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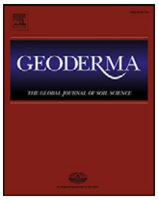

Does hydrocarbon contamination induce water repellency and changes in hydraulic properties in inherently wettable tropical sandy soils?

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ABSTRACT

Hydrophobicity influences soil hydrological and ecological functions. Compared to naturally-occurring and fire-induced hydrophobicity, limited information is available on the impacts of hydrocarbon contamination on water repellency and hydraulic properties. Water repellency and hydraulic properties were measured on laboratory simulated, and field contaminated soils, 1 and 5 years after an accidental petroleum hydrocarbon spill. The objectives were; (1) to compare the water droplet penetration test (WDPT) to the molarity of ethanol droplet (MED) test, (2) to investigate the effect of hydrocarbon contamination on water repellency and hydraulic properties, and (3) to evaluate the performance of pedotransfer functions for hydraulic properties. The WDPT and MED tests gave qualitatively similar water repellency results as evidenced by a significant positive correlation ($p < 0.05$, $r^2 = 0.95$) between the mean time for the two methods. Laboratory simulated hydrocarbon contamination induced soil water repellency. Saturated hydraulic conductivity (K_s) increased linearly with level of contamination ($p < 0.05$; $r^2 \approx 0.8$), indicating that rapid flow of water attributed to a reduction of the dielectric constant, and hence water–soil matrix interactions. No water repellency was observed in contaminated field soils (WDPT < 3 s), but the residual signature of hydrocarbon contamination was evident in other soil properties particularly electrical conductivity. This indicates that natural soils were inherently wettable and that hydrocarbon-induced hydrophobicity could be transient. This non-persistence was attributed to high decomposition rates stimulated by tropical conditions and nutrients added to promote revegetation. Predictions of pedotransfer functions were comparable to measured hydraulic data ($p < 0.05$, $r^2 > 0.8$), confirming their general validity for water and solute transport modeling even on contaminated soils. The study confirmed the hypothesis that hydrocarbon contamination induces water repellency and reduces soil moisture retention at low suction (<100 kPa) for laboratory contaminated soils, but effects were inconsistent for field samples. However, the increased saturated hydraulic conductivity associated with laboratory contaminated soils contradicted the original hypothesis. The findings imply that storms falling on initially dry recently contaminated soils may trigger contaminant transport and erosion via enhanced surface runoff, and rapid spreading of contaminants once they reach the groundwater systems. These hydrological impacts are critical for remediation of contaminated sites. Future research could use a contamination chronosequence/gradient to provide comprehensive information on the temporal evolution of water repellency and hydraulic properties under field conditions.

1. Introduction

Hydrophobicity or water repellency is a well-recognized phenomenon influencing soil hydrological behavior and agricultural productivity. Water repellent soils resist wetting, and inhibit infiltration (Dekker and Ritsema, 1994). Naturally-occurring and fire-induced water repellency has been the subject of several studies conducted in Mediterranean environments in Australia, Spain, Portugal and Chile (Doerr et al., 2000, 2006) and sandy dunes in Netherlands (Dekker and Ritsema, 1994), where water repellency appears more widely reported than in other regions. Besides fire and antecedent soil moisture, the occurrence and severity of water repellency are also influenced by soil type and properties (Badía et al., 2013; Doerr et al., 1996), vegetation type, soil management and land use practices (Harper et al., 2000; Zavala et al., 2009). For example, although water repellency has often been associated with coarse-textured soils, several studies have shown that severe water repellency also occurs in various soil types including those that are fine-textured, aggregated, acidic and alkaline soils (Doerr et al., 2000; Jordán et al., 2013; Mataix-Solera and Doerr, 2004).

