



Relative roles of resuspended particles and pore water in release of contaminants from sediment

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Abstract: Sediment layers containing contaminants play a significant role in environmental hydrodynamics. Experiments were conducted in order to characterize the relative roles of resuspended particles and pore water under different flow and sediment conditions. A conservative tracer (NaCl) and a reactive tracer (phosphate) were used as contaminants in the bottom sediment in a laboratory flume. The mixing between the overlying water and pore water occurred over a short time while the desorption of contaminants from fine-grained resuspended particles lasted a relatively long time. The effects of resuspended particles and pore water on the variations of release flux and concentration of contaminants in water with time under different hydrodynamic conditions were quantified. The results show that pore water dominated the initial release flux, which could be several orders of magnitude greater than the flux due to molecular diffusion. Flux contribution of desorption from sediment particles in the latter release could be equal to what was seen from pore water in the initial stage.

Key words: sediment resuspension; resuspended particle; pore water; release of contaminants; release flux

1 Introduction

Sediment-water interactions in lakes and rivers have become very important since bottom sediments are large repositories of contaminants (Corbett 2010). When sediment resuspension occurs, the contaminants may not be permanently stored in the bottom sediments and may repeatedly be recycled (Chung et al. 2009). Resuspended particles may be an additional source of solutes if they react in the water column (Fig. 1) (Tengberg et al. 2003). If these particles stay in flowing water, they can be oxidized, releasing dissolved contaminants into the water column (Kalnejais et al. 2007, 2010). The impact of resuspension on water quality depends on both hydrodynamic conditions and sediment features (Li et al. 2002). The sediment's cohesive

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strength depends on multiple factors, including grain size, mineralogical properties, and the activity of benthic organisms, which determine the ability of sediment to resist the shear stress imposed by the overlying water (Dey and Papanicolaou 2008). Contaminants exist in both dissolved and solid phases. The conservative solutes have nothing to do with sediment particles. For reactive solutes, the degree of particle association has to be taken into consideration as chemical reactions can contribute to medium-term or long-term immobilization (Li et al. 2004).

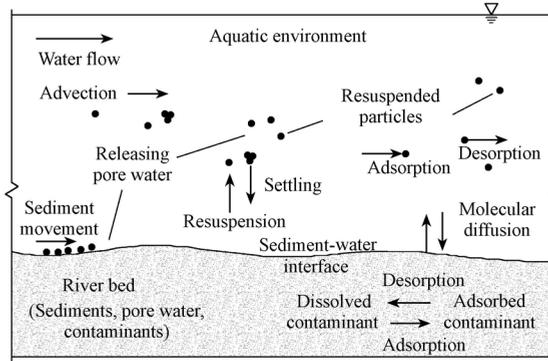


Fig. 1 Mechanisms of interaction between particles and contaminants in conceptual model

The purpose of this study was to investigate the characteristics of conservative and reactive contaminants released from cohesive and non-cohesive sediments over a range of bottom shear stress values. Laboratory experiments were conducted to predict the release flux. This combination of contaminants, sediment properties, and hydrodynamics will enable a more complete understanding of roles of resuspended particles and pore water in the release of pollutants from sediment.

2 Materials and methods

2.1 Experimental apparatus

A circulating flume (Li et al. 2008) consisting of a rectangular test section with 6 m in length, 0.2 m in width, and 0.45 m in depth was used to investigate the response of bottom sediments over a range of flow velocities (Fig. 2). Surface water flow was controlled by a variable frequency pump and a tail valve. The flow velocity varied from 0.1 m/s to 0.3 m/s. The maximum design water depth was 0.4 m.

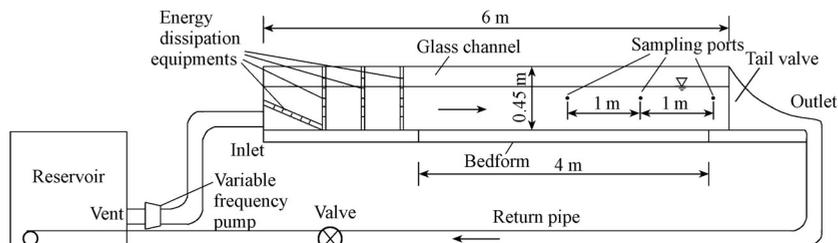


Fig. 2 Schematic diagram of experimental apparatus

A micro-propeller velocimeter was used to measure the flow velocity. A layer of sediment containing tracers of about 5 cm in thickness was gently and uniformly laid on the bottom of the flume. Three horizontal arrays of sampling ports with 1 m intervals were installed through the sides of the flume for parallel sampling.

