

The main Removal Mechanism of Organic micropollutants and Organisms in an Irrigation System using Untreated Wastewater

Alma C. Chávez-Mejía*, Rafael Magaña-López, Juan Carlos Durán-Álvarez and Blanca Elena Jiménez-Cisneros

Subdirección de Hidráulica, Coordinación de Ingeniería Ambiental, Instituto de Ingeniería, Universidad Nacional Autónoma de México, México, Av. Universidad 3000, C.P. 04510, Coyoacán, Ciudad de México, México.

Email: AChavezM@iingen.unam.mx

Abstract— *The presence of organic micropollutants on residual water used for irrigation is common in development countries, and their human impacts and the ecological consequences are still completely unknown. The aim of this study was to determine the main mechanisms involved during removal of three organic compounds (carbamazepine, ibuprofen, and 4-nonylphenol), and three usually pathogenic organisms (Escherichia coli, Giardia lamblia and Ascaris lumbricoides) contained in untreated wastewater dumped to agricultural soil, based on laboratory studies with inoculated soil, as well as the corroboration with field measurements. The results suggest adsorption and biodegradation as the predominant processes responsible for the removal of all contaminants analyzed, reporting efficiencies greater than 95% during the first 30 cm of the soil depth. Nevertheless, the efficiency depends on the type of pollutant, and also the physicochemical characteristics of the soil. For ibuprofen and 4-nonylphenol occurs mostly by biodegradation, and the adsorption of carbamazepine is associated with the organic matter content, while E. coli is adsorbed to the clay fraction of the soil. Finally, G. lamblia and A. lumbricoides removal is due to different processes from adsorption such as colloidal filtration.*

Keywords—*Endocrine disrupting chemicals, pharmaceuticals, removal mechanisms, soil, unintentional wastewater reuse.*

I. INTRODUCTION

The maintenance of the cities implies the supply of a large volume of water, which implies a model of complicated sustainability. The reuse of raw or treated wastewater for specific cases is an ancient and common practice around the world, especially in countries with emerging economies. One of the main uses is for agricultural irrigation which is estimated to amount to 29

183 897 m³/d (Jiménez and Asano, 2008). Most of developed countries practice the irrigation with wastewater in a controlled way, contrasting the situation in developing countries, owing to a lack of appropriate infrastructure, irrigation is seen as an economic and feasible way of disposing of raw wastewater. Each of these two ways of management has advantages and disadvantages. For the former, risks to health are minimized as the concentration of pollutants and nutrient content like nitrogen, phosphorus, and organic material is controlled, although, for the firsts, depending on its subsequent uses can be added. In the latter case, crop production is favoured as a result of the abundance of nutrients present in the irrigation water, but there is a greater risk to the health of the farm workers as well as consumers, nevertheless in most cases these menaces have not been quantified. The pollutants that represent the greatest danger to the population include heavy metals, pathogenic organisms, organic compounds and other not regulated with metabolic implications. However, the content not removed during the pre-treatment process ends in the environment, where their impacts and environmental implications in the medium and long term are still unknown

In Mexico, irrigation of crops with raw wastewater is a common practice estimated in 40% of the 208 m³/s of wastewater collected in the whole country, although the goal is to achieve 60% by 2012, although there is still no updated official data available. The most well-known area where this practice occurs is the Tula Valley, where the irrigation uses raw wastewater from Mexico City. Information on the irrigation is available from 1912 (the rate was 2 m³/s over 14 000 ha of land), and records how the increasing availability of wastewater as the population of the city has grown has involved in use of greater volumes of wastewater (such that today 45 m³/s is used for irrigation of 85 450 ha of land). Previous studies

Gibson *et al.*, (2007), Jimenez and Asano, (2008), Gibson *et al.*, (2010), and Chávez *et al.*, (2011) have shown that the infiltration of raw wastewater through the soil has functioned as a natural purification system on the organic, inorganic, and biological pollutants presents in wastewater. The rates of removal of diverse contaminants are due to a complex series of natural processes that can act in a synergistic manner, as mention Yamamoto *et al.*, (2009). The mentioned author emphasizes that, in the case of organic micropollutants; the natural mechanisms involved in the degradation and removal of the water soluble material are adsorption, biodegradation, and to a lesser extent photodegradation.

