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Tracking movement dynamic of fenitrothion and thiobencarb in rice paddy using a field lysimeters at different levels of soil depth

Mohamed R. Fouad[a*], **Ahmed F. El-Aswad**[a] and **Maher I. Aly**[a]

[a]*Department of Pesticide Chemistry and Technology, Faculty of Agriculture, Alexandria University, Aflaton St., 21545, El-Shatby, Alexandria, Egypt*

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ABSTRACT

In this study, the movement dynamic of fenitrothion (50% EC) and thiobencarb (50% EC) was investigated using the field lysimeter in the presence of rice plant at four different levels of soil depth. Iodide was used as an indicator of the mobility of these pesticides through the soil in the field lysimeter. Iodide was detected in the leachates collected at level 1 and 2 only, the concentration of iodide collected from level 2 was more than those collected from level 1. The highest breakthrough curve for fenitrothion or thiobencarb was produced from the level 4 (deep level) followed by level 3 while the breakthrough curve of level 1 was the lowest peak. Significant differences were observed among the cumulative amounts of fenitrothion or thiobencarb collected from different depth levels. The pesticide residues in the leachates increase with the depth of soil profile increase. The cumulative amounts of the two tested pesticides were compatible with the concentration of treatments, and were higher in high-treatment (50 µg/g soil) compared with that in low-treatment (25 µg/g soil). Our results obtained leaching of thiobencarb was slightly higher than the leaching of fenitrothion. These results are useful in understanding the movement of pesticides and agrochemicals in the agricultural environment.

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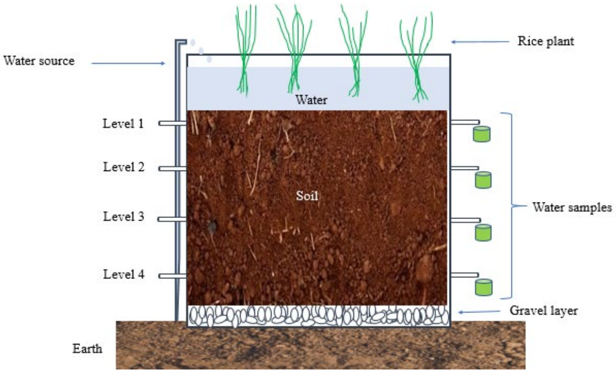

Graphical Abstract

1. Introduction

The controlling pesticide runoff from paddy fields was developed by using outdoor lysimeters.[1] Directive 91/414 of the European Union stressed the use of lysimeters for registration of crop protection products.[2] In Japan, the new registration guidelines for rice pesticides have stipulated the use of paddy lysimeters to predict environmental pesticides concentration. However, lysimeters of undisturbed soil were costly due to their larger size.[3] Thus, a lysimeters system that is easy to set

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* Corresponding author
E-mail address mohammed.riad@alexu.edu.eg (M. R. Fouad)

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up and can provide reliable data is of very interest. The intensive use of pesticides in rice fields has been responsible for making paddy fields a significant contributor for pollution by non-point sources. Fate and transport of agrochemicals in paddy fields were monitored through many studies. Nevertheless, observations of the transport and fate of these chemicals under field conditions are usually laborious, expensive, and time consuming.[2,4-6] In many lysimeter studies, lysimeters have been proven to be effective tools in predicting, simulating flow, assessing and solute transport in the soil profile.[7]

Lysimeter experiments were used to study leaching models of pesticides according to the Working Group Modeling of the European Program (Fate of Pesticides in the Soil and the Environment).[2] The dataset is the obtained result of a three-year lysimeter experiment carried out near Rome, Italy on a clay loam calcareous fluvisol, to assess the risk of groundwater contamination from commonly used herbicides in the Mediterranean area.[2] The potential of a pesticide to contaminate soil and groundwater a stepwise testing scheme has been developed involving laboratory experiments, outdoor experiments such as fate studies using undisturbed lysimeters, mathematical model calculations and field experiments.[8] The German Authorities passed a test guideline in 1990, for the performance of lysimeter experiments. Therefore, some recent experiences included focussing on the topics of; comparative fate assessment based on laboratory tests, mathematical model calculations and lysimeter experiments. Indoor and outdoor lysimeter tests are carried out to compare influence of water regime and climate on the pesticide leaching and influence of soil properties on the leaching process.[9] A set of packed micro-paddy lysimeters, located in a greenhouse, was used to simulate the dissipation of thiobencarb and simetryn. Data from a field study were used in the simulation of dissipation. In the intermittent water irrigation management scenario, the patterns of the dissipation in the surface water of the field followed the first-order kinetic.[10-11] In the continuous water irrigation management scenario, similarity was recorded in concentrations of lysimeter and field. Disappearance curves of simetryn and thiobencarb in the surface water of the two simulated irrigation scenarios were not significantly different from the field results.[11] The highest concentrations of the two herbicides were found on the topsoil layer (0–2.5 cm) depth. Only a small amount of compounds moved down to the deeper soil layers. Thus, micro-paddy lysimeters are a good alternative for the pesticide dissipation study in the paddy environment.[11]

