

## **Influence of Olive Oil Mill Waste Amendment on Fate of Oxyfluorfen in Southern Spain Soils**

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### **Abstract**

Oxyfluorfen herbicide residues have been previously reported in surface and ground water in the Guadalquivir river basin in Spain. Soil factors and processes (sorption, dissipation and leaching) influencing the potential offsite transport of oxyfluorfen to surface and ground water were characterized in laboratory experiments for two soils from Southern Spain. The influence of olive-oil mill waste amendment on the soil processes was also determined. Oxyfluorfen sorption in unamended soils was not significantly affected by soil active components (organic matter and clay fraction). However, a significant increase in soil sorption of the herbicide was observed with upon amendment. Laboratory half-life values for oxyfluorfen in unamended sandy clay loam and silty clay soils at 40% moisture content and 25°C were 29 and 19 days, respectively, and it increased by a factor of 4 upon amendment. Oxyfluorfen's mobility along soil profile increased with the amendment in leaching studies at 25°C, suggesting that dissolved organic matter of amendment can play an important role enhancing soil vertical movement of the pesticide. The increase in soil adsorption and persistence observed with olive-oil mill waste amendment can promote its losses by runoff following winter application, whereas vertical movement of oxyfluorfen can be enhanced following spring application when olive-oil mill waste amendment is also applied.

**Keywords:** Crop, Degradation, Herbicide, Leaching, Sorption

**Abbreviations:** DOC, dissolved organic carbon; DOM, dissolved organic matter; DT<sub>50</sub>, half-life value; HPLC, high pressure liquid chromatography; OC, organic carbon OM, organic matter;

## 1. Introduction

Olive orchard is one of the most important crops in the Mediterranean area, with Spain, Italy and Greece being the main producers of olive oil. In Spain, olive tree cultivation occurs primarily in the Southern region (Jaen, Córdoba and Seville provinces) along the Guadalquivir River basin. The rainfall period, at the end of fall and the start of winter, coincides with herbicide application before harvesting, which can enhance the water contamination problems associated with pesticide use in this regional crop system [1].

Countries with a high production of olive oil generate very large amounts of olive-oil mill waste. This organic residue is generally promoted as a natural, low-cost, carbon-recycling amendment in olive cultivar regions of Southern Europe [2, 3] and is particularly valuable in semiarid locations of this cultivation [4]. The addition of organic residues has also been shown to greatly influence the behavior of herbicides and other organic chemicals in soils; however, these effects depend upon the specific herbicide compound, soil and amendment characteristics [5--7].

Oxyfluorfen, an herbicide applied in several **crop** cultivations as e.g., sunflower, onion, citrus and asparagus, has been detected in ground and surface waters in Southern Spain mainly associated with olive crops [1]. The concentration levels were three to five times the drinking water limit (0.1 µg L<sup>-1</sup>). This contamination was largely related to runoff and leaching processes in winter and spring time, respectively. However, no studies relating soil processes of oxyfluorfen in water have been reported so far, except for that which was conducted under very different climatic and agronomic application conditions [8].

The soil factors or processes involved in the fate of pesticides in soil are well known and assessed elsewhere [9-11]. Soil processes that decrease potential water contamination by pesticides are those that contribute to their disappearance: i) as bound residues, irreversibly adsorbed or entrapped in small soil pores [12, 13], and ii) disappearance by any type of degradation route [9] i.e., photodegradation in top surface soil layers [14, 15], and biodegradation [16].

The objective of this research was to assess the soil factors and processes influencing the behavior of oxyfluorfen as related to its use on olive cropped soils in Southern Spain. The effect of olive-oil mill waste, added as soil organic amendment, was also determined since this agronomic management has been shown to either decrease or enhance herbicide contamination potential, depending on the soil and molecule [5, 17].

## 2. Materials and methods

### 2.1. Herbicide and soil characteristics

Oxyfluorfen (2-chloro-4-trifluoromethylphenyl-3-ethoxy-4-nitrophenyl ether) is a yellowish crystalline compound, which has a vapor pressure of 0.0267 mPa at 25°C and solubility in water of 0.1

**Comentario [PH1]:** crop cultivations?  
"applied in crops" makes no sense

mg L<sup>-1</sup>. The herbicide was purchased from Dr. Ehrenstorfer (Augsburg, Germany) with a purity of 98%. <sup>14</sup>C-labeled oxyfluorfen (oxyfluorfen-benzene-Ph-UL-<sup>14</sup>C, specific activity 10.2 mCi mmol<sup>-1</sup>, radiochemical purity 99.6%) was graciously provided by Dow Agrosiences LLC, Indianapolis, IN.

