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Title
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Permalink

Journal

ISSN

Authors
Wang, C
Wang, R
Huo, Z
et al.

Publication Date

DOI

Peer reviewed

WIREs WATER WILEY

Colloid transport through soil and other porous media under transient flow conditions—A review

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Abstract

Understanding colloid transport in porous media under transient-flow conditions is crucial in understanding contaminant transport in soil or the vadose zone where flow conditions vary constantly. In this article, we provide a review of experimental studies, numerical approaches, and new technologies available to determine the transport of colloids in transient flow. Experiments indicate that soil structure and preferential flow are primary factors. In undisturbed soils with preferential flow pathways, macropores serve as main conduits for colloid transport. In homogeneously packed soil, the soil matrix often serves as filter. At the macroscale, transient flow facilitates colloid transport by frequently disturbing the force balance that retains colloids in the soil as indicated by the offset between colloid breakthrough peaks and discharge peaks. At the pore-scale and under saturated condition, straining, and attachment at solid–water interfaces are the main mechanisms for colloid retention. Variably saturated conditions add more complexity, such as immobile water zones, film straining, attachment to air–water interfaces, and air–water–solid contact lines. Filter ripening, size exclusion, ionic strength, and hydrophobicity are identified as the most influential factors. Our review indicates that microscale and continuum-scale models for colloid transport under transient-flow conditions are rare, compared to the numerous steady-state models. The few transient flow models that do exist are highly parameterized and suffer from a lack of a priori information of required pore-scale parameters. However, new techniques are becoming available to measure colloid transport in real-time and in a nondestructive way that might help to better understand transient flow colloid transport.

This article is categorized under:
    Science of Water > Hydrological Processes
    Science of Water > Water Quality

KEYWORDS

colloid transport, transient flow, vadose zone, unsaturated soil, preferential flow, porous media, particle transport

1 | INTRODUCTION

Particles with an effective diameter of 1 nm to 10 μm are considered colloids (Chrysikopoulos & Sim, 1996; Gao, Cao, Dong, Luo, & Ma, 2011; Sirivithayapakorn & Keller, 2003a; Smith et al., 2007; Vasiliadou & Chrysikopoulos, 2011; Wan & Wilson, 1994b; Wang, Schneider, Parlange, Dahlke, & Walter, 2018). In the subsurface environment colloids comprise many naturally occurring substances including clays, metal oxides, mineral precipitates, and organic compounds as well as biological organisms such as pathogenic bacteria (Wang et al., 2017, 2018; Wang, Schneider, et al., 2018), viruses (Torkzaban, Hassanizadeh, Schijven, Bruijn, & Husman, 2006; Torkzaban, Hassanizadeh, Schijven, & van den Berg, 2006; Zhang, Hassanizadeh, Raoof, van Genuchten, & Roels, 2012), and protozoa (Bradford, Wang, Kim, Torkzaban, & Šimůnek, 2014; Sen, 2011). Many known environmental pollutants can attach to and move with colloids, a process referred to as colloid-facilitated transport, which has been identified as one of the most important mechanisms responsible for the mobilization of reactive heavy metals in soils (Barton & Karathanasis, 2003; Gao et al., 2011; Grolimund & Borkovec, 2005; Kretzschmar & Schafer, 2005). Other contaminants transported by colloid-facilitated transport include radionuclides from mines (Artinger et al., 2002; Malkovsky et al., 2015; Severino, Cvetkovic, & Coppola, 2007), heavy metals (Chekli et al., 2016; Ouyang, Shinde, Mansell, & Harris, 1996), organic compounds such as pesticides and herbicides from agricultural land (Barton & Karathanasis, 2003), or excess nutrients such as phosphorous from farmland (de Jonge, Moldrup, Rubæk, Schelde, & Djurhuus, 2004). Biocolloids (i.e., colloidal-sized microbes and pathogens) and many of the pollutants transported with colloids listed above are known to pose a threat to public and environmental health (e.g., eutrophication from excess nutrients, poisoning by heavy metals, disease outbreaks from waterborne pathogens; Haygarth et al., 2006; Heathwaite, Haygarth, Matthews, Preedy, & Butler, 2005; Kouznetsov et al., 2007; Ren & Packman, 2005). Therefore, understanding colloid transport in surface and subsurface environments is crucial to protect public health and aquatic ecosystems from potential contaminants of concern (Masliyah & Bhattacharjee, 2006).

To date, several decades of research have been devoted to improve understanding of the fate and transport of colloids in porous media (Anderson, 1989; Bradford & Torkzaban, 2008; de Jonge et al., 2004; Elimelech, Nagai, Ko, & Ryan, 2000; Masliyah & Bhattacharjee, 2006; McCarthy & McKay, 2004; Wan & Wilson, 1994a). It has been found that colloid transport and retention in porous media are generally governed by advection, dispersion, and inactivation, as well as by colloid interactions with various interfaces (Keswick & Gerba, 1980; Schijven & Hassanizadeh, 2000). Based on years of research, it is widely accepted that due to their small size, colloidal suspensions moving through porous media are excluded from pores through which they physically cannot fit (i.e., straining; Bradford & Bettahar, 2005), they have limited diffusivity (James, Wang, & Chrysikopoulos, 2018; Soto-Gómez, Perez-Rodriguez, Vazquez Juiz, Lopez-Periago, & Paradelo Perez, 2019; Yu et al., 2019), and are generally electrostatically repelled from like-charged porous media surfaces (Johnson, Rasmuson, Pazmino, & Hilpert, 2018; Rasmuson, VanNess, Ron, & Johnson, 2019; VanNess, Rasmuson, Ron, & Johnson, 2019). Because of these features, their transport is both enhanced and impeded in the subsurface, compared to non-sorbing solutes (Bradford, Šimůnek, Bettahar, van Genuchten, & Yates, 2003; McCarthy & Zachara, 1989; McDowell-Boyer, Hunt, & Sitar, 1986). Research has also shown that depending on the flow regime (e.g., steady-state vs. transient) and water content of the porous media, the dominant processes controlling colloid transport, attachment, retention, and remobilization can vary substantially influencing the arrival time, peak concentration, and travel distance of colloids transported in porous media (Carstens, Bachmann, & Neuweiler, 2017; Cheng & Saiers, 2010; Flury & Aramrak, 2017; Keller & Auset, 2007; Knappenberger, Flury, Mattson, & Harsh, 2014; Sen, 2011). It is further understood that solution chemistry including pH (Chotpantarat & Kiatvarangkul, 2018; Ma et al., 2018; Rastghalam, Yan, Shang, & Cheng, 2019), ionic strength (Jahan, Alias, Bin Abu Bakar, & Bin Yusoff, 2019; Magal, Weisbrod, Yechieli, Walker, & Yakirevich, 2011; Rod et al., 2018; Zhuang et al., 2010), and organic matter (Ma et al., 2018; Ma, Guo, Weng, et al., 2018; Yang et al., 2019) as well as the texture and soil grain surface roughness (Kretzschmar & Sticher, 1997; Rasmuson et al., 2019; Redman, Grant, Olson, & Estes, 2001) enhance or impede these processes.

In principle, the Derjaguin–Landau–Verwey–Overbeek (DVLO) theory is widely accepted in describing colloid attachment and detachment processes and general interactions between colloids and soil grains in porous media (Hahn, Abadzic, & O'Melia, 2004; Muller, 1994; Redman, Walker, & Elimelech, 2004; Tufenkji & Elimelech, 2005). Colloids may be attached or deposited to grain surfaces when the net attractive forces (sum of the interactions of electrical double layers and van der Waals–London forces) fall into either the primary or secondary energy minimum (Gao et al., 2011). However, solution chemistry, grain size, pore flow velocity, and colloid concentration all influence the shape of the DLVO interaction energy curve, the magnitude of the energy barrier, and the depth of the primary and

secondary minimum (Carstens et al., 2017; Liu et al., 2018; Rasmuson et al., 2019; Rastghalam et al., 2019; Xu et al., 2016; Yang et al., 2019) and with that the deposition efficiency of colloids from bulk solution. These parameters have also been found to control colloid detachment, the release of deposited colloids from the soil grain, which is dependent on the balance of repulsive forces between colloid and grain surfaces (Chequer, Bedrikovetsky, Carageorgos, Badalyan, & Gitis, 2019; Ma et al., 2016; Ma, Guo, Lei, et al., 2018; Mohanty, Saiers, & Ryan, 2016; Sepehrnia, Fishkis, Huwe, & Bachmann, 2018; VanNess et al., 2019). In ideal porous media and saturated conditions, colloid retention is mainly controlled by the net rate of colloid deposition and release. This mechanism, also known as the classic "clean-bed" filtration model (see Yao, Habibian, & Omelia, 1971), describes the removal of suspended colloids by first-order kinetics and assumes an exponential decrease in colloid retention with transport distance in porous media (Logan, Jewett, Arnold, Bouwer, & O'Melia, 1995; Tufenkji & Elimelech, 2004b). Despite its widespread use in modeling and experimental studies (Albinger, Biesemeyer, Arnold, & Logan, 1994; Baygents et al., 1998; Bolster, Mills, Hornberger, & Herman, 2000; Camesano & Logan, 1998; Martin, Logan, Johnson, Jewett, & Arnold, 1996; Redman, Estes, & Grant, 2001), observations have shown hyperexponential (a decreasing rate of deposition coefficient with distance; Albinger et al., 1994; Baygents et al., 1998; Bolster et al., 2000; Bradford & Bettahar, 2005; Bradford, Yates, Bettahar, & Simunek, 2002; DeFlaun & Condee, 1997; Redman, Estes, & Grant, 2001; Simoni, Harms, Bosma, & Zehnder, 1998) or nonmonotonic (a peak in retained colloids away from the injection source; Bradford, Yates, Bettahar, & Simunek, 2002; Tong, Kashima, Shirai, Suda, & Matsumura, 2005) deposition profiles, which have been attributed to soil grain surface and physical properties, surface charge heterogeneity, interfacial electrodynamics, and colloid deposition in the secondary DLVO energy minimum (Bradford & Torkzaban, 2008; Gao et al., 2011). Other processes controlling colloid transport in porous media that are not included in the filtration theory are pore straining (i.e., deposition of colloids in small pore spaces such as those formed at grain-to-grain contacts; Bradford, Simunek, Bettahar, van Genuchten, & Yates, 2006; Bradford, Simunek, & Walker, 2006), thin-water films (i.e., film straining; Wan & Tokunaga, 1997), air–water–solid interfaces (i.e., capillary retention; Gao et al., 2008), and immobile waters (i.e., immobile-water trapping; Gao, Saiers, & Ryan, 2006).

Motivated by the need to advance understanding of contaminant transport through groundwater systems (Grolimund & Borkovec, 2005; Roy & Dzombak, 1997; Saiers & Hornberger, 1999; Turner, Ryan, & Saiers, 2006; Zhuang, Flury, & Jin, 2003), much of the existing research on colloid transport has focused on saturated porous media (Vasiliadou & Chrysikopoulos, 2011; Wang et al., 2012; Yang et al., 2019; Zhuang et al., 2003). Fewer studies have centered on colloid transport in unsaturated porous media, despite the recognition that substantial amounts of colloids are either generated in or passed through the vadose zone (Cheng & Saiers, 2010; Ranville, Chittleborough, & Beckett, 2005; Seaman, Bertsch, & Strom, 1997). The vadose zone plays a crucial role in mitigating the risk of contamination of drinking-water aquifers and surface water bodies (Cey, Rudolph, & Passmore, 2009; Flury, Fluhler, Jury, & Leuenberger, 1994). Colloid transport in the unsaturated zone (i.e., vadose zone) is more complicated than under saturated conditions due to the presence of air, capillary forces, and transients in flow and chemistry, which in part due to heterogeneity in soil structure often restrict the water flow and colloid transport to smaller pore space regions in the vadose zone (Bradford & Torkzaban, 2008; Cheng & Saiers, 2010; Denovio, Saiers, & Ryan, 2004). The physical and geochemical heterogeneity present in the unsaturated zone can cause greater velocity variation and more dispersion, since only a fraction of the soil moisture is involved in the flow and colloid transport (Bradford & Torkzaban, 2008; Germann, Alaoui, & Riesen, 2002; Keller & Sirivithayapakorn, 2004; Krupp & Elrick, 1968). Research has further shown that the presence of air creates additional potential retention sites at the solid–water interfaces (SWI; Chu, Jin, Flury, & Yates, 2001; Lance & Gerba, 1984; Torkzaban, Hassanizadeh, Schijven, Bruijn, & Husman, 2006) and the air–water interfaces (AWI; Cherrey, Flury, & Harsh, 2003; Mitropoulou, Syngouna, & Chrysikopoulos, 2013; Schafer, Harms, & Zehnder, 1998; Torkzaban, Hassanizadeh, Schijven, & van den Berg, 2006; Wan & Wilson, 1994a, 1994b). Colloids can also be retained in water films enveloping the soil grains (i.e., film straining; Saiers & Lenhart, 2003a, 2003b; Wan & Tokunaga, 1997) and at solid–air–water triple points (Chen & Flury, 2005; Crist et al., 2004; Crist, Zevi, McCarthy, Throop, & Steenhuis, 2005; Steenhuis et al., 2006; Zevi, Dathe, McCarthy, Richards, & Steenhuis, 2005).

