Benchmark Calculations with COMSOL of the Transport of Radionuclides through Clay and Bentonite Barriers in a Geological Repository

Manuel Lorenzo Sentís[*1], Felix Altorfer[1], Martin Herfort[1], Andreas Jakob[2], Georg Kosakowski[2], Sven Friedel[3]

[1]Swiss Federal Nuclear Safety Inspectorate (ENSI)
[2]Paul Scherrer Institute (PSI)
[3]COMSOL Multiphysics GmbH
*Corresponding author: ENSI, Disposal and Safety Analyses Section (TISA), CH-5232 Villigen-ENSI, Switzerland, manuel.sentis@ensi.ch

Abstract:

A benchmark study was carried out by the Swiss Nuclear Safety Inspectorate (ENSI) in collaboration with the Laboratory for Waste Management (LES) of the Paul Scherrer Institute (PSI) in order to evaluate the capabilities of the program COMSOL for the calculation of the transport of radionuclides through clay and bentonite barriers in a geological repository for radioactive waste. In this study results of four different codes were compared. The results of the transport calculations for a selection of key radionuclides are shown in this paper. The good agreement of the results obtained with the different codes is a verification of COMSOL as modeling tool for a geological repository of radioactive waste in a clay formation.

Keywords: Geological repository, radioactive waste, safety assessment, benchmark, porous medium.

1. Introduction

The Swiss Nuclear Safety Inspectorate ENSI is the regulatory authority for nuclear safety and radiation protection in nuclear facilities in Switzerland. It also supervises the preparations for the disposal of radioactive waste, which includes the review of safety assessments for geological repositories for all types of radioactive waste submitted by the Swiss implementer Nagra.

The sectoral plan for the selection of appropriate sites for geological repositories for all kind of nuclear waste (spent fuel, vitrified high-level and long-lived intermediate-level waste as well as for low-level waste) in Switzerland started in 2008 and will last about ten years. ENSI examines and evaluates the proposals put forward by the implementer Nagra from the point of view of safety. In order to perform independent safety analysis calculations and thus to review the safety assessments of the implementer Nagra, ENSI decided to use the program COMSOL [1,2] in particular the "Earth Science Module". To gain confidence in the quality of the results of the safety analysis, especially since the program must provide results for very long timescales, it is general practice to perform a verification of the code. This is done typically by performing benchmark analyses in which the results of different codes are compared to each other. With this objective a benchmark study was undertaken in collaboration with PSI, in which the results of four different codes were compared, namely PICNIC, FRAC3DVS, COMSOL (Version 3.4) and Tough2-EOS9nT [3,4]. PICNIC calculations were published by Nagra [5] for a conceptual model of a geological repository within a feasibility study (project "Opalinus Clay"). The results were reproduced by PSI [6] with help of FRAC3DVS and an independently developed COMSOL model as well as by ENSI with COMSOL and Tough2-EOS9nT [7,8].

For the feasibility study a potential site located in the geological formation Opalinus Clay in northern Switzerland was selected by Nagra.

2. Conceptual Model

To estimate the release of radionuclides to the biosphere, the complex system of caverns and tunnels was simplified into a one-dimensional (PICNIC) or into a two-dimensional (COMSOL, FRAC3DVS, Tough2-EOS9nT) conceptual model as shown in Figure 1. As a first simplification the horizontal distances between the can-