Two soils devoted to olive crop use were chosen for this study: A sandy clay loam soil classified as calcic cambisol [18] from an experimental farm located in Coria del Rfo (Seville, SW Spain) and a silty clay soil (chromic luvisol, [18]) from an experimental farm located in Mengibar (Jaen, SE Spain). Physicochemical properties of the soils are shown in Table 1 and were determined by the Analysis Service of IRNAS. Soil pH was measured as soil/deionized water 1:2.5 (w/w) suspension. The texture was determined by a Boyoucos hydrometer method, carbonates by the Bernard calcimeter method and organic matter (OM) by the Walkley and Black method [19]. Dissolved organic carbon (DOC) was extracted from soils with 0.01 M CaCl<sub>2</sub> 1:20 (w/v) and extracts were analyzed in a Shimadzu TOC-V<sub>CSH</sub> model. For laboratory experiments, soil field samples were taken from the top 0-20 cm. Sandy clay loam and silty clay soils were amended in the laboratory (5% w/w) in order to observe possible changes in pesticide behavior in soil. The properties of the composted olive-oil mill waste were pH 6.7, 80.9% organic matter, and DOC = 6820 mg kg<sup>-1</sup>. The pH was determined as soil/deionized water suspension 1:5 (w/w) and DOC was extracted with 0.01 M CaCl<sub>2</sub> 1:5 (w/v) and analyzed in a Shimadzu TOC-V<sub>CSH</sub> model. Original and amended soil samples were air dried, sieved through a 2 mm mesh sieve, and stored at 4°C until experiments were carried out.

## 2.2. Soil porosity

The soil pore-size distribution (from 600 to 0.004 μm) was determined by mercury intrusion porosimetry using a Micromeritics Autopore IV mercury intrusion porosimeter. Porosity was determined in soil samples from the soil column leaching experiment described below, once they were air-dried. Triplicate soil cores of approximately 1 g were sampled and dried at 90 °C for 24 h before analyzed. The surface tension of mercury,  $\gamma = 0.48 \text{ N m}^{-1}$ , and a mercury solid contact angle of  $\theta = 171.3^\circ$  were assumed in the Washburn equation (1) to calculate the pore size distributions:

$$p r = -2 \gamma \cos \theta \quad (1)$$

where  $p$  is the capillary pressure,  $r$  is the radius of the pore where mercury intrudes,  $\gamma$  is the surface tension of mercury and  $\theta$  is the contact angle of mercury on the surface of the solid sample.

## 2.3. Sorption study

<sup>14</sup>C-labeled oxyfluorfen was dissolved in methanol and aliquots were used to prepare aqueous herbicide solutions in 0.01 M CaCl<sub>2</sub> for the sorption study (0.064, 0.035, 0.015 and 0.006 μg mL<sup>-1</sup>). In the adsorption experiment, 1 g of each soil (unamended and amended soils) was weighed in triplicate into glass centrifuge tubes and 30 mL of the four different herbicide solutions were added. Blank samples (without soils) were also used as a control. The samples were shaken at  $20 \pm 2^\circ\text{C}$  for 24 h and then centrifuged at  $370 \times g$  for 30 min. The supernatants were analyzed and the differences between initial and final solution concentrations were assumed to be due to sorption.

Oxyfluorfen desorption was determined at different initial concentrations for each soil in triplicate (0.064 μg mL<sup>-1</sup> for clay loam soils, and 0.035 μg mL<sup>-1</sup> for silty clay soils). Desorption was performed

immediately after adsorption by replacing 20 mL of the supernatants with 20 mL 0.01 M CaCl<sub>2</sub>. The final volume of 30 mL was mixed using a vortex mixer, shaken for 24 h, centrifuged, and the supernatant was analyzed. This was done for three desorption steps.

For the adsorption--desorption analysis, a 3 mL aliquot of each supernatant was added to 15 mL scintillation cocktail (EcoLite, cocktail, ICN Biomedicals, Costa Mesa, CA) and the amount of <sup>14</sup>C-oxyfluorfen was determined by liquid scintillation counting with a Packard 1500 counter (Packard Instruments, Downer Grove, IL.)