An important distinction between studies of colloid transport in unsaturated porous media versus colloid transport in transient flow conditions is the state of the hydrologic regime. Most unsaturated porous media studies have been conducted under unsaturated but steady water content conditions (i.e., constant trickling flow; Keller & Sirivithayapakorn, 2004), while transient flow studies specifically investigate colloid transport during nonsteady flow transients (i.e., variable flow, wetting-drying cycles; Cheng & Saiers, 2010). Transient pore water flow most closely resembles the flow occurring in the unsaturated zone in response to rainfall, snowmelt or irrigation events, when the

water potential and the energy required to move water in the soil changes over time between the wilting point, field capacity, and saturation (O'Geen, McDaniel, & Boll, 2002; O'Geen, McDaniel, Boll, & Brooks, 2003). Transient flow has been shown to mobilize more colloids, because the variation in soil water content (e.g., during infiltration/imbibition or drainage) can lead to sudden changes in the local soil water potential, pore water saturation, AWI surface area, AWI locations, and thickness of water films (Zhuang, McCarthy, Tyner, Perfect, & Flury, 2007). The dynamic nature of non-steady state flow can cause shifts in chemical factors (e.g., ionic strength, pH, surface charge, and chemical composition of the pore water) and physical factors (e.g., pore size distribution, shrinking, and swelling of the soil) in the unsaturated zone, which might fundamentally influence the importance and magnitude of some of the processes assumed to be primary controls on colloid transport in steady-state flow systems (Saiers, Hornberger, Gower, & Herman, 2003; Saiers & Lenhart, 2003a, 2003b; Torkzaban, Hassanizadeh, Schijven, Bruijn, & Husman, 2006; Torkzaban, Hassanizadeh, Schijven, & van den Berg, 2006; Wang et al., 2019). Despite the fact that colloid transport in transient flow has received the least attention in colloid research to date (Baumann, 2007; Lazouskaya & Jin, 2008; Lazouskaya, Jin, & Or, 2006; Sang et al., 2013; Wan & Wilson, 1994a), transient flow experiments are often considered suitable for making inferences on the mobilization of colloids, in addition to the transport and retention mechanisms (Cheng & Saiers, 2010; Chequer et al., 2019; Ma et al., 2016; Mohanty et al., 2016; Sepehrnia et al., 2018). This is particularly important for nonpoint source pollution mitigation since transient flow more closely mimics the phenomenon of colloid transport and colloid-facilitated transport of contaminants as it occurs in real vadose zone environments (Gao et al., 2006; McCarthy & McKay, 2004; Saiers & Lenhart, 2003a; Zhuang et al., 2007).

The study of colloid transport under transient flow conditions is also of interest because the soil structure and soil heterogeneity play a much greater role in these conditions. Macropores and naturally occurring soil layers have been shown to have profound effects on colloid transport (Bond, 1986). Depending on soil structure, pore connectivity, and the presence of macropores, preferential flow (e.g., short-circuiting flow, finger flow, funnel flow; Kung, 1990a; Nimmo, 2012; Seyfried & Rao, 1987) can occur which can increase the vertical transport distance of colloids and attached contaminants (Kung, 1990b). Although colloid and contaminant mobility has been shown to decline with decreasing volumetric moisture content because of a decrease in colloid mobility and an increase in contaminant desorption from mobile colloids (Chen, Flury, Harsh, & Lichtner, 2005; Harter, Wagner, & Atwill, 2000; Lenhart & Saiers, 2002), during re-saturation (i.e., imbibition) when the water film thickness around the solid phase is increasing, retained colloids can be remobilized into the mobile phase and recovered in the outflow, showing that the retention of colloids is reversible and mainly controlled by the degree of water saturation (Chen & Flury, 2005; Crist et al., 2005). Colloid remobilization is increasing with water saturation (El-Farhan, Denovio, Herman, & Hornberger, 2000; Kjaergaard, Moldrup, de Jonge, & Jacobsen, 2004; Kjaergaard, Poulsen, Moldrup, & de Jonge, 2004; Saiers & Lenhart, 2003a). However, during flow transients even higher colloid concentrations have been observed (El-Farhan et al., 2000; Flury, Mathison, & Harsh, 2002; Litaor et al., 1998; Ryan, Illangasekare, Litaor, & Shannon, 1998; Saiers & Lenhart, 2003a; Shang, Flury, Chen, & Zhuang, 2008; Totsche, Jann, & Kogel-Knabner, 2007; Zhuang et al., 2007), highlighting the importance of and need to study the short-lived but often abrupt changes in soil moisture content, flow velocity, and pore water chemistry.

The above literature indicates that many colloid retentions and remobilization processes in flow transients are still poorly understood and quantified. This is in part due to a lack of laboratory studies able to observe these processes at the spatial–temporal time scales they occur at. However, the incomplete understanding of colloid transport processes in transient flow systems has major implications for contaminant transport mitigation as it presents a challenge to numerical modeling (van Genuchten & Šimůnek, 2004). Most colloid transport models simulate systems in steady-state conditions and only a few attempt to simulate transient flow conditions. This is because, in saturated steady-state systems, water flow is a function of location only for which analytical solutions are available (Denovio et al., 2004; Flury & Qiu, 2008). In contrast, under transient flow conditions, water flow is a function of both location and time. In this case, not many analytical solutions are available, hence most models turn to numerical solutions for these systems (van Genuchten & Šimůnek, 2004).

In this article, we review our current understanding of the main transport and retention mechanisms associated with colloid transport in transient flow. Transient flow here is defined as flow undergoing temporal change due to transients in soil moisture content and flow velocity. We begin our review by summarizing recent literature findings from laboratory experiments at the column scale (i.e., macroscale) and investigate the role that soil structure (e.g., homogeneous vs. intact/undisturbed/heterogeneous soils) and saturation of different subregions within the soil play in colloid recovery and breakthrough. We then review the main mechanisms and factors influencing colloid transport and retention at the microscale and explore the importance of these mechanisms in transient flow systems. We

exclude the role of biota, microbes, and particulate organic matter on colloid fate (growth, inactivation, and degradation) and transport in this review. Finally, we continue with a survey of current mathematical and numerical models that describe colloid transport in transient flow and conclude with a summary of research gaps and recommendations for future research.

2 | MECHANISTIC UNDERSTANDING GAINED FROM EXPERIMENTS

Most of the mechanisms controlling colloid transport have been discovered through laboratory and field experiments, however, the understanding gained depends on the scale at which the transport phenomenon has been observed. In this first part of our review, we summarize important experimental discoveries made at the macroscale (e.g., column and lysimeter experiments) and microscale (e.g., pore and interface scale). We will first summarize discoveries from macroscale experiments, which mostly analyze breakthrough patterns from systems subject to different transient flow conditions. We will then focus on microscale processes by starting with a review of colloid transport mechanisms under saturated conditions before reviewing the complexity added through the variably saturated conditions.

2.1 | Macroscale phenomena

Review of literature studies on colloid transport through soil under transient flow conditions reveals that most column scale experiments can be grouped into two categories: Colloid transport (a) through homogeneously packed soil (Table 1) and (b) through intact/undisturbed or artificially packed heterogeneous soil columns (Table 2). The latter soil column experiments are often characterized by a dual permeability system consisting of fast preferential flow pathways and the slower flow and transport through the soil matrix (Kinsall et al., 2000). Homogeneously packed soil is often seen as an equivalent to the soil matrix in dual permeability systems. Among known preferential pathway types (e.g., short-circuiting flow, funnel flow, finger flow; Gerke, 2010), the preferential pathways covered in this review mainly include the flow through continuous macropores and the flow at the interface of two soil layers comprised of two types of porous media (Wang, Bradford, & Simunek, 2014).

Most macroscale (i.e., column scale) experiments investigating colloid transport processes in transient flow infer governing processes and drivers from the breakthrough curve, specifically, the magnitude and timing of the colloids peak in the discharge compared to the discharge peak itself. Early arrival or peaks in colloid concentration in the effluent are typically attributed to the occurrence of preferential flow (Kinsall et al., 2000; Majdalani, Michel, Di Pietro, Angulo-Jaramillo, & Rousseau, 2007; Wang et al., 2014) (Figure 1a), or the occurrence of sharp changes in water flow caused by, for example, (a) the onset of rainfall or irrigation (Liu, Flury, Zhang, et al., 2013; Mohanty, Bulicek, et al., 2015; Zhuang et al., 2007; Figure 1b) or (b) a sharp increase or decrease in discharge or soil water content due to the movement of the wetting or drying front (Cheng & Saiers, 2010; El-Farhan et al., 2000; Liu, Flury, Harsh, Mathison, & Vogs, 2013; Shang et al., 2008; van den Bogaert et al., 2016; Wang et al., 2019; Zhuang et al., 2007; Figure 1c). Colloids that are transported through macropores have been observed to result in the highest recovery rates, while colloids transported through the soil matrix have very low recovery rates (Mohanty, Bulicek, et al., 2015; van den Bogaert et al., 2016). Auset et al. (2005), Kinsall et al. (2000), and Rousseau et al. (2004) observed colloid concentration peaks that were concurrent with discharge peaks or soil water content peaks (Figure 1d). They concluded that these colloids likely moved through the soil matrix in a similar way as conservative solutes. Finally, the occurrence of colloid concentration peaks during drainage (Figure 1b) has been interpreted to be indicative of colloids moving with the drying front (Kinsall et al., 2000; Powelson & Mills, 2001; Zhuang et al., 2007).

2.2 | Microscale mechanisms

The column scale phenomena discussed above are macroscale demonstrations of pore scale or interface scale colloid transport mechanisms. The microscale mechanisms influencing colloid transport include (a) the basic colloid transport mechanisms under fully saturated condition and (b) mechanisms added due to variably saturated condition. Note that unless explicitly stated otherwise, all mechanisms discussed in Section 2.2 were demonstrated through direct experimental observations in the original research. We list a few examples where pore-scale mechanisms were inferred by parameter fitting.

TABLE 1 Colloid transport experiments through homogeneously packed soil under transient flow conditions

Experiment	Colloid type	Colloid concentration peak timing	Key processes, forces or factors
Powelson and Mills (2001)	*Escherichia coli*	Out of phase with soil water content peaks	Moving air–water–solid (AWS) contact lines
Auset, Keller, Brissaud, and Lazarova (2005)	*Escherichia coli*	Concurrent with discharge peaks	Attachment to and release from air–water interfaces (AWIs) and film straining; attachment to solid–water interfaces (SWIs) was irreversible
Keller and Sirivithayapakorn (2004)	Carboxylate-modified polystyrene latex spheres (3.0 μm and 50 nm); bacteriophage MS2 (~25 nm)	After discharge peak	Retention at AWIs; film straining; cluster formation
Crist et al. (2004)	Hydrophilic negatively charged microspheres in two different sizes (0.3 and 0.8 μm)	No breakthrough curve	Trapping at or near AWS interface
Gao et al. (2006)	Carboxylate-modified polystyrene latex microspheres with a mean diameter of 2.0 μm	No breakthrough curve	Persisted attachment to air bubble surfaces and SWIs; trapped by and released from AWIs of the pendular rings and thin-water films; retained in and released from immobile water zones
Zhuang et al. (2010)	Fluorescent latex microsphere (980 nm in diameter)	No breakthrough curve	Coupled effects of solution surface tension and ionic strength
Zhuang et al. (2007)	In situ colloids	Primary peak soon after the beginning of imbibition; drainage induced secondary peaks	Moving AWIs and extension/shrinkage of water films; capillary, electrostatic, van der Waals and hydrophobic forces; ionic strength
Shang et al. (2008)	In situ colloids	Concurrent with sharp increases of discharge (i.e., the arrival of infiltration fronts)	Capillary force dominated
Cheng and Saiers (2009)	In situ colloids	Concurrent with passage of wetting or drying fronts	Moving wetting and drying fronts; ionic strength; flow rate
Cheng and Saiers (2010)	In situ colloids	Concurrent with sharp changes of discharge and moisture content, both sharp increases and sharp decreases	During drainage, AWIs scouring; during imbibition, expansion of thin-water films, reconnection of stagnant water with water flow, increased shear forces and redeposition
Zhuang, Tyner, and Perfect (2009)	Montmorillonite clay	Primary peak soon after the beginning of imbibition; drainage induced secondary peaks	Coupled effects of water content, flow velocity and ionic strength
Liu et al. (2013)	Eu-hydroxy-carbonate colloids	Correlated with precipitation/irrigation	Accumulation of dispersive colloids by physical straining during drying period; moving AWIs created by transfer between wetting and drying

Note: In situ colloids are soil mineral particles that are being mobilized during transient flow. AWS and AWI are abbreviations for air–water–solid and air–water interface.

