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Environmental impact of biochar and wheat straw on mobility of dinotefuran and metribuzin into soils

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Abstract. *Fouad MR, El-Aswad AF, Aly MI, Badawy ME-T. 2024. Environmental impact of biochar and wheat straw on mobility of dinotefuran and metribuzin into soils. Asian J Agric 8: 57-63.* The environmental effect of 5% biochar and wheat straw on the mobility of dinotefuran (DNF) and metribuzin (MBZ) in alluvial soil (A) and calcareous soil (B) was studied. The breakthrough (BTC) curves of DNF and MBZ were delayed in soil columns compared to that of iodide. The amendment of soil A or B columns with biochar and wheat straw increased the leaching of DNF collected in leachates, consequently reducing the breakthrough time. Biochar amendment increased the leaching of MBZ in soil A, whereas it significantly reduced the leaching of MBZ in soil B. Wheat straw amendment also significantly reduced the MBZ cumulative percentage. It is indicated that the downward of DNF in soil A is more rapid than that in soil B, while MBZ was faster in soil B compared to soil A; the leaching rate of MBZ is higher than DNF in all soil columns. The calculated GUS indices of DNF and MBZ were lower than 2.8, which means these pesticides are non-lechers. DNF required more water to leach from soil B than from soil A. In contrast more water was needed to leach MBZ from soil A compared to soil B. In addition, the DNF required more water for leaching from soil A or B than MBZ.

Keywords: Dinotefuran, metribuzin, mobility, soil amendments

INTRODUCTION

The pesticide leaching potential in soil is described by the pesticide sorption characteristics. The attenuation factor and retardation factor are two indexes frequently used in predicting the environmental risk assessment of pesticide groundwater contamination (Rao et al. 1985; Mulla et al. 1996; Abdel-Raheem et al. 2023; Abd-Eldaim et al. 2023; El-Aswad et al. 2023a). The attenuation factor thus estimates the mass emission of pesticides in groundwater. The retardation factor is a numerical representation of the slowing down of pesticide leaching to the water flow within the soil. The leaching delay is caused by the soil's sorption and degradation of pesticides (El-Aswad et al. 2024a, b) and the soil diffusion of pesticides in gaseous and aqueous forms. The graph of the breakthrough curve represents the relationship between the relative concentration and time evolution concentration (Paraiba and Spadotto 2002; El-Aswad et al. 2023b). The mobility of the pesticides has been related to the total OM content, with the nature of the OM having little apparent influence on sorption processes (Bekbölet et al. 1999; Fouad 2023a). Conventional pesticide formulations are applied at rates exceeding the minimum threshold concentration to mitigate losses from sorption (Fouad et al. 2023a, 2024a, b) photodecomposition, chemical and microbial degradation, volatilization (Fouad et al. 2023b), and leaching (Fouad 2023b, c).

Leaching is one of the important factors affecting the herbicidal activity of soil-applied herbicides. Some herbicides may disappear from the soil's upper layers where most weed seeds are located by excessive leaching when heavy rainfall occurs or when large amounts of irrigation water are applied. Leaching data is valuable for predicting and comprehending pesticide behavior in diverse soil types under varying rainfall conditions. There was a direct correlation between the pesticides' water solubility and the depths of leaching in mineral soils; the leaching depth decreased as the soil clay content increased. Leaching depth also decreased as OM increased (Gray and Weierich 1968). A set of column experiments was conducted to assess the leaching potential of dinotefuran (DNF), thiamethoxam, and imidacloprid insecticides in two experimental conditions: mixed and individual pulse modes. In both cases, the pesticide breakout pattern in the predominantly acidic to neutral vineyard soil illustrates their medium to high leachability. DNF has shown a high tendency to leach and has passed through the column with fewer pore volumes, while imidacloprid was retained for a longer period, indicating lower leachability (Kurwadkar et al. 2014). The potential for leaching into surface water associated with the widespread use of neonicotinoids, particularly near water bodies, is a significant concern (Anderson et al. 2015; Fouad 2023d).

