

MIGRATION AND RETENTION OF PHENOL IN THREE NATURAL SOILS AND EFFECT OF SOLVENT

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Abstract. Soil contaminated with phenol poses a serious threat to the groundwater and soil organisms. In this paper, column transport experiments were used to investigate the retention and the mobility of phenol in three types of natural soils from northern Algeria. The effect of methanol, as an organic solvent, on migration of phenol through soil columns was also investigated. Transport parameters such as the retardation factor and the partition coefficient were calculated using “Laboratory Flow-Through”. The use of “Laboratory Flow-Through” method showed that phenol was more retained in Corso: a soil with higher organic matter content (7.22%) than Chorfa and Maâtkas (2.72 and 1.87%, respectively) and the retention increased with soil depth. The retardation of the transport of phenol was reduced in the presence of methanol which increased remarkably the mobility of phenol and its retention was much reduced especially at a depth of 5 cm by 25.55% in Corso, 19.05% in Chorfa and 9.18% in Maâtkas.

Keywords: *Laboratory Flow-Through (LFT) method, methanol, mobility, soil pollution*

Introduction

Phenolic compounds are persistent organic pollutants which have been widely used in chemical plants, metallurgical plants and petroleum refineries (Comninellis and Pulgarin, 1991; Li et al., 2005; Zhang et al., 2011), with a great variety of functions, in the production of many and different products: plastics, explosives, medicines, paints, detergents, pesticides, anti-oxidants, among others; in fact, phenol is considered among the most serious environmental contaminant which can affect air, groundwater, surface water and soil. It is very harmful even at low concentrations (Din et al., 2009; Pal et al., 2011), since it is toxic, carcinogenic, teratogenic and mutagenic to humans (Zhang et al., 2011).

Phenol is a colourless to light pink crystalline solid liquefies upon contact with water. It is moderately volatile at room temperature and has an acrid odour (Earl et al., 2003). Generally phenol is soluble in most organic solvents and it has a high solubility within water (from 67 g L⁻¹ at 16 °C to 84 g L⁻¹ at 20 °C) (Earl et al., 2003). Phenol in the aqueous phase is subject to chemical oxidation and adsorption (Sivasubramanian et al., 2012) and it is readily biodegraded under aerobic conditions in all environmental media, including soil and sediment; however, phenol degradation is slower under anaerobic conditions than under aerobic conditions and acclimation times are longer (Earl et al., 2003). A review of biodegradation studies in groundwater indicates that phenol is degraded under methanogenic, sulphate-reducing, iron-reducing and nitrate reducing conditions, with a half-life range of 22 to 533 days (Earl et al., 2003). As a result and based on its low adsorptivity to soil, phenol trickles down through different strata and migrates along with groundwater flow due to tortuosity and favourable

hydraulic conductivity of soil (Pal et al., 2014); however, information on levels of phenol in soil is limited (Earl et al., 2003).

Natural soil means soil from the ground (without further modification in the composition), which is distinguished from commercial potting soil. It is composed of mineral component and organic material or humus. Scientists usually test disturbed natural soil samples for soil texture and type and contaminant analysis, among other evaluations; it is easier to collect.

Phenol migration through natural soil has already been experimentally studied by researchers using laboratory batch adsorption test and vertical soil column tests. Smith et al. (1999) showed that phenol was relatively mobile in loamy sand and organic carbon content was found to be the most important soil component controlling retention potential of soils followed by clay mineral content (Khan and Anjaneyulu, 2005; Pal et al., 2014). Different studies showed that addition of other compounds may have an effect on phenol mobility. It is anticipated that the addition of Heavy metals may substantially alter pollutants mobility (Smith et al., 1999); however, compounds such as organic solvents, which were widely used to extract organic contaminants from contaminated soils (Osman and Saim, 2013), may have the opposite effect by increasing the mobility of phenol in soil. Nevertheless, experimental studies of the effect of methanol on the phenol vertical migration through soil media are lacking.

This paper aims to study retention and migration of phenol in three types of natural soils from northern Algeria. For this study, column experiments were used. “Laboratory Flow-Through” (LFT) method (United States, 1999) was used to determine the important parameters governing phenol migration. The effect on phenol mobility of the use of methanol, as an organic solvent, was also established.