TABLE 2 Colloid transport experiments through intact/undisturbed or artificially packed heterogeneous soil under transient flow conditions
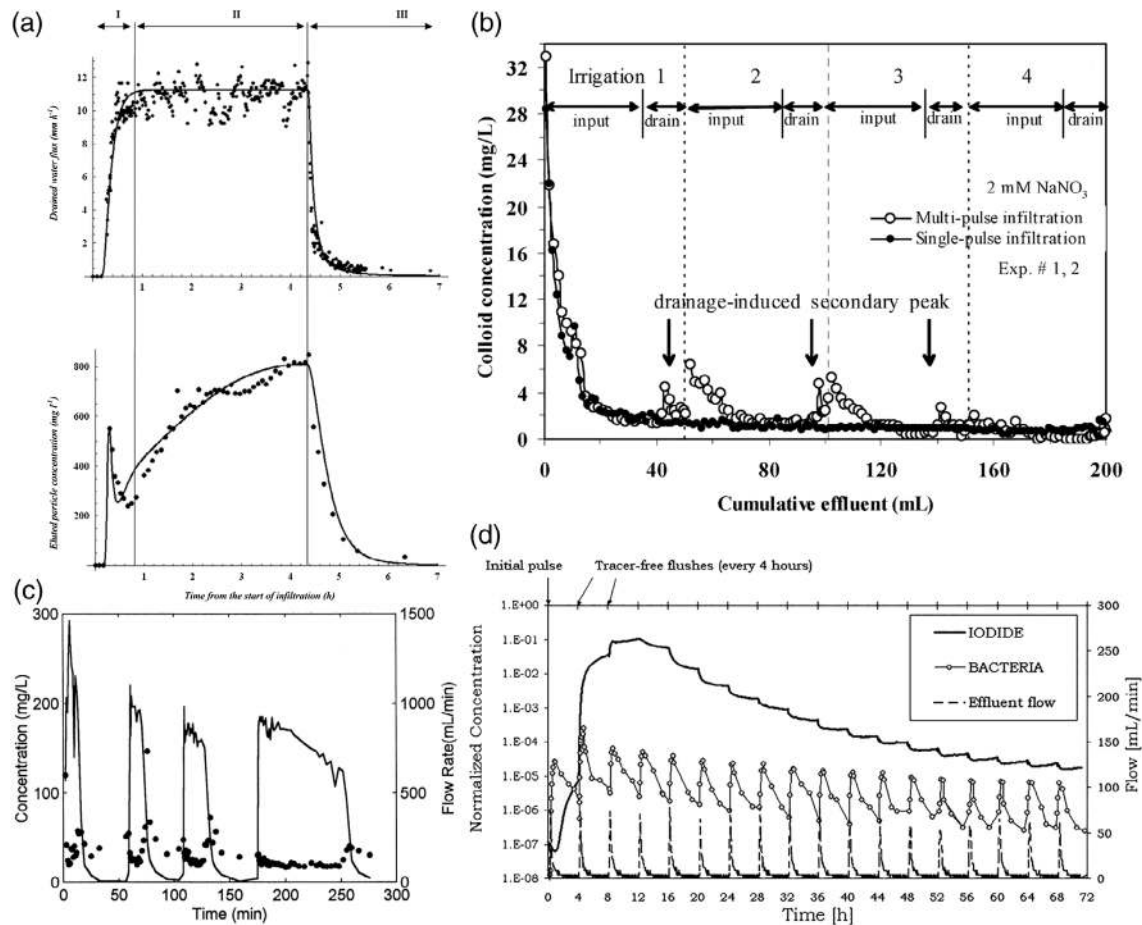
Experiment	Colloid type	Colloid concentration peak timing	Key processes or forces
Kinsall, Wilson, and Palumbo (2000)	GFP bacteria (*Pseudomonas putida* modified by the addition of the green fluorescent protein [GFP] gene)	The first short lasting peak was concurrent with discharge peak; the second, long lasting peak was during drainage	Dual pore system: Preferential flow pathways and pores in soil matrix
El-Farhan et al. (2000)	In situ colloids	Concurrent with sharp changes of discharge, both sharp increases and sharp decreases	Moving AWIs
Rousseau, Di Pietro, Angulo-Jaramillo, Tessier, and Cabibel (2004)	In situ colloids	Concentration correlated with discharge	Hydrodynamic shear; ionic strength, size exclusion
Liu, Flury, Harsh, et al. (2013)	In situ colloids	Concurrent with sudden changes of discharge	Physical disturbance
Mohanty, Saiers, and Ryan (2015)	In situ colloids	Peaked at the start of rainfall events	Preferential flow and matrix flow; colloid generated mainly from fractures; colloid released by moving wetting and drying fronts
Zhang et al. (2016)	In situ colloids	Concurrent with the flow generation (including surface runoff, interflow at the interface of soil and mudrock, and fracture flow at the interface of mudrock and sandstone)	Four types of preferential pathways: surface runoff, macropore flow, interflow, and fracture flow; size selection effect: <10 μm colloids dominated in surface runoff, and >10 μm colloids dominated in subsurface flow; moving AWI; hydrodynamic shear force
Mohanty, Bulicek, et al. (2015)	Fluorescent polystyrene microspheres of two sizes (0.5 and 1.8 μm)	Peaked at the start of rainfall events	Preferential flow induced by macropores; during wetting periods, shear, water film expanding, and AWI scouring; during drying periods, AWI scouring; less redeposition in macropores; size exclusion
van den Bogaert, Cornu, and Michel (2016)	Rain-applied fluorescent colloids	Concurrent with a sharp increase of discharge	Preferential flow; during drying periods, water drained faster in macropores, and moved towards narrower pores; during wetting periods, water saturated the soil matrix, and infiltrated into macropores
Wang et al. (2019)	DNA-labeled microsphere (~850 nm)	Peaked at the start of rainfall events	Straining; physical–chemical filtration; preferential flow along capillary barrier; size exclusion; cluster formation

Note: AWS and AWI are abbreviations for air–water–solid and air–water interface.

2.2.1 | Colloid transport mechanisms under fully saturated condition

In saturated soil columns, straining and attachment are the two main mechanisms that hinder colloid transport. Straining is the retention at the wedge-shaped, grain to grain contacts (Zhang et al., 2010). Colloids collide with soil grains by three mechanisms: sedimentation, interception, and Brownian motion (McDowell-Boyer et al., 1986; Sakthivadivel, 1969). Whether a collision leads to an attachment is determined by the colloid-surface interaction forces, mainly electrostatic and London–van der Waals forces. The Derjaguin–Landau–Verwey–Overbeek (DLVO) theory is the fundamental theory describing the forces involved in the attachment mechanisms (McDowell-Boyer et al., 1986). However, other forces can also act on colloids, such as the hydrophobic force, Born repulsion force, among others. The

FIGURE 1 Different colloid discharge concentration peak timings compared to water transients. (a) Occurrence of colloid concentration peaks as a result of preferential flow (note the occurrence of the colloid concentration peak before the discharge peak; Reprinted with permission from Majdalani et al. (2007). Copyright 2007 John Wiley and Sons); (b) colloid concentration peaks soon after the onset of irrigation and during drainage (i.e., drainage-induced secondary peaks; Reprinted with permission from Zhuang et al., 2007); (c) occurrence of colloid concentration peaks concurrent with sharp changes in discharge (the line indicates flow rate and the dots indicate concentrations; Reprinted with permission from El-Farhan et al. (2000). Copyright 2000 American Chemical Society); (d) colloid concentration peaks concurrent with discharge peaks (Reprinted with permission from Auset et al. (2005). Copyright 2005 John Wiley and Sons)

inclusion of these forces results in various kinds of extended DLVO theories (Bradford & Torkzaban, 2008; Lazouskaya et al., 2013). Auset and Keller (2006), for example, demonstrated through direct pore-scale observation that when the pore throat to colloid diameter ratio (T/C) is less than 1.8, straining dominates; if T/C is larger than 2.5, attachment dominates; while if T/C is between 1.8 and 2.5, both mechanisms are important. And, both can lead to filter ripening.

Straining

In contrast to mechanical filtering and attachment, straining of colloids occurs in soil pores that are defined by the intersection of at least two solid–water interfaces (SWIs) such as grain-to-grain contacts or surface roughness (Auset & Keller, 2006; Bradford & Bettahar, 2006; Bradford, Simunek, Bettahar, et al., 2006). Straining often only occurs in a fraction of the soil pore space and is largely dependent on the structure of the porous media skeleton, the relative size of the colloid to the pore throats, and the surface roughness of the media grains (Auset & Keller, 2006). Auset and Keller (2006) demonstrated through optical microscopy that straining happens mainly in the first one or two pore throats as the colloids enter the porous media. They also found that straining is not influenced by flow rate. Sang et al. (2013) more recently demonstrated that straining occurs irrespective of changes in ionic strength or saturation conditions. Tufenkji and Elimelech (2004a, 2004b) also found that straining increases with increasing surface roughness. As such, straining is of significant importance for colloid retention in natural soil and vadose zone environments, for contaminated waste sites, and during riverbank filtration. Approximating soil pores as a bundle of tortuous cylindrical capillary

tubes, Bradford, Simunek, Bettahar, et al. (2006) estimated the straining of common viruses, bacteria, and protozoa will occur in 10–92% of the pore space depending on soil texture. Straining can be released by reversing the flow direction (Foppen, Herwerden, & Schijven, 2007) or by breaking down the porous media skeleton (Sang et al., 2013). However, a broader definition of straining may lead to other influencing factors as discussed by Bradford and Torkzaban (2008) in their "pore scale" section.

Attachment at the solid–water interface

One of the main mechanisms of colloid deposition is the collision with and subsequent retention of colloids at the solid–water interface. A solid–water interface (SWI) is the part of a soil particle surface that is in contact with water (Auset et al., 2005; Gao et al., 2006). In saturated porous media or saturated regions in partly saturated porous media, colloids often solely attach to SWI. According to DLVO theory or the extended DLVO theories, a colloid particle can be held at the secondary minimum or primary minimum depending on its distance to the SWI. Retention at the secondary minimum is often an intermediate state, as the colloid can be re-entrained into the bulk fluid or it can overcome the energy barrier and become attached to the SWI at the primary minimum (Johnson & Hilpert, 2013). Retention at the secondary minimum can be released by sharply reducing the ionic strength, for example, through flushing with deionized water (Sang et al., 2013). However, colloid attachment to SWIs in the primary minimum is irreversible under saturated conditions (Auset et al., 2005; Gao et al., 2006; Figure 2a). Once colloids attached to a SWI, the variation in local pore flow velocity cannot mobilize them under the experimental conditions as long as the local pore remains saturated.

Filter ripening

Colloids attached to SWIs can serve as extended attachment sites for bypassing colloids in solution (Keller & Auset, 2007). This phenomenon is called filter ripening, clogging, or blocking. Several colloids attached at the same location may even form a cluster (Keller & Auset, 2007; Figure 2a). Filter ripening hinders colloid transport and removes colloidal and colloidal borne contaminants. As a result of filter ripening, the permeability of the porous media can be decreased (McDowell-Boyer et al., 1986). Filter ripening is largely dependent on the flow velocity, and zeta potential and the size of the colloid (Salkar & Tembhurkar, 2016).

Size exclusion

Size exclusion refers to the phenomenon when colloids are unable to move into soil pores due to the ratio of pore throat to colloid size. Particles that do not physically fit into smaller pores tend to be transported at larger flow velocities along with more conductive flow pathways that are physically accessible. Sirivithayapakorn and Keller (2003a) directly observed this phenomenon when the throat to colloid diameter ratio was 1.5 and colloids preferentially entered larger pore throats where most of the flow was directed. They found, the larger the colloid particles, the stronger the size exclusion (Bradford & Torkzaban, 2008; Mohanty, Bulicek, et al., 2015; Sirivithayapakorn & Keller, 2003a; Wang et al., 2019). When macropores are present, smaller colloid particles are attached more strongly to macropore walls because of the relatively large specific surface area and relatively large surface charge, while larger colloid particles are more susceptible to hydrodynamic shear and capillary forces (Rousseau et al., 2004). Wang et al. (2014) also found that the size exclusion effect was more pronounced under low ionic strength (≤1 mM) conditions.

2.2.2 | Colloid transport mechanisms under variably saturated condition

Under variably saturated condition, the water and air distribution within a porous medium is controlled by capillary forces (Bradford & Torkzaban, 2008). Wetting-drying cycles together with the soil physical heterogeneity add complexity to straining and attachment. Immobile water zones emerge. Besides the straining at the grain-to-grain contacts, film straining occurs. Similarly, besides attachment at SWIs, attachment at air–water interfaces (AWIs) and air–water–solid (AWS) contact lines occur.

