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Review article

The Role of Soil Clays in Mitigating or Exacerbating Impacts to Fertility in Crude Oil-contaminated Sites

Gerónimo Álvarez-Coronel[1], Verónica-Isidra Domínguez-Rodríguez[1]*, Randy Howard Adams[1], David Jesús Palma-López[2] and Joel Zavala-Cruz[2]

[1]*División Académica de Ciencias Biológicas, Carretera Villahermosa-Cárdenas Km 0.5, Villahermosa, Tabasco, C. P. 86150, México*

[2]*Colegio de Postgraduados, Campus Tabasco, Periférico Carlos A. Molina s/n. AP 24, C. P. 86500, Cárdenas, Tabasco, México*

ABSTRACT

About two-thirds of crude oil is produced in countries with tropical and subtropical climates. Many sites in these regions have been threatened by oil spills that can adversely affect soil physical, chemical and biological properties. In some tropical countries, such as Mexico, Venezuela, India, and Nigeria, studies have been conducted to evaluate the effects of petroleum spills on soil fertility, often by monitoring pasture germination or contaminant toxicity. It has been observed that most common impacts to petroleum-contaminated soil occur by two mechanisms: a) by the formation of a thin layer of hydrocarbons on soil particles that results in a reduction in field capacity and causes soil water repellency; and b) by the formation of macro-aggregates (agglomeration) of fine soil particles into coarse particles, thus causing compaction and reduced porosity in the soil. In these studies, it appears that the type and quantity of soil clays influence how severe these impacts may be, being mitigated in the presence of higher contents of smectite clays and being more intense in soils with other fine materials (silts, kaolinite clays, Fe/Al oxides). However, these results have been observed as circumstantial evidence in natural soils. To better understand the relationship between petroleum hydrocarbons and soil clays, an artificial soil system is suggested in which the type and amount of soil clay can be controlled.

Keywords: Compaction, kaolinite, petroleum, smectite, tropics, toxicity, water-repellency

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E-mail addresses:
ing.alvarezcoronel@gmail.com (Gerónimo Álvarez-Coronel)
tazvro@gmail.com (Verónica-Isidra Domínguez-Rodríguez)
drrandocan@hotmail.com (Randy Howard Adams)
dapalma@colpos.mx (David Jesús Palma-López)
zavala_cruz@colpos.mx (Joel Zavala-Cruz)
* Corresponding author

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INTRODUCTION

In this article, the interaction between the type and amount of clay in soil, with respect to negative impacts to soil fertility caused by petroleum hydrocarbon contamination was discussed, especially with respect to water repellency, compaction and toxicity, and a strategy for investigating these interactions systematically in an artificial soil system was proposed.

Petroleum Production in Tropical and Subtropical Regions and Environmental Regulation

The petroleum industry one of the industries that causes considerable impacts to agriculture and livestock raising (Palma-Cruz et al., 2016). According to the International Energy Agency (IEA) (2016), in 2016 almost two-thirds of petroleum was produced in countries with tropical and subtropical climates (Figure 1). These include countries in the tropics with land-based operations and nearby areas with subtropical climates and petroleum production.

In Mexico, as in other parts of the world, environmental regulations are only developed with respect to the concentration of hydrocarbons in the soil, without regard to the types of soil and the potential impacts (Hernández-Valencia & Mager, 2003; Louisiana Department of Nature Resources [LDNR], 1986; McMillen et al., 2002; Michelsen & Petito Boyce, 1993; Secretaria de Medio Ambiente y Recursos Naturales [SEMARNAT], 2013). This focus on hydrocarbon concentration exclusively, is

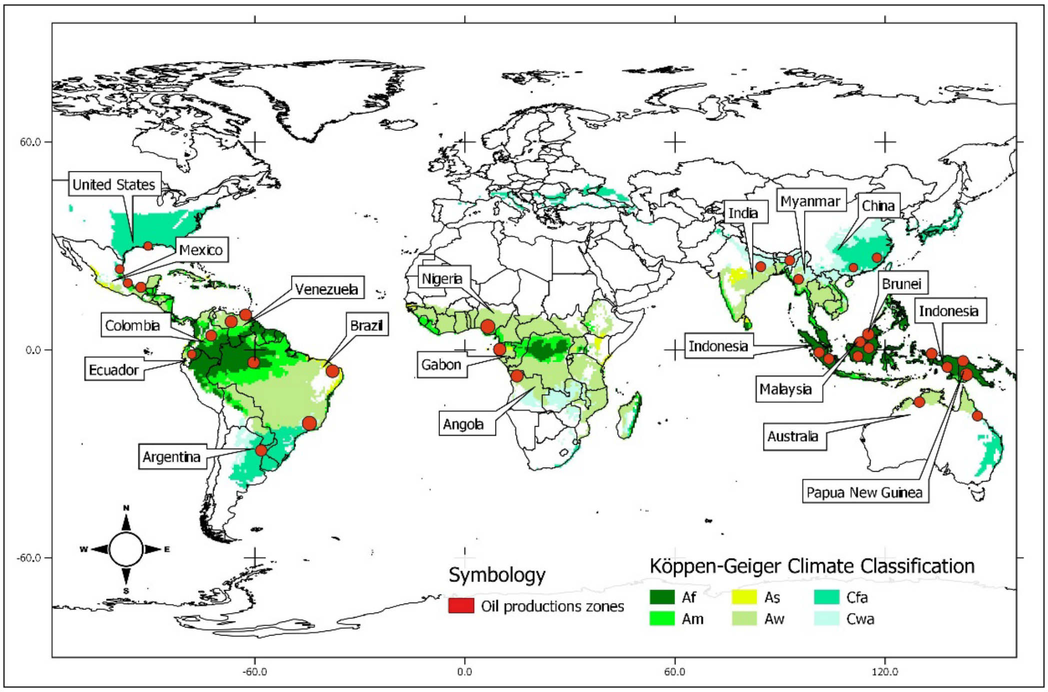


Figure 1. Petroleum producing areas in tropical and subtropical regions. Adapted from Rubel and Kottex (2010) using QGIS ver. 3.8. Open code free access to geographical information system (GIS)

