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Äspö Hard Rock Laboratory

Deconvolution of breakthrough curves from TRUE-1 tracer tests (STT-1 and STT-1b) with sorbing tracers

Äspö Task Force, Task 4E

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November 1999

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Abstract

The report presents deconvolution of the experimental results from the sorbing tracer experiments STT-1 and STT-1b performed within the Tracer Retention Understanding Experiments (TRUE) programme at the Äspö Hard Rock Laboratory. Deconvolution is a mathematical treatment of the tracer test data where the experimental tracer injection curves and breakthrough curves are used to evaluate what the breakthrough curve would have looked like if the injection was performed as a pulse with unit mass and zero duration (Dirac function). A short description of the experiments and a discussion of different methods for deconvolution are given. Deconvolution appears to be a useful approach to evaluate tracer experiments in order to identify features in the breakthrough curves caused by transport processes. In particular the deconvoluted curves can be used for comparison with unit response curves obtained from model predictions. The method presently used for deconvolution has successfully deconvoluted most of the tracers used in the STT-1 and STT-1b tests.

Sammanfattning

Inom programmet Tracer Retention Understanding Experiments (TRUE) vid Äspölaboratoriet genomförs spårämnesförsök i olika skalor. I den första fasen, TRUE-1, har en serie spårämnesförsök med icke-sorberande och sorberande spårämnen genomförts mellan avgränsade sektioner i borrhål som penetrerar en enskild spricka. Dessa försök har använts av Äspös arbetsgrupp för modellering av grundvattenflöde och transport som ett underlag för prediktiv modellering. I Task 4C och 4D genomfördes prediktiv modellering av radiellt konvergerande försök (RC-1) och dipolförsök (DP-1 – DP-4) som genomförts med icke-sorberande spårämnen. I Task 4E och 4F genomför arbetsgruppen för modellering prediktiv modellering av spårämnesförsök med sorberande spårämnen (STT-1, STT-1b och STT-2).

För att möjligöra en utvärderingen av transportegenskaperna i berget från ett spårämnesförsök eftersträvar man en väldefinierad injektionspuls som är kort i förhållande till spårämnets gångtid. I praktiken är detta svårt att åstadkomma, vilket till exempel var fallet med de första försöken med icke-sorberande spårämnen där en avklingande pulsinjektion användes. Metoden för injektion har förfinats för de försök som utförts med sorberande spårämnen. I dessa tester byttes spårämnet i injektionssektionen ut mot vatten utan spårämne efter några timmar. På detta sätt erhölls en finit puls med endast begränsad avklingning pga utspädning.

För att kunna utesluta effekter orsakade av injektionspulsens utsträckning i tiden vid utvärdering av genombrottskurvor kan en matematisk metod användas på de experimentella data. Metoden innebär en inversfiltrering (deconvolution) och använder de experimentella genombrotts- och injektionskurvorna för att utvärdera hur genombrottskurvan skulle se ut om injektionen genomförts som en puls med enhetsmassa utan utsträckning i tiden (en Dirac-puls). Idealt så påverkas denna genombrottskurva – enhetsresponsfunktionen – endast av processer som sker under transporten. I verkligheten orsakar experimentella fel oscillationer och andra matematiska artefakter i enhetsresponsfunktionen. Därför krävs ofta en förbehandling av de experimentella kurvorna i form av filtrering eller kurvanpassning.

I denna rapport presenteras en inversfiltrering av de experimentella resultaten från spårämnesförsöken STT-1 och STT-1b som utförts med sorberande spårämnen. En kortfattad beskrivning görs av försöken och olika metoder för inversfiltrering redovisas. Det konstateras att inversfiltrering är en användbar metod för att utvärdera spårämnesförsök i syfte att identifiera företeelser i genombrottskurvorna orsakade av transportprocesser. Den kan också användas för att göra jämförelser med enhetsresponsfunktioner som beräknats med matematiska modeller. Metoden som använts för inversfiltrering har fungerat väl för de flesta spårämnen som använts i STT-1 och STT-1b försöken. Det finns dock ett behov för ytterligare förbättringar av metoden att hantera resultat med stora experimentella fel.

Executive Summary

The Tracer Retention Understanding Experiments (TRUE) programme includes tracer tests at different experimental scales at the Äspö Hard Rock Laboratory. Within the first stage, TRUE-1, a series of tracer experiments has been performed between packed off boreholes in a single feature using both non-sorbing and sorbing tracers. In support of the TRUE-1 tracer tests the Äspö Task Force on Modelling of Groundwater Flow and Transport of Solutes has performed predictive modelling of the TRUE-1 tracer tests. Tasks 4C and 4D comprised predictive modelling of the radially converging tracer tests (RC-1) and dipole tracer tests (DP-1 – DP-4) performed within the TRUE-1 tests using non-sorbing tracers. In Tasks 4E and 4F of the Äspö Modelling Task Force predictive modelling of the sorbing tracer tests (STT-1, STT-1b and STT-2) is performed.

In order to facilitate the evaluation of the transport properties from the tracer tests it is important to have a well-defined injection source term that is short in comparison with the tracer travel time. In practice this may be difficult to achieve, which was the case for the non-sorbing tracer tests where a decaying pulse injection was used. The injection methods have been improved for the tests with sorbing tracers by introducing a finite pulse injection. In these tests the tracer solution was replaced by unlabeled water after a few hours. This resulted in a finite pulse with only a slight decay due to dilution.

To evaluate the breakthrough curves excluding the effects caused by the injection procedure a mathematical treatment of the tracer test data can been done. This treatment, called deconvolution, uses the experimental tracer injection curves and breakthrough curves to evaluate what the breakthrough curve would have looked like if the injection was performed as a pulse with unit mass and zero duration (Dirac function). Ideally, all features of the resulting curve - the unit response function - are caused by processes occurring during the transport. In reality experimental errors may cause oscillations or mathematical artefacts in the unit response function. Therefore, mathematical manipulation of the experimental curves in the form of smoothing or curve fitting may be needed.

In this report a deconvolution of the experimental results from the sorbing tracer experiments STT-1 and STT-1b is presented. A short description of the experiments and a discussion of different methods for deconvolution are given. It is concluded that deconvolution is a useful approach to evaluate tracer experiments in order to identify features in the breakthrough curves caused by transport processes. In particular they can be used for comparison with unit response curves obtained from model predictions. The method presently used for deconvolution has successfully deconvoluted most of the tracers used in the STT-1 and STT-1b tests. However, there is a need for further improvement of the method in order to handle curves with large experimental errors.

Contents

List of Figures

List of Tables

1 Introduction

1.1 Background

The Äspö Hard Rock Laboratory (HRL) is an underground research facility situated on the east coast of Sweden operated by the Swedish Nuclear Fuel and Waste Management Company (SKB). The Äspö HRL provides opportunities to perform studies of behaviour and properties of the natural geological barriers, investigate interactions between engineered barriers and the host rock, and perform development and demonstration of technology for deep repository systems.

Within the Äspö Hard Rock Laboratory project a programme called Tracer Retention Understanding Experiments (TRUE) has been defined for tracer tests at different experimental scales. The overall objective of the TRUE programme is to increase the understanding of the processes which govern retention of radionuclides transported in crystalline rock, and to increase the credibility in the computer models for radionuclide transport which will be used in the licensing of a repository. Within the first stage, TRUE-1, a series of tracer experiments have been performed in a single feature using both non-sorbing and sorbing tracers.

1.2 Äspö Modelling Task Force

The Äspö Task Force on Modelling of Groundwater Flow and Transport of Solutes was initiated by SKB in 1992 as a forum for the organisations supporting the Äspö HRL Project. The purpose of the Task Force is to interact in the area of conceptual and numerical modelling of groundwater flow and solute transport in fractured rock. In particular, the Task Force proposes, reviews, evaluates and contributes to such work in the HRL Project.

Task 4 of the Äspö Modelling Task Force consists of modelling exercises in support of the TRUE-1 tracer tests. Tasks 4C and 4D comprised predictive modelling of the radially converging tracer tests (RC-1) and dipole tracer tests (DP-1 – DP-4) performed within the TRUE-1 tests using non-sorbing tracers. The tests were performed between packed off boreholes penetrating a water-conducting geological feature with a simple structure (Feature A). These tests have been followed by tests with sorbing radioactive tracers (STT-1, STT-1b and STT-2) in the same feature. In Tasks 4E and 4F of the Äspö Modelling Task Force predictive modelling of the sorbing tracer tests is performed.

1.3 Deconvolution

In order to facilitate the evaluation of the transport properties from the tracer tests it is important to have a well-defined injection source term that is short in comparison with the tracer travel time. In practice this may be difficult to achieve. In the radially converging test tracers a decaying pulse injection was used. Tracers were injected in the borehole sections and initially circulated in order to achieve a rapid homogenisation. Thereafter, the tracer concentration in the injection section decreased slowly due to the induced flow obtained by the pumping. Due to the low flow rates and the relatively large volumes of the injection sections, the decay of the injection pulse was slow. The time until the concentration reached half its original value was of the same magnitude as the median breakthrough time of the tracers (Figure 1-1).

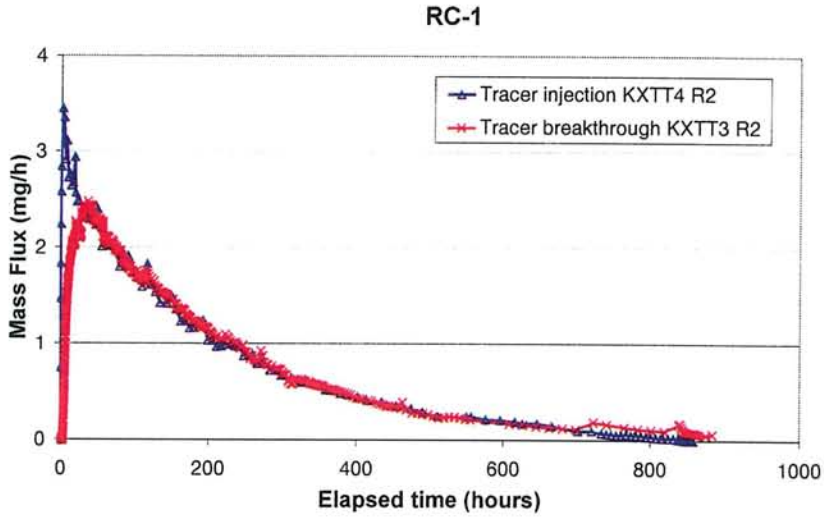

Figure 1-1 Tracer injection and breakthrough in RC-1.

The same methodology was also used in the dipole tests. However, the decay of the pulse was much faster due to the flow induced by the injection of unlabeled water in the injection sections (Figure 1-2).

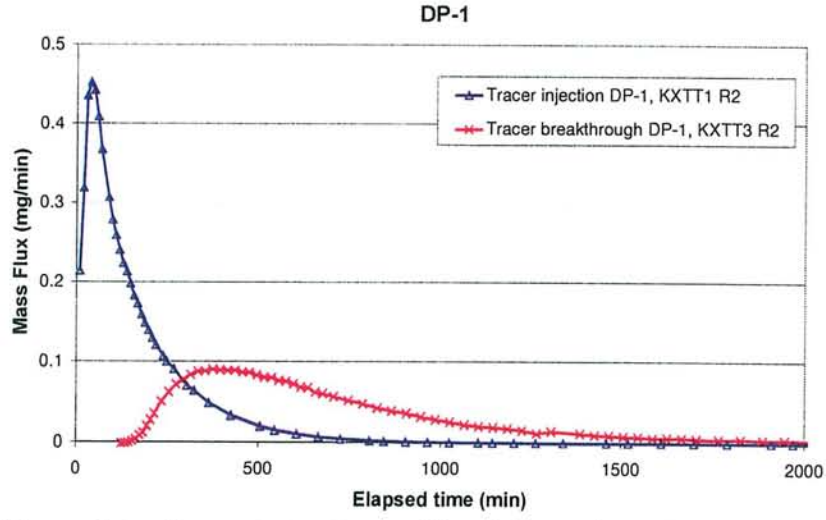

Figure 1-2 Tracer injection and breakthrough in DP-1.

The injection methods have been improved for the tests with sorbing tracers by introducing a finite pulse injection. In these tests the tracer solution was replaced by unlabeled water after a few hours, after which the tracer concentration in the injection section decreased rapidly by a factor of 30. This resulted in a finite pulse with only a slight decay due to dilution. After the replacement of the injection fluid the tracer concentration increased slightly again (Figure 1-3).

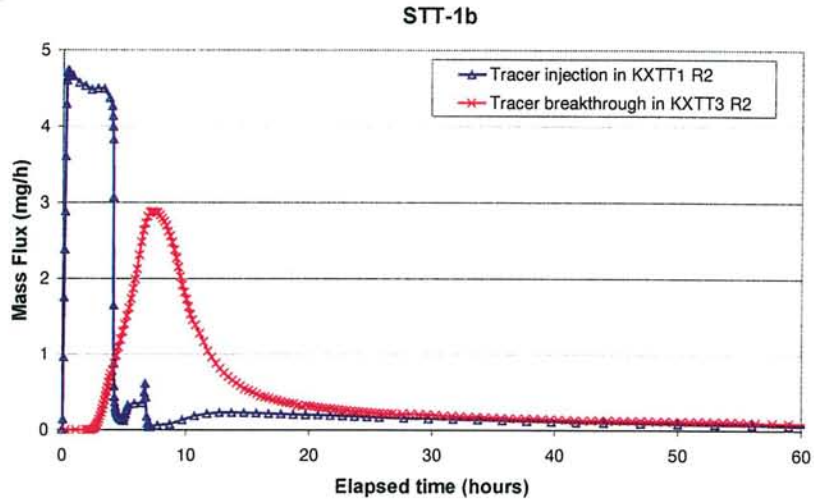

Figure 1-3 Tracer injection and breakthrough in STT-1b.

To evaluate the breakthrough curves excluding the effects caused by the injection procedure a mathematical treatment of the tracer test data can been done. This treatment, called deconvolution, uses the experimental tracer injection curves and breakthrough curves to evaluate what the breakthrough curve would have looked like if the injection was performed as a pulse with unit mass and zero duration (Dirac function). Ideally, all features of the resulting curve - the unit response function - are caused by processes occurring during the transport. In reality experimental errors may cause oscillations or mathematical artefacts in the unit response function. Therefore, mathematical manipulation of the experimental curves in the form of smoothing or curve fitting may be needed.

In this report deconvolution of the experimental results from the sorbing tracer experiments STT-1 and STT-1b is presented. The report also gives a short description of the experiments and discusses different methods for deconvolution.

2 Tracer tests with sorbing tracers (STT-1, STT-1b)

The two tracer tests STT-1 and STT-1b were performed between different borehole sections penetrating Feature A. A detailed experimental description of STT-1 is given in Andersson et al. (1998) and of STT-1b in Andersson et al., (1999). The main objective for the tracer tests with sorbing tracers was to test equipment and procedures for tests with radioactive sorbing tracers to be performed in later stages of the TRUE Project. Secondly, to increase the understanding of transport and retention of sorbing species in crystalline rock and to obtain in-situ sorption data.

STT-1 was performed in Feature A in a radially converging flow geometry between borehole sections KXTT4 R3 → KXTT3 R2, see Figure 2-1. The pumping rate was 0.4 l/min. In total eight tracers, two conservative (Uranine and tritiated water) and six weakly to moderately radioactive sorbing tracers (^{22}Na, ^{47}Ca, ^{85}Sr, ^{133}Ba, ^{86}Rb, ^{137}Cs) were mixed and injected as a finite pulse with a duration of four hours. Tracer breakthrough in the pumping section was monitored for all eight tracers injected. The breakthrough curves show one distinct and high peak and a lower secondary peak reflecting the shape of the injection function.

