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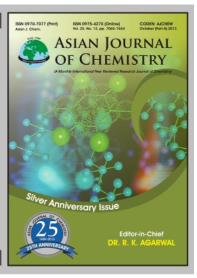

Mobilization and Leaching of Trace Elements (Fe, Al and Mn) in Agricultural Soils as Affected by Simulated Acid Rain

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In this study, the mobilization and leaching of iron, aluminum and manganese were evaluated as influenced by different rates of acid deposition on three agricultural soils (Korat, Pak Chong and Phon Pisai) of Thailand. In laboratory experiments, acidic solutions (pH 3.5, 3.0, 2.5 and 2.0) and unpolluted water (pH 7.0) were applied to soil columns over a period of 45 days. Basic properties of the investigated soil were determined before the experiment, while levels of target metals (Fe, Al and Mn) were determined before and after the experiment. The effluent was collected after every 5 days and analyzed for the target elements. Acid rain caused highly significant (p value < 0.001) leaching of all the investigated metals. Mobilization and leaching of Fe, Al and Mn increased with an increase in the acidity of applied solutions. Highest leaching of Fe occurred in Korat soil under all the applied acidic solutions. Leaching of Al was very high in Pak Chong and Phon Pisai soils. Quite high leaching of Mn was found in all the soils under treatment with a highly acidic solution (pH 2.00). Extractable contents of all the investigated elements changed considerably in all the soils. Behaviour of the target elements depended on soil texture, cation exchange capacity, soil reaction (acidity), sesquioxides, organic matter content, and the sulfate and nitrate adsorption capacity of the soil. Mobility of these elements can cause rhizotoxicity and their leaching by acid rain poses a threat to groundwater quality and environmental contamination.

Key Words: Acid deposition, Aluminum, Iron, Manganese, Leaching, Rhizotoxicity, Tropical soils, Thailand, Southeast Asia.

INTRODUCTION

Acid rain is a broad term which refers to a mixture of wet and dry deposition (deposited material) from the atmosphere containing higher than normal amounts of nitric acid and sulfuric acid. The precursors of acid rain formation result from both natural sources (volcanoes and decaying vegetation) and man-made sources such as primary emissions of sulfur dioxide and nitrogen oxides (NO_x) resulting from fossil fuel combustion[1]. Due to rapid industrialization in Asian countries, emissions of various air pollutants have been increasing annually. Urban air pollution is increasing which is a serious threat to soil system and consequently food security[2]. These increasing developmental activities have led to higher levels of atmospheric pollutants which have severely affected natural ecosystems[3,4].

According to this study acidic subsoil under acid rain treatment (pH 3.5) inhibited above- and below-ground growth of young model forest trees (S. *viminalis*). Acidic treatment (pH 3.5) of soil increased the concentrations of Mn, Pb and Cd in the soil system and decreased the soil pH significantly[5]. Significant reduction in acid buffering capacity of the soils has been observed under nitrogen deposition[6]. Mitani and Ogawa[7] found high leachability of exchangeable cadmium caused by acid rain in activated sludge applied to soil. Soil sorption capacity of heavy metals is related to different soil properties, such as soil particle size, organic matter, base saturation, soil pH, adsorbed SO_4^{2-}, exchangeable Al^{3+} and H^+, and the Al_2O_3 and Fe_2O_3 content. Generally decreased sorption capacity of soil is caused by increased soil acidification. The ground water and crops in contaminated areas are susceptible to heavy metals, especially Zn and Cd, which have high sensitivity to acid rain[8]. Acid rain causes leaching of soil base cations[9,10] and dissolved organic matter from the soils[11] and mobilization of selenium in the soils threatening groundwater quality[12]. High concentrations of toxic elements have been found in the mountainous areas of Thailand[13] and Thai-Laos Mekong River[14]. Acid deposition in such areas can accelerate the mobility of toxic elements. Acid rain has also been considered one of the major issues of transboundary air pollution in the Southeast

Asian countries[15-17]. Effect of simulated acid rain on base cations in highly weathered soils have already been studied by Nawaz *et al.*[18]. The main objective of this paper is to determine the effects of simulated acid rain on the behaviour of Al, Fe and Mn in three different tropical soils of Southeast Asia at different pH levels of acidic deposition.

