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# Disposition of Wastewater from Oil Wells in Venezuelan Savannahs and Their Effects on Percolation Waters

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**Abstract:** In the eastern Venezuelan Llanos, the drilling fluids composed of wastewater (WW), accompanying hydrocarbons and wastes from oil activity are deposited in a system of waterproofed pits. Later, by means of siphons, the hydrocarbon phase of the WW is separated. WW are typically very salty and contain suspended and dissolved solids, residual hydrocarbons, and chemicals used in hydrocarbon extraction. So they are transported to a pit and treated with a flocculating agent and lime. Once carried out, the flocculation-coagulation operation and pH correction, WW are released to the environment. The savannas where the treated water are irrigated, in addition to the oil operation, present an intense agricultural and livestock activity, and rest on aquifers that are partially replenished with the percolation waters near the treatment pits. Therefore, this research will aim to analyze: i) the levels of soluble salts and heavy metals in the percolation waters collected in lysimeters located in savanna soils adjacent to oil wells after irrigation with WW treated with flocculating agents, and ii) the estimation of time which these salts take to be dislodged from the soil. The waters from the lysimeters did not report high levels of soluble aluminum or heavy metals since their soluble forms were precipitated by liming. Likewise, the coagulation-flocculation process removed the high levels of barium from the untreated WW. The contributions of sodium and calcium from the WW have a liming effect on the acidity of these soils. The sodium levels contributed by the WW could represent an environmental risk, fortunately, the good internal drainage of these sandy Ultisols allows their removal. Studies with lysimeters showed that the percolation waters of areas irrigated with flocculated water have a similar sodium content to that of control lysimeters six months after treatment.

**Keywords:** Sodium, Barium, Aluminum, Contamination, Savannah

## 1. Introduction

The oil industry, through its different activities, uses and generates a set of substances, many of them potential agents of environmental deterioration, so that the contamination of terrestrial ecosystems by oil or its derivatives is an event that can occur with certain frequency, either by the disposal of waste in the environment, or accidentally due to the rupture in the different containers of oil and their derivatives [1-6]. In oil well exploitation areas, disused materials and wastes are generated from oil exploration, production, refining and commercialization activities, which due to non-compliance with environmental law, become environmental liabilities. These liabilities degrade the physical, natural and social environment, causing risks to health and ecosystems [7-10].

Hydrocarbon pits are excavations carried out in the soil, which were used when there were no appropriate technologies to temporarily store effluents and waste generated by oil and gas exploration, production and refining activities. They can be classified according to their status or type of operation in: i) drilling pits (DP), excavations equipped to store drilling fluids, well drilling waste and drilling cuts (cuttings). Its surface can vary between 1500 $m^2$ and 2000 $m^2$ and its depth between 2 to 3 m, and ii) production pits, built with retaining walls, which occupy surfaces greater than 2000 $m^2$ and volumes of 7500 $m^3$, where salty water from crude dehydration processes is stored [7]. These pits contain a layer of emulsified hydrocarbon or crude that can be recovered (Figure 1). According to Arellano [7] in 2008 in Venezuela there were a total of 12367 hydrocarbon pits without cleaning, of which 37% corresponded to drilling pits. Therefore, among the environmental liabilities generated by the oil activity on land, the drilling pits stand out.

