minerals

MDPI

Supplementary data

Ex and In Situ Reactivity and Sorption of Selenium in Opalinus Clay in the Presence of a Selenium Reducing Microbial Community

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Rarefaction curves

A DNA-based analysis of the microbial community in the sampled solutions from Interval 3 was performed. For this, DNA was extracted and a high-throughput amplicon sequencing of the V3 – V4 hypervariable region of the 16S rRNA gene was performed with the Illumina® MiSeq platform. The number of individual OTUs found in each sample is shown in the rarefaction curves (Figure S1).

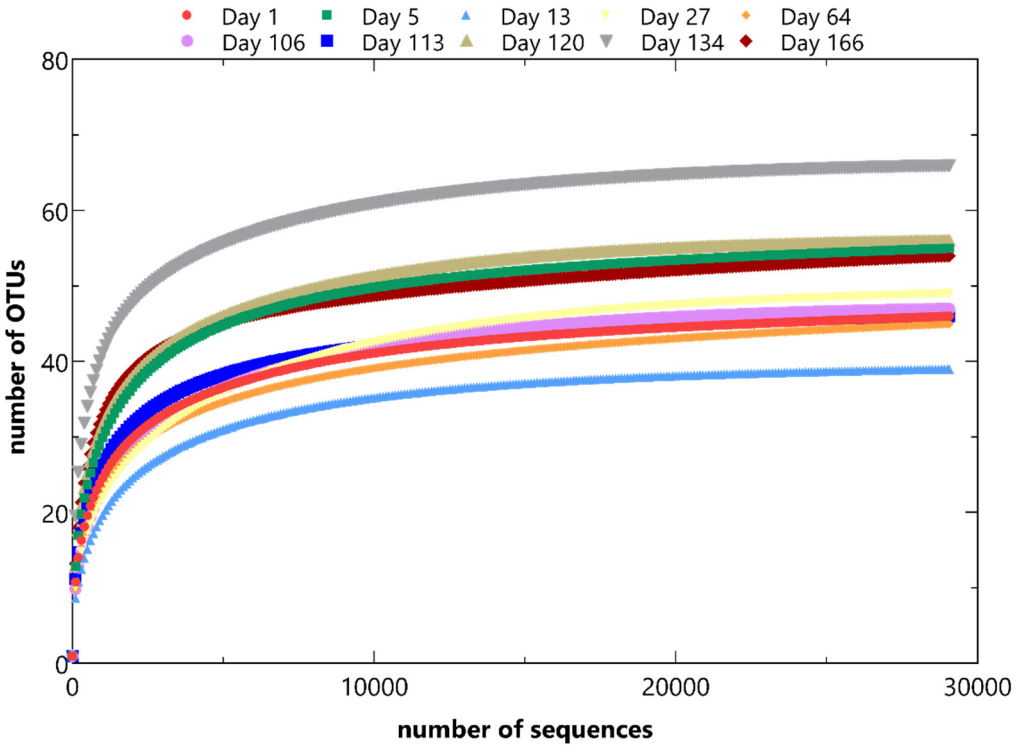


Figure S1. Rarefaction curves for the Illumina® 16S rDNA gene amplicon sequencing data showing for each sample the number of OTUs in function of the number of reads.

Fitting of Br^- and HDO diffusion from Intervals 1 and 2

In 2011, tracer diffusion tests with Br^- and HDO were performed in Intervals 1 and 2 of the BN experiment [23]. In the present study, the Br- and HDO data was fitted using the PHREEQC radial diffusion model by pore diffusion coefficients (Dp) of 3.0×10^{-11} m^2 s^{-1} and 1.8×10^{-10} m^2 s^{-1} respectively assuming a total porosity of 0.17 for both Br^- and HDO, as shown in Figure S2.

