

Reply to comment by William P. Johnson et al. on “Transport and fate of bacteria in porous media: Coupled effects of chemical conditions and pore space geometry”

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Received 7 November 2008; revised 22 July 2009; accepted 7 August 2009; published 25 September 2009.

Citation: Torkzaban, S., S. L. Walker, and S. A. Bradford (2009), Reply to comment by William P. Johnson et al. on “Transport and fate of bacteria in porous media: Coupled effects of chemical conditions and pore space geometry,” *Water Resour. Res.*, 45, W09604, doi:10.1029/2008WR007576.

1. Introduction

[1] We are grateful for this opportunity to address the comment of *Johnson et al.* [2009] on our manuscript. Their comment concerns the nature of colloid association with solid surfaces via the secondary energy minimum. There are currently different opinions in the literature on this topic. *Johnson et al.* [2007] have assumed that colloids associated with collector surfaces via the secondary minimum are “freely mobile” in the presence of fluid drag and can therefore only be immobilized in stagnation regions. In contrast, we have applied a torque balance approach to determine locations where colloid retention is theoretically possible when the driving (hydrodynamic) torque is less than the resisting (adhesive) torque [*Bradford et al.*, 2007; *Torkzaban et al.*, 2007, 2008]. Each of these points of view will be discussed below.

2. Are Secondary Minimum Colloids “Freely Mobile” in the Presence of Fluid Drag?

[2] Colloid attachment on a solid surface is controlled by the summation of external forces and torques that act on colloids at a given location. Traditional approaches have solved the force and torque balance equations to determine the trajectories of colloids in flowing fluid under favorable attachment conditions [*Rajagopalan and Tien*, 1976; *Prieve and Lin*, 1980; *Yang et al.*, 1998]. If the trajectory of the colloid intercepted the collector surface, then the colloid was assumed to be attached. This is likely to be a reasonable assumption under typical groundwater flow conditions when colloids are strongly associated with the solid surface in the primary minimum at a close separation distance [*Bergendahl and Grasso*, 2000; *Torkzaban et al.*, 2007]. In contrast, colloids may be weakly associated with the solid surface via the secondary minimum at a greater separation distance under unfavorable attachment conditions, and only a fraction of colloids that approach the solid surface will be retained [*Ryan and Elimelech*, 1996]. In this

case, the assumption of colloid immobilization in the presence of fluid drag and Brownian forces is not always justifiable [*Simoni et al.*, 1998; *Dong et al.*, 2002; *Torkzaban et al.*, 2007], and greater care must be taken to select the proper force and torque balance equations adjacent to the solid surface.

[3] Traditional trajectory analysis under favorable conditions has implicitly assumed that a frictional force or resisting (adhesive) torque acts to completely immobilize the colloid on the solid surface. Similarly, *Johnson et al.* [2007] have neglected any frictional force or resisting torque due to adhesion under unfavorable attachment conditions. Under this assumption no colloid attachment should occur because the two major forces (adhesive and fluid drag) acting on the colloids are perpendicular and do not cancel each other. Other forces are negligible (lift force is negligible under laminar flow conditions, the gravitational force is negligible when the density of the colloid is close to that of water, and the Brownian force acts in a random direction). Hence, the trajectory analysis of *Johnson et al.* [2007] results in secondary minimum associated colloids that are “freely mobile” in the presence of fluid drag.

[4] Hydrodynamic correction functions have been used in traditional trajectory analysis to adjust the magnitude of the hydrodynamic forces and torques that act on colloids near solid surfaces [*Rajagopalan and Tien*, 1976; *Johnson et al.*, 2007]. *Johnson et al.* [2009] have implied that the hydrodynamic correction functions provide a frictional viscous force for colloid immobilization. However, the hydrodynamic corrections should not change the direction of the tangential hydrodynamic force, and this quantity should still be nonzero during fluid flow. Hence, the force and torque balance implemented by *Johnson et al.* [2007] indicates that colloids associated with the secondary minimum will only be immobilized in porous media in stagnation regions where the fluid velocity is zero. An accurate solution of the Navier-Stokes equations reveals that there are just two (front and rear) stagnation points for each collector in simple and dense cubic sphere packs when water is flowing. Therefore, the contribution of these stagnation points on colloid retention under unfavorable conditions should be insignificant if the notion of “freely mobile” associated colloids in secondary energy minimum is correct.