based on the supposition that the primary impacts of petroleum in soil are toxicity and potential to leach hydrocarbons to groundwater. However, it has been shown that there are other impacts to soil fertility caused by petroleum hydrocarbons that affect the ability of the soil to maintain a vegetative cover and to be agriculturally productive (Adams et al., 2015; Guzmán-Osorio & Adams, 2015; Marín-García et al., 2015). It is worth mentioning that the personnel in environmental agencies are generally ignorant of the behavior of hydrocarbons in the soil. Thus, essential criteria for the conservation of fertility in petroleum contaminated soils have been overlooked when proposing methods for evaluating site contamination and the effectiveness of remediation projects.

Effects of Petroleum on Soil Surfaces and Significance for Fertility in Tropical Regions

Certain molecules in petroleum are of very low toxicity, but cause changes in the chemical and physical properties of the soil. They are more prevalent in heavy crude or old spills with weathered oil (Adams et al., 2008a). These contaminants cover soil particle surfaces, interfering in the normal soil-water-plant relationship, causing water repellency and reduced moisture content at field capacity.

Tropical regions have intense sunlight, high temperatures and humid climates, favorable for the natural processes involved in petroleum weathering–volatilization, photolysis, partial biodegradation, chemical oxidation/condensation, and sequestration in soil clays and organic material. Thus, in the tropics these kinds of hydrocarbon molecules are most likely to be produced, have the greatest effect on soil surfaces and fertility, and negatively impact agriculture and cattle-raising (Adams et al., 2008a; Ndimele et al., 2018).

In Mexico, Venezuela, India, and Nigeria research has been carried out to evaluate the effects of petroleum on soil fertility using a grass seed-germination bioassay or contaminant toxicity assay (Barua et al., 2011; Hernández-Valencia et al., 2017; Osuji & Nwoye, 2007; Oyedeji et al., 2012; Vázquez-Luna et al., 2010). This helps in understanding the behavior of these contaminants in soil, and may be an important conceptual tool for decision making concerning the remediation processes of contaminated sites in the tropics. The magnitude of the impacts depends on not only the type and concentration of the oil spilled (Ataikiru & Okerentugba, 2018; Ndimele et al., 2018), but also the kind of soil (De Silva & van Gestel, 2009; Marín-García et al., 2015) (Table 1).

Soil types presented in Table 1 can also be found in other tropical petroleum-producing areas such as the Faja del Orinoco in Venezuela (Hernández-Valencia et al., 2017); in the Niger River Delta in Nigeria (Osuji & Nwoye, 2007); in the upper Assam region in India (Barua et al., 2011); in the southern part of Sumatra (Indonesia) (Yudono et al., 2010); and in regions of Borneo and Papua.

Table 1
Common soils contaminated in the petroleum-producing zone of Southeastern Mexico

Soil type		Description	Clay	
USDA	WRB		%	Type
Psamment	Arenosol	Coarse texture, high permeability, low nutrient storage capacity. Found principally in coastal zones (dunes).	Very low (< 3)	NA
Fluvent	Fluvisol	Deep soils with good permeability, medium texture and poor soil horizon development, good superficial drainage, high in nutrients and organic material. Found principally in natural river levees.	Medium (~40 %)	2:1
Vertisol	Vertisol	High concentration of expandable clays. Principally in tropical and subtropical climates. Vegetation is predominately pasture for cattle and/or forest.	High (~60-65%)	2:1
Aquent	Gleysol	Saturated with water for the major part of the year, shallow water table. From low-lying areas in alluvial plains. High organic matter and nutrients.	High (~50-55%)	2:1
Ultisol	Acrisol	Strongly acidic, degraded soils, low base saturation at depth, high concentration of low reactivity clays. Found principally in humid tropical and subtropical areas.	High (~40-50%)	1:1

Note. USDA = United States Department of Agriculture (2014), WRB = World Reference Base for Soil Resource (2014)

It is a key to consider that petroleum-contaminated soils are affected principally by a) by the production of thin hydrocarbon laminates on soil surfaces, that produce reduced field capacity and water repellency; and b) by the formation of macro-aggregates, from the agglomeration of smaller soil particles, affecting porosity and compaction (Figure 2). The importance of soil clays on these processes are discussed in the following sections.