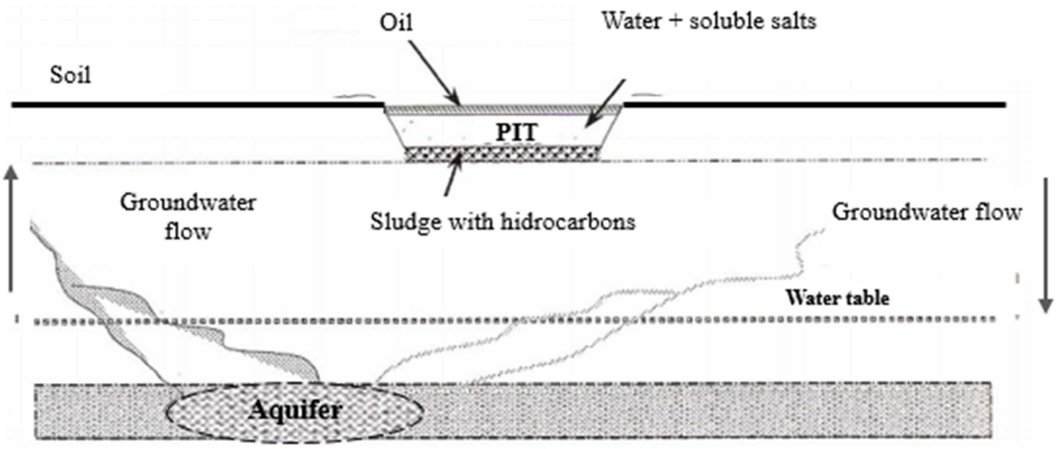


***Figure 1.*** *Diagram of an oil production pit located in a Venezuelan savannah (Modified from Arellano 2008 [7]. The diagram is not in scale.*

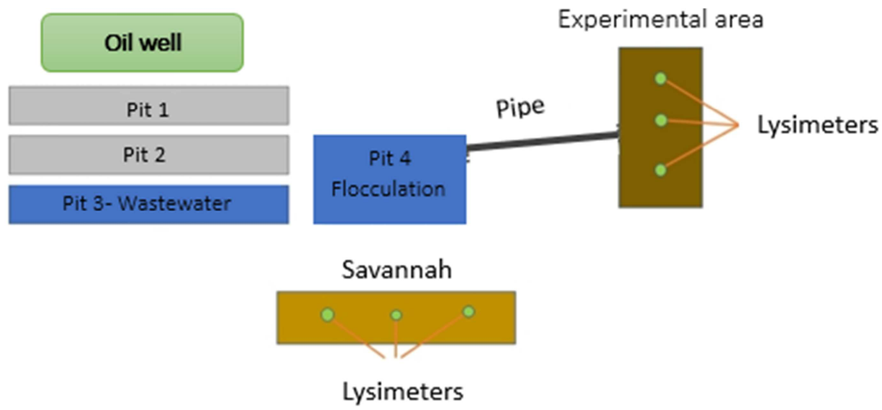


***Figure 2.*** *Diagram of the arrangement of an oil well (Ful-15) showing the fluid treatment pits (Pits 1-3), the flocculation pit (Pit 4), and the pipe that connects this to the Experimental Area with the lysimeters. In an adjacent area the control is located (natural savannah). The diagram is not in scale.*

Drilling fluids, in addition to solid waste materials and hydrocarbon, contain an abundant proportion of produced water. Water comes from the exploitation process (e.g. formation water, usually very salty) or from the fresh water that is injected to maintain oil and gas production [11]. The abundance of DP in oil activity sites are of sufficient magnitude to represent an important environmental and aesthetic problem in terrestrial environments [11, 12]). In this context, the Instituto de Tecnología Venezolana para el Petróleo (Intevep) (Venezuelan Institute of Technology for Petroleum-Intevep), together with the subsidiaries of Petroleos of Venezuela, in order to accomplish with the security measures established by current environmental regulations, began since the 80s [13], a study on treatment and disposal of mud from drilling fluids [14, 15].

The treatment for this waste is illustrated in Figure 2 for the Ful-15 oil well. The fluids (sewage, accompanying hydrocarbons and wastes from the drilling activity) are deposited in a system of pits (see Pits 1 and 2, figure 2) previously waterproofed with a layer of clay and a synthetic polyethylene membrane in order to avoid the percolation of diesel and other pollutants to the subsoil [7, 11, 12, 14, 15]. Subsequently, through the use of siphons, the hydrocarbon phase is separated from the wastewater (Wastewater Pit 3, Figure 2). The WW produced is usually very salty and contains suspended and dissolved solids, residual hydrocarbons, numerous organic species, heavy metals, naturally-occurring radioactive substances, and chemicals used in the extraction of hydrocarbons [14-16]. Therefore, these WW are transported to a flocculation pit (FP) (Pit 4,

Figure 2) where they are treated with a flocculating agent (usually aluminum sulfate) and lime in order to agglomerate the suspended solids and control acidity, respectively [15, 16]. After the flocculation-coagulation and pH correction (liming) operation is carried out, the treated water is discharged into the environment.

