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PORFLOW Simulations Supporting the Saltstone Performance Assessment Revision

G. P. Flach

T. Hang

December 13, 2018

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Ralph Nichols provided van Genuchten parameters for granular materials based on WSRC-STI-2006-00198.

EXECUTIVE SUMMARY

The most recent Performance Assessment (PA) of the Z-Area Saltstone Disposal Facility (SDF) was performed in 2009, and three Special Analyses were subsequently conducted. Currently the SDF PA analysis is being revised by the Savannah River Remediation (SRR) Waste Disposal Authority group to reflect updated input information and assumptions, and improved understanding of disposal system performance. In support of the PA revision, Savannah River National Laboratory has developed revised methods for simulating the transport of I-129 and Tc-99 through porous media in a shrinking core model using the PORFLOW code. The revised methods are based on updated reduction capacity, I-129 distribution coefficient, and Tc-99 solubility values from SRR. PORFLOW simulations of aquifer transport have been revised to reflect a recent update to the General Separations Area groundwater flow model and improved treatment of plume dispersion. Hydraulic properties for some soils / sediments have been revised based on E-Area PA maintenance work. Example simulation results are provided to illustrate the behavior of the new I-129 and Tc-99 shrinking core models.

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CA	Composite Analysis
GSA	General Separations Area
PA	Performance Assessment
SA	Special Analysis
SDF	Saltstone Disposal Facility
SDU	Saltstone Disposal Unit
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation LLC

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1.0 Introduction

The most recent Performance Assessment (PA) of the Z-Area Saltstone Disposal Facility (SDF) was performed in 2009 (SRR 2009) and three Special Analyses (SAs) were subsequently conducted (SRR 2013, 2014, 2016). Currently the SDF PA analysis is being revised by the Savannah River Remediation (SRR) Waste Disposal Authority group to reflect updated input information and assumptions, and improved understanding of disposal system performance.

In support of the PA revision, Savannah River National Laboratory (SRNL) has developed revised methods for simulating the transport of I-129 and Tc-99 through porous media in a shrinking core model using the PORFLOW code developed by Analytic & Computational Research, Inc. (https://www.acricfd.com/software/porflow/). The revised methods are based on updated reduction capacity, I-129 distribution coefficient, and Tc-99 solubility values from Hommel and Dixon (2018), Lester (2018a), and Lester (2018b), respectively. Also, PORFLOW simulations of aquifer transport have been revised to reflect a recent update to the General Separations Area (GSA) groundwater flow model (Flach 2018c) and improved treatment of plume dispersion based on recommendations of Flach (2018a). Other updates to the PA inputs include Hommel (2018a), Flach (2018b), and Watkins (2018).

The scope of the efforts described herein are defined by Task Technical Request G-TTR-Z-00012 and Task Technical and Quality Assurance Plan SRNL-RP-2018-00620 (Hang 2018).

1.1 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in Manual E7 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2. Danielson (2018) performed a Quality Assurance design check of the present work.

2.0 Shrinking Core Model Revisions

In the SDF Special Analyses, Tc-99 transport through a Saltstone Disposal Unit (SDU) was simulated through shrinking core models described by Jordan and Flach (2013), Flach and Taylor (2014), and Flach (2017). Reducing cementitious materials, concrete and grout containing ground blast furnace slag, are assigned an initial solid phase reduction capacity that is consumed over time by transport of dissolved oxygen into the SDU. At intermediate times, the core of an SDU remains in a reduced chemical state while the outer fringes have become oxidized. The reduced core shrinks over time due to ongoing oxygen ingress, hence the descriptor "shrinking core". In the reduced region, the aqueous concentration of Tc-99 is controlled by a solubility limit (mol/L). In the oxidized region, Tc-99 concentration is controlled by a distribution coefficient (K_d, mL/g).

The original shrinking core method has been revised to reflect laboratory studies of actual releases from saltstone and saltstone simulants (Seaman et al 2018). Release modeling for Tc-99 considers two solubility limits under reduced conditions (Lester 2018b): a higher value for early times when pH > 11 (e.g. 9.7e-07 mol/L) and a lower value for later times when pH < 11 (e.g. 4.5e-07 mol/L). The shrinking core model has also been extended to I-129 transport using the recommendations of Lester (2018a). Under reduced conditions, two distribution coefficients are implemented for grout: a lower value of K_d until the first pore volume is flushed (e.g. 0.07 mL/g), followed by a higher value (e.g. 0.71 mL/g). After a region becomes oxidized, a third K_d is operative (e.g. 4 mL/g). Each shrinking core model is further described below.

2.1 Tc-99 Shrinking Core Model

Under reduced conditions the effective distribution coefficient resulting from a solubility limit (${K_d}^{solubility}$) is given by Equation (12) in Flach and Taylor (2014) as

$$ {K_d}^{solubility} = max\left[\frac{c_T - nSc_{sol}}{\rho_b c_{sol}}, K_{d,Re}\right] \quad (1) $$

where c_T = total bulk Tc-99 concentration (m_{Tc}/V) (i.e., mass of Tc-99 divided by total volume), n = porosity, S = saturation, c_{sol} = Tc solubility limit under reducing conditions, and ρ_b = bulk density. Two, pH dependent, solubilities are implemented by blending the end-members according to a weighting fraction $0 \leq x_{pH} \leq 1$

$$ c_{sol} = x_{pH}{c_{sol}}^{pH>11} + (1 - x_{pH}){c_{sol}}^{pH<11} \quad (2) $$

where $x_{pH} = 1$ when $pH > 11$ and $x_{pH} = 0$ when $pH < 11$.

Figure 17 of Lester (2018b) indicates that the solubility transition at $pH = 11$ occurs at about 6 pore volume flushes through an SDU2A Saltstone core sample, which is also consistent with findings from Dyer (2018). Pore volume flushes can be monitored in PORFLOW by creating a hypothetical solid phase buffering capacity and introducing a hypothetical dissolved tracer that consumes the buffer. The buffer-tracer system simulates evolving pH conditions in a shrinking core manner analogous to the reductant-oxygen system for Eh.

The concentration of the tracer is arbitrarily set to 1.0e-3 mol/mL, which is similar in numerical magnitude to the oxygen concentration of 1.06e-3 meq e-/mL in the Eh shrinking core model. Based on this reference tracer concentration, the buffering capacity of Saltstone grout is calculated as $c_{buffer,0}$ = 4.22e-3 mol/g. Lacking experimental data to the contrary, the $pH = 11$ condition for concrete is assumed to occur at 6 pore volumes too, and the corresponding buffering capacity is $c_{buffer,0}$ = 0.303e-3 mol/g. The weighting fraction x_{pH} is then defined by

$$ x_{pH} = \frac{c_{buffer}}{c_{buffer,0}} \quad (3) $$

The remainder of the Eh shrinking core model described by Flach and Taylor (2014) is unaltered.

In summary, the Tc-99 transport simulation now includes two shrinking core models: an Eh model representing reduced versus oxidized conditions, and a pH model representing regions where $pH > 11$ and $pH < 11$. The pH model controls blending of the two solubilities for $pH > 11$ and $pH < 11$ under reduced conditions. The Eh model controls blending between solubility control and K_d control, as before. The combined shrinking core models can be termed “dual-solubility + redox”.