Wetting–drying cycles

The dynamic changes in flow velocity, flow region as well as the configuration of AWIs are the main drivers of colloid transport under transient flow conditions (Liu, Flury, Harsh, et al., 2013; Zhuang et al., 2007). Transient flow induces movement of AWS contact lines and AWIs, extension/shrinkage of water films, and connection/disconnection of
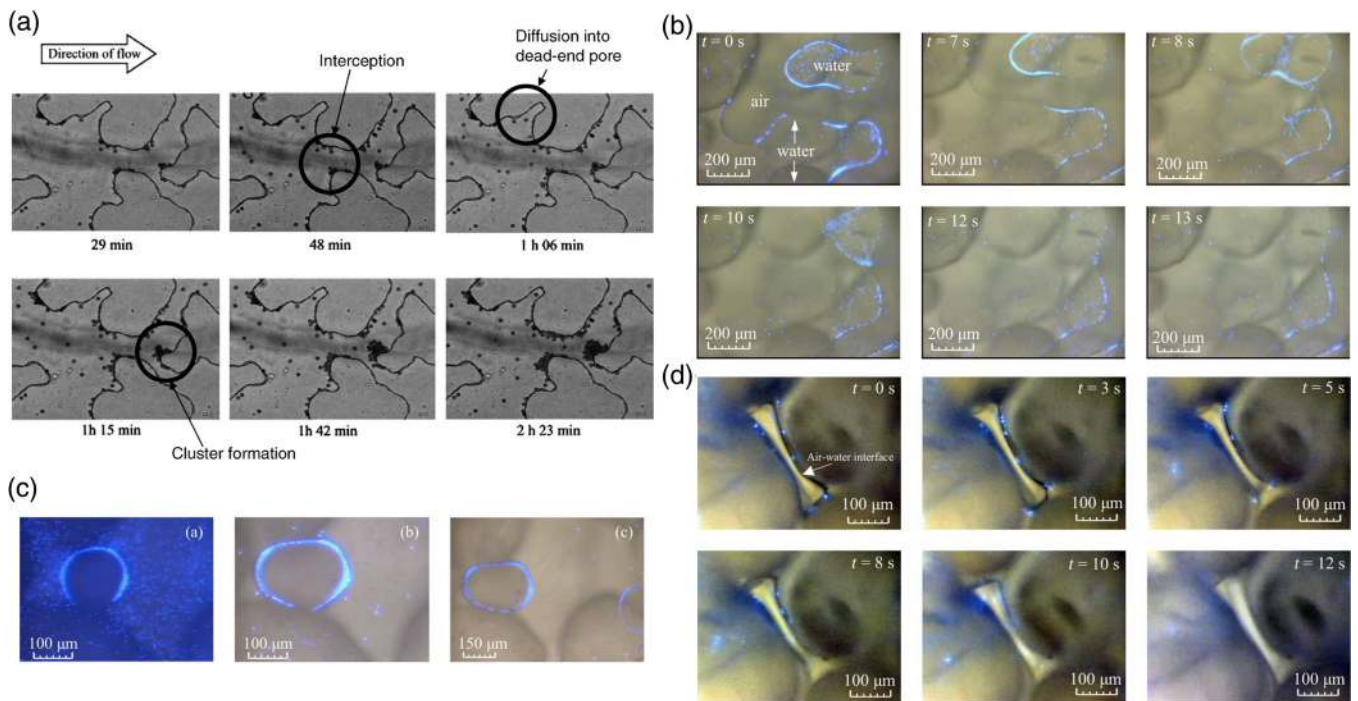
FIGURE 2 The mechanisms of colloid retention at SWIs, release from an immobile/stagnant water zone, and retention at and release from air–water interfaces (AWIs). In the filter ripening process, (a) colloids attached to SWIs can serve as extended retention sites (Reprinted with permission from Keller & Auset (2007). Copyright 2007 Elsevier). (b) Colloids are released from an immobile water zone when the wetting front passes and the immobile water zone reconnects to the bulk water flow (Reprinted with permission from Gao et al. (2006). Copyright 2006 John Wiley and Sons). (c) Colloids continuously retained at an air bubble surface (Reprinted with permission from Gao et al. (2006). Copyright 2006 John Wiley and Sons), but (d) colloids can be released from pendular rings when water invades the pore space (Reprinted with permission from Gao et al. (2006). Copyright 2006 John Wiley and Sons)

pendular rings and immobile water zones. During wetting periods, colloids are mobilized by shear (as a result of the flow increase), expanding water films, reconnecting pendular rings and immobile water zones, and AWS contact lines/ AWI scouring. During drying periods, colloids can only be mobilized by AWS contact lines/AWI scouring (Mohanty, Bulicek, et al., 2015).

Transient flow also induces frequent reconfiguration of electrostatic, van der Waals, hydrodynamic shear, capillary forces, and surface tension. As such, the force equilibrium, which retains colloids in the soil by the pore-scale mechanisms reviewed above, experiences frequent disturbances under transient flow conditions, and, as a result, more colloids can be mobilized than under steady-state conditions (Zhuang et al., 2007). Under transient flow conditions, the hydrodynamic shear force is the dominant force during imbibition (Cheng & Saiers, 2010; Rousseau et al., 2004) and more colloids are mobilized by a higher flow rate (Shang et al., 2008). During drainage, the capillary force is the dominant force (Gao et al., 2008; Shang et al., 2008; Shang, Flury, & Deng, 2009).

Soil physical heterogeneity

During our review, the existence or absence of macropores in porous media emerged as the main soil physical heterogeneity factor influencing colloid transport in transient flow (based on a comparison of studies listed in Tables 1 and 2). In contrast, soil texture (e.g., clay-rich soil vs. sandy soil) was found to be insignificant irrespective of whether macropores existed or not. When water is flowing through macropores, colloids are mobilized by the moving water (Kinsall et al., 2000; van den Bogaert et al., 2016); however, when the water content is too low to allow flow in macropores, colloids move with the remaining water to smaller pores and are filtered by the soil matrix along the way (Kinsall et al., 2000; van den Bogaert et al., 2016). By direct observation, Wang et al. (2014) discovered that it was the length and connectivity of macropores that mainly impacted the preferential transport of colloids. And, this impact was more pronounced under high ionic strength ($\geq$20 mM).

Soil physical heterogeneity was found to enhance the effect of wetting-drying cycles, especially the drying duration and drying condition (Majdalani, Michel, Pietro, & Angulo-Jaramillo, 2010; van den Bogaert et al., 2016). For colloids

applied with rainfall, van den Bogaert et al. (2016) observed that when preferential flow dominates, colloid retention increases with increased drying duration. Whereas, when matrix flow dominates, extensive colloid retention occurs, and the effect of drying duration is negligible. Based on these results, van den Bogaert et al. (2016) speculated that at the beginning of a drying period, water drains faster in macropores and redistributes toward the narrower pores in the soil matrix due to capillary force (Graphical Abstract). During the next rainfall event, when the soil matrix is saturated with the colloid suspension and the soil matrix permeability is exceeded by the rainfall intensity, the colloid suspension infiltrates into the macropores, preferential flow occurs, and colloids are transported with the higher velocity flow in the macropores (Graphical Abstract). When drying duration increases, the colloid suspension passes through an increasing number of increasingly narrower pores in the soil matrix (Graphical Abstract). As a result, the colloids get in contact with an increasing area of SWIs and are more likely to attach to the soil matrix due to the larger contact area (Kretzschmar, Robarge, & Amoozegar, 1994).

By testing the mineral composition of the effluent colloids, Mohanty, Saiers, and Ryan (2015) proved that in situ colloids (i.e., colloids naturally present in soil) were generated mainly from the macropore walls in the intact soil. They also observed that in regions with abundant macropores, increasing the drying duration helped to generate more colloids until a critical drying duration was reached (2.5 days), after which less colloids were generated with longer drying duration. Whereas, at the regions without abundant macropores, the colloid generation was kept at a low level, which is not affected by drying duration. The generated colloids, regardless of the sources, were available for mobilization during the subsequent wetting period. Mohanty, Saiers, and Ryan (2014) also demonstrated that a freezing–thawing cycle during a drying period could create more preferential pathways, which in turn mobilized more in situ colloids during the subsequent wetting period. In addition, if the drying–wetting cycle was repeated several times, even more colloids could be mobilized during later wetting periods (Mohanty et al., 2014).

The role of immobile water zones

Immobile water (or stagnant water) is the pore water that connects to the bulk pore water only through very thin water films (Auset et al., 2005; Gao et al., 2006). Colloids retained in an immobile water zone are released when the wetting front passes through and the immobile water zone reconnects with the bulk flow (Auset et al., 2005; Cheng & Saiers, 2010; Gao et al., 2006; Figure 2b).
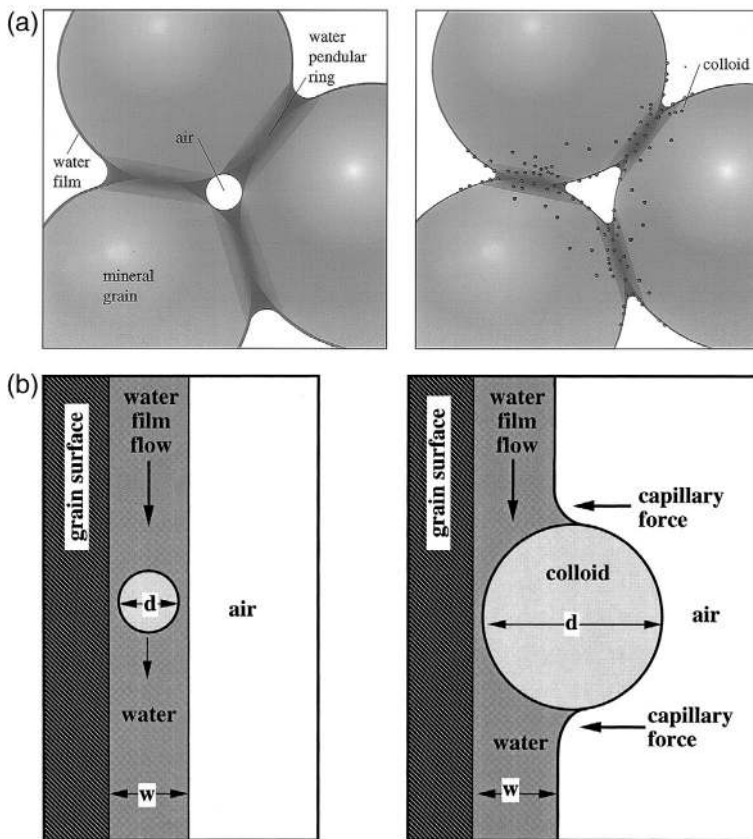
Film straining

Film straining is caused by either disconnected pendular rings or very thin water films (Wan & Tokunaga, 1997; Figure 3a). A water film is a thin layer of water covering a soil grain surface that is retained on the grain surface by adsorption (Tokunaga, 2011; Figure 3a). A pendular ring forms at the contact of two-grain particles by capillarity. When the local saturation is below a critical threshold, a pendular ring can become disconnected from adjacent pendular rings (Wan & Tokunaga, 1997; Figure 3a). When the local saturation is above the critical saturation, pendular rings are connected by water films to neighboring pendular rings. The colloids entrapped in a disconnected pendular ring cannot move to other regions (Gao et al., 2006; Wan & Tokunaga, 1997). Water films expand/thicken during imbibition and shrink/thin during drainage (Auset et al., 2005; Cheng & Saiers, 2010; Gao et al., 2006). When the film thickness is less than the hydrodynamic diameter of the colloid particle, the colloid particle is film strained due to surface tension (also known as capillary force; Lazouskaya et al., 2013; Figure 3b). Surface tension has also been identified as the dominant force responsible for colloid mobilization (Lazouskaya et al., 2013). When the water film expands, the colloid particle can be released from the soil grain surface due to surface tension (Lazouskaya et al., 2013; Sharma, Flury, & Zhou, 2008). When the water film is thick enough to connect the adjacent pendular rings, the colloids entrapped in the pendular rings can move again (Wan & Tokunaga, 1997). Overall, more colloids are deposited as the water content decreases, the grain size decreases and the colloid size increases, creating the characteristic hyperexponential colloid deposition profiles (Wan & Tokunaga, 1997).

Air–water interfaces

An air–water interface is the interface of the two immiscible fluids—air and water—in soil pores (Auset et al., 2005; Cheng & Saiers, 2010; Gao et al., 2006; Powelson & Mills, 2001; Zhuang et al., 2007). In unsaturated flow systems, as the water content of the porous medium decreases, the average size of water-filled pores decreases, while the air–water interfacial area tends to increase (Bradford & Torkzaban, 2008). Changes in the water content of the porous media have been found to increase interfacial sorption of colloids onto AWIs (Saiers & Lenhart, 2003b). Once colloids are attached to air bubble surfaces, they remain attached (Gao et al., 2006). Transient flow cannot detach colloids from air bubble surfaces under experimental conditions (Gao et al., 2006; Figure 1c). When water invades the pore space and the air

FIGURE 3 Illustration of film straining mechanisms. Illustration of pendular rings and water films, and colloids retained in disconnected pendular rings and thin water films (a) (Reprinted with permission from Wan & Tokunaga (1997). Copyright 1997 American Chemical Society) and illustration of the thin water film straining mechanism (b) (Reprinted with permission from Wan & Tokunaga (1997). Copyright 1997 American Chemical Society)

bubble dissolves, the colloids attached to the air bubble can form a cluster (Sirivithayapakorn & Keller, 2003b). Colloids attached to the AWIs of pendular rings (i.e., capillarity retained corner water ducts around the contact of two soil particles) can be released when water invades the pore space and the AWI configuration changes (Gao et al., 2006; Keller & Auset, 2007; Wan & Tokunaga, 1997) (Figures 2d and 3a). Some research suggests that colloids preferentially attach to AWIs relative to SWIs in transient flow systems (Wan & Wilson, 1994a).

Air–water–solid contact lines

An air–water–solid (AWS) contact line, or a triple-phase contact (TPC), is the contact line of a solid–water interface and an air–water interface (Crist et al., 2004; Powelson & Mills, 2001). During drainage, the AWS contact lines scour the solid surfaces and release the attached colloids into suspension. During imbibition, suspended colloids in the newly added soil water attach to available attachment sites on solid surfaces, which were scoured by the AWS contact lines during the previous drainage (Powelson & Mills, 2001). Some researchers demonstrated that trapping at or near the AWS contact lines, rather than the previously suggested attachment to air–water interfaces (AWIs), is a main colloid retention mechanism in unsaturated soil (Crist et al., 2004; Gao et al., 2008; Powelson & Mills, 2001).

Redeposition

Redeposition refers to a deposited colloid particle mobilized at a certain location, which is then subsequently deposited at a deeper location (Cheng & Saiers, 2010). By comparing short and long soil columns, it was found that colloids mobilized during imbibition were more susceptible to redeposition at a deeper location than during drainage (Cheng & Saiers, 2010). Mohanty, Bulicek, et al. (2015) found that because of less chances to collide with a grain particle, less colloids redeposit in macropores.