Lysimeters are valuable to study the transport and fate of agrochemicals in soil. Large-scale field lysimeters are used to investigate radionuclide transport as well as pesticide behaviour under natural field conditions better than laboratory soil columns. Also, usually field lysimeters are characterized by a free-draining lower boundary.[12] Usually field experiments are labor intensive and expensive; thus, alternative methods to investigate the fate of pesticides in rice paddies is required. The micro-paddy lysimeter has proved to be a useful tool to assess the transport and fate of pesticides in the rice paddy system. Micro-paddy lysimeters can perform multiple experiments per year and was able to simulate the fate of pesticides, yielding good agreement with the data in an actual field experiment.[11,13]

The water contamination potential of rice pesticides is of great concern to researchers in particular environmentalists. Many studies have pointed out that pesticide runoff from paddy fields is responsible for rivers contamination in Japan.[14-15] While most of the detected pesticides was the herbicide group, the presence of other groups such as insecticides or fungicide illustrated that their potential risk cannot be neglected.[14,16] The micro-paddy lysimeter was developed to simulate solute movement under laboratory conditions. Two plow soil types; sieved and unsieved soils were compared. To ponding water, an inert tracer was used with controlled boundary conditions to evaluate the soil hydraulic characteristics reproducibility. It was proved that micro-paddy lysimeters can use a useful tool to simulate transport of solute in a paddy environment.[13] The micro-paddy lysimeter can conduct multiple experiments per year since it is independent from natural conditions such as location and weather. Also, the preparation and experimental procedure of micro-paddy lysimeter are inexpensive, quick, and less laborious compared to that of field experiments.[2]

A method to construct the micro-paddy lysimeter to simulate the transport and fate of solute in a paddy field was developed. The micro-paddy lysimeter was proven to be an effective tool to predict and simulate the transport and fate of solutes in a paddy environment. The micro-paddy lysimeter has similar properties of hydraulic soil profile with the paddy field such as, size distribution, soil bulk densities, percolation rate and hydraulic properties. A micro-paddy lysimeter was used to monitor the behaviour of imidacloprid granules in the rice paddy system using two treatment methods (at sowing and before transplanting). Tested application rates were 3-folds the field recommended rate. Under a water management scenario like an actual field, the behaviour of the insecticide in micro-paddy lysimeters (paddy water and paddy soil) was comparable with field data.[17-18]

In general, the lysimeter systems have several advantages compared with other methods because they almost exactly reproduce the field environment and agricultural practice, they are easier to monitor than field experiments. But their main disadvantages are expense.[19-21] In recent years, the lysimeter technique was used to determine the behaviour of specific pollutants and their risk assessment. Therefore, it was used to study the leaching of different pesticides such as chlorantraniliprole and bispyribac-sodium,[22] flufenacet,[23] acetochlor, alachlor, S-metolachlor, butachlor and metalaxyl,[24] herbicide dichlobenil,[25] alachlor, ethoprophos, carbofuran, napropamide, tebuconazole, and etridiazole,[26] Glyphosate,[27] Metribuzin,[28] new insecticide cyantraniliprole,[29-30] bentazone, terbuthylazine and S-metolachlor,[31] fenitrothion and thiobencarb.[32]

2. Materials and methods

2.1. Tested pesticides

Fenitrothion

UPAC name: O, O-dimethyl-O-4-nitro-m-tolyl phosphorothioate. Available formulations: EC 50%. Chemical structure: **Fig. 1**. Production Company: DuPont, Kafr el zayat pesticide (KZ). Usage: For controlling chewing and sucking insects on rice, orchard fruits, vegetables, cereals, cotton, and forest. Also fly, mosquito, and cockroach control in public health programs.[32,33]

Thiobencarb

UPAC name: S-4-chlorobenzyl diethyl thiocarbamate. Available formulations: EC 50%. Chemical structure: **Fig. 1**. Production Company: Mitsuichemical Agro. Inc., AGROCHEM. Usage: It is a pre-emergence and early post-emergence herbicide for weed control in rice paddy fields and other situations.[32,33]