EXPERIMENTAL

Materials preparation: A laboratory experiment was conducted on three tropical soils (Korat, Pak Chong and Phon Pisai soil series) of the Northeast region of Thailand. Collected soil samples (0-30 cm) were air dried under shade, ground, mixed thoroughly and sieved by 2 mm mesh. The soil samples were analyzed for basic soil properties including soil pH, particle size distribution by the hydrometer method[19], cation exchange capacity (CEC) by 1 N ammonium acetate[20] and soil organic matter by the wet-oxidation method[21]. Plastic cylinders (15 cm × 35 cm) were used to contain 30 cm long soil columns. At the bottom, filtering material was placed on porous plates to filter the leachate. The prepared soil samples were poured into the cylinders. The experiments were carried out on 45 soil columns *i.e.*, 3 soil types × 5 acid loading rates × 3 replications.

Simulation of acid rain in the laboratory: Analytical grade HNO_3 and H_2SO_4 reagents were used to prepare the stock acidic solutions in the laboratory. The working solutions with pH 3.5, 3.0, 2.5 and 2.0 were simulated by diluting the stock acidic solutions with deionized water. In order to represent unpolluted rain, deionized water was used as a control (non-acidic) treatment with pH 7.0. The total volume of applied solution (24.38 L) was calculated by an equation ($V = \pi r^2 h$, where "V" is the total volume of applied solution, "r" is the radius of the soil columns *i.e.*, 75 mm and "h" is the average annual rainfall *i.e.*, 1379.1 mm).

SAR application and analysis of effluent and soil samples: Solutions were applied to the columns through vessels with perforated bottoms for a period of 45 days. Leachate samples were collected for every five days and analyzed with inductively coupled plasma-optical emission spectrometry (ICP-OES). After the leaching experiments, the soil was removed and cut into three equal layers (10 cm). These soil samples were air dried and prepared for extraction of aluminum by 1 N KCl[22] and for extraction of iron and manganese by 0.01 N $CaCl_2$[23] and analysis by ICP-OES. An F-test was performed for the data obtained from the leachate analysis with a confidence level at $\alpha = 0.01$.

RESULTS AND DISCUSSION

The basic soil properties of the selected soils are presented in Table-1. Pak Chong (Pc) and Phon Pisai (Pp) soil series have higher cation exchange capacity due to their clay and organic matter contents as compared to the sandy Korat soil series (Kr). Phon Pisai is slightly alkaline, while Korat and Pak Chong soils are acidic in nature.

Leaching of elements from the soils: Acidic treatments caused highly significant (p value < 0.0001) leaching of elements, as shown in Table-2. Leaching behaviour of iron under non-acidic treatment (pH 7.0) was almost similar in all the

TABLE-1
BASIC PROPERTIES OF THE INVESTIGATED SOILS

Soil	Depth (cm)	Bulk density (mg m^{-3})	Particle size distribution (%)			Soil texture	pH (1:1)	SOM (%)	CEC (cmol kg^{-1})
			Sand	Silt	Clay				
Kr	0-30	1.53	80.30	12.43	7.27	Loamy sand	5.82	0.19	2.75
Pc	0-30	1.18	34.30	18.43	47.27	Clay	6.31	2.94	17.72
Pp	0-30	1.32	38.15	20.50	41.34	Clay	7.14	0.75	17.56

