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Research Paper

Comparison of fate and transport of atrazine in corn and rubber plantation soils

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ABSTRACT

The rubber and corn plantation soils in Nan province were investigated for the fate and transport behavior of the atrazine. The biodegradation, adsorption, photodegradation, and infiltration of atrazine in both soils were conducted. The kinetic rates of each experiment were calculated and reported. Results illustrated that the microbials in the soils have a high tendency in consuming the atrazine in both corn and rubber soils. The corn soils with the biodegradation rate as 0.2523 Kg/mg-min had higher kinetics rate than rubber soil with 0.1357 Kg/mg-min. Langmuir isotherm best described the adsorption of atrazine in both corn and rubber soils. The sorption capacity of atrazine adsorption on the rubber soil (1/qm = 9.7095) was much higher than the corn soil (1/qm = 0.5195). The amount of herbicide residue bonded on soil after application in the rubber soil was slightly higher than the corn soil, and the photolytic rate in the rubber soil is much higher than that in the corn soil. From the mass balance, atrazine in water was detected in the higher amount than in the soil for both corn and rubber soils. The infiltration rate in corn soil was 13.1 mm/hr while that in rubber soil was 11.8 mm/hr. The mass of atrazine in the leachate is in the range of 32.1-75.3% in both corn and rubber soils during 36 hours of infiltration. This experiment is the scientific evidence supporting that the contamination of atrazine in groundwater has high tendency to occur in the extreme application of the atrazine to the agriculture soil.

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1. Introduction

Nan is the province in the north of Thailand. The most people is the farmer. The areas of agriculture are the slope. The most of the plantation is corn and rubber. The product of agriculture in Nan is 319 million kilograms, in which the corn is 95 million kilogram, and rubber is 16 million kilograms. The farmers used chemicals in farming by chemicals imported 1274.1 tons per year, some which are chemicals used herbicide up to 1172.7 tons.

Atrazine (2-chloro-4-ethylaminuteso-6-isopropylamine -1,3, 5-triazine) has height stability in soil but ubiquitous and unpreventable water contamination. It is easily leached out; thus, it can be contaminutesated with underground water. Despite being banned in most European countries, atrazine is the most widely used herbicide in the United States and is registered in more than 70 countries worldwide. It is also widely used in Thailand.

This study is focused behavior of atrazine fate and transport, biodegradation, adsorption, photo-degradation and infiltration in corn and rubber plantation.

2. Materials and methods

2.1 Experimental site

The experimental site was situated in Du-Phong Sub-District, Sunti-Suk District, Nan province, Thailand. The soil was collected from corn and rubber plantations in the mountaineouss area.

Fig. 1. Study area at Du-Pong Sub-District, Santi-Suk District, Nan province, Thailand. (Data economics of plantation in Nan province, 2015).

2.2 Soil samples

Soil physico-chemical properties at each site are given in Tab. 1. Soil textures from both corn and rubber sites are classified as clay according to US Soil Taxonomy. (USDA., 2014) All soil samples were collected from a deep soil profile 0–65 cm layer and homogenized. Further, the soil samples were air-dried and sieved to a particle size ≤ 2 mm to remove stones, roots and other plant materials, then placed in polyethylene bags and stored at room temperature.

2.3 Chemicals

Atrazine powder with purity 90% weight was purchased from Syngenta group company in Thailand. Atrazine(6-chloro-N-ethyl-N´-(1-methylethyl)-1,3,5,triazine -2,4-diaminutese), calcium chloride ($CaCl_2$), sodium sulfate anhydrous (Na_2SO_4) were lab grade and obtained from Fisher Chemical. Hexane (C_6H_{14}) was analytical grade and obtained from RCI Labscan. All chemicals were used as received.

2.4 Method experimental

2.4.1 Biodegradation experimental

For both corn and rubber soils, six containers were prepared with 1550 g (sieved ≤ 2 mm) soil for various concentrations of atrazine. The 100 mL of atrazine solutions were mixed up into the soil in five various concentrations 10, 20, 40, 80, and 100 ppm for atrazine, respectively. For the control experiment, 100 mL sterile water were mixed up with the soils. Additional 350 mL distilled water was added to the corn and rubber soils to obtain a soil moisture to about 20 – 40 % of the water holding capacity. The room temperature was controlled at 25°C.