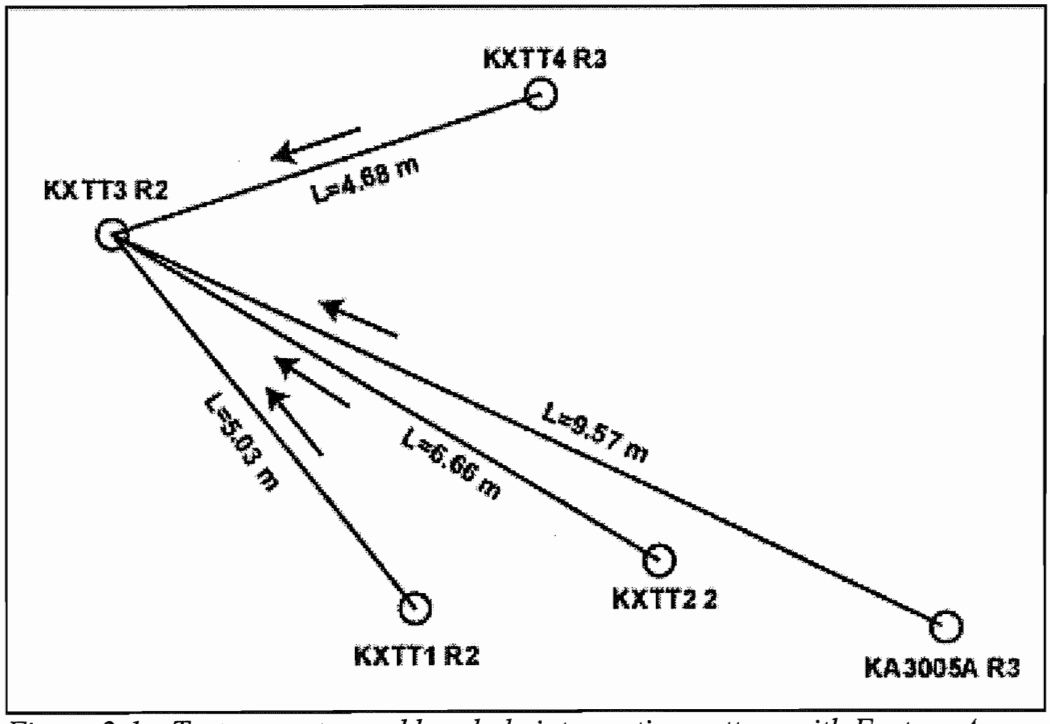

Figure 2-1 Test geometry and borehole intersection pattern with Feature A.

The STT-1b experiment was performed in a radially converging configuration where tracers were injected in KXTT1 R2 with pumping in KXTT3 R2 at a rate of 0.4 l/min (Figure 2-1). A solution containing ten different tracers (Uranine, HTO, Br, I , ^{22}Na, ^{42}K, ^{85}Sr, ^{86}Rb, ^{58}Co, ^{99m}Tc) were injected simultaneously as a finite pulse with a duration of 4 hours. The radioactive tracers were recovered in-line with a HpGe detector.

2.1 Experimental setup

2.1.1 Equipment

Each borehole in the TRUE-1 array is instrumented with inflatable packers such that 4-5 borehole sections are isolated. The borehole sections in Feature A were also equipped with volume reducers (dummies). The equipment was tested for sorption of the tracers used in STT-1 and no significant sorption could be detected. The borehole fluid is circulated in the injection borehole in order to obtain a homogeneous tracer concentration inside the borehole and to be able sample the tracer concentration outside the borehole. Circulation is controlled by a pump with variable speed and measured by a flow meter.

2.1.2 Tracers used

During STT-1, a mixture of in total eight different tracers, both conservative and sorbing, was injected. The sorbing tracers used were six radioactive, gamma-emitting, isotopes of mono- and divalent cations, see Table 2-1. These tracers needed to be injected in such low concentrations that the chemical conditions were kept unchanged in Feature A. Another restriction was the maximum permitted dose. The conservative tracers were Uranine (Sodium Fluorescein) and tritiated water (HTO).

Table 2-1 Tracers used in STT-1.

Name	Isotope	Half-life
Uranine	-	-
HTO	^{3}H	12.3 a
Sodium	^{22}Na	2.6 a
Calcium	^{47}Ca	4.5 d
Rubidium	^{86}Rb	19 d
Strontium	^{85}Sr	65 d
Barium	^{133}Ba	10.5 a
Caesium	^{137}Cs	30.2 a

For STT-1b some modifications were made of the tracers, see Table 2-2.

Table 2-2. Tracers used in STT-1b.

Name	Isotope	Half-life
Uranine	-	-
HTO	^{3}H	12.3 a
Bromine	-	-
Iodine	-	-
Sodium	^{22}Na	2.6 a
Potassium	^{42}K	12.4 h
Cobalt	^{58}Co	70.9 d
Rubidium	^{86}Rb	19 d
Strontium	^{85}Sr	65 d
Technetium	^{99m}Tc	6 h

2.1.3 Injection and sampling

The tracer solution (3.5 litres) containing all eight tracers was mixed with synthetic groundwater and mixed into a tracer stock solution with the non-sorbing tracers HTO and Uranine. The injection of tracer was made in the circulation loop as a finite pulse injection with a length of four hours. After four hours of injection the tracer solution was exchanged with unlabelled water. The exchange procedure lasted for 72 minutes.

The tracer concentration in the injection loop is measured both in situ using a HPGe-detector and by sampling and subsequent analysis in the laboratory. The sampling is made by continuously extracting a small volume of water from the system through a flow controller (constant leak) to a fractional sampler. The decrease in injection concentration was measured by sampling for Uranine and HTO with samples taken every 2nd minute during the initial 40 minutes of injection, and then every 30 minutes up to four hours. The sampling frequency was then increased again to one sample every 4th minute during the exchange procedure. After that samples were taken once every hour. After some months the sampling frequency was gradually decreased to one sample per week. The radionuclides were measured with the on-line detector with a somewhat higher frequency than the discrete sampling during the first five hours. After that activity measurements were made over a period of one hour. At later stages of the sampling, measurements of Cs activity were made over 12-hour periods.

The sampling system in the pumping borehole is based on the same principle as the injection system, namely a circulating system with a circulation pump and a flow meter. The sampling was made with two independent systems, a "constant leak" system producing 8 ml samples integrated over a period between 5 and 100 minutes, and a 24-valve sampling unit producing 1 litre samples discrete in time. Both systems took samples every 10th minute during the first two hours, thereafter the decreasing sampling frequency gradually decreased down to one sample per day after two months.

2.2 Modelling Task 4E

The modelling Task 4E was defined with the overall objectives of developing the understanding of radionuclide migration and retention in fractured rock and evaluating the usefulness and feasibility of different approaches to model radionuclide migration of sorbing species based on existing in situ and laboratory data from the TRUE-1 site.

The task includes predictive modelling of the sorbing tracer experiments STT-1 and STT-1b. At the 11th Äspö Task Force Meeting in September 1998 11 modelling teams had submitted predictions of the tracer experiments.

3 Deconvolution approach

3.1 Introduction

Convolution and deconvolution techniques are used in signal processing and in other problems where the purpose is to analyse a linear system where the output function is given by an input function and a unit response function. For evaluation of tracer transport convolution can be used to obtain a breakthrough curve for a given injection curve when the unit response function is known, see Figure 3-1. Convolution is often used in mathematical modelling of solute transport. In this case a single unit response function for a system is calculated and then used to derive breakthrough curves for arbitrary input functions.

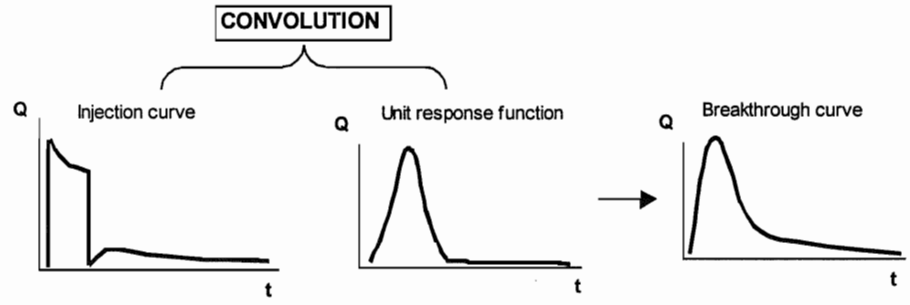

Figure 3-1 Principle of convolution.

The inverse of convolution is called deconvolution and can be used to find the unit response function from the injection curve and the breakthrough curve. The unit response function describes how the breakthrough curve would look like if the injection curve was a Dirac delta function, i.e. a pulse with unit mass and zero duration. In Figure 3-2 the deconvolution principle is described schematically.

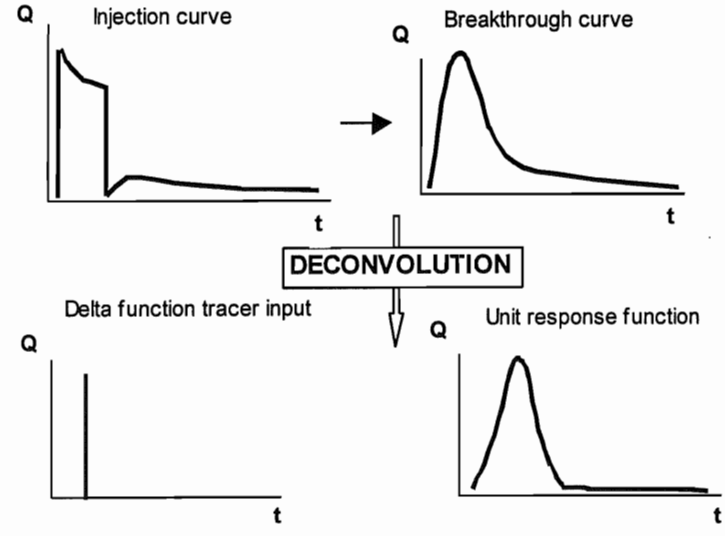

Figure 3-2 Principle of deconvolution.

The unit response function describes the characteristic features of the particular flow and transport problem determined by the transport mechanisms along the flowpaths. The unit response function is unaffected by the boundary conditions for the tracer, i.e. injection concentration as a function of time. Since any effects due to the shape of the injection curve is removed, the unit response function can be used as a diagnostic tool for identification of the relevant transport mechanisms, for example early time diagnostics, tailing and presence of multiple peaks. It can also be used for parameter estimation (porosity, dispersivity, etc.).

The convolution and deconvolution is based on the superpositioning of solutions. It is therefore required that the transport processes are linear as regard to concentration and that they are invariant in time, i.e. processes such as non-linear sorption, etc cannot be treated.

3.2 Deconvolution techniques

Deconvolution is an ill-posed problem, small measurement errors in the injection and breakthrough curves may cause numerical problems (e.g. oscillations) and large errors in the calculated response functions. The physical constraints on the unit response function can be used to improve the solution. For example requirements on the stability of the obtained solution, the exclusion of negative values of the unit response function as they have no physical meaning.

A large range of techniques has been applied to problems in signal processing, astrophysics, geophysics and image processing. A basic method is the use of Fourier Transforms including various types of filtering techniques. However, Fourier transforms are not very suitable for deconvolution, e.g. due to spectral leakage (Ilvonen et al., 1994). Other techniques use the Kalman filter that gives an optimal estimate of a given process allowing for updates based on output measurements. However, the Kalman filter requires an accurate model of the system in order to achieve a good performance.

Several techniques have been used for deconvolution in connection with solute transport. In a method called regularisation a solution is found by minimising an objective function containing two terms, one that measures the fit of the solution and a second that measures other properties of the solution, e.g. smoothness (cf. Skaggs et al., 1998). Deconvolution of tracer test data from Finnsjön has been made using the Extreme Value Estimation Method (EVE) (Ilvonen et al., 1994) and the Toeplitz method (Tsang et al., 1991). The EVE method solves a linear set of equations where all unknowns are required to be non-negative. Estimates are given of the upper and lower bands of the unknowns.

The deconvolution technique used in this report is based on the Toeplitz method (Tsang et al., 1991). The tracer injection mass flow rate m_j is given in discrete steps where j is the index for successive times. Also the tracer mass breakthrough M_i is discretisized with i as the index for successive times. For each input element m_j at time t_j a unit response function a_{ij} is defined. The response function must fulfil some special properties. The response time i must be later than the input time j and the response function must be time invariant and therefore the response function a_{ij} is only a function

of t_i-t_j which means that a_{ij}=a_{i-j}. Then the output M_i can be given as the superposition for all the input elements (m_j) times the response function a_{i-j}:

$$M_i = \sum_j a_{ij} \cdot m_j$$

where

$a_{ij} = a_{i+n,\, j+n}$ (time invariant)
$a_{ij} = 0$ for $i \leq j$

The transfer coefficients a_{ij} defined as above is a Toeplitz matrix and because of its time invariance *(i-j)* it can be described as a vector a_s where $s = i\text{-}j$:

$$M_i = \sum_s m_{is} a_s$$

where
$a_s = a_{ij}$ $\quad s = i\text{-}j,\ i > j$
$m_{is} = m_j$ $\quad j = i\text{-}s$
$m_{is} = 0$ $\quad i \leq s$

The vector a_s, the response function to a unit pulse injection at the first time interval, and the m_{is} coefficients become in matrix form:

$\mathbf{M} = \mathrm{m} \cdot \mathbf{a}$

If this expression is inverted it gives the unit response function as:

$\mathbf{a} = \mathrm{m}^{-1}\,\mathbf{M}$

The different methods available in Matlab for matrix inversion have been tested:

- the "backslash" operator where A\B is the matrix division of A into B. For square systems (A is an N-by-N matrix and B is a column vector with N components), X = A\B is the solution to the equation A*X = B computed by Gaussian elimination. For an under- or overdetermined system of equations A*X = B, the effective rank, K, of A is determined from the QR decomposition with pivoting. A solution X is computed which has at most K nonzero components per column.

- the function `nnls` (Non-negative least-squares) which calculates the vector X that minimizes NORM(A*X - b) subject to X greater or equal to zero. Thus the non-physical solutions with negative unit response functions can be avoided.

- the function `pinv` which uses the Moore-Penrose pseudoinverse of a matrix non-square matrix.

- the function `cgs` which uses the Conjugate Gradients Squared method to iteratively solve the system of linear equations A*x = b for x.

The Toeplitz method has proved to be considerably more stable than using the deconvolution routine (`deconv`) included in Matlab. With this routine it was not possible to obtain stable solutions for the unit response function with the original experimental data for any of the tracers.

3.3 Mathematical treatment of experimental data

3.3.1 Fitting of injection curves

Some of the injection curves were highly irregular and to facilitate the deconvolution they were smoothed with curve fitting. Because of the complexity of the injection curves they were divided into several parts and to each part a simple function (e.g. linear fit, logarithmic fit) was fitted. New injection values were calculated with fitted functions and used in the deconvolution calculations.

3.3.2 Filtering of breakthrough curves

Also some of the breakthrough curves were irregular. The result of the deconvolution is very sensitive to the detailed shape of the breakthrough curve. Curve fitting may either eliminate details that are present or introduce artificial details in the curves. Therefore, irregular breakthrough curves, e.g. spiky curves, were filtered. Although filtering also is a manipulation of the results it was considered to create less potential problems than curve fitting. The Matlab function *filter (b,a,x)* was used as a moving average filter on the breakthrough concentration values. Below is an example of the algorithm presented for a case when averaging over a four hour window:

$$y(n) = \frac{1}{4}x(n) + \frac{1}{4}x(n-1) + \frac{1}{4}x(n-2) + \frac{1}{4}x(n-3)$$

x are the original breakthrough values
y are the filtered breakthrough values
n is the index of current sample

The algorithm described above is averaging backwards and therefore the filtered concentration/flow vector had to be shifted forward, in this case two steps, relative to the time vector.

