

24 **Abstract:** Dissolved-phase contaminants experiencing enhanced diffusion (i.e., “super-diffusion”)
25 with a pronounced leading plume edge can pose risk for groundwater quality. The drivers for
26 complex super-diffusion in geological media, however, are not fully understood. This study
27 investigates the impacts of hydrofacies’ mean lengths and the initial source geometry, motivated
28 by a hydrofacies model built recently for the well-known MADE aquifer, on the spatial pattern of
29 super-diffusion for two-dimensional alluvial aquifer systems. Monte Carlo simulations show that
30 the bimodal velocity distribution, whose pattern is affected by the hydrofacies’ mean lengths, leads
31 to super-diffusion of solutes with a bi-peak plume snapshot in alluvial settings where advection
32 dominates transport. A larger longitudinal mean length (i.e., width) for hydrofacies with high
33 hydraulic conductivity (K) enhances the connectivity of preferential pathways, resulting in higher
34 values in the bimodal velocity distribution and an enhanced leading front for the bi-peak plume
35 snapshot, while the opposite impact is identified for the hydrofacies’ vertical mean length (i.e.,
36 thickness) on the bi-peak super-diffusion. A multi-domain non-local transport model is then
37 proposed, extending upon the concept of the distributed-order fractional derivative, to quantify the
38 evolution of bi-peak super-diffusion due to differential advection and mobile-mobile mass
39 exchange for solute particles moving in hydrofacies with distinct K . Results show that the bi-
40 peak super-diffusion identified for the MADE site and perhaps the other similar aquifers, which is
41 affected by the initial source geometry at an early stage and the thickness and width of high- K
42 hydrofacies during all stages, can be quantified by the mobile-mobile fractional-derivative model.
43 Scale dependency, porous medium dimensionality, and stochastic model comparison are also
44 discussed to further explore the nature of bi-peak super-diffusion in alluvial systems.

45 **Keywords:** Super-diffusion; Alluvial aquifer; Hydrofacies model; Monte Carlo simulation

46 **1. Introduction**

47 Super-diffusive transport (defined by the faster than linear growth of a solute plume's second
48 central moment (or variance) in time) in heterogeneous aquifers, which is usually characterized by
49 an apparent leading edge of the solute plume, can pose a high risk to groundwater quality (Benson
50 et al., 2001; Schumer et al., 2003a; Zhang et al., 2009). Super-diffusion differs significantly from
51 sub-diffusion (defined by the slower than linear growth of the plume variance in time) which is
52 mainly characterized by solute retention and whose transport behavior can be characterized by
53 various stochastic models (Haggerty and Gorelick, 1995; Schumer et al., 2003b; Berkowitz et al.,
54 2006; Zhang et al., 2010; Dentz et al., 2015; Tyukhova et al., 2016; among many others). Super-
55 diffusion has mainly been identified for a few field tracer tests, including those conducted at the
56 well-known MAcroDispersion Experiment (MADE) site (a heterogeneous alluvial depositional
57 aquifer system) focused by hydrogeological modelers for over three decades (Adams and Gelhar,
58 1992; Zheng et al., 2011). Dynamics of super-diffusion in real-world aquifer systems, including
59 the MADE site, have not been fully developed or well understood, motivating this study.

60 Efforts have been made to identify the mechanisms controlling super-diffusion in
61 groundwater for decades. Numerical and analytical analyses were carried out first, revealing that
62 the extensive “heavy” tailing behavior (especially the power-law distributed) and long-range
63 correlation of hydraulic conductivity (K) can lead to early arrivals of solutes in heterogeneous
64 porous media (Sahimi, 1993; Benson et al., 2001; Herrick et al., 2002; Saadatfar and Sahimi, 2002;

65 [Kohlbecker et al., 2006](#); [Dentz and Bolster, 2010](#)). Spatial moments analysis also showed that
66 super-diffusive transport via random walk motion can be driven by power-law distributed,
67 correlated velocities ([Dentz and Bolster, 2010](#)) or as a result of layered media with specific velocity
68 distributions ([Bouchaud et al., 1990](#)). [Salamon et al. \(2007\)](#) conducted numerical experiments and
69 also found that the strong variation and continuity of K in space caused the heavy (extended)
70 leading edge of the tracer plumes observed at the MADE site. Most of these studies revealed the
71 physical condition for super-diffusion, i.e., a random K field with a large variance (and long
72 correlation lengths for most cases). Particularly, the wide contrasts in hydraulic properties of the
73 sediments forming typical alluvial systems can promote super-diffusion, because the
74 interconnected, high- K deposits (such as ancient channels) surrounded by relatively low- K
75 deposits, representing the common internal architecture of an alluvial setting, can guarantee the
76 highly correlated K with a large variance ([Fogg and Zhang, 2016](#)).

77 This typical alluvial structure can be reliably captured by the transition probability based
78 geostatistical tool called “T-PROGS” ([Carle and Fogg, 1996, 1997](#); [Carle, 1999](#)), which provides
79 a feasible way to systematically explore anomalous transport in alluvial systems. T-PROGS can
80 capture major properties of hydrofacies, including their global volumetric proportion, mean
81 lengths (i.e., thickness/width along the vertical/longitudinal direction), and juxtaposition tendency,
82 motivating the numerical exploration of anomalous transport for models of hydrofacies or
83 lithofacies (representing the assemblage of deposits with similar hydrological properties).

84 Hydrofacies models built by T-PROGS in [Zhang et al. \(2013\)](#) and [Bianchi and Zheng \(2016\)](#),

85 however, led to contrasting conclusions describing the generation of super-diffusion. [Zhang et al.](#)
86 [\(2013\)](#) found that although the hydrofacies models do capture the strong spatial variation and
87 continuity of K , they cannot generate super-diffusion with a heavy leading plume front, although
88 apparent sub-diffusion with extensive late-time concentration tailing behavior dominates the solute
89 breakthrough curves (BTCs). Contrarily, the recent work by [Bianchi and Zheng \(2016\)](#)
90 successfully captured super-diffusion with an obvious and pronounced leading edge in the plume
91 snapshots observed at the MADE site (at the sample cycles after ~ 132 days), using the T-PROGS-
92 generated hydrofacies model. A percolated hydrofacies with a significantly high K (two orders of
93 magnitude higher than the other hydrofacies) was found to cause the rapid movement of the leading
94 plume edge ([Bianchi and Zheng, 2016](#)). In T-PROGS, the hydrofacies' mean lengths significantly
95 affect the hydrofacies' connectivity, while the estimation of the hydrofacies' mean lengths
96 (especially the horizontal mean length) contains high uncertainty due to the typically limited
97 number of boreholes. The impacts of hydrofacies' mean lengths along different directions on super-
98 diffusion were not systematically addressed in [Bianchi and Zheng \(2016\)](#), except for a preliminary
99 sensitivity test. To reliably identify the major geological mechanisms controlling super-diffusion
100 in typical alluvial systems, discrepancy between these two studies needs be explored by
101 systematically extending the hydrofacies model for addressing the facies' mean length uncertainty
102 on solute transport. Therefore, the exploration of such mechanisms is the major focus of this study.

103 The following content of this work is organized as follows. **Section 2** introduces the Monte
104 Carlo simulations with multiple scenarios of hydrofacies models generated by T-PROGS to

105 explore anomalous transport in the alluvial settings. **Section 3** presents the numerical results for
106 the scenarios, by focusing on the behavior and mechanism of super-diffusion with a bi-peak plume
107 snapshot. **Section 4** discusses the hydrogeological mechanisms dominating bi-peak super-
108 diffusion by analyzing the velocity distribution, the impact of major properties of hydrofacies on
109 super-diffusion, and the pattern of plume snapshots changing with the initial source geometry. A
110 novel mobile-mobile transport model is then proposed to quantify and interpret the bimodal
111 distribution of the pollutant snapshots. **Section 5** presents the main conclusions of the study. The
112 super-diffusion with bi-peak snapshots and its hydrogeological interpretation, as well as its
113 stochastic quantification, are the new contributions of this work and improve our understanding in
114 the nature of anomalous transport through complex aquifers on the scale of a few hundred meters.

115 **2. Method of Monte Carlo simulations from hydrofacies model to pollutant transport**

116 To explore the dynamics of tracers transport in alluvial settings with intrinsic heterogeneity,
117 a numerical approach with three main steps was used by adopting the procedures in [Zhang et al.](#)
118 [\(2013\)](#) and [Bianchi & Zheng \(2016\)](#). First, T-PROGS was used to generate two-dimensional (2-*d*),
119 different alluvial settings with various hydrofacies structures ([Carle and Fogg, 1997](#)). Monte Carlo
120 simulations of the hydrofacies distribution lead to the random *K* fields. Second, steady-state
121 groundwater flow fields were calculated using a block-centered finite difference model ([Harbaugh,](#)
122 [2006](#)). Third, conservative tracer transport was simulated using the MT3D program ([Zheng et al.,](#)
123 [2010](#)). The following subsections briefly describe these steps.

124 2.1 Modeling hydrofacies structures and hydraulic conductivity fields

125 T-PROGS (Carle, 1999; Carle and Fogg, 1997) was used to generate hydrofacies structures
126 that can be representative of different alluvial aquifers. To construct numerical models representing
127 major properties of real-world aquifers, the geostatistical characters/properties of the MADE
128 aquifer identified by Bianchi and Zheng (2016) is used as a reference. As in the previous work, the
129 generated aquifers are therefore characterized by five hydrofacies, including (1) a highly
130 conductive gravel (HCG), (2) gravel with sand (GS), (3) sand gravel and fines (SGF), (4) sand and
131 gravel (SG), and (5) well-sorted sand (S). The hydrofacies properties, including the hydraulic
132 conductivity, mean lengths, and the volumetric proportion listed in **Table 1** are also consistent with
133 the lithological model proposed by Bianchi and Zheng (2016).

134 A 2-*d* vertical profile of the aquifer with a dimension of 300 m in length and 40 m in thickness
135 is used, with the grid size of 10 m and 0.5 m along the longitudinal and vertical directions,
136 respectively. The same model and grid dimensions are used for the following flow and transport
137 models. The sensitivity of transport dynamics to the grid size is discussed in the supplementary
138 material (section S4), to evaluate the feasibility of the grid resolution selected in this study.

139 There are three modifications of the hydrofacies modeling in Bianchi and Zheng (2016). *First*,
140 multiple scenarios containing different mean lengths along the vertical direction (i.e., thicknesses)
141 or the longitudinal direction (i.e., widths) for hydrofacies are developed to account for the
142 uncertainty of hydrofacies mean lengths and their potential impact on transport. *Second*, no hard-
143 conditional data are used in most scenarios when running T-PROGS, so that the resultant

144 hydrofacies models can capture the maximum spatial variation of hydrofacies necessary for a
145 systematic analysis. *Third*, the 3-*d* models used by [Bianchi and Zheng \(2016\)](#) are simplified to 2-
146 *d* models, since the 2-*d* models provide the simplest framework to control the longitudinal and
147 vertical correlations of *K* and evaluate the impact of *K* structures on solute transport. The impact
148 of model dimension on super-diffusion will be discussed in the supplementary material (section
149 S2).

