

**Unsaturated Flow Effects on Solute Transport in Soils**

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**Introduction**

This supporting information provides brief description of the gamma transmission method (Text S1), ADE transport parameters estimated from the BTCs of sands S1 and S2 at different depths (Tables S1-S6), MIM transport parameters estimated from the BTCs of sands S1 and S2 at different depths (Tables S7-S12), and ADE transport parameters obtained from the BTCs of sand S1 using the small sandbox (Table S13).

## Gamma Transmission Method

### Text S1.

The gamma system consisted of a radioactive source  $^{241}\text{Am}$ , with an energy peak of 59 keV. A schematic view is shown in Figure S1. Gamma radiation attenuation in an unsaturated soil sample can be described using Beer-Lambert's equation as follows

$$I = I_0 \exp(-\mu_s l_s - \mu_w l_w) \quad (\text{S1})$$

where  $I$  and  $I_0$  denote measured and corresponding reference intensities,  $\mu_s$  and  $\mu_w$ , are the solid and water attenuation coefficients, respectively,  $l_s$  and  $l_w$  denote the overall length of the solid and water phases along the path of the gamma-ray beam, respectively.

The diameter of the gamma-ray beam was 6 mm. Measured intensities hence were average values over the cross section of the beam and the soil thickness. The attenuation coefficients  $\mu_s$  and  $\mu_w$  were commonly assumed linearly related to the length of the solid and water phases, respectively. The attenuation coefficients  $\mu_s$  and  $\mu_w$  were measured and calculated beforehand. Detailed information has been reported in the literature (Fritz, 2012). When only deionized water is present without any solute, the reference intensity  $I_0$  was set equal to the measured intensity of the empty sandbox. For saturated samples, the total thickness of the soil sample was known as the width of the sandbox (i.e., 2 cm). The total thickness under saturated conditions is the sum of  $l_s$  and  $l_w$ . Therefore, the values of  $l_w$  and  $l_s = 2 - l_w$  can be calculated from the measured intensity for the saturated samples using Equation S1. Sand porosity ( $\phi$ ) and water saturation ( $S$ ) can then be calculated as:

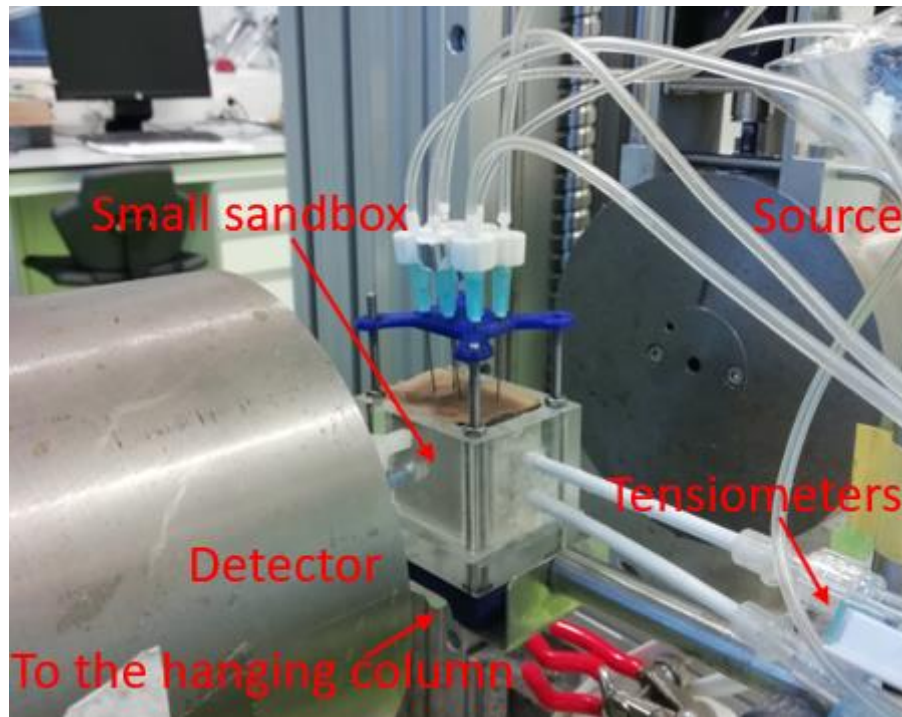
$$\phi = \frac{l - l_s}{l} \text{ and } S = \frac{l_w}{l - l_s} \quad (\text{S2})$$

respectively, in which  $l$  is the total thickness of the soil sample (i.e. 2 cm, being the width of our sandbox). For one set of solute transport experiments, the value of  $l_s$  was fixed at its value for the initial saturated sample, assuming no change of the porosity during the experiment. For each unsaturated condition, the value of  $l_w$  can be calculated based on the measured value of  $I$ .