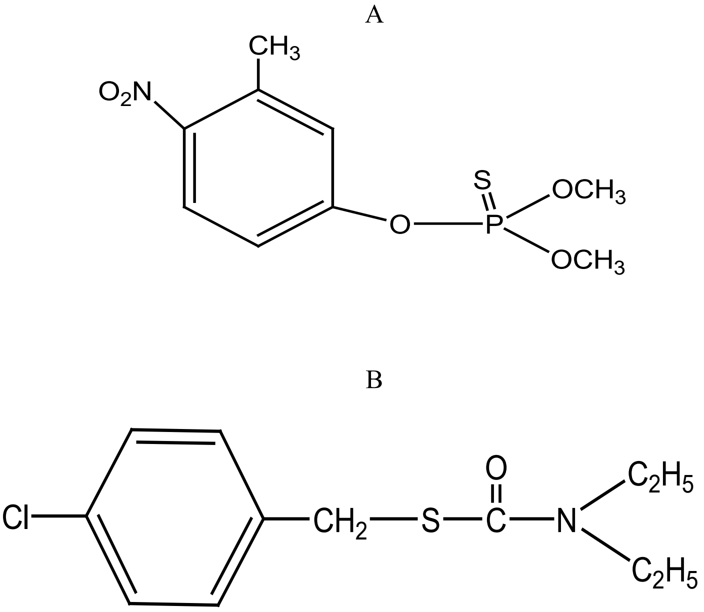


Fig. 1. Chemical structures of fenitrothion (A) and thiobencarb (B).

2.2. Tested plant

Rice plant (*Oryza sativa*, variety Giza 101) was cultivated in the field lysimeter (500 g seeds/lysimeter) to study the leaching and transport of fenitrothion and thiobencarb under field conditions.[32]

2.3. Pesticide determination by UV-Spectrophotometer

Fenitrothion and thiobencarb residues in aqueous samples of leaching experiments were quantitatively analyzed by UV-VIS Spectrophotometer (Thermo Corporation, Nicolet, evolution 100, Germany). The leachates were filtered, then measured at the appropriate λ_{max} for fenitrothion (λ_{max} = 266 nm) and thiobencarb (λ_{max} = 233 nm). The concentrations of the pesticides were calculated using k value for each tested pesticide.[32]

2.4. Field lysimeter

The field lysimeter experiments were conducted in the Agricultural research station, Abis farm of the Faculty of Agriculture, Soil and Water Sciences Dept., University of Alexandria. The lysimeters are built above the ground with bricks, 1.1 m length × 1.1 m width x 2.5 m depth. A coarse aggregate layer of 50 cm thickness was placed, then the lysimeter was filled with calcareous soil from the Bangar Elsokar area, and a network of 4 cm diameter PVC pipes were connected with it to drain the leachate's water at different depths every 30 cm. After plowing the soil of the lysimeter and leveling, the rice plant was cultivated at a rate of 500 g seeds per lysimeter. Three irrigation times were carried out in the first week, the first was on the day of applying the pesticides and planting (50 L of water), the second was on 3rd day of planting (10 L), and the third at 5th day after planting (5 L per lysimeter). The experiment lasted for 12 weeks, the irrigation was conducted twice weekly, the total amount of irrigation water every week was 50 L for each lysimeter. The experiment was conducted from May-August 2020.[34] Before cultivation immediately, the tested pesticides as formulations; fenitrothion and thiobencarb were applied to cover the soil surface (two concentrations that are equivalent to 25 and 50 µg/g soil). Good agriculture practice was applied through the season. The leachate samples at different depths (1 L for each) were collected and analyzed to detect the leaching of tested pesticides in paddy environment.[17,34]

3. Results and discussion

Ten lysimeters located in the open field were used in this experiment; Lys 1 and Lys 2 were treated with 25 mL fenitrothion EC 50% (equivalent to 25 µg/g soil), Lys 3 and Lys 4 were treated with 50 mL fenitrothion EC 50% (equivalent to 50 µg/g soil), Lys 5 and Lys 6 were treated with 25 mL thiobencarb EC 50% (equivalent to 25 µg/g soil), Lys 7 and Lys 8 were treated with 50 mL thiobencarb EC 50% (equivalent to 50 µg/g soil), and two lysimeters were used as control. Mean of the two lysimeters in the same treatment was calculated and presented in **Figs. (2-5)**. In addition, iodide has been used as a water tracer at a rate of 100 mL KI 1M for each lysimeter. The field lysimeters connected with a network of 4 cm diameter PVC pipes to drain the leachate water at different depths every 30 cm (four levels, level 1 at a depth of 30 cm from the soil surface). The lysimeters were cultivated by rice plants. The experiment lasted for 12 weeks, the irrigation was conducted twice weekly, the total amount of irrigation water every week was 50 L for each lysimeter. The experiment was conducted from May-August 2020.[34] The leachate samples at different levels (1 L for each) were collected and analyzed.[17]

