Evaluation of two remediation techniques applied to a site impacted by petroleum production waters
Evaluación de dos técnicas de remediación aplicadas a un sitio impactado por aguas de producción petrolera

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SUMMARY

Salinity in soils is a problem that has increased in recent years, in the Mexican southeast one of the main sources associated with these effects is the oil production water, affected congenital water. Although the chemical composition changes between one site and another, it is documented that its components can cause harmful effects on health and ecosystems. To recover the vocation to use recovery areas, different treatments have been established that eliminate standardized parameters, but others that may influence soil quality are not considered. Therefore, two treatment techniques of a soil contaminated experimentally with congenital waters were evaluated, these were; cation exchange and natural attenuation, the evaluation consists in comparing the physical and chemical properties of the control soil, treated and treated after one year, some heavy metals are also determined in all of them. In the results, it is modified that the pollutant causes changes in the soil, such as, pH reduction (neutral to acid), porosity (20%), field capacity (50%) and organic matter (50%), as well as an increase in salinity (without saline to saline) and densities (10%), it is also increased that the concentrations of Pb, Zn, Ni increase. Na, Fe and V and Ca and K were reduced. Both treatments reduce salinity, but natural attenuation shows better results than cation exchange, mainly in pH, field capacity and textures, but with higher concentrations of sodium with respect to the witness in both cases, the metals are below the regulatory limit before and after the treatment, but when compared with control soils a level of increase in V and Pb is noted, because the area is immersed in oil activities, it is advisable to carry out long-term bioaccumulation studies.

Index words: *natural attenuation, cation exchange, vocation of use, impacted site.*

RESUMEN

La salinidad en suelos es un problema que se ha incrementado en los últimos años, en el sureste mexicano una de las principales fuentes asociadas a estos efectos es el agua de producción petrolera, denominada agua congénita. Aunque la composición química varía entre un sitio y otro, está documentado que sus componentes pueden causar efectos nocivos sobre la salud y los ecosistemas. Para recuperar la vocación de uso de zonas afectadas, se han establecido diferentes tratamientos que remueven parámetros normados, pero no consideran a otros que pueden influir en la calidad del suelo. Por esto, se evaluaron dos técnicas de tratamiento de un suelo contaminado experimentalmente con aguas congénitas, estas fueron; intercambio catiónico y atenuación natural,

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la evaluación consistió en comparar las propiedades físicas y químicas de suelo testigo, tratado y tratado después de un año, también se determinó algunos metales pesados en todos ellos. En los resultados, se observó que el contaminante provoca cambios en el suelo, tales como, reducción del pH (neutro a ácido), porosidad (20%), capacidad de campo (50%) y de materia orgánica (50%), así como aumento de salinidad (no salino a salino) y densidades (10%), también se observó que aumentaron las concentraciones de Pb, Zn, Ni. Na, Fe y V y se redujeron las de Ca y K. Ambos tratamientos reducen la salinidad, pero atenuación natural muestra mejores resultados que intercambio catiónico, principalmente, en pH, capacidad de campo y texturas, pero con concentraciones más altas de sodio con respecto al testigo. En ambos casos, los metales están por debajo del límite normativo antes y después del tratamiento, pero al compararlos con suelos testigos se nota un leve aumento en V y Pb, debido a que la zona se encuentra inmersa en actividades petroleras, es recomendable realizar estudios a largo plazo de bioacumulación.

Palabras clave: *atenuación natural, intercambio catiónico, vocación de uso, sitio impactado.*

INTRODUCTION

Several studies have shown the importance of soil conservation for the species that inhabit it since they obtain the nutrients necessary for its full development, however, these conditions can be altered by the discharge of external agents from the environment. Due to the magnitude of the associated effects, the anthropogenic sources are the ones that they consider of highest important (Porta *et al*., 2003). Generally, it has been observed that during the processes of manufacturing and use of natural resources, accidents occur in which spills of components succeed that can interrupt physical, chemical and biological processes of soil, for these reasons, the fertility of a site it is lost and, consequently, there is low access feeding or negative effects on public health (Becerra-Castro *et al.,* 2015). Also, there are other investigations that have documented negative impacts in areas surrounding facilities of the different productive sectors, many of them have not been served for a long time and, in the case of soils, the dimension of the effects depends on the conditions environmental of each particular site (Adams *et al*., 2008a; Labra-Cardón *et al*., 2012; Cuero, 2012).

