The variation of particle size of colloids in the overlying water due to hyporheic exchange

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Abstract

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38   Colloids are often in forms of inorganic and organic particles whose particle sizes (d_p) are ranging

39   from 1.0 nm to 10.0 μm. On the one hand, their transport processes in the hyporheic zone such as

40   deposition, resuspension, clogging and release are substantially affected by hyporheic exchange. On

41   the other hand, the existence of colloids can modify the hydraulic structure of the hyporheic zone

42   due to clogging. Despite the general acknowledgement that particle size plays an important role in

43   contaminant adsorption and clogging, it remains unclear how the particle size of colloids changes

44   during their transport processes in the hyporheic zone. This study aims to investigate the variation of

45   the particle size of colloids in the overlying water and the effects of settlement and convection-

46   diffusion via laboratory experiment and numerical simulation. The results show that both settlement

47   and convection-diffusion play roles in the exchange of colloids between the stream and the

48   streambed. More specifically, settlement is the dominant factor affecting the exchange process of

49   large-sized particles ($d_p > 3.06$ μm) in the hyporheic zone as the high settling velocity dominates the

50   outflux of colloids in the overlying water; the exchange process of small-sized particles ($d_p < 1.10$

51   μm) is more affected by convection-diffusion and some of them can be released from the streambed

52   to the overlying water; while the exchange process of middle-sized particles (1.10 μm $< d_p < 3.06$

53   μm) is affected by both convection-diffusion and settlement. Thus, this study may provide important

54   insights into the variation of the particle size of colloids in the overlying water and the effects of

55   settlement and convection-diffusion.

56   **Keywords**: Hyporheic exchange; Particle size distribution; Settlement; Colloids

57

1 Introduction

The hyporheic zone is broadly defined as a saturated area beneath the streambed where the overlying water and groundwater exchange in a river system [*Boulton et al.*, 1998; *Triska et al.*, 1989a; *Triska et al.*, 1989b], and it is an area where myriad hydrological processes are coupled in a complex manner [*Cardenas*, 2015] and thus provides an excellent habitat for benthic and interstitial organisms [*Brunke*, 1997; *Baxter*, 2000; *Besemer*, 2015]. Hyporheic exchange induced by hydraulic gradient and local morphology of the streambed plays a critical role in maintaining the biological conditions of the hyporheic habitat and even the whole river ecosystem [*Fraser & Williams*, 1996; *Boano et al.*, 2014; *Fox et al.*, 2014]. Hyporheic exchange enhances the flux of mass (such as water, solutes, colloids and nutrients) and energy between the stream and the subsurface [*Packman et al.*, 2000; *Cardenas*, 2006; *Jin et al.*, 2015] and thus has a direct impact on a wide variety of aquatic organisms from benthic microorganisms to invertebrates, aquatic plants and fish [*Findlay*, 1995; *Wagner et al.*, 2014; *Stubbington*, 2012].

Colloids are microscopically dispersed insoluble particles ranging from 1.0 nm to 10.0 μm in particle size and exist widely in natural rivers in forms of inorganic and organic particles [*Huettel et al.*, 1996]. The inorganic colloids include metal oxide, clay minerals, silicate and carbonate, while the organic ones include corrosive materials, viruses, bacteria and synthetic organic macromolecules [*Pretty et al.*, 2006]. Colloids can significantly affect the aquatic environment and ecosystem in the hyporheic zone. The sedimentation of colloids in the hyporheic zone may affect the diversity and metabolism of streams [*Mathers et al.*, 2014; *Partington et al.*, 2017; *Wharton et al.*, 2017], because colloids are more likely to be retained in the porous media due to filtration, which can clog the streambed and thus reduce the bed permeability and porosity [*Rehg et al.*, 2005; *Jin et al.*, 2019a].

80   This may reduce the connectivity between streams and aquifers [*Fox et al.*, 2018] and consequently

81   have an impact on the hyporheic ecosystem functioning and river resilience [*Boano et al.*, 2007;

82   *Benjankar et al.*, 2016; *Stubbington*, 2012]. Colloids can also act as important carriers of

83   contaminants in the hyporheic zone [*Jin et al.*, 2019a; *McCarthy & Zachara*, 1989], as their large

84   specific surface area makes it possible to adsorb more heavy metals, radioactive substances and

85   organic pollutants [*Bradl*, 2004]. Many studies have demonstrated that colloidal particles in the solid

86   phase can move in subsurface environments and thus carry contaminants to the groundwater [e.g.

