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TEBOTRIONE HALF-LIFE IN SOILS WITH DIFFERENT ATTRIBUTES

Meia-Vida do Tembotrione em Solos com Diferentes Atributos

ABSTRACT - Before recommending an herbicide, it is necessary to know its interactions with the soil attributes that will influence its sorption and half-life in the soil. This knowledge is an essential condition for minimizing any negative effects that may result from its application. However, due to the great diversity of soil and climate in Brazil, especially for products of recent use in the country, such as the herbicide tembotrione, this knowledge is often not available. This may be the main cause of occurrence of intoxication reports in crops carried out in succession to corn sprayed with this herbicide. In order to reduce possible impacts on successor crops and the contamination of surface and groundwater using the high-performance liquid chromatography, the tembotrione half-life was determined in soils with different attributes. The evaluated soils were a Red-Yellow Latosol with and without liming (Viçosa, MG), Red-Yellow Latosol (Rio Paranaíba, MG), Yellow Latosol (Sooretama, ES), and Red-Yellow Latosol with and without liming (Gurupi, TO). The results showed that liming might favor tembotrione degradation in the soil, as well as direct relationships between values of tembotrione half-life and contents of clay and soil organic matter. Tembotrione half-life in samples of Red-Yellow Latosol (without liming) collected in Viçosa and Rio Paranaíba, MG, was higher than 90 days, indicating carryover risks in successive crops to corn sprayed with tembotrione in these regions.

Keywords: herbicide, environmental impact, persistence in soil.

RESUMO - Antes de fazer a recomendação de um herbicida, é preciso conhecer suas interações com os atributos do solo que irão influenciar a sua sorção e a meia-vida no solo. Esse conhecimento é condição essencial para serem minimizados os eventuais efeitos negativos que possam resultar de sua aplicação. Todavia, devido à grande diversidade de solo e clima do Brasil, principalmente para os produtos de uso recente no País, como é o caso do herbicida tembotrione, esse conhecimento na maioria das vezes não está disponível. Essa pode ser a principal causa da ocorrência de relatos de intoxicação em cultivos realizados em sucessão a cultura do milho pulverizada com esse herbicida. A fim de reduzir possíveis impactos em culturas sucessoras e a contaminação de águas superficiais e subterrâneas, utilizando a cromatografia líquida de alta eficiência, neste trabalho foi determinada a meia-vida do tembotrione em solos com diferentes atributos. Os solos avaliados foram: Latossolo Vermelho-Amarelo de Viçosa-MG com e sem calagem, Latossolo Vermelho-Amarelo de Rio Paranaíba-MG, Latossolo Amarelo de Sooretama-ES e Latossolo Vermelho-Amarelo de Gurupi-TO com e sem calagem. Conclui-se que a calagem pode favorecer a degradação do tembotrione no solo e que existem relações diretas entre valores da meia-vida do tembotrione e teores de

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argila e matéria orgânica dos solos. A meia-vida do tembotrione em amostras do Latossolo Vermelho-Amarelo coletadas nos municípios de Viçosa (que não recebeu calagem) e Rio Paranaíba, MG, foi superior a 90 dias, indicando riscos de carryover em cultivos sucessivos ao milho pulverizados com o tembotrione nessas regiões.

Palavras-chave: herbicida, impacto ambiental, persistência no solo.

INTRODUCTION

Knowing the environmental factors that directly or indirectly affect herbicide efficiency and its interactions with soil attributes is an essential condition to minimize any negative effects that may result from its application. This is most important when the herbicides have a long period of activity in the soil, guaranteeing weed control. These products should preferably be applied to crops that have a long critical period of interference prevention (Oliveira Jr and Regitano, 2009; Silva and Silva, 2013). The use of these products without the knowledge of their interactions with soil colloids can result in serious agronomic problems, such as intoxication of crops in succession to the crop treated with the herbicide, in addition to contamination of surface and groundwater resulting from leaching (Andrade and Stigter 2009; Andrade et al., 2010).