3.4 Convolution of the unit response function

To verify the accuracy of the unit response functions, calculated with the Toeplitz method, convolution has been done on the response functions and the corresponding injection curves. The original breakthrough curves from the tracer experiments are compared to the calculated breakthrough curves from the convolution. The convolution procedure used in this report is based on the Matlab function *conv (a,b)* which convolves vectors a and b:

$$c(k) = \sum_j a(j) \cdot b(k+1-j)$$

where
a is the unit response function of length m
b is the discrete injection mass flow rate of length n
c is the convoluted breakthrough curve of length $m+n-1$

4 Results of deconvolution

4.1 Experimental results

4.1.1 Tracer test SST-1

The injection concentrations versus time for the different tracers in STT-1 are shown in logarithmic scale Figure 4-1. There is a sharp increase in concentration during the initial part of the injection, followed by a decline in concentration due to dilution by the flow through the borehole section. The decline is more pronounced for the sorbing radionuclides due to sorption on the borehole walls. After 4 hours the injection fluid was exchanged with unlabelled water which gives a rapid decrease in concentration by a factor 30. This is followed by an increase in concentration probably due to tracers remaining in a stagnant zone within the injection section. Finally, the concentration declines due to dilution.

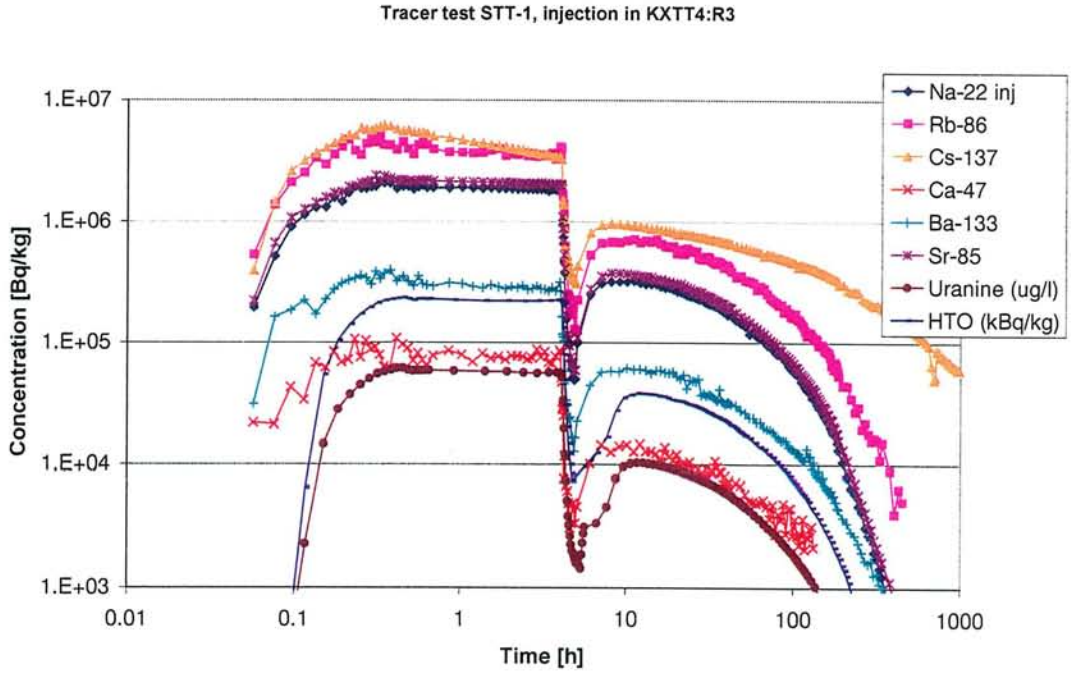

Figure 4-1 Injection concentration versus time for STT-1.

In STT-1 all of the eight injected tracers were recovered in the pumping section KXTT3 R2. Figure 4-2 shows the breakthrough concentration as a function of time in a logarithmic scale. For Na-22, Sr-85, HTO and Uranine breakthrough occurs at a similar time and the curves also have a similar shape. For these tracers there is an indication of a second peak occurring after the primary. This is likely due to the shape of the injection curve. The tracers Ba-133, Rb-86, Cs-137 show a varying degree of retardation in the breakthrough.

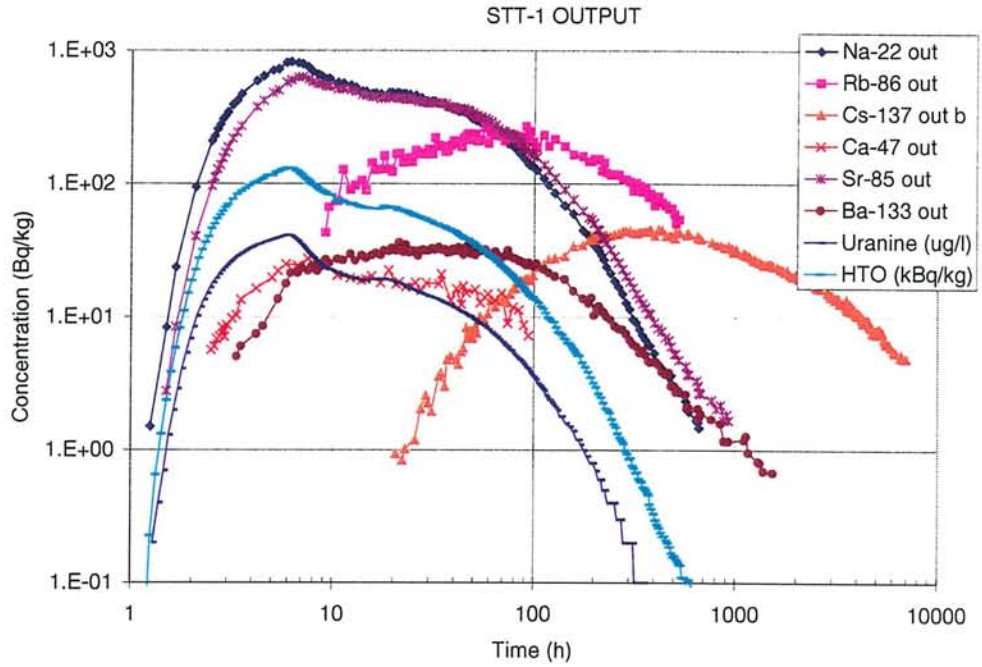

Figure 4-2 Breakthrough concentrations for STT-1.

4.1.2 Tracer test STT-1b

The injection concentration curves in STT-1b have a shape similar to those of STT-1, see Figure 4-3, an initial peak with a duration of 4 hours ending with a rapid decline in concentration due to the exchange of water in the injections section. This is followed by an increase in concentration probably due to tracer present in stagnant water in the injection section. The main difference is that in STT-1b there is an additional decline in concentration after about 7 hours, caused by a second exchange of water in the injection section.

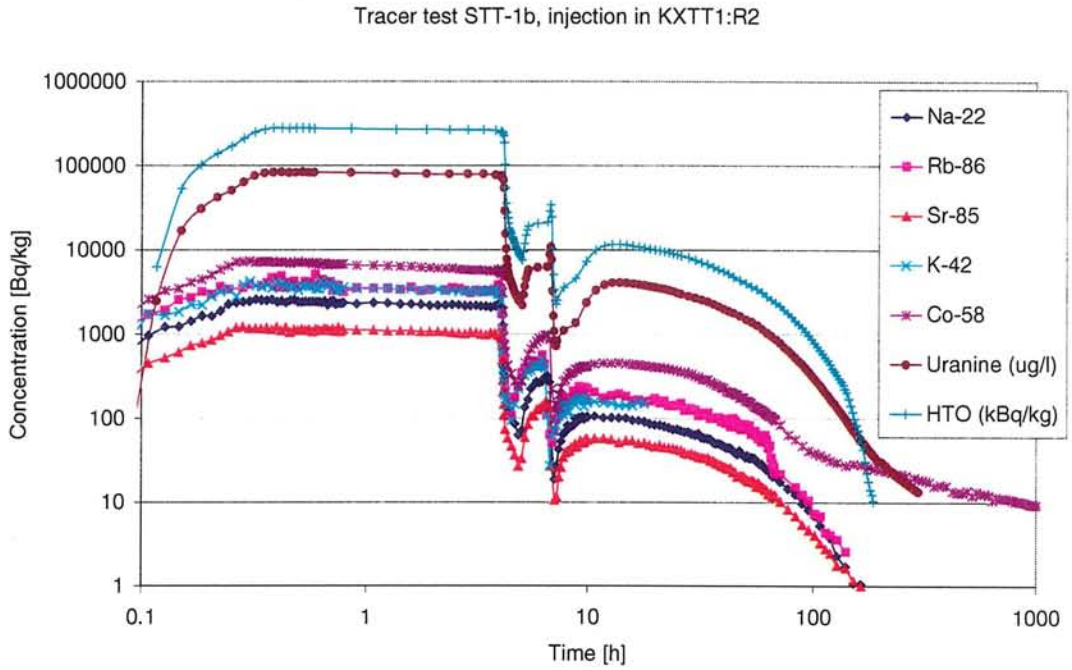

Figure 4-3 Injection concentration for STT-1b.

In tracer test STT-1b recovery was obtained for nine of the injected tracers, but not for Tc-99m. Figure 4-4 shows the breakthrough curves as function of time in a logarithmic scale. Also in this test the curves for HTO, Uranine and Na-22 have a similar shape. A somewhat more pronounced delay of Sr-85 than in STT-1 can be observed. The breakthrough curve of K-42 is rather oscillating and contains only few points. A marked delay of Rb-86 and Co-58 can be observed.

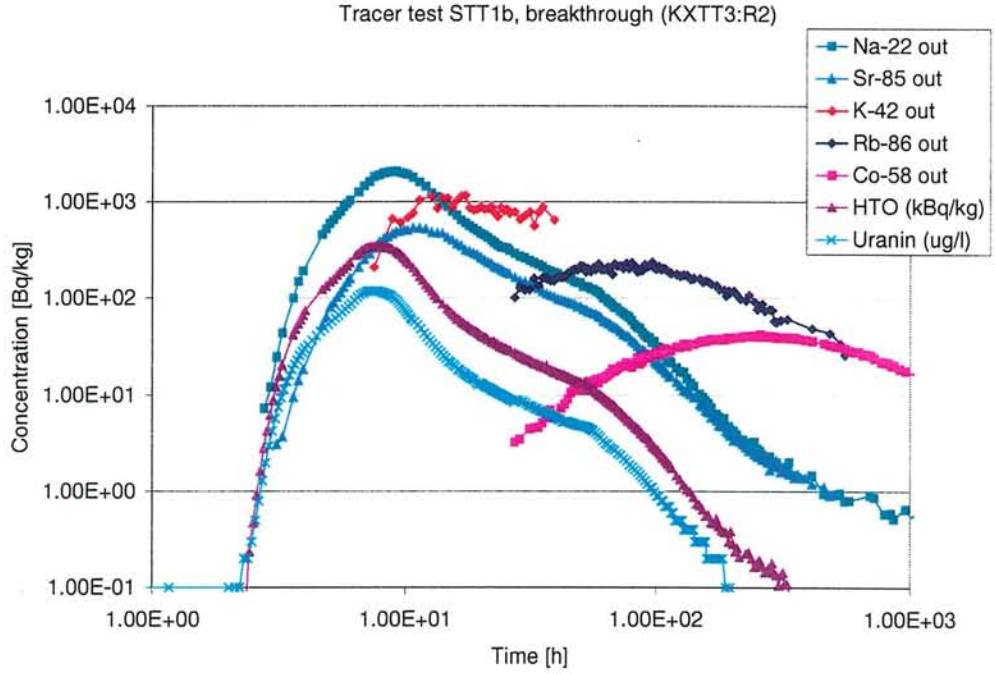

Figure 4-4 Breakthrough concentrations for STT-1b.

4.2 Deconvolution of STT-1 test

Deconvolution of the experimental results from the STT-1 test has been performed on the tracers: Uranine, HTO, Sr-85, Na-22 , Ba-133, Rb-86, Ca-47, Cs-137. However, for Ca-47 it was not possible to make a successful deconvolution. In the case of Ca-47 the breakthrough curve was too short to be able to obtain a successful deconvolution.

For Uranine, HTO, Na-22 and Sr-85 the curve fitting could be performed on the original experimental data. For Ba-133, Rb-86 and Cs-137 the experimental curves were too rough to perform deconvolution of the original data and therefore a curve fitting was made of the injection curve and a smoothing of the breakthrough curve. The size of the time step, the treatment of data and the matrix inversion method used for the different tracers is summarised in Table 4-1. A trial and error methodology was chosen to find the smallest time step that could be used without obtaining problems with oscillations.

Table 4-1 Time steps and methods used for deconvolution of the STT-1 test.

	Start time (h)	Time step (h)	Injection curve	Breakthrough curve	Inversion method
Uranine	0.21	2.1	experimental	experimental	nnls
HTO	0.13	2.01	experimental	experimental	backslash
Na-22	0.05	2.04	experimental	experimental	backslash
Sr-85	0.06	2.04	experimental	experimental	backslash
Ba-133	0.0001	2.05	curve-fitted	smoothed	backslash
Rb-86	0.05	2	curve-fitted	smoothed	pinv
Ca-47			Incomplete breakthrough curve		-
Cs-137	0.2	1	experimental	smoothed	cgs

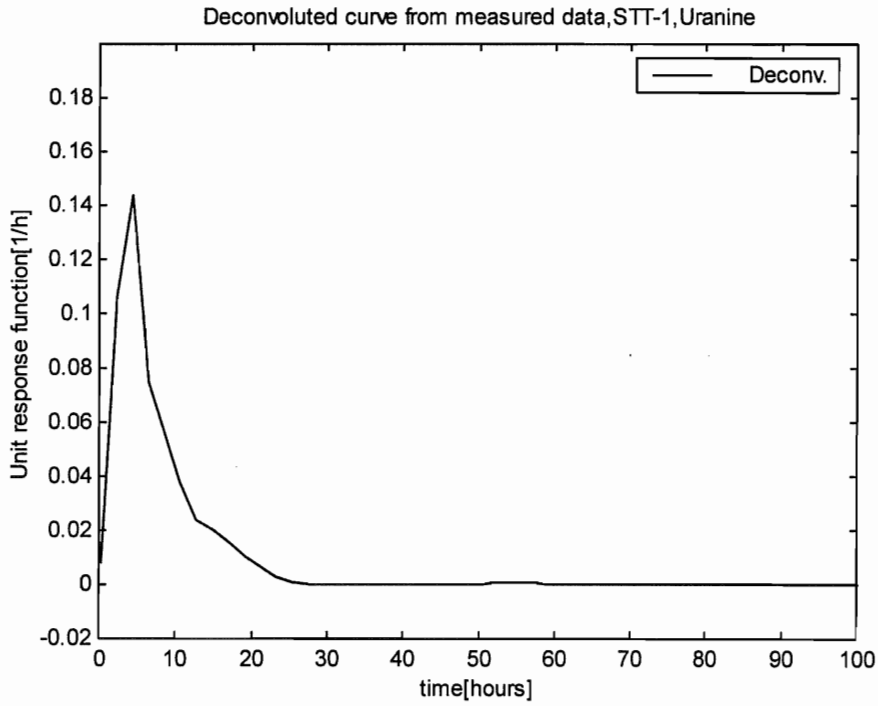

Figure 4-5 Unit response curve for Uranine in STT-1.