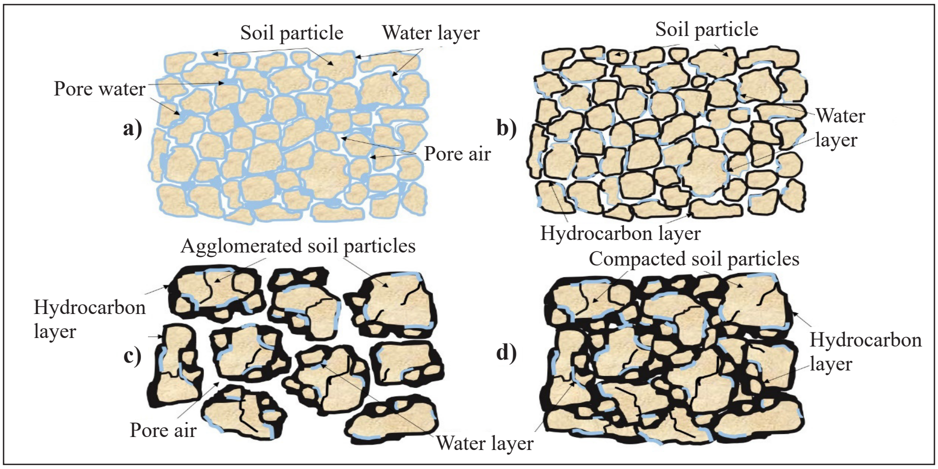


Figure 2. Representation of crude oil impacts to soil: a) clean soil at field capacity; b) contaminated soil-formation of hydrocarbon laminates on particle surfaces; c) formation of macro-aggregates – agglomeration of fine particles; d) compaction of contaminated soil-union of macro-aggregates due to an external force

Importance of Clays in Soil Water Repellency (Production of Crude Oil Laminates)

Soil water repellency prevents soil from moistening (principally at the start of the rainy season), and interrupts the free flow of water through the soil, affecting field capacity and moisture content. The principal cause of repellency is the accumulation of hydrophobic substances on particle surfaces (Hajabbasi, 2016; Jaramillo, 2006). Principally, products resulting from the partial decomposition of vegetable matter (Doerr et al., 2000); due to fires (DeBano, 2000; Dekker & Ritsema, 2000); as well as the contamination from petroleum (Marín-García et al., 2015; Roy & McGill, 1998).

Fertility loss in petroleum contaminated soil begins when hydrocarbons cover soil surfaces (Akinwumi et al., 2014; Litvina et al., 2003; Osuji & Nwoye, 2007; Walter & Omasirichi, 2015). These form a thin layer of hydrocarbons that generate water repellency and reduce the soils' ability to be moistened (Adams et al., 2008b). The normal soil-water relationship is altered and plant productivity affected by the obstruction of roots due to the hydrocarbon layer (Barua et al., 2011; Hernández-Valencia et al., 2017; Li et al., 1997; Quiñones-Aguilar et al., 2003; Figure 2b).

Litvina et al. (2003) developed a conceptual model for this phenomenon in petroleum-contaminated soil (Figure 3). The hydrocarbon layer comprises several components. The mineral component of the soil is overlain and to some extent intermixed with the natural organic (plant-derived) components in the soil, by the interaction between the negative charges in soil clays and the positive charges in the soil organic matter (SOM). The SOM is then overlain with partially degraded petroleum hydrocarbons. During the chemical or biodegradation of the hydrocarbons, polar functional groups are produced, such as alcohols, ketones, aldehydes and carboxylic acids. These groups then interact by hydrogen type bonds with similar functional groups in the SOM. The non-polar parts of

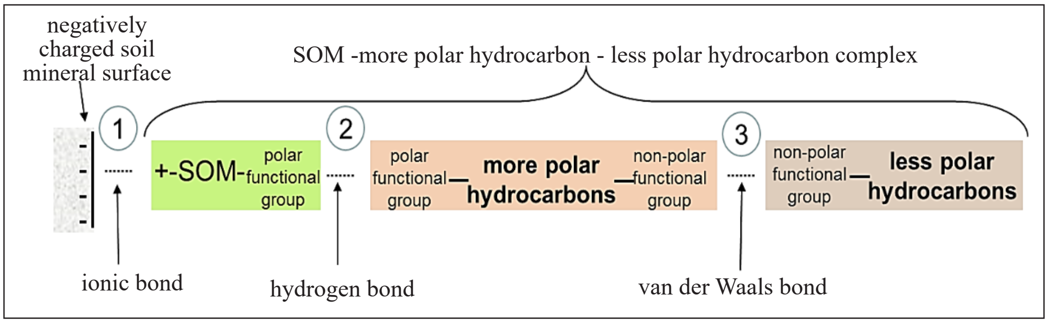


Figure 3. Interaction between soil and petroleum: the soil organic matter – hydrocarbon complex. Developed from Domínguez-Rodríguez and Adams (2011). SOM: Soil Organic Matter. 1) Negative charges in clays are attracted to cationic groups in the SOM. 2) Polar groups in the SOM are attracted by hydrogen-type bonds to polar groups in partially oxidized hydrocarbons. 3) The non-polar functional groups in the partially oxidized hydrocarbons are attracted to most hydrocarbons in the mixture, which do not have oxidized groups, by van de Waals type bonds

the partially biodegraded hydrocarbons are then free to interact with the vast majority of non-polar hydrocarbons in the mixture of contaminating oil – by van der Waal forces.

In this model, soils contaminated with hydrocarbon mixtures rich in polar functional groups (such as very heavy crude or weathered oil), would be more apt to from extensive hydrocarbon laminates on the soil surfaces and present more severe water repellency. This is because these kinds of hydrocarbon mixtures have more of the "bridging" compounds that bind the non-polar hydrocarbons to the SOM more effectively. This tendency was confirmed by Morales-Bautista et al. (2016). In agreement with this, the SOM-hydrocarbon complex can be de-stabilized by applying solutions with soluble cations that displace the complex from soil surfaces and thereby reduce or eliminate water repellency. This was confirmed by Alejandro Álvarez (2015) and Contreras-Pérez (2017). These researchers used solutions of sodium hydroxide to restore wettability to oil-contaminated soil, reducing water repellency by up to 75%.

