Leaching of plant nutrients from an acid forest soil after nitrogen fertilizer application

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Summary In a laboratory experiment, the effect of nitrogen fertilizers on the leaching patterns and input-output budgets of plant nutrients in an acid soil (dystric cambisols) was studied. An amount of N corresponding to 80 kg/ha was applied to the top surface of undisturbed soil columns of 30 cm layers with a diameter of 14.4 cm as $Ca(NO_3)_2$ or $(NH_4)_2SO_4$. After establishing steady state flow condition, a flux of 0.5 cm solution/column was applied daily for 60 days. The leachates were collected and analysed every four days. It was found that not only the amount and quality of the applied cation influences the leaching pattern of plant nutrients in an acid soil but also the accompanying anion. To interpret leaching data properly, it was necessary to know the magnitude of N transformed in the different processes. Although the two applied salts influenced plant nutrient transport in different ways, total outputs of cations and anions were somewhat similar though outputs after $(NH_4)_2SO_4$ were slightly higher than after $Ca(NO_3)_2$ addition.

Introduction

Ltudies on the leaching of plant nutrients from soils after nitrogen rertilizer application are scarce. Previous studies on nitrogen behaviour in soils^{14,19}, have been limited to N transport and transformations without considering the effect it has on the other elements in the soil. An application of a salt to the soil followed by a percolating solution may lead to a different chemical behaviour in soil and may even change the chemistry of the soil solution^{4,9,13,29}. Studies to show the effects of an acid rain on the leaching of plant nutrients especially in acid soils have recently received much attention^{1,4,23,27,29}. One potential longterm effect of continuous leaching of nutrients from the soil is a decrease in soil fertility. This may lead even to a reduction of plant productivity especially in soils of low buffering capacity.

Applying NO_3^- to an acid soil, the accompanying cation can induce ion exchange which may result to high H⁺ ion-concentrations in soil solution. Uptake of NO_3^- by plants and micro-organisms, and its reduction to gaseous compounds will consume one proton per $NO_3^$ removed from the soil solution, causing soil alkalinization. An addition of NH_4^+ into a soil may be involved in oxidation to nitrates, in cation exchange and in assimilation by micro-organisms. During its oxidation, two moles of H⁺ ions are produced per mole of NH_4^+ whereas its uptake creates one of H⁺. The degree of NH_4^+ oxidation and its uptake will therefore affect the H⁺ dynamics and other related soil processes²⁶.

This study was carried out to observe and to quantify some of the effects of nitrogen fertilizers on the leaching patterns and input-output budgets of plant nutrients in an acid soil.

Materials and methods

Undisturbed soil columns from an upper 30 cm layer with a diameter of 14.4 cm of an acid soil (dystric cambisols) were used for this study. The soil was sampled from a forested area of mountaineous region of Solling district in W. Germany, occupied by 120 year old beech trees. The soil is derived from loess (0-60 cm) and lies over solifluction masses which consist of weathered triassic sandstone. The rooting zone of this soil is strongly acidic. The soil has low bulk density, well aggregated and has a high physical stability. Table 1 shows some of the physical and chemical properties of the soil.

In order to collect the leachates completely, the experiment was set in a column study¹⁵. Unsaturated steady state flow of soil solution in the columns was established. This condition was maintained for 60 days by a flux of 0.5 cm solution/column/day. This solution was prepared and had a concentration composition of ions similar to a concentration from a saturated extract of a 30 cm layer of this acid soil (Table 2). Each column was treated with 130.3 mg N (an amount corresponding to 80 kg/ha) as $Ca(NO_3)_2$ or $(NH_4)_2SO_4$ in dissolved form. This amount was applied in one lot and to the top surface of the columns. The study took place at $23^{\circ}C$.

Under a tension of $100 \text{ ml H}_2\text{O}$ column, the effluents were collected from the bottom of every column once every four days and analysed for pH, main cations and anions. The analysis was done according to Ulrich²⁵. Na, K, Ca, Mg and Mn were determined by atomic absorption, Fe by Calorimetric as phenanthrolin complex⁵, Al by calorimetric with aluminon¹¹, S by potentiometric titration with EDTA after BaSO₄ precipitation⁸, Cl by titrating against AgNO₃, NH₄-N and NO₃-N by distillation-titration method according to Fleige⁶, and C according to Wösthoff³⁰.

