**Appendix A: Expanded methodology for studies and surface complexation modeling of Ra sorption**

**Experimental Methods:** Ferrihydrite naturally exists as a hydrated mineral,1 thus it was important to prepare and handle the mineral such that its hydration was preserved. Here, a ferrihydrite slurry was synthesized by adding 0.4 M NaOH to a 50 mM ferric chloride hexahydrate solution, and then centrifuging and washing the resulting precipitate three times with 18 MΩ water, resulting in a thick paste of ferrihydrite. A small volume (<100 mL) of DI water was added to create a slurry consistency. The iron content of the prepared ferrhydrite slurry was determined through colorimetry (ferrozine method)2 and stirred slurry aliquots were added directly to the experiments to achieve the desired mineral mass.

Goethite was prepared through slow air-oxidation of a 50 mM Fe2+ and 100 mM bicarbonate solution over two days, and then centrifuged and washed three times with 18 MΩ water. The resultant slurry was then oven dried at 70 °C for two hours, resulting in a mineral powder that was then gently ground with a mortar and pestle to homogenize the sample. This dried sample was added directly to experimental bottles.

There are well established methods for preparing natural clay mineral samples for use in experimental work.3 The STx-1b montmorillonite ordered from the Clay Minerals Society was originally a calcium montmorillonite, but was converted to sodium montmorillonite for easier comparison to other studies. This was achieved by first dispersing the clay with 1 M NaCl, then siphoning the suspended <0.2 µm clay fraction by successively centrifuging and suspending the clay eight times in DI water. The suspension was flocculated with saturated NaCl, and excess solution removed via centrifugation. The clay suspension was then treated with a 1 M sodium acetate solution (pH 5) to remove residual carbonate minerals. The resulting Na-equilibrated montomorillonite was then centrifuged and equilibrated with the experimental background solution (10 mM NaCl), centrifuged again, dried at 70 °C overnight, and then gently powdered using mortar and pestle. This dried powder was added directly to the experiments. Results of the kinetic experiments involving sodium montmorillonite are shown in figure A1, showing that over a 24 hour period, the water Ra concentrations stabilize.

All prepared minerals’ surface area was measured using a BET surface area analyzer (table A1). Ferrihydrite was dried for this analysis, and pyrite was held anaerobically until analysis.

Displacement of H+ from mineral surfaces can cause sorption experiments to drift in pH over time, requiring some pH adjustment. Following the experimental period (24 hours), pH was checked and re-titrated to the desired value if necessary; if the pH deviated more than 0.1 pH units, the bottle was allowed to re-equilibrate for 15 minutes after titration, and the re-titration process repeated. This process was sufficient to ensure the experimental pHs described.

The composition of the various background solutions used in sorption experiments are reported in table A2. Artificial groundwater, brackish water, and seawater were diluted from a 2x artificial seawater stock. The ratio of individual cations was not selected to match a specific location, but rather to simulate common groundwater and seawater cations in “typical” environmental ratios.4 The cations selected for single competing cation experiments reported in figure 2 of the main text were selected based on the composition of the artificial waters. Only chloride salts of these cations were used to make solutions to limit interference from different anions.

Control experiments without any mineral were also performed to examine Ra sorption to the serum bottle surface. Some pH dependent sorption was observed, but was limited in comparison to the minerals studied. Given the limited observed sorption within controls, no corrections to data were made. Serum bottles and caps were scrubbed in detergent solution, rinsed, soaked in 2% HNO3 for at least 1 hour, rinsed 3 times with 18 MΩ water, and then air dried before usage in experiments. This procedure was sufficient to prevent Ra and mineral cross contamination across experiments.

**Analytical Methods:** Background concentrations Ra were measured through scintillation counting (N = 47) to develop a limit of blank of 1.4 counts per second (cps). From this, we developed a blank derived minimum detection limit of 0.209 Bq, as defined by the EPA (EPA 821-R-16-006).5 A Canberra low energy germanium detector and multichannel analyzer was calibrated using a multinuclide standard from Eckert and Ziegler ([www.ezag.com)](http://www.ezag.com)), and 226Ra activities for scintillation counting standards were determined using Canberra Genie software using the 186 keV peak.

**Comparison of Sorption Experiment Results:** Fitted Kd and Ksa values developed from isotherm experiments are reported in table S1, with the corresponding fits plotted for each mineral in figure A2. Errors reported for Kd values are the standard error of regression produced by the least squares fitting function, linregress, from the python package SciPy, and Ksa the fitted Kd normalized by mineral specific surface area. These values were also used for error bar plots in the main text (figure 1). The pH and pH error are the average and standard deviation of all data points used for a given regression.

Our results are compared with literature K­d values (Table A3), which are mostly derived from experimental isotherms. In some cases, a Kd value was not reported, thus a Kd value was fit directly from the reported experimental data. The greatest number of reported Kd values are found for the iron (hydr)oxides; however, there are significant differences in the experimental conditions (solid-solution loading, background electrolyte composition, etc.). Notable similarities and differences are explained in the main text.

**Surface Complexation Modeling:** Experimental sorption data was fit by varying Ra sorption reaction constants.6,7 Surface area and site densities, while fittable parameters in the models, were not varied, and were constrained with surface areas reported in table S1 and site densities previously determined in the literature (table A4). For exchange sites used montmorillonite modeling, the CEC value given by the The Clay Minerals Society (clays.org) was used. Solution complexation behavior was accounted for using the SIT database, which includes Ra carbonate, sulfate, chloride, and hydroxide complexes, albeit these solution complexes had little impact over the experimental conditions considered.

The surface complexes used to model Ra sorption and impact of competing ions are reported in table A4. Ra complexation constants were fitted to match sorption isotherm data or circumneutral, varied background cation data, and are also reported for both methods of fitting in table A4. The resultant visual fits to the isotherm data are shown in figures A3, A4, and A5, using the typical means of comparing the isotherm pH vs the fraction of Ra sorbed, and plotting both the SCM result and the experimental data. For fitting the competiting cation data, another approach was used, where data points are plotted by their experimental value, and the value produced by the SCM (Figures A6, A7, and A8). To evalulate the whole fit, the RMSE of the simulation results vs the experimental data was calculated, and is reported in the main text. As discussed in the main text, the log K values generated using either fitting method were also evaluated using the complimentary method, as plotted in the supplementary figures S3-S8. Constants for the other surface complexes are drawn from the literature, however many of the models used in the literature use alternative representations of the mineral surface (ie. CD-MUSIC, or extended triple layer models), thus the constants are not readily applicable to the generalized two layer models used here. Only constants fit using the same type of mineral specific model and similar surface complexes were used to model background electrolyte competition. Carbonate surface complexes were only included in the SCM for goethite, as the goethite synthesis method involves high concentrations of carbonate to buffer pH, resulting in carbonate surface complexes that persist even after washing.8 Some cations used experimentally (e.g. Na+, K+) did not have published constants, but clearly demonstrated differential amounts of sorption (Figure 2, main text, Figures A6, A7, A8). In these cases, no constants were fit or used, underscoring a potential limitation for using these models for predicting Ra transport in natural systems.

We also considered a few other mineral-specific SCMs to attempt replication of results reported in the main text. For the Fe (hydr)oxides, single site models featuring tetradentate surface coordination have been used in both generalized double layer models, and extended triple layer models to fit divalent cation sorption.7,9,10 For montmorillonite, many models have been proposed, but no standardized SCM has been adopted, some which follow the non-electrostatic method used here, others using various means of representing the permanent negative charge of the clay surface.11,12 While in some cases use of these models improved fits to our data, the lack of spectroscopic studies for Ra combined with the limited database of constants for competing ions effectively precluded the use of these alternative models. The non-standardization of these different SCMs and their calibration also impeded any efforts to test their viability for predicting transport of Ra in the different geochemical conditions tested here.13 Thus, we elected to use relatively “simple” models that are well established in the literature, and have a wider variety of available reaction constants that can be used to model the effect of different competing cations. In the case of pyrite, no model specific to an unoxidized surface was available, and testing a few different reactions with free Ra did not produce satisfactory fits. A previous SCM used to model Eu and Sr sorption to pyrite used both unoxidized and oxidized sites to replicate experimental data, but that model did not fit here either.14 Additionally, since there is no experimental evidence of oxidation of the pyrite surface, a model that included such types of sites would be inappropriate. That these simpler models failed to provide meaningful prediction of the impact of different solution conditions (main text), and the no models were able to fit the pyrite experimental data found here suggests that further detailed study of Ra sorption to mineral surfaces is needed to constrain these SCMs. In particular, constants to explain the differences in Na and K sorption, as well as better understanding of mineral specific Ra surface complexes would improve these models’ predictive capability.

