

Effect of some Additives on the hydrolysis, persistence and the downward Movement of Glyphosate and Fluazifop-butyl Herbicides

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Abstract: The first objective was to determine the effect of additives and water type on the degradation of glyphosate and fluazifop-butyl herbicides. Both glyphosate and fusillade were strongly hydrolysed in hard water more than Nile water and distilled water. Most additives were decreased glyphosate persistence in all types of water. Glyphosate plus glue was more persistent than glyphosate alone and when mixed with the following additives (urea, glycerine, dioleate, monooleate, paraffin and mineral oil), respectively. Fusilade alone was more persistent than its mixture with the additives rape seed oil, mineral oil, paraffin oil, dioleate and monooleate, respectively. The second objective was to deal with the effect of additives soil texture interactions on persistence and leaching of glyphosate and fluazifop-butyl herbicides. Persistence of glyphosate in clay soil was lower than in sandy soil. Most additives increased half life of glyphosate in clay soil, while glyphosate additive mixtures decreased its stability in sandy soil. Fluazifop-butyl was rapidly dissipated in clay soil more than the sandy soil. Whereas most of the used adjuvant decreased fluazifop-butyl half life in each soil. Glyphosate was not detected in the soil leachate of clay and sandy soil column. The majority of glyphosate and fluazifop-butyl amount was located in the top layer of soil column followed by fewer amounts in the successive layers. Whereas glyphosate and fluazifop-butyl could be transferred from the sub- surface to the following layer from sandy soil with some additives (surfactants) to the following layer. Meanwhile fusillade with surfactants was detected in sandy soil column leaching with non significant amount.

Key words: Glyphosate, Fusilade, Hydrolysis, Persistence, The downward Movement, Leaching and Additives.

INTRODUCTION

Many factors influencing the effectiveness of herbicide applied to the foliage of plants, decomposition, physico-chemical characteristics of spray solution that governing the herbicide deposit and retention, absorption by the leaf surface. Hydrolysis is the reaction of pesticide with water usually resulting in the cleavage of the

molecule into smaller, more water-soluble portions and in the formation of new C-OH or C-H bonds. The degradation rate of a given organic compound is dependent on pH, temperature and the presence of certain metal ions or other catalyzing substances. Meanwhile, leaching is a fundamental soil process whereby constituents are lost from the soil profile by the action of percolating liquid water. Leaching is responsible

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for the transport of herbicide from agriculture areas and can lead to the subsequent contamination of ground and surface water. The dissipation of fluazifop-butyl (Fusilade at 0.39 kg a.i./ha) and fluazifop in a sandy loam soil in a soyabean plot and in the same soil under controlled laboratory conditions. Fluazifop-butyl was found to be completely transformed fluazifop within 3 d, and fluazifop was not detected 32 d after application (Negre *et al.*, 1993). Glyphosate, ¹⁴C-atrazine and ¹⁴C-alachlor at 8.34 and 9.64 mM, resp., were applied to soil columns with 0.88 M potassium bromide (conservative tracer for water) following 2, 13 or 26 weeks of root decomposition.

Most of the herbicide remained in the top 9 cm of soil following leaching at 17 ml/min for 9 h on 2 consecutive days (Zins *et al.*, 1991). The vertical mobility glyphosate and imazapyr in two soils with different physical and chemical compositions was studied. Glyphosate leaching in the studied soils was very low; and the imazapyr presented high leaching although was higher in the sandy loam texture soil than in the one of clay (Souza *et al.*, 1999). The environmental fate of a polyacrylamide thickening agent, PATA, (NALCOTROL II) formulated alone and in combination with glyphosate + surfactant herbicide, in distilled-deionized, 3 surface and 2 groundwater samples and were tested in outdoor conditions. The results of leaching experiments with PATA-herbicide solutions indicated that acrylamide leached fastest from columns containing sand only (peak at 2 days), followed by sandy loam, then silt loam (peak at 2 days followed by constant leaching for 5 days); acrylamide was not detected in runoff water from any treatment >4 days after exposure (Smith *et al.*, 1997).