Herbicide recommendations without the knowledge of their half-life under environmental conditions (soil attributes and climate) of the site to be applied are common in Brazil. Among these herbicides, tembotrione stands out. It belongs to the chemical group of tricetones, which inhibits the enzyme 4-hydroxyphenylpyruvate dioxygenase (HPPD), important in carotenoid synthesis, and controls many species of grass weeds and some dicotyledonous (Abit et al., 2009; Silva and Silva, 2013). This herbicide is used in extensive areas cultivated with corn in Brazil and in recent years, there have been reports of carryover of this compound in succession crops to corn.

The persistence and movement of herbicides in the soil are dependent on the sorption and physical, chemical, and biological degradation, contributing to the biogeochemical transformation of the molecule (Ahmad et al., 2001; Andréa and Luchini, 2002; Oliveira Jr and Regitano, 2009). All these variables are affected by soil chemical, biological, and physical characteristics and the chemical properties of herbicides (Inoue et al., 2003; Lourencetti et al., 2008). The wide use of herbicides with a long persistence in the soil has caused residue accumulation in the environment (Pareja et al., 2012; Delwiche et al., 2014). The main damages are the alteration of the soil biota, volatilization of toxic compounds, surface runoff, and leaching of these compounds (Sarmah et al., 2004).

The half-life of an herbicide in the soil refers to the time to the dissipation of 50% of its initial amount in the soil (Silva and Silva, 2013). This parameter depends on factors such as organic matter, physical properties, and soil pH, in addition to microbial degradation and light, among others (Pires et al., 2005; Sarmah and Close, 2009). The higher the half-life is, the greater the risk of environmental problems, such as the contamination of soils and surface and underground water springs (Inoue et al., 2003; Pires et al., 2005; Andrade et al., 2010). This variable is essential for estimating the herbicide persistence in the environment and the potential for contamination of groundwater (Cohen et al., 1984; Gustafson, 1989).

For success in the use of herbicides in weed management, it is important to know the factors that influence their activity and soil stability (Zabaloy et al., 2011). Tembotrione is registered for weed control in corn (Karam et al., 2009), mainly grasses (Dan et al., 2010) and was classified as highly mobile and with a variable persistence in the soil (PMRA, 2012). However, for Brazil, with a great diversity of soils and climate conditions, it is necessary to study the dissipation kinetics of this herbicide in soils with different attributes (Dong et al., 2015). Considering the importance of using tembotrione and the few studies on its behavior in the soil, other researches are needed to elucidate its dynamics in the soil. In the search for this knowledge, this study was carried out in order to determine, by high-performance liquid chromatography, tembotrione half-life in different soil types cultivated with corn in Brazil.

MATERIAL AND METHODS

Soil collection and preparation

The experiment was carried out in a greenhouse at the Federal University of Viçosa. Samples of the soils Red-Yellow Latosol from Viçosa, MG (LVAV), Red-Yellow Latosol from Rio Paranaíba, MG (LV), Yellow Latosol from Sooretama, ES (LA), and Red-Yellow Latosol from Gurupi, TO (LVAG) were collected at a depth of 0-20 cm. Soil samples of the Red-Yellow Latosol from Viçosa, MG, and Gurupi, TO, were submitted to liming with CaCO_3 using an acid neutralization curve, which resulted in two more samples, totaling six soils (Table 1).

Table 1 - Results of chemical and physical analyses of soils used in this study*

Solo	pH	Ca ²⁺	Mg ²⁺	Al ³⁺	H+Al	(t)	V	m	OM
	(H ₂ O)	(cmol _c dm ⁻³)					(%)		
LVAG ⁽¹⁾	5.70	0.99	0.52	0.00	4.70	1.71	26.70	0.00	3.00
LVAG cc ⁽²⁾	6.50	1.30	0.70	0.00	4.50	2.30	33.50	0.00	3.00
LVAV ⁽³⁾	5.10	0.04	0.06	1.60	5.30	1.66	2.90	90.70	2.07
LVAV cc ⁽⁴⁾	6.50	0.34	0.15	0.00	2.52	0.79	60.00	0.00	2.07
LVAR ⁽⁵⁾	6.50	1.20	0.40	0.00	2.64	1.70	39.00	0.00	2.18
LA ⁽⁶⁾	6.30	2.90	1.00	0.00	1.32	4.18	76.00	0.00	2.20
	Coarse sand	Fine sand		Silt	Clay		Textural class		
	(dag kg ⁻¹)								
LVAG	25.00	30.00		6.00	39.00		Sandy clay		
LVAV	11.00	10.00		17.00	62.00		Very clayey		
LVAR	10.00	33.00		16.00	41.00		Clay		
LA	60.00	19.00		1.00	20.00		Sandy loam		

