Radionuclide retardation in water conducting systems - lessons learned in the research programme in the Grimsel Test Site

W. Russell Alexander and Wolfgang Kickmaier

Cover: Main entrance to Nagra's Grimsel Test Site (GTS) in the winter. The GTS lies in the central Swiss Alps at 1740m above sea level and the Grimsel Pass (visible in the background) is 2100m.

Sanierung der Hinterlassenschaften des Uranerzbergbaus (Chancen und Grenzen der geochemischen und Transport-Modellierung bei der Verwahrung von Uranbergwerken und bei der Endlagerung radioaktiver Abfälle).
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Radionuclide retardation in water conducting systems - lessons learned in the research programme in the Grimsel Test Site

Radionuklidretardation in wasserführenden Systemen - Erfahrungen aus den Untersuchungsprogrammen im Felslabor Grimsel

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ABSTRACT

The Grimsel Test Site (GTS) is an underground rock laboratory which has been operated since 1984 by the Swiss National Cooperative for the Disposal of Radioactive Waste (Nagra) in the crystalline rock of the Aare Massif, central Switzerland. With increasing experience in the implementation of in-situ experiments, improved process understanding and more mature repository concepts, the experimental programmes at the GTS have gradually become more complex and more directly related to open questions defined by performance assessors or by regulatory bodies. The aspect of demonstrating disposal concepts by performing large- or full-scale, long-term experiments has also become a key aspect of investigations in the rock laboratory. In this paper, a synthesis of the Nagra/JNC approach to the testing of radionuclide transport models is presented (details can be found in McKinley et al., 1988, Alexander et al., 1992, 1996, Frick et al., 1992, Ota et al., 1998, Eikenberg et al., 1998; Frieg et al., 1998, Kickmaier et al., 2000 and Smith et al., 2000a, b) with examples from a large programme of field, laboratory and natural analogue studies based around the GTS. The successes and failures are discussed as is the general approach to the thorough testing of predictive transport codes which will be used in repository PA.

Some of the work is still ongoing and this represents a preliminary presentation of an unique set of results and conclusions. Although this work has never been presented before, this paper represents a synthesis of two papers which are currently in press, namely Kickmaier et al., (2000) and Smith et al., (2000a), three reports which are also in press, namely Alexander et al., (2000) and Möri et al., (2000a,b) along with some new information.

1. INTRODUCTION

Over the last decade, Nagra (Swiss National Co-operative for the Disposal of Radioactive Waste) has developed a modelling approach to radionuclide transport in fractured rocks that integrates input from geological studies, hydrogeological data and geochemical data on the composition of the rock and groundwater. This approach has been applied, for example, in the Kristallin-I performance assessment (PA), that considered a repository for high-level waste sited in the crystalline basement of Northern Switzerland (Nagra, 1994).

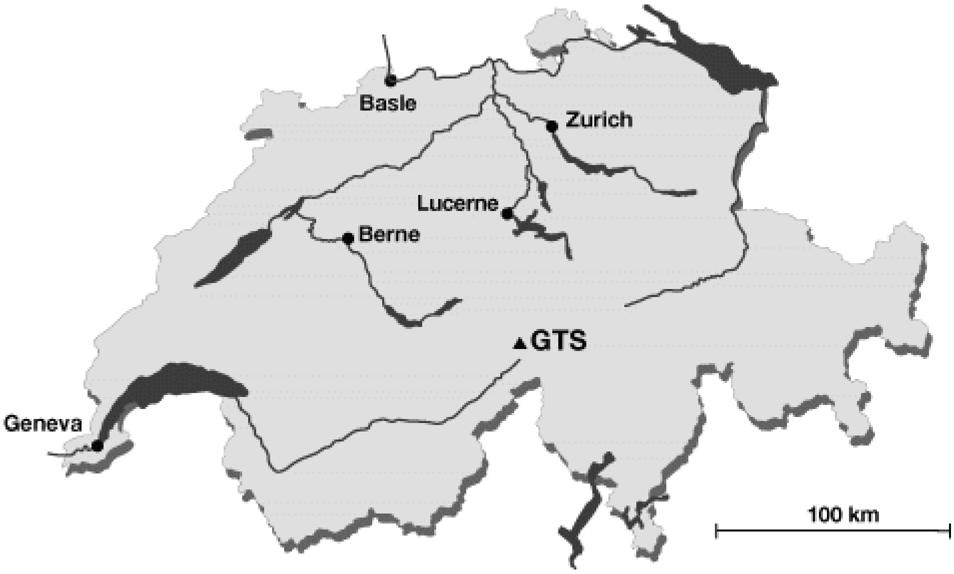

Figure 1: Map of Switzerland. Nagra's Grimsel Test Site (GTS) lies in the crystalline Aare Massif in the central Swiss Alps.

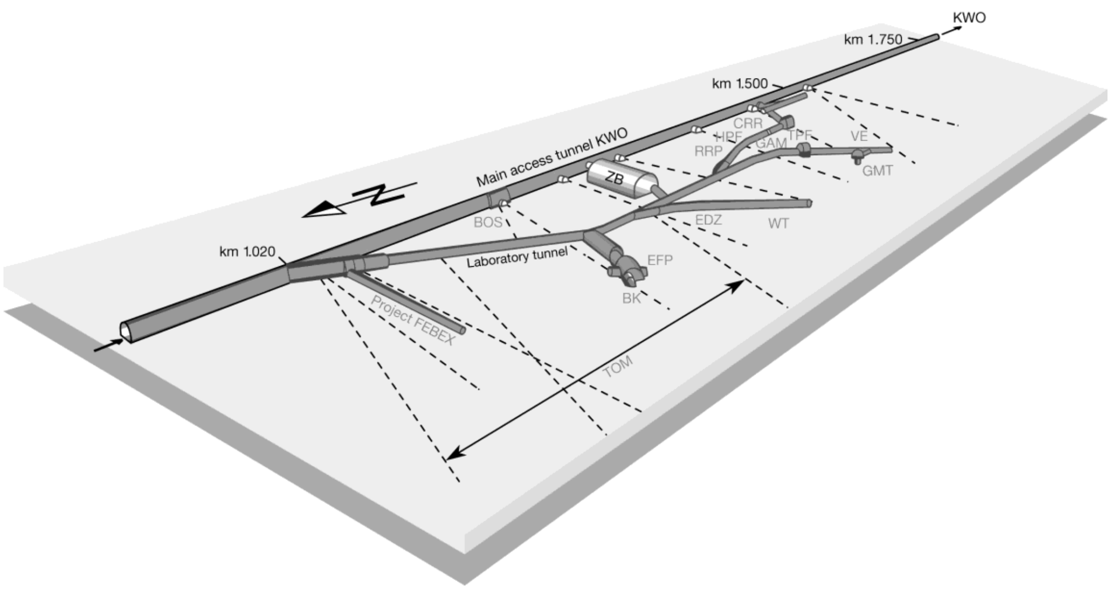

Figure 2: The MI, RRP, CRR and HPF experimental sites in the GTS tunnel system.

The model and databases developed by this approach, in common with all models of geosphere transport in fractured crystalline rock, are simplifications of reality insofar that the processes that are incorporated and their representation in the model, and the geometry (structures in the rock) within which these processes operate.

The direct testing of the results of a model, as used in PA, is impossible, due to the scales of space and time involved. Rather, confidence is developed through the consistency of the model assumptions, and associated databases, with a large number of diverse observations and experiments. Furthermore, the model should have the capability to make predictions, or at least bounding estimates, that can be tested, even if the scales differ from those relevant to PA. Few people, even those involved in the disposal of radioactive waste, fully appreciate the difference between blind testing of model *predictions* and testing if a model can *simulate* particular observations - as can be clearly seen in the literature. This is a crucial point, as pointed out by Pate et al, (1994) "This aspect of blind (ie *predictive*) testing is particularly important as, in many cases, the manner in which the simulation is carried out can be very objective and, if the "answer" is known, can be biased either consciously or subconsciously". In a repository PA, simulation of data brings little or no confidence that the models involved can later predict repository evolution: confidence can be much better built by carrying out a series of predictive modelling exercises followed by experimental runs and a final assessment of the accuracy of the predictions.

In the present paper, a general modelling approach is briefly described and examples from the joint Nagra/JNC (Japan Nuclear Cycle Development Institute) Radionuclide Migration Programme are taken from the wide-ranging experimental programme at Nagra's Grimsel Test Site (GTS; see Figure 1) in Switzerland (for details see Alexander et al., 1992; Eikenberg et al., 1998; Frieg et al., 1998; Ota et al., 1998; Kickmaier et al., 2000).

Work on transport modelling began at the GTS in 1985 with the hydrogeological characterisation of a water conducting shear zone in granodiorite (see Figure 3) and has continued with a large series of *in situ* tracer migration experiments, increasing in complexity from simple, non-sorbing tracers (fluorescein dye, ^{82}Br, ^{123}I, ^{3}He and ^{3}H) through various weakly sorbing tracers (^{22}Na, ^{24}Na, ^{85}Sr and ^{86}Rb) to a long-term experiment with strongly sorbing ^{137}Cs (Frick et al., 1992; Heer and Hadermann, 1996; Smith et al., 2000b). Most recently, the construction of an IAEA level B radiation controlled zone in the GTS tunnels (see Alexander et al., 1996 for details) has allowed the use of chemically complex tracers (^{99}Tc,^{113}Sn, ^{75}Se, ^{234,235,238}U, ^{237}Np, ^{60}Co, ^{152}Eu and stable Mo) and this was followed by the physical excavation of a part of the experimental shear zone to recover these strongly retarded radionuclides (Alexander et al., 1996, 2000, Möri et al., 2000a).

Currently, two experimental programmes are being performed in the radiation controlled zone of the GTS. Both the Hyperalkaline Plume in Fractured rock (HPF) and the Colloid and Radionuclide Retardation (CRR) experiments have similarities with the previously carried out experiments, but now specifically address the retardation behaviour of safety-relevant nuclides in the presence of near-field colloids (CRR) and the effect of hyperalkaline cementitious leachates on the host rock and the retardation behaviour of radionuclides (HPF).