isters in a tunnel are conservatively neglected. Although the geological repository consists of a series of tunnels, based on symmetry arguments of the geometrical configuration it is only necessary to consider one tunnel in the study. The interactions between neighboring tunnels are considered by setting no-flow conditions at the two vertical boundaries. The vertical dimensions are defined based on the location of the nearest aquifer. After the above-mentioned considerations the conceptual model for the benchmark study is simplified as shown in the Figure 1 [6,9].
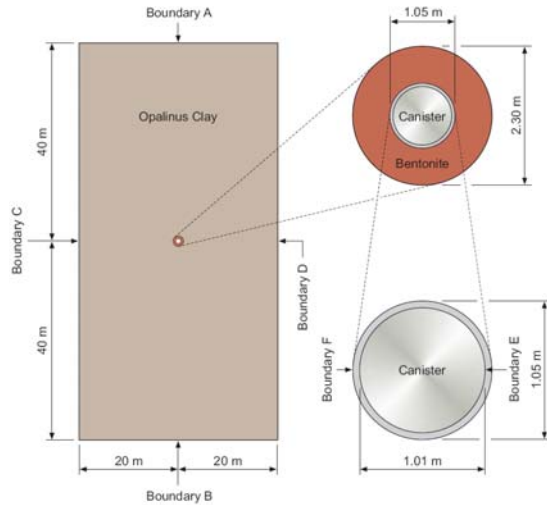
Figure 1. Conceptual model for the simulations [**6**].

The radionuclides are confined in the canister until the corrosion of the steel canister allows them to be released into the bentonite backfill. The thickness of the canister wall is about 1 cm. Based on a steel corrosion rate of 1μm/a it is assumed in the calculations performed in this paper that the radionuclides start being released from the canister after 10000 years. Afterwards the radionuclides are transported through the low-permeable bentonite and clay barriers based on advective and mainly diffusive processes. Due to the small values of the Darcy velocities, the dispersive transport of radionuclides was not considered in the calculations. The bentonite backfill and the Opalinus Clay, assumed to be completely saturated, were modeled as saturated homogeneous porous media without fractures. The sorption coefficients of the radionuclides in the clay and bentonite layers are considered,

solubility limits were conservatively neglected in these calculations.

The purpose of this paper is to show the numerical estimation by different codes of the radionuclide mass flow on the top and bottom boundary of the model. This is obtained by integrating the mass flux over the top and bottom boundaries. These mass flows are used within the biosphere model to obtain the effective dosis to the population.

3. Mathematical Model

To study the transport of different nuclides through the different barriers of the geological repository the program COMSOL Multiphysics and in particular the "Earth Science Module" was used. The study concentrated on some critical radionuclides that show only small retention in the different barriers of the geological repository: I-129, Ca-41, Cl-36, Se-79 and C-14. The Model is based on the following three equations [1]:

Mass conservation of water

$$\frac{\partial(\phi s_f \rho_f)}{\partial t} + \nabla(\phi s_f \rho_f \vec{v}) = Q_f \qquad (1)$$

with ρ_f [kg m^{-3}] for the density of water, $\vec{v}$ [m/s] the average velocity of the flow in the porous medium, s_f [-] the saturation of the medium, ϕ [-] the porosity of the medium and Q_f [kg m^{-3}s^{-1}] a source term for the flow of water.

Darcy's law for the mass flux of water

$$\vec{j} = \rho_f \phi \vec{v} = -\rho_f K \phi \nabla \varphi \qquad (2)$$

with K [m/s] for the hydraulic conductivity and φ [m] for the hydraulic potential.

The equation (1) is simplified with the elimination of the time derivative (stationary flow) and of the source term for water. Furthermore the medium considered in this report is fully saturated and that means:

$$s_f = 1 \qquad (3)$$

Taking into account these considerations and equation (2), equation (1) becomes:

$$- \rho_f K \phi \Delta \varphi = 0 \qquad (4)$$

Mass conservation of the radionuclides or transport equation

$$\frac{\partial}{\partial t}(\phi c) + \frac{\partial}{\partial t}(\rho_t c_s) + \nabla \cdot [-\phi \overline{\overline{D}} \nabla c + \phi \vec{v} c] =$$

$$\sum R_f + \sum R_s + Q \qquad (5)$$

with ρ_t [kg m^{-3}] for the bulk density, c [kg m^{-3}] the concentration of the nuclide in the liquid phase and c_s [-] (the dimensions are mass of adsorbed contaminant per dry unit weight of solid) the concentration of the nuclide in the solid phase, $\overline{\overline{D}}$ [m^2/s] the hydrodynamic dispersion tensor, which is given by the following expression:

$$\overline{\overline{D}}_{ij} = (\alpha_L - \alpha_T)\frac{v_i v_j}{|\vec{v}|} + \alpha_T |\vec{v}| \delta_{ij} + D_p \qquad (6)$$

where α_L and α_T are the longitudinal and transverse dispersivity and D_p is the pore diffusion coefficient related to the molecular diffusion coefficient D_0 in water through a form factor f:

$$D_p = f D_0 \qquad (7)$$

α_L and α_T are considered equal to zero (dispersion term negligible in comparison to the diffusion term) for the calculations in this document and then the dispersion tensor reduces to D_p. R_f [kg m^{-3} s^{-1}] and R_s [kg m^{-3} s^{-1}] are the terms for the reactions (sum over the different reactions, for example chemical reactions, radioactive decay) of the nuclides in water and in the solid phase. For the calculations shown in this document only radioactive decay was considered and the two terms reduce to:

$$\sum R_f = \lambda c \qquad (8)$$

$$\sum R_s = \lambda c_s \qquad (9)$$

with λ [s^{-1}] for the decay constant of the considered radionuclide. For the calculations shown in this document c_s is related to c through a linear isotherm with a sorption coefficient K_d [m^3/kg]:

$$c_s = K_d c \qquad (10)$$

The last term Q [kg m^{-3}s^{-1}] in equation (5) represents the solute added per unit volume of soil and per unit time. The release of the nuclides from the canister is implemented in COMSOL as a boundary condition at the canister boundary. Therefore the term Q is neglected in (5). After all these considerations the equation (5) becomes:

$$[\phi + \rho_g K_s]\frac{\partial c}{\partial t} + \nabla \cdot [-\phi D_p \nabla c + \vec{u} c] =$$

$$[\phi + \rho_g K_s]c\lambda \qquad (11)$$

The Darcy velocity u is defined as the product:

$$u = \phi \cdot v \qquad (12)$$

and it is often referred to as specific discharge.

The hydraulic boundary conditions are fixed potentials of 0 m at the top and 80 m at the bottom (boundaries A and B in Figure 1), that corresponds to a hydraulic gradient of 1 m/m. On the left and right part of the boundary (boundaries C and D in Figure 1) a no-flow boundary for both, flow and transport was defined. As consequence of the corrosion of the waste container the nuclides are released from the boundary E. The volume between E and F was defined in order to implement a source term in the codeTough2-EOS9nT. In COMSOL this bentonite volume does not play an active role since the release rates from the canister are implemented as boundary conditions. The values of the release rate originally in a tabular format are implemented into COMSOL by means of an interpolation function. As initial condition for the hydraulic potential a linear variation from the bottom to the top of the domain was set (upward flow). The initial pressure in the bentonite filling is $0.14 \cdot 10^6$ Pa.

The flow and the transport model are symmetric with respect to a vertical line passing through the centre of the canister and therefore only the right part was considered in the calculations. This reduces the time needed for the solution. However, the computational time was not a critical aspect in the simulations because this time was of the order of some minutes for each nuclide with COMSOL as well as with Tough2-EOS9nT. The values of all parameters mentioned in this document are listed in table 1 in the appendix.

4. Results

A two-step approach with COMSOL version 3.4 was used to solve the above-mentioned equations. In a first step the stationary water flow equation (4) (Darcy's law model) is solved and the velocity obtained is used as input for the second step, in which the transport equation (11) is solved (solute transport model). Integration coupling variables were defined on the top and bottom part of the domain in order to integrate the flux of each nuclide over these boundaries.