* Analyses carried out at the Laboratório de Análises de Solo Viçosa, according to the methodology of the Brazilian Agricultural Research Corporation (Embrapa, 1997). (t) = effective cation exchange capacity; V = base saturation; m = Al³⁺ saturation; OM = organic matter.

⁽¹⁾ Red-Yellow Latosol from Gurupi, TO; ⁽²⁾ Red-Yellow Latosol after liming from Gurupi, TO; ⁽³⁾ Red-Yellow Latosol from Viçosa, MG;

⁽⁴⁾ Red-Yellow Latosol after liming from Viçosa, MG; ⁽⁵⁾ Red-Yellow Latosol from Rio Paranaíba, MG; ⁽⁶⁾ Yellow Latosol from Sooretama, ES.

Experimental design

The experiment was conducted in a randomized block design, in a split-plot arrangement. Plots consisted of six soil types, subplots consisted of seven evaluation times (0, 15, 30, 45, 60, 75, and 90 days after herbicide application - DAA), and the sub-subplots consisted of two managements (without and with herbicide application). Four replications were used, and each pot had a capacity of 1 dm³. In the second experiment, the experimental design was a randomized design in a split-plot arrangement. Plots consisted of three tembotrione doses (0, 100.8, and 302.4 g ha⁻¹) and subplots consisted of seven evaluation times (0, 15, 30, 45, 60, 75, and 90 days after herbicide application - DAA), being used as a substrate a Red-Yellow Latosol from Rio Paranaíba, MG.

Herbicide application and sampling

Tembotrione was applied at doses of 302.4 g ha⁻¹ (three times the commercial dose) in all soils and 100.8 g ha⁻¹ (commercial dose) in the Red-Yellow Latosol from Rio Paranaíba, MG, using a high-precision CO₂-pressurized sprayer (TTI 110 02 tips calibrated to apply 150 L ha⁻¹). The used containers had 1 dm³ and were filled with 1 kg of soil, which was homogenized after application and also at the sampling time (factor A). Sampling was carried out in triplicate and the samples were transferred to dark-colored containers and stored in a freezer for laboratory analyses.

Extraction and determination of tembotrione by chromatography

For tembotrione extraction from soils, 5.0 mL of a 0.5 mol L⁻¹ KCl solution in was added to polypropylene tubes containing 2 g of soil. The tubes were then placed under vertical stirring at 80 rpm for one hour at a temperature of 27 ± 2 °C. After stirring, the samples were centrifuged at 2,260 x g for six minutes. Subsequently, the supernatant was removed from each sample and 5.0 mL of a 0.5 mol L⁻¹ KCl solution was added to the tubes, which were again stirred for one hour and centrifuged for six minutes. The supernatant was added to the previous one (5 + 5 mL) and an aliquot of 1.5 mL was taken from this mixture, being filtered on a Millipore filter with a 0.45 µm PTFE membrane for further analysis by high-performance liquid chromatography (HPLC).

The tembotrione determination was carried out in a Shimadzu LC 20AT high-performance liquid chromatography system equipped with a DAD detector (Shimadzu SPD 20A) and stainless steel column (Shimadzu VP-ODS Shim-pack 280 mm x 4.6 mm I.D. x 4.6 µm PS). The linearity of this equipment was evaluated before the linearity of the proposed method. For this, solutions containing tebuthiuron at concentrations of 1.0 to 120.0 µg L⁻¹ were injected in methanol. The obtained areas allowed constructing an analytical curve for each concentration, with correlation coefficients of 0.99.