TABLE-2
ANALYSIS OF VARIANCE FOR ELEMENTS LEACHING FROM SOILS UNDER DIFFERENT TREATMENTS

SOV	D.F	MSS	F-Value	p-Value	MSS	F-Value	p-Value	MSS	F-Value	p-Value
Korat soil			Iron leaching			Aluminum leaching			Manganese leaching	
Acid rain pH (T)	4	47744.4	8004.8	<0.0001	35692.6	9657.0	<0.0001	5674.1	1575.0	<0.0001
Time intervals (t)	8	4389.9	736.0	<0.0001	1317.4	356.4	<0.0001	220.8	61.3	<0.0001
Interaction (T × t)	32	2537.5	425.4	<0.0001	1367.9	370.1	<0.0001	911.1	252.9	<0.0001
Error	88	6.0	–	–	3.7	–	–	3.6	–	–
CV	–		9.5			11.3			10.5	
Pak Chong soil			Iron leaching			Aluminum leaching			Manganese leaching	
Acid rain pH (T)	4	1605.0	1958.7	<0.0001	25076.9	5759.7	<0.0001	12085.5	2964.8	<0.0001
Time intervals (t)	8	343.2	418.8	<0.0001	6444.2	1480.1	<0.0001	2228.4	546.7	<0.0001
Interaction (T × t)	32	202.8	247.6	<0.0001	4901.5	1125.8	<0.0001	1010.5	247.9	<0.0001
Error	88	0.82	–	–	4.4	–	–	4.1	–	–
CV	–		19.7			12.5			14.1	
Phon Pisai soil			Iron leaching			Aluminum leaching			Manganese leaching	
Acid rain pH (T)	4	277.5	914.8	<0.0001	36135.1	10892.0	<0.0001	7024.3	2465.1	10.5
Time intervals (t)	8	60.6	199.8	<0.0001	4100.5	1236.0	<0.0001	992.5	348.3	10.5
Interaction (T × t)	32	32.8	108.0	<0.0001	3510.0	1058.0	<0.0001	401.1	140.8	10.5
Error	88	0.3	–	–	3.3	–	–	2.8	–	–
CV	–	–	26.0	–	–	10.2	–	–	15.4	–

Where, SOV: Source of variation, D.F: Degree of freedom, MSS: Mean sum of squares, CV: Coefficient of variance.

