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Short Communication

Assessment of the application of percolation theory to a water repellent soil

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Abstract. A few hydrophobic grains in otherwise hydrophilic sand render the soil hydrophobic and can completely alter the flow of water through unsaturated sands. In this paper we examine whether percolation theory can explain the phenomenon. Percolation theory has been used to describe the dependence of large-scale flow phenomena on heterogeneities found at the pore scale and should, therefore, be able to explain the water flow behaviour in hydrophobic soil. We show that the theory is valid, in general, for a hydrophilic soil into which a small but increasing fraction of highly hydrophobic grains is mixed. However, the application of percolation theory is limited by the complex interactions of matric potential and contact angle effects due to the introduction of hydrophobic particles.

Introduction

Microscopically examining a Dutch soil that was macroscopically water repellent, Bisdom *et al*. (1993) identified only a few hydrophobic particles. Likewise, Bauters *et al*. (1998) observed that as few as 3 hydrophobic grains in 100 total grains could change the infiltration wetting pattern from a wide horizontal wetting front to an unstable finger-like pattern. Crist *et al*. (2004) showed that even 1 grain in 100 could alter the flow behaviour. But it is not clear how so few hydrophobic particles can change the hydraulic properties of a porous medium. In this contribution, we assess how well critical path analysis and percolation theory can explain these changes. Specifically, we examine whether the theory can predict the dramatic differences in flow behaviour observed by Bauters *et al*. (1998) as a result of replacing a few hydrophilic grains by hydrophobic grains.

Percolation theory has been used extensively to model various aspects of flow through porous media to describe the dependence of large-scale flow phenomena on heterogeneities found at the pore scale (Hunt 2001). The change in flow behaviour at the Darcy scale by a small fraction of water-repellent particles therefore falls within the domain for which the theory was developed. Percolation theory applications to transport in heterogeneous systems have been described in the physics literature since the 1970s (Pike and Seager 1974; Seager and Pike 1974), and a wide range of porous media flow problems have also drawn on percolation concepts (DiCarlo *et al*. 2000; Blunt 2001; Simms and Yanful 2002). Percolation theory can also be used to predict soil unsaturated hydraulic conductivity (Hunt 2001; Hunt and Gee 2002).

The experiment of Bauters *et al*. (1998) used blasting silica sand (W.F. Saunders & Sons, Inc.) as the substrate to analyse the effect of water repellency. The water repellency in this sand was made such that, unlike natural sandy soils, the hydrophobic portion would not leach out or degrade as observed by Ritsema and Dekker (1996) in the coastal region of The Netherlands. Table 1 shows the textural composition of the sand used. Initial infiltration experiments showed that this hydrophilic sand produced a normal wetting front. Different degrees of water repellency were added to the sand through the following procedure. An ethanol solution containing 4.8% octadecyltrichlorosilane (OTS) was mixed with 25 kg of blasting sand in a cement mixer for 5 h, yielding an extremely water repellent sand after drying (Bradford and Leij 1996). A basis sand batch was made by mixing 3 kg of this hydrophobic sand with 45 kg of regular (hydrophilic) sand. This basis sand was then diluted with regular sand to achieve repellency fractions ranging from 0 to 10% water repellent grains. As mentioned above, fingers formed when the sand contained 3% or more water repellent grains. When the water repellent sand/regular sand ratio was increased to 5–6%, water would no longer infiltrate spontaneously, and a positive water pressure was required for infiltration. These differences in infiltration were directly related to the shape and location of the primary wetting curve of the soil. Wetting curves were measured using

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Table 1. Screen analysis (by weight) of the blasting silica sand provided by manufacturer (W. F. Saunders & Sons, Inc.)

Screen opening (mm)	Cumulative (%)
0.840	Trace
0.590	0.8
0.425	14.9
0.297	53.5
0.212	81.9
0.149	92.9
Through 0.150	100.0

sands with different degrees of water repellency, letting water infiltrate from the bottom in a segmented column and measuring the water content in each segment (Fig. 1). At 3.1%, the matric potential at which the water content increased is still negative, but at fractions >5.5%, the water entry pressure becomes positive. In Fig. 2, the drying curves from the hydrophobic sands all fell approximately on the same line.