150 Eight scenarios are designed, with 100 realizations for each scenario, to investigate the
151 influence of the internal aquifer structure on super-diffusion. Scenario 1 is the basic case
152 representing the general statistics obtained from the MADE aquifer, with parameters estimated by
153 [Bianchi and Zheng \(2016\)](#) (**Table 1**). Scenarios 2, 3, and 4 have the thickness (i.e., the vertical
154 mean length) of 1.5, 2.0, and 2.5 times larger than that of the base scenario for all the hydrofacies,
155 respectively. Scenarios 5, 6, and 7 have a longitudinal mean length or width for each hydrofacies
156 1.25, 1.5, and 2.0 times larger than the base case, respectively.

157 Scenario 8 is designed to investigate the influence of the contaminant source vertical
158 extension on solute transport. Four vertical line sources with the length increasing from 2, 5, 20,
159 to 40 m (i.e., 1/20, 1/4, 1/2 and 1.0 time of the aquifer thickness, respectively) are considered in
160 this study. To decrease the impact of low-*K* zones on the injection of the initial pollutant source,
161 an aggregate of high-*K* HCG facies located at $z=20\sim 22$ m and $x=5$ m is used as the hard conditional
162 data (configuration) when building the realizations for Scenario 8, and this zone is selected as the
163 midpoint of the line source. The same strategy was used by [Zhang et al. \(2013\)](#) in a similar Monte

164 Carlo study. **Fig. 1** shows one realization arbitrarily selected (realization #1) for each scenario.

165 **2.2 Modeling of groundwater flow and solute transport**

166 The steady-state flow fields are calculated using MODFLOW (Harbaugh, 2006), with the
167 specified head boundary condition defined for the two vertical boundaries (left and right
168 boundaries) and the no-flow boundary condition for the two horizontal boundaries (top and bottom
169 boundaries). A ratio between the vertical and longitudinal K , 0.13, based on the pumping test
170 conducted at the MADE site (Bianchi and Zheng, 2016; Boggs et al., 1990), is used in the
171 groundwater flow modeling. The general hydraulic gradient of 0.006 is used in this study, which
172 is similar to the one (0.0058) used by Guan et al. (2008). The average K (Table 1) is assigned for
173 each hydrofacies when calculating groundwater flow.

174 The finite difference transport code MT3DMS (Zheng et al., 2010) is then used to calculate
175 solute transport. A vertical instantaneous line source with the uniform concentration is used in
176 Scenarios 1 to 7. In Scenario 8, various lengths (2, 5, 20, and 40 m) of the vertical line source is
177 considered to investigate the impact of the initial source scale on solute transport. The initial source
178 for all scenarios is located at $x = 5$ m. The downgradient boundary along the longitudinal direction
179 is defined as the zero-value Neumann boundary (i.e., the free exit boundary), while the top and
180 bottom boundaries are no-flux boundaries. The molecular diffusion coefficient is 1.16×10^{-9} m²/s,
181 representing the diffusivity for tritium in water (Bianchi and Zheng, 2016). The effective porosity
182 for each hydrofacies is listed in Table 1. The longitudinal dispersivity α_L is 1 m (which is 1/10

183 of the grid dimension), and the vertical dispersivity α_V is two orders of magnitude lower than α_L
184 (Llopis-Albert and Capilla, 2009; Bianchi and Zheng, 2016). The sensitivity of super-diffusion on
185 dispersivity will be addressed in the supplementary material (section S4).

186 **3. Results**

187 The ensemble average of the simulated plume snapshots for all the 100 realizations for each
188 scenario is calculated and shown below (**Fig. 2~5**). The 1-*d* normalized longitudinal mass
189 distribution at 27, 132, 224 and 328 days (after the release of the contaminant source), which
190 present the four sample snapshots during the MADE-2 experiment (Bogg et al.,1990), is also
191 plotted for further analysis. The variance of solute plumes is also calculated (shown by **Fig. S4**),
192 providing clear evidence for super-diffusion of the simulated transport. The following subsections
193 introduce the plume snapshots for each scenario in order to explore the impact of medium
194 architecture and the initial concentration distribution on solute transport in alluvial systems.

195 **3.1 Impact of hydrofacies' vertical mean length (i.e., thickness) on super-diffusion**

196 The calculated mass distribution for scenarios 1~4 with different thicknesses for hydrofacies
197 is depicted in **Fig. 2**. Super-diffusive transport is characterized by the plume's apparent leading
198 front moving quickly along preferential pathways, while a large portion of solute remains near the
199 initial source location.

200 The transport simulations also show that a smaller thickness of hydrofacies leads to a better
201 connectivity of the high-*K* zones, which can enhance the downstream expansion of the plume front.

202 In addition, the different shape (i.e., skewness) of the simulated plume snapshots (**Fig. 2**) imply
203 that the hydrofacies' thickness also affects the mass ratio of contaminants in the relatively high and
204 low velocity zones. Particularly, the scenario with thicker hydrofacies (such as scenario 4) tends
205 to delay (retain) more contaminants near the source and release less mass downgradient.

206 Therefore, the hydrofacies' thickness has the opposite impact on the two edges of the plume
207 snapshot. A thicker hydrofacies, in fact, enhances the trailing edge due to the longer path for slow
208 advection, while a thinner hydrofacies promotes the longitudinal facies/flow connection and hence
209 enhances the downgradient migration of the plume's leading edge.

210 **3.2 Impact of hydrofacies' longitudinal mean length (i.e., width) on super-diffusion**

211 **Fig. 3** depicts the simulated evolution of contaminant snapshots for scenarios 1, 5, 6, and 7,
212 with different widths for hydrofacies. Results show that a larger width for the hydrofacies causes
213 faster transport of the plume's leading edge, which is opposite to the effect of hydrofacies'
214 thickness on super-diffusion. A similar result was found by [Bianchi and Pedretti \(2017\)](#), who
215 showed that a larger horizontal mean length led to a more slant distribution for solute particles'
216 arrival times. In addition, the hydrofacies' width only slightly affects the plume's trailing edge,
217 which is different from the result shown in **section 3.1** whereby the hydrofacies' thickness can
218 affect both edges of the plume snapshot.

219 **3.3 Impact of the size of the initial contaminant source on super-diffusion**

220 **Fig. 4** shows the simulated plume snapshots for scenarios 8 with different sizes for the initial

221 source. The initial source condition can significantly impact super-diffusion by affecting the
222 overall pattern of plume snapshots. Strong super-diffusion is identified for all of the initial source-
223 lengths evaluated in this study. On one hand, a larger initial source (in which orientation is
224 perpendicular to the general flow direction) causes more solute particles to remain in the low
225 velocity zones around the initial source, resulting in a more (positively) skewed plume snapshot.
226 On the other hand, when the source size is much smaller (i.e., ≤ 2 m) and can be approximated as
227 a point source (all mass located in the high- K zone initially), solute particles can move fast and
228 form a distinct plume peak.

229 **3.4 Two-dimensional plume snapshot**

230 To directly view the solute transport process in heterogeneous structures, the spatial
231 distribution of K and the corresponding 2- d snapshot of the plume front at different times for one
232 realization in scenario 1 are plotted in **Fig. 5a** and **Fig. 5b**, respectively. The results show that the
233 preferential flow path generated by continuous high- K hydrofacies has a great impact on plume
234 evolution and is the main reason for super-diffusion. Meanwhile, a larger fraction of solute located
235 at the relatively less permeable zone moves slowly. Another interesting phenomenon shown in **Fig.**
236 **5b** is that while the preferential path generates super-diffusion, a front peak can also arise when a
237 fraction of solutes moves faster along the preferential path. This bi-peak solute transport in alluvial
238 aquifers will be further discussed and modeled in **section 4**.

239 4. Discussion

240 Super-diffusion for conservative tracers observed in the Monte Carlo simulations is mainly
241 driven by the internal structure of the alluvial aquifer settings, where the detailed mechanism is
242 discussed below. Particularly, we analyze the relationship among the medium's architecture, the
243 velocity distribution, and anomalous transport characteristics. A novel physical model is then
244 proposed to quantify the observed anomalous transport in the alluvial aquifers. The applicability
245 of another popular stochastic model (the time nonlocal transport model) is discussed in the
246 supplementary material to further explore the nature of bi-peak super-diffusion in alluvial systems.

247 4.1 Statistics of the Eulerian velocity

248 To calculate the velocity distribution for each scenario, we adopted the approach proposed by
249 Hyman et al. (2019). Particularly, we define the magnitude of the Eulerian velocity
250 using $v_e(x, z) = \|u(x, z)\|$, where $u(x, z)$ is the velocity simulated by our flow model. The PDF
251 of the Eulerian velocity v in the flow domain Ω_e , denoted as $p_e(v)$, is given as:

$$252 \quad p_e(v) = \frac{1}{U_e} \sum_{\Omega_e} \delta[v - v_e(x, z)] dU, \quad (1)$$

253 where U_e is the domain's volume, δ is the Dirac delta function, dU is the volume of the cell in
254 the groundwater flow model, and x and z are the longitudinal and vertical coordinates, respectively.

255 Since we consider an instantaneous line source in a rectangular aquifer under the condition
256 of no-flow bottom/top boundaries, the longitudinal velocity is the main factor that may control the
257 solute plume's longitudinal mass distribution. Therefore, to reveal the dominant mechanism of

258 solute transport in alluvial structures, the longitudinal velocity is analyzed in detail herein. For
259 each scenario, velocities of $\sim 24,000$ grids are counted, resulting in a solid PDF. **Fig. 6** shows that
260 there are two peaks in the calculated velocity distribution with a broad spectrum ranging mostly
261 between 1.0×10^{-4} m/d and 1.0×10^1 m/d. To provide a more quantitative view, several key statistics
262 of the velocity distribution are listed in **Table 2**. Notably, the percentage of the slow velocity zone
263 (represented by “ $P(v \leq v_m)$ ” in **Table 2**) is close to the total volumetric proportion (~ 0.88) of the
264 hydrofacies excluding the high- K HCG, and the two velocity zones may be separated by the poor
265 connectivity of some HCGs. A larger width for hydrofacies results in a slightly smaller fraction of
266 the low velocity zone (**Table 2**), since the model with a larger width for hydrofacies can produce
267 better longitudinal connectivity for high- K hydrofacies which in turn increases the proportion of
268 high velocities in the velocity PDF. The opposite impact can be found for the hydrofacies thickness:
269 thicker hydrofacies slightly increase the fraction of the low velocity zone (**Table 2**), likely due to
270 the decreased longitudinal connectivity.

