Impact of spreading olive mill wastewater on soil characteristics: laboratory experiments

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Agronomie 21 (2001) 749–755

Original article

Impact of spreading olive mill wastewater on soil characteristics: laboratory experiments

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(Received 21 May 2001; revised 20 July 2001; accepted 20 September 2001)

Abstract – A dynamic of soil pollution with olive mill wastewater (OMW) was investigated in the laboratory. Organic and inorganic components of OMW were removed at the first infiltration by the upper layer of the soil, whose chemical properties changed. Besides organic carbon, Kjeldahl N, total phosphorus and potassium concentration increase, electrical conductivity, phenol concentration and salinization level increased, which could compromise soil fertility. At the second infiltration carried out 2 months later, the properties of the soil samples appeared to be neatly altered.

OMW / soil / impact / phenols / organic matter / salinity

Résumé – Impact du rejet des effluents d'huileries sur le sol. L'impact de l'épandage d'effluents d'huileries (margines) sur un échantillon de sol au laboratoire a fait l'objet de cette étude. La majorité des micro-polluants organiques et inorganiques des margines ont été éliminés après la première infiltration ; ceci entraîne le changement des caractéristiques physico-chimiques du sol. La concentration en carbone organique, en azote Kjeldahl, en phosphore total et en potassium augmente. L'augmentation de la conductivité électrique, des phénols et de la concentration du sodium, surtout au niveau de la couche supérieure (0–20 cm), jusqu'aux limites de salinisation du sol peut entraîner la diminution de la fertilité de celui-ci. Une deuxième application de ces effluents sur les mêmes colonnes, après deux mois de repos, a montré une diminution des capacités épuratrices du sol.

margines / sol / impact / phénols / matière organique / salinité

1. INTRODUCTION

The manufacturing process of olive oil yields three phases; an oily phase, a solid residue and an aqueous phase. The latter, when combined with the washing water process forms an olive mill wastewater (OMW). The amount of this waste depends on the process used for oil extraction. The average volume ranges from 0.5 to 1.5 m^3 per ton of processed olives [16, 17].

The disposal of OMW causes serious environmental problems related to its high organic content made up largely of simple phenolic compounds [24], that have been described as having both antimicrobial and phytotoxic effects [18]. Indeed, many rivers in Morocco (Sebou and Fez rivers) and in Spain (Guadalquivir river) have become anoxic [10]. For that reason, the public health authority has prohibited the discharge of OMW in the rivers and subsidised the construction of ponds for storage during the milling period and evaporation of water during the summer. However, the ponds caused a serious negative environmental impact due to the foul odours, insect proliferation and groundwater contamination [11].

Communicated by Gérard Guyot (Avignon, France)

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Nevertheless, the main problem is their insufficient capacity because of the large volume of produced OMW.

One alternative solution to the problems of OMW disposal is land treatment/utilisation, especially in arid areas. OMW can be considered as a source of water and nutrients. Land treatment depends on the soil characteristics and climatic conditions.

This paper reports the results of laboratory experiments carried out to determine the pollution removal capacity of local soil irrigated with OMW. The effect of successive OMW treatment on chemical properties of the soil profiles was also evaluated in the present work.

2. MATERIALS AND METHODS

Soil samples were collected at 1 m depth from an olive grove in the province of Marrakech (Morocco) near an olive oil mill. This soil has never received OMW. The properties of the soil samples used in this study are summarised in Table I.

The OMW used for irrigation was collected from an olive oil production plant near Marrakech, screened through a 1 mm sieve to eliminate large fragments and then used for irrigation.

2.1. Experimental device

Air-dried soil samples were packed in 6 columns with PVC pipes (7 cm in diameter and 120 cm in length). The columns were irrigated daily with OMW (750 ml) until filled. Three columns were cut into five 20 cm length layers. The remaining columns (3) were left to dry for about 2 months in the laboratory and then irrigated in the same way to measure the effect of an additional application of the OMW. All leachates were collected every day for a period of 3 days for the first experiment and 4 days for the second experiment.

2.2. Chemical analysis

Input and output liquids were analysed for pH, electrical conductivity (EC), COD, Kjeldahl N (Kj-N) and total P, according to AFNOR methods [2]. NO_3 and NH_4 were analysed by distillation according to the Bremner and Keeney [9] method. Na and K were determined by the flame photometry method.

