The transport of three emerging pollutants through an agricultural soil irrigated with untreated wastewater

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ABSTRACT

The aim of this work was to determine the mobility of naproxen, carbamazepine, and triclosan through a wastewater-irrigated agricultural soil. Transport experiments were carried out using undisturbed soil columns taken at 10 and 40 cm depths. The mobilization of the pollutants was evaluated using two hydrological regimes transient flow for superficial columns and steady-state conditions for the sub-superficial columns. Results demonstrated that preferential flows are present in the superficial soil, and transient flow conditions facilitate the movement of the pollutants through the soil. Conversely, displacement of the contaminants in the sub-superficial soil columns was slower than that observed in the superficial soil. Triclosan was not found in the leachates of the soil columns at the two depths, indicating the strong retention of the compound by the soils. Conversely, naproxen and carbamazepine were determined in leachates of the soil columns at both depths. Retardation in the transport of carbamazepine was higher than that observed for naproxen in the two tested soils. Naproxen and triclosan showed some degree of dissipation, while carbamazepine was recalcitrant. It was concluded that the natural depuration system studied is capable of retaining and removing the studied pollutants and thus the risk of groundwater pollution is minimized.

Key words | pharmaceuticals, solute transport, sorption, undisturbed soil columns, wastewater reuse

INTRODUCTION

Wastewater reuse for agricultural irrigation is a practice gaining popularity worldwide, notably in developing countries where, due to the scarcity of treatment facilities, untreated wastewater is frequently used (Jiménez & Asano 2008). In the near future, an increment is expected in the volume of wastewater reused for agricultural irrigation because: (1) it represents an easy and cheap way to dispose of wastewater; (2) it is an option to relieve water stress in arid and semiarid zones; and (3) wastewater is a source of nutrients for the receiving soils, increasing crop yields and thus the profits of farmers (Jiménez 2006). However, wastewater reuse can spoil the quality of surface and subterranean water sources in and nearby the irrigated area. It is well known that wastewater is the main route of so-called emerging pollutants to enter into the environment. These pollutants are residues of substances used in everyday consumer products, such as pharmaceuticals, personal care products, flame retardants, plasticizers, additives, etc. (Petrovic et al. 2008). Conventional wastewater treatment systems have demonstrated the partial removal most emerging pollutants, and thus a fraction of the original compound along with a group of its by-products are released into the environment (Ratola et al. 2012). Despite emerging pollutants being present in water bodies at trace levels (concentrations are reported in the range of ng/L to μg/L), their occurrence is related to endocrine disruption effects as well as to systemic damage in vulnerable organisms (Naidoo et al. 2009; Brausch & Rand 2011; Vandenberg et al. 2012).
In zones where treated wastewater is used for agricultural purposes, emerging pollutants have been found in wastewater-irrigated soil and in the water bodies nearby (Gottschall et al. 2012). One example of a long-term irrigation system reusing untreated wastewater can be found in Tula Valley, central Mexico. In this zone, a flow of up to 5.2 million m$^3$/s of untreated wastewater, produced by the 22 million people living in Mexico City’s metropolitan zone, is used to irrigate 85,000 ha of agricultural fields (Jiménez & Chávez 2004). Even though wastewater used for irrigation does not receive any kind of treatment, a significant decay in the concentration of some contaminants, e.g., heavy metals, microbiological agents, and organic compounds (e.g., some emerging pollutants) has been observed during the transportation, storage, and application of wastewater on the soil, as reported by Jiménez & Chávez (2004) and Chávez et al. (2011). Wastewater depuration has demonstrated to be so efficient that the aquifer of the irrigated area, which has been recharged with the infiltrated wastewater for several decades, is used as the only drinking water source for the 500,000 people living in the Tula region, only chlorination being applied as disinfection prior to distribution. The removal and dissipation of emerging pollutants in the Tula Valley region may be attributed to natural attenuation processes, such as photodegradation, biodegradation, and sorption onto the soil. In this sense, it is necessary to elucidate how these pollutants mobilize or degrade in the soil in order to understand how depuration works, as well as its potential limitations. Soil column experiments are useful to describe the mobility of contaminants through the soil porous media, assessing separately the factors that influence this process. The vertical movement of the emerging pollutants toward the aquifer may depend on: (1) the chemical properties of the solutes; (2) the hydrological regime at which water infiltrates through the soil; and (3) the physical and chemical properties of the soil. The polarity of the molecules as well as the structure and texture of the soil also have a decisive role in delaying the transport of pollutants. On the other hand, preferential flows within the soil structure can accelerate the migration of pollutants into the aquifer (Wehrer & Totsche 2008). The aim of this work was to determine the transport of three emerging pollutants, namely the non-steroidal anti-inflammatory drug naproxen, the antiepileptic carbamazepine, and the antimicrobial agent triclosan, through a long-term wastewater-irrigated agricultural soil; the method being by undisturbed soil column experiments performed using two hydrological regimes.

