

Cation Leaching from Soils Percolated with Simulated Sulfuric Acid Rain

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人工酸性 빗물에 의한 여러 土壤으로부터의 이온 洗脫

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ABSTRACT

Soils of four combinations, sand with high content of organic matter(SL), sand with low content of OM(SS), silty loam with high content of OM(LL) and silty loam with low content of OM(LS), were filled in column and then percolated with simulated sulfuric acid rain with pH 5.6, 4.0, 3.5, 3.0 and 2.5. From soil leachates, pH and concentrations of basic cations and Al were determined. Cation concentrations in the leachates increased as pH of the rain decreased. The orders of buffering capacity of soil, leachability of cation from soil, leaching sensitivity of ion and base saturation sensitivity of soil to acidity of the rain water were $SS < LS < SL < LL$, $Ca < Mg \ll Na < K$, $Na \leq K \ll Mg < Ca$, and $LS < SS \leq LL < SL$, respectively. Basic cations were leached more sensitively by acid rain from soil with high content of OM than low content of OM and from sand than silty loam.

Key words: Simulated sulfuric acid rain, Cation leaching, Aluminium mobilization, Soil sensitivity to acid rain, Buffering capacity of soil.

INTRODUCTION

The forest decline syndrome was first noted in *Abies alba* in West Germany in the early 1970s, and then this was apparent in *Picea abies* and *Fagus sylvatica* with a large scale in Europe in 1980s(Krause *et al.* 1986, Nilsson and Duinker 1987). Forest declines also have been reported from North America, Canada, China and Japan(CRIEPI 1989, Ma 1990/1991). The primary causes of forest decline have been focused on not only natural factors such as drought and diseases, but also anthropogenic factors such as acid rain, gaseous pollutants and excessive nitrogen(Rehfuss 1989, Kadza 1990). Out of them, the soil acidification by acid rain has been thought as an important causal factor of forest de-

cline(Zottle *et al.* 1989, Ulrich 1990). As soil is acidified, basic cations in soil are replaced by hydrogen ions. On the one hand, the replaced basic cations combine with soluble anions, such as SO_4^{2-} and NO_3^- , be deposited from polluted atmosphere, and these combined cations are leached through runoff or into ground water(Abrahamsen and Dollard 1978). Therefore, plants growing on acidic soil show cation deficiency symptoms in most acid rain area. One example is leaf chlorosis occurring as a result of Mg deficiency in forest decline area(Kandler and Miller 1990/1991, Ma 1990/1991). On the other hand, acid soluble metals, Al^{3+} and Fe^{2+} from acid soil, increase in soil solution, and excessive soluble Al^{3+} inhibits root growth(Huttermann and Ulrich 1984).

Effects of acid rain on soils were thought to vary depending upon soil properties, i.e. texture and organic matter content of soil. The purpose of this study is to characterize and compare the leachate properties of four soils, which differ in soil textures(sand and silty loam) and contents of organic matter, percolated with simulated sulfuric acid rains with varying pH values.

METHODS

Preparation of soils and simulated sulfuric acid rain

The soil was collected from A layer of forest soil(granite) in *Quercus mongolica* community at Mt. Kwanak, and screened through 1 mm and 0.062 mm screen sieve. Each of the four soils used for the experiment had four combinations of soil texture and content of organic matter(OM) as shown in Table 1.

The four soils are sand with high content of organic matter(SL), sand with low content of OM(SS), silty loam with high content of OM(LL) and silty loam with low content of

Table 1. Properties of soils used for experiment

Soil properties	SL	LL	SS	LS
O.M. content(%)	16	16	5	5
pH	6.7	6.6	6.4	6.5
Sand(%)	94	46	90	47
Silt(%)	5	50	9	49
Clay(%)	1	4	1	4
Soil texture	sand	silty loam	sand	silty loam
Exchangeable cation(meq / 100g)				
K	9.0	12.5	1.0	2.5
Na	3.2	3.4	0.3	0.6
Ca	18.6	22.5	6.7	9.5
Mg	10.5	12.5	3.1	4.2
Cation exchangeable capacity (meq / 100g)	42.6	59.1	24.0	27.8
Base saturation(%)	96.9	86.1	46.2	60.4

OM(LS). The pH values of simulated sulfuric acid rain(SSAR) were adjusted to 4.0, 3.5, 3.0 and 2.5 by the diluted H_2SO_4 and pH 5.6 of control rain was adjusted by dissolving CO_2 in deionized water.