Materials and methods

Sampling and soil characterization

Three types of soil were taken from three different regions of northern Algeria, namely, Chorfa (CH) situated in the province of Bouira (SE Algiers), Corso (CO) situated in the province of Boumerdès (E Algiers) and Maâtkas (MA) situated in the province of Tizi Ouzou (E Algiers). These three regions are representative of northern Algeria, as 90% of the population living in northern regions which are most likely to be polluted by phenol used in industrial and agricultural activities especially pesticides. Fifteen (15) soil samples were collected, few hours before the characterization, from multiples spots of each site, by using an Edelman hand auger and then mixed. Only the top 0-20 cm soil layer was sampled. In the laboratory, soil was air dried at ambient temperature, pre-treated by removing any large pieces of debris (e.g. Gravel, plant residues, etc.), gently crushed to break aggregates, homogenized and then screened through a 2 mm stainless steel sieve prior to analysis. These analyzes do not lead to any changes in the composition of the soil sample; therefore, it is still natural. The particle size distribution of soil was measured by laser diffraction (Malvern Mastersizer 2000 instrument). Soil textures were determined according to USDA textural classes (United States, 1993). Organic matter content (OM) was measured by calcination at 550 °C. Organic carbon content (OC) was estimated by dividing the organic matter content by assumed constant conversion factor 1.724; in addition porosity and bulk density (ρ) were also determined.

Chemicals

Phenol, C_6H_6O was obtained in crystal form (99% purity) from Merck Co. (Darmstadt, Germany). Its solubility in water is 76.04 g L^{-1} . Two stock solutions of phenol were prepared by dissolving 70 mg of phenol in 1000 ml of distilled water for the first one and 70 mg of phenol in 1000 ml of a mixture of methanol/distilled water (50:50, v/v) for the second to examine the effect of methanol, as an organic solvent, on transport of phenol. 100 mg of Mercury (II) Chloride ($HgCl_2$) was added to each stock solution as biocide to avoid phenol degradation. As phenol degrades when in contact with air and sunlight, care was taken to keep the stock solutions away from both. $NO_3^- [KNO_3]$ is used as inorganic non-reactive tracer, prior to each non-conservative phenol transport experiment to test columns performance and to determine retardation factor (R) of phenol within soil columns. Nitrate is widely used as tracer to study solute mobility and transport process (Braun et al., 2015; Zhen et al., 2015). A solution of KNO_3 (100 mg L^{-1}) was prepared using distilled water.

Column leaching experiments

Column leaching experiments were carried out in eighteen vertical propylene columns (1.3 cm inner diameter \times 6.5 cm length), containing CH-soil for six of them, CO-soil for six others and MA-soil for the last six ones. Nine of these columns were used to study the transport of phenol dissolved in distilled water and the last nine ones were used to study the transport of phenol dissolved in a mixture of methanol/distilled water (50:50, v/v). A 2 mm thick polystyrene filter was inserted in the base of each column. This filter also served as support for the soil at the bottom of the column. Negligible adsorption of phenol onto the column material was observed in phenol tracer test of the empty system. The columns were packed with soil in layers and were shaken to insure uniform mixing between layers. The soil was approximately 1.7, 3.3 and 5 cm thick for each type of soil (Fig. 1).

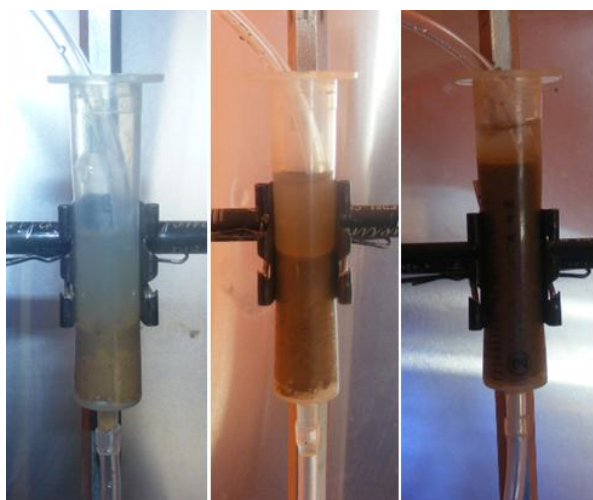


Figure 1. Soil columns: 1.7 cm thick (left), 3.3 cm thick (middle) and 5 cm thick (right)

