Density effects on solute release from streambeds

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Funding information
111 Project, Grant/Award Number: B17015; National Natural Science Foundation of China, Grant/Award Numbers: 51421006, 51679065; Social Development - Major Demonstration of Science and Technology of Science and Technology Projects of Jiangsu Province, Grant/Award Number: BE2018737; The Basic Research Programs (Natural Science Foundation) of Jiangsu Province, Grant/Award Number: BK20171436

Abstract
Contaminants that entered the streambed during previous surface water pollution events can be released to the stream, causing secondary pollution of the stream and impacting its eco-environmental condition. By means of laboratory experiments and numerical simulations, we investigated density effects on the release of solute from periodic bedforms. The results show that solute release from the upper streambed is driven by bedform-induced convection, and that density effects generally inhibit the solute release from the lower streambed. Density gradients modify the pore water flow patterns and form circulating flows in the area of lower streambed. The formation of circulating flows is affected by density gradients associated with the solute concentration and horizontal pressure gradients induced by stream slope. The circulating flows near the bottom of the streambed enhance mixing of the hyporheic zone and the ambient flow zone.

KEYWORDS
circulating flow, mixing, release, solute transport, variable density

1 | INTRODUCTION

Most contaminants, such as metals, colloids and/or nanoparticles, enter streams due to point and non-point sources (Butler, 2009; Palumbo-Roe, Wragg, & Banks, 2012; Ruiping, Huijuan, Dongjin, & Min, 2008). There are numerous studies on the input of metal, phosphorus, organic contaminants, gases, nanoparticles and microorganisms to streams (Butler, 2009; Feijó et al., 2018; Fuller & Harvey, 2000; Jin, Jiang, Tang, Li, & Barry, 2018; Park, Pachepsky, Hong, Shelton, & Coppock, 2017; Quick, 2018; Saup et al., 2017), with consideration of different environmental factors such as pH, ionic strength and particle size (Butler, 2009, 2011; Cheng, Zhu, Zhong, & Wang, 2015). A portion of the contaminants in the stream are transported into the hyporheic zone by convection and diffusion associated with hyporheic flow (Cardenas & Wilson, 2006, 2007; Elliott & Brooks, 1997; Packman et al., 2006; Packman & Jerolmack, 2004; Salehin, Packman, & Paradis, 2017).
Contaminants entering the streambed can be (partly) released back to the stream (Thibodeaux & Boyle, 1987). Nutrients and contaminants released from groundwater into streams can have an adverse impact on the ecological health of surface waters (Gandy, Smith, & Jarvis, 2007; Smith et al., 2008). For example, the exchange between the bottom sediments and the water column induces contaminant release from the streambed into the water column (Linnik & Zubenko, 2000), which can further pollute the stream and impact its eco-environmental condition (Burton & Johnston, 2010). The release of highly toxic chemicals can be long-lasting with a tail of solute concentration within the water column (Gandy et al., 2007).

Pressure gradients due to stream slope affect hyporheic flow (Cardenas & Wilson, 2006; Cardenas, Wilson, & Haggerty, 2008), as does the bed morphology (Cheng, Song, Wang, & Zhang, 2019; Gomez-Velez, Wilson, Cardenas, & Harvey, 2017; Song et al., 2019). Advection induced by stream morphology drives the solute transport more quickly in and from the hyporheic zone than diffusion (Jin, Tang, Li, & Barry, 2011). In addition, other factors (e.g., low hydraulic conductivity blocks in streambed) were also considered to influence contaminants release from the streambed in the hyporheic zone (Herzog, Higgins, Singha, & McCray, 2018). Indeed, the resulting density gradients were found to drive significant hyporheic exchange between a model stream and its flat streamed in a flume experiment (Boano, Poggi, Revelli, & Ridolfi, 2009). More recently, small density variations due to low solute concentrations were demonstrated to influence stream–streambed exchange (Jin et al., 2015). This gravity-driven flow provides an additional advective solute transport mechanism that enhances stream–streambed exchange (Jin et al., 2011, 2015).

