

SUPPLEMENTARY INFORMATION FOR

An evaluation of the consistency of data and models for performance assessment: Divalent metal sorption on montmorillonite

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TABLES:Table S1. Estimated Mg + Ca concentrations ($\text{mol}\cdot\text{kg}^{-1}$) with 2SPNE SC/CE and minimalist models.

Reference	Dataset number	Estimated Mg + Ca concentrations ($\text{mol}\cdot\text{kg}^{-1}$)	
		2SPNE SC/CE model	Minimalist model
Akafia et al. (2011)	1	0.58	0.4
Baeyens and Bradbury (1997)	2	0.007	0.007
Bogolepov (2009)	3	0.48	0.4
Businelli et al. (2003)	4	0.12	0.1
Fernandes and Baeyens (2019)	5	0.007	0.007
Garcia-Miragaya and Page (1976)	6	0.007	0.007
Ghayaza et al. (2011)	7	0.4	0.4
Gu et al. (2010)	8	0.29	0.29
Lothenbach et al. (1997)	9	0.4	0.4
Zachara et al. (1993)	10	0.009	0.005
Marcussen et al. (2009)	11	0.28	0.2
Morton et al. (2001)	12	0.007	0.007
Strawn and Sparks (1999)	13	0.007	0.007
Green-Pedersen and Pind (2000)	14	0.9	0.7
Shao et al. (2009)	15	0.48	0.4
Chen and Dong (2013)	16	0.48	0.4
Orucoglu et al. (2018)	17	0.007	0.007
Tertre et al. (2005)	18	0.4	0.4
Yu et al. (2016)	19	0.48	0.4
Chen et al. (2016)	20	0.38	0.38
Wang et al. (2009)	21	0.28	0.28
Hu et al. (2018)	22	0.38	0.3
Yang et al. (2010)	23	0.28	0.28
Wen et al. (2011)	24	0.007	0.007
Hu and Tan (2012)	25	0.38	0.38
Wu et al. (2011)	26	0.58	0.4
Li et al. (2009)	27	0.58	0.4

Table S2. Distribution for the measured and predicted $\log_{10} K_D$ after removal of the outliers (the Shapiro-Wilk normality test results). “+” indicates that the data distribution is normal distribution, and “-” indicates that the distribution is not normal.

Metal	Full pH range	Low pH range	Middle pH range	High pH range
Measured $\log_{10} K_D$				
Ni	+	+	-	+
Pb	-	-	-	-
Cd	-	-	-	+
Co	-	-	-	+
Cu	-	-	+	+
Predicted $\log_{10} K_D$				
Ni	-	-	-	-
Pb	-	-	-	-
Cd	-	-	-	+
Co	-	-	-	+
Cu	-	-	-	-

Table S3. Spearman's rho correlation coefficient calculation results

		Spearman's rho			
		Full pH range	Low pH range	Middle pH range	High pH range
Ni sorption					
2SPNE SC/CE model	No correction	0.52	0.75	0.73	-0.15
	Ca+Mg correction	0.56	0.76	0.75	-0.23
Minimalist	Ca+Mg correction and mineral precipitation	0.86	0.76	0.77	0.72
	Ca+Mg correction and mineral precipitation	0.88	0.80	0.77	0.72
Pb sorption					
2SPNE SC/CE model	No correction	0.63	0.86	0.86	0.43
	Ca+Mg correction	0.66	0.82	0.87	0.41
Minimalist	Ca+Mg correction and mineral precipitation	0.86	0.82	0.87	0.64
	Ca+Mg correction and mineral precipitation	0.87	0.84	0.89	0.64
Cd sorption					
2SPNE SC/CE model	Ca+Mg correction and mineral precipitation	0.90	0.94	0.94	0.57
Minimalist	Ca+Mg correction and mineral precipitation	0.92	0.96	0.93	0.40
Co sorption					
2SPNE SC/CE model	Ca+Mg correction and mineral precipitation	0.88	0.91	0.83	0.71
Minimalist	Ca+Mg correction and mineral precipitation	0.82	0.93	0.68	0.74
Cu sorption					
2SPNE SC/CE	Ca+Mg correction and	0.87	0.86	0.84	0.78

model	mineral precipitation				
Minimalist	Ca+Mg correction and mineral precipitation	0.93	0.90	0.83	0.80

Table S4. Aqueous speciation reactions and related log K values for some species.

SOLUTION SPECIES	log K
$\text{Cu}^{+2} + \text{H}_2\text{O} = \text{Cu}(\text{OH})^+ + \text{H}^+$	-7.95 ^a
$\text{Cu}^{+2} + \text{Cl}^- = \text{CuCl}^+$	0.83 ^a
$\text{Cu}^{+2} + \text{HCO}_3^- = \text{CuCO}_3 + \text{H}^+$	-3.56 ^a
$\text{Acetate}^- + \text{Pb}^{+2} = \text{Pb}(\text{Acetate})^+$	2.87 ^b

a: PHREEQC_ThermmoddemV1.10_06Jun2017, b: Minteq database provided with PHREEQC

Table S5. General performance rating for NSE.