271 The bimodal distribution of velocity (**Fig. 6**) sheds light on the formation of super-diffusion
272 and may explain the plume snapshots generated for different hydrofacies scenarios. The second
273 peak of the velocity PDF, representing the high velocity, is associated with preferential flow paths
274 for solute particles that exhibit the super-diffusion behavior. The proportion of the high to low
275 velocity zones is approximately 10% to 90%, respectively (**Table 2**), indicating that most of the
276 solute particles are transporting in the low velocity zone while the remaining particles move fast
277 in the high velocity zone, generating the positively skewed plume snapshot and the overall super-

278 diffusive transport behavior.

279 The vertical and longitudinal mean lengths of hydrofacies have significant and varied
280 influences on the distribution of the longitudinal velocity. On one hand, as shown in **Fig. 6**, the
281 hydrofacies' thickness affects the distribution of both the low and high velocities of moving solutes.
282 Thicker hydrofacies lead to a higher percentage of the low velocity zones and shorter distances
283 between the two peaks of the velocity PDF. On the other hand, the hydrofacies' width mainly
284 affects the distribution of the high velocities with little associated impact on the low velocities (**Fig.**
285 **6**). Wider hydrofacies produce higher velocities and therefore separate the two contrasting velocity
286 zones further in the velocity PDF. These results are consistent with the Monte Carlo simulations
287 shown in **section 3** and provide further insight on the interpretation of the impact of alluvial
288 setting's architecture on solute transport.

289 **4.2 Correlation between hydraulic conductivity and velocity field**

290 The relationship between the velocity field and the hydraulic conductivity field had been
291 explored by various studies. For example, [Veneziano and Tabaei \(2004\)](#) and [Veneziano and Essiam](#)
292 [\(2004\)](#) found a clear relationship between the velocity field and statistics of the K field. [Herrick et](#)
293 [al. \(2002\)](#) and [Kohlbecker et al. \(2006\)](#) investigated the relationship between the heavy-tailed
294 logarithm K and the heavy tailed log velocity distribution, and an empirical equation was proposed
295 by [Kohlbecker et al. \(2006\)](#) to predict the tail of the velocity PDF based on the K distribution. A
296 recent work in revealing the relationship between K and v ([Bianchi and Pedretti, 2017](#)) used a

297 novel metric, *geological entropy*, and established a quantitative relationship between structure
298 settings and the velocity distribution. The geological entropy was found to be a promising way to
299 describe heterogeneous structure and predict solute transport (Bianchi and Pedretti, 2018).

300 In this section, we investigate the origin of the bimodal velocity distribution and focus on the
301 influence of medium architecture on the relationship between the distributions of K and v . The
302 frequency of the random K distribution (Fig. S5) clearly shows a bimodal pattern with the majority
303 of K near $1 \times 10^{0.5}$ m/d and a smaller peak but a much larger K around $1 \times 10^{2.5}$ m/d. The similar
304 bimodal characteristics of the random K field and the velocity's spatial distribution imply a direct
305 correlation between these two random fields.

306 The spatial distributions of K and v of one realization for scenario 1 are shown in Fig. 7,
307 illustrating a positive correlation between the spatial distribution of high K and large velocities.
308 We calculate the coefficient of the spatial correlation between K and v using

$$309 \quad C_r(K', v') = \frac{\text{Cov}(K', v')}{\sqrt{\text{Var}(K') \text{Var}(v')}} \quad , \quad (2)$$

310 where K' and v' are the spatial distributions of the normalized $\log_{10}(K)$ and the longitudinal
311 velocity, respectively. The calculated correlation coefficient C_r between K and v is shown in Fig.
312 8 for the scenarios with different mean lengths of hydrofacies. The porous medium's architecture
313 setting has a significant impact on the correlation between K and v distributions. For example,
314 when the width for hydrofacies is doubled, C_r increases from 0.737 to 0.784. This positive impact
315 may be due to the better connectivity of K with wider hydrofacies. Contrarily, when the hydrofacies'
316 thickness increases by 2.5 times, C_r decreases from 0.737 to 0.691. This negative impact may be

317 due to the fact that thicker hydrofacies result in more high- K zones surrounded by the low- K zones,
 318 resulting in an overall lower proportion of high velocity zones (or isolation of high K zones) for
 319 solute transport.

320 **4.3 Quantify bimodal super-diffusion using a distributed-order fractional-derivative model**

321 To capture super-diffusion with multiple peaks in the plume snapshot, we adopt the concept
 322 of the distributed-order fractional derivative ([Lorenzo and Hartley, 2002](#); [Chechkin et al., 2002](#)):

$$323 \int_1^m a(r) \mathfrak{D}_+^r u(x) dr = f(x) , \quad (3)$$

324 where the variable r ($1 < r \leq 2$) denotes the order of the space fractional derivative, $a(r)$ is the
 325 weight for order r , and the symbol \mathfrak{D}_+^r denotes the positive Riemann-Liouville fractional
 326 derivative ([Miller and Ross, 1993](#)):

$$327 \mathfrak{D}_+^r u(x) = \frac{\partial^r}{\partial x^r} u(x) = \frac{1}{\Gamma(2-r)} \frac{\partial^2}{\partial x^2} \int_{-\infty}^x u(y) (x-y)^{1-r} dy , \quad (4)$$

328 where $\Gamma(\cdot)$ represents the Gamma function. The multi-term (i.e., discrete components) version of
 329 the distributed-order fractional derivative equation takes the form ([Diethelm and Ford, 2009](#)):

$$330 \sum_{i=1}^k a_i \mathfrak{D}_*^{r_i} u(x) = f(x) , \quad (5)$$

331 which can be used to quantify the impact of multiple mobile zones (each with a distinct memory
 332 kernel or index) on the material dynamics. The distributed-order time fractional-derivative models,
 333 which replace the space fractional derivative in the above formula using the time fractional
 334 derivative, have been applied to decelerating solute sub-diffusion and the other random processes
 335 with multiple scaling ([Mainardi et al. 2008](#); [Eab and Lim, 2011](#); [Gorenflo et al., 2015](#)). To the best

336 of our knowledge, the distributed-order space fractional-derivative model has not been applied for
 337 real-world or synthetic groundwater solute transport problems.

338 The multi-term distributed-order fractional derivative equation leads logically to the 1-*d*,
 339 Multi-Domain, tempered space Fractional-Derivative Model (MD-tsFDM) proposed by this study.
 340 The total mass M_t of the MD-tsFDM within a representative elementary volume (REV) is
 341 calculated by

$$342 \quad M_t(x, t) = \sum_{i=1}^N C_i(x, t) \theta_i \varphi_i U_{REV} \quad , \quad (6)$$

343 where $C_i(x, t)$ [M/L^3] is the concentration within the *i*-th domain at location x and time t , θ_i
 344 [-] and φ_i [-] are the average porosity and the proportion of the *i*-th domain, N [-] is the total
 345 number of domains, and U_{REV} [L^3] is the volume of the REV. To capture the strong super-diffusive
 346 transport observed in the Monte Carlo simulations discussed above, the space fractional-derivative
 347 equation with a truncation parameter proposed by [Baeumer and Meerschaert \(2010\)](#) is selected:

$$348 \quad \partial_t C_1(x, t) = -v_1 \partial_x C_1(x, t) + D_1 \partial_x^{\alpha_1, \lambda_1} C_1(x, t) - W_1(x, t) \quad (7a)$$

$$349 \quad \partial_t C_2(x, t) = -v_2 \partial_x C_2(x, t) + D_2 \partial_x^{\alpha_2, \lambda_2} C_2(x, t) + W_2(x, t) \quad (7b)$$

350 where $W(x, t)$ is the mass exchange between the two mobile domains:

$$351 \quad W_i(x, t) = \frac{\omega}{\varepsilon_i \varphi_i} [C_1(x, t) - C_2(x, t)] \quad (8)$$

352 and the operator $\partial_x^{\alpha, \lambda}$ in (7) denotes the tempered fractional derivative and can be calculated by
 353 ([Baeumer and Meerschaert, 2010](#)):

$$354 \quad \partial_x^{\alpha, \lambda} f(x, t) = e^{-\lambda x} \partial_x^\alpha [e^{\lambda x} f(x, t)] - \lambda^\alpha f(x, t) - \alpha \lambda^{\alpha-1} \partial_x f(x, t) \quad (9)$$

355 where $\alpha \in (1, 2]$ (dimensionless) is the fractional index; v [LT^{-1}] and D [$L^\alpha T^{-1}$] are the

356 average velocity and the effective dispersion coefficient, respectively; $\lambda > 0$ [L^{-1}] is the
357 truncation parameter in space; and ω [$ML^{-3}T^{-1}$] is the mass transfer rate between the two domains.
358 The space fractional derivative in (7) is used here since it was proved to be an efficient tool in
359 capturing super-diffusion with a leading edge or positive skewness for the plume snapshot, due to
360 nonlocal transport along preferential flow paths (Zhang et al., 2015). The truncation parameter λ
361 in model (7) describes the maximum displacement of solute particles due to the finite size of the
362 interconnected, preferential pathways.

363 The MD-tsFDM (7), which is a simplified distributed-order FDM, assumes dual mobile zones
364 with distinct advective capacities. Model (7) can be conveniently generalized to account for
365 multiple mobile-immobile zones using the distributed-order, spatiotemporal FDM, which however,
366 may not be necessary here. This is because, as discussed in **sections S1** and **S3**, solute transport is
367 dominated by advection in the Monte Carlo models built in this study. It is also noteworthy that
368 the concept of “multiple mobile zones” in model (7) is consistent with that in the mobile-mobile
369 mass exchange model proposed firstly by Ginn (2018) and Lu et al. (2018).

370 The boundary conditions for the stochastic model (7) are the same as those used for the
371 MT3DMS model discussed in section 2.2. The finite difference method proposed by Baeumer and
372 Meerschaert (2010) is used to solve the MD-tsFDM in this study. To decrease the uncertainty of
373 model parameters, we fix the volumetric proportion of each domain representing the low and large
374 velocity zones, φ_i ($i=1, 2$), as 0.88 and 0.12, respectively. The porosity of each domain is also
375 assumed to be equal in the MD-tsFDM, following the assumption in Llopis and Capilla (2009) and

376 [Guan et al. \(2008\)](#). It is noteworthy that the proportion corresponds to the volumetric fraction of
377 lithofacies HGC (=0.12) estimated from the borehole logs. A similar conclusion was drawn by
378 [Bianchi and Zheng \(2016\)](#), who found that the volumetric fraction of HCG corresponds to the
379 calibrated ratio between the mobile and total porosities of the dual-domain transport model.
380 Therefore, in real-world applications, this proportion (φ_i) can be approximated firstly by the
381 volumetric ratio of low- and high- K deposits gleaned from cores, drillers' logs, and/or outcrops.