Extraction of soil solution

Five gram of air-dried soil was filled up a column of cation exchange capacity(CEC) apparatus with 1.2cm in inner diameter and 11.0cm in length. The SSAR of 100 ml was poured over the soil and the leachate from it was collected with polyethylene bottle. Such procedure was repeated consecutively 10 times until 1,000ml of SSAR was collected. For 100ml of SSAR to be leached, it took 3~8 hours in the sand column, and about 20 hours in early period and about a week in late period in the silty loam column. Two to five replicates were used for leaching procedure of SSAR.

Physico-chemical analyses of soil and soil leachate

Soil texture was determined by hydrometer method(Black *et al.* 1973). The air-dried soils before and after SSAR leaching were analyzed chemically; OM content of soil was determined by loss on ignition at 550°C for 4 hr; the basic cations in soil were extracted with 1 N ammonium acetate(pH 7.0); the leachates from the soils were stored at 4°C until analysis; chemical analyses of both extracts and leachates were made using a flame-photometer(Coleman 51) for K and Na and using an atomic absorption spectrophotometer(GBS 903) for Ca, Mg and Al. The pH of leachate and soil(soil:water = 1:5, w/v) was measured using a pH meter(Fisher 230). Cation exchange capacity(CEC) was determined by Moore and Chapman(1986) and base saturation(BS) was calculated as a percentage of CEC to basic cations(Ca + Mg + K + Na).

Leachability of ion from soil by simulated sulfuric acid rain

Leachability of an ion from soil by acid rain water(pH 4.0~2.5) was defined as a regression coefficient(W_m / L_{max}) in Michaelis-Menten's reciprocal function:

$$\frac{1}{L} = \frac{W_m}{L_{max}} \times \frac{1}{W} + \frac{1}{L_{max}}$$

L : leached amount of an ion

L_{max} : total leached amount of an ion

W : percolated amount of simulated sulfuric acid rain

W_m : amount of simulated sulfuric acid rain percolated when a half of the total amount of an ion was leached

RESULTS AND DISCUSSION

pH buffering capacity of soils to simulated sulfuric acid rain

When SSAR with pH 2.5 were percolated to soil columns filled with SL, LL, SS and

LS, pH of leachates from soil rapidly decreased in leachates of 400, 600, 200 and 300 ml respectively and kept constantly thereafter (Fig. 1).

