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Kinetics and Leaching Study of Nitrate and Nitrite on Urea Hydrolysis in Alkaline Soil

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Authors' contributions

This work was carried out in collaboration between two authors. Author BS designed this study, literature search, collected soil samples, preparation of soil column and conduct all experimental works, wrote the protocol and wrote the first draft of the manuscript. Author AR managed the interpretation of results and analyses of the study. All authors read and approved the final manuscript.

Research Article

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ABSTRACT

Aims: The present research study describes leaching kinetics of NO_2^- and NO_3^- at different added urea concentrations on undisturbed columns of agriculture soil.
Study Design: This study design to understand solute transport in soil.
Place of Study: Samples are collected from the agriculture fields of Kota region and research work related to analysis is carried out at University of Kota. Rajasthan. India.
Methodology: The clay loam soil collected from agriculture fields of Kota, Rajasthan. India was dried and sieved for uniform particle size. Initial leaching rates of NO_2^- and NO_3^- ions have been calculated at different added urea concentrations and linear dependence of LR_{obs} and $[ion]_i$ has been derived. The leaching kinetics of nitrate and nitrite have been studied by determining the concentrations of NO_2^- and NO_3^- in the leachate with time using standard spectroscopic method by measuring the wavelength at 520 nm and 410 nm respectively. The leaching was carried out till ions are completely removed. Effect of Na and Ca level of percolating water have been studied on nitrate and nitrite leaching. Graphical equation of different kinetic models applied on results were tested with least square regression analysis.
Results: The log-log plot of $[ion]_i$ vs LR_{obs} are found to be second order for NO_2^- leaching and fractional order of more than one for NO_3^- leaching. On increasing Ca level of

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percolating water LR_{obs} found to decrease while in case of Na results are found to be reverse.

Conclusion: It is evident that urea-soil interaction generate nitrate and nitrite but initially soil was dried and incubation period of urea is only 24 hours therefore the conversion of added urea N into NO_3-N and NO_2-N is only less than 2 and 0.2% respectively. Fraction order rate in case of nitrate suggest that another path of conversion of nitrate into nitrite simultaneously occurs with its generation. Effect of irrigation water quality is also found to have significant effect on leaching rates of both the ions.

Keywords: Leaching; kinetics; urea; nitrate; nitrite; nitrification-denitrification; kinetic models.

1. INTRODUCTION

Urea is a miraculous agro chemical being applied worldwide to increase the production of rice and other crops enormously [1]. Urea consumption in Indian agriculture exceeded up to 20 metric tons during 2001 to 2002 [2] accounting for 80–83 percent of the total fertilizer N – consumption. When urea is applied to most aerobic and flooded soil, it hydrolyses to $(NH_4)_2CO_3$ by the enzyme urease, principally produced by soil microbes [3] depending upon the alkalinity and pH buffering capacity of soil, $(NH_4)_2CO_3$ decomposes and NH_3 gas escapes to atmosphere resulting in volatilization of ammonia [4]. In an another transformation, biochemical oxidation i.e. nitrification of NH_4^+ ion occurs in aerobic soil, during nitrification, ammonia is first converted to nitrite then from nitrite to nitrate by autotropic bacteria's viz nitrosomonas and nitrobactor respectively [5].

The use of nitrogen fertilizers at rate higher than the rate uptake by the plants, increase the potential for increased nitrate leaching as has been shown by several studies [6], it is evident that irrigated agriculture drains below the root zone is required to maintain the salt balance [7] The flow below the root zone can produce nitrate losses [8]. However the water flow and nitrate leaching depend on soil structure [9] i.e. porosity and aggregation, markedly affecting the cultivability and workability of clay soil [10]. Formation and the stability of clay aggregates are important factors for the reduction of slaking, generation of surface runoff, erodibility of soil and the transport of particular element in clay fields [11]. Leaching of solute also depend on amount of water applied by irrigation or natural precipitation and the amount, timing and species of fertilizer applied [12].

Studies conducted in different parts of the world have shown that farmers often use amount of nitrogen fertilizers that exceed the nitrogen requirement of crops [13]. Most of the studies conducted on nitrate leaching are from the regions where rain fall is abundant and well distributed [14] but studies under semiarid condition are scares.

As nitrate is a wide spread contaminant of ground and surface water worldwide, it posses a potential threat to human health specially to infant causing the condition known as methemoglobinmea [15]. Chronic consumption of high level of nitrate may also cause other health problems such as cancer and teratogenic effect, data are inconclusive but cause of concern [16].