2.2 Field sites and sampling

The sediment used in the experiment was collected from Dianshan Lake, China (Lin et al. 2010). Natural sediments with $d_{50} < 0.05$ mm and $d_{50} = 0.35$ mm (d_{50} is the sediment median size) were respectively used as fine-grained and coarse-grained particles, and also as cohesive silt and non-cohesive sand. Prior to the experiment, the sediment was prepared following a three-step procedure: (1) the sediment was washed three times with hydrogen peroxide and distilled water to remove impurities, (2) the sediment was washed in a concentrated hydrochloric acid for approximately 12 hours to remove adsorbed contaminants on particles, and (3) the sediment was washed again with distilled water. After being filtered and dried, the sediment was then stored for the laboratory experiment. A DDS-11A conductivity meter was used to measure the concentration of NaCl. A 722N visible spectrophotometer was used to measure phosphorus concentrations following the method proposed by Zhu et al. (2011).

2.3 Experimental methods

In this study, phosphate was used as a reactive tracer (Su et al. 2011). The fate of phosphate in aquatic environments is highly dependent on the sorptive behavior of sediment. The equilibrium adsorption capacity of sediment should be measured experimentally. The adsorption capacity C is defined as the mass of phosphorus adsorbed per unit mass of sediment, with a unit of mg/g. The adsorption capacity depends on the surface area and quality of sediment. The adsorption and desorption experiments were first conducted in glass vessels. A monopotassium phosphate solution was first added to the sediment. Samples were taken at different time intervals. An agitator was used during the experiments to make sure that the sediment stayed in suspension. The dimensionless concentration C_w/C_0 was used in this study, where C_0 is the initial concentration of the contaminant in pore water and C_w is the concentration of the contaminant in the overlying water.

3 Results and discussion

3.1 Adsorption and desorption experiments

Migration of phosphorus in the sediment depends on the interactions between sorption characteristics of sediment particles and pore water. The results of the sorption experiments are shown in Fig. 3 and Fig. 4.

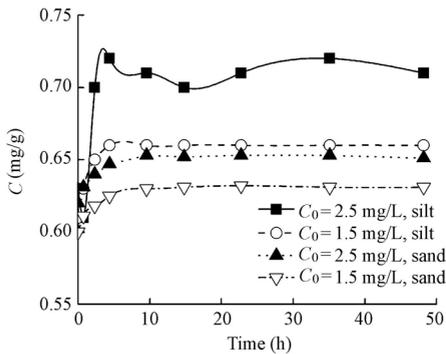


Fig. 3 Dynamic adsorption experiments with respect to different initial concentrations of phosphate C_0

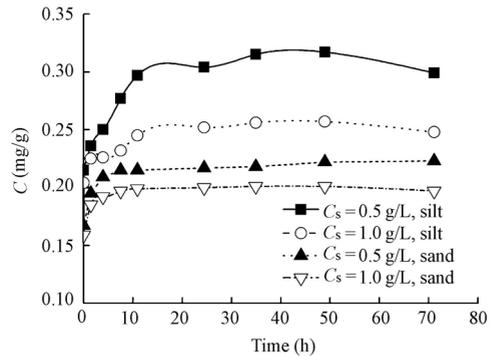


Fig. 4 Dynamic desorption experiments with respect to different sediment concentrations C_s

