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Leaching of pyrazosulfuron-ethyl in a sandy loam soil under natural rains in field lysimeters

Shobha Sondhia**Abstract**

A study was conducted to determine pyrazosulfuron-ethyl mobility in a sandy loam soil and distribution of its residues at various depths in field lysimeters. Pyrazosulfuron-ethyl was applied at a recommended dose 25 g/ha after onset of rains. Soil samples were taken at 0-225 cm depths after rains on various days. Approximately 1234 mm of rain caused leaching of pyrazosulfuron-ethyl through the soil and resulted in 0.163 mg to 1.64 µg/L of residues in the leachates. Pyrazosulfuron-ethyl residues were detected in significant amount from the soil collected at various depths up to 10 days. A positive correlation was found between amount of rain received at different days and a negative significant correlation of residues with the depth and EC. This study demonstrated higher mobility of pyrazosulfuron-ethyl in sandy loam soil. Results obtained are of interest to improve pyrazosulfuron-ethyl management in soil, and to predict risk of ground water contamination.

Keywords: Pyrazosulfuron-ethyl, Lysimeter, Leaching, Residues, Sandy loam soil

Introduction

Herbicide use has increased tremendously in last two decades in India. Repeated applications of herbicides for weeds control during crop growth period especially in rainy season enhanced risk of leaching of these chemicals to the lower soil profile and results in several environmental problems and harmful effects on non-targeted organisms (Ritter *et al.* 1996; Sondhia, 2007, 2008) [15, 17, 18]. The potential for contamination of water bodies and persistence in soil and waters due to pesticides use is high in paddy cultivated areas (Sondhia and Yaduraju 2005; Müller *et al.* 2007; Sondhia *et al.* 2013) [21, 13, 20, 23]. In general, herbicide leaching and risk of groundwater contamination is greatly affected by formulation, water solubility, mode of pesticide application, interaction of the chemical with soil, and irrigation and/or rain to the field (Jury 1986; Sondhia 2008, 2013) [10, 18, 20, 23]. However, timing, intensity, and amount of precipitation play a significant role in the eventual behavior of chemical in the soil environment (Sondhia 2008; Cessna *et al.* 2009) [18, 6].

Pyrazosulfuron ethyl is a sulfonylurea herbicide and widely used in rice crop to control annual and perennial weeds (Table 1). It inhibits an important enzyme, acetolactate synthase (ALS) which catalyzes biosynthesis of branched amino acids, valine, leucine, and isoleucine in the plants. Legumes and pastures are highly sensitive to the residues of sulfonylurea herbicides in soils (Brown 1990; Sondhia, 2008) [4, 18]. Several studies demonstrated frequent detection of residues of sulfonylurea herbicides in surface and groundwater that is mainly due to runoff and leaching after their application in the agricultural lands (Sondhia 2008, 2013) [18, 20, 23].

Higher adsorption reduces the amount of chemical available to plants and thereby slows the process of leaching. The higher percentage of organic matter and clay composition in the soil increases extent of adsorption. As sulfonylureas herbicides are acidic in nature and are adsorbed in moderate amounts on organic matter and in relatively low amounts on clay soils. Adsorption of sulfosulfuron in soil increased with decreasing soil pH (Sondhia *et al.* 2013) [20, 23]. Sulfonylurea herbicides degraded more slowly in alkaline, cold, dry and low-organic matter soils. Mobility of sulfonylureas increases with increasing pH and decreasing soil organic matter (Kearney and Kaufinan 1988) [11]. Sulfonylurea herbicides do not show any remarkable volatility and photodegradability. In certain pedo-climatic conditions very long persistence of some sulfonylureas caused risk of leaching and/or damage to successive crops (Sondhia *et al.* 2013) [20, 23]. Assessment of leaching of some other sulfonylurea, tribenuron methyl, thifensulfuron methyl, and rimsulfuron indicated that they were leached into groundwater (Cessna *et al.* 2009) [6].

