Supplementary material

Potential Uranium Migration Within the Geochemical Gradient of the Opalinus Clay System at the Mont Terri

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Supplementary material S1

Table S1. Summary of the pore water components used for model calibration. Concentrations are given as averaged values from borehole analyses at the underground laboratory Mont Terri (Switzerland).

Parameter & Unit \ Borehole		BK[a] (Dogger aquifer)	BWS-A3[a,b] (OPA sandy)	BWS-A2[a] (OPA sandy)	BGP-1[a] (OPA carbonate-rich)	BWS-A1[a,b] (OPA shaly)	BPC-C1[c] (OPA shaly)	E6[a] (Liassic shales)	JM[a] (Liassic shales)	G2[a] (Liassic aquifer)
pH	-	7.38	7.39	7.55	7.28	7.49	7.13	7.78	7.96	7.44
Na^+	mmol/L	0.83	121	197	195	230	276	281	43.8	1.29
K^+	mmol/L	0.38	0.87	1.14	0.73	1.47	1.93	1.28	0.29	0.11
Mg^{2+}	mmol/L	2.65	5.91	11.01	15.89	16.79	21.97	24.71	0.71	0.67
Ca^{2+}	mmol/L	1.99	6.73	8.23	16.37	16.03	18.91	19.28	0.69	2.09
Sr^{2+}	mmol/L	0.23	0.35	0.42	0.44	0.47	0.46	0.35	0.05	0.03
Fe_{total}	µmol/L	1.27	9.81	10.57	n.a.	8.37[c]	29.62	n.a.	0.23	n.a.
Cl^-	mmol/L	0.09	121	321	242	273	327	321	22.19	0.18
SO_4^{2-}	mmol/L	0.97	6.94	15.34	16.03	12.29	16.79	15.63	3.01	0.49
U_{total}	nmol/L	1.22	2.52	0.63	n.a.	2.28[c]	n.a.	n.a.	0.04	n.a.
pCO_2	bar	$10^{-1.7}$	$10^{-2.3}$	$10^{-2.2}$	$10^{-2.4}$	$10^{-2.8}$	$10^{-1.9}$	$10^{-2.5}$	$10^{-2.3}$	$10^{-1.9}$
I[d]	mol/L	0.02	0.16	0.23	0.28	0.32	0.38	0.39	0.05	0.01
Distance[e]	m	0	50	65	85	105	135	165	176	210

n.a. = no data available; OPA = Opalinus Clay; [a] Data from Pearson et al. [1]; [b] Data from Wersin et al. [2]; [c] Data from Vinsot et al. [3]; [d] calculated in PHREEQC; [e] Distance from contact between Dogger limestone and Opalinus Clay

Supplementary material S2

The solubility of calcium increases in solutions containing magnesium compared to the pure mineral due to the exchange of calcium with magnesium ions on the surface of the calcite lattice [4]. Therefore, the stability constant of calcite given in the PSI/Nagra thermodynamic database Version 12/07 [5]:

Calcite (pure)