**METHODS**

In this section, the methodology followed to obtain the soil columns, perform the transport experiments, and determine the target pollutants in the liquid and soil samples is described.

**Chemicals**

All of the target compounds as well as the surrogate standards ketoprofen and $[^2\text{H}_{16}]$ bisfenol-A and the internal standards $[^2\text{H}_4]$ 4-n-nonylphenol and clofibrac acid were purchased from Sigma-Aldrich (St Louis, MO). The solvents used for the analysis were high-performance liquid chromatography (HPLC) grade and purchased from Burdick and Jackson (Morristown, NJ). The derivatizing agents $N\text{-}t\text{ert}$-butyldimethylsilyl-$N\text{-}$methyltrifluoroacetamide (MTBSTFA) with 1% of $t\text{-}t\text{butyldimethylsilyl}$trifluoroacetamide and $N\text{-}t\text{hexadecylsilyl}$trifluoroacetamide (BSTFA) with 1% of trimethylsilylchlorane (TMSCI) were also obtained from Sigma-Aldrich. Oasis HLB extraction cartridges (200 mg, 60 cc) were bought from Waters (Milford, MA). For experiments, standard solutions containing the mixture of the target pollutants were prepared at concentrations of 10, 1, and 0.1 ng/$\mu$L using methanol as solvent. For the analytical procedure, internal and surrogate standards solutions were prepared, in methanol, at 10 ng/$\mu$L. Table 1 displays the relevant chemical properties of the target compounds.

**Soil columns’ collection and soil characterization**

Undisturbed soil columns were taken from a cropland where irrigation using untreated wastewater has occurred for the last 85 years.

Soil columns were collected in triplicate at two soil depths: 5–30 and 40–55 cm depth. The superficial soil columns were 25 cm high and 15 cm wide, while the sub-superficial columns were 15 cm high and 9 cm wide. To
obtain the soil columns, a pit of 90 cm deep was dug and the soil monoliths were carved directly in the wall of the pit. Once shaped, the monoliths were put into stainless steel cases, and liquid paraffin (at 60°C) was poured in the free space between the soil and the stainless steel case, in order to both fix the monolith to the column and avoid the border effect. The soil columns were stored in plastic bags at 4°C until the transport assays.

Before experiments, 50 g soil samples were taken at the top of the three columns corresponding to each soil depth and were combined to obtain one composite sample. Parameters shown in Table 2 were determined for the soil samples using standard methodologies (Carter & Gregorich, 2008).

The pH and the electrical conductivity were measured in a 1:1 soil:0.01 M CaCl2 suspension using a sensION156 HACH pH/conductivity meter. Concentration of soil organic carbon was determined using the Walkley-Black chromic acid wet oxidation method. Soil bulk density was obtained by weighing soil cores, which were taken directly in the field. The determination of the soil particle size distribution and texture was done using the Bouyoucos hydrometer method. Additionally, the specific surface area of the soil particles was measured using the Brunauer-Emmett-Teller (BET) nitrogen adsorption method. Lastly, the background concentration of the target compounds in the tested soils was quantified prior to the experiments.

### Transport experiments

The transport assays were carried out separately for superficial and sub-superficial soil columns by simulating irrigation events using a 0.01 M CaCl2 solution as irrigation water. Water was applied at the top of the soil columns using two hydrological regimes: (1) transient flow for the superficial soil columns, in order to emulate the infiltration of water through the soil during irrigation; and (2) steady-state flow conditions for the sub-superficial columns, aimed to simulate the slow infiltration of the irrigation water through the sub-soil. All the soil columns were weighed prior to the experiment in order to determine the initial water content.