**Table A1. Mineral Surface Areas and Fitted Kd and Ksa values**

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Mineral | Surface Area (m2/g) | pH | Kd (mL/g) | Ksa (mL/m2) |
| Ferrihydrite | 383 | 3 | 405±72 | 1.06±0.19 |
|  | 5 | 526±48 | 1.37±0.13 |
|  | 7 | 2420±210 | 6.33±0.55 |
|  | 9 | 302000±29000 | 789±76 |
| Goethite | 146 | 3 | 2.77±7.86 | 0.019±0.054 |
|  | 5 | 400±42 | 2.73±0.29 |
|  | 7 | 679±52 | 4.64±0.36 |
|  | 9 | 12400±700 | 84.8±4.8 |
| Sodium Montmorillonite | 50.2 | 3 | 7740±860 | 154±17 |
|  | 5 | 20000±2000 | 398±40 |
|  | 7 | 22600±300 | 451±6 |
|  | 9 | 25400±1500 | 507±29 |
| Pyrite | 0.07 | 3 | 156±18 | 2280±260 |
|  | 5 | 400±29 | 5840±420 |
|  | 7 | 579±16 | 8450±230 |
|  | 9 | 574±29 | 8380±430 |

**Table A2: Major Salt Concentrations in the different background treatments**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Experiment** | **Na (mM)** | **K (mM)** | **Mg (mM)** | **Ca (mM)** | **Sr (mM)** | **Cl (mM)** |
| NaCl | 10 |  |  |  |  | 10 |
| KCl |  | 10 |  |  |  | 10 |
| MgCl2 |  |  | 10 |  |  | 20 |
| CaCl2 |  |  |  | 10 |  | 20 |
| SrCl2 |  |  |  |  | 10 | 20 |
| AGW | 5 | 2 | 0.5 | 0.5 | 0.0001 | 9 |
| ABW | 50 | 20 | 5 | 5 | 0.001 | 90 |
| ASW | 400 | 160 | 40 | 40 | 0.008 | 720 |

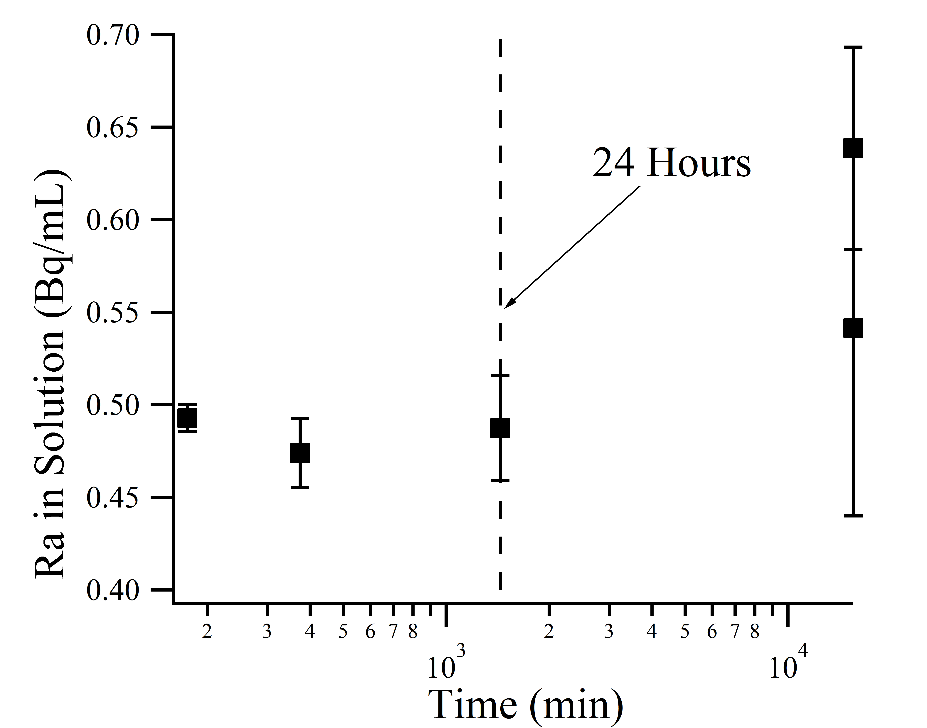
**Table A3. Comparison of Literature Ra Sorption Experiments and Fitted Kd Values**

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Mineral | Solid/Solution Ratio (mg/L) | Background Solution | pH | Kd (mL/g) | Source |
| Ferrihydrite | 300 | 10 mM NaCl | 7 | 2420 | This study |
| 300 | 10 mM NaCl | 9 | 302000 | This study |
| 25000 | Seawater | 8.25 | 1535 | 15 |
| 10000 | 100 mM NaClO4 | 7 | 1440 | 7 |
| Goethite | 300 | 10 mM NaCl | 3 | 2.77 | This study |
| 300 | 10 mM NaCl | 5 | 400 | This study |
| 300 | 10 mM NaCl | 7 | 679 | This study |
| 300 | 10 mM NaCl | 9 | 12400 | This study |
| 25000 | Seawater | 8.25 | 20 | 15 |
| 10000 | 100 mM NaClO4 | 7 | 50.6 | 7 |
| 500000 | “pH 1 solution” | 1 | 0.752 | 16 |
| 500000 | “pH 10 solution” | 10.1 | 544 | 16 |
| Lepidocrocite | 25000 | Seawater | 8.25 | 174 | 15 |
| Hematite | 25000 | Seawater | 8.25 | 75 | 15 |
| Sodium Montmorillonite | 300 | 10 mM NaCl | 5 | 20000 | This study |
| 300 | 10 mM NaCl | 7 | 22600 | This study |
| 3333 | 10 mM NaCl | 5.25 | 9700 | 17 |
| 50000 | 10 mM NaCl | 6.5 | 3724 | 18 |