In some of these regions, the most representative source of pollution are the derivatives and residues of the oil industry, of which, crude oil, fuels and water of formation or production called congenital water stand out, due to the composition they possess, different studies they are considered as dangerous, in some cases, below by normative limits (Khodaveisi *et al*., 2011; Cavazos-Arroyo *et al*., 2014; Ordóñez and Schroeder, 2014). The reports of the emergencies of these spills mention that most of them occur due to ruptured pipelines related to the lack of maintenance or natural disasters, but in some countries, there has recently been an increasing tendency that these are provoked, mainly during the fuel theft (Romo, 2016; Enamorado *et al*., 2016; Oswald, 2017). To remedy these sites, different proposals have been carried out, some have managed to re-establish the vocation of use, despite this, and there still problems after restoration such as toxicity and water repellency (Fawzy, 2008; Yongming, 2009; Arao *et al*., 2010). In this sense, several investigations have shown the importance of evaluating the bioaccumulation of metals and aromatic compounds after remediation work and have emphasized that the behavior of the pollutant depends on the specific conditions of each region, in some cases, the contaminants can be transferred in the middle and in this way reach the human being, there are also sites in which the hydrocarbons are below the regulatory limit and have negative impacts on the development of some plants, while others are above this limit, they do not present affectations; different researchers have emphasized that in order to restore a site, different alternatives must be evaluated and the long-term efficiency of each of them must be compared, in such a way that the information generated can serve as a decision-making tool to restore the properties of the site, optimally, feasibly and adequately (Liang *et al*., 2011; Hou and Al-Tabbaa, 2014; Bolan *et al*., 2014; Marín-García *et al*., 2016; Lu *et al*., 2017).

In México, until date no real monitoring to soil pollution, it is estimated that the annual deterioration has impacted around 64% of the surface of the national territory due to various processes, chemical degradation being an important factor, since it is considered as contamination by action of residues of chemicals compounds, many of them classified as dangerous according to NOM-052-SEMARNAT-2005, it is important to note that some of the residues of the oil industry are within this standard (SEMARNAT, 2005; Riojas-Rodríguez *et al*., 2013; Surendra *et al*., 2014). For this sector, until a few years ago who established the statutes to determine environmental impacts was the Secretary of Environment and Natural Resources (SEMARNAT, by its acronym in spanish), but after the law reform in energy matters in 2013, the Safety, Energy Agency was created and Environment (ASEA), currently this body has established guidelines to diagnose negative effects depending on the activity to be performed, however, only certain areas of interest have been studied, mainly those where there is private capital investment, in general, in the reports presented as "Environmental Base Line", some sites have been declared with presence of hydrocarbons and heavy metals, but there are no records of what are or will be the mitigation measures for each zone (Mendoza-Cantú and Ize-Lezama, 2017; Lázaro-Sánchez, 2017).

For the southeast region of this country, one of the pollutants that has been little studied is congenital water, which usually varies its components according to the source of generation and final disposal, therefore, also effects it has on soils (Li *et al.,* 2016). This waste, generated in dehydration plants or separation batteries, contains hydrocarbons in low concentrations, solids in suspension, heavy metals (mainly chromium and vanadium), high electrical conductivity associated with sodium precursor salts, whose concentration is usually variable (between 180 000 and 300 000 mg L^{-1}), all these parameters are governed by NOM 143 SEMARNAT-2003 for final disposal (SEMARNAT, 2003). However, for various reasons spills of this waste have been caused during its transport, affecting soil and water, especially in the states of Tabasco and Veracruz (Morales-Bautista *et al*., 2011; Ortiz-Salinas *et al*., 2012). To remedy the affected areas has been taken as a basis salinity, the literature states that this occurs when there are excessive accumulations of sodium precursors in living tissues, however, there are other metals that are usually present in the contaminant and also are deposited on the ground, for example, in the case of plants, some reports suggest that the response depends not only on the concentration but also on the composition of the salts present and the properties of the soil, even some species may be tolerant to these conditions and allow bioaccumulate the pollutants, in several cases of studies, it is mentioned that many of these species are used in some sites as an alternative for remediation, however, some problems have occurred in which the risk to health increases because the restored areas are used for agricultural or cattle raising, mainly due to ignorance of the population (Mulligan *et al*., 2001; Ruyters *et al*., 2011; Yao *et al*., 2012).