This area of the GTS is particularly suitable for in-situ experiments using radionuclides as it offers:

i) steady-state hydrochemistry known from the MI experiments
ii) steady-state hydrology (constant discharge rates, stable pressures)

iii) saturated flow conditions
iv) practical experimental timescales
v) an extensive database on the internal structure of the shear zones

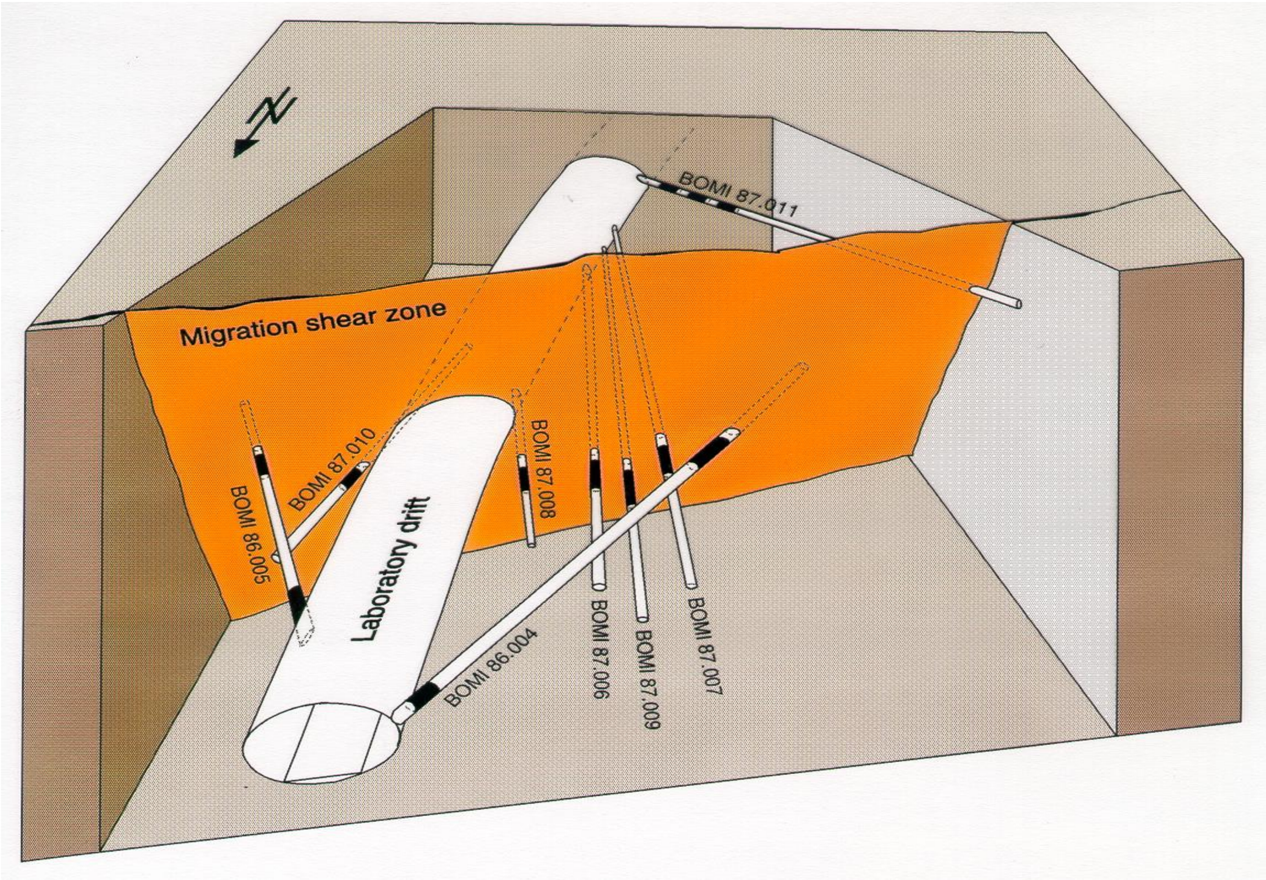

Figure 3: Schematic layout of the experimental shear zone in the GTS

As noted above, both new experiments intend to employ safety relevant radionuclides with CRR considering the behaviour of $^{242,244}Pu$, ^{237}Np, $^{233,238}U$, ^{232}Th, ^{241}Am, ^{226}Ra, ^{99}Tc, ^{75}Se, and ^{134}Cs and HPF examining the retardation of ^{134}Cs, ^{60}Co, ^{152}Eu and $^{129,131}I$.

2. THE BASIS OF GEOSPHERE TRANSPORT MODELLING FOR FRACTURED CRYSTALLINE ROCK

In accordance with current understanding of geosphere transport processes (see, for example, NEA, 1999), the dominant processes governing solute transport in fractured crystalline rocks are generally assumed to be (see also Figure 4):

- advection and dispersion within the water-conducting features and
- retardation due to matrix diffusion into the rock matrix and sorption onto mineral surfaces.

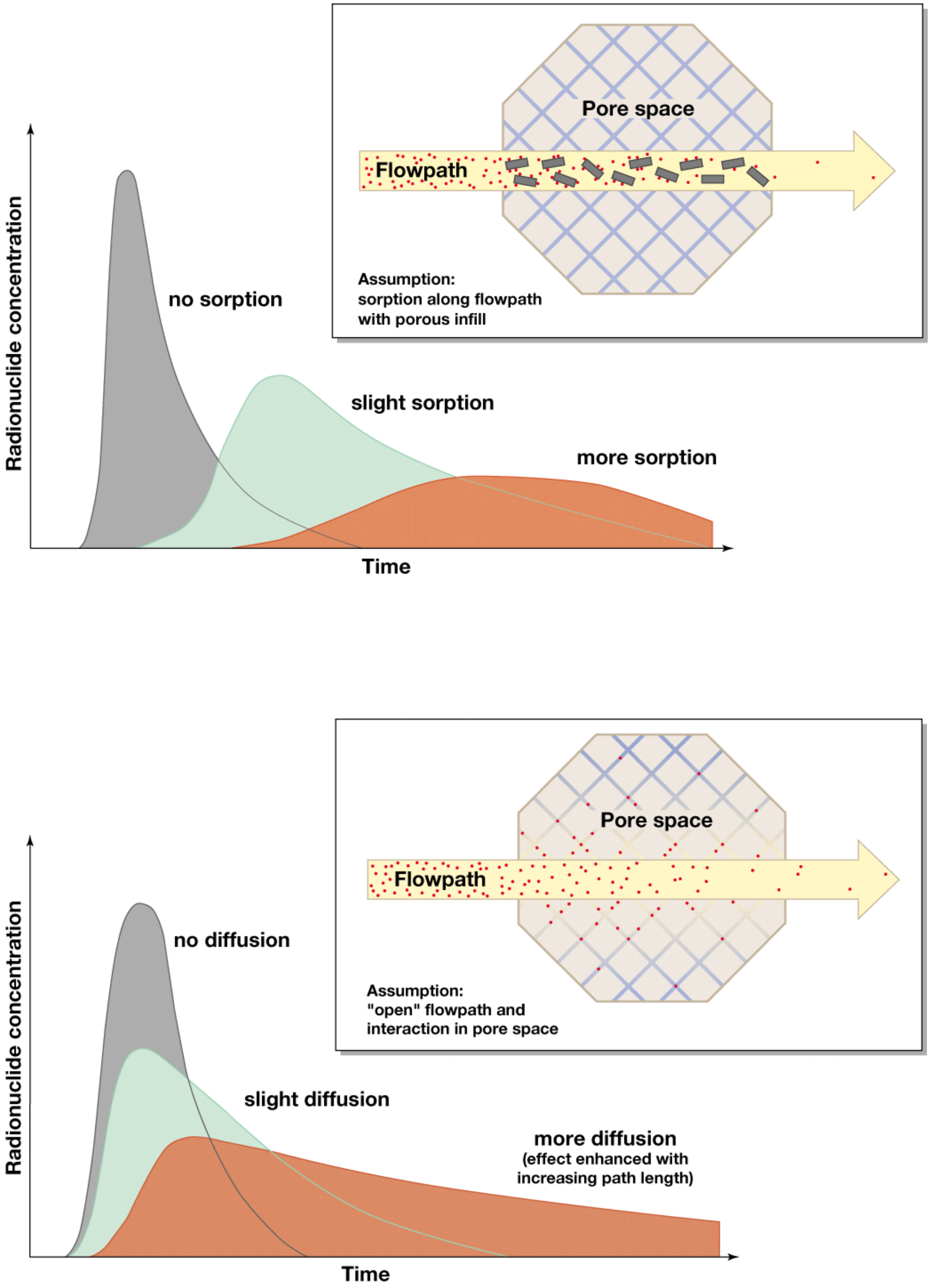

Figure 4: Illustration of the effects of different retardation mechanisms on radionuclide tracers in the geosphere.

In PA, these processes are assumed to operate in extensive heterogeneous networks of water-conducting features, although a detailed, small-scale understanding of the structure of the features is also required in order to model matrix diffusion (see, for example, Nagra, 1994). As illustrated in Figure 5, the processes (including the geometrical parameters and the parameters that are used to define the rates and spatial extent of processes) are derived from a broad base of information which includes the results of a range of characterisation techniques and general scientific understanding. The information is interpreted, in terms of transport-model parameters, by means of various supporting hypotheses and models (*eg* measured transmissivities are converted to advection parameters via groundwater flow models; sorption measurements are converted to transport model parameters via a K_d or sorption isotherm model: see, for example, Mazurek et al.,1992; McKinley and Alexander, 1992).

It should be noted that there may be many specific differences in the structures that are relevant and the parameter values used depending on whether a model is applied in performance assessment or to the modelling of a field experiment. For example, in the experimental shear zone at the GTS, the focus was on an individual feature – a shear zone – rather than on a network of water-conducting features and the scales may differ - the MI and RRP experiments were carried out over 2 - 15 metres, CRR and HPF will be carried out over 2 - 4 metres whereas distances of 100s to 1000s of metres are more applicable to a repository performance assessment. Nevertheless, the modelling approach outlined above, and in Figure 5, applies equally in all applications.

3. THE MIGRATION EXPERIMENT (MI)

Aims with respect to transport model testing

MI applied a model and parameter set, developed according to the above approach, to the results of radiotracer transport experiments in a shear zone at GTS (see Figures 3, 4 and 6). The ability of the model to reproduce, or, still more convincingly, to predict in advance the outcome of the experiments, provides a test of all the components illustrated in Figure 5, *ie*:

- the appropriateness of current scientific understanding that provided the basis, for example, of the catalogue of relevant processes incorporated in the model;
- the characterisation techniques employed;
- supporting hypotheses and interpretative models;
- the synthesis of this information into a transport model.

MI therefore provides a test of the general approach and specific methods and model assumptions that are applied in performance assessment.

Overview of experimental and modelling procedure

The central component of MI is a series of radiotracer-transport tests performed in well-defined dipole flow fields, supported by a range of field and laboratory investigations and modelling studies aimed at understanding and, ultimately, predicting the results of the radiotracer tests. The tests were performed within a single, approximately planar, shear zone that was selected on the basis of criteria given in Frick et al., (1992).

Models of this system were developed by independent researchers at JNC, PSI (Paul Scherrer Institute) and ETH (Federal Technical High School, Zürich) in Switzerland. Although developed independently, the models differed only in detail and most findings of the model testing exercises were compatible (*cf* Umeki et al., 1995 and Smith et al., 2000b) and, as such, the results can be summarised by considering the details of only one particular model, that of PSI.