87   *Ryan & Elimelech*, 1996; *Guo & Chorover*, 2003, *Zhang et al.*, 2005].

88   Due to the important roles of colloids in the hyporheic zone, many studies have been done to

89   investigate their transport and retention in the stream and streambed. There is some evidence that the

90   presence of colloids facilitated the transport of hazardous chemicals such as radiative materials and

91   even viruses in porous media [e.g. *Zhuang et al.*, 2003; *Syngouna & Chrysikopoulos*, 2013]. *Kan &*

92   *Tomson* [1990] found that dissolved organics and colloids could increase the transport distance of

93   some hazardous chemicals, such as DDT, by a thousand times or more in groundwater. *Karathanasis*

94   *et al.* [1999; 2000] found that colloids accelerated the transport speed of heavy metals in porous

95   media by one to two orders of magnitude. *O'Carroll et al.* [2012] found that colloids affected by

96   natural organic matter could be more mobile in the hyporheic zone and thus enhanced the transport

97   process of arsenic. Many researches have also investigated the function and distribution of colloids

98   in river systems via field investigation. *Zhang et al.* [2016] found that most colloids in the overlying

99   flow were fine particles of 1-10 μm, while those in the fracture flow (deeper underground) were 1 or

100   2 orders of magnitude larger. *Chupakova et al.* [2018] found that the transformation of dissolved

101   organic matter was mainly affected by organo-mineral colloids in boreal and subarctic rivers.

Drummond et al. [2014, 2017] conducted injection experiments at different reaches and found that sediments and macrophytes on the streambed were important reservoirs for fine particles. In terms of laboratory experiments, *Packman et al.* [2000] investigated the exchange flux of kaolinite colloids at the surface of stream-streambed, and they proposed a tentative model to describe colloid and solute exchange and verified that colloids were extensively trapped in the streambed compared with that of conservative solute. *Ren & Packman* [2004] found that the immobility of metal ions was significantly enhanced in the presence of colloids in the hyporheic zone via flume experiments. *Fox et al.* [2018] investigated the effects of the deposition of kaolinite colloids on hyporheic exchange under losing and gaining flow conditions through flume experiments. *Jin et al.* [2018] found that the transport of nano-sized colloids in the hyporheic zone was affected by gravity. *Jin et al.* [2019a] investigated the transport and distribution of colloids in the hyporheic zone via laboratory experiment and found that most of colloids were retained near the shallow layer (less than 5 cm) of streambed. Furthermore, numerical simulations and modified models were built previously. *Karwan & Saiers* [2012] conducted simulations to examine hyporheic exchange and streambed filtration of suspended particles but without considering particle settling. *Camarena et al.*, [2016] modified a process-based multiphase exchange model for the p,p-DDE kinetic adsorption to and desorption from the bed colloids and verified it by experiments. *Jin et al.* [2019b] modified the transport model of colloids by taking into consideration the settlement and pore clogging in the hyporheic zone. *Preziosi-Ribero et al.* [2020] developed a stochastic process deposition numerical particle tracking model in the hyporheic zone verified by previous colloids deposition experiments in flumes, and found that the spatial pattern of particle deposition was mainly affected by pore water velocity and the particle concentration was mainly affected by the filtration dynamics.

124   However, it is important to note that the particle size is often assumed to be uniform in most

125   previous studies. It remains unclear how the particle size of colloids changes during their transport

126   processes in the hyporheic zone. In order to investigate the variation of colloidal particle size in the

127   overlying water, we conducted flume experiments and simulations to analyze the effects of settling

128   velocity and mass transfer coefficient on the exchange process of colloids. This study may elucidate

129   1) the mechanisms of the variation of the particle size during the transport process of colloids in the

130   overlying water; and 2) how different-sized particles are affected by settlement and convection-

131   diffusion in the overlying water.

132   **2 Laboratory experiment**

133   In order to investigate the transport process of colloids in the hyporheic zone and the variation

134   of colloidal particle size induced by hyporheic exchange, a flume experiment was conducted in this

135   study [*Elliott and Brooks*, 1997; *Packman et al.*, 2000; *Ren and Packman*, 2004; *Jin et al.*, 2019a].

136   A recirculating flume was used to model the 2D longitudinal section of a river with uniform

137   triangular bedforms (Fig. 1a). The wall of the flume is made of toughened transparent glass for easy

138   observation. A transducer is installed at the tail end of the sink to monitor the temperature,

139   conductivity, pH and salinity of the overlying water.