**Table A4: Surface complexation reactions and constants from data fitting**

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Reactions | Sites (mol/g) | log K Isotherm | Lg K  Salinity | Source |
| Ferrihydrite  Protonation  ≡FhysOH + H+ ⇄ ≡FhysOH2+  ≡FhysOH ⇄ ≡FhysO- + H+  ≡FhywOH + H+ ⇄ ≡FhywOH2+  ≡FhywOH ⇄ ≡FhywO- + H+  Radium  ≡FhysOH + Ra2+ ⇄ ≡FhysOHRa2+  ≡FhywOH + Ra2+ ⇄ ≡FhywORa++ H+  ≡FhywOH + Ra2+ + H2O ⇄ ≡FhywORaOH + 2H+  Competing Ions  ≡FhysOH + Ca2+ ⇄ ≡FhysOHCa2+  ≡FhywOH + Ca2+ ⇄ ≡FhywOCa++ H+  ≡FhywOH + Mg2+ ⇄ ≡FhywOMg++ H+  ≡FhysOH + Sr2+ ⇄ ≡FhysOHSr2+  ≡FhywOH + Sr2+ ⇄ ≡FhywOSr++ H+  ≡FhywOH + Sr2+ H2O ⇄ ≡FhywOSrOH + 2H+ | 1.87E-3  4.67E-5 | 7.92  -8.93  7.92  -8.93  6.7  -2.8  -15  4.97  -5.85  4.6  5.01  -6.58  -17.6 | 7.92  -8.93  7.92  -8.93  5.7  -11.0  -9.4  4.97  -5.85  4.6  5.01  -6.58  -17.6 | A  A  A  A  Fitting  Fitting  Fitting  A  A  A A  A  A |
| Goethite  Protonation  ≡GoeOH + H+ ⇄ ≡GoeOH2+  ≡GoeOH ⇄ ≡GoeO- + H+  Radium  ≡GoeOH + Ra2+ ⇄ ≡GoeORa+ + H+  ≡GoeOH + Ra2+ ⇄ ≡GoeOHRa2+  Competing Ions  ≡GoeOH + Ca2+ ⇄ ≡GoeOCa+ + H+  ≡GoeOH + Ca2+ ⇄ ≡GoeOHCa2+  ≡GoeOH + Mg2+ ⇄ ≡GoeOMg+ + H+  ≡GoeOH + Mg2+ ⇄ ≡GoeOHMg2+  ≡GoeOH + Sr2+ ⇄ ≡GoeOSr+ + H+  ≡GoeOH + Sr2+ ⇄ ≡GoeOHSr2+  ≡GoeOH + 2H+ + CO32- ⇄ ≡GoeOCOOH+ H2O  ≡GoeOH + H+ + CO32- ⇄ ≡GoeOCOO- ­­+ H2O | 4.87E-4 | 6.93  -9.65  -3.9  3.5  -6.48  3.98  -3.02  5.24  -5.44  3.59  20.78  12.71 | 6.93  -9.65  -2.5  -6.6  -6.48  3.98  -3.02  5.24  -5.44  3.59  20.78  12.71 | B  B  Fitting  Fitting  B  LFER, B  B  LFER, B  B  LFER, B  C  C |
| Sodium Montmorillonite  Exchange  2 ≡Clay-Na + Ra2+ ⇄ ≡Clay2-Ra + 2 Na+  2 ≡Clay-Na + Ca2+ ⇄ ≡Clay2-Ca + 2 Na+  2 ≡Clay-Na + Mg2+ ⇄ ≡Clay2-Mg + 2 Na+  ­Surface Protonation  ≡ClaysOH + H+ ⇄ ≡ClaysOH2+  ≡ClaysOH ⇄ ≡ClaysO- + H+  ≡Clayw1OH + H+ ⇄ ≡Clayw1OH2+  ≡Clayw1OH ⇄ ≡Clayw1O- + H+  ≡Clayw2OH + H+ ⇄ ≡Clayw2OH2+  ≡Clayw2OH ⇄ ≡Clayw2O- + H+  Radium  ≡ClaysOH + Ra2+ ⇄ ≡ClaysORa+ + H+  ≡Clayw1OH + Ra2+ ⇄ ≡Clayw1ORa+ + H+  Competing Ions ≡ClaysOH + Mg2+ ⇄ ≡ClaysOMg+ + H+  ≡Clayw1OH + Mg2+ ⇄ ≡Clayw1OMg+ + H+  ≡ClaysOH + Ca2+ ⇄ ≡ClaysOCa+ + H+  ≡Clayw1OH + Ca2+ ⇄ ≡Clayw1OCa+ + H+  ≡ClaysOH + Sr2+ ⇄ ≡ClaysOSr+ + H+  ≡ClaysOH + Sr2+ + H2O ⇄ ≡ClaysOSrOH+ 2H+  ≡Clayw1OH + Sr2+ ⇄ ≡Clayw1OSr+ + H+  ≡Clayw1OH + Sr2+ + H2O ⇄ ≡Clayw1OSrOH+ 2H+ | 8.45E-4  2E-6  4E-5  4E-5 | 0.2  0.5  0.4  4.5  -7.9  4.5  -7.9  6.0  -10.5  0.0  -2.1  -2.4  -5.2  -3.4  -5.2  -3.9  -16.7  -6.8  -20.8 | 0.2  0.5  0.4  4.5  -7.9  4.5  -7.9  6.0  -10.5  -0.9  -1.9  -2.4  -5.2  -3.4  -5.2  -3.9  -16.7  -6.8  -20.8 | Fitting, D  E  F  G  G  G  G  G  G  Fitting  Fitting  LFER, H  LFER, H  LFER, H  LFER, H  LFER, H  LFER, H  LFER, H  LFER, H |

Notes: LFER: Indicates constants was fitted using a linear free energy relationship from the denoted source. Sources: A: From Dzombak and Morel, 1990.19 B: From Mathur and Dzombak, 2006.20 C: From Van Geen, Robertson, and Leckie, 1994.21 D: Number of exchange sites from CEC reported by Clays.org for STx-1b Calcium Montmorillonite. Exchange reaction fitted to match low pH sorption (ie, sorption without surface site impacts). E: From Tournassat et al., 2004.22 F: From Charlet and Tournassat, 2005.23 G: From Bradbury and Baeyens, 1997.24 H: From Bradbury and Baeyens, 2005.25

Figure A1. Ra sorption to Sodium Montmorillonite as a function of time.