It is important to know the chemical composition of these WW before and after flocculation, and what effects they have when they are spread on the surrounding terrestrial ecosystems. Studies of this nature are particularly convenient in the savannas of eastern Venezuela, ecosystems where a large number of DP occur. The savannas where the treated water is irrigated, apart from the oil operation, present an intense agricultural and livestock activity, and rest on aquifers that are partially replenished with the percolation waters near the treatment pits (Figure 1). A recent study carried out on the levels of soluble salts and heavy metals in the soils of savannas adjacent to drilling rigs of oil wells located in eastern Venezuela after irrigation with WW showed [15] that when irrigated to the environment, the treated WW has a liming effect on the natural acidity of these soils. In general, the soils treated with the effluents had a higher concentration of sodium. However, the good internal drainage of the soils and the precipitation of the area may allow the displacement of salts. None-the-less, a strict monitoring is recommended, in particular of the pH levels of the wastewater before being irrigated to the surrounding savannas and its control in the percolation waters.

This study will have the following objectives: i) to analyze the levels of soluble salts and heavy metals in the treated and untreated wastewater produced in oil well operations, ii) to analyze the levels of soluble salts and heavy metals in the percolation waters of lysimeters that were located in irrigated savannas with wastewater treated with the flocculation-coagulation technique, and iii) to study the time it takes for these soluble salts to be dislodged from the lysimeters.

## 2. Materials and Methods

### *2.1. Study Area*

The study was carried out in the surroundings of El Furrial, Monagas State, Eastern Venezuela (9 ° 41' N and 63 ° 27' W, altitude 120 meters above sea level), in a savannah area affected by the drilling and management of oil wells. The climate of the Monagas´ savannahs is of the Awg'i type according to Koeppen, corresponding to a dry tropical climate with temperatures and average annual precipitation of 26.42° C and 1480 mm, respectively.

The alluvial deposits that cover much of the Llanos of Monagas have experimented a long time of pedogenesis evolution giving rise to Oxisols and Ultisols of sandy textures, acidic pH and low natural fertility [15]. The soils of the savannah studied are located within the Oxic-Paleustults Group, sandy loam within the Order of the Ultisols. The dominant landscape is of High Plateau, while the natural vegetation, is constituted mainly by grasses of savannahs dominated by *Trachypogon sp*. and *Axonopus sp*.

### *2.2. Sampling of the Irrigation Water Before and After Flocculation*

For the physicochemical characterization of the WW from the oil wells, composed triplicate samples were taken from the wastewater pits of the Ful-5 and Ful-7 oil wells before being treated with the flocculation-coagulation procedure.

Additionally, in order to study the effect of the flocculation-coagulation treatment on the levels of soluble salts and heavy metals, a sampling was carried out that included the taking of composite samples in crude water (non treated) and flocculated (treated) wastewater of the waters of the flocculation pit of Ful-15 oil well (Figure 2).

### *2.3. Irrigation Experiments and Location of Lysimeters*

Tension-free lysimeters were placed in hollows to collect deep drainage waters [17] in the area watered with flocculated water, and in a nearby savannah, not affected by the irrigation, the same number of lysimeters were located as a control (Figure 2). These percolation waters were sampled from lysimeters in two kind of experiments:

### *2.4. Experiment with Controlled Irrigation*

For eight hours an area (200 $m^2$) of savannah located 400 m apart from the oil well Ful-15 (Figure 2) was watered with flocculated water from the flocculation pit (Pit 4, Figure 2). To irrigate the experimental sub-plots with the FW, a pipe system was placed to transfer the water from the FP to the experimental area (Figure 2), sprinklers allowed to irrigate the plot for 8 continuous hours. This irrigation time in such a small area corresponds to a high dose of flocculated water.

