Supporting Information to: A Holistic Approach to Multicomponent EXAFS: Sr and Cs Complexation in Clayey Soils

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Electronic Supplementary Information with 14 pages, 15 tables and 4 figures.

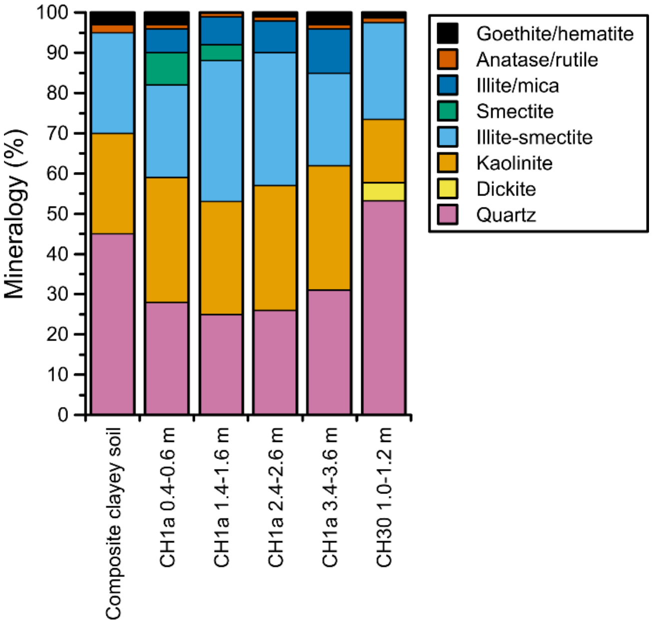

Fig. S1 Summary of the mineralogical composition of samples used for the adsorption experiments on mixed phase solids (Fig. 1 and 2), this includes the composite clayey soil sample and the Little Forest Legacy Site core samples from two site locations: CH1a[1] and CH30, and their respective depth interval[2, 3]

Table S1 Summary of the variables used during the shell by shell fitting strategy to obtain information on the surface species of Sr.

	Anatase pH 5	**Anatase pH 7**	**Anatase pH 8**	**Illite-smectite pH 5**	**Illite-smectite pH 7**	**Illite-smectite pH 8**	**Kaolinite pH 5**	**Kaolinite pH 7**	**Kaolinite pH 8**	**Soil pH 5**	**Soil pH 7**	**Soil pH 8**
$S_0^{2\#}$	1	1	1	1	1	1	1	1	1	1	1	1
ΔE_0*	E0ana5	E0ana7	E0ana8	E0ill5	E0ill7	E0ill8	E0kao5	E0kao7	E0kao8	E0soil5	E0soil7	E0soil8
		Sr-O			**Sr-O**			**Sr-$O_{(1)}$**			**Sr-O**	
C.N.*	CNOana5	CNOana7	CNOana8	CNOill5	CNOill7	CNOill8	CNO1kao5	CNO1kao7	CNO1kao8	CNOsoil5	CNOsoil7	CNOsoil8
R*	ROana5	ROana7	ROana8	ROill5	ROill7	ROill8	RO1kao5	RO1kao7	RO1kao8	ROsoil5	ROsoil7	ROsoil8
σ*	ssOana5	ssOana7	ssOana8	ssOill5	ssOill7	ssOill8	*ssOkao*	*ssOkao*	*ssOkao*	ssOsoil5	ssOsoil7	ssOsoil8
		Sr-Ti			**Sr-Si**			**Sr-$O_{(2)}$**			**Sr-Si**	
C.N.*	CNTiana5	CNTiana7	CNTiana8	CNSiill5	CNSiill7	CNSiill8	CNO2kao5	CNO2kao7	CNO2kao8	CNSisoil5	CNSisoil7	CNSisoil8
R*	*RTiana*	*RTiana*	*RTiana*	*RSiill*	*RSiill*	*RSiill*	*RO2kao*	*RO2kao*	*RO2kao*	*RSisoil*	*RSisoil*	*RSisoil*
σ*	*ssTiana*	*ssTiana*	*ssTiana*	*ssSiill*	*ssSiill*	*ssSiill*	*ssOkao*	*ssOkao*	*ssOkao*	*ssSisoil*	*ssSisoil*	*ssSisoil*
								Sr-Si				
C.N.*							CNSikao5	CNSikao7	CNSikao8			
R*							*RSikao*	*RSikao*	*RSikao*			
σ*							*ssSikao*	*ssSikao*	*ssSikao*			

The amplitude correction factor (S_0^2) was fixed to 1 to enable the determination of the coordination numbers for the Sr scattering paths.

* C.N. represents the coordination number (fixed, the errors on the C.N. are estimated to be ~25%), ΔE_0 the energy shift, R the radial distance and σ^2 the Debye-Waller factor, the variables with the same name (in italic) were fitted simultaneously to the same value

Table S2 Example of the holistic fitting strategy (Sr at pH 7) and the respective variables used to determine the speciation within the composite clayey soil samples

	Anatase pH 7	Illite-smectite pH 7	Kaolinite pH 7	Soil pH 7
$S_0^{2\#}$	1	1	1	1
ΔE_0*	E0ana	E0ill	E0kao	**E0soil~**
Anatase				
Sr-O				
C.N.*	8.7			8.7·$\boldsymbol{f_{Anatase}}$~
R*	*ROana*			*ROana*
σ²*	*ssOana*			*ssOana*
Sr-Ti				
C.N.*	0.7			0.7·$\boldsymbol{f_{Anatase}}$~
R*	*RTiana*			*RTiana*
σ*	*ssTiana*			*ssTiana*
Illite-smectite				
Sr-O				
C.N.*		9.3		9.3·$\boldsymbol{f_{Illite}}$~
R*		*ROill*		*ROill*
σ²*		*ssOill*		*ssOill*
Sr-Si				
C.N.*		0.9		0.9·$\boldsymbol{f_{Illite}}$~
R*		*RSiill*		*RSiill*
σ²*		*ssSiill*		*ssSiill*
Kaolinite				
Sr-O(1)				
C.N.*			8.9	8.9·$\boldsymbol{f_{Kaolinite}}$~
R*			*RO1kao*	*RO1kao*
σ²*			*ssO1kao*	*ssO1kao*
Sr-O(2)				
C.N.*			3.1	3.1·$\boldsymbol{f_{Kaolinite}}$~
R*			*RO2kao*	*RO2kao*
σ²*			*ssO2kao*	*ssO2kao*
Sr-Si				
C.N.*			5.7	5.7·$\boldsymbol{f_{Kaolinite}}$~
R*			*RSikao*	*RSikao*
σ²			*ssSikao*	*ssSikao*

* C.N. represents the coordination number (fixed, the errors on the C.N. are estimated to be ~25%), ΔE_0 the energy shift, R the radial distance and σ^2 the Debye-Waller factor, the amplitude correction factor (S_0^2) were fixed, and the variables with the same name (in italic) were fitted simultaneously to the same value. C.N. were fixed to the results from the shell-by-shell fits (Table S10)

~ The variables in bold highlight the unique variables required to fit the EXAFS of the soil samples using this holistic fitting strategy and represent the fraction of Sr adsorbed to anatase ($f_{Anatase}$), to illite-smectite (f_{Illite}) and to kaolinite ($f_{Kaolinite}$)

Table S3 Summary of the variables used during the shell-by-shell and holistic (Cs at pH 7) fitting strategy to obtain information on the speciation within the composite clayey samples.