The experimental setup used for the superficial soil columns is shown in Figure 1(a). In the first step, 950 mL of the 0.01 M CaCl2 solution, containing the conservative tracer bromide (0.1 M) – as KBr salt – and a mixture of the emerging pollutants (10 μg/L, each), were applied to each column. Water was poured in the 10 cm head space of the columns, and water was allowed to infiltrate through soil by gravity. After infiltration, the soil in the columns was allowed to dry for the next 4 days. Subsequently, eight irrigation events consisting only of the application of the 0.01 M CaCl2 solution were carried out in order to displace both the tracer and the target pollutants. The time lapse between

<table>
<thead>
<tr>
<th>Compound</th>
<th>Chemical structure</th>
<th>(^{a}pK_a)</th>
<th>(^{b}\log K_{ow})</th>
<th>(^{c}\text{Water solubility (mg L}^{-1})</th>
</tr>
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<tr>
<td>Naproxen</td>
<td><img src="" alt="Naproxen structure" /></td>
<td>4.1</td>
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</tr>
<tr>
<td>Carbamazepine</td>
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<td>2.3</td>
<td>17.7</td>
</tr>
<tr>
<td>Triclosan</td>
<td><img src="" alt="Triclosan structure" /></td>
<td>8.4</td>
<td>4.8</td>
<td>9.6</td>
</tr>
</tbody>
</table>

\(^{a}\text{Acidic ionization constant.}\)
\(^{b}\text{Log of the octanol-water partition coefficient.}\)
\(^{c}\text{Determined at 25°C.}\)

<table>
<thead>
<tr>
<th>Soil sample depth</th>
<th>Parameter</th>
<th>10 cm</th>
<th>40 cm</th>
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</thead>
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<tr>
<td></td>
<td>pH</td>
<td>8.01</td>
<td>8.14</td>
</tr>
<tr>
<td></td>
<td>Electrical conductivity (μS/cm)</td>
<td>1,546</td>
<td>1,601</td>
</tr>
<tr>
<td></td>
<td>Total organic carbon (mg/g)</td>
<td>25</td>
<td>18</td>
</tr>
<tr>
<td></td>
<td>Sand (%)</td>
<td>12</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td>Silt (%)</td>
<td>43</td>
<td>38</td>
</tr>
<tr>
<td></td>
<td>Clay (%)</td>
<td>45</td>
<td>49</td>
</tr>
<tr>
<td></td>
<td>Bulk density (g/cm³)</td>
<td>1.1</td>
<td>1.3</td>
</tr>
<tr>
<td></td>
<td>Texture</td>
<td>Clay loam</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Specific surface area (m²/g)</td>
<td>66.4</td>
<td>115</td>
</tr>
<tr>
<td></td>
<td>Naproxen (ng/g)</td>
<td>2.7</td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>Carbamazepine (ng/g)</td>
<td>6.2</td>
<td>1.4</td>
</tr>
<tr>
<td></td>
<td>Triclosan (ng/g)</td>
<td>9.4</td>
<td>6.3</td>
</tr>
</tbody>
</table>
each irrigation event was 4 days at a controlled temperature of 25 °C. Throughout the irrigation events, leachates were recovered at the bottom of the soil columns and discretized into 100 mL sub-samples.

For the sub-superficial columns, transport experiments under steady-state flow conditions were set up as shown in Figure 1(b). The irrigation solution (0.01 M CaCl₂) was supplied to the soil columns using a Gilson Miniplus 3 peristaltic pump at a constant flow rate of 0.125 mL/min. Water was sucked from the bottom of the columns using the same peristaltic pump (the outflow rate was therefore equal to the inflow rate). The pH and electrical conductivity of the leachate was measured online throughout the experiment and recorded in a data-logger. Leachate was discretized into 7.5 mL sub-samples, using a GE Frac-912 fraction collector, and stored in glass tubes. The transport experiment in the sub-superficial columns was divided into three stages. In the first stage, which took 48 h, irrigation was performed using the 0.01 M CaCl₂ solution alone until the equilibrium conditions were achieved (i.e., pH and electrical conductivity values in leachate were constant, stabilization of the flow rate at the entry and at the exit of the column). In the second stage, the mixture of the emerging pollutants (10 μg/L, each) and the conservative tracer bromide (0.1 M) were applied as a pulse. The pulse was carried out for 27 h, corresponding to 0.5 pore volume. In the third stage, the displacement of the compounds and the tracer was carried out by applying the 0.01 M CaCl₂ solution sole for 780 h, equivalent to 13 pore volumes.