2.2 I-129 Shrinking Core Model

In current PORFLOW modeling, I-129 transport is simulated in a manner much like Tc-99. Under reducing conditions, the partition coefficient for I-129 takes on one value through the first pore volume flush (K_{d1}), and a second value after that (K_{d2}) per Lester (2018a). Analogous to Equation (2) for Tc-99, K_d^{Re} under the range of pH conditions can be computed as

$$K_d^{Re} = x_{pH}K_{d1} + (1 - x_{pH})K_{d2} \quad (4)$$

where x_{pH} is defined by Equation (3). However, buffering capacities for grout and concrete in the I-129 simulation are computed as $c_{buffer,0}$ = 0.704e-3 mol/g and 0.050e-3 mol/g, respectively, based on a transition at 1 instead of 6 pore volumes. Note that 1 pore volume is assumed instead of 6 pore volumes based on an analytical evaluation of actual I-129 releases per Lester (2018a).

The transition between reduced and oxidized conditions is handled by blending K_d^{Re} and K_d^{Ox}:

$$K_d = x_{Re}K_d^{Re} + (1 - x_{Re})K_d^{Ox} \quad (5)$$

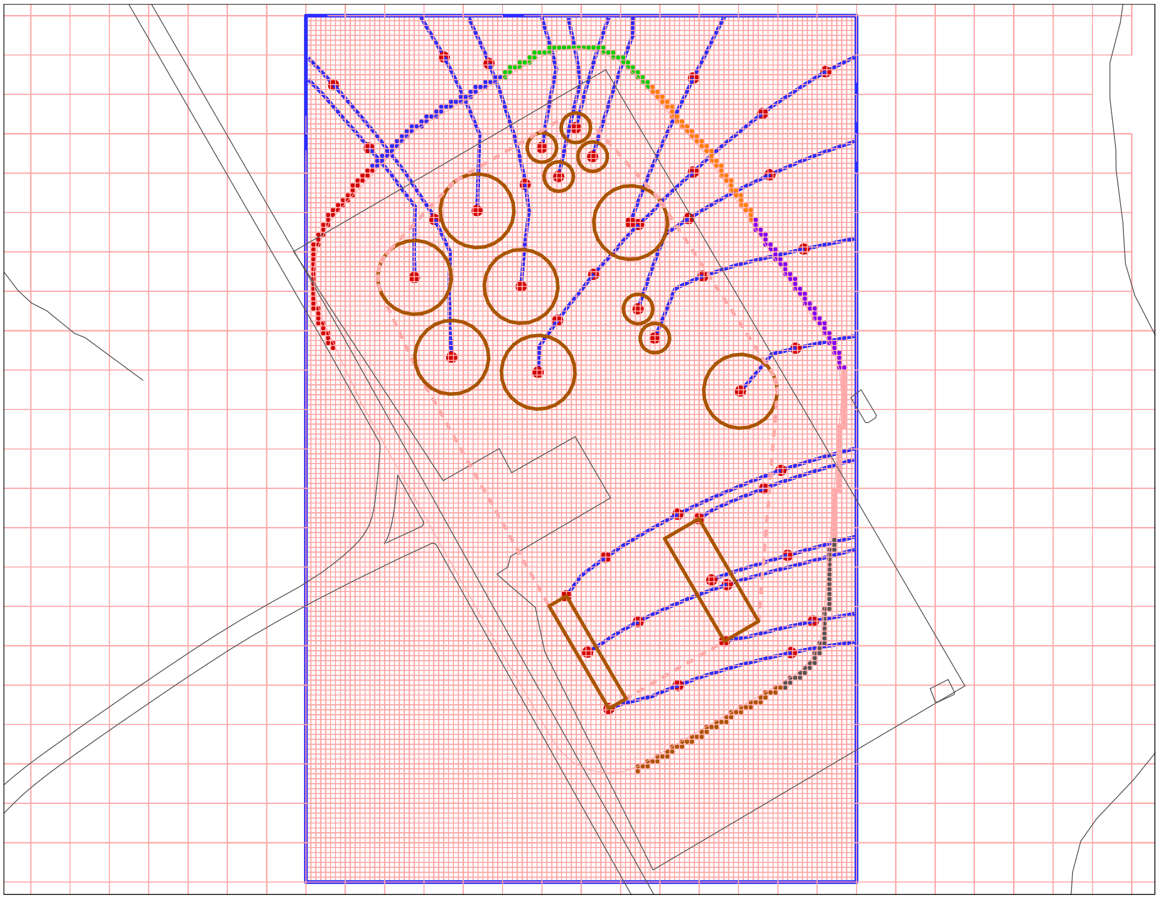
where x_{Re} is the fraction remaining of the initial reductant (slag concentration).

In summary, the I-129 transport simulation now includes two shrinking core models: an Eh model representing reduced versus oxidized conditions, and a pH model representing regions of higher and lower pH. The pH model controls blending of the two K_d values for higher and lower pH under reduced conditions. The Eh model controls blending between the effective K_d under reduced conditions and K_d under oxidized conditions. The combined shrinking core models can be termed "dual-K_d + redox".

