Colloids and Colloid-Facilitated Transport of Contaminants in Soils: An Introduction

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ABSTRACT

Until some two decades ago, it was believed that only the soil liquid and gaseous phases were mobile and could facilitate the transport of chemicals and nutrients through the vadose zone. It is now generally accepted that also part of the soil solid phase is mobile, and that mobile organic and inorganic soil colloids may facilitate chemical transport. However, the magnitude and significance of these colloidal transport processes are yet to be determined. It is essential to examine whether current models for transport and fate of chemicals in soil and groundwater need to be revised. The collection of papers in this special section of *Vadose Zone Journal* mainly take their origin, but not exclusively, from an international workshop "Colloids and Colloid-Facilitated Transport of Contaminants in Soil and Sediments" held at the Danish Institute of Agricultural Sciences, Denmark, 19–20 Sept. 2002. The workshop was organized to review our present knowledge of colloid behavior and transport in porous media and the possibility of colloid-bound transport of contaminants and nutrients in soil and groundwater. Here we will first give a brief introduction to the topic of mobilization and transport of colloids in the vadose zone, and highlight previous evidence of colloid-facilitated transport. We then introduce the review and technical papers in the special section. We hope that the information provided in this special section will lead to improvements in our understanding and associated conceptual models of contaminant transport and fate in soil.

THE SOMETIMES RAPID APPEARANCE of environmental contaminants in groundwater reflects limitations in our current understanding of transport processes in soils. Traditional approaches to describe and predict the movement of nonvolatile contaminants treat soil and groundwater as a two-phase system in which contaminants partition between immobile solid constituents and the mobile water phase. Contaminants that are sparingly soluble in water and have a strong tendency to bind to the solid phase are assumed to be retarded in the soil. Many contaminants readily adsorb to the soil solid phase and are therefore considered to present little danger in risk assessments of surface water or groundwater pollution. However, it has become evident that predictions and results obtained in laboratory batch systems may not always be applicable to field situations. Spatial variability or preferential flow phenomena have been hypothesized to be responsible for some of the inconsistencies between predictions and actual leaching of contaminants (Jury and Flühler, 1992). In addition, colloids in the solid phase may be mobile and can therefore act as a third phase, thus enhancing the transport of strongly sorbing contaminants (McCarthy and Zachara, 1989).

SOURCES OF MOBILE SOIL COLLOIDS

The tendency of soil colloids to disperse from soil aggregates in response to infiltration of water is a natural phenomenon, sometimes even leading to the development of illuvial subsurface horizons with higher contents of clay compared with the upper eluvial horizons. Micromorphological features showing deposits of clay skins on ped faces and at the interface of water-conducting pores represent evidence of such colloid translocation (Buol and Hole, 1961). Dispersion of colloids is also suspected to be responsible for affecting soil physical properties such as surface crusting, surface erosion, water infiltration, and hydraulic conductivities (e.g., Miller and Baharuddin, 1986; Shainberg et al., 1992). The source of mobile colloids in the vadose zone is generally considered to be the in situ release of water-dispersible colloids. Colloids are operationally defined as particles between 1 to 10 nm and 2 to 10 μm in diameter (e.g., Stumm, 1992; Buffle and Leppard, 1995), and include layer silicates, sesquioxides (Fe- and Al-oxyhydroxides), organic macromolecules, bacteria, and viruses. Because of their high specific surface area, colloids have a high sorptive capacity and can be effective sorbents of low solubility, strongly sorbing contaminants. Transport of colloids through the vadose zone thus causes an increased risk of leaching of contaminants generally regarded as relatively immobile.

EVIDENCE OF COLLOID-FACILITATED TRANSPORT

Several experimental investigations have suggested that colloids may influence the transport of contaminants in the vadose zone. Laboratory column studies have demonstrated co-transport of contaminants sorbed to suspended colloids, or simultaneous leaching of in situ colloids and contaminants, while field studies have revealed the association of contaminants with colloids in drain or groundwater. Using homogeneously packed soil columns, Vinten et al. (1983) obtained evidence of vertical transport of DDT adsorbed to suspended montmorillonite colloids. Grolimund et al. (1996) demonstrated that suspended in situ mobilized colloids can provide a pathway for rapid transport of Pb. In another study, Flury et al. (2002) found that colloids mobilized in flow experiments with packed sediments carried Cs along.