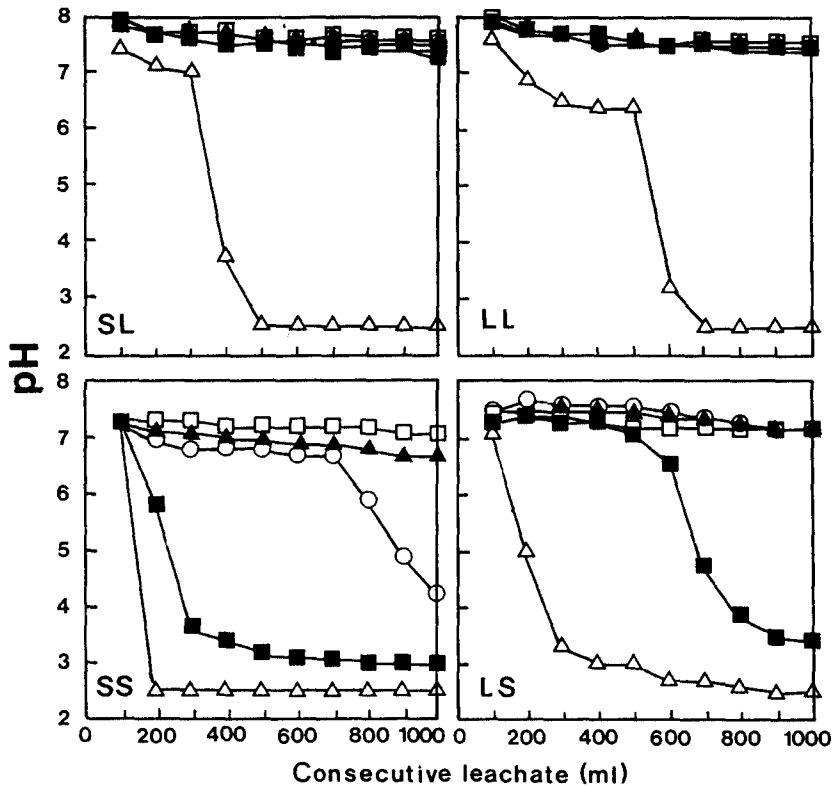


Fig. 1. Changes of pH of consecutive leachates from soils after percolating with simulated sulfuric rain water with pH 5.6(Δ), pH 4.0(\blacksquare), pH 3.5(\circ), pH 3.0(\blacktriangle) and pH 2.5(\square)

The order of buffering capacity of the soil, ability to be changed less pH, to SSAR was $SS < LS < SL < LL$. Such buffering capacity decreased more rapidly in soil with low content of OM than with high content of OM, and also more in sand than in silty loam. Thus the buffering capacity of soil was profoundly affected by content of OM than texture of soil. This result was consistent with that of Wilklander's work (1978). The pHs (pH 7.1~8.0) of first leachate were higher than the pHs (pH 6.4~6.7) of original soils (Table 1) (Hutchinson 1979). High values of pH in initial leachates were thought that acid additions were initially neutralized by release of basic cations and sulfate adsorption (Dahlgren *et al.* 1990).

Changes of soil pH with simulated sulfuric acid rain

The pHs of sand percolated with SSAR of 300 ml decreased by 1.0~1.4 unit and those

of silty loam by 0.0~0.4 unit compared with those of original soils regardless of content of OM in soil (Table 2).

Decrease of pH of original soil by SSAR was profoundly affected by the difference in soil texture compared with that of OM content. Rapid decrease of pH after leaching of 1,000ml SSAR occurred below pH 3.0, pH 3.0 and pH 3.5 of SSAR in soils of SL, LL and SS, respectively. Soil acidification by SSAR occurred faster in sand with low OM content than in sand with high OM content. If it were the same content of OM in soils, sand might be more acidified than silty loam by SSAR percolating. Lau and Mainwaring (1985) insisted that soil texture and base saturation were closely linked and together play an important role in governing buffering activities in a soil system.

Table 2. pH values of original soil and soil after percolated with 300ml and 1,000ml of simulated sulfuric acid rain(SSAR)

Soil	Amount of SSAR	pH of SSAR				
		5.6	4.0	3.5	3.0	2.5
SL	original soil	6.7	6.7	6.7	6.7	6.7
	300ml	5.7	5.5	5.6	5.6	5.3
	1,000ml	5.7	5.5	5.4	4.8	4.1
LL	original soil	6.6	6.6	6.6	6.6	6.6
	300ml	6.6	6.5	6.6	6.6	6.4
	1,000ml	5.6	5.4	5.8	4.8	4.0
SS	original soil	6.4	6.4	6.4	6.4	6.4
	300ml	5.1	5.1	5.1	5.1	5.1
	1,000ml	4.9	5.0	4.4	3.5	3.0
LS	original soil	6.5	6.5	6.5	6.5	6.5
	300ml	6.1	6.1	6.1	6.1	6.1

Changes in the amount of cations leached from soil by simulated sulfuric acid rain

Amount of K in 100 ml of leachate percolated from soils of SL, LL, SS and LS were 75~80%, 78~80%, 39~68% and 40~67%, respectively. Leaching of K in soil increased as OM content of soil increased but as pH of SSAR decreased regardless of soil texture (Fig. 2). In both cases of SL and LL, the K concentration of leachate was almost constant over 200 ml of leachate percolated with different pHs of SSAR.