The concentrations of cations and anions analysed from 5 replicates for the 60 days (2.3 pore volumes), plotted against the corresponding volumes of effluents collected led to the construction of breakthrough curves (BTCs). A breakthrough curve is a standard form of presenting concentration changes of solutes in the effluent with time from a soil column. Such curves can give information about the behaviour of solutes in porous media because their shapes are influenced by physical, chemical and bio-chemical reactions during their passage in soil.

Results

Fig. 1 (a-k) shows the BTCs after Ca(NO₃)₂ and (NH₄)₂SO₄ application. The curves obtained after Ca(NO₃)₂, particularly those for K⁺, Ca²⁺, Al³⁺, Mn³⁺ and NO₃ show distinct peaks. The concentrations of these ions in the soil solution at the beginning were low but as leaching continued, their concentrations steadily increased and reached the maximum after 28 days. Except for K⁺, the peaks of other ions almost coincided with the exchange of the first pore volume, *i.e.* after 26 days.

After (NH₄)₂SO₄ application, a different behaviour was expected

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_ 5	H (meg.kº	Na (1-,	К	Mg	Ca	Al	Fe	Mn	C.E.C.	J	Z %	C/N	вр g/cm ³
1 20	p u	, u d	p u	p u	n d.	n.d.	n d	- p.u	n d.	45.1	1.63	27.67	0.18
(90.0										(5.25)	(0.05)	(3.22)	(0.08)
68.	55.2	1.40	6.37	0.40	15.2	80.7	5.45	1.65	166.4	11.1	69.0	16.09	0.73
0.07)	(20.8)	(0.29)	(3.19)	(0.22)	(8.40)	(8.03)	(2.05)	(0.53)	(41.9)	(5.22)	(0.35)	(3.27)	(0.31)
.03	29.7	1.00	2.45	0.14	4.0	74.1	3.23	1.11	115.7	4.5	0.26	17.31	1.13
(60.0	(10.2)	(0.13)	(0.51)	(0.05)	(1.13)	(8.91)	(1.31)	(0.44)	(13.6)	(1.35)	(0.08)	(3.59)	(0.15)
.18	17.9	0.9	2.04	0.08	2.42	76.7	1.78	2.34	104.2	3.1	0.17	18.23	1.29
0.08)	(6.3)	*0.15)	(0.22)	(0.02)	(0.43)	(11.68)	(0.91)	(69.0)	(6.1)	(0.63)	(0.03)	(2.68)	(0.07)
.34	10.3	0.9	1.70	0.05	1.55	77.4	0.79	2.74	95.6	2.31	0.14	16.5	1.33
0.69)	(3.3)	(0.11)	(0.12)	(0.005)	(0.20)	(9.13)	(0.46)	(0.73)	(6.5)	(0.24)	(0.01)	(1.51)	(0.08)
3.49	5.9	1.00	1.56	0.05	1.44	80.6	0.36	2.71	93.6	2.25	0.13	17.31	1.23
(60.0)	(1.5)	(6.0)	(0.18)	(0.01)	(0.05)	(8.00)	(0.11)	(0.73)	(6.7)	(0.38)	(0.01)	(2.16)	(0.01)
3.62	3.9	0.9	1.34	0.04	1.15	76.0	0.20	1.99	85.6	1.73	0.12	14.42	1.18
(0.1)	(1.0)	(0.12)	(0.21)	(0.004)	(0.31)	(0.25)	(0.05)	(0.4)	(2.3)	(0.39)	(0.007)	(3.6)	(0.12)
3.71	3.0	1.0	1.42	0.03	1.52	72.3	0.14	1.59	81.0	1.89	0.11	17.18	1.14
(0.11)	(1.0)	(0.01)	(0.42)	(00.006)	(0.35)	(4.08)	(0.05)	(0.44)	(5.3)	(0.50)	(0.008)	(3.76)	(0.10)