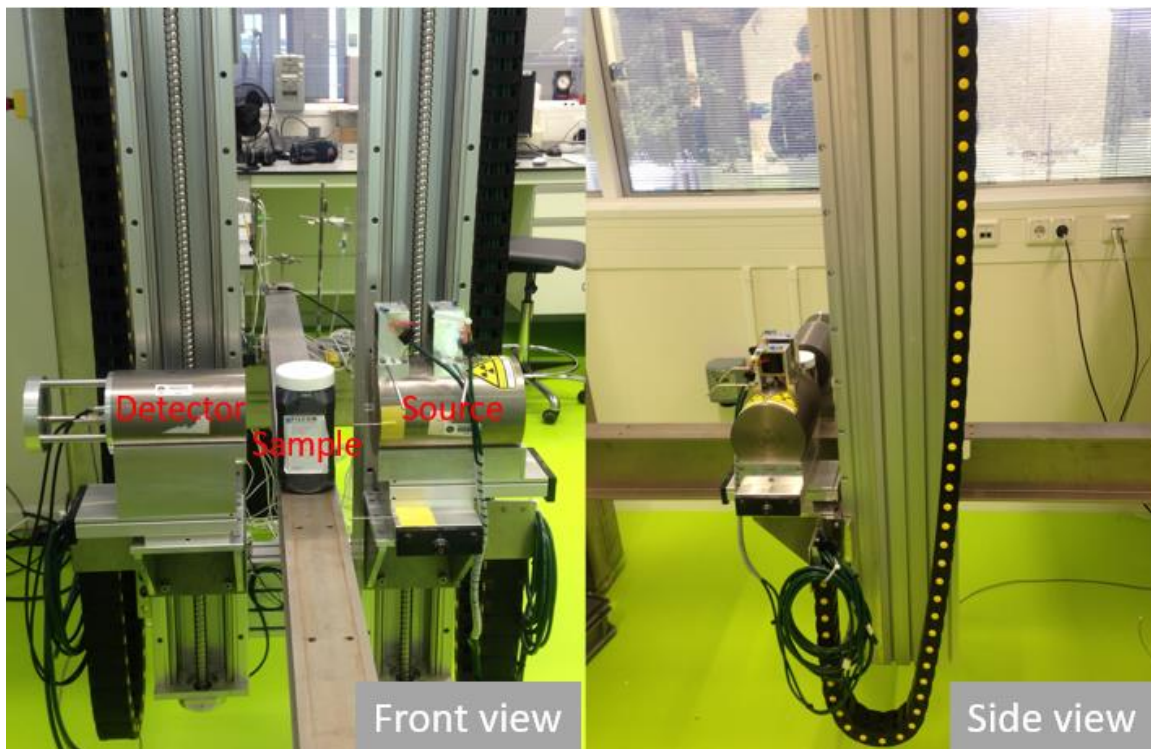
After establishing the unit-gradient flow, the value of  $l_w$  was fixed. When the solution was applied, the variation of  $I$  was only caused by the different concentration of the solution. Equation (S2) is modified to

$$I = I_0 \exp(-\mu_s l_s - \mu_w l_w - \mu_c l_w) \quad (\text{S3})$$

where  $\mu_c$  is the attenuation coefficient of the solution, which is commonly a linear function of solute concentration (Udagani, 2013). We determined the relationship between  $\mu_c$  and solute concentration prior to the experiments. Under the unit-gradient flow condition, the values of  $u_s$ ,  $l_s$ ,  $u_w$ , and  $l_w$  were fixed. The values of  $\mu_c$  can be calculated based on the variation of the intensity  $I$ . For each gamma measurement, the value of solute concentration can be derived from the value of  $\mu_c$ .