Another reason for soil water repellency mentioned in the literature is forest fires (Ferreira et al., 2005). With more intense fires, soil water repellency becomes more severe and spatially uniform. In contaminated soils, water repellency is more common after catastrophic spills and fires or extended dry periods (Adams et.al. 2008a; Roy & McGill, 1998). This appears to be related to the production of more polar hydrocarbons from the fire itself, and/or the elimination of water from the soil due to evaporation, and the subsequent and intense binding between hydrocarbons and soil surfaces (without an intervening water layer impeding this binding).

Adams et al (2008a, 2015) also found that one of the consequences of soil water repellency was a reduction in field capacity. The hydrocarbons interfered in the interaction between soil particles (especially clays) and water due to the formation of a thin layer of the contaminant on the surfaces, thus reducing the ability to retain water. They also found a negative and lineal correlation between contaminant concentration and field capacity.

Likewise, other authors (Ávila Acosta, 2011; de la Cruz Morales, 2014; Marín García, 2012; Montero Vélez, 2016; Morales Bautista, 2014) have studied the impacts of diverse types of petroleum in different types of soil (Table 2). Among the impacts found, water repellency was common and was strongly associated with the type of petroleum as well as soil type, principally the type and amount of clay in the soil (Morales Bautista, 2014).

Although water repellency is more common in sandy soils (Roy & McGill, 1998), it can also occur in clayey soils. Marín García (2012) found water repellency in a Vertisol contaminated with light, medium and heavy crude (1-8%) and the repellency could be calculated according to the concentration and the type of oil (°API, also known as API degrees or API gravity - a classification system of the American Petroleum Institute). With light crude, the effects were very moderate (only slight repellency at 8%). However, with heavy petroleum, a strong repellency was found, even at a lower concentration (2%). The heavier crudes, contained more hydrocarbons with polar functional groups serving as chemical "bridges" between the SOM (which also had charged/polar functional groups) and the majority of (non-polar) hydrocarbons in the petroleum, thus enabling more a complete covering of the soil surfaces and more severe water repellency.

The clays, most prevalent in this soil (Vertisol), are smectites, with high shrink-swell capacity. The degree of water repellency was less in this soil than in a medium-textured soil with the same kind of clays (Fluvent) (Morales Bautista, 2014). Thus, with increasing amounts of smectite clays, the intensity of water repellency is reduced. A possible explanation: as the smectites shrink and swell, the thin layer of hydrocarbons on soil particles is ruptured, permitting more contact between the (wettable) mineral surfaces of the soil and water.

With respect to clayey soils with kaolinite, de la Cruz Morales (2014) found that in an Ultisol, the severity of water repellency increased with higher concentrations of crude, and with heavier crudes, coinciding with results reported by Ávila Acosta (2011) and Marín García (2012). Although the kind of clay predominant in this kind of soil (kaolinite) is not expandable and therefore generally has less surface area than in Vertisols, they have more surface area than in sandy soils (Psamments), and tend to present water

Table 2
Examples of studies related to the effects of petroleum on soil fertility

Soil	Petroleum	Concen-tration	Objetive	Scale	Reference
Psamment (Arenosol)	Light, Medium, Heavy and Extraheavy	1, 2, 4, and 8%	Evaluate petroleum type and concentration on soil fertility/ toxicity	Labor-atory	Ávila Acosta, 2011
Vertisol	Light, Medium, Heavy and Extraheavy	1, 2, 4, and 8%	Evaluate petroleum type and concentration on soil fertility/ toxicity	Labor-atory	Marín García, 2012
Psamment (Arenosol), Aquent (Gleysol) and Fluvent (Fluvisol)	Medium, and Heavy	0.25, 0.5, 1, 2, 4, and 8%	Evaluate petroleum weathering on soil properties	Experimen-tal patio (open air)	Ávila Acosta, 2014
Vertisol, Aquent (Gleysol) and Fluvent (Fluvisol)	Light, Medium, Heavy and Extraheavy	1, 2, 4, and 8%	Evaluate the type of petroleum (°API) vs. negative effects on soil fertility	Laboratory	Morales Bautista, 2014
Ultisol (Acrisol)	Light, Medium, Heavy and Extraheavy	1, 2, 4, and 8%	Evaluate petroleum type and concentration on soil fertility/ toxicity	Laboratory	de la Cruz Morales, 2014

repellency in an intermediate range. With light crude, the water repellency went from slight (1% oil) to severe (4% and 8% oil). With medium crude, at only 1% oil there was already severe water repellency and with heavy crude at 1% oil there was very severe water repellency. In general, there has been greater water repellency with heavier crudes than with lighter crudes and more water repellency at high concentrations of oil, but the degree of water repellency also depends upon the type and amount of clay in the soil (Table 3).

Water repellency is presented in its two most common manifestations: persistence and severity. Persistence refers to how long it takes a drop of water to penetrate dry soil. It is measured in seconds and reported as the Water Drop Penetration Time (WDPT).