140   The bed sand used in this study was silica sand collected from the Yangtze River and sieved to

141   0.25 - 0.60 mm with a median of d_s = 0.387 mm. The bed sediment was required to be clean with no

142   impurities, metals and organic matters, and bed sand was washed before the experiment as follows:

143   1) deionized water for four times, 45 mins each time; 2) an acidic solution at pH 3.5 for 8-9 h; 3)

144   repeat step 1 three more times; 4) repeat step 2 with a solution at pH 10.5; and 5) repeat step 1 three

145   times. In the experiment, the pH of the flume effluent was 6.8-7.1. After washing, the bulk porosity

146　(θ = 0.33) and hydraulic conductivity (K = 8.84 × 10^{-4} m s^{-1}) were tested by the water evaporation

147　method and the constant-head method with a Darcy column, respectively. These coefficients were

148　assumed to be constant throughout the experiment and simulation [*Jin et al.*, 2019].

149　　Kaolinite colloids (Jihong Factory, Nanjing, China) were pretreated with 2M NaCl solution

150　prior to experiment to avoid coagulation during the experiment [*Jin et al.*, 2019a]. Kaolinite was

151　converted to homoionic sodium kaolinite by stirring for a day in NaCl solution [*Packman et al.*,

152　1996], and then rinsed several times with deionized water until the NaCl concentration was 20 mM

153　in the rinse solution. The particle size distribution was measured after pretreatment. No significant

154　change was observed over time, indicating no coagulation of colloids [*Packman et al.*, 1996].

155　　Clean sand was packed in the flume and triangular bedforms were made to enhance the

156　hyporheic exchange flux. The effective length of the sand bed is about 10 m; the length of a single

157　bedform is 15.5 cm; and the thickness varied from 12.0 cm at the trough to 14.0 cm at the crest

158　(Table 1). Before the experiment, the stability of the bedforms was tested at different flow velocities

159　and water depths of the overlying water. The velocity of the overlying flow was set to 13.3 cm s^{-1},

160　and the average depth was set to 10.75 cm (Table 1). Subsequently, NaCl was mixed with the

161　overlying water to reach an initial ionic strength of 5 mM based on the previous work [*Packman et*

162　*al.*, 2000; *Jin et al.*, 2019a]. Similarly, kaolinite was added to obtain an initial colloid concentration

163　of 0.2 kg m^{-3} in the overlying water. Note that both NaCl and colloids were initially absent in the

164　bed. NaCl and colloid concentrations were determined from samples collected at different times

165　during the experiment. The experiment would continue until (quasi-) stable NaCl and colloid

166　concentrations were obtained in the overlying water.

167　　In order to monitor colloid concentration in the overlying water, samples were collected more

168 frequently at the beginning of the experiment, at which the concentration would change more

169 rapidly [*Packman et al.*, 2000; *Jin et al.*, 2019a]. The samples were collected from three different

170 positions (5 cm to the two tail ends of the flume and the very middle of the flume) in the overlying

171 water within 30 min (Fig. 1a), and after 30 min as the concentration of colloids was basically the

172 same in the flume. The concentration of colloids was measured using an ultraviolet

173 spectrophotometer. The light absorbance was determined for each sample at 20°C, which was then

174 converted to the colloid concentration based on the calibrated linear relationship between them [*Jin*

175 *et al.*, 2019a].

176 Samples were selected at 0, 2.0, 7.8, 17.6, 26.0 and 39.2 h for measurement of particle size

177 distribution using an automatic laser particle size analyzer (LS13320). To validate the accuracy of

178 the analyzer, the same colloid samples were measured three times, and the maximum mean variance

179 of the three measurements was only 0.113%.

3 Colloid exchange theory

181 The transient storage model, which is widely use in the hyporheic exchange [eg. *Bencala and*

182 *Walters*, 1983; *Marion et al.*, 2003; *Marion et al.*, 2008; Briggs *et al.*, 2009; *Zaramella et al.*, 2006]

183 is used to simulate the transport of colloids in the hyporheic zone. The model is described as

184 follows:

185
$$\frac{dC}{dt} = \alpha(S - C)$$
(1)

186 where C (kg m^{-3}) is the concentration of colloids in the overlying water, S (kg m^{-3}) is the colloids in

187 the pore water, t (s) is the time, and α (s^{-1}) is the mass transfer coefficient, which takes into the

188 concentration convection-diffusion induced exchange consideration (Fig. 1b).