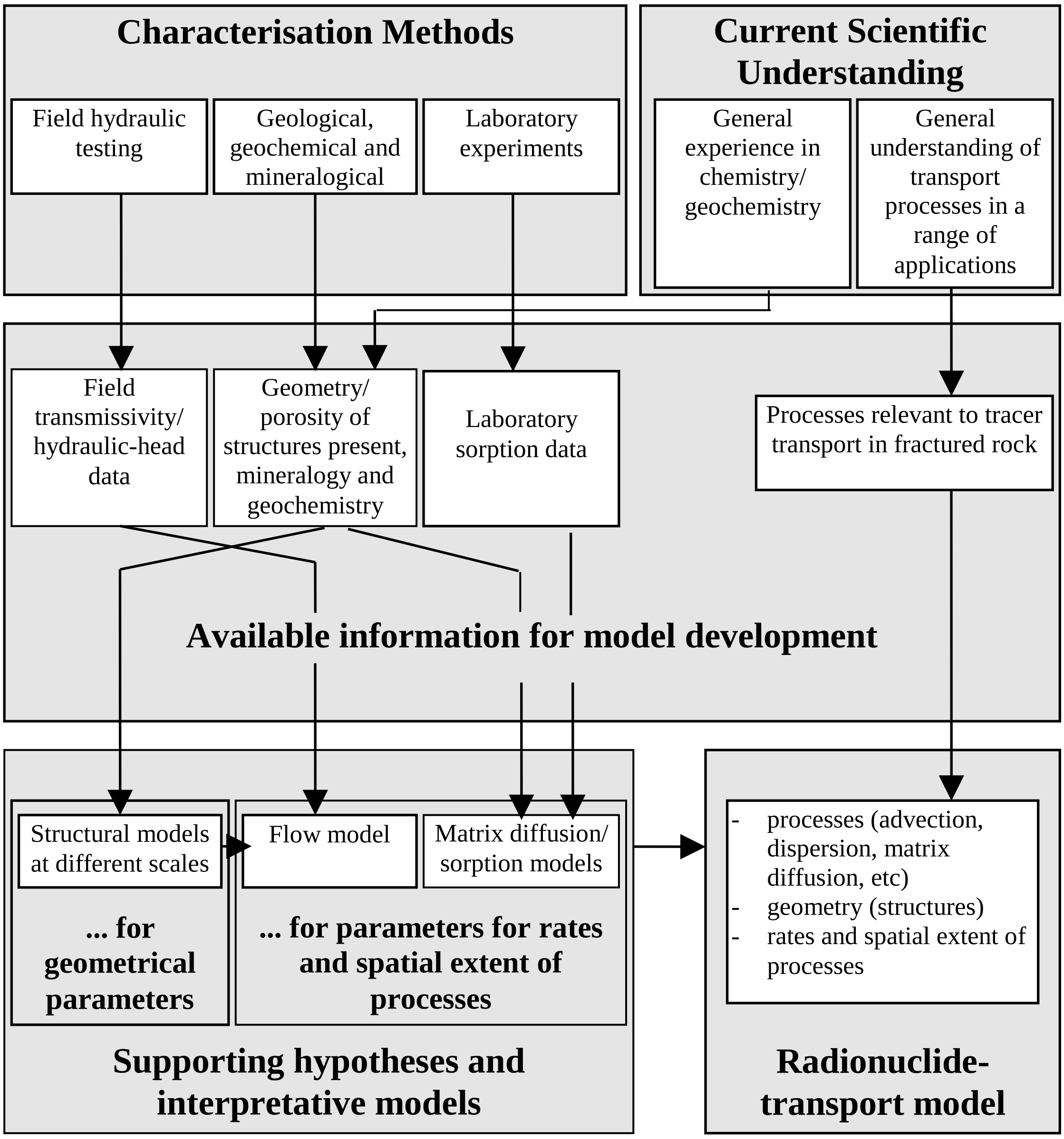

Figure 5: The use of supporting hypotheses and interpretative models to interpret field and laboratory data in terms of input parameters of a transport model.

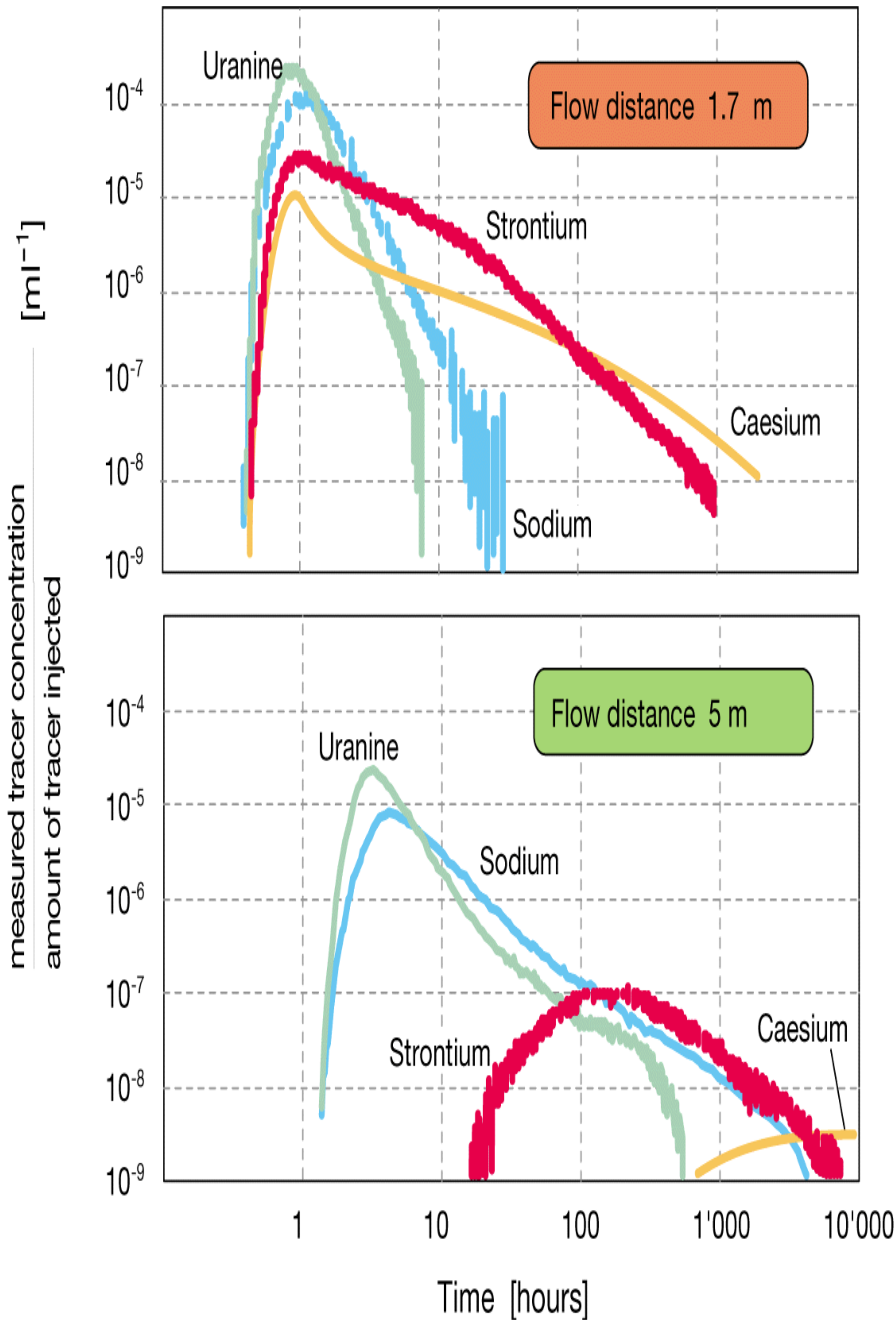

Figure 6: Examples of measured breakthrough curves from the MI experiment (cf Figure 4)

Here, the methodology for model testing was:

1. Inverse modelling
   For a suitable experimental flow field, the model was fitted to the experimental break-through curves for non-sorbing and sorbing tracers, by a process of inverse modelling.

2. Consistency of derived parameter values
   Parameter combinations were derived from the fits. Since the number of individual physico-chemical parameters exceeds the number of parameter combinations that can be fitted, additional information (*eg* from independent observations and experiments) is required in order to obtain these individual parameters. Consistency of derived parameters with all available information was sought and, where necessary, the details of the model concept were modified.

3. Predictive (or blind) modelling
   The model was calibrated using parameters that were expected to be independent of the experimental flow field in a given shear zone. Predictions were then made, using the calibrated model, of the experimental break-through curves for a different flow field or different tracers, in which the various processes are expected to be weighted differently.

Such predictive modelling, in particular, provides a sensitive test of the model concept and the numerical procedures precisely because the modeller is given only limited information. The method of testing is crucial: as noted above (and worth repeating here) few people, even those involved in the disposal of radioactive waste, fully appreciate the difference between blind testing of model *predictions* and testing if a model can *simulate* particular observations This coming together of transport modellers, field and laboratory experimenters and (to a lesser extent) performance assessors has been the hallmark of the MI experiment.

Main findings

Relation to the overall R+D programme

The MI experiment has been the single biggest experiment to date in the Nagra R+D programme and had a web of connections to other areas of the Swiss and Japanese programmes. There was direct input into Nagra`s laboratory sorption programme where an assessment was made of the relevance of laboratory produced sorption data to *in situ* retardation of radionuclides and, in addition, there was much cross-fertilisation between the MI experiment and site characterisation/PA in the field of flow path description.

In the PR field, initial use of the MI experiment was limited but this changed with the production of a Nagra Bulletin on the GTS which included an extensive article on the MI experiment (Frick et al, 1988). Some 45,000 people have now visited the GTS and the MI experiment is a routine stop on the tour of the laboratory. Currently, Nagra is producing a video on the GTS which will include footage on the MI experiment and its successor the Radionuclide Retardation Project (or RRP). Finally, the Federation of Electric Power Companies of Japan recently shot footage for inclusion in a PR video about underground rock laboratories worldwide. Arguably, more remains to be done.

Uses and extrapolation of the so obtained information

The most important use of the MI experiment has been the development of testing methodologies and the application of those methods to confidence building within PA. Indeed, the Kristallin-1 safety assessment (Nagra, 1994) carried out by Nagra specifically mentions the contribution of the MI experiment to model testing in general. Further, it was noted that ″...the results provide confidence in the dual-porosity concept as an appropriate foundation for a model of transport in fractured porous media″. In addition, it was noted that ″...the model provides a satisfactory interpretation of the measured data and no evidence has been found which would indicate that processes relevant to safety assessment and not accounted for in the model are operating .″

With respect to model testing, the main findings of the MI include:

- Although the dual-porosity concept provides a highly simplified representation of a complex natural system, it is nevertheless able to describe, with reasonable accuracy, experimental break-through curves forfluorescein dye, Na, Sr and Cs in a 4.9 m dipole flow field (Heer and Hadermann, 1996).
- The physical parameters derived by inverse modelling are consistent with values from independent observations and measurements, supporting the underlying model concepts.
- Of special note is the agreement between values for the radionuclide distribution coefficients (Kd), obtained by different laboratory and field techniques. In particular, for tracers that sorb rapidly and exhibit a reversible cation exchange on fault gouge, the results of laboratory experiments can be extrapolated reasonable well to field conditions, provided adequate care is taken in selecting and preparing the rock samples, so as to ensure that they properly reflect the geological character of the site (Baeyens and Bradbury, 1989).
- The success of the model in predicting the break-through for uranine, sodium, and strontium in a 1.7 m dipole flow field, where the different transport mechanisms are weighted differently to the larger dipole flow field, builds confidence in the underlying model concepts (Smith et al., 2000a,b).