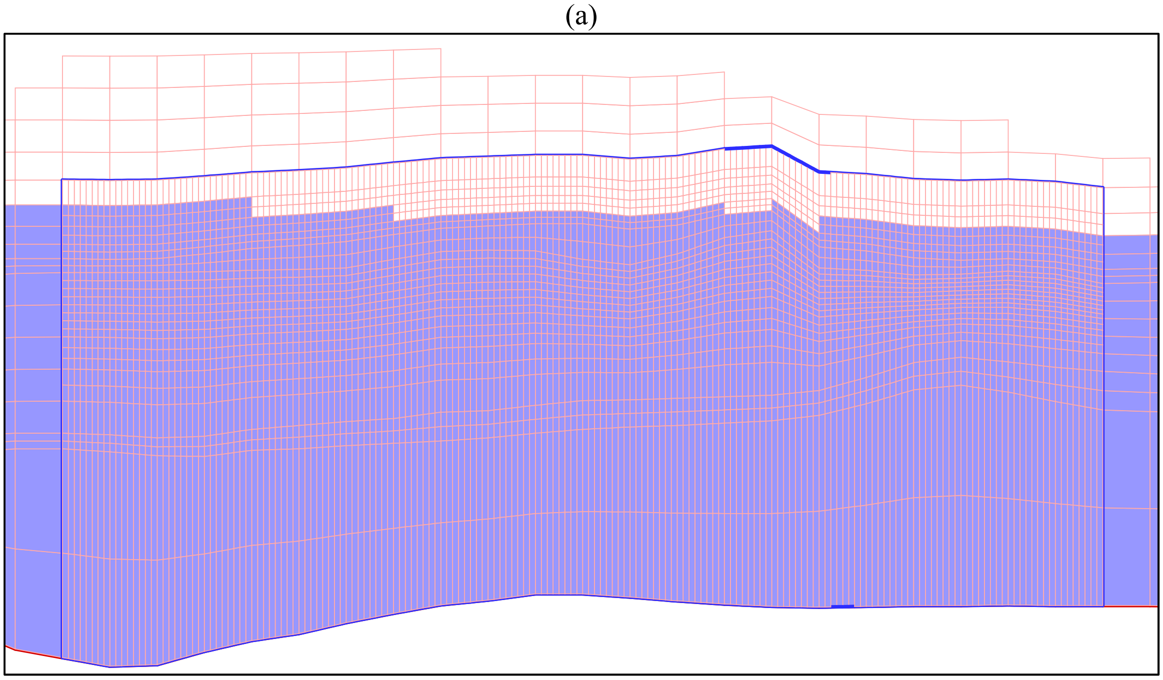
3.0 Aquifer Transport Updates

The groundwater flow model supporting Performance Assessments (PAs) and Composite Analyses (CAs) at the Savannah River Site was significantly revised in 2016 and 2017 using new hydrostratigraphic surfaces, updated well water level calibration targets, and semi-automated model calibration with the PEST optimization code (Flach et al. 2017). This model is referred to as "GSA_2016". The GSA_2016 model was further refined in 2018 to incorporate additional updates to model calibration targets, closure of the H-Area Ash Basin, construction of E-Area Slit Trench operational covers, and plume information from the Mixed Waste Management Facility and Low-Level Radioactive Waste Disposal Facility. Another objective was to lower hydraulic head residuals by adding another calibration zone. The resulting model is referred to as "GSA_2018", or "GSA_2018.LW" in explicit reference to a Layer-cake hydraulic conductivity field and Weighted calibration targets in the optimization objective function to be minimized (Flach 2018c). The groundwater flow field from the GSA_2018 model was adopted for aquifer transport simulations.

However, to minimize numerical dispersion and grid size, a Z-Area sub-region of the GSA_2018 coarse grid is cut out and refined using the MESH3D code (Flach 2012). Figure 3-1 shows the selected horizontal and vertical extents of the Z-Area grid. Also shown is the horizontal and vertical grid refinement relative to the original, coarse mesh, GSA_2018 grid. The selected horizontal grid resolution is 25 ft based on recommendations by Flach (2018a). The selected vertical resolution near the water table is approximately 3 ft per Flach (2018a). Note that the simulated water table is visually depicted in Figure 3-1 as the interface between the cells (or nodes) that are shaded in blue and those that are not. The selected model settings avoid significant numerical dispersion in comparison to specified physical dispersion in PORFLOW transport simulations. Representative stream traces emanating from each SDU are shown to indicate general groundwater flow direction and speed.



(a)



Blue shading indicates the saturated zone, no shading the vadose zone

(b)

Figure 3-1. Cutout and refined Z-Area grid: (a) plan view with 5-year streamtrace markers and (b) N-S cross-sectional view.

4.0 Hydraulic Property Updates for Soils

Hydraulic properties for native sediments and control compacted backfill were previously taken from Phifer et al. (2006). The previous properties have been retained with two notable exceptions.

First, water retention and relative permeability curves in Phifer et al. (2006) are defined through data tables. For the Saltstone PA revision, van Genuchten (1980) / Mualem (1976) functions were fit to the sample data underlying these tables, for more convenient generation of characteristic curves in plots and material property blending. The van Genuchten / Mualem analytical curves closely approximate the original tabular curves. As an example, the water retention curve from the WSRC-STI-2006-00198 data table for "Sand" is compared to the analytical van Genuchten curve in Figure 4-1.

Second, the sample data set underlying the original "Control Compacted Backfill" was modified to be more representative of the material classification. Figure 4-2 compares the original water retention data curve to the revised van Genuchten analytical curve.

Table 4-1. van Genuchten parameters for sediments and soils.

Material name	θs	θr	α (1/cm)	n	m	Name
Upper Vadose Zone	0.3890	0.2214	0.0144	1.3881	0.2796	UpperVadoseZone
Lower Vadose Zone	0.3905	0.1604	0.0259	1.4048	0.2882	LowerVadoseZone
E-Area Operational Soil Cover Before Dynamic Compaction	0.4560	0.2596	0.0185	1.3881	0.2796	OscBefore
E-Area Operational Soil Cover After Dynamic Compaction	0.2700	0.1537	0.0082	1.3879	0.2795	OscAfter
Control Compacted Backfill	0.37	0.224	0.0219	1.659	0.3972	CCbackfill
IL Vault Permeable Backfill	0.4100	0.1605	0.0199	2.1063	0.5252	ILVbackfill
Single Vadose Zone	0.38996	0.18556	2.21E-02	1.38402	0.277467	SingleVadoseZone
Sand (<25% Mud)	0.3709	0.1074	0.048829	1.312885	0.23832	Sand
Clay-Sand (25-50% Mud)	0.2489	0.1058	0.0212	1.2376	0.1919	ClaySand
Clay (>50% Mud)	0.2157	0.1182	0.0125	1.1902	0.1598	Clay
Gravel	0.2505	0.0189	0.1266	1.5001	0.3334	Gravel

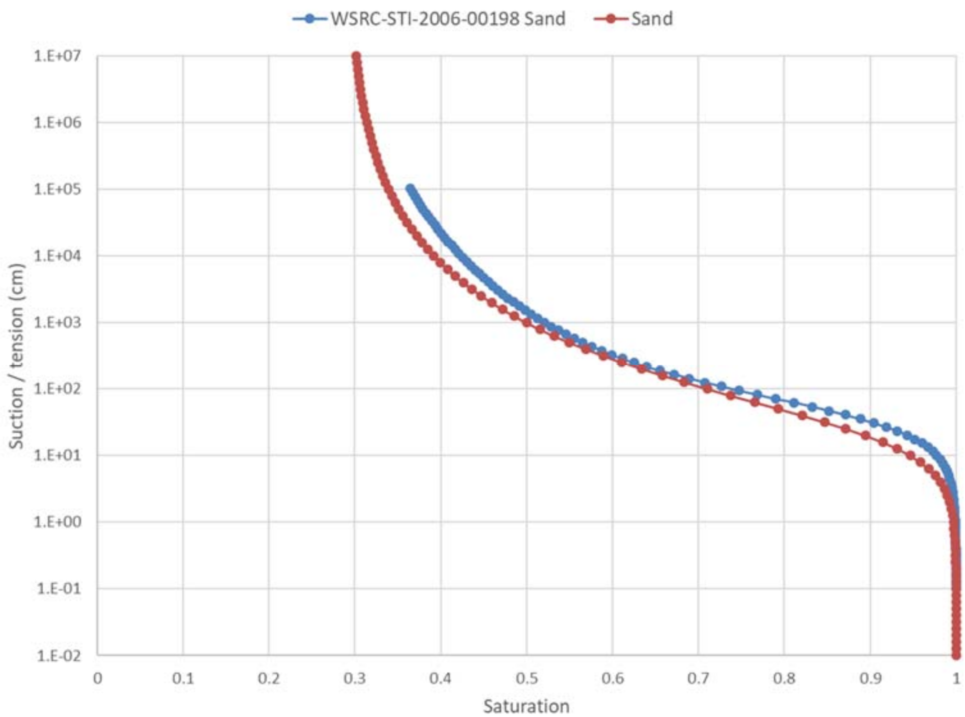


Figure 4-1. Water retention curve for "Sand" from WSRC-STI-2006-00198 data tables compared to van Genuchten / Mualem fit of underlying sample data.