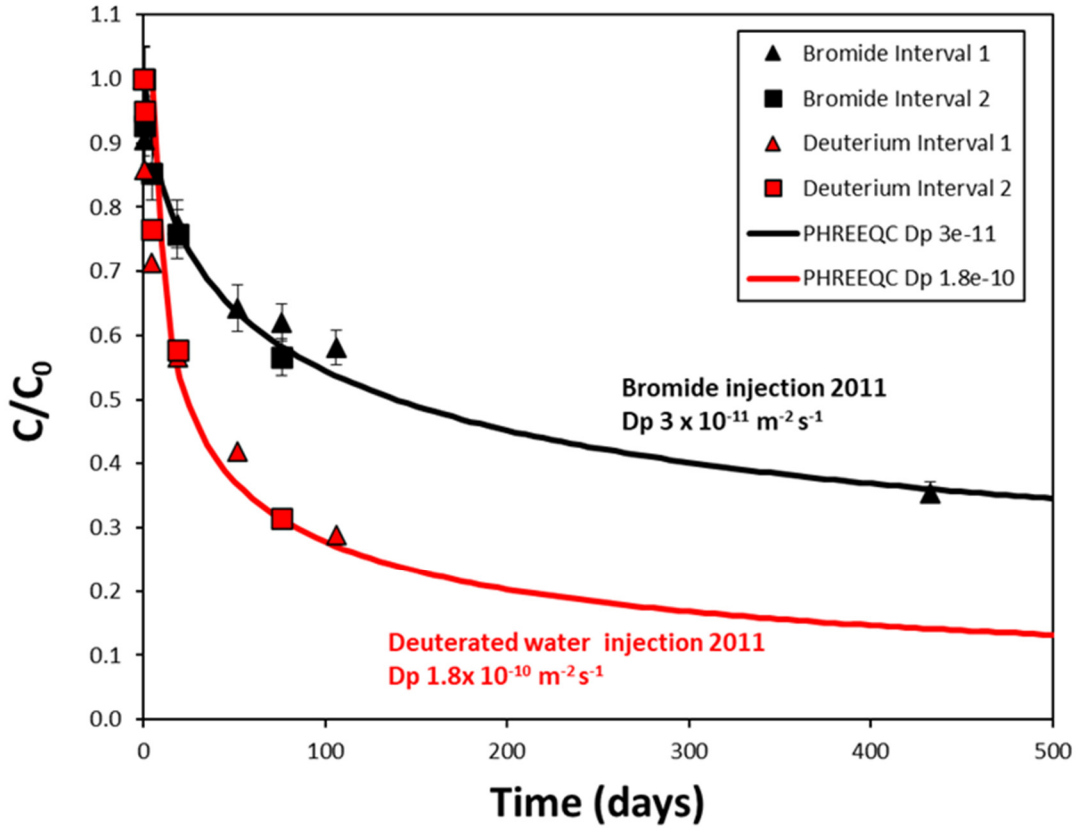


Figure S2. PHREEQC radial diffusion model fitted to bromide and deuterium diffusion tests in Interval 1 and Interval 2 of the BN experiment assuming a porosity of 0.17 for both tracer species [23].

SEM-EDX studies

Scanning Electron Microscopy coupled with Energy Dispersive X-ray spectroscopy (SEM-EDX) was performed on the filter pieces and Opalinus clay pieces added to the abiotic batch tests (Table 2). The samples were analyzed with a Phenom ProX table top SEM equipped with an EDX element identification probe (Benelux Scientific, Belgium). The results are shown in Figure S3.

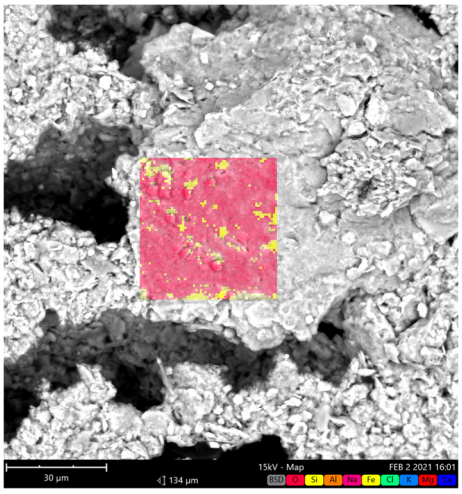
Three types of solid phases are analyzed by SEM-EDX:

- Stainless steel filter piece: filter pieces removed from the suspensions and dried in anaerobic conditions under argon atmosphere in an anaerobic glove box.
- Clay captured on filter: Suspended clay captured by transferring the suspension through a syringe over a Whatman® Nuclepore Track-Echted membrane (Sigma-Aldrich, Belgium) placed in a Swinnex holder (EMD Millipore, Germany).
- Intact clay piece: larger pieces of clay that were retrieved.