The streambed may be divided into two different parts: a hyporheic zone in direct exchange with the overlying stream (interfacial exchange zone) and an ambient flow zone affected by regional groundwater conditions (Jin, Tang, Gibbs, Li, & Barry, 2010). However, the extent to which the interaction between hyporheic and ambient flow affects solute release from the streambed is unclear. How does the variable density affect the release of solute? Does the mixing of the hyporheic zone and the ambient zone become stronger? This study was designed to address these questions through laboratory experiments and numerical simulations.

2 | LABORATORY EXPERIMENTS AND NUMERICAL SIMULATIONS

2.1 | Laboratory experiments

The experiments aimed to investigate solute release from the ‘polluted’ streambed with stationary triangular bedforms. A recirculating, tilting flume consisting of an 11.5-m long, 30-cm wide and 50-cm deep rectangular channel was used in the experiments (Jin et al., 2010, 2015). In this closed system, stream water was continuously recirculated over the sediment bed and then through a return pipe by means of a stainless steel pump. The artificial sand used for the experiment was composed of grains between 0.25 and 0.60 mm in diameter, and was made of 99% pure silicon dioxide. The streambed’s bulk porosity was measured to be 0.46 using the water evaporation method and the saturated hydraulic conductivity, measured using the constant-head method (Chinese National Standard, 1999), was found to be around 4.35 × 10⁻³ m s⁻¹. The flume was filled with a salt solution with a concentration of \( C_0 = 1.32 \text{ kg m}^{-3} \) to the desired channel depth. The sand was poured into the flume channel and triangular bedforms were made manually using a perforated template. Pumping was subsequently started and in order to maintain a stable bedform the overlying water was kept at constant flow over the streambed for 1 day. Following this phase, the overlying water in the flume was replaced quickly with deionised water to start the solute release from the streambed (Jin et al., 2015). The background concentration in the overlying water was \( C_1 = 0.14 \text{ kg m}^{-3} \) after replacing the stream water with deionised water.

Pore water concentrations were measured from samples withdrawn from two vertical arrays of sampling ports (with 1-cm intervals between neighboring ports, Figure 1c) at the stoss and lee slopes of a bedform. The 0.1-ml samples were diluted 51 times with 5 ml of deionised water, and then measured by an electrical conductivity (EC) probe (DDSJ-308A model, Shanghai Precision and Scientific Instrument Company). A self-logging probe (MPTROLL 9500) was installed to measure EC (salt concentration) and temperature in the surface water at the end of the flume throughout the experiment. The depth was measured by ruler with a precision of 0.1 mm. Flow velocity is calculated from \( v = Q/A \), where \( v \) is the flow velocity, \( Q \) is the discharge, and \( A \) is the cross-sectional area for water. The precision of discharge is 0.001 m³ h⁻¹ measured by an electromagnetic flowmeter. The calculation of precision of discharge is: 278 mm³ s⁻¹. Therefore, the precision of the flow velocity measurement is 6.5 mm s⁻¹, that is, 0.1 cm s⁻¹. The average flow velocity and water depth in the stream water were set to be 13.3 cm s⁻¹ and 6.91 cm, respectively.

2.2 | Numerical simulations

The stream water flow was first computed using the CFD (Computational Fluid Dynamics) package FLUENT-based on the Reynolds-averaged Navier–Stokes equations together with the \( k-\omega \) turbulence closure scheme (Jin et al., 2010). The simulated pressure at the streambed surface was then input into a COMSOL-based model as boundary condition to drive coupled pore water flow and solute transport in the bed (Jin et al., 2010).

Density-dependent flow in the bed is governed by the following equation (Bear, 1979):

\[
\rho S_1 \frac{\partial h_1}{\partial t} + \frac{\partial (\rho C)}{\partial x} + \nabla \cdot \left[ -\rho K_1 \left( \nabla h_1 + \frac{\rho - \rho_1}{\rho_1} \nabla k_2 \right) \right] = 0, \tag{1}
\]
where $h_f$ (m), the equivalent freshwater hydraulic head, is given by $\frac{p}{\rho_f g} + x_2$ with $p$ (Pa) the pore water pressure, $\rho_f$ (1000 kg m$^{-3}$) the freshwater density, $g$ (m s$^{-2}$) is the gravitational acceleration and $x_2$ (m) the vertical coordinate directed upward; $C$ (kg m$^{-3}$) is the solute concentration; $\rho$ (kg m$^{-3}$) is the fluid density; $S_f$ (m$^{-1}$) is the equivalent freshwater specific storativity, representing storage due to compressibility of the fluid and porous medium; $t$ (s) is time; $\theta$ is the porosity; and $K_f$ (m s$^{-1}$) is the equivalent freshwater hydraulic conductivity. The density of the fluid as a function of solute concentration is given by:

$$\rho = \rho_f + \frac{\partial \rho}{\partial C} C = \rho_f + \gamma C,$$

where $\gamma$ is 0.7143 (Langevin, Shoemaker, & Guo, 2003).

The second term on the left side of Equation (1) represents the change in storage due to concentration variations and is likely to dominate the compressible storage term due to the relatively small compressibility of the stream sand used in the study (Jin et al., 2010). Coupled with the pore water flow, the salt solute transport in the porous bed is governed by the conservative mass transport equation (Jin et al., 2015):

$$\frac{\partial C}{\partial t} = \nabla \cdot \left( D_i \frac{\partial C}{\partial x_i} - u_l C \right)$$

where $u_l$ ($l = 1, 2$) (m s$^{-1}$) is pore water flow velocity component in the $x_l$ ($l = 1, 2$) (m) direction,
\[ u_1 = -K f \frac{\partial h f}{\partial x_1} \]  
\[ u_2 = -K f \frac{\partial h f}{\partial x_2} + \rho - \rho f \frac{\rho f}{\rho_f} \]  

\( D_{ij} \) (m² s⁻¹) is the component of the 2D dispersion coefficient tensor:

\[ D_{ii} = \alpha_L \frac{u_i^2}{|u|} + \alpha_T \frac{u_i^3}{|u|^2} + D_e \]  
\[ D_{ij} = D_{ji} = \alpha_L - \alpha_T \]  

where \( \alpha_L \) (m) is the longitudinal dispersivity, a property of the porous medium describing dispersive transport in the flow direction; and \( \alpha_T \) (m) is the transverse dispersivity, describing dispersive transport normal to the flow direction. The absolute value \( |u| = \sqrt{u_x^2 + u_y^2} \) (m s⁻¹) is the magnitude of the pore water flow velocity. \( D_e \) (m² s⁻¹) is the effective molecular diffusion coefficient in the porous medium.

The model formulated here is often applied in studies of density-dependent groundwater flow (e.g. Langevin et al., 2003) and involves standard assumptions such as the validity of Darcy's law, the expression for specific storage, the diffusive approach to dispersive transport based on Fick's law and isothermal conditions.

The (homogeneous and isotropic) porous medium is assumed to be fully water-saturated.

The initial and boundary conditions for both the stream water flow model, and pore water flow and solute transport model were set according to Jin et al. (2010, 2011) (Figure S1), simulating the conditions in the laboratory experiments. The computed pressure at the streambed surface from the stream water flow simulation was converted to the equivalent freshwater head for its use in the top boundary condition for the pore water flow simulation. As the solute concentration changes with time due to solute exchange across the bed surface, the converted equivalent freshwater head varied temporally. Periodic boundary conditions were applied to the lateral (vertical) boundaries for velocity, pressure, concentration and solute flux (Figure S1a,b). The bottom boundary was impermeable and subjected to zero-flux conditions. The initial solute concentration was \( C_0 = 1.32 \text{ kg m}^{-3} \) for the pore water and \( C_1 = 0.14 \text{ kg m}^{-3} \) for the overlying stream water (measured background concentration after replacing the stream water with deionised water). The concentration