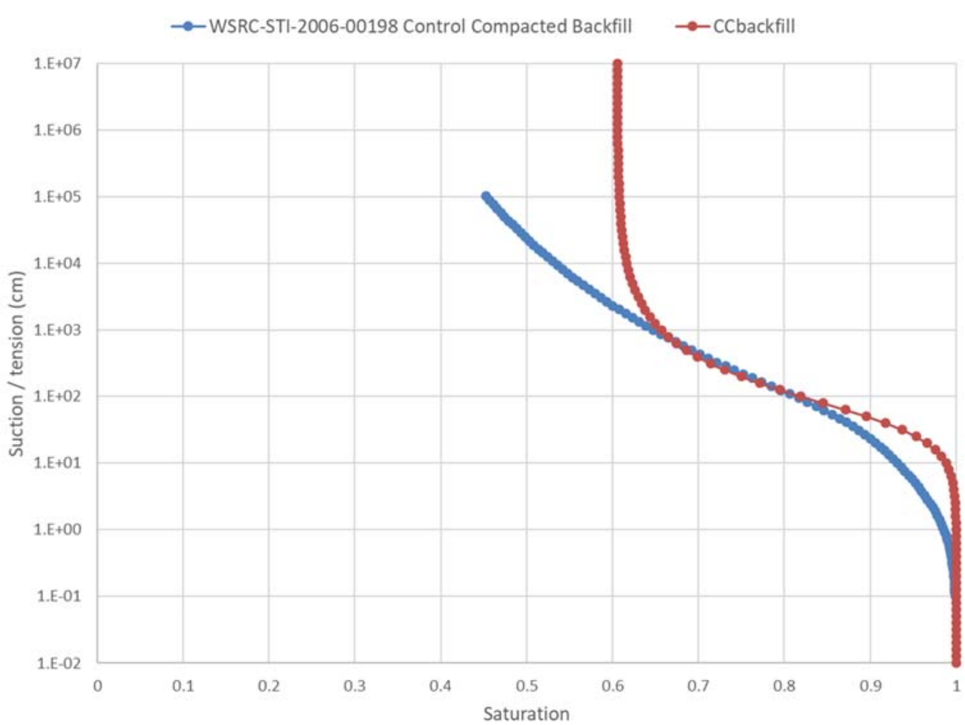


Figure 4-2. Water retention curve for "Control Compacted Backfill" from WSRC-STI-2006-00198 data tables compared to van Genuchten / Mualem fit of a <u>modified</u> sample data.

5.0 Example Simulation Results

Section 2.0 describes revisions to the shrinking core model previously applied to Tc-99 in Special Analyses following the 2009 SDF PA, and extension of the shrinking core model to I-129. This section presents example simulation results for SDU 7 illustrating the general behavior of these models. As a point of reference, simulation results are first presented for Cl-36, followed by the I-129 and Tc-99 results.

The results presented here reflect a representative simulation case labeled "CaseCV.5_100k." In support of the SDF PA, multiple modeling cases are run, each providing additional insights with respect to the behavior of the disposal system. "CaseCV.5" was developed as the "Compliance Case", that is, the case used to establish that waste disposal at the SDF will not exceed the applicable performance objectives. As the Compliance Case, "CaseCV.5" assumes "compliance values" for all model inputs, hence the "CV" in the case identifier. These compliance values were generally selected to represent a compromise between the most probable and most defensible inputs. The ".5" appended to the case identifier indicates that the PORFLOW model has undergone five iterations during model development to resolve various input, and implementation issues. Finally, the "100K" at the end of the case identifier indicates that transport for this specific modeling case was simulated out to 100,000 years after closure; as the default model setting, the other modeling cases supporting the SDF PA were simulated out to 20,000 years after closure.

Other modeling cases that have been developed include "CaseBE," or the "Realistic Case," which assumes "Best Estimate" inputs, and "CaseCE" or the "Defense-In-Depth Case," which assumes "Conservative Estimate" inputs. Additional cases include 54 parametric flow cases ("CaseF01" through "CaseF54"), which simulated various combinations select parameters to evaluate different flow conditions (Hommel 2018b); and the sensitivity analysis cases ("CaseSA##" series). In general, the sensitivity analysis cases are copies of "CaseCV," except for a single parameter, or small number of parameters, varied to provide a comparative result for analysis.

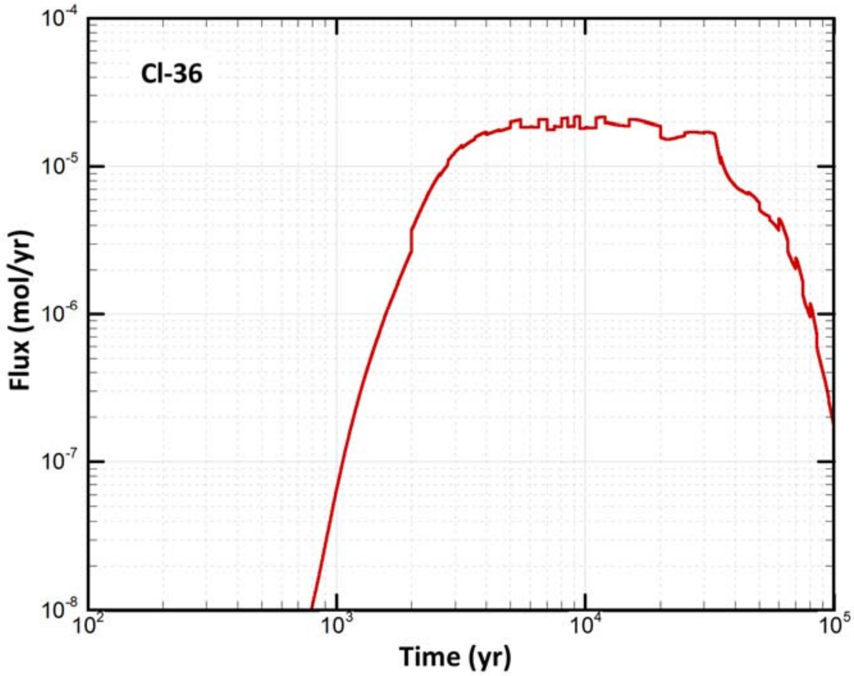
Figure 5-1 shows the molar flux of Cl-36 leaving the vadose zone and entering the saturated zone (i.e., crossing the water table) for CaseCV.5_100k. In this simulation the distribution coefficient for Cl-36 in Saltstone transitions at 32,462 years from K_d = 0 mL/g for "reducing young" conditions to K_d = 1 mL/g for "reducing old" conditions. The timing of this transition is driven by the chemical evolution of the Saltstone waste form as infiltrating water passes through it, as described in Dyer 2018 (i.e., six pore volume exchanges). Otherwise, changes in Cl-36 flux are a result of flow field changes and source depletion.