For EDX analysis, disabled elements were: Bi, Br, Dy, I, Re, Sb, Sr, Te, Ti, V, W, the equipment was set to 15 kV in map mode, with BDS detector. Both spot analysis and EDX mapping was performed.

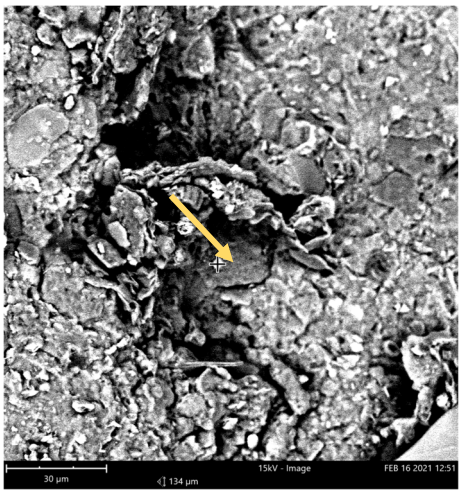
Ab_Se(VI)-C

Clay Captured on Filter



Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
O	69.47	55.69
Si	13.57	19.10
Al	9.05	12.23
Na	2.54	2.92
Fe	0.96	2.70
Cl	1.38	2.45
K	1.05	2.06
Mg	1.44	1.75
Ca	0.54	1.09

Intact Clay Piece



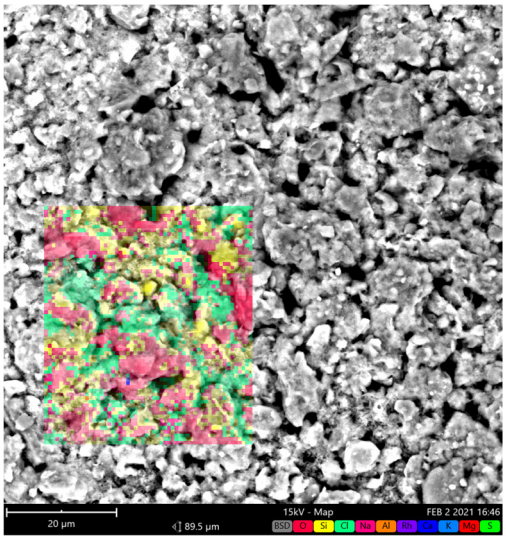
Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
Fe	19.69	44.20
O	40.70	26.17
C	24.53	11.84
Si	6.73	7.60
Al	5.27	5.72
Ca	1.80	2.89
K	0.58	0.91
Mg	0.68	0.67

Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
O	77.70	64.99
Si	18.73	27.50
Al	2.81	3.97
Sr	0.73	3.33

Figure S3. *Cont.*

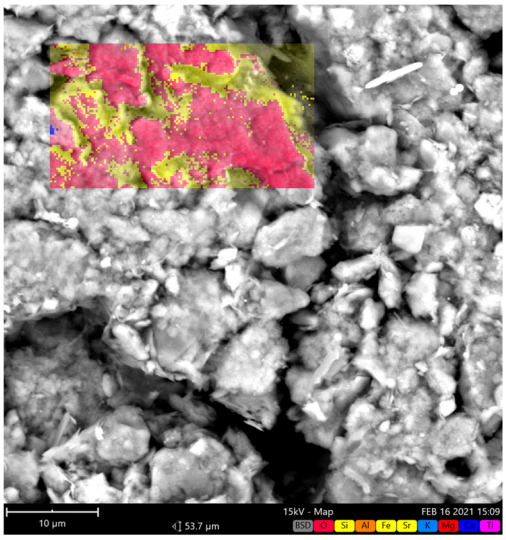
Ab_Se(IV)-C

Clay Captured on Filter



Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
O	59.33	43.72
Si	10.59	13.70
Cl	8.10	13.23
Na	10.27	10.87
Al	6.90	8.57
Rh	0.78	3.69
Ca	1.13	2.09
K	0.93	1.68
Mg	1.29	1.45
S	0.68	1.00