() = standard deviations

n.d. = not determined + BD = Bulk density









Fig. 1. 1-k. Breakthrough curves (BTCs) of cations and anions following Ca(NO₃)₂ and (NH₄)₂SO₄ application. (mean values, n = 5). pages 20-23 a) H⁺ b) Na⁺; c) K⁺ d) Ca²⁺;

a) 11	0) Ha ,	C) IX	u)ca,
e) Mg ^{2 +}	f) Al ³⁺ ;	g) Mn ²⁺ ;	h) $SO_{4}^{-}S^{2}^{-};$
i) Cl ⁺ ;	k) NO ₃ -N ⁻ .		

Table 2. The composition of the input solution (mg/l)

pН	Na	K	Ca	Mg	Al	Fe	Mn	SO₄-S	C1
3.65	0.50	0.38	1.40	0.22	0.33	0.26	0.09	3.88	4.89

due to the possible transformation of NH_4^+ in soil. This indeeed was the case as the curves show. In general, there were no clear peaks except for Ca^{2+} . Instead there was distinct retardation of ions such as H^+ , Al^{3+} , Mn^{2+} and SO_4^{2+} in the soil solution. The low concentrations were especially pronounced immediately before and after the first pore volume. In both treatments, the concentrations of Mg^{2+} , Na and Cl^- tended to decrease with time while that of K⁺ tended to increase.

The input-output balances of the analysed cations and anions are shown in Table 3. After Ca^{2+} application, the leaching of cations was highest with Al^{3+} followed by Mg^{2+} , Mn^{2+} , Na^+ , $K^+ NH_4^+$ in that decreasing order. Ca^{2+} , Fe^{2+} and H^+ were retained in the soil. Consequently, the output of both cations and anions was slightly less by about 7 and 2% for cations and anions, respectively, than the input. After NH_4^+ application, a similar behaviour was found with the following leaching order of decreasing: $Al^{3+} > Mg^{2+} > Ca^{2+} > Na^+ > K^+$. In this case Ca^{2+} was released while NH_4^+ and Fe_3^+ were retained in the soil. The input-output balance shows that about 39% of the anions,

Table 3. 1	Input-out	out balances	s of cations	and anions	(Keq.ha ⁻¹	t; mean val	tues $(n = 5)$							
	Н	Na	Х	NH4-N	Ca	Mg	AI	Fe	Mn	SO4-S	ច	NO. _• ON	+	1
$Ca(NO_3)$	applicati	on												
Input	0.67	0.1	0.03	I	5.55	0.06	0.11	0.4	0.01	0.66	0.41	5.72	6.93	6.79
Output	0.31	0.43	0.23	0.19	0.97	1.29	2.61	0.02	0.4	1.11	1.12	4.43	6.45	6.66
	(0.11)	(0.05)	(0.1)	1	(0.32)	(0.19)	(1.2)	600.0)) (0.14)	(0.31)	(0.12)	(1.40)		
Input -Outnut	+0.36	-033	-0.20	-019	+458	-173	-250	+038	-030	-0.45	-0.71	+1 29	+0.48	+013
					2	241	2			2.0	1.1.0			
(NH ⁴) ₂ S() ₄ applica	tion												
Input	0.67	0.1	0.03	5.72	0.21	0.06	0.11	0.4	0.01	12.08	0.41	I	7.31	12.49
Output	0.27	0.32	0.19	0.22	1.05	1.64	3.3	0.02	0.34	2.69	1.02	3.93	7.35	7.64
	(0.12)	(0.04)	(0.13)	I	(0.25)	(0.29)	(1.46)	(0.005	(0.08)	(0.67)	(0.18)	I		
Input														
-Output	+0.40	- 0.22	-0.16	+ 5.50	-0.84	-1.58	-3.19	+0.38	-0.33	+ 9.39	-0.61	- 3.93	-0.04	+4.85
() = Stan	dard devia	tions + - (Sint in soil	- Source	e from coil	+ - Cum	of ations	C mins	f anian					

= Sum of cations, - Sum of amons. = Source from soil, + = Sink in soli, -() = Standard deviations, +

24

mainly sulphates were retained in soil whereas for cations it was almost zero. In the side of anions, the soil released Cl^- and SO_4^- and retained NO_3^- after NO_3^- application whereas NO_3^- and Cl^- were released and SO_4^- retained after SO_4^- application. The phosphates were negligible in the leachates.