382 *4.3.1 Bimodal mass distribution*

383 The best-fit results of the mean snapshots for scenario 1 using MD-tsFDMs (7) are shown in
384 **Fig. 9**. The MD-tsFDM (7) with $n=2$ (two domains) can capture the bimodal plume snapshots
385 better than the single-domain model ($n=1$). The MD-tsFDM also captures the plume evolution in
386 each domain (shown by the green dashed and dotted line in **Fig. 9**). Compared with the plume in
387 domain 1 ($i=1$, transport slowly) with a small velocity, the plume in domain 2 ($i=2$, along the
388 preferential paths) moves faster and dominates the plume front at early time. Additionally, due to
389 the fast advection, the plume moving in the preferential flow path expands quickly in space.
390 Therefore, at late time (i.e., $t > 300$ days), the 2nd peak (formed by the fast motion) of the plume
391 snapshot tends to be smeared (**Fig. 9**).

392 Extending upon the previous analysis, we check whether the biomodal (transport or
393 distribution of the plume) snapshot is due to the initial source size. Previous studies have found
394 that the initial condition of pollutants affects solute transport in heterogeneous media (i.e., [Zinn
395 and Harvey, 2003](#)), because different spatial distributions (uniform or flux-weighted) of the initial

396 source can assign different initial velocities to solute particles, thereby impacting subsequent
397 transport behavior (Morales et al., 2017; Puyguiraud et al., 2019). Here we explore another critical,
398 unsolved question of whether a point source can produce bimodal super-diffusion. To explore the
399 impact of initial source conditions on the bimodal transport in alluvial aquifers, we calculate the
400 mass distribution with different initial source lengths at the sampling time of 27 days and 132 days,
401 respectively. Two types of initial source geometry are considered, including a “point” source with
402 a relatively short (2 m) vertical length and a line source with a 40-m vertical length (**Fig. 10**).
403 Results show that the plume for the case with a line source exhibits the bimodal shape at both 27
404 and 132 days. The plume resulting from the initial point source exhibits a single peak for a
405 relatively short travel distance, which can be fitted by the MD-tsFDM (7) with a single domain.
406 However, when the travel distance increases, even the plume with an initial point source begins to
407 show the significant bimodal snapshot. This result indicates that the velocity field (or the related
408 porous medium architecture setting) is the key factor that controls the bimodal shape of the plume
409 snapshot, while the initial source condition only affects solute transport at early times.

410 The generally well fit (**Fig. 9**) shows that the MD-tsFDM can capture the bimodal snapshot
411 in alluvial settings with strong heterogeneity (the best-fit results for the other scenarios are shown
412 in **section S6** in the supplementary material) and provides physical interpretation of solute transport
413 in bimodal structure media. The best-fit parameters of the MD-tsFDM (listed in **Table 3**) show the
414 impact of medium architecture on solute transport within each domain. The velocity of the
415 preferential flow path domain (v_2) is much higher than that of the slow domain (v_1), and a larger

416 horizontal mean length leads to a larger v_2 . This result is consistent with the analysis in **section 3.2**
417 and **section 4.1**. The dispersivity for transport along the preferential flow domain is much larger
418 than that of the slow domain, (which is expected) and expands quickly the plume 2 (**Fig. 9**). The
419 fractional index, α , of both domains is relatively small (ranging from 1.1 to 1.4), indicating strong
420 heterogeneity for each domain (Benson et al., 2001). The small value of λ demonstrates a long
421 correlation of high- K hydrofacies, and generally decreases with an increasing horizontal mean
422 length for hydrofacies (Meerschaert et al., 2008).

423 *4.3.2 Bimodal super-diffusion at the MADE-2 site and the other sites*

424 As discussed above, the MD-tsFDM (7) can successfully capture the bimodal mass
425 distribution in complex alluvial aquifer systems. To check the applicability of model (7) in real-
426 world aquifers, snapshots of the MADE-2 experiment are fitted using the mathematical model (7)
427 proposed herein. The MADE-2 experiment was conducted in the alluvial aquifer located in
428 Columbus, Mississippi, USA, under natural hydraulic gradient conditions. Tracer snapshots were
429 sampled at 27, 132, 224, and 328 days after the injection of 9.7 m³ tritium solution. For this analysis,
430 we use the last two plume snapshots which were considered to be more reliable and had been
431 thoroughly analyzed in previous studies (Sun et al., 2014; Zhang et al., 2007). Detailed information
432 and review about the MADE-2 experiment can be found in Zheng et al. (2011).

433 The longitudinal mass distribution for tritium is calculated using the same method described
434 in Bianchi and Zheng (2016). The observed and best-fit mass distributions are plotted in **Fig. 11**.
435 The obvious bimodal pattern of the MADE-2 snapshots provides the field evidence for bi-peak

436 super-diffusion and validates the applicability of the MD-tsFDM (7) to interpret anomalous
437 transport in real-world alluvial aquifer systems on the scale of a few hundred meters.

438 Multi-peak plume snapshots or tracer BTCs were also observed at the other study sites. For
439 example, [Guihéneu et al. \(2017\)](#) conducted a series of convergent and push-pull tracer experiments
440 and identified various bimodal BTCs. The bimodal snapshots and BTCs were also observed by [Hu
441 and Huang \(2002\)](#) for transport in stochastic heterogeneous dual-permeability media. The MADE-
442 1 tracer test also identified the very similar bimodal snapshots for tracers as those observed for the
443 MADE-2 test ([Adams and Gelhar, 1992](#)). The bimodal transport behavior for pollutants was also
444 widely investigated using column or sand-box experiments and numerical experiments ([Coppola
445 et al., 2009](#); [Leij and Bradford, 2013](#); [Pedretti et al., 2016](#); [Golfier et al., 2011](#)). The multi-domain
446 models, such as the MD-tsFDM proposed by this study, shed light on the reliable simulation and
447 prediction of pollutant transport in the complex structures mentioned above.

448 **4.4 Impact of hydrofacies HCG on bimodal super-diffusion**

449 It is critical to explore the dominant properties of hydrofacies defined by the transition
450 probability model that generate super-diffusion conditions for solute transport in aquifers. So far,
451 we find that the mean lengths of the hydrofacies affect super-diffusion. In this analysis we explore
452 how other properties of the hydrofacies may impact solute transport and potential super-diffusion
453 observations. Two additional scenarios (scenarios A1 and A2 listed in **Table 4**) were conducted to
454 explore the impact of the volumetric proportion of the coarse grain hydrofacies (i.e., HCG) on

455 super-diffusion. **Fig. 12a** shows the simulated mass distribution at 224 days for different scenarios,
456 in which the global volumetric proportion of HCG increases from 7% to 17%. The proportion of
457 HCG has an obvious impact on super-diffusion by controlling the proportion of the high-velocity
458 zone in the bimodal velocity PDF. A smaller proportion of HCG reduces the size of the high-
459 velocity zone, resulting in a weaker plume front edge and more mass delayed (retained) near the
460 source location. In addition, the simulated mass distribution for the scenario with the lower
461 proportion for HCG (i.e., 7%) also contains more fluctuations of solute mass than the other
462 scenarios (**Fig. 12a**). This result is consistent with the conclusion in [Zhang et al. \(2013\)](#) that found
463 when the proportion of the ancient channel deposits is small (e.g., less than 12%, which is below
464 the percolation threshold (0.14) suggested by [Harter \(2005\)](#) for the 3-*d* model), the high-*K* deposits
465 are not interconnected throughout the entire model domain. Thus, the discontinuous preferential
466 pathways may lead to more local mass peaks in the plume snapshot (**Fig. 12a**).

467 Second, to further explore the impact of HCG on super-diffusion, we built four additional
468 scenarios (Scenarios A3~A6 listed in **Table 4**) with various horizontal or vertical mean lengths for
469 HCG and the other hydrofacies. The results show that the mean lengths (both thickness and width)
470 of HCG have a prominent impact on bimodal super-diffusion in the alluvial aquifer (**Fig. 12b** and
471 **12c**). A larger width of HCG (with the other hydrofacies' mean lengths remaining unchanged)
472 enhances the second peak of solute mass, resulting in a heavier (more pronounced) plume front
473 transport (**Fig. 12b**) because of the enhanced connectivity of the most preferential flow pathways.
474 Contrarily, a thicker HCG (while keeping the other hydrofacies's mean lengths unchanged) results

475 in a lighter (less pronounced) plume leading edge (**Fig. 12c**) because of the truncation of the most-
476 permeable transport pathways. This conclusion further expands upon the results provided in
477 [Bianchi and Zheng \(2016\)](#).

478 Contrarily, the modeled mass distributions are similar for the scenarios where the fine-grain
479 materials were changed to incorporate different mean lengths (thickness or width) while keeping
480 the HCG's mean lengths unchanged, even though the variation of the fine-grained hydrofacies'
481 mean lengths is large (i.e., 1.5 times the hydrofacies' width and 2.5 times the thickness). This result
482 provides a robust explanation as to why none of the thousands of hydrofacies models developed
483 by [Zhang et al. \(2013\)](#) captured the power-law leading edge behavior: there were no extremely
484 high- K HCG zones used in their hydrofacies models, which could not produce the 2nd peak in the
485 velocity PDF or the heavy (enhanced) leading plume front of the snapshot.

486 Overall, these analyses showed that, three primary properties of the hydrofacies HCG
487 impacted super-diffusion. A relatively high K distribution, a volumetric proportion of HCG higher
488 than the percolation threshold, and a sufficiently large HCG width for the generated porous
489 medium systems similar to a fracture/matrix system are essential for producing the bimodal super-
490 diffusive snapshots observed at the MADE site. It is noteworthy that, of the three HCG properties,
491 the HCG's mean width contains the highest uncertainty (due to the discontinuous cores/drillers'
492 logs along the longitudinal direction), revealing that additional techniques are needed to reliably
493 define the high- K lithofacies' width for such field applications.

494 5. Conclusion

495 This study explored the impacts of hydrofacies' mean lengths and the initial source size on
496 bimodal super-diffusion for conservative tracer transport through alluvial aquifers captured by the
497 hydrofacies models built upon the well-known geostatistical tool T-PROGS (Carle and Fogg, 1996,
498 1997; Carle, 1999). Various scenarios of the hydrofacies models significantly expanded upon the
499 original hydrofacies model for the MADE aquifer developed by Bianchi and Zheng (2016). This
500 expanded analysis was conducted to address the following two questions: (1) there is a historical
501 debate about whether the hydrofacies models for alluvial settings can produce super-diffusion
502 (Zhang et al., 2013); and (2) the detailed impacts of the hydrofacies' thickness/width and the initial
503 source size on super-diffusion remain obscure (Bianchi and Zheng, 2016). By combining Monte
504 Carlo simulations and stochastic model analysis, this study yielded the following five main
505 conclusions, not previously identified, that improve our understanding for the characteristics and
506 description of super-diffusion processes in complex alluvial aquifer settings.