Performance	NSE
Very good	$0.75 < \text{NSE} \leq 1$
Good	$0.65 < \text{NSE} \leq 0.75$
Satisfactory	$0.5 < \text{NSE} \leq 0.65$
Unsatisfactory	$\text{NSE} \leq 0.5$

FIGURES

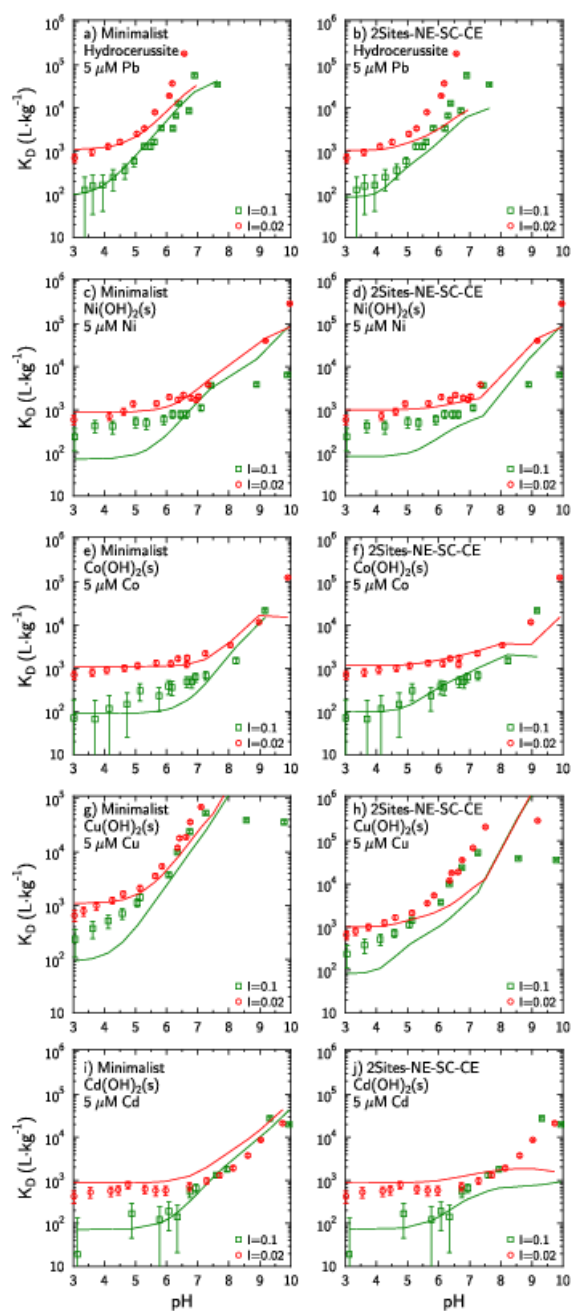


Figure S1. Modeling of Pb^{2+} , Ni^{2+} , Co^{2+} , Cu^{2+} , and Cd^{2+} sorption data from Akafia et al. (2011) using the minimalist (first column) and the 2SPNE SC/CE (second column) models for 5 μM metal sorption. Experimental conditions are available in the inner captions of the figure.

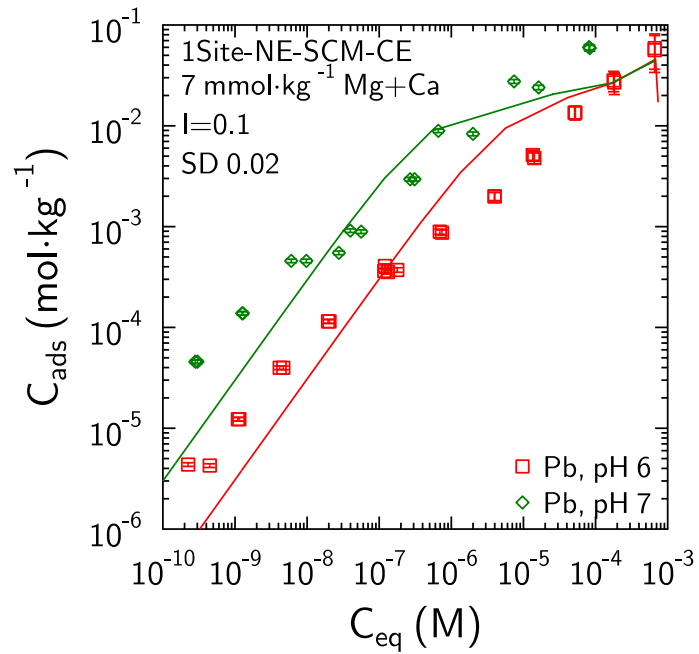


Figure S2. Model prediction (lines) of Pb^{2+} adsorption data at pH 6 and pH 7 from Marques Fernandes and Baeyens (2019), using the minimalist model. The total Mg concentration was taken at $7 \text{ mmol}_{\text{Mg}}/\text{kg}_{\text{clay}}$ because the montmorillonite mineral was acid-washed, and dissolved organic carbon was considered insignificant. Additionally, Pb precipitation was not taken into consideration.