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Hitherto, information of leaching potential of pyrazosulfuron-ethyl in the soil environment is poorly documented. Zheng *et al.* (2008) [27] studied chemical hydrolysis kinetics and pathway of pyrazosulfuron-ethyl in water. Several laboratory studies were reported to evaluate mobility of some pesticides in soils (Sharma and Singh 2001; Sondhia 2007) [16, 17]. As most of the herbicides formulation are soluble in water and thus enhanced risk of ground water contamination under saturated moisture regime. Therefore this study was undertaken to determine (i) impact of varying rain on pyrazosulfuron-ethyl leaching and effect of leaching on soil physico-chemical properties.

Materials and Methods

Study was undertaken in field lysimeters of 1 and 2 meters depths that receiving natural rains. Lysimeters contained sandy loam soil having sand 67.32 to 66.31%, silt 9.2 to 10.00%, clay 22.68 to 21.88% and organic carbon 0.369 to 0.280% at 0-150 cm depths. Pyrazosulfuron-ethyl (EC 10%) was applied to individual lysimeter with a hand sprayer at field rates equivalent to 25 g/ha. Experiment was replicated thrice. After each rain, leachates were collected in individual plastic canes. Soil samples at 3, 5, 10, 20, 30, 60 and 90 days were taken from 0-25, 25-50, 50-75 cm and 0-25, 25-50, 50-75, 75-100, 100-125, 125-150 cm depths from lysimeters.

Pyrazosulfuron-ethyl residues were extracted and analysed as described by Sondhia *et al.* (2013) [20, 23] with a limit of detection (LOD) and limit of quantification (LOQ) of 0.001 and 0.01 µg/mL, respectively. Pyrazosulfuron-ethyl reference analytical standard (Purity 99%) was procured from AccuStandard Inc, USA and used in the study. All other chemicals and solvents used for extraction and analysis were of analytical grade.

A total of 1233.9 mm of rain on the experimental lysimeter between the date of pyrazosulfuron-ethyl application and termination of experiment was recorded. Maximum and minimum temperature was found to be 34.5 to 22.6 °C and humidity varied between 96 and 51%, respectively. Data was analysed using variance of analysis technique (ANOVA). Significant interaction among application rate of pyrazosulfuron-ethyl, rainfall events and soil physico-chemical properties at various depths were calculated using two ways ANOVA table. Standard deviation was calculated with Excel.

Results and discussion

Pyrazosulfuron-ethyl residues in soil

Random distribution of pyrazosulfuron-ethyl residues was found in soil at various depths following its application at g/ha rates. After 1 day of experiment, 63.4 mm of precipitation was received in experimental field and mean of 0.039 and 0.058 µg/g pyrazosulfuron-ethyl residues were found in upper depth (0-25 cm) of 1 and 2 m lysimeter, respectively and 0.027 and 0.053 µg/g pyrazosulfuron-ethyl residues were found into 0-25 and 25-50 cm depths in 1 and 2 m lysimeter, respectively. After 3 days, 0.0223 and 0.0210 µg/g and 0.0212, and 0.0127 µg/g pyrazosulfuron-ethyl were found in 0-25, and 25-50 cm depths in 1,2m lysimeters, respectively after 3 days that received 5.7 mm rainfall (Figure 1 and 2).

After application on the soil surface, herbicides undergo several processes including degradation and transport (Vink and Robert 1992; Sondhia 2012; Sondhia *et al.* 2013) [24, 19, 20, 23]. Herbicides with high persistence and weak sorption rate may be readily leached through the soil and are more likely to

contaminate groundwater. After ten days, about 412 mm precipitation resulted in leaching and degradation of pyrazosulfuron-ethyl by cleavage of Sulfonamide Bridge by hydrolysis. Pyrazosulfuron-ethyl residues were not found in lower depths (50-150 cm) in soil in initial days of sampling. After 10 days of experiment, about 412 mm of precipitation resulted in movement of 0.067, 0.12 µg/g of pyrazosulfuron-ethyl residues at 25-50 and 50-75 cm depths, in 1 m lysimeter, respectively. However in 2 m depths lysimeter 0.029 µg/g of pyrazosulfuron-ethyl residues were moved down to 125-150 cm depths.