Shell-by-shell	**Illite-smectite pH 7**	**Kaolinite pH 7**	**Soil pH 7**
$S_0^{2\#}$	1	1	1
ΔE_0	E0ill7	E0kao7	E0soil7
Cs-O(1)			
C.N.#	8	7	8
R (Å)	RO1ill7	RO1kao7	RO1soil7
σ (Å²)*	*ssOill7*	*ssOkao7*	*ssOsoil7*
Cs-O(2)			
C.N.#	4	5	4
R (Å)	RO2ill7	RO2kao7	RO2soil7
σ (Å²)*	*ssOill7*	*ssOkao7*	*ssOsoil7*
Cs-Si			
C.N.#	5	6	5.5
R (Å)	RSiill7	RSikao7	RSisoil7
σ (Å²)	ssSiill7	ssSikao7	ssSisoil7
Holistic	**Illite-smectite pH 7**	**Kaolinite pH 7**	**Soil pH 7**
$S_0^{2\#}$	1	1	1
ΔE_0	E0ill7	E0kao7	**E0soil7~**
Illite-smectite			
Cs-O(1)			
C.N.*	8		7*$\boldsymbol{f_{Illite}}$~
R (Å)*	*RO1ill7*		*RO1kao7*
σ (Å²)*	*ssOill7*		*ssOkao7*
Cs-O(2)			
C.N.#	4		5*$\boldsymbol{f_{Illite}}$~
R (Å)*	*RO2ill7*		*RO2kao7*
σ (Å²)*	*ssOill7*		*ssOkao7*
Cs-Si			
C.N.*	5		6*$\boldsymbol{f_{Illite}}$~
R (Å)*	*RSiill7*		*RSikao7*
σ (Å²)*	*ssSiill7*		ssSikao7
Kaolinite			
Cs-O(1)			
C.N.*		7	7*(1-$\boldsymbol{f_{Illite}}$)~
R (Å)*		*RO1kao7*	*RO1kao7*
σ (Å²)*		*ssOkao7*	*ssOkao7*
Cs-O(2)			
C.N.*		5	5*(1-$\boldsymbol{f_{Illite}}$)~
R (Å)*		*RO2kao7*	*RO2kao7*
σ (Å²)*		*ssOkao7*	*ssOkao7*
Cs-Si			
C.N.*		6	6*(1-$\boldsymbol{f_{Illite}}$)~
R (Å)*		*RSikao7*	*RSikao7*
σ (Å²)*		*ssSikao7*	ssSikao7

The amplitude correction factor (S_0^2) was fixed to 1.

* C.N. represents the coordination number (fixed, the errors on the C.N. are estimated to be ~25%), ΔE_0 the energy shift, R the radial distance and σ^2 the Debye-Waller factor, the variables with the same name (in italic) were fitted simultaneously to the same value and the C.N. were fixed based on the C.N. for O and Cs were fixed based on Molecular Dynamics for Cs complexation at the Si vacancy sites of the basal plane of Illite[4] or kaolinite[5] and EXAFS analyses on Cs interaction with a clayey soil.[6]

~ The variables in bold highlight the unique variables required to fit the EXAFS of the soil samples using this holistic fitting strategy and represent the fraction of Cs adsorbed to illite-smectite (f_{Illite}) and to kaolinite (1-f_{Illite})

Table S4 Summary of the results from the adsorption experiments at trace concentrations of Sr-85 and Cs-137 (at ~20 Bq/ml), including the solid/solution ratio (g/L), the measured initial and final pH, and Sr-85 and Cs-137 concentrations in solution (in Bq/ml), the % removed from solution (adsorbed), the Sr-85 and Cs-137 concentrations on the solids (in Bq/g), and the respective distribution coefficients (K_D, in ml/g). The description of the experiments includes the solid phase and the composition of the solutions, the graphical representation of the data is shown in Fig. 1.

				Strontium					Caesium				
Experiments	Solid (g/L)	pH_i	pH_{final}	Initial conc. (Bq/ml)	Final conc. (Bq/ml)	% adsorbed	Solid conc. (Bq/g)	K_D (ml/g)	Initial conc. (Bq/ml)	Final conc. (Bq/ml)	% adsorbed	Solid conc. (Bq/g)	K_D (ml/g)
Anatase: 20 Bq/ml Sr-85 and Cs-137	9.66	4.05	4.25	21.04	14.93	29.0	632	42	23.08	21.97	4.8	114	5
	9.67	5.04	5.45	20.96	8.03	61.7	1337	166	23.14	21.30	7.9	190	9
	9.70	5.98	6.26	20.95	2.27	89.2	1926	849	23.14	20.17	12.8	306	15
	9.67	7.04	7.08	20.92	0.75	96.4	2086	2774	23.12	20.52	11.3	270	13
	9.66	7.94	7.92	20.95	0.25	98.8	2143	8549	23.12	21.63	6.5	155	7
Illite-smectite: 20 Bq/ml Sr-85 and Cs-137	9.68	4.23	4.43	20.89	6.90	67.0	1446	210	23.13	1.80	92.2	2204	1224
	9.69	5.08	5.09	20.99	4.80	77.2	1672	349	23.12	1.26	94.5	2256	1784
	9.66	5.93	6.01	20.90	2.63	87.4	1891	720	23.11	2.10	90.9	2175	1033
	9.70	7.06	7.04	20.64	2.91	85.9	1827	628	23.15	2.61	88.7	2117	811
	9.65	8.45	7.83	20.98	2.67	87.3	1897	710	23.15	1.06	95.4	2288	2154
Kaolinite: 20 Bq/ml Sr-85 and Cs-137	9.70	4.27	4.45	20.98	17.18	18.1	392	23	23.19	10.18	56.1	1341	132
	9.70	4.94	5.14	20.99	13.08	37.7	816	62	23.15	5.34	76.9	1835	344
	9.66	5.87	6.43	20.97	11.28	46.2	1003	89	23.18	5.03	78.3	1878	374
	9.68	7.06	7.31	20.94	10.54	49.7	1075	102	23.14	4.39	81.0	1938	441
	9.69	8.04	8.03	20.97	10.89	48.1	1041	96	23.07	5.21	77.4	1844	354
Soil: 20 Bq/ml Sr-85 and Cs-137	9.73	4.33	4.45	22.78	11.71	48.6	1138	97	23.53	1.11	95.3	2306	2077
	9.75	5.14	5.14	22.92	10.00	56.3	1324	132	23.51	1.72	92.7	2235	1303
	9.75	6.09	5.60	22.89	8.30	63.7	1496	180	23.50	3.13	86.7	2089	668
	9.76	7.04	7.00	22.90	5.75	74.9	1758	306	23.55	1.21	94.9	2290	1891
	9.79	8.28	7.80	22.92	7.02	69.4	1623	231	23.55	2.02	91.4	2199	1090

Table S5 Summary of the results from the adsorption experiments at elevated concentrations of Sr and Cs (at 25 µg/ml [ppm]), including the solid/solution ratio (g/L), the measured initial and final pH, and Sr and Cs concentrations in solution (in µg/ml [ppm]), the % removed from solution (adsorbed), the Sr and Cs concentrations on the solids (in µg/g), and the respective distribution coefficients (K_D, in ml/g). The description of the experiments includes the solid phase and the composition of the solutions, the graphical representation of the data is shown in Fig. 1.