Leaching of glyphosate (N-(phosphonomethyl glycine) and /or its metabolite AMPA (aminomethylphosphonic acid) in sandy loam soil with 13-14% clay. The mean yearly concentration of leached glyphosate and/or AMPA was significantly below 0.1 µg/l. A significant difference between the soil residual concentrations of AMPA was seen, the higher concentration was found where low-tillage had been practiced and where round up had been used several times in the years before sampling of the sandy soil (Fomsgaard *et al.*, 2003). Both glyphosate and AMPA can leach through structured soils; they thereby pose a potential risk to the aquatic environment (Kjær *et al.*, 2005). Fluazifop-butyl was rapidly hydrolyzed by a mixed culture enriched from a landfill leachate. The hydrolysis product, fluazifop, was further degraded by the same mixed culture to 4- (5-trifluoromethyl-2-pyridyloxy) phenol. The metabolite accumulated when fluazifop was in the medium as a sole source of carbon and energy whereas it was further degraded when sodium acetate or sodium succinate was added to fluazifop as cometabolites (Gennari *et al.*, 1991). The aim of this work was to study the effect of additives and water type on the degradation behavior of glyphosate and fluazifop-butyl herbicides. Secondly to demonstrated the additives soil type interactions on persistence and leaching of glyphosate and fluazifop-butyl herbicides

MATERIALS AND METHODS

Herbicides; Round up 48%WSC (Glyphosate) supplied by Monsanto and Fusilade 12.5 EC (Fluazifop-butyl) supplied by Syngenta.

Additives; Glue, Glycerine, Rape seed oils, Paraffin oils, Monoleate (Monoethylene glycol mono - oleate) and Diolate (Monoethylene glycol Di-oleate).

Table (1). Chemical analysis of soils and water types.

Chemical properties of the used soils and water										
Samples Type	PH	EC Ds/m	Cation meq /L				Anion meq/L			
			Na+	K+	Ca2+	Mg2+	Co32-	Hco3-	CL-	So42-
El-Gharbia (Clay)	7.57	5.81	16.15	0.39	13.7	3.86	-	2.85	59.3	15.95
El-Arish (Sand)	7.12	1.14	6.05	0.15	3.20	1.70	-	1.70	5.80	3.60
Nile water	7.95	0.43	2.95	0.07	0.85	0.43	-	0.60	2.70	1.00
Tap water	7.43	0.44	2.96	0.07	0.90	0.47	-	0.50	2.75	1.15

Mechanical and physical properties of used soils					
Location	Texture	Clay (%)	Silt (%)	Sand(%)	WHC (%)
El-Gharbia (Clay)	Silty clay	50.1	40.6	10.35	40
El-Arish (Sand)	Sandy	10.0	4.45	85.64	20

Effect of additives and water type on herbicide persistence in water

The persistence of the tested herbicides was determined in three types of water: Distilled , Nile and hard water. Each treatment was carried out under lab conditions. Two litters from each type of water were adjusted to the recommended rate, and additives at the tested rate. Three replication samples of 50 ml were taken from each treatment at different time ;1,3, 5, 10, 15, 21, 30 days (Glyphosate) and 0, 1, 3, 5, 10, 15, 21 (fluazifop-butyl) (Negre *et al.*, 1988) for extraction, and residue determination.

Fate of herbicide in soil in the presence and absence of additives.

Herbicides as spray volumes were added to the two selected soils at their recommended rates and mixed thoroughly. Five hundred grams of tested soils were placed in plastic bag and water was added to reach the soil to 65% of their water field capacity (F.C). The bags were closed and incubated at 25 °C and water was added when need to maintain the soils at 65% F.C. Each treatment was done in three replicates, after incubation period 0,1, 5,10, 20, 40, 60, and 80, days (glyphosate)and 0,3 ,7,14, 21and 42, days with fluazifop-butyl (Negre *et al.*, 1988). Fifty gram soil samples were taken from clay soil and soil and prepared for pesticide residue analysis

Effect of additives and soil type on the downward movement of herbicides.