The adsorption--desorption coefficients ( $K_f$  and  $N_f$ ) were obtained by plotting the amount of oxyfluorfen adsorbed ( $C_s$ ,  $\mu\text{g g}^{-1}$ ) versus the equilibrium concentration ( $C_e$ ,  $\mu\text{g mL}^{-1}$ ) and fitting the isotherms to the Freundlich equation, Eq. (2):

$$C_s = K_f C_e^{N_f} \quad (2)$$

$C_s$  for adsorption was calculated from the differences between initial and equilibrium oxyfluorfen concentrations in solution. Organic C-normalized  $K_f$  values ( $K_{f-oc}$ ) were calculated by dividing  $K_f$  by the organic carbon content of the soils. The hysteresis value  $H$ , Eq. (3), was calculated as a measure of the sorption reversibility:

$$H = N_{f_{des}}/N_{f_{ads}} \quad (3)$$

The  $H$  value ranges from 0, which corresponds to total irreversibility, to 1, which implies full reversibility, although some values  $>1$  has been reported [20].

#### 2.4. Dissipation study

Amended and unamended soils (500 g) were placed in glass bottles (inner diameter 9 cm) and treated with 6 mL of an ethanol solution of oxyfluorfen ( $100 \text{ mg L}^{-1}$ ) to approximately achieve the field mean dose ( $4 \text{ L ha}^{-1}$  of commercial oxyfluorfen 24% (w/v) a.i.). After 1 h equilibration, water was added to the soils to obtain two different soil moisture contents, 20 and 40%. The soils were incubated at 25°C in the dark. The soil moisture content was maintained during the experiment adding the appropriate amount of water. 10 g soil samples were removed from the bottles in duplicate, ten times, from 0 up to 84 days and extracted with 20 mL methanol. The soil samples were centrifuged ( $5724 \times g$ , 10 min) and the supernatants were filtered (0.45  $\mu\text{m}$ ) prior to analysis of oxyfluorfen residues by high pressure liquid chromatography (HPLC).

#### 2.5. Leaching study

Soil movement of oxyfluorfen was studied in glass columns (20 cm length  $\times$  3 cm id), with glass wool at the bottom of the column to avoid soil particle losses. These columns were hand packed with 180 g of amended and unamended soils. Sea sand was added on the top of the column to allow uniform distribution of the herbicide and water in the soil column. The soil column was saturated with 0.01 M CaCl<sub>2</sub> and 0.68 mL of a methanolic solution of oxyfluorfen ( $100 \text{ mg L}^{-1}$ ) was applied to roughly reach the field mean dose ( $4 \text{ L ha}^{-1}$  of commercial oxyfluorfen 24% (w/v) a.i.). 15 mL 0.01 M CaCl<sub>2</sub> were applied daily and the leachates were collected, filtered and analyzed by HPLC. After 20 days, the soil columns were divided into 5 cm increments of depth and the soil was extracted with methanol. Leaching experiments were carried out in duplicates at 5 and 25°C, a temperature range typical for autumn to late

**Comentario [PH2]:** what does this mean? two columns? or are these special columns?

spring in Southern Spain.

## 2.6. Oxyfluorfen soil extraction and analytical conditions

Oxyfluorfen soil extraction during dissipation and leaching studies was accomplished in a soil/methanol solution 1:2 (w/v). Previously, it was checked that this extraction method gave a recovery efficiency of 98.5% and a relative standard deviation (RSD) value of 1.68% ( $n = 4$ ). Samples from the soil dissipation and leaching experiments were analyzed by HPLC with photodiode array detection (2996 PDA). A Waters (Milford, MA) 1525 system equipped with a 717 autosampler was used. The column was a Nova-Pack C18 column (150 mm length  $\times$  3.9 mm id) with a mobile phase of acetonitrile/water 70:30 at a flow rate of 1 mL min<sup>-1</sup>, 25  $\mu$ L injection volume and UV detection at 215 nm. External calibration curves with standard solutions between 0.05 and 1 mg L<sup>-1</sup> were used in the calculations. The linear calibration resulted in correlation coefficients  $>0.99$  and mean RSD values  $<20\%$  ( $n = 3$ ). The limit of quantification (LOQ) of the analytical method for the determination of oxyfluorfen was 0.05 mg L<sup>-1</sup>.