				Strontium					Caesium				
Experiments	Solid (g/L)	pH_i	pH_{final}	Initial conc. (µg/ml)	Final conc. (µg/ml)	% adsorbed	Solid conc. (µg/g)	K_D (ml/g)	Initial conc. (µg/ml)	Final conc. (µg/ml)	% adsorbed	Solid conc. (µg/g)	K_D (ml/g)
Anatase: 25 ppm Sr and Cs	9.92	3.95	3.91	23.7	22.9	3.2	81	3	24.1	23.7	1.8	40	2
	9.94	5.09	4.79	23.7	21.3	10.2	241	11	24.2	23.6	2.4	60	2
	9.95	5.95	5.49	23.8	19.5	18.0	432	22	24.2	23.6	2.5	60	3
	9.94	6.98	6.50	23.7	15.3	35.7	845	56	24.2	23.3	3.4	91	4
	9.95	7.81	7.16	23.7	10.9	54.2	1286	119	24.1	23.3	3.2	80	3
Illite-smectite: 25 ppm Sr and Cs	9.93	4.05	4.02	23.7	12.8	45.9	1098	85	24.1	17.5	27.5	665	38
	9.92	5.00	4.80	23.7	11.0	53.6	1280	117	24.2	16.8	30.4	746	44
	9.89	5.94	5.69	23.6	6.3	73.2	1749	276	24.0	14.3	40.5	981	69
	9.87	6.68	6.55	23.6	5.8	75.4	1803	310	24.0	13.5	43.9	1064	79
	9.84	7.08	6.67	25.0	6.4	74.5	1890	296	25.0	13.4	46.5	1179	88
	9.92	7.54	7.25	23.7	5.5	76.9	1835	335	24.1	13.2	45.1	1099	83
Kaolinite: 25 ppm Sr and Cs	9.92	3.96	4.02	23.7	23.1	2.3	60	2	24.1	20.7	14.1	343	17
	9.94	4.93	4.82	23.7	21.0	11.7	272	13	24.2	19.1	20.9	513	27
	9.95	5.90	5.88	23.8	20.0	15.8	382	19	24.2	18.6	23.1	563	30
	10.00	7.19	6.94	23.8	19.9	16.3	390	19	24.2	18.6	23.2	560	30
	9.91	6.94	6.79	25.2	20.3	19.2	494	24	25.2	18.6	26.2	666	36
	9.92	8.07	7.58	23.7	19.5	17.9	423	22	24.2	18.3	24.1	595	32
Goethite 25 ppm Sr and Cs*	9.98	3.92	3.97	23.7	23.9	-0.9%	-22	-1	24.1	24.0	0.7%	18	1
	9.88	5.03	5.04	23.7	23.5	1.0%	25	1	24.1	23.6	2.0%	49	2
	9.88	5.90	5.87	23.7	22.3	5.9%	141	6	24.1	22.9	5.2%	127	6
	9.95	7.16	7.01	23.6	22.5	4.7%	111	5	24.1	24.4	-1.3%	-31	-1
	9.94	8.11	7.82	23.6	22.4	5.0%	119	5	24.0	23.4	2.6%	63	3
Quartz 25 ppm Sr and Cs*	9.91	3.89	3.89	23.7	25.3	-6.7%	-161	-6	24.1	25.0	-3.7%	-91	-4
	9.90	4.87	4.97	23.8	25.3	-6.4%	-154	-6	24.2	25.0	-3.6%	-88	-4
	9.89	6.07	6.09	23.7	25.6	-7.6%	-183	-7	24.2	25.5	-5.5%	-133	-5
	9.99	7.08	6.86	23.7	24.4	-2.9%	-68	-3	24.2	24.6	-1.7%	-41	-2
	9.96	7.62	7.29	23.7	24.8	-4.6%	-110	-4	24.1	25.2	-4.2%	-102	-4
Soil: 25 ppm Sr and Cs	9.92	4.09	4.11	23.7	21.2	10.3	252	12	24.1	21.7	9.9	242	11
	9.97	5.09	4.88	23.8	17.3	27.3	652	38	24.2	20.2	16.3	401	20
	9.95	6.21	6.07	23.7	14.8	37.6	894	61	24.1	19.9	17.5	422	21
	9.95	6.90	6.76	23.7	14.2	40.1	955	67	24.1	19.5	19.3	462	24
	9.73	7.24	7.03	24.7	16.4	33.7	853	52	24.7	19.8	19.7	504	25
	9.90	7.88	7.49	23.6	12.7	46.4	1101	87	24.1	18.5	23.3	566	31

* The Sr and Cs adsorption experiments to goethite and quartz were performed during a set of preliminary experiments to develop the appropriate experimental procedures, as such the errors on the aqueous analyses (and consequently on the calculated values: % adsorbed, solid concentration and K_D values) are larger compared to results from the other adsorption experiments.

Table S6 Summary of the results from the adsorption experiments at elevated concentrations of Sr (at 25 µg/ml [ppm]), including the solid/solution ratio (g/L), the measured initial and final pH, and Sr concentrations in solution (in µg/ml [ppm]), the % removed from solution (adsorbed), the Sr concentrations on the solids (in µg/g), and the respective distribution coefficients (K_D, in ml/g). The description of the experiments includes the solid phase and the composition of the solutions, the values in bold represent the samples used for X-ray Absorption Spectroscopy analyses and the graphical representation of the data is shown in Fig. 1.

Experiments	Solid (g/L)	pH_i	pH_{final}	Strontium Initial conc. (µg/ml)	Final conc. (µg/ml)	% adsorbed	Solid conc. (µg/g)	K_D (ml/g)
Anatase: 25 ppm Sr	9.92	3.96	3.94	24.2	23.2	4.2	101	4
	9.97	**5.03**	**4.66**	**25.0**	**22.3**	**10.6**	**271**	**12**
	9.92	5.11	4.81	24.3	21.4	11.6	292	13
	9.95	5.99	5.50	24.3	19.7	18.9	462	23
	9.91	6.95	6.49	24.2	15.6	35.5	868	55
	9.93	**7.49**	**6.85**	**24.8**	**14.4**	**42.0**	**1047**	**73**
	9.92	7.83	7.17	24.2	12.1	49.9	1220	100
	9.85	**8.75**	**7.59**	**25.3**	**11.5**	**54.4**	**1401**	**121**
Illite-smectite: 25 ppm Sr	9.93	4.01	3.99	24.2	12.8	47.0	1148	89
	9.86	**5.02**	**4.77**	**24.7**	**10.6**	**56.9**	**1430**	**134**
	9.92	5.01	4.83	24.3	10.5	56.8	1391	132
	9.89	5.96	5.76	24.1	5.7	76.5	1860	329
	9.88	6.70	6.56	24.2	5.2	78.3	1923	365
	9.89	**7.25**	**6.74**	**24.7**	**5.3**	**78.6**	**1962**	**371**
	9.94	7.51	7.18	24.2	5.0	79.2	1932	383
	10.08	**8.60**	**7.86**	**25.9**	**4.5**	**82.5**	**2123**	**468**
Kaolinite: 25 ppm Sr	9.97	3.98	4.03	24.2	23.0	5.0	120	5
	9.91	4.92	4.88	24.2	18.3	24.5	595	33
	9.84	**5.05**	**4.89**	**24.6**	**20.8**	**15.6**	**386**	**19**
	9.95	5.87	5.89	24.3	19.0	21.9	533	28
	9.93	**7.01**	**6.85**	**24.8**	**19.3**	**22.2**	**554**	**29**
	9.97	6.93	6.85	24.3	18.8	22.6	552	29
	9.92	**7.99**	**7.33**	**25.6**	**19.2**	**24.7**	**645**	**33**
	9.94	7.70	7.99	24.3	18.6	23.4	573	31
Goethite 25 ppm Sr*	9.98	3.92	3.98	24.2	24.7	-2.0%	-50	-2
	9.90	5.03	5.05	24.2	22.0	9.3%	229	10
	9.90	5.90	5.88	24.2	24.8	-2.2%	-55	-2
	9.88	7.13	6.95	24.2	23.2	3.8%	92	4
	9.86	8.08	7.79	24.1	23.0	4.6%	113	5
Quartz 25 ppm Sr*	9.96	3.91	3.88	24.2	25.4	-4.9%	-120	-5
	10.00	4.89	4.96	24.3	25.4	-4.6%	-111	-4
	9.94	6.07	6.08	24.3	24.8	-2.2%	-53	-2
	9.91	7.01	6.86	24.2	24.8	-2.4%	-59	-2
	9.94	7.58	7.18	24.2	25.1	-3.6%	-89	-4
Soil: 25 ppm Sr	9.92	4.09	4.13	24.2	21.3	12.0	292	14
	9.92	5.06	4.87	24.3	16.2	33.3	817	50
	9.79	**5.11**	**4.90**	**24.5**	**17.2**	**29.8**	**746**	**43**
	9.93	6.16	6.06	24.2	13.3	45.0	1098	82
	9.92	6.94	6.84	24.2	13.6	43.8	1069	79
	9.85	**7.08**	**6.90**	**24.6**	**14.2**	**42.4**	**1056**	**75**
	9.93	7.82	7.46	24.2	12.8	47.0	1148	89
	9.68	**8.16**	**7.49**	**24.9**	**15.1**	**39.4**	**1012**	**67**

* The Sr adsorption experiments to goethite and quartz were performed during a set of preliminary experiments to develop the appropriate experimental procedures, as such the errors on the aqueous analyses (and consequently on the calculated values: % adsorbed, solid concentration and K_D values) are larger compared to results from the other adsorption experiments.