3.1 Modified control equation with settlement

The original model focuses on only one coefficient, the mass transfer coefficient (α). However, the settlement of colloids is also an important factor for the hyporheic exchange [*Jin et al*. 2019a]:

$$\frac{dC}{dt} = \alpha(S - C) - \frac{v_s}{H}C \tag{2}$$

where v_s (m s^{-1}) is the settling velocity of colloids, and H (m) is the depth of the overlying water. According to Eq. 2, the hyporheic exchange of colloidal particles is affected mainly by 1) particle characteristics (settling velocity) and 2) hydraulic characteristics (mass transfer coefficient).

The mathematical derivation steps of Eq. 2 are shown below: 1) Consider only settlement, and the overlying water is divided into several vertical columns (blue dashed boxes in Fig. 1c.) and the bottom area of each column is A, and its height (water depth) is H. 2) Assuming that the column is filled with particles, and the concentration is C_1 at time t_1 and C_2 at t_2 after Δt, respectively. If only settlement is considered, the mass of particles settled on the top of the streambed or penetrated into the streambed during Δt is $\Delta t \, v_s A C_1$. Therefore, the mass in the overlying water is $\left(H - \Delta t \, v_s \right) A C_1$.

3) As shown in the right blue dashed box, the mass in the overlying water can be represented as HAC_2. Then, $\left(H - \Delta t \, v_s \right) A C_1 = HAC_2$, which can be converted into $\dfrac{C_2 - C_1}{\Delta t} = -\dfrac{v_s}{H}C_1$ and written as a differential form $\dfrac{dC}{dt} = -\dfrac{v_s}{H}C$. 4) Consider both settlement and convection-diffusion by linear

205 additivity, and then Eq. 2 is obtained.

206   The relationship between the concentrations of particles in the overlying water and pore water

207 can be described as follows:

208
$$SA_s + CA_c = M_0 \tag{3}$$

209 where A_s (m²) is the cross-sectional area of the storage zone, A_c (m²) is the cross-sectional area of the

210 overlying water, and M_0 (kg m⁻¹) is the total mass of particles per length of the stream-streambed

211 system. If we focus on the concentration in the overlying water (C), a new equation can be obtained

212 by combining Eq. 2 and Eq. 3:

213
$$\frac{dC}{dt} = \alpha \frac{M_0}{A_s} - \left[\alpha \left(\frac{A_c}{A_s} + 1 \right) + \frac{v_s}{H} \right] C \tag{4}$$

214   Thus, The control equation for the transport of colloids in the overlying water can be obtained

215 (Eq. 4).

216 **3.2 Analytical solutions and characteristic parameters**

217   Assuming that:

218
$$\begin{cases} K_1 = \alpha \left(\frac{A_c}{A_s} + 1 \right) + \frac{v_s}{H} \\ K_2 = \alpha \frac{M_0}{A_s} \end{cases} \tag{5}$$

219   Eq. 4 can be simplified into:

220
$$\frac{dC}{dt} = K_2 - K_1 C \tag{6}$$

221   Under the initial conditions, $C(t = 0) = C_0$ (where C_0 is the initial concentration of particles in

222 the overlying water), the final analytical solution for the concentration in the overlying water is

$$C = \frac{K_1 C_0 - K_2}{K_1} \exp(-K_1 t) + \frac{K_2}{K_1}$$

223 (7)

224   Several parameters are defined here to characterize the transport process of particles in the

225 hyporheic zone. The final stable concentration (equilibrium concentration) C_f (kg m^{-3}) is defined as

226 the final stable concentration in the overlying water ($C_f = \lim_{t \to \infty} C$). Thus,

$$C_f = \frac{K_2}{K_1} = \frac{M_0}{(A_c + A_s) + A_s \left(\frac{v_s}{\alpha H}\right)}$$

227 (8)

228   A dimensionless number (settlement-transfer number) N_s ($N_s = \frac{v_s}{\alpha H}$) is defined to characterize

229 the effects of settlement and convection-diffusion on the transport process of particles. Thus,

$$C_f = \frac{K_2}{K_1} = \frac{M_0}{(A_c + A_s) + A_s N_s}$$

230 (9)

231 The arrival time of half equilibrium concentration t_h (s) is defined as the time when $C(t_h) = \frac{C_0 + C_f}{2}$