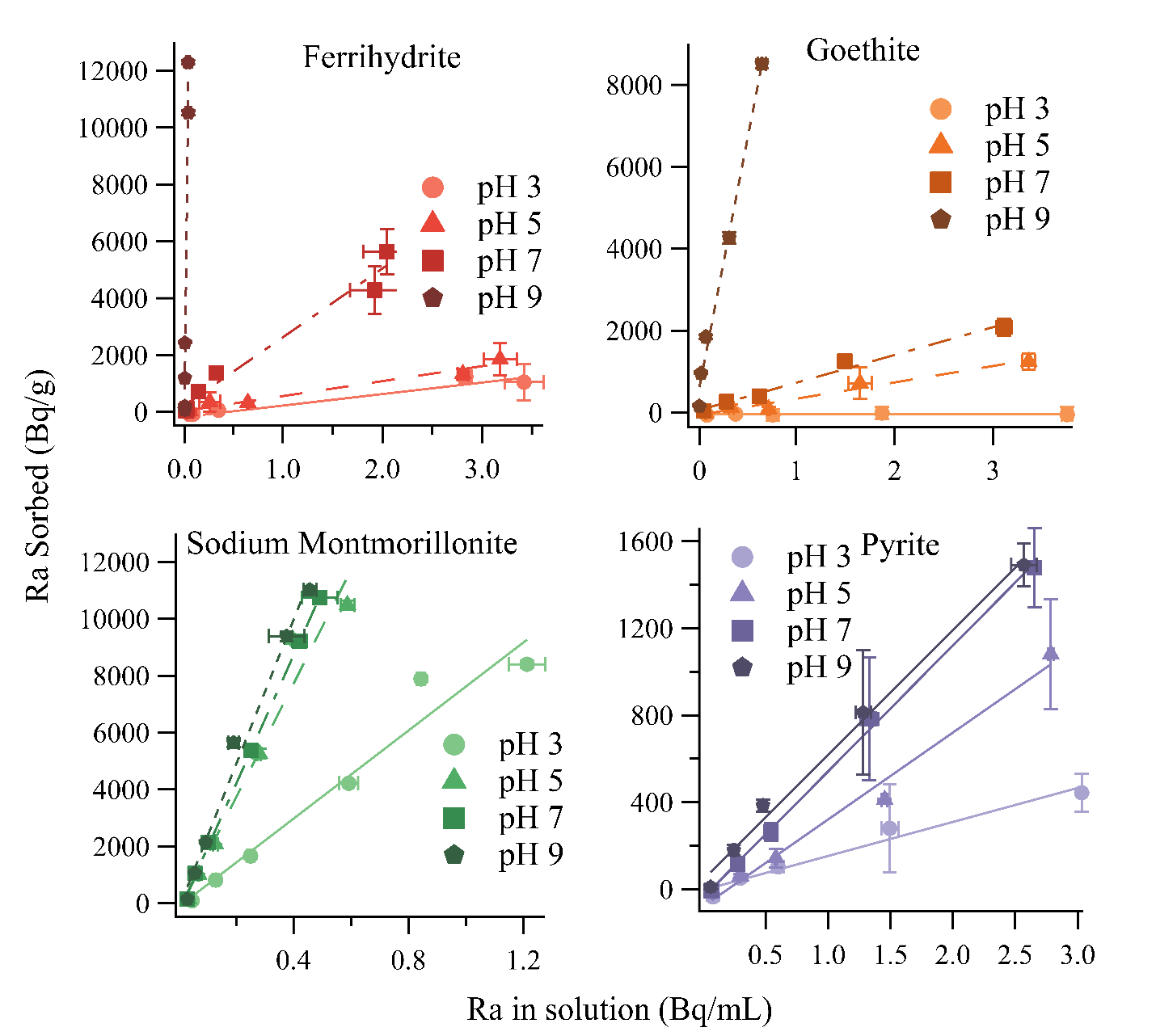
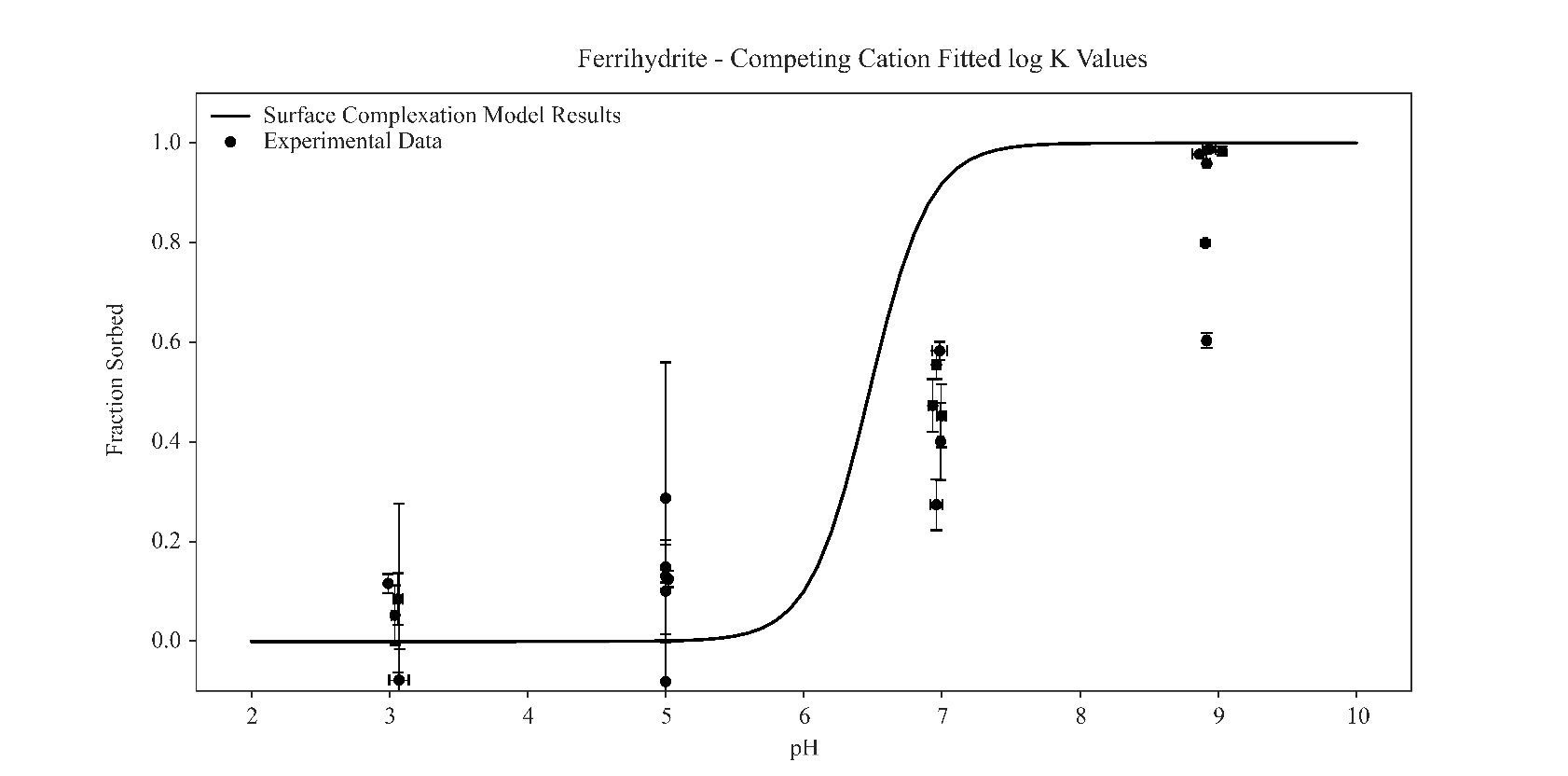
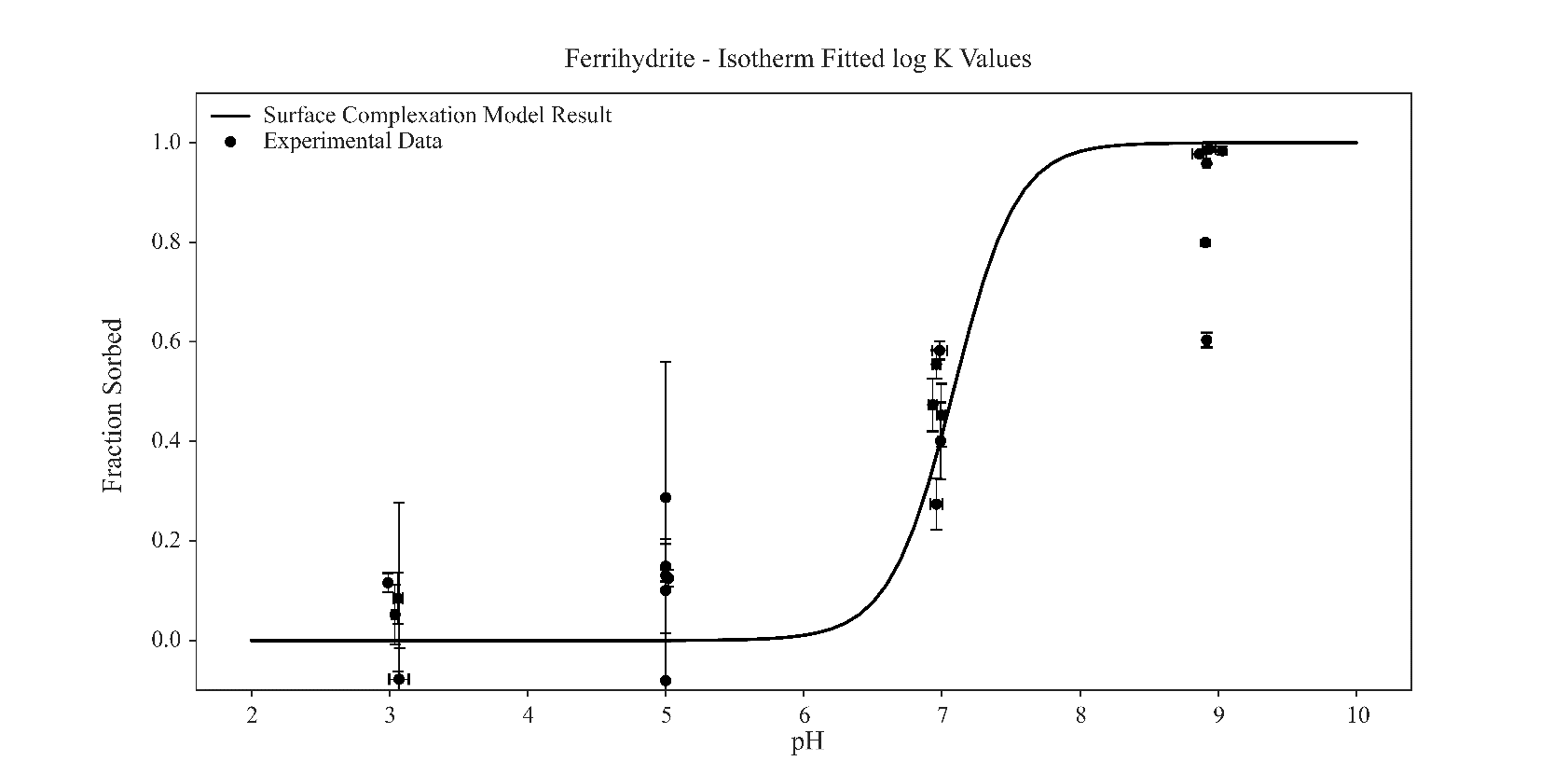


Figure A2: Results of isotherm experiments with linear fits.

Figure A3: Comparison of ferrihydrite SCM results generated from either fitting isotherm data or fitting competing cation data. Experimental data plotted is from the isotherm experiments with 10 mM NaCl background.