One of the effects of adding hydrophobic grains is that water will use different pathways to flow through the porous media. We could say that the original pathway is blocked at the location of the hydrophobic grain at the same matric potential as the hydrophilic sand. If sufficient grains are added, all paths are blocked. From Fig. 1, we see that in the Bauters *et al.* (1998) experiments, a positive pressure is needed for water to enter the soil when 5–6% of the grains are water-repellent. With fewer hydrophobic particles, the matric pressure at saturation is negative, meaning that the water can infiltrate immediately after it is applied without ponding. If >5–6% of the grains are hydrophobic, water will not infiltrate unless ponded. In other words, at saturation at zero matric potential, all flow paths are blocked. To describe this type of blocking, percolation theory is being used. The main objective of this paper is to examine to what degree percolation theory can be used for describing the flow phenomena in hydrophobic soils.

Conceptual model

An important distinction must be made before proceeding. Henceforth, we use the term 'allowable' to designate a pore which could be filled with water at a given matric potential. In order to reach and fill that pore, however, water must find a continuous, connected path through the allowable pore space. If such a path exists, we designate the pore as 'accessible.' In the absence of hydrophobic particles, if water is to access the pore in question, this pathway must be composed of wettable pores small enough so that they are water-filled at the given matric potential. In the presence of hydrophobic particles, the same path can only be filled with water at a higher matric potential. The matric potential must increase to produce contact angle changes required to allow the pathway to be water-filled.

Replacing regular hydrophilic sand particles by hydrophobic sand particles does not affect the structure of the soil itself, but at a given matric potential it changes the distance between the flow paths and, thereby, the means to get water to the *allowable pore space* at that particular matric potential. We will assume that the matric potential has to increase before pores will accept water if they border a water repellent grain. But, by increasing the matric potential, other pathways with larger pores will also open and soil water content will increase. This is consistent with the increase in water content with matric potential in the wetting curve.

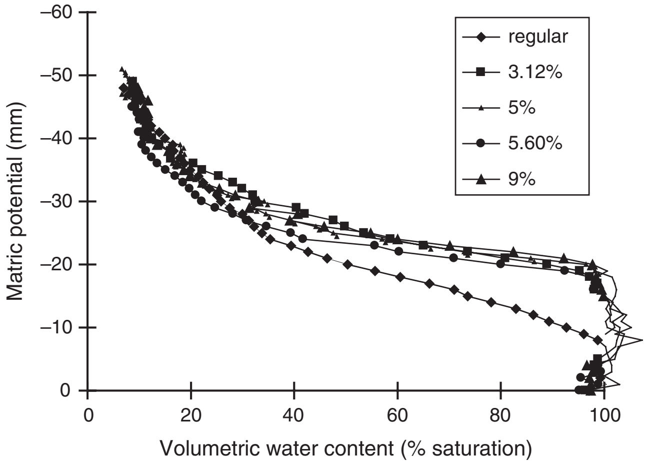

Fig. 1. Wetting curves for identical sands with different degrees of hydrophobicity.

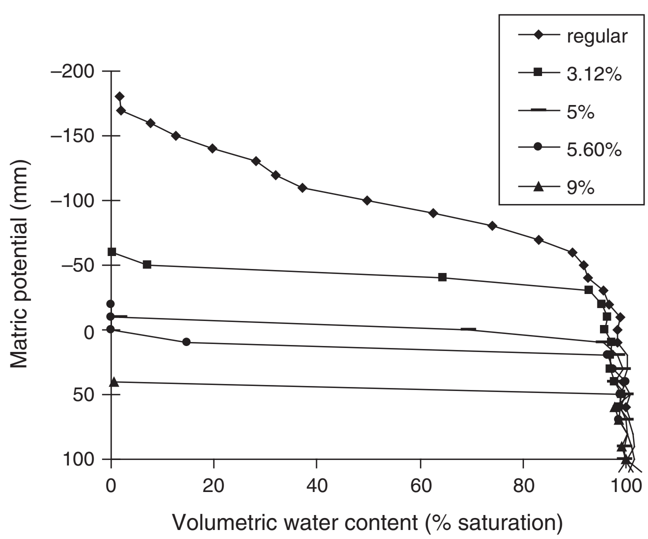

Fig. 2. Drying curves for identical sands with different degrees of hydrophobicity.

Percolation theory application

The critical question is: Is there sufficient *allowable pore space* to form an interconnected path? To answer that question, we need to set criteria for water to enter the accessible pore space. We will assume that a significant volume of water can begin to enter the system after the distance between flow paths, χ, diminishes below the typical separation distance of hydrophobic particles, L_0. This parallels the approach of Raikh and Ruzin (1990), who treated *longitudinal* conduction in systems with one physical dimension much shorter than the others.