Severity refers to how much surfactant is needed to overcome the water repellency, using ethanol as a week surfactant. The Molarity Ethanol Drop (MED) value refers the concentration (molarity) of ethanol in water that permits a drop of solution to penetrate the soil in ≤10s. In general, soils with no or low clay contents (for example Psamments) have very little surface area, which is completely covered with oil even at low hydrocarbon concentrations, showing high persistence (long times for water penetration). As in Table 3, the Psamment showed extreme persistence and a severity classified as very severe. However, under more moderate hydrocarbon concentrations, this persistence is easily overcome by relatively low concentrations of surfactants, since very little surface area is involved (Adams et al., 2008b). Conversely, soils with high clay contents (such as Vertisols or clayey Aquents) tend to show much lower persistence, but once affected, much higher

Table 3
Concept table on the degree of water repellency (WDPT and MED) with different types and quantities of clay

Crude oil	Type of Water Repellency	Quantity of clay and soil type			
		Very low (Psamment/ Arenosol)*	Medium (Fluvent/ Fluvisol)**	High (Vertisol, Aquent/ Gleysol)***	High (Ultisol/ Acrisol)****
Light	WDPT	Extreme[a]	Null[a]	Null[a]	Extreme[a]
	MED	Very severe[b]	Low[b]	Null[b]	Very severe[b]
Medium	WDPT	Extreme[a]	Slight to strong[a]	Slight[a]	Strong to extreme[a]
	MED	Very severe[b]	Low to severe[b]	Low to very severe[b]	Low to very severe[b]
Heavy	WDPT	Extreme[a]	Slight to strong[a]	Slight to strong[a]	Extreme[a]
	MED	Very severe[b]	Low to very severe[b]	Very severe[b]	Very severe[b]
Extra-heavy	WDPT	Extreme[a]	Null to strong[a]	Null to strong[a]	Severe[a]
	MED	Very severe[b]	Low to very severe[b]	Null to Very severe[b]	Very severe[b]

Note. *Ávila Acosta (2011), **Ávila Acosta (2014) and Morales Bautista (2014), ***Marín García (2012) and Morales Bautista (2014); ****de la Cruz Morales (2014), WDPT = water repellency persistence, MED = water repellency severity, a = water repellency persistence, b = water repellency severity

severity. This is especially pronounced in soils with 2:1 clays (expandable) due to the internal surface areas available in addition to the external surfaces (such as in Vertisols, Aquents, and to a lesser degree, Fluvents). Meanwhile, in clayey soils that are not expandable (in Ultisols, for example, with high kaolinite contents), the lack of internal surfaces in the clays appear to reduce the ability of the soil to overcome water repellency. Although not as repellent as soils without clays (Psamments), in comparison to alluvial soils, they are much more likely to develop both persistence and severity.

In a comparison between alluvial soils (Fluvent, a Vertisol and an Aquent), Morales Bautista (2014) found that the Vertisol and Aquent showed much less impact from petroleum to soil water repellency. The proposed reason was the shrink-swell capacity of the 2:1 clay being more abundant in the Vertisol and the Aquent. The frequent shrinking and swelling in these soils were postulated to break apart hydrocarbon laminates and expose hydrophilic soil surfaces, thus reducing water repellency. Therefore, soils with higher amounts of smectite clays tend to show less water repellency, congruent with that observed by Marín García (2012).

In summary, soils without clays (Psamments) are most vulnerable, followed by soils basically without 2:1 clays (Ultisols), while soils with moderate amounts of 2:1 clays (Fluvents) tend to have moderate water repellency when contaminated, and soils with abundant 2:1 clays (Vertisols and clayey Aquents) tend to have little to no water repellency when contaminated by petroleum hydrocarbons.

Compaction (Particle Agglomeration) in Clayey Soils

Hydrocarbons also result in the formation of aggregates caused by the viscosity of hydrocarbons adhered to particle surfaces (Marín García, 2012). Adams et al. (2008b) explained how the chemical structures of residual hydrocarbons might have "sticky" terminals that act as agglomeration agents, uniting fine particles (clays) into larger (sand-sized) particles, causing soil compaction. During petroleum weathering, alcohols, keto-groups, aldehides and carboxylic acids, are produced, similar to the kinds of functional groups common in asphaltenes. These give the resulting hydrocarbon mixture strong binding characteristics. According to Montero Vélez (2016), contamination with petroleum mixtures with these kinds of compounds results in permeability changes and percolation of water through the soil (Figures 2c and 2d). A conceptual model of this is shown in Figure 4. As shown, the partially oxidized hydrocarbons play a crucial role in both the formation of hydrocarbon laminates and agglomeration of small particles into larger clusters.

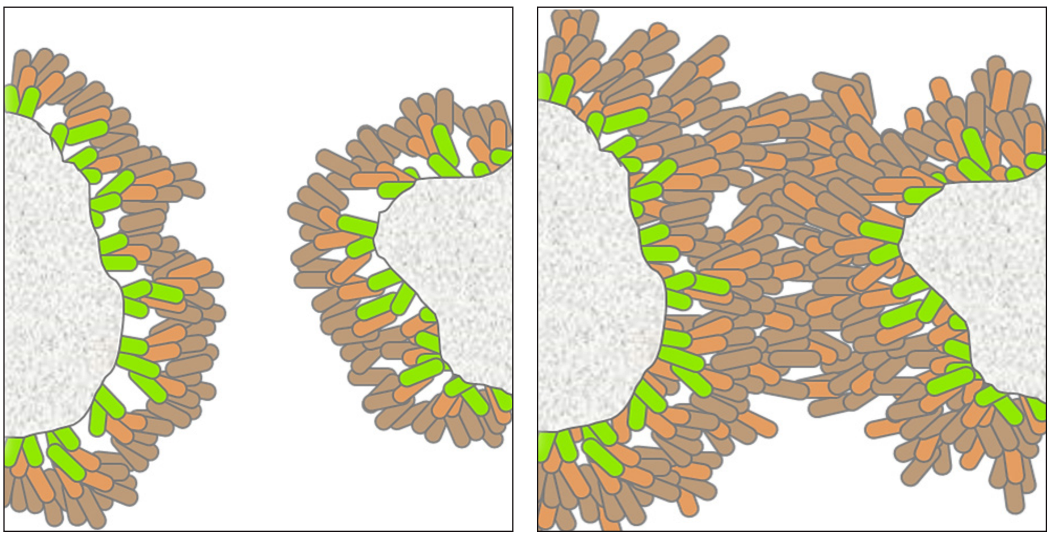
Compaction results in increased bulk density and reduced pore spaces, reducing the movement of water and air, and limiting biological activities (Barik et al., 2011; Hajabbasi, 2016; Nawaz et al., 2013). In agricultural soils, compaction is caused by the constant movement of heavy machinery

(Adams et al., 2008b; Palma-López et al., 2007), or by intensive hoof pounding by cattle.