On the other hand, regarding to the pathogenic organisms, adhesion and deprecation are the dominant removal processes, particularly in agricultural systems using wastewater for irrigation (Stevik *et al.*, 2004). In this sense, it is important to consider the dependence of removal of pollutants, as much on the physicochemical properties of the compounds, as on the particular characteristics of the area (solar radiation, humidity, temperature, properties of the receiving soil, and the wastewater itself). The Tula Valley is well known as the oldest and largest example in the world of an irrigation system that uses raw wastewater (more than 100 years). Despite this, there are few studies at a regional level that have explored the processes of removal and/or degradation of pollutants involved. In addition, little is known about the capacity of this system, or the strategies that might be necessary in order to manage it efficiently, and whether could be applied to other similar areas.

The first step is to recognize in a global manner the processes by which this reduction in the concentration of pollutants occurs. The objective of this work is to understand the natural mechanisms of reduction in infiltrated water by comparison to the irrigation water, in particular the adsorption and biodegradation of pollutants in the agricultural soils of the area, and that cause the drop in concentration of the pollutants (the anti-inflammatory ibuprofen, the anti-epileptic carbamazepine, and the surfactant metabolite 4-nonylphenol), and three groups of organisms, including bacteria (*Escherichia coli*), protozoa (*Giardia lamblia*), and helminth (*Ascaris lumbricoides*) eggs. This was carried out by means of a field study complimented by laboratory studies for the different groups.

II. METHODS

Studies are focused on the determination of adsorption and biodegradation of organic micropollutants and pathogenic organisms were carried out in the laboratory while a field monitoring study was used to add weight to the laboratory data. Methodology used is described in the

following sections.

2.1 Sampling and characterization of the soil

A soil from the Phaeozem class was studied. For the laboratory tests, samples from 0 to 30 cm depth were collected by triplicate. For the field monitoring study, samples were taken from three soil horizons of each soil profile. All of the samples were stored in plastic bags at 4° C. In the laboratory, the soils were gently broken up and sieved to 2 mm and then characterized in terms of pH, humidity, cations (Ca²⁺, Mg²⁺, K⁺ and Na⁺), cationic exchange capacity, organic and total carbon, and the composition by its particle size, determining particularly the sand, silt, and clay contents). The details and techniques are listed below.

2.2 Batch adsorption assays

Organic compounds evaluated: 2 g of dried soil sterilized with a dose of gamma rays of 25 kGy was weighed into amber vials, then 10 mL of CaCl₂ [10 mM] were added (the final ratio of soil to solution was 1:5). Then carbamazepine, ibuprofen, and 4-nonylphenols were added to the vials at nine different concentrations (10, 50, 75, 100, 200, 400, 500, 1.000 and 5.000 µg/L); blanks without the micropollutants, and without soil were included to evaluate liberation of the contaminants from the soil, and the possibility of sorption to the walls of the vials were made, respectively. The flasks were shaken at 150 rpm for 24 hours at 25 °C in absence of light as possible (to avoid photodegradation). After shaking, the vials were centrifuged (3000 rpm for 10 min), and the supernatant was transferred to a clean vial and stored at -18 °C until chromatographic analysis.

Organisms evaluated: These assays were carried out by varying the quantity of soil instead of the concentration of organisms under test due to the difficulty of establishing and maintaining exact their concentrations. Samples of soil (5 masses) were tested for *E. coli* ATCC 700078 WG5 (0.5, 1, 1.5, 2 y 2.5 g), while for *G. lamblia* and *A. lumbricoides* eggs a mass of 0.1, 0.5, 1, 1.5, 2 y 2.5 g of soil were used. Then the soils were weighed into Teflon centrifuge tubes and the organisms were added using 20 mL of a solution of 0.85% NaCl.

The concentrations tried were 8x10⁸ CFU/mL for *E. coli*, 2.5 cysts/mL for *G. lamblia* and 2.5 eggs/mL for *A. lumbricoides*. The tubes were shaken at a speed of 110 rpm for 30 min for *E. coli*, and during 2.5 hours and during the same time for the other two species, then each the suspension was centrifuged (3000 rpm for 15 min), quantifying organisms from supernatant. All assays were carried out in triplicate; blanks (without soil and with no organisms) were made in parallel.