507 First, Monte Carlo simulations revealed a bimodal velocity distribution with two peaks, which
508 may explain the bimodal distribution of the plume snapshots observed at the MADE site. The
509 bimodal velocity distribution is likely caused by the contrasting K between the hydrofacies HCG
510 and the other hydrofacies at the MADE site (where the K for HCG is two orders of magnitude
511 higher than the other hydrofacies). The 1st peak of the velocity distribution (representing the
512 contribution from the fine grain hydrofacies and not the HCG) captures the relatively low velocity
513 zones that are primarily responsible for delayed transport and the positively skewed plume

514 snapshots. The 2nd peak of the velocity distribution (due to the HCG distribution) accounts for fast
515 motion of solute particles along the preferential flow pathways and the resultant enhanced “heavy”
516 leading plume front observed for the MADE tests. Super-diffusive transport due to the 2nd peak of
517 the velocity distribution can dampen quickly in space and time due to the finite size of the
518 preferential flow paths, making the detection of super-diffusion difficult in real-world aquifers.

519 Second, the thickness and width of the hydrofacies (especially the high-*K* HCG) can exhibit
520 different impacts on the spatial pattern of bimodal super-diffusion and associated solute transport.
521 A larger width for the hydrofacies enhances the connectivity of high-permeability deposits
522 (“channels”), resulting in higher values in the bimodal velocity distribution and the enhanced
523 “heavier” plume front. The opposite impact on super-diffusion is identified for the hydrofacies’
524 mean thickness; i.e., a thicker hydrofacies can retard more solutes near the source and shrink the
525 plume’s leading front.

526 Third, the size of the initial source affects dynamics of bimodal super-diffusion, due to the
527 fact that the initial source size controls the initial velocity distribution of solute particles.
528 Particularly, a larger initial source results in a more positively skewed plume snapshot, as the
529 particles can experience a wider distribution of velocities (covering both slower and larger
530 velocities). Contrarily, a point source tends to generate a single peak in the plume mass snapshot
531 at early time due to the relatively narrow range of starting velocities and then transitioning to a
532 bimodal pattern in the later plume snapshots after particles sample (experience) greater local
533 velocity variation over time and space..

534 Fourth, the multi-domain spatial non-local transport model (7), which can be extended from
535 the distributed-order fractional derivative model, can quantify the bimodal super-diffusive
536 transport obtained from the Monte Carlo simulations and the bi-peak tracer snapshots observed
537 at the MADE-2 site. This model may also be applicable for the other aquifers where bi- or multi-
538 peak plumes and/or BTCs were observed. Notably, the slow and fast transport in different domains
539 account for the two peaks of the bimodal velocity distribution, and therefore the stochastic model
540 can capture the negatively skewed plume for pollutants undergoing super-diffusion. The generally
541 well fit shows the applicability of the MD-tsFDM model proposed by this study.

542 Fifth, as discussed in the supplementary material, additional Monte Carlo simulations and
543 stochastic model analyses are needed to expand the hydrofacies model dimension and capture
544 mixed super- and sub-diffusion processes in alluvial aquifer systems. Our preliminary experiments
545 (shown in the supplementary material) show that the 3-*d* hydrofacies models can enhance the 2nd,
546 fast peak in the bimodal super-diffusion and generate enhanced “heavier” leading plume edges
547 (fronts) than the 2-*d* models, since the lateral extension of the high-*K* hydrofacies enhances the
548 interconnection of high-*K* materials and generates more preferential flow paths for an extended,
549 more pronounced plume front. In addition, the time nonlocal transport components need to be
550 added in the transport model to account for solute retention in complex alluvial aquifers where the
551 Peclet number is small and molecular diffusion controls solute retention. Extensions of both the
552 hydrofacies model and the mobile-mobile model will be discussed in the next study.

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723 **Table 1.** Geostatistics and hydraulic properties of each of the five hydrofacies. In the legend, “*K*” denotes
 724 the hydraulic conductivity, “HCG” represents Highly Conductive Gravel, “GS” represents gravel with
 725 sand, “SGF” represents Sand Gravel and Fines, “SG” represents Sand and Gravel, “S” represents well-
 726 sorted Sand, and “*Pe*” denotes the Peclet number.

Hydrofacies	Width (m)	Thickness [m]	Proportion [-]	Porosity [-]	<i>K</i> [m/d]	<i>Pe</i> [-]
SGF	39	0.9	0.35	0.259	2.52	1.68×10^4
S	35	1.7	0.21	0.415	5.65	5.64×10^4
HCG	30	1	0.12	0.265	303.39	4.54×10^6
SG	25	0.4	0.14	0.298	7.74	8.27×10^4
GS	31	0.5	0.18	0.257	6.76	6.97×10^4

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728 **Table 2.** Characteristic parameters of the velocity PDF. In the legend, v_{p1} and v_{p2} denote the peak of
729 the low and high velocity zones, respectively; v_m is the velocity with the lowest probability distributed
730 between v_{p1} and v_{p2} ; and $P(v \leq v_m)$ denotes the percentage of the low velocity zone. The units for
731 velocity are m/d.

Mean Length	Scenario 1 (1.0Z)	Scenario 2 (1.5Z)	Scenario 3 (2.0Z)	Scenario 4 (2.5Z)	Scenario 5 (1.25X)	Scenario 6 (1.5X)	Scenario 7 (2.0X)
v_{p1}	0.32	0.32	0.37	0.37	0.32	0.24	0.28
v_{p2}	3.89	3.39	1.95	1.95	4.47	5.89	6.76
v_m	1.70	1.70	1.70	1.70	1.94	1.94	2.24
$P(v \leq v_m)$	0.91	0.92	0.94	0.92	0.90	0.90	0.89

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733 **Table 3.** The best-fit parameters of the MD-tsFDM (7) for all scenarios. The superscript “*” denotes the
 734 hydrofacies simulated conditionally, and “SL” means the vertical source length.

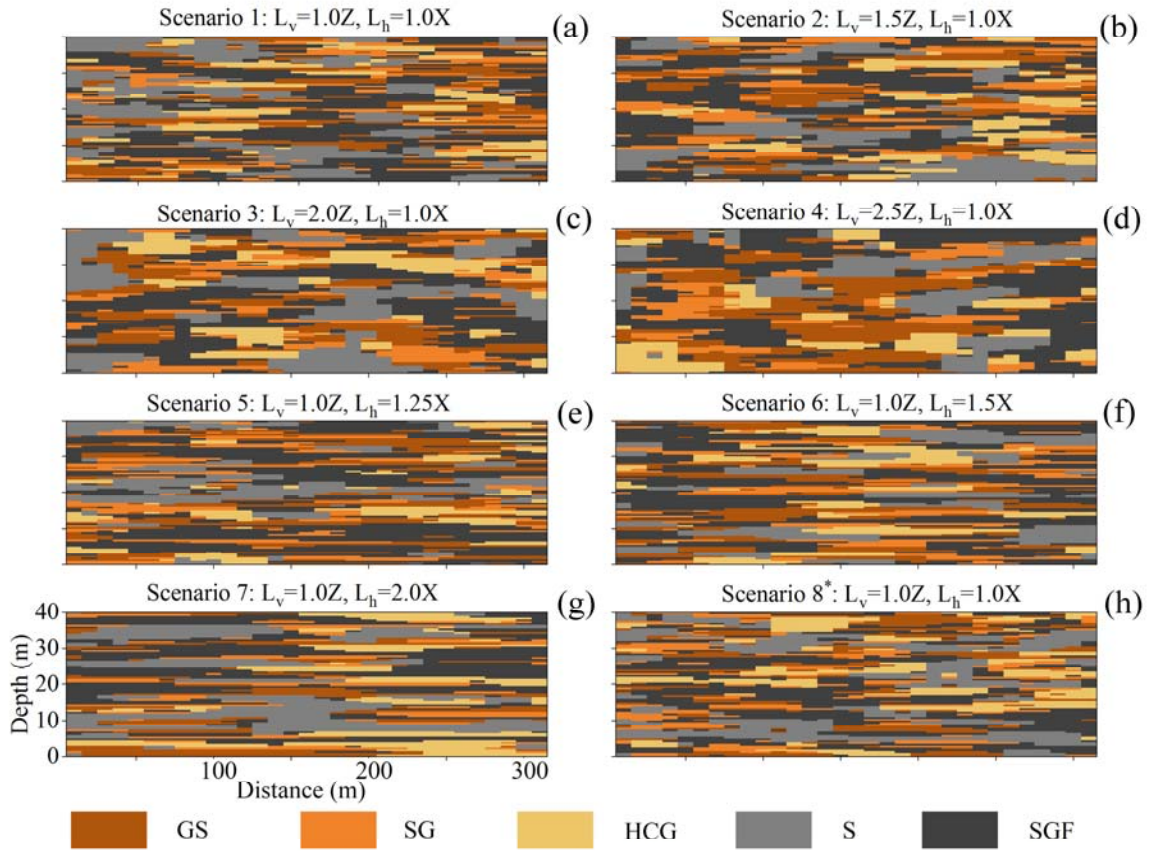
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Scenario	Mean length	v_1 [m/d]	v_2 [m/d]	D_1 [m ² /d]	D_2 [m ² /d]	α_1 [-]	α_2 [-]	λ_1 [m ⁻¹]	λ_2 [m ⁻¹]	ω [mgL ⁻¹ d ⁻¹]
1	1.0X 1.0Z	0.15	0.6	0.5	1.9	1.3	1.25	0.005	0.03	0.0005
2	1.5Z	0.13	0.6	0.45	2.1	1.3	1.3	0.007	0.025	0.0006
3	2.0Z	0.11	0.55	0.5	1.2	1.1	1.3	0.01	0.03	0.0004
4	2.5Z	0.11	0.5	0.55	1.4	1.1	1.3	0.02	0.025	0.0007
5	1.25X	0.15	0.85	0.5	1.9	1.3	1.3	0.008	0.02	0.0007
6	1.5X	0.25	0.9	0.6	2.1	1.1	1.4	0.000 1	0.015	0.0008
7	2.0X	0.17	1.45	0.55	1.8	1.1	1.4	0.003	0.000 01	0.0012
8* (SL=40m)	1.0X 1.0Z	0.1	0.8	0.3	2.4	1.25	1.15	0.01	0.03	0.001
8* (SL=2m) 27 days	1.0X 1.0Z	-	1.05	-	2.7	-	1.5	-	0.02	-
8* (SL=2m) 132 days	1.0X 1.0Z	0.45	1.25	1.25	1.9	1.25	1.25	0.015	0.06	0.0006

736 **Table 4.** Additional scenarios built in **section 4.4** to explore the impact of HCG properties on bimodal
 737 super-diffusion.