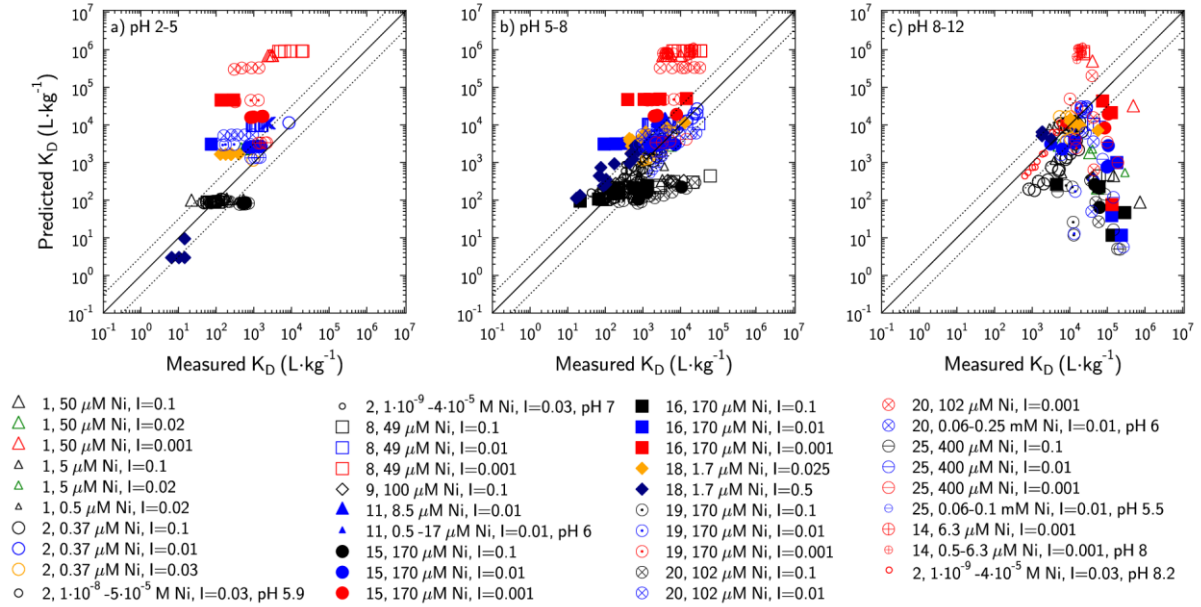


Figure S3. Comparison of measured K_D values with values predicted with the 2SPNE SC/CE model for Ni (a-c) in three different pH ranges without corrections. The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in log K_D values. Symbols and numbers in the inner caption point to the references listed in **Erreur ! Source du renvoi introuvable.**