Chromatographic conditions for the analysis were as follows: mobile phase composed of water and acetonitrile at a proportion of 40:60 (v/v), flux of 1.4 mL min⁻¹, injection volume of 20 µL, and wavelength of 225 nm. Tembotrione retention time was approximately 5.9 minutes under these conditions. Herbicide concentration was estimated by means of analytical curve parameters obtained by chromatography by the external calibration method.

Used models

A non-linear model (biexponential) was used to interpret the data, following a methodology proposed by Blumhorst (1996). The half-life time (T_{1/2}) was calculated as the time required for 50% of the initial concentration to be degraded.

Statistical program

The regression analyses were performed using the SigmaPlot 12.0 program (Exact Graphs and Data Analysis) for Windows.

RESULTS AND DISCUSSION

The modified method of extraction and analysis of tebuthiuron was validated for the main figures of merit: selectivity, linearity, limits of detection, quantification, precision, and accuracy, according to the recommendations of INMETRO (2003) and ANVISA (2003).

Tembotrione degradation was faster in the Yellow Latosol. The lowest herbicide concentration was observed at 90 days after application, with a half-life of 32 days (Figure 1 and Table 2).

The highest tembotrione degradation in the Yellow Latosol is due to the lower sorption of the herbicide in this soil, which is related to the higher sand content and lower clay and organic matter contents, as reported for polar herbicides in a Cambisol in Germany (Dechene et al., 2014). In general, the highest binding of a substance with soil colloids occurs by surface adsorption, hydrogen bonds, and Van der Waals interactions (Silva and Silva, 2013), with less availability of binding sites in sandy soils. Because the herbicide is less sorbed, more of it is available for degradation by microorganisms (Andrighetti et al., 2014) because sorbed molecules are protected from microbial degradation (Villaverde et al., 2008). In general, half-life depends on sorption, leaching, degradation and/or biological transformation regulating the concentration and flux in the soil (Oliveira Jr and Regitano, 2009), which are affected by soil chemical, biological, and physical characteristics (Lourencetti et al., 2008). This relationship between sorption and degradation is evident for other compounds, such as mesotrione, an herbicide of the same mechanism of action of tembotrione, in which the fast degradation became limited by the molecules adsorption rate in the soil (Shaner et al., 2012).