From the results shown in Fig. 3, it can be seen that the initial concentration of phosphate had a large effect on the adsorption process. The adsorption capacity of sediment increased with the initial concentration of phosphate. Generally, both sand and silt would take two to four hours to reach the equilibrium condition, and the adsorption capacity still slightly increased after a period of time. Interestingly, because silt was made of smaller particles, which had larger specific surface areas, it reached equilibrium more slowly than sand, and had a relatively high phosphate content. This may be explained by the fact that adsorption can be divided into two steps: the first step is rapid surface adsorption (over minutes to hours); and the second step is slow reaction (over days to months), which is a process of adsorbed phosphate in the solute phase diffusing to the inner grains (Wang et al. 2007). In the dynamic desorption experiments, described in Fig. 4, the time to reach the equilibrium state was approximately one more hour at the sediment concentration of 0.5 g/L than the time at 1 g/L, which indicated that, with the same grain size, a higher sediment concentration corresponded to a faster arrival at the equilibrium state. Meanwhile, with the same sediment concentration, a smaller particle size corresponded to more phosphate being desorbed from the sediment, as it did with adsorption.

3.2 Molecular diffusive flux and resuspension flux

The flux across the sediment-water interface has traditionally been assumed to be dominated by molecular diffusion when the sediment is at a steady state or in steady flow (Steinberger and Hondzo 1999). Diffusion experiments were conducted using sand and silt with different NaCl concentrations. The variation of solute density due to the increase of the NaCl concentration was not considered in this study.

Fig. 5 shows that the concentrations of NaCl in the overlying water increased with time. As the molecular diffusion was very slow, only 30% of the initial quantity was released into the water in 24 hours. A comparison of the solute proportions in sand and silt showed that although the general trend was almost the same, solutes were released faster from silt than from sand. Silt may have low porosity, resulting in a higher concentration gradient across the

sediment-water interface. This indicates that the physicochemical characteristics of sediment may also play an important role in the contaminant release process.

Based on the results shown in Fig. 6, the fluxes due to bulk resuspension could be several orders of magnitude greater than those due to molecular diffusion. Almost all the conservation solute (NaCl) in pore water entered the overlying water when sediment resuspension occurred. As phosphorus can be stored in the aqueous phase (dissolved total phosphorus, DTP) and solid phase, there was an equilibrium sorption process, and only 12% of the phosphorus, with the resuspension of pore water, contributed to the release flux to the overlying water. The resuspension was a significant term in release of solutes from sediment. However, some strong reactive solutes might not have been affected because most of them were stored in solid phase in sediments.

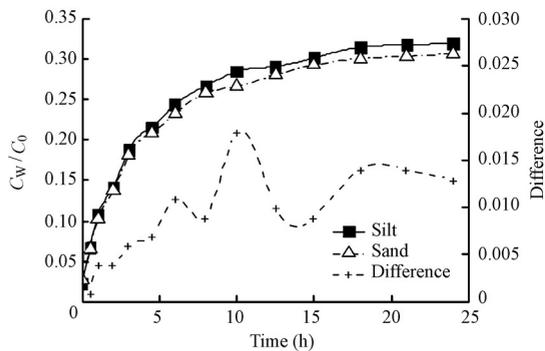


Fig. 5 Non-dimensional concentration of NaCl at steady state in sand and silt and their difference

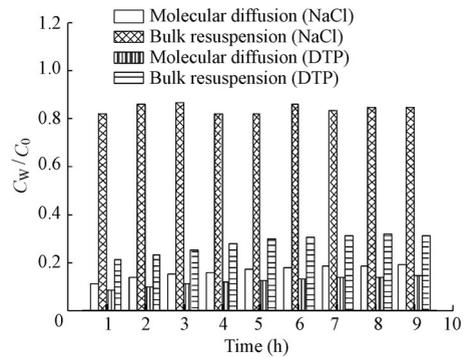


Fig. 6 Non-dimensional concentrations of NaCl and DTP at steady and suspended states

3.3 Contributions from suspended particles and pore water

A series of experiments were conducted under typical flow conditions using fine-grained sediment. When the bottom shear stress increased, more particles and pore water would enter the overlying water. As can be seen from Fig. 7, the concentration of contaminants depended on the shear stress. The release of NaCl was faster than that of phosphate with the increase of shear stress. The NaCl concentration in the water body increased immediately when shear stress exceeded the critical shear stress. The contaminant release flux might have been due to the contaminants in the liquid phase stored in the surficial sediment. Pore water might have dominated the release at the initial stage of resuspension. The DTP concentration also increased at the early stage, but its ratio to its initial value was less than that of the NaCl concentration.