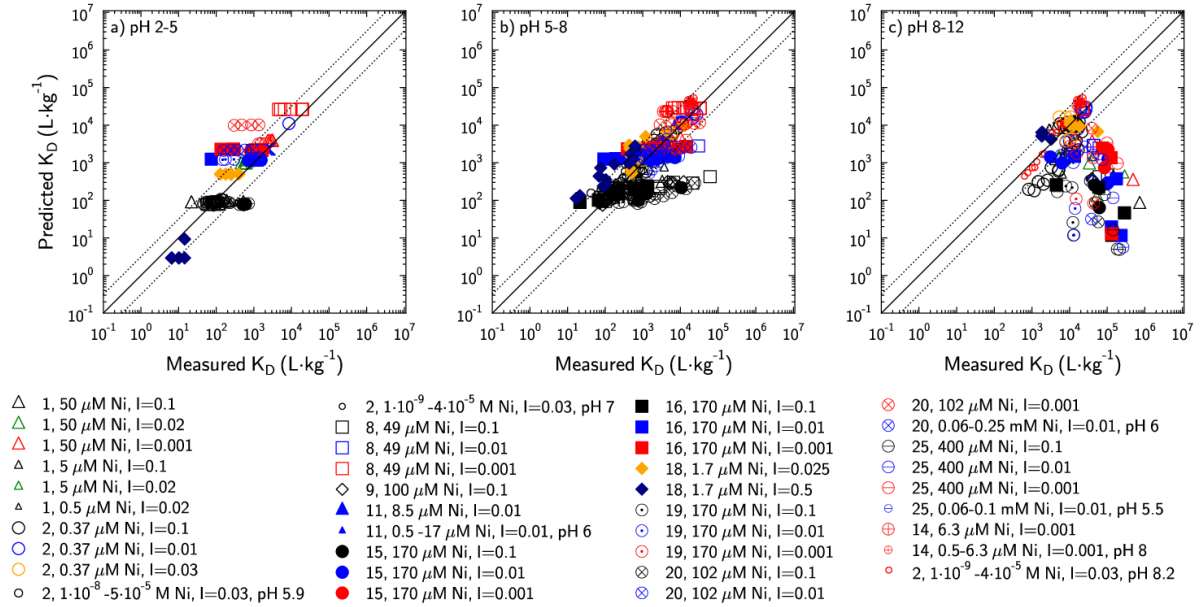


Figure S4. Comparison of measured K_D values with values predicted with the 2SPNE SC/CE model for Ni (a-c) in three different pH ranges with the consideration of Ca+Mg concentration correction. The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in log K_D values. Symbols and numbers in the inner caption point to the references listed in **Erreur ! Source du renvoi introuvable.**

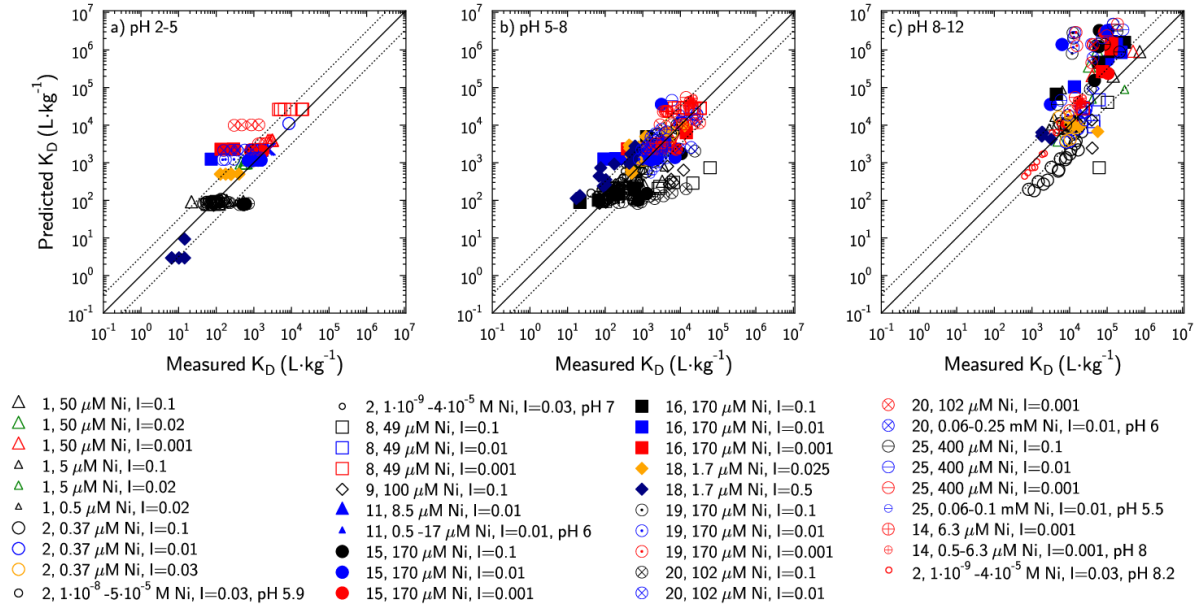


Figure S5. Comparison of measured K_D values with values predicted with the 2SPNE SC/CE model for Ni (a-c) in three different pH ranges with the consideration of corrections related to the Ca+Mg concentration to the precipitation of metal hydroxides at high pH ($\text{Ni}(\text{OH})_2(\text{s})$). The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in log K_D values. Symbols and numbers in the inner caption point to the references listed in **Erreur ! Source du renvoi introuvable.**