To study the leaching, PVC columns (45 cm * 6.25cm.i.d.) were used. Each column was segmented to 5 segments. The segment height was 10, 5, 10, 10 and 10 cm from top to bottom, respectively. For all treatments double folds of water field capacity was used. Water was added to the top of the columns at the rate of 20ml/min. 50 ml from the elute was taken for residue analysis, After eluting all the water quantity, columns were allowed to stand for overnight, after which columns were carefully segmented and (50) grams soil were taken from the 0-5, 5-10, 10-20, 20-30, 30-40 cm layers, extracted and then prepared for chemical analysis.

Glyphosate herbicide

Extraction from water; the 50 ml of water was extracted with dichloromethane /2-propanol (1:1), acidified with H₂SO₄ and evaporated to dryness as described by (Gauch *et al.*, 1989, Feng and Thompson 1990).

Extraction from soils; Glyphosate was triplicate extracted with 0.2 M KOH, 50gm with 150 ml KOH solution triplicate and filtrated through Whatman paper No 1 after then concentrated to 10 ml as described by (Miles and Moye 1988) .

Clean Up: 10 grams of analytical grade anion exchange resin were pre-rinsed with 0.2M NH₄HCO₃ (200ml) and deionized water (100 ml),

and packed into a column. The column was rinsed with deionised water (100 ml). The crud extract solution of each sample was applied to the column, rinsed with deionized water 100 ml, then eluted with 50 ml 0.5 N HCL in test tube (Lundgren 1986, Thompson *et al.*, 1989).

Determination: Glyphosate was oxidized to orthophosphate by Mo (V)-Mo (VI) reagent, which measured calorimetrically as the phosphomolybdate heteropoly blue complex on spectrophotometer at 830 nm (Glass 1981). The procedure used for preparing Mo (V)-Mo (VI) reagent was recently described by (Yoza *et al.*, 1977).

Fusilade herbicides

Soil extraction; 50 grams of air dried soil were put in a bottle and placed in 60°C water bath and shaken for 60 min at 1500 rpm. The alkaline extract and filtered under vacuum. An additional 100 ml of 0.1N Na OH was added to the centrifuge bottle and the process repeated (30 min in water bath). After being rinsed with 20ml of water, the entire alkaline extract was transferred with 50ml of hexane, which was discarded. The aqueous alkaline fraction was then acidified with ca 25 ml of H₂SO₄ to pH <2 and 100ml of saturated NaCl was added. The phenoxalkanoic acids were extracted twice with 50 ml of dichloromethane by shaking for 5min. The dichloromethane extracts were drained through dichloromethane pre washed and dried cotton into 250 ml receiver. Isooctane (0.5 ml) was added and the samples were concentrated on a rotary evaporator at 50 °C to insipient dryness (Clegg 1987). Extraction from water; Samples of 50 ml water were transferred to a separating funnel, acidified to pH 2-3 with 1 M hydrochloric acid and extracted with (50 ml) dichloromethane three times. The pooled extract was dried by filtration over anhydrous sodium sulphate, concentrated to about 5 ml in a rotary evaporator (water-bath at 30 °C), then added to 5 ml of methanol. The final solution was concentrated to about 1 ml in a rotary evaporator. The residue was transferred to a volumetric flask and brought to volume (2-20 ml) with methanol (Negre *et al.*, 1987).

Determination: Photometric determination of fluazifop-butyl as ferrihydroxamate was measured at wavelength 520 nm, as described by (Zhemchuzhin and Kononova 1988a, b). The $t_{1/2}$ values for each parameters were calculated mathematically through trend line equation of each treatments

RESULTS AND DISCUSSION

Effects of tested additive-water type on degradation behavior of glyphosate and fusilade herbicides

Stability of glyphosate when used alone or in mixtures with the additives such as glue, urea, glycerine, dioleate, monoleate, paraffin and mineral oil was changed with type of water. The $t_{1/2}$ in days being 9, 10.25, 7.6, 8.42, 8.5, 6.5, 8 and 6.83 days in distilled water of these treatments, respectively. Whereas in Nile water 9.83, 8.20, 9, 9.63, 9.5, 9.63 and 9.5 days respectively. In case of hard water was 5.5, 5, 4.5, 4.5, 4.83, 4.4 and 4 days respectively (Table 2). This indicates great deterioration of glyphosate residues in hard water with and without additive additives. Comparison between the regression coefficients of the tested additives in water showed that the rate of degradation of glyphosate plus surfactant in the three type of water occurred more rapidly, these results are supported with that obtained by (Zaranyika and Nyandoro 1993, Wang 1999).

