## Supplementary Information

### $^2$H and $^{27}$Al Solid-State NMR Study of the Local Environments in Al-Doped 2-Line Ferrihydrite, Goethite, and Lepidocrocite

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### Table S1

| Configuration    | Unscaled $^{27}$Al shifts / $10^5$ ppm | Scaled $^{27}$Al shifts / ppm |
|------------------|----------------------------------------|-------------------------------|
|                  | Al atom 1 | Al atom 2 | Al atom 1 | Al atom 2 |
| Goethite         |           |           |           |           |
| Single           | 3.02      | 1.67      | 662       | 366       |
| 1NN              | 1.67      | 1.67      | 366       | 366       |
| 2NN              | 3.70      | 3.70      | 813       | 813       |
| 3NN              | 2.42      | 2.58      | 532       | 565       |
| Lepidocrocite    |           |           |           |           |
| Single           | 5.20      | 5.20      | 4760      | 4700      |
| 1NN              | 5.20      | 5.21      | 4690      | 4700      |
| 2NN              | 5.27      | 5.33      | 4750      | 4810      |
| 3NN              | 1.61      | 1.60      | 1450      | 1450      |

Table S1. Calculated $^{27}$Al unscaled and scaled hyperfine shifts for Al-doped goethite and lepidocrocite configurations with 35% Hartree-Fock exchange (results for 20% HF exchange displayed in main manuscript).
Figure S1. Results from the first principles calculation of the \(^2\)H hyperfine shifts using 35% Hartree-Fock exchange in (a) goethite and (b) lepidocrocite. The shifts for the unique \(^2\)H nucleus in the pristine material are quoted while the individual \(^2\)H shifts for sites surrounding a single Al dopant are labeled on the associated structures.

Figure S2. Magnetization curves for all of the goethite series as a function of temperature.