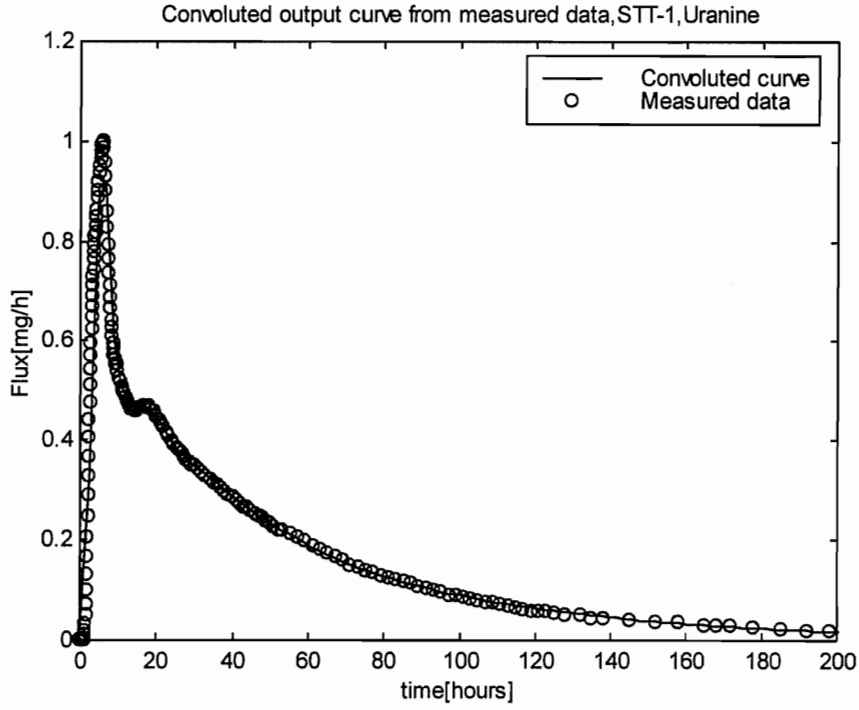

Figure 4-6 Convoluted unit response curve for Uranine in STT-1 compared with measured data.

The deconvoluted curve for Uranine shows a single sharp peak with some tailing. However, the time step used (around 2 hours) was too large to reveal all details of the peak. In particular it gives a less sharp rise in the curve. Smaller time steps generated unstable solutions. The nnls function was necessary to use in order to avoid having a negative unit response function after the peak. A convolution of the unit response curve

with the injection curve gives results very similar to the measurements from the experiment, which indicates a successful deconvolution.

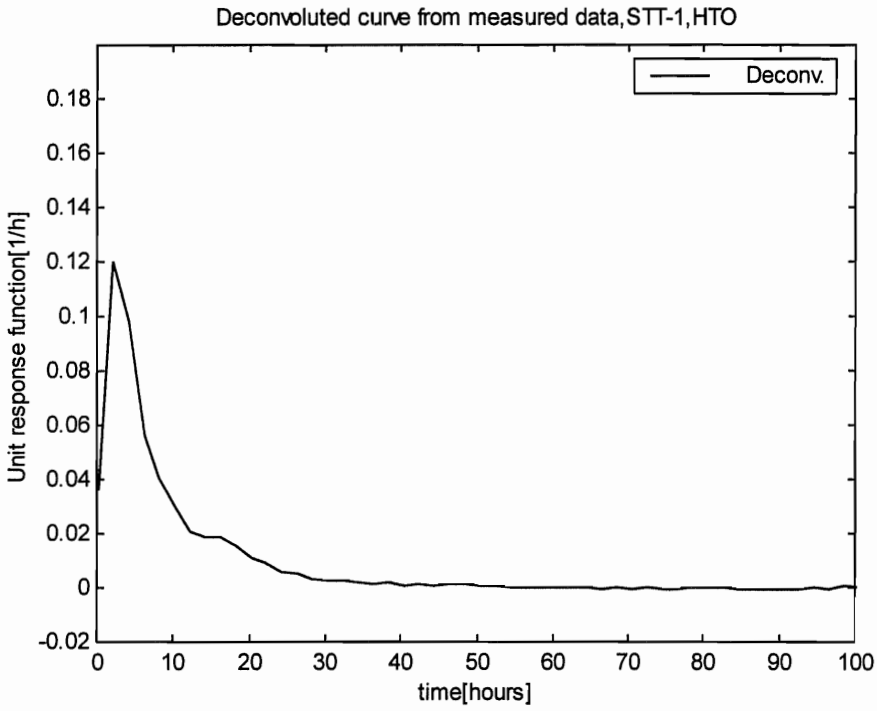

Figure 4-7 Unit response curve for HTO in STT-1.

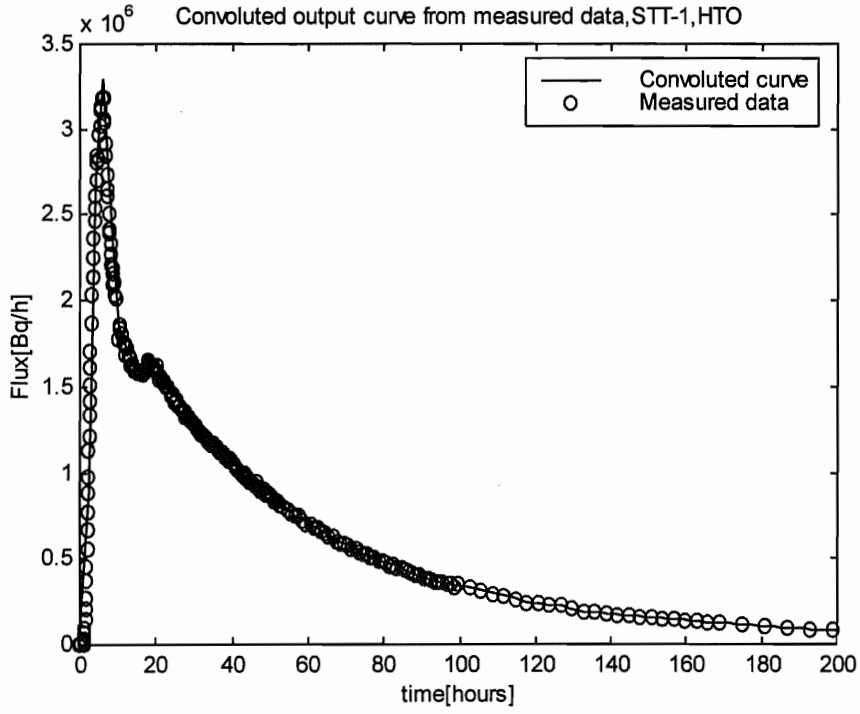

Figure 4-8 Convoluted unit response curve for HTO in STT-1 compared with measured data.

The unit response function for HTO has a similar appearance to that of Uranine. There are indications of a smaller second peak after 15 hours. However, this may be a mathematical artefact.

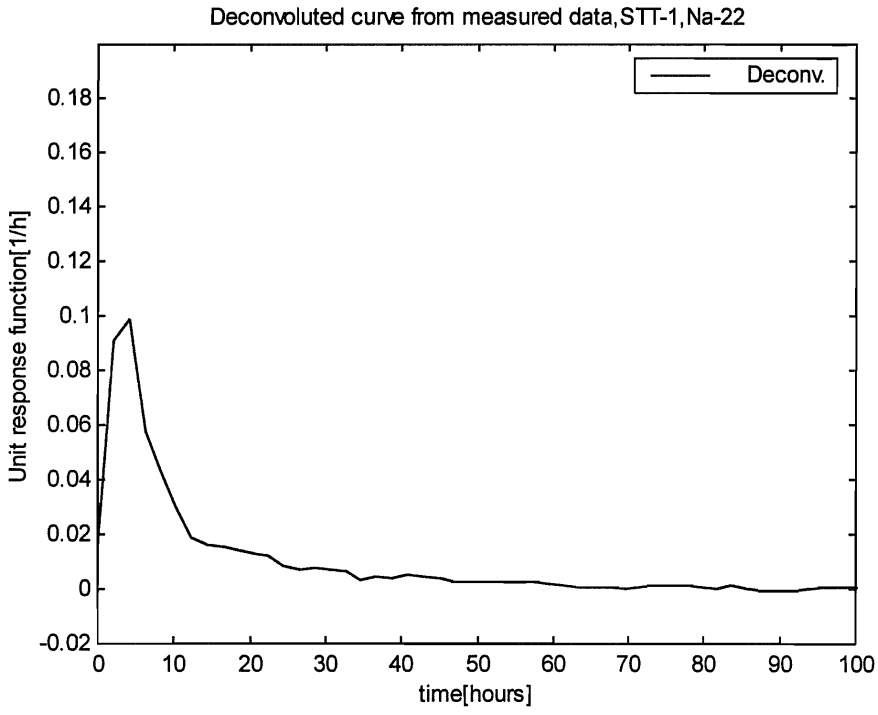

Figure 4-9 Unit response curve for Na-22 in STT-1.

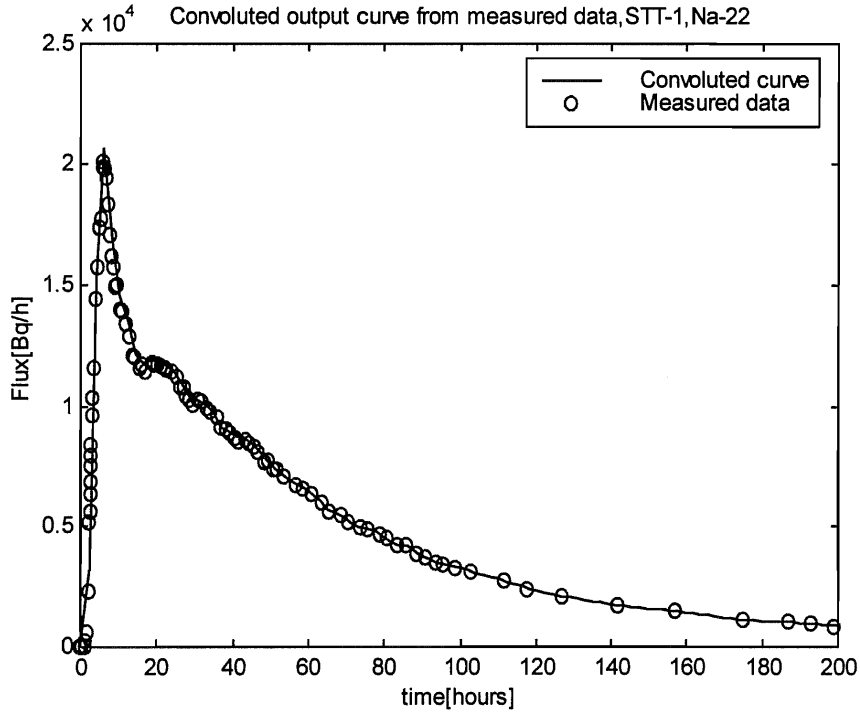

Figure 4-10 Convoluted unit response curve for Na-22 in STT-1 compared with measured data.

The curve for Na-22 is nearly identical to that of HTO. The difference seen near the actual peak is probably caused by the different time step used. There is also a somewhat more pronounced tailing for Na-22.

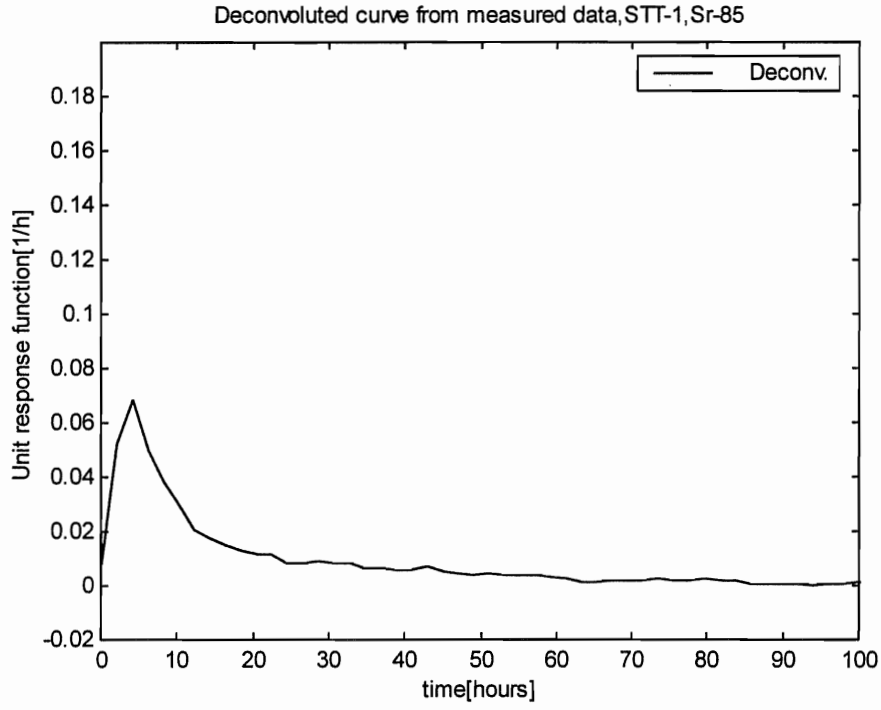

Figure 4.11 Unit response curve for Sr-85 in STT-1.

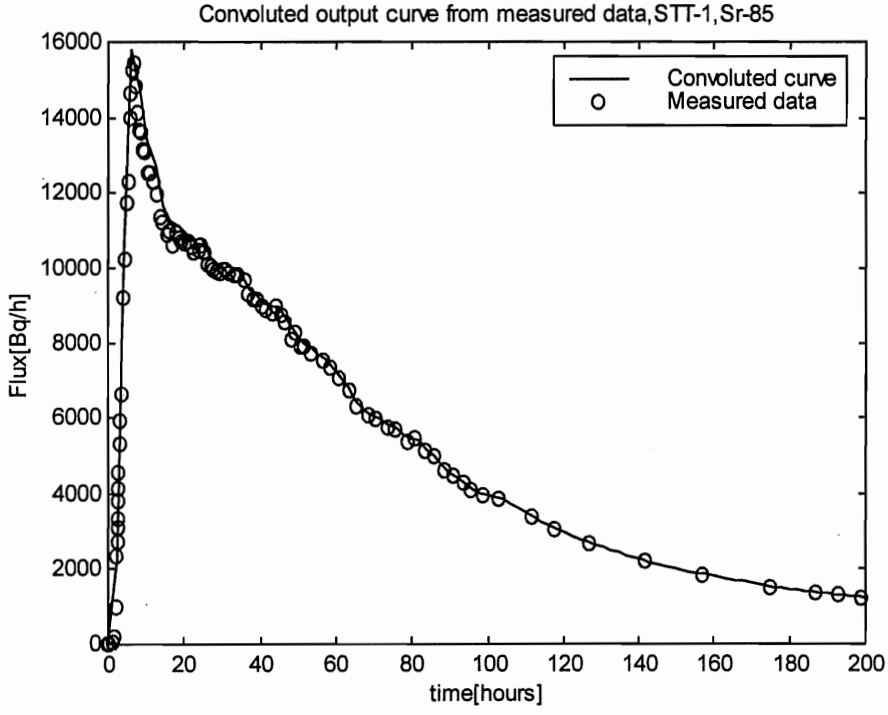

Figure 4-12 Convoluted unit response curve for Sr-85 in STT-1 compared with measured data.