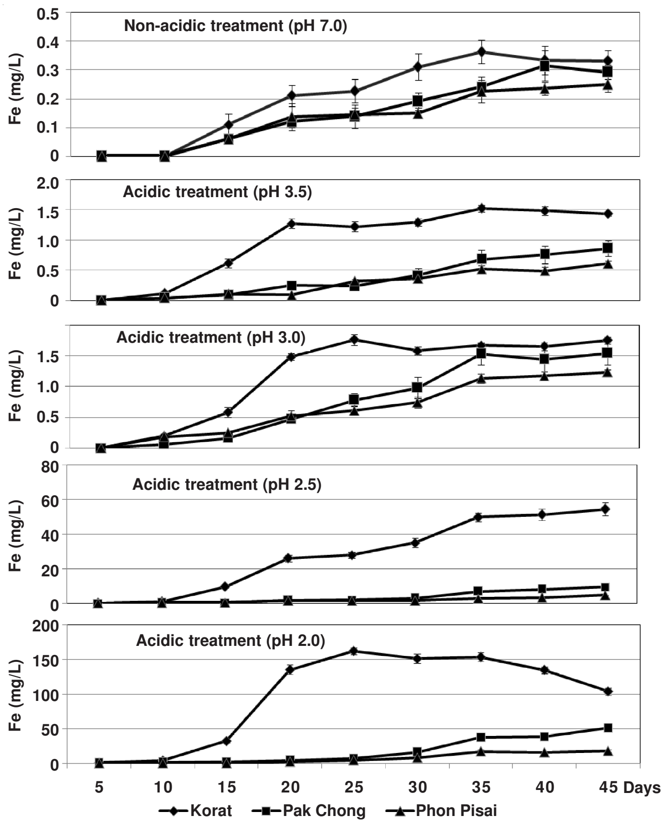

Fig. 1. Leaching of iron under different acidic treatments

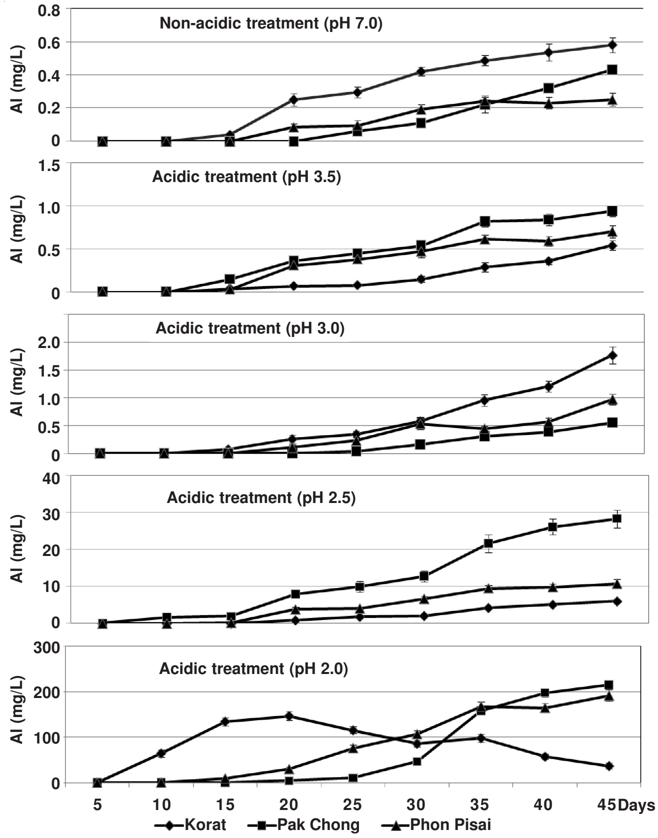

Fig. 2. Leaching of aluminum under different acidic treatments

investigated soils (Fig. 1). Iron concentrations in the leachate samples increased significantly ($p < 0.001$) with a decrease in the pH of applied simulated acid rain. The highest levels of iron leaching occurred in Korat soil under all the applied acidic solutions as compared to those of Pak Chong and Phon Pisai soil. This is because of the sandy texture, low cation exchange capacity and the acidic nature of Korat soil. Highly acidic solutions (pH 2.5 and 2.0) induced profound leaching of iron from the soils *i.e.*, 162.4, 51.5 and 18.5 mg/L, respectively from Korat, Pak Chong and Phon Pisai soils under pH 2 treatments.

Similar but reduced leaching patterns of aluminum were found in all the soils under non-acidic (pH 7) and moderate acidic treatments (pH 3.5 and 3), as given in Fig. 2. However, leaching of aluminum increased substantially under highly acidic treatments (pH 2.5 and 2) in all the investigated soils. This is because of accelerated process of weathering of soil minerals.

Highly acidic treatment can break solid-phase alumino-organics and release Al^{3+} [24]. Under acidic treatment with pH 2.5, aluminum concentrations reached 28.3, 10.8 and 6.0 mg/L in Pak Chong, Phon Pisai and Korat soils, respectively. The concentrations of soluble Al in the effluent samples increased as the pH of the applied solution decreased, resulting to faster decrease in soil reactive Al under higher acidic treatments[11]. Very high aluminum leaching occurred under acidic treatment with pH 2.0 *i.e.*, 215.8, 191.8 and 146.8 mg/L in Pak Chong, Phon Pisai and Korat soils, respectively. Higher leaching in clayey soils is due to have higher concentrations of sesquioxides in Pak Chong and Phon Pisai soils as compared to that of Korat soil having sandy texture and low organic matter. Soil acidification developed by acid deposition increases the solubility of aluminum in the soil[25] water which can cause phytotoxicity to crops. Mobilized aluminum, which is highly toxic to many aquatic organisms, also enters into the adjacent water bodies[1].

Leaching patterns of manganese from the soils under different acidic treatments are presented in Fig. 3. Similar leaching trends of manganese were observed for acidic treatments with pH 3.5, 3.0 and 2.5, reaching 40.2, 24.9 and 22.5 mg/L in Korat, Pak Chong and Phon Pisai soils under acidic treatment with pH 2.5. Quite high leaching of manganese was found in all the soils under highly acidic solution (pH 2) *i.e.*, 101.9, 70.3 and 82.3 mg/L in Pak Chong, Phon Pisai and Korat soil, respectively.