The majority of soil treatment techniques base their objectives according to the risk represented by the pollutant, for saline soils, in Mexico the criteria of NOM-021-SEMARNAT-2000 are used, most of them have managed to restore the main indicators to regulate levels, however, several studies show that other properties such as pH, organic matter (% MO) and field capacity (% CC) are determinants in the survival and development of a plant, as well as the metals present that can be absorbed (SEMARNAT, 2002; Jayasekera and Hall, 2007; Prieto *et al*., 2009; Dastgheib *et al*., 2011) Different lines of research agree that the relevance of studying each variable involved in the remediation systems will allow knowing the level of risk of each component and, in this way, to be able to determine the measures for the prevention of impacts when they are significant (Kadali *et al*., 2012; Morales-Bautista *et al*., 2016). there are methods to eliminate the most dangerous part of the pollutant, while others are based on stabilization and as a result the disposition to the environment is inhibited, consequently, its toxicity; all of the above can be carried out through biological, physical and chemical processes. When compared to each other, not only the cost-benefit of each of them can be evaluated, but also the long-term environmental viability (Peng *et al*., 2009; Saint-Laurent *et al*., 2010).

Due to the above, this paper addresses two techniques of remediation of soils affected by salinity, considering the effects of environmental conditions in tropical areas which play an important role in the behavior of soils, mainly, alluvial. These technologies are cation exchange (IC) and natural attenuation (AN) (Lakhdar *et al*., 2009; Tang *et al*., 2012; Etim, 2012; Song *et al*., 2017). Within the study, the soil was experimentally contaminated with congenital water, treated by these alternatives and subjected to one year of weathering in order to observe whether the variations of rainfall and temperature of the Tabasco state influence after the treatment process. As response variables, the physical and chemical properties of the control, contaminated and treated soils were evaluated, in the same way, the content of metals in congenital water and in treated soil after a period of expose to weathering.

MATERIALS AND METHODS

Targeted sampling was performed on land located between the Samaria II separation battery and the Samaria river bank, the facilities belong to Petróleos Mexicanos (PEMEX, exploration and production) within the municipality of Cunduacán, Tabasco, México; Although industrial development is evident, the area still retains the vocation of agricultural livestock (Ruiz-Álvarez *et al*., 2012); For this reason, in the margins of the oil infrastructure, the inhabitants develop activities such as grazing, planting and breeding of domestic animals, mainly for self-consumption (Gutiérrez and Zavala, 2002; Ruiz-Álvarez *et al*., 2012; Pinkus-Rendón and Contreras-Sánchez, 2012; Maldonado-Sánchez *et al*., 2016).

During the sampling, fodder grass "chontalpo" (*Brachiaria brizantha* (Hochst. Ex A. Rich.) Stapf) and trees used as a live fence, mainly of "cocoite" (*Gliricidia sepium* (Jacq.) Kunth ex Walp.), could also be identified, and some poultry and feeding cattle were also observed. The sampling was carried out using shovels and steel nucleators, three composite geological strata were taken, which were collected from the rhizosphere to the water table (70-80 cm), the locations of the points are denoted in the Table 1.

In general, the samples showed sand-clayey texture to the touch, medium humidity, high reaction to hydrogen peroxide and medium to acetic acid, colour orange mottles were observed (7.5 YR 10/8) and soil base was identified as color light-brown (10 YR 4/4) (according to the Munsell color chart), few deep roots (25-20 cm) but abundant, as well as earthworms were observed. Due to the horizons being homogenized in most of the soil remediation tasks, in the present work, composite (homogenized) samples were always analyzed. All the extracts were placed on a canvas and homogenized with shovels, a quartering was performed and three samples of approximately 3 kg each were taken, these were labeled as control soil, kept in black and fridge with ice, the rest was stored in bags and, in this way, all were transferred to the laboratory where they were treated separately. On the one hand, the witnesses were dried in a stove with a stable atmosphere, the roots and rocks were removed, and finally, they were ground and screened, instead, those of the sacks, only the roots and rocks were removed, were manually homogenized and then placed in plastic containers (45 cm diameter and, 25 cm deep) with holes in the bottom (to allow drainage of contaminants and water after each amendment) which were called experimental units (a total of nine with approximately 10 kg of soil in each) and were left so for 10 days in an indoor place.to allow the drainage of pollutants and water after each amendment

Table 1. Reference of samples of control soil.

Point	Location UTM 15 Q ± 3 m		
1	484 513.54 E	1 996 107.86 N	9 msnm
2	484 388.87 E	1 996 079.81 N	11 msnm
3	484 530.05 E	1 995 961.33 N	8 msnm

UTM = universal transverse mercator; msnm = meters above sea level.

After this time, six of the units were contaminated experimentally with approximately 15 liters of congenital water from the Samaria Battery II (UTM 15Q, 490531E, 1989895 N), it was left to rest for 10 more days and take one sample homogenized of 500 g of each were, they all joined and homogenized a quartering was made and three samples of 2 kg were taken, these were named contaminated soil.