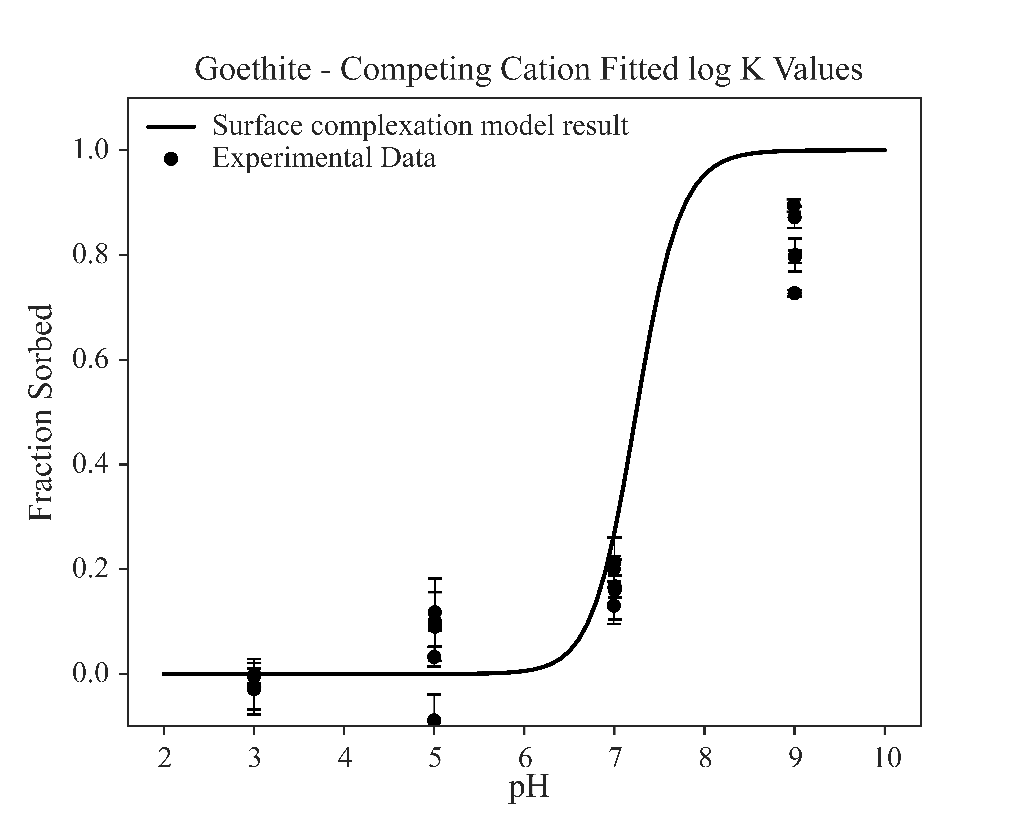
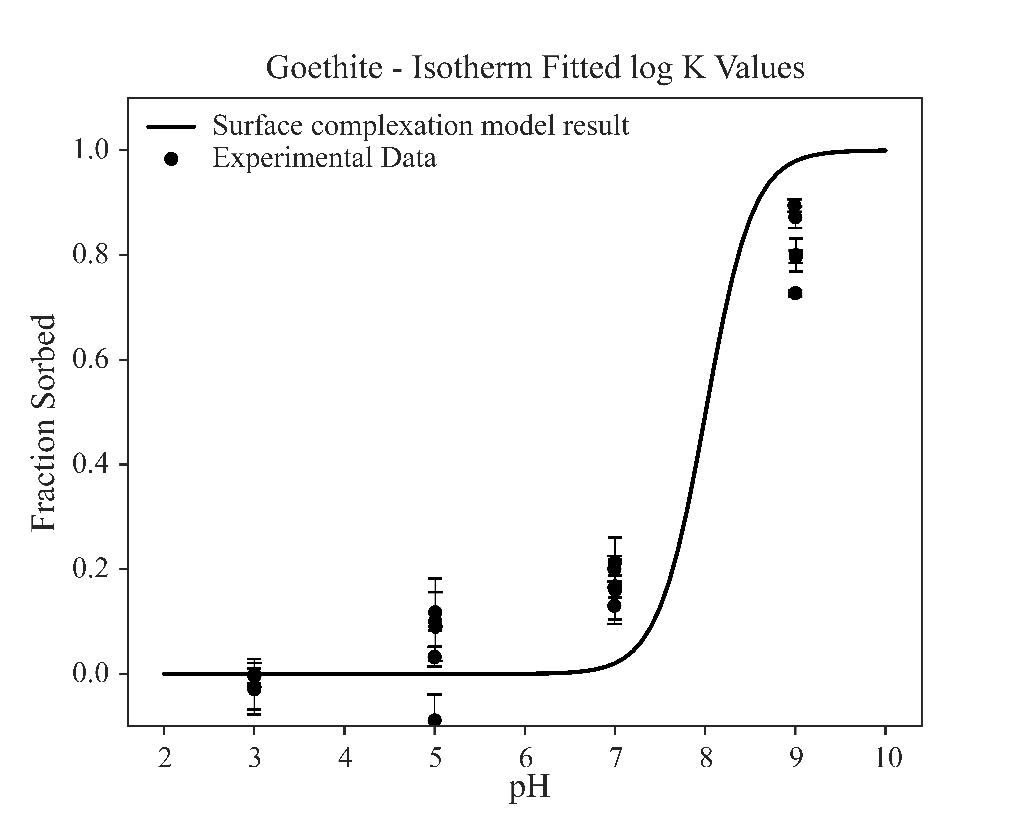


Figure A4: Comparison of goethite SCM results generated from either fitting isotherm data or fitting competing cation data. Experimental data plotted is from the isotherm experiments with 10 mM NaCl background.

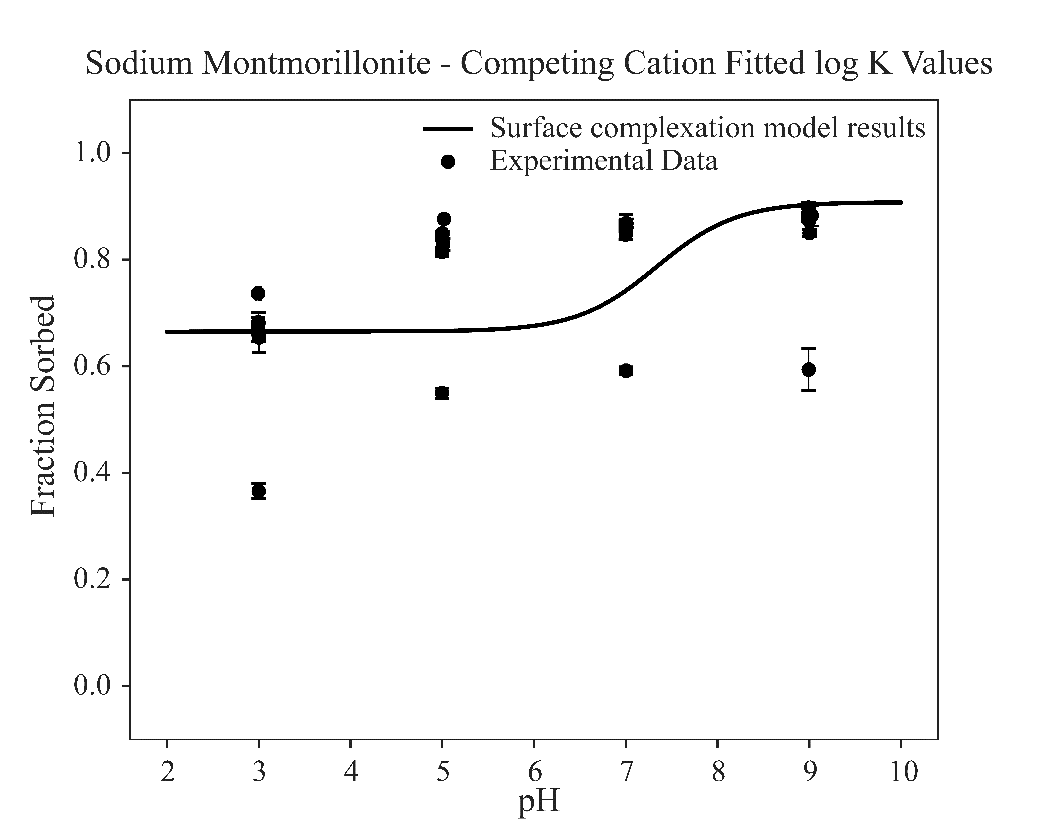
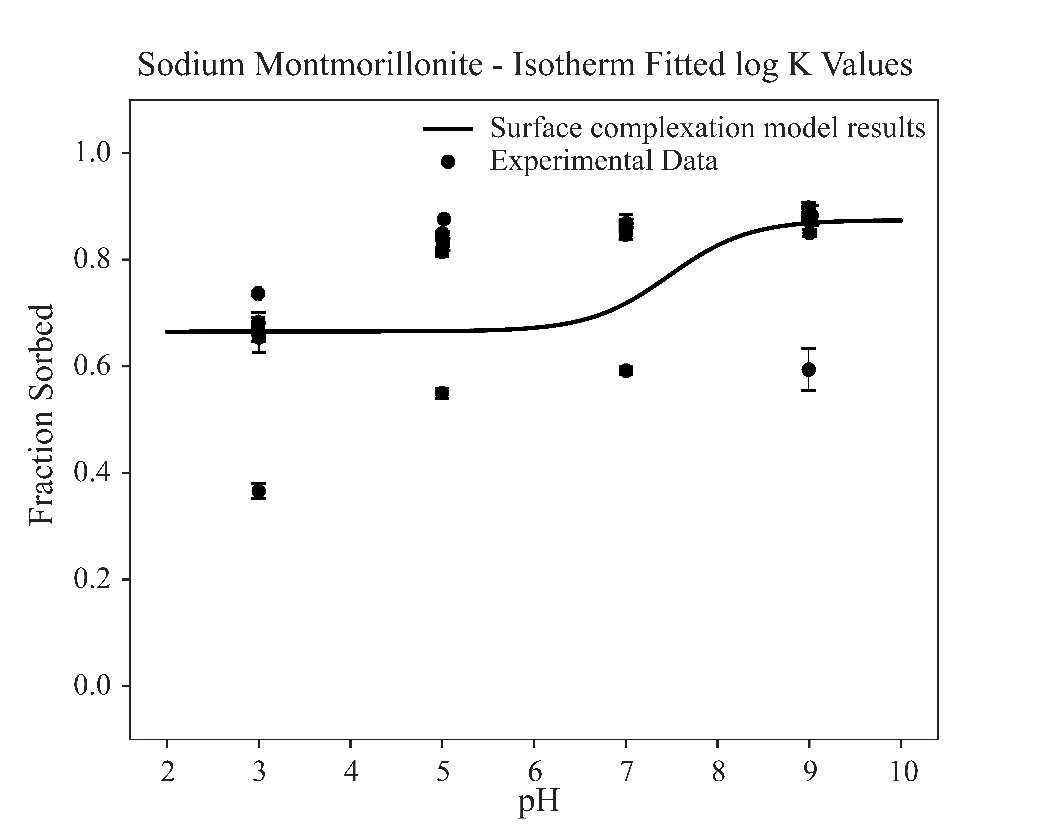