Stability of fluazifop-butyl was increased in Nile water when mixed with dioleate, while the contrary was noticed when mixed with monoleate. Stability was lower in distilled water as well as hard water when fusilade at $1/2$ rates was mixed with both surfactants. The $t_{1/2}$ value were being more than >21 in (distilled water). Whereas the fusilade herbicide alone and its mixture with the additive dioleate and monoleate (15.5, 20 and 15), respectively. More ever rape seed oil, mineral oil and paraffin oil more than 21 days (Nile water). Meanwhile in The fusilade herbicide alone and its mixing with additive rape seed oil, mineral oil and paraffin oil dioleate and monoleate showed $t_{1/2}$ by 3.2, 4.83, 4.5, 4.5, 3.42 and 4.2 days (hard water), respectively (Table 2). Comparison between the

regression coefficients of the tested herbicide in water showed that the rate of degradation of fusilade in three type of water occurred more rapidly. This is in accordance with the results of half-life periods for the fusilade. Our results supported with (Negre et al., 1993, Martinez et al., 2000). Reviewing the aforementioned results, it could be concluded that a rapid degradation of glyphosate and fusillade in hard water than Nile water and distilled water may be explained due to the water properties, i.e. EC and pH followed by lower and gradual losses by the lapse of time until the end of experiment (Table 1).

Degradation of herbicide in soil.

The intercept of the regression lines were greater for glyphosate than for glyphosate – glue mixture in the two tested soil types. It is clearly evident to notice that glyphosate plus glue was more persistent than glyphosate when used alone and with its mixture with the additives urea, glycerine, dioleate monoleate, paraffin and mineral oil. The half life of glyphosate alone and its mixtures with the additives glue, urea, glycerin, dioleate, monoleate, paraffin and mineral oil in clay soil in days were being 28,42,28,32,33,28,36 and 42 days respectively. Meanwhile the corresponding

$t_{1/2}$ in sandy soil was 57, 57, 44, 53, 50, 50, 53 and 54 days respectively (Table, 2). Data indicate that fluazifop-butyl when used alone and in its mixture with the additive rape seed oil, mineral oil, paraffin oil, dioleate and monoleate showed the $t_{1/2}$ in days of 9,7, 4.5, 6.5,4 and 5 days, in clays soil, respectively. Meanwhile, the corresponding $t_{1/2}$ in sandy soil reached 13.42, 6.5, 11, 10.5,8 and 7days with the same treatment, respectively. It is clearly evident to notice that the intercepts of the regression lines were greater for fusilade than for fusilade plus paraffin. Comparison between the regression coefficient of the two tested soil showed that the rate of degradation of fusillade plus rape seed oil occurred more rapidly.

The differences in glyphosate and fusillade degradation between sandy and clay soil may be explained due to the soil properties, i. e. total clay content, pH and organic matter (Table, 1). Degradation occurred faster within the 1 st 2 weeks, followed by lower and gradual losses by the lapse of time until the end of experiment by this incubation period, a measurable quantity of the two tested chemicals was detected in all soil treatments.

Table (2). Effect of additives on half life periods (days) of glyphosate (Gly.) and fusillade (Fus.) herbicides in soil and in water under laboratory conditions.