The tracer solution containing $NO_3^- [KNO_3]$ (100 mg L^{-1}) was applied on the top of each first nine soil columns at a constant pressure head under steady state flow conditions, allowing the influents to flow through the soil under positive pressure head

(Rodriguez-Liebana et al., 2014). The transport of phenol dissolved in distilled water (70 mg L^{-1}) was performed in the same way as the transport of tracer and in the same soil columns. Transport of phenol dissolved in a mixture of methanol/distilled water (50:50, v/v) (70 mg L^{-1}) was performed in the last nine soil columns in the same way as the transport of the two first solutions to investigate the effect of methanol on phenol transport. Leachate samples were collected and analyzed for NO_3^- and phenol concentrations by UV-VIS spectrophotometer at wavelength of 220 nm and 510 nm, respectively. The pore volume (PV) or V_0 of the packed columns was estimated as follows: $V_0 = \text{column total porosity} \times \text{column volume}$. The breakthrough curves (BTCs) of NO_3^- and phenol from the packed columns were generated by plotting the relative concentration (concentration of effluent/ concentration of influent: C/C_0) in the leachate versus the relative pore volume (V/V_0) (Fig. 2).

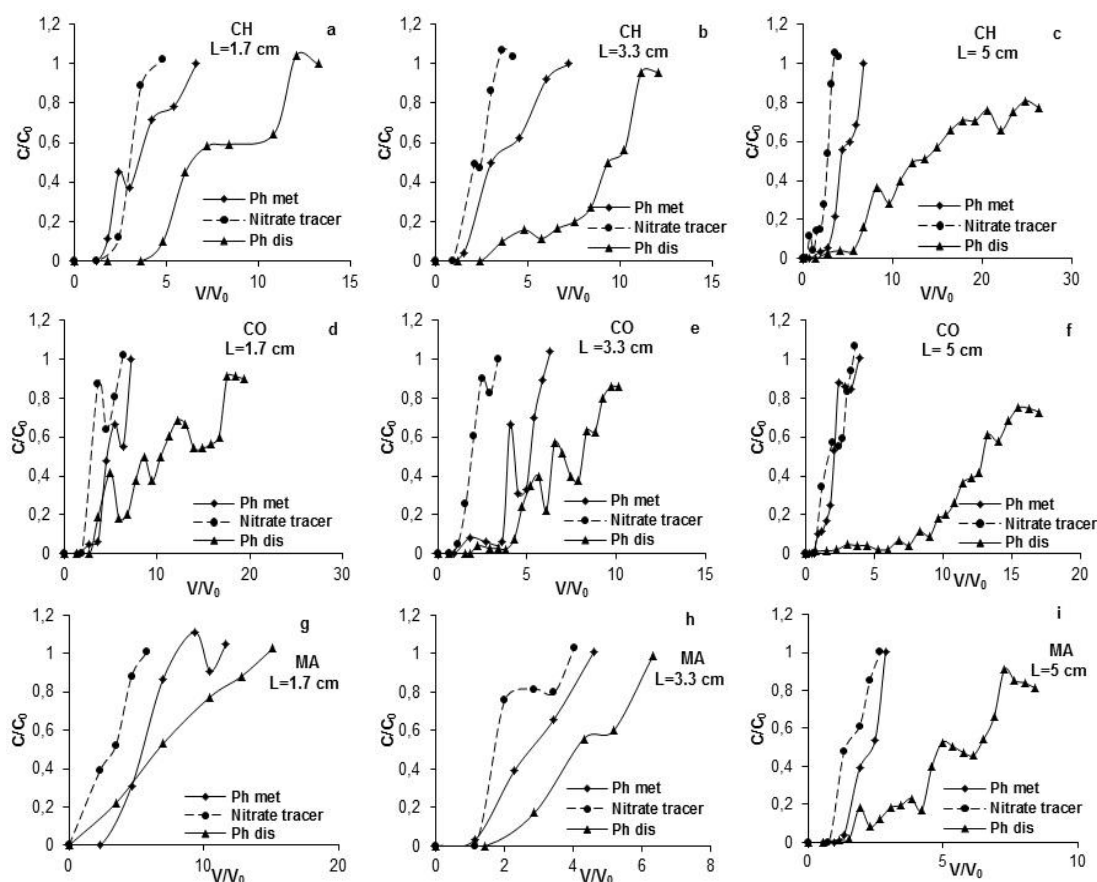


Figure 2. Measured breakthrough curves of phenol and tracer for various soil columns at different depths: CH at $L = 1.7 \text{ cm}$ (a); CH at $L = 3.3 \text{ cm}$ (b); CH at $L = 5 \text{ cm}$ (c); CO at $L = 1.7 \text{ cm}$ (d); CO at $L = 3.3 \text{ cm}$ (e); CO at $L = 5 \text{ cm}$ (f); MA at $L = 1.7 \text{ cm}$ (g); MA at $L = 3.3 \text{ cm}$ (h); MA at $L = 5 \text{ cm}$ (i). Ph_{dis} and Ph_{met} refer to phenol dissolved in distilled water and phenol dissolved in a mixture of methanol/ distilled water (50:50, v/v), respectively

Mathematical modelling

The solute transport model, which is considered the dominant mechanism of pollutant transport in porous media, was used in our study. This model is based on the one-dimensional convection-dispersion Richards equation (Eq. 1):

$$R \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} \quad (\text{Eq.1})$$