The corn and rubber soil samples were taken from each container during the time interval. Duplicate samples of 15 g for soil extraction were conducted. In each sample, 40 mL of hexanes was added and mixed by the ultrasonic processor following the method 3550 from US. EPA. The 13 g of sodium sulfate anhydrous and the additional of 10 mL of hexanes were added to the previous extracted solutions and mixed by the ultrasonic processor. Then, the aliquots were poured into 250 mL Kuderna-Danish Concentrator (US. EPA., 2007) to expel the solvent during the extraction. The samples after Kuderna-Danish were measured with UV spectrometer at 229 nm (US.EPA.,1996). Approximately 1 - 5 g of soil were analyzed for moisture content and another 1 g soil for bacteria count.

For bacteria counting, 1 g of soil from each concentration was put into a test tube containing 9 mL of sterile deionized water. The dilution of soil solutions was performed with sterile deionized water. Further samples were diluted and pipetted into a petri dish.

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Table1. Physical-chemical characteristics.

Plan station	Clay (%)	Sand (%)	Silt (%)	Texture	BD (g/cm^3)	N (%)	K (ppm)	P (ppm)	Ca (%)	CEC (%)	OM (%)	pH
Corn	70.2	8.8	20.8	clay	1.15	0.10	17.6	11.9	0.004	35.4	9.79	5.4
Rubber	83.1	3.0	13.8	clay	1.19	0.11	22.8	6.4	0.004	26.7	4.25	4.3

***Remark:** BD = Bulk density
K = Potassium
Ca = Calcium,
OM = Organic matter
N = Nitrogen
P = Phosphorus
CEC = Cation exchange capacity

2.4.2 Adsorption experimental

The adsorption of atrazine in corn and rubber soil was studied by batch experiment technique. The 0.5 g sieved soil samples were shaken with 11.25 mL of 0.01 M $CaCl_2$ for 12 hours. The $CaCl_2$ solution is used as the aqueous solvent phase to improve centrifugation and minimize cation exchange. The 1.25 mL of the stock solution of atrazine was added to adjust the final volume to 12.5 mL. The bottles with the soil solution mixtures were shaken for 0-30 minutes at 120 rpm. After agitation, the mixtures were centrifuged and the supernatant solutions were analyzed by UV spectrometer at 229 nm (US.EPA.,1996). The results of the spectrophotometer are used to create isotherms of atrazine sorption for the corn and rubber soils.

2.4.3 Photodegradation experimental

This experiment set was conducted under the strong radiation of the sun at Naresuan University (16.7483° N, 100.1923° E) from 1 pm to 3 pm. The aluminum tray (40 x 40 x 4 cm) containing the 1,200 gram of atrazine contaminated soils were used for this experiment. The 300 mL with 50-4,000 mg/L of atrazine solution was poured into the soils and mixed homogeneously. The tray was covered with polyethylene film to prevent the volatilization of atrazine and to let the sunlight passing through the film and get into the contaminated soils. For sample analysis, 15 g of contaminated soil samples were mixed with four teaspoons of sodium sulfate anhydrous. The 40 mL of hexane was added before extracting by the ultrasonic processor. The 13 g of sodium sulfate anhydrous and the additional of 10 mL of hexanes were added to the previous extracted solutions and mixed by the ultrasonic processor. Then, the aliquots were poured into 250 mL Kuderna-Danish Concentrator to expel the solvent during the extraction. The samples after Kuderna-Danish were measured with UV spectrometer at 229 nm (US.EPA.,1996).