	$T_{1/2}$ days				
	Soil type		Water type		
	Clay	Sand	Distilled	Nile	Hard
Glyphosate	28.00	57.00	9.00	9.20	5.12
Gly. Plus glue	42.00	57.00	10.60	9.20	5.00
Gly. Plus urea	28.00	44.00	7.60	8.20	4.12
Gly. Plus glycerine	32.00	53.00	8.10	9.00	4.12
Gly. Plus dioleate	33.00	50.00	8.12	9.15	4.20
Gly. Plus monoleate	28.00	50.00	6.12	9.12	4.00
Gly.plus paraffin oil	36.00	35.00	8.00	9.15	4.00
Gly. Plus mineral oil	42.00	54.00	6.20	9.12	4.00
Fusilade	9.00	13.10	33.00	15.50	3.40
Fus. plus rap oil	7.00	6.12	42.00	21.00	4.20
Fus. plus mineral oil	4.12	11.00	42.00	21.00	4.12
Fus. plus paraffin oil	6.12	10.10	42.00	21.00	4.12
Fus. plus dioleate	4.00	8.00	25.60	20.00	3.10
Fus. plus monoleate	5.00	7.00	31.00	17.50	3.40

Effect of additives - soil type interactions on downward movement and leaching of tested herbicides.

Data in figure (1) showed that there were differences in the total recovered amount of glyphosate in the two soils, e.i. 86.00 and 87.42 % sandy soil and clay soil, respectively. The results concluded that the majority of glyphosate herbicide amount was located in the top layer of soil column, while less amounts in the leachate were followed by gradual and lower values in the other soil depths of the treated soil. Glyphosate was not found in the leachate from the two soils. There were significant differences between additives in the determined amount of herbicide in soil column. Our results are in agreement with (de Jonge *et al.*, 2000, Fomsgaard *et al.*, 2003, Kjær *et al.*, 2005). Data indicate that the majority of the added fusillade herbicide was located in the top layer of soil column (0-5 cm), followed by gradual movement but in lower values in the successive depths figure (1). No residues were detected in the leachate from both soil types. There were significant differences between additives role in diminishing the movement of herbicide in soil column. The differences in herbicide movement between sandy and clay soil may be explained due to the differences of soil properties, i. e. total clay content (50.1, and 10%), pH 7.57 and 7.12) in clay and sandy soil, respectively. The majority of fusilade amount was located in the top layer of soil column while fewer amounts were detected in the leachate followed by gradual lower values in the other depth. Leaching of fusilade was increased in sandy soil. Stability of fusilade was increased in sandy soil. Our results are in agreement with (Rick *et al.*, 1987, Gennari *et al.*, 1991). Glyphosate and fusilade downward movements in clay and sandy soils were very slowly due to the great adsorption of the herbicide in the top soil layer. Whereas glyphosate and fusilade could be transferred from the sub-surface to the following layer with some additives (surfactants) to the following layer. The majority of glyphosate amount was located in the top layer of soil column and gradual lower values in the other depths of soil. Glyphosate not appeared in water leachate from the two soils.

There were significant differences between additives in determining the movement of herbicide alone.