Figure A5: Comparison of sodium montmorillonite SCM results generated from either fitting isotherm data or fitting competing cation data. Experimental data plotted is from the isotherm experiments with 10 mM NaCl background.

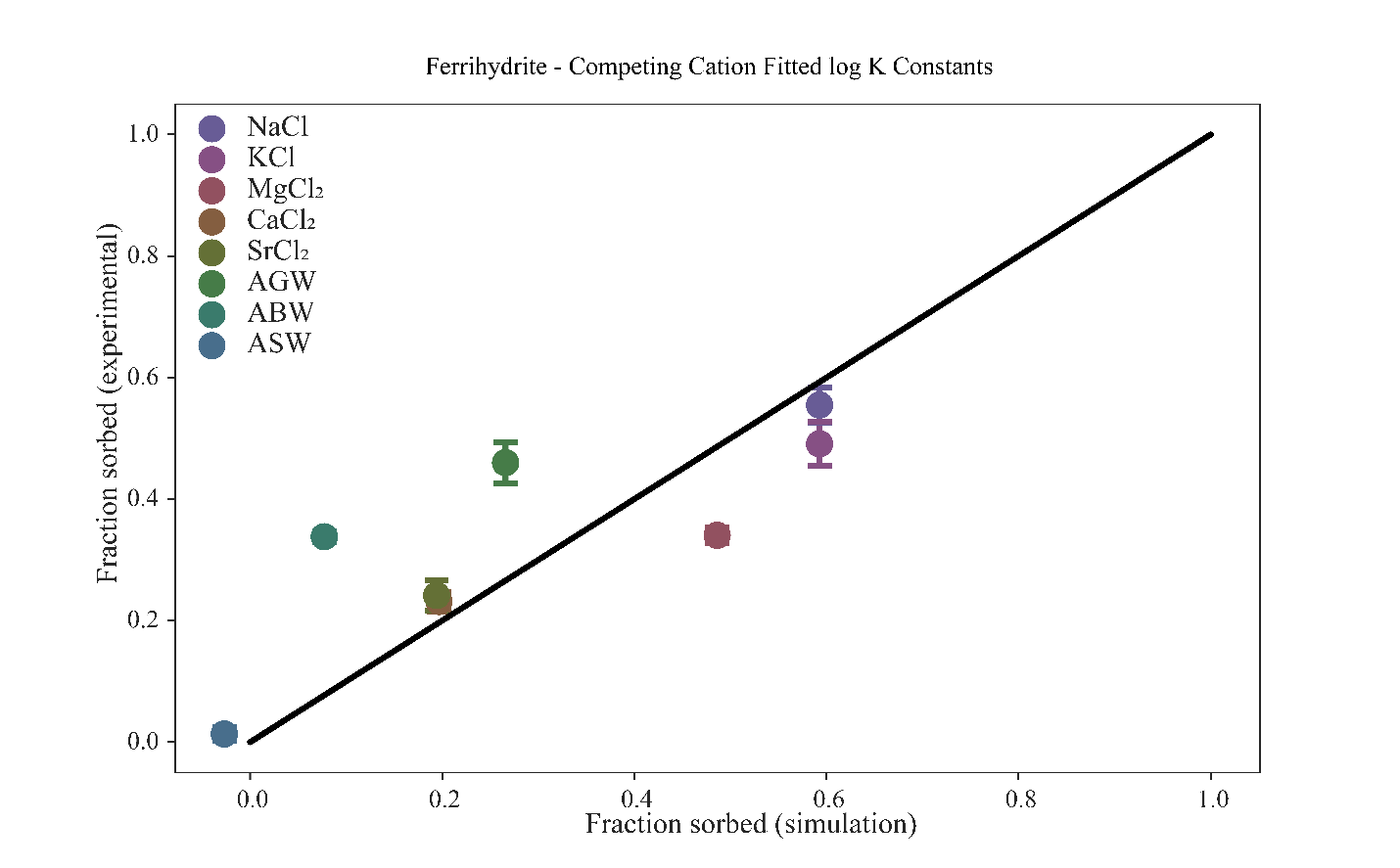
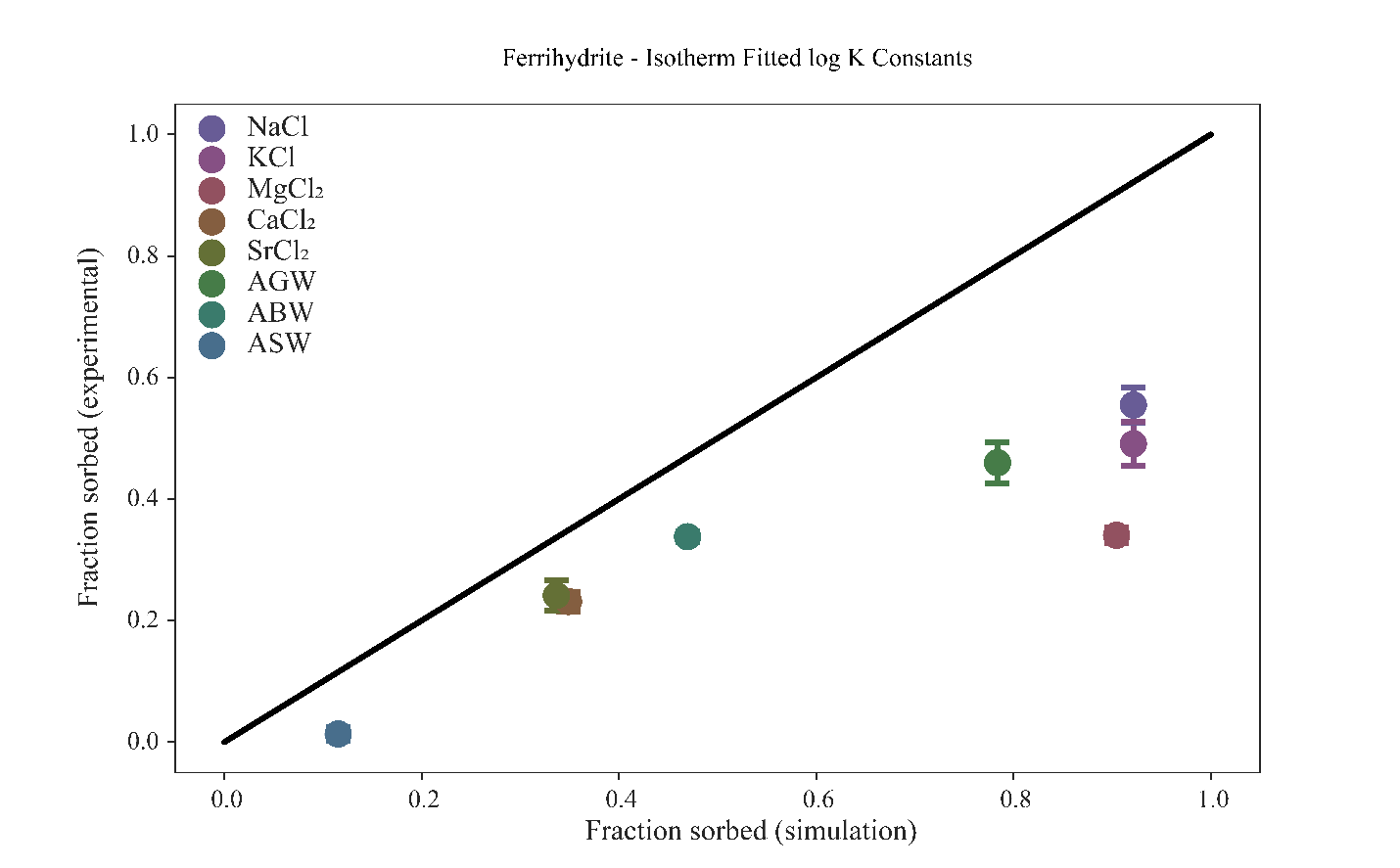
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Figure A6: Ferrihydrite SCM results from either fitting to isotherm data or competing cation data. The simulation results are compared against the relevant competing cation expeirments.