The unit response curve of Sr-85 has a lower peak and a more pronounced tailing than Uranine, HTO and Na-22.

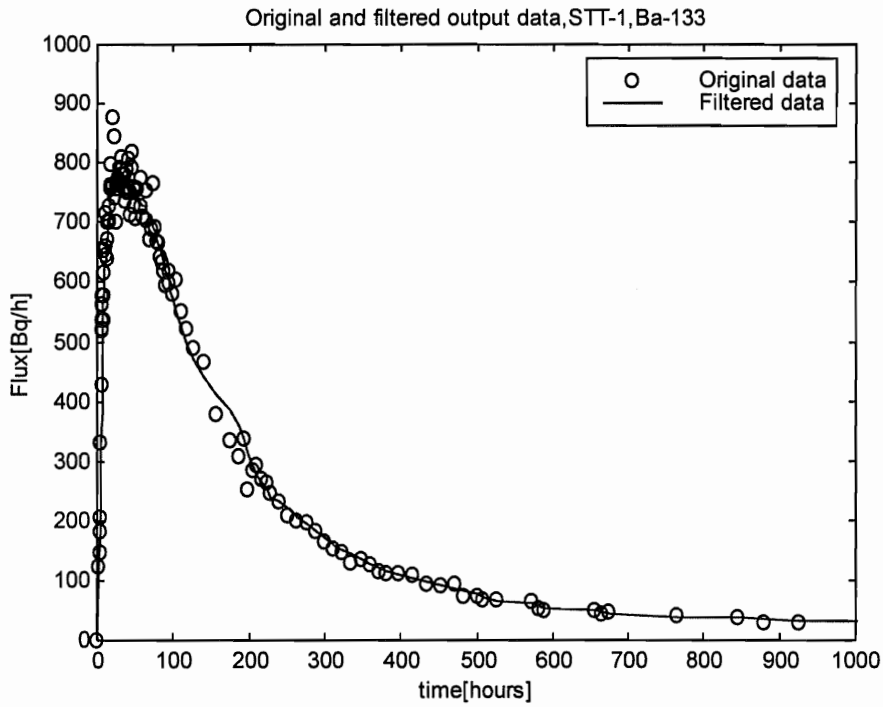

Figure 4-13 Original and filtered output data for Ba-133 in STT-1.

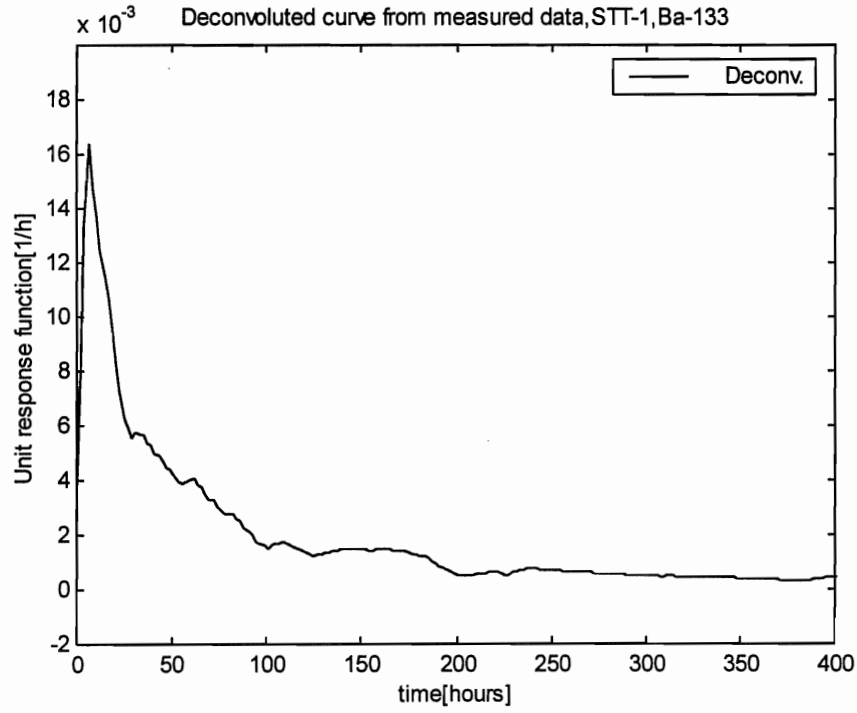

Figure 4-14 Unit response curve for Ba-133 in STT-1.

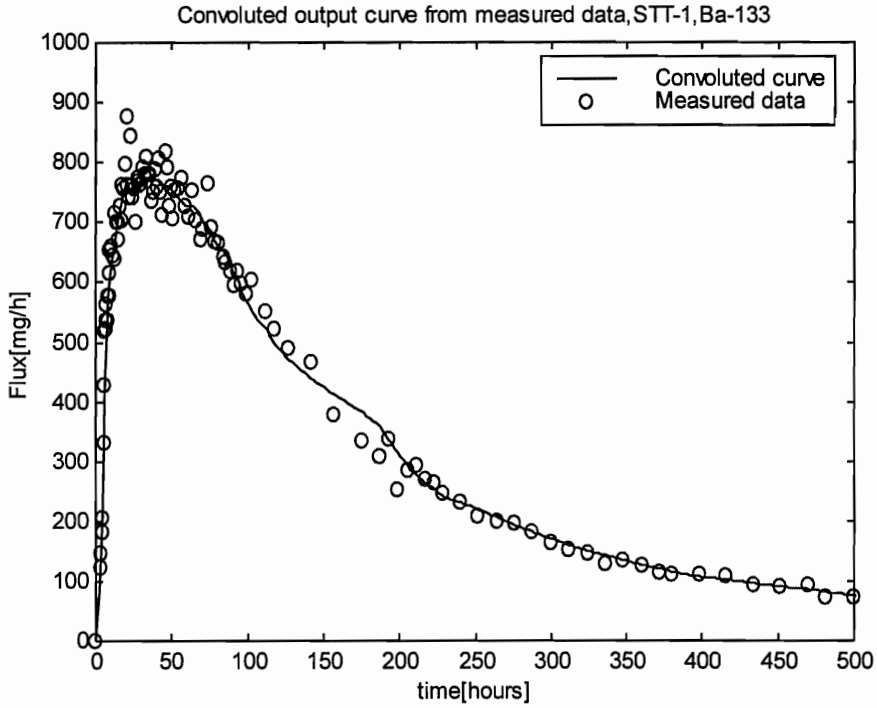

Figure 4-15 Convoluted unit response curve for Ba-133 in STT-1 compared with measured data.

For Ba-133 there was so much noise in the injection curves and breakthrough curves that the deconvolution could not be made with the original experimental data. For the injection curve a curve fitting was made where the injection curve was divided into 5 segments. The segments were fitted to a polynomial, logarithmic, exponential, polynomial and logarithmic function, respectively. The breakthrough curve was filtered according to the method described in Section 3.3. The resulting filtered curve and the original data are shown in Figure 4-13. The resulting unit response function (Figure 4-14) is delayed compared to the non-sorbing tracers, but has the same general shape - a sharp peak with tailing. The features of the curve appearing after 100 hours are most likely artefacts of the data treatment. The convoluted unit response function compares well with the measured data, see Figure 4-15.

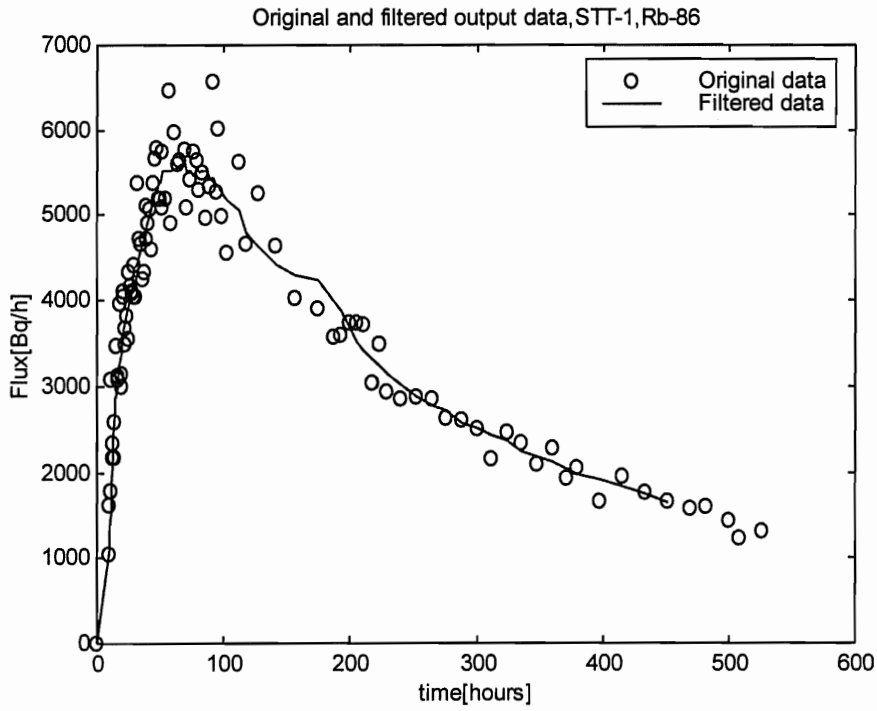

Figure 4-16 Original and filtered output data for Rb-86 in STT-1.

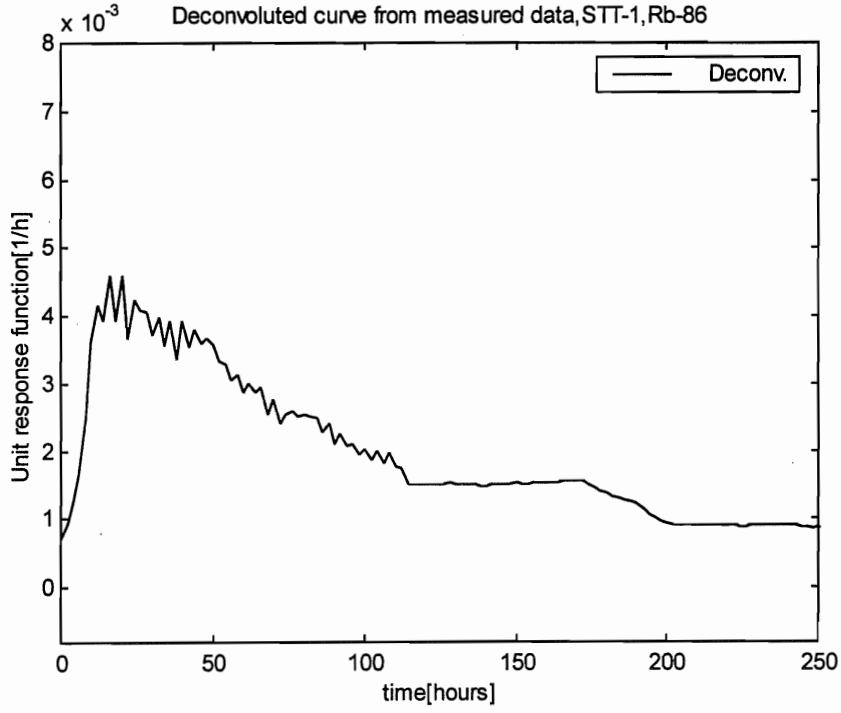

Figure 4-17 Unit response function for Rb-86 in STT-1.

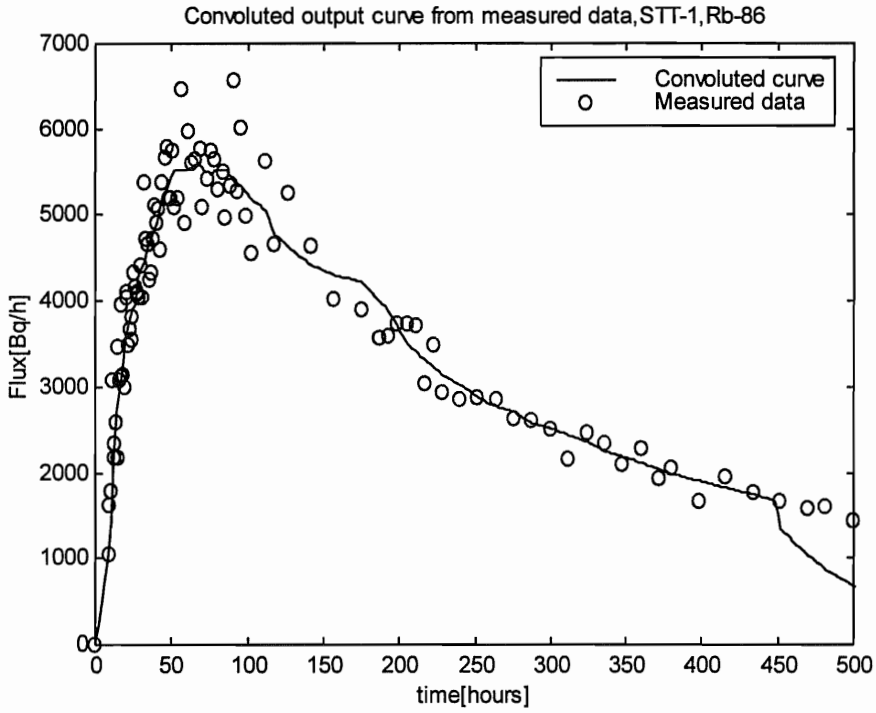

Figure 4-18 Convoluted unit response curve for Rb-86 in STT-1 compared with measured data.

The deconvolution of Rb-86 was not completely successful. As both the injection and breakthrough curves were spiky the curve fitting and filtering procedures were applied. The injection curve was divided into 5 segments, which were fitted to a linear, logarithmic, exponential, polynomial and logarithmic function, respectively. The filtered curve follows the measured data quite well, cf. Figure 4-16, except for a section between 150 and 200 hours with relatively few measurements. In this section there is a "bump", where it appears that the filtered curve is too high. The same "bump" also appears in the unit response curve obtained from the deconvolution Figure 4-17.

The deconvolution using the "backslash"-method was not very successful. Instead the pseudo-inverse method (pinv) was used. A filtered curve was used up to 450 hours, which causes the failure of the deconvolution after that time.

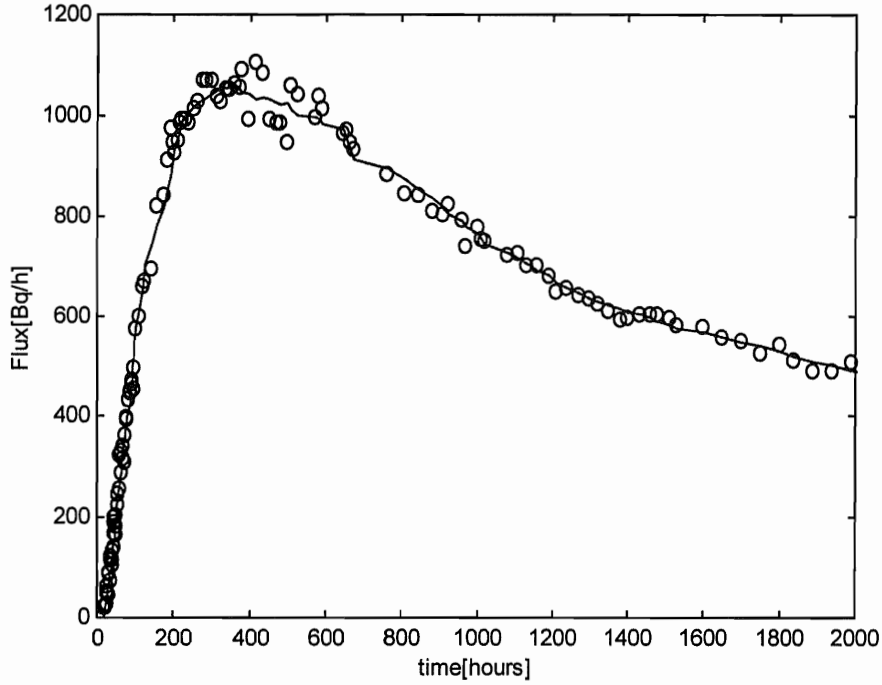

Figure 4-19 Original and filtered output data for Cs-137 in STT-1.