The abrupt stair-step changes in Cl-36 flux are a result of switching from one steady-state flow field to another at discrete times. Although material properties change gradually with time, the flow simulation is highly non-linear and severely challenges the ability of PORFLOW to achieve accurate convergence for some time intervals. The jagged portions of the Cl-36 flux curve, particularly centered around 10,000 years, reflect a level of non-uniqueness in the numerical solution, and can be considered noise. The Cl-36 results provide a baseline reference to which I-129 and Tc-99 shrinking core results may be compared.

Figure 5-2 illustrates the simulated flux of I-129 crossing the water table. At the early times the I-129 flux exhibits the same character as Cl-36, reflecting mostly changes in the steady-state flow fields underlying the transport simulation. However, the I-129 release is longer and peaks later than Cl-36 due to a gradual increase in K_d in the shrinking core model from 0.07 mL/g (reducing conditions, higher pH) to 0.71 mL/g (reducing conditions, lower pH) to 4.0 mL/g (oxidized conditions). Figure 5-3 plots the concentrations of reductant (slag, Eh control, labeled as C3), buffer (pH control, labeled as C5), and I-129 (labeled as C) early in the 100,000 year simulation (250 years). Figure 5-4 plots the same variables at 20,000 years. The buffering capacity of Saltstone with respect to pH control (C5) is lower than the reducing capacity (C3). Thus, the high pH core of Saltstone has shrunk significantly more than the low Eh core. The core of high

I-129 concentrations is smaller than the buffer core because I-129 is relatively mobile in this region (reducing conditions + high $pH \rightarrow K_d$ = 0.07 mL/g).

Figure 5-5 through Figure 5-7 provides the same series of plots for the Tc-99 shrinking core model. While both models have the same amount of reductant (slag), the Tc-99 model has a larger amount of buffer so that the pH transition occurs at 6 pore volumes instead of 1 pore volume for I-129. Late time results are shown for 50,000 years, to see comparable shrinkage in the buffer core as I-129. The Tc-99 concentration plot shows two distinct solubility control regions. Tc-99 concentration in the intersection of the reductant and buffer cores is 9.7e-7 mol/L. Tc-99 concentration within the reductant core but outside the buffer core is 4.5e-7 mol/L. Outside of the reductant core, Tc-99 concentration is controlled by K_d = 0.5 mL/g. Because Tc-99 is easily swept away when under K_d control, concentrations are very low.



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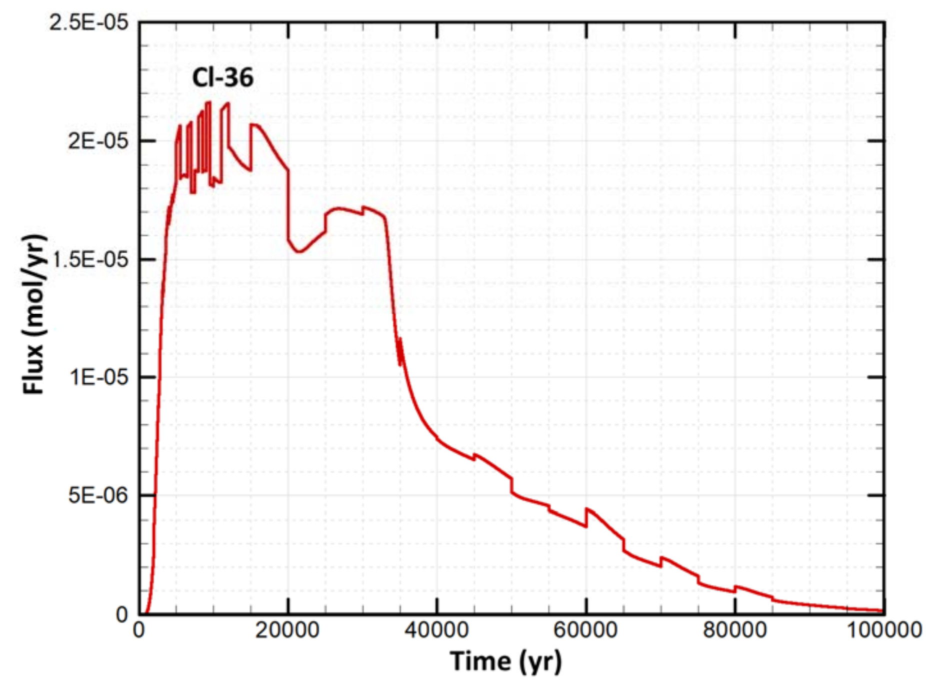
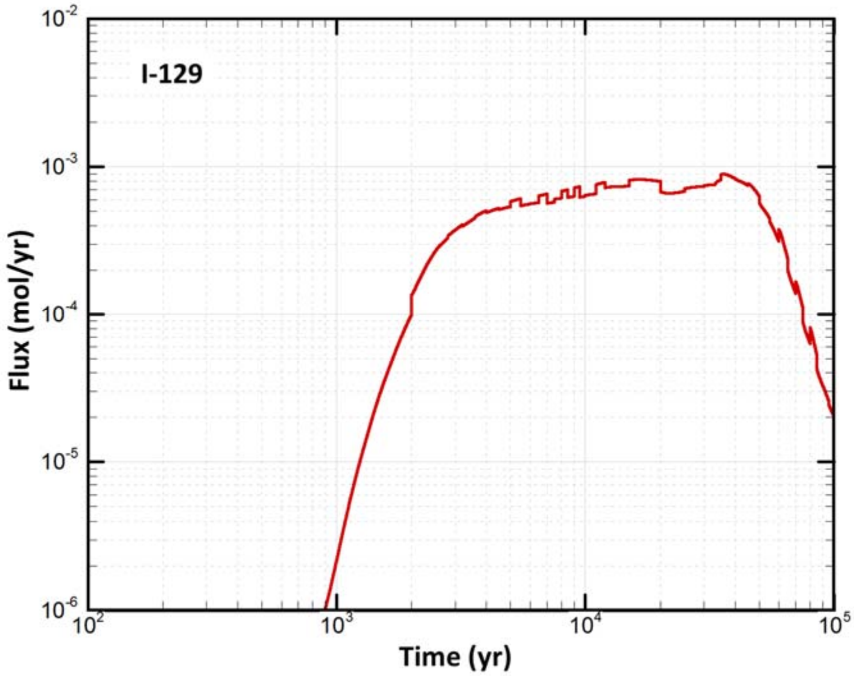


(b)

Figure 5-1. Molar flux of Cl-36 at the water table for CaseCV.5_100k using (a) logarithmic and (b) linear scales



