Reply to “Comments on ‘Pore-Scale Visualization of Colloid Transport and Retention in Partly Saturated Porous Media’”

Wan and Tokunaga’s (2005) comments give us an opportunity to relate our recent findings to our initial paper (Crist et al., 2004). The main purpose of our first paper (Crist et al., 2004) was “to describe a novel pore-scale visualization technique” and “help reconcile some inconsistencies among earlier studies,” in particular by demonstrating the need to refine the widely held assumption used to explain observed breakthrough curves, namely that colloids are retained at air–water (AW) interfaces in unsaturated media. Contrary to the statement of Wan and Tokunaga (2005), attempting “to provide pore-scale insights into mechanisms responsible for controlling colloid transport in unsaturated porous media” was not the aim of the initial Crist et al. (2004) paper. We discussed insights into retention mechanisms in a subsequent paper (Crist et al., 2005) under conditions in which evaporation effects on colloid retention were greatly reduced by using steady-state flow. Furthermore, the length of the unsaturated zone in this subsequent paper was increased by using larger diameter sand particles and employing a steeper chamber slope. We are therefore disappointed that Wan and Tokunaga restricted their comments “for brevity” mainly to our 2004 paper.

It is of interest that we did not observe the colloid straining conceptualizations that Wan and Tokunaga (2005) depicted in their Fig. 2b (“with evaporation,” where colloids form a wide band at air–water–solid (AWS) interfaces in pendular rings) unless colloid diameter and water film thickness were approximately the same. With some slight adjustments, we observed phenomena similar to their Fig. 2a, with one difference: while Wan and Tokunaga (1997, p. 2418) envisioned that “ colloids are constrained to rolling along the grain surfaces...” below critical saturation, in our case colloids were stagnant after they attached at the AWS interface, indicating a slightly different mechanism. Note that by “AWS interface” we refer to the location where the meniscus either meets the dry grain surface or diminishes in thickness to a thin water film covering the grain surface. In our recent work (Zevi et al., 2005) we have proposed the term air–water meniscus–solid interface (abbreviated AWmS) for this concept, denoting the region where the water meniscus ends by thinning to a thin film, which more correctly represents the interfaces observed in unsaturated media. Our equipment at the time of the Crist et al. (2004) experiments could not observe thin films. Wan and Tokunaga’s (2005) reasons for distinguishing whether a thin film exists at the AWS interface are not clear to us at this time; although it could prove to be important in the future.

We are also puzzled by Wan and Tokunaga’s (2005) comment that the “magnitude of [our] observations [of colloid retention at the AWS] are not likely to occur anywhere except within the most superficial few centimeters of soils.” In this initial paper we did not claim (nor understand) that it was an important mechanism, noting that we “fully expected” not to observe such retention in the vadose zone. The authors have chosen not to cite our findings of retention at the AWS in Crist et al. (2004), both real and significant.

The comment of Wan and Tokunaga (2005) that evaporation increases the advection of contact lines where they are subsequently defined is an oversimplification. In our subsequent work with both better experimental and fewer evaporation effects, we proved with our equipment that the findings of retention at the AWS in Crist et al. (2004) were both real and significant.

In our work with both better experimental and fewer evaporation effects, we proved that bands in the Fig. 6 of the original paper [Crist et al., 2004] were visible bands at the AWS surfaces, it is clear that evaporation is a process that influences colloid retention at the AWS.

In the flowing water experiments described by Wan and Tokunaga (2005), the same pattern of immobilization at the AWS interface was clearly seen under conditions in which any effect of evaporation would be negligible. To further determine the relative significance of evaporation, we repeated the Crist et al. (2004) experiment in a larger chamber using a bright field microscope, but this time with a cover plate on the chamber to prevent evaporation. The image in Fig. 1 (which is of moderate quality due to the optical effects of the cover plate) clearly shows that colloids are attached at the AWS (AWmS) interface. More importantly, colloids occurred as soon as the colloids were added and we were not to see any opportunity for evaporation to exert an effect.

To further determine the relative significance of evaporation, we repeated the Crist et al. (2004) experiment in a larger chamber using a bright field microscope, but this time with a cover plate on the chamber to prevent evaporation. The image in Fig. 1 clearly shows that colloids are attached at the AWS interface. More importantly, colloids occurred as soon as the colloids were added, indicating a slightly different mechanism. Note that by “AWS interface” we refer to the location where the meniscus either meets the dry grain surface or diminishes in thickness to a thin water film covering the grain surface. In our recent work (Zevi et al., 2005) we have proposed the term air–water meniscus–solid interface (abbreviated AWmS) for this concept, denoting the region where the water meniscus ends by thinning to a thin film, which more correctly represents the interfaces observed in unsaturated media. Our equipment at the time of the Crist et al. (2004) experiments could not observe thin films. Wan and Tokunaga’s (2005) reasons for distinguishing whether a thin film exists at the AWS interface are not clear to us at this time; although it could prove to be important in the future.

We feel that the Wan and Tokunaga (2005) conclusion that bands in the Fig. 1 are a poor analog for observations on evaporation of a static 1-μL droplet of a colloid suspension are not likely to occur anywhere except at a much larger scale and with flowing water. We continually replenish any small amount of water that could be lost to evaporation. In our experiments the contact line recedes during evaporation, resulting in the attachment of colloids at the contact line as shown in their Fig. 6. In our water experiments, the contact line is not at the location of the constant inflow of water. In these circumstances, bands in the Fig. 6 are a poor analog for observations on evaporation of a static 1-μL droplet of a colloid suspension.