Soil samples were air dried, ground and sieved (2 mm). Soil organic matter was determined by the Anneas method described by Aubert [3]. TKj-N was determined by the micro-Kjeldahl digestion and distillation method, and P by the Bongonzo method [6]. pH and electrical conductivity (EC) were determined respectively on a mixture of soil/water (1:2.5 and 1:5). Na, K, Ca and Mg were determined in the saturated paste extracts of soil, prepared according to the method described by Richard [20].

The total phenols were soxhlet extracted from the soil samples according to the modified Blouin's method [5]. The extraction was carried out with distilled water for 2 h at 100 °C. Phenols were quantified by spectrophotometry using the Folin reagent [23].

2.3. Statistical analyses

The mean and standard deviation values of the triplicate samples were calculated. The data collected were analysed with a parametric three-way analysis of variance (ANOVA) to test the significant differences between treatments ($PL_{0.05}$).

3. RESULTS AND DISCUSSION

Physico-chemical analysis (Tab. II) showed that the OMW was an acidic effluent and contained a high level of organic matter and phenols. This also showed that the OMW contained a high level of sodium (5.2 g) different from the contents found in the literature which did not exceed 2 g/L [7]. This can be explained by the excessive

Table I. Granulometric characteristics of soil.

Layer (cm)	Clay %	Silt		Sand	
		Fine silt %	Coarse silt %	Fine sand %	Coarse sand %
0–30	12.52	23.20	8.25	18.83	32.80
30–60	25.21	25.76	7.12	13.26	28.92
60–80	34.33	26.72	6.65	12.36	19.66
80–100	35.68	23.23	6.25	11.32	20.13

Table II. Removal efficiency of soil after the first and the second infiltration of OMW (data are means of three replicates).

	Input (OMW)	Output 1 (EP1)	Removal %	Output 2 (EP2)	Removal %
pH	3.93	7.61±0.65	–	6.21±0.95	–
EC ($\mu S \cdot cm^{-1}$)	25 000	1056±122	–	7500±232	–
TSS ($mg \cdot L^{-1}$)	20000	425±50.2	97.87	2468±225.5	87.66
COD($mg \cdot L^{-1}$)	60 000	436 ±85.5	99.2	9800±532	83.6
PO_4 ($mg \cdot L^{-1}$)	45	0.189±0.02	99.5	3.60±1.02	92
Total P($mg \cdot L^{-1}$)	462	5.28±0.25	98.8	46.54±8.35	89.9
TKj-N ($mg \cdot L^{-1}$)	784	28.87 ±3.22	96.3	108.68±42.51	86.1
Na^+($mg \cdot L^{-1}$)	5200	11.51 ±1.85	99.7	1220±82.1	76.5
K^+($mg \cdot L^{-1}$)	6800	3.54±1.02	99.4	750±55.3	88.9
Ca^{++}($mg \cdot L^{-1}$)	1200	130 ±10.5	89.16	1700±222	–29.41
Mg^{++}($mg \cdot L^{-1}$)	450	65±13	85.55	1436±186.26	–68.66
Total phenols ($mg \cdot L^{-1}$)	3500	3.54 ±0.19	98.9	187.5±53.5	94.64

use of salt (NaCl) for the conservation of olives before extraction.

3.1. Soil purifying capacity

Results of the physico-chemical analyses of input and output liquids after the first infiltration (OL1) and the second infiltration (OL2) are presented in Table II. It can be observed that the OMW underwent important changes in composition after passing through the soil.

The OMW became limpid after the first infiltration. Indeed, 98% of the total suspended solids (TSS) were eliminated. According to Ranalli [19], the pigments responsible for the dark colour of OMW are phenolic compounds. Thus, the particulate part of phenols was eliminated by sifting on the filter solid mass.

Furthermore, more than 99% of the OMW (COD, TP, TKj-N, Na, K, Ca, Mg) was removed after percolating through the soil columns. These concentrations in the leaches were very low suggesting the effectiveness of the soil in removing all OMW nutrients.

The electrical conductivity (EC) of OMW decreased during its application on the soil as a consequence of adsorption phenomena. In contrast, the pH of OMW increased by 3 units, which can be due to the fixation on clay particles of organic acids, responsible for a low pH value.