2.4.4 Infiltration experimental

The corn and rubber plantation soils were sieved through less than 2 mm screen and dry at the room temperature. The soils were packed into the body of the columns with an inner diameter of approximately 2.6 cm and 50 cm long. The soil depth in the column was 40 cm. The stock solution of 1,000 mg/L atrazine solutions were prepared in DI water. The 10 mL of the solution was added onto the surface of the soil in the column. The 0.01 M of the calcium chloride was dropped to the column with the rate 18 mL/hr for 24 hours. The liquid samples that pass through the soil layer were analyzed during 36 hours time interval. At the end of the experiment, every 5 cm depth of the soil layers were extracted to determine the atrazine residue in the soil layers. Then, the extracted solutions were measured by UV spectrometer at 229 nm (US.EPA.,1996).

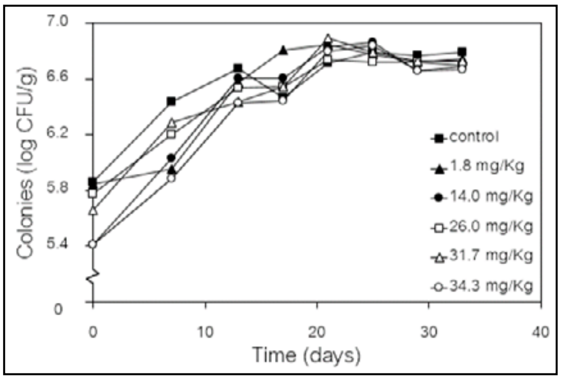
3. Results

3.1 Biodegradation experimental

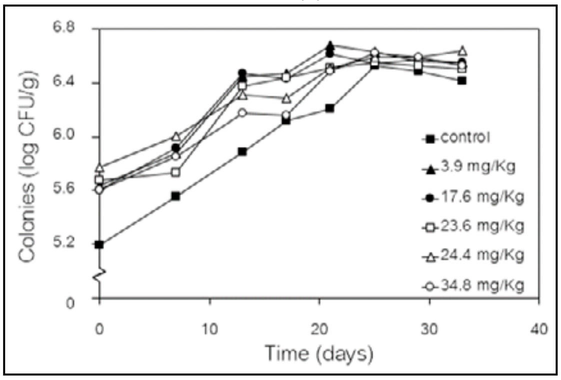
To enumerate aerobic microorganisms, the dilution plate method was commonly used. The results were calculated and plotted by logarithmic transformation and shown in CFU (colony forming unit) colonies per gram. **Figs 2a and 2b** show the colonies grown during 33 days in corn and rubber soils. In the time course study, it was found that there was a visible growth of the microorganisms in contaminated soil experiments and the control experiment.

In the rubber soil, the higher growth of bacteria in atrazine contaminutesated soil was predominant than the control soil. The growth of colonies seems to be stable in corn and rubber soil after 21 days.

In **Figs 3a and 3b** depict the concentration of atrazine in the soil during the biodegradation experiments. Results show that the atrazine concentration substantially decreased within time. **Fig. 4** exhibited the kinetics of the biodegradation of atrazine of corn and rubber soil. The biodegradation rate increase with increasing residual of atrazine concentration. The kinetic rate constants for corn and rubber soil were 0.2523 and 0.1357 Kg/mg-min, respectively.

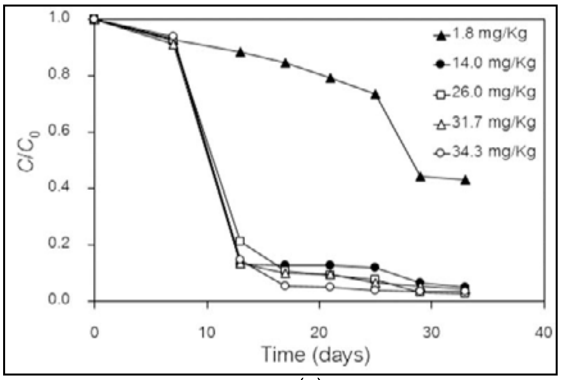


(a)

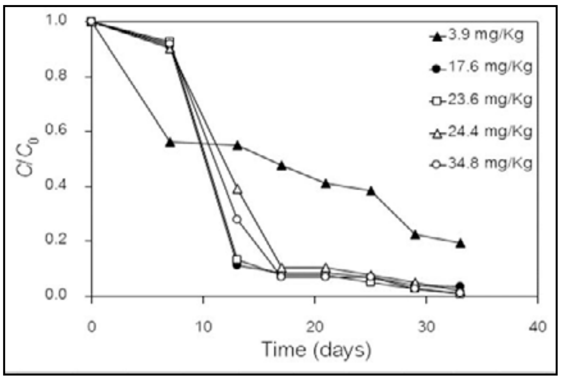


(b)

Fig. 2. Bacteria colonies log CFU g^{-1} in (a) corn and (b) rubber soils.