CaCO3	=	+1.000Ca+2	-1.000H+	+1.000HCO3-	logk	1.8490

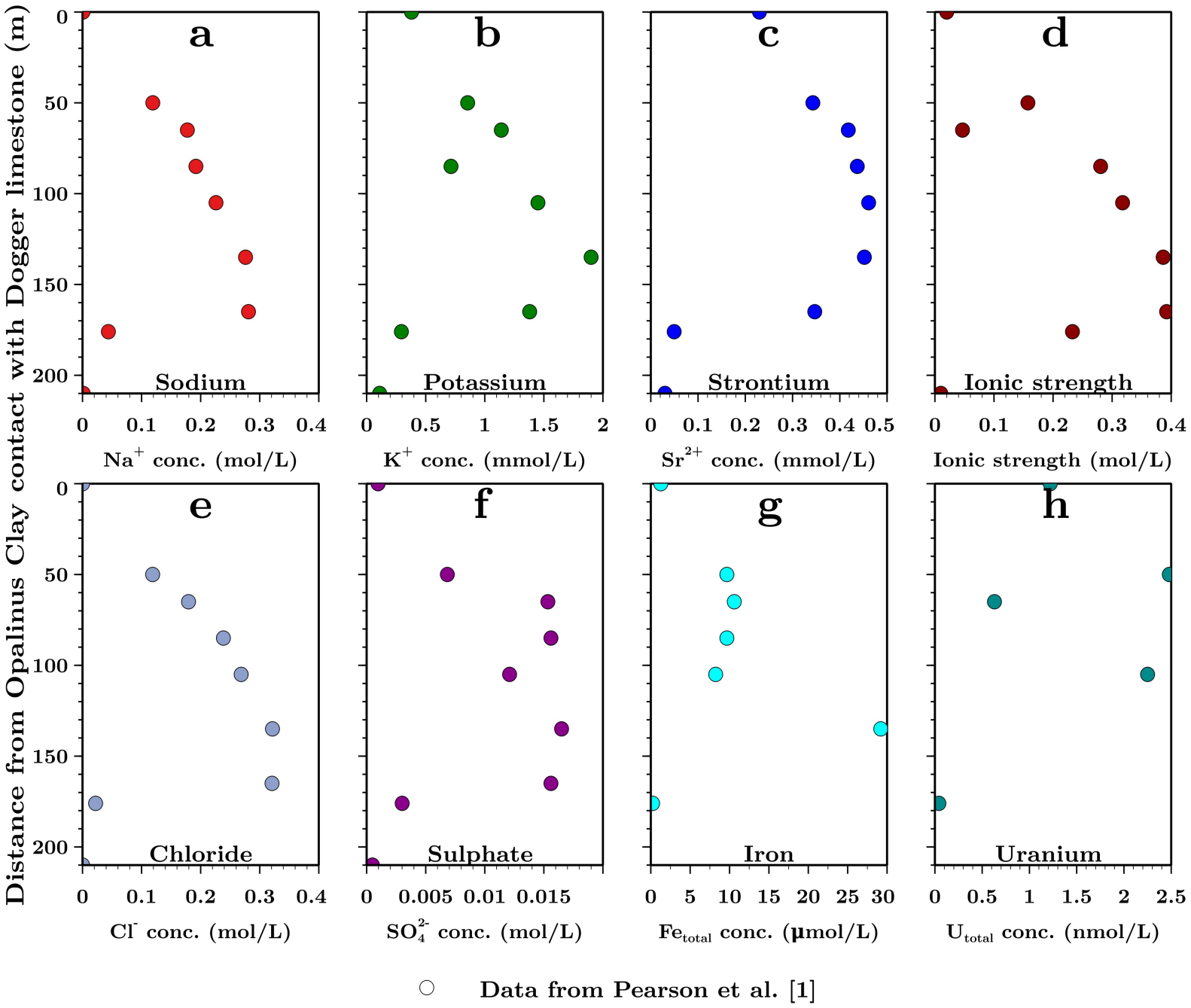
is adapted based on the Mg^{2+}/Ca^{2+} ratio in the pore waters of the Opalinus Clay formation at Mont Russelin representing old chemical signatures [6,7]. According to the approach presented in Möller and De Lucia [4], the new stability constant for calcite is about 0.2 log units higher:

MgCalcite

CaCO3	=	+1.000Ca+2	-1.000H+	+1.000HCO3-	logk	2.01672

Supplementary material S3

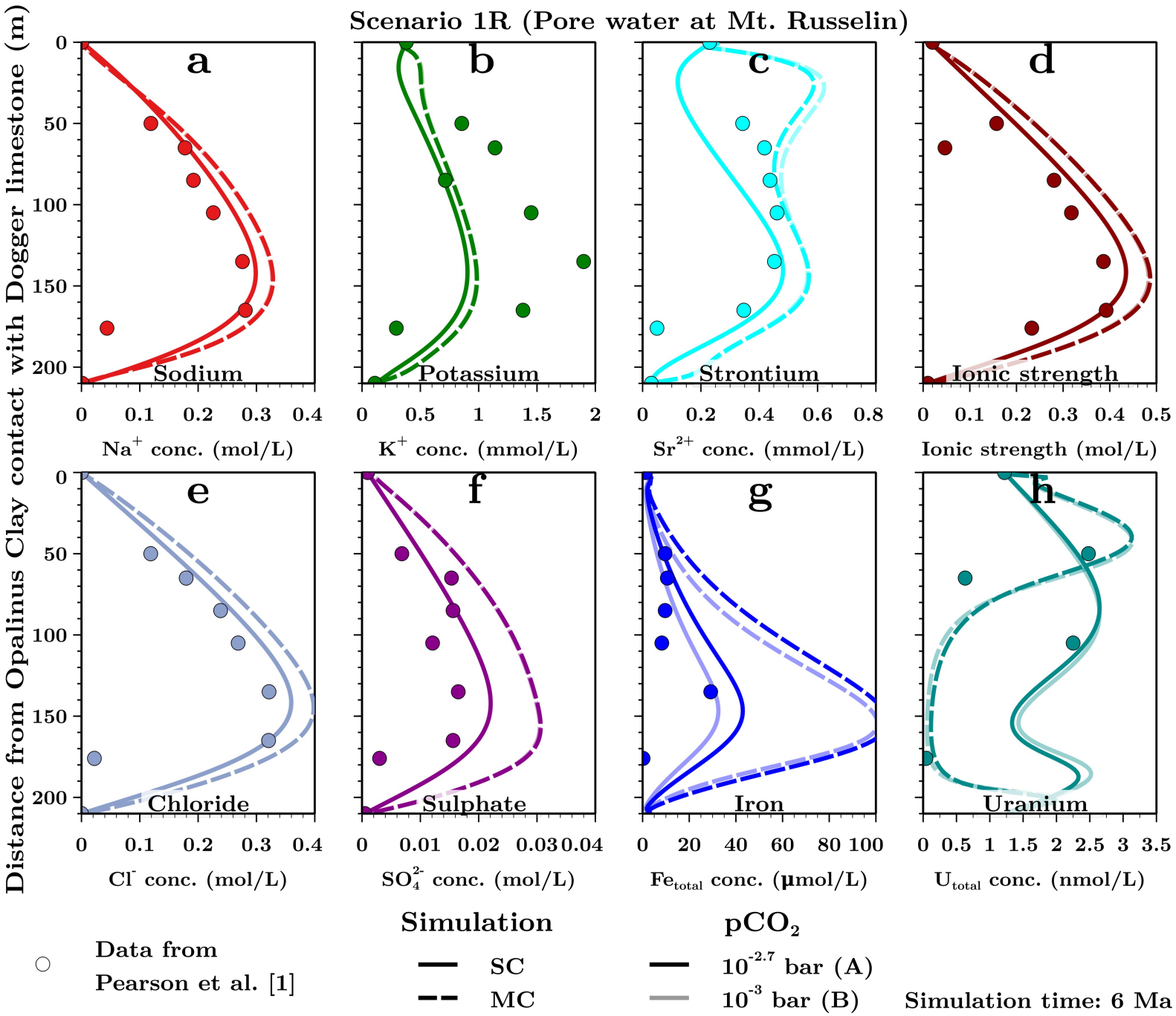
Figure S3.
Concentration profiles of pore water components used for model calibration. The data stems from borehole analysis at the underground laboratory Mont Terri. The y-axis is the distance perpendicular to the contact between Opalinus Clay and Dogger limestones.



Supplementary material S4

Figure S4.