Le Cadre and his co-worker [17] measure ammonia volatilization in the laboratory. Major concern has been given to the leaching losses of applied nitrogen from conventional nitrogenous fertilizers. The leaching losses of nitrogen from urea can occur by two processes (1) leaching of unhydrolyzed non toxic urea molecule when rainfall immediately follows urea application (2) hydrolysis and nitrification and denitrification and leaching as nitrate and nitrite. Among three major forms of N-fertilizer, nitrate ions are more susceptible to leaching while NH_4^+ ions is least and urea is moderately susceptible. Urea leaches at lower rate than nitrate because it can be weakly adsorbed on to soil and simultaneously hydrolyzed to ammonium compounds [18].

Rates of leaching and downward movement of urea N and NH_4^+-N in fine coarse textured soils of pH 7.8 and 7.9 have been measured by Singh and his co-workers [6]. However, studies on kinetics of leaching on undisturbed vertical columns are still lacking. Keeping the soil transformation reactions in mind the present study has been conducted to find out the conversion of urea into nitrate and nitrite in agriculture soil column during 24 hr. incubation. The detail kinetics of leaching of nitrite and nitrate during urea application has been investigated and compared using various kinetic models.

2. MATERIALS AND METHODS

The clay loam soil collected from agriculture fields of Kota, an industrial city of Rajasthan, India, was dried and sieved for uniform particle size. The physico-chemical properties of the experimental soil analyzed using Indian standard method (IS 2720) are given in Table.1.

Table 1. Physico-chemical characteristics of the column soil

Soil Properties	Values
pH	8.2
E. Ce (dsm$^-$	0.67
Bulk Density (mg m^{-3})	1.35
Colour	Grayish black
Sp. Gravity	2.57
Type	Clay-clay loam
Sand %	21.2
Silt %	26.2
Clay %	48
Cation exchange capacity	46.01
O.C. (g kg $^{-1}$)	0.65
Available N (kg ha^{-1})	232
Available P (kg ha^{-1})	17
Available K (kg ha^{-1})	291
Exchangeable cations (mg / 100 g	
a. Ca^{+2}	21.4
b. Mg^{+2}	12.8
c. Na^+	3.2
d. K^+	1.2

Agriculture soil used for experimental work was rich in leachable nitrate. A column of 60g of soil contained a 3.82 mg/kg leachable NO_2^- and 82 mg/kg NO_3^-. Soil with particle size ≥ 500 μm was used in the column as lower particle sized soil was rich in clay content thus restricted the preferential flow and resulted in clogging. Detailed leaching kinetics of nitrate and nitrite have been discussed in the following pages.

2.1 Experimental Set Up

Column of 60×3 cm^2 were prepared surrounded by glass jackets of continuously flowing thermo stated water,60 g soil was gently filled in the column. Clay loam soil was collected from agriculture fields of Kota region of chambal command area. Initially soil was dry, a fixed volume of urea solution with desired urea concentration was added on the top of the soil column in each experiment. The urea solution was allowed to get adsorbed uniformly for 24 hours, after which column was continuously leached with de-ionized water.

Fertilizer urea (NH_2CONH_2) of SD fine grade with 46% N was used, spectrophotometric method (Systonic-118) and ion selective electrode (6500 ion meter-Eutech) were used for analysis of nitrate and nitrite present in leachates. $CaCO_3$ and NaCl of A.R. grade and SD fine make were used for changing the Ca and Na levels of extractant water. The flow rate of extractant water was kept at 20 ± 2 ml/10min, water filled porosity (θ), at 0.34 cm^3cm^{-3}. pH and temperature of the percolating water were also kept effectively constant during the entire kinetic work. The results were found to be reproducible with in ± 10%.Water filled porosity determined by following formula.

θ = Volume of water retained by dry soil /Total volume of soil column

$$= \pi r^2 h / \pi r^2 h \qquad (1)$$
$$= 3.14\times1.5\times1.5\times2.9/3.14\times1.5\times1.5\times8.6 = 0.34\ cm^3$$

The pore volume of column soil can be calculated as

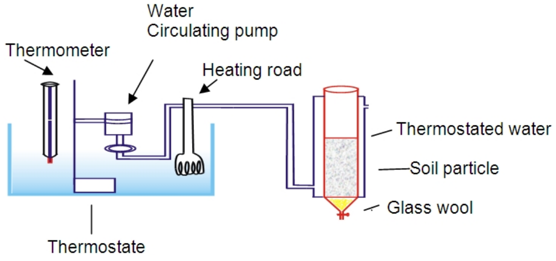
$$Pv = q't / \theta V \qquad (2)$$

Where

q' =Volume of effluent collected per unit time i.e. flow rate $cm^3\ h^{-1}$.
t =Time that has elapsed since the slug was introduced.
θ =water filled porosity cm^3cm^{-3}.
V =Total volume of soil column.