C is the solute concentration in the aqueous phase [ML⁻³], t is the time [T], D is the hydrodynamic dispersion coefficient [L²T⁻¹], X is the spatial coordinate [L], and v is the average pore water velocity [L T⁻¹].

The retardation factor (R) can be defined as (Eq. 2):

$$R = 1 + \frac{\rho}{\theta} K_d \quad (\text{Eq.2})$$

where θ is the volumetric water content [L³ L⁻³], ρ is the bulk density of soil [ML⁻³] and k_d is the partition coefficient [L³M⁻¹] describing the partitioning of aqueous phase constituents to a solid phase.

For a solute transport without adsorption: $k_d = 0$ then $R = 1$. R can be calculated from the breakthrough curves using “Laboratory Flow-Through” (LFT) method (Eq. 3):

$$R = \frac{V_2}{V_1} \quad (\text{Eq.3})$$

where V_2 is the volume of the eluted phenol solution when C/C_0 (phenol) = 0.5 and V_1 is the volume of the eluted tracer solution when C/C_0 (tracer) = 0.5 (United States, 1999); therefore, k_d is determined using Equation 2.

Results

Soils properties

The measured soils properties are summarized in Table 1. Based on texture, CH is loamy sand soil and CO and MA are sandy soils.

Table 1. Properties of the natural soils (CH, CO and MA) used in study

Soil	Sand (%)	Silt (%)	Clay (%)	Texture	OM (%)	OC (%)	Porosity (%)	ρ (g cm ⁻³)
CH	73.31	24.69	1.98	Loamy sand	1.87	1.08	37.64	0.97
CO	90.49	8.88	0.62	Sand	7.22	4.18	50.21	0.91
MA	91.15	8.84	0.008	Sand	2.72	1.57	39.39	1.03

CH, CO and MA, refer to soils collected from Chorfa, Corso and Maâtka, respectively; OM, OC and ρ refer to organic matter content, organic carbon content and bulk density, respectively

Transport of phenol in soil columns using “Laboratory Flow-Through Method”

Phenol and tracer breakthrough curves (BTCs) for the soil columns are presented in Figure 2 and transport parameters calculated using “Laboratory Flow Through” (LFT) method are listed in Table 2. The same parameters of columns are considered for the transport of both phenol dissolved in distilled water and phenol dissolved in a mixture of methanol/distilled water (50:50, v/v). The recovery rates of tracer were approach 100% in almost all soil columns, which can be considered ideal (Gibert et al., 2014).

