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

Recent studies by Crist et al. (2004, 2005) attempted to provide pore-scale insights into mechanisms responsible for controlling colloid transport in unsaturated porous media. However, their key observation of colloids being trapped at air–water–solid (AWS) contact lines relied on images obtained along surfaces that were open to the atmosphere and thus subject to evaporation. Our analyses using their procedure show that because of exposure to the atmosphere, evaporation-driven artifacts account for most of their observed colloid trapping at AWS contact lines. In the following analysis of their work, we show how evaporation resulted in colloid deposition at AWS contact lines. Our comments were originally submitted in response to Crist et al. (2004) before the publication of Crist et al. (2005). The evaporation problem that we noted previously, in response to the first paper, was not rectified in Crist et al. (2005). For brevity, we will restrict our present comments to Crist et al. (2004), while noting that the visualization results presented in Crist et al. (2005) also suffer from the same artifacts because the obtained images were of evaporating surfaces.

As described in Crist et al. (2004), in their experimental setup “the front panel was removed to avoid light reflections that obscured the view and, thus, exposed one side of the sand column to air”. Here, we show that removal of the front panel results in creating a sequence of three critical artifacts: (i) significant evaporation; (ii) drying of thin films on grain surfaces, causing formation of AWS contact lines; and (iii) advection of colloids to AWS contact lines, where they are deposited. As explained below, these artifacts so drastically disturbed their system that the magnitude of their observed colloid entrapment is not likely to occur anywhere except within the most superficial few centimeters of soils.

Before explaining these artifacts, we note that while the trapping of colloids at AWS contact lines, as reported in Crist et al. (2004, 2005), is largely an artifact of evaporation, colloid filtration within perimeters of pendular rings is indeed a key prediction of the film straining model (Wan and Tokunaga, 1997). In that model, colloid filtration is predicted to be more efficient below a critical water saturation when capillary connections between pendular rings become separated, connected only by thin water films. In that paper, we stated, “Retardation of ideal, nonabsorbing colloids can occur at two locations: trapped within individual pendular rings due to exclusion from entry into surrounding thin films and within films” (Wan and Tokunaga, 1997). Thus, while Crist et al. (2004, 2005) implied that the film straining model applies only to retardation of colloid transport within thin films, colloid retention within perimeters of pendular rings is in fact a critical feature of our model.

Significance of Evaporation

To determine the significance of evaporation, we constructed a flow chamber having the dimensions presented in Crist et al. (2004) and repeated their wetting and drainage procedures, with and without covering the sand pack. Our tests were conducted on acid-washed Unimin sand of the same grain size (0.43–0.60 mm), at room temperature (21.5–23.5°C), under relative humidity (RH) values ranging from 24 to 40%, and without lighting or heating from an illuminating device other than ordinary fluorescent lights along the laboratory ceiling. Our tests on covered sand packs were conducted in the manner described in Crist et al. (2004), except that the cover plate remained on until the time at which moisture-content sampling was performed. The air phase in the upper portion of the covered flow chamber was vented to atmospheric pressure via a pair of syringe needles. As in Crist et al. (2004), outflow was undetectable in all of our tests beyond a few minutes after setting the sand at a 35° inclined angle. However, in open systems such as that used in Crist et al. (2004), hydrostatic conditions cannot be assumed simply because of undetectable drainage. Various moisture profiles obtained in our tests, as well as the profile at 2 h drainage reported in Crist et al. (2004, estimated from Fig. 2 of their paper) are shown in Fig. 1. The moisture profiles obtained in our open system at 2 h of drainage are in rough agreement with Crist et al. (2004), although they obtained drier sand through most of their profiles (40–170 mm from the top) and slightly wetter sands within the upper section (0–40 mm). Their generally lower water contents could be the result of additional heating from their lighting system (Crist, 2002, p. 62). In contrast to the moisture profiles obtained from open flow chambers, those obtained on the covered sands remained nearly saturated, even in the upper sections. Water contents in the upper region (down to the 70-mm depth along the 35° incline) of our open system were clearly much drier than in our covered system. The water loss from the flow chamber during the 2-h period (shaded area between curves in Fig. 1) normalized to the open sand area is equivalent to an evaporation rate of 83 ± 7 μm h⁻¹. Pan evaporation rates of 82 to 120 μm h⁻¹ were measured in the laboratory adjacent to the flow chamber during these experiments.

The profile resulting from the 11-h exposure of the sand surface further illustrates the impact of evaporative water loss. Evaporation significantly disturbed moisture contents during and after the 2-h time frame used for obtaining images. Thus, based on our findings, the assumption made by Crist et al. (2004) that after 2 h, “The moisture content did not vary significantly thereafter, as the drainage from the bottom of the column was minimal to undetectable,” is incorrect. While the disturbance was obvious in these “bulk” measurements obtained on the full 5-mm thickness of the slab sections, artifacts from drying had to be much more severe along the surficial monolayer of sand grains, where evaporation took place. Advancement of the drying front downward from this open boundary amplified the magnitude of the evaporation artifact because all of the photographic images were obtained within micrometers of this surface layer.

Air–Water–Solid Contact Lines Are Artifacts Caused by Evaporation

The term AWS interfaces appears throughout Crist et al. (2004) and is central to their results. However, because dry air–solid interfaces need to be present for AWS contact lines to exist, the significance of such boundaries in most vadose zone environments is questionable. Below depths of a few centimeters, RH values in soils are typically higher than 95%, so thin water films coat surfaces of mineral grains lacking hydrophobic organic coatings. The development of AWS in-

Abbreviations: AWS, air–water–solid; RH, relative humidity.