**TABLE 1** Parameters in the experiment and numerical simulations

| Parameters | Experiment | Simulation |
|------------|------------|------------|
| Average velocity for overlying water (U) | 13.3 cm s⁻¹ | 13.3 cm s⁻¹ |
| Average water depth for overlying water (H) | 6.91 cm | 6.91 cm |
| Initial concentration in bedform (C₀) | 1.32 kg m⁻³ | Varies by cases |
| Slope (J) | 7.1 × 10⁻⁵ | Varies by cases |
| Initial concentration in bedform (C₁) | 0.14 kg m⁻³ | 0 (for discussion) 0.14 kg m⁻³ (for experiment) |
| Bedform length (L) | 15.55 cm | 15.55 cm |
| Stoss length (Lₜ) | 11.05 cm | 11.05 cm |
| Lee length (Lₜ) | 4.5 cm | 4.5 cm |
| Bedform height (Hₖ) | 2.19 cm | 2.19 cm |
| Streambed depth (dₖ) | 13.16 cm | 13.16 cm |
| Hydraulic conductivity (K) | 4.35 × 10⁻³ m s⁻¹ | 4.35 × 10⁻³ m s⁻¹ |
| Bulk porosity (θ) | 0.46 | 0.46 |
| Water density (ρ) | 1000 kg m⁻³ | 1000 kg m⁻³ |
| Water dynamic viscosity (μ) | 0.001 Pa s | 0.001 Pa s |
| Longitudinal dispersivity (αₗ) | 0.001 m | 0.001 m |
| Transverse dispersivity (αₜ) | 0.0001 m | 0.0001 m |
| Effective diffusion coefficient (Dₑ) | 10⁻⁹ m² s⁻¹ | 10⁻⁹ m² s⁻¹ |

**FIGURE 2** Comparison of experimental data and modelled solute concentration in the overlying water. DE, density effect

**FIGURE 3** Comparison of experimental data and modelled solute concentrations in pore water with depth and at different times. (a) N1 array; (b) N2 array; DE, density effect
in the overlying water, $C_t$, changes with solute release from the streambed into the flume water according to:

$$C_t = \frac{\partial C_0 BA + C_1 V_s - B \int_{x_1}^{x_2} \partial C(x_1, x_2, t) dA}{V_s} \tag{8}$$

where $V_s$ ($m^3$) is the volume of overlying water, $C(x_1, x_2, t)$ ($kg \ m^{-3}$) is the simulated solute concentration of pore water at location $(x_1, x_2)$ at time $t$, $B$ ($m$) is the width of the flume, and $A$ ($m^2$) is the area (on the $x_1-x_2$ plane) of the sand bed in the flume. However, the solute concentration of the overlying water in the simulations for extension is 0. In terms of the simulation, to simplify simulation and analysis, the upstream is assumed to be clean water, so it is set to zero in order to avoid the disturbance of varying pressure caused by the change of the concentration in the overlying water.

Two groups of simulations with ($\gamma \neq 0$) and without ($\gamma = 0$) density effects were conducted to examine solute release from the streambed, and to compare with the experimental data. The aim was to ascertain the effects of density gradients on hyporheic flow and solute release from the streambed.

3 | RESULTS

In the experiment, the flume is a closed system. As discussed above, the solute concentration in the overlying water increases as the solute releases from the bed. The values of relevant parameters are given in Table 1, including hydraulic conductivity, porosity and dispersivity. The results from both the experiment and numerical simulations are described and discussed in the following sections. The flow simulation

![Flow simulation](image)

**FIGURE 4** Comparisons of the solute plume, streamlines and flow directions with and without DE. The parameters were set as for the flume experiment at $t = 1440$ min (Table 1)

![Solute plume](image)

**FIGURE 5** Solute plume with different slopes and different initial concentrations in a streambed.  
(a) $t = 3492$ min, $J = 0$, $C_0^* = 9.4 \times 10^{-4}$.  
(b) $t = 3492$ min, $J = 9.8 \times 10^{-4}$, $C_0^* = 1.1 \times 10^{-3}$
results were essential for predictions of solute transport and concentration variations in both the stream and bed. The performance of the flow model is evaluated by the comparison of the predicted and measured concentration profiles presented below.