After 30 days, 119.5 mm of precipitation resulted in 0.0059 to 0.0073 µg/g pyrazosulfuron-ethyl residues at 0-75 cm depths in 1 m lysimeter. However, 0.0046 to 0.0088 µg/g of pyrazosulfuron-ethyl residues were leached to 25 to 125 cm depths. Approximately 367.4 mm of precipitation at 60 days resulted in leaching of pyrazosulfuron-ethyl and an amount of 0.001 to 0.0022 µg/g residues were found in upper to 0 to 50 cm depths after 60 days. After 90 days pyrazosulfuron-ethyl residues were not detected in any soil depth.

Statistically significant correlation ($P < 0.05$) and interaction were found at rains at various days and application of pyrazosulfuron-ethyl. Sampling depth also showed significant relation in all phases of the sampling. Higher concentrations of pyrazosulfuron-ethyl were found in surface soil followed by successively lower amounts at the other depths. In general, results suggested that there was a positive correlation between application rate of pyrazosulfuron-ethyl and amount of rainfall.

Pyrazosulfuron-ethyl application and subsequent precipitation had a significant impact on mobility. Weber and Miller (1989) [26] reported that leaching of herbicides in soils was governed largely by the amount of rainfall and soil type. Similar results were observed in this study. Experimental soil was having more than 67% of sand and low organic matter content at various depths (0.369 to 0.280%) which might have provided more opportunity for pyrazosulfuron-ethyl leaching. Blasioli *et al.* (2008) [1] reported that organic matter have no effects on pyrazosulfuron-ethyl leaching, while they observed that presence of potassium ions reduced leaching of pyrazosulfuron-ethyl due to increased soil adsorption of inner sphere complex, with a potassium ions formed at the carboxyl group by cation bridge. Similar finding was also reported by Jhala *et al.* (2012) [9]. Sharma and Singh, (2001) [16] demonstrated positive correlation between precipitation and herbicide leaching. They showed that leaching of norflurazon increased from 19.6 to 105.4 cm with increasing amount of rainfall from 6.25 to 12.5 cm/ha. By comparing results of this study with previous leaching studies of herbicides commonly used in rice (Sharma and Singh 2001; Nelson and Penner 2007) [16, 14] pyrazosulfuron-ethyl can be considered as a moderate leaching herbicide. Amounts of leaching and runoff are largely dependent on total rainfall specially the first few days after an application of herbicide. Bovey *et al.* (1978) [2] reported that low soil adsorption capacity, high rates of application and high rainfall increased total runoff and contamination of local waterways.

Effect of pyrazosulfuron-ethyl on soil pH and EC

To evaluate effect of herbicide mobility on soil chemical properties, two important parameters viz. pH and EC were evaluated and interaction between dose verses herbicide movement at various depth at varying precipitation was observed. Result showed that pyrazosulfuron-ethyl application on soil lysimeter increased pH of soil. Change in

pH of soil at 0-90 d was found from 7.1 to 8.4 (Table 3). Precipitation did not caused significant difference on soil pH at various depths due to downward movement of pyrazosulfuron-ethyl through soil profile of lysimeter after receiving pyrazosulfuron-ethyl application at probability $P < 0.05$ level (Table 4). Continuous sporadic changes of soil pH were observed at soil depths of 0-150 cm depths.

EC in sampled soil at various depths ranged between 17.1 to 196 $\mu\text{S}/\text{cm}$ with the greatest amount and variation in the upper soil (0-25 cm). There was change in EC of soil at various depths and it was found higher where considerable amount of pyrazosulfuron-ethyl was moved down. These values were within the range typical for vertisol (Virman *et al.* 1982) [25]. As EC of soil is closely related to organic matter and clay content, it might be expected that a high EC would give rise to increased adsorption. Precipitation caused significant difference ($P = 0.05$ level) on soil EC at various depths due to leaching of pyrazosulfuron-ethyl after receiving of pyrazosulfuron-ethyl application. Prediction of movement and fate of herbicides in soils represent an important strategy in limiting their environmental impact. Organic matter influences mobility of herbicides by complex interactions that can facilitate or reduce movement of chemicals along the soil profile (Braschi *et al.* 2011). Knowledge of soil phase characteristics and mechanisms involved in herbicide transformation can help to understand the fate of herbicides in soil. Braschi *et al.* (2011) found that leaching was dependent on herbicide solubility, precipitation and degree of adsorption encountered in soils. Greater downward mobility was found with more soluble herbicides and in soils with low herbicide adsorption.