Three of the contaminated units were treated by cationic exchange with CaO, for this, 0.5 moles of Ca were added for every 1 mole of excess Na in the contaminated soil with respect to the control. The treatment agent was added to the contaminated soil and homogenized with a spatula, subsequently washed with drinking water distributed in three amendments of five liters (one per day; Morales-Bautista *et al*., 2011, 2016). The other three, were remediated by biological method adding potable water washes (10 liters distributed in two days), subsequently, added organic matter (4% by weight with respect to contaminated soil and, with composition of 13% Nitrogen, 2% Phosphorus, 44% Potassium) homogenizing with a spatula and, finally, one last water amendment (five liters; Ebuehi *et al*., 2005; Tang *et al*., 2012). The treated soils were allowed to stand for 10 days for the water to drain; after this time, they were homogenized and from each cation exchange cell 1 Kg were taken and placed on a canvas, homogenized, a quartering was made to extract three composite samples of approximately 1 kg, these were called fresh cation exchange (ICF), all the previous, was also made for the units treated for biological method and, these were called fresh natural attenuation (ANF).

The experimental design was carried out in completely randomized blocks (Table 2), to each soil, was analyzed parameters specified in Table 3, in the same way, in Table 4 the climatic conditions to which they were subjected are shown treated soils. All the results obtained were analyzed by R-Project Statistical Computing (version 3.5.3) to assess significant differences between treatments (Boqué and Maroto, 2004).

RESULTS AND DISCUSSION

In Table 5 present the parameters determined to the congenital water of the Samaria II battery are presented according to the methods of NOM-143-SEMARNAT-2003 (SEMARNAT, 2003).

According to the results obtained from the analyzes carried out on congenital water, it is observed that the values of the components are below the maximum permissible limit (LMP) for discharge in coastal waters as established by NOM-143-SEMARNAT-2003, however, when comparing with the limits of NOM-001-SEMARNAT-1996 for discharges in this same source, it was detected that some parameters are above the LMP, these are floating matter corresponding to the suspended solids and Pb, the latter considering daily discharge. Several studies have shown that one of the problems of metals is that, although they can be in low concentrations and adequate comprehensive management is carried out through various treatments to reach regulatory, are bioaccumulables (Diya'uddeen *et al*., 2011); several reports show that in sites exposed to spill conditions or emissions of these compounds, the population is in constant risk, for example, there are jobs that mention that when dumped into the sea the species that develop there can fix metals, so the toxicity in the whole trophic chain is always latent by the biomagnification (Azetsu-Scott *et al*., 2007; Hobman and Crossman, 2015). Also, high values were observed for CE and sodium, which is why it is considered that if this waste is spilled in the soil it can be associated with salinity problems, although some reports have emphasized that these effects depend on the amount spilled and the type of water and soil (Qin *et al*., 2012; Li *et al*., 2016).

On the other hand, in Table 6 presents the results of the physical and chemical properties of the control soils, contaminated, treated and weathered both by ion exchange and by environmental attenuation.

Table 2. Experimental design.

Experimental design			
Contaminated	R1	R2	R3
Treatment 1 (IC)	R1	R2	R3
Treatment 2 (AT)	R1	R2	R3

Table 3. Parameters to be determined in soils.

Soil		Metals	
pH		Lead	Pb
CE	dS m^{-1}	Nickel	Ni
CC	%	Zinc	Zn
Texture	%	Cadmium	Cd
DA	g cm^{-3}	Chrome	Cr
DR	g cm^{-3}	Sodium	Na
Po	%	Potassium	K
CIC	Cmol + kg^{-1}	Calcium	Ca
MO	%	Iron	Fe
		Magnesium	Mg
		Vanadium	V
		Copper	Cu

CE = electrical conductivity; CC = field capacity; Textures = arenas (% A), clays (% R) and limo (% L); DA = apparent density; DR = real density; % Po = porosity; CIC = cationic exchange capacity; % MO = organic matter.

Table 4. Environmental conditions in weathering period (CNA, 2019).

Month	1	2	3	4	5	6	7	8	9	10	11	12
T	22.1	25.8	27.5	28.3	29.3	29	29.4	28.5	28.6	27.9	26.2	24.3
Pr	323.9	58.3	49.3	109.5	59.5	129.9	109.5	195.7	218.1	253.7	358.8	99.4

T is the average monthly temperature in ° C; Pr is the average monthly rainfall in mm.

Table 5. Characterization of congenital water of battery of separation Samaria II.