Effect of ionic strength

High ionic strength (≥ 1 mM) can reduce the electrical double layer repulsion between the negatively charged colloids and like-charged AWIs or soil grain surfaces (Mitropoulou et al., 2013; Zhuang et al., 2007). Several studies have found that higher ionic strength leads to higher colloid attachment to AWIs and SWIs and lower colloid mobility (Mitropoulou et al., 2013; Shein & Devin, 2007; Zhuang et al., 2007, 2009). However, Magal et al. (2011) observed that

after reaching a critical value (15.8 mM), a further increase in ionic strength would not lead to more retention. At the highest natural ionic strength, that is, Dead Sea brines (7.94 M), about 30% of the colloids were still mobile despite favorable conditions for colloid–surface interaction. Under transient flow conditions, a high enough water content ($\geq$0.38 water saturation) and flow velocity ($\geq$6.6% saturated hydraulic conductivity) can compensate for high ionic strength (up to 100 mM) and mobilize colloids (Zhuang et al., 2009). Moreover, via direct observation, Bradford and Kim (2010) demonstrated that not only the ionic strength but also the ion species and the order and magnitude of the ionic strength sequence matters. For instance, by first using multivalent ions to exchange with the monovalent ions at soil grain and colloid surfaces, and then decreasing the solution ionic strength to 0, they could dramatically enhance colloid mobilization. This was because first the zeta potential of both soil grains and colloids was increased after which the thickness of the electrical double layers was expanded (Bradford & Kim, 2010). In addition, changing both the ionic strength and input colloid concentration can influence colloid retention (Zhang et al., 2010). Zhang et al. (2010), for example, observed that greater colloid input concentrations resulted in increased colloid retention at ionic strength above 0.1 mM, which they attributed to filter ripening at SWIs and AWS contact lines. In contrast, when using deionized water (i.e., 0 mM ionic strength), colloid retention was not affected by input colloid concentration.

Effect of hydrophobicity

Colloid surface hydrophobicity can be characterized by measuring the water contact angle at the junction of a thin layer of dry colloid particles, air, and a water drop (Wan & Wilson, 1994a). Hydrophobic colloids generally have contact angles larger than 90° (Lazouskaya et al., 2013; Wan & Wilson, 1994a). The larger the contact angle, the higher the hydrophobicity (Lazouskaya et al., 2013; Wan & Wilson, 1994a). Colloid surface hydrophobicity enhances colloid attachment to AWIs and SWIs, and hence colloid retention in both saturated and unsaturated porous media (Wan & Wilson, 1994a). In the DLVO theory, the secondary-minimum attachment is a weak reversible association of colloids compared to the irreversible strong primary-minimum attachment (Sang et al., 2013). Using confocal microscopy with sulfate (hydrophobic) and carboxylate-modified (hydrophilic) polystyrene microspheres, Lazouskaya and Jin (2008) evaluated hydrophobic interaction in the calculation of total interaction energy. Through experimental data, they proposed that retention of colloids at AWIs occurs mainly via secondary-minimum retention due to hydrophobic attraction. They further observed that the secondary-minimum retention serves as a means to deliver colloids to AWS contact lines. When ionic strength was low (e.g., in deionized water), hydrophilic colloids did not attach to AWIs; and those that had attached to SWIs, could be mobilized by AWS contact lines (Lazouskaya et al., 2011).

3 | FACTORS AND NUMERICAL APPROACHES FOR COLLOID FATE AND TRANSPORT MODELING

Accurate simulation of the retention and mobilization of colloids in the vadose zone, in particular under transient flow conditions, is currently limited by the lack of basic understanding of colloid retention processes at the interface, collector, pore, and continuum scale. The below summary provides a short overview of our current knowledge of conceptual quantitative and numerical approaches available at the microscale and continuum scale to simulate the fate and transport of colloids in transient flow. We will start with a review of quantitative concepts and models at the interface scale, which describes the interaction energy and hydrodynamic forces and torques that act on colloids near interfaces. Next, we will describe the collector scale models, summarizing the numerical methods available to describe colloid transport and interaction processes on a single solid grain or air bubble collector. The pore scale model section will describe the same process system while considering the role of several collectors and multiple interfaces. Finally, the continuum scale model section will review models predicting colloid breakthrough curves and/or soil retention profiles. For each type, we recommend one or two models that are advanced but also easy to understand and use.

3.1 | Microscale models

3.1.1 | Interface scale models

According to Bradford and Torkzaban (2008), at the interface scale, the transport and retention of colloids depend not only on the force balance of the adhesion force (F_A), capillary force (F_σ, also known as surface tension force), lift force

(negligible), drag force (F_D), and Brownian diffusion force, but also on the torque balance of the adhesive torque and the applied torques (Figure 4). The equations of the above forces and torques are readily available in the comprehensive review by Bradford and Torkzaban (2008).

Recently, Lazouskaya et al. (2013) analyzed the forces and torques acting on colloids at the imbibition and drainage fronts (i.e., AWS contact lines) during colloid mobilization under transient flow, considering the hydrophobic and hydrophilic surface characteristics of the colloids and the media surface. Besides the electrostatic, van der Waals, and hydrophobic forces, they added the born repulsion force in the adhesion force (F_A) calculation. According to Lazouskaya et al. (2013), consideration of Born repulsion in the DLVO calculation results in a finite depth of the primary minimum and a more accurate prediction of the force required for colloid mobilization. As a result, the maximum adhesion force, F_A, can be found as the maximum attractive (negative) force in the DLVO force profile.

Lazouskaya et al. (2013) further adjusted the drag force, F_D, to reflect the varying shear flow conditions during imbibition or drainage (the drag force is smaller for a partially submerged particle):

$$F_D = \pm 1.701 \left(6\pi\mu G r^2\right) \tag{1}$$

where μ is fluid viscosity [$ML^{-1}T^{-1}$], r is colloid radius [L], G is shear rate [LT^{-1}], and the sign indicates drainage ($-$) or imbibition ($+$).

Likewise, as the liquid advances (or recedes) during transient flow, the angle, φ, determining the AWI position on the colloid surface, changes from 180° to 0° (or 0°–180°), and the surface tension force assumes two magnitude maxima if the surface tension force, F^*_σ, is directed away from or directed towards the liquid (Lazouskaya et al., 2013; Figure 4). The surface tension force acts along the contact between the colloid and the AWI. As such, Lazouskaya et al. (2013) proposed to calculate the surface tension force in two directions (F^z_σ and F^y_σ), while also taking the substrate dynamic contact angle, α, into account:

When $\phi < \theta$ (when surface tension force is directed away from the liquid, and maximum occurs at $\phi = \theta/2$):
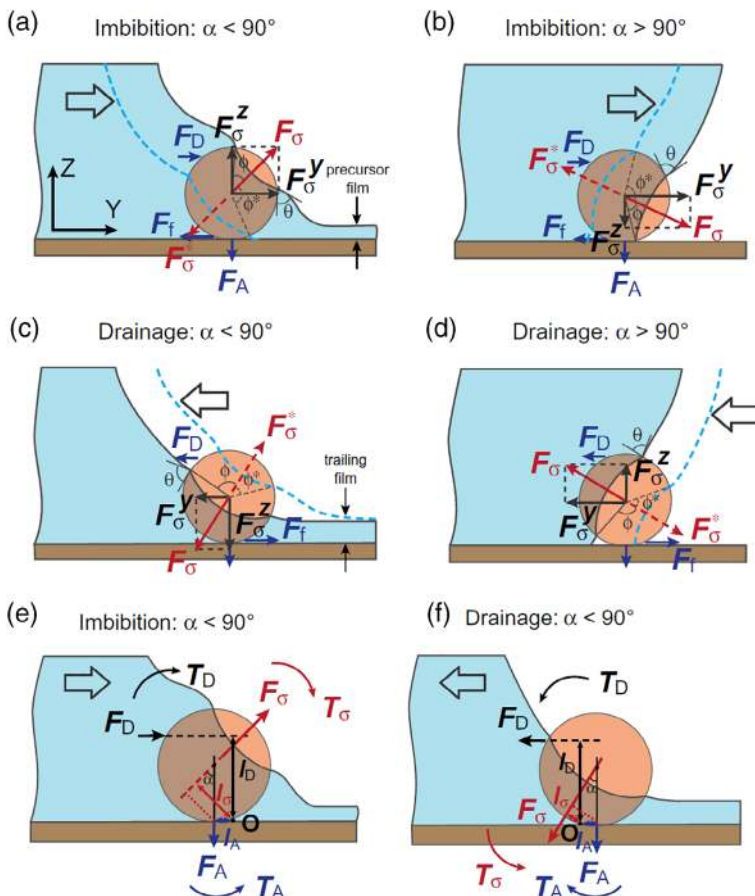
FIGURE 4 Colloids interacting with imbibition (a, b, e) and drainage (c, d, f) fronts on a hydrophilic and a hydrophobic substrate (Reprinted with permission from Lazouskaya et al. (2013), Figures 1 and 2). Two interface positions on the particle (for the two surface tension force directions, that is, $\varphi > h$ and $\varphi < h$) are shown (left and right column). The earlier interface position is represented with the dashed line; only the surface tension force F^*_σ and position angle φ^* are shown for this interface. For the later interface position, the direction and components of the surface tension force and other forces are shown. Force arrows do not represent force magnitudes. Panels e and f show the corresponding torques denoted as $T_D = F_D l_D$, $T_A = F_A l_A$, and $T_r = F_r l_r$. The torques are analogous for a hydrophobic substrate and are not shown. Point O denotes the point of rotation and all torques are calculated with respect to O. The large arrows indicate flow direction

$$F_\sigma^z = 2\pi r \sigma \sin^2(\theta/2)\cos\alpha \tag{2a}$$

$$F_\sigma^y = 2\pi r \sigma \sin^2(\theta/2)\sin\alpha \tag{2b}$$

When $\phi > \theta$ (when surface tension force is directed toward the liquid, and maximum occurs at $\phi = 90° + \theta/2$):

$$F_\sigma^z = -2\pi r \sigma \sin^2(90° + \theta/2)\cos\alpha \tag{2c}$$

$$F_\sigma^y = -2\pi r \sigma \sin^2(90° + \theta/2)\sin\alpha \tag{2d}$$

where ϕ is the angle determining the AWI position on the colloid surface, θ is the dynamic contact angle of the liquid on the colloid surface, σ is the liquid surface tension [MT^{-2}], and α is the substrate dynamic contact angle.

The friction force, F_f, acts at the colloid contact with the substrate and is proportional to the net normal (downward) force, F_N, holding the particle and substrate together (Lazouskaya et al., 2013; Figure 4). For the transient flow conditions shown in Figure 4, the friction force, F_f, is:

$$F_f = \pm\mu_f F_N = \pm\mu_f(F_\sigma^z + F_A), F_\sigma^z + F_A < 0 \tag{3}$$

where μ_f is the coefficient of static friction, F_N is the net normal force [MLT^{-2}]. The sign indicates drainage ($-$) or imbibition ($+$), and F_A has a negative value.

Lazouskaya et al. (2013) were one of the first to consider the mechanisms of colloid mobilization (i.e., lifting, sliding, and rolling) with receding and advancing wetting fronts (corresponding to drainage and imbibition, respectively). Colloid mobilization due to rolling can occur when a net nonzero torque occurs (Figure 4e,f). The torques identified for imbibition and drainage fronts include the resisting torque due to the adhesion force, $T_A = F_A l_A$, and applied torques due to the drag force, $T_D = F_D l_D$, and surface tension forces, $T_\sigma = F_\sigma l_\sigma$.

Lazouskaya et al. (2013) further provided criteria determining whether mobilization due to particle lifting would happen. Lifting happens when $F_\sigma^z + F_A > 0$, while sliding occurs when $F_\sigma^z + F_A \leq 0$ and $|F_\sigma^y + F_D| > |F_f|$. Rolling occurs when $|F_D l_D| + |F_\sigma l_\sigma| > |F_A l_A|$. Otherwise, the colloid remains attached. A detailed analysis of forces and torques under chemically heterogeneous conditions can be found in Bradford, Saeed, and Alexander (2013).

3.1.2 | Collector scale models

At the collector scale, the liquid flow field around a solid grain or an air bubble and the rate of mass (i.e., colloid) transfer to a simple collector surface can be simulated with the Navier–Stokes equation (Bradford & Torkzaban, 2008):

$$\rho\left(\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v}\cdot\nabla\mathbf{v}\right) = -\nabla p + \mu\nabla^2\mathbf{v} + \rho g \tag{4}$$

where ρ is the fluid density [ML^{-3}], v is the velocity vector [LT^{-1}], t is time [T], p is pressure [ML^{-1} T^{-2}], and g is the gravitational acceleration vector [LT^{-2}].

For ideal systems, composed of a spherical collector with a smooth surface and assuming a perfect sink at the collector boundary, the rate of mass transfer to a solid grain or an air bubble can be obtained by solving the advection–diffusion equation:

$$\frac{\partial C}{\partial t} = \nabla\cdot(\mathbf{D}\cdot\nabla C) - \nabla\cdot(\mathbf{v}C) - \nabla\cdot\left(\frac{\mathbf{D}\cdot\mathbf{F}}{k_B T_k}C\right) \tag{5}$$

where C is the aqueous colloid concentration [NL^{-3}; N denotes the number of colloids], D is the colloid diffusion tensor [L^{2}T^{-1}], F is the external force vector [MLT^{-2}], k_B is the Boltzmann constant ($= 1.38 \times 10^{-23}$ JK^{-1}), and T_K is the absolute temperature.