Before irrigation in the area to be treated, and in the non-irrigated savannah, three lysimeters were placed at a depth of 50 cm.

### *2.5. Experiment with Uncontrolled Irrigation*

In four oil wells named Ful-2, Ful-6, Ful-8 and Lagoven-5, the adjacent savannahs to the FP were watered with flocculated wastewater. The irrigation was originally carried out by the personnel of the oil company, as the usual practice of evacuating the FP to the surrounding savannah, so no record was kept of the irrigation time, nor of the irrigation volumes. Therefore, the trial corresponds to an *ex post facto* experiment. In this experiments, lysimeters were located at 20 and 100 cm depth in both irrigated and savannah control.

The collection of the percolation waters was carried out at 24 months (oil well Ful-2), 6 months (oil well Ful-6), 1 month (oil well Ful-8) and 7 days (oil well Lagoven-5) after irrigation of the adjacent savannah.

### *2.6. Chemical Analysis of Flocculated (FW) and Non-flocculated (NFW) Waters*

The analysis of the elements in the non-flocculated wastewater (NFW), the flocculated wastewater (FW), and the waters collected in the lysimeters were carried out by flame atomic absorption in a Varian Techtron AA6 apparatus [18].

### *2.7. Statistics*

The means of the treatments and their respective controls were compared with t tests, statistical package SPSS 13.0 with a level of significance $p < 0.05$. Analysis of variance and comparison of means between treatments were performed by Duncan's test using the Statistical Analysis System (SAS).

## 3. Results and Discussion

### *3.1. Characterization of the Non-flocculated Wastewater (Raw Water)*

In drilling oil wells in the eastern plains of Venezuela, the main chemical additives used in drilling operations are: barite (barium sulfate), bentonite clay, lime (calcium hydroxide), diesel, caustic soda (sodium hydroxide), chloride of calcium and different polymers [14, 15, 19], so the abundance of calcium and sodium followed by potassium and magnesium found in the water from the untreated flocculation pits of the Ful-5 and Ful-7 wells (Table 1), while the levels of heavy metals (zinc, copper, chromium, cadmium, iron and manganese) are very low, in some cases at trace levels. In turn, the aluminum content in solution in the case of the Ful-7 well was high (4.4 mg $L^{-1}$), in accordance with the higher acidity (4.91) of its waters.

***Table 1.*** *Physicochemical characteristics of the raw water (before flocculation) of oil wells Ful-5 and Ful-7. Contents of the elements in mg $L^{-1}$.*

| Parameter | Ful-5 | Ful-7 |
|---|---|---|
| pH | 6.20 | 4.91 |
| Conductivity (µS/cm) | 503 | 590 |
| Na | 34.6 | 41.7 |
| K | 1.7 | 3.4 |
| Ca | 70.1 | 92.4 |
| Mg | 1.2 | 1.7 |
| Al | 0.87 | 4.4 |
| Fe | 0.0 | 0.37 |
| Mn | 0.07 | 0.11 |
| Zn | 0.016 | 0.012 |
| Cu | tr | tr |
| Cr | tr | tr |

### *3.2. Effects of the Coagulation-Flocculation Process on the Chemical Composition of Wastewater*

The levels of some of the elements studied had a significant drop after WW treatment with aluminum sulfate and pH correction with lime, particularly the values of barium, iron and aluminum (Table 2). The drastic precipitation of barium is evidenced as barium sulfate, which is a compound with very low solubility [20]. Ba levels in water without flocculating are very high (213.2 mg $L^{-1}$), well above the values accepted by the environmental standard [14]. However, these levels were reduced to traces after the coagulation-flocculation treatment.