Pyrazosulfuron-ethyl content in leachates

Amount of chemicals received from leachates reveal leaching potential and risk of ground water contamination. Thus after each rain, leachates were collected and analyzed for pyrazosulfuron-ethyl content. The concentrations of the pyrazosulfuron-ethyl in the leachates (applied at 25 g/ha) ranged from 0.00169 to 0.00055 mg/L. These concentrations were much lower than those detected in the soil at various depths when herbicide was applied to the surface of lysimeters at rates ranging from 25 to 50 g/ha. Carabias

Martinez *et al.* (2000) monitored concentration of fifteen herbicides owing to their frequency and amounts used toxicity and persistence in river basins in the provinces of Zamora and Salamanca (Spain). After six months presence of six out of fifteen herbicides monitored was detected at levels ranging from the detection limit to 1.2 $\mu\text{g}/\text{L}$. Presence of these herbicides was related to agricultural activities as well as the kind of crop and its treatment period. Higher mobility of sulfosulfuron and degradation products might be due to its lower adsorption capacity and higher solubility (Sondhia *et al.* 2013; Loganayagi and Ramesh 2014) [20, 23, 12].

The presence of pyrazosulfuron-ethyl in the initial leachates samples collected from the lysimeter provide direct evidence for the preferential flow of this herbicide with the infiltrating water (Sondhia 2008) [18]. Under field situations, preferential flow occurs via preferential flow paths, such as earthworm burrows, channels created by live drying or and cracks or fissures in the soil. Mobility of pyrazosulfuron residues in the increasing soil depths are positively correlated with the pH and rain (Table 4).

Because rainfall commence immediately after herbicide application, the elevated concentrations in the initial water to reach the leachates most likely occurred because the surface deposits of this herbicide was not strongly sorbed to the soil and readily moved through larger pores (preferential routes) with the infiltrating rain water (Sondhia 2007, 2008;) [17, 18]. Then, as previously reported by Kladvikova *et al.* (1991) and Elliott *et al.* (2000), concentrations decreased rapidly as flow through the tile drains continued.

Leaching of the herbicide was most pronounced for the 1m lysimeter where the maximum concentration of pyrazosulfuron-ethyl was detected from leachates on 17 and 26 July. These concentrations were the highest measured in the study and were 0.66 $\mu\text{g}/\text{L}$ for 1m depth lysimeter and 0.2 $\mu\text{g}/\text{L}$ for 2m depth lysimeter. Despite having almost similar drain flow of approximately 179.53 and 177.9 mm in the 1 and 2m lysimeter, peak concentrations in the leachates from the 2 m lysimeter were smaller and more diffuse and was 0.56 $\mu\text{g}/\text{L}$ in comparison to 1.69 $\mu\text{g}/\text{L}$ occurring for pyrazosulfuron-ethyl. Rainfall of 139 mm in first and second week of September resulted in 0.088 and 0.078 $\mu\text{g}/\text{L}$ pyrazosulfuron-ethyl leaching from 1 to 2 m lysimeter.

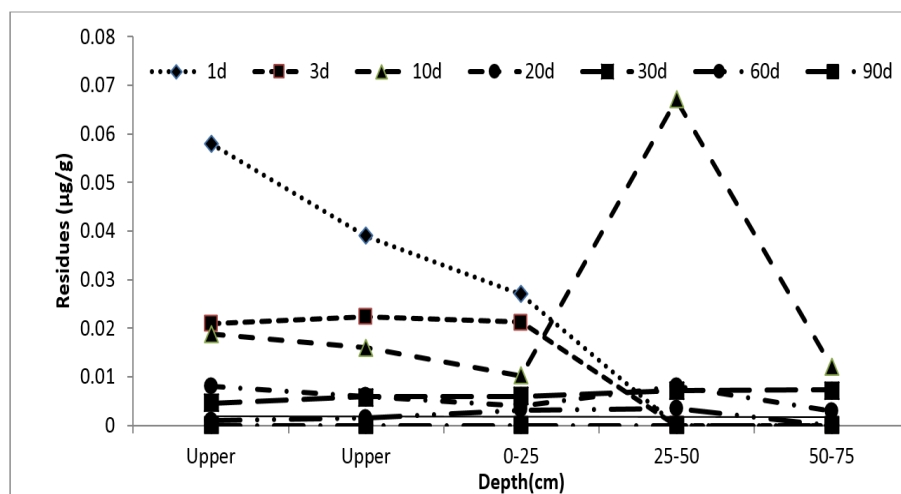


Fig 1: Residues of pyrazosulfuron in soil at various depths in lysimeter of 1m depths