Metribuzin (MBZ) is one of the most important contaminants in ground and surface waters (López-Piñeiro et al. 2013). MBZ is characterized by its high water solubility (1,050 mg/L) and low soil adsorption (Koc = 60); therefore, it has a high potential for soil profile movement. The US EPA maximum advisory concentration for MBZ in drinking water is 175 μg/L (Singh 2009). The mobility of MBZ within the soil profile is of concern because crop phytotoxicity levels depend on the quantity of MBZ absorbed by the root system and possible groundwater contamination. The movement of herbicides through soil columns is analogous to chromatography, and the extent of movement in soil could be predicted from the distribution or adsorption coefficient, the void volume, water content, and OM content (Savage 1976). MBZ is readily leached in sandy soils low in OM content, but leaching potential is reduced on finer textured soils (Milburn et al. 1991).

MATERIALS AND METHODS

Tested pesticides

Dinotefuran

IUPAC name: (RS)-1-methyl-2-nitro-3-(tetrahydro-3-furylmethyl) guanidine. Structure: Shown in Figure 1. Solubility (20°C): Water 54.3 g/L, methanol 57.2 g/L and dichloromethane 60.9 g/L. Chemical class: Neo-nicotinoid. Pesticide type: Insecticide.

Metribuzin

IUPAC name: 4-amino-6-terbutyl-3-methylsulfanyl-1,2,4-triazin-5-one. Structure: Shown in Figure 1. Solubility (20°C): Water 1,200 mg/L, methanol (several hundred times greater than in water). Chemical class: Triazinone. Pesticide type: selective systemic herbicide.

Tested soils

This study tested two types of Egyptian soil: clay loam and sandy loam. The samples were collected from the topsoil profile layers from different locations. The physical and chemical properties were determined at the Department of Soil and Water Sciences, Faculty of Agriculture, University of Alexandria, Egypt, and the data are presented in Table 1.

Tested soil amendments

The study tested four amendment substances. The commercial substances' forms were obtained from the Faculty of Agriculture, University of Alexandria, Egypt.

Mobility study

The bench-scale soil columns were employed to assess the potential mobility of the pesticides tested in both soil and amended soil with 5% biochar or wheat straw. Each column was uniformly filled with 3 kg of soil to achieve a known bulk density (Weber et al. 1986; Fouad 2023b, c). The bottom end cap supported a porous stainless-steel plate, and the control column soil was similarly mixed for consistency. The columns were pre-treated with 0.01 M $CaCl_2$ before applying the pesticide and KI (Thanos and Maniatis 1995). The flow rate was chosen to give saturated flow conditions and to limit leaching experiment timescale, thereby reducing potential pesticide degradation. Next, 10 mL of KI solution with a concentration of 0.2 molar was used as a water tracer. Dinotefuran or metribuzin was then added to each column to achieve a soil concentration of 10 µg/g.

Table 1. Physical properties and chemical properties of tested soils

Properties	Soil A	Soil B
Soil type	Alluvial	Calcareous
Texture class	Clay loam	Sandy loam
Water holding capacity (mL/100g)	46	35
EC (m mohs/cm) at 25°C	1.32	2.34
Soil pH	8.25	8.20
Organic matter content (%)	3.31	1.32
Total carbonate (%)	7.87	40.09
Soluble cations conc. (meq/L):		
Ca^{++}	3.8	8.8
Mg^{++}	5.0	7.0
Na^{+}	9.4	15.3
K^{+}	0.5	2.4
Soluble anions conc. (meq/L):		
CO_3^{-}	1.6	1.4
HCO_3^{-}	2.6	3.4
Cl^{-}	8.5	16.5
SO_4^{-}	0.6	1.8

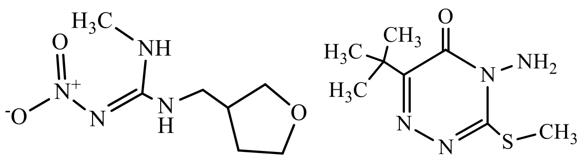

Figure 1. Chemical structure of dinotefuran and metribuzin

Next, we applied the $CaCl_2$ solution and collected the leachates (25 mL leachate). The KI was determined in all leachate samples using the iodimetric method (Mendham et al. 2000). Additionally, the tested pesticides were identified by analyzing all leachates.