In earlier studies, researchers investigated the origin and characteristics of water repellency (Doerr et al., 2000; Jordán et al., 2013), evaluation methods (e.g. Doerr, 1998; King, 1981; Letey, 1969; Letey et al., 2000; Watson and Letey, 1970), impacts on hydrological behavior including preferential flow (Dekker and Ritsema, 1994; Wallach, 2010;

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Wallach and Jortzick, 2008) and amelioration and management practices (e.g. Dlapa et al., 2004). The causes of hydrophobicity include plant derived waxes, humic and fulvic acids and organic compounds from forest fires (Arcenegui et al., 2007; Doerr et al., 2000; Huffman et al., 2001; Ritsema et al., 1993; Scott, 2000). Water repellency influences water redistribution via reduced infiltration, enhanced surface runoff and erosion, and preferential flow or fingering (Doerr et al., 2000). These changes in hydrological balance, may in turn impact on soil–plant water relations, resulting in impeded seed germination, stunted plant growth and reduced plant productivity (Mainwaring et al., 2004). Other researchers have investigated the potential to ameliorate water repellency and the associated impacts through localized irrigation, tillage, and application of clays and surfactants or wetting agents (Buczko et al., 2006; Dlapa et al., 2004; Kostka, 2000). Several methods exist for evaluating the occurrence and severity of soil water repellency, the molarity of ethanol droplet (MED) and the water droplet penetration test (WDPT) being the most prominent (Dekker and Jungerius, 1990; Dekker and Ritsema, 1994; King, 1981). However, comparative studies on their performance particularly on hydrocarbon contaminated soil are limited. Therefore uncertainty exists about the sensitivity and comparability of results between the two methods.

In comparison to other areas, little is known about the occurrence of water repellency in the predominant tropical soils of southern Africa. The reason for this lack of information is unclear, but could be indicative of the general lack of hydrological research in the region. An exception is a study by Scott (2000) documenting water repellency and reduced infiltration and enhanced runoff in an exotic eucalyptus timber and pine plantation, and natural *Acacia* dominated miombo woodland in South Africa. The miombo woodlands are the dominant native vegetation type in southern Africa, covering over 3.6 million km^2 across 11 countries (Timberlake and Chidumayo, 2011). The miombo woodlands consist predominantly of deciduous broad-leaved leguminous trees with a well-developed grass understory, giving rise to frequent and widespread veld fires. Although documented cases of naturally-occurring or fire-induced water repellency are scarce in the region, soil contamination through anthropogenic activities could potentially cause water repellency. In particular, wastewater irrigation, soil application of biosolids and hydrocarbon contamination may introduce hydrophobic organic compounds into the soil system. However, compared to naturally-occurring and fire induced hydrophobicity, little is known about the impacts of contamination on water repellency and soil hydraulic properties.

Aislabie et al. (2004) noted that few studies exist on the impacts of hydrocarbon contamination and associated additives on water repellency and moisture retention. In an arid region, wastewater irrigation has been reported to cause water repellency (Wallach et al., 2005). A study conducted in Canada on weathered oil-contaminated sites showed that some long-chain and polycyclic aliphatic organic compounds of petroleum origin were water repellent (Roy et al., 1999). On Barrow islands in Australia, George et al. (2011) observed that flowline additives associated with oilfield installation had no effect on water repellency. In the Antarctic region, hydrocarbon-contaminated soils were weakly hydrophobic, but impacts on moisture retention were negligible (Aislabie et al., 2004). Elsewhere, hydrocarbon contamination was also reported to alter soil field capacity, porosity, soil bulk density and optimum water content even at low hydrocarbon contamination levels (Adams and Cruz, 2008; Adams et al., 2008; Caravaca and Rolda, 2003; Rahman et al., 2010). In other studies, soil contamination by petroleum hydrocarbons was reported to increase the moisture retention of soil at high suction values (Burckhard et al., 2004; Hyun et al., 2008), while a decline in water retention was observed by Roy and McGill (1998). These changes often result in reduced plant growth and productivity (Adams and Cruz, 2008). In summary, the findings of these earlier studies are inconsistent, and often contradicting. Moreover, the bulk of these studies were drawn from cool and humid temperate and arctic conditions (Adams and Cruz, 2008; Balks et al., 2002; Foght and Waterhouse, 2004; Quyum, 2000). By contrast, there is a paucity of information on the impacts of hydrocarbon contamination on water repellency and hydraulic properties in tropical environments typical of southern Africa. Unlike temperate and Arctic environments, the tropics experience distinct warm to hot and seasonally dry climatic conditions, resulting in diverse soil types. These unique climatic and soil characteristics constrain the extrapolation and generalization of findings obtained in other environments.