Nonetheless, in soils with petroleum contamination, this problem intensifies. This is due to the sticky ends in asphaltenes, resins and polar compounds found in weathered oil that increases the viscosity of the oil and its adherence to soil, thus generating agglomeration between soil particles (Adams et al., 2008a, 2008b, 2015).

Nawaz et al. (2013) mentioned compaction as physical soil degradation causing changes in soil structure and the soil-water-air dynamic, as well as reduced plant growth from less root penetration, lowering crop production (Adams et al., 2015; Batey, 2009; Suuster, 2011; Trujillo-Narcía et al., 2012). Likewise, Adams et al. (2015), found a 56% reduction in biomass production in petroleum-contaminated soil. This was attributed to compaction from the oil caused by the more polar compounds found in the petroleum hydrocarbon mixture (Batey, 2009).

Others, such as de la Cruz Morales (2014), Morales Bautista (2014), and Montero Vélez et al. (2016) found a tendency in petroleum-contaminated soils where the degree of the negative compaction impacts depended not only on the type of oil (light, medium and heavy) and its concentration, but also the type of soil, principally to the type and quantity of clays. Nawaz et al. (2013) showed that soil compaction varied based on the type of soil, principally, soil particle size. For example, soils with



(a) (b)

Figure 4. Formation of hydrocarbon laminates on soil surfaces and particle agglomeration by petroleum. From the Litvina et al. (2003) conceptual model. a) formation of laminates and water repellency; b) agglomeration of small particles into larger clusters. Partially oxidized hydrocarbons are key to these interactions, forming chemical bridges between the soil organic matter and the mass of non-oxidized hydrocarbons, thus facilitating the laminates formation even at low concentrations (a). Also, due to their asphaltene-like nature, they are strong binders of particles into larger agglomerations at higher concentrations (b)

kaolinites (for example Ultisols) tend to compact naturally, while soils with smectites (Vertisols, clayey Aquents, and to a lesser extent Fluvents) tend to expand (constant shrink-swell). Due to these different intrinsic properties of soil clays, industrially they find different uses. Kaolinites are used for the production of ceramics and bricks, while smectites are employed as agglomerating agents for heat resistant materials (Díaz Rodríguez & Torrecillas, 2002) and in the petroleum industry, for oil well sealers.

As mentioned, compaction problems can be more be severe with heavier crudes. de la Cruz Morales (2014) observed that in an Ultisol (1:1 clays), compaction problems seemed greater with heavier crudes, suggesting that the type and concentration of petroleum also plays an important role. Although potentially almost all soils could suffer compaction, in contaminated soils it appears to be much more problematic in Ultisols contaminated with heavy crude or weathered oil.

Toxicity in Clayey Soils

Some researchers (Ávila Acosta, 2011; de la Cruz Morales, 2014; Marín García, 2012; Montero Vélez, 2016; Morales Bautista, 2014) have found that the type of soil plays a key role in the toxic impact of chemical compounds spilled in soil. Petroleum is a complex mixture of compounds mostly containing hydrogen and carbon, but also small amounts of nitrogen, sulfur, oxygen and some metals. The hydrocarbons cause alterations in soil by several mechanisms: direct toxicity to soil organisms, reduction in soil humidity/nutrient availability, and changes to pH and salinity (Adams et al., 2008b). The toxicity varies according to the type of petroleum. The lighter hydrocarbon fractions (with lower molecular weight) are more toxic, while the heavier fractions (with higher molecular weight) present less toxicity (Adams et al., 2008b; Tang et al., 2011).

For example, Ávila Acosta (2011) found that in a Psamment, the toxicity of four kinds of petroleum increased with concentration. The most toxic contaminant was light crude at 2-8%, followed by medium crude at 8%, while heavy crude did not show toxicity even at the 8% level. Thus, the lighter fractions cause greater toxicity, and this diminishes as the oil type becomes heavier. However, Marín García (2012) found that in a Vertisol, light, medium, heavy and extra-heavy crudes did not produce toxicity at concentrations of 1-8%. These kinds of soils with 2:1 clays (smectites) have a much greater surface area and large shrink-swell capacity that provide for a higher adsorption. Likely, this characteristic mitigated toxicity, since the petroleum hydrocarbons were adsorbed onto the soil clays and therefore, less bioavailable. In contrast, Ávila Acosta (2011) found that in a Psamment, a coarse-textured soil with very low clay content (thus low surface area), much higher toxicities were found.

Likewise, de la Cruz Morales (2014) found that in an Ultisol, only light crude caused toxicity in concentrations of 2-8%, while the medium, heavy and extra-heavy crudes did not cause toxicity. In this kind of soil, the clays are not expandable (type 1:1), with a limited surface area. However, they do have greater surface area than sandy soils (like Psamments). This may help adsorb contaminants and thus show lower toxicity than that found by Ávila Acosta (2011). In these studies tendencies are observed: toxicity depends on the type of soil, with soils with very low quantities of clays (Psamment) showing the highest toxicity, kaolinite-rich soils (Ultisol) with intermediate toxicities, and smectite-rich soils (Vertisol) with relatively low toxicities.

