Original Research Article

Organic amendments addition on changing the sorption, vertical movement and leaching behavior of herbicides in soil

Abstract

Herbicides are applied indiscriminately to achieve maximum weed control in the agriculture production. This might contaminate the food chain through the bio-augmentation of residue concentration in soil. One of the management suggested to overcome the herbicide contamination is the application of various organic amendments. Hence the laboratory column leaching experiment was conducted in TNAU, Coimbatore to investigate the leaching behavior of herbicides viz., pendimethalin, atrazine and imazethapyr in soil as influenced by various organic amendments applied @ 5t/ha viz., FYM, Vermicompost, composted coir pith, maize crop residue and biochar. Results showed that leaching and movement behavior of the studied herbicides in soil was modified by the applied organic amendments particularly the biochar and maize crop residue. Biochar treated soil retained 29.76, 40.20, and 38.07 percent of the applied pendimethalin, atrazine and imazethapyr in top surface 0-15 cm depth soil and hence reduced its movement to lower depths and to leachates. All the applied organic amendments reduced the leaching by increasing their sorption when compared to control and was highly significant with pendimethalin and atrazine and non significant with imazethapyr. This showed the effect of herbicide nature on its movement beside the organic amendments type. It is concluded that the application of biochar @ 5t/ha as organic amendment is efficient in decreasing the herbicides movement viz., pendimethalin, atrazine and imazethapyr and modifying their sorption and leaching potential to groundwater and was followed by the maize crop residue.

Key Words : Herbicides, biochar, maize crop residue, movement, leaching

1. INTRODUCTION

In the present agro-ecosystem, herbicides are considered to be an efficient and economic tool to control weeds. The increased input cost and non-availability of manual labour for timely weeding warrant the herbicides application as a significant option (Janaki *et al.*, 2019). The behaviour of herbicides in the environment is complex and controlled by inter-dependent physical, chemical and biological processes. The most important processes determining the fate of herbicides in environment are photo degradation, hydrolysis, microbial degradation, volatilization, and sorption and transport process. The movement of herbicides from soil into water, air and the food chain is directly controlled by these processes. The wide use of herbicides with a long persistence in the soil has causes residue to accumulate 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).

Atrazine, pendimethalin and imazethapyr are the commonly used herbicides applied as pre-emergence or early post emergence for weed management in field crops of India. Atrazine is moderate to highly mobile in soil and does not adsorb strongly to soil particles and has a high potential for groundwater contamination (Wauchope *et al.*, 1992). While the dinitroaniline herbicides (pendimethalin) are highly volatile and undergo little leaching in soil (Anderson, 1996), they have persisted up to 50 weeks (Pritchard and Stobbe, 1980) and cause toxicity to the succeeding crops (Zimdalh *et al.*, 1984; Smith *et al.*, 1995).

Due to continuous and frequent use, the residue of these herbicides is magnified and might contaminate the soil, water and food chain due to bio-accumulation and increased the persistence (Farmer and Aochi, 1987) in soil environment. Apart from this, the carryover of herbicide residue to the follow-up crops is also an issue when the crop received these herbicides failed due to other management or climatic issues and that necessitates the cultivation of next crop immediately. For managing the herbicides residue issues in soil environment, many techniques and practices are in adoption (Janaki et al., 2015) are irrigation and ploughing, exposing soil to sunlight, application of excess quantities of organic manures and amendments, immobilizing agents etc. One of the immediate and safe practices is the incorporation of organic amendments as they not only immobilize the herbicides residue temporarily but also enhance the biodegradation of residues in soil environment into metabolites and non toxic products. Hence the present study was undertaken to evaluate the efficiency of various organic amendments on modifying the movement and leaching behavior of atrazine, pendimethalin and imazethapyr in soil.

2. MATERIALS AND METHODS

The surface (0-15 cm) soil samples were collected from pesticide free zone in the eastern block farm, TNAU, Coimbatore, air dried, processed and used for movement and leaching and sorption study after passing through 2 mm sieve. The soil was calcareous sandy clay loam in texture having pH 8.35, EC 1.1 dS/m, OC 0.46 %, available N,P,K of 260, 19.8 and 593 kg/ha. The study was conducted in lab under controlled condition by imposing the treatments following the standard OECD guidelines for pesticides dynamics in soil.

The leaching columns and sorption experiments were conducted as described by Janaki et al. (2013). For column movement and leaching study, each herbicide was applied at two rates viz., pendimethalin @ 750 and 1500 g/ha, atrazine @ 500 and 1000 g/ha, and imazethapyr @ 75 and 150 g/ha to the column of soil incorporated with different organic amendments viz., FYM 5 t/ha, vermicompost 5 t/ha, composted coir pith 5 t/ha, maize crop residue 5 t/ha and biochar 5 t/ha along with control (no amendment). Imposed treatments combination was replicated thrice. Each soil column was leached with 300 ml of water at a regular interval of two days. After leaching soil columns for 14 days, they were dissected into different sections as 0-15 and 15-30 cm from top. Soil sample from each section and leachate from each soil column was collected and pooled on day 14 for herbicides residue analysis. Sorption study was also conducted using the above organic amendments treated soils and each herbicides were applied at different concentrations, (0, 0.5, 0.75, 1.5, 2.0 and 2.5 mg/kg soil for pendimethalin, 0.25, 0.5, 1.0, 2.0 and 2.5 mg/kg soil for atrazine, 0.05, 0.1, 0.2, 0.5 and 1.0 mg/kg soil for pendimethalin,) and the experiment was duplicated. The soil: solution ratio used for this study was 1:5 and were shaken for 24hrs on an end over end shaker to attain equilibrium. After equilibration, the soil water suspension was centrifuged at 2000 rpm for 15 minutes and the herbicide concentration was measured in the supernatant (Janaki et al., 2013). All the samples from leaching and sorption experiments were analyzed for the herbicides residue in HPLC (Babu et al. 2015; Janaki et al., 2012; Janaki et al., 2015a).