Amounts of Na in 100 ml of leachate percolated from soils of SL, LL, SS and LS were 78~83%, 92~98%, 46~60% and 39~56%, respectively (Fig. 2). Almost all of Na in soils of SL and LL was leached by low amount of SSAR. The values above 100% were not expressed with symbols in Fig. 2. The Na leaching pattern by acidity of SSAR was similar to that of K leaching.

Amount of Ca in 100 ml of leachates increased as pH of SSAR decreased, and as amount

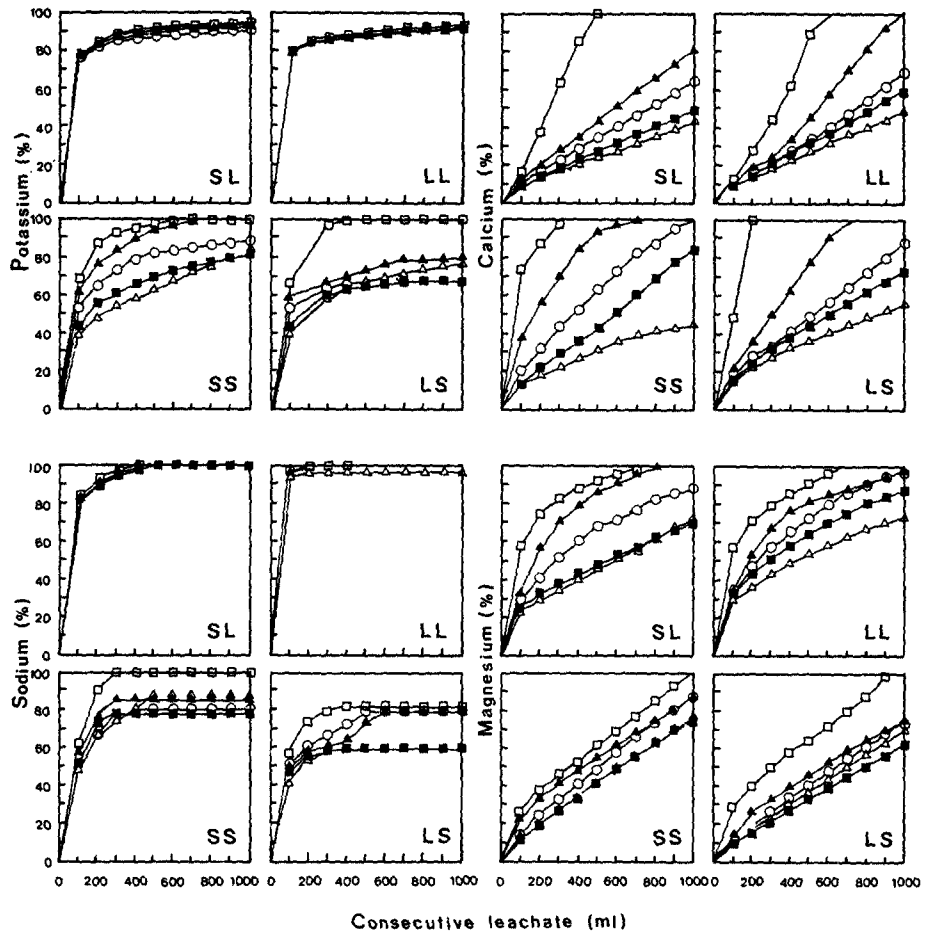


Fig. 2. Cumulative content of potassium, sodium, calcium and magnesium in leachates as percent of original soil after percolating with simulated sulfuric rain water with pH 5.6(Δ), pH 4.0(\blacksquare), pH 3.5(\circ), pH 3.0(\blacktriangle) and pH 2.5(\square). Each ion content in the original soil extracted with 100 ml of 1N ammonium acetate solution was expressed as 100%. The values above 100% were not expressed with symbols in figure.

of SSAR increased without regard to both OM content and texture of soil(Fig. 2). Exchangeable Ca from soils of SL, LL, SS and LS was perfectly exhausted by percolation of 500, 600, 400 and 200 ml of SSAR with pH 2.5, respectively. Ca leaching from soil by SSAR increased as both OM content in soil and pH of SSAR decreased but it was not profoundly affected by soil textures.