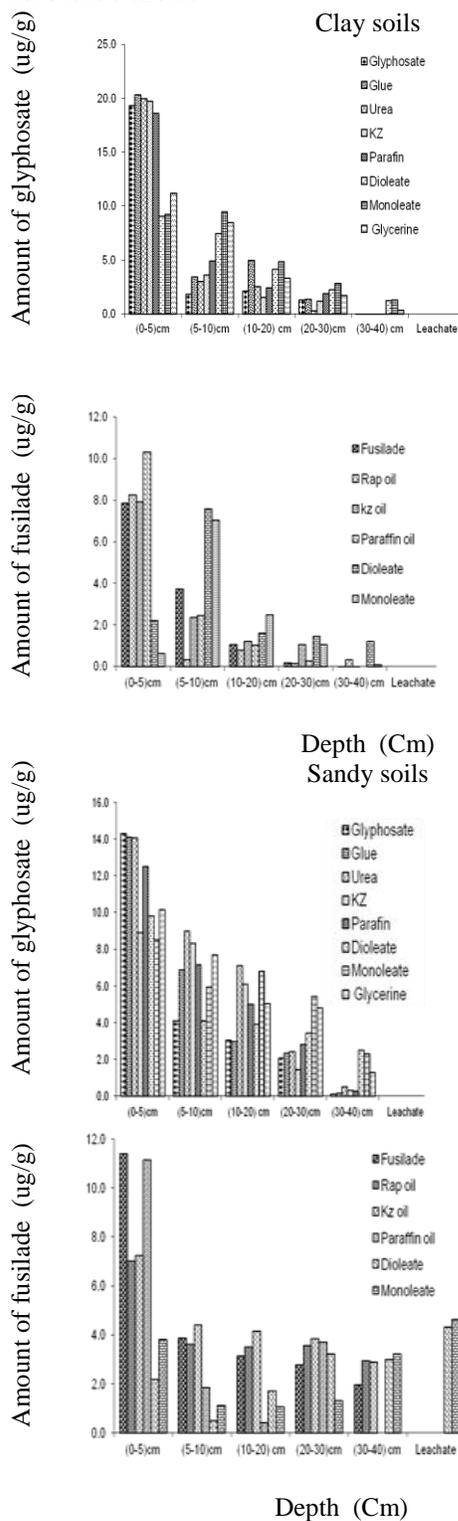


Fig (1). Effect of tested additives on herbicides downward movement in soils.

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تأثير بعض المواد الإضافية على التحلل المائي والثبات و الحركة الراسية لمبيدي الأعشاب الجليفوسات و الفلزي فوب بيوتيل

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المستخلص: استهدفت الدراسة أولاً تقييم تأثير المواد الإضافية ونوع الماء على التحلل المائي لمبيدي الأعشاب الجليفوسات والفيوزيليد، حيث أوضحت النتائج أن الجليفوسات يتحلل مائياً بدرجة كبيرة في الماء العسر عن ماء النيل ثم الماء المقطر، بإضافة الغراء إلى الجليفوسات زاد من ثباته مقارنة باستخدام المبيد منفرداً. بينما خفضت المواد الإضافية الأخرى مثل اليوريا، الجلوسرين، الذي أوليت، المونواوليت، زيت البارافين، الزيت المعدني من ثبات الجليفوسات في كل أنواع الماء المختبر على الترتيب. أوضحت النتائج أن الفلزي فوب بيوتيل ثابت نسبياً في الماء المقطر خلال جميع فترات الدراسة عن ماء النيل في حين أظهر انهياراً سريعاً في الماء العسر، وعموماً كان الفلزي فوب بيوتيل منفرداً أكثر ثباتاً عن خلطه بالمواد الإضافية مثل زيت الراب، الزيت المعدني، زيت البارافين، الداى أوليت ثم المونواوليت على الترتيب. دراسة تأثير نوع التربة والمواد الإضافية على درجة الثبات والحركة الراسية إلى الأسفل في عمود التربة للمبيدات المستهدفة حيث أظهر الجليفوسات والفلزي فوب بيوتيل انهياراً سريعاً في التربة الرملية عن الرملية، كما أدت المواد الإضافية إلى زيادة ثبات الجليفوسات في التربة الطينية وانخفاض ثباته في التربة الرملية، على العكس من ذلك أدت المواد الإضافية إلى انخفاض فترة نصف عمر الفلزي فوب بيوتيل في التربة الطينية والرملية. كما أظهرت النتائج تأثير المواد الإضافية على الحركة الراسية للمبيدين في عمود التربة في حين أن أكبر كمية من مبيدي الجليفوسات والفلزي فوب بيوتيل كانت متواجدة في الطبقة السطحية من عمود التربة الطينية والرملية عند استخدام المبيدين منفردين ومع بعض المواد الإضافية ولكن عند خلط المبيدات بالمواد النشطة سطحياً انتقلت أجزاء من المبيدات إلى الطبقات المتتالية وبخاصة في التربة الرملية عن الطينية في حين وصلت كمية بسيطة من مبيد الفلزي فوب بيوتيل إلى راسح العمود أمكن الكشف عنها.

الكلمات المفتاحية: غليفوسات، فوسيلاد، التحلل، المقاومة، الحركة النزولية، الرش والمواد المضافة.

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