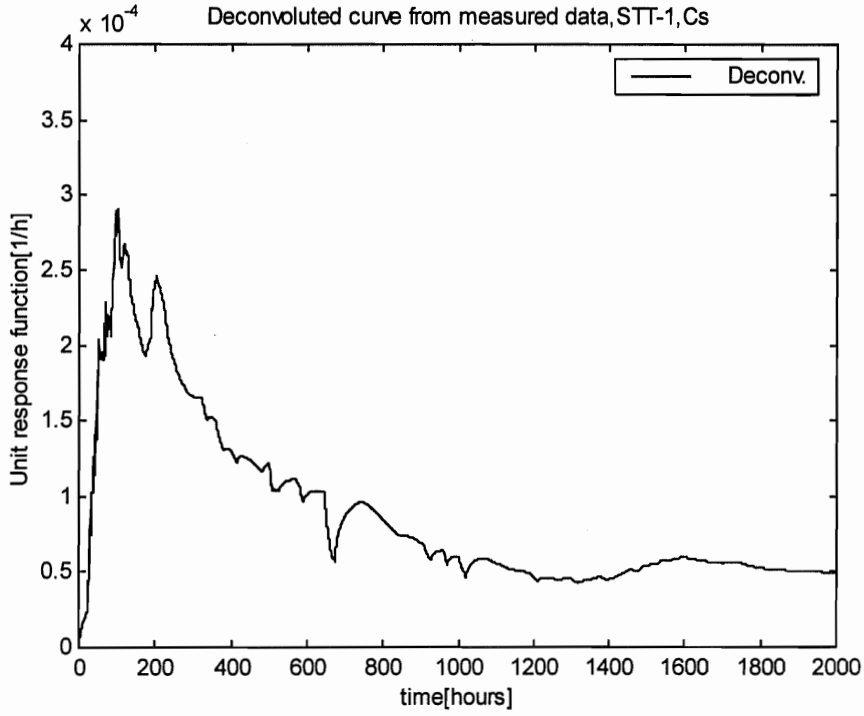

Figure 4-20 Unit response function for Cs-137 in STT-1.

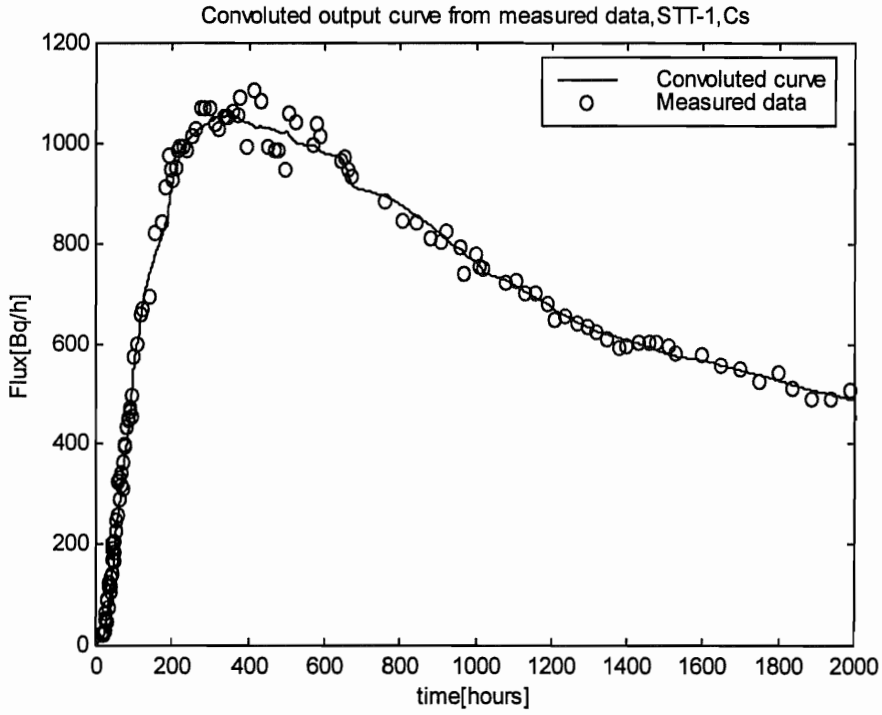

Figure 4-21 Convoluted unit response curve for Cs-137 in STT-1 compared with measured data.

The breakthrough curve for Cs-137 could not be deconvoluted using the ordinary matrix inversion methods, although filtering was applied. For Cs-137 the breakthrough curve was very long compared to the breakthrough curve. If the time step was chosen small enough to describe the details of the injection curve, too many points were obtained for the breakthrough curve. Instead the cgs-function (conjugated gradient squared method) was tried. The method was not completely successful and a full convergence to the desired tolerance could not be achieved. The unit response curve also shows marked artefacts (e.g. at 200, 650 and 1000 hours), probably caused by features in the smoothed breakthrough curve.

4.3 Deconvolution of STT-1b test

Deconvolution of the experimental results from the STT-1b test has been performed on the tracers: Uranine, HTO, Sr-85, Na-22 , Co-58, Rb-86 and K-42. However, for K-42 it was not possible to make a successful deconvolution since the breakthrough curve was too short to be able to obtain a successful deconvolution.

For Uranine, HTO, Na-22 and Sr-85 the curve fitting could be performed on the original experimental data. For Co-58 and Rb-86 the breakthrough curve was filtered in order to remove some of the spikes. The time steps used and the treatment of data performed for the different tracers are summarised in Table 4-2. A trial and error methodology was chosen to find the smallest time step that could be used without obtaining problems with oscillations.

Table 4-2 Time steps and methods used for deconvolution of the STT-1b test.

	Start time (h)	Time step (h)	Injection curve	Breakthrough curve	Inversion method
Uranine	0.185	1.8	experimental	experimental	nnls
HTO	0.14	2.	experimental	experimental	backslash
Na-22	0.065	2.04	experimental	experimental	backslash
Sr-85	0.06	2.04	experimental	experimental	backslash
Co-58	0.0001	2.5	experimental	smoothed	backslash
Rb-86	0.1	1.8	experimental	smoothed	pinv
K-42	Incomplete breakthrough curve				

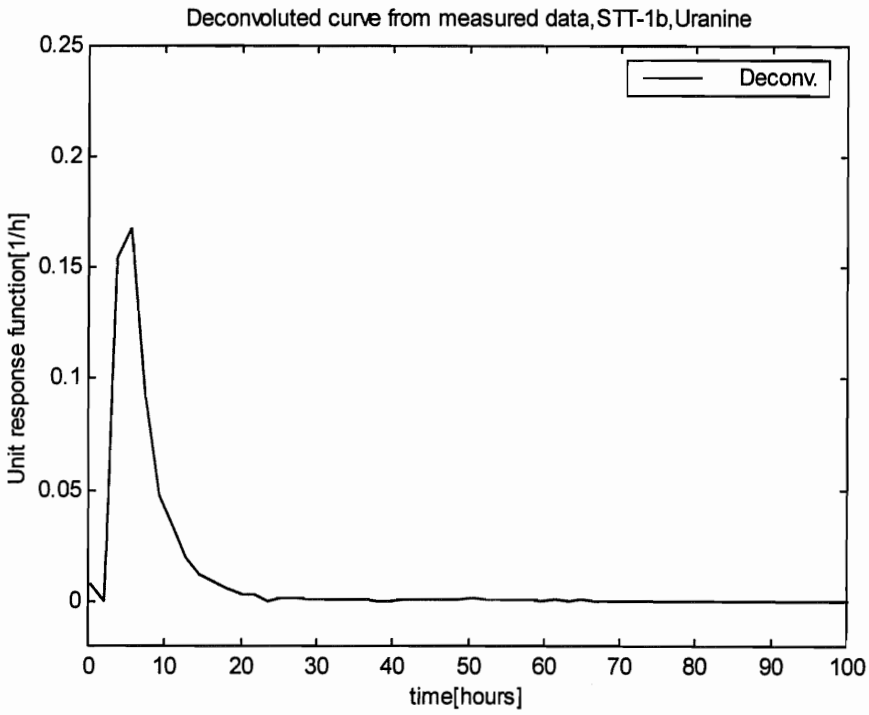

Figure 4-22 Unit response function for in Uranine STT-1b.

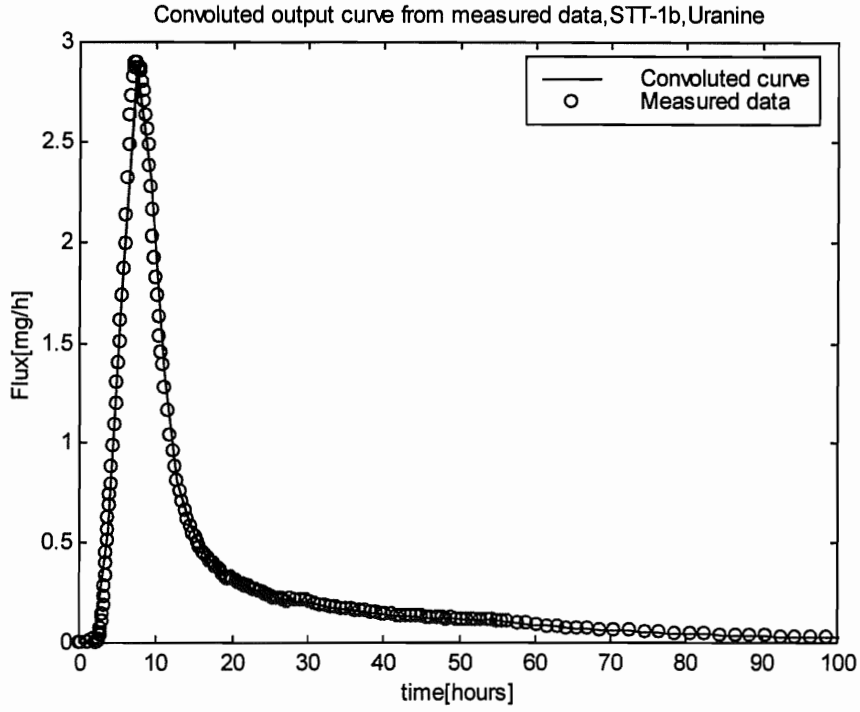

Figure 4-23 Convoluted curve for Uranine in STT-1b compared with measured data.

The nnls function was used to obtain the unit response curve for Uranine in order to avoid negative values. The unit release curve for Uranine in the STT-1b experiment comes somewhat later in time, has a higher peak and less pronounced tailing than that from the STT-1 experiment. The occurrence of a non-zero value at time zero is a mathematical artefact from the deconvolution.

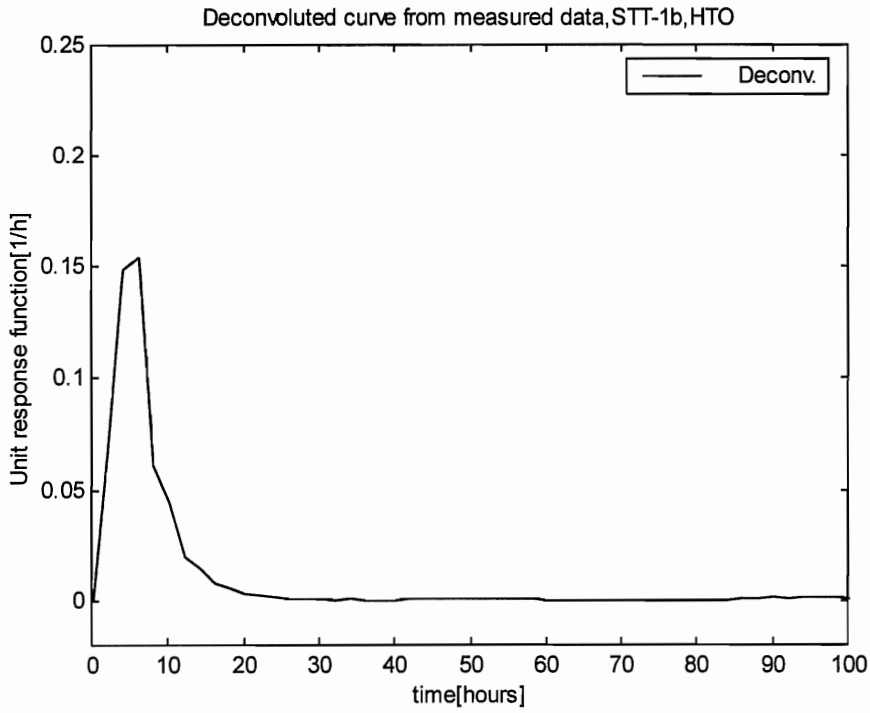

Figure 4-24 Unit response function for HTO in STT-1b.

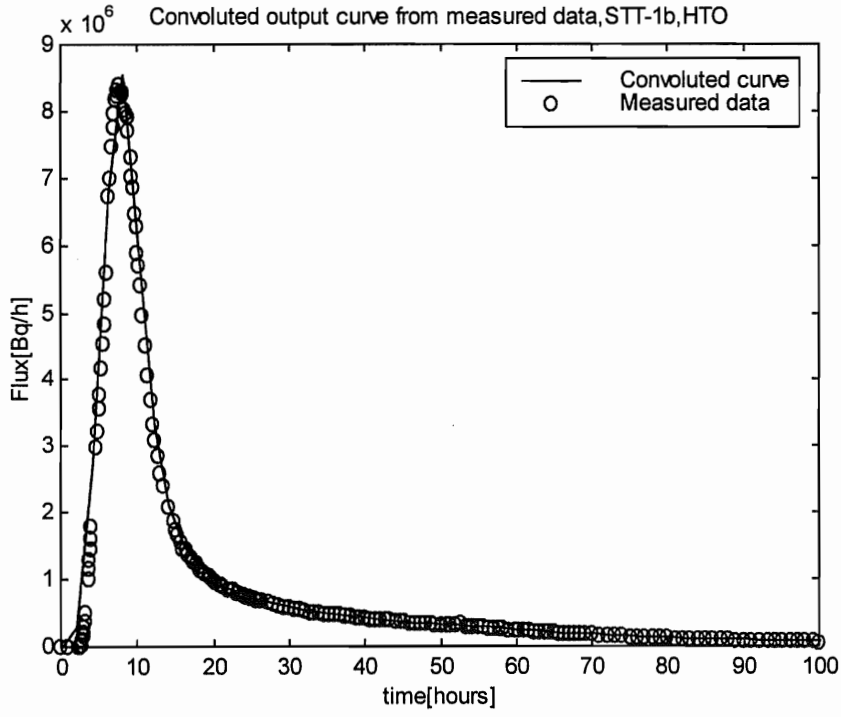

Figure 4-25 Convoluted curve for HTO in STT-1b compared with measured data.

The unit response curve for HTO in STT-1b is almost identical to that of Uranine, except for a different appearance of the top of the peak. This may be caused by differences in the time step.