Intact Clay Piece

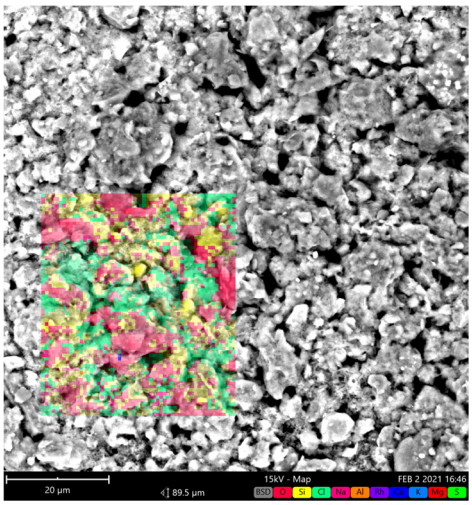


Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
O	69.99	54.50
Si	15.10	20.64
Al	9.76	12.82
Fe	1.33	3.60
Sr	0.80	3.40
K	1.12	2.13
Mg	1.12	1.33
Ca	0.65	1.27

Figure S3. *Cont.*

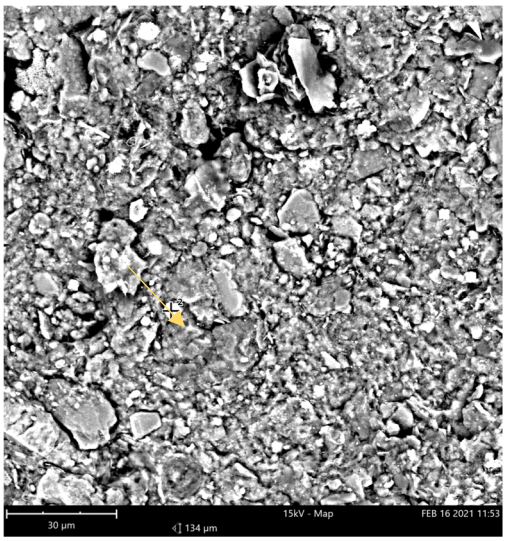
Ab_Se(VI)-CF

Clay Captured on Filter

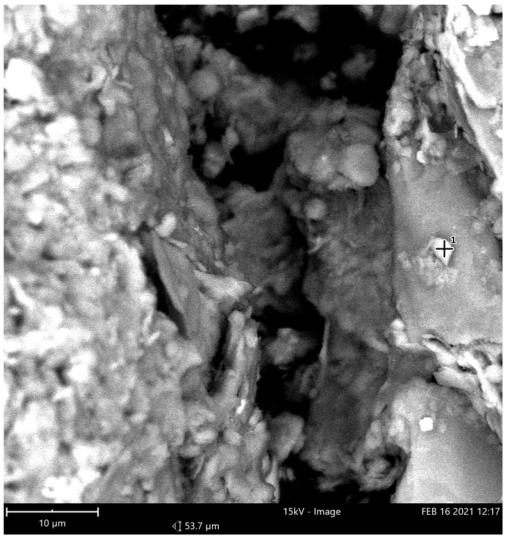


Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
O	59.33	43.72
Si	10.59	13.70
Cl	8.10	13.23
Na	10.27	10.87
Al	6.90	8.57
Rh	0.78	3.69
Ca	1.13	2.09
K	0.93	1.68
Mg	1.29	1.45
S	0.68	1.00

Intact Clay Piece



Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
O	62.62	38.17
Fe	13.60	28.94
S	15.97	19.52
Br	2.22	6.76
Si	4.83	5.17
Te	0.19	0.93
Mg	0.57	0.52