Once transport experiments were concluded, the soil columns were weighed in order to determine the mass of water retained by the soil. Then, the soil monoliths were cut into three equal sections, soil was air-dried for 24 h and homogenized by sieving through a 2 mm pore sized metallic mesh. Concentration of the target pollutants was determined in both leachate samples and the soil collected at the end of the experiment. Both the distribution of the target pollutants throughout the soil column at the end of the experiment and the mass balance was obtained from the analysis of the soil samples.

Emerging pollutants’ analysis

Concentrations of the target pollutants were determined in the leachates following the procedure developed and validated by Gibson et al. (2007). The pH of the liquid samples was adjusted to 2 using sulfuric acid (98%); then, the surrogate standards ketoprofen and [²H₁₆] bisphenol-A were added. Samples were passed through the Oasis HLB cartridges, which were previously conditioned twice with
5 mL of acetone and once with 5 mL of 5% acetic acid solution, using a vacuum flow equivalent to 5 mL/min. After passing the liquid samples, naproxen and carbamazepine were eluted from cartridges with 5.5 mL of a mixture 40:60 of acetone:sodium bicarbonate 0.1 M, adjusted to pH 10; then, the cartridges were dried for 1 h with vacuum at a flow rate of 10 mL/min. Following the drying step, triclosan was eluted with 5 mL of acetone. Both phases were concentrated using a gentle atmosphere of ultra high purity nitrogen and the water remaining in the sample was removed by adding anhydrous sodium sulfate. Lastly, the internal standards clofibric acid and [2H4]4-nonylphenol were added to samples and derivatization was performed, through the production of methyl-silyl derivatives, using the agents MTBSTFA for naproxen and carbamazepine, and BSTFA for triclosan.

Analysis of soil was performed using the method proposed and validated by Durán-Álvarez et al. (2009). Extraction of the analytes was done through the pressurized liquid extraction technique, using a Dionex ASE® 200 device. For this, 5 g of soil and 2 g of diatomaceous earth, the latter used as dispersing agent, were accurately weighed in 22 mL ASE stainless steel cells and extracted using the hexane:acetone:acetic acid (49:49:2 v/v/v) mixture. Surrogate standards were added to samples prior to extraction. The extraction conditions were as follows: two cycles, 100 °C, 10.34 MPa, 0 min of pre-heat, 5 min of heating time, 5 min of static time, and flush at 60%. The extracts were evaporated to reach a volume of ~3 mL. After evaporation, 20 mL of HPLC-grade water was added to the concentrates. The resulting solutions were passed through the Oasis HLB cartridges, previously conditioned as stated above. Naproxen and carbamazepine were eluted from cartridges using the 40:60 acetone:sodium bicarbonate 0.1 M solution, while triclosan was eluted using 5 mL of a 50:50 acetone:dichloromethane solvent mixture. After elution, the sample preparation procedure was the same as described for liquid samples.

Separation and quantification of the analytes were carried out using a HP 6890 gas chromatograph in tandem with a HP 5397 mass selective detector. The chromatographic column was a fused silica capillary column (30 m × 0.25 mm, 0.25 μm of film thickness). The carrier gas was helium at a constant flow of 1 mL/min, and 1 μL of sample was injected in the splitless mode. The oven temperature program was as follows: 100 °C for 1 min, ramp of 20 °C/min to 280 °C, and 280 °C for 10 min. The detector was used in the selected ion monitoring (SIM) mode. The temperature of the electron impact source was 230 °C, with electron energy of 70 eV. Calculation of the analytes' concentration was done through the internal standard method. Quality assurance was guaranteed by the use of surrogate standards. The recoveries obtained were 90–96% for ketoprofen and 88–91% for [2H16] bisfenol-A.