**Figure S1.** View of the small sandbox



**Figure S2.** View of the gamma ray system used for the small sandbox BTCs

**ADE transport parameters estimated from the BTCs of sands S1 and S2 at different depths**

**Table S1.** ADE transport parameters estimated from the BTCs of sand S1 at  $x=10.5$  cm.

<b>ADE model, <math>x=10.5</math> cm</b>					
Exp.	$S$	$\nu$	$D$	$\lambda$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-
S1L-1	1.00	0.760	0.0125	0.017	0.9869
S1L-2	1.00	0.096	0.0019	0.020	0.9328
S1L-3	0.80	0.882	0.0447	0.051	0.9992
S1L-4	0.70	0.679	0.0361	0.053	0.9996
S1L-5	0.60	0.346	0.0289	0.083	0.9989
S1L-6	0.50	0.134	0.0099	0.074	0.9990
S1L-7	0.44	0.051	0.0047	0.093	0.9828
S1L-8	0.35	0.014	0.0013	0.090	0.9964

**Table S2.** ADE transport parameters estimated from the BTCs of sand S1 at  $x=18.0$  cm.

<b>ADE model, <math>x=18.0</math> cm</b>					
Exp.	$S$	$\nu$	$D$	$\lambda$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-
S1L-1	1.00	0.806	0.0132	0.016	0.9989
S1L-2	1.00	0.101	0.0018	0.018	0.9618
S1L-3	0.80	0.827	0.0576	0.070	0.9984
S1L-4	0.70	0.687	0.0466	0.068	0.9993
S1L-5	0.60	0.372	0.0333	0.089	0.9961
S1L-6	0.50	0.141	0.0161	0.114	0.9974
S1L-7	0.44	0.055	0.0071	0.128	0.9920
S1L-8	0.35	0.016	0.0020	0.128	0.9953

**Table S3.** ADE transport parameters estimated from the BTCs of sand S1 at  $x=25.5$  cm.

<b>ADE model, <math>x=25.5</math> cm</b>					
Exp.	$S$	$\nu$	$D$	$\lambda$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-
S1L-1	1.00	0.813	0.0114	0.014	0.9994
S1L-2	1.00	0.103	0.0015	0.015	0.9878
S1L-3	0.80	0.792	0.0513	0.065	0.9954
S1L-4	0.70	0.667	0.0463	0.069	0.9946
S1L-5	0.60	0.351	0.0307	0.087	0.9957
S1L-6	0.50	0.135	0.0162	0.120	0.9892
S1L-7	0.44	0.055	0.0074	0.134	0.9917
S1L-8	0.35	0.016	0.0021	0.132	0.9917

**Table S4.** ADE transport parameters estimated from the BTCs of sand S2 at  $x=10.5$  cm.

<b>ADE model, <math>x=10.5</math> cm</b>					
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Exp.	$S$	$\nu$	$D$	$\lambda$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-
S2L-1	1.00	0.173	0.007	0.042	0.9833
S2L-2	1.00	0.319	0.010	0.033	0.9992
S2L-3	0.81	7.804	0.540	0.069	0.9977
S2L-4	0.68	6.473	4.000	0.618	0.9887
S2L-5	0.58	3.417	5.063	1.482	0.9774
S2L-6	0.49	2.186	3.208	1.468	0.9855
S2L-7	0.44	0.678	0.715	1.053	0.9886
S2L-8	0.35	0.394	0.354	0.897	0.9914
S2L-9	0.30	0.216	0.218	1.012	0.9806
S2L-10	0.27	0.080	0.055	0.680	0.9894

**Table S5.** ADE transport parameters estimated from the BTCs of sand S2 at  $x=18.0$  cm.

ADE model, $x=18.0$ cm					
Exp.	$S$	$\nu$	$D$	$\lambda$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-
S2L-1	1.00	0.177	0.008	0.045	0.9806
S2L-2	1.00	0.326	0.013	0.039	0.9987
S2L-3	0.81	8.458	0.552	0.065	0.9916
S2L-4	0.68	5.518	2.431	0.441	0.9987
S2L-5	0.58	3.488	5.826	1.670	0.9787
S2L-6	0.49	2.272	2.453	1.080	0.9943
S2L-7	0.44	0.775	0.804	1.038	0.9960
S2L-8	0.35	0.411	0.352	0.858	0.9936
S2L-9	0.30	0.212	0.208	0.979	0.9882
S2L-10	0.27	0.087	0.077	0.883	0.9862