For uniform distribution of hydrophobic grains, the separation distance of hydrophobic particles, L_0, is given by:

$$L_0 = C_2 d N^{-\frac{1}{3}} \tag{1}$$

where N is the fractional volume of the grains that are hydrophobic, d is the typical particle diameter, and C_2 is the constant of proportionality. For a cubic packing we find that when $N \Rightarrow 1$, ${L_0}^3 = d^3$; hence, C_2 is of the order 1.

To find the mean path separation between water accessible pores, χ, the geometrical extent of the interconnected allowable pore spaces has been derived (Stauffer 1979; Berkowitz and Ewing 1998). Since the mean path separation distance is expected to behave like a power law, and the separation distance becomes infinite near the critical moisture content, a behaviour of the form:

$$\chi = C_1 d(\theta - \theta_c)^{-0.88} \tag{2}$$

can be expected (eqn 7 in Berkowitz and Ewing 1998) where θ is the volumetric moisture content and represents the bond probability; C_1 is an unknown numerical quantity that should be of the order unity (Stauffer 1979), and the equation is written for $\theta > \theta_c$, where θ_c is the 'critical volume fraction for percolation'.

To find θ_c, it is reasonable to assume that the continuous network for capillary flow breaks up at the moisture content on the drainage curve (Fig. 2) where the matric potential begins to decrease dramatically without an appreciable change in moisture content (Hunt and Gee 2002). According to Fig. 2, the moisture content, θ_c, where this occurs is approximately 10% of saturation or 0.04 cm^3/cm^3. Having identified this parameter from the drainage branch of the soil characteristic curve, we can then use it in predictions involving either wetting or drying. In order to find the water content, a rule concerning the blockage of pores is needed. The simplest rule is that if a pore is bordered by a hydrophobic grain, the pore is blocked for water entry independent of the matric potential. Based on this the water content, θ', at which the accessible pore space forms an unblocked network and percolation can take place can be found by setting the mean path of separation, χ, in Eqn 2 equal to the average separation length, L_0, of the hydrophobic particles in Eqn 1:

$$C_2 d N^{-\frac{1}{3}} = C_1 d(\theta' - \theta_c)^{-0.88} \tag{3}$$

Solving Eqn 3 for θ' gives the effective critical volume fraction for percolation as a function of the fraction, N, of the hydrophobic grains:

$$\theta' = \theta_c + C_3 N^{\frac{1}{2.64}} \tag{4}$$

with $C_3 = (C_2/C_1)^{1/0.88}$. Both values C_1 and C_2 are of the order unity, so the combination of constants, C_3, is also of the order unity.

To assess the validity of Eqn 4, we will use the experiment of Bauters *et al.* (1998) by checking if the constant C_3 is approximately equal to 1. As mentioned before, the water does not enter the soil without ponding when 5.5% of the grains are water repellent, i.e. $N = 0.055$. In other words, at a matric potential of zero, i.e. no ponding, the critical volume fraction for percolation is saturation, i.e. $\theta' = 0.38$. Using these values and the earlier estimate $\theta_c = 0.04$ we find with Eqn 4 that:

$$C_3 = \frac{0.38 - 0.04}{0.055^{\frac{1}{2.64}}} \tag{5}$$

Thus $C_3 = 0.33/0.34$ or 0.98, which is, indeed, of the order unity. This means our assumption that pores are blocked when they are bordered by a hydrophobic grain is reasonable. Since we made the calculations at zero potential, we can be more specific. When the effective contact angle is 180°, water will not infiltrate into pores that are bordered by hydrophobic grains. Conceptually, this seems to be acceptable too.

We have shown that percolation theory explains well why a few hydrophobic grains can prevent water entry into the soil under nonponded conditions. What percolation theory cannot predict is at what matric potential the water will infiltrate into a hydrophobic soil. To do so, more information would be needed about the matric potential at which water will enter into pore spaces that are bordered by hydrophobic grains.

In summary, based on percolation theory, we have demonstrated that, in agreement with experimental observations, small percentages of hydrophobic grains can drastically change the flow behaviour in a soil. Two practical applications of our findings bear mentioning. First, if a native soil is strongly water repellent only at the surface, mixing the hydrophobic material in with the rest of the soil profile will merely result in making the entire profile water-repellent. Second, our results provide an explanation of why relatively small amounts of hydrophobic peat mixed into a hydrophilic potting soil can significantly increase the water-holding capacity of these mixes by blocking paths through which the water otherwise would drain.

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