(a)



(b)

Fig. 3. Atrazine residue C/C_0 on (a) corn and (b) rubber soil.

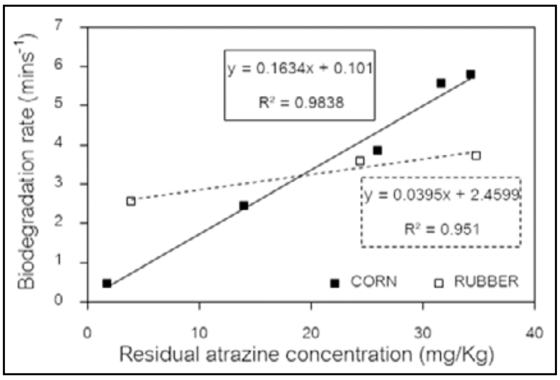


Fig. 4. Kinetics of the biodegradation of atrazine in corn and rubber soils.

3.2 Adsorption experimental

Figs. 5 and 6 show the batch experimental equilibrium data for the atrazine sorption in mg/Kg on corn and rubber soil within 30 minutes. At the certain adsorption times, the equilibrium between the solid-phase concentration and the residual liquid-phase concentration was reached and stayed virtually constant for each concentration. The higher initial concentrations of atrazine led to the higher adsorbed values of atrazine on corn and rubber soil. At 875 mg/kg the adsorbed atrazine in the corn soil is much higher (108.11 mg/kg) than that in the rubber soil (54.79 mg/kg). The maximum adsorption (q_{max}) of atrazine was shown narrow values after 30 minutes on corn soil (q_{max} for 125 mg/kg = 16.86 mg/kg, q_{max} for 250 mg/kg = 26.98 mg/kg, q_{max} for 375 mg/kg = 42.29 mg/kg) and on rubber soil (q_{max} for 125 mg/kg = 20.49 mg/kg, q_{max} for 250 mg/kg = 33.91 mg/kg, q_{max} for 375 mg/kg = 49.46 mg/kg).

For rubber soil, the equilibrium time was longer for the higher contaminated soil. For example, the 25 minutes were required for the high concentration of 375-1000 mg/kg of atrazine in soils, while the 15 minutes were required for the 250 mg/kg of atrazine.

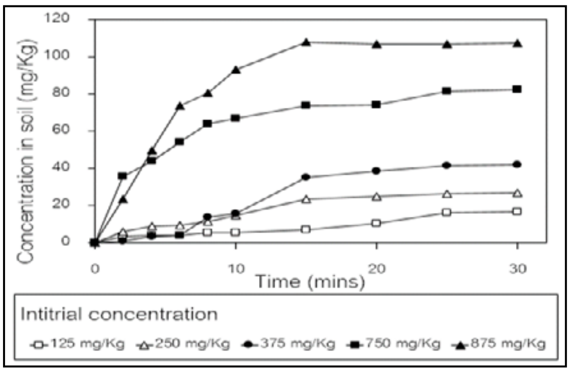


Fig. 5. Atrazine sorption mg/Kg on corn soil.

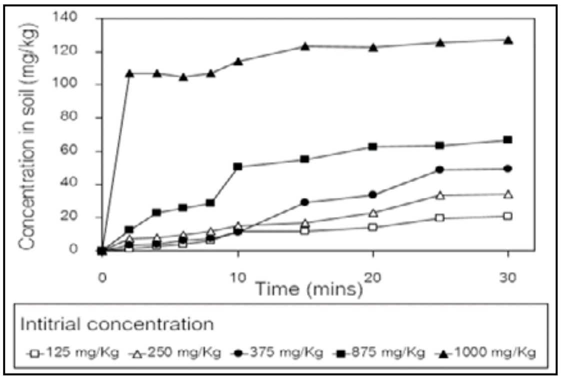


Fig. 6. Atrazine sorption mg/Kg on rubber soil.