**Table S6.** ADE transport parameters estimated from the BTCs of sand S2 at  $x=25.5$  cm.

ADE model, $x=25.5$ cm					
Exp.	$S$	$\nu$	$D$	$\lambda$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-
S2L-1	1.00	0.177	0.009	0.049	0.9894
S2L-2	1.00	0.324	0.015	0.046	0.9976
S2L-3	0.81	8.550	0.638	0.075	0.9961
S2L-4	0.68	6.243	1.751	0.280	0.9954
S2L-5	0.58	3.908	2.681	0.686	0.9852
S2L-6	0.49	2.373	1.679	0.708	0.9918
S2L-7	0.44	0.862	0.682	0.792	0.9929
S2L-8	0.35	0.430	0.283	0.657	0.9956
S2L-9	0.30	0.235	0.131	0.559	0.9949
S2L-10	0.27	0.101	0.062	0.617	0.9900

**MIM transport parameters estimated from the BTCs of sands S1 and S2 at different depths**

**Table S7.** MIM transport parameters estimated from the BTCs of sand S1 at  $x=10.5$  cm.

<b>MIM model, <math>x=10.5</math> cm</b>							
Exp.	$S$	$\nu$	$D$	$\lambda_m$	$\beta$	$\omega$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-	min <sup>-1</sup>	-
S1L-3	0.80	0.879	0.0382	0.043	0.986	$4.0 \times 10^{-3}$	0.9993
S1L-4	0.70	0.677	0.0302	0.045	0.982	$4.5 \times 10^{-3}$	0.9996
S1L-5	0.60	0.343	0.0239	0.069	0.981	$8.2 \times 10^{-4}$	0.9992
S1L-6	0.50	0.132	0.0082	0.062	0.978	$1.7 \times 10^{-4}$	0.9994
S1L-7	0.44	0.050	0.0036	0.072	0.967	$8.0 \times 10^{-5}$	0.9837
S1L-8	0.35	0.014	0.0009	0.068	0.960	$1.5 \times 10^{-5}$	0.9974

**Table S8.** MIM transport parameters estimated from the BTCs of sand S1 at  $x=18.0$  cm.

<b>MIM model, <math>x=18.0</math> cm</b>							
Exp.	$S$	$\nu$	$D$	$\lambda_m$	$\beta$	$\omega$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-	min <sup>-1</sup>	-
S1L-3	0.80	0.820	0.0423	0.052	0.978	$2.5 \times 10^{-3}$	0.9989
S1L-4	0.70	0.683	0.0279	0.041	0.962	$7.2 \times 10^{-3}$	0.9998
S1L-5	0.60	0.370	0.0185	0.050	0.944	$5.2 \times 10^{-3}$	0.9966
S1L-6	0.50	0.139	0.0100	0.072	0.961	$2.8 \times 10^{-4}$	0.9993
S1L-7	0.44	0.054	0.0032	0.058	0.941	$1.8 \times 10^{-4}$	0.9958
S1L-8	0.35	0.015	0.0012	0.078	0.953	$1.8 \times 10^{-5}$	0.9974

**Table S9.** MIM transport parameters estimated from the BTCs of sand S1 at  $x=25.5$  cm.

<b>MIM model, <math>x=25.5</math> cm</b>							
Exp.	$S$	$\nu$	$D$	$\lambda_m$	$\beta$	$\omega$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-	min <sup>-1</sup>	-
S1L-3	0.80	0.789	0.0391	0.049	0.980	$4.6 \times 10^{-3}$	0.9926
S1L-4	0.70	0.617	0.0365	0.059	0.923	$2.4 \times 10^{-4}$	0.9962
S1L-5	0.60	0.346	0.0239	0.069	0.980	$2.6 \times 10^{-4}$	0.9970
S1L-6	0.50	0.126	0.0110	0.087	0.927	$7.1 \times 10^{-5}$	0.9930
S1L-7	0.44	0.053	0.0040	0.075	0.954	$6.6 \times 10^{-5}$	0.9967
S1L-8	0.35	0.015	0.0013	0.085	0.937	$8.1 \times 10^{-6}$	0.9968

**Table S10.** MIM transport parameters estimated from the BTCs of sand S2 at  $x=10.5$  cm.

<b>MIM model, <math>x=10.5</math> cm</b>							
Exp.	$S$	$\nu$	$D$	$\lambda_m$	$\beta$	$\omega$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-	min <sup>-1</sup>	-
S2L-3	0.81	7.752	0.0798	0.010	0.868	$5.2 \times 10^{-1}$	0.9981
S2L-4	0.68	5.932	0.8133	0.137	0.795	$6.7 \times 10^{-2}$	0.9887
S2L-5	0.58	2.814	2.4370	0.866	0.801	$4.3 \times 10^{-3}$	0.9850
S2L-6	0.49	1.585	1.6720	1.055	0.721	$1.3 \times 10^{-3}$	0.9893