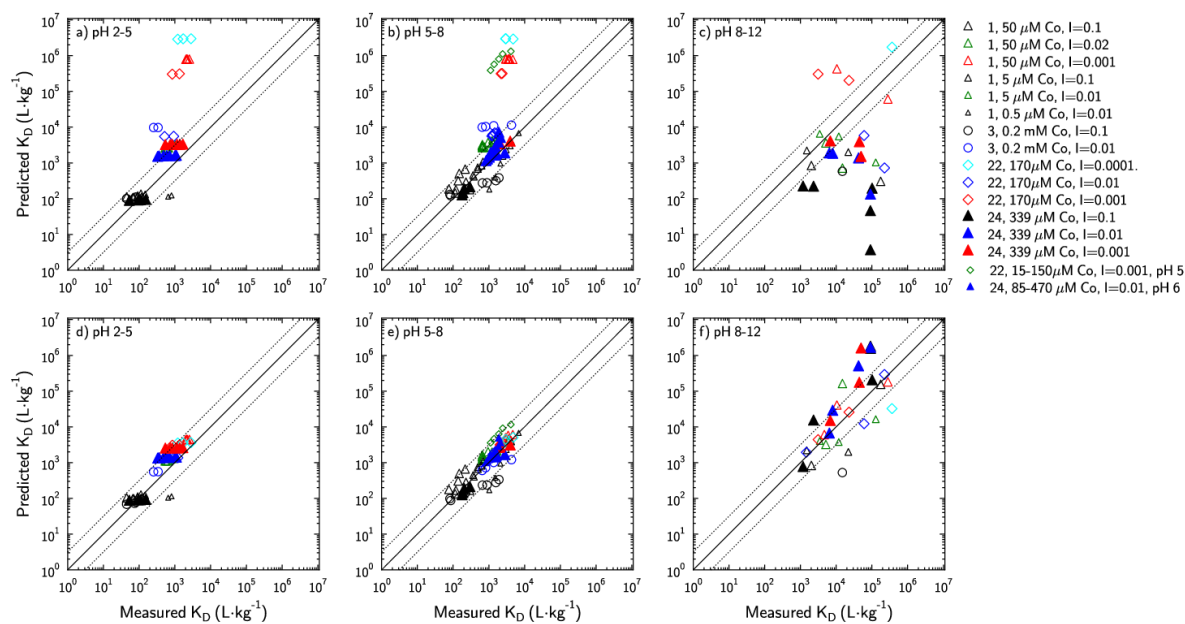


Figure S6. Comparison of measured K_D values with values predicted with the 2SPNE SC/CE model for Co in three different pH ranges without the consideration of corrections (a-c) and with the consideration of corrections (d-f) with Ca+Mg concentration and metal hydroxides precipitation at high pH. The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in $\log K_D$ values. The colors of the symbols are representative of the ionic strength. Symbols and numbers in the inner caption point to the references listed in Table 2.

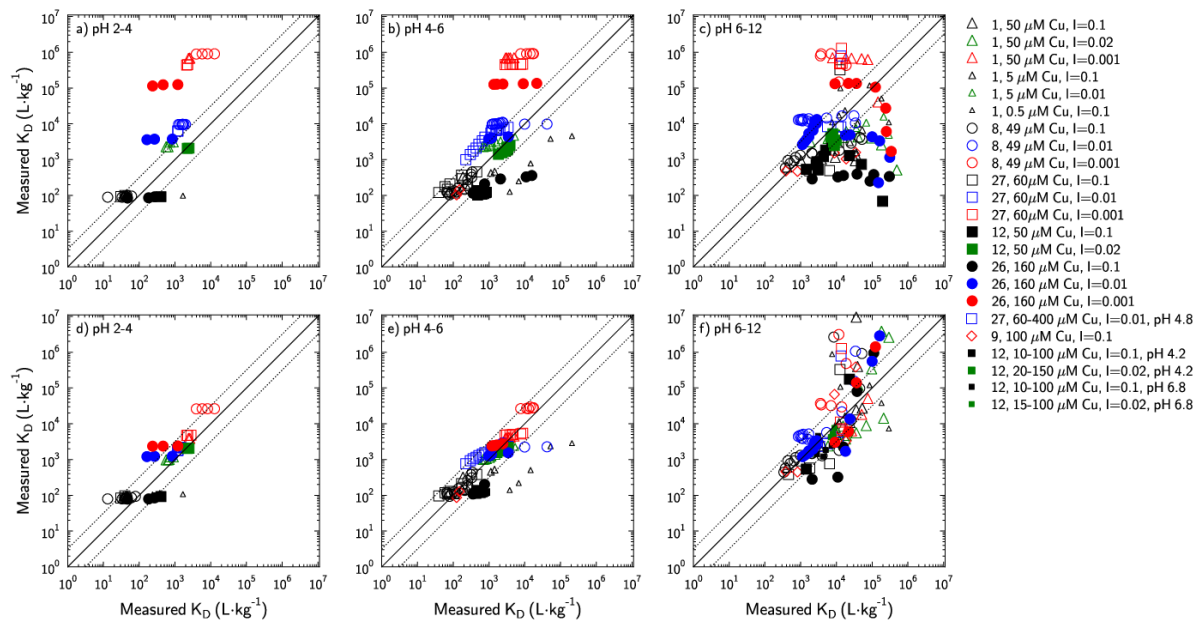


Figure S7. Comparison of measured K_D values with values predicted with the 2SPNE SC/CE model for Cu in three different pH ranges without the consideration of corrections (a-c) and with the consideration of corrections (d-f) with Ca+Mg concentration and metal hydroxides precipitation at high pH. The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in $\log K_D$ values. The colors of the symbols are representative of the ionic strength. Symbols and numbers in the inner caption point to the references listed in Table 2.