The results of the simulations are shown in the figures 2 - 6 and were represented with the help of *Tecplot* [10].They represent the sum of the integrated mass flux over the top and bottom boundaries. The red points represent the values of the source term used in the calculations. The maxima of the mass flow values and the times of these maxima are summarized in Table 2 for the different codes and for the different radionuclides. The results for Ca-41 are shown in Figure 2. Two intervals can be distinguished, the one before the peak and the one after the peak. In the interval before the peak Tough2-EOS9nT and Frac3dvs limits the maximal and minimal mass flows respectively.
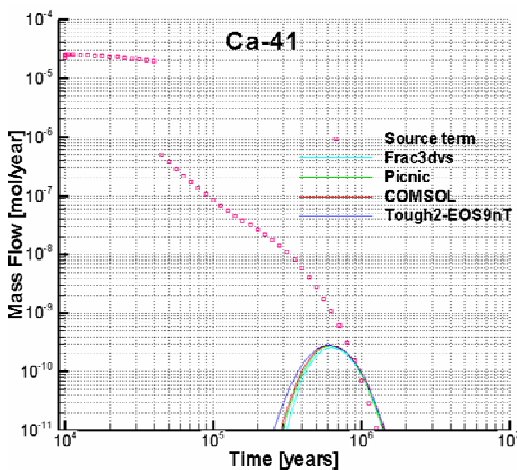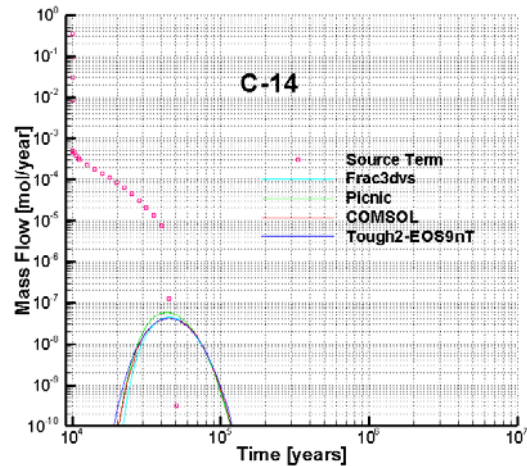

Figure 2. Geosphere release rates for Ca-41.

The different approach to the interpolation of the source term by the different codes and also the fast decrease of the source term at about 40000 years explain the differences in the rising edge of the mass flow for the different codes. The differences in the first part of the breakthrough curve reflect also the one-dimensional approach of Picnic in comparison with the other codes and the different time steps of the simulations. After the peak the agreement among the results for the different codes is excellent.

Figure 3 shows the results for the non-sorbing radionuclide C-14 (organic).
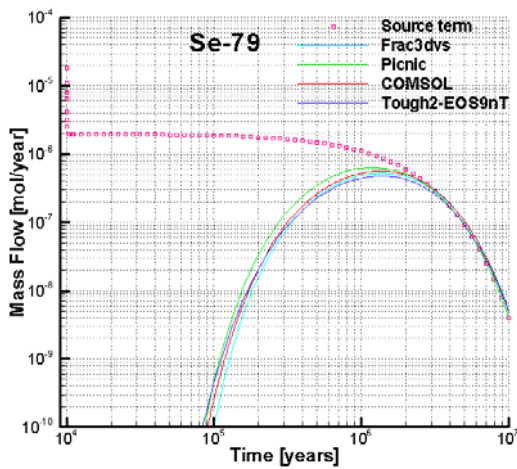

Figure 3. Geosphere release rates for C-14.

Ca-41 and C-14 have the same values for the diffusion constant and for the effective porosity. The differences are the data of the source term, which are about 10000 times larger for C-14; the half life, which is about 100 times longer for Ca-41; and the sorption coefficient K_d, which is zero for C-14. Due to zero sorption of C-14, the peak for C-14 is reached earlier than the one for Ca-41 and the peak concentration is about 100 times higher for C-14 than for Ca-41. At a first glance a peak concentration for C-14 10000 times higher would have been expected but the half life, the sorption coefficient and the faster decline of the source term for C-14 compensate the larger values of the source term.

