Supplemental Material

Phosphorus containing water dispersible nanoparticles in arable soil

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Figure S1. Size-calibration of AF4.

Figure S2. AF4-fractograms of fine colloids of soil from fallow area dispersed in the aqueous soil extract, after oxalate and DCB treatments.

Table S1. Soil physicochemical characteristics.
AF4-ICP-MS calibration

Collision cell ICP-MS was operated in He-Mode monitoring $^{31}$P, $^{27}$Al, $^{28}$Si, and $^{57}$Fe. Internal standard solution (as 100 µg L$^{-1}$ Rh in 0.5% HCl) was added via a T-piece post-channel before entering the ICP-nebulizer. Briefly, calibration was performed while AF4 was operated at 0.5 mL min$^{-1}$ detector flow without sample injection by replacing the internal standard solution by calibration standards containing the same amount of Rh and hydrochloric acid, but different levels of the elements of interest. Based on the obtained ICP-MS intensities for the calibration standards and on the flow rates the fractograms of the samples were converted into mass flow data.

TEM-EDX Characterization

In order to allocate the elements within and on the nanoparticles, the soil suspension ($d_{\text{average}} = 180$ nm) before and after DCB treatments was characterized by TEM. The soil suspension was centrifuged to obtain a suitable concentration of nanoparticles for TEM measurement. The resulting pellet was then diluted in deionized water so that a turbid suspension was obtained. One to two drops of the suspension were deposited on a Cu specimen grid coated with an ultrathin layer of carbon (Plano GmbH) and air dried. After evaporation of the water, micrographs were obtained using a Philips CM 20 FEG operating at 200 keV outfitted with an EDAX-detector (EDAX, Genesis) for EDX. With this equipment high resolution TEM images of the nanoparticles were obtained as well as chemical identification and quantification of various elements (Ca, Si, Al, P, and Fe) in nanoparticles were achieved.
Supplemental Fig. S1. Asymmetric flow field-flow fractionation (AF4)-fractograms of sulfate latex nanoparticles (Postnova Analytics) with a nominal diameter of 20, 60, 100, and 200 nm with the same AF4 method as soil nanoparticles with average size of 180 nm. The fractograms show the ultraviolet (UV) absorbance and the hydrodynamic diameters by dynamic light scattering (DLS).

Comparison between cropped and bare fallow soils

The pH values of these two soils were similar (7.64 for the arable soil and 7.54 for the bare fallow soil) (Table S1). The contents of TOC, TN, Fe, Al, and Ca were higher in the cropped soil than in the bare fallow soil (Table S1). However, the P content was slightly lower in the cropped soil compared to the bare fallow soil. Ca contents in both two soils were significantly lower than Fe and Al, which means that Fe and Al minerals were important carriers for P compared to Ca-minerals in these two soils. When comparing the AF4-UV-ICP-MS result of cropped site with that of the bare fallow site, similar findings were made. As shown in Supplemental Fig. S2, the same shifts in peak size and element distributions among nanoparticles were observed in the bare fallow site as in the cropped soil (Fig. 1d-f). Hence, 10 years of cropping, or, vice versa, 10 years of bare fallow did not affect the amount and properties of water-dispersible soil fine colloids in the Luvisol topsoils. This finding strongly indicates that the formation and fate of water dispersible nanoparticles and fine colloids were
independent from the recent growth of vegetation, possibly because these particles were older than 10 years.

Supplemental Fig. S2. Asymmetric flow field-flow fractionation (AF4)-fractograms of fine colloids of soil from fallow area (without plants for at least 10 years) dispersed in the aqueous soil extract, in the same extract after oxalate treatment and in the same extract after dithionite (DCB) treatment, and the Fe, Al, P, and Si mass flow monitored by inductively coupled plasma mass spectrometer (ICP-MS) in the aqueous soil extract (a), in the same extract after oxalate treatment (b) and in the same extract after DCB treatment (c).
### Supplemental Table S1. Soil physicochemical characteristics.

<table>
<thead>
<tr>
<th>Soil Type</th>
<th>pH</th>
<th>TOC (g kg⁻¹)</th>
<th>TN (g kg⁻¹)</th>
<th>TP (g kg⁻¹)</th>
<th>TFe (g kg⁻¹)</th>
<th>TAl (g kg⁻¹)</th>
<th>TCa (g kg⁻¹)</th>
<th>P_ox (g kg⁻¹)</th>
<th>Fe_ox (g kg⁻¹)</th>
<th>Al_ox (g kg⁻¹)</th>
<th>P_DCBI (g kg⁻¹)</th>
<th>Fe_DCBI (g kg⁻¹)</th>
<th>Al_DCBI (g kg⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cropped soil</td>
<td>7.64</td>
<td>11.8±0.20</td>
<td>1.48±0.03</td>
<td>0.75</td>
<td>23.4</td>
<td>46.8</td>
<td>4.1</td>
<td>0.41±0.01</td>
<td>3.75±0.10</td>
<td>0.88±0.03</td>
<td>0.53±0.02</td>
<td>11.6±0.5</td>
<td>1.03±0.08</td>
</tr>
<tr>
<td>Bare fallow soil</td>
<td>7.54</td>
<td>8.8±0.31</td>
<td>1.31±0.09</td>
<td>0.88</td>
<td>20.2</td>
<td>42.7</td>
<td>3.8</td>
<td>0.54±0.04</td>
<td>3.70±0.10</td>
<td>0.93±0.07</td>
<td>0.62±0.04</td>
<td>10.1±0.6</td>
<td>0.95±0.06</td>
</tr>
</tbody>
</table>

- a Soil : water =1 : 2.5.
- b Total organic carbon and total nitrogen values of bulk soil are average of three different experiments ± standard deviation.
- c Total P, Fe, Al, and Ca of bulk soil.
- d Oxalate extractable P, Fe, and Al of bulk soil.
- e Dithionite (DCB) extractable P, Fe, and Al of bulk soil.