Experiments with undisturbed soil columns have shown that suspended colloid–contaminant complexes may facilitate the transport of atrazine (e.g., Seta and Karathanasis, 1997b), and Cu and Zn (Karathanasis, 1999). In addition, it was demonstrated that in situ mobilized colloids were able to facilitate the transport of

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prochloraz (de Jonge et al., 1998) and glyphosate (de Jonge et al., 2000) in undisturbed macroporous soil columns. Jørgensen and Frederecia (1992) found indications of the transport of DDT adsorbed to clay minerals in a fractured clayey till. In roadside soils receiving deicing salts, Amrhein et al. (1993) found effluent concentrations of Cu, Pb, Ni, and Cr in the colloidal size fraction. Grant et al. (1996) observed significant amounts of particulate P in drainage from arable catchments, while Laubel et al. (1999) found significant transport of particulate matter and particulate P in both field plot experiments and tile-drained catchment studies. Sprague et al. (2000) similarly found that a measurable amount of total atrazine transport in a field lysimeter occurred in association with mobile soil colloids. In other studies Villholth et al. (2000) noticed evidence of colloid-facilitated transport of prochloraz to subsurface drains, while Uusitalo et al. (2001) observed that most of the P leached as drain flow occurred as particulate P. Petersen et al. (2003) observed a positive correlation between drainage water concentration of strongly sorbing pendimethalin and particle turbidity. Hence, there is ample evidence that colloids can facilitate the movement of strongly sorbing contaminants in soils. Despite this evidence, current models are largely unable to predict in a quantitative manner the importance of colloid-facilitated transport of contaminants or assess the risk of colloid-facilitated contaminant transport under field conditions. There is a clear need for understanding the processes controlling in situ mobilization and transport of colloids in the vadose zone.

SOIL PROPERTIES INFLUENCING IN SITU COLLOID MOBILIZATION AND TRANSPORT

A comprehensive amount of literature exists on the topic of colloid mobilization and transport in model systems of packed sand (e.g., Tan et al., 1992; Ryan and Gschwend, 1994; Roy and Dzombak, 1996) or packed soil columns (e.g., Grolimund et al., 1998; Kretzschmar and Sticher, 1997; Grolimund and Borkovec, 1999; Noack et al., 2000; Flury et al., 2002). Several reviews also addressed this topic (McDowell-Boyer et al., 1986; McCarthy and Zachara, 1989; Swanton, 1995; Ryan and Elimelech, 1996; Kretzschmar et al., 1999). These studies have demonstrated the importance of physical and chemical perturbations on the mobilization and transport of colloids. In natural, structured soils, however, the processes of colloid mobilization and transport may be further complicated by the profound effects of pore structure on the active flow pathways of water, thus affecting both in situ colloid mobilization and subsequent transport. To date few studies have addressed the complex issue of colloid mobilization and transport in natural structured soils (Jacobsen et al., 1997; Seta and Karathanasis, 1997a, 1997b; de Jonge et al., 1998, 2000; Ryan et al., 1998; Karathanasis, 1999; Laegdsmand et al., 1999; El-Farhan et al., 2000; Villholth et al., 2000; Schelde et al., 2002; Petersen et al., 2003).

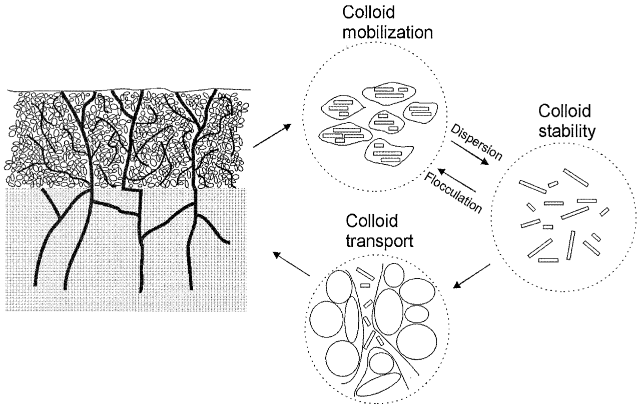

Fig. 1. Conceptual illustration of the main processes to be considered when investigating in situ colloid mobilization and transport.