2.3 Biodegradation and adsorption column experiments for the organic micropollutants

PVC columns (5.08 cm internal diameter by 30 cm

height) were packed with dry soil. Layers of sand and then silica (mean 5 mm and 1 mm diameter) were added to the bottom of the columns above a mesh of steel (pore size 0.2 mm). The soil was packed manually 1 cm at a time to a depth of 10 cm; the total amount packed was 197 g, and the soil bulk density reached was 0.97 g/cm³. The columns were covered with aluminum foil to avoid possible photodegradation of the analytes. For both, the biodegradation and adsorption experiments were used four columns (three replicates and a blank), making a total of eight columns in total; for the adsorption tests the columns were packed using sterile soil. Six irrigation events were simulated in each column, the mixture of organic micropollutants being only 4-nonylphenols (36.4 µg/L) and ibuprofen (6.5 µg/L). Each irrigation was carried out manually with 13.6 cm³ of water (275 mL) poured in a single event, similar to rates used for irrigation of maize in the valley. For the biodegradation experiments a sterile solution of 29 mg/L of nitrogen as NH₄Cl, and 39 mg/L of phosphorus as KH₂PO₄ were added, similar to concentrations found in the local wastewater. The columns were saturated with a solution of [10 mM] of CaCl₂ before the first irrigation. For each one, the water was allowed to pass through the packed soil without the application of a vacuum. The leachates were collected in glass vessels and stored at -18 °C until their analysis. The irrigation events were applied every 21 days, simulating the time periods between irrigations in the field. At the end of the tests, the soil columns were taken apart under antiseptic conditions and the concentrations of the organic micropollutants determined.

2.4 Chemical reagents

All the analytes, internal and the recovery standards, as well as the derivatising agents N-tert-butyltrimethylsilyl-N-methyltrifluoroacetamide (MTBSTFA), and N,O-bis(trimethylsilyl) trifluoroacetamide (BSTFA) were obtained from Sigma-Aldrich (St. Louis MO, USA). The solvents used were HPLC grade, bought from Burdick and Jackson (Morristown, NJ, USA); the OASIS HLB extraction cartridges (200 mg) were supplied by Waters (Milford, MA, USA).

2.5 Analytical methods

Organic compounds evaluated: The determination of organic micropollutants in wastewater and leachates was done according to the method validated by Gibson *et al.*, (2007). Briefly, the leachates and aqueous solutions were acidified to pH = 2 with concentrated H₂SO₄. After passage through the Oasis HLB cartridges (conditioned with 2 x 5 mL of CH₃(CO)CH₃ followed by 5 mL of 5% CH₃COOH. The acidic pharmaceuticals were recovered by elution with 5.5 mL of a solution of CH₃(CO)CH₃:NaHCO₃ (40:60 at pH = 10), and the phenolic compounds were eluted with 5 mL of

CH₃(CO)CH₃. The derivatives of the acidic pharmaceuticals were produced using MTBSTFA as the derivatising agent while derivatives of the phenols were obtained by reaction with BSTFA. For soils, the analytes were extracted by accelerated solvent extraction and details of the procedure can be found in Durán-Álvarez *et al.*, (2009). The extracts (20 mL) were evaporated to approximately 3 mL, diluted to 15 mL with water, and then passed through the Oasis cartridges. From this point the procedure was the same as described for the aqueous samples. The final analysis and quantification of the analytes was carried out using an Agilent 6890 gas chromatograph coupled to an Agilent 5973 mass selective detector. 2,3-dichlorophenoxyacetic acid (2,3-D), [2H4] 4-n-nonylphenol, and [2H16] bisphenol-A were used as internal standards and 3,4-dichlorophenoxyacetic acid (3,4-D), 4-n-nonylphenol, and 10-11, dihydrocarbamazepine were used as recovery standards. In addition, solvent blanks were analyzed with each batch of samples.