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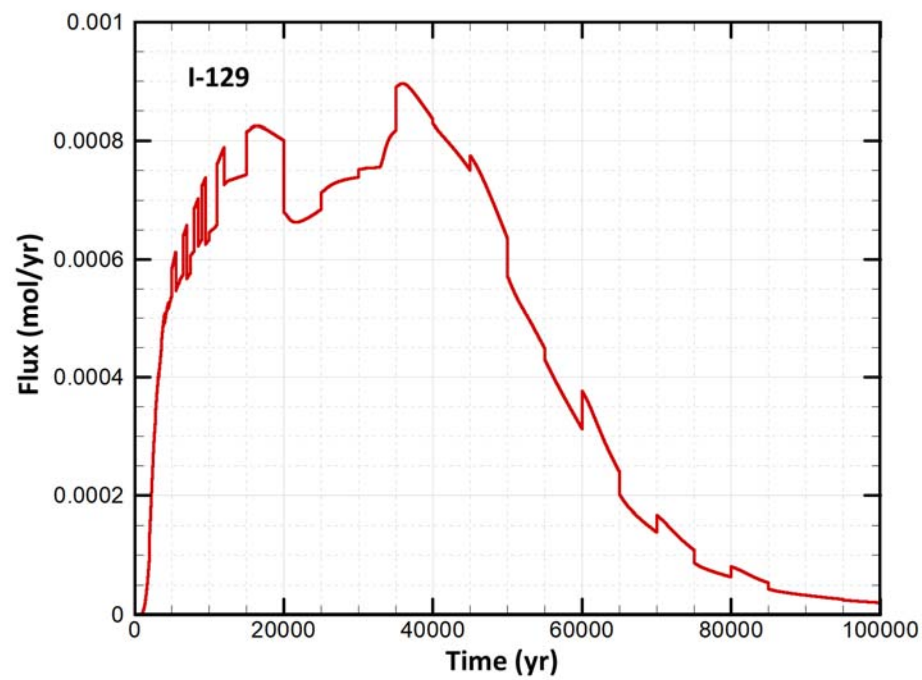


(b)

Figure 5-2. Molar flux of I-129 at the water table for CaseCV.5_100k using (a) logarithmic and (b) linear scales

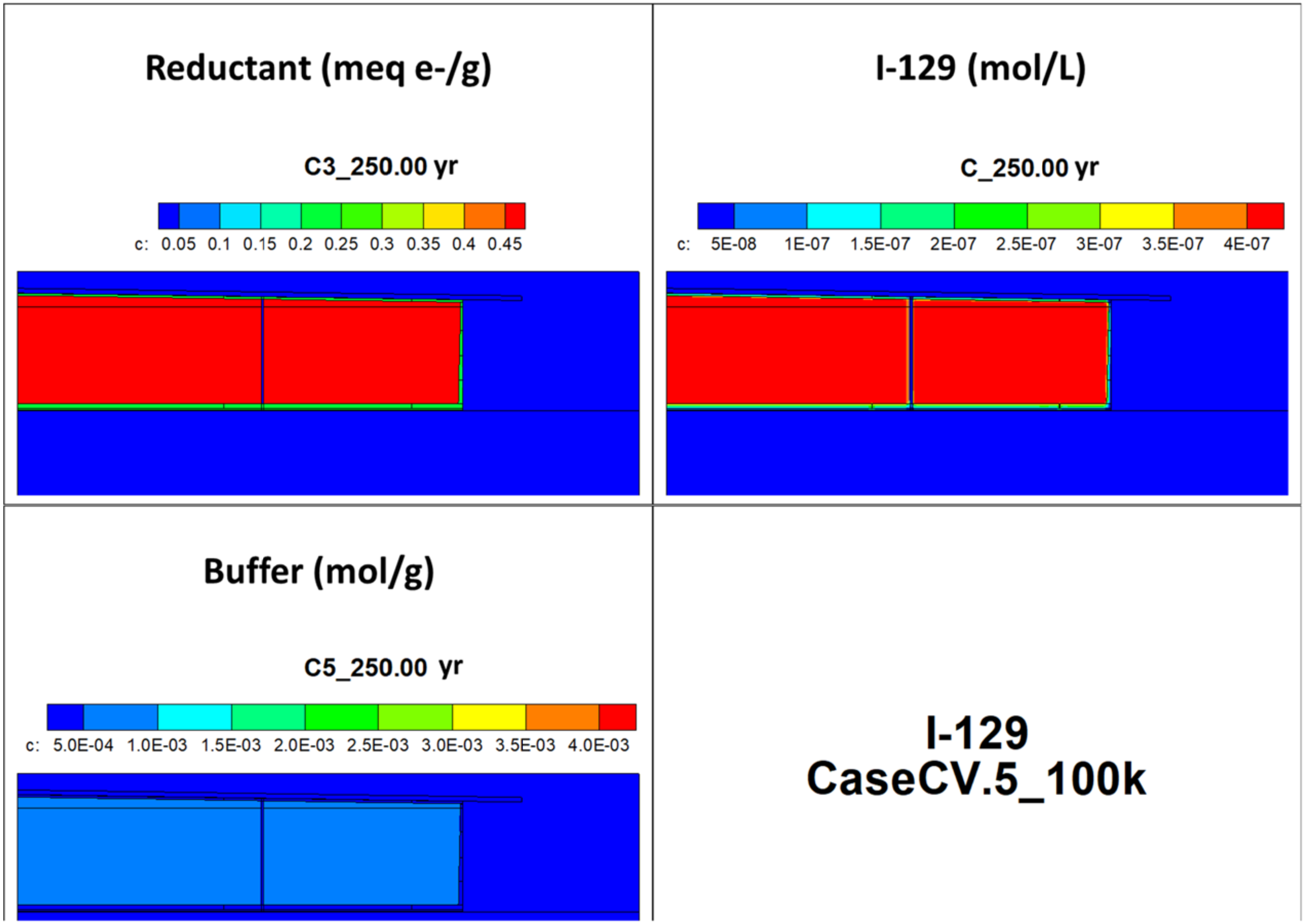


Figure 5-3. Concentrations of reductant (C3), buffer (C5), and I-129 (C) at 250 years for SDU 7.