**Data analysis**

The transport parameters for target emerging pollutants and tracer (i.e., retardation factor, solute dispersion, and determination coefficient) were determined by the analysis of the breakthrough curves. For the sub-superficial soil columns, the solution of the classic convection-dispersion equation for the solutes was achieved using the CXTFIT 2.1 code (Toride et al. 1999). The code was used in the inverse mode for either equilibrium or non-equilibrium conditions. The solution of equation was fitted to the experimental data by the least-squares optimization method. Due to the non-symmetry of the breakthrough curves obtained for the superficial columns, the CXTFIT code could not be executed using these results. Therefore, determination of the retardation factor in the superficial soil columns was done by the visual analysis of the breakthrough curves.

**RESULTS AND DISCUSSION**

In this section, the discussion of the most relevant results of the transport experiments is shown. The results obtained for the superficial and sub-superficial soil columns are displayed separately.

**Transport of emerging pollutants in superficial soil columns**

Figure 2 shows the breakthrough curves of both the conservative tracer bromide and the pharmaceutical compounds naproxen and carbamazepine. In this figure, the relative...
concentration of the target emerging pollutants in leachates is plotted with the total volume of leachate normalized to the pore volume of the soil column ($V_{\text{accumulated}}/V_{\text{pore}}$; pore volume = 1,035 mL). The asymmetry observed in the bromide breakthrough curve indicates the rapid movement of water and solutes through the soil columns (Melamed et al. 1997). This behavior may be attributed to: (1) the occurrence of preferential paths within the soil monolith; and (2) the repulsion forces between the bromide anion molecule and the negatively charged soil components (i.e., soil organic matter and clay), which is known as anionic exclusion (Gvirtzman & Gorelick 1994). Within the soil porous network, preferential paths occur as the result of both the presence of roots and stones, the activity of macrobiota (e.g., worms, ants, or beetles) and the tillage activities. The preferential paths may consist of either a single channel through the soil profile or the complex connectivity of the pores within the soil matrix. In any case, preferential paths result in the fast movement of the solutes (the tracer and pollutants) through the soil monolith (Kung et al. 2000). Regarding anion exclusion, this phenomenon has been previously reported in similar soils (García-Gutiérrez et al. 2004; Prado et al. 2011). Anion exclusion may be patent not only for bromide but also for the negatively charged contaminants such as naproxen and the dissociated fraction of triclosan at the pH value of the soil (see $pK_a$ values of triclosan in Table 1). Mass balance for bromide showed that neither degradation nor accumulation nor production occurred throughout the soil column experiment.

Regarding the target emerging pollutants, naproxen and carbamazepine were found in the leachate samples at different times throughout the experiment, while triclosan was not observed in leachate samples. This behavior may be caused by two phenomena, which may occur simultaneously: (1) the irreversible sorption of the antimicrobial agent onto the soil particles; and (2) its rapid biodegradation by the native microorganisms of the soil. Retardation factors of the compounds detected in the leachates could be determined by the analysis of the breakthrough curves, establishing the value of this factor as the center or gravity of the curve. Retardation factor was 3.8 for carbamazepine and 1.5 for naproxen. Higher retardation of carbamazepine compared to naproxen has been reported by others (Chefetz et al. 2008), and may be explained by the higher affinity of the antiepileptic drug to the soil organic matter. Conversely, naproxen molecules, occurring in their negative dissociated form at the pH values of soils (Table 1), may be repelled to an important extent by the soil organic matter and clay particles.

In spite of the molecules of naproxen undergoing anion exclusion by the soil particles, the mobilization of this pharmaceutical through the soil was not as rapid as that observed for the bromide. This is explained by the organic nature of the molecule, which maintains an organic domain consisting of a double ring structure. Through the organic moiety, naproxen molecules may be sorbed by both the soil organic matter and clay (Chefetz et al. 2008).

According to the mass balance, 14.5% of the amount of naproxen entering into the soil columns was degraded, while for carbamazepine this value was zero. In the case of triclosan, the compound was not found either in the leachate samples or the soil at the end of the experiments, indicating that the compound was degraded at some point of the experiment. Ready biodegradation of triclosan in aerobic soils has been reported by Cha & Cupples (2010). In general, little is known about the microorganisms leading to the biodegradation of emerging pollutants in soil. Recent evidence points to bacteria *Pseudomonas putida* and *Sphingomonas* sp. and fungus *Trametes versicolor* as the microorganisms responsible for the biodegradation of pharmaceutically active substances in the soil (Kagle et al. 2009; Girardi et al. 2011).