Organisms evaluated. Bacteria were detected by the membrane filtration method (using 0.45 µm pore size membrane filters Millipore Corp., Bedford, MA selective agar), and most probable number with selective broths), methods according to Standard Methods for the examination of water and wastewater mentioned in APHA (1998). For the case of *G. lamblia*, the sample was concentrated through filtration in a polycarbonate membrane (5 µm pore and 20 mm diameter) incubated with polyclonal antibodies (rabbit sera and anti-rabbit immunoglobulin G) at 37 °C for 30 minutes, for each equipped with epifluorescence and phase contrast, according to APHA (1998). The concentration and further identification of helminth eggs in wastewater and water supply was performed using continual washing, combined with diverse filtration stages (sieve pore 150 and 20 mm) and flotation (utilizing a saturated solution of ZnSO₄; density 1.3 mg/cm³) and continual centrifugation concentration.

III. RESULTS AND DISCUSSION

3.1 Sampling and soil characterization

The physicochemical properties of the soil are recorded in Table 1. There is an elevated concentration of organic material in the first 30 cm of the soil profile (in comparison with others) as a result of the intensive use of wastewater for irrigation. In addition, there is a prevalence of expandable clays in the soils dominated by the smectite type (Friedel *et al.*, 2000). The soil moisture was well below the field capacity (34%) as the soils were sampled in the dry period between irrigations (which occur every three weeks approximately), a time when microbial activity is also reduced.

Table.1: Physicochemical characterization of the soil

Horizon (cm)	pH (pH units)	Exchangeable Cations (mg/L)				CEC (cmol/kg)	OC (mg/g)	TC (%)	TN (%)	Particle size		
		Ca ²⁺	Mg ²⁺	K ⁺	Na ⁺					Sand (%)	silt (%)	Clay (%)
0-10	6.63	37	6.6	1.8	1.2	47	40.8	4.6	0.4	39	35	26
10-20	6.73	38	6.6	1.8	1.2	48	41.5	4.6	0.4	40	35	25
20-30	6.61	39	6.5	1.8	1.3	49	42.2	4.5	0.4	38	36	26
>30	7.33	38	6.0	1.6	1.3	47	13.9	1.6	0.1	45	32	23

CEC = cationic exchange capacity; OC = organic carbon, TC = total carbon; TN = total nitrogen

3.2 Batch adsorption experiments

Organic compounds evaluated: Adsorption isotherms for the pollutants studied are shown in Fig. 1, and adsorption parameters for the three compounds are evidenced in Table 2. Ibuprofen was adsorbed less than either carbamazepine or 4-nonylphenol, possibly because the pH of the soil would encourage dissociation of the anti-inflammatory drug (pKa = 4.15) leading to greater aqueous solubility and also less adsorption due to repulsive forces as the organic material, additionally because the clays have negative charges. Carbamazepine is present in the soil in its non-ionized form (pKa = 14), and adsorption is favored because of its hydrophobicity. Chefetz *et al.*, (2008) suggested that the aromatic rings present in carbamazepine can form π - π interactions with the aromatic rings of the highly humified organic material of the soil, resulting in rapid irreversible adsorption. Therefore, a high degree of adsorption of carbamazepine can be expected in soils deeper than the first 30 cm of the soil profile. A similar pattern could be expected for ibuprofen, but with less adsorption overall due to its ionized state.

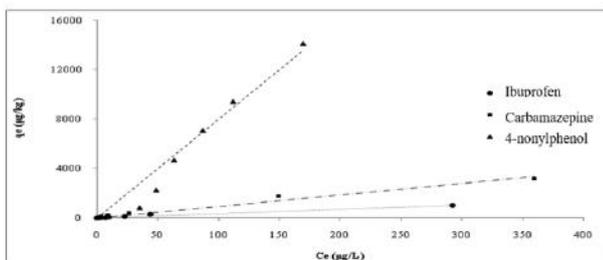


Fig. 1: Adsorption isotherms obtained for the organic micropollutants in the tested soils

Bi *et al.*, (2006) found a removal of carbamazepine when studying its adsorption in smectic clays similar to those found abundantly in the Tula Valley. In case of 4-nonylphenol showed the strongest affinity for the soil studied consistent with literature reports such as Düring *et al.*, (2002) who reported high distribution constants for 4-nonylphenol in soils with diverse characteristics (8.5-321 L/kg). The high value of the adsorption constant (normalized for organic carbon content) suggests that this adsorption occurs principally by means of non-specific hydrophobic interactions. The analysis of blanks without soil and blanks without the presence of the contaminants

showed that adsorption to the walls of the vials and their liberation into the aqueous solution was not significant.