Table S7 Summary of the results from the adsorption experiments at elevated concentrations of Cs (at 25 µg/ml [ppm]), including the solid/solution ratio (g/L), the measured initial and final pH, and Cs concentrations in solution (in µg/ml [ppm]), the % removed from solution (adsorbed), the Cs concentrations on the solids (in µg/g), and the respective distribution coefficients (K_D, in ml/g). The description of the experiments includes the solid phase and the composition of the solutions, the values in bold represent the samples used for X-ray Absorption Spectroscopy analyses and the graphical representation of the data is shown in Fig. 1.

				Caesium				
Experiments	Solid (g/L)	pH_i	pH_{final}	Initial conc. (µg/ml)	Final conc. (µg/ml)	% adsorbed	Solid conc. (µg/g)	K_D (ml/g)
Anatase: 25 ppm Cs	9.90	3.95	3.97	24.3	23.4	3.9	91	4
	9.94	5.08	5.10	24.4	22.9	6.0	151	6
	9.95	6.00	6.01	24.4	22.0	9.6	241	11
	9.88	7.00	6.91	24.9	22.0	11.6	294	13
	9.94	6.98	6.93	24.4	22.4	8.1	201	9
	9.90	7.85	7.81	24.3	21.6	11.4	273	13
Illite-smectite: 25 ppm Cs	9.93	4.05	4.08	24.4	16.3	33.0	816	49
	9.96	5.02	4.97	24.4	14.6	39.9	984	67
	9.87	5.95	5.90	24.3	11.6	52.2	1287	110
	9.95	6.67	6.60	24.3	11.1	54.3	1327	119
	9.82	**6.96**	**6.67**	**24.8**	**10.9**	**56.1**	**1415**	**130**
	9.91	7.54	7.38	24.3	10.4	57.1	1403	134
Kaolinite: 25 ppm Cs	9.94	3.96	4.04	24.3	19.7	18.8	463	23
	9.99	4.94	4.98	24.4	17.4	28.6	701	40
	9.92	5.89	6.01	24.4	16.4	32.8	806	49
	9.88	**6.94**	**6.86**	**25.0**	**16.1**	**35.8**	**901**	**56**
	9.92	7.11	7.03	24.4	16.1	33.9	837	52
	9.94	7.90	7.57	24.4	16.0	34.5	845	53
Goethite 25 ppm Cs*	9.98	3.92	3.98	24.4	24.5	-0.7%	-18	-1
	9.88	5.02	5.07	24.3	24.6	-0.9%	-23	-1
	9.93	5.90	4.91	24.3	25.3	-3.9%	-97	-4
	9.88	7.14	7.11	24.3	24.2	0.2%	4	0
	9.86	8.13	7.92	24.2	24.4	-0.6%	-15	-1
Quartz 25 ppm Cs*	9.99	3.90	3.90	24.4	24.6	-0.9%	-22	-1
	9.97	4.88	5.02	24.4	24.8	-1.6%	-40	-2
	9.97	6.02	6.11	24.4	25.3	-3.7%	-90	-4
	9.96	7.03	6.87	24.4	24.5	-0.5%	-11	0
	9.98	7.68	7.27	24.4	24.9	-2.4%	-58	-2
Soil: 25 ppm Cs	9.87	4.10	4.17	24.3	20.6	15.4	375	18
	9.97	5.08	5.05	24.4	18.4	24.4	602	32
	9.98	6.22	6.26	24.4	17.1	29.7	731	42
	9.94	6.97	6.91	24.3	16.5	32.2	785	48
	9.92	**7.23**	**7.01**	**25.1**	**16.7**	**33.3**	**847**	**50**
	9.90	7.82	7.63	24.3	15.0	38.1	939	62

* The Cs adsorption experiments to goethite and quartz were performed during a set of preliminary experiments to develop the appropriate experimental procedures, as such the errors on the aqueous analyses (and consequently on the calculated values: % adsorbed, solid concentration and K_D values) are larger compared to results from the other adsorption experiments.

Table S8 Summary of the results from the adsorption experiments at trace concentrations of Sr-85 and Cs-137 (at ~20 Bq/ml) to the Little Forest Legacy Site core samples (Fig. S1), including the solid/solution ratio (g/L), the measured initial and final pH, and Sr-85 and Cs-137 concentrations in solution (in Bq/ml), the % removed from solution (adsorbed), the Sr-85 and Cs-137 concentrations on the solids (in Bq/g), and the respective distribution coefficients (K_D, in ml/g). The description of the experiments includes the site locations: CH1a and CH30, and their respective depth interval[2, 3] and the composition of the solutions, the graphical representation of the data is shown in Fig. 2.

				Strontium					Caesium				
Experiments	Solid (g/L)	pH_i	pH_{final}	Initial conc. (Bq/ml)	Final conc. (Bq/ml)	% adsorbed	Solid conc. (Bq/g)	K_D (ml/g)	Initial conc. (Bq/ml)	Final conc. (Bq/ml)	% adsorbed	Solid conc. (Bq/g)	K_D (ml/g)
CH1a 0.4-0.6 m 20 Bq/ml Sr-85 and Cs-137	11.24	3.00	3.16	18.25	16.94	7.2	116.4	7	18.18	1.02	94.4	1527.0	1500
	10.29	3.96	4.13	17.94	13.40	25.3	441.7	33	19.01	0.64	96.6	1784.2	2768
	10.15	4.97	4.89	18.20	6.16	66.1	1185.8	192	19.32	0.39	98.0	1865.4	4819
	10.20	6.06	5.80	18.30	3.89	78.8	1413.1	364	19.62	0.88	95.5	1837.3	2099
	10.17	7.10	6.85	18.41	4.29	76.7	1387.2	323	19.46	1.33	93.2	1782.3	1344
	10.35	8.10	7.40	18.71	2.07	88.9	1608.0	776	19.90	0.70	96.5	1855.9	2662
CH1a 1.4-1.6 m 20 Bq/ml Sr-85 and Cs-137	10.27	3.02	3.06	16.26	13.73	15.6	246.2	18	18.33	1.38	92.5	1649.7	1194
	10.21	4.03	4.07	17.15	8.63	49.7	834.5	97	19.33	0.98	94.9	1796.9	1835
	10.29	5.00	4.85	16.92	4.21	75.1	1235.5	293	19.07	1.02	94.6	1754.8	1716
	10.42	6.01	5.64	16.77	1.63	90.3	1453.8	891	18.90	1.08	94.3	1711.7	1591
	10.22	7.00	6.64	16.77	2.75	83.6	1370.9	498	18.90	2.16	88.6	1637.6	759
	10.12	8.10	7.52	16.86	2.27	86.5	1441.6	635	19.00	2.07	89.1	1673.3	809
CH1a 2.4-2.6 m 20 Bq/ml Sr-85 and Cs-137	10.34	3.09	3.11	15.69	12.93	17.5	266.1	21	18.33	1.93	89.5	1585.9	822
	10.35	4.04	4.08	15.79	8.07	48.9	745.5	92	19.75	1.36	93.1	1777.3	1308
	10.22	4.93	4.85	15.67	6.88	56.1	860.1	125	18.92	2.74	85.5	1584.6	579
	10.25	6.07	5.74	15.81	2.76	82.6	1273.0	462	18.98	2.14	88.7	1642.5	769
	10.39	7.08	6.92	15.95	1.73	89.1	1367.8	790	19.37	2.00	89.7	1672.2	838
	10.14	8.03	7.55	15.62	1.91	87.8	1353.1	710	18.87	2.69	85.8	1596.0	594
CH1a 3.4-3.6 m 20 Bq/ml Sr-85 and Cs-137	10.13	3.01	3.05	18.05	13.85	18.2	325.0	22	19.09	0.89	95.3	1797.0	2022
	10.20	3.99	4.03	18.22	8.65	49.4	882.3	96	19.19	0.51	97.4	1831.2	3605
	10.00	4.92	4.94	18.11	3.43	79.8	1445.5	396	19.11	0.41	97.8	1869.2	4534
	10.23	6.01	5.89	16.96	1.25	92.1	1526.9	1145	19.16	0.46	97.6	1827.9	3974
	10.09	6.93	6.80	18.18	1.05	93.8	1690.9	1510	19.18	0.55	97.1	1847.0	3377
	10.17	7.99	7.50	18.20	0.71	95.8	1714.2	2252	19.13	0.51	97.3	1830.0	3582
CH30	10.11	4.53	4.30	7.84	5.15	34.2	265.6	52	22.19	3.84	82.7	1815.4	473
1.0-1.2 m	10.16	5.50	4.97	7.46	4.58	38.6	283.8	62	22.76	5.47	76.0	1702.1	311
20 Bq/ml	10.13	5.53	4.98	7.85	3.70	52.9	409.8	111	22.22	3.04	86.3	1893.6	622
Sr-85 and	10.14	6.52	6.52	7.26	2.16	70.2	505.6	234	22.14	3.18	85.6	1879.6	591
Cs-137*	10.10	5.52	6.64	19.27	7.46	61.3	1170.1	157	22.34	2.77	87.6	1938.0	699