232 , which can be obtained:

$$t_h = \frac{\ln 2}{\alpha \left(\frac{A_c}{A_s} + 1\right) + \frac{v_s}{H}}$$

233 (10)

234

14

3.3 Modified control equations with release

For small-sized ($d_p < 1.10$ μm) colloids, an obvious "time-lapse release" phenomenon is seen during the transport process, as these particles would be retained instantly by the sand bed and later released over a long period. The release process is assumed to be a Poisson process [*Anderson & Kurtz*, 2011]. The probability of the release at τ is:

$$P(\tau) = \frac{e^{-\lambda\tau}(\lambda\tau)^k}{k!}$$

(11)

where τ (s) is the duration, k is the time for the release of particles, and λ (s^{-1}) is the Poisson process parameter. A new function $\varphi(t)$ is defined from Eq. 11 to represent the probability density of released colloidal particles.

$$\varphi(t) = \lambda t e^{-\lambda t}$$

(12)

Combining Eq. 4, the control equation for small-sized colloidal particles is:

$$\frac{dC_s}{dt} = \alpha_s \frac{M_{0s}}{A_s} - \left[\alpha_s\left(\frac{A_c}{A_s}+1\right) + \frac{v_{ss}}{H}\right]C_s + k_s \int_0^t \frac{\partial S_s}{\partial t_*}\cdot\varphi(t-t_*)dt_*$$

(13)

where α_s (s^{-1}) is the mass transfer coefficient of small-sized particles induced by convection-diffusion, v_{ss} (m s^{-1}) is the settling velocity, C_s (kg m^{-3}) is the concentration in the overlying water, S_s (kg m^{-3}) is the concentration in the pore water, t (s) is the time, k_s (s^{-1}) is the release coefficient, t_* is the time between 0 and t, and M_{0s} (kg m^{-1}) is the initial mass of small-sized particles per length.

251   The release term $k_s \int_0^t \frac{\partial S_s}{\partial t_*} \cdot \varphi(t - t_*)\mathrm{d}t_*$ can be obtained according to Fig. 2. First, we assume

252   that the retained small-sized colloids S_s would be released, and the released concentration is

253   positively related to $\frac{\partial S_s}{\partial t_*}$ (the retained concentration at a given time between 0 to t). The probability

254   density to initial time is $\varphi(t - 0)$ for the initial time (black line in Fig. 2). Similarly, it is $\varphi(t - t_{*1})$, $\varphi(t -$

255    $t_{*2})$ and $\varphi(t - t_{*3})$ for t_{*1}, t_{*2}, and t_{*3}, respectively. Note that the probability density to time t is $\varphi(t - t) =$

256    $\varphi(0) = 0$ for the present time (t). Then $\frac{\partial S_s}{\partial t_*}$ is multiplied by probability densities. The product is

257   taken by integral of t_* from 0 to t , and $\int_0^t \frac{\partial S_s}{\partial t_*} \cdot \varphi(t - t_*)\mathrm{d}t_*$ is obtained. Finally, we time a coefficient

258   k_s which is between 0 to 1 to balance the total released mass and dimension.

259   **4 Results and discussion**

260   **4.1 Variation of concentration and particle size in the overlying water**

261   The transport of colloids was examined by experiment and simulation in this study. The results

262   of this study (Fig. 3) and previous ones [*Packman et al.*, 2000; *Jin et al.*, 2019a] show that the

263   colloid concentration in the overlying water decreases quickly at first and then more slowly, and the

264   final stable concentration is around zero, which verifies the reliability of the flume experiment.

265   Fig. 3 shows the variation of colloid concentration in the overlying water, where the blue stars

266   indicate measured colloid concentrations and the green line is fitted by Eq. 7 by varying α and v_s.

267   The other two lines will be discussed in Section 5.2. The colloid concentration in the overlying

268 water decreases very rapidly within the first 20 h but more slowly from 20 h to 80 h, after which it

269 remains stable (Fig. 3). Within the first 20 h, there is a large difference in the colloid concentration

270 between the overlying water and the streambed, and as a result colloids can be transported to the

271 streambed easily and rapidly. From 20 h to 80 h, the streambed is clogged with colloids and thus the

272 colloid concentration in the overlying water becomes lower, resulting in a decrease in the exchange

273 rate of colloids [*Packman et al.* 2000; *Jin et al.* 2019a; 2019b]. After 80 h, the exchange of colloids

274 between stream and streambed reaches a dynamic equilibrium. This trend can also be deduced from

275 the exponential function described in Eq. 7.