Table.2: Adsorption parameters for the compounds studied in tested soil

Compound	K _d (L/kg)	R ²	K _d literature (L/kg)	K _{oc} (L/kg)	R (theoretical)
Ibuprofen	3.6	0.9789	3.7 ^a	84	12
Carbamazepine	9.4	0.9812	12.3 ^b	218	30
4-nonylphenols	80	0.9538	8.5-321 ^c	1862	250

K_d = Distribution coefficient; K_{oc} = Organic carbon adsorption coefficient; R² = correlation coefficient; R = Delay factor

^aXu *et al.*, 2009; ^b Stein *et al.*, 2008; ^c Düring *et al.*, 2002

Organisms evaluated: The adsorption isotherm for *E. coli* is presented in Fig. 2 adjusted to the Freundlich model. It can be seen that its concentration fits the model used, and the value of the factor “n” suggests that adsorption of this microorganism can be considered linear. The value of constant of Freundlich (KF) obtained was 1.8×10^3 , less than that reported by other researchers who have worked with soils with similar organic carbon contents but different amounts and types of clays as Smith and Badawy, (2007). It could be inferred that the adsorption of this microorganism is determined by the clay fraction of the soil, and it has been suggested that the production of extracellular polymeric substances which contain polyvalent cations help to bind bacteria with soil minerals, notably clays (Stevik *et al.*, 2004). In this sense, Guber *et al.*, (2005) found that the presence of natural organic material in the soil was not favourable for the process of adsorption of microorganisms.

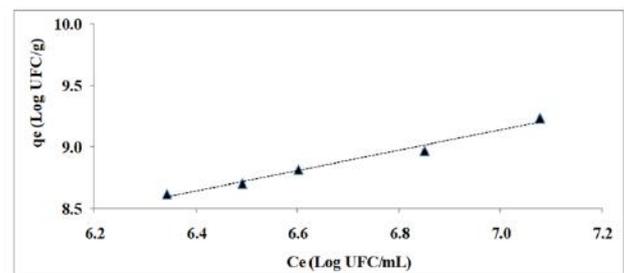


Fig.2: Adsorption isotherm for *E. coli*, adjusted to the Freundlich model

Data generated for *G. lamblia* and *A. lumbricoides* eggs could not be adjusted to either Freundlich or Langmuir models (R²<0.4) and so the isotherms are not shown. This could be due to reasons such as (a) the size of the organisms, (b) the complexity and heterogeneity of the soil which resulted in a multifarious adsorption process that could not be described by these models, or (c) the existence of other dominant processes such as colloidal filtration. In fact, last process has been reported by Brusseau *et al.*, (2005), but only for *G. lamblia*. Searcy *et al.*, (2005) found that cysts could adhere to different substrate, however, Dai and Boll, (2003) showed no

interaction between cysts and soil particles. For helminth eggs there is a paucity of information about their removal and inactivation in soil, although Capizzi and Schwartzbrod, (2001) have reported its capacity to adhere to silica particles present in clays and sand in soil, notably in clays of the smectite type abundant in the soil studied. In general terms, through this assay it was possible to determine removal rates in the liquid phase of 1 log unit for *E. coli*, 65% for *G. lamblia*, and 97% for *A. lumbricoides* eggs.

3.3 Biodegradation and adsorption of organic micropollutants in columns

Fig. 3 shows the concentration of the micropollutants in the leachates from the six irrigations. In the non-sterile columns less 4-nonylphenol was leached during the irrigations. The concentration of 4-nonylphenol recovered in the leachates was less after the third irrigation compared with previous, indicating that biodegradation of the compound was less after 60 days of residence. In the sterile soil columns, a large amount of leaching of 4-nonylphenol was observed until the third irrigation, after which the concentration in the leachate dropped by 55%. This could be due to greater adsorption of the soil or intra-particle diffusion where the compound enters into the mesopores and micropores of the soil, making it progressively more difficult for the compound to move through the soil. This last hypothesis is backed up by the fact that for the sixth irrigation the concentration of 4-nonylphenol leaching from the column was less than that found in the previous three irrigations, which were approximately constant.