### 3.1 Concentration variation of solute in the overlying water

The solute concentration in the overlying water increased quickly during the first 200 min, and then the rate of increase reduced (Figure 2). This behaviour shows that the near-surface plume in the streambed was released quickly initially because flow velocity was large near the interface (Cardenas & Wilson, 2007; Jin et al., 2010). This rapid initial change is similar to the reverse situation (solute in the stream, not the streambed) (Jin et al., 2010). The results from the simulation with density effect (DE) in Figure 2 compare well with experimental data, but the simulated concentrations without DE are obviously too large, that is, due to gravity, DE reduces the rate of solute release from the streambed. If the experiment and simulation continued to run for a sufficiently long time, the solute concentration in the pore water would become the same as that in the overlying water, that is, the system would reach a steady state, no net exchange between the overlying water and the pore water. The asymptotic dimensionless solute concentration in both the overlying water and pore water can be determined according to \((\theta \cdot C_0A + C_1V_s)/(V_s + V_p)/C_0\) (\(V_p\) is the total volume of the pore water in the streambed). The final concentration is 0.38, which, as already noted above, is reached more slowly with DE.

### 3.2 Concentration variation of solute in the streambed

Pore water samples were measured at arrays N1 and N2 (Figure 1c), and simulating results are compared with the measured in Figure 3. The fit of the simulation with DE is much better than without DE at both array N1 and N2 in Figures 3 and S2. The releases in the two cases show the same trend but with different values in both measurement arrays at each given time. Obviously, there is a sharp change in concentration around a narrow depth range (e.g., 1–4 cm in Figure 3a), the location of which is always deeper without DE. The concentration with DE is higher in the transition zone since, as seen above, the flux of solute from the bedform is reduced. However, above this zone, the concentration with DE is slightly lower because the concentration is also lower in the overlying water (Figure 2). The concentrations (Figures 3 and S2) in the pore water are slightly decreasing with depth.
above the transition zone, because the concentration in the overlying water, which exchanges with the pore water, is increasing with time. This trend is obvious especially in the first 143 min when the hyporheic exchange has not reached a stable trend.

At the beginning of the experiment, the solute released quickly and mainly in the upper and middle layers of the streambed (Figures 4 and S3). In these areas, the hyporheic flow is higher than that on two lateral sides of a bedform and lower streambed (Cardenas & Wilson, 2007; Jin et al., 2010). The solute concentration was high in the streamed at the two lateral sides of the bedform and the lower streamed. The depths of the solute release with and without DE were almost the same in the first 180 min. However, the concentration distributions were much different after 1440 min and the released section without DE is deeper than with DE. Density gradients induced by the higher concentration at the two lateral sides of the bedform drive downwards flow, and change the flow field in the streamed, that is, change the vertical to the horizontal flow. Therefore, the thickness of the ambient flow section is larger with DE than without DE at each specific time. The DE plays an important role in the solute release and prevents the solute releasing from streamed.

**4 | DISCUSSION**

As mentioned above, the initial concentration and the pressure between the two lateral sides of the streamed have an effect on solute release from streamed. Thus, we are interested in the effects of the density and the pressure on the solute release from streamed, which we explore using numerical simulations. The solute concentration of the overlying water in these simulations for extension is 0 in Section 4.

**4.1 | Effects of pressure difference (slope) and initial concentration on circulating flows**

The pressure difference between two vertical boundaries $P$ (Pa) that is induced by the slope can be expressed as $P = \rho g L J$, where $L$ (m) is the length of a bedform, and $J$ is the slope. Thus, $J = P/(\rho g L)$ can be considered as a dimensionless coefficient of $P$. The density gradients induced variable concentration $C$ is $\gamma C$ and its normalisation by pure water density $\gamma_c C/\rho_f$. So, the normalised concentration is $C^* = \gamma C/\rho_f$. Thus, initial concentration $C_0$ could be nondimensionalised into $C_0^* = \gamma C_0/\rho_f$, which is also a dimensionless coefficient.

The pressure gradients between the vertical sides of the bedforms (nondimensionalised into slope) and initial concentration have a great impact on solute transport (Figures 5 and S4). Circulating flows form cells within the bedform. They are induced by density gradients, and may occur if the slope is small (Figure 5). The cells are in the deeper part of the streamed and below the hyporheic flow section that are induced by the interaction between surface flow and bedforms. The circulating flow cells are distinct from the hyporheic flows, even though the two coexist. The existence of circulating flows may mix the hyporheic and ambient flows, which can strongly enhance the exchange flux between shallow and deeper parts of the bedform. When the $J$ is very small, there are two circulating flow cells in the deeper streamed of one bedform. However, there is only one circulation with larger $J$ (e.g. $J = 1.3 \times 10^{-5}$), and then, with very large $J$, the circulation disappears and the lower section of the streamed is

![Comparison of the effects of circulating flow or no circulating flow on the transport of another solute X](image)
occupied by ambient flow (Figure S4a). With larger \( C_0^* \), the circulating flows in the ambient zone become more obvious (Figure S4b).