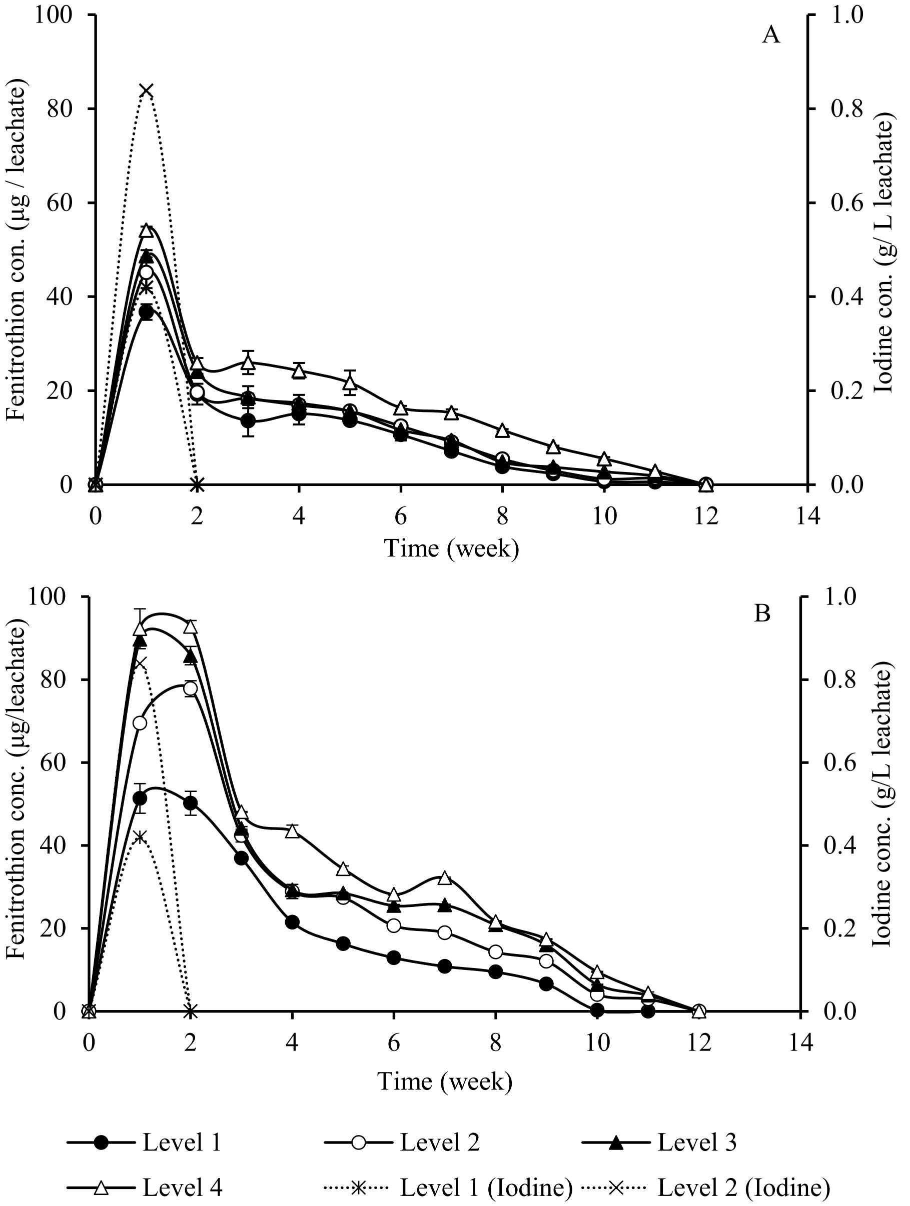


Fig. 2. Breakthrough curves of fenitrothion and water tracer I⁻ using filed lysimeters. A: Treatment of 25 µg/g soil, B: Treatment of 50 µg/g soil.