Under unfavorable attachment conditions (i.e., if both the colloid and the collector show surface charge heterogeneity), if the zeta potential distributions of either the collector or the colloid is known or assumed, it is possible to determine the fraction of the surface area that is accessible for attachment for a given solution chemistry when the charge heterogeneity (colloid or collector) is uniformly distributed across the collector surface into N categories. In this case, the fraction of available attachment sites on a grain surface, S_f, can be calculated as the sum of the fractional contribution of each surface charge category of colloids, $f_i S_i$, whereby the charge heterogeneity is uniformly distributed across the collector surface into N categories (Bradford & Torkzaban, 2008).

3.1.3 | Pore-scale models

In contrast to the collector scale, the pore scale is defined by an ensemble of collectors and the presence of multiple interfaces such as SWIs, AWIs, and contact points (e.g., grain-to-grain contacts, solid–water–air triple points) that are mainly defined by the pore space geometry. At the pore scale, the pore structure strongly influences the aqueous flow field, the mass transfer rate, and the forces and torques acting on colloids. Bradford and Torkzaban (2008) provide a detailed overview of the types of retention at the pore scale, but only provide equations for determining the AWI shape (Equation 6) for a given capillary pressure, while Tuller, Or, and Dudley (1999) provided relationships to determine the saturation that corresponds to a given interface curvature:

$$\rho g\psi = \sigma\left(\frac{1}{R_1} + \frac{1}{R_2}\right) \tag{6}$$

where R_1 and R_2 are principal radii of curvature of the interface [L], ψ is matric potential head [L], and g is the gravitational acceleration [LT^{-2}].

In unsaturated conditions, the thickness of the water film around grains is changing depending on water content and capillary pressure. Iwamatsu and Horii (1996) quantified the equilibrium thickness of water films (w [L]) at a given capillary pressure:

$$w = \sqrt[3]{\frac{A_{saw}}{6\pi\rho g\psi}} \tag{7}$$

where A_{saw} is the Hamaker constant for the AWS system [ML^{2}T^{-2}].

Pore-scale models are often a part of a multi-scale model, or provide parameters for continuum scale models (Johnson & Hilpert, 2013). For instance, Smith et al. (2007) quantified the attachment and detachment rates through pore-scale observations under saturated conditions. Recently, Ryan and Tartakovsky (2011) and Hilpert, Rasmuson, and Johnson (2017) developed more advanced pore-scale models that can be upscaled to continuum scale models.

Ryan and Tartakovsky (2011) divided the pore-scale simulation domain into two nonoverlapping subdomains: The pore space subdomain, Ω_A, and the solid grain subdomain, Ω_S. Under saturated conditions, the Ω_A subdomain consists purely of the aqueous phase, at which point colloid transport in the domain can be simulated by solving the Navier–Stokes (Equation 8) and the advection–diffusion (Equation 9) equations, similar to the above collector scale model.

$$-\nabla p + \mu\nabla^2 v + \rho g = 0 \tag{8}$$

$$\frac{DC(\mathbf{x},t)}{Dt} = \nabla\cdot(D(\mathbf{x})\cdot\nabla C(\mathbf{x},t)) \tag{9}$$

where D is the diffusion coefficient [L^{2}T^{-1}], and x is the position vector [L].

The pore-scale model provides the effective diffusion coefficients (D^{IGD} [L^{2}T^{-1}]) for the hybrid model, which is able to simulate colloid transport in connected macropores (Ryan & Tartakovsky, 2011). The hybrid model divides the simulation domain into two nonoverlapping subdomains in a slightly different way: a macropore domain, Ω_F, and a soil matrix domain, Ω_M. Compared to the macropore domain, advection is negligible in the soil matrix domain. Therefore,

the same Navier–Stokes equation (Equation 8) is used to describe the flow field in Ω_F; while a diffusion equation (Equation 10) is used to describe the diffusion in both the macropore and soil matrix domains.

$$\frac{D[\varepsilon(\mathbf{x}) \cdot C(\mathbf{x}, t)]}{Dt} = \nabla \cdot \left(D^{\text{IGD}}(\mathbf{x}) \cdot \varepsilon(\mathbf{x}) \cdot \nabla C(\mathbf{x}, t)\right) \tag{10}$$

where ε is the porosity. In the soil matrix domain, ε is equal to the actual porosity of Ω_M, while in the macropore domain ε is set to 1. D^{IGD} is the effective diffusion coefficient obtained by the pore-scale model in Ω_M, and the molecular diffusion coefficient in Ω_F.

More recently, Hilpert et al. (2017) conceptualized the porous media as a series of unit cells. A unit cell is composed of a grain and a pore, and it contains a near-surface fluid domain (NSFD), where the distance between the colloid and the grain surface corresponds to the secondary energy minimum. A colloid can enter a unit cell through the bulk pore water (State p), or through the NSFD of the upstream unit cell (State n). A colloid can also overcome the energy barrier and attach to a grain surface via the primary energy minimum (State a). Hilpert et al. (2017) assumed that a colloid trajectory is a Markov chain, that is, the next state depends on the current state but not on previous states. The transition from one state to another has a certain probability and takes a certain amount of time. In Equations 11a and 11b below, each of the p_{pa}, p_{pp}, p_{pn}, p_{na}, p_{np}, p_{nn} denotes the transition probability, p, from the first state to the second. Accordingly, each of the t_{pa}, t_{pp}, t_{pn}, t_{na}, t_{np}, t_{nn} denotes the required time to transit from the first state to the second.

Assuming a chain of a total number of N_{tot} unit cells, the pore-scale model can be upscaled to a continuum scale model. A colloid moving through the chain experiences the same total number of p and n states (i.e., $N_p + N_n = N_{tot}$). The number of times the colloid transitions from state p to state n along the chain is denoted as N_c. Then, the continuum scale residence time of the colloid can be described as:

$$t^{(p)}(N_{\text{tot}}, N_n, N_c) = (N_p - N_c - 1)t_{pp} + N_c t_{pn} + (N_n - N_c)t_{nn} + N_c t_{np} \tag{11a}$$

$$t^{(n)}(N_{\text{tot}}, N_n, N_c) = (N_p - N_c)t_{pp} + N_c t_{pn} + (N_n - N_c)t_{nn} + (N_c - 1)t_{np} \tag{11b}$$

where $t^{(P)}$ and $t^{(n)}$ means the trajectory ends in state p and state n, respectively. According to Hilpert et al. (2017), application of the model is not limited to systems in which the immobilization of colloids is due to irreversible attachment in the primary energy minimum. Other mechanisms such as straining can also be simulated with the model. The Hilpert model is one of the first models that explicitly consider the longitudinal colloid transport in the near-surface fluid domain, in particular processes such as the grain-to-grain transport of colloids and the re-entrainment of colloids into the bulk fluid domain via diffusion or via expulsion at the rear-flow stagnation zone. All of these processes are important to explain the residence time of colloids at the column scale, specifically the extended tailing (Schijven, Hoogenboezem, Hassanizadeh, & Peters, 1999), and the hyper-exponential and nonmonotonic retention profiles observed (Bolster, Haznedaroglu, & Walker, 2009; Molnar, Johnson, Gerhard, Willson, & O'Carroll, 2015). In spite of the recent advances made in multi-scale models, pore-scale models for unsaturated or transient flow conditions are still largely lacking.

3.2 | Continuum scale models

To study colloid transport under controlled but nearly natural conditions, continuum scale models that can predict colloid concentration in discharge or the soil profile are most useful. Most macroscale models for colloid transport have only been validated under steady-state flow conditions, and only few models were derived and have been validated under transient flow conditions (Chen, Liu, Tawfiq, Yang, & Banks, 2009; Denovio et al., 2004; Engstrom, Thunvik, Kulabako, & Balfors, 2015; Flury & Qiu, 2008). In this review, we introduce the macroscale models available in the HYDRUS software package (Šimůnek, van Genuchten, & Šejna, 2008) that can simulate colloid transport under transient flow conditions. We further present two other macroscale models that were particularly derived for and validated with colloid transport experiments under transient flow conditions. For simplicity, in this review, we neglect models that account for inactivation and degradation of microorganisms (i.e., bio-colloids), and only focus on the transport processes of nonbiological colloids. But, for most models, inactivation and degradation of microorganisms can be included by simply adding a sink term.

3.2.1 | Models available in HYDRUS

The finite element model HYDRUS (Šimůnek et al., 2008) offers an attachment-detachment model or two-site kinetic sorption model for colloid transport:

$$\frac{\partial \theta c}{\partial t} + \rho \frac{\partial s_e}{\partial t} + \rho \frac{\partial s_1}{\partial t} + \rho \frac{\partial s_2}{\partial t} = \lambda_L \theta v \frac{\partial^2 C}{\partial x^2} - \theta v \frac{\partial C}{\partial x} \tag{12}$$

$$\rho \frac{\partial s_1}{\partial t} = \theta k_{a1} \psi c - k_{d1} \rho s \tag{13}$$

$$\rho \frac{\partial s_2}{\partial t} = \theta k_{a2} \psi c - k_{d2} \rho s \tag{14}$$

where c is the colloid concentration in the aqueous phase [NL^{-3}], s is the concentration of colloids at kinetic sorption sites [NL^{-3}], v (=q/θ) is the average interstitial water velocity [LT^{-1}], k_a and k_d are first-order attachment and detachment rate coefficients, respectively [T^{-1}], subscripts e, 1, and 2 refer to the equilibrium and two kinetic sorption sites, and ψ is a dimensionless colloid retention function. The colloid transport model assumes that for the two kinetic sorption sites, the attachment and detachment follow first-order kinetic processes.

Note that this model only allows simulation of colloid transport, not colloid-facilitated solute transport which is implemented in the C-Ride module (Šimůnek, He, Pang, & Bradford, 2006). The full version of the model uses two kinetic sorption sites to account for two different processes such as straining and physical–chemical filtration, or attachment to AWIs and SWIs. But, one or two of the $\rho \frac{\partial s}{\partial t}$ terms can be dropped based on the simulated system, and the model can be easily reduced to a one-site kinetic sorption model and equilibrium sorption model, respectively.

The attachment rate coefficient, k_a, in the model can be calculated with the filtration theory model (Bradford, Yates, Bettahar, & Simunek, 2002; Logan et al., 1995; Rajagopalan & Tien, 1976), which is able to account for colloid size and the efficiency of different physical–chemical filtration mechanisms (e.g., Brownian diffusion, interception, and gravitational sedimentation; Bradford et al., 2002):

$$k_a = \frac{3(1-\theta)}{2d_c} \eta \alpha v \tag{15}$$

where d_c is the diameter of the soil particles [L], α is the sticking efficiency (the ratio of the rate of colloids sticking to a collector and the rate at which they collide with the collector), v is the pore water velocity [LT^{-1}], and η is the single-collector efficiency, which composed of three terms, representing colloid removal from the aqueous phase by diffusion, interception and gravitational sedimentation, respectively (see Bradford et al., 2002 for details).

The dimensionless colloid retention function, ψ, can account for colloid clogging or filter ripening, that is, the process that earlier attached colloids could influence the attachment of later colloids, and it has several different formats (see Adamczyk, Siwek, Zembala, & Belouschek, 1994; Bradford & Bettahar, 2006; Bradford et al., 2003; Johnson & Elimelech, 1996; and Wang et al., 2012 for details).

The attachment-detachment model (i.e., the two-site kinetic sorption model or its reduced formats—the one-site kinetic sorption model or the equilibrium sorption model) has wide application in colloid transport under steady-state conditions. For instance, using one site for straining and the other site for attachment, Foppen et al. (2007) successfully simulated the straining and attachment of *Escherichia coli* in saturated packed sand columns of various grain sizes, column lengths, injected colloid concentrations, and flow rates. They used Equation 15 to calculate the attachment rate coefficient, k_a, and used the Langmuir format of ψ suggested by Adamczyk et al. (1994) to describe the filter-ripening phenomenon. Using one site for reversible attachment–detachment and the other site for irreversible straining and favorable deposition, Cornelis, Pang, Doolette, Kirby, and Mclaughlin (2013) were able to capture the transport and retention of silver nanoparticles in saturated natural soils of various physical and chemical properties very well. Using one attachment–detachment site and the Langmuir format of ψ, Balkhair (2016) successfully described the transport and retention of fecal bacteria in both saturated and partially saturated soils of different physical and chemical properties. Using the one-site kinetic sorption model and the Bradford and Bettahar (2006) format of ψ, Rahmatpour,

Mosaddeghi, Shirvani, and Šimůnek (2018) simulated the transport and retention of silver nanoparticles in both saturated and unsaturated undisturbed calcareous soils of different textures.

However, testing of the attachment–detachment model with experimental data under transient flow conditions has been very limited. The only example we could find was the study of Schijven and Šimůnek (2002). Using the two-dimensional two-site kinetic sorption model, they successfully captured the measured transport and retention of virus colloids under transient flow conditions in a dune recharge field experiment conducted by Schijven et al. (1999) and in a deep well injection field experiment conducted by Schijven, Medema, Vogelaar, and Hassanizadeh (2000). More studies such as the ones listed here are needed to test the applicability of the attachment–detachment model to predict colloid transport under transient flow conditions.

In addition, the equations used for solute transport in the dual-porosity models (Philip, 1968; Šimůnek, 2008; Šimůnek, Jarvis, van Genuchten, & Gärdenäs, 2003; van Genuchten, 1981; van Genuchten & Wagenet, 1989; van Genuchten & Wierenga, 1976) and dual permeability models (Gerke & van Genuchten, 1993, 1996, 2010; Pot et al., 2005; Šimůnek, 2008; Šimůnek et al., 2003; van Genuchten & Dalton, 1986) in HYDRUS, although not explicitly written for colloid transport, could be adjusted to simulate colloid transport through soil with the existence of preferential flow.