Characterization of organic amendments

The organic amendments used for the study was obtained from the central farm, TNAU, Coimbatore except biochar which was purchased from AEC&RI, TNAU, Coimbatore. The biochar used was prepared by pyrolysis of the coconut shells. Maize crop residue was powdered using Willey mill and then used in the experiments. All the amendments were analyzed for their physico-chemical properties and the results are presented in Table 1.

Table 1.Physico-chemical properties of the organic amendments used in the study

S.No	Organic amendments used	FYM	Vermi-compost	Composted Coir pith	Maize crop residue	Bio-char
1.	pH (1: 5 dilution)	7.21	5.98	6.21	5.81	7.98
2.	EC (dS/m) (1: 5 dilution)	0.52	0.37	1.85	1.56	0.41
3.	Organic carbon (%)	10.2	13.8	32.0	35.8	31.8
4.	Total N (%)	0.65	0.52	0.34	0.81	0.07
5.	Total P (%)	0.21	0.24	0.21	0.13	0.21
6.	Total K (%)	0.47	0.92	0.87	1.54	0.98

Data analysis

The value of K_d were determined either from the slope of the linear plots of sorbed Vs aqueous herbicide concentrations or from the single solution concentration of the herbicide was calculated as described by Janaki et al. (2013). The amount of herbicide sorption per unit soil organic carbon (SOC), K_{oc}, was also determined: $K_{oc}=(K_d/SOC) \times 100$ which is a more intrinsic property of the compound particularly for non-ionized chemicals. Adsorption isotherm parameters were calculated for each sorbent using the normalized Freundlich equation:

$$\text{Log } x/m = l/n \log C + \log K_f$$

Where x/m = micromoles of herbicide adsorbed per kilogram of material, C = micromoles of herbicide per liter of solution after equilibration, K_f is the Freundlich coefficient (ml/g) and 1/n is a dimensionless parameter commonly less than unity. The K_f values were normalized for organic carbon content.

Pearson correlations were calculated for sorption coefficients (K_d) vs. all soil textual and chemical properties with a significance level of $P< 0.05$.

3. RESULTS AND DISCUSSION

Influence of organic amendments on adsorption of herbicides

The movement of herbicides viz., pendimethalin, atrazine and imazethpyr were analyzed at two soil depths viz., 0-15 and 15-30 cm as well as in leachates at the end of study (Table 2 to 4). It was found that the herbicides residue concentration has decreased with increased soil depth under both the levels of application and was also detected in leachates. Increased rate of application enriched the soil with herbicides molecule besides transporting considerable quantity to lower depth also. At the end of experiment, irrespective of the organic amendments application, 28.9, 3.5 and 2.1 percent of the applied pendimethalin, 18.7, 13.8 and 2.3 percent atrazine and 19.5, 17.2 and 14.4 percent of the applied imazethapyr retained in soil at 0-15 cm, 15-30 cm and leachate, respectively.

Table 2. Effect of different organic amendments and pendimethalin rates on its retention in soil (mg/kg) at different depths and in leachate (mg/L)

Soil depth (cm) / Amendments	0-15 cm		15-30 cm		Leachate	
	750 g/ha	1500 g/ha	750 g/ha	1500 g/ha	750 g/ha	1500 g/ha
FYM	0.206	0.416	0.023	0.036	0.020	0.033
Vermicompost	0.191	0.406	0.030	0.048	0.021	0.037
Composted Coir pith	0.201	0.424	0.031	0.054	0.025	0.046
Maize crop residue	0.224	0.460	0.024	0.047	BDL	BDL
Biochar	0.284	0.571	0.025	0.061	BDL	BDL
Control	0.159	0.391	0.032	0.063	0.033	0.057

The pendimethalin retention in top soil (0-15 cm depth) was high (Fig 1A) and is ranged from 25.51-38.07 % at surface when compared to 2.37-4.27 % at subsurface (15-30 cm depth) irrespective of organic amendments and rate applied. The pendimethalin residue was not detected in leachates of soil applied with biochar and maize crop residue amendments (Fig 2). This showed that these amendments sorbed the molecules tightly and hence not subjected to much leaching whereas other amendments treated soil leachates recorded residue level of 2.20 - 4.40 % irrespective of the rate applied.

The atrazine retention in top soil (0-15 cm depth) was high (Fig 1B) and is ranged from 6.0-40.2 % at surface when compared to 1.38-19.2 % at subsurface (15-30 cm depth) irrespective of organic amendments and rate applied. Control treatment recorded higher residue in subsurface (0.074 and 0.119 mg/kg) than the surface soil (0.048 and 0.063 mg/kg) at both the rates of application. The atrazine residue was detected in leachates of soil in the range of 0.74 – 3.6 % across different amendments and rates of application (Fig 2). The quantity measured in biochar was low and it was 0.74 and 0.80 % respectively at 500 and 1000 g/ha rate of application. Control showed higher quantity in leachates with the values of 3.40 and 2.45 percent respectively at 500 and 1000 g /ha rate of application.