* The adsorption experiments on the samples from CH30 1.0-1.2 m were performed using single spike from a stock solution containing both Sr-85 and Cs-137 (instead of two separate stock solutions) and were performed on different dates; due to the difference in half-life of Sr-85 (65 days) and Cs-137 (30 yr), a selection of the adsorption experiments were performed at 7.2-7.9 Bq/ml as initial Sr-85 concentration instead of ~20 Bq/ml.

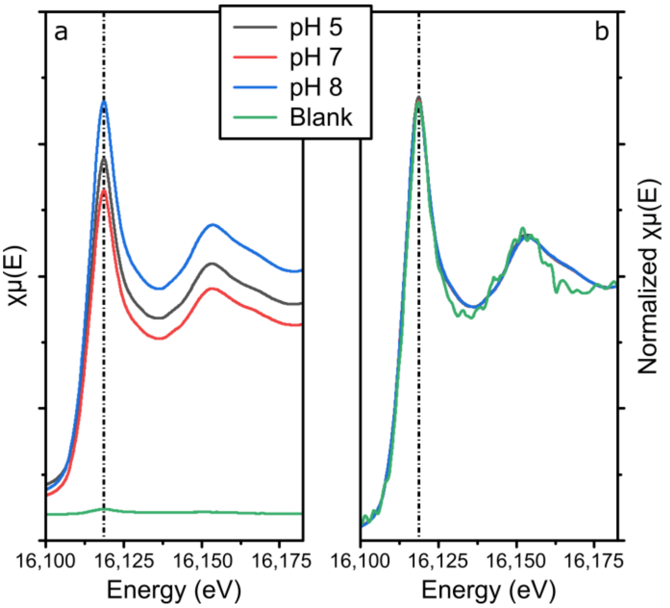

Fig. S2 Comparison of the XANES from the adsorption experiments with kaolinite and a blank kaolinite sample plotted as non-normalized χμ(E) (a), and normalized χμ(E) (b) to emphasize the difference in step size of the adsorption samples compared to the blank kaolinite, and the difference in the shape of the XANES between the adsorption samples and the blank kaolinite.

Table S9 Results from Linear Combination Fitting (LCF)[7] on the Sr XANES and EXAFS spectra, the LCFs were performed on the XANES of the soil samples (from -20 eV to 80 eV relative to the inflection point of the XANES) and on the EXAFS of the soil samples in k-space (from 3 Å^{-1} to 11 Å^{-1}) using the spectra for aqueous Sr^{2+}, $SrCO_3$ and the Sr adsorption samples at the same pH values as the respective standards, included are the top 5 fits (with the lowest R-factor) where all included spectra/species had a fitted percentage higher than zero and sorted left to right from a low to high R-factor, numbers in parenthesis are the errors as determined by Athena.[7]

Standard	**Contribution of the standards to the XANES of the soil sample at pH 5 (%)**					**Contributions of the standards to the EXAFS of the soil sample at pH 5 (%)**				
Aqueous Sr^{2+}				58(2)		18(5)			22(7)	
$SrCO_3$					12(4)				4(1)	4(1)
Anatase	26(7)		63(2)		37(15)		6(4)			8(5)
Kaolinite	36(2)	36(2)	37(2)	42(2)		25(2)	25(2)	25(2)		
Illite-smectite	38(8)	64(2)			52(15)	62(4)	69(4)	75(2)	74(5)	82(5)
R-factor	0.00036	0.00039	0.00042	0.00069	0.00146	0.016	0.017	0.017	0.026	0.027
Reduced χ^2	$6.2\cdot10^{-5}$	$6.7\cdot10^{-5}$	$7.2\cdot10^{-5}$	$1.1\cdot10^{-4}$	$2.4\cdot10^{-4}$	0.076	0.080	0.081	0.13	0.13
Standard	**Contribution of the standards to the XANES of the soil sample at pH 7 (%)**					**Contributions of the standards to the EXAFS of the soil sample at pH 7 (%)**				
Aqueous Sr^{2+}				55(2)			14(6)			
$SrCO_3$					5(4)			1(1)		3(1)
Anatase	2(7)		60(2)		20(14)	20(6)				18(6)
Kaolinite	39(2)	39(2)	41(2)	45(2)		20(3)	20(3)	18(3)	19(3)	
Illite-smectite	60(7)	62(2)			76(14)	63(6)	71(5)	81(3)	81(2)	77(6)
R-factor	0.00039	0.00039	0.00057	0.00068	0.00149	0.014	0.015	0.015	0.015	0.018
Reduced χ^2	$6.7\cdot10^{-5}$	$6.7\cdot10^{-5}$	$9.7\cdot10^{-5}$	$1.1\cdot10^{-4}$	$2.5\cdot10^{-4}$	0.069	0.072	0.074	0.074	0.087
Standard	**Contribution of the standards to the XANES of the soil sample at pH 8 (%)**					**Contributions of the standards to the EXAFS of the soil sample at pH 8 (%)**				
Aqueous Sr^{2+}			15(10)		53(2)				23(9)	
$SrCO_3$										
Anatase	14(5)		40(10)	54(2)		17(7)		14(8)	56(8)	77(4)
Kaolinite	43(2)	43(2)	45(2)	46(2)	48(2)	21(3)	20(3)		33(4)	35(4)
Illite-smectite	44(5)	58(2)				66(7)	80(3)	84(7)		
R-factor	0.00024	0.00025	0.00035	0.00036	0.00039	0.022	0.023	0.028	0.035	0.037
Reduced χ^2	$4.2\cdot10^{-5}$	$4.4\cdot10^{-5}$	$6.2\cdot10^{-5}$	$6.3\cdot10^{-5}$	$6.8\cdot10^{-5}$	0.11	0.11	0.14	0.17	0.17

Table S10 Summary of the results from the shell-by-shell fitting strategy to the EXAFS of Sr adsorbed to anatase, illite-smectite, kaolinite and the composite clayey soil samples, the fits are visualized in Fig. S3, the fitting strategy is summarized in Table S1 and the fit ranges and goodness of fit parameters (R-factor, reduced χ^2 and the results from the f-test for EXAFS[8]) are summarized in Table S10.