Scenario	HCG	HCG Width	HCG	Other hydrofacies	Other hydrofacies
	Thickness		proportion	thickness	width
A1	1.0Z	1.0X	7%	1.0Z	1.0X
A2	1.0Z	1.0X	17%	1.0Z	1.0X
A3	1.0Z	1.5X	12%	1.0Z	1.0X
A4	1.0Z	1.0X	12%	1.0Z	1.5X
A5	2.5Z	1.0X	12%	1.0Z	1.0X
A6	1.0Z	1.0X	12%	2.5Z	1.0X

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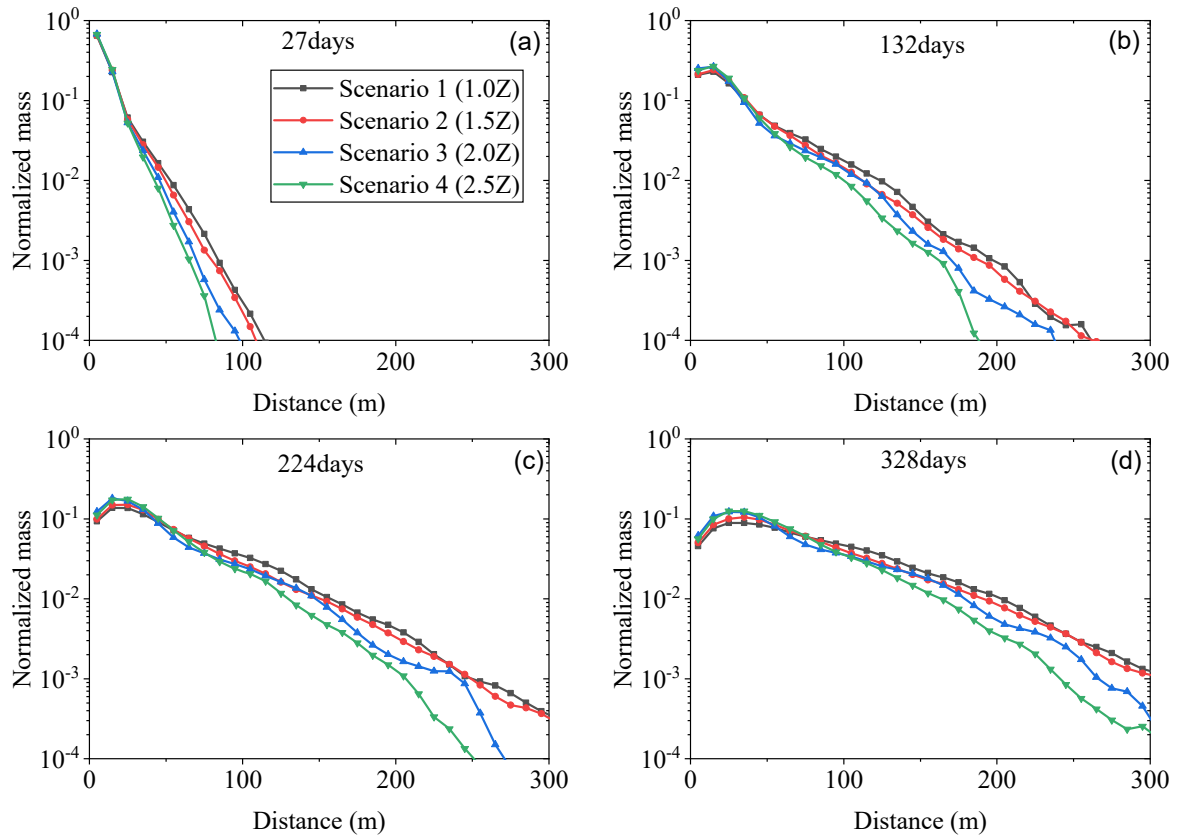
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Fig. 1. One realization of the hydrofacies model for each scenario (8 scenarios total). L_v and L_h are mean length of hydrofacies in vertical and horizontal direction, Z and X denote the mean thickness and longitudinal mean length for the hydrofacies for the base case (which is Scenario 1), respectively.

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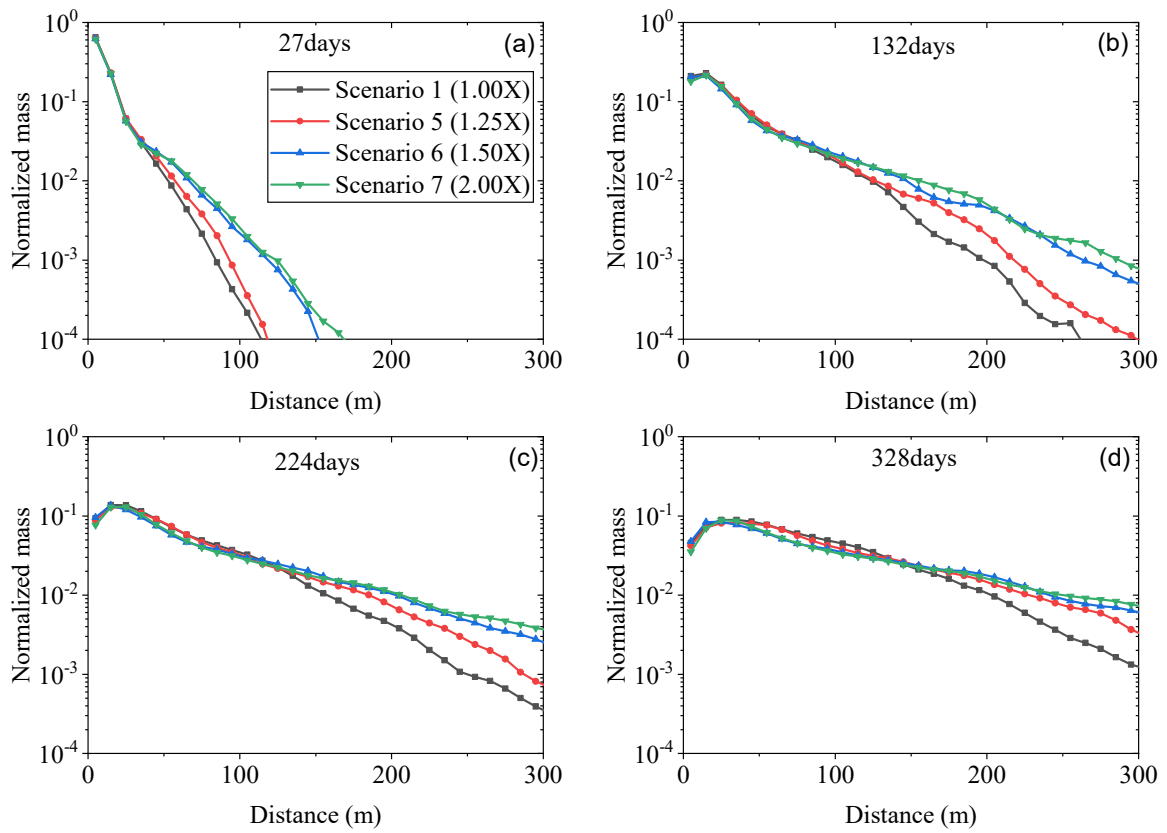
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Fig. 2. The simulated normalized mass distribution at 27 days (a), 132 days (b), 224 days (c), and 328 days (d) after the instantaneous source was released, for four scenarios of hydrofacies models with different vertical mean lengths. In the legend, “1.0Z, 1.5Z, 2.0Z, and 2.5Z” denote that the vertical mean length is 1.0, 1.5, 2.0, and 2.5 times of the base case, respectively.



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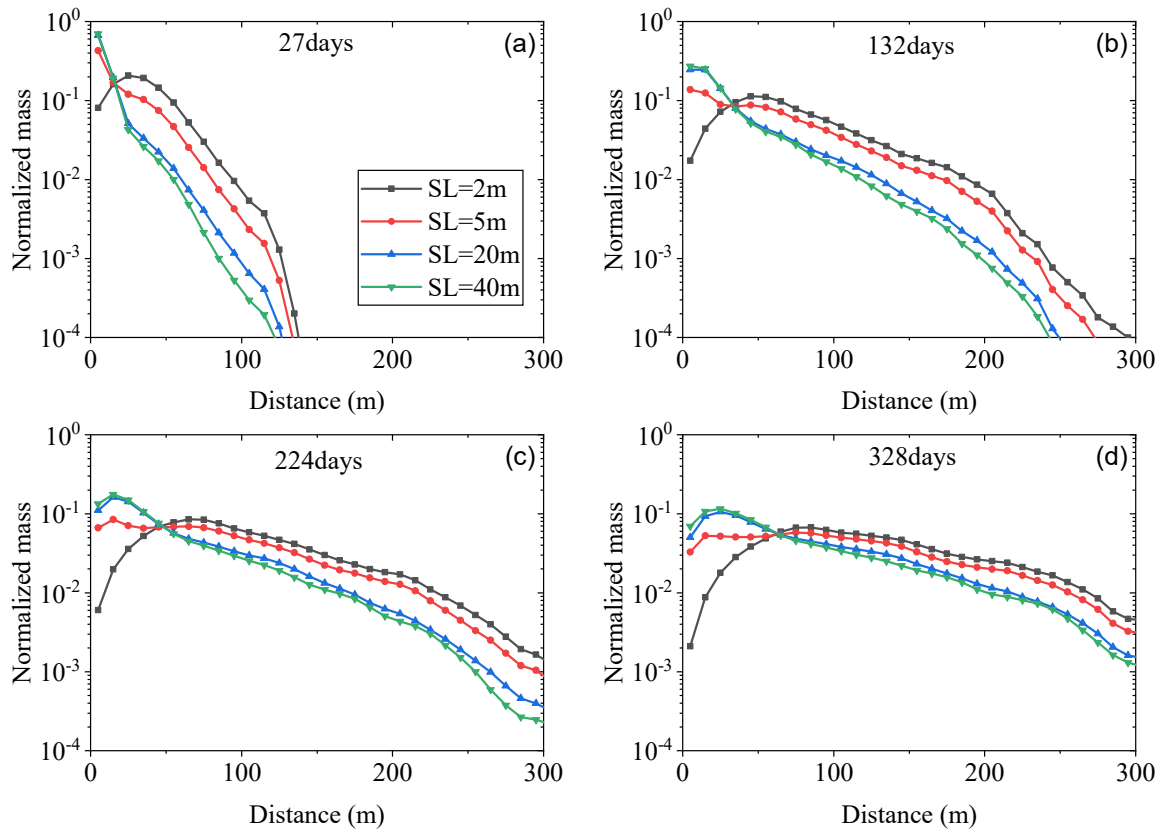
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Fig. 3. The simulated evolution of (normalized) plume snapshots at 27 days (a), 132 days (b), 224 days (c), and 328 days (d) after the release of an instantaneous source for scenarios of hydrofacies models with different longitudinal mean lengths. The legend “X” denotes the longitudinal mean length, and the number “1.0, 1.5, 2.0 and 2.5” denotes the ratio of the longitudinal mean length between the scenario and the base case.