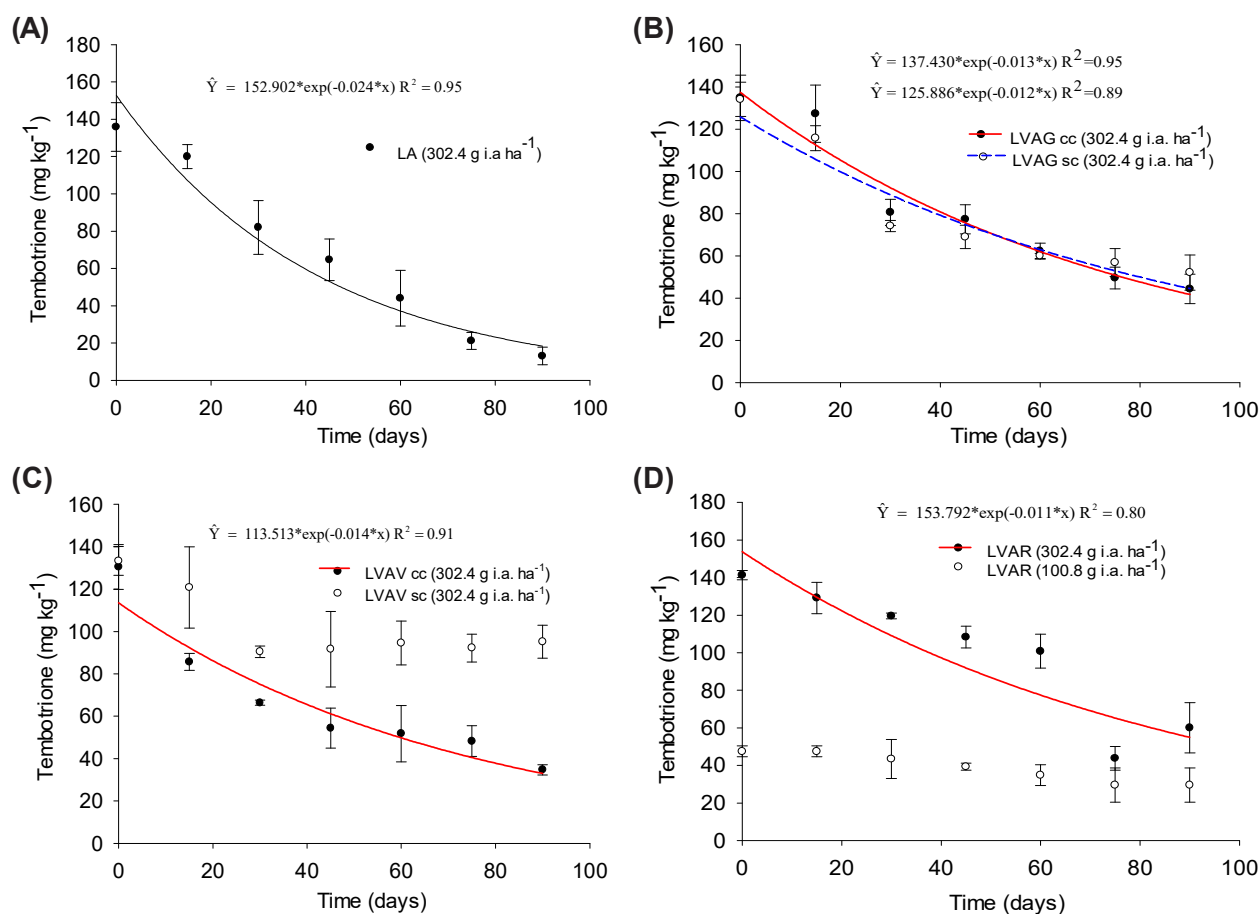


Figure 1 - Tembotrione concentration ($\mu\text{g kg}^{-1}$) in samples of (A) Yellow Latosol from Sooretama, ES (LA), (B) Red-Yellow Latosol with and without liming from Gurupi, TO (LVAG), (C) Red-Yellow Latosol with and without liming from Viçosa, MG (LVAV), and (D) Red-Yellow Latosol with and without liming from Rio Paranaíba, MG (LVAR) collected from 0 to 90 days after tembotrione application.

Table 2 - Half-life ($T_{1/2}$) values determined as a function of tembotrione concentration ($\mu\text{g kg}^{-1}$) over time in samples of Yellow Latosol from Sooretama (LA), Red-Yellow Latosol from Rio Paranaíba (LVAR), Red-Yellow Latosol with and without liming from Viçosa (LVAV), and Red-Yellow Latosol with and without liming from Gurupi (LVAG) collected from 0 to 90 days after tembotrione application

Soil	Equation	$T_{1/2}$ (days)
LA ⁽¹⁾ (302.4 g ha ⁻¹)	$\hat{Y} = 152.902 \cdot \exp(-0.024 \cdot x) R^2 = 0.95$	32
LVAG CC ⁽²⁾ (302.4 g ha ⁻¹)	$\hat{Y} = 137.430 \cdot \exp(-0.013 \cdot x) R^2 = 0.95$	52
LVAG SC ⁽³⁾ (302.4 g ha ⁻¹)	$\hat{Y} = 125.886 \cdot \exp(-0.012 \cdot x) R^2 = 0.89$	60
LVAV CC ⁽⁴⁾ (302.4 g ha ⁻¹)	$\hat{Y} = 113.513 \cdot \exp(-0.014 \cdot x) R^2 = 0.91$	51
LVAV SC ⁽⁵⁾ (302.4 g ha ⁻¹)	No adjustment	>90
LVAR ⁽⁶⁾ (302.4 g ha ⁻¹)	$\hat{Y} = 153.792 \cdot \exp(-0.011 \cdot x) R^2 = 0.80$	61
LVAR ⁽⁶⁾ (100.8 g ha ⁻¹)	No adjustment	>90

* Application of three times or the dose of 100.8 g ha⁻¹ of tembotrione. ⁽¹⁾ Yellow Latosol from Sooretama, ES; ⁽²⁾ Red-Yellow Latosol after liming from Gurupi, TO; ⁽³⁾ Red-Yellow Latosol from Gurupi, TO; ⁽⁴⁾ Red-Yellow Latosol after liming from Viçosa, MG; ⁽⁵⁾ Red-Yellow Latosol from Viçosa, MG; ⁽⁶⁾ Red-Yellow Latosol from Rio Paranaíba, MG.