Component	Value	Metal	Concentration
			mg L^{-1}
Total solids	101 mg L^{-1}	Pb	0,3
Dissolved solids	95%	Ni	0,075
Sedimentable solids	1%	Zn	0,146
Total suspended soils	4%	Cd	<LD
HTP	10 mg L^{-1}	Cr	<LD
pH	6	Na	6639
DBO	9 mg L^{-1}	K	128,9
CE	140.14 dS m^{-1}	Ca	48,2
Colour	550 Upt Co^{-1}	Fe	0,877
CaCO3	420 mg L^{-1}	V	0,059
Total coliforms	990 NMP	Mg	38,475
Total chorides	40 mg L^{-1}	Cu	<LD

HTP = total petroleum hydrocarbons; COD = biochemical oxygen demand; CE = electrical conductivity; UPt / Co = units in platinum/cobalt and NMP scale = probable number and L.D = detection limit.

Based on the results of table 5, the pH of soil is passed from neutral to acid classification when contaminated with congenital waters, when remediating, both treatments favor the recovery of this indicator, however, the classification of the soil remediated by Cationic exchange is moderately alkaline and remains so after one year of weathering since no significant differences were observed when comparing them ($P > 0.05$) and may be related to the aggregate CaO; in contrast, the soil treated by attenuation presents a neutral pH in fresh and these conditions remain after weathering, when comparing these two conditions with that of the control soil, no significant differences were observed ($P > 0.05$), so it can be deduced that AN recovers this value to conditions similar to the initial ones; some investigations mention that the spillage of acidic or alkaline materials contribute to the variation of pH and CE, also affirms that these factors are determinants in the disposition of some nutrients and, although some species are usually tolerant, their development or growth can be affected, in the case of pH, but there may also be metal disposition, mainly to acidic conditions (Morales Bautista *et al*., 2011; Paz *et al*., 2014).

In this sense, the CE values were also contrasted, it was found that the control soil has negligible salinity effects, but when contaminated, the classification changes to saline soil (CE of 12 dS m^{-1}), when comparing the variations of this property after treating, it was obtained that this parameter decreases to conditions less than 4 dS m^{-1} in both cases, considering them without salinity problems. However, when contrasting these values with that of the control soil, significant differences were found ($P < 0.05$), for example, treatment by cationic exchange (CI) is 1 dS m^{-1} and decreases 0.5 dS m^{-1} after of one year of weathering, on the other hand, remediation by natural attenuation (AN) after treatment has a value of 2.5 dS m^{-1} and is reduced to a value of 1.5 dS m^{-1}; In relation to these results, in some restoration work, it has been observed that chemical treatments tend to be faster and more efficient in terms of the removal of an inorganic contaminant due to the compatibility between the treating agent in comparison with the biological agents, the latter they tend to be slower and their efficiency depends on the type of material added since all processes involved are,

Table 6. Physical and chemical parameters of control, contaminated, treated and weathered soils.

Soil	pH	CE	CC	% A	% L	% R	DA	DR	% Po	% MO
T	7.20 ± 0.09	0.13 ± 0.002	35 ± 1.11	55 ± 2.01	10 ± 0.11	35 ± 0.61	1.50 ± 0.03	1.98 ± 0.01	24.24	2.0 ± 0.05
C	5.33 ± 0.09	12 ± 0.100	18 ± 0.92	60 ± 2.11	20 ± 0.31	20 ± 0.41	1.60 ± 0.01	2.02 ± 0.02	20	0.10 ± 0.004
ICF	8.41± 0.12	1.0 ± 0.010	30 ± 0.91	80 ± 3.00	10 ± 0.29	10 ± 0.71	1.75 ± 0.03	2.21 ± 0.02	20.45	0.25 ± 0.01
ICI	8.37± 0.08	0.55 ± 0.010	29 ± 1.01	79 ± 1.99	11± 0.31	10 ± 0.11	1.80 ± 0.02	2.18 ± 0.01	18.09	0.5 ± 0.01
ANF	7.30± 0.07	2.5 ± 0.10	32 ± 1.21	58 ± 2.00	12 ± 0.25	30 ± 0.21	1.51 ± 0.01	1.99 ± 0.03	24.12	2.4 ± 0.09
ANI	7.21± 0.05	1.5 ± 0.019	33 ± 1.01	57 ± 1.89	13 ± 0.08	30 ± 0.31	1.49 ± 0.03	2.01 ± 0.02	25.87	2.3 ± 0.12

T = control; C = contaminated; ICF = cation exchange after treatment; ICI = cation exchange after one year of treatment; ATF = natural attenuation after treatment; ATI = natural attenuation after one year of treatment. The porosity calculation it was from the averages of DA and DR, by the equation% Po = 100 ((DR-DA) / DR).

mainly, metabolic; In the case of common plantings in the Tabasco region, it has been observed that certain grass such as humidicola (*Brachiaria humidicola*) is tolerant to these levels of salinity and pH, although the nutrients it provides to livestock is very low (Palma-López *et al*., 2007; Salgado-García *et al*., 2016).