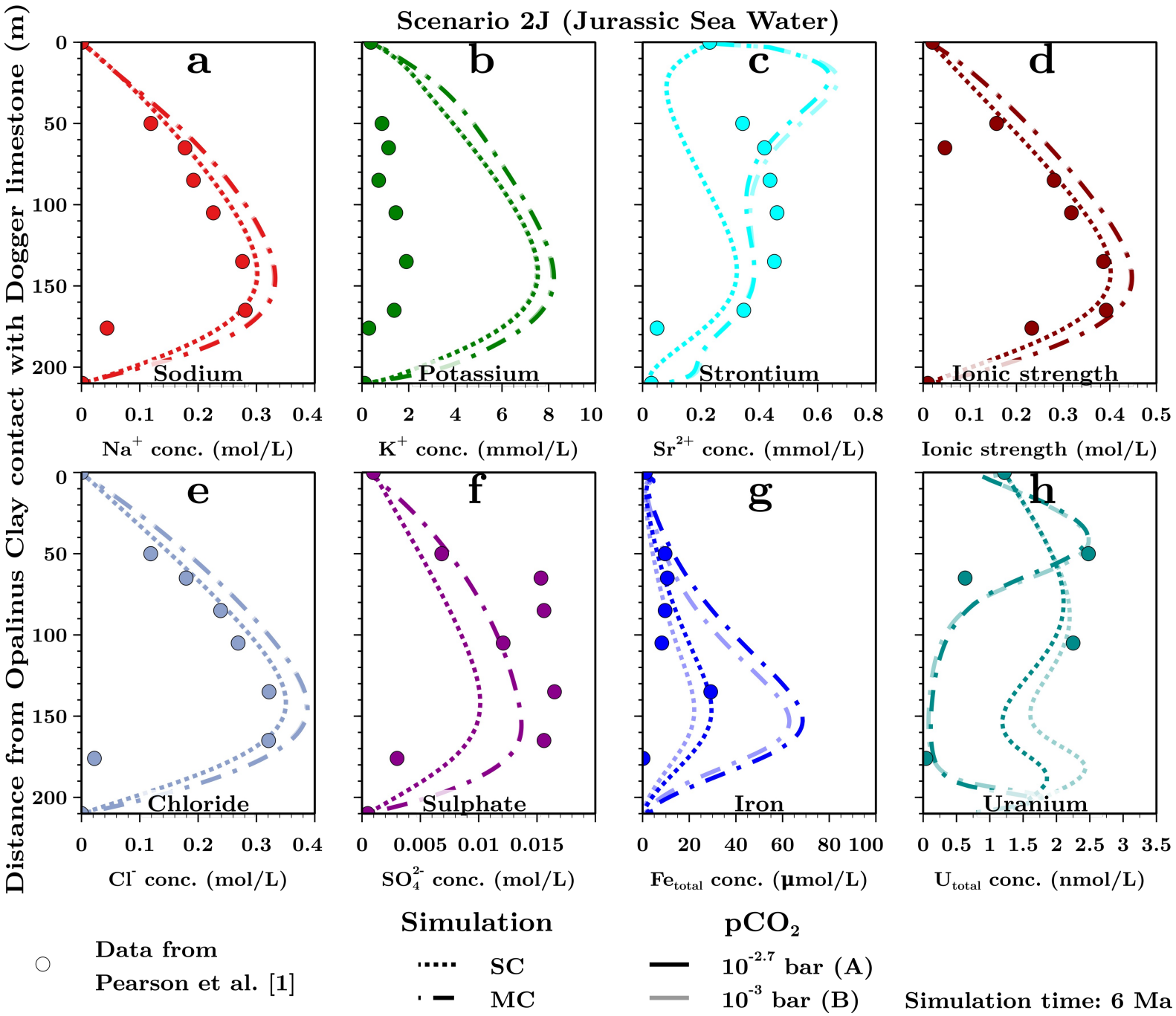
Modelled concentration profiles of SC simulations coincide with measured data using initial pore water composition according to scenario 1R. With MC diffusion, transport towards the embedding aquifers is decreased leading to higher concentrations, especially for sulphate and iron.



Supplementary material S5

Figure S5.

Modelled concentration profiles of SC simulations coincide with measured data using initial pore water composition according to scenario 2J, except for potassium and sulphate. With MC diffusion, transport towards the embedding aquifers is decreased leading to higher concentrations, especially for potassium and iron.



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