Dissolved barium in the aquatic environment may pose a risk to aquatic organisms such as daphnids (water fleas), but the risk appears to be lower for fish and aquatic plants, although data in the literature are limited [21]). No adverse effects have been reported in ecological assessments of terrestrial plants or wild flora and fauna, although some plants are capable of bioaccumulate barium from the soil [21]. On the other hand, raising the pH with liming, a common practice to reduce high levels of aluminum and heavy metals in acidic environments [22], apart from slightly increasing the pH, allowed to reach the solubility product of compounds of aluminum and iron [23], for which there was a significant reduction in the levels of these elements (Table 2). It should be noted that in both flocculated and untreated waters, no detectable levels of chromium, nickel, lead and cadmium were found, however, the levels of aluminum in solution in untreated waters (5 mg $L^{-1}$) were within the limit allowed by the environmental standard [24].

Sodium levels in NFW are important (182.7 mg $L^{-1}$), although below the environmental norm (200 mg $L^{-1}$), while calcium levels do not represent any danger to the environment, both elements decrease significantly after coagulation-flocculation. The solids precipitated by the flocculation process carry part of the alkali metals (sodium and potassium) and the alkaline earth metals by trapping them in the exchange complex of the flocculating clays [25]; this phenomenon is corroborated by the significant decrease in the content of salts represented by the conductivity values after treatment (Table 2).

***Table 2.*** *Physicochemical characteristics of the waters of the oil well Ful-15, before and after flocculation. Contents of the elements in mg $L^{-1}$, values in parentheses correspond to standard deviations. Mean values with different letters differ significantly.*

| Parameter | Ful-15 non flocculated | Ful-15 flocculated |
|---|---|---|
| pH | 6.45a | 6.98b |
| Conductivity (µS/cm) | 1150 (95)a | 825 (80)b |
| Na | 182.7 (70.1)a | 88.1 (7.8)b |
| K | 3.2 (0.5)a | 1.2 (0.3)b |
| Ca | 64.4 (2.7)a | 49.2 (7.5)b |
| Mg | 0.7 (0.2)a | 1.0 (0.2)b |
| Al | 5.0 (0.5)a | 0.0b |
| Fe | 6.2 (0.5)a | 0.5 (0.1)b |
| Ba | 213.2 (93.9)a | 0.0b |

### *3.3. Content of Elements in Drainage Water in Irrigated Savannas Without Irrigation Control*

Table 3 shows the content of the elements analyzed in lysimeters placed at 20 and 100 cm depth in soils that had been irrigated with water from flocculation pits recently (1 week) until 24 months. At the beginning of the irrigation time, stand out higher levels of sodium, calcium and potassium in the waters collected at a greater depth (100 cm) with respect to the lysimeter placed superficially (20 cm), which indicates a dilution effect in the surface soil due to subsequent rains to irrigation. However, lysimeters on irrigated areas have higher levels of sodium and calcium only at the beginning of irrigation, as time passes (e.g. after 24 months) there is no major difference between soils irrigated with FW and the values of the control, where only rainwater is collected (Table 3).

***Table 3.*** *Physicochemical characteristics of percolation waters in savanna soils watered without irrigation control. Lysimeters placed at 20 and 100 cm depth. Contents of the elements in mg L$^{-1}$, values in parentheses correspond to the standard deviations. Mean values with different letters differ significantly.*