The logarithmic transformation of the Freundlich isotherms of atrazine sorption on corn and rubber soils is shown in **Fig 7a and 7b,** respectively. The Langmuir-I isotherms for atrazine sorption on corn and rubber soils are illustrated in **Fig 8a and 8b,** respectively.

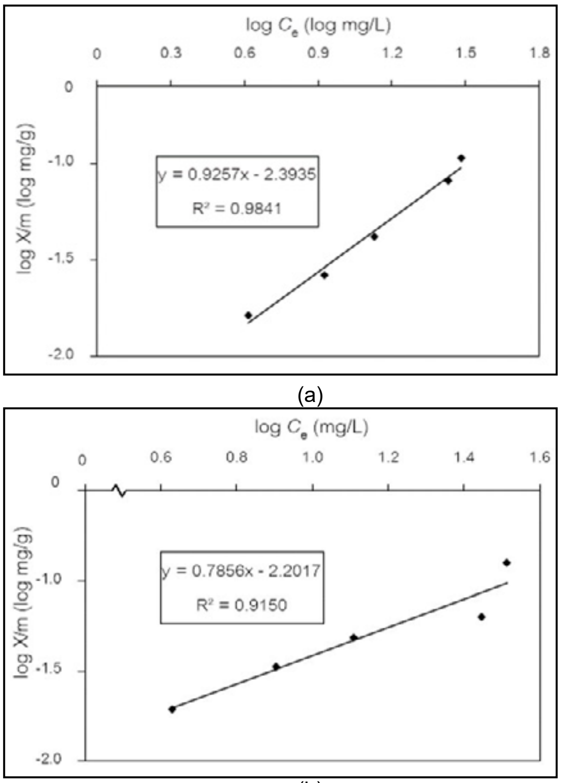


Fig. 7. Freundlich Linear Isotherm of atrazine in (a) corn and (b) rubber soils.

Both soils exerted a high correlation between the adsorbed concentration on the soil and the adsorbed concentration in the solution, and this confirms the best fit to the Langmuir isotherm rather than the Freundlich Linear isotherm. Table 2 shows the linearized expression of Freundlich isotherms of atrazine sorption with the Freundlich parameters; determination coefficient r^2, Freundlich constant K_F, and Freundlich exponent 1/n.

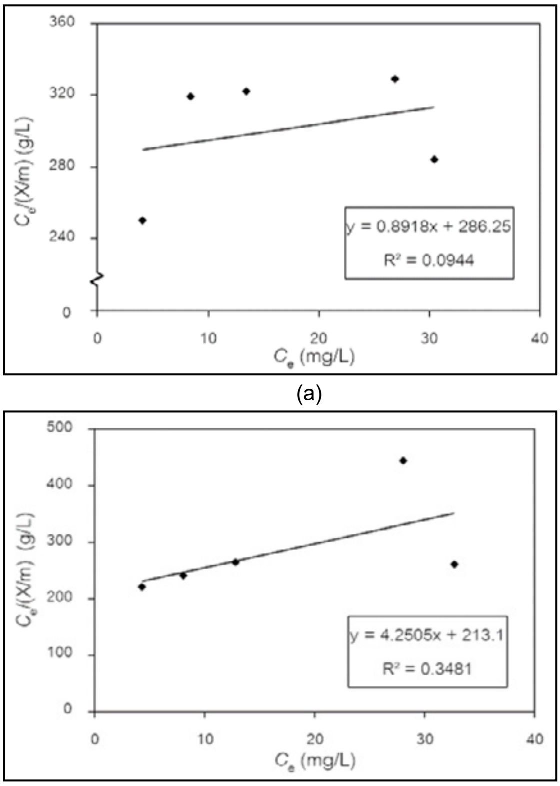


Fig. 8. Langmuir isotherm of atrazine on (a) corn and (b) rubber soils.