The failure of the model to predict the break-through peak of caesium in the 1.7 m dipole flow field, while more successfully representing last part of the tail, suggests that sorption kinetics, which was not included in the models tested, may be a significant factor for this particular experiment (the effects of the sorption kinetics of caesium were not observed in the case of the 4.9 m dipole field and are, in any case, not relevant in PA; Bradbury and Baeyens, 1992).

Some effort has gone into extrapolating data on retardation mechanisms from MI to repository relevant host rocks, but this has been limited in some instances. For example, it was noted in Kristallin-1 that ″The key mechanism of matrix diffusion has, in many experiments, been identified as important and its existance and effectiveness are much better founded than 10 years ago. ″. Despite this, the diffusion constants for the rock matrix used in Kristallin-1 were ″...selected on the basis of a survey of (*laboratory*) experimentally determined diffusion constants for crystalline rocks. ″ (authors` italics). Further, no reference is made to evidence from the MI experiment (*eg* Heer and Hadermann, 1996) when depths of accessible wall rock are considered other than in the case of one parameter variation where data from MI supporting experiments (see Alexander et al., 1990a,b) are used to define a minimum depth of diffusion.

While the work on investigating the connection between laboratory measured sorption data and field retardation has shown that, with enough background information on the flow field, it is possible to show reasonable agreement within the MI experiment between field and laboratory data, this has been taken no further as yet and has most certainly not been utilised in recent Nagra or PNC assessments.

4. THE RADIONUCLIDE RETARDATION PROJECT (RRP)

Aims with respect to transport model testing

The RRP expands on MI to include (i), the Excavation sub-project (EP in figure 2), and (ii), the Connected Porosity (CP in figure 2) sub-project.

The main aim of EP was to extend the form of model testing developed in MI (Alexander et al., 1998a). At present, studies such as MI produce data for the overall flow system 'seen' by the radionuclides. These data are then modelled to produce a set of 'best fit' bulk or average values for the retardation parameters, such as rock/water distribution coefficient (or Kd) values, flow-wetted surface area and matrix diffusion depth, by comparing calculated model curves with the experimental break-through curves. However, the model values obtained may not be an unique solution and, from this process, no detailed information is obtained about the actual sites of retardation. Thus is it very difficult to extrapolate from one site or flow system to another where properties are different. This is a key issue as, in practice, the geosphere around a radioactive waste repository will not be exhaustively explored, due to the necessity to maintain favourable characteristics in as unperturbed a state as possible. Hence it is essential that extrapolations from nearby or similar sites, where such restrictions do not apply, can be justified by a thorough understanding of the factors influencing radionuclide transport (Alexander et al., 2000).

The contribution of CP within RRP to model testing is more specfic than MI. In fractured media, the modelling of the retardation mechanism of matrix diffusion pre-supposes the existence of an interconnected porosity in the rock matrix adjacent to the fractures. The definition of the depth and nature of the interconnected porosity has been ongoing in the GTS for over a decade with data gathered from natural analogues (Alexander et al., 1990a,b) and from the MI experiment (Frick et al., 1992; Heer and Hadermann, 1996). Some effort has gone into extrapolating data on retardation mechanisms from MI to repository relevant host rocks, but this has been limited (see comments above on the use of the MI experiment data on matrix diffusion).

Apart from ignoring the available data from the GTS, the approach in Kristallin-1 is also open to criticism insofar that, despite the wide range of laboratory techniques available to measure matrix diffusion, all have one common uncertainty: the effects of stress release, drilling and sample preparation on the results remain largely unquantified (outwith the pioneering work of Chernis, 1981, 1983, 1984). The main aim of CP was to constrain this uncertainty and to compare and integrate all other matrix diffusion data from the GTS.

Overview of experimental procedure

1. The Excavation Project (EP)
   This focusses on the structure of the shear zone and on the behaviour of radionuclides that are relevant to repository post-closure safety (see Figure 7), but are so strongly retarded by interaction with the shear-zone rock that they are not expected to pass through the dipole flow field in experimentally reasonable times. Consequently, a "post-mortem" analysis of the shear zone has been performed, in which the entire dipole flow field has been

Element	Half-life (years)
Technetium	210'000
Selenium	0.3
Europium	13.4
Neptunium	2.1 mio.
Tin	0.3
Cobalt	5.8
Uranium	700 mio.
Uranium	240'000

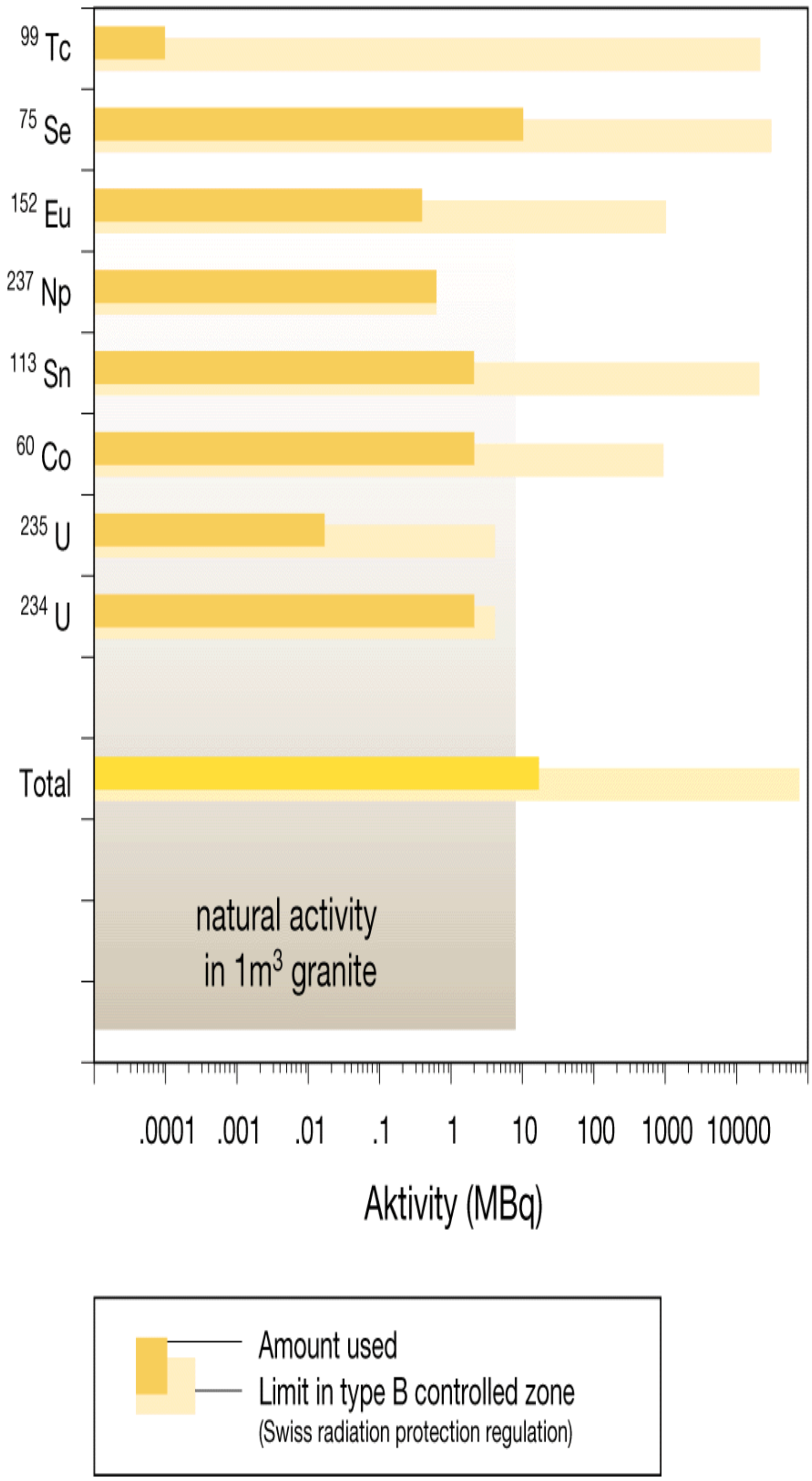

Figure 7: Activities of the safety relevant radionuclides injected into the experimental shear zone in the EP experiment.