276 Six samples were collected at 0, 2.0, 7.8, 17.6, 26.0 and 39.2 h in the overlying water for the

277 measurement of particle size distribution (red squares in Fig. 3). Fig. 4a & b show the particle size

278 distribution, where the mass percentage is multiplied by the relative concentration (the sum of mass

279 percentages for all-sized particles is $C/C_0 \times 100\%$). A unimodal skewed distribution is observed at 0

280 h (initial distribution), 2.0 h, and 7.8 h; while a bimodal distribution is observed at $t = 17.6$, 26.0 and

281 39.2 h, respectively. As a large amount of overlying water is needed in the measurement of particle

282 size distribution, the sampling time is designed before experiment to avoid systematic errors. Thus,

283 the transitional samples between unimodal and bimodal distribution could not be obtained and we

284 simulated the transitional distribution, as shown in Fig. S1. However, it is important to note that the

285 total mass of small-sized particles is increased from 7.8 h to 17.6 h (Fig. 4c).

286 In order to better understand the particle size distribution, particles are divided into small-sized

287 ($d_p < 1.10$ μm), middle-sized ($d_p = 1.10$-3.06 μm), and large-sized ($d_p > 3.06$ μm), as shown in Fig.

288 4c. The dividing value between small-sized and middle-sized particles ($d_p = 1.10$ μm) corresponds to

the trough of the bimodal distribution (Fig. 4b & c), while that between large-sized and middle-sized

particles (d_p = 3.06 μm) corresponds to the zero value in the bimodal distribution (Fig. 4b & c). In

the following three sections, we will discuss small-, middle- and large-sized particles, respectively.

4.2 Concentration of large-sized particles in the overlying water

The initial mass proportion of large-sized colloidal particles is around 36% (Table 2, Fig. 5a).

The concentration in the overlying water drops very rapidly within the first 10 h, and it is close to

zero after 30 h (Fig. 5a). Given the high settling velocity, the function of convection-diffusion is not

obvious for the large-sized colloidal particles, and it is difficult for retained particles to resuspend

(Table 2).

4.3 Concentration of middle-sized particles in the overlying water

The initial mass proportion of middle-sized colloidal particles is around 39% (Table 2, Fig.

5a). The concentration in the overlying water decreases rapidly within the first 20 h and more slowly

from 20 h to 80 h, after which a stable concentration higher than 0 is seen. The influx of colloidal

particles is induced by both convection-diffusion and settlement, while the outflux is induced by

only convection-diffusion. However, a stable concentration would be reached in the overlying water

when there is a dynamic equilibrium between convection-diffusion and settlement. It is important to

note that convection-diffusion induced exchange has a more significant effect on the transport of

middle-sized colloids compared with the large-sized ones.

Note that a special crest is observed from the particle size distribution at around d_p = 1.8 μm

(Fig. 4b & c, t 1055 min), which means that colloids are weakly attached to the sand bed around

309   d_p = 1.8 μm. The attachment coefficient k_{att} (s⁻¹) is defined to quantify the removal rate of fine

310   particles due to deposition, which can be calculated by the traditional filtration theory [*Bradford et*

311   *al.*, 2003; *Yao et al.*, 1971]:

312
$$k_{att} = \frac{3(1 - \bar{\varepsilon})}{2d} c_e \eta u_c \qquad (14)$$

313   where d (m) is the mean diameter of streambed sediments; $\bar{\varepsilon}$ is the average porosity of streambed

314   surface; u_c (m s⁻¹) is the characteristic pore water velocity defined as $Kh_m / (L\bar{\varepsilon})$ [*Jin et al.*, 2010],

315   in which h_m (m) is the variation of hydraulic head along the sand-water interface, L is the length of a

316   single bedform; c_e is the collision efficiency; η is the collector efficiency which is related to the

317   particle size d_p and can be calculated following the method of *Rajagopalan and Tien* [1976].

318   The larger the k_{att} value is, the larger the removal rate of colloids in the overlying water will

319   be. The relationship between k_{att} and particle size (d_p) for middle-sized colloids is shown in Fig. 6.

320   For the middle-sized particles, the bottom of attachment coefficient and the crest of mass percentage

321   (t = 17.6 h) are almost overlapped around d_p = 1.7~1.8 μm (Fig. 4b & 6). This confirms that a

322   smaller attachment coefficient k_{att} makes it difficult for colloids to be retained in the streambed, and

323   thus the mass in the overlying water is relatively higher.