	Anatase pH 5	Anatase pH 7	Anatase pH 8	Illite-smectite pH 5	Illite-smectite pH 7	Illite-smectite pH 8	Kaolinite pH 5	Kaolinite pH 7	Kaolinite pH 8	Soil pH 5	Soil pH 7	Soil pH 8
S_0^2*	1	1	1	1	1	1	1	1	1	1	1	1
ΔE_0 (eV)*	-3.6(4)	-3.0(2)	-2.9(2)	-1.8(3)	-1.8(4)	-1.7(4)	-6.1(13)	-6.4(12)	-6.8(12)	-2.9(3)	-2.8(3)	-2.9(3)
		Sr-O			**Sr-O**			**Sr-O$_{(1)}$**			**Sr-O**	
C.N.*	9.3(4)	8.7(2)	8.5(2)	8.9(6)	9.3(7)	9.1(7)	8.8(10)	8.9(10)	9.0(10)	10.6(3)	10.2(3)	10.1(4)
R (Å)*	2.595(4)	2.596(2)	2.597(3)	2.602(5)	2.604(5)	2.604(5)	2.60(1)	2.60(1)	2.60(1)	2.610(3)	2.608(3)	2.609(4)
σ (Å²)*	0.0095(7)	0.0101(4)	0.0098(4)	0.0080(8)	0.0088(9)	0.0087(9)	0.011(1)	0.011(1)	0.011(1)	0.0105(5)	0.0100(5)	0.0100(5)
		Sr-Ti			**Sr-Si**			**Sr-O$_{(2)}$**			**Sr-Si**	
C.N.*	0.5(3)	0.7(3)	0.9(3)	1.0(8)	0.9(8)	0.9(8)	3.6(12)	3.1(12)	2.9(13)	1.1(5)	1.2(5)	1.2(6)
R (Å)*	3.58(1)	3.58(1)	3.58(1)	3.12(2)	3.12(2)	3.12(2)	3.12(1)	3.12(1)	3.12(1)	3.79(2)	3.79(2)	3.79(2)
σ (Å²)*	0.016(5)	0.016(5)	0.016(5)	0.014(10)	0.014(10)	0.014(10)	0.011(1)	0.011(1)	0.011(1)	0.013(6)	0.013(6)	0.013(6)
								Sr-Si				
C.N.*							6.6(32)	5.7(29)	5.5(29)			
R (Å)*							3.79(2)	3.79(2)	3.79(2)			
σ (Å²)*							0.014(10)	0.014(10)	0.014(10)			

* C.N. represents the coordination number, ΔE_0 the energy shift, S_0^2 the amplitude correction factor (fixed to 1 to enable determining the C.N.), R the radial distance and σ^2 the Debye-Waller factor, the numbers in parentheses are the uncertainties as calculated by Artemis,[7] see Table S1 for details on the fitting strategies.

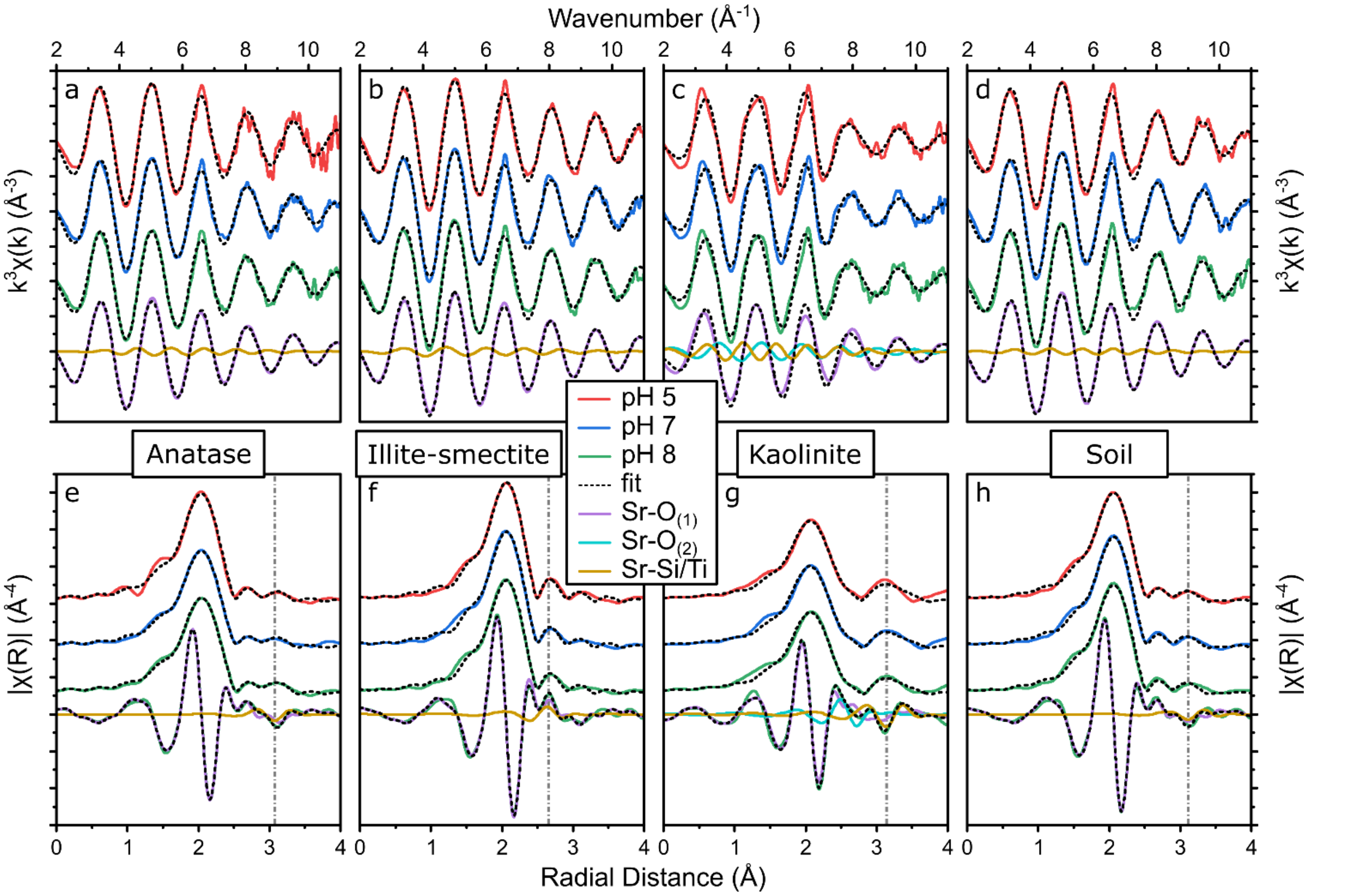

Fig. S3 Visualization of EXAFS in k-space (a-d) and the respective Fourier Transform (FT, e-h) from the samples on Sr adsorption to anatase (a and e), illite-smectite (b and f), kaolinite (c and g) and soil (d and h) and the respective fit to the spectra from the shell-by-shell fitting strategy (Table S9), including the contribution of each scattering path to the EXAFS and the FT of the fit to the adsorption sample at pH 8.

Table S11 Summary of the fit ranges used for the shell-by-shell strategy to fit the Sr EXAFS, and the respective goodness of fit parameters (R-factor, reduced χ^2 and the results from the f-test for EXAFS[8]), the fitting strategy is summarized in Table S1 and the results are summarized in Table S9.