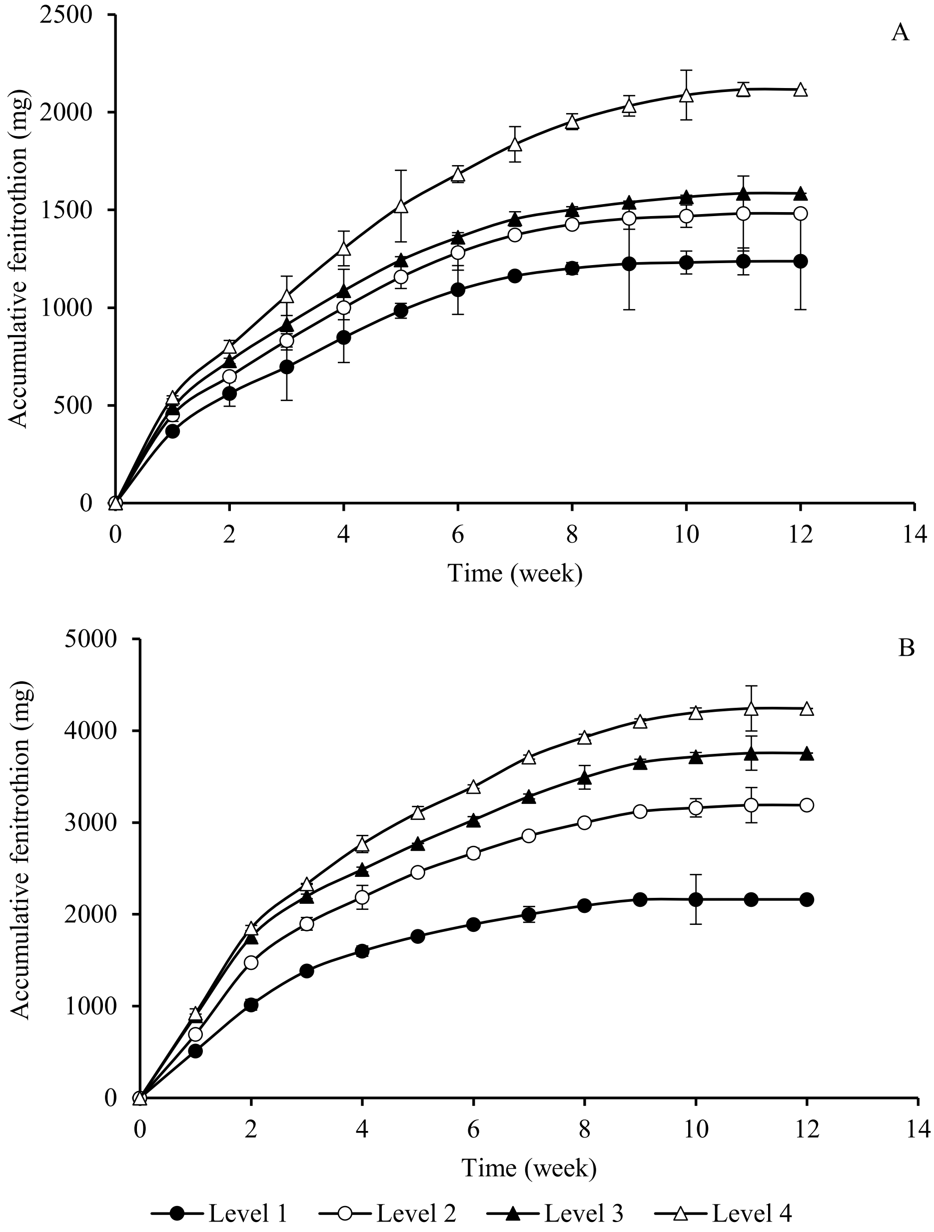
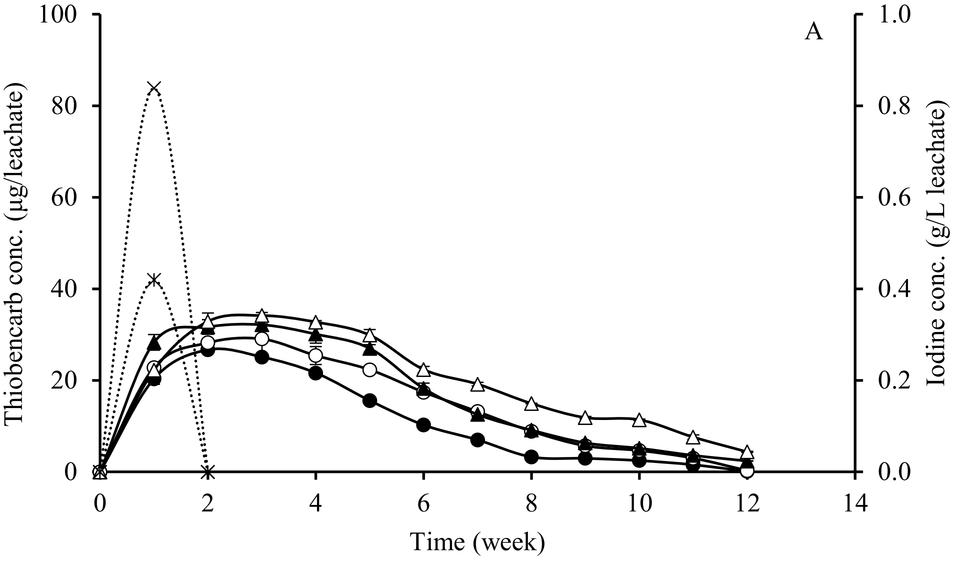


Fig. 3. Cumulative leachate curves of fenitrothion (±SE) using filed lysimeters. A: Treatment of 25 µg/g soil, B: Treatment of 50 µg/g soil.