Transport of phenol dissolved in distilled water

The transport of phenol dissolved in distilled water was retarded in almost all soil columns with late breakthrough curves (BTCs) especially at a depth of 5 cm and in all CO-soil columns (*Fig. 2*). The transport of phenol in the sandy soil MA was the greatest with recovery rates (RR) ranging from 90.97 to 102.77%; however, the recovery rates (RR) of phenol were poor in the sandy soil CO especially at a depth of 5 cm with 75.07% and in the loamy sand soil CH at a depth of 5 cm with 80.80% (*Table 2*).

Transport of phenol dissolved in a mixture of methanol/distilled water (50:50, v/v)

The (BTCs) showed nearly conservative transport behaviour especially at a depth of 5 cm in CO-soil and in MA-soil, where they were slightly delayed relative to the (BTCs) of the tracer (*Fig. 2*).

Table 2. Retention and transport parameters of phenol in natural soils calculated using column experiments

Phenol	Soil	Column number	L (cm)	Transport parameters calculated using LFT method		Recovery rate (RR, %)
				k_d (L kg ⁻¹)	R [-]	
Phenol _{dis}	CH	1	1.7	0.38	2	99.90
		2	3.3	1.3	4.42	95.64
		3	5	1.46	4.85	80.80
	CO	4	1.7	1.33	3.43	91.25
		5	3.3	1.25	3.29	85.84
		6	5	3.63	7.62	75.07
	MA	7	1.7	0.37	2	102.77
		8	3.3	0.46	2.23	98.94
		9	5	0.88	3.33	90.97
Phenol _{met}	CH	1	1.7	0.08	1.23	99.90
		2	3.3	0.16	1.42	99.92
		3	5	0.21	1.57	99.85
	CO	4	1.7	0.31	1.57	100.01
		5	3.3	0.60	2.11	104.05
		6	5	0.06	1.12	100.62
	MA	7	1.7	0.24	1.66	104.64
		8	3.3	0.24	1.66	100.74
		9	5	0.24	1.66	100.15

Subscripts _{dis} and _{met} refer to phenol dissolved in distilled water and phenol dissolved in a mixture of methanol/distilled water (50:50, v/v), respectively. CH, CO and MA, refer to soils collected from Chorfa, Corso and Maâtka, respectively. L, k_d , R and LFT refer to soil depth, partition coefficient, retardation factor and ‘laboratory flow-through’ method, respectively

The recovery rate increased to reach approximately 100% in almost all soil columns (*Table 2*) and the highest reductions in the retention of phenol were observed at a depth of 5 cm and reached 19.05, 25.55 and 9.18% in CH, CO and MA, respectively (*Fig. 3*).

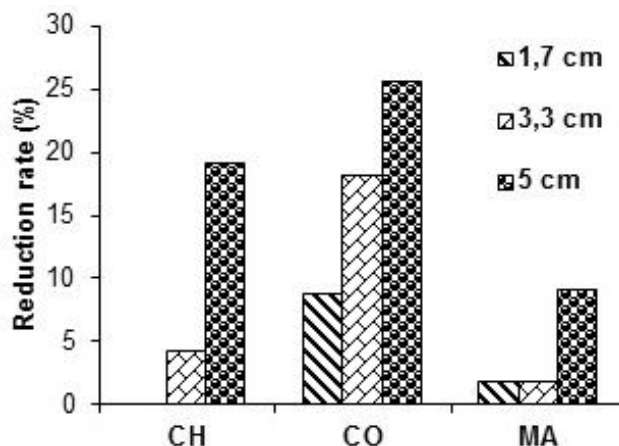
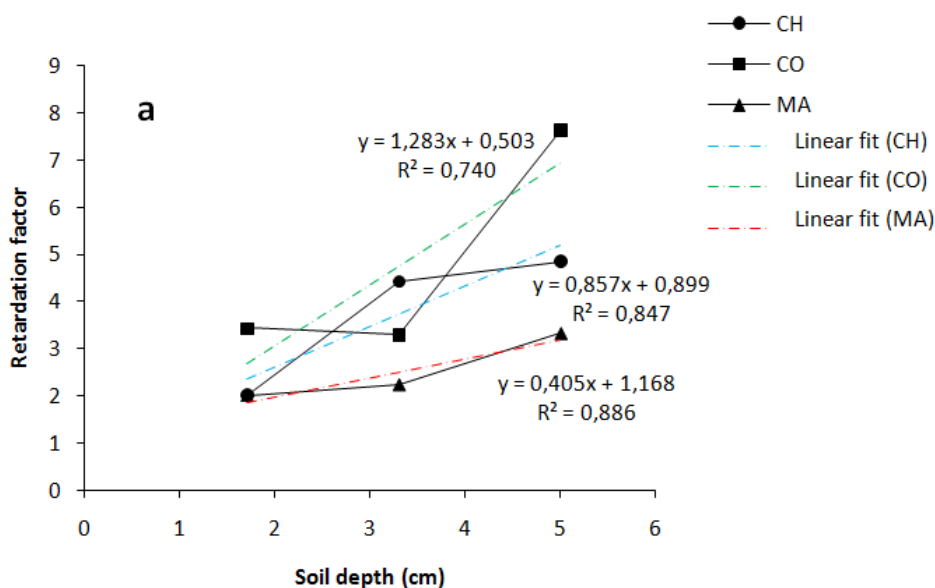