Discussion and conclusions

During percolation, cations accompanying anions undergo cation exchange, precipitation and dissolution reactions. This applies also to some anions except for Cl^- and NO_3^- which are seldom involved in chemical reactions and rarely interact with the solid part of the soil.

The formation of peaks by K⁺, Ca^{2+} , Al^{3+} and Mn^{2+} after Ca^{2+} application demonstrates the quick participation and ability of Ca²⁺ in exchanging other cations from the soil complex. While the leachability of Ca²⁺, Al³⁺ and Mn²⁺ was faster, thus their maximum concentrations coincided with the first pore volume *i.e.* when the water in the soil was replaced one time, that of K⁺ showed some resistance. This behaviour of K^+ to leaching has also been observed by others^{4,9,22}. Hartikainen⁹ from a leaching experiment with an acid soil, reported that K⁺ showed highest resistance to leaching while Na⁺ showed the least. Nevertheless, in acid soils where exchange sites depend on pH, K⁺ leachability may be enhanced^{15,28}. This shows therefore that leaching of nutrients from soils does not depend only on the availability of the nutrient but also on other factors such as the nature of the element itself, environmental variables and the history of the soil. For anions, NO₃ exhibited the expected pattern of transport *i.e.* with minimum interaction in the soil. Its maximum concentration in the output almost coincided with the replacement of one pore water volume, *i.e.* after 26 days. This is comparable to the maximum outputs of Ca²⁺, Al³⁺ and Mn²⁺. This could therefore suggest that cations do follow the same pattern as the sum of the anions. The BTCs shown in Fig. 2 could therefore support the assumption of electroneutrality because peaks of NO_3^- and those of Ca^{2+} , Al^{3+} and Mn^{2+} are exhibited after 28 days of leaching. With respect to other anions (Cl^{-} and SO_{4}^{2-}), the situation is different. The chloride showed a slight increase in concentration when NO₃ had passed its maximum, whereas SO_4^2 concentration was slightly depressed.

The absence of distinct peaks after $(NH_4)_2SO_4$ application demonstrates the slow participation of NH_4^+ in exchanging other cations from the soil complex. This is because NH_4^+ is submitted to various reactions in soil such as oxidation, fixation in clay mineral lattices and immobilization by microbes. In this case however, not only NH_4^+ is affected but also SO_4^{2+} . The change involves two different processes. NH_4^+ is oxidized to nitrates. During this reaction, two strong acids are formed, *i.e.*

$$(NH_4)_2SO_4 + 4O_2 \rightarrow 2 HNO_3 + H_2SO_4 + 2H_2O$$
 (1)

The sulphuric acid that is formed can react with aluminium hydroxide in soil solution to form insoluble $Al(OH)SO_4$.

$$Al(OH)_3 + 2H^+ + SO_4^{2+} \rightarrow Al(OH)SO_4 + 2H_2O$$
⁽²⁾

The possibility of Al(OH)SO₄ formation in acid soils was reported by van Breemen³. He observed that there were changes of aluminium ion activity in solutions of different pH values. His findings have been confirmed by others^{2,24}. The formation of this compound is pronounced when there is abundant SO_4^{2+} ions. The reverse situation occurs when the SO_4^{2+} concentration decreases in soil solution or when the compound goes into solution through hydrolysis. The formation and dissolution of the compound therefore govern the behavioural pattern of H⁺, Al³⁺ and SO₄²⁺ movement in soil. This therefore supports the described retention and release mechanism of sulphates in acid soils²⁰. Prenzel²⁰ came up with a mechanism which demonstrates the temporary SO_4^{2+} retention in acid soil.