When evaluating others values in contaminated soil with respect to the control, a decrease of approximately 50% CC was observed (with values close to the point of permanent wilting) and, of % MO of ~ 95%, which coincides with the results of some studies of salinity impacts in which it has been observed that sodium precursors are usually incorporated in the organic matter as well as in hydroxyl ions of the clays, also they can form complexes with the available water, in all cases there is a displacement of H^+ ions and, consequently, low water potential and destruction of organic matter is obtained mainly by nutrient entrainment, in the case of pH, it tends to be more acidic, therefore, in places frequently influenced by salts, there are usually reduction effects in Field Capacity (% CC) and Organic Matter (% MO) (Wang and Peng, 2010; Stoica *et al*., 2016). When restoring the soil, these properties are recovered according to the type of treatment, in the case of CI, % CC = 30, only ~ 14% lower compared to the control and this remains constant after one year (% CC = 29 %, $P > 0.05$), also % MO in these samples increases, the case of ICF is 0.25% and ICI = 0.5 %, both well below the value of the control, but a recovery is estimated since the values are higher compared to the contaminated sample that was 0.1%, it is worth mentioning that after the remediation, no organic material was added, but the increase in this property may be related to the removal of sodium, which allowed greater moisture retention and the establishment of some type of vegetation after one year, although it is of great benefit, the values of % MO of the soil treated by CI (very low) did not reach the same classification of the control (medium) soil according to NOM-021-SEMARNAT-2000 (SEMARNAT, 2002; Callejas-Rodríguez *et al*., 2012).

Similar to the previous, the samples treated by natural attenuation also present recovery in % CC and % MO, for example, % CC in ANF is 32% and ANI of 33%, when comparing them with each other and the control, no differences significant ($P > 0.05$); however, the values of % MO for ANF and ANI did not differ between them ($P > 0.05$), but area different with the control ($P < 0.05$) since they are around 50% higher, but they have the same classification as the control according to the aforementioned norm, stands out that 4% of % MO was added and the contaminated soil had 0.1%, so it can be associated that only 1% can remove CE to conditions below the normative limit. Several studies have emphasized the importance of organic matter in soils, most agree that most of the biogeochemical processes in soil depend on them, mainly, the survival and development of plants (López, 2000; García *et al*., 2010; Jaramillo and Restrepo, 2017), in this sense, natural attenuation has values of % CC and % MO closer to the control, the previous thing recovers importance, due to the fact that in the study area the seeding is practiced by temporary and not by assisted risk (Zavala-Cruz *et al*., 2016).

Other properties evaluated were the content of textures, densities and porosity, these parameters are intrinsically related to soil compaction and respiration (González-Cueto *et al*., 2009); under this argument, the results of apparent and real density expressed in Table 5 were contrasted, it was observed that when polluting, both have a slight increase (DA of ~ 7% and DR of ~ 2%) but with significant differences with respect to control ($P < 0.05$), when applying the treatments, cation exchange DA and DR increases ~ 17% and ~ 11% respectively in comparison with non-contaminated soil, however, the values obtained for this same variables in natural attenuation decrease and do not they change after of weathering ($P > 0.05$), when comparing these results with those of control soil; no significant differences were found ($P > 0.05$). In relation to the above, there are studies that mention that changes in soil densities after being contaminated may be related to the formation of higher molecular weight aggregates as compounds are deposited mainly in clays (Suthersan *et al*., 2016; Marín-García *et al*., 2016), these effects can be measured in the increase of densities (compaction), decrease in clay content and increase in sands (decrease in field capacity, as well as significant changes in porosity (generally decrease; Gutiérrez and Zavala, 2002; Khamehchiyan *et al*., 2007; Maldonado-Chávez *et al*., 2010); all these investigations can justify the decrease in porosity (% Po), the slight increase in sands (% A) and decrease in clays (% R) in contaminated soil with respect to the control, after the two restoration processes, it can be seen that there are different behaviors, in the case of ICF, there is a change in textures where % A goes from ~ 60% in contaminated to ~ 80% in treated and, thus, remains in ICI (79%, $P > 0.05$), reflecting in the decrease of % R (from 20% in contaminated to 10% in ICF and ICI, $P > 0.05$), in the case of % Po, this is lower by ~ 15% after treatment and drops to ~ 25% after one year, it is important to mention that the textural classification in the samples treated by CI is sandy-loam compared to the control that is loam-clay-sandy, also this phenomenon can be related that in the study zone there is forage grass so it does not trampling by livestock is ruled out (Rucks *et al*., 2004; Zamora *et al*., 2012). In discrepancy, when evaluating the results of % Po and textures of the treatment by AN, we obtained what % Po recovers to similar values of the control and, although there are significant differences in the textures ($P < 0.05$), the classification coincides with the witness. One of the great challenges of soil impact assessments is to determine the level of effects according to the time of year, mainly in alluvial soils, of which, vertisols or soils with similar properties are the ones that represent the greatest complexity, Palma-López *et al.* (2007) reports these soils for the study area, Adams *et al.* (2008b) and Morales-Bautista *et al.* (2016) mention that in these soils the effects on porosity may not be as representative, since usually show variations in the expansion and contraction of the clays according to the seasons of the year (self-tillage), but factors highlight that factors such as the content of interchangeable cations, pH and % MO, are fundamental properties for the establishment, survival and development of the plants, mainly, if the zone is of vocation of agricultural use, reason why the long-term monitoring allows to determine the behavior of the residual components deposited in the a soil matrix, especially, because depending on the environmental conditions it will be the degradation, leaching and disposal of the pollutants (Mulligan *et al*., 2001; Kavamura and Esposito, 2010).