[5] The conceptual picture described above of colloid interaction via the secondary minimum is not always consistent with experimental observations. In particular, significant amounts of colloid retention have been observed

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under unfavorable conditions [Franchi and O'Melia, 2003; Walker et al., 2004; Redman et al., 2004; Tufenkji and Elimelech, 2004, 2005; Hahn and O'Melia, 2004; Hahn et al., 2004], and this is not consistent with the "freely mobile" assumption of colloid retention only in stagnation points. Furthermore, a sharp peak of colloid release has been observed once the eluting solution was switched to a lower ionic strength and the secondary minimum was reduced or eliminated [Redman et al., 2004; Tufenkji and Elimelech, 2004, 2005; Bradford et al., 2007]. In contrast, release of colloids will be diffusion controlled when they are retained in stagnation points ("freely mobile" assumption) and this will produce low levels of "concentration tailing." We encourage the authors of Johnson et al. [2007] to further investigate this issue using their colloid trajectory model by simulating the retention of colloids in the secondary minimum, and then the subsequent release of colloids when the secondary minima is removed.

3. Can the Torque Balance Approach Be Used to Quantify Adhesion and Detachment?

[6] Inspection of the comment by Johnson et al. [2009] and the above information reveals their concern, whether the torque balance approached can be used to quantify adhesion and detachment. Specifically, the authors believe that colloids associated with the secondary minimum are "freely mobile" and that they do not experience any resistance to colloid motion from adhesive forces because there is no physical contact. Hence, they do not approve of our torque balance approach used to generate Figure 5 of Torkzaban et al. [2008].

[7] A rigorous theoretical attempt to quantify the adhesion of colloids was made by Johnson et al. [1971], comprising the JKR theory. This theory forms the basis of modern approaches to quantify colloid adhesion [Pollock et al., 1978], and has been applied to investigate the influence of hydrodynamics on particle detachment under favorable [Bergendahl and Grasso, 1999, 2000] and unfavorable [Bradford et al., 2007; Torkzaban et al., 2007] attachment conditions. In the JKR theory the net adhesive force between a spherical colloid at a particular separation distance from a flat surface acts over a contact radius that produces a lever arm that is given by [Israelachvili, 1992]

$$l_x = \left(\frac{F_A r_c}{K} \right)^{1/3} \quad (1)$$

where r_c is the colloid radius, F_A is the net adhesive force, l_x is the lever arm that is associated with F_A (i.e., the distance on the solid surface over which F_A acts), and K is the composite Young's modulus. It should be noted that equation (1) has been proposed for cases when there is no physical contact between the colloid and collector. This occurs because the adhesive force acts over a contact radius, not just a single point [Israelachvili, 1992; Hoek et al., 2003; Duffadar and Davis, 2007]. The contact radius therefore increases with net adhesive force and colloid size. We do not claim this equation is ideal, but to the best of our knowledge this is the only expression available for determining the contact radius for colloids interacting with the solid phase at a separation distance. Torkzaban et al.

[2008] have therefore applied equation (1) to estimate l_x when colloids interact with the solid phase by a secondary minimum.

[8] It should be mentioned that in our paper [Torkzaban et al., 2007] we employed a slightly different functional form for l_x at separation that had a 4 in the denominator of equation (1) as a result of an algebraic error. Furthermore, if physical contact between the colloid and collector is assumed in JKR theory, then the value of the lever arm is given as

$$l_x = \left(\frac{4F_A r_c}{K} \right)^{1/3} \quad (2)$$

These differences in l_x have undoubtedly led to some confusion in the literature, and we acknowledge that the comment by Johnson et al. [2009] has given us the opportunity to clarify this issue. However, relatively minor differences are predicted for the various forms of l_x (0.63 to 1.59 times l_x given by equation (1)) and torque balance calculations produce similar trends with regard to ionic strength and velocity when using these formulations.