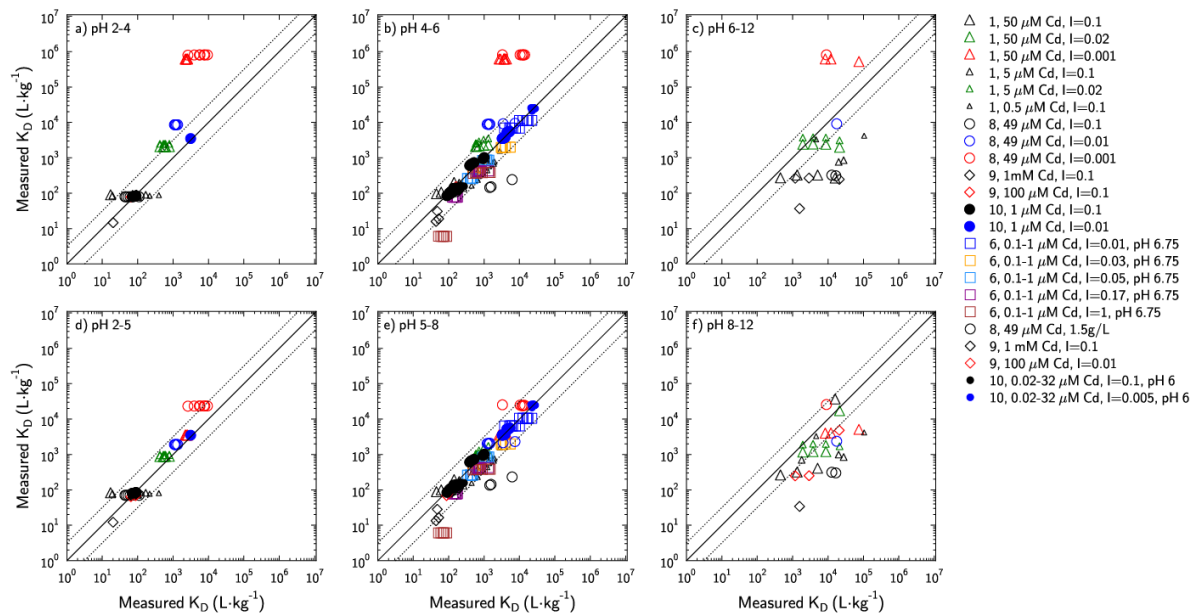


Figure S8. Comparison of measured K_D values with values predicted with the 2SPNE SC/CE model for Cd in three different pH ranges without the consideration of corrections (a-c) and with the consideration of corrections (d-f) with Ca+Mg concentration and metal hydroxides precipitation at high pH. The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in log K_D values. The colors of the symbols are representative of the ionic strength. Symbols and numbers in the inner caption point to the references listed in Table 2.