After the second infiltration, the concentrations of the OMW (COD, TP, TKj-N, Na, K) were decreased in leaches. This result suggested that the soil's capacity to absorb/adsorb anions had been exhausted. Only the concentration of soluble Ca and Mg were higher in leachates than in the OMW. This could be due to the dissolution of the soil carbonates by the acidic OMW. This result has been demonstrated during the application of OMW on calcareous soil [11].

More than 99% of phenols were removed after the first infiltration. In fact, the increased pH of the OMW during the experiment may have transformed phenols into phenolates which are well retained by soil cations [14].

After the second infiltration, the phenol concentration was increased in the leachates which can present a risk of contamination of the groundwater. Indeed, Spandre and Dellomonac [21] reported a link between the wastewater spreading and local high concentrations of phenolic compounds in groundwater. The OMW was recognised as being the source of local groundwater pollution by phenols.

3.2. Effect of OMW on soil

The vertical distribution of nutrients in each soil column was investigated. The results revealed that the chemical properties of soil were changed.

3.2.1. Soil pH

Figure 1a shows the pH values of the various layers of soil during the first and second infiltration. The addition of OMW to the soil caused a change in the soil pH. The surface layer of samples seems to have a more acidic pH value (6.95) than their respective bottom layers. Although, it has been reported that addition of OMW sewage material would cause a decrease in soil pH value [8].

The reduction of soil pH may affect the capacity of soil to immobilise heavy metals [23]. In the same way,

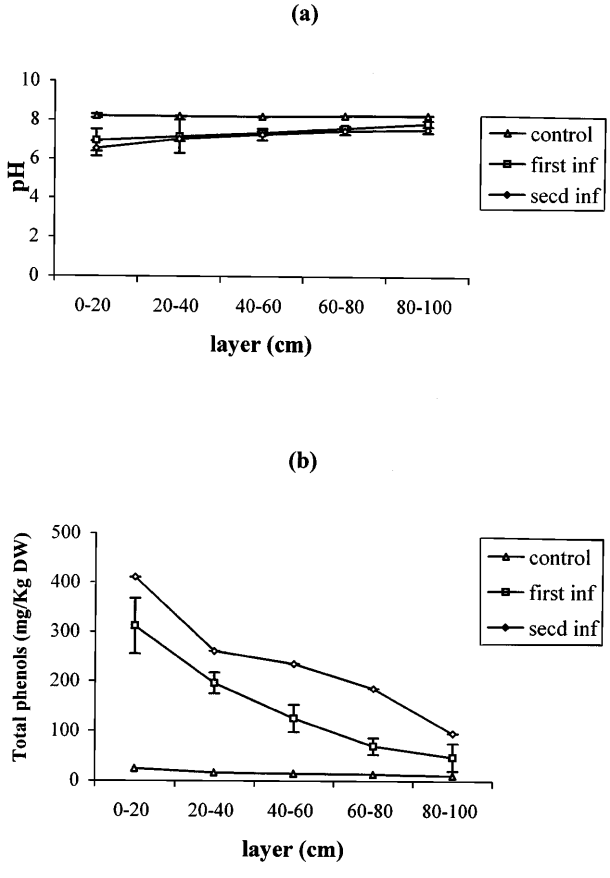

Figure 1. Effects of OMW on the profile of soil pH (a) and total phenols (b) after the first and second infiltration (inf.). Data are means of 3 replicates.