On the other hand, the results of the metals analyze performed on the samples of control, contaminated and treated soil are listed in Table 7.

In the results of Table 7, it is observed that in all the samples there is absence of Cd, Cr, and Mn, but that, if they found the metals Pb, Zn, Ni. Fe and V, but are below the regulatory limit and, according to some reports, below the tolerance levels for some crops, when comparing the concentrations found in control and contaminated soil, significant differences were found ($P < 0.05$), this may be due to the fact that congenital water also contains these metals (SEMARNAT, 2007; Abioye, 2011; Chambi *et al*., 2012). Also, these metals coincide with the work of De la Cruz *et al.* (2012), who conducted a monitoring of nearby soils similar to the study area located in Cárdenas, Tabasco but the reported concentrations by authors, are much larger in comparation with the present work, this may be due to the fact that Cárdenas has greater anthropogenic influence than Cunduacán (study area), although there are no major comparative studies, also can be linked to some studies of atmospheric emissions in the area, which specifies what impacts for anthropogenic activities they are more ageddate in the Activo Cinco Presidentes (to which Cárdenas belongs) in compared to those of the Activo Samaria (o which Cunduacán belongs), there are also other authors who mention that the metals in soil they can increase for deposition of emissions or effluent carryover in the lower part of the hydrological basin (Pérez-Vidal *et al*., 2010; Machado *et al*., 2018).

Table 7. Metals present in contaminated and treated soils.

Soil	Pb	Zn	Ni	Cd	Cu	Cr	Mn	Fe	V	Mg	Na	Ca	K
	mg kg^{-1}									Cmol+kg^{-1}			
T	0.105	0.042	0.017	<LD	<LD	<LD	<LD	4.151	0.044	2.521	2.400	5.621	0.398
C	0.131	0.054	0.019	<LD	<LD	<LD	<LD	4.512	0.051	3.852	7.930	5.642	0.199
ICF	0.133	0.041	0.020	<LD	<LD	<LD	<LD	4.491	0.025	1.781	2.068	9.782	0.300
ICI	0.131	0.040	0.022	<LD	<LD	<LD	<LD	4.497	0.021	1.692	1.988	9.113	0.298
ANF	0.129	0.052	0.021	<LD	<LD	<LD	<LD	4.422	0.049	2.801	3.730	5.341	0.981
ANI	0.100	0.050	0.019	<LD	<LD	<LD	<LD	4.420	0.047	2.751	3.010	4.742	0.980

CV ≤ 2 %; LD = limit of detection (Pb = 0.0197; Zn = 0.0033; Ni = 0.0138; Cd = 0.0014; Cu = 0.0037; Cr = 0.0086; Mg = 0.001; Mn = 0.0020; Na = 0.0011; Ca = 0.0028; K = 0.0028; Fe = 0.0061; V = 0.048, all in mg L^{-1}).