In situ mobilization and transport of colloids in natural, structured soils depends on complex interactions among soil characteristics controlling the inherent dispersibility of colloids, and on dynamic properties determining the prevailing conditions for colloid mobilization and transport. Three main issues should be considered when evaluating the overall process of in situ colloid mobilization and transport (Fig. 1). The first issue to be considered involves dispersion or release of in situ colloids. Once the colloids have been mobilized, they can be transported through the vadose zone, with transport being determined by both the size and stability of the dispersed colloids in the soil solution, and by the pore size and geometry of the actively conducting pore system.

A variety of soil properties influence the dispersibility of colloids from aggregates. Besides the well-documented effects of clay mineralogy (e.g., Seta and Karathanasis, 1996) and solution ionic strength and pH (e.g., Grolimund and Borkovec, 1999; Flury et al., 2002) on colloid dispersion, total clay content (e.g., Brubaker et al., 1992), soil moisture conditions (e.g., Pojasok and Kay, 1990), soil management (e.g., Watts et al., 1996a, 1996b), and interactions among these properties have also been shown to affect colloid dispersibility. In addition, the amount of soil volume conducting water vs. soil volume bypassed by water flow, defining the degree of preferential flow, may be an important property influencing both in situ colloid mobilization and subsequent translocation. Preferential flow may accelerate the transport of externally applied colloid–contaminant complexes, or colloid–contaminant complexes located close to the preferential pathways, but decrease the in situ mobilization and leaching of colloids located at some distance from the preferential flow paths.

The above discussion provides a brief and, we acknowledge, in many respects incomplete introduction to the exciting and rapidly emerging area of colloid and colloid-facilitated transport in porous media. We have pointed out that a large number of soil physical and chemical factors act in combination with colloid properties and soil structural heterogeneity to govern the occurrence and magnitude of colloid- and colloid-facilitated transport in the vadose zone. At present, we have reasonably good understanding of many of the individual soil physical and chemical factors and their influence on colloid mobilization and transport in water-saturated, typically homogeneous, porous systems in the laboratory. Especially for unsaturated porous media, however, the effects of both individual and simultaneous changes in soil physical and chemical parameters and the effects of water–air and water–air–solid interfaces on colloid and colloid-facilitated transport in unsaturated porous media are still poorly examined and understood. We also need to improve our understanding of colloid release and transport processes at the pore scale, and to upscale these processes to obtain more mechanistically based descriptions of the transport of colloids and colloid-bound chemicals in the vadose zone. To be able to use our knowledge for predicting the processes in natural vadose zone systems, it is essential to extend our investigations to more physically and chemically heterogeneous media systems at both the laboratory and field scale. In perspective, risk assessment models that include transport and fate of chemicals in soil will eventually need to include the quantitatively significant processes related to colloid and colloid-facilitated transport. This special issue on colloid and colloid-facilitated transport reviews and furthers our knowledge within this research area. We trust that the 15 papers in this issue will serve as an inspiration for numerous future studies on colloid and colloid-facilitated transport in soil.

BRIEF OVERVIEW OF SPECIAL SECTION PAPERS

This issue of *Vadose Zone Journal* opens with two reviews that examine recent advances in process-based understanding and elucidate concerns and challenges within the field of colloid transport in the vadose zone. Both reviews outline future research needed to improve our understanding of colloid transport and fate in variably saturated, porous media. McCarthy and McKay (2004) describe the evolution in our understanding of colloids, including especially their role as important and potentially mobile constituents in subsurface systems. The authors summarize some of the key challenges in predicting colloid transport in the vadose zone, including the issues of sampling, colloid surface properties, soil water solution chemistry, interfaces (air–water and air–water–solid), kinetic- and flow-controlled processes, and soil structural heterogeneity. DeNovio et al. (2004) similarly provide an overview of recent developments, experimental data, and models for colloid transport in unsaturated soil, with special emphasis on air–water interfaces and the effects of rapid fluctuations in pore water flow rates and soil solution chemistry.