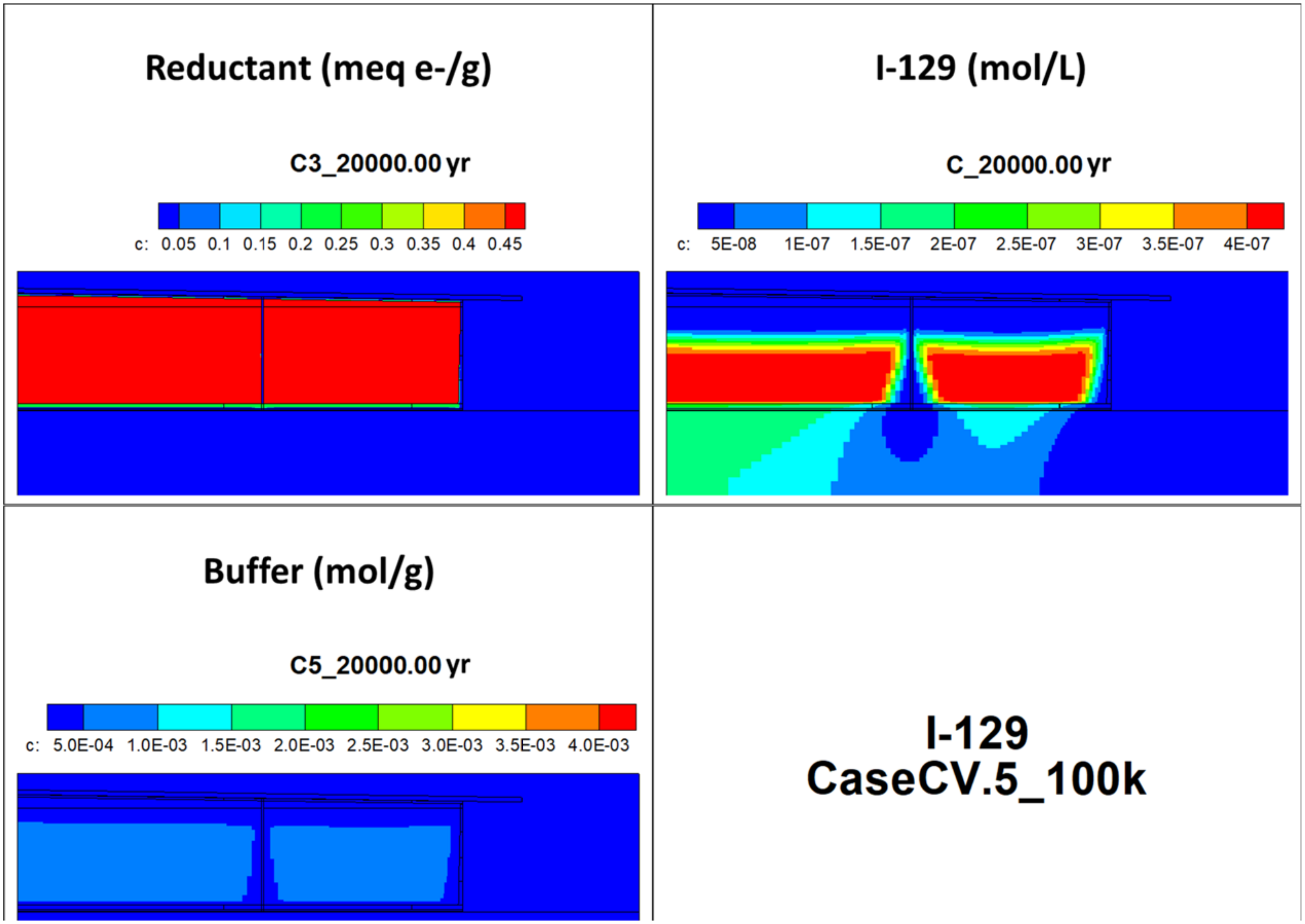
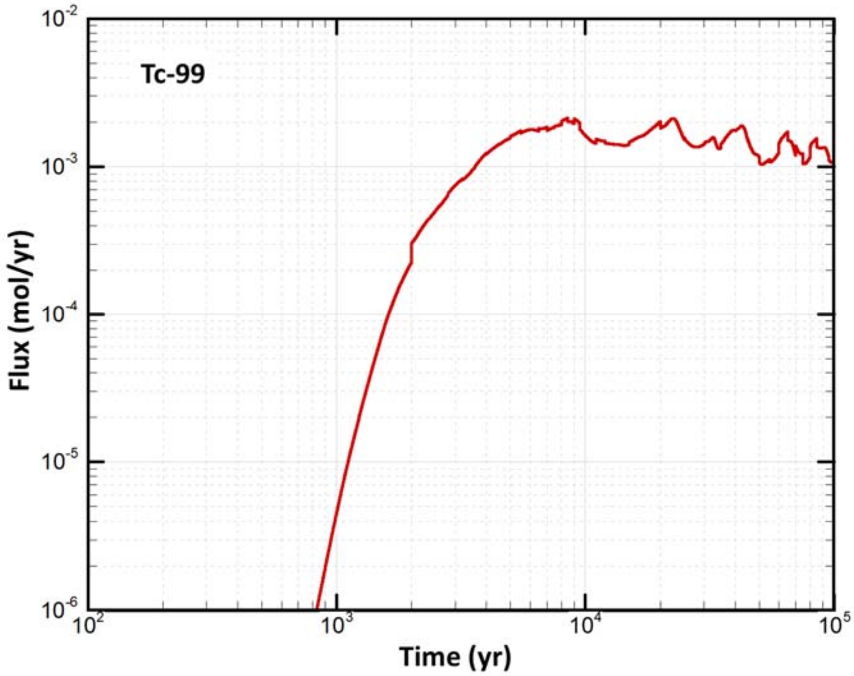
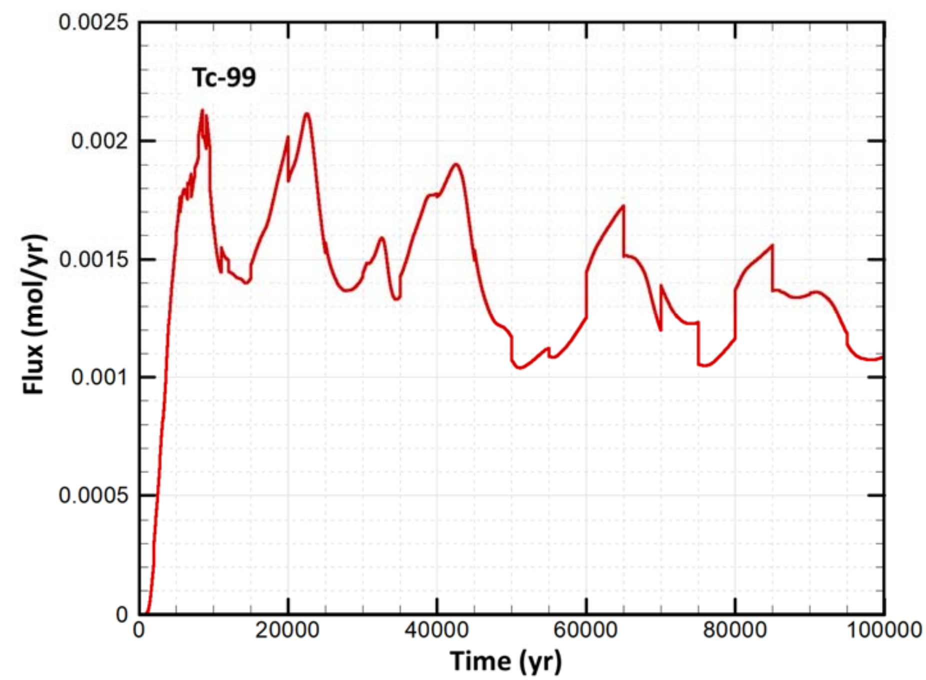


Figure 5-4. Concentrations of reductant (C3), buffer (C5), and I-129 (C) at 20,000 years for SDU 7.