Lastly, Morales Bautista (2014) found that among alluvial soils (Fluvent, Vertisol, and Aquent), the greatest toxicity was observed in the Fluvent contaminated with light and medium petroleum. At only 1% of light or medium crude, the Fluvent showed considerable toxicity, while the Vertisol and the Aquent did not show toxicity at this level. This is congruent with that reported by Marín García (2012) for a Vertisol. Likewise, Morales Bautista (2014) found that with increasing clay content, the toxicity decreased, as also found by Ávila Acosta (2011), Marín García (2012), and de la Cruz Morales (2014). These authors observed a tendency in which the soil texture played in important part in toxicity, such that among the alluvial soils, the most affected by toxicity was the soil with the least amount of clay, in contrast to the Vertisol and Aquent (with a high clay content).

Plant Yields in Clayey Soils

Although fertility is one of the soil qualities that leads to the necessary conditions for plant development, this may be strongly affected by physical-chemical changes in soil. One of the principal causes is oil spills, which reduce plant yields. This effect was shown by Montero Vélez (2016), who observed a reduction in plant yield of Humidicola grass (*Brachiaria humidicola*) in several soils: Psamment, Fluvent, Vertisol, Aquent, and Ultisol, all contaminated with 1% of heavy crude. In these soils, reductions in pasture production were found to be 22%, 51%, 23%, 10%, and 67%, respectively.

Montero Vélez (2016) considered that the relative amount of reduction in plant yield was associated with the following: 1) the impact is relatively low in sandy soil (Psamment), basically without clay and few problems from compaction; 2) relatively low impacts in soils with large amounts of smectites (Vertisol, Aquent), in which the shrink-swell properties rupture hydrocarbon laminates that may form on the soil (thus mitigating problems from water repellency and compaction); and 3) relatively high impacts in soils with large amounts of fine particles other than smectites; silts in the Fluvent and kaolinte (+amorphous Fe/Al oxides) in the Ultisol.

In the last few years, other researchers have also carried out studies on the effect of petroleum on fertility in pastures. For example, Vázquez-Luna et al. (2010) found that high hydrocarbon concentrations damaged plants and reduced growth due to the hydrocarbons covering plant roots and interfering in nutrient uptake. Likewise, Hernández-Valencia et al. (2017) observed impacts in the germination of pastures

(*Megathyrsus maximus* and *Urochloa brizantha*) that was related to the type and concentration of petroleum. With higher °API (lighter oils), and higher concentrations, reduction in germination was more severe, due to the higher content of aromatics and saturates in the oil (more toxicity). Conversely, Barua et al. (2011), mentioned the inhibition of several herbaceous species (*Axonopus compressus*, *Cynodon dactylon*, *Cyperus brevifolius*, and *Eclipta prostrate*), and considered that it was due to: 1) reduced pore space in the soil that makes gas exchange more difficult; and 2) to the hydrophobic properties of petroleum covering seeds, acting as a physical barrier, reducing oxygen and water availability, and reducing gas interchange. This is in agreement with Osuji and Nwoye (2007): the partial covering of soil particles with hydrophobic hydrocarbons could reduce the soil water retention capacity (field capacity) due to a significant reduction in clay bonding.

Alternatively, Morales-Bautista et al. (2016) indicated that the relative amount of polar functional groups in the petroleum caused compaction-agglomeration, reduction in field capacity, reduction in cationic exchange capacity and the formation of soil water repellency. These functional groups are present principally in heavy and extra-heavy crude. These findings are in agreement with Adams et al. (2015) and Montero Vélez (2016), who observed a larger reduction in pasture biomass (*Brachiaria humidicola*) with increasing concentrations of extra-heavy crude. This could be from soil compaction interfering with root penetration, reducing the capacity of the pasture to obtain sufficient moisture and nutrients. Likewise, Oyedeji et al. (2012) mentioned the reduction in plant height and thickness (*Abelmoschus esculentus*) in contaminated soil that could be due to the lack of available water, and thus the mobility and absorption of plant nutrients.

Méndez-Natera et al. (2007) found that petroleum covered root surfaces in a thin layer, altering the absorption of water and nutrients, and reducing plant growth (reduction in respiration and photosynthesis rates). In this sense, some research confirms that the light fraction of petroleum (naphtha) is 20 times more toxic than the heavy fraction (Chaineau et al., 1997). However, Ferrera-Cerrato (1995) mentioned that some plant species could grow in hydrocarbon contaminated soils, and furthermore, actively contributed to hydrocarbon degradation in the rhizosphere. Likewise, Zavala-Cruz et al. (2005) mentioned that pastures had phytoremediation potential. Their adaptation to petroleum-contaminated soil was able to reduce the total petroleum hydrocarbon (TPH) content by 48% after 3.5 months of cultivation with Humidicola grass (*Brachiaria humidicola*) in two soils, an anthropic Entisol and an Ultisol, with Aleman (German) grass (*Echinochloa polystachya* (H. B. K.) Hitchcok) in an Aquent, and with Egyptian grass (*Brachiaria mutica* (Forksskal) Stapf). Thus, measurement of plant growth in contaminated soils may provide better knowledge of the degree of soil fertility impacts.