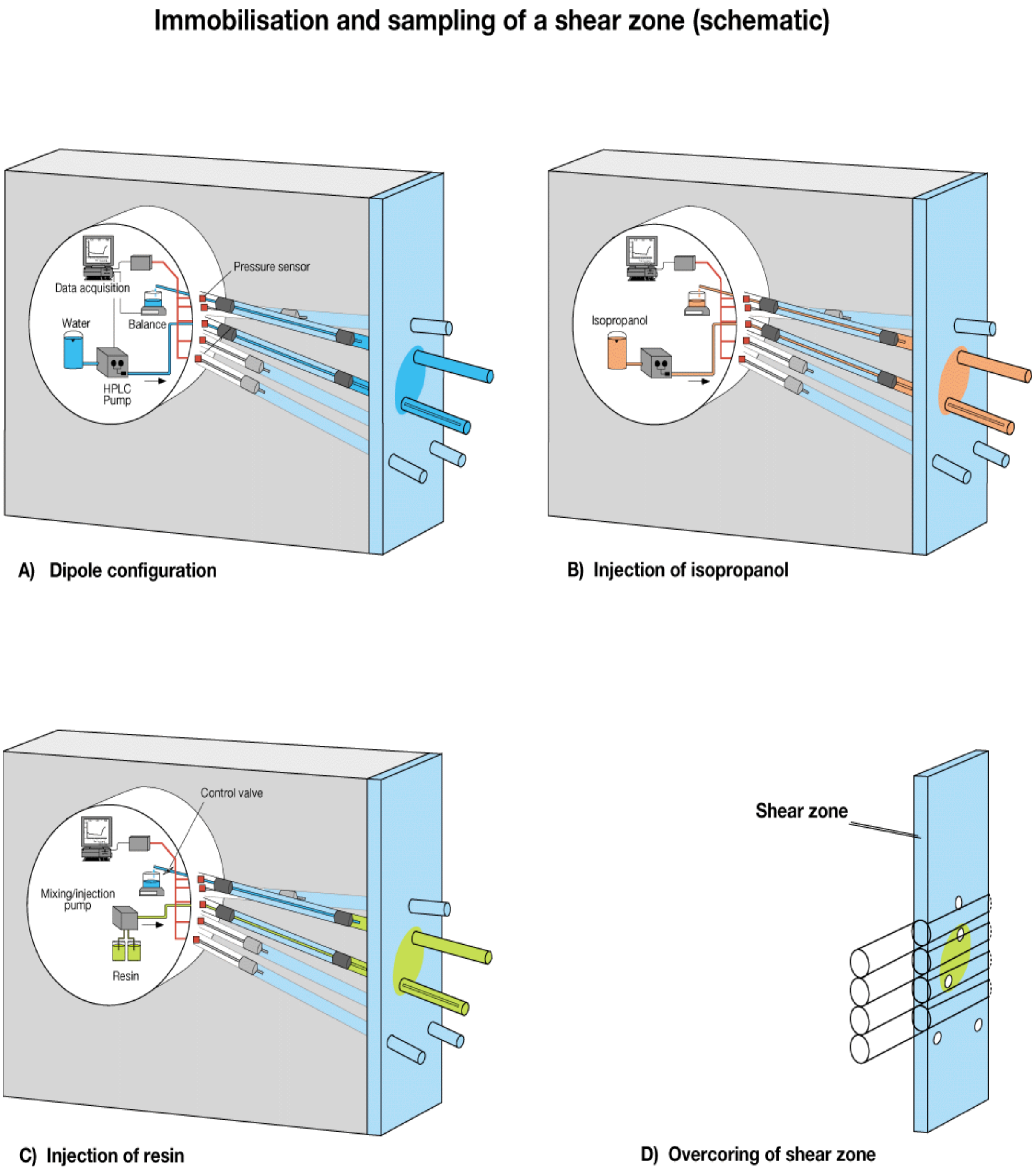

Figure 8: Excavation of the experimental shear zone during the EP experiment. A: injection of the radionuclides into a dipole flow field. B: the water in the dipole is replaced by isopropanol to ensure adequate wetting of the rock by the resin which follows. C: specially developed resin is injected into the experimental shear zone and allowed to polymerise and harden. D: the immobilised (radioactive) part of the shear zone is recovered for analysis by overlapping triple barrel drill cores, drilled parallel to the shear zone.

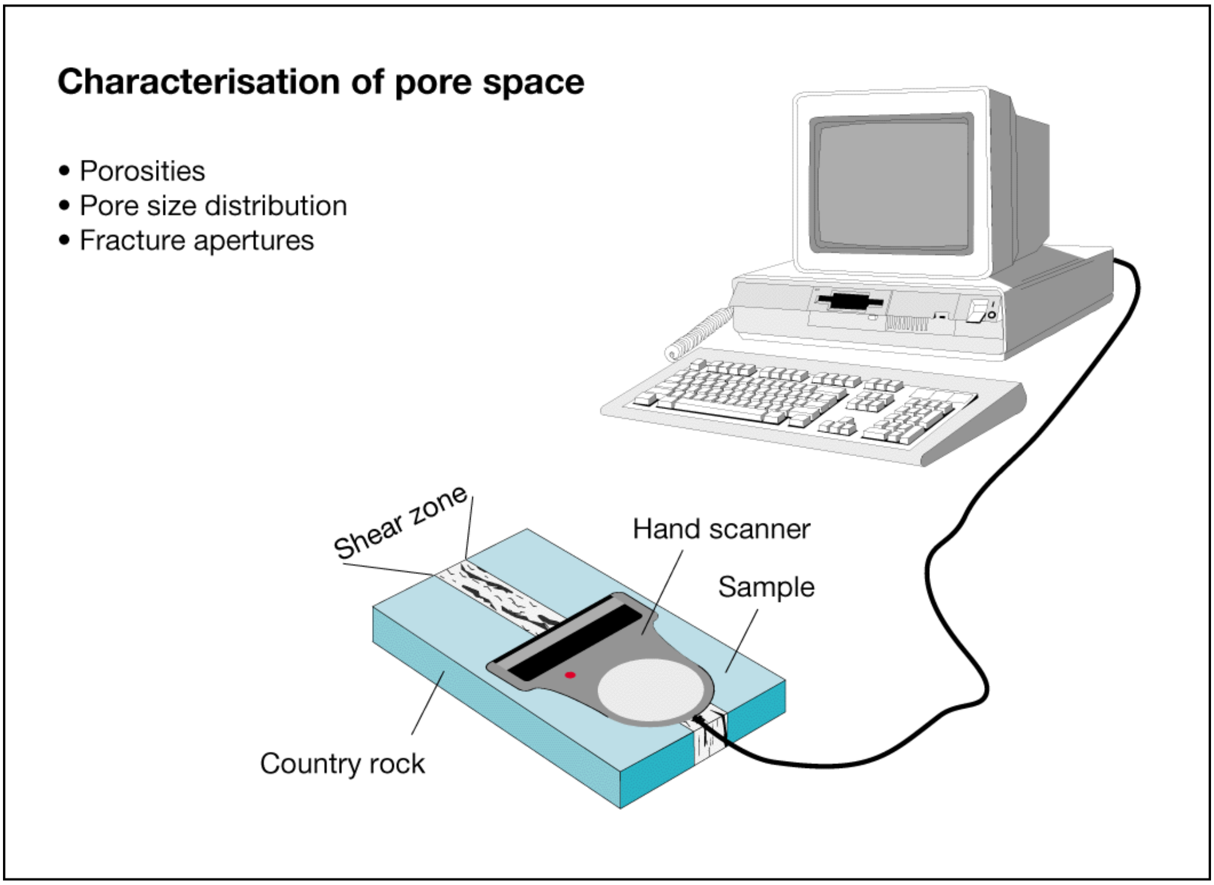

Figure 9: Analysis of the shear zone pore structure

immobilised using a newly developed resin injection technique, excavated (see Figure 8) and taken back to the laboratory for analysis (see Figure 9) of its three-dimensional structure and of the distribution of injected radionuclides within the flow field (see Alexander et al., 1996, 2000, Frieg et al., 1998 and Möri et al., 2000a for details)

2. The Connected Porosities (CP) Project

This focusses on the form and distribution of porosity in the undisturbed rock matrix at depth (up to several metres) normal to the plane of a fracture. Although radionuclides would not be expected to penetrate this porosity significantly on experimental time scales, wall-rock porosity may be a key feature affecting radionuclide retardation over performance-assessment time scales (see, for example, Nagra, 1994, JNC, 2000). In CP, a specially developed resin has been injected into the rock matrix under a small over-pressure, hardened *in situ* and wall-rock samples have been excavated (see Figure 10) and the pore structure of the impregnated material has been compared in the laboratory with material excavated by conventional drilling (see Möri et al., 2000b for details).

Main findings of the RRP experiment

The analyses of the results of the EP project is still ongoing (see Alexander et al, 2000; Möri et al., 2000a), but the main implications of the study for model testing may be briefly concluded to be:

- The basic modelling approach employed in MI was shown to reasonably predict the spatial distribution of the strongly sorbing radionuclides along the experimental flow path (namely in the vicinity of the injection borehole and at a short distance along the flow path). Again, this demonstrates, at least qualitatively, that laboratory derived Kd values may be used with care in transport modelling if the laboratory system can be set up to mimic the *in situ* conditions appropriately. Of course, this was not the case for all radionuclides: ^{75}Se, although injected into the flow path in reducing form (selinite), appeared to behave as a non-sorbing tracer (*ie* as selenate). Unlike the case for Cs in the MI experiment (above), where kinetics was shown to be the problem, here it seems more likely that equipment problems allowed oxidation of the Se (but this is still under investigation).

- Although the experimental shear zone was known to be highly heterogeneous (Bossart et al., 1991 and see Figure 11), the degree to which channels in the water conducting features controlled radionuclide migration was surprising and this would be worth considering further in any future *in situ* tests of transport codes. Dead-end channels, short-cuts between adjacent channels and merging and bifurcating of migration pathways characterised radionuclide flow on the metre and centimetre scale. Interestingly, not all the seemingly open channels were involved in radionuclide transport.

- Matrix diffusion played a significant role in radionuclide retardation (see Figure 4) although most material was not trapped in the rock matrix *senso stricto*, rather in the highly porous fault gouge present in the system. This has serious implications for repository site characterisation drilling techniques: unless double-, or better still, triple-barrel core recovery technology is used, such fault gouges are frequently lost from deep drillholes and there will, therefore, be a tendency to under-estimate potential geosphere retardation (never mind the impact on the hydrogeological assessment of the site).

Although the final results of CP are currently being reported (Möri et al., 2000b), the main findings of the project of relevance to model testing may be summarised as:

- The architecture of the matrix porosity was studied under the microscope and was found to be similar, but not identical to that observed in samples that were impregnated in the laboratory. It appears that stress release, and the preparation of samples in the laboratory, while not creating new types of pores, does visibly alter the existing porosity.

Matrix pore apertures have not yet been measured with confidence under the microscope, but JNC is currently carrying out method development in an attempt to remedy this problem. Qualitatively, however, apertures are generally smaller in samples impregnated *in situ*, when compared to laboratory impregnations. Thus, stress release and sample preparation may result in the widening of existing pores. In laboratory experiments, this may have the effect of increasing the values of measured diffusion coefficients and underestimating the role of size (and charge) exclusion effects in the rock matrix. Although this will probably be a rock specific effect, further work is currently ongoing to assess the consequences for PA via comparison of data from the MI, EP and CP experiments.