Adsorption experiments	k-range (Å^{-1})	R-Range (Å)	No. of independent data points*	Description of parameters in:		No. of variables Fit2	R-factor Fit2	Reduced χ^2 Fit2	Statistical significance that Fit1 is better than Fit2#
				Fit1	Fit2				
Anatase pH 5-8	**3-10.5**	**1.5-3.5**	**28.37**		**All**	17	0.00092	10.7	
				All	**No Sr-Si**	12	0.00283	46.1	98.6%
Illite-smectite pH 5-8	**3-11**	**1.5-3.5**	**30.28**		**All**	17	0.00163	60.7	
				All	**No Sr-Ti**	12	0.00459	141.9	99.0%
Kaolinite pH 5-8	**3.5-11**	**1.5-4**	**35.36**		**All**	19	0.0064	95.9	
				All	**No Sr-O$_{(2)}$**	15	0.0150	168.1	99.4%
				No Sr-Si	**No Sr-O$_{(2)}$,Si**	10	0.0302	300.4	99.0%
Soil pH 5-8	**3-11**	**1.5-4**	**37.73**		**All**	17	0.0019	45.7	99.7%
				All	**No Sr-Si**	12	0.0043	103.2	

* Number of independent points (NIDP) in the EXAFS spectra is calculated by Artemis:[7] $NIDP = \sum \frac{2}{\pi}(k_{max} - k_{min})(R_{max} - R_{min})$[9]

\# The top row of each fit is empty as the top row describes the fit with all/most fitted variables to which the subsequent fits have been compared

Table S12 Summary of the fit ranges used for the holistic strategy to fit the Sr EXAFS, and the respective goodness of fit parameters (R-factor, reduced χ^2 and the results from the f-test for EXAFS[8]), an example of the fitting strategy is summarized in Table S2. The fits described as "All" are summarized in Table S13, and the fits described as "no $f_{Anatase}$" are summarized in Table 2 and Fig. 5.

Adsorption experiments	k-range (Å^{-1})	R-Range (Å)	No. of independent data points*	Description of parameters in:		No. of variables Fit2	R-factor Fit2	Reduced χ^2 Fit2	Statistical significance that Fit1 is better than Fit2#
				Fit1	Fit2				
pH 5					**All**	21	0.00349	64.83	
Soil				**All**	**No $f_{Anatase}$**	20	0.00343	72.26	0.0%
	3-10.5	**1.5-4**		**All**	**No f_{Illite}**	20	0.00373	63.88	76.6%
				All	**No $f_{Kaolinite}$**	20	0.00429	78.94	96.3%
			42.49	**No $f_{Anatase}$**	**No $f_{Anatase}$, f_{Illite}**	19	0.00714	141.35	100.0%
				No $f_{Anatase}$	**No $f_{Anatase}$, $f_{Kaolinite}$**	19	0.00400	90.65	93.4%
Anatase	**3-10.5**	**1.5-3.5**		**No $f_{Anatase}$**	**No Sr-Ti**	18	0.00367	67.91	53.9%
Kaolinite	**3.5-11**	**1.5-4**		**No $f_{Anatase}$**	**No Sr-O$_{(2)}$**	18	0.00828	138.48	100.0%
				No Sr-O$_{(2)}$	**No Sr-O$_{(2)}$,Si**	16	0.01225	208.54	99.2%
Illite	**3-10.5**	**1.5-3.5**		**No $f_{Anatase}$**	**No Sr-Si**	18	0.00573	171.01	99.7%
pH 7					**All**	21	0.00195	42.38	
Soil				**All**	**No $f_{Anatase}$**	20	0.00195	40.50	3.7%
	3-10.5	**1.5-4**		**All**	**No f_{Illite}**	20	0.00261	59.16	98.7%
				All	**No $f_{Kaolinite}$**	20	0.00215	49.66	85.2%
			42.49	**No $f_{Anatase}$**	**No $f_{Anatase}$, f_{Illite}**	19	0.00521	86.88	100.0%
				No $f_{Anatase}$	**No $f_{Anatase}$, $f_{Kaolinite}$**	19	0.00219	48.85	88.8%
Anatase	**3-10.5**	**1.5-3.5**		**No $f_{Anatase}$**	**No Sr-Ti**	18	0.00247	50.36	93.0%
Kaolinite	**3.5-11**	**1.5-4**		**No $f_{Anatase}$**	**No Sr-O$_{(2)}$**	18	0.00509	77.19	100.0%
				No Sr-O$_{(2)}$	**No Sr-O$_{(2)}$,Si**	16	0.00860	121.09	99.8%
Illite	**3-10.5**	**1.5-3.5**		**No f_{ana}**	**No Sr-Si**	18	0.00348	93.91	99.9%
pH 8					**All**	21	0.00232	44.48	
Soil				**All**	**No $f_{Anatase}$**	20	0.00232	42.50	0.0%
	3-10.5	**1.5-4**		**All**	**No f_{Illite}**	20	0.00321	59.74	99.1%
				All	**No $f_{Kaolinite}$**	20	0.00250	48.10	79.6%
			42.49	**No $f_{Anatase}$**	**No $f_{Anatase}$, f_{Illite}**	19	0.00542	74.10	100.0%
				No $f_{Anatase}$	**No $f_{Anatase}$, $f_{Kaolinite}$**	19	0.00257	47.21	86.7%
Anatase	**3-10.5**	**1.5-3.5**		**No $f_{Anatase}$**	**No Sr-Ti**	18	0.00332	54.69	98.3%
Kaolinite	**3.5-11**	**1.5-4**		**No $f_{Anatase}$**	**No Sr-O$_{(2)}$**	18	0.00469	60.73	100.0%
				No Sr-O$_{(2)}$	**No Sr-O$_{(2)}$,Si**	16	0.00796	90.69	99.8%
Illite	**3-10.5**	**1.5-3.5**		**No $f_{Anatase}$**	**No Sr-Si**	18	0.00412	102.82	99.8%

* Number of independent points (NIDP) in the EXAFS spectra is calculated by Artemis:[7] $NIDP = \sum \frac{2}{\pi}(k_{max} - k_{min})(R_{max} - R_{min})$[9]

\# The top row of each fit is empty as the top row describes the fit with all/most fitted variables to which the subsequent fits have been compared.

Table S13 Summary of the results from the holistic fitting strategy to the EXAFS of Sr adsorbed to anatase, illite-smectite, kaolinite and the composite clayey soil samples (including refining the fraction of anatase [$f_{Anastase}$] during the fitting strategy). The fitting strategy and respective parameters are summarized in Table S2 and the fit parameters and goodness of fit parameters (R-factor, reduced χ^2 and the results from the f-test for EXAFS[8]) are summarized in Table S12.

	Anatase pH 5	**Anatase pH 7**	**Anatase pH 8**	**Illite-smectite pH 5**	**Illite-smectite pH 7**	**Illite-smectite pH 8**	**Kaolinite pH 5**	**Kaolinite pH 7**	**Kaolinite pH 8**	**Soil pH 5#**	**Soil pH 7#**	**Soil pH 8#**
S_0^2*	1	1	1	1	1	1	1	1	1	1	1	1
ΔE_0 (eV)*	-3.6(8)	-3.0(4)	-2.8(4)	-1.7(4)	-1.9(3)	-1.8(3)	-7.0(8)	-6.3(7)	-5.9(9)	-3.8(6)	-2.7(4)	-2.7(4)
		Sr-O			**Sr-O**			**Sr-O(1)**			**$f_{Anatase}$~**	
C.N.*	9.5	8.6	8.5	8.9	9.3	9.1	8.8	8.9	9.0			
R (Å)*	2.600(9)	2.596(4)	2.598(5)	2.602(4)	2.603(3)	2.603(3)	2.603(8)	2.600(7)	2.606(8)	0.49(26)	0.02(23)	0.00(24)
σ (Å²)*	0.0099(7)	0.0099(3)	0.0099(4)	0.0079(2)	0.0087(2)	0.0087(2)	0.0111(4)	0.0110(4)	0.0110(4)			
		Sr-Ti			**Sr-Si**			**Sr-O(2)**			**f_{Illite}~**	
C.N.*	0.5	0.7	1.0	1	0.9	0.9	3.6	3.2	2.9			
R (Å)*	**3.42(10)**#	3.57(4)	3.59(3)	3.13(2)	3.11(2)	3.11(2)	3.11(1)	3.12(1)	3.12(2)	0.24(26)	0.74(19)	0.76(19)
σ (Å²)*	0.020(18)	0.017(6)	0.017(2)	0.013(2)	0.014(3)	0.014(3)	0.011(2)	0.012(2)	0.012(3)			
								Sr-Si			**$f_{Kaolinite}$~**	
C.N.*							6.6	5.7	5.4			
R (Å)*							3.79(2)	3.80(2)	3.80(2)	0.30(13)	0.25(12)	0.25(15)
σ (Å²)*							0.020(2)	0.020(2)	0.020(2)			

* S_0^2 represents the amplitude correction factor (fixed to 1), C.N. the coordination number (fixed, the errors on the C.N. are estimated to be ~25%), ΔE_0 the energy shift, R the radial distance and σ^2 the Debye-Waller factor, the numbers in parentheses are the uncertainties as calculated by Artemis,[7] the C.N. were fixed based on the shell by shell fits (Table S10).