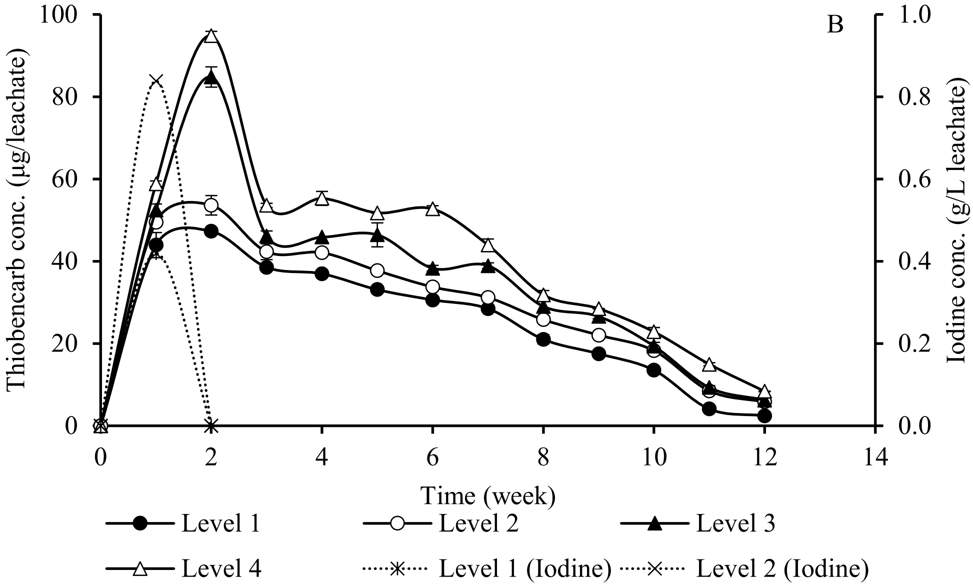


Fig. 4. Breakthrough curves of thiobencarb and water tracer I⁻ using filed lysimeters. A: Treatment of 25 µg/g soil, B: Treatment of 50 µg/g soil.

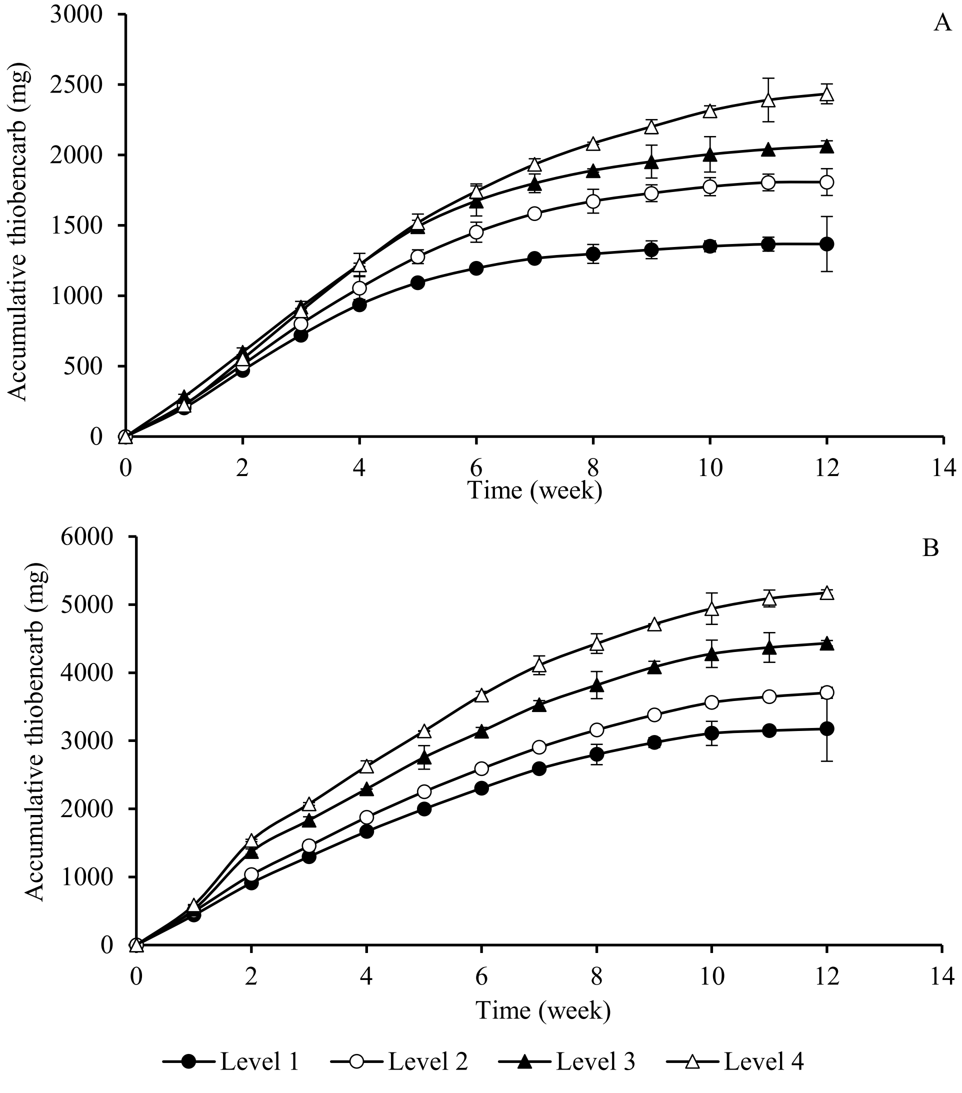


Fig. 5. Cumulative leachate curves of thiobencarb (±SE) using filed lysimeters. A: Treatment of 25 µg/g soil, B: Treatment of 50 µg/g soil.

