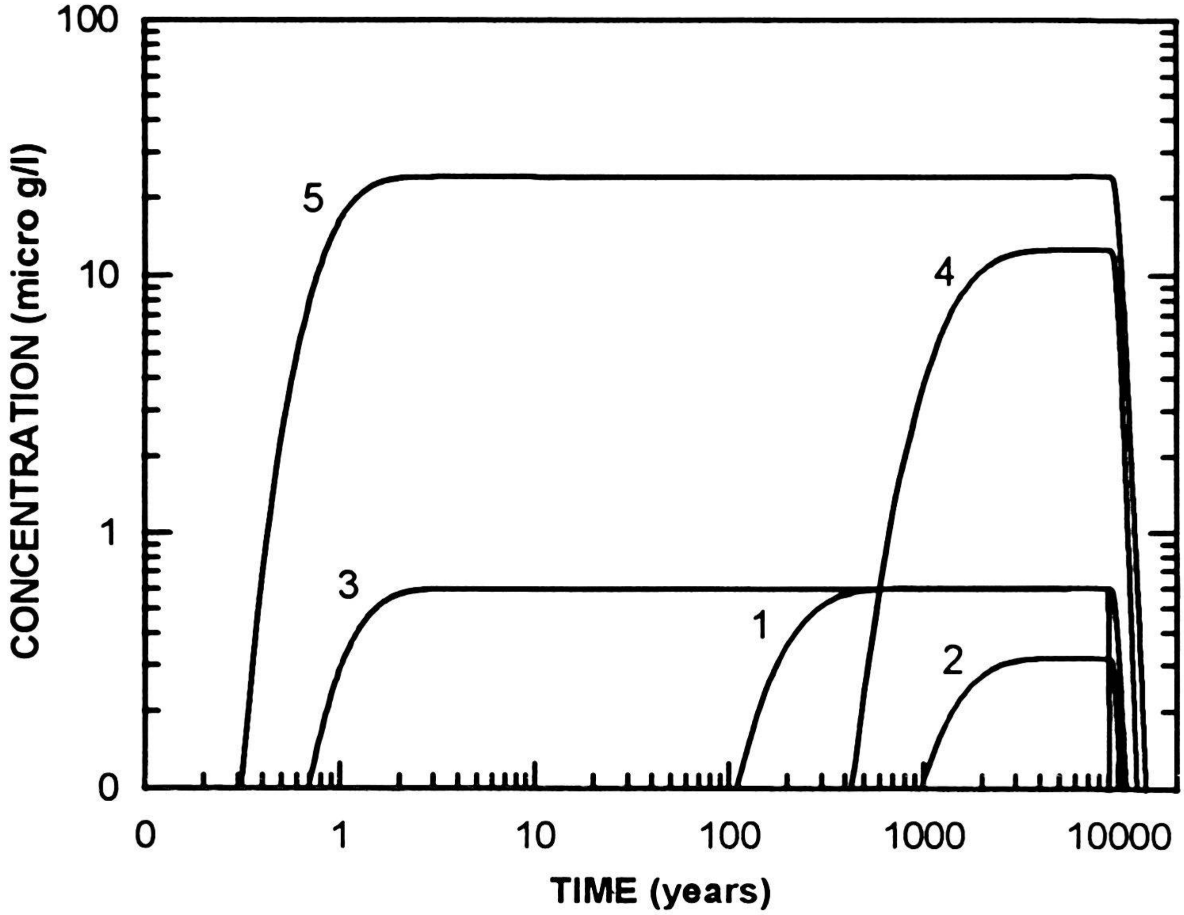

Fig. 6. Results of sensitivity analysis.

5. Conclusions

Engineered barrier systems alone are not sufficent for the long-term safety of toxic waste disposal sites because their life expectancies are most probably smaller than the durations of contaminant release times. Although the mechanical stability of engineered barriers may be influenced by bulk (effective) hydraulic properties of the surrounding rock body, once contaminants reach the geosphere, local heterogeneity rather than bulk hydraulic properties control transport and the long-term impact of the disposal site. Identification and characterization of the individual transport pathways is therefore essential for a realistic safety analysis.

As demonstrated by fluid logging, a method particularly well suited for the localization and characterization of preferential pathways for flow and transport, most geological barriers considered in the context of waste disposal in Switzerland are heterogeneous. Small-scale heterogeneities can decrease contaminant travel times and increase peak concentrations in aquifers by several orders of magnitude. Existing regulations for landfill site characterization (such as the TVA) do not explicitly consider such transport criteria.

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