A higher peak value is observed for C-14 from the calculations performed with Picnic in comparison with the other codes. Namely $5.9 \cdot 10^{-8}$ mol/a for Picnic and about $(4.2-4.5) \cdot 10^{-8}$ mol/a for the other codes as it can be seen in Table 2 in the Appendix. Picnic is a one-dimensional computer code and therefore neglects the diffusive flow in the transverse direction, that explains the higher values of the release rates.

Figure 4 shows the results of the simulations for Se-79, which has a zero sorption coefficient in both Opalinus clay and bentonite. The agreement between the different results is good, only Picnic shows a small difference to the results of the other codes. The half life of Se-79 is $1.1 \cdot 10^6$ years, that explains the width of the break-through curve reaching from 10^5 to 10^7 years. The weak decline of the breakthrough curve for long times in comparison with C-14 and Ca-41 is caused by the same decline of the source term.
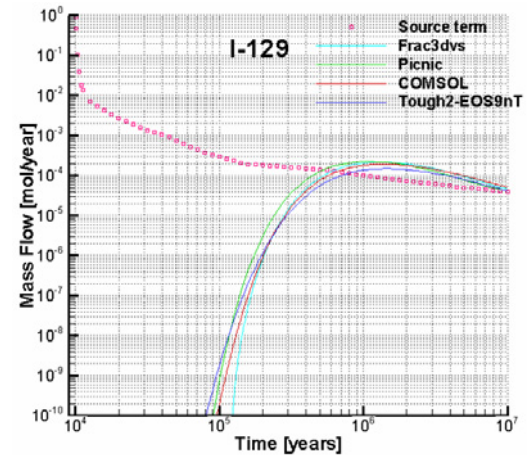

Figure 4. Geosphere release rates for Se-79.

Figure 5 shows the breakthrough curve for Cl-36. The only differences between the input of Cl-36 and that of Se-79 are the half life, which is one order of magnitude larger for Se-79 and the details of the source term. The source term has higher values for Cl-36 but the decrease is also faster. The faster decrease and the lower half life explain that the breakthrough curve extends over a broader range on the time-axis. The higher values of the peak reflect the larger values of the source term for Cl-36 at the beginning.
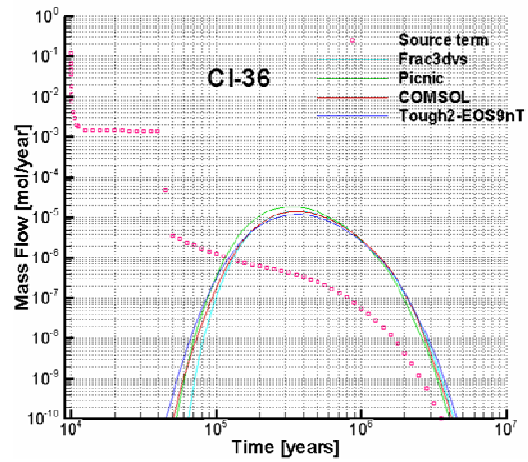
Figure 5. Geosphere release rates for Cl-36.

Finally, Figure 6 shows the results of the calculations for the radionuclide I-129, which migrates through the clay layers as an anion and it is therefore weakly retained by the negatively charged clay layers.

Figure 6. Geosphere release rates for I-129.

I-129 has the longest half life of all considered radionuclides namely $1.57 \cdot 10^7$ years. That to-gether with the slow decrease and the high values of the source term explains that the solute breakthrough curve extends over a very long range of time values even beyond 10^7 years. The retardation effect as result of the sorption coefficient of I-129 is partially compensated by the high values of its source term, what in turn explains also the higher values of the peak for I-129 in comparison with Se-79 and Cl-36.

Small variations in the results obtained with the codes can be explained by differences in the conceptualization and because of different numerical methods implemented in the codes. Picnic uses a one-dimensional approximation for the transport in the geosphere, while the other codes use a two-dimensional approximation. Besides, Tough2-EOS9nT uses the integral finite difference method whereas Picnic and Comsol use the finite element method and Frac3dvs uses both the finite volume and finite difference methods. For all the radionuclides considered in these calculations the results of COMSOL are always in between the results of all the other codes.