Figure 3. The reduction rate of the retention of phenol in various soil columns at different depths. Reduction rate is the difference between recovery rate of phenol dissolved in distilled water and recovery rate of phenol dissolved in a mixture of methanol/ distilled water (50:50, v/v)

Variation of the retardation factor with soil depth

As shown in Figure 4, the changes in the retardation factor values (R) measured using (LFT) method were found to be positively correlated with soil depth, for transport of phenol dissolved in distilled water (a), and followed the linear equations $R = 0.857 L + 0.899$ ($r^2 = 0.847$), $R = 1.283 L + 0.503$ ($r^2 = 0.740$) and $R = 0.405 L + 1.168$ ($r^2 = 0.886$) for CH, CO and MA, respectively, which suggested that the retardation of the transport of phenol increased with soil depth.

However, for transport of phenol dissolved in in a mixture of methanol/distilled water (50:50, v/v) (b), there is no significant correlations between retardation factor values (R) and soil depth, except for CH, where the retardation factor value (R) increased with soil depth and followed the linear equation $R = 0.102 L + 1.063$ ($r^2 = 0.992$). Therefore the retardation of the migration of phenol through CH increased with soil depth.



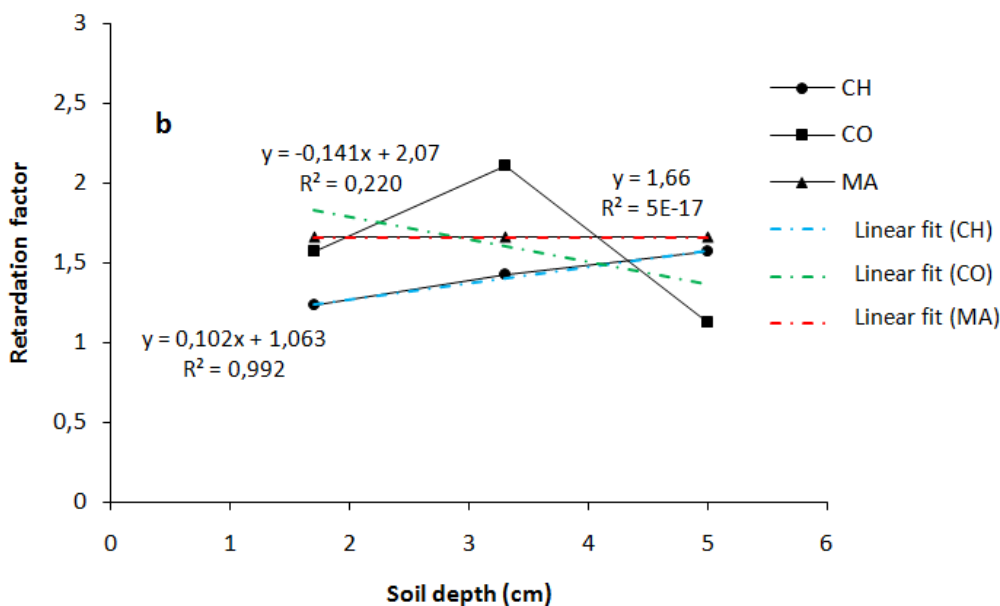


Figure 4. Relationship of the retardation factor (R) measured using (LFT) method for phenol dissolved in distilled water (a) and phenol dissolved in a mixture of methanol/ distilled water (50:50, v/v) (b) with soil depth