Besides Al(OH)SO₄ formation, SO₄²⁺ in the soil can be adsorbed. This can be either through the neutralization of positive charges, usually referred as non-specific²¹ or through the ligand exchange reaction (specific type) whereby the adsorbed ion must be able to donate or accept protons depending on the soil reaction^{7,10}. Other findings^{12,16} show that SO₄²⁺ adsorption is higher at lower soil layers than at the top (0–10 cm), a layer rich in humus.

Due to the retardation of the anion applied, the movement of the accompanying cations are also retarded to fulfill the electroneutrality. As a consequence, the BTCs of cations and anions differ considerably after the application of the two salts.

The salt balances (on charge basis) exhibit differences which can be explained by the internal transformations of matter in the soil. The input of the salts was dominated by the fertilizers which was in one case Ca^{2+} and NO_3^- and in the other case NH_4^+ and SO_4^{2+} with percentages of total salt input ranging between 81 and 96.

After 60 days, the outputs following the $Ca(NO_3)_2$ application was slightly lower than the input (0.43-0.13 Keq.ha⁻¹). This on a first view indicates that the soil behaved like an ion exchanger modifying the concentrations of the ions in the output according to their fractions in and at the binding strength to the soil. However from

LEACHING OF NUTRIENTS FROM FOREST SOIL

	Ca(NO ₃) ₂ a	application	(NH ₄) ₂ SO ₄	application
	N	H+	N N	H+
Input	5.72		5.72	
Fixation	-	-	1.5	_
Incorporation	0.86	- 0.86	2.0	+ 2.0
Denitrification	2.14	- 2.14	1.79	+ 1.79
Percolation Mineralization, nitrification	2.71	_	0.36	1.72
and percolation	1.70	+ 1.70	3.5	+ 3.5

Table 4. Internal nitrogen transformations, proton production and - consumption (kmol_c/ha)

the findings¹⁷ (see Table 4), we know that other processes occurred in the soil during the study. Only 2.7 Kmol_c/ha of the applied N was leached through the soil. 1.70 Kmol_c/ha of the nitrate came from the soil organic matter after mineralization and nitrification, forming an equivalent amount of protons. In the same time, 0.86 and 2.14 Kmol_c/ha H⁺ were consumed by organic regime and gaseous N respectively. This therefore leads to a net proton consumption or OH⁻ production of 1.30 Kmol_c/ha. In the output, these OH⁻ ions are completely displaced by SO₄²⁺ and Cl⁻ ions.

After application of $(NH_4)_2SO_4$, the situation was however different. Table 4 shows that most of the applied NH_4 -N was transformed to other compounds. The processes involved combined with proton formation, induce cation exchange and other chemical reactions. Due to the formation of HNO_3 , there was an excess H^+ production of $0.36 \text{ Kmol}_e/ha$. This, however, was only a small fraction of the possible maximum proton production of $5.72 \text{ Kmol}_e/ha$, which could have been produced had all the NH_4 -N applied been transformed to NO_3 -N. Besides the protons which came from the fertilizer, there was a production of $3.5 \text{ Kmol}_e/ha$ of HNO_3 from organic matter through mineralization and nitrification. This led to a total internal production of protons of $3.86 \text{ Kmol}_e/ha$ which in turn led to an increase of cation outputs of only $0.04 \text{ Kmol}_e/ha$ which means 3.82 Kmol of the total protons produced was consumed internally.

From this study, however, one is not able to give an accurate answer as to what type of processes took place in soil. Nevertheless, the results obtained showed that the processes and reactions induced by the fertilizer application are much more complex as the charge balances indicate.

In conclusion, the results of this experiment show that

1. not only the amount and quality of the applied cation influences

the leaching pattern of actions in an acid soil but also the accompanying anion.

2. nitrogen compounds both from fertilizer and soil are submitted to different processes in soil. Without the knowledge of the quantitites of N transformed in different processes, it is practically impossible to interpret leaching data properly.

3. the approach used, *i.e.* the measurement of change through breakthrough curves and the formation of a charge balance combined with tracer studies, gives a better insight of the multiple processes involved in N-fertilizer transformations and transport in soils.

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