To determine the behavior of these metals, each soil sample was compared and it was observed that each behaves differently, for example, there are no significant differences ($P > 0.05$) between the samples of Pb for C, ICF and ICI, but if with T ($P < 0.05$) since it increases by 24%, so it can be deduced that the treatment does not eliminate this metal but if it stabilizes it, the rest (Zn, Ni and Fe) present differences with T and C ($P < 0.05$), but when comparing the values with the ICF and ICI, no significant differences were found, because they are also stabilized ($P > 0.05$), however, V presents discrepancies between these samples ($P < 0.05$) with lower concentrations after weathering (~ 16%), which may indicate that the metal does not stabilize and, due to weather conditions, can be leached or absorbed by a plant. On the contrary, the treatment by natural attenuation, except for Pb, the rest of the metals do not show significant difference between C and ANF ($P > 0.05$) and this is maintained after weathering, however there are differences between the concentration of Pb of ANF and ANI ($P < 0.05$) being 23% lower in the latter, so this treatment could promote the mobility of this metal. The differences between the concentration of metal found between a treatment coincide with reports that establish that these are more leachable or are available at a more acidic pH, therefore, AN has lower concentrations than IC, the latter may contain stabilized metals in the clays or in the micelles formed by $Ca(OH)_2$; however, although this coincides with the literature, it is only a hypothesis since the weathering was done outdoors and there is no information on the content of metals in the rain, In addition to the fact that it is important to carry out a detailed study of the arrangement of the metal phases according to the variations of the properties over a longer period since some metals such as Pb and V have different trends depending on the type of treatment (Torres *et al*., 2012; Reyes-Rodríguez *et al*., 2014).

Like the rest of the metals, the interchangeable cations present in the soil were contrasted, it was observed that when contaminating Mg and Na, compared with the control they increased by 50 and 300% respectively, which can be related to the concentration in the congenital water and that the ions, in the case of the lowering of K, may be part of the organic matter that is lost when contaminated, it was also observed that there is no difference in the concentration of Ca between T and C ($P > 0.05$), when treating the soil, IC allows the displacement of the other three cations which are different to the values of the control ($P < 0.05$) (Mg, Na and K are less ~ 30%, ~ Na 14 % and K ~ 25% less than T), instead Ca is greater by ~ 75% in ICF and reaches up to 64% in ICI, these values coincide with the report of Morales-Bautista *et al*. (2011) who establish that these changes are related with the change in pH due to the physical and biological factors of the weather. However, when evaluating the changes in the concentration of these cations in the remediation by natural attenuation, it is observed that Ca in ANF is found in values similar to the control ($P > 0.05$), but drops to ~ 6% lower than T after weathering, this may also be associated with the decrease of Na that goes from ~ 55% higher to control in ANF to 25% in ANI, also Mg decreases until reaching values higher than 10% in ANF with respect to the control, this does not vary, because when comparing with ANI they do not present significant differences ($P > 0.05$), on the other hand potassium that is added in the treating material represents a 240% greater than the control in ANF but reaches values analogous to T in ANI since they did not find differences between them. For all the previously obtained, AN has higher recoveries and similar values in K and Mg to the control, and Ca and Na do not represent changes between ± 25% with respect to the control soil, in contrast, only IC represents values between this range, considering that these factors are also determinants for implementing endemic plants and that they can adapt to these variables after the restoration (Jiménez-Heredia *et al*., 2010; Ruiz-Álvarez *et al*., 2011; Trujillo-Narcía *et al*., 2012).

In summary and, based on the analysis of the results, we build Table 8 which lists the parameters recovered after weathering the treated soils in comparison with the control.

In Table 8, Although EC recovers to non-saline conditions (< 4 dS m^{-1}), it should be considered that some plants are not very tolerant to changes in this parameter, so although in AN a greater number of physical and chemical parameters are recovered, especially, CE values are not similar to the witness, even after the weathering. CC does not exceed PMP and the Na, Mg, K and Ca cations with estimates between ± 25% with respect to the control, as well as the concentration of the metals Zn, Ni, Cd, Cu, Cr, Mn and V, the LMP and tolerance levels for some plants are low, with reservations of the mobility assessment

Table 8. Parameters recovered according to the type of treatment in soil contaminated by congenital waters after one year of weathering.

Soil	pH	CE	% CC	% MO	DA	DR	% Po	% A	% R	% L	Na	Ca	Mg	K
IC	Not	Yes	Yes	Not	Not	Not	Not	Not	Not	Not	Yes	Not	Not	Not
AN	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes

considerations of the V for IC and Pb in AN. A recommendation would be to carry out a review of the plants to be planted that although they are recommended are preferably endemic, but that also do not allow the fixation of these two metals.

CONCLUSIONS

It is concluded that, when a soil is contaminated, both the contaminant and the treatment agent cause changes in the physical and chemical properties and, although remedies are made based on the reduction of the regulated parameters, there will always be others that will be affected both by the contaminants as by the components that are added during the treatment. In addition, there are other conditions that can cause negative effects, in the case of oil production waters, there are heavy metals, which although they are in low concentrations it was observed that depending on the type of remediation, they vary their availability to the environment.

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