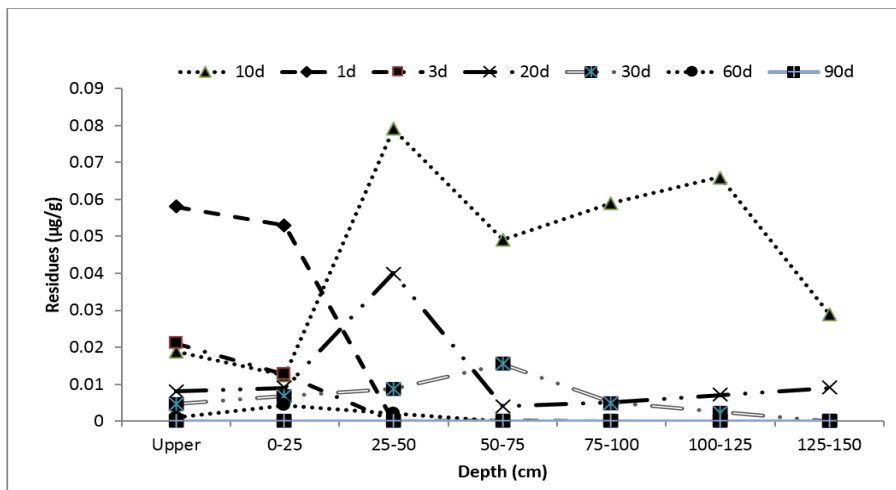


Fig 2: Residues of pyrazosulfuron in soil at various depths in lysimeter of 2 m depths

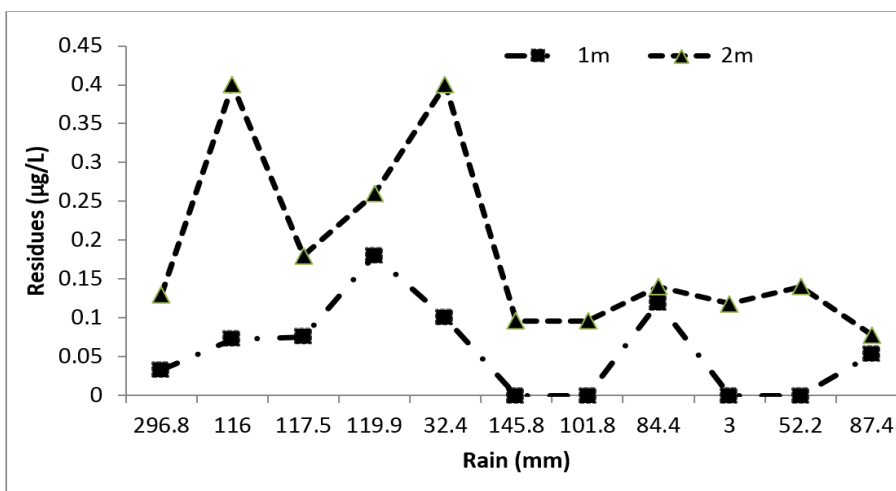


Fig 3: Effect of rainfall on residues in the leachates due to pyrazosulfuron-ethyl application on the surface of lysimeters

Table 1: Some important physico-chemical properties of pyrazosulfuron-ethyl

Chemical structure	IUPAC name	ethyl-5-[(4, 6-dimethoxypyrimidin-2-ylcarbamoyl)sulfamoyl]-1-methyl pyrazole-4-carboxylate]
	Molecular formula	C ₁₄ H ₁₈ N ₆ O ₇ S
	Molecular weight	414.4 g/mol
	Formulation	WP10%
	Solubility in water	14.5 mg/L
	Vapor pressure	4.2x10 ⁻⁵ Pa at 25 °C
	Henry's constant	1.72 X 10 ⁻⁷ Pa m ³ /mol at 25°C
	Partition coefficient Log P _{ow}	1.3