Table 3. Effect of different organic amendments and atrazine rates on its retention in soil (mg/kg) at different depths and in leachate (mg/L)

Soil depth (cm) / Amendments	0-15 cm		15-30 cm		Leachate	
	500 g/ha	1000 g/ha	500 g/ha	1000 g/ha	500 g/ha	1000 g/ha
FYM	0.045	0.079	0.086	0.171	0.016	0.023
Vermicompost	0.047	0.104	0.069	0.193	0.016	0.023
Composted Coir pith	0.108	0.236	0.054	0.129	0.018	0.022
Maize crop residue	0.159	0.295	0.062	0.151	0.007	0.012
Biochar	0.186	0.402	0.065	0.141	0.004	0.008
Control	0.048	0.063	0.074	0.119	0.017	0.025

The imazethapyr retained in top soil (0-15 cm depth) was high ((Fig 1C) and is ranged from 12.06-29.76 % at surface when compared to 10.37-22.32 % at subsurface (15-30 cm depth) irrespective of organic amendments and rates applied. The imazethapyr residue was also detected in leachates of soil in the range of 10.33 – 28.67 % across different amendments and rates of application. The quantity measured in biochar was low and it was 17.3 and 18.0 % respectively at 75 and 150 g/ha rate of application. Control showed higher quantity in leachates (Fig 2) irrespective of rates applied and the mean quantity detected in leachate was equal to that in sub surface (15-30 cm). This showed that the imazethapyr is highly leaching herbicide when compared to atrazine and pendimethalin.

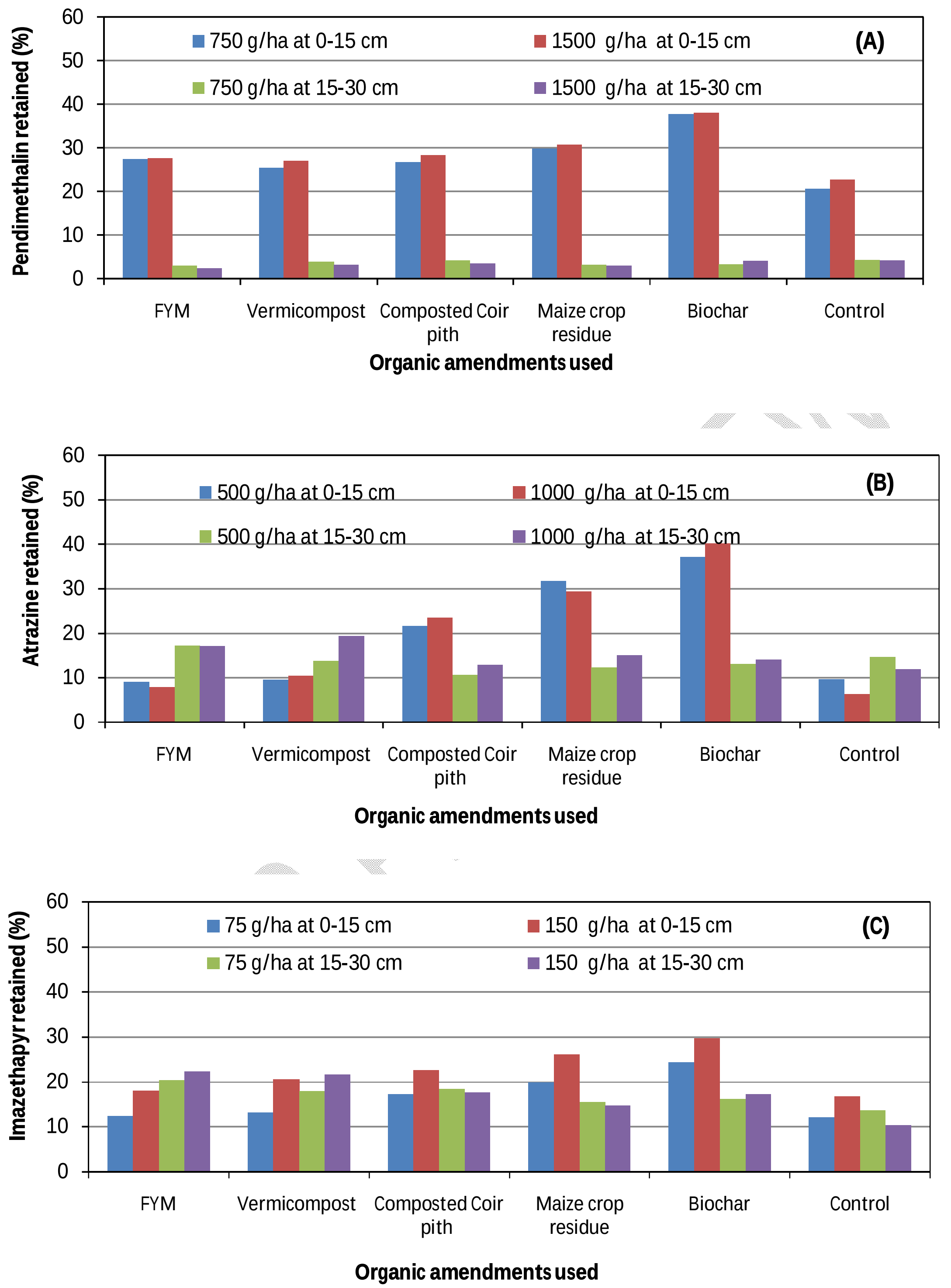

Fig 1. Effect of different organic amendments and rates of herbicides application on their retention in soil at different depths (A. pendimethalin; B. Atrazine; C. Imazethapyr)