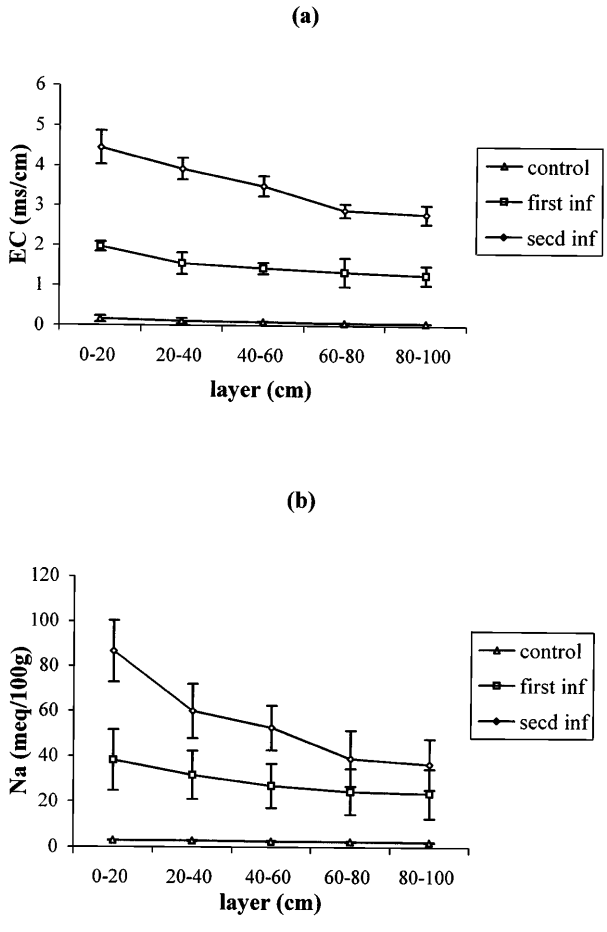

Figure 2. Effects of OMW on the profile of soil conductivity (a) and sodium content (b) after the first and second infiltration (inf.). Data are means of 3 replicates.

Bejarano and Madrid [4] showed that the joint effect of pH and the presence of OMW is the release of amounts of metals which are comparable to the metal fractions attributed to exchangeable and bound to carbonates ones.

The soil pH did not show any difference between the first and second infiltration; the value was near neutrality. This can be explained by the soil buffer capacity [22].

3.2.2. Total phenol in soil

Few studies have approached, until now, the phenol concentration of OMW in the various soil horizons. Our results (Fig. 1b) showed that the OMW increased the content of total phenols in all soil layers. Some phenolic compounds, such as flavonoids, are abundant in OMW; they are not assimilated by the plants and their degradation rate is often low [7, 15]. The concentration of phenols increased significantly in the surface layer. The phenol immobilised in this layer can act on the germination of seeds and inhibit the activity and the growth of the micro-organisms responsible for organic matter mineralisation [15]. In contrast, Dommergues [12] has explained that a neutral pH value and the presence of oxygen in the soil favour the decomposition of phenolic compounds.

After the second infiltration, the concentration of phenols increased in all layers, which can contribute to the sterility of soil.

3.2.3. Effect of OMW on the soil salinity

The application of OMW induced an increase in the electrical conductivity (EC) of all soil layers compared to the control (Fig. 2a). After the second application, the soil EC increased and the surface soil layers had much

higher values of conductivity. The value reached 4 ms/cm corresponding to the limit of saline soils. The increase of soil salinity involved the alteration of the composition of the CEC complex [10]. This alteration was gradual with an upward trend for exchangeable K and Na. At the same time, the treatment involved the enrichment of the soil by soluble Na (Fig. 2b) because of its high concentration in OMW. Moreover, Na migrated to the major horizons, which could present a risk of contamination for groundwater.

3.2.4. Effect on soil fertilising value

The analysis of the organic matter content in the various soil horizons (Fig. 3a) showed that the OMW induced an increase in organic matter content in higher soil layers with a slight migration towards the subjacent horizons. The immobilisation of the OMW organic matter in the upper layers facilitated its mineralisation even when the OMW was applied at a lower dose (3731 $m^3 \cdot y^{-1}$) [11]. According to the same authors, this mineralisation could be explained by the proportion of biodegradable organic matter (BOD) of OMW (BOD/COD ca. 0.45). After the second application, the organic matter was not maintained in the higher layers any more but it migrated to deep layers (80–100 cm). The content of organic matter in the surface layer did not change significantly between the first and second infiltration. It is possible that the added organic carbon had been degraded biologically by aerobic and anaerobic micro-organisms [22].

Total nitrogen content in the various horizons increased after the second application, compared to the first contributing to the soil fertility (Fig. 3b). The high percentage of TKj-N in the upper layer can be at the origin of production of nitrates by mineralisation of the TKj-N, which could contribute to groundwater contamination [1].

As for nitrogen, the phosphorus content increased after the second infiltration. The application of the OMW involved the enrichment of the higher soil layers by phosphorus with a small quantity which migrated towards the subjacent horizons, even after the second application (Fig. 4).