As a general conclusion, Picnic tends to overestimate the peak of the mass flow when compared with the results of the other codes. Such a trend can be explained based on the one-dimensionality of Picnic. Tough2-EOS9nT overestimates in comparison with the other codes the mass flow at rising edge of the breakthrough curves. The reason for that is not yet clear but presumably it is due to the different interpolation techniques of the tabular data of the source term. Such an interpolation can be made in many different ways, for example as a linear interpolation between two consecutive points or in terms of a step function in each interval.

6. Conclusions and Outlook

The good agreement between the results of the transport of the radionuclides C-14, Ca-41, Cl-36, Se-79, I-129 can be considered as a successful verification test of COMSOL as a simulation tool for the safety analysis about a nuclear waste geological repository in an homogeneous rock like clay. Moreover, COMSOL is a very user-friendly code that can be used to obtain a quick assessment of a conceptual model for a geological repository to gain insight in how the simulated system works.

ENSI will use COMSOL in combination with other hydrogeological and transport codes to evaluate the calculations of the implementer Nagra within the sectoral plan in Switzerland that started in 2008. In this framework the code will be tested to be used as a tool for the evaluation of fractured systems and for radionuclide decay chains.

7. References

1. COMSOL, *Earth Science Module (Comsol Multiphysics 3.3),* Comsol AB, 2006.
2. COMSOL, *Comsol Multiphysics 3.3, User's Guide*, 2006.
3. Pruess K., Oldenburg C., Moridis G., *Tough2 User's Guide, version 2.0*, 1999.
4. Moridis G.J., Wu Y., Pruess K., *EOS9nT, A Tough2 module for the simulation of water flow and solute/colloid transport in the subsurfaces*, 1999.
5. Nagra NTB 02-05, *Project Opalinus Clay – Safety Report Demonstration of Disposal feasibility for spent fuel; vitrified high-level waste and long-lived intermediate level waste (Entsorgungsnachweis)*, 2002.
6. Kosakowski G., *Time-dependent Flow and Transport Calculations for Project Opalinus Clay (Entsorgungsnachweis),* PSI-Report 04-10, 2004.
7. Sentis M.L., Altorfer F., Herfort M., *Benchmarkberechnungen für die Simulation der Ausbreitung ausgewählter Nuklide innerhalb von Bentonit- und Opalinustonschichten mit den Rechenprogrammen Tough2-EOS9nT und COMSOL,* ENSI-AN-6877, 2009.
8. Sentis M.L., Altorfer F., Kosakowski G., *Benchmark calculations with Tough2-EOS9nT of the transport of radionuclides through clay and bentonite barriers in a geological repository.* Proceedings of Tough Symposium 2009.
9. Nagra NTB 02-03, *Projekt Opalinuston Synthese der geowissenschaftlichen Untersuchungsergebnisse - Entsorgungsnachweis für abgebrannte Brennelemente, verglaste hochaktive sowie langlebige mittelaktive Abfälle*. 2002.
10. Tecplot 360, *User's Guide.* Tecplot, Inc. 2006.

8. Appendix

Table 1. *Material parameters for the different radionuclides used in this benchmark study, (Nagra, NTB 02-06) and (Kosakowski, 2004).*