Table 4. Effect of different organic amendments and imazethapyr rates on its retention in soil (mg/kg) at different depths and in leachate (mg/L)

Soil depth (cm) / Amendments	0-15 cm (ppm)		15-30 cm (ppm)		Leachate (ppm)	
	750 g/ha	1500 g/ha	750 g/ha	1500 g/ha	750 g/ha	1500 g/ha
FYM	0.009	0.027	0.015	0.033	0.016	0.022
Vermicompost	0.010	0.031	0.014	0.033	0.014	0.020
Composted Coir pith	0.013	0.034	0.014	0.026	0.016	0.020
Maize crop residue	0.015	0.039	0.012	0.022	0.015	0.019
Biochar	0.018	0.045	0.012	0.026	0.014	0.016
Control	0.009	0.025	0.010	0.016	0.022	0.028

Retention of herbicides is high in biochar applied soil at both the rate of application and the retention order was influenced by the different organic amendments and soil depth irrespective of rates of application at 0-15 cm depth is Biochar> maize crop residue> composted coir pith> Vermicompost>FYM and at 15-30 cm depth is Biochar ≥composted coir pith≥Vermicompost> maize crop residue >FYM.

Among the different organic amendments, biochar has retained more quantity in top soil which showed the efficiency of the biochar to increase the herbicides sorption in soil. This could be attributed to the higher sorption capacity enabled by the increased surface area, higher CEC besides having more ash content and organic functional groups. These made the biochar a best absorbent to retain the applied herbicides in the surface soil itself (Aldana et al., 2020). Similarly Ouyang et al. (2016) reported that the addition of 5% of corn cob and straw biochar increased the removal rate of 1 mg/L atrazine solution from 27.02 to 76.65%. Carpio et al., (2021) reported that the greater or lesser leaching of pesticides in amended soils may not be due solely to the presence of additional SOM or DOM in amended soil, but also to structural changes in soil porosity induced by the higher soil OC content (Worrall et al., 2001). Increased imazapyr retention in top soil with biochar was also reported by Yavari et al. (2021). In general, a decrease in pesticide leaching was observed because of enhanced pesticide sorption as the influenced by the organic amendments in soils (Carpio et al., 2021).

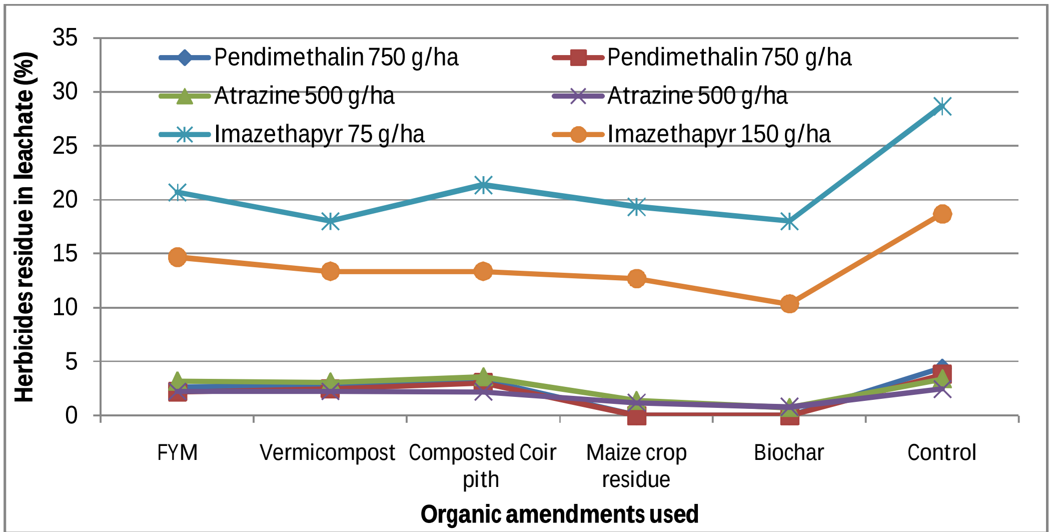

Fig 2. Effect of different organic amendments and rates of herbicides application on their residual concentration in leachates

The per cent retention of herbicides viz., pendimethalin and atrazine is more in double the recommended dose (2x) applied column (Fig 1). Their retention is high in biochar applied soil at both the rate of applicaiton. The retention order influenced by different organic amendments and soil depth irrespective of application rates is at 0-15 cm depth is biochar> maize crop residue> composted coir pith> vermicompost>FYM and at 15-30 cm depth is vermicompost> FYM> maize crop residue>biochar>composted coir pith.

The per cent retention of imazethapyr is more in double the recommended dose (2x) applied column ((Fig 1). Retention of all the three herbicides is high in surface (0-15 cm depth) in biochar applied at both the rate of application and the retention order influenced by different organic amendments and rates of application in surface 0-15 cm depth soil is biochar> maize crop residue> composted coir pith> vermicompost>FYM. Similar higher retention of imazethapyr in surface soil was reported by Yavari et al. (2021) due to biochar application. In sub surface soil, the herbicides retention was significantly influenced by the organic amendments. The order of herbicides retention in 15-30 cm depth soil for pendimethalin is control>composted coir pith≥biochar>maize crop residue> vermicompost>FYM. Order of retention for atrazine is vermicompost>FYM> biochar ≥maize crop residue> control>composted coir pith and imazethapyr is composted coir pith>FYM>vermicompost>biochar>maize crop residue> control.