3.2.5. Incidence on the soil physical properties

The principal risk incurred by soil during the OMW spreading was the filling-in which occurred on the 2nd day after the first application. The soil sweating became slow and was cancelled on the 3rd day. This filling-in can be generated by the suspended matter which was abundant in the OMW (20 g/l) and which can seal

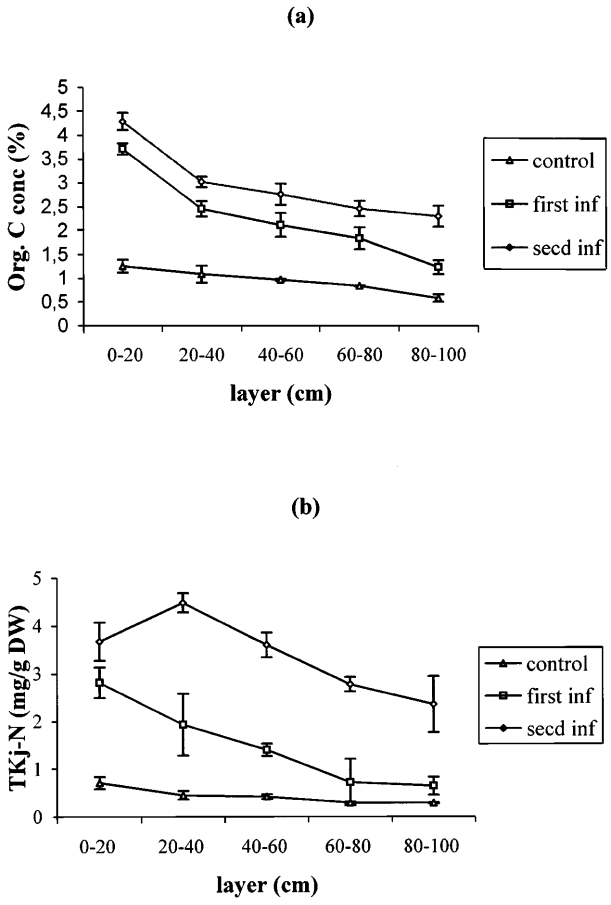

Figure 3. Effects of OMW on the profile of soil organic carbon (a) and total nitrogen (b) after the first and second infiltration (inf.). Data are means of 3 replicates.

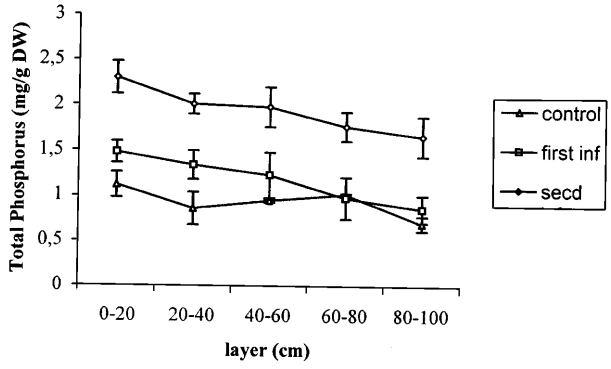

Figure 4. Effects of OMW on the profile of soil total phosphorus after the first and second infiltration (inf.). Data are means of 3 replicates.

the large pores of the soil. Cox et al. [11] showed that OMW affected the soil porosity by reducing large pores ($r > 10$ µm) and increasing small pores ($r < 0.1$ µm).

We also observed a swelling of soil particles especially after the second application due to the high sodium content of the soil which reacted with the alkaline-earth elements (Ca and Mg), with consequences for soil permeability reduction.

The soil also had a bad smell due to the anaerobic conditions in the columns related to an overload of the soil by organic matter.

4. CONCLUSION

The present study indicates that the clay soil used in this work has a very effective adsorption/absorption capacity. Over 99% of nutrients were removed after the first infiltration with the OMW. On the contrary, after the second infiltration the soil capacity to absorb/adsorb the anions had been exhausted.

When applied to the soil, the OMW induced its enrichment by fertilisers as well as negative effects, like fast filling-in of the soil and contamination by phenols. The latter biodegrade with difficulty, especially those immobilised in deep layers. Alteration of soil physical properties by swelling of soil argillaceous particles is mainly due to the presence of salt.

To solve these problems, we first suggest minimising the quantity of salt used for the conservation of olives, which is responsible for the high content of sodium in OMW and to increase the time between irrigations so that the soil can recover its purifying capacities. In the same way, special attention must be paid to the amount of irrigation, which depends on the physico-chemical characteristics of the OMW used, and on the hydrogeological conditions of soil irrigated in order to avoid the possible contamination of groundwater.

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