It is also important to note that aluminum and manganese leaching became highest at the middle stage in the sandy (Korat) soil and at the last stage of sprinkling in the clayey (Pak Chong and Phon Pisai) soils, respectively. This is because of lower sulfate and nitrate adsorption capacity and earlier depletion of aluminum and manganese contents in sandy soil in comparison with clayey soils, as indicated in Tables 4 and 5. Under normal conditions, these toxic metals are bound to the soil. However, due to additional dissolving action of H^+ ions (from acid rain) small bound soil particles break releasing toxic metals into the soil solution.

Status of extractable elements in the soils: Table-3 shows the status of iron in different soil layers after application of treatments in the leaching experiments. It is clear from the table that mobilization of iron increased quite significantly with an increase in acidity of applied solutions in all the investigated soils. The concentration of extractable Fe^{2+} increased

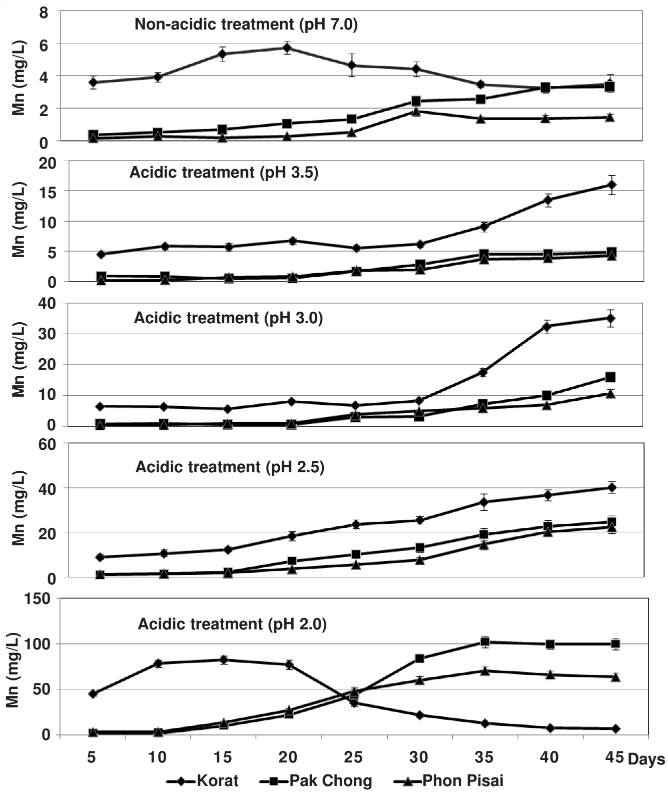

Fig. 3. Leaching of manganese under different acidic treatments

with a decrease in pH of the applied acid rain solution, as found by Liu *et al.*[26]. The highest iron mobilization occurred under the most highly acidic treatments (pH 2.0) in Pak Chong soil followed by the Korat soil series. This is because of the acidic nature of the Pak Chong and Korat soil series *i.e.*, 6.31 and 5.82, respectively[27]. Lower mobilization of iron was found in Phon Pisai soil series due to its alkaline nature (pH 7.14). Maximum extractable iron was found to be 128.9, 54.7 and 15.5 mg/kg of soil in Pak Chong, Korat and Phon Pisai soil, respectively. Higher mobilization of iron was observed in the upper layers of Pak Chong soil series, while in the lower layers of the Korat soil series. This is because of different soil texture which affects the sulfate and nitrate adsorption capacity. Higher iron contents in soils can decrease the activities of urease and acid phosphatase[28].

Extractable aluminum also increased due to higher mobilization with a decrease in rain pH. Aluminum mobilization occurred quite significantly in all the soils under highly acidic treatments (pH 3.0, 2.5 and 2.0). After treatment with acid rain (pH 2.5), very high extractable aluminum levels were found in the upper layers of Korat (108.6 mg/kg), Pak Chong (516.1 mg/kg) and Phon Pisai soil series (341.8 mg/kg). Maximum extractable aluminum in entire soil columns were found after highly acidic treatment (pH 2.0) ranging 98.7-163.7, 659.3-1162.0 and 666.7-957.4 mg/kg in Korat, Pak Chong and Phon Pisai soil series, respectively (Table-4). Benchmark (threshold) concentrations for the phytotoxicity of aluminum in soil and soil solution are 50 mg/kg and 0.3 mg/L, respectively[29]. Extractable aluminum was found to be higher than the threshold level (50 mg/kg) in all soils (particularly in upper layers) after highly acidic treatments (pH 2.5 and 2).