Artificial Soil for Systematic Investigation of Soil Clay - Petroleum Interactions (Strategy)

Many research gaps exist in our understanding of how clays influence soil fertility in petroleum-contaminated soils. No current information can thoroughly explain how the type and concentration of clays affect plant yields in petroleum-contaminated soils. Although previous studies showed a tendency in the behavior of water repellency, compaction, toxicity and plant yield concerning the type and quantity of soil clays, these were observed in natural soils. By studying natural soils, a more precise evaluation is made of the type and magnitude of the impacts caused by petroleum in regional soils of interest. None-the-less, these are limited by the type and quantity of clays in naturally occurring soils in a particular region. To date, there has been no systematic study of how the kind and amount of clay affect common problems in petroleum-contaminated soils. The studies on natural soils do not allowing for a systematic evaluation, where the type and quantity or soil clay can be varied, to determine with greater certitude, how clays influence the mitigation (or exacerbation) of impacts in contaminated soils.

The formation of soil is very complex (Zavala-Cruz et al., 2011), and generates a heterogeneous distribution of organic compounds, minerals, water and gases. This complicates the understanding of the processes that are carried out in the soil and the interactions between components (Guenet et al., 2011). This is because it is impossible to manipulate the proportions of the different components that interact with each other in a natural soil. However, by constructing an artificial soil, one can better understand the interactions in a systematic manner.

To determine how, and in what degree, the type and quantity of soil clay affects water repellency, compaction, toxicity and plant yield with greater certainty and precision, and thus to overcome the research gap, it is necessary to study an artificial soil system. In such a system, the type and quantity of soil clay can be controlled, as well as the quantity of sand and organic matter. In this way, the interactions between the diverse petroleum types and the different types of soil can be better understood. The information generated by such a study could be a useful tool for decision making in remediation projects, and to establish soil restoration criteria and techniques.

On the international level, several authors had conducted studies with artificial soils to systematically study toxicity of different contaminants, and in which at least one soil component was controlled (De Silva & van Gestel, 2009; Shanmugasundaram et al., 2014). Other research had focused on using artificial soils as a tool for analyzing and better understanding biological processes in soil, systematically modifying at least one variable of interest (Ellis, 2004; Guenet et al., 2011). It is worth mentioning that there already exist methods for the preparation of artificial soils, proposed by various agencies, including the American Society for Testing and Materials (ASTM)

(2004), the International Organization for Standardization (ISO) (1993, 1998), the Organization for Economic Co-operation and Development (OECD) (1984), the United State Environmental Protection Agency (USEPA) (1989) that are used for acute and subchronic toxicity bioassays. One advantage of making an artificial soil is that it allows for the creation of an environment with characteristic that are similar to those present in natural soils (Ellis, 2004; Saberi et al., 2018). Furthermore, such systems can be used to evaluate future contamination scenarios such as petroleum spills.

The components of a systematic study to evaluate the interaction between clays and impacts to soil could include a system similar to one of the currently used methods for making artificial soil, for example that proposed by the OECD (Protocol No. 207, OECD 1984). According to the OECD, artificial soil is defined as a mixture of 70% fine quartz sand (50% of the particles between 0.05 – 0.2mm), 20% kaolinite clay and 10% *Sphagnum* moss, finely crushed (Hofman et al., 2009). This artificial soil was developed to be a standardized medium "similar to soil and was introduced as a substrate for acute toxicity test with earthworms (Hofman et al., 2009).

Based on this method, modifications could be included to study the impacts of petroleum in soil, according to the type and quantity of clays, as well as the type and concentration of petroleum (see Table 4).

By systematically varying the amount and type of clay in the artificial soil, as well as the type and concentration of petroleum in the soil, the role that clays play with respect to water repellency, field capacity, toxicity and plant yield can be determined. Some overall tendencies may be found that predict what kinds of soils are most vulnerable, what remediation criteria are most important to restore, and what could be the most effective remediation or restoration techniques. As seen by Montero Vélez (2016), for vulnerable soils (for example Ultisols) the concentrations of heavy crude that do not limit plant growth are very, very

Table 4
Experimental design for use of artificial soil in the evaluation of clay-petroleum interactions in contaminated soil (proposed)

Independent Variables	Constants	Dependent Variables	Method
Type of clay (kaolinite or smectite)	Quantity and type of organic matter (10% *Sphagnum* moss)	Water repellency and field capacity	MED, WDPT, FC (Adams et al., 2008a)
Quantity of clay	Type and quantity of sand (percent to complete 100%)	Compaction	Penetrometer (American Society of Agricultural and Biological Engineers [ASABE], 1999)
Type of crude petroleum (light, medium, heavy)		Acute toxicity (earthworm bioassay)	Direct contact modification of OECD (Domínguez-Rodríguez et al., 2020)
Concentration of petroleum		Plant yield	*Brachiaria humidicola* grass biomass (Montero Vélez, 2016)

low (<100 mg Kg^{-1}) and not technically achievable or economically feasible. A better strategy may be to develop techniques to overcome the soil impacts (by adding soil conditioners, for example) rather than reduce the hydrocarbon concentration.

Additionally, the results of this kind of systematic study will need to be compared with real regional soils and validated under field conditions. Such scale-ups may provide additional information to better understand and improve the conceptual or descriptive models developed using the artificial soils.

CONCLUSION

The negative impacts of crude oil on soil fertility in terms of toxicity, soil water repellency and plant yields depend not only on the type and concentration of oil spilled, but also on the type and quantity of soil clay. There is abundant circumstantial evidence in the published literature on soil compaction and the formation of soil water repellency in petroleum-contaminated soil. In soils with higher contents of expandable clays (smectites), these kinds of impacts tend to be less with fewer problems for soil fertility. These investigations have all been carried out on natural soils; however, there is a need for a systematic study in which the type and amount of soil clay can be experimentally controlled, using an artificial soil system.

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