More recently, Bradford, Wang, Torkzaban, and Šimůnek (2015) developed a model (Bradford model) that explicitly considers colloid attachment to SWIs and colloid mobilization by moving AWI or AWS contact lines in transient flow, especially during drainage and imbibition cycles. Detailed equations of the mathematical model can be found in Bradford et al. (2015).

In the Bradford model, variably saturated water flow is described using Richards' equation and colloid transport in the aqueous phase is described using the advection–dispersion equation with two additional terms that allow quantification of the exchange of colloids between the aqueous phase and SWIs and AWIs, respectively (Bradford et al., 2015). Furthermore, the model allows estimating the mass balance for colloids retained and released from SWI and AWIs. Retention and release of colloids from solid–water interfaces are estimated using a retention rate coefficient to the SWI and a steady-state release rate coefficient as well as a colloid release term that partitions colloids from the SWI to the AWI. Colloid retention and release from air–water interfaces is estimated by simulating the changes in the area of AWIs. During drainage, the area of AWIs increases and a fraction of the colloids attached to the SWIs are partitioned to the aqueous phase and the AWIs or AWS contact lines. During imbibition, the model simulates the decrease in area or the destruction of AWIs, and the partitioning of colloids from the AWIs or the AWS contact lines to the mobile water.

The Bradford model was successfully used to simulate *E. coli* release during drainage and imbibition cycles when saturation and AWIs configuration varied (experimental results from Wang, Bradford, and Šimůnek (2015)). According to Bradford et al. (2015) the developed equilibrium, kinetic, and combined equilibrium and kinetic models provided a reasonable description of the colloid release behavior for various changes in the fraction of the solid surface area that contributes to colloid retention when parameters were optimized to the experimental data. Experimental data further indicated that the microscopic surface roughness and grain-to-grain contacts played a larger role in *E. coli* retention, even when the primary minimum and secondary minimum were eliminated by a decrease in solution ionic strength. Further, temporal changes in water velocity were found to not have a large influence on bacteria release, which they again attributed to the microscopic surface roughness and grain-to-grain contacts that decreased the hydrodynamic torque. The largest weakness of the Bradford model is the complexity of the model, which requires a precise definition of the initial conditions, the removal efficiency, and the specific transient conditions of the release model, for which often only limited a priori information exists. The Bradford model is available in HYDRUS-1D, which will promote its wider application in the coming years.

3.2.2 | Other continuum scale models

One of the numerical models that couples transient pore water flow with the mobilization, transport, and redeposition of colloids in unsaturated porous media is the Saiers–Cheng–Zhang model. Saiers and Lenhart (2003a) couple the advection–dispersion equation with an equation that estimates the temporal change in the total concentration of immobile colloids (Equation 16). This is done by discretizing the immobile-phase colloid population into compartments, each of which is characterized by its critical (θ_{cri}) and actual (θ) moisture content (Equations 19a, 19b), which determines the kinetics of colloid release. The critical moisture content distribution is described by a piecewise linear density

function. Only when the moisture content in a compartment exceeds its critical moisture content, colloids in that compartment can be mobilized.

Colloid transport and mass transfer:

$$\frac{\partial(\theta C)}{\partial t} + \rho_b \frac{\partial S_T}{\partial t} = \frac{\partial}{\partial z}\left[D\theta \frac{\partial C}{\partial z} \right] - \frac{\partial(qC)}{\partial z} \tag{16}$$

$$\frac{\partial S_T}{\partial t} = \sum_{i=1}^{NC} \frac{\partial S_i}{\partial t} \tag{17}$$

$$\frac{\partial S_i}{\partial t} = \frac{\theta}{\rho_b} \frac{1}{NC} k_f C - \alpha_i S_i \tag{18}$$

$$\alpha_i = 0 \text{ for } \theta < \theta_{cri} \tag{19a}$$

$$\alpha_i = N v^\kappa \text{ for } \theta \geq \theta_{cri} \tag{19b}$$

where S_T is the total concentration of immobile colloids [MM^{-1}, mass of colloids per unit mass of sand], and D is the hydrodynamic dispersion coefficient [$L^2 T^{-1}$], NC is the number of compartments, S_i is the concentration of colloids held within the ith compartment [MM^{-1}, mass of colloids per unit mass of liquid], k_f is a rate coefficient for colloid removal by straining [T^{-1}] and it can be quantified by an empirical relationship depending on volumetric moisture content and average pore water velocity, α_i is a rate coefficient for colloid release from compartment i [T^{-1}], and θ_{cri} is the critical moisture content associated with compartment I, and k and N [$L^{-k}T^{k-1}$] are empirical coefficients quantifying the kinetics of colloid release.

The Saiers and Lenhart model was successfully applied to capture the colloid mobilization in packed sand during a series of imbibition events (Saiers & Lenhart, 2003a). And, a more detailed pore water velocity function may further improve the accuracy. After all, the model provides a fundamental framework to simulate real-world variably saturated conditions.

Cheng and Saiers (2009) modified the Saiers–Lenhart model to account for colloid mobilization during drainage and imbibition and to quantify the colloid mobilization kinetics according to the porous medium water retention characteristics (Saiers–Cheng model). The main difference to the Saiers–Lenhart model is that Cheng and Saiers (2009) compared the pressure head (Ψ) with the snap off pressure head of compartment i (Ψ_{S_i} [L]), instead of comparing the volumetric water content (θ) with the critical moisture content of compartment i (θ_{cri}). According to Cheng and Saiers (2009), pore snap off occurs in pores that fill with water during imbibition or empty during drainage as a result of changes in pressure head during transient flow. Changes in pressure head are not only dependent on water content but also on pore size and since pore sizes in any porous medium are nonuniform, the model assumes a distribution in snap off pressures and subsequently a distributed colloid mobilization response (α_i [T^{-1}]) that is different for drainage (Equations 20a, 20b) and imbibition (Equations 20c, 20d).

During drainage and before the next imbibition:

$$\alpha_i = 0 \text{ for } \Psi > \Psi_{S_i} \tag{20a}$$
$$\alpha_i = N_d \left| \frac{\partial \theta}{\partial t} \right| \text{ for } \Psi \leq \Psi_{S_i} \tag{20b}$$

During imbibition:

$$\alpha_i = 0 \text{ for } \Psi < \Psi_{S_i} \tag{20c}$$
$$\alpha_i = N_{imb} \frac{\partial \theta}{\partial t} \text{ for } \Psi \geq \Psi_{S_i} \tag{20d}$$

where α_i is the coefficient that quantifies the rate of colloid mobilization from the ith compartment and N_d and N_{imb} are empirical coefficients quantifying the kinetics of colloid mobilization during a drainage event and an imbibition event, respectively. The Saiers–Cheng model captured well in situ colloid mobilization and transport in packed sand during both drainage and imbibition events at various ionic strength and flow rates (Cheng & Saiers, 2009). Both the experimental results and the theoretical simulation demonstrate that the moving drainage and imbibition fronts (AWS contact lines at microscale) could substantially mobilize colloids.

Zhang et al. (2012) further extended the Saiers–Cheng model by regarding the attachment to and detachment from AWIs as an equilibrium sorption process. The size of the available AWI for colloids to attach to is formulated to be linearly dependent on saturation (Saiers–Cheng–Zhang model):

$$\frac{\partial \theta R C_{\mathrm{w}}}{\partial t} = \frac{\partial}{\partial x}\left(\theta D \frac{\partial C_{\mathrm{w}}}{\partial x}\right) - \frac{\partial q C_{\mathrm{w}}}{\partial x} - \theta k_{\mathrm{att}}^{s} C_{\mathrm{w}} + \rho_{\mathrm{b}} k_{\mathrm{det}}^{s} C_{\mathrm{s}} \tag{21}$$

$$R = 1 + \frac{A k_{D}^{a}}{\theta} \tag{22}$$

$$K_{D}^{a} = \theta k_{\mathrm{att}}^{a} / A k_{\mathrm{det}}^{a} \tag{23}$$

$$A = A_{o}(1 - S_{\mathrm{w}})^{3} S_{w} \tag{24}$$

where C_{w} [NL^{-3}] is the colloid concentration in water, C_{aw} [NL^{-3}] is the concentration of colloids attached to the AWI in terms of number of colloids per unit air–water interfacial area, A [L^{2}L^{-3}] is the air–water interfacial area per unit volume of porous medium, q [LT^{-1}] is the Darcy-Buckingham flow rate, $k_{\mathrm{att}}{}^{s}$ and $k_{\mathrm{det}}{}^{s}$ [T^{-1}] are attachment and detachment rate coefficients, respectively, of colloids to and from the SWI, $k_{\mathrm{att}}{}^{a}$ and $k_{\mathrm{det}}{}^{a}$ are similar attachment and detachment rate coefficients, respectively, associated with the AWI, $K_{\mathrm{D}}{}^{a}$ [L] is the equilibrium distribution coefficient for attachment to the AWI, and S_{w} is the degree of fluid saturation.

The Saiers–Cheng–Zhang model successfully captured observed virus transport behaviors under both saturated and unsaturated conditions and during both drainage and imbibition periods (experimental results from Torkzaban, Hassanizadeh, Schijven, Bruijn, and Husman (2006) and Torkzaban, Hassanizadeh, Schijven, and van den Berg (2006); Zhang et al., 2012). When comparing the saturation-dependent attachment coefficient to a constant–attachment model simulation, they found that the constant-attachment model simulation performed surprisingly well, which they attributed to the large values of the transient detachment coefficients that might have obscured any variation. Zhang et al. (2012) further point out that the ability of the model to simulate virus transport through porous media needs to be further tested for undisturbed, and medium- and fine-textured soils.

Majdalani et al. (2007) developed a model (Majdalani model) to simulate colloid transport through structured soil. The preferential water flow is described by a kinematic–dispersive wave model, and colloid detachment from macropore walls is simulated by first-order kinetics. One novelty that the Majdalani model has, is that they introduce a detachment efficiency term that captures the dynamic nature of the activation/deactivation of colloids in the porous media due to detachment, clogging, and particle filtration depending on changes in water content and ionic strength. The frequency and amount of colloid detachment are made dependent on the cumulative mass removed from a particular location in the porous medium. Because particles cannot infinitely be released into the bulk fluid flow, as particle stocks get exhausted over time. Further, the detachment efficiency is characterized by hysteresis, instead of the commonly used constant detachment efficiency.

The preferential water flow in macropores (noncapillary pores) is simulated by a nonlinear kinematic dispersive wave model developed by Pietro, Ruy, and Capowiez (2003):

$$\frac{\partial u(z,t)}{\partial t} + c[u(z,t)]\frac{\partial u(z,t)}{\partial z} = v[u(z,t)]\frac{\partial^{2} u(z,t)}{\partial z^{2}} \tag{25}$$

where $u(z, t)$ is the water flux at instant t and depth z, $c[u(z,t)]$ and $v[u(z,t)]$ are, respectively, the convective celerity and the dispersivity distorting the advancing water front.

The colloid transport in macropores is described by a convection–dispersion equation with two source (S_1 and S_2) and one sink term (P), which represent the colloid detachment and colloid deposition, respectively.

$$\frac{\partial(\theta_{\mathrm{m}}(z,t)C(z,t))}{\partial t} = -\frac{\partial(C(z,t)u(z,t))}{\partial z} + \frac{\partial}{\partial z}\left(\theta_{\mathrm{m}}(z,t)D(z,t)\frac{\partial C(z,t)}{\partial z}\right) + S - P \tag{26}$$

$$P = k_{\mathrm{att}}\theta_{\mathrm{m}}(z,t)C(z,t) \tag{27}$$

$$S_{1} = k_{\mathrm{acc}}\frac{\partial u(z,t)}{\partial t} \text{ with } \left\{\begin{array}{l} k_{\mathrm{acc}} > 0, \quad \text{if } t \leq t_{\mathrm{s}} \\ k_{\mathrm{acc}} = 0, \quad \text{if } t > t_{\mathrm{s}} \end{array}\right\} \tag{28}$$

$$S_2 = k_{\text{det}} E(z,t) \theta_{\text{m}}(z,t) u(z,t) C^*(z,t) \qquad (29)$$

where $C(z, t)$ [ML^{-3}] is the mass concentration of colloids in the flowing solution at depth z and time t, $D(z, t)$ [L^{2}T^{-1}] is the particle dispersion coefficient, S [ML^{-3} T^{-1}] is the source term that accounts for colloid detachment, P [ML^{-3} T^{-1}] is the sink term that describes colloid deposition along macropore walls, k_{acc} [M V^{-1} T L^{-1}] is a flux acceleration coefficient, k_{det} [L^{-1}] is the particle detachment coefficient, and $C^*(z, t)$ [M V^{-1}] is the crust particle concentration.

The Majdalani model performed well when simulating the mobilization and transport of in situ soil particles in unsaturated undisturbed soils, especially those with macropores (experimental results from Rousseau et al. (2004)). During model validation, Majdalani et al. (2007) noted that the attachment and detachment coefficients (k_{att}, k_{det}) were mainly influenced by the nature of the soil, incoming solution ionic strength, and rainfall intensity, while the water flux acceleration coefficient (k_{acc}), as well as the shape of the cumulative particle detachment curve over time (determined through α and β) depend more on the macropore network structure and column history. In later studies, Majdalani et al. (2007) showed that wetting/drying cycles in unsaturated porous media generate erosive-like releases of colloids that increase with increasing drying period length. Mohanty, Saiers, and Ryan (2015) note that this mechanism was more important for colloid release than ionic strength or rainfall intensity.