Phosphates existed both in solid and liquid phases in the bottom sediment. Only those stored in pore water were released into the overlying water at first, while others were still adsorbed in resuspended particles. Time is needed for the adsorption-desorption process to reach an equilibrium condition. As shown in Fig. 6, when bulk resuspension occurred, the NaCl concentration did not increase much after one hour while the DTP concentration was as 1.5 times that of ten hours before.

3.4 Long-term release from suspended particles

The concentration distribution of suspended sediment shows two different mechanisms. Coarse-grained sediment may resuspend many times, if the properties of the bottom sediment do not change with time. For cohesive, fine-grained sediment, the distribution of sediment particle sizes with depth and the time-changed coherency must be considered (Rubin and Akinson 2001).

A typical flow condition was applied to the fine-grained sediment. Fig. 8 shows the released amount of total phosphorus (TP) and sediment concentration under a flow rate of 10 cm/s at water depths of $h = 0.1$ m and $h = 0.2$ m. Sediment concentration and TP concentration slowly approached a stable value within about 12 hours. The TP concentration rapidly declined while sediment concentration declined in the overlying water. After about 6 to 8 hours, the release amount gradually stabilized, which means that all release processes reached stable states. It can also be seen that under a specific flow rate, the depth of the overlying water also influence the release of pollutants. The resuspension release flux of pollutants decreased with increasing water depth. This might be due to the fact that deeper water resulted in a smaller bottom shear stress, which caused a decrement in the amount of suspended particles.

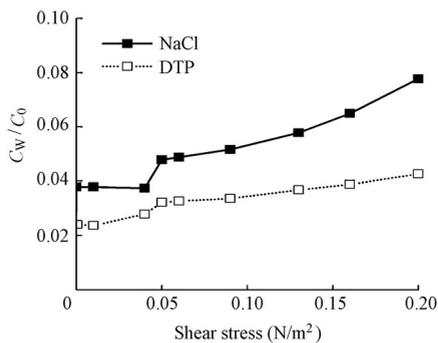


Fig. 7 Variations of non-dimensional concentrations of NaCl and DTP with shear stress

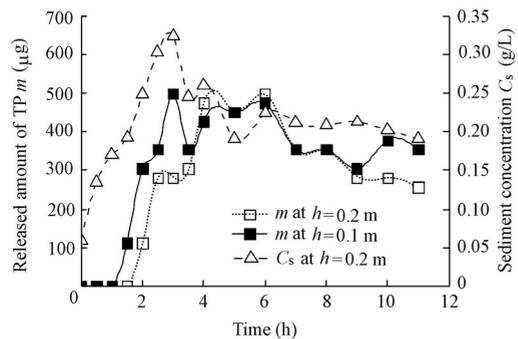


Fig. 8 Released amount of TP and sediment concentration at certain flow conditions

4 Conclusions

Through experimental studies and theoretic analyses, the mechanisms of release of contaminants from bottom sediment were examined deeply. The adsorption capacities of silt were larger than those of sand because silt has a larger specific surface area. When the sediment was static, only molecular diffusion on the sediment-water interface was considered. When the sediment resuspension occurred, all the conservative contaminants were released from pore water resuspended with sediment. When reactive tracer was contained alone, pore water played a dominant role at the early stage of pollutant release, and then the adsorbed tracer was desorbed from resuspended particles, controlling successive pollutant release. The mixing of overlying water and pore water occurred in a short time while the adsorption and desorption of contaminants from resuspended particles occurred over a relatively long period.

The release flux of contaminants depended on hydrodynamic conditions, including flow velocity and water depth. Variations in velocity and depth, which lead to an increase in bottom shear stress, will result in an increase in a large quantity of contaminants.