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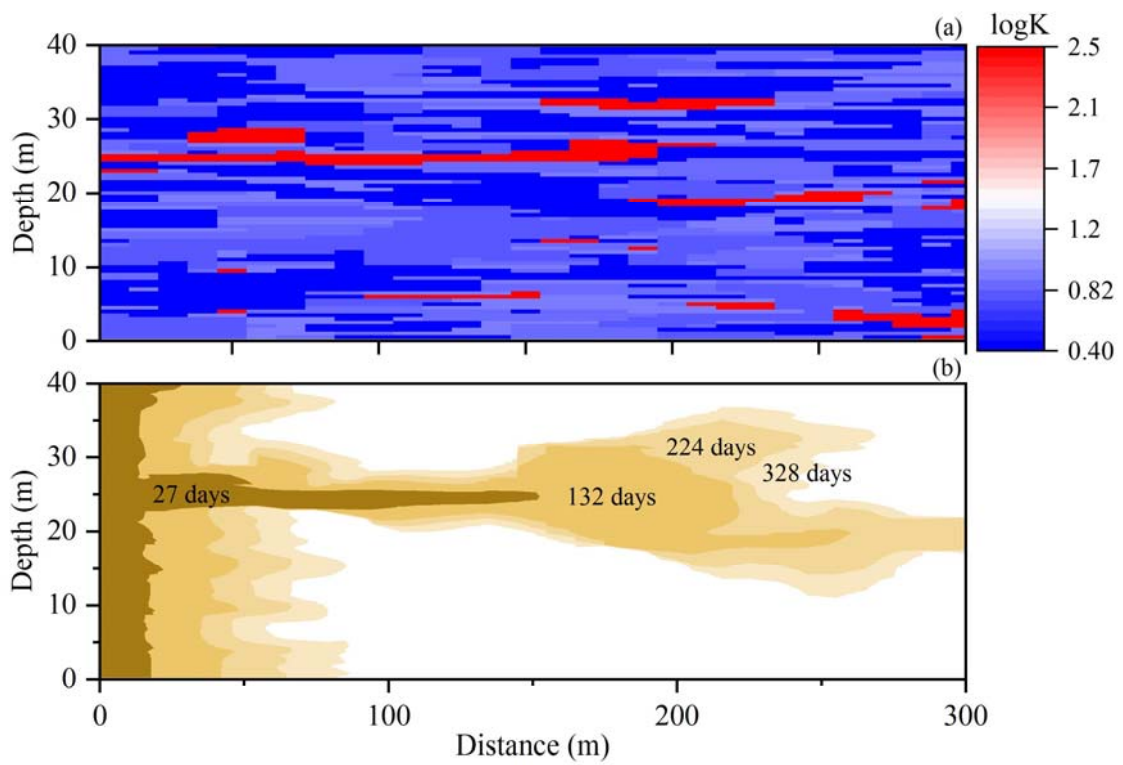
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Fig. 4. The simulated and normalized mass distribution for contaminants at 27 days (a), 132 days (b), 224 days (c), and 328 days (d) after the source release for scenarios with various sizes of the initial line source (scenarios 8). The legend “SL=2m” means that the source length is 2 m.



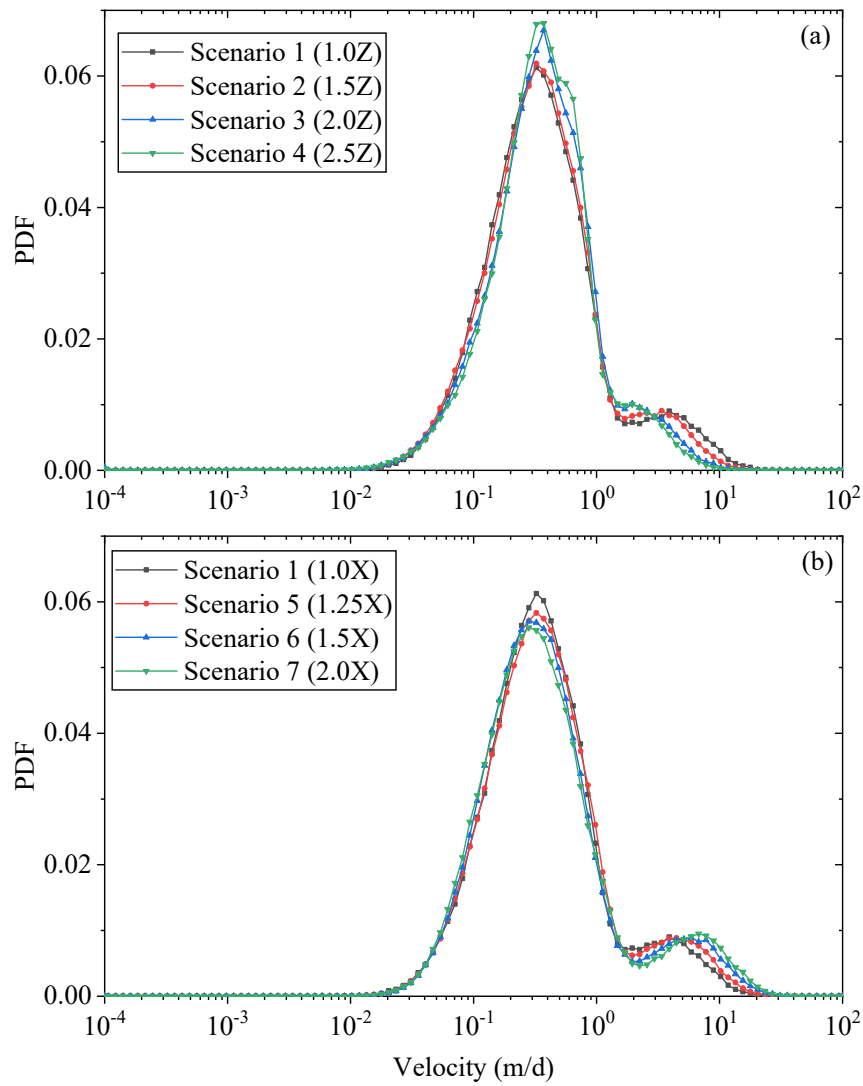
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Fig. 5. The simulated spatial distribution of $\log_{10}(K)$ field (a) and the temporal evolution of the simulated plume front ($C = 0.01C_0$) (b), for one realization of Scenario 1.

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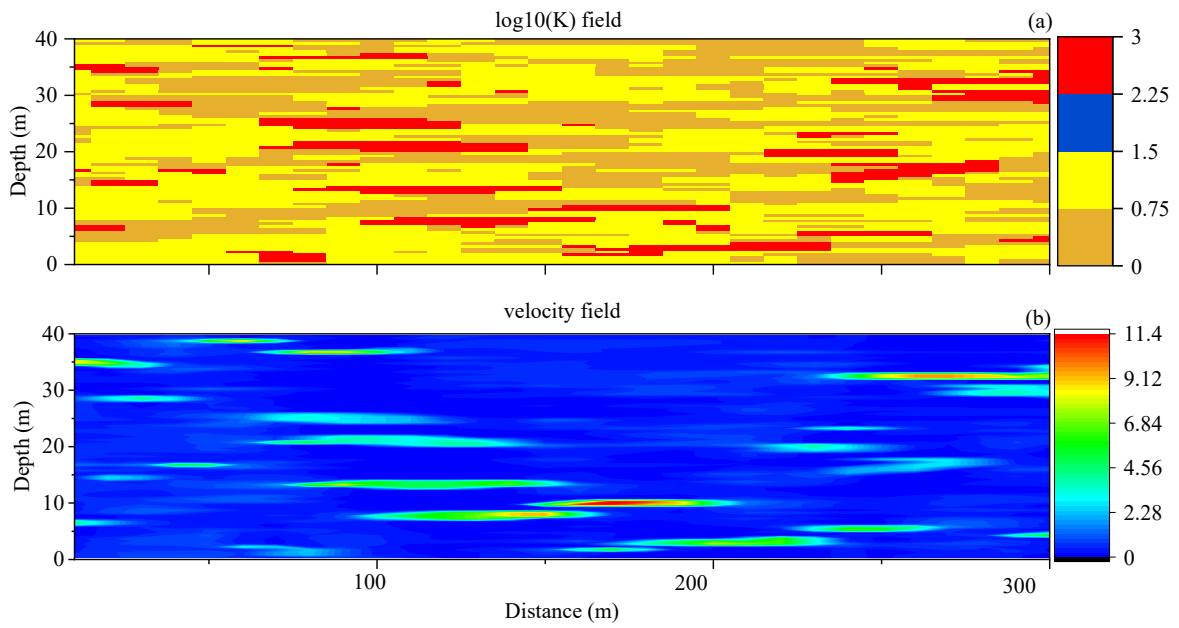
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Fig. 6. The PDF of the velocity for scenarios with different thicknesses (a) and different widths (b).

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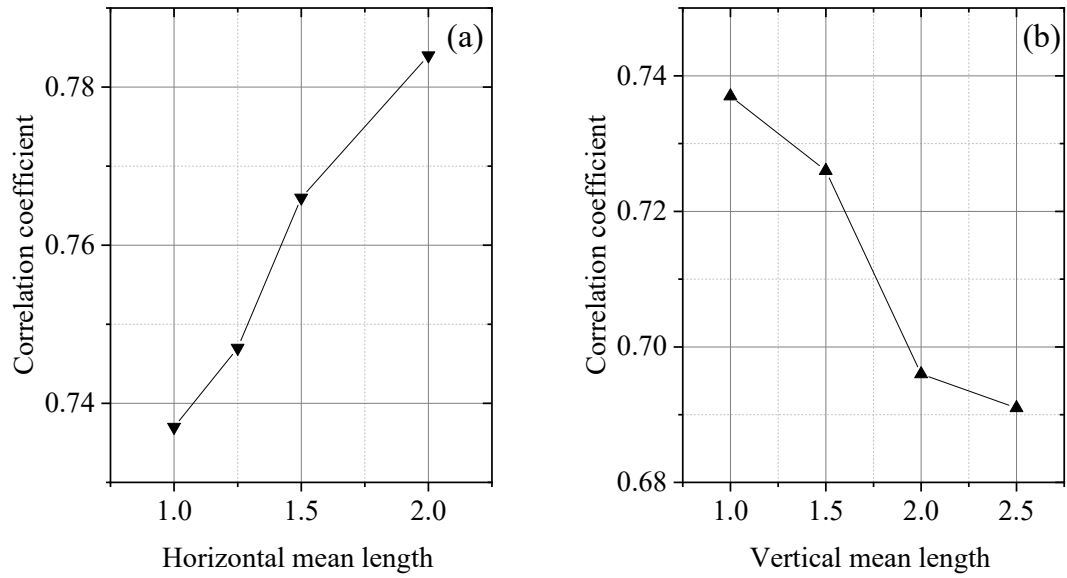
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Fig. 7. One realization of the simulated spatial distribution of hydraulic conductivity and the longitudinal velocity for scenario 1: the $\log_{10}(K)$ field (m/d) (a) and the corresponding spatial distribution of velocity (m/d) (b).