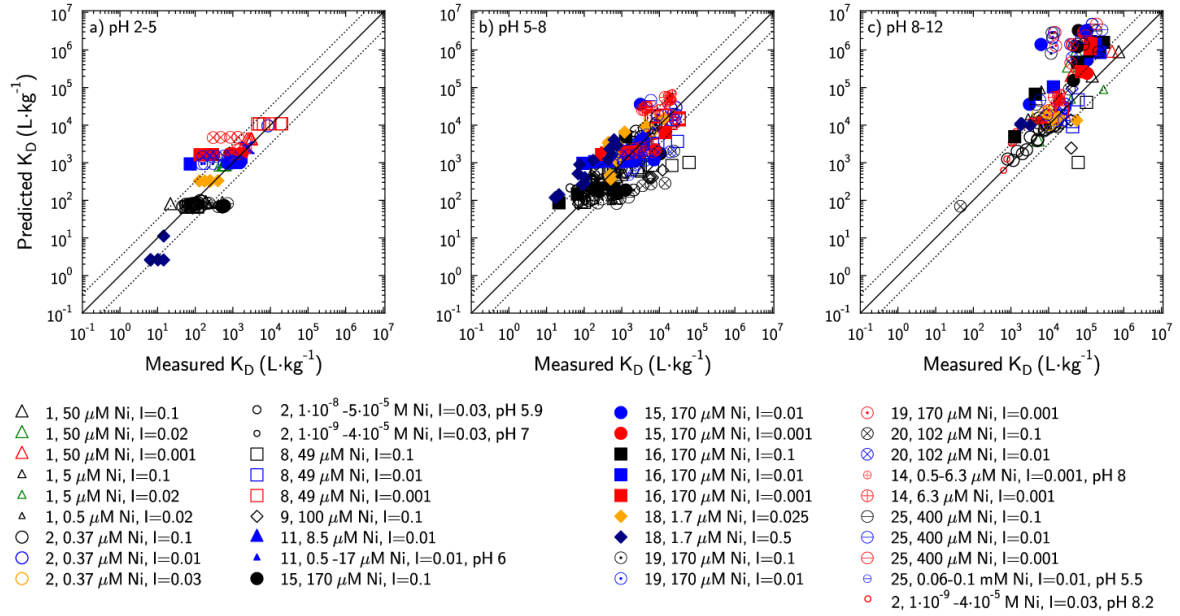


Figure S9. Comparison of measured K_D values with values modeled with the minimalist approach for Ni (a-c) in three different pH ranges with the consideration of corrections related to the Ca+Mg concentration to the precipitation of metal hydroxides at high pH ($\text{Ni}(\text{OH})_2(\text{s})$). The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in log K_D values. Symbols and numbers in the inner caption point to the references listed in **Erreur ! Source du renvoi introuvable.**

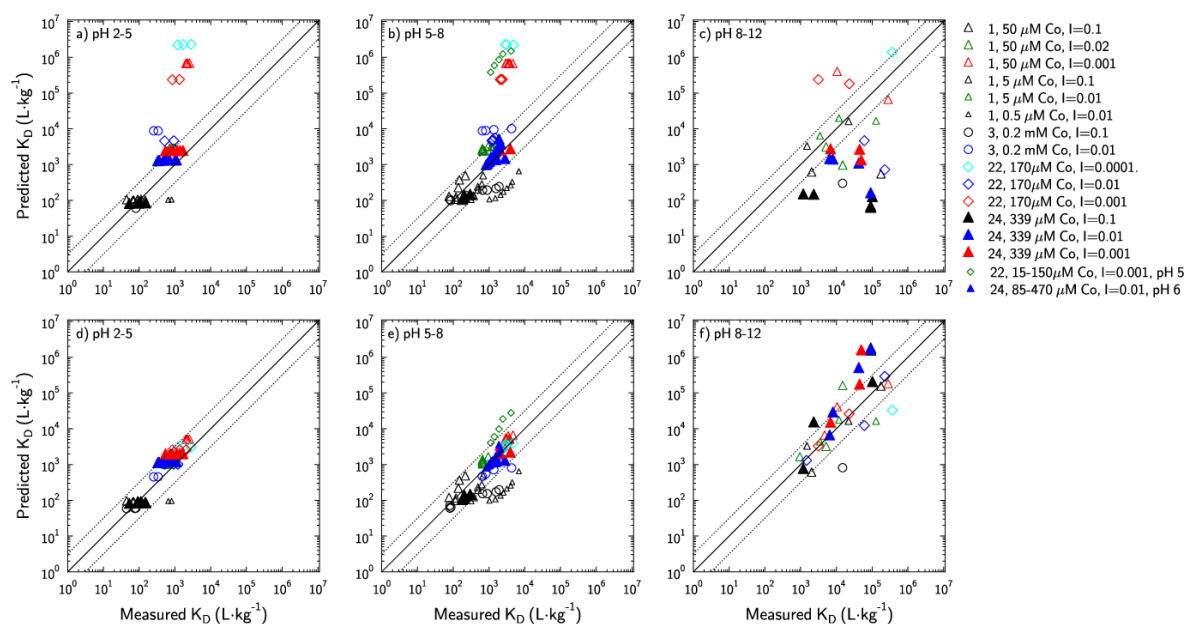


Figure S10. Comparison of measured K_D values modeled with the minimalist approach for Co in three different pH ranges without the consideration of corrections (a-c) and with the consideration of corrections (d-f) with Ca+Mg concentration and metal hydroxides e precipitation at high pH. The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in log K_D values. The colors of the symbols are representative of the ionic strength. Symbols and numbers in the inner caption point to the references listed in Table 2.