Tembotrione half-life was higher in the Red-Yellow Latosol without liming from Viçosa, being higher than 90 days, while in the corrected soil, it was 51 days (Figure 1 and Table 2).

The highest tembotrione half-life in the Red-Yellow Latosol without liming from Viçosa, when compared to that observed in the same soil with liming, can be attributed in large part to the action of edaphic microorganisms (Andrighetti et al., 2014).

The reduction of half-life with an increasing soil pH can be explained by a decrease in the number of cationic molecules present in the soil (Trigo et al., 2014), thus reducing tembotrione sorption. Because this herbicide has an acid character and is negatively charged in solution, it is repulsed in situations where soil pH is increased. Thus, only compounds in the molecular form are adsorbed (Trigo et al., 2014), making it difficult the degradation by microorganisms.

Tembotrione degradation was faster in the soils where liming was carried out (Red-Yellow Latosols from Viçosa and Gurupi), with lower half-life values (Figure 1 and Table 2).

The higher tembotrione degradation in corrected soils is due to the fact that liming favored degradation since soils with lower pH values have a higher availability and toxicity of mineral elements, such as Mn and Al (Table 1), and deficiency of metal cations, such as Ca, Mg, and K, which may impair the microbiota (Andrighetti et al., 2014). Soil pH correction and higher fertility favored diuron degradation, which was attributed to higher microbial activity under higher fertility conditions (Rocha et al., 2013) since soils with better fertility may have higher microbial activity (Silva et al., 2010). Soil microorganisms can use herbicides as a source of nutrients and energy or even modify the chemical structure of the compound without obtaining energy for its growth in a process of cometabolism. This was observed for diuron, which presented a half-life in sterilized soil or not of 129 and 15 days, respectively (Barra Caracciolo et al., 2005). In addition to the lower tembotrione sorption, which favors availability to microorganisms, a high positive correlation is usually observed between soil pH and degradation of acid herbicides (Quan et al., 2015).

Tembotrione half-life is lower when applied at higher doses and higher when applied at lower doses, with values of 61 days in the Red-Yellow Latosol from Rio Paranaíba when applied at a dose of 302.4 g ha⁻¹ and higher than 90 days when applied at a dose of 100.8 g ha⁻¹ (Figure 1 and Table 2).

The lower tembotrione half-life when applied at higher doses (Figure 1 and Table 2), when compared to the application of lower dose in the Red-Yellow Latosol from Rio Paranaíba, can be explained by its higher initial concentration, which can be influenced by frequency and application rate, weather (sunlight, temperature, humidity, and wind), microorganisms, soil and water pH, and plant species (Lu et al., 2014; Zhang et al., 2015). A higher initial herbicide concentration may potentiate the results due to the development of an abnormal capacity of microbial communities to use the herbicide as a carbon source, but in some cases, it may also delay degradation by hindering the adaptation and even present toxicity to native microbial communities (Mercurio et al., 2015). On the other hand, lower herbicide concentrations may limit microbial community adaptation in order to herbicide metabolization due to a lower carbon availability (Ahtiainen et al., 2003). This can also occur when herbicide sorption is fast and high, leading to slow degradation. Therefore, in future experiments, commercial doses should be used for each soil to determine the herbicide agronomic persistence.

Tembotrione half-life in samples of Red-Yellow Latosol collected in Viçosa (the sample that did not receive liming) and Rio Paranaíba, MG, was higher than 90 days, indicating carryover risks in successive crops to corn sprayed with this herbicide in these regions. A direct relationship was observed between the values tembotrione half-life and contents of clay and soil organic matter. In addition, soil liming may favor the degradation of this herbicide.

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