The value in bold deviates significantly from the value refined during the shell-by-shell fitting strategy (Table S10)

~ The fitting parameters to the soil samples (besides the energy shift, ΔE_0) represent the fraction of Sr in the sample complexed with anatase ($f_{Anatase}$), illite-smectite (f_{Illite}) and kaolinite ($f_{Kaolinite}$; Table S2) rather than single scattering paths

Table S14 Summary of the results from the shell-by-shell fitting strategy to the EXAFS of Cs adsorbed to illite-smectite, kaolinite and the composite clayey soil samples, for comparison this table includes published coordination environments obtained through Molecular Dynamics for Cs complexation at the Si vacancy sites of the basal plane of Illite[4] or kaolinite[5]. The fits are visualized in Fig. S4, the fitting strategy is summarized in Table S3.

	Illite-smectite pH 7	**Kerisit et al.[4]**	**Kaolinite pH 7**	**Vasconcelos et al.[5]**	**Soil pH 7**
S_0^2*	1	1	1	1	1
ΔE_0 (eV)*	-0.6(52)		4.8(63)		0.0(67)
			Cs-O(1)		
C.N.*	8*	7.8	7*	5.4	8*
R (Å)*	3.18(6)	3.15	3.15(8)	3.12	3.14(4)
σ (Å²)*	0.022(5)		0.016(6)		0.018(3)
			Cs-O(2)		
C.N.*	4*	3.9	5*	5.1	4*
R (Å)*	3.55(10)	3.45	3.37(10)	3.41	3.43(6)
σ (Å²)*	0.022(5)		0.016(6)		0.018(3)
			Cs-Si		
C.N.*	5*	5-6	6*	5.4	5.5*
R (Å)*	4.11(8)	3.85-3.95	4.14(8)	4.06	4.12(8)
σ (Å²)*	0.016(4)		0.013(3)		0.014(2)

* C.N. represents the coordination number, ΔE_0 the energy shift, S_0^2 the amplitude correction factor (fixed to 1 to enable determining the C.N.), R the radial distance and σ^2 the Debye-Waller factor, the numbers in parentheses are the uncertainties as calculated by Artemis,[7] see Table S3 for details on the fitting strategies.

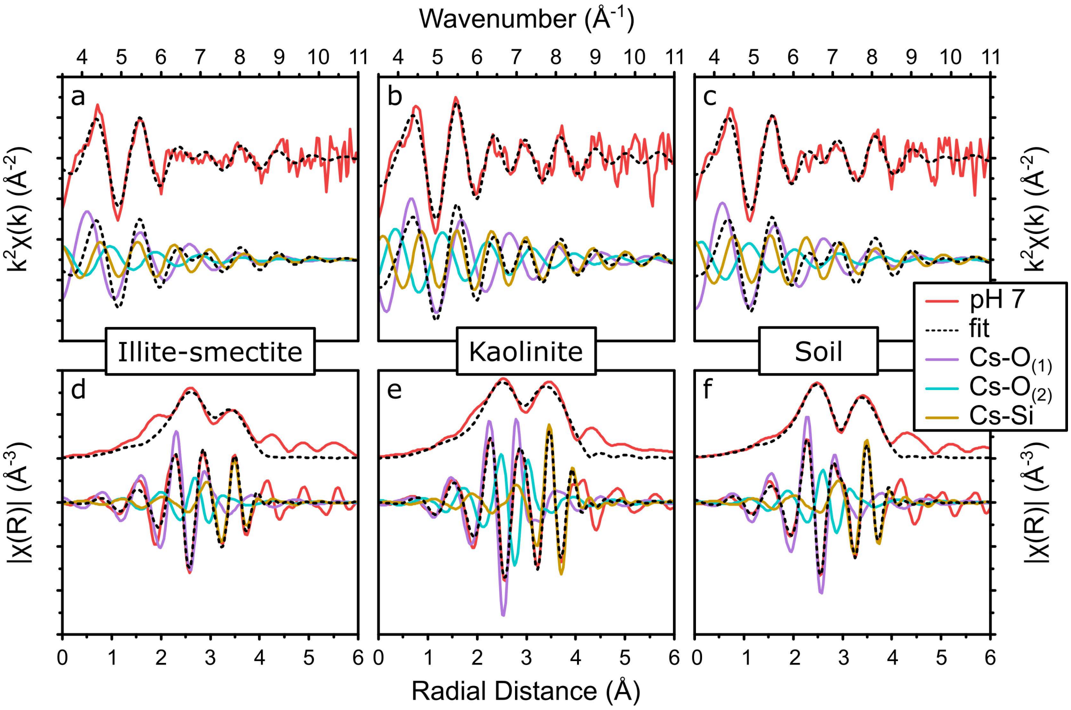

Fig. S4 Visualization of EXAFS in k-space (a-d) and the respective Fourier Transform (FT, e-h) from the samples on Cs adsorption at pH7 to illite-smectite (e and d), kaolinite (b and e) and soil (c and f) and the respective fit to the spectra from the shell-by-shell fitting strategy (Table S14), including the contribution of each scattering path to the EXAFS and the FT of the fit to the adsorption sample at pH 8.

Table S15 Summary of the fit ranges used for the holistic strategy to fit the Cs EXAFS, and the respective goodness of fit parameters (R-factor, reduced χ^2 and the results from the f-test for EXAFS[8]). The fitting strategy is summarized in Table S3 and the results are summarized in Table 3.

Adsorption experiments	k-range (Å^{-1})	R-Range (Å)	No. of independent data points*	Description of parameters in: Fit1	Fit2	No. of variables Fit2	R-factor Fit2	Reduced χ^2 Fit2	Statistical significance that Fit1 is better than Fit2[#]
Soil pH 7	**3.5-9.5**	**2.2-4**	**20.22**		**All**	14	0.028	72.0	
				All	**No f_{kao}**	14	0.031	78.3	n/a^
				All	**No f_{ill}**	14	0.033	86.6	n/a^
Illite-smectite pH 7	**3.5-9.5**	**2.2-4**		**All**	**No Cs-O$_{(2)}$**	13	0.036	78.3	77.8%
				No Cs-O$_{(2)}$	**No Cs-O$_{(2)}$,Si**	11	0.127	227	98.7%
Kaolinite pH 7	**3.5-9.5**	**2.2-4**		**All**	**No Cs-O$_{(2)}$**	13	0.036	80.3	76.0%
				No Cs-O$_{(2)}$	**No Cs-O$_{(2)}$,Si**	11	0.188	301	99.7%

* Number of independent points (NIDP) in the EXAFS spectra is calculated by Artemis:[7] $NIDP = \sum \frac{2}{\pi}(k_{max} - k_{min})(R_{max} - R_{min})$[9]

[#] The top row of each fit is empty as the top row describes the fit with all/most fitted variables to which the subsequent fits have been compared

^ Because f_{ill} was defined as 1-f_{kao}, the number of independent variables between both fits were identical and no f-test could be performed, instead the reduction in both the reduced χ^2 and the R-factor when including both, f_{ill} and f_{kao}, instead of either one suggests both, f_{ill} and f_{kao}, need to be included for the best fit.

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