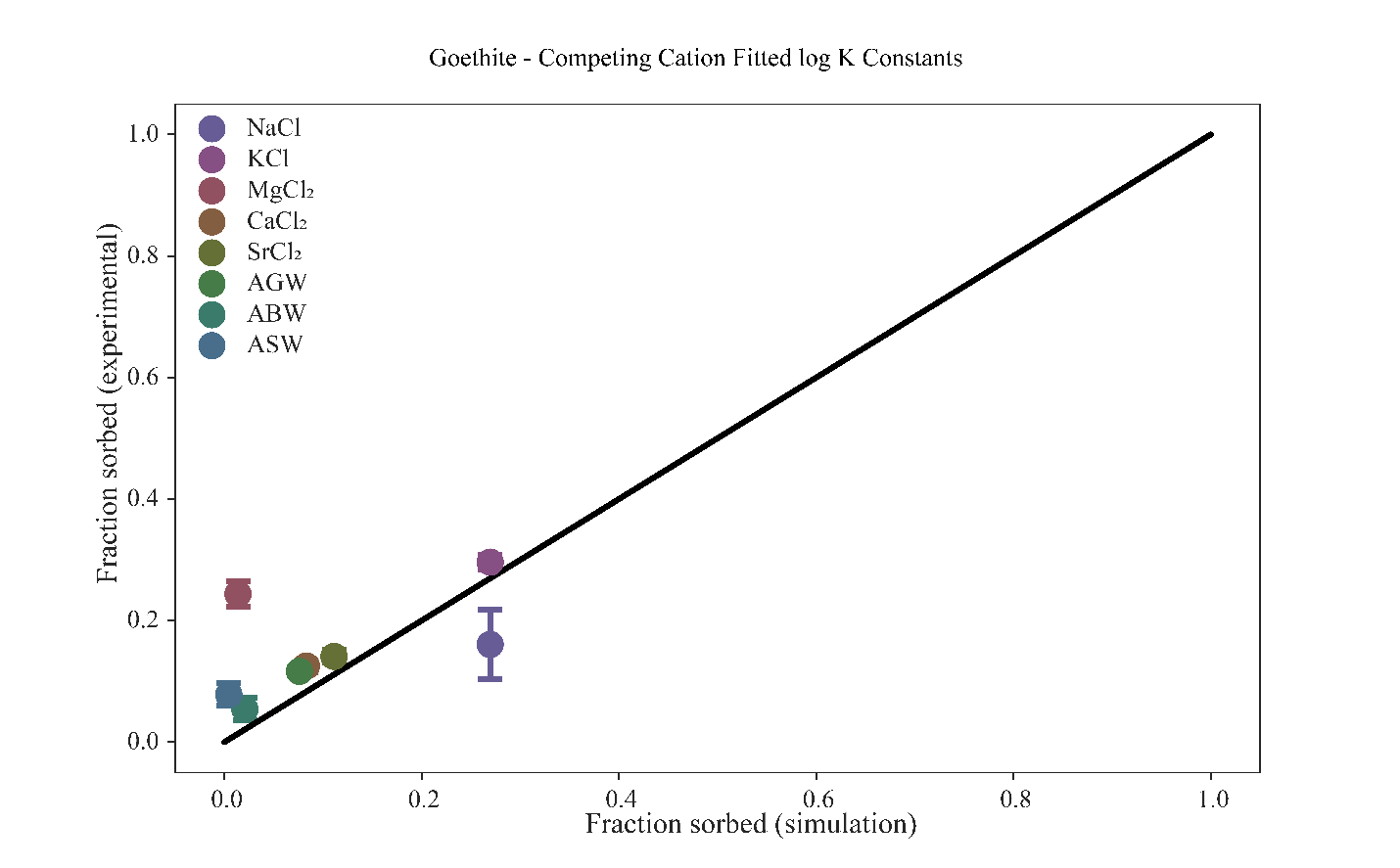
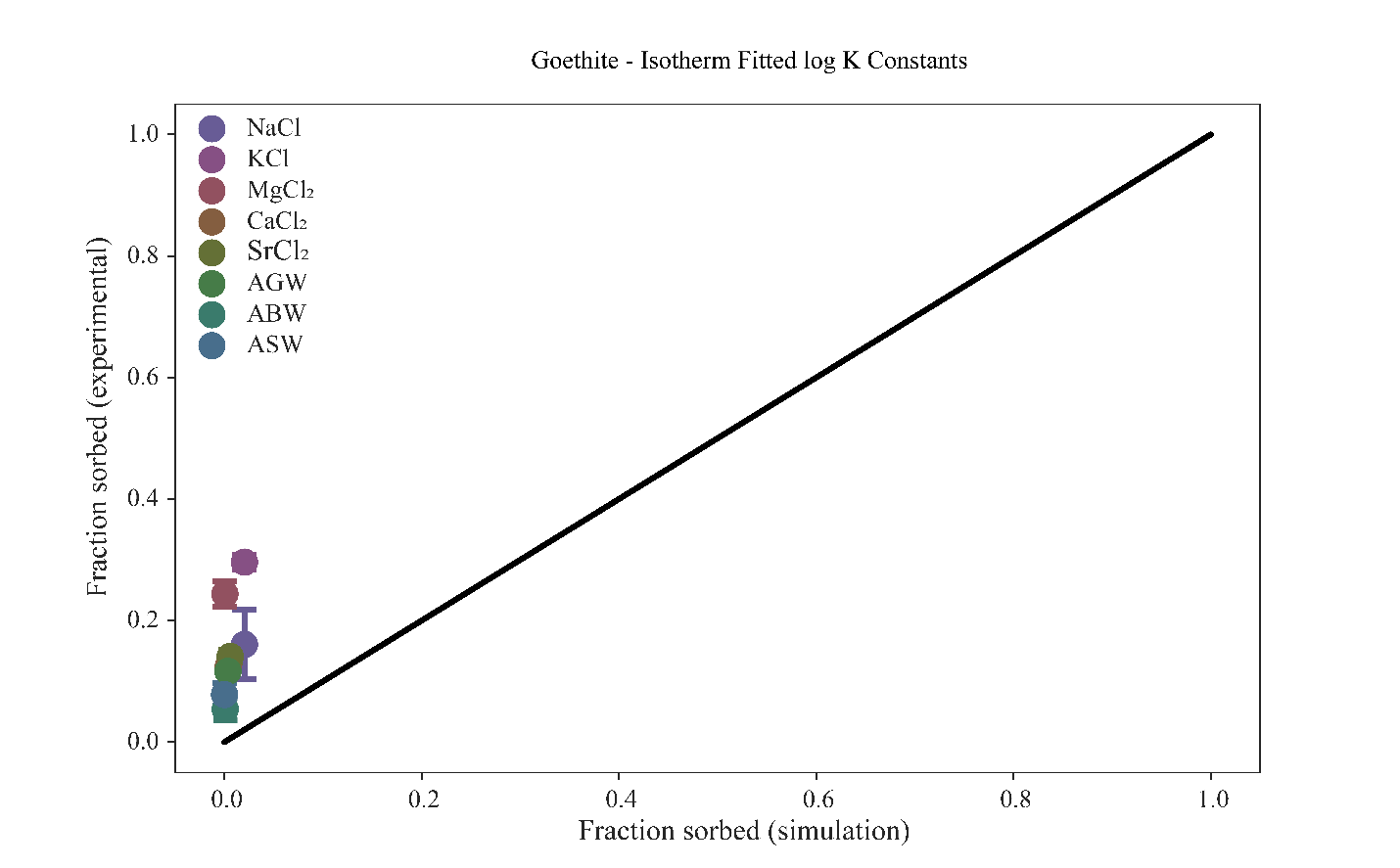
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Figure A7: Goethite SCM results from either fitting to isotherm data or competing cation data. The simulation results are compared against the relevant competing cation expeirments.