Discussion

Effect of soil composition on retention and migration of phenol

For transport of phenol dissolved in distilled water, the calculated values of k_d (Table 2) denote that retention of phenol was higher in CO-soil (1.33, 1.25 and 3.63 L kg⁻¹ at a depth of 1.7, 3.3 and 5 cm, respectively) than in MA-soil (0.37, 0.46 and 0.88 L kg⁻¹ at a depth of 1.7, 3.3 and 5 cm, respectively), which were both sandy soils, and it was even higher in CO-soil (1.33, and 3.63 L kg⁻¹ at a depth of 1.7 and 5 cm, respectively) than in CH-soil (0.38 and 1.46 L kg⁻¹ at a depth of 1.7 and 5 cm, respectively): soil which has the greatest clay content (Table 1), except at a depth of 3.3 cm (1.3 L kg⁻¹ for CH and 1.25 L kg⁻¹ for CO); it is well known that natural clays have good capacity for the adsorption of organic molecules (Djebbar et al., 2012). This fact is supported by high organic matter content (OM) of CO-soil (7.22%) compared to the two other soils (1.87% for CH and 2.72% for MA; Table 1); it is well known that, once in soil, organic pollutants bound to organic matter (OM) have reduced mobility due to their low solubility (Valentín et al., 2013). In spite of the high solubility of phenol compared to the other organic pollutants which are less hydrophilic, OM may be of great importance for adsorption of polar organic compounds like phenol (Delle Site, 2001). Based on retardation factor values (R), the soils can be presented in the following orders of retardation of phenol migration: CO > CH > MA at a depth of 5 cm, CH > CO > MA at a depth of 3.3 cm and CO > CH = MA at a depth of 1.7 cm (Table 2).

Effect of methanol on retention and migration of phenol in natural soils

The retention of phenol was very low with values of k_d ranging from 0.06 to 0.6 L kg⁻¹, which were much lower than those obtained from transport of phenol dissolved in distilled water. Therefore, the retardation factor values (R) were

remarkably lower than those observed from transport of phenol dissolved in distilled water (*Table 2*). It is well known that organic solvents such as methanol are often used to extract phenols from environmental samples (Afghan and Chau, 1989) and types of environmental matrices such as soils (Mahugo Santana et al., 2009); in addition, methanol, as one of the most polar organic solvent, was used by Osman and Saim (2013) to extract phenol, as one of the most polar contaminant, from soil. As a conclusion, our results confirm the opposite effect of methanol on the retention of phenol in our soils. In our case of study, this characteristic of methanol accelerated phenol vertical migration process through soil.

Conclusion

LFT method based on the one-dimensional convection-dispersion equation (*Eq. 1*) was used to investigate migration and retention of phenol in three types of natural soils. Our results confirm that organic matter can decrease the risk of groundwater contamination (Rodriguez-Liebana et al., 2014). The use of LFT method indicates that phenol was more retained in sandy soil “CO” rich in organic matter than in sandy soil “MA” and in loamy sand soil “CH” both poor in organic matter. The mobility of phenol decreased with increasing soil depth. The effect of methanol, as an organic solvent, on transport of phenol was also investigated. Results indicate that methanol increased remarkably the mobility of phenol through soil columns and decreased its retention. As a negative effect, methanol can increase the risk of groundwater contamination, but it can also be used for cleaning up soils contaminated with phenol, as a positive effect. These results can be generalized and utilized in practice to estimate migration of phenol at large scales in the northern regions of Algeria to predict the gravity of the risk of contamination of groundwater and soil. Further research can be conducted with undisturbed soil samples. In addition, other factors that may influence phenol migration in natural soil, such as pH, should also be considered for future studies.

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