Parameter	Unit	Ca-41	C_{org}-14	Cl-36	I-129	Se-79
Molecular diffusion coefficient D_0	$m^2\,a^{-1}$	$1.75 \cdot 10^{-2}$	$1.75 \cdot 10^{-2}$	$1.89 \cdot 10^{-3}$	$1.89 \cdot 10^{-3}$	$1.89 \cdot 10^{-3}$
Decay constant λ	a^{-1}	$6.73 \cdot 10^{-6}$	$1.21 \cdot 10^{-4}$	$2.31 \cdot 10^{-6}$	$4.41 \cdot 10^{-8}$	$6.3 \cdot 10^{-7}$
Half life $T_{1/2}$	a	$1.03 \cdot 10^{5}$	$5.73 \cdot 10^{3}$	$3 \cdot 10^{5}$	$1.57 \cdot 10^{7}$	$1.1 \cdot 10^{6}$
Bentonite	**Unit**	**Ca-41**	**C_{org}-14**	**Cl-36**	**I-129**	**Se-79**
Effective porosity ϕ_t	-	0.36	0.36	0.05	0.05	0.05
Effective diffusion coefficient D_e	$m^2\,s^{-1}$	$2 \cdot 10^{-10}$	$2 \cdot 10^{-10}$	$3 \cdot 10^{-12}$	$3 \cdot 10^{-12}$	$3 \cdot 10^{-12}$
Distribution coefficient for sorption K_s	$m^3\,kg^{-1}$	$3 \cdot 10^{-3}$	0	0	$5 \cdot 10^{-4}$	0
Tortuosity τ	-	1.0	1.0	1.0	1.0	1.0
Opalinus Clay	**Unit**	**Ca-41**	**C_{org}-14**	**Cl-36**	**I-129**	**Se-79**
Effective porosity ϕ_t	-	0.12	0.12	0.06	0.06	0.06
Effective diffusion coefficient D_e	$m^2\,s^{-1}$	$1 \cdot 10^{-11}$	$1 \cdot 10^{-11}$	$1 \cdot 10^{-12}$	$1 \cdot 10^{-12}$	$1 \cdot 10^{-12}$
Distribution coefficient for sorption K_s	$m^3\,kg^{-1}$	$1 \cdot 10^{-3}$	0	0	$3 \cdot 10^{-5}$	0
Tortuosity τ	-	0.15	0.15	0.278	0.278	0.278

Table 2. *Comparison of the maximum values of the release rates for the different codes.*

Nuclide	Frac3dvs		Picnic		Comsol		Tough2-EOS9nT	
	Max. [mol/year]	$t_{max.}$ [year]	Max. [mol/year]	$t_{max.}$ [year]	Max. [mol/year]	$t_{max.}$ [year]	Max. [mol/year]	$t_{max.}$ [year]
Ca-41	$2.6 \cdot 10^{-10}$	$6.3 \cdot 10^{9}$	$2.8 \cdot 10^{-10}$	$6.3 \cdot 10^{5}$	$2.8 \cdot 10^{-10}$	$6.2 \cdot 10^{5}$	$2.8 \cdot 10^{-10}$	$6.2 \cdot 10^{5}$
C_{org}-14	$4.5 \cdot 10^{-8}$	$4.5 \cdot 10^{4}$	$5.9 \cdot 10^{-8}$	$4.5 \cdot 10^{4}$	$4.2 \cdot 10^{-8}$	$4.6 \cdot 10^{4}$	$4.2 \cdot 10^{-8}$	$4.5 \cdot 10^{4}$
Se-79	$5.3 \cdot 10^{-7}$	$1.4 \cdot 10^{6}$	$6.3 \cdot 10^{-7}$	$1.3 \cdot 10^{6}$	$5.6 \cdot 10^{-7}$	$1.4 \cdot 10^{6}$	$4.8 \cdot 10^{-7}$	$1.4 \cdot 10^{6}$
Cl-36	$1.4 \cdot 10^{-5}$	$3.6 \cdot 10^{5}$	$1.9 \cdot 10^{-5}$	$3.2 \cdot 10^{5}$	$1.4 \cdot 10^{-5}$	$3.6 \cdot 10^{5}$	$1.2 \cdot 10^{-5}$	$3.6 \cdot 10^{5}$
I-129	$2.2 \cdot 10^{-4}$	$1.3 \cdot 10^{6}$	$2.3 \cdot 10^{-4}$	$1.3 \cdot 10^{6}$	$1.9 \cdot 10^{-4}$	$1.4 \cdot 10^{6}$	$1.5 \cdot 10^{-4}$	$1.4 \cdot 10^{6}$