[9] Johnson et al. [2009] state in their comment that a physical contact is established when a colloid associates with a surface via the primary energy minimum. This statement implies that these authors accept the existence of a lever arm (l_x) for particles retained in the primary minimum, but not in the secondary minimum. So, we would like to point out the similarities between these two energetic conditions for attachment. In aquatic environments, colloids retained in either the primary or secondary minima do not have direct physical contact with the surface. Rather there always exists a film of water between the colloid and collector surface creating a minuscule separation distance [Elimelech et al., 1995; van de Ven, 1989]. Theoretical modeling of the water structure at interfaces with solids, experimental determinations of surface speciation, force measurements between interacting particles, and studies of the rheological properties of concentrated suspensions all indicate that for separations of a few nanometers or less, non-DLVO forces will often produce strong repulsive forces that prevent the formation of physical contact in aquatic systems [Elimelech et al., 1995; Hahn et al., 2004]. Indeed experimental work confirms the existence of this water layer and minor separation distance, in that colloids in the primary minimum can be released by altering the solution chemistry (i.e., increase in pH or decrease in ionic strength) [Redman et al., 2004; Tufenkji and Elimelech, 2004, 2005]. If direct physical contact existed between the colloid and the surface then these colloids should not have been released by changes in the pH or ionic strength. Therefore, the notion of physical contact with the surface in the primary minimum seems to be inappropriate. Figure 1 clarifies the point further. Note in this schematic that colloids retained in the primary and secondary minimum are simply colloids interacting with a surface. The only difference is the magnitude of the adhesive force and the separation distance between the collector and the colloid, which in this case is assumed to be 1 nm and 8 nm for the primary and secondary minima, respectively. If we accept that there is a lever arm (adhesive contact area) on which the forces act for colloids held in the primary minimum, then there should also be a

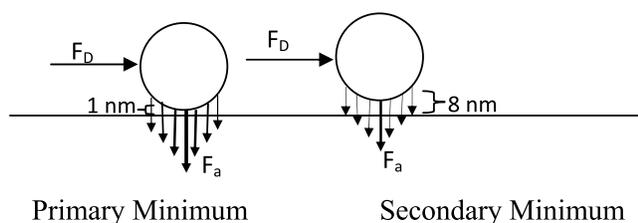


Figure 1. A schematic of colloids retained in the primary and secondary minimum. Here F_D is the net fluid drag force and F_a is the distribution of adhesive forces between the colloid and the solid surface.

lever arm on which forces act for colloids attached in the secondary minimum.

[10] Similar to Bergendahl and Grasso [1999, 2000], Torkzaban et al. [2008] have assumed that colloid attachment and detachment on the solid surface is controlled by the torque balance (consideration of the applied hydrodynamic torque and the resisting adhesive torque) instead of the force balance equation. When the torque balance results in disequilibrium at a particular location, hydrodynamic forces cause colloids to roll and colloid attachment is prevented [Bergendahl and Grasso, 1999, 2000; Bradford et al., 2007; Torkzaban et al., 2007]. It should be noted that our technique does not imply that the colloid was previously immobilized; rather it assesses the fraction of the collector surface that may potentially contribute to colloid attachment, as previously described by Torkzaban et al. [2007].

[11] In summary, the notion of attachment of secondary and primary minima associated colloids on the solid surface in the presence of fluid drag without assuming any resisting torque and force due to adhesive is not consistent with the basic physical laws of force and torque balances. If a resisting torque (lever arm) did not exist for colloids in the secondary (or primary) minimum, then in contrast to literature findings [Franchi and O'Melia, 2003; Walker et al., 2004; Redman et al., 2004; Tufenkji and Elimelech, 2004, 2005; Hahn and O'Melia, 2004; Hahn et al., 2004] no colloid attachment would occur in the presence of fluid drag because the two major forces (adhesive and fluid drag) acting on the colloids are perpendicular and do not cancel each other.

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