**SupplementaryMaterials:** Nuclear magnetic resonance analysis of changes in dissolved organic matter composition with successive layering on clay mineral surfaces

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**Figure S1**. Solution-state <sup>1</sup>H nuclear magnetic resonance (NMR) spectra of unbound dissolved organic matter (DOM) remaining in solution after sorption to kaolinite over ten successive DOM loading experiments. For comparison, all spectra are scaled to the same maximum intensity. The resonance at 2.50 ppm is from the NMR solvent (DMSO-d<sub>6</sub>).

## **Montmorillonite**



**Figure S2**. Solution-state <sup>1</sup>H nuclear magnetic resonance (NMR) spectra of unbound dissolved organic matter (DOM) remaining in solution after sorption to montmorillonite over ten successive DOM loading experiments. For comparison, all spectra are scaled to the same maximum intensity. The resonance at 2.50 ppm is from the NMR solvent (DMSO-d<sub>6</sub>).



**Figure S3**. Solution-state <sup>1</sup>H nuclear magnetic resonance (NMR) spectra of unbound dissolved organic matter (DOM) remaining in solution after sorption to gibbsite over ten successive DOM loading experiments. For comparison, all spectra are scaled to the same maximum intensity. The resonance at 2.50 ppm is from the NMR solvent (DMSO-d<sub>6</sub>).

## Kaolinite



**Figure S4**. Solution-state diffusion-edited <sup>1</sup>H nuclear magnetic resonance (NMR) spectra of unbound dissolved organic matter (DOM) remaining in solution after sorption to kaolinite over ten successive DOM loading experiments. For comparison, all spectra are scaled to the same maximum intensity. The NMR solvent is DMSO-d<sub>6</sub>.

## **Montmorillonite**



**Figure S5**. Solution-state diffusion-edited <sup>1</sup>H nuclear magnetic resonance (NMR) spectra of unbound dissolved organic matter (DOM) remaining in solution after sorption to montmorillonite over ten successive DOM loading experiments. For comparison, all spectra are scaled to the same maximum intensity. The NMR solvent is DMSO-d<sub>6</sub>.

## **Gibbsite**



**Figure S6**. Solution-state diffusion-edited <sup>1</sup>H nuclear magnetic resonance (NMR) spectra of unbound dissolved organic matter (DOM) remaining in solution after sorption to gibbsite over ten successive DOM loading experiments. For comparison, all spectra are scaled to the same maximum intensity. The NMR solvent is DMSO-d6.



**Figure S7**. Integration results for seven types of dissolved organic matter (DOM) components in the solution-state <sup>1</sup>H nuclear magnetic resonance (NMR) spectra of the initial DOM and the unbound DOM samples following sorption to (a) kaolinite, (b) montmorillonite and (c) gibbsite over ten successive DOM loading experiments. The peak area of each region is expressed as a percentage of the total <sup>1</sup>H NMR signal across the seven regions.



**Figure S8**. Integration results for seven types of dissolved organic matter (DOM) components in the solution-state diffusion-edited <sup>1</sup>H nuclear magnetic resonance (NMR) spectra of the initial DOM and the unbound DOM samples following sorption to (a) kaolinite, (b) montmorillonite and (c) gibbsite over ten successive DOM loading experiments. The peak area of each region is expressed as a percentage of the total <sup>1</sup>H NMR signal across the seven regions. Diffusion-edited <sup>1</sup>H NMR experiments provide information about relatively large molecular components within the DOM and complements information obtained from <sup>1</sup>H NMR spectra.



**Figure S9**. Percent change in integrated solution-state diffusion-edited <sup>1</sup>H nuclear magnetic resonance (NMR) signal area of seven classes of dissolved organic matter (DOM) components between the initial DOM and unbound DOM samples following sorptive fractionation by (a) kaolinite, (b) montmorillonite and (c) gibbsite over ten successive DOM loading experiments. A positive percent change indicates sorption of a component to the organo-clay complex, whereas a negative percent change indicates that a component accumulated in solution and was not sorbed. Diffusion-edited <sup>1</sup>H NMR experiments provide information about relatively large molecular components within the DOM and complements information obtained from <sup>1</sup>H NMR spectra.