The Freundlich exponent 1/n is for corn soil = 0.9257 and rubber soil = 0.9257. Further, the r^2 values are listed: corn soil (0.9841) and rubber soil (0.9841). This result indicates a high correlation between the adsorbed concentration in the soil and the adsorbed concentration in the solution. Table 3 shows the calculated Langmuir isotherm parameters $1/q_m$, $1/K_a{*}q_m$ and r^2. The determination coefficient r^2 for corn soil = 0.0944 and for rubber soil = 0.0944, a high correlation is observed. For $1/q_m$ in corn soil = 0.8918 and in rubber soil = 4.2505, whereby q_m described the sorption capacity of atrazine sorption on the soil.

3.3 Photodegradation experimental

Fig. 9a and 9b show the photodegradation of atrazine in corn and rubber soils, respectively. The residual of atrazine in corn and rubber soil at 25 minutes was quickly decreased. After 30 minutes residual of atrazine was slowly decreased and then stable after 60 minutes.

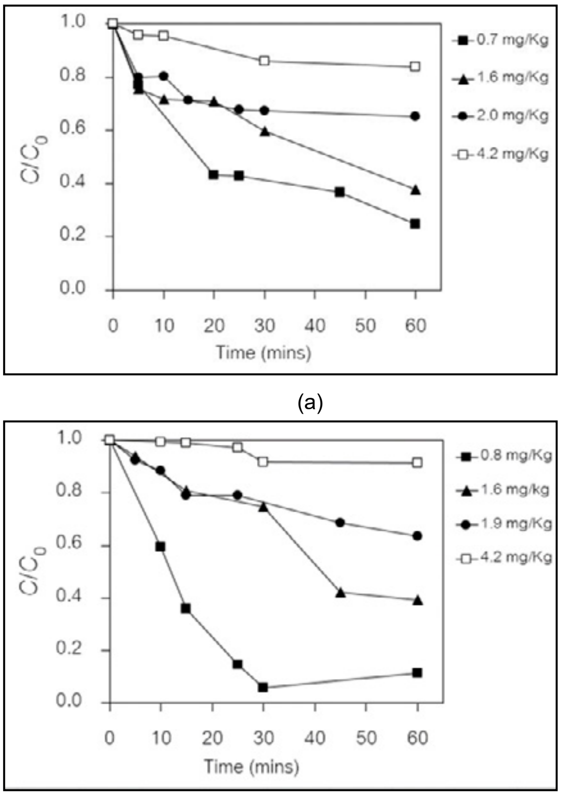
Fig. 10 shows the kinetics of the photodegradation of atrazine of corn and rubber soils. The photodegradation rate decrease with increasing residual of atrazine concentration. The kinetic rate constants for corn and rubber soil were 0.0061 and 0.0281 kg/mg-min, respectively.

Table 2. Freundlich Isotherm equation with r^2, 1/n and K_F for atrazine on corn and rubber soils.

Sample	Equation	Graph equation	r^2	K_F	1/n
Corn	$\log(q_e) = (1/n)\log C_e + \log K_F$	y = 0.9257x - 2.3935	0.9841	0.0040	0.9257
Rubber	$\log(q_e) = (1/n)\log C_e + \log K_F$	y = 0.7856x - 2.2017	0.9150	0.0063	0.7856

Table 3. Langmuir isotherm equation with r^2, 1/qm, and 1/(K_a*qm) of atrazine on corn and rubber soils.

Sample	Equation	Graph equation	r^2	$1/q_m$	$1/K_a*q_m$
Corn	$C_e/q_e = (1/q_m)*C_e + (1/K_a*q_m)$	y = 0.8918x – 286.25	0.0944	0.8918	286.25
Rubber	$C_e/q_e = (1/q_m)*C_e + (1/K_a*q_m)$	y = 4.2505x – 213.10	0.3481	4.2505	213.10



(a)

(b)

Fig. 9. Photodegradation of atrazine in C/C_0 with the different concentration in (a) corn and (b) rubber soil.