- The porosity of samples impregnated *in situ*, as well as the porosity of non-impregnated samples, was quantified in the laboratory. Non-impregnated samples typically yield porosities around 0.6 - 0.7 vol%, irrespective of the method used (water saturation gravimetry, mercury injection etc). The porosity of *in-situ* impregnated samples is 0.2-0.3 vol%. It appears that around half of the porosity measured by standard laboratory

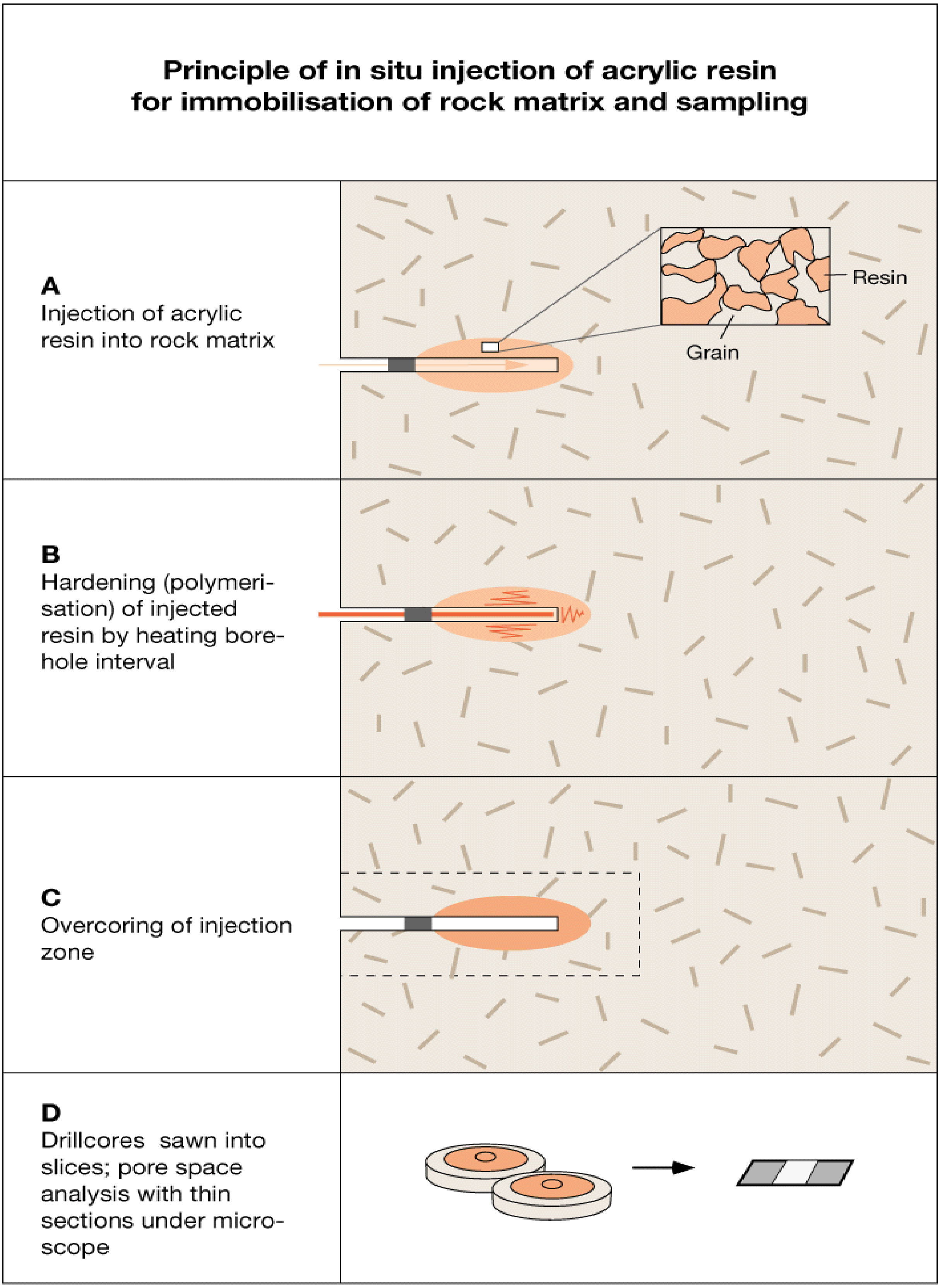

Figure 10: CP experiment methodology. A: injection of the specially developed acrylic resin into the rock matrix. B: polymerisation of the resin by means of a heat shock. C: overcoring of the resin impregnated zone. D: pore space analysis by application of object recognition and 3-D plotting software to thin section images.

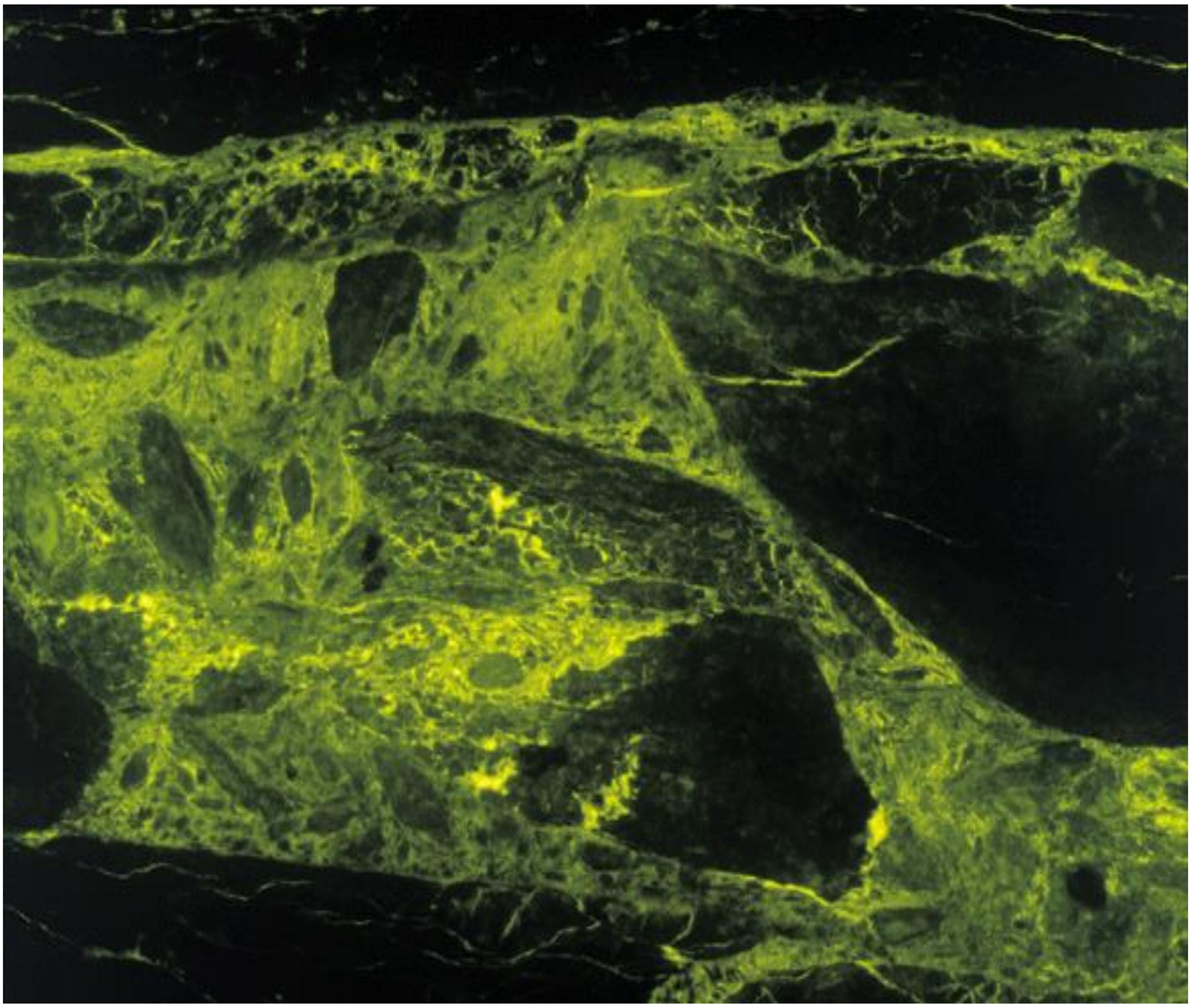

Figure 11: An example of the flow path porosity in the experimental shear zone in the GTS (photomicrograph taken under UV-light and is approximately 3mm wide). Particles of ground up wall rock (porosity <1%) can be seen isolated in the fine grained fracture infill (porosity of 30-40%)

techniques is probably an artifact of stress release and sample preparation, leading to an over-estimation of the *in situ* retardation potential. Clearly this is non-conservative, from a PA viewpoint, and further work is currently ongoing, comparing the values for matrix diffusion depths and calculated diffusion coefficients from the various matrix diffusion studies carried out to date within the Nagra/JNC joint programme at the GTS. It is hoped that recommendations for conservative values for matrix diffusion parameters can be calculated from this work and an attempt will then be made to test these values via new *in situ* tracer experiments planned in the current phase of work in the GTS (see Kickmaier et al., 2000).

5. The new work: HPF and CRR

Hyperalkaline plume in fractured rock (HPF)

It has long been recognised (eg Ewart et al., 1985) that the use of large quantities of cementitious materials in an underground (deep or near-surface) radioactive or toxic chemical waste repository could, in the long term, give rise to a plume of hyperalkaline water displaced by groundwater flow from the repository into the host rock formation. Interaction of the rock with this hyperalkaline porewater could be detrimental to the performance of the host rock as a barrier to radionuclide migration (e.g Bath et al., 1987). While some potential processes, such as enhancement of fracture porosity by dissolution of host rock minerals and sealing of matrix porosity due to secondary mineral precipitation, are likely to be deleterious, other simultaneous processes may counteract these or even outweigh them to produce a positive effect on safety. Such positive processes include precipitation of secondary minerals with improved sorption characteristics compared to the primary mineralogy and sealing of fractures by secondary minerals inhibiting water movement and radionuclide transport.

Modelling, laboratory experiments and natural analogue studies carried out over the last few years have produced much useful, information and increased understanding of the various effects of a so-called high pH plume. However, these studies have been inconclusive in predicting the overall result with respect to host rock performance. This has been, at least in part, because each study concentrated on a particular aspect of the problem more or less in isolation and no one study attempted to consider all the information in an integrated manner (Alexander et al., 1998a).

The extensive use of cementitious materials is a feature of many repository designs for the geological disposal of radioactive and toxic wastes. For example, in the design for a low- and intermediate-level radioactive waste (L/ILW) repository in Switzerland, the near-field will be dominated by cement-based materials (current design concepts envisage the use of up to 1.5 million tonnes of concrete, approximately 85-90% by weight of the total repository). This concept should ensure considerable radionuclide retention in the near-field through the predicted low solubility of some key radionuclides of concern under the *in situ* hyperalkaline conditions allied to sorption on the large surface area of minerals and gels in cement (Hodgekinson and Robinson, 1987). Hence, failure of the physical containment, which may take place quite early, for example due to tectonic activity causing cracking of the cavern lining concrete, does not result in a catastrophic loss of safety of the engineered barriers.

However, experiments have shown that interaction of the host rock groundwater with concrete/cement will produce hyperalkaline leachates (often called a "hyperalkaline plume"), which are predicted to interact with the repository host rock. This will potentially affect the original retardation properties of the host rock by altering existing geochemical and hydrological conditions. The conceptual model for the potential formation of a hyperalkaline plume in a repository host rock is illustrated in Figure 12; also shown is the close link between the HPF experiment and the natural analogue studies performed in Jordan, which provide additional long-term data (Alexander, 1995; Smellie et al., 1997).

From a safety assessment viewpoint, the extent of formation of such an altered zone in the geosphere and the associated changes (positive or negative) in radionuclide retardation properties of the disturbed host rock must be carefully assessed.