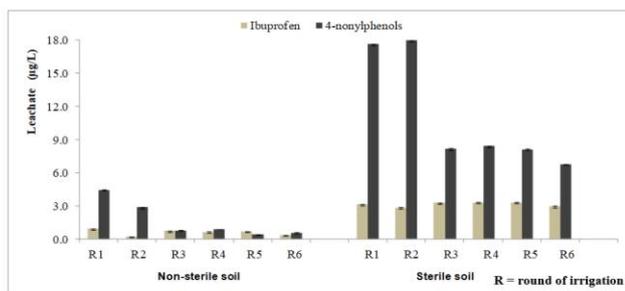


Fig.3: Concentration of the organic micropollutants in the leachates from columns packed with sterile and non-sterile soils

The leaching of ibuprofen from the sterile columns was constant over the six irrigations, indicating a low affinity for the soil; this was similar to the results for adsorption obtained from the batch tests. It is important to consider that ibuprofen and 4-nonylphenol could be adsorbed to the dissolved organic material and migrate through the soil column associated with this dissolved material and so arrive in aquifers (Hollrigl-Rosta *et al.*, 2003). In the non-sterile columns only a small amount of ibuprofen was

seen in the leachates, indicating that biodegradation is the process that defines the fate of this compound in the irrigation system. Ibuprofen has been reported to degrade rapidly in water and solid matrices (Xu *et al.*, 2009; Yamamoto *et al.*, 2009). However, it is necessary to evaluate the biodegradation of compounds that are poorly adsorbed into soil in irrigation systems where periods of dryness occur; causing large drops in the microbial activity of the soil, and this follows irrigation events that cause anaerobic conditions in the soil for short periods.

In general terms the removal of ibuprofen and 4-nonylphenol was between 66% and 89% in sterile columns, and between 85% and 93%, respectively in non-sterile columns. This indicates that biodegradation is the most important mechanism that determines the fate of both compounds in the irrigation system studied.

3.4 Field study

Organic compounds evaluated: Fig. 4 shows the concentration of organic micropollutants in the upper horizons of the soils studied. It could be observed an accumulation of ibuprofen in the three of them, indicating the occurrence of degradation together with a poor affinity for the soil, and data that reinforces the findings from the batch and column studies. Carbamazepine was retained in the first 30 cm of the soil profile, which suggests that it has a greater affinity for the labile organic material present in the upper profile. In order to clarify this, it is necessary to explore deeply about the quality of the organic material in the soil, and its qualitative and quantitative properties. The concentrations of carbamazepine in the deeper horizons studied could be attributed to the lower content (17 mg/g) or different quality of the organic material present, or to the presence of preferential flows caused by “alfalfa” (*Medicago sativa* L.) and roots of maize (the most common crops in the zone). but not to the biodegradation of this compound that although not evaluated in this study, nevertheless is known to be slow in soil compared with other cultivated species. Similar results have been reported by Kinney *et al.*, (2006) (although for soils irrigated with treated wastewater), where acidic pharmaceuticals similar to ibuprofen were degraded before migration to lower soil layers, while carbamazepine was retained in the first centimeters of the soil. In the light of these results, it appears contradictory that concentrations of tens of ng/L of carbamazepine are found in underground water; it suggests the necessity to study in the field the movement of this recalcitrant compound.

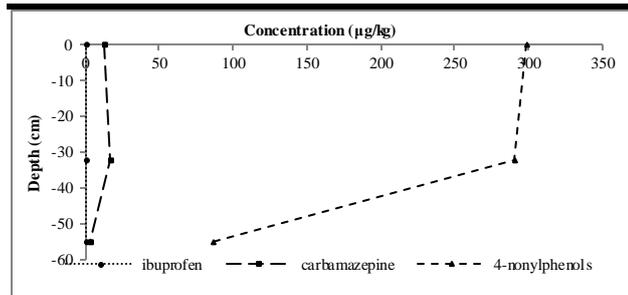


Fig.4: Concentration of organic micropollutants in the first three horizons of the soil profile.