Levels of extractable manganese in different soil layers are given in Table-5. Extractable manganese increased under almost all the acidic solutions applied. Highly acidic treatments (pH 2) induced higher mobilization of manganese in clayey soils (Pak Chong and Phon Pisai) as compared to that in sandy soil (Korat). This indicates that clayey soils have higher reservoirs of manganese due to their higher organic matter content. Under the highly acidic treatment (pH 2), maximum extractable manganese was found to be 75.2 and 58.1 mg/kg of soil in Pak Chong and Phon Pisai soil, respectively. In Korat soil, maximum extractable manganese was found under acidic solutions with pH 3.0 (16.9 mg/kg) and 2.5 (20.8 mg/kg) in the lower layers of the soil columns. Increased concentration of soil Mn can cause Fe deficiency to plants in soils with sufficient Fe by inhibiting the root absorption and translocation of Fe to leaves[30]. Amounts of manganese extracted decreased

TABLE-3
STATUS OF EXTRACTABLE IRON AT DIFFERENT SOIL DEPTHS AFTER ACIDIC TREATMENTS

Soil	Depth		Iron (mg kg^{-1}) T0	T1	T2	T3	T4	T5
Kr	00-10 cm	Mean	0.15	0.13	2.67	6.44	10.65	21.17
		SD (±)	0.03	0.01	0.24	0.27	0.32	2.10
	10-20 cm	Mean	0.15	0.12	1.07	1.13	13.18	51.36
		SD (±)	0.02	0.03	0.10	0.07	0.64	3.67
	20-30 cm	Mean	0.15	0.12	0.76	1.60	24.76	54.68
		SD (±)	0.04	0.06	0.07	0.10	1.21	3.04
PC	00-10 cm	Mean	0.12	0.15	0.78	1.25	8.62	15.53
		SD (±)	0.02	0.03	0.07	0.11	0.57	1.06
	10-20 cm	Mean	0.12	0.08	0.43	1.07	5.76	6.38
		SD (±)	0.02	0.05	0.06	0.19	0.34	0.93
	20-30 cm	Mean	0.12	0.06	0.14	0.64	2.78	4.46
		SD (±)	0.03	0.02	0.02	0.11	0.23	0.25
PP	00-10 cm	Mean	0.22	0.08	1.87	4.57	30.57	128.96
		SD (±)	0.03	0.02	0.30	0.50	1.81	4.96
	10-20 cm	Mean	0.22	0.07	1.04	2.10	16.71	43.54
		SD (±)	0.04	0.02	0.05	0.11	1.63	2.33
	20-30 cm	Mean	0.22	0.06	0.67	0.98	9.78	23.11
		SD (±)	0.03	0.02	0.06	0.10	0.57	2.16

T0 = before exp., T1 = pH 7.0, T2 = pH 3.5, T3 = pH 3.0, T4 = pH 2.5, T5 = pH 2.0.

TABLE-4
STATUS OF EXTRACTABLE ALUMINUM AT DIFFERENT SOIL DEPTHS AFTER ACIDIC TREATMENTS

Soil	Depth		Aluminum (mg kg^{-1})					
			T0	T1	T2	T3	T4	T5
Kr	00-10 cm	Mean	2.50	1.80	21.09	83.20	108.62	163.70
		SD (±)	0.14	0.12	2.39	3.82	4.49	5.57
	10-20 cm	Mean	2.50	2.10	10.12	77.90	86.78	111.80
		SD (±)	0.14	0.08	1.24	3.49	5.11	6.92
	20-30 cm	Mean	2.50	2.20	4.30	7.60	37.04	98.70
		SD (±)	0.14	0.04	0.17	0.74	2.61	5.63
PC	00-10 cm	Mean	1.60	1.20	57.04	130.80	341.76	957.40
		SD (±)	0.17	0.09	3.07	5.01	7.75	9.48
	10-20 cm	Mean	1.60	1.40	37.03	21.00	208.90	824.70
		SD (±)	0.17	0.07	2.97	2.40	11.40	10.88
	20-30 cm	Mean	1.60	1.70	5.76	6.80	198.73	666.70
		SD (±)	0.17	0.09	0.64	0.60	9.95	14.21
PP	00-10 cm	Mean	1.40	1.10	51.08	107.00	516.05	1162.00
		SD (±)	0.18	0.22	3.24	5.97	11.04	20.82
	10-20 cm	Mean	1.40	1.30	43.72	12.70	401.90	847.40
		SD (±)	0.18	0.07	3.60	1.82	11.07	10.71
	20-30 cm	Mean	1.40	1.30	3.60	3.80	308.34	659.30
		SD (±)	0.18	0.11	0.20	0.25	14.21	11.51