Comentario [PH3]: explanation

## 3. Results and discussion

### 3.1 Soil properties and porosity

The soil properties relevant to pesticide soil behavior [9-11] and their changes upon amendment are summarized in Table 1. The increase in organic carbon (OC) with the olive-oil mill waste amendment was slightly higher in the sandy clay loam soil than in the silty clay one. The soil pH decreased in the sandy clay loam soil and it did not change in the silty clay soil due to the presence of carbonate. Large differences were observed in the DOC content between the two original soils with greater DOC in the silty clay soil compared to sandy clay loam soil. Addition of amendment increased the DOC of both soils, but the increase was much greater for sandy clay loam soil as compared to silty clay soil, indicating that a large part of the added DOC from olive-oil mill waste amendment was strongly sorbed to the soil surfaces of silty clay soil. This is very important with respect to the nature of the soil surfaces exposed to the solute molecules such as herbicides [7, 21].

Total cumulative soil pore volumes and their distribution versus pore radius are shown in Fig. 1. The higher porosity of sandy clay loam soil is mainly due to the higher amount of large pores ( $>10 \mu\text{m}$ ), or transmission pores, which accounted for 85% of the total porosity. Whereas in silty clay soil, the pores  $>10 \mu\text{m}$  only accounted for 56% of the total porosity. The inverse relationship between the high porosity at high pore radii and clay contents has been previously reported for other soils [22, 23].

Figure 1 shows that the total porosity increased upon organic amendment in both soils. This increase, mainly due to the development of large size or transmission pores ( $>10 \mu\text{m}$ ) is higher in silty clay than in sandy clay loam soil. The different amounts of DOC in amended sandy clay loam and silty clay soil may be related to the different porosity development in each soil. The increase in DOC in both soils after amendment (Table 1) is lower in the case of silty clay than in sandy clay loam soil. This suggests a stronger interaction of dissolved organic matter (DOM) with the components of silty clay soil, possibly with its high clay fraction, which could be related to the greater development of  $>10 \mu\text{m}$  pores. Furthermore, this clay-DOM interaction in amended silty clay soil may have implications for the nature of the surfaces of soil components and pores [7, 20], and hence in the pesticide-soil particles interactions

[7] and in the pesticide movement with water [17] as it will be discussed below.

### 3.2. Sorption study

Oxyfluorfen sorption isotherms on unamended and amended soils were well described by the Freundlich equation ( $R^2 > 0.99$ ) (Table 2). Values of the Freundlich slopes ( $N_f$ ) were  $<1$ , indicating a high dependence of adsorption on initial solution concentration. Similar  $N_f$  values allowed comparison of the  $K_f$  values. Those values for unamended soil did not correspond to the soil clay fraction and OM content, soil components described previously as relevant to adsorption. The lower  $K_f$  of unamended silty clay soil could be related to its higher DOC content; it is known that DOM molecules (of polar character and small size) may compete with herbicide molecules for the sorption sites in soils [7, 20] especially at the clay surfaces, which are abundant in this soil. However, the amendment with olive-oil mill waste increased the adsorption of oxyfluorfen in both soils,  $K_f$  increased from 28 to 72 (sandy clay loam soil) and from 17 to 56 (silty clay soil). This was mainly due to the increase in the OC content after the addition of the organic amendment (Table 1). The increase in  $K_f$  was similar for both soils (44 for sandy clay loam soil and 39 for silty clay soil) as it was directly related to the increase in OM (2.4 for sandy clay loam soil and 1.9 for silty clay soil) upon amendment.

An increase in the herbicide adsorption upon soil amendment with olive-oil mill waste was also found for simazine and diuron [5, 7]. Values of  $K_f$  normalized by the OC content of the soils ( $K_{f-oc}$ ) showed considerable variability (Table 2), indicating that organic C alone cannot account for the different adsorptive properties of the soils [20]. Addition of the olive-oil mill waste had contrasting effects on the  $K_{f-oc}$  values of the two soils, decreasing in sandy clay loam soil and increasing in silty clay soil. In this case, DOM can interact with the more abundant clay fraction in silty clay soil and enhances the efficacy of OM added to soil for oxyfluorfen adsorption, as compared to sandy clay loam soil [20, 23]. This is confirmed by the different amounts of DOC found in both soils (Table 1). The small increase in measured DOC for silty clay soil, as compared with that for sandy clay loam soil seems to support this argument. By the contrary, the higher amount of DOC in the amended sandy clay loam soil contributes to interactions in solution with oxyfluorfen molecules, decreasing the efficacy of added OM, as observed in the different  $K_f$  values for the unamended soils. The interactions of DOM with pesticides are influenced by many different factors, e.g. pH, soil properties, chemical and physical properties of the pesticide, and interactions between pesticide--soil, pesticide--DOM, and DOM--soil. Thus often yields inconsistent results with regard to sorption and pesticide transport [24].