Patterns of Mg leaching from soils with high content of OM were different from those with low content of OM. Mg leaching from the former increased as amount and acidity of SSAR increased along parabolic curve which was similar patterns to K and Na leaching. However, that from the latter increased in proportion to leaching amount of SSAR along

linear curve(Fig. 2).

These characteristics of cation leachings from soils were consistent with those of cation leachings observed by Haynes and Swift(1986) in which levels of Ca and Mg in soil solution increased exponentially as the pH of soil with sulfuric acid was lowered from 6.5 to 3.8, while levels of K in soil solution increased at a more-or-less linear fashion and those for Na tended to increase. Also Lee(1985) reported that Mg and Ca in top soil decreased but K and Na were not greatly changed by treatments of pH 4.0, 3.5 and 3.0 of sulfuric acid rain compared with that of control rain. Bergkvist(1986) observed the significant decrease of Ca and Mg from soil by soil acidification. Abrahamsen and Stuanes(1986) and Johnson and Reuss(1984) observed that the lower pH of rain water was, the more basic cations were rapidly leached with small amount of rain water.

Aluminium mobilization from soil with simulated sulfuric acid rain

By percolating SSAR of pH 2.5 consecutively, Al mobilization from soil started from 600, 300, 100 and 300 ml of leachate in soils of LL, SL, SS and LS, and by percolating SSAR of pH 3.0 it initiated from 900, 200 and 700 ml of leachate in soils of SL, SS and LS,

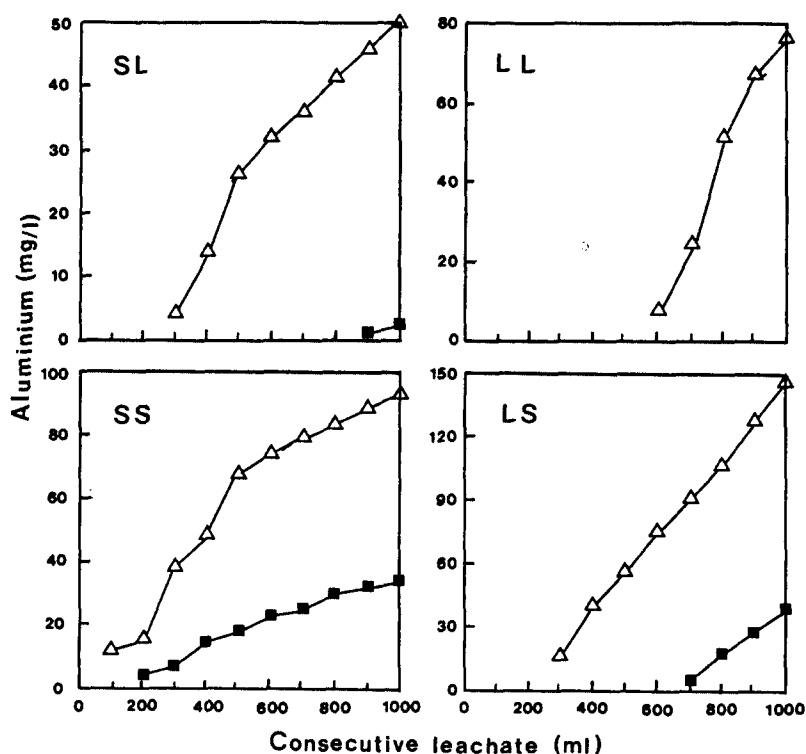


Fig. 3. Cumulative curves of content of aluminium in leachates from soils after percolating with simulated sulfuric rain water with pH 5.6(△), pH 4.0(■), pH 3.5(○), pH 3.0(▲) and pH 2.5 (□)

respectively. Thus Al mobilization from soil was inversely proportional to pH of SSAR, and was faster in sand than in silty loam and in soil with low content of OM than with high content of OM (Fig. 3).

The extent of Al mobilization from soil was inversely proportional to buffering capacity of soil and the order was SS>LS>SL>LL. It was known that the more acidified the soil was, the more Al mobilization occurred (Giddings and Galloway 1976, Huttermann and Ulrich 1984).