Table 2: Effect of pyrazosulfuron application on pH and EC of soil at various depths in 1m depths

Depths (cm)	pH and EC (µS/cm) of pyrazosulfuron at various depths													
	Precipitation (mm)													
	63.4		5.7		412		117.5		119.5		367.4		150.6	
	1d		3d		10d		20d		30d		60d		90d	
	pH	EC	pH	EC	pH	EC	pH	EC	pH	EC	pH	EC	pH	EC
Upper	7.8	28.5	7.5	29.6	7.9	109.2	8.2	59.2	8.3	65.2	8.2	39.8	8.29	26.6
0-25	7.2	21.4	7.4	24.1	8.0	113.3	8.1	105.6	8.4	65.9	8.0	65.6	8.31	60.7
25-50	7.4	29.6	7.2	27.9	8.1	112.5	8.2	93.6	8.2	87.7	8.1	72.5	8.21	34.1
50-75	7.1	24.1	7.52	29.0	7.9	89.78	7.9	88.7	8.5	104.6	8.0	58.4	8.14	35.4

Table 3: Effect of pyrazosulfuron application on pH and EC of soil at various depths in 2m depths

Depths (cm)	pH and EC($\mu\text{S}/\text{cm}$) of pyrazosulfuron at various depths													
	Precipitation (mm)													
	63.4		5.7		412		117.5		119.5		367.4		150.6	
	1d	3d	10d	20d	30d	60d	90d							
Upper	7.5	36.8	7.6	79.2	7.9	90.2	7.6	84.2	7.7	98.8	7.8	41.4	8.0	23.3
0-25	7.3	24.6	7.5	72.1	7.8	87.5	7.6	196.8	8.0	101.1	7.6	65.6	8.5	27.8
25-50	7.4	29.1	7.6	54.5	7.4	90.7	7.5	112.9	7.7	110.3	7.8	82.7	8.8	34.6
50-75	7.2	26.9	7.61	66.7	8.1	91.2	7.8	107.4	7.8	84.4	8.0	85.8	8.4	26.8
75-100	7.3	29.2	7.5	94.5	8.0	110.8	7.9	102.6	8.0	112.7	8.2	101.2	8.1	30.2
100-125	7.2	36.5	7.6	56.7	8.0	88.9	7.8	124.3	8.1	87.6	8.0	75.6	7.4	30.4
125-150	7.1	29.8	7.8	82.5	8.3	127.2	7.8	84.5	7.9	108.8	8.0	62.7	8.24	29.7

Table 4: Correlation matrix between residues and related parameters

	Variable	Correlation matrix				
		(1)	(2)	(3)	(4)	(5)
(1)	Depth	1				
(2)	pH	(0.85)	1.00			
(3)	EC	0.19	(0.61)	1.00		
(4)	Rain	0.35	0.42	(0.62)	1.00	
(5)	Days	(0.94)	(0.65)	0.10	0.26	1.00
	Mean	75.14	7.93	0.80	76.59	0.57
	SD	53.78	0.39	1.42	53.44	33.09

Note: Correlation greater than 0.50 is given in parenthesis

Conclusion

Movement of pyrazosulfuron-ethyl was determined largely by the amount of water moving through the soil with the soil characteristics altering this movement. Since ionic solutions affect many chemical reactions, they might also be expected to affect herbicide movement. Because of the potential need for such information, this study was initiated to observe any trends in the mobility and distribution of pyrazosulfuron-ethyl residues as affected by pyrazosulfuron-ethyl application and precipitation. Higher rains in initial days of experiments caused mobility of pyrazosulfuron-ethyl and significant amount of residues in soil after 3 to 10 days. Therefore, it can be concluded that at higher precipitation rates and saturated soil moisture conditions, pyrazosulfuron-ethyl can move to subsurface soil. Nonetheless, under the conditions of this field study, there was evidence for leaching of pyrazosulfuron-ethyl to the lower depths with the infiltrating rain water. This herbicide was present in the leachates in low (micrograms per liter) concentration. The leaching of pyrazosulfuron-ethyl below the root zone in each lysimeter of the study site was significant in terms of ground water contaminations under saturated soil moisture condition.

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