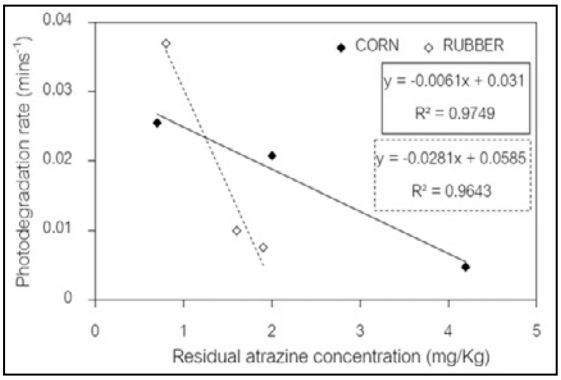


Fig. 10. Kinetics of the photodegradation of atrazine in corn and rubber soil.

3.4 Infiltration experimental

Fig. 11 show the leached atrazine at 10 mg for both corn and rubber soil. Results represented in percent of atrazine leaching in each hour from the initial amount of spraying atrazine. The percentage leached atrazine in corn soil (8.80 %), and rubber soil (12.43%) were showed at 18 hours. Apparently, the percentage of leached atrazine in corn soil was 32.07% which was lower than that of rubber soil (75.33%) during 30 hours experimental period.

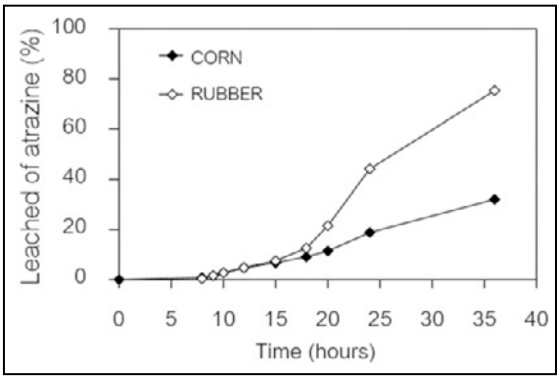


Fig. 11. Leached atrazine in percent each hour in corn and rubber soil at 10 mg.

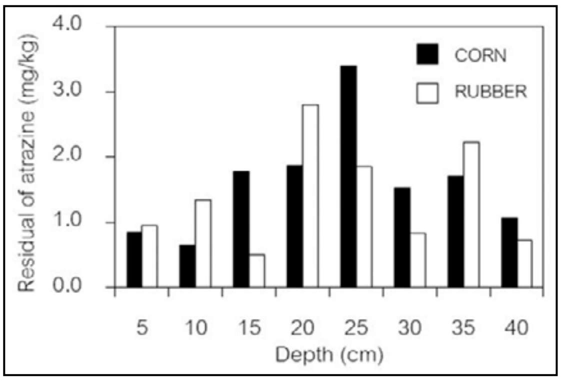


Fig. 12. Atrazine residue in mg/kg with soil depth in cm on corn and rubber soils.

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Table 4. Infiltration rate of atrazine in corn and rubber soil.

Sample	Concentration (mg)	volume (mL)	area (cm^2)	time (hour)	infiltration rate (cm^3/hr)	velocity rate (cm^3/hr)
Corn	10	315.5	4.91	36	8.79	1.79
Rubber	10	415.0	4.91	36	11.53	2.35

Fig. 12 exhibited the atrazine residues in corn and rubber soils in mg/kg. The initial concentration of atrazine in this experiment set was 1,000 mg/kg. Results show the different behaviors of both soils under different layers of the soil depth. The depths of corn and rubber soils in the column for this experiment were 25 and 20 cm, respectively.

Table 4 shows the infiltration rates in cm^3/hr and velocity rate in cm/hr for corn and rubber soils at 1,000 mg/Kg of atrazine. The infiltration rates of atrazine in corn and rubber soils were 8.79 and 11.53 cm^3/hr, respectively. The velocity rates of atrazine in corn and rubber soils were 8.79 and 11.53 cm^3/hr, respectively.