S2L-7	0.44	0.549	0.4239	0.773	0.800	$4.2 \times 10^{-4}$	0.9920
S2L-8	0.35	0.245	0.1900	0.776	0.622	$5.2 \times 10^{-5}$	0.9935
S2L-9	0.30	0.118	0.0865	0.736	0.552	$4.3 \times 10^{-5}$	0.9877
S1L-10	0.27	0.040	0.0217	0.540	0.501	$9.4 \times 10^{-6}$	0.9653

**Table S11.** MIM transport parameters estimated from the BTCs of sand S2 at  $x=18.0$  cm.

MIM model, $x=18.0$ cm							
Exp.	$S$	$\nu$	$D$	$\lambda_m$	$\beta$	$\omega$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-	min <sup>-1</sup>	-
S2L-3	0.81	8.447	0.1859	0.022	0.881	$6.9 \times 10^{-1}$	0.9916
S2L-4	0.68	5.363	0.3544	0.066	0.749	$1.5 \times 10^{-1}$	0.9993
S2L-5	0.58	2.881	0.7935	0.275	0.711	$1.0 \times 10^{-2}$	0.9958
S2L-6	0.49	2.133	0.5201	0.244	0.747	$1.8 \times 10^{-2}$	0.9961
S2L-7	0.44	0.727	0.0542	0.074	0.650	$9.2 \times 10^{-3}$	0.9970
S2L-8	0.35	0.391	0.0665	0.170	0.739	$3.5 \times 10^{-3}$	0.9955
S2L-9	0.30	0.201	0.0176	0.088	0.693	$1.5 \times 10^{-3}$	0.9923
S1L-10	0.27	0.083	0.0150	0.180	0.717	$6.3 \times 10^{-4}$	0.9837

**Table S12.** MIM transport parameters estimated from the BTCs of sand S2 at  $x=25.5$  cm.

MIM model, $x=25.5$ cm							
Exp.	$S$	$\nu$	$D$	$\lambda_m$	$\beta$	$\omega$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-	min <sup>-1</sup>	-
S2L-3	0.81	8.514	0.1836	0.022	0.875	$6.4 \times 10^{-1}$	0.9961
S2L-4	0.68	6.171	0.8514	0.138	0.900	$8.5 \times 10^{-2}$	0.9934
S2L-5	0.58	3.535	0.8977	0.254	0.855	$6.2 \times 10^{-3}$	0.9969
S2L-6	0.49	2.271	0.4420	0.195	0.852	$1.0 \times 10^{-2}$	0.9964
S2L-7	0.44	0.823	0.1822	0.221	0.840	$3.6 \times 10^{-3}$	0.9965
S2L-8	0.35	0.417	0.1093	0.262	0.872	$1.6 \times 10^{-3}$	0.9965
S2L-9	0.30	0.228	0.0289	0.127	0.824	$1.7 \times 10^{-3}$	0.9976
S1L-10	0.27	0.098	0.0208	0.213	0.853	$3.8 \times 10^{-4}$	0.9918

**ADE transport parameters obtained from the BTCs of sand S1 using the small sandbox**

**Table S13.** ADE parameters obtained from the BTCs of sand S1 using the small sandbox

ADE model, $x=1.5$ cm					
Exp.	$S$	$\nu$	$D$	$\lambda$	$R^2$
	-	cm min <sup>-1</sup>	cm <sup>2</sup> min <sup>-1</sup>	cm	-
S1S-1	1.00	2.585	0.7504	0.290	0.9891
S1S-2	1.00	1.386	0.5228	0.377	0.9493
S1S-3	0.80	1.149	0.5236	0.456	0.9176
S1S-4	0.70	0.677	0.2974	0.439	0.9802

S1S-5	0.60	0.372	0.2295	0.617	0.9043
S1S-6	0.50	0.180	0.1175	0.652	0.9240
S1S-7	0.41	0.065	0.0300	0.459	0.9594
S1S-8	0.28	0.024	0.0293	1.222	0.9164

## SI References

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