T0 = before exp., T1 = pH 7.0, T2 = pH 3.5, T3 = pH 3.0, T4 = pH 2.5, T5 = pH 2.0.

TABLE-5
STATUS OF EXTRACTABLE MANGANESE AT DIFFERENT SOIL DEPTHS AFTER ACIDIC TREATMENTS

Soil	Depth		Aluminum (mg kg^{-1})					
			T0	T1	T2	T3	T4	T5
Kr	00-10 cm	Mean	3.50	2.93	6.07	1.65	3.03	0.21
		SD (±)	0.30	0.15	0.40	0.17	0.13	0.05
	10-20 cm	Mean	3.50	4.36	8.65	9.23	14.87	0.17
		SD (±)	0.40	0.43	0.50	0.84	1.16	0.03
	20-30 cm	Mean	3.50	4.73	11.76	16.87	20.76	2.13
		SD (±)	0.39	0.41	1.46	1.82	1.36	0.30
PC	00-10 cm	Mean	0.53	0.57	2.03	5.29	9.62	26.09
		SD (±)	0.08	0.09	0.12	0.36	0.94	1.92
	10-20 cm	Mean	0.53	0.39	1.83	1.18	6.71	21.48
		SD (±)	0.06	0.07	0.10	0.08	0.37	1.68
	20-30 cm	Mean	0.53	0.24	0.79	0.20	2.09	12.80
		SD (±)	0.07	0.08	0.08	0.04	0.11	1.07
PP	00-10 cm	Mean	2.39	1.02	3.30	7.85	12.98	75.22
		SD (±)	0.14	0.10	0.19	1.01	1.69	3.52
	10-20 cm	Mean	2.39	1.26	1.06	3.83	10.89	44.96
		SD (±)	0.08	0.04	0.07	0.22	1.18	3.30
	20-30 cm	Mean	2.39	1.35	0.67	1.06	7.06	54.08
		SD (±)	0.12	0.06	0.13	0.07	0.92	3.14

T0 = before exp., T1 = pH 7.0, T2 = pH 3.5, T3 = pH 3.0, T4 = pH 2.5, T5 = pH 2.0.

under highly acidic solution (pH 2) due to depletion of limited manganese content. As the investigated soils are acidic upland soils of Thailand with large extractable Al and Mn contents, Al and Mn together would cause rhizotoxicity. The mobilized iron, due to acidic treatments, would also cause toxicity to plants in soils because of the low cation exchange capacity of the soils[31].

Conclusion

This study indicated that leaching concentrations of all the investigated elements increased with an increase in the acidity of the applied solution. Acid rain also increased the remobilization of iron, aluminum and manganese in the soils, particularly in the upper soil layers. Extractable aluminum was found to be higher than the phytotoxicity threshold level under highly acidic treatments. Increased solubility of these metals in agricultural soils would also cause increased rhizotoxicity. Leaching of iron, aluminum and manganese out of the soil is associated with unfavorable environmental consequences due to contamination of underground water resources. There is a demand for developing effective countermeasures to reduce or at least control the emission of acid rain precursors in developing Asian countries. Energy conservation together with alternative energy sources can play a major role in reducing acid rain in the future. Energy efficiency should also be improved to reduce atmospheric pollutant emissions. Strong economic growth in Southeast Asia is expected to increase the demand for energy which emphasizes the need to shift from non-renewable to renewable energy sources.

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