Leachability of ion from soil to simulated sulfuric acid rain

Leachabilities of ions which indicated the extent of amount of rain water used until the half of total amount of an ion in soil was leached were shown in Table 3.

Table 3. Leachabilities (W_m/L_{max}) of ions calculated from Michaelis-Menten's reciprocal function ($1/L = [W_m/L_{max}].[1/W] + 1/L_{max}$).

pH	SL	LL	SS	LS	Mean (pH 4.0~2.5)
	W_m/L_{max}	W_m/L_{max}	W_m/L_{max}	W_m/L_{max}	
Potassium					
5.6	0.247	0.176	1.517	1.299	
4.0	0.280	0.189	1.227	0.907	
3.5	0.260	0.195	0.866	0.475	
3.0	0.262	0.194	0.716	0.495	
2.5	0.254	0.214	0.507	0.531	
Mean	0.264	0.198	0.829	0.620	0.473
Sodium					
5.6	0.356	0.032	1.174	0.928	
4.0	0.297	0.066	0.719	0.961	
3.5	0.374	0.076	0.819	0.853	
3.0	0.339	0.088	0.678	1.049	
2.5	0.341	0.136	0.694	0.617	
Mean	0.337	0.091	0.727	0.870	0.506
Calcium					
5.6	10.394	10.175	5.954	5.801	
4.0	10.306	9.676	6.612	5.662	
3.5	8.664	10.248	4.350	5.126	
3.0	7.169	9.316	1.932	4.273	
2.5	5.723	7.807	0.668	1.232	
Mean	7.965	9.261	3.390	4.073	6.173
Magnesium					
5.6	3.346	2.238	7.878	8.676	
4.0	2.618	2.029	8.761	10.212	
3.5	2.656	2.147	6.344	8.707	
3.0	2.256	1.950	3.561	6.268	
2.5	0.847	0.919	3.024	2.698	
Mean	2.094	1.761	5.422	6.971	4.062

Leachabilities of the various cations were greatly dependent on difference of content of OM of soil than that of soil texture : K, Na and Mg were more easily leached from soil with high content of OM than that with low content, while Ca was more easily leached from soil with low content of OM. The order of soils in K, Na and Mg leaching was $LS \leq SS < SL < LL$ but that in Ca leaching was $LL < SL < LS < SS$. Mean values of leachabilities from the various soils were 0.473, 0.506, 4.062 and 6.173 in K, Na, Mg and Ca ion, respectively and lower in monovalent ions(K and Na) than in divalent ions(Ca and Mg). The order of ion leachability to acid rain was $Ca < Mg \ll Na \leq K$. This order was inverse to that of ion leaching sensitivity shown by Haynes and Swift(1986).

Changes of residual ions, cation exchange capacity and base saturation

Content of residual ions, cation exchange capacity(CEC) and base saturation(BS) of various soils percolated with SSAR of 1,000ml decreased with decreasing pH of SSAR (Table 4).

Table 4. Content of residual ions, cation exchangeable capacity(CEC) and base saturation(BS) of soils leached or unleached with 1,000ml of simulated sulfuric acid rain