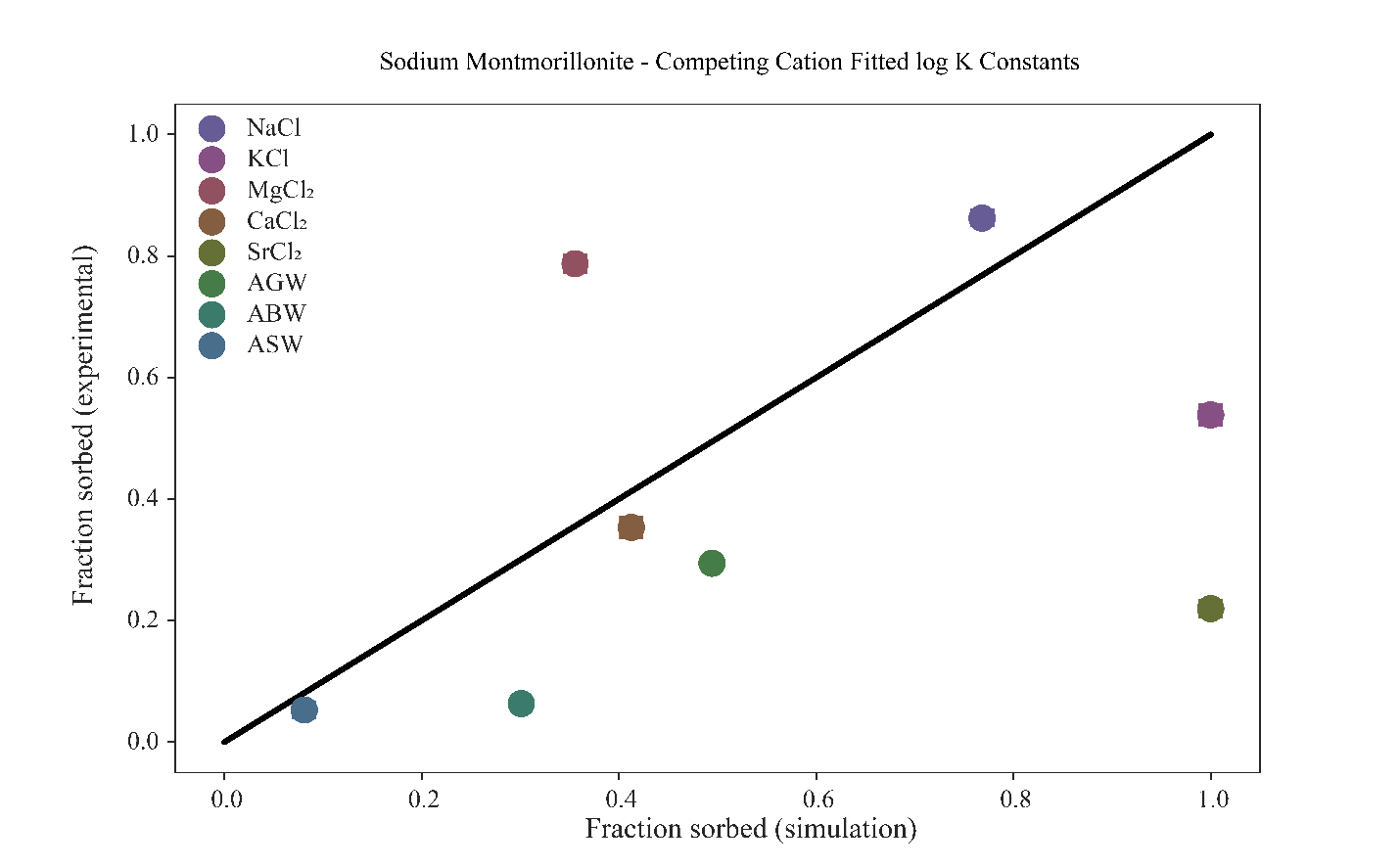
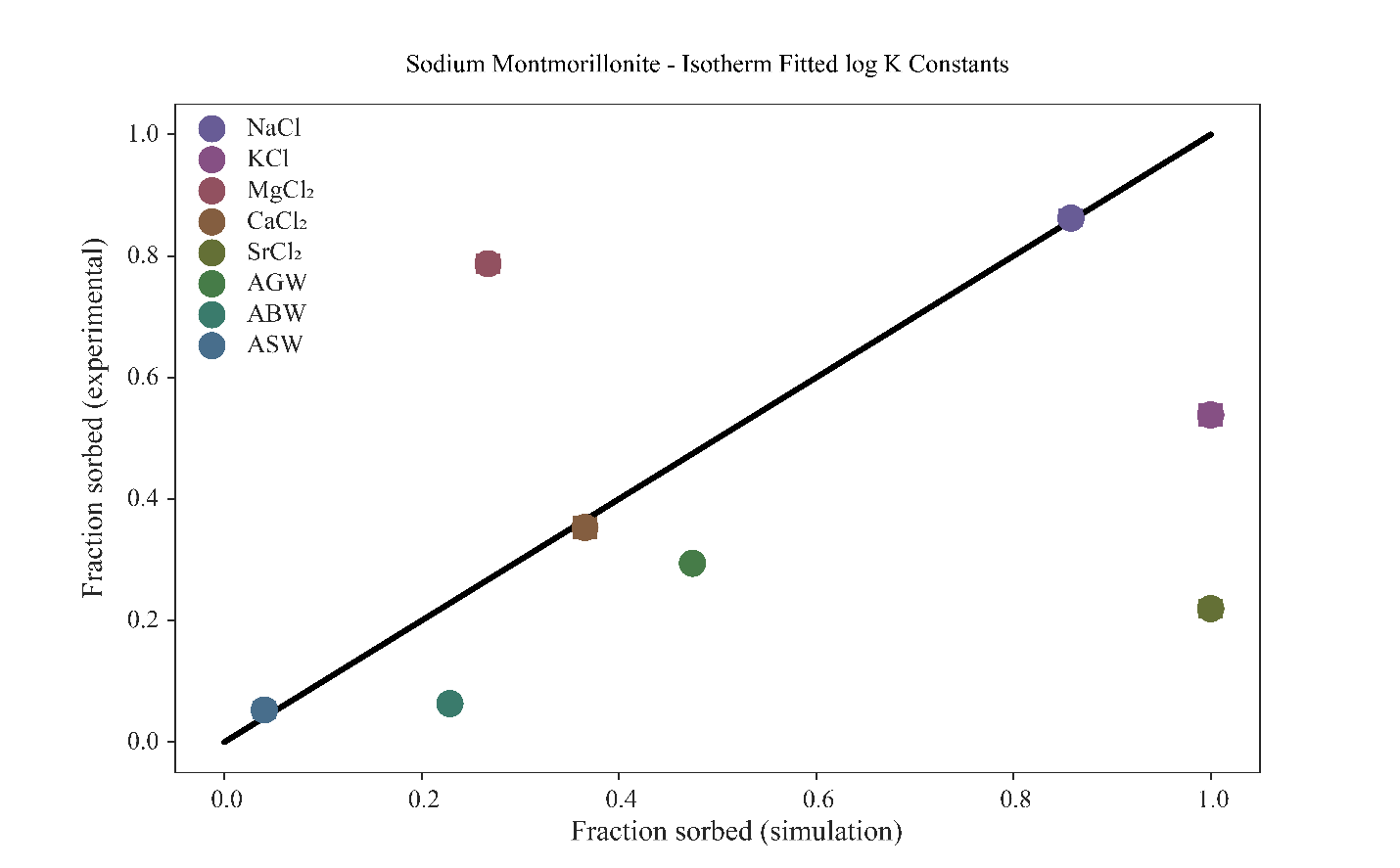
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Figure A8: Sodium Montmorillonite SCM results from either fitting to isotherm data or competing cation data. The simulation results are compared against the relevant competing cation expeirments.

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