Statistical analysis

Experimental data are presented as mean ± standard error, and the statistical analysis was performed by the SPSS program (ver. 21.0, USA).

RESULTS AND DISCUSSION

Figure 2 represents DNF's breakthrough (BTC) curves compared to iodide as a water tracer in columns of unamended soil A and B and 5% of biochar and 5% of wheat straw amended soil A and B. The BTC curves of DNF were delayed in soil A and soil B columns compared to that of iodide; the top of the BTC corresponding to the maximum concentration of DNF was obtained after percolating 8.2 L of leachates. All BTC of DNF needed about 1.1 L of cumulative volume. The amendment of soil A or B columns with biochar or wheat straw increased DNF's leaching, reducing the breakthrough time. The maximum concentration of DNF was recorded after percolating 1,000 mL and 3,000 mL of leachates cumulative volume of biochar amendment soil A and soil B columns, respectively. Also, the top of the breakthrough peak of DNF was obtained after percolating 3,000 mL leachates cumulative volume of wheat straw amended soil A and soil B columns. All BTC need about 6 L of leachates. Accordingly, there was no decrease in the DNF leachate obtained from biochar and wheat straw amended columns. The BTC curve is a specific form that depends on the chemical's soil solids, physicochemical properties, and the soil structure through which the chemical moves (Shipitalo et al. 1990).

In addition, 97.2 % and 91.5 % of applied DNF in the unamended soil A and B columns were recovered in the leachate after percolating about 12 L of $CaCl_2$ solution. About 89.2%, 98.0%, 93.2%, and 89.3% of the applied DNF were recovered in the leachates of biochar-amended soil A and B columns and wheat straw amended soil A and B columns, respectively. However, adding soil A and B with biochar or wheat straw significantly increased the cumulative percentage of DNF collected in leachates (Figure 3). It was observed that the data indicated that the leaching of DNF was low in the soil columns. Furthermore, adding soil columns with 5% biochar or wheat straw increased the DNF leachability.

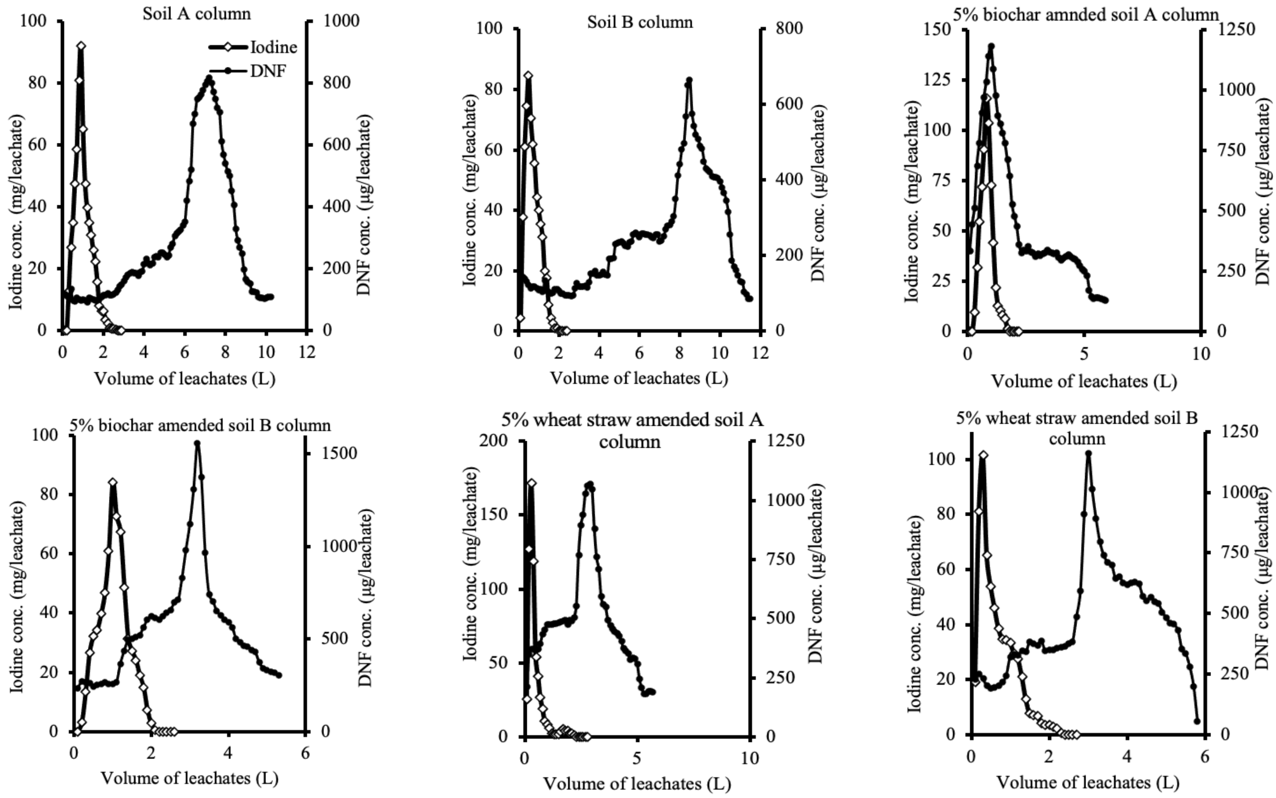

Figure 2. Breakthrough curves of insecticide DNF and water tracer I- in unamended and amended soil columns

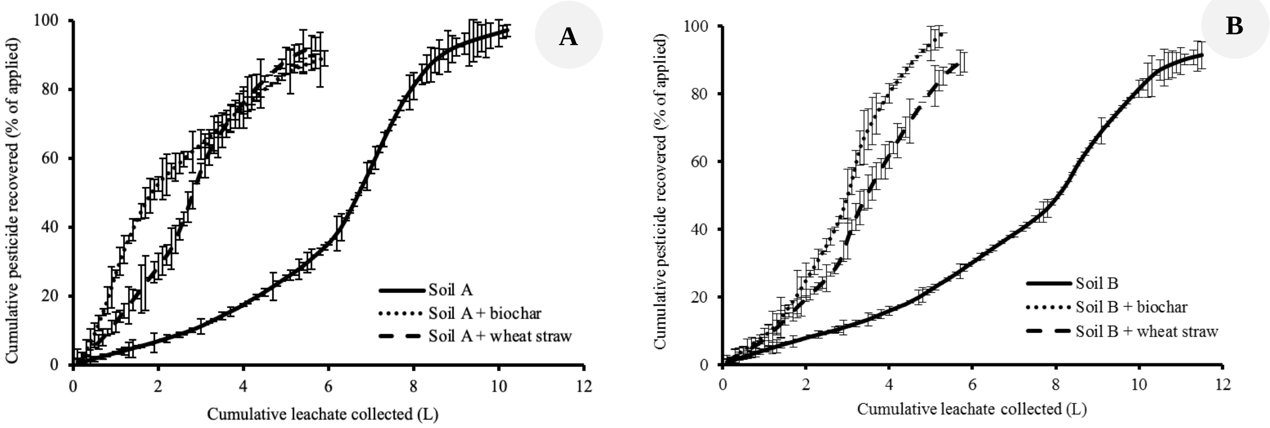

Figure 3. Cumulative leachate curves of DNF in unamended and amended soil A (upper) and B (lower) columns

Therefore, the leaching of pesticides in the soil depends on the sorptive characteristics of the compound and soil. Sorptive characteristics significantly affect the physical leaching of pesticides within a soil-water matrix (Perry et al. 1988). Dinetofuran is highly water soluble but exhibits high percent sorption (Kurwadkar et al. 2013). Also, this observation is similar to the findings reported by Kinney et al. (2006), who reported that acetaminophen with higher water solubility tended to accumulate more in the soil than other compounds with low water solubility (Kinney et al. 2006). Moreover, DNF is highly leachable compared to the other neonicotinoids (Kurwadkar et al. 2014).

Figure 4 exhibited the breakthrough curves of metribuzin in unamended soil A and B and 5% biochar and wheat straw amended soil columns. The iodide BTCs observed from the release of these compounds started with the first leachates. The accumulated volume of the leachates until the top of BTC was about 2.6 L with a concentration of 120 µg/mL for soil A column, whereas about 0.6 L with 190 µg/mL for soil B column. It is indicated that the downward of MBZ in sandy loam soil is more rapid than in clay loam soil. The BTCs of MBZ for soil A and B columns required nearly 5 L accumulation volume. The BTCs of MBZ were retarded relative to the I^- movement and exhibited a flatter peak in soil A and tailing in soil B column. The highest concentration of MBZ was delayed by about 500 mL to the highest concentration of iodide in biochar and wheat straw soil A columns. The highest concentration of MBZ in biochar soil A column was 148 µg/mL of leachate, corresponding to 1.5 L cumulative volume. The concentration decreased to 57 µg/mL of leachate, corresponding to 2.3 L, which remains constant until the 5.5 L cumulative volume. Almost the same shape of BTC was obtained in wheat straw amended soil A columns but with broader. The highest concentration of MBZ in the wheat straw column was 100 µg/mL of leachate. Then, the concentration of MBZ decreased to a minimal value after the release of 5.5 L leachates.

The breakthrough curves of MBZ in biochar and wheat straw amended soil B showed two peaks. The first peak in each case was small and approximately identical to that of iodide, while the second peak was much larger. The second peak in biochar-amended soil B was symmetrical, and its maximum concentration of MBZ was 68 µg/mL, corresponding to about 4.5 L cumulative volume. The second peak in wheat straw amended soil B was flatter, and its maximum concentration of MBZ was 82 µg/mL, corresponding to a 2.5 L cumulative volume. The BTC of MBZ contained two peaks needs 5.5 L leachates cumulative volume neither in biochar amended soil B nor in wheat straw amended soil B. El-Aswad et al. (2002) suggested that in the case of BTC contained two peaks, some pesticide molecules might be leached through macropores without interference with soil matrix, giving rise to a small peak and leaching of pesticide in the soil matrix was occurred by diffusion through micropores, giving a greater peak.

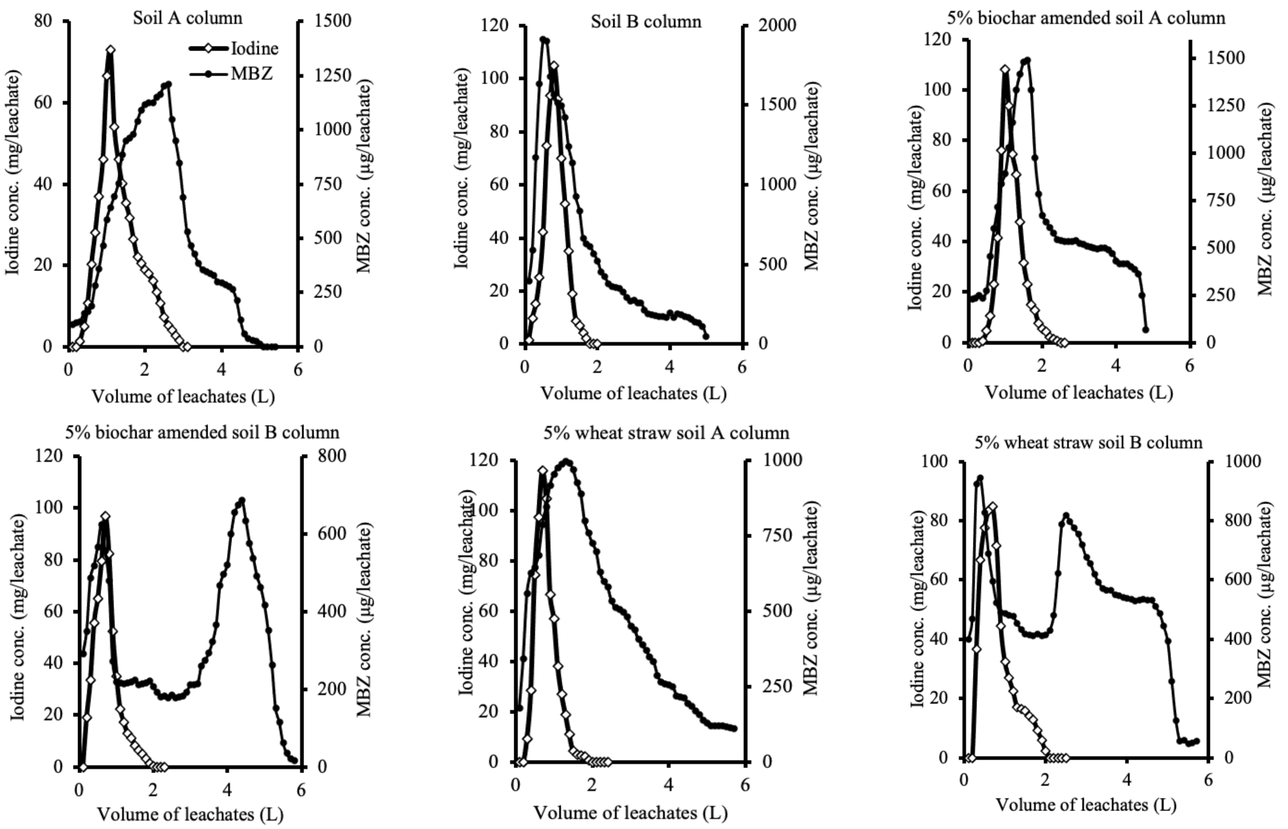

Figure 4. Breakthrough curves of insecticide MBZ and water tracer I- in unamended and amended soil columns

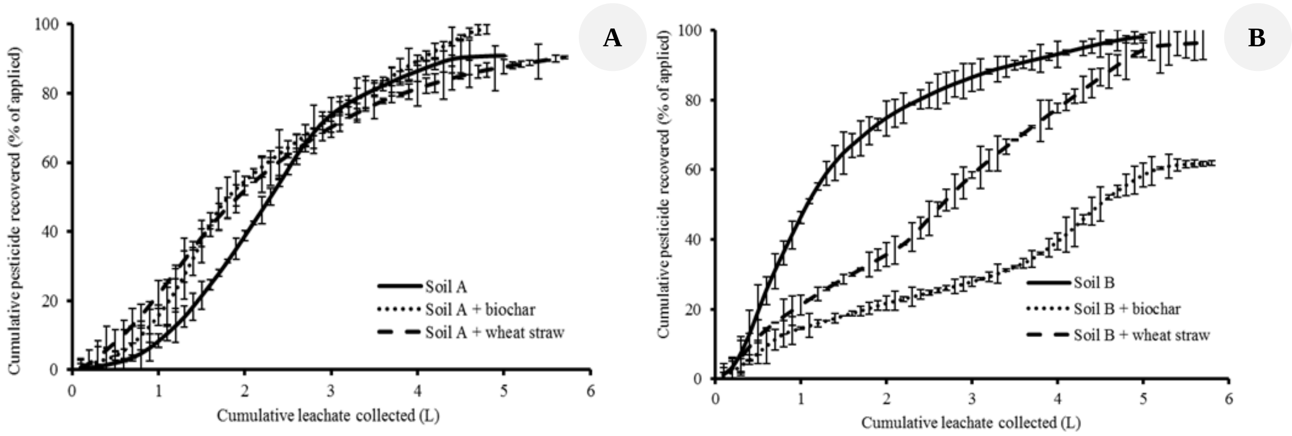

Figure 5. Cumulative leachate curves of MBZ in unamended and amended soil A (upper) and B (lower) columns.

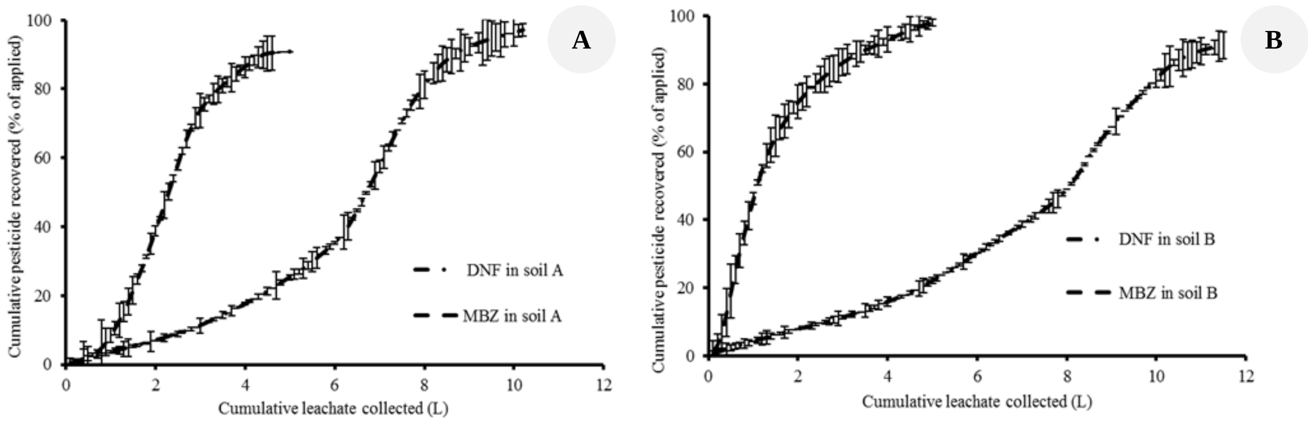

Figure 6. Comparison of tested pesticides cumulative leachate curves in unamended and amended soil A (upper) and B (lower) columns

The cumulative percentages of MBZ collected in leachates of unamended soil A and B columns were 90.8% and 98.0%, respectively. Biochar amendment increased the leaching of MBZ in soil A and significantly reduced the leaching of MBZ in soil B (Figure 5). Compared to 98.0% leaching losses of MBZ from unamended soil B columns, leaching losses of MBZ were significantly reduced to 61.8% after biochar amendment. The amendment of soil A with wheat straw increased the cumulative percentage of MBZ until 3 L percolated, then it was amended, and unamended soils were symmetrical. BTCs of MBZ and I indicated that the cumulative percentage of herbicide decreased until 6 L percolated. Therefore, 90.4% of the initially applied MBZ was recovered from the wheat straw amended soil A column leachates. The wheat straw amendment also significantly reduced the MBZ cumulative percentage; about 96.6% of the applied MBZ was recovered in the wheat straw amended soil B column leachates. A previous study showed that both animal manure and fly ash are highly effective in decreasing the downward movement of metribuzin in packed soil columns of sandy loam soil (Majumdar and Singh 2007). Jones et al. (2011) also demonstrated that biochar decreased the leaching of simazine from the soil due to the strong binding capacity of biochar. Peter and Weber (1985) found that 82.3% of the applied metribuzin was recovered in the leachate, and they observed that MBZ is highly mobile in soil. Consequently, the activity of MBZ increased, in which leaching was prevented, indicating that leaching may be important in the loss of activity of MBZ (Peek and Appleby 1989).

Moreover, comparing the leachability of tested pesticides in columns of soil (A) and soil (B), the recovered amounts were standardized to 100% to compare the tested pesticides (see Figure 6) directly. The leaching rate can be arranged in the following order in soil A and B columns: MBZ > DNF. The differences detected in pesticide leaching were more significant in the columns containing soil A than in those containing soil B, except for DNF, which had the lowest leachability. There were notable variations found in the cumulative percentages of DNF and MBZ. It was observed that the arrangement of the pesticides according to their cumulative percentage in soil A columns corresponding to the arrangement depends upon their water solubility, DNF (54.3 g/L) > MBZ (1.2 g/L). Harris (1966) and Rodgers (1968) studied several herbicide leaching and found water solubility is crucial, but adsorption data provided a better indicator of mobility.

Savage (1976) and Sharom and Stephenson (1976) found that MBZ mobility in soils was inversely related to the soil sorption capacity of the metribuzin molecules. Greater movement of MBZ was observed in soil, particularly in coarser textured column soil (Peek and Appleby 1989; Beck et al. 1993). In addition, DNF has exhibited high leaching potential in soil columns (Kurwadkar et al. 2014). Morrissey et al. (2015) indicated that DNF has high leaching and runoff potential. Therefore, due to the high water solubility, DNF risks water contamination higher, particularly by leaching into the groundwater (Sánchez-Bayo and Hyne 2014).

Generally, data from leaching are valuable for predicting and comprehending the behavior of pesticides in various soil types (Gray and Weierich 1968). Moreover, the drinking water directive (98/83/EC) requires pesticide concentration in drinking water not to exceed 0.1 µg/L for a single pesticide and 0.5 µg/L for total pesticides. Not only does groundwater pollution harm human health since it is used for drinking, but when used for irrigation, it contaminates the food chain (Navarro et al. 2007). The study by Si et al. (2006) suggested that organic amendment could be an efficient strategy for regulating pesticide leaching. Incorporating organic amendments into calcareous and sandy soils may gradually diminish the risk of groundwater contamination by pesticides, as evidenced by research from Cox et al. (1997), Tatarková et al. (2013) and Fouad et al. (2024c, d).

The Gustafson model is based on the pesticide partition coefficient between soil organic carbon and water (Koc) and pesticide soil disappearance (half-life DT50, required for 50% dissipation of initial concentration) (Gustafson 1989). The relationship between these two parameters is called the Groundwater Ubiquity Score (GUS) index.

$$\text{GUS index} = [\log DT_{50}] \times [4 - \log Koc]$$

Higher values of the GUS index indicate the pesticide will have higher mobility, posing a greater threat to groundwater resources. Gustafson has ranked many tested pesticides into leachers, non-leachers, and transient, which have intermediate properties (Bottoni et al. 1996). Accordingly, the calculated GUS indices of DNF and MBZ were lower than 2.8, meaning the tested pesticides in soil A and B are non-lechers. After calculating Kd values for the tested pesticides, indicating that the Kd value of DNF = 1.972 and 2.438 mL/g and MBZ = 0.505 and 0.105 mL/g in soil A and B, respectively. These results illustrated that MBZ required more water to leach from soil A than from soil B; in contrast, more water was needed to leach DNF from soil B than from soil A. In addition, the compound DNF required more water for leaching from soil A or B than MBZ (El-Aswad et al. 2024c; Fouad et al. 2024e).

In conclusion, the water tracer I^- was leached fast, and their BTCs in unamended soil A and B and biochar and wheat straw amended soil columns are symmetrical. The BTC curves of DNF were delayed in soil A and soil B columns compared to that of iodide. The amendment of soil A or B columns with biochar or wheat straw increased DNF's leaching, reducing the breakthrough time. The downward of MBZ in sandy loam soil is more rapid than in clay loam soil. The BTCs of MBZ in biochar and wheat straw amended soil B showed two peaks. The first was small, it attributed to leaching through macropores, while the second peak was much larger, it attributed to leaching through micropores. The soil A and B column's leaching rates can be sorted as follows: MBZ>DNF. The evaluated pesticides' computed GUS indices were less than 2.8. This indicates that the tested soils' DNF and MBZ are not leachers. The Kd values for the pesticides tested in soil columns showed that more water was needed for MBZ to leach from both soils. Conversely, compared to soil A, more water was required for soil B to drain DNF. Furthermore, compared to MBZ, the compound DNF needed more water to leach from soil A or B.

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