![Breakthrough curves of the bromide tracer and the target emerging pollutants corresponding to the superficial soil columns.](https://iwaponline.com/jwrd/article-pdf/4/1/9/377993/9.pdf)
Transport in sub-superficial columns

For the sub-superficial soil columns, bromide breakthrough curves were shown to be symmetrical (Figure 3). This indicates that transport of water and solutes was carried out without the occurrence of preferential flows. This may be due to the high content of expansive clays in the soil. Bromide retardation factor was below 1 (0.85), indicating the occurrence of an anion exclusion phenomenon, caused by the negative charge that dominates in the soil. The slight tailing observed in the bromide breakthrough curve may be caused by an intra-particle diffusion phenomenon, which may occur in soils where micro pores are abundant, e.g., clayey soils (Brusseau & Rao 1990).

The presence of naproxen and carbamazepine in leachates was observed prior to the application of the pulse containing the target compounds. This indicates the occurrence of a readily translocable initial mass of these compounds in the studied soils, which can move into the aquifer during rain events. Triclosan was not observed in the leachate samples, indicating that this compound can be irreversibly retained by soil or degraded within the soil matrix. In accordance with this behavior, it is recommended that the metabolites generated by the biodegradation of triclosan as well as their potential to reach the groundwater should be studied.

According to the breakthrough curves (Figure 4), naproxen was more displaceable than carbamazepine in the sub-superficial soil columns. The same behavior was observed by Chefetz et al. (2008) in transport experiments using disturbed soil columns. Table 3 shows the transport parameters of the target compounds and the tracer, obtained using the CXTFIT mode.

Similar to that observed in the superficial soil columns, the retardation factor of naproxen was lower than obtained for carbamazepine. The rapid transport of naproxen through the sub-superficial soil may be caused by the same anion exclusion observed for the bromide tracer. On the other hand, the low mobility of carbamazepine and triclosan through the soil can be explained by their sorption onto the soil organic material, as has been demonstrated by Williams et al. (2006) and Lozano et al. (2010). The notorious dispersion observed in the carbamazepine breakthrough curves is indicative that there are strong interactions between the soil components and the pharmaceutical compound, such as sorption and probably intra-particle diffusion, as was suggested by the tracer tests.

Mass balance showed losses of naproxen and triclosan (13 and 98%, respectively) during the experiment, proving the degradation of the target compounds, notably for

Table 3 | Transport parameters of the tracer and the target pollutants for the sub-superficial soil

<table>
<thead>
<tr>
<th>Solute</th>
<th>Retardation factor</th>
<th>Dispersion coefficient</th>
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</thead>
<tbody>
<tr>
<td>Bromide</td>
<td>0.85</td>
<td>2.7 × 10⁻³</td>
</tr>
<tr>
<td>Naproxen</td>
<td>1.8</td>
<td>3.6 × 10⁻³</td>
</tr>
<tr>
<td>Carbamazepine</td>
<td>8.2</td>
<td>4.3 × 10⁻⁴</td>
</tr>
</tbody>
</table>

* Determined by the least-squares optimization method by comparing the modeled and the experimental data.
triclosan. Degradation of naproxen and triclosan was similar for both soil depths. On the other hand, high recovery of carbamazepine in leachate and the soil (>97%) was indicative of its recalcitrance, which is consistent with those reported in the literature (Clara et al. 2004). In the case of carbamazepine, degradation was higher in the sub-superficial soil than in the superficial soil.

CONCLUSION

Infiltration of wastewater through soil proved to be efficient in retaining the emerging pollutants tested. Natural attenuation mechanisms, such as sorption onto the soil organic matter and biodegradation, occurred in the soil porous media, delaying the arrival of these contaminants to the aquifer and thus minimizing the risk of groundwater pollution. The mobilization of the target emerging pollutants was higher in the superficial soil due to both preferential paths through the soil column and the anion exclusion of the negatively dissociated compounds. Conversely, delay in the transport of the pollutants in the sub-superficial soil was higher than that observed in the superficial soil, indicating that the closure of preferential paths upon the humidification of soil in the steady-state hydrological regime contributes to the higher retention of these compounds in the soil matrix as well as to their degradation.

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