Knowledge of soil hydraulic properties is crucial for understanding the hydrology and remediation of contaminated sites (Gwenzi et al., 2011). Soil hydraulic properties particularly saturated hydraulic conductivity (K_s) and soil moisture retention (SMR) influence soil moisture storage, deep drainage, runoff and infiltration, and provide key inputs for water balance and solute transport models (Gwenzi, 2010; Gwenzi et al., 2011; Holländer et al., 2009). Most existing water and solute transport models rely on hydraulic properties estimated from pedotransfer functions derived for uncontaminated natural soils (Bohnhoff et al., 2009; Holländer et al., 2009). Hydrocarbon contamination could potentially cause water repellency and associated changes in hydraulic properties. Consequently, PTFs for K_s and SMR developed for uncontaminated natural soils may fail to predict field measurements on such contaminated soils. Therefore, there is need to evaluate the capacity of existing pedotransfer functions to predict saturated hydraulic conductivity and soil moisture retention for hydrocarbon contaminated soils. In the current study we investigated the hypothesis that hydrocarbon contamination induces water repellency and reduces moisture retention and saturated hydraulic conductivity in inherently wettable tropical sandy soils. The objectives of the study were; (1) to compare the water droplet penetration test (WDPT) to the molarity of ethanol droplet (MED) as water repellency tests, (2) to investigate whether hydrocarbon contamination induces water repellency and changes in soil hydraulic properties, and (3) to evaluate the performance of pedotransfer functions for soil moisture retention curve and saturated hydraulic conductivity.

2. Materials and methods

2.1. Description of study sites

The study was conducted on two field sites in Zimbabwe; Ruwa (E 031° 13′ 04.0″, S 17°52′ 52.7″, altitude: 1521 m asl) and Goromonzi (E 031° 24′ 10.9″, S 180° 07′ 54.0″, altitude: 1609 m asl). The sites were located along the Mutare highway, approximately 10 (Ruwa) and 30 km (Goromonzi) from Harare, the capital city of Zimbabwe. The highway links Zimbabwe to the international seaport of Beira in Mozambique, and is frequently used by oil tankers for the transport of oil and other petroleum products. The two sites were approximately 10 km apart, and had similar soils, vegetation types and climatic conditions. The climate of the area is tropical, characterized by distinct warm wet summers (27 °C) and cool dry winters (17.5 °C). Average annual rainfall is about 800 mm, occurring mainly in summer stretching from November to February. Soils are predominantly in-situ sands derived from granites. They are classified as Harare 6G.2 according to the Zimbabwe soil classification system, corresponding to Udic Kandiustalf (USDA, 1994) and Gleyic Luvisol (FAO, 1988) (Nyamapfene, 1991). Natural vegetation in the study area is miombo woodlands consisting of deciduous trees and a grass understorey.

Two sampling sites representing hydrocarbon-contaminated and uncontaminated soils (control) were selected within each study site. Contaminated soils were selected from sites that experienced a large spillage of petroleum hydrocarbons through road accidents involving oil tankers. Petroleum hydrocarbon contamination occurred in 2007 at Ruwa and 2012 at Goromonzi. Sampling was conducted between January and April 2012, approximately 5 and 1 year later, respectively. No clean-up or remediation was conducted at the Ruwa site, while at Goromonzi, post-contamination remediation involved