The schematic diagramme of glass column used for experimental work is given below.

Glass Column



3. RESULTS

3.1 Data Interpretation

The treatment of data is based on the calculations of the following parameters as defined below.

$[Ion]_s$ = leachable ions naturally present in the column soil.
$[NH_2CONH_2]_{ad}$ = urea concentration introduced in the soil column.
$[Ion]_i$ = total leachable ions present initially.
$[Ion]_t$ = leached concentration of ion at time t.
$[Ion]_t$ = $[Ion]_i – [Ion]_t$ = leachable concentration remaining at time t.

3.2 Estimation of Ions Generated from Soil-NH_2CONH_2 Interaction

Different concentrations of NH_2CONH_2 were added on identical columns. After 24 hr. incubation ions such as nitrate and nitrite were estimated in leachates collected by extracting the column with de-ionized water. NO_3^- ions are determined spectrophotometrically at a wavelength of 410 nm whereas NO_2^- ions are determined at a wavelength of 520 nm using standard procedure [19]. Total leachable concentration of nitrate $[NO_3^-]_i$ and nitrite $[NO_2^-]_i$ were estimated , N contents of NH_2CONH_2, NO_2^- and NO_3^- and its percentage conversion of added N into leachable N were calculated .Detailed results are given in Table 2.

Table 2. Added N levels of NH_2CONH_2 and their percentage conversion into leachable Nitrate $[NO_2^-]_i$ and Nitrite $[NO_3^-]_i$

$[NH_2CONH_2]_{add}$ g kg^{-1}		NO_2^- mg kg^{-1} s^{-1}			NO_3^- mgkg^{-1}s^{-1}		
Added	**Added N**	**$[NO_2^-]_i$**	**Leached N**	**% conversion**	**$[NO_3^-]_i$**	**Leached N**	**% conversion**
0.00	0.00	3.82	1.16	-	82	18.51	-
1.20	0.56	6.46	1.96	0.14	122	27.45	1.59
2.40	1.12	9.87	3.01	0.16	139	31.27	1.13
2.60	1.21	11.63	3.54	0.19	157	35.32	1.38
4.80	2.24	15.20	4.62	0.15	173	39.92	0.95
6.00	2.80	17.82	5.42	0.17	190	42.75	0.86
7.20	3.36	20.17	6.13	0.15	212	47.72	0.87

The values of total leachable concentrations of $[NO_2^-]_i$ and $[NO_3^-]_i$ at different $[Urea]_{ad}$ are given in Table 2. The soil itself has leachable NO_2^- and NO_3^- present in it. In the sequential calculations the total leachable content has been taken to be equal to the leachable content present initially for the determination of initial leaching rates. On adding NH_2CONH_2 from outside the concentration of leachable ions increases due to conversion of NH_2CONH_2 into NO_2^- and NO_3^- by multi step pathways.

3.3 Leaching Rate Profiles

For calculating initial leaching rates, the concentration of anions were determined in leachates collected periodically at an interval of 5 min. The leaching was carried out till the soluble anions were completely removed. After completely removing the soluble ions from the column, the soil of column was transfer into the beaker and a suspension was made with a known volume of de-ionized water. This soil suspension was stirred for two hours.

Table 3. Variation of $[NO_2^-]_i$ and $[NO_3^-]_i$ with LR_{obs}, for different $[NH_2CONH_2]_{ad}$

$[NH_2CONH_2]_{ad}$	0.00	1.20	2.40	3.60	4.80	6.00	7.20
$[NO_2^-]_i$	3.82	6.46	9.87	11.63	15.20	17.82	20.17
LR_{obs}, $mgkg^{-1}s^{-1}$	4.2×10^{-3}	1.06×10^{-2}	2.45×10^{-2}	3.56×10^{-2}	15.20×10^{-2}	10.39×10^{-2}	14.79×10^{-2}
$[NO_3^-]_i$	82	122	139	157	173	190	212
LR_{obs}, $mgkg^{-1}s^{-1}$	3.28	4.74	6.95	7.85	8.65	12.66	14.13

The ionic concentration were further determined in the filtrate of soil suspension to ensure the complete removal of soluble ion from the soil. Table 3 gives the variation of $[NO_2^-]_i$ and $[NO_3^-]_i$ with LR_{obs}, for different $[NH_2CONH_2]_{ad}$. LR_{obs} values were obtained from the initial slopes of the plots between $[Ion]_i$ and time as shown in Figs. 1 and 2 by Latshow method. It was calculated manually by using plane mirror.