4. Discussion

4.1 Biodegradation discussion

Results illustrated that the microbials in the soils have a high tendency in consuming the atrazine in both corn and rubber soils. The microbials in both soils had a higher amount in colonies than the control experiment (without atrazine). Microbial colonies in corn soil were 6.72 log CFU/g which is slightly higher than that in rubber soil with 6.42 log CFU/g. This finding is in good agreement with the previous works (Neumann, G., et *al*., 2004; Sene, L., et *al*., 2010). Both researches have also detected a microbial degradation of atrazine. It was reported that the pure bacterial culture Pseudomonas sp. strain ADP was able to degrade atrazine as a sole source of nitrogen. In a comparison of atrazine biodegradation in corn and rubber soils, the corn soils with the biodegradation rate as 0.1634 kg/mg-min had higher kinetics rate than rubber soil which had the biodegradation rate as 0.0395 kg/mg-min.

4.2 Adsorption discussion

Langmuir isotherm best described the adsorption of atrazine in both corn and rubber soils. The sorption capacity of atrazine adsorption on the rubber soil ($1/q_m$ = 4.2505) was much higher than the corn soil ($1/q_m$ = 0.8918). Also, the amount of herbicide residue bonded on soil after application in the rubber soil was slightly higher than the corn soil. However, the adsorption of the atrazine can be different by the type of soils. In this work, the adsorption of atrazine in both corn and rubber soils are best described by Langmuir, but Deghani and his group detected the best fit of Freundlich isotherm for atrazine sorption on corn-wheat rotation soils in Iran. (Deghani, M., et *al*., 2005). This difference in the adsorption isotherm may arise from the soil characteristics from different places.

4.3 Photodegradation discussion

Atrazine can be degraded by the sunlight as shown in this work. In comparison, the kinetics rate of atrazine photodegradation in rubber soil was higher than that in corn soil. The kinetic rate constants for photodegradation of atrazine in corn and rubber soils were 0.0061 and 0.0281 kg/mg-min, respectively. The percentage of clay in both soils ply the major role. The clay portion in the soil has high tendency to capture the pollutant in the soil.

Our results were in good agreement with previous work (Zhen, et *al*., 2005). The photolytic rate in thin silt soil is larger than that in the coarser the soil is, the less the solar light penetrates. In our work, the rubber soil has higher in clay percentage than that in the corn soil. Consequently, the photolytic rate in the rubber soil is much higher than that in the corn soil.

4.4 Infiltration discussion

Atrazine was detected in both soils and water during the infiltration process. A high percentage of atrazine was detected in the filtrated water from both corn and rubber soils. From the mass balance, atrazine in water was detected in the higher amount than in the soil for both corn and rubber soils. Although atrazine can bound to organic matter or soil particles and adsorb in the soil (Gevao, B., et *al*., 2000; Paraniape, et *al*., 2004). The high solubility of atrazine causes the leaching of this chemical to the water (Gevao, B., et *al*., 2000; Paraniape, et *al*., 2004). From our work, the infiltration rate of atrazine from soil to water for corn soil was slightly higher than that for rubber soil. The infiltration rate in corn soil was 13.1 mm/hr while that in rubber soil was 11.8 mm/hr. The mass of atrazine in the leachate is in the range of 32.1-75.3% in both corn and rubber soils during 36 hours of infiltration. This experiment is the scientific evidence supporting that the contamination of atrazine in groundwater has high tendency to occur in the extreme application of the atrazine to the agriculture soil.

5. Conclusions

Atrazine can be degraded by the microbial activity, the photodegradation by the sunlight. The residual of atrazine in both corn and rubber soils has high tendency to be adsorbed in monolayer in the soil as best explained by Langmuir isotherm. The sorption capacity of atrazine adsorption on the rubber soil ($1/q_m$ = 9.7095) was much higher than the corn soil ($1/q_m$ = 0.5195). The atrazine to the soil can be leached out into the water during the infiltration process. The infiltration rate in corn soil was 13.1 mm/hr while that in rubber soil was 11.8 mm/hr. The mass of atrazine in the leachate is in the range of 32.1-75.3% in both corn and rubber soils during 36 hours of infiltration.

Acknowledgements

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