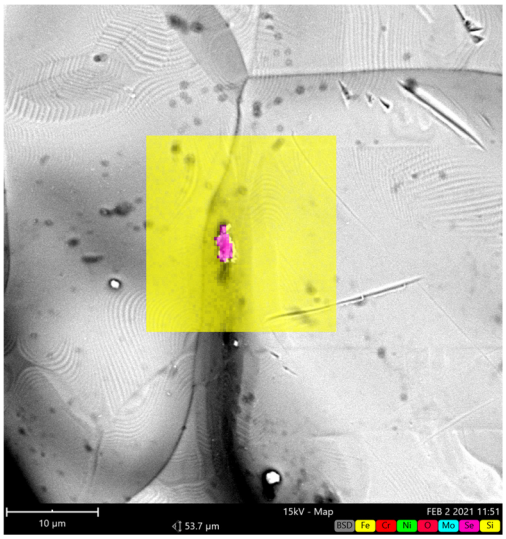


Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
O	75.04	63.32
Si	12.05	17.85
Al	11.14	15.85
K	0.85	1.76
Mg	0.89	1.14

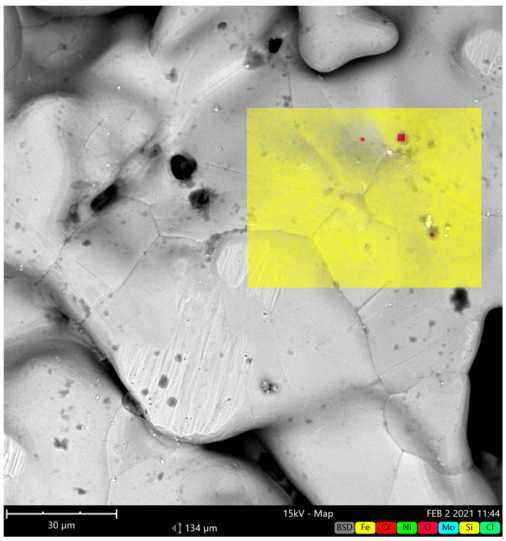
Figure S3. *Cont.*

Ab_Se(VI)-CF

Stainless Steel Filter Piece



Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
Fe	54.10	60.95
Cr	15.42	16.17
Ni	10.27	12.16
O	15.71	5.07
Mo	1.70	3.30
Se	0.74	1.18
Si	2.06	1.16

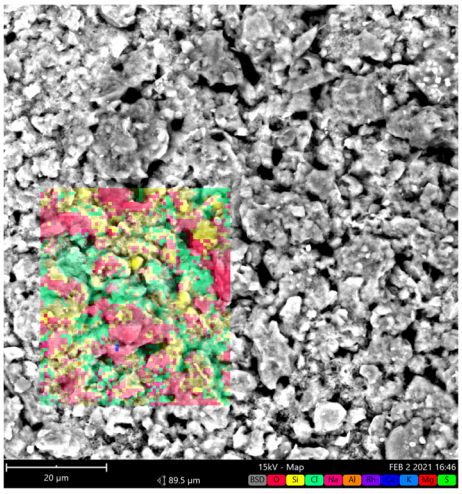


Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
Fe	57.88	63.50
Cr	16.32	16.68
Ni	10.37	11.95
O	11.62	3.65
Mo	1.54	2.91
Si	1.85	1.02