The circulating flow and no circulating flow for all simulation cases with different \( J \) and \( C_0^* \) are shown in Figure 6. A new non-dimensionalised coefficient \( J_c^* = J/C_0^* \) combining the pressure gradients \( J \) and initial concentration \( C_0^* \) is defined to show the critical condition for the onset of circulating flow. The cases for with or without circulating flows are distinguished by a grey belt (0.0541 < \( J_c^* \) < 0.0690). At the left side of the belt in Figure 6, the points data represent the cases there are circulating flows, vice versa. The belt is transition of cases with or without circulating flows. However, what is the function of the circulating flow? This will be analysed in Section 4.2.

### 4.2 Effects of circulating flows on solute release time

To illustrate the effect of density (initial concentration) and pressure difference (slope) on solute release, especially circulating flow induced by the density gradient, we focused on analysing the relationship between the 5\% residual time \( T_{5\%} \) and slope with different initial concentrations (Figure 7a). \( T_5 \) defines the time when the solute mass in the bedform is 5\% of the total solute mass initially in the bedform. Firstly, the greater the slope the greater is \( T_5 \) for the same initial concentration. At lower slopes (from 0 to \( 2 \times 10^{-4} \)), the \( T_5 \) values vary little, and at the higher slopes (> \( 1.5 \times 10^{-3} \)), they increase slightly increased. However, at slopes between \( 2 \times 10^{-4} \) and \( 1.5 \times 10^{-3} \), \( T_5 \) shows a dramatic increasing trend with slope. Second, \( T_5 \) for different initial concentrations reduces with increasing slope, which means that at a higher slope, the initial concentration has only a slight influence on the transport of solute from the bedform. Third, the greater the initial concentration is, the greater the time to reach \( T_5 \) for the same slope (Figure 7a).

\( T_5 \) can be normalised as \( T^* = T_5/T_0 \), where \( T^* \) is the normalised time, and \( T_0 \) is \( T_5 \) for when the slope is 0. \( T^* \) does not change with \( J_c^* \) when \( J_c^* < 0.0541 \) (left side of the transition, grey belt, Figure 7b) because the circulating flows mix well the hyporheic and ambient zones, which keeps \( T_5 \) constant. \( T^* \) increases with \( J_c^* \) when \( J_c^* > 0.0690 \) (right side of the transition, grey belt). However, when \( 0.0541 < J_c^* < 0.0690 \), that is, in the transition (grey belt), \( T^* \) slightly increases with \( J_c^* \) since there are cases with or without circulating flows (Figures 6 and 7b).

### 4.3 Effects of circulating flows on solute mixing and retention in the streambed

To study the mixing induced by a circulating flow, a conservative tracer (solute X, nonreactive and no density effect) was put in the bedform for 900 min (Figure 8). The background flow field in the ‘No circulating flow’ case is induced by interaction between the current and bedform \( (C_0^* = 0 \text{ and } J = 0) \), while the flow field in the ‘circulating flow’ case is induced by the previous salt solution \( (C_0^* = 1.1 \times 10^{-3} \text{ and } J = 0) \) and the interaction between bedforms and surface flow.

In order to evaluate the degree of mixing, an entropy mixing index \( (M, \text{ whose value increases with increased mixing}) \) whose value is between 0 and 1 is chosen (Guida, Nienow, & Barigou, 2010), where:

\[
M = -\frac{1}{\log n_c} \sum_{i=1}^{n_c} p_i \log p_i
\]

\[
p_i = \frac{C_{xi}}{\sum_{i=1}^{n_c} C_{xi}}
\]

where \( n_c \) is total number of grid cells, \( p_i \) is the proportion of the mass in the \( i \)th cell and \( C_{xi} \) (kg m\(^{-3}\)) is the concentration of solute X in the \( i \)th cell. In this simulation \( n_c = 179535 \). The entropy mixing index variation of solute X at different times is presented in Table 2.