19   17

4.4 Concentration of small-sized particles in the overlying water

324

325   The initial mass proportion of small-sized colloidal particles is around 25% (Table 2, Fig. 5a).

326   The concentration in the overlying water drops from 0 to 7 h and then increases slightly. After 15 h,

327   it decreases again and finally the C/C_0 is kept at around 0.05 (Fig. 5a). Different from the

328   continuous dropping trend of the concentration of middle- and large-sized colloids in the overlying

329   water, the concentration of small-sized colloids is increased at 7-15 h (Fig. 5a). Based on the

330   assumption in Section 3, the variation trend of small-sized colloids can be well explained (Fig. 5a).

331   The simulated concentrations of small-sized colloids in Fig. 5a (green line) are obtained from

332   Eq. 13, and all coefficients are shown in Table 2. The reason for the decrease of the colloid

333   concentration within the first 7 h is that most colloids are present in the overlying water, and few

334   colloids retained in the streambed could be released back to the overlying water. The increase in the

335   colloid concentration from 7 h to 15 h is attributed to the release of small-sized particles retained in

336   the streambed due to "time-lapse release". The peak of "time-lapse release" is reached at 15 h, after

337   which the concentration of small-sized colloids in the overlying water decreases. Finally, the

338   exchange induced by convection-diffusion and settlement reaches a dynamic equilibrium.

339   The "time-lapse release" results in an increase in the mass of colloids in the overlying water. If

340   "time-lapse release" is not considered ($k_s = 0$), the results are indicated by the black dashed line in

341   Fig. 5a, which is significantly lower than that when the release is considered. In order to analyze the

342   effect of increment of colloids in the overlying water, the incremental ratio of small-sized colloids (

343 $R_s = \dfrac{C_s - C_{s0}}{C_{s0}}$, where C_s is the concentration of small-sized colloids considering release, C_{s0} is the

344 concentration of small-sized colloids without considering release) and to all colloids ($R_a = \dfrac{C_a - C_{a0}}{C_{a0}}$,

345 where C_a is the concentration of all colloids considering release, C_{a0} is the concentration of all

346 colloids without considering release) are calculated and presented in Fig. 5b. It is obvious that the

347 incremental ratio of the released mass to small-sized and all colloids in the overlying water show

348 similar trends. The ratio increases at first until a peak is reached at 30 h, after which it decreases and

349 then becomes stable after 100 h. The ratio to small-sized colloids is close to 1.0 at its peak, which

350 indicates that the maximum proportion of released mass reaches nearly half of all small-sized mass.

351 In addition, the peak ratio to all colloids is around 0.4, which also indicates that there is considerable

352 proportion of released mass. However, the correlation coefficient between measured and fitted data

353 is not significantly increased when the "time-lapse release" is considered, because after 30 h (when

354 the incremental ratio of release is close to peak) the total concentration is not very high and thus less

355 effects on the correlation coefficient (Fig. 5a & b, Section S1).

356 **4.5 Effects of settling velocity v_s and mass transfer coefficient α**

357   According to Eq. 8, assuming that A_s, A_c and M_0 are constant, and the final stable concentration

358 C_f (kg m^{-3}) is determined by the settlement-transfer number N_s ($N_s = \dfrac{v_s}{\alpha H}$). Fig. 7a shows that the

359 final stable concentration C_f differs substantially depending on N_s. The green line (labeled "Fitted

360   curve") in Fig. 7a is fitted with measured data (blue stars) by Eq. 9, and the parameters are shown in

361   Table 2. When $N_s = 0$ ($v_s = 0$ and $\alpha > 0$), the transport of colloids is driven only by convection-

362   diffusion induced exchange, and the final stable concentration in the overlying water ($C_{f\alpha}$) reaches

363   a maximum (blue line in Fig. 7a). When $N_s = \infty$ ($v_s > 0$ and $\alpha=0$), the transport of colloids is

364   driven only by settlement induced exchange, and the final stable concentration in the overlying

365   water (C_{fv}) reaches a minimum of 0 (red line in Fig. 7a). Another dimensionless number N_{se} (equal

366   settlement-transfer number) is defined, which can be considered a special case of N_s when settlement

367   and convection-diffusion play an equal role in the exchange of colloids. Assuming that

368    $C_f = \dfrac{C_{f\alpha} + C_{fv}}{2}$, and combining Eq. 9, the equal settlement-transfer number is $N_{se} = \dfrac{A_c + A_s}{A_s}$, which

369   only relates to A_s and A_c. The blue dashed line in Fig. 7a shows the case when $N_s = N_{se}$. It shows

370   that the larger the N_s value is, the lower the C_f value will be. If N_s is a constant, the final stable

371   concentration C_f will remain the same regardless of the mass transfer coefficient α (Fig. 7b).