The mobility of both abiotic and biotic organic colloidal matter in the vadose zone is addressed in two subsequent reviews. Based on numerical case studies, Totsche and Kögel-Knabner (2004) illustrate how mobile organic constituents affect chemical transport in soils and show that mobile organic sorbents can both enhance and reduce chemical mobility, depending upon the dominant sorption processes. In some cases, the colloid itself can pose a risk of contamination, such as in the case of bacteria and virus transport. In a review of methods for modeling coupled microbial and transport processes in porous media and soil, Rockhold et al. (2004) illustrate the many complex interactions that take place between microbial processes and flow and transport processes.

Porous media type, colloid properties, water flow velocity, and solution chemistry, among many other factors, are known to influence colloidal transport. Experimental and modeling results by Bradford et al. (2004) illustrate the influence of pore size distribution relative to colloid size on leaching and distribution of retained model colloids (latex microspheres) in repacked, physically heterogeneous sand columns. A comparison of the transport of natural, soil-extracted colloids, as compared with a model colloid (kaolinite) in two types of repacked porous media, is provided by Zhuang et al. (2004).

Additional key factors in controlling colloid mobilization and leaching in undisturbed soil systems are soil structure and stability, and the amount of potentially mobile colloids. Kjaergaard et al. (2004a) investigate the amount of water-dispersible clay in six soils sampled along a natural clay gradient. The influence of physical factors such as soil-water content, wetting rate and clay content on water-dispersible clay, measured by either a high-energy or low-energy input method, is shown. For the same six soils, the two companion papers by Kjaergaard et al. (2004b, 2004c) address the effect of pore structure and structural stability on the active flow pathways of water, and illustrate the important role of preferential flow and colloid dispersion on in situ colloid mobilization and leaching.

Further understanding of the processes governing colloid mobilization and mobility in soil, may come from detailed visualization of the processes involved, especially at the pore scale. Visualization studies of colloid transport through porous media are provided by Baumann and Werth (2004) and Christ et al. (2004). Using light transmission and epifluoresent microscopy techniques, respectively, these two papers investigate colloid transport phenomena and colloid entrapment at air–water–solid interfaces.

The final three papers in this special issue concern colloid-facilitated transport. Sorption–desorption processes are hypothesized to play an important role in colloid-facilitated transport. Laegdsmand et al. (2004) examine the effects of soil solution chemistry and indigenous organic matter on pyrene sorption onto water-dispersible colloids as extracted by three different methods. The results, used in combination with a simple pyrene leaching model, suggest that colloid-facilitated transport dominates pyrene leaching.

A study by de Jonge et al. (2004) shows that soil spatial heterogeneity can strongly affect actual colloid and colloid-facilitated transport in the vadose zone. Their study focuses on field-scale variations in the transport of colloids in conjunction with a strongly adsorbing compound (P). They found that soil structure controlled to a large extent the leaching of colloids and colloid-bound P.

Detailed knowledge of chemical processes influencing colloid-facilitated transport is valuable in designing remediation methods for soil sites polluted with strongly sorbing contaminants. Mansfeldt et al. (2004) investigate leaching of both dissolved and colloidal cyanide as affected by citrate addition, which is of interest in cleanup operations at manufactured gas plants and coal coking plants.

We hope that this special issue may guide our research approaches and model concepts with respect to the transport and fate of soil colloids and strongly sorbing chemicals and nutrients in the vadose zone, and will serve as a resource for future studies. We would like to thank Drs. Markus Flury and John E. McCray for their effort as associate editors on this special section along with the reviewers and staff of *Vadose Zone Journal*. Finally, we thank the authors for preparing such high quality, scientific papers.

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