(a)



(b)

Figure 5-5. Molar flux of Tc-99 at the water table for CaseCV.5_100k using (a) logarithmic and (b) linear scales

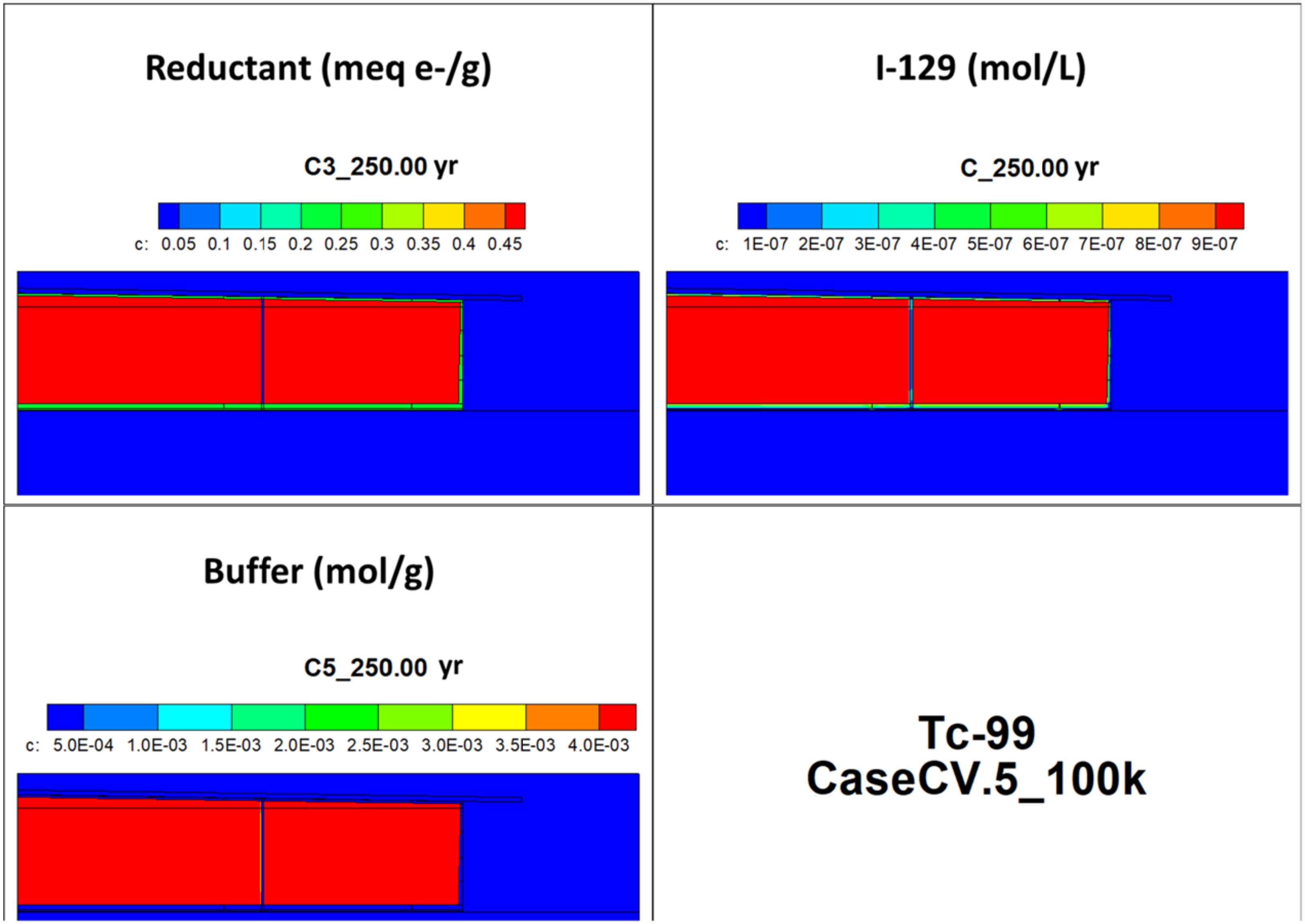


Figure 5-6. Concentrations of reductant (C3), buffer (C5), and Tc-99 (C) at 250 years for SDU 7.

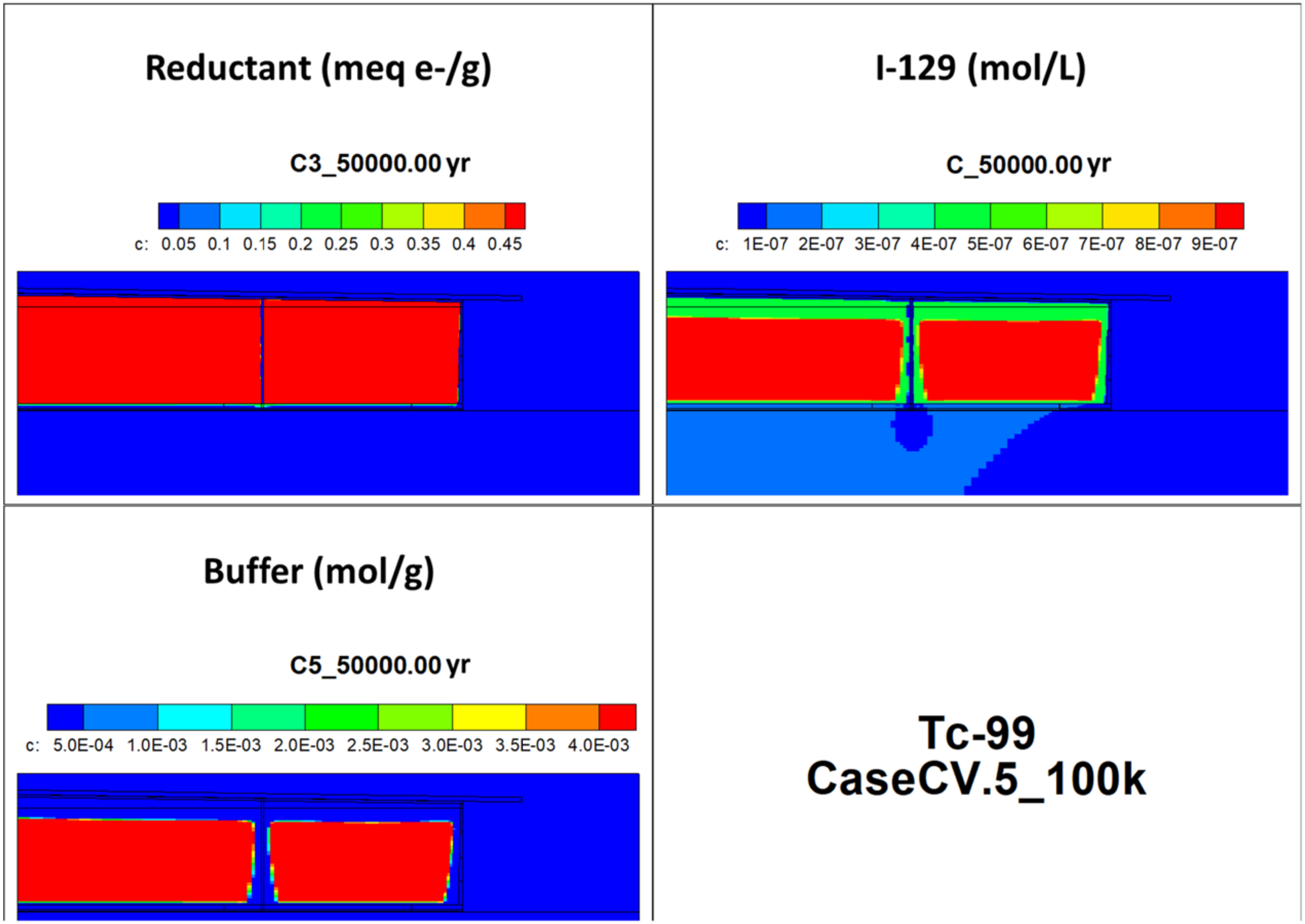


Figure 5-7. Concentrations of reductant (C3), buffer (C5), and Tc-99 (C) at 50,000 years for SDU 7.

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