References

- Chung, E. G., Bombardelli, F. A., and Schladow, S. G. 2009. Modeling linkages between sediment re-suspension and water quality in a shallow, eutrophic, wind-exposed lake. *Ecological Modelling*, 220(9-10), 1251-1265. [doi:10.1016/j.ecolmodel.2009.01.038]
- Corbett, D. R. 2010. Re-suspension and estuarine nutrient cycling: Insights from the Neuse River Estuary. *Biogeosciences*, (7), 3289-3300. [doi:10.5194/bg-7-3289-2010]
- Dey, S., and Papanicolaou, A. 2008. Sediment threshold under stream flow: A state-of-the-art review. *KSCE Journal of Civil Engineering*, 12(1), 45-60. [doi:10.1007/s12205-008-8045-3]
- Fang, H. W., Chen, M. H., and Chen, Z. H. 2009. *Surface Characteristics and Model of the Environment Sediment*. Beijing: Science Press. (in Chinese)
- Kalnejais, L. H., Martin, W. R., Signall, R. P., and Bothner, M. H. 2007. Role of sediment resuspension in the remobilization of particulate-phase metals from coastal sediments. *Environmental Science and Technology*, 41(7), 2282-2288. [doi:10.1021/es061770z]
- Kalnejais, L. H., Martin, W. R., and Bothner, M. H. 2010. The release of dissolved nutrients and metals from coastal sediments due to resuspension. *Marine Chemistry*, 121(1-4), 224-235. [doi:10.1016/j.marchem.2010.05.002]
- Li, B., Zhang, K., Zhong, B. C., and Wang, D. Z. 2008. An experimental study on release of pollutants from sediment under hydrodynamic conditions. *Chinese Journal of Hydrodynamics*, Ser. A, 23(2), 126-133. (in Chinese)
- Li, J. C., Chu, J. D., and Feng, H. L. 2002. Experimental investigation of impact way of scouring and suspending of river bottom sediment on water quality. *Resources and Environment in the Yangtze Basin*, 11(2), 137-140. (in Chinese) [doi:10.3969/j.issn.1004-8227.2002.02.008]
- Li, Y. P., Jiang, Y., Lü, J., Zhang, G., Ding, L., Peng, J. P., Wang, C., and Fan, L. L. 2004. On the relation between the release rate of TN, TP from sediment and water velocity. *Journal of Lake Sciences*, 16(4), 318-324. (in Chinese) [doi:10.3321/j.issn:1003-5427.2004.04.005]
- Lin, W. Q., Lu, S. Q., and Chen, Y. Z. 2010. An application of eco-dynamic model in evaluating eutrophication control measures for Dianshan Lake in Shanghai. *Shanghai Environmental Science*, 29(1), 1-10. (in Chinese)
- Rubin, H., and Atkinson, J. 2001. *Environmental Fluid Mechanics*, 652-695. Abingdon: Marcel Dekker.
- Steinberger, N., and Hondzo, M. 1999. Diffusional mass transfer at sediment-water interface. *Journal of Environmental Engineering*, 125(2), 192-200. [doi:10.1061/(ASCE)0733-9372(1999)125:2(192)]
- Su, N., Du, J. Z., Ji, T., and Zhang, J. 2011. ²²⁶Ra and ²²⁸Ra tracer study on nutrient transport in east coastal waters of Hainan Island, China: A case of nutrients. *Water Science and Engineering*, 4(2), 157-169. [doi:10.3882/j.issn.1674-2370.2011.02.004]
- Tengberg, A., Almroth, E., and Hall, P. 2003. Resuspension and its effects on organic carbon recycling and nutrient exchange in coastal sediments: In situ measurements using new experimental technology. *Journal of Experimental Marine Biology and Ecology*, 285-286, 119-142. [doi:10.1016/S0022-0981(02)00523-3]
- Wang, X. Q., Li, Z., Lü, P. Y., and Guo, J. S. 2007. Adsorption and desorption of phosphorus on suspended particles in Three Gorges area. *Resources and Environment in the Yangtze Basin*, 16(1), 31-36. (in Chinese) [doi:10.3969/j.issn.1004-8227.2007.01.007]
- Zhu, H. W., Zhang, K., Zhong, B. C., and Wang, D. Z. 2011. Effects of particles and pore water in release of pollutants due to sediment resuspension. *Chinese Journal of Hydrodynamics*, Ser. A, 26(5), 631-641. (in Chinese) [doi:10.3969/j.issn1000-4874.2011.05.015]

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