The herbicides quantity detected in leachate was significantly influenced by the organic amendments. In general biochar application reduced the leaching of all the three herbicides and was followed by the maize crop residues when compared to other amendments and control (Fig 3). Leaching reduction effect was found to be 0.95-4.10 times higher for pendimethalin, 0.03-2.16 times higher for atrazine and 1.34-1.68 times higher for imazethapyr due to the organic amendments application when compared to control. For all the herbicides biochar incorporation showed higher efficiency of leaching reduction and was followed by maize crop residue. Similar positive correlation between the mobility level of pesticide and the biochar addition was reported by Hui et al. (2017) who found that the biochar from cassava wastes application enhanced retention of atrazine in soil by increasing its sorption.

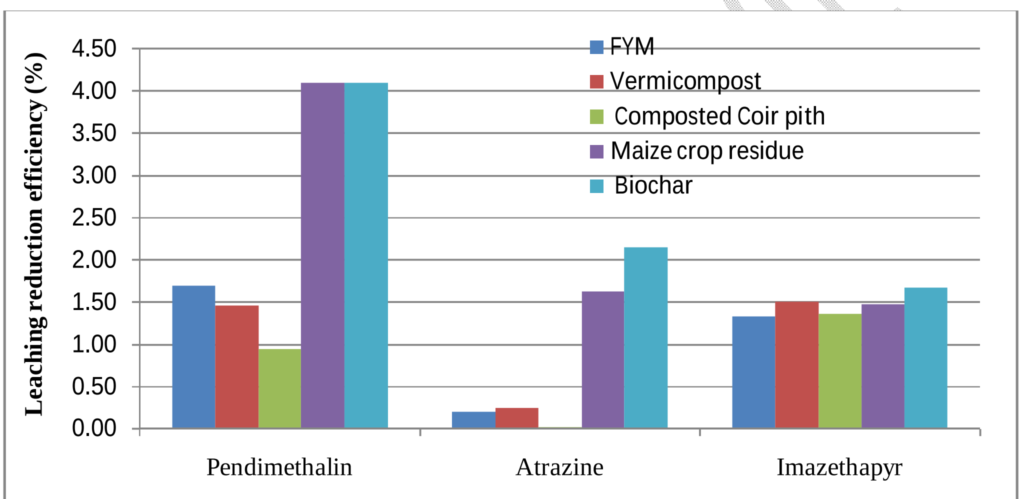

Fig 3. Efficiency of different organic amendments on reducing the leaching of herbicides in soil compared to control

Lower efficiency of leaching reduction was seen with composted coir pith for pendimethalin and atrazine and FYM for imazethapyr. Overall, all the organic amendments showed less leaching efficiency for imazethapyr and could be ascribed to the nature of the compound. Since imazethapyr is an amphoteric compound, its anionic form pyridine nitrogen predominates in the present high pH soil and might cause low sorption due to repulsive forces (Stougaard et al., 1990; Oufqir et al., 2017) and hence leaching was high. Further the organic matter addition to the soil through different sources did not have significant influence on reducing the leaching behavior of imazethapyr since pH was not altered much due to the buffering capacity of the soil. However biochar showed little higher efficiency on altering the leaching behavior of imazethapyr when compared to other organic amendments. This could be attributed to the strong binding of pesticides to biochar elemental composition and increased sorption (Oufqir et al., 2017) and hence might decrease its leaching in the present soil.

Influence of organic amendments on adsorption of herbicides

The influence various organic amendments on the adsorption pendimethalin, atrazine and imazethapyr was studied at different concentrations including the recommended rate. Results showed that an increased herbicides concentration increased their adsorption (Fig 4 to 6) and adsorption was found to be higher with biochar and was followed by the maize crop residues and other amendments. The difference in herbicide adsorption with respect to soil type is attributed to the chemical properties of the organic amendments used and the herbicide molecules studied. Like other polar ionisable pesticides, the sorption of imidazolinone herbicides is highly sensitive to soil pH, because of the amphoteric characteristics of these herbicides (Oufqir et al., 2017).

The shapes of the adsorption isotherm for all the three herbicides were assessed at different concentrations as influenced by the various organic amendments (Fig 4 to 6). It was found that the isotherm of three herbicides viz., pendimethalin, atrazine and imazethapyr expressed an increasing trend in the adsorbed content C_s (mg/kg) with respect to increase in the equilibrium concentration C_e (mg/L) in solution. It was seen that the shape of both pendimethalin and atrazine adsorption was 'L' type with all organic amendments except biochar where they showed 'H' type sorption. This confirmed the increased availability of sites with increased pendimethalin concentration. Irrespective of organic amendments treatment, the imzaethapyr sorption showed 'S' type which showed that the solid has high affinity for the solvent. Similar shapes for the sorption of atrazine, pendimethalin and imazethapyr was reported by Janaki et al. (2019) in different soils of Tamil Nadu.

Table 5. Adsorption of herbicides (mg/kg) as influenced by different organic amendments

Concentration added / Amendments	Pemdimethalin		Atrazine		Imazethapyr	
	0.75 mg/kg	1.50 mg/kg	0.50 mg/kg	1.00 mg/kg	0.075 mg/kg	0.150 mg/kg
FYM @12.5 t/ha	0.380	0.460	0.320	0.790	0.056	0.121
Vermicompost @5t/ha	0.420	0.573	0.190	0.670	0.058	0.123
Composted Coir pith@5t/ha	0.470	0.654	0.210	0.690	0.058	0.131

Maize crop residue @ 5 t/ha	0.260	0.431	0.410	0.860	0.065	0.135
Biochar @ 5 t/ha	0.080	0.210	0.420	0.890	0.068	0.140
Control	0.450	0.685	0.350	0.790	0.054	0.090

A linear fit (straight line) of Freundlich adsorption isotherm was obtained with experimental data for all the three herbicides irrespective of organic amendments (Fig 4 to 6) with the correlation coefficient 90 % and above. The distribution coefficient values (K_d) indicate (Table 6) higher adsorption of all the three herbicides in biochar treated soil followed by maize crop residue and composted coir pith which confirms the relevance of organic amendments and their nature on modifying the herbicides sorption in soil. The Koc values were found to be high for biochar as that of Kd which showed the increased sorption sites with biochar, maize crop residue than the composted coir pith, vermicompost and FYM. Similar effect of biochar on herbicides sorption was reported by Hui et al. (2017) and Ouyang et al. (2016).

Table 6. Koc and Kd values for herbicides sorption as influenced by different organic amendments

Organic amendments	Pendimethalin		Atrazine		Imazethapyr	
	K_{OC}	Kd	K_{OC}	Kd	K_{OC}	Kd
FYM @12.5 t/ha	36.18	20.98	41.73	24.20	5.56	3.22
Vermicompost @5t/ha	35.27	20.46	36.49	21.16	6.13	3.56
Composted Coir pith @ 5t/ha	42.69	24.76	34.56	20.04	6.29	3.65
Maize crop residue @ 5 t/ha	57.48	33.34	56.52	32.78	13.68	7.94
Biochar @ 5 t/ha	225.49	130.78	184.07	106.76	22.44	13.02
Control	11.56	6.70	10.00	5.63	5.16	3.00

The KOC values were calculated and used to assess the mobility of studied herbicides in soil according to FAO Soil mobility classification (<10-higly mobile; 10 to 100- mobile; 100 to 1000- moderately mobile; 1000 to 10000- slightly mobile; 10000 to 100000 – Hardly mobile; >100000 – Immobile). Based on the control treatment data, the imazethapyr is classified as highly mobile one and atrazine and pendimethalin are classified as mobile herbicides. When organic amendments were applied, the KOC values for atrazine and pendimethalin were increased towards moderate mobility particularly in biochar treated soil. However the imazethapyr mobility was not affected much by the organic amendments except biochar which changed its mobility class from highly mobile to mobile category. Such a decrease in herbicides mobility due to the addition of organic amendments in soil was also reported by Hui et al. (2017) and Ouyang et al. (2016).

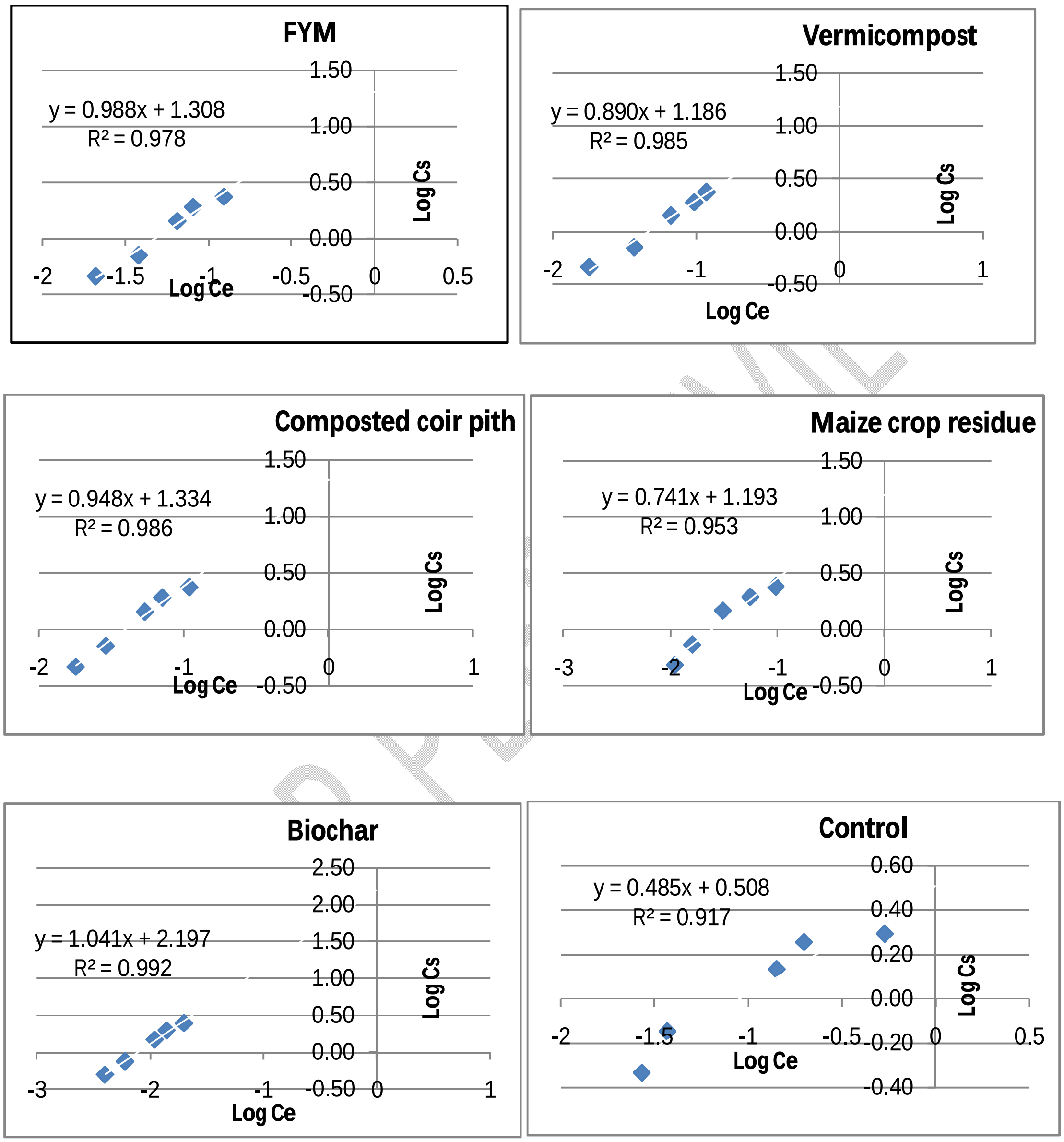

Fig 4. Fruendlich Sorption isotherm of pendimethalin as influenced by the various organic amendments

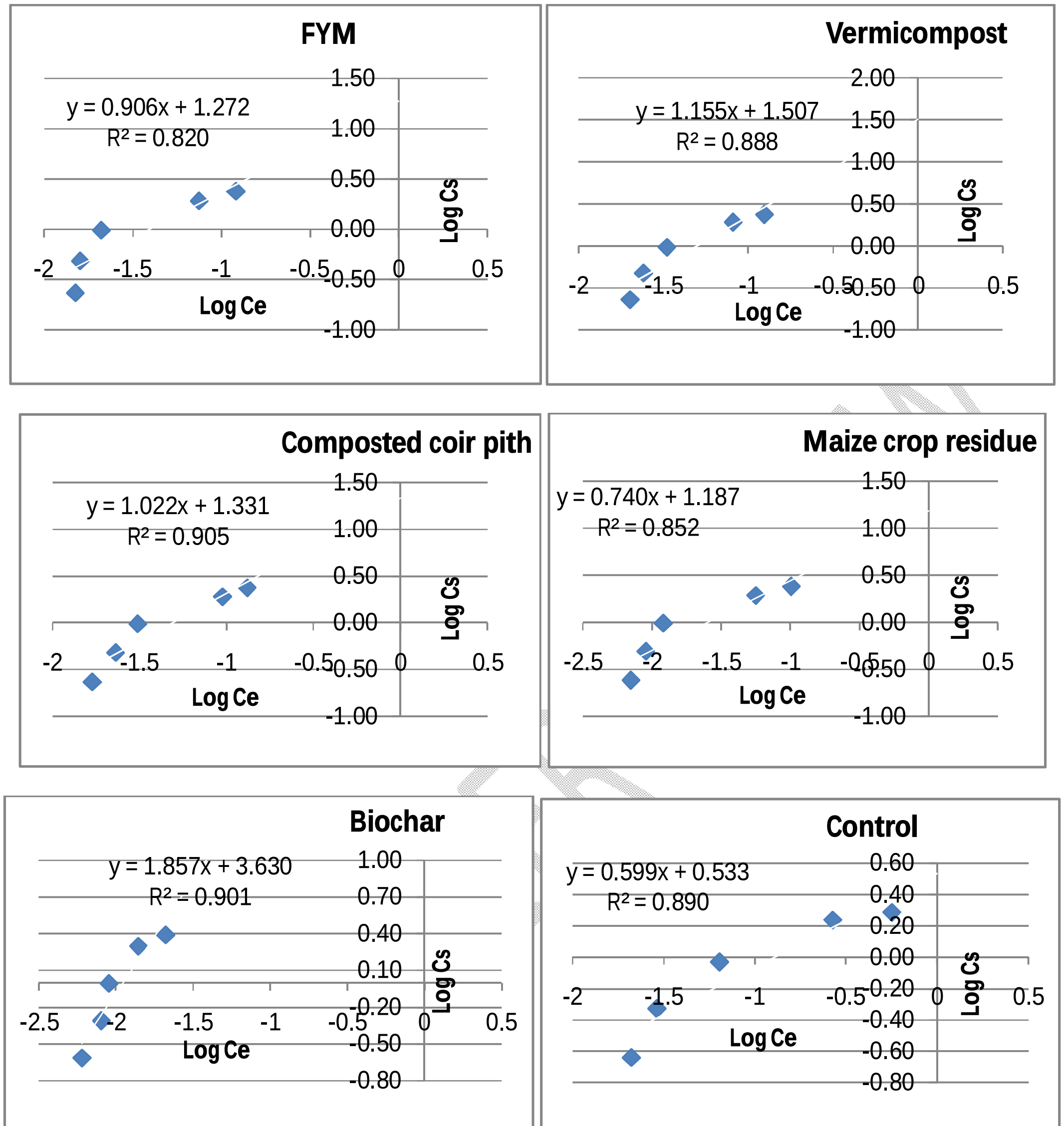

Fig 5. Fruendlich Sorption isotherm of atrazine as influenced by the various organic amendments

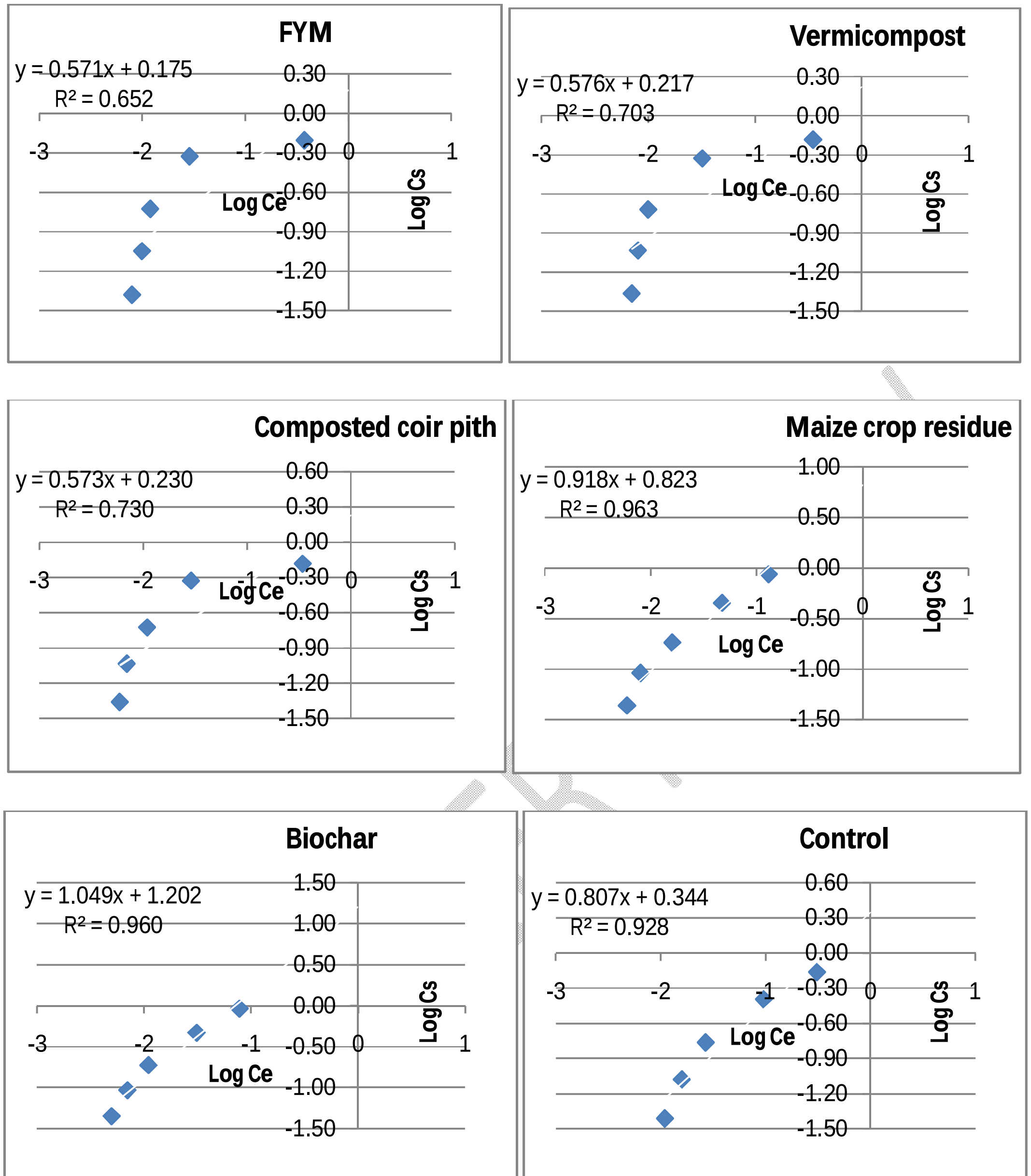

Fig 6. Fruendlich Sorption isotherm of imazethapyr as influenced by the various organic amendments

Conclusion

One of the management suggested to overcome the herbicide contamination is the application of various organic amendments. Hence the influence of organic amendments on the movement and leaching behavior of commonly used herbicides was investigated. Results showed that the leaching and movement and sorption behavior of the herbicides viz., pendimethalin, atrazine and imazethapyr in soil was modified by the organic amendments particularly the biochar and maize crop residue. Biochar treated soil retained 29.76, 40.20, and 38.07 percent of the applied pendimethalin, atrazine and imazethapyr in surface 0-15 cm depth soil and hence reduced its movement to lower depths and to leachates. All the applied organic amendments reduced the leaching by increasing their sorption when compared to control and was highly significant with pendimethalin and atrazine and non significant with imazethapyr showed the effect of herbicide nature on its movement beside the organic amendments type. The present study shows the effect of various organic amendments on herbicide dynamic behavior in soil under controlled condition keeping all external factors constant. Hence future studies are needed to assess the influence of various organic amendments particularly the biochar and maize crop residue on modifying the herbicides sorption, movement and leaching behvaiour on long run under actual field conditions and their influence on the environmental fate of herbicides.

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