Figure S3. *Cont.*

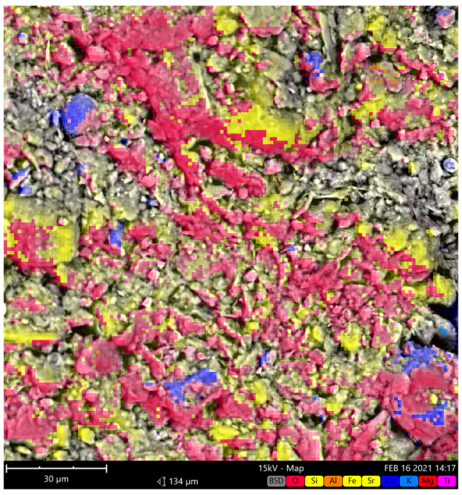
Ab_Se(IV)-CF

Clay Captured on Filter



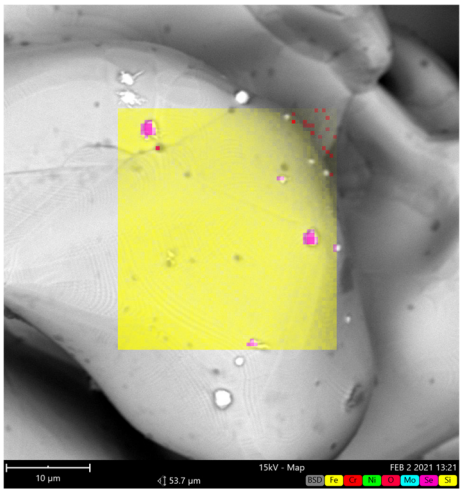
Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
O	59.33	43.72
Si	10.59	13.70
Cl	8.10	13.23
Na	10.27	10.87
Al	6.90	8.57
Rh	0.78	3.69
Ca	1.13	2.09
K	0.93	1.68
Mg	1.29	1.45
S	0.68	1.00

Intact Clay Piece



Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
O	69.94	53.82
Si	15.01	20.27
Al	8.54	11.08
Fe	1.70	4.57
Sr	0.78	3.29
Ca	1.52	2.93
K	1.24	2.33
Mg	1.06	1.24

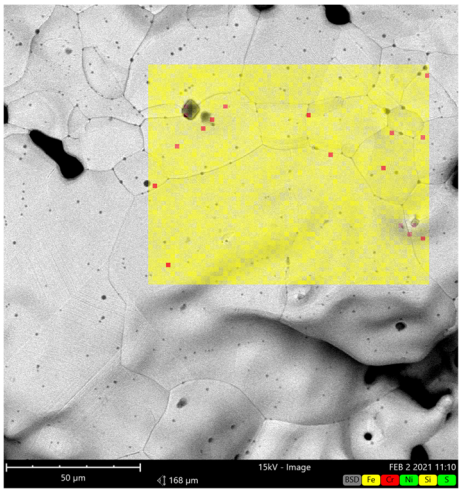
Stainless Steel Filter Piece



Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
Fe	56.42	62.02
Cr	16.60	16.99
Ni	10.09	11.66
O	12.44	3.92
Mo	1.46	2.76
Se	0.99	1.54
Si	1.99	1.10

Figure S3. *Cont.*

Ab_Se(VI)-F
Stainless Steel Filter Piece

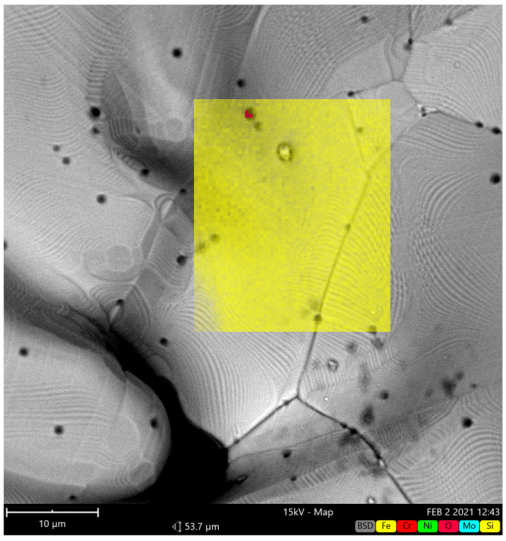


Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
Fe	65.76	67.22
Cr	18.65	17.75
Ni	12.33	13.24
Si	1.85	0.95
S	1.42	0.83