4 | COMMENTS AND FUTURE RESEARCH NEEDS

Although colloid transport has been heavily investigated over the last decades, our understanding of colloid transport under transient flow remains limited. Specifically, a clear understanding of retention and mobilization mechanisms, controlling factors and their interaction during imbibition and drainage is needed to accurately characterize and predict colloid transport in transient flow systems. In this section, we summarize our comments and vision for this field after reviewing the experimental needs, new technologies available, and modeling advances made in the last few years.

4.1 | Experimental studies

In spite of the many experimental studies conducted so far, processes of colloid transport during the transitional stages of transient flow, when specific discharge and volumetric moisture content abruptly increase or decrease, remain still incompletely understood and quantified. During rewetting of the soil, for example, the conversion of immobile water to mobile water contributes to the release of colloids from immobile zones into the bulk fluid flow, a process that last on the order of seconds and therefore is difficult to observe and quantify (Gao et al., 2006). Other studies suggest that the colloid release behavior changes during repeated cycles of drainage and imbibition. For example, colloid concentration on the AWI may increase during imbibition due to retention of colloids released from SWIs; during rewetting, air entrapment, preventing full saturation of the porous media, may prevent complete release of colloids from AWIs (Bradford et al., 2015). These effects are expected to be even more pronounced for hydrophobic and/or larger colloids that have a propensity to more readily attach to AWIs, and for colloid transport in finer textured soils that have a larger interfacial area (Bradford et al., 2015; Bradford & Leij, 1997). More recently, Zhou et al. (2019) pointed out that most transient flow studies (particularly those studying wetting-drying cycles) used very simplified experimental designs, such as ideal porous media (e.g., sand, sand quartz), that present a gross oversimplification of the dynamic, heterogeneous conditions found in most subsurface environments. In order to more accurately capture colloid transport mechanisms in transient flow systems, they postulate for experimental designs where physical and chemical conditions vary with time and space reflecting the vertical variations and horizontal variations in the composition of soil porous media due to rainwater and evaporation (Mohanty et al., 2016; Shang et al., 2008).

As indicated in our review, although existing experiments at the column scale provided valuable insights into colloid transport through soil under transient flow conditions, they provide only limited information about the processes involved, especially if limited to breakthrough curve analysis only. While important for understanding the role of scale and soil structure, there is a need to augment column scale studies with more intricate theoretical and experimental pore-scale studies to more precisely determine the underlying processes responsible for colloid detachment, attachment, and remobilization during transient imbibition and drainage periods. Unfortunately, to date, there exist only a few studies that quantified nanoscale physical and chemical parameters for developing these theoretical approaches

(e.g., Lazouskaya et al., 2013; Mohanty, Saiers, & Ryan, 2015; Zhou et al., 2019), hence more research is needed to overcome these data limitations. In particular, more experiments are needed to test the behavior of colloids of different sizes and surface properties, from different source locations (e.g., autochthonous, injected), and of different ages in the system to better understand colloid behavior in the natural vadose zone and its underlying transport processes. With respect to colloid fate and transport mechanisms, the role of colloid hydrophobicity, transient changes in water chemistry, soil texture, soil structure, and frequency and duration of wetting/drying cycles and associated imbibition/drainage dynamics on the fate and transport of different-sized colloids in natural soil systems is needed.

4.2 | New technologies

To properly track transport pathways of colloids and attached contaminants in the environment without the risk of infection or field contamination, nontoxic particle tracers should mimic as closely as possible the transport behavior of the particle suspension(s) of concern. To simultaneously address the issues of limited tracer varieties and unrepresentativeness for suspensions, novel synthetic-free DNA tracers and DNA-labeled particle tracers (Dahlke et al., 2015; Liao, Yang, Wu, Luo, & Yang, 2018; McNew, Wang, Walter, & Dahlke, 2018; Sharma, Luo, & Walter, 2012; Wang et al., 2019) could potentially provide a new experimental tool. DNA-labeled particle tracers deliver an unlimited number of distinguishable colloids of desired sizes and surface properties that could be used to investigate the transport pathways of colloids from different source locations that entered the transient flow soil system at different wetting–drying stages (Figure 5).

New visualization techniques, such as confocal microscopy (e.g., spectral or laser scanning) (Dathe et al., 2014; Gao et al., 2006; Sang et al., 2013; Smith et al., 2007; Zevi et al., 2005, 2009, 2012; Zevi, Dathe, Gao, Richards, & Steenhuis, 2006), epi-fluorescent microscopy (Auset & Keller, 2006; Baumann & Werth, 2004; Gao et al., 2006), and X-ray microtomography (Li, Lin, Miller, & Johnson, 2006) have greatly aided colloid transport research over the past two decades. At the column scale, magnetic resonance imaging has provided real-time noninvasive visualization of bacteria transport in saturated porous media (Olson, Ford, Smith, & Fernandev, 2004; Sherwood et al., 2003). Synchrotron radiation X-ray fluorescence has been used to measure the in situ colloid concentration and moisture content in soil column experiments (DiCarlo et al., 2006). More recently, X-ray computed tomography (CT) has been used to extract macropore networks in soil columns (Carrel et al., 2017, 2018; Soto-Gómez, Pérez-Rodríguez, Vázquez-Juiz, López-Periago, & Paradelo, 2018) allowing to link the characteristics of the extracted macropore networks to the transport characteristics of colloids. Development and wider application of existing as well as new techniques such as the 19-port sampling system by Mohanty, Bulicek, et al. (2015) (Figure 6) to visualize and measure in situ moisture content and colloid concentration in porous media (Crist et al., 2004; DiCarlo et al., 2006; Keller & Auset, 2007; Soto-Gómez et al., 2018, 2019), and to differentiate different flow pathways and colloid attachment sites (Auset & Keller, 2006; Baumann &
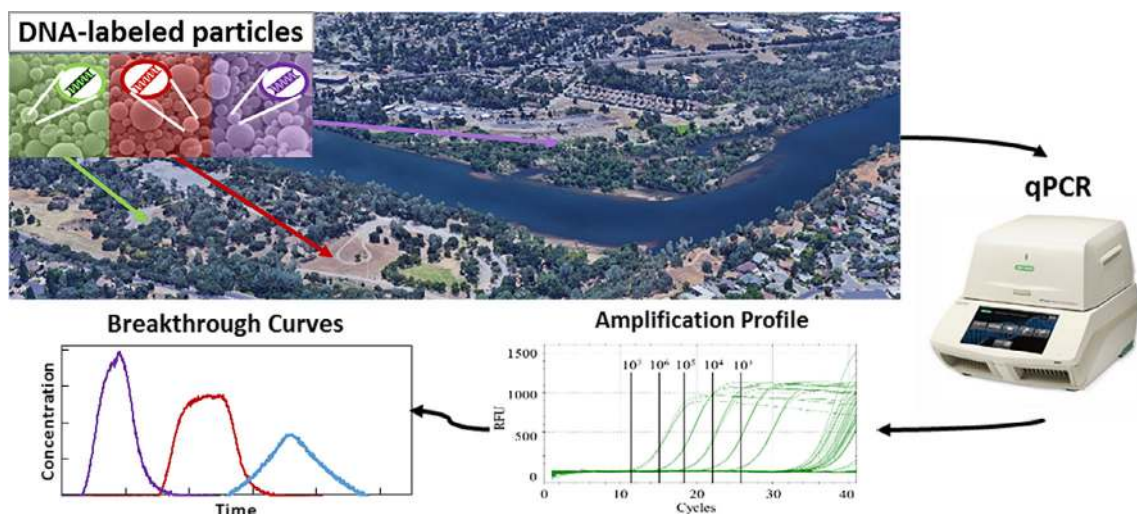
FIGURE 5 Illustration of the novel DNA-labeled particle tracing technique (Reprinted with permission from McNew et al. (2018). Copyright 2018 Elsevier)
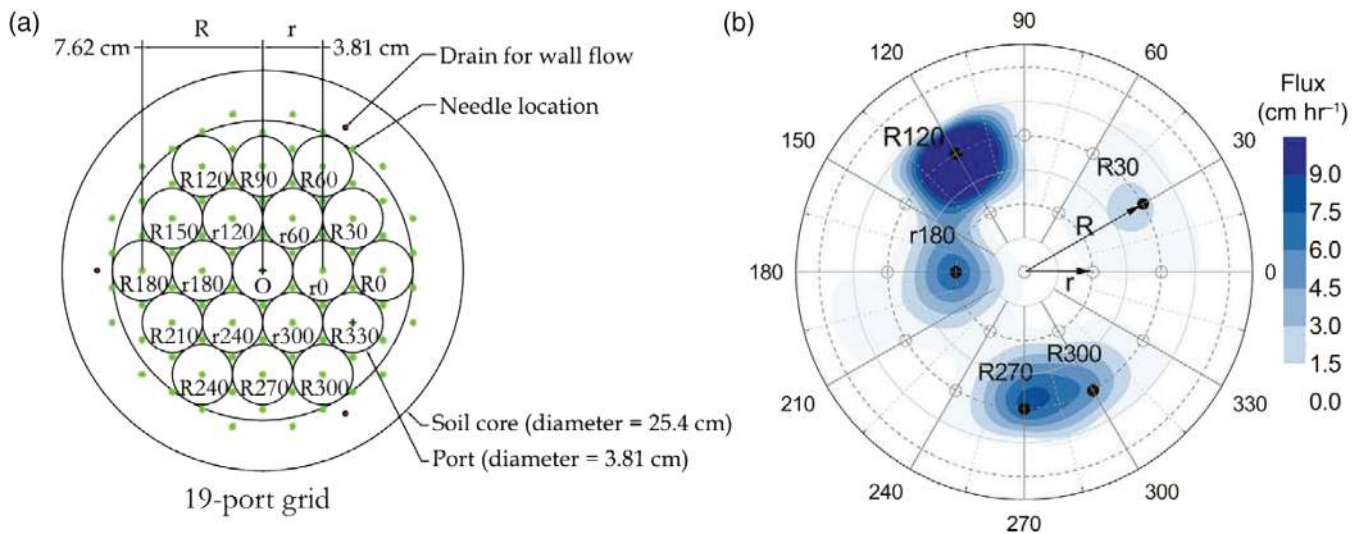
FIGURE 6 The 19-port sampling system at the bottom of an intact/undisturbed soil column to differentiate different flow pathways (a) and the water flux collected by the 19-port sampling system (b) (Reprinted with permission from Mohanty, Bulicek, et al. (2015). Copyright 2015 John Wiley and Sons)

Werth, 2004; Lanning & Ford, 2002; Zhang, Hassanizadeh, Liu, Schijven, & Karadimitriou, 2014) offer promising methods and technologies to advance understanding of the fate and transport of colloids in transient flow systems.

4.3 | Modeling

Microscale models have advanced rapidly after the review provided by Bradford and Torkzaban (2008). Most advances have been made in the areas of interface scale models and upscaling models that link the pore scale and continuum scale. To date, interface scale models are the main ones considering more complicated transient flow conditions, such as AWS movement and transients in solution chemistry. Upscaling of models that link the pore scale and the continuum scale are the future research trend of pore-scale models. However, one of the biggest gaps in microscale and continuum scale modeling continues to be the formulation of colloid transport mechanisms in structured soils and representation of flow conditions in two- or three-dimensions.

Among the continuum scale models, the HYDRUS software package can simulate colloid transport through soil under transient conditions due to (a) its ability to simulate transient water flow, (b) availability of add-on options to modify the attachment rates of the one- or two-site kinetic sorption models, and (c) availability of dual-porosity and dual-permeability models to account for preferential flow. Availability of the Bradford model within HYDRUS explicitly allows simulation of pore-scale colloid transport processes and simulation of colloid transport through packed soil.

Besides the HYDRUS software package, the Saiers–Cheng–Zhang model also simulates colloid transport through packed soil well; whereas, the Majdalani model is best applied to simulate colloid transport through undisturbed soil. These two models include explicit descriptions of physical transport mechanisms, but whether they can be used to broadly simulate colloid transport through soil under different transient conditions needs to be tested with more experimental data. Moreover, they are not readily available to the hydrologic community and public, thus, limiting their use to advance understanding of the fate and transport of colloids in variably saturated systems. Often researchers need to write their own code to implement the models.

Due to the complexity of colloid transport in transient flow, there is a need to more openly share existing source code so that new numerical or analytical solutions of physical colloid transport and retention mechanisms can be added and tested for different soil material configurations (different porous media types, dual permeability system) and water flow transients to advance colloid transport through soil under transient flow conditions. In addition, application of colloid transport models in field studies of natural soil or vadose zone systems remains an enormous challenge. This, in part requires more studies on the importance of colloid transport and retention mechanisms in transient flow systems at different scales (e.g., nano to column, plot, and field scale).

CONFLICT OF INTEREST

The authors have declared no conflicts of interest for this article.

AUTHOR CONTRIBUTIONS

Chaozi Wang: Conceptualization; funding acquisition; investigation; resources; visualization; writing-original draft; writing-review and editing. **Ruoyu Wang:** Conceptualization; investigation; resources; writing-original draft; writing-review and editing. **Zailin Huo:** Conceptualization; funding acquisition; investigation; resources; supervision; writing-original draft; writing-review and editing. **En Xie:** Conceptualization; resources; writing-original draft. **Helen Dahlke:** Conceptualization; funding acquisition; investigation; resources; supervision; visualization; writing-original draft; writing-review and editing.

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