Desorption of oxyfluorfen from both soils was hysteretic; very little, if any, of the adsorbed chemical desorbed. The addition of olive-oil mill waste did not affect oxyfluorfen soil desorption; the high hysteresis of oxyfluorfen sorption--desorption remained, as revealed by the H values, close to zero (Table 2).

### 3.3. Dissipation study

Dissipation curves obtained during the 84 days of incubation were fitted to first-order kinetics and the parameters for these equations are summarized in Table 3. In unamended soils, half-life ( $DT_{50}$ ) values at 20% moisture levels were 39 days for sandy clay loam soil and 75 days for silty clay soil.

Increasing the soil moisture to 40%,  $DT_{50}$  decreased to 19 days for sandy clay loam soil and 29 days for silty clay soil and are in general agreement with those reported by other authors [25--27]. Pesticide dissipation is the result of a complex combination between different processes and depends on soil and pesticide properties [28]. Despite the photolysis process reported for oxyfluorfen dissipation in soil [14], we observed a very low photodegradation of the herbicide after 96 h Xenon lamp irradiation (22% in sandy clay loam soil and 10% in silty clay soil) (data not shown). Thus, we considered that photodegradation did not play an important role in the dissipation of oxyfluorfen in our soils and other dissipation processes should be considered. Oxyfluorfen dissipation can be attributed to microbial degradation [29, 30] and depends on microbial species and temperature [29]. Some authors have found that an increase in soil moisture level consistently decreased pesticide persistence [31, 32] and it has been related either to an increase in pesticide bioavailability [33] or/and to an enhancement of soil microbial activity [34]. Differences in degradation rates with soil moisture could be more related to changes in microbial population activity. However, the disappearance of the herbicide could be also contributed by the formation of bound residues [35], which could be supported by the high irreversibility measured in the adsorption of oxyfluorfen in our soils. That process is recognized to have a relevant contribution to the fate of pesticides in soils.

Amending the soils with olive-oil mill waste resulted in an increase in the persistence of oxyfluorfen in both soils (Table 3). In amended soils at 40% moisture,  $DT_{50}$  values were 88 and 120 days for sandy clay loam and silty clay soils, respectively. Although  $DT_{50}$  values are not merely related to the sorption coefficient ( $K_f$ ), as other authors have observed [28], those values seem to confirm that soil OM and clay content and organic amendment increase herbicide soil persistence. The increase on oxyfluorfen sorption upon amendment, observed in both soils, protects the herbicide against biodegradation. The chemical-physical interactions (bonds) between pesticide and OM added could mask sites of actions where pesticide degradation can take place, thus resulting in its higher persistence.

### **3.4. Leaching experiment**

No oxyfluorfen was recovered in any soil leachate, which suggests a low leaching potential of oxyfluorfen, in accordance with other authors results [12, 36, 37]. The oxyfluorfen residue distribution in soil columns at 5°C (Fig. 2) showed higher amounts of residual herbicide in the first 5 cm, indicating very low mobility and low occurrence of other dissipation processes at this temperature. However, a higher mobility was observed in sandy clay loam soil as compared to silty clay soil. No significant differences appear evident in oxyfluorfen soil distribution with the amendment and in both soils at 5°C. This result points out a high persistence of oxyfluorfen in the top soil and hence an increase in its runoff potential when applied to soil at winter, which is the case of olive crop in Southern Spain, and could be facilitated by olive-oil mill waste amendment.

Higher amounts of oxyfluorfen residues were found in silty clay soils than in sandy clay loam soils at 25°C (Fig. 2). This could be due to the higher volume of small pore sizes found in the first one (Fig. 1). Some authors observed that herbicide molecules can be entrapped in this type of pores, enhancing its soil retention [13]. With respect to oxyfluorfen distribution along the soil column at 25°C, higher depths were reached in amended soils than in unamended soils. The increase in DOC after soil amendment (Table 1)

could have some effect on oxyfluorfen soil movement at this temperature. DOM water movement has resulted in the development of new pores in amended soils in the medium size region (100--10  $\mu\text{m}$ ; Fig. 1), which could facilitate downward movement of oxyfluorfen molecules. In fact, the downward movement resulted more relevant in the amended silty clay soil at 25 °C (Fig. 2). Hence, olive-oil mill waste soil amendment is not a recommended management practice when oxyfluorfen is applied to soil because it enhances downward mobility of the herbicide.