Kinds of soil	pH of rain	Content of residual ions(meq /100g)				CEC (meq /100g)	BS (%)
		K	Na	Ca	Mg		
SL	original soil	9.03	3.24	18.68	10.53	42.64	93.1
	5.6	0.61	0.52	10.06	3.88	21.07	71.5
	4.0	0.54	0.57	8.73	3.93	21.00	65.4
	3.5	0.54	0.38	6.24	2.27	20.14	46.8
	3.0	0.24	0.32	3.55	1.02	20.00	25.7
	2.5	0.19	0.18	0.25	0.82	15.00	6.0
LL	original soil	12.58	3.47	22.52	12.02	59.15	86.1
	5.6	0.46	0.42	12.18	5.02	27.29	66.3
	4.0	0.47	0.40	8.35	3.88	26.86	48.8
	3.5	0.53	0.42	9.03	3.62	26.43	51.5
	3.0	0.25	0.29	1.07	3.56	26.43	19.6
	2.5	0.16	0.22	0.54	2.06	20.79	14.3
SS	original soil	1.06	0.36	6.78	3.16	18.57	46.2
	5.6	0.19	0.10	3.97	0.71	9.14	54.4
	4.0	0.20	0.10	2.03	0.70	8.71	34.8
	3.5	0.12	0.10	0.15	0.74	9.14	12.1
	3.0	0.09	0.09	0.09	0.38	7.43	8.7
	2.5	0.09	0.08	0.09	0.20	6.79	6.8
LS	original soil	2.55	0.63	9.55	4.21	27.82	60.4
	5.6	0.30	0.09	3.99	1.14	13.57	40.7
	4.0	0.26	0.10	3.32	1.22	13.29	36.9
	3.5	0.30	0.09	1.49	0.84	11.43	24.1
	3.0	0.10	0.08	0.23	0.40	10.00	8.1
	2.5	0.10	0.06	0.18	0.35	10.00	6.9

Basic cations remaining in soils, even though percolated with SSAR of pH 5.6, decreased compared with those in unpercolated soils. Such decreasing patterns were more conspicuous in K and Na than in Ca and Mg. In this study, regression coefficient (b) of log function, BS or $ion = a + b \log(pH)$, was calculated from both the BS or ion of percolated soil and the pHs of SSAR. Regression coefficient was used as BS sensitivity of soil or leaching sensitivity of ion to SSAR. The orders of them were $LS < SS \leq LL < SL$ in BS of soils, and $Na \leq K \ll Mg < Ca$ in ions, respectively (Table 5).

Table 5. Regression coefficients of log function calculated from BS or ion of percolated soil and the pHs of SSAR. In case of regression coefficient of ion, the values represent means of four soils, and the values in parentheses represent range of regression coefficients of four soils.

Base saturation	Regression coefficient	Ion	Regression coefficient	Range
SL	84.39	K	0.34	(0.15~0.56)
LL	67.84	Na	0.18	(0.02~0.44)
SS	64.17	Ca	8.56	(1.15~15.51)
LS	47.83	Mg	3.37	(0.64~4.41)

This result indicated that the amount of basic cations leached from soils of SL and LL increased as the pH of SSAR decreased, and that basic cations were more sensitively leached from soil with high content of OM than with low content of OM and from sand than from silty loam. The order of leaching sensitivity of ions was inverse to that of ion leachability as shown previously. Lee (1985) found that for soils unsaturated with sulfate the order of the sensitivities of major cations to leaching by simulated sulfuric acid rain was $K < H < Ca \leq Mg$ while no sensitivity was found in Na. Also Haynes and Swift (1986) shown experimentally that an increased acidity of simulated sulfuric acid rain most significantly increased the Ca and Mg loss from soil; K appeared to be less sensitive. Therefore, the order of leaching sensitivity of ions was $Na < K < Mg < Ca$. But Bergkvist (1986) reported that leaching of K and Na were more sensitive than that of Ca and Mg to soil acidity.

摘 要

토성과 유기물함량이 다른 4가지 토양을 유리관에 충전하고 인공산성빗물을 연속적으로 관류시켜 그 세탈액의 pH, K, Na, Ca, Mg 및 Al를 정량한 결과 빗물의 pH가 낮을수록 세탈되는 이온량이 많았다. 산성빗물에 대한 토양의 완충능은 유기물이 많은 미사질점토(LL) > 유기물이 많은 사토(SL) > 유기물이 적은 미사질 점토(LS) > 유기물이 적은 사토(SS) 순이었고, 토양으로부터의 이온세탈도는 $Ca < Mg \ll Na \leq K$ 순이었으며, 이온세탈 민감도는 $Na \leq K \ll Mg < Ca$ 순이었다. 염기포화 감소로 판단한 산성빗물에 대한 토양 민감도는 $LS < SS \leq LL < SL$ 순이었다. 즉 산성빗물에 의하여 유기물이 적은 토양보다 많은 토양에서, 미사질점토보다 사토에서 그리고 산도가 낮은 빗물일수록 더욱 많은 이온이 세탈되었다.

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