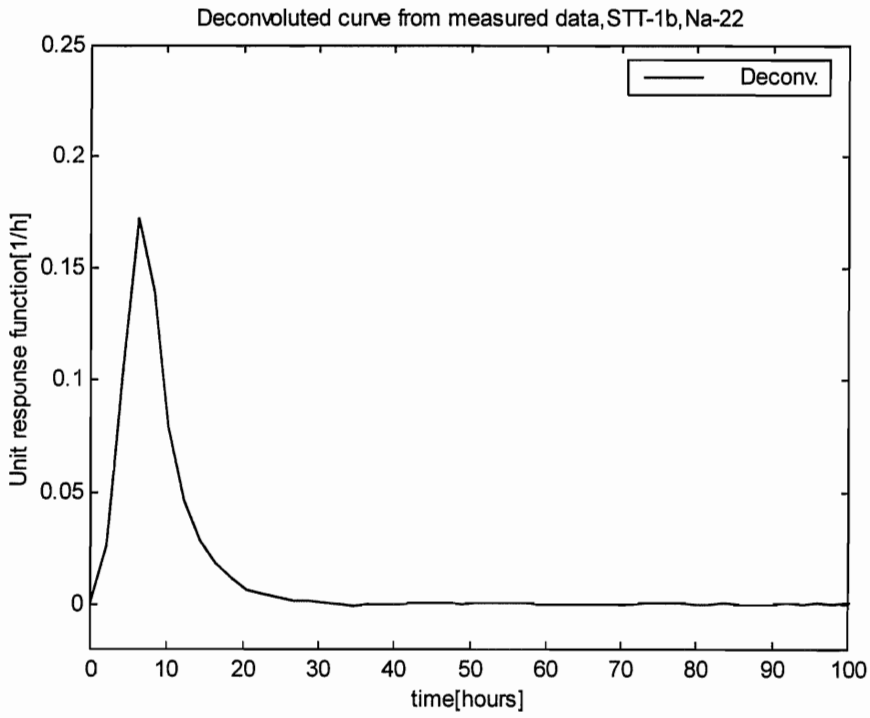

Figure 4-26 Unit response function for Na-22 in STT-1b.

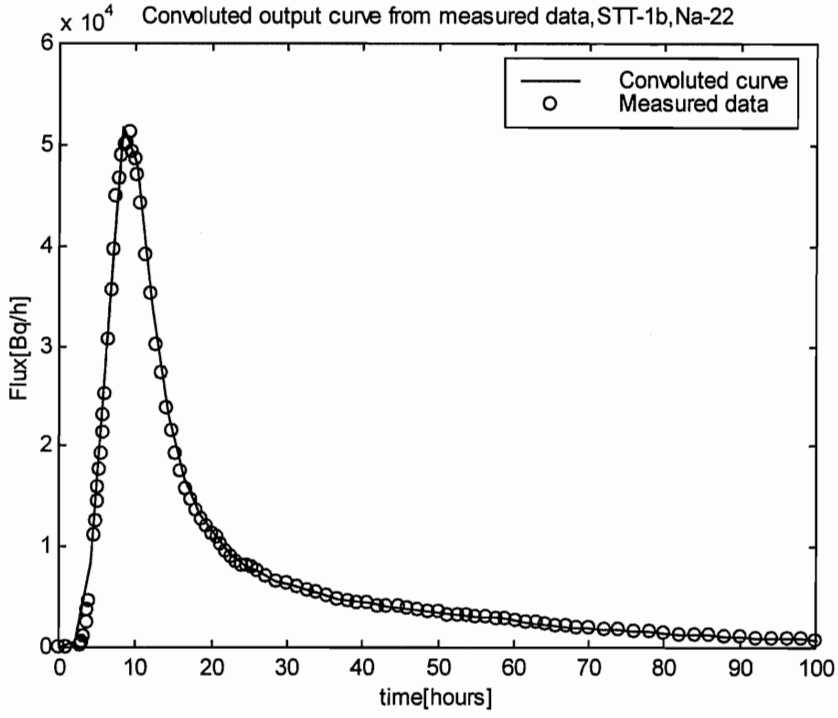

Figure 4-27 Convoluted curve for Na-22 in STT-1b compared with measured data.

The unit response curve of Na-22 has a similar shape to that of Uranine and HTO, but is to some extent delayed in time.

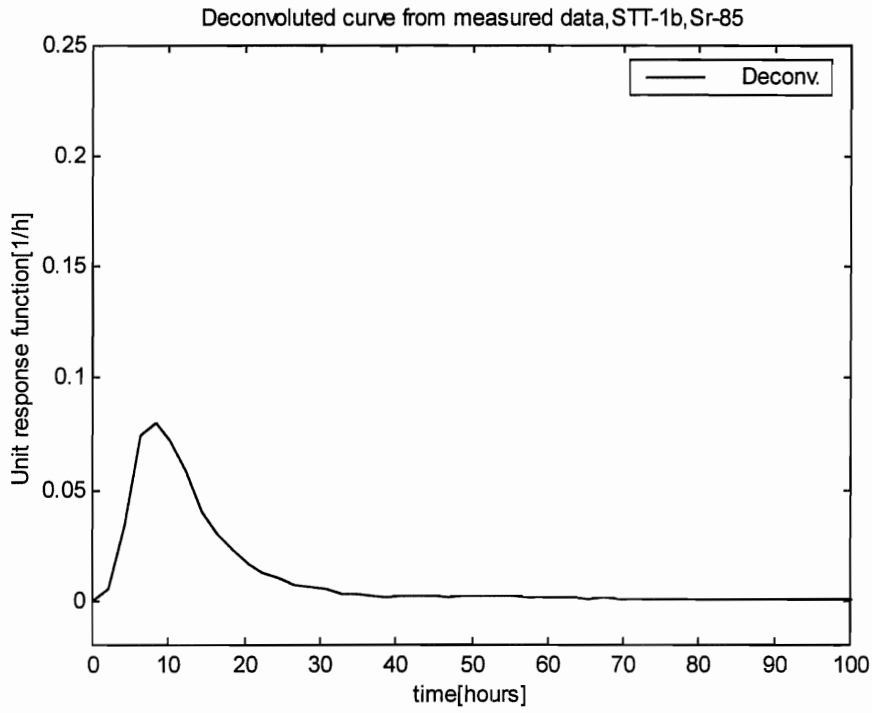

Figure 4-28 Unit response function for Sr-85 in STT-1b.

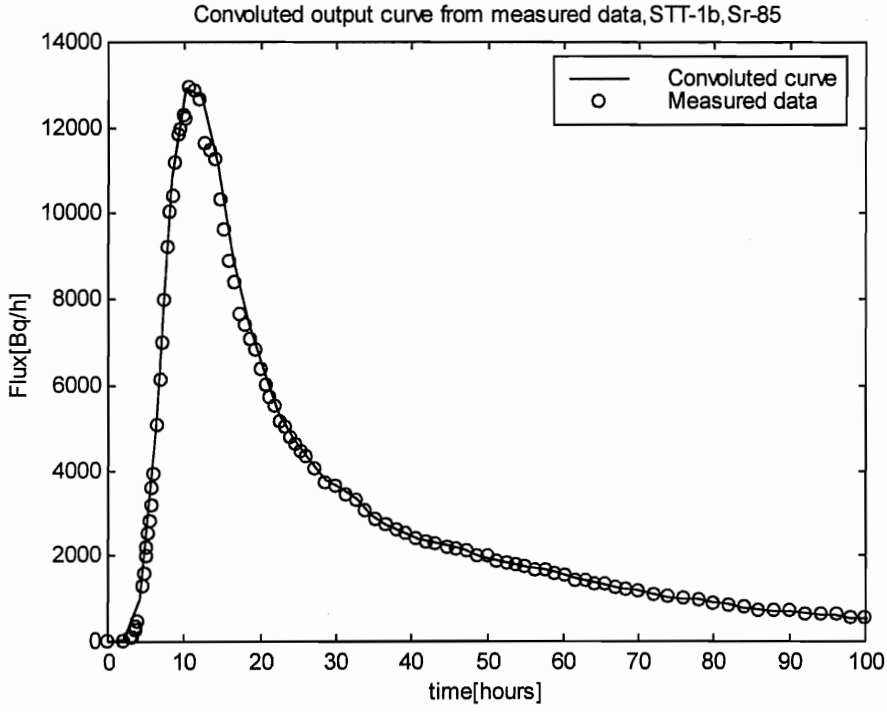

Figure 4-29 Convoluted curve for Sr-85 in STT-1b compared with measured data.

The unit response curve of Sr-85 even more delayed than that of Na-22 and the peak is considerably lower.

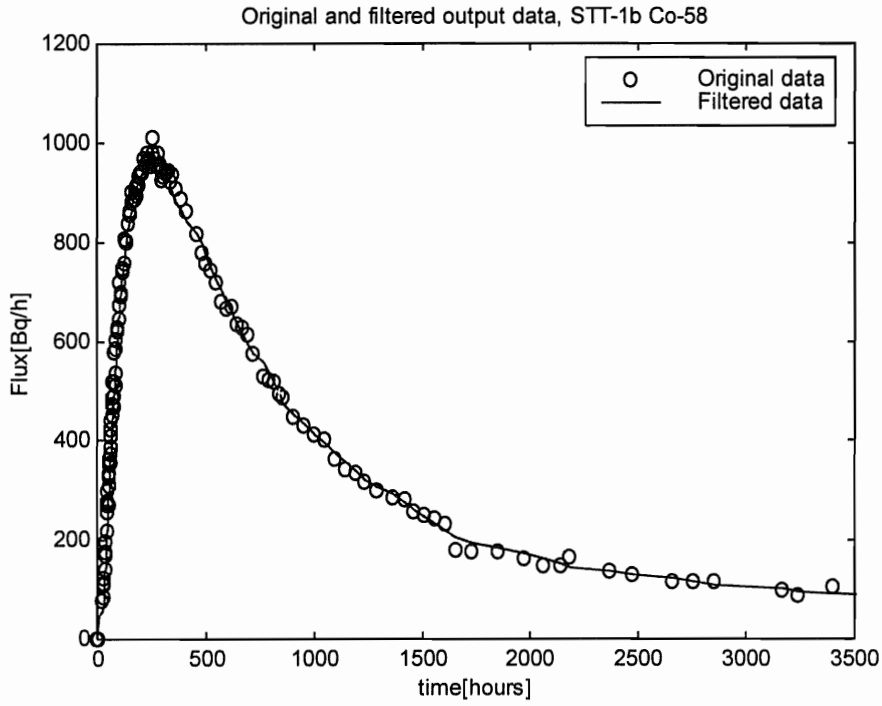

Figure 4-30 Original and filtered output data for Co-58 in STT-1b.

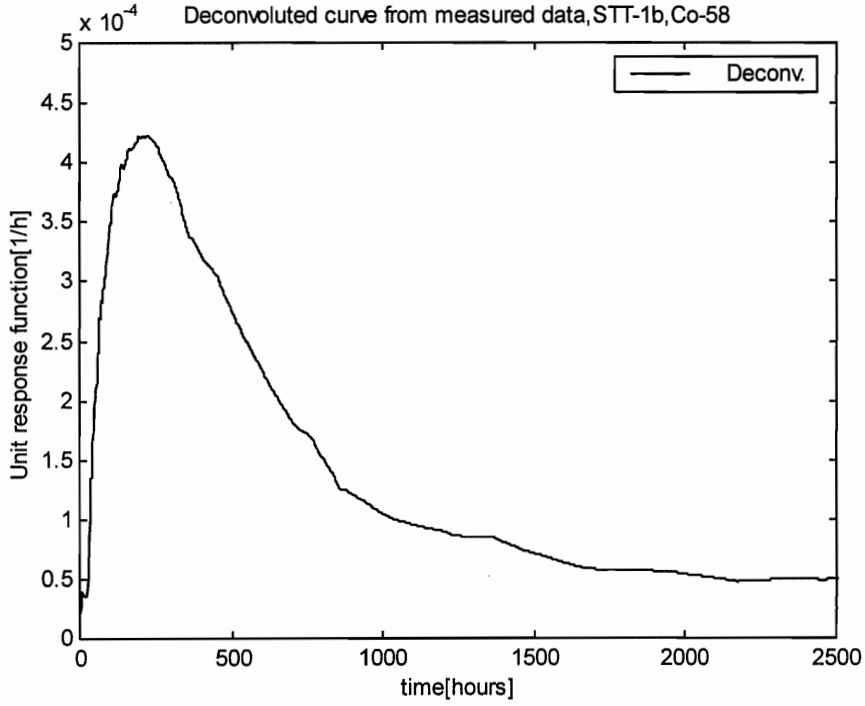

Figure 4-31 Unit response function for Co-58 in STT-1b.

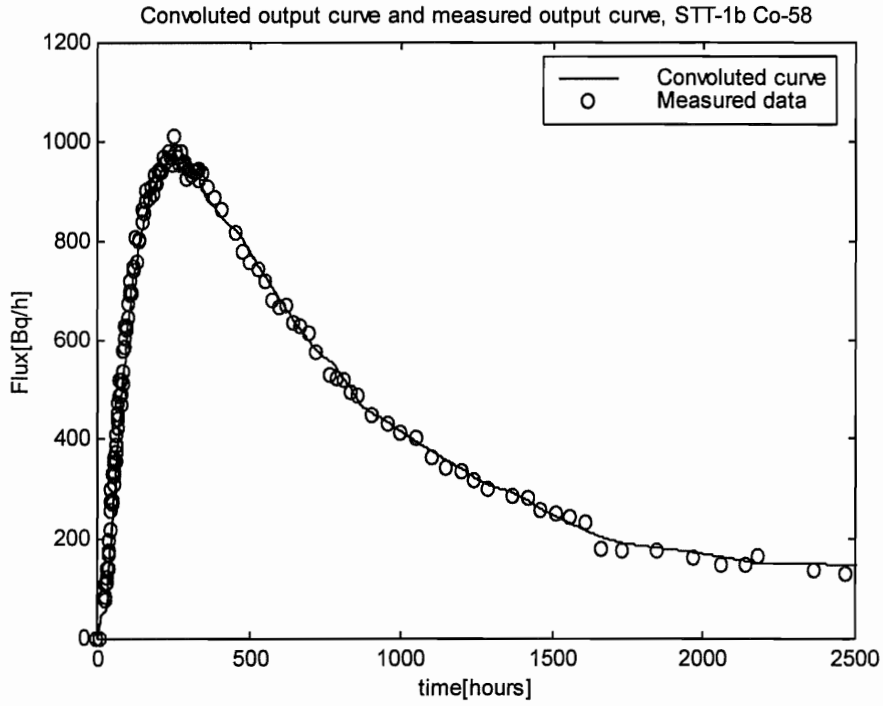

Figure 4-32 Convoluted curve for Co-58 in STT-1b compared with measured data

The breakthrough curve of Co-58 was filtered before deconvolution. The unit response curve shows considerable tailing. The curve decreases initially rapidly after the peak, but the decrease seems to level out after about 900 hours.