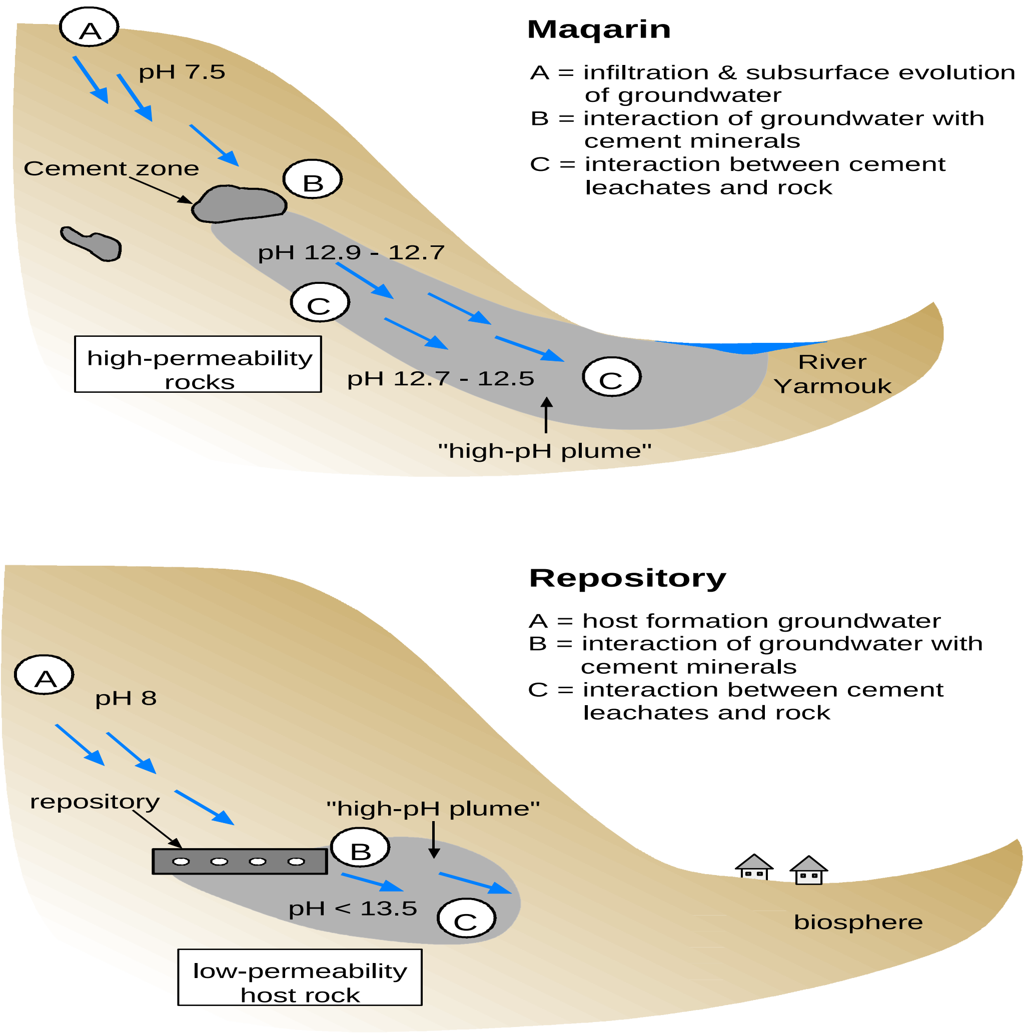

Figure 12: The potential formation of a hyperalkaline plume
Above: observations from the Maqarin Natural Analogue site, northern Jordan
Below: conceptual model for a repository host rock.

To date, model predictions of the potential effects vary enormously, some indicating that the host rock will be sealed by secondary mineral phases, thus minimising the groundwater flux, while others suggest significant damage to the host rock, with consequent increased groundwater flow through the system. This wide range of predictions is partly a reflection of the overall lack of data (laboratory and natural analogue) on hyperalkaline leachate/host rock interaction and is partly due to the fact that it is difficult to extrapolate the available data to in-situ host rock conditions.

Based on laboratory studies, modelling and detailed flow-field characterisation, a test field in the shear zone will be selected by the end of 1999 and a liquid hyperalkaline source and a suite of radionuclides will then be injected (see list of likely candidates above). Immobilisation of the altered zone by resin injection and subsequent sample recovery for detailed definition of radionuclide/cement sinks, in conjunction with definition of the flow system (à la RRP, see above) is foreseen for early 2002.

Colloid and radionuclide retardation (CRR)

The Engineered Barrier System (or near-field) of a repository will slowly degrade and eventually fail, potentially releasing small quantities of radionuclides to the host rock. It is expected that most nuclides will decay within the EBS or be retarded in the host rock (or far-field) surrounding the repository. However, failure of the engineered barriers is also likely to produce colloids, which could take up some of the released radionuclides and transport them through the host rock to the biosphere (see Figure 13). For many performance assessors (and regulators[1]), the effect of colloid-facilitated transport of strongly sorbing radionuclides generated at the near-field/far-field interface remains very much an open question. The broad objective of the CRR experiment is to provide an improved understanding of the stability and in-situ retardation of colloid associated, safety relevant radionuclides in the vicinity of the near-field/far-field interface.

Before starting the field tests in the summer of 2000, a programme of laboratory and modelling work has been performed with the intention of asessing the feasibility of the project and in providing important supporting data on *in situ* radionuclide solubility, the stability of bentonite colloids and the sorption behaviour of the various radionuclides on the bentonite and host rock. The results of this first phase of work have been very promising (and are summarised in Figure 14), indicating that no fundamental problems exist to prevent the field phase of work going ahead immediately.

During the ensuing *in situ* phase, radionuclide and colloid solutions will be injected into a dipole flow-field and the migration/retardation behaviour observed via tracer breakthrough. As with HPF, immobilisation of the experimental shear zone by resin injection and subsequent sample recovery for detailed definition of radionuclide/colloid sinks, in conjunction with definition of the flow system (à la RRP, see above) is foreseen for early 2002.

6. DISCUSSION AND CONCLUSIONS

The value of field experiments in model testing

Given the scales of time and space that must be considered in a PA, direct testing of the realism, or conservatism, of a model in the system of interest is impossible. Field experiments, such as those performed at the GTS, are consequently of great value in that:

- the fundamental transport processes that operate in the system are expected to be the same or similar to those relevant to any fractured repository host rock;
- the structures present, though differing in detail, are also similar to those of potential fractured repository host rocks;
- the scales of time and space over which the experiments operate, though often considerably smaller than those of performance assessment, are larger than those achievable in the laboratory;

[1] HSK, the Swiss Federal Nuclear Safety Inspectorate, noted in 1998 that "...the generation of colloids at the boundary of the bentonite package cannot be excluded...." and that "...colloid facilitated transport of strongly sorbing radionuclides at the near-field/far-field boundary remains very much an open question.."

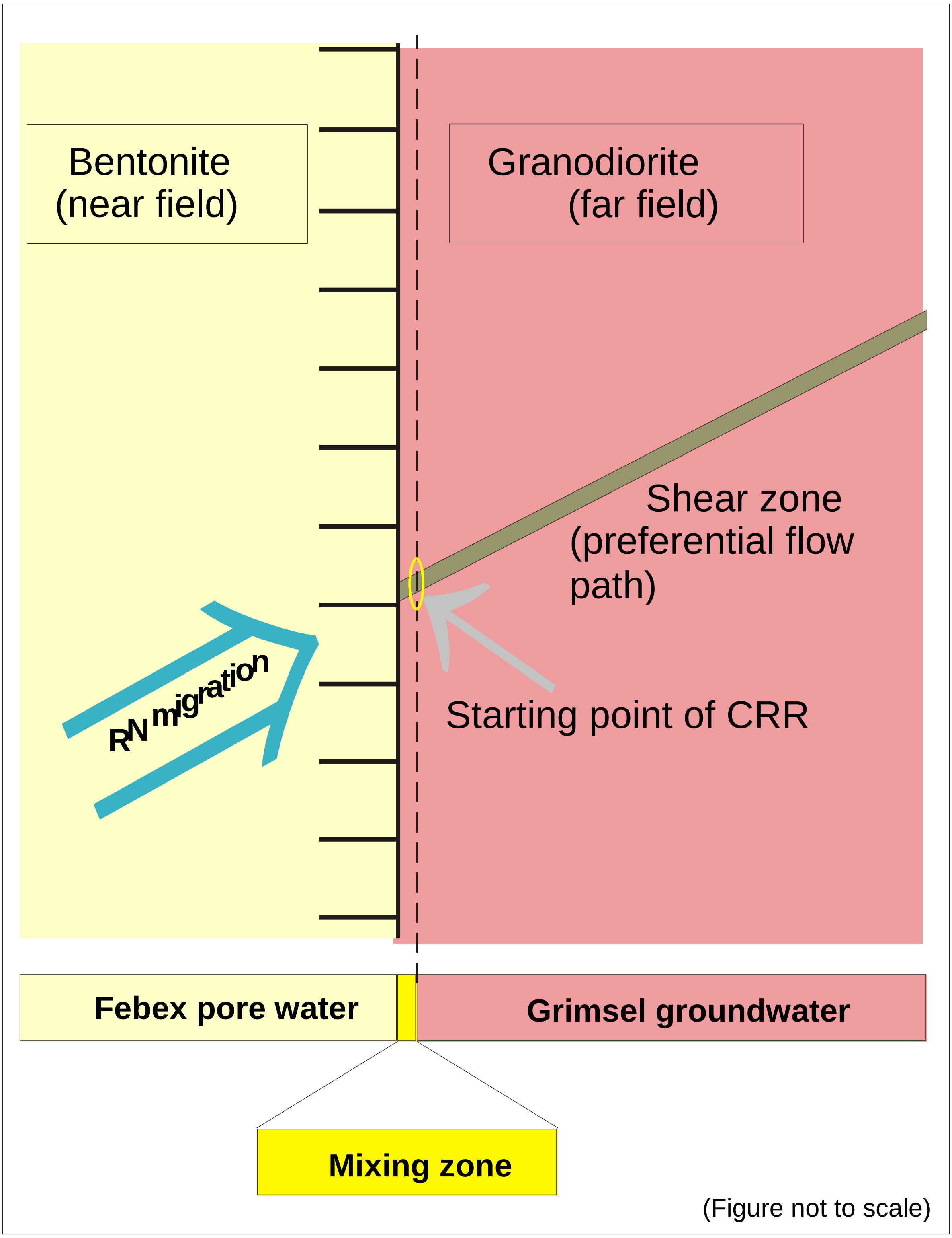

Figure 13: Concept behind the CRR project: radionuclides (RN) released from the bentonite could be transported through the far-field by colloids eroded from the bentonite backfill