Figure S3. *Cont.*

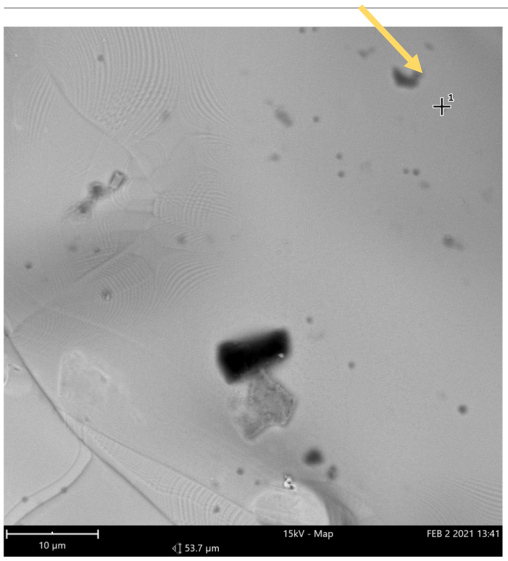
Ab_Se(IV)-F

Stainless Steel Filter Piece



Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
Fe	54.59	61.45
Cr	16.43	17.21
Ni	10.23	12.10
O	15.01	4.84
Mo	1.66	3.22
Si	2.08	1.18

Stainless Steel Filter Piece



Element Symbol	Atomic Conc. (%)	Weight Conc. (%)
Fe	63.00	66.41
Cr	17.96	17.63
Ni	10.34	11.46
O	6.62	2.00
Mo	1.09	1.97
Si	1.00	0.53

Figure S3. Overview of the SEM-EDX images and data of stainless steel and clay samples from the abiotic batch tests. The test codes are indicated on the top of each page and correspond to Table 2.

Equilibrium between Se(0) and aqueous Se(IV) species

PHREEQC calculations were performed to determine the aqueous concentration of Se(IV) in equilibrium with Se(0), as shown in Figure S4.

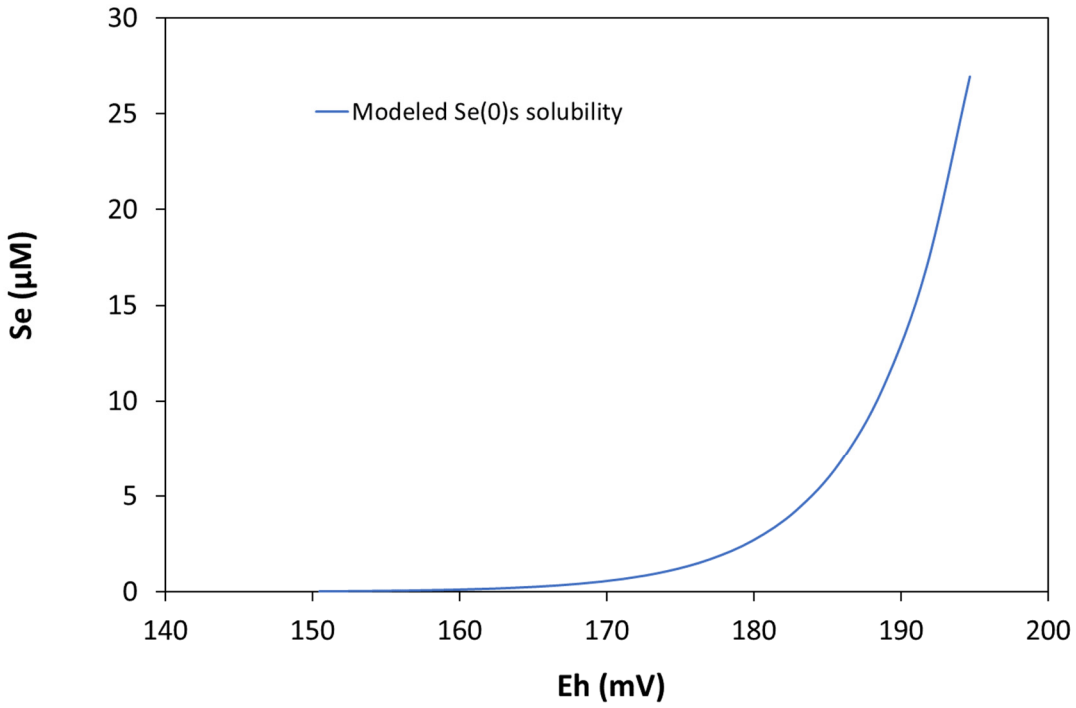


Figure S4. Modeled solubility of Se(0)s in equilibrium with Se(IV) as a function of Eh, at pH 7.