Elevated concentrations of 4-nonylphenol suggest that it is not so rapidly degraded in the soil (as was seen in the column study). However, consideration should be given to the fact that the mass of 4-nonylphenol entering the soil in the wastewater (58.2 g/ha year) is greater than that of ibuprofen or carbamazepine (11.4 y 0.53 g/ha year). The decrease in concentration of 4-nonylphenol below 50 cm depth indicates a fall in the adsorption of this compound that explains its presence at trace levels in the local aquifer (Gibson *et al.*, 2007).

Organisms evaluated: Similar to the concentration of the organic pollutants, the organisms showed a decrease in concentration down the profile of the soil studied. The eggs of *A. lumbricoides* seemed to be completely retained in the first 30 cm of the soil horizon. Other studies have found that the removal of helminth eggs from water by filtration through soil occurs in the first 15 cm of the soil profile (O'Lorcain, 1994). However, this still entails a risk to health as these eggs can remain viable for several years. The same process appears to occur with the cysts of *G. lamblia*, whose concentrations tended towards zero with increasing depth. These results are greater than those reported by Mawdsley *et al.*, (1996), who found removal rates of 73% of oocysts of *Cryptosporidium parvum* applied to the soil, mostly by retention in the first 2 cm of the soil profile. The mechanisms that govern the transport of protozoa through the soil have not been completely understood, although Brusseau *et al.*, (2005) suggested, that this could be controlled by processes of colloidal filtration, although studies in this area are still scarce. The removal of *E. coli* reaches 1.5 log units, and this result is in line with the presence of total coliforms (up to 1×10^3 CFU/100 mL) in the water of the local aquifer, as well as with observations seen in the adsorption tests (removal of 1 log unit) (Fig. 5).

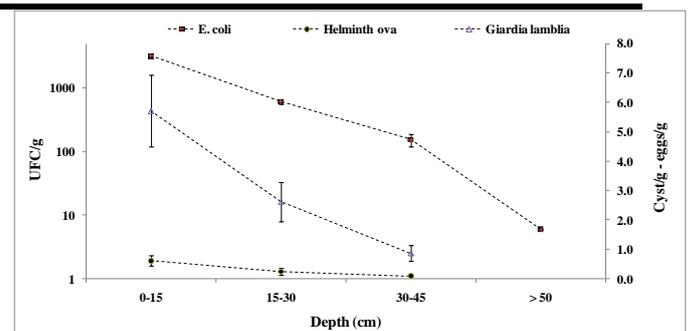


Fig. 5: Concentration of organisms in the first three horizons of the soil profile studied.

Previously mentioned authors have suggested that bacteria (such as *E. coli*) are retained in the top 8 cm of the soil during infiltration, although others comment that could be seen at much greater depths, but both are agree with their removal. In contrast, cysts of *G. lamblia* and eggs of *A. lumbricoides* can be removed from the water by simple filtration compared with *E. coli* that cannot be eliminated by that process, another difference is its survival time of the species (2 to 3 months), which is function of soil moisture and temperature as well the effect of the particular microfauna (Gerba and Lance, 1978). Soil clay content is a determining factor in the removal of bacteria from wastewater, so it is important to define which soils are suitable for wastewater irrigation and which pollutants should be eliminated as a priority before the agricultural irrigation.

IV. CONCLUSIONS

The physicochemical properties of the soil (significantly the content of clay and organic matter) promote organic micropollutants and pathogens removal by adsorption and biodegradation. Laboratory tests indicate for ibuprofen and 4-nonylphenol the biodegradation as the dominant removal process when wastewater is used for irrigation. On the other hand, carbamazepine is adsorbed in the soil, presumably in the organic matter content. For the case of organisms, the results show that *E. coli* is removed through adsorption to clay particles, while the removal of *G. lamblia* and *A. lumbricoides* cannot be described by typical means of adsorption, implying other mechanisms such as filtration colloidal occur in the system.

Field studies have established that the removal of both organic pollutants and organisms occur in the first 30 cm depth of the soil, which is probably related to strong adsorption of these contaminants to soil organic matter, except for ibuprofen where the main factor for its removal is biodegradation. *E. coli* removal is associated with the adsorption capacity of soils, which is correlated mainly to clay content.

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