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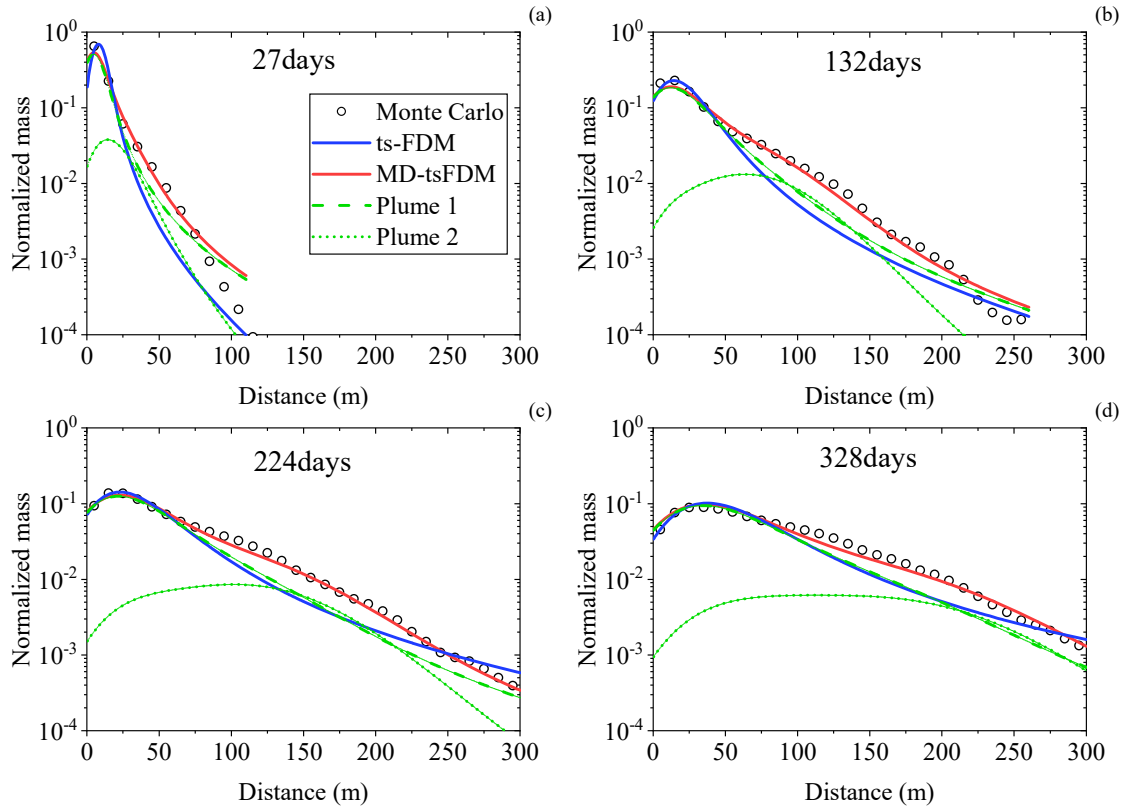
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Fig. 8. The correlation coefficient between the spatial distribution of hydraulic conductivity and the longitudinal velocity for scenarios with different longitudinal mean lengths (Scenarios 1, 5, 6, and 7) (a) and mean thicknesses (Scenarios 1, 2, 3, and 4) (b).



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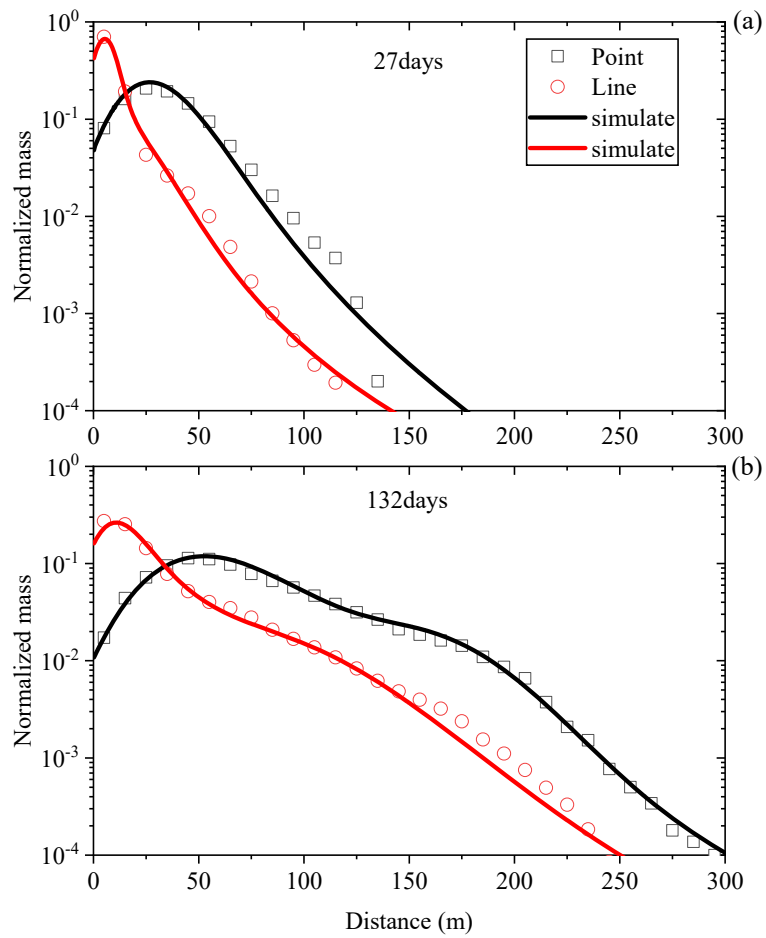
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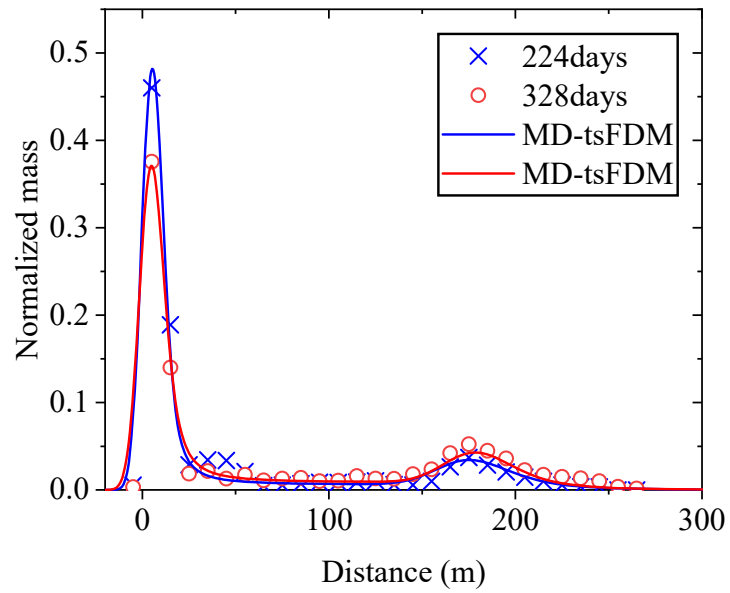
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Fig. 9. The Monte Carlo results for scenario 1 (symbols) versus the best-fit snapshots for the tempered space fractional-derivative model with a single domain (the blue line, e.g., tsFDM) or two domains (the red line, e.g., MD-tsFDM) at 27 days (a), 132 days (b), 224 days (c), and 328 days (d) after the source release. Plume 1 (green dash line) and plume 2 (green dot line) denote the plume of MD-tsFDM within domain 1 and domain 2, respectively.



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Fig. 10. The Monte Carlo results (symbols) versus the best-fit solutions (lines) using the MD-tsFDM for the plume snapshot at 27 days (a) and 132 days (b), respectively, after releasing an instantaneous point source (the black rectangles) or a line source (the red dots) for Scenario 8.



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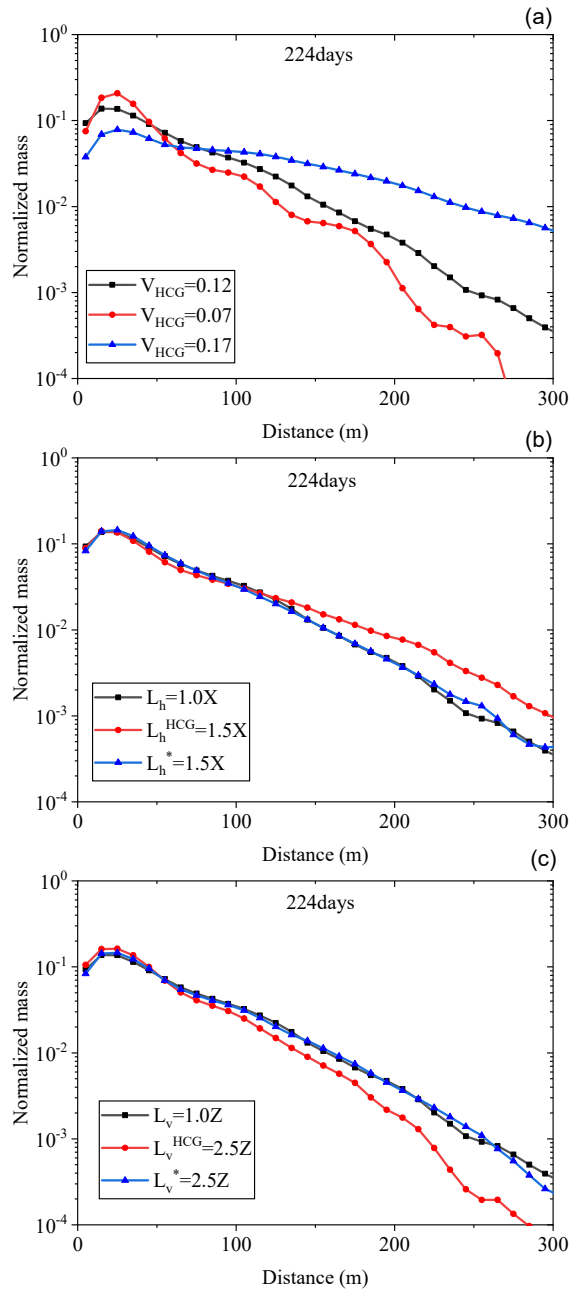
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Fig. 11. The longitudinal mass distribution of tritium observed in the MADE-2 experiment (symbols) and the best-fit results (lines) using the MD-tsFDM (7) at the sampling cycle 224 days (blue) and 328 days (red), respectively.

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799 **Fig. 12.** Major factors controlling super-diffusion, including the volumetric proportion of HCG (a), the

800 horizontal mean length of HCG (denoted by L_h^{HCG}) and the other hydrofacies (denoted by L_h^*) (b), the

801 vertical mean length of HCG (denoted by L_v^{HCG}) and the other hydrofacies (denoted by L_v^*) (c).