372   The arrival times (t_h) of the half equilibrium concentration are different for different mass

373   transfer coefficients (α) (Fig. 7b). This is because t_h is related to both mass transfer coefficient (α)

374   and settling velocity (v_s), as shown in Eq. 10. If $N_s = \dfrac{v_s}{\alpha H}$ is considered, t_h can be described as:

$$t_h = \frac{\ln 2}{\alpha \left[\left(\dfrac{A_c}{A_s} + 1 \right) + N_s \right]}$$

375   (15)

$$t_h = \frac{\ln 2}{v_s \left[\dfrac{1}{HN_s} \left(\dfrac{A_c}{A_s} + 1 \right) + \dfrac{1}{H} \right]}$$

376   or   (16)

377   Assuming that N_s is a constant, the sensitivity curves of parameters α and v_s to t_h can be described

378   by an inverse proportional function, as shown in Fig. 8a. This is because the curve in Fig. 8a is

379   obtained from Eq. 15 and Eq. 16, where α is inversely proportional to t_h if N_s, A_s and A_c are

380   constants, and a similar relationship is obtained between v_s and t_h. Assuming that α and v_s are

381   constants, the sensitivity curve of parameter N_s to t_h is shown in Fig. 8b. The red line in Fig. 8b is

382   plotted by Eq. 15, and α is assumed to be a constant α_0 (the α in an authentic case), which can be

383   approximately described by an inverse proportional function. The blue line is plotted by Eq. 16, and

384   v_s is assumed to be a constant v_{s0} (the v_s in an authentic case). It shows a slightly increasing trend,

385   because N_s and t_h are positively correlated if v_s remains unchanged. It is also noted that v_{s0} has a

386   significant effect on t_h, because the sensitivity of N_s to t_h is weak (Fig. 8b, Eq. 16). Fig. 8c is plotted

387   according to Eq. 10. The red line represents the sensitivity of α to t_h when v_s equals v_{s0}, while the red

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388 line represents the sensitivity of v_s to t_h when α equals α_0. The blue curve is approximately described

389 by the inverse proportional function, while the trend of the red curve is very moderate. Thus, α_0 is

390 relatively small and has little effect on t_h, while v_{s0} is relatively large and has a more significant

391 effect on t_h (Fig. 8c, Eq. 10).

5 Conclusions

393   In this study, both experiment and simulation were conducted to better understand the particle

394 size variation of colloids in the overlying water due to hyporheic exchange between the stream and

395 the streambed. The main conclusions are:

396   1) The particle size of colloids varies during the hyporheic exchange processes, and colloids

397 with different sizes follow different patterns. The concentration of large-sized colloids decreases

398 rapidly and finally gets close to zero in the overlying water, while the concentration of small-sized

399 ones drops more slowly and a considerable amount of them are remained in overlying water.

400   2) The effects of convection-diffusion and settlement are distinct for different particles. Overall,

401 the larger the particle size is, the stronger the effect of settlement will be; while the smaller the

402 particle size is, the stronger the effect of convection-diffusion will be. For large-sized ($d_p > 3.06$ μm)

403 and middle-sized (1.10 μm $< d_p < 3.06$ μm) particles, the effect of settlement is relatively stronger;

404 while for small-sized ($d_p < 1.10$ μm) particles, both settlement and convection-diffusion have

405 obvious effects, and some of colloids can be released from the streambed to the stream.

406   3) The exchange process of colloids is affected by both settling velocity (v_s) and mass transfer

407 coefficient (α). The final stable concentration (C_f) is determined by the settlement-transfer number

408 N_s ($= \dfrac{v_s}{\alpha H}$), and the arrival time of half equilibrium concentration (t_h) is negatively correlated with

409 v_s and α.

410   This study provides some insights in the particle size variation of colloids in the overlying

411 water during the exchange between the stream and the streambed and the effects of settlement and

412 convection-diffusion. More studies are need to elucidate how the particle size of colloids vary in the

413 streambed, and how the porosity and hydraulic conductivity of the streambed change when it is

414 clogged with different sized colloids.

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