| Element | Location of lysimeters | 1 week Lagoven-5 | 1 month Ful-8 | 6 months Ful-6 | 24 months Ful-2 | Control |
|---|---|---|---|---|---|---|
| Na | 20 cm | 6.83b (1.69) | 6.33 (9.40) | 9.33 (3.17) | 4.70 (5.49) | 6.18 (7.91) |
| | 100 cm | 21.1a (29.3) | 6.74 (2.61) | 9.32 (8.87) | 6.76 (3.85) | 5.40 (3.20) |
| Ca | 20 cm | 0.93b (0.43) | 0.69 (0.39) | 3.26 (5.08) | 1.44 (0.68) | 0.35 (0.25) |
| | 100 cm | 9.25a (11.82) | 1.08 (1.28) | 1.85 (2.87) | 5.10 (3.20) | 1.37 (1.25) |
| K | 20 cm | 1.15b (1.37) | 1.70 (0.44) | 2.57 (3.88) | 1.50 (1.99) | 0.86 (0.94) |
| | 100 cm | 5.42 (3.37) | 1.72 (0.67) | 0.75 (0.24) | 2.32 (0.92) | 1.09 (0.51) |
| Mg | 20 cm | 0.17 (0.15) | 0.34 (0.18) | 2.06 (0.91) | 0.63 (0.84) | 0.11 (0.09) |
| | 100 cm | 1.72 (1.99) | 0.86 (1.56) | 3.62 (3.61) | 0.65 (0.15) | 0.28 (0.15) |
| Fe | 20 cm | 0.54 (0.51) | 0.43 (0.66) | 0.33 (0.53) | 0.07 (0.15) | 0.25 (0.42) |
| | 100 cm | 0.68 (0.62) | 0.22 (0.17) | 0.20 (0.40) | 0.22 (0.34) | 0.33 (0.36) |
| Al | 20 cm | 1.70 (1.77) | 2.77 (5.67) | 0.33 (0.66) | 0.10 (0.14) | 0.81 (0.02) |
| | 100 cm | 1.88 (1.73) | 0.19 (0.20) | 0.92 (2.04) | 0.22 (0.25) | 2.67 (4.11) |
| Zn | 20 cm | 0.00 | 0.00 | 0.07 (0.09) | 0.05 (0.07) | 0.01 (0.01) |
| | 100 cm | 0.045 (0.004) | 0.02 (0.04) | 0.01 (0.01) | 0.05 (0.10) | 0.00 |

### *3.4. Content of Elements in Drainage Water in Irrigated Savannas with Irrigation Control*

In the test with irrigation for 8 hours, important levels of sodium and calcium are incorporated into the savannah soils by FW when they are compared with the data from the lysimeters located in the control savanna. (Figure 3). For the rest of the elements analyzed (potassium, magnesium and iron) there are no major differences between the two treatments (Table 4). Sodium hydroxide, calcium chloride and lime (calcium carbonate) are materials commonly used in oil drilling and exploitation activities [26, 27], so it is not surprising that these percolation waters present important levels of sodium and calcium when flocculated waters have recently been incorporated to the savannah, values that decrease with the time elapsed after irrigation.

***Table 4.*** *Physicochemical characteristics of percolation waters in savannas with controlled irrigation. Lysimeters placed at 50 cm deep. Contents of the elements in mg L$^{-1}$, values in parentheses correspond to standard deviations. Average values with different letters differ significantly over time.*

| Element | 1 week | 1 month | 2 months | 4 months | Control |
|---|---|---|---|---|---|
| K | 1.85 (0.41) | 1.06 (0.41) | 2.15 (1.18) | 0.45 (0.12) | 1.06 (0.41) |
| Mg | 1.89 (0.38) | 1.93 (0.32) | 0.32 (0.15) | 3.23 (1.36) | 0.88 (0.90) |
| Fe | 0.96 (0.48)a | 0.58 (0.17)a | 0.39 (0.03)b | 0.73 (0.12)a | 0.37 (0.01)b |

### *3.5. Effects of Time After Irrigation on Soluble Salt Levels in Percolation Waters*

From an environmental point of view, the time in which high levels of soluble salts and other polluting agents may occur in the different compartments of terrestrial ecosystems (soils and percolation waters) is of concern, since as the soils are sinks production could be affected. Therefore, once these waters are spread out to the savannas, the lysimeters register high levels of soluble salts of sodium and calcium (Figure 3). One month after irrigation, significant levels of these elements still persist in the environment, but after 2 to 4 months of irrigation, the levels of sodium and calcium salts in the percolation waters drastically decreased. In summary, half a year after the savannas have been irrigated with flocculated waters, the sodium and calcium values in the lysimeters do not differ from the values reported in lysimeters control (Figure 3 and table 3).