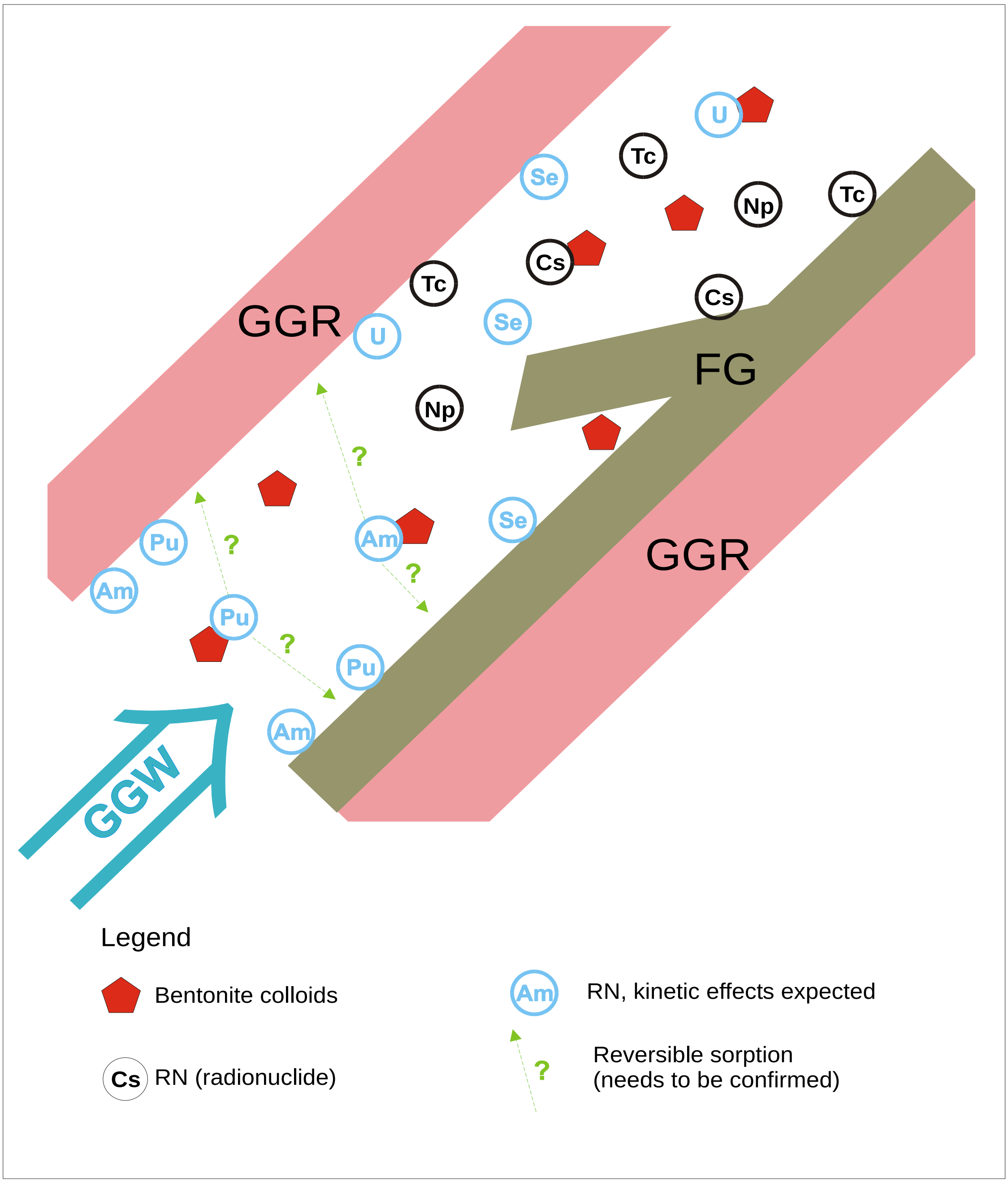

Figure 14: Summary of the preliminary laboratory results from CRR. The above diagram is a representation of a water-conducting fracture in the GTS. As an example of the radionuclide and colloid behaviour, it can be seen that Pu is strongly associated with the bentonite colloids, the host rock (GGR) and the fracture infill (FG) whereas Np stays preferentially in solution. In addition, the bentonite colloids are stable in the groundwater (GGW).

- the degree of characterisation of the system that is possible is greater than that achievable at a repository site (due to the smaller spatial scales involved in many field experiments and the need, at a repository site, to avoid perturbing the favourable properties of the system);
- field experiments, being performed *in situ*, are less subject to experimental artefacts than laboratory experiments.

Field experiments can never show beyond doubt that a model applied in performance assessment is "correct", given, for example, the differences in scales of space and time involved. They can, however, add to the body of observations and experiments with which the model is consistent and thus build confidence in its application in performance assessment. This can be further strengthened by carrying out a series of complementary and inter-related experiments as has been the case at the GTS with the MI, RRP (EP/CP) experiments (and the ongoing HPF and CRR projects) and their wide range of supporting field, laboratory and natural analogue studies (see also the comments on this approach in Alexander et al., 1998b).

Specific achievements in model testing

In the case of MI, the success of the modelling exercises gives confidence that relevant structures and processes are understood and adequately represented in the modelling approach, that no relevant processes have been overlooked and that, with appropriate consideration of the differences in conditions, laboratory data (*eg* on sorption and diffusion) can be applied to field-scale experiments, giving some support to their application in performance assessment.

Apart from the comments noted above, the greatest success has been the rigorous testing of the PSI developed PA transport code RANCHMD. Of particular note have been the attempts to minimise the number of free parameters in the code by including as many hard data on the shear zone structure, flow paths, tracer sorption values etc as possible (see Heer and Hadermann, 1996, for details). In this way, it has been possible to identify the in situ retardation mechanisms in a more thorough manner than has previously been the case.

In the case of RRP, the EP experiment has shown that the basic modelling approach developed in MI can be extended to the case of more chemically complex and more strongly sorbing radionuclides. It was also clearly shown that matrix diffusion played a significant role in radionuclide retardation in this particular system, as had been proposed by Frick et al. (1992) and Heer and Hadermann (1996) based on the modelling results. However, EP showed that rather than diffusion in the the rock matrix *senso stricto*, the radionuclides were preferentially retarded in the highly porous fault gouge present in the experimental shear zone. In addition, the complex effects of channelling, even over the relatively short scale (2 metres) of the experiment have clearly been shown to play a role in transport.

In line with a large number of natural analogue and laboratory studies, the CP experiment substantiates the existence of interconnected matrix porosity in crystalline rocks but clearly indicates the non-conservative (in the PA sense) nature of studying diffusion on laboratory samples due to the effects of stress release on the rock samples.

Limitations

The successful modelling of the MI and RRP radiotracer tests gives support to the model representation of structures and processes that exist in, or operate on, spatial and temporal scales that are similar to, or smaller than, the tests themselves. A major difficulty with the tests lies in the extrapolation of this conclusion to the larger scales that are relevant to PA: no information is provided on processes that, though irrelevant on the spatial and temporal scales of field tracer tests, may be important over scales relevant to PA. Natural analogues and palaeohydrogeology have an important role in this respect (see discussion in Miller et al., 1994).

With respect to the above comments, the greatest failure has been in the rigorous testing of the PA transport code RANCHMD. The problem here is not the code, rather the limitations imposed on the code by the experiment. PA transport codes such as RANCHMD are specifically developed to calculate the long-term, slow movement of radionuclides in the groundwaters of a repository site. Unfortunately, outwith experiments such as the RRP (see Alexander et al., 1996, 1998a), field tracer migration experiments cannot provide analogous conditions against which to test such transport codes. In the case of MI, for example, most experiments lasted days to weeks and even the longest experiment conducted, the last ^{137}Cs migration, no more than 20 months or so. This means that testing matrix diffusion within a code such as RANCHMD is relegated to observations based on highly porous matrix (or fracture fill) which may not be of much relevance to a given repository host rock. Also, kinetic effects may play an important role in the experiment but, obviously, are of no relevance to a repository PA.

Although CP has clearly shown the limitations in the dependence on laboratory derived diffusion parameters, it has not yet been possible to produce quantitative data on *in situ* processes as this work is still ongoing (final results are expected later in the year).

Although both HPF and CRR have not yet reached the field stage, the ongoing laboratory tests have already provided valuable data on *in situ* bentonite colloid stability and radionuclide solubility and, equally important, have produced laboratory retardation data which will be compared with the *in situ* data to further provide indications on the applicability of laboratory sorption data to the real conditions of a repository host rock

Future work

To look at something in hindsight is always an easy way to build an experimental programme and in MI/RRP several things would almost certainly be changed (for example, in RRP a more complete hydrological characterisation of the experimental site was produced before beginning the *in situ* work and, for HPF/CRR, better structural and petrological descriptions of the flow paths have been produced). However, a more realistic question might be `knowing now as little as you knew over ten years ago, at the beginning of MI/RRP, would you do it differently?`. In this case, it is likely that we would change much less: the entire experiment has been a learning experience for most of the people involved and has certainly contributed to our views on blind predictive testing and the development and testing of conceptual models of groundwater flow. One weakness, which has perhaps only now been acknowledged, is that, while the field experimenters, laboratory experimenters and transport modellers were in it together from the very beginning, the performance assessors were remarkable only by their absence. This would probably be the single greatest improvement possible to ensure the production of PA relevant data from any field tracer experiment - and the eventual inclusion of such data in a repository PA.

A full synthesis of the model testing carried out in the joint Nagra/JNC Radionuclide Migration Programme is not possible as the final conclusions of RRP are not yet available. This is certainly an area where future investment of effort would be profitable and it is hoped that this can be carried out in the final year of the millenium (*ie* 2000). This should clearly identify the lessons learnt during model testing and indicate those areas of potential concern for PA assessors everywhere.

Otherwise, the direction for future *in situ* experiments is clear: there must be greater integration of field experiments with appropriate natural analogues and laboratory data, everything being pulled together by the modelling of the systems. At the moment, the HPF experiment offers the clearest example of this approach but this must not be allowed to be the only one.

Conclusions

Overall, the field experiments at the GTS have enhanced confidence that the methodology adopted for:

- geological and hydrological characterisation of water-conducting features;
- simplification of this characterisation for modelling purposes;
- the adaptation of laboratory data (particularly sorption data) to field conditions;
- numerical solution of the governing equations for solute transport in dual-porosity media;

is indeed applicable to the modelling of solute transport through a fractured crystalline rock (note that some of the methodology developed in this programme has been, at least in part, further applied in the Kamaishi and Aspö rock laboratories).

In addition, limitations to our current understanding of matrix diffusion have been highlighted (and will be studied further) as well as the role of channelling in radionuclide retardation *in situ* in a repository host rock.

Acknowledgements

A task as large as the joint Nagra/JNC Radionuclide Migration Programme has many players and the authors would like to extend their thanks to all of our colleagues worldwide who have contributed in any form to the lessons learnt over the last 15 years. While the CRR and HPF projects would not now be in as advanced a state as they currently are without the input from their forefathers (namely MI and RRP), the teams involved here (CRR: ANDRA, BMWi/FZK, ENRESA, JNC, Nagra and USDoE/Sandia. HPF: ANDRA, JNC, Nagra and SKB) are warmly thanked for their efforts over the last two years.

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