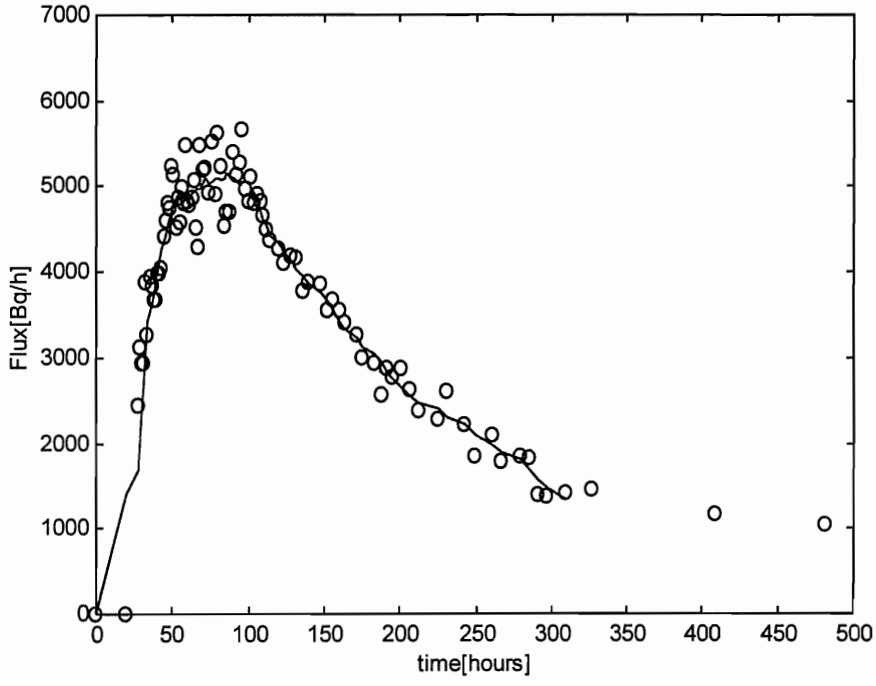

Figure 4-33 Original and filtered output data for Rb-86 in STT-1b.

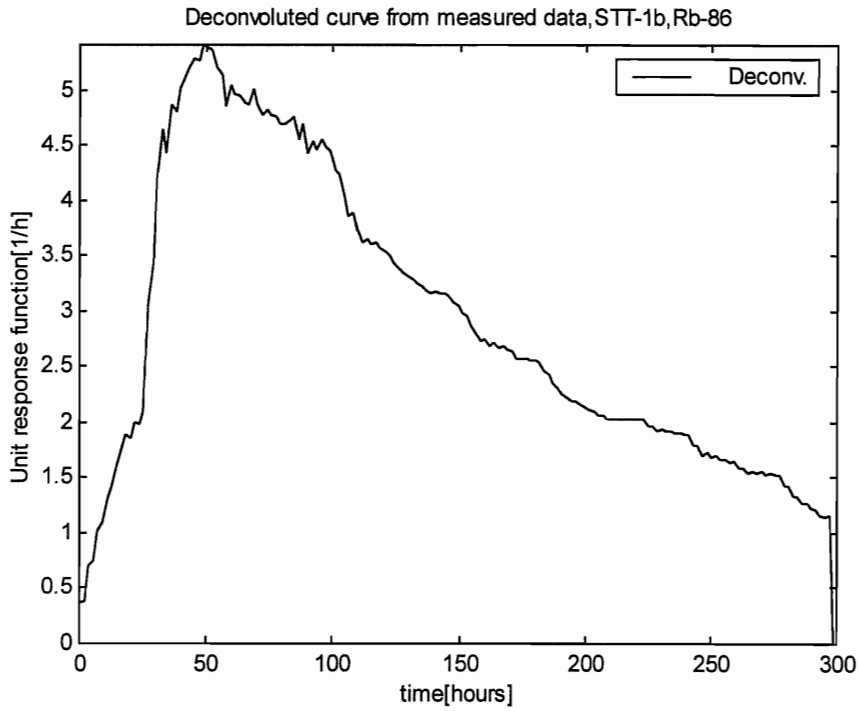

Figure 4-34 Unit response function for Rb-86 in STT-1b.

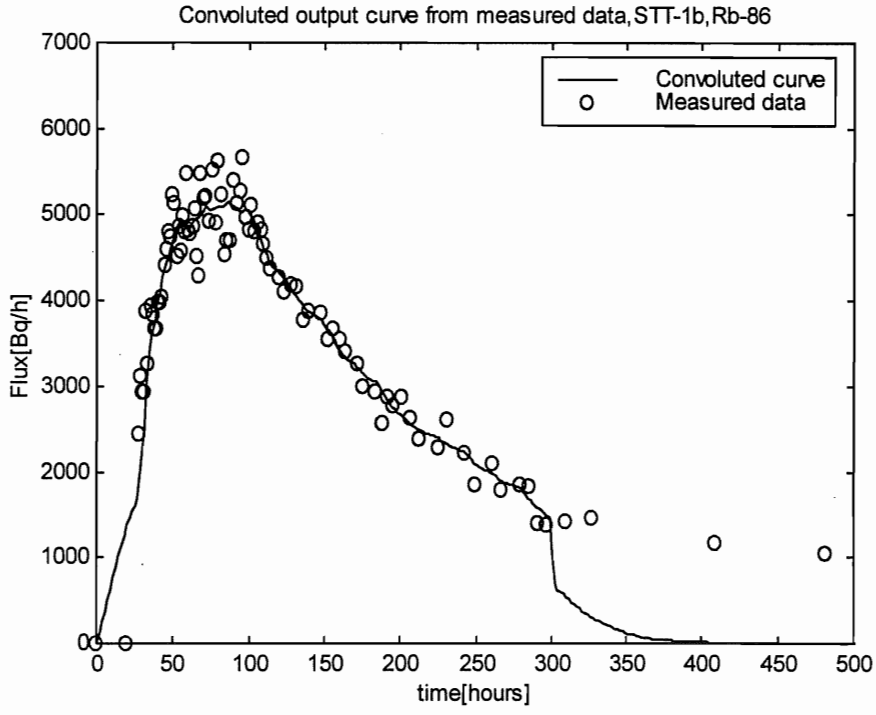

Figure 4-35 Convoluted curve for Rb-86 in STT-1b compared with measured data.

As was the case for STT-1, the deconvolution required treatment of the breakthrough curve and the use of the pinv-function. In this case the deconvolution broke down after 300 hours. The first part of the unit response curve shows a increase from the time zero. This is due to the filtering of the breakthrough curve.

4.4 Comparison of results

In Figure 4-36 the unit response curves of all tracers from STT-1 have been compiled in one figure in order to facilitate the comparison between the different tracers. The compiled results for the tracers used in STT-1b are shown in Figure 4-37.

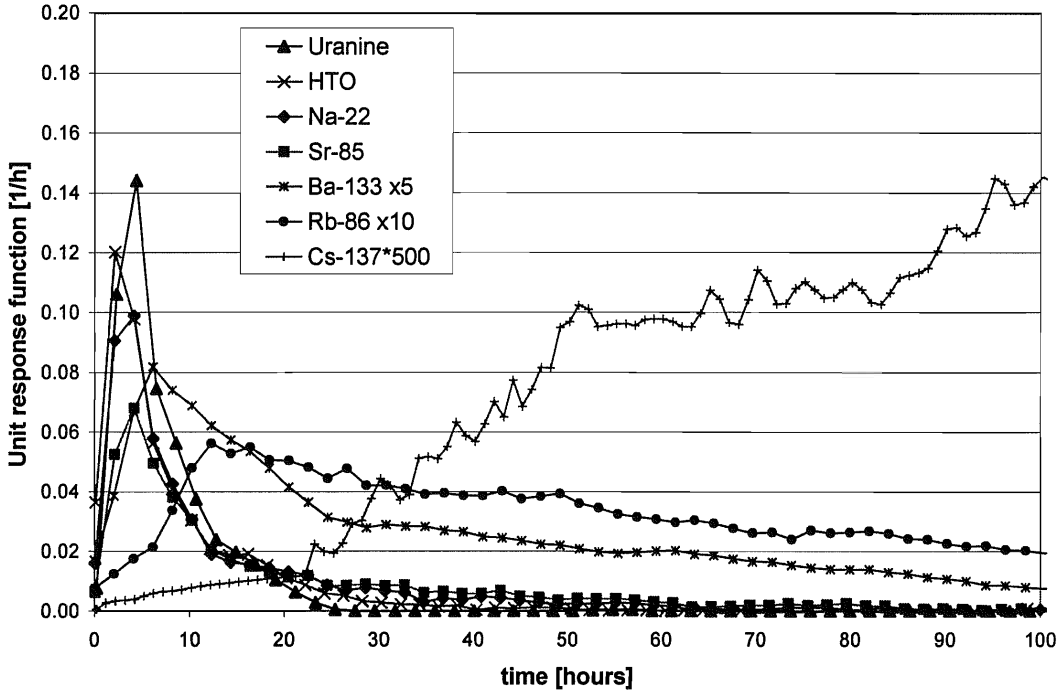

Figure 4-36 Unit response curves for tracers in STT-1.

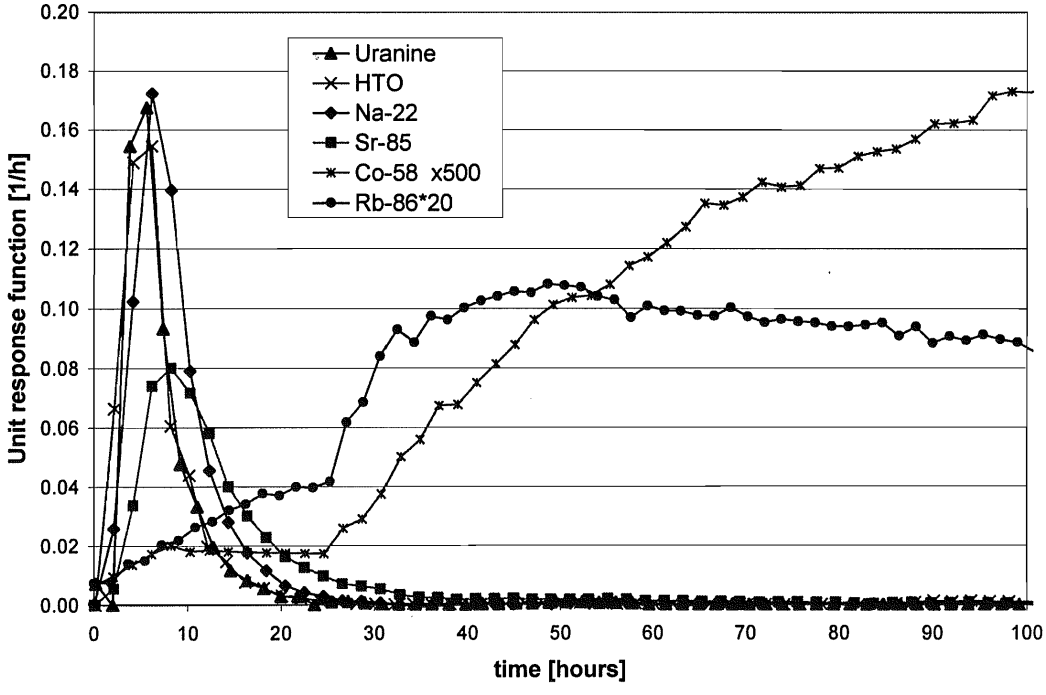

Figure 4-37 Unit response curves for tracers in STT-1b.

5 Discussion and conclusions

5.1 Deconvolution approach

The use of deconvolution to obtain unit response functions for tracers can be very helpful in evaluating tracer experiments, since the unit response functions only contain features which are due to transport processes and not affected by the shape of the injection curve. Thus, the effects such as dispersion and mass transfer may be visualised even with long injections.

Unit response curves can be helpful in a comparison between model predictions and experimental curves. In comparisons of breakthrough curves for tests with long injection periods it may be difficult to distinguish the features that are introduced by the shape of the injection curve from those due to transport processes. Many of the mathematical models used within Task 4E derive unit response functions as a part of the transport calculations, or may give such functions as output if desired. A comparison between unit response functions predicted by models and those derived from the experimental data will give information on how well the models describe the actual transport processes.

The disadvantage is that the procedure involves a mathematical manipulation of the data, and since deconvolution is an ill-posed mathematical problem this may give rise to numerical problems. It may therefore be difficult to distinguish between physically relevant features from mathematical peculiarities. In the method used for the present study most problems were found for sorbing tracers where the length of the breakthrough curves is considerably larger than that of the injection curve.

The injection method has been significantly developed during the TRUE-1 tracer tests. The initial radially converging tests had a very long injection curve. For the dipole tests the injection of water in the source section lead to a faster dilution and considerably shorter injection curve. The injection procedure used for the sorbing tracer tests within TRUE-1, were the injection section is flushed with unlabeled water after a few hours injection, has lead to distinct peaks with a duration of only a few hours. Thus, the shape of the injection curve has a pronounced effect on the breakthrough curves of the non-sorbing or very weakly sorbing tracers. In STT-1 and STT-1b tests these tracers had a t_5 of around 5 hours and a t_{50} of about 40 hours. However, also for the sorbing radionuclides (e.g. Co-58 in STT-1b) it can be seen that the peak of the unit response curve is less spread out than the peak of the breakthrough curve.

Unit response curves obtained by deconvolution can also be used to compare tracer tests performed within the same geometry, but with a different source term.

5.2 Deconvolution methods

The deconvolution method used in this study is relatively simple. More sophisticated methods have been developed, for example the Extreme Value Estimation (EVE). However, the majority of the experimental curves were of very good quality, with only few or no outlying values. Thus, successful deconvolution could be made of most of the tracer tests. However, the present method may be developed to improve the obtained unit response functions and to ensure a successful deconvolution also of the remaining tracer tests:

- The use of discrete time steps equal for both the injection curve and the breakthrough curve causes problems for sorbing tracers (e.g. Cs-137) where the length of these curves may be very different. A small time step is needed in order to have a good description of the injection curve, but this leads in some cases to a enormous matrices that needs to be inverted. The conjugated gradient square method used in this report produced reasonable results, but the methods need further development.

- In the case of non-sorbing radionuclides a much shorter time step than proved to be numerically stable would be needed to determine the detailed characteristics of the peaks. Improvement of the stability of the method or the use of more stable methods is therefore desirable.

- The method of filtering spiky output data needs to be developed.

- The optimisation of the deconvolution is to a large degree performed manually, which may be very time consuming. It would be a great advantage to have a more automated method.

5.3 Conclusions

Deconvolution of experimental breakthrough curves using the injection curve in order to obtain a unit response function is a useful approach to evaluate tracer experiments in order to identify features in the breakthrough curves caused by transport processes and not caused by the shape of the injection curve. In particular they can be used for comparison with unit response curves obtained from model predictions.

The method presently used for deconvolution has successfully deconvoluted most of the tracers used in the STT-1 and STT-1b tests. However, there is a need for further improvement of the method in order to handle curves with oscillations or large experimental errors.

A possible way of achieving this is to put further restrictions on the unit response functions derived from the deconvolution. The physical models for transport along a single flow path (e.g. advection-dispersion models, matrix diffusion models) give rise to unit response functions of a certain form. A possibility would be to use the knowledge about these forms to put restrictions on the impulse response functions. Furthermore, the methods used for deconvolution in other disciplines needs to be investigated further.

References

Andersson P, Johansson H, Nordqvist R, Skarnemark G, Skålberg M, Wass E, 1998. TRUE 1st stage tracer test programme, Tracer tests with sorbing tracers, STT-1, Experimental description and preliminary evaluation. Äspö Hard Rock Laboratory Technical Note TN-98-10t, Swedish Nuclear Fuel and Waste Management Co.

Andersson P, Johansson H, Skarnemark G, Skålberg M, Wass E, 1999. TRUE 1st stage tracer test programme, Tracer tests with sorbing tracers, STT-1, Experimental description and preliminary evaluation. Swedish Nuclear Fuel and Waste Management Co.

Ilvonen M, Hautojärvi A, Paatero P, 1994. Intraval project phase 2, Analysis of Stripa 3D data by a deconvolution technique. Nuclear Waste Commission of Finnish Power Companies, YJT-94-14.

Skaggs T H., Kabala Z J ,Jury W A, 1998. Deconvolution of a nonparametric transfer function for solute transport in soils, Journal of Hydrology, 207, 170-178.

Tsang C F, Tsang Y W, Hale F V, 1991. Tracer transport in fractures: Analysis of field data based on a variable-aperture channel model, Water Resources Research, 27, 3095-3106.