Of the elements analyzed, sodium and calcium were the only ones that presented statistically significant differences between lysimeters located in soils irrigated with treated water and their respective control in both controlled and non controlled experiments (Table 3 and Figure 3). The addition of calcium to the environment does not constitute a problem, on the contrary, due to the low pH of these soils, this nutrient acts as a liming substance, improving the natural fertility of the soil and decreasing the levels of soluble aluminum which is potentially toxic. Petroleum fluids containing high concentrations of salts have been reported to cause significant effects on soil and vegetation [19, 28]. The levels of sodium that are incorporated could constitute a delicate environmental problem if salts are accumulated in soils with poor drainage, and could induce sodium salinization [29]. However, in the specific case of the study area, although significant levels of sodium were found in the treated soils [15], the danger of salinization is ruled out, since this element does not tend to remain for a long time in the profile of these savanna soils, due to their good internal drainage and adequate rainfall in the region (greater than 1400 mm). As the salts are diluted by meteoric water (rainwater) the contaminants will move laterally gradient down [30], as shown in the information in Table 3 and Figure 3.

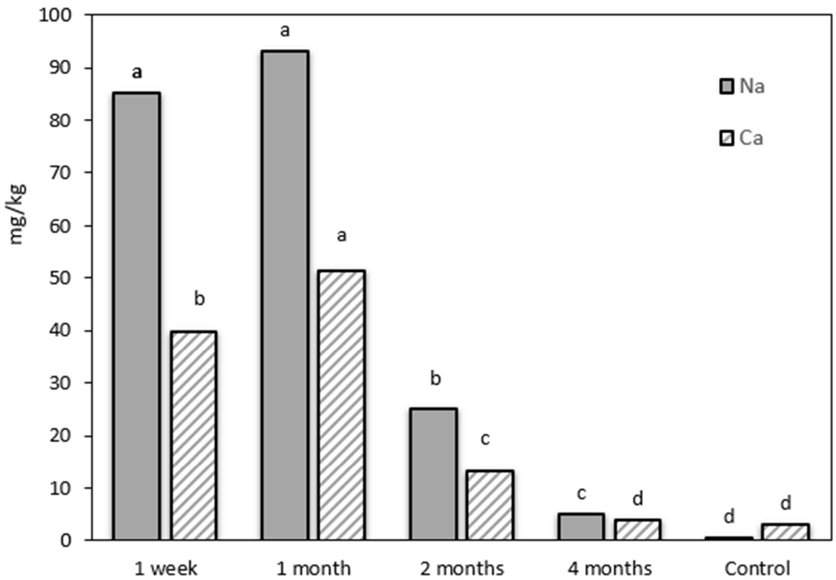


***Figure 3.*** *Changes in the Ca and Na contents in the percolation waters as a function of the elapsed irrigation time. Element averages at different times followed by different letters differ significantly.*

# 4. Conclusion

The wastewater generated in the disposal of oil residues has a high electrical conductivity and high contents of barium, sodium, calcium, aluminum and suspended solids. Once WW are treated with the coagulation-flocculation technique, agglomeration and precipitation of the suspended particles occurs and a concomitant decrease in the levels of soluble salts. In particular, the high levels of barium and aluminum are reduced to traces after the flocculation treatment. There is also a slight decrease in the levels of sodium and calcium since the soluble forms of these elements are trapped in the exchange complex of the solids that precipitate.

The sodium content of the lysimeters at the beginning of irrigation is high, and then decreases as a function of the time elapsed after irrigation. However, the danger of sodium salinization is ruled out, since this element does not tend to remain for a long time in the soil profile, due to its good internal drainage, and the adequate precipitation of the region (greater than 1400 mm), thus, the values of the controls and treatment do not differ significantly at 6 months after irrigation.


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