The low recovery of oxyfluorfen in the sandy clay loam soil columns can be attributed to microbial degradation and differences in the microbial population of soils and/or formation of bound residues. This assumption is also in accordance with the results of the dissipation study, shorter  $DT_{50}$  were observed in the sandy clay loam soil.

#### 4. Concluding remarks

The results of this study show the increase in soil sorption of oxyfluorfen when olive-oil mill waste amendment is used, and how this affects processes related to environmental behavior. Persistence was related to bound residues formation and biodegradation processes increasing  $DT_{50}$  values upon amendment by a factor of 4. Leaching through the soil profile depended on soil composition, temperature and soil porosity, which increased upon amendment. At low temperature, oxyfluorfen accumulated in the top cm of soils. In contrast, at 25°C oxyfluorfen moved through the soil profile, showing greater vertical mobility upon amendment.

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The authors have declared no conflict of interest

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**Figure 1.** Total porosity and pore-size distribution of sandy clay loam and silty clay soils before and after amendment, measured by mercury intrusion porosimetry

**Figure 2.** Oxyfluorfen residue distribution along soil column after leaching experiment in unamended and amended sandy clay loam and silty clay soils.

**Table 1.** Physicochemical properties of original and amended soils

	pH	OC (%)	DOC (mg kg <sup>-1</sup> )	Clay (%)	Silt (%)	Sand (%)	CaCO <sub>3</sub> (%)
Sandy clay loam soil	8.1	0.6	50	20.4	8.9	70.7	0.6
Silty clay soil	8.1	1.0	112	51.6	42.1	6.3	33.0
Amended sandy clay soil	7.6	2.0	230	20.4	8.9	70.7	0.6
Amended silty clay soil	8.1	2.2	176	51.6	42.1	6.3	33.0

**Table 2.** Freundlich coefficients for oxyfluorfen sorption and desorption on soils

	$K_f$	$N_f$	$R^2$	$K_{f-oc}$	$K_{fd}$	$N_{fd}$	$R^2$	$H$
Sandy clay loam soil	28 (26--30) <sup>a)</sup>	0.82 (0.81--0.83)	0.999	4375	1.57 (1.38--1.78)	0.06 (0.03--0.09)	0.710	0.073
Silty clay soil	17 (14--22)	0.79 (0.74--0.84)	0.993	1598	0.84 (0.88--0.80)	0.02 (0.01--0.03)	0.647	0.025
Amended Sandy clay loam soil	72 (61--85)	0.87 (0.84--0.90)	0.998	3547	1.77 (1.66--1.89)	0.02 (0.01--0.03)	0.592	0.023
Amended silty clay soil	56 (41--78)	0.85 (0.80--0.90)	0.991	2605	1.12 (1.07--1.18)	0.02 (0.01--0.03)	0.603	0.023

<sup>a)</sup> Values in parentheses correspond to the standard errors applied to the coefficients

**Table 3.** First-order degradation constant ( $K$ ), half-lives ( $DT_{50}$ ) and regression coefficients ( $R^2$ ) of oxyfluorfen dissipation data in sandy clay loam and silty clay soils incubated at 25°C and two different soil moisture contents (20 and 40%)

	Moisture (%)	$K$	$DT_{50}$ (day)	$R^2$
Sandy clay loam soil	20	-0.0174	39 (38.0--41.9) <sup>a)</sup>	0.981
	40	-0.0370	19 (18.0--19.5)	0.986
Silty clay soil	20	-0.0092	75 (65.9--87.4)	0.864
	40	-0.0235	29 (26.3--33.4)	0.899
Amended sandy soil	20	nc	nc	nc
	40	-0.0078	88 (77.6--102.6)	0.928

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Amended	20	nc	nc	nc
silty clay soil	40	--0.0059	120 (102.5--145.3)	0.893

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<sup>a)</sup> Numbers in parentheses correspond to the standard errors applied to the coefficient

nc, not calculated because no dissipation was observed during the experiment