Regarding Lys 1-4, the bromide amount was recovered in the leachates during the first two weeks. The leaching of fenitrothion was started at the same time with the release of iodide. Iodide was detected in the leachates collected at level 1 and 2 only, producing symmetrical peaks, the concentration of iodide collected from level 2 was more than those collected from level 1. The concentrations of bromide detected at level 3 and 4 were negligible. The top of the fenitrothion-breakthrough curves for all levels appeared during the first two weeks but their tails delayed to about 10 weeks. The highest breakthrough was corresponding to level 4 followed by level 3 while the breakthrough of level 1 was the lowest peak. The peaks of the breakthrough curves for fenitrothion were obtained at percolation of 2nd week corresponding to the maximum concentrations of 55, 52, 50 and 37 µg/L for low concentration-treatment (**Fig. 2A**) and 98, 95, 78 and 55 µg/L for high concentration-treatment (**Fig. 2B**) at levels 4, 3, 2 and 1, respectively. Moreover, the significant differences were observed among the cumulative amounts of fenitrothion collected from different depth levels. While no statistical differences between the lysimeters were observed neither in relation to the occurrence of the bromide peak or the amount of leached pesticides.[35] It can be seen in **Fig. (3)** the cumulative amount of fenitrothion recovered in leachates at each level was more about twice in the case of Lys 7 and 8 that treated with high concentration of fenitrothion than that leachate from Lys 5 and 6 that treated with low concentration of fenitrothion. The cumulative amounts of fenitrothion for leachates in low concentration-treatment contained 1200, 1500, 1600 and 2100 mg while the leachates in high concentration-treatment contained 2200, 3200, 3700 and 4200 mg from 1st, 2nd, 3rd and 4th level, respectively. However, the percentages of cumulative leachates of low concentration-treatment and high concentration-treatment were 51.4 and 53.4%, respectively (**Table 1 and Fig. 6**). The cumulative amounts of fenitrothion recovered from lowest level (level 4) that equivalent of 33.0 and 31.8% followed by the amount of level 3 (24.7 and 28.1%) were more than the amounts released from level 2 (23.1 and 23.9%) followed by the amount released from level 1 (19.3 and 16.2%) for low concentration-treatment and high concentration-treatment. Many studies for other pesticides have shown a statistical difference between the chemically extractable amount, residual pesticide amount and the bioavailable amount.[36-37]

Concerning thiobencarb, the symmetrical shape of bromide-breakthrough curves was obtained within about three weeks. The breakthrough curves of thiobencarb were flatter with some tailing. The maximum top of the peaks was at 31 and 47 µg/L for level 1, 32 and 53 µg/L for level 2, 33 and 85 µg/L for level 3, and 34 and 95 µg/L for level 4 from low concentration-treatment (**Fig. 4A**) and high concentration-treatment (**Fig. 4B**), respectively. In addition, statistical differences were detected among the cumulative amounts of thiobencarb collected from different levels, 1st to 4th for low concentration-treatment 1400, 1800, 2000 and 2400 mg (**Fig. 5A**) and that for high concentration-treatment 3200, 3700, 4400 and 5200 mg (**Fig. 5B**), respectively. Data in **Table (1)** and **Fig. (6)** indicated that the percentages of thiobencarb in cumulative leachates for low concentration-treatment and high concentration-treatment were 61.4 and 65.9%, respectively. The cumulative amounts of the pesticide recovered from each level for the two treatments were almost the same. According to the Danish legislation a pesticide or its metabolite for which lysimeter studies must not be detected in the leachate in a concentration per year above 0.1 mg/L.[35]

It was observed that the cumulative amounts of the two tested pesticides were compatible with the concentration of treatments, and were higher in the treatment (50 µg/ g soil) compared with that in treatment (25 µg/g soil). The residues of the two tested pesticides in the leachates increase with the depth of soil profile increase. The leaching of thiobencarb was slightly higher than the leaching of fenitrothion. This result agrees with the previous results obtained from adsorption-desorption exerimentand leaching using soil columns and laboratory lysimeters for both tested pesticides, fenitrothion and thiobencarb.[32,34]

Table 1. Cumulative leachates (%) of tested pesticides and its distribution in different depth levels of field lysimeters