In order to evaluate the degree of mass retaining in the streambed, residual mass ratio \( (R) \) whose value is between 0 and 1 is chosen. The \( R \) represents the mass proportion retaining in the bedform, and \( R \) can be defined as:

\[
R = \frac{\int_{\Omega} C_X dV}{\int_{\Omega} C_{X0} dV}
\]

where \( C_X \) (kg m\(^{-3}\)) is the concentration of solute X at a certain time, \( C_{X0} \) (kg m\(^{-3}\)) is the concentration of solute X at initial time, \( \Omega \) (m\(^3\)) is the total volume of bedform, and \( dV \) (m\(^3\)) is differential volume. The residual mass ratio variation of solute X at different times is presented in Table 2.

For the ‘No circulating flow’ case, after 4400 min, some of the solute X leaves the bedform along the pore streamline, while most of the solute X is released from the streambed after 8500 min, only 13.6\% of mass left. For the ‘Circulating flow’ case most of the solute

| Time       | 900 min | 4400 min | 8500 min |
|------------|---------|----------|----------|
| Entropy mixing index | No circulating flow | 0.491 | 0.530 | 0.489 |
|             | Circulating flow | 0.491 | 0.550 | 0.572 |
| Residual mass ratio | No circulating flow | 1.000 | 0.512 | 0.136 |
|             | Circulating flow | 1.000 | 0.999 | 0.974 |
X is retained in the streambed, and even at 8500 min 97.4% of solute X is retained and is mixed strongly by two circulating flows near the bottom of bedform (Table 2 and Figure 8).

For the ‘No circulating flow’ case, the entropy mixing index (M) increases from 900 to 4400 min and then drops back from 4400 to 8500 min. This occurs because solute X is partly retained in the pore water at 4400 min and most of the solute X leaves the bedform by 8500 min. For the ‘Circulating flow’ case, the entropy mixing index (M) increases from 900 to 8500 min in all cases because solute X stays almost at the bottom of bedform (where the circulating flows are strong) and the degree of mixing increases with time. Moreover, M is always slightly higher in the ‘Circulating flow’ than the ‘No circulating flow’ case at 4400 and 8500 min (Table 2 and Figure 8).

Overall, although the circulating flows enhances the degree of mixing slightly, a more important function of circulating flows is that they retain solute X and postpone the release from the streambed.

5 | CONCLUDING REMARKS

We conducted experiments and numerical simulations to examine solute release from the hyporheic zone of a streambed. The combined experimental/numerical approach shows that:

1. The pressure gradients between two lateral sides of the bedform and the initial concentration influence the process of solute release. Due to gravity, the release is reduced with increasing density of the pore water.
2. DE can induce the circulating flow in the ambient zone. The horizontal pressure gradient between two lateral sides and the initial concentration determines whether there would be circulating flows. Such flows enhance mixing between the ambient and hyporheic zones.
3. Circulating flows can affect the releasing and mass transport processes in streambed: keeping the residual time (T_r) the same, enhancing the mixing of the hyporheic zone and the ambient zone, and retained other solute in the deeper area of streambed.

The present study focused on the behaviour of solutes released from a polluted streambed. Circulating flows induced by DE could be controlled by boundary conditions. The possibility of so-called secondary pollution remains an issue even after long periods. This study may offer a new perspective, and a quantitative guide on control and treatment of secondary pollution.

ACKNOWLEDGMENTS

his research has been supported by the Natural Science Foundation of China (51421006, 51679065), the Basic Research Programs (Natural Science Foundation) of Jiangsu Province (BK20171436), Social Development - Major Demonstration of Science and Technology of Science and Technology Projects of Jiangsu Province (BE2018737), and the 111 Project (B17015), Ministry of Education and State Administration of Foreign Experts Affairs, P. R. China.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of this article.

How to cite this article: Jin G, Yang W, Xu H, et al. Density effects on solute release from streambeds. *Hydrological Processes*. 2020;34:1144–1153. https://doi.org/10.1002/hyp.13655