SUPPLEMENTAL MATERIALS

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Direct Injection of Aluminum–Organic Matter Flocs to Reduce Soil Permeability and Create a Vertical Flow Barrier In Situ

J. Zhou, S. Laumann, and T. J. Heimovaara

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Text S1. 1D COLUMN EXPERIMENTS

14 **Experimental set-up and testing procedure**

The column used for the column experiments is made of polyether ether ketone (PEEK), with a height of 22 cm and an inner diameter of 5.4 cm. In order to prevent the spill of sand while allowing the passage of injected Al-OM flocs, circular filters with a diameter of 23.90 mm were placed at both ends of the column. Five pressure sensors were installed at various heights of the column in order to measure the local pressure in the sand column and thus enable the determination of the local hydraulic conductivity continuously. The configuration of the pressure sensors is shown in Figure S1.

The testing procedure is summarised as: 1) packing the column; 2) initial tracer test (using 1 M NaCl solution); 3) injection of the Al-OM floc suspension; 4) resting the column for 1 hour; 5) flushing the column at low flow velocity with background solution (1 mM NaCl) and 6) Extracting the sand material out of the column for visual inspection of floc distribution.

The column is dry-packed with Dorsilit nr. 8 (99.1 % SiO2, grain size: 0.3-0.8 mm, average 26 density: 2.63 g/cm³, Eurogrit, The Netherlands), with an increase in the sand level of 1 cm at a 27 time. Therefore, the packing of the entire column was divided into 22 steps. The mass of sand in 28 each column is kept as constant at a value of 846.64 g. After the packing, the sand column was 29 connected to a vacuum pump with the aim to remove all air. Subsequently, the sand column was 30 flushed with $C O_2$ for two t imes. The column was then saturated by injection of water from the 31 bottom at a low rate, i.e., 3.15 m/d. Tracer test using 1 M of NaCl solution at an injection velocity 32 of 25.2 m/d was performed with each column to determine the respective effective porosity. The 33 effective porosity of the sand column was in the range of 0.37-0.38. 34

³⁵ A high-precision syringe pump (260D ISCO, Beun De Ronde, The Netherlands) was used to ³⁶ supply the Al-OM flocs suspension and to perform tracer and flushing te st. In order to test the ³⁷ effect of different injection velocities on the hydraulic conductivity reduction, experiments were ³⁸ carried out with four different injection velocities ranging from 12.6 to 66.7 m/d. For each column ³⁹ injection, 2 1 of Al-OM flocs suspension at an input concentration of 1 g/l was i njected. This

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40 corresponds to approximately 10 pore volumes. All injection tests were performed in duplicate.

Another set of column experiments was designed to test the effect of different input concentration. In this set of tests, the column was packed with sand took from the field site (grain size distribution of this sand can be found in Figure S4) and the injection velocity was set at 61.9 m/d. Al-OM flocs suspension was prepared at concentrations of 0.1, 0.5, 1, 3, and 5 g/l. Due to the limited availability of field sand, this set of tests was performed without duplicate. Therefore, results from this set were used qualitatively.

47 Data acquisition and analysis

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⁴⁸ During the experiments the change in hydraulic conductivity was continuously monitored. The ⁴⁹ pressure sensors (measurement interval of 1 s) monitored the pressure drop along the column. The ⁵⁰ hydraulic conductivity is calculated by applying the following equation:

$$K = \frac{Q}{A} * \frac{L}{dH}$$

where Q is the volumetric injection rate $[L^3/T]$; A is the cross-sectional area perpendicular to the flow $[L^2]$, L is the length of the transport length [L], and dH is the average gradient between the two measurement points along the column [L].

⁵⁵ The reduction in hydraulic conductivity (HCR) is calculated as follows:

$$HCR = \frac{K_{Ini}}{K_{End}}$$

where K_{Ini} is the initial hydraulic conductivity [L/T] and K_{End} the hydraulic conductivity [L/T]at the end of the injection.









Notes	Derived from falling head tests in the lab with sample recovered from drilling	Representative value	Representative value	Representative value	Collected from field observation	Collected from field observation	Fitted against field monitoring data	Fitted against field monitoring data	Fitted against field monitoring data
Units	m/d	·	1/m		ш	ш	1/d	1/d	1/d
Value	9	0.33	3.2	1.8	6.5	0.5	0.06	0.114	6
Description	Hydraulic conductivity of the sand	Porosity of the sand	van Genuchten parameter of the sand for unsaturated flow	van Genuchten parameter of the sand for unsaturated flow	External hydraulic head at the water reservoir	External hydraulic head at the adjacent ditch	Initial conductance term at the boundary related to the water reservoir	Conductance term at the boundary related to the water reservoir after dredging	Conductance term at the boundary related to the adjacent ditch
Parameters	K_c	$\mathcal{E}_{\mathcal{C}}$	a	u	$H_{ext,w}$	$H_{ext,d}$	$R_{w,ini}$	$R_{w,dredging}$	R_d

TABLE S1. Summary of model parameters



Fig. S3. Satellite image of the water reservoir de Gijster and the location of the pilot site (marked in yellow). (Imagery ©2022 Google, Imagery ©2022 Aerodata International Surveys, CNES/ Airbus, Landsat/Copernicus, Maxar Technologies, Map data ©2022.)



Fig. S4. Grain size distribution of sand material took from the field location.



Fig. S5. Illustration of the field installation



Fig. S6. Particle size distribution of the Al-OM flocs under various shear conditions.



Fig. S7. Set-up of the column experiment. a) is the schematic illustration and b) gives a picture of the column with the pressure sensors.



Fig. S8. Measured change in hydraulic head (dH) during infiltration tests performed in zone A. a) and b) show results from infiltration test in well A3 before and after the injection of flocs; c) shows the result of an infiltration test performed in well A10 before the injection and d) denotes the test performed in well A7 after the injection.



Fig. S9. Measured change in hydraulic head (dH) during infiltration tests performed in zone B. a) and b) show results from infiltration test performed in well B2 before and after the injection of flocs; c) and d) denote the infiltration test performed in well B10 before and after the injection.



Fig. S10. Measured electrical conductivity (EC) (a and b) and change in hydraulic head (dH) (c and d) during the injection of the Al-OM flocs suspension in zone A. Figures a) and c) show the results at the end of the first injection of flocs of a day (I7), and b) and d) show the last injection of the day (I13). The injection sequence of that day is I7->I6->I8->I11->I13.



Fig. S11. Illustration of the effect of a flow barrier on the infiltration test. a) is without a flow barrier and 2) includes a flow barrier. The color denotes the hydraulic head and black line denote the streamline.



Fig. S12. The value of the reduced hydraulic conductivity as a function of the assumed thickness of the flow barrier.