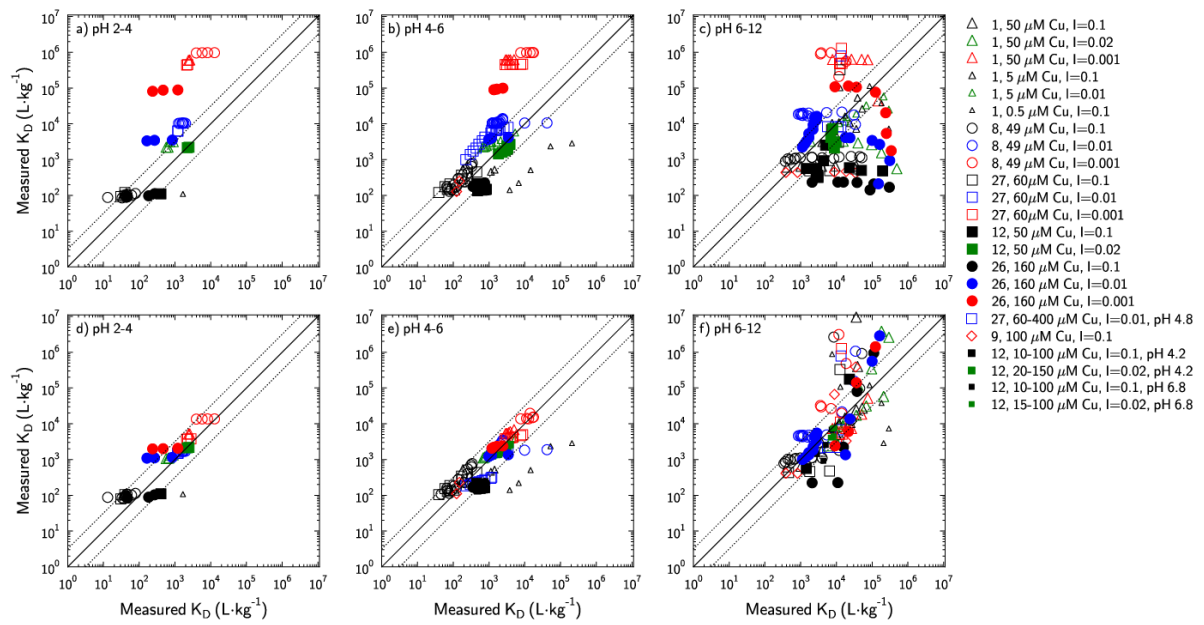


Figure S11. Comparison of measured K_D values modeled with the minimalist approach for Cu in three different pH ranges without the consideration of corrections (a-c) and with the consideration of corrections (d-f) with Ca+Mg concentration and metal hydroxides precipitation at high pH. The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in log K_D values. The colors of the symbols are representative of the ionic strength. Symbols and numbers in the inner caption point to the references listed in Table 2.

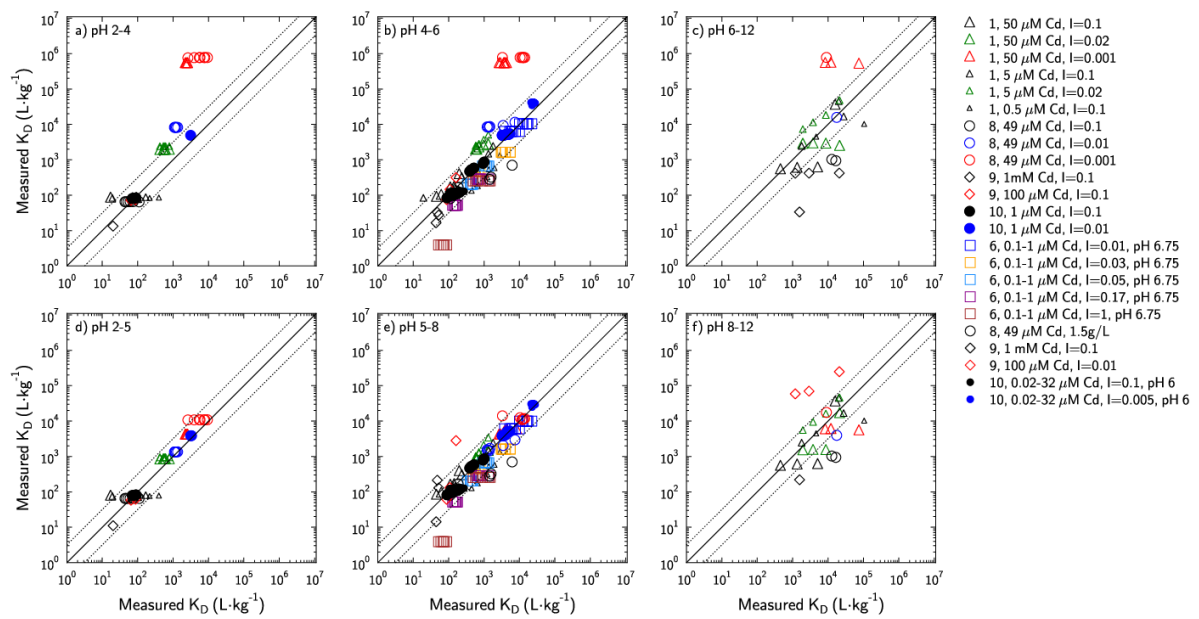


Figure S12. Comparison of measured K_D values modeled with their minimalist approach for Cd in three different pH ranges without the consideration of corrections (a-c) and with the consideration of corrections (d-f) with Ca+Mg concentration and metal hydroxides precipitation at high pH. The plain line shows the 1:1 relationship between the predicted and the measured K_D . The dashed lines are indicative of an ± 0.5 difference in $\log K_D$ values. The colors of the symbols are representative of the ionic strength. Symbols and numbers in the inner caption point to the references listed in Table 2.