Parameters	Fenitrothion		Thiobencarb	
	Low-treatment*	High-treatment**	Low-treatment	High-treatment
Cumulative (%)	51.4	53.4	61.4	65.9
Level 1	9.9	8.7	10.9	12.7
Level 2	11.9	12.8	14.5	14.8
Level 3	12.7	15.0	16.5	17.7
Level 4	17.0	17.0	19.5	20.7

* Treatment of 25 µg/g soil, ** Treatment of 50 µg/g soil

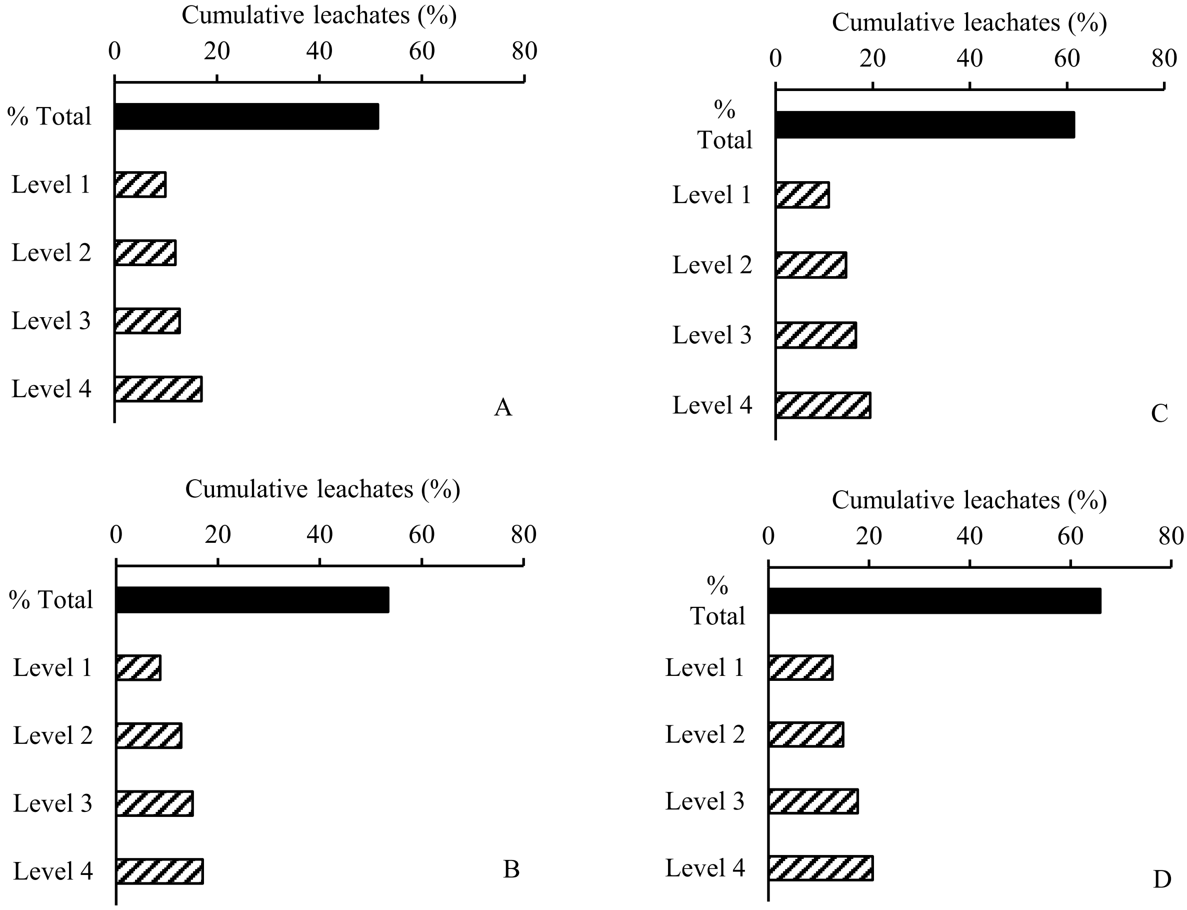


Fig. 6. Cumulative leachates (%) of tested pesticides and its distribution in different depth levels of field lysimeters. A: Treatment of 25 µg/g soil for fenitrothion, B: Treatment of 50 µg/g soil for fenitrothion, C: Treatment of 25 µg/g soil for thiobencarb, D: Treatment of 50 µg/g soil for thiobencarb.

4. Conclusion

The leachates collected at levels 1 and 2 were the only ones that included iodide, and the level 2 concentration was higher than the level 1 concentration. The highest concentration of fenitrothion and thiobencarb was obtained at level 4 followed by level 3 while the lowest concentration was at level 1. Significant differences were observed among the cumulative amounts collected from the different levels. The cumulative amounts of fenitrothion and thiobencarb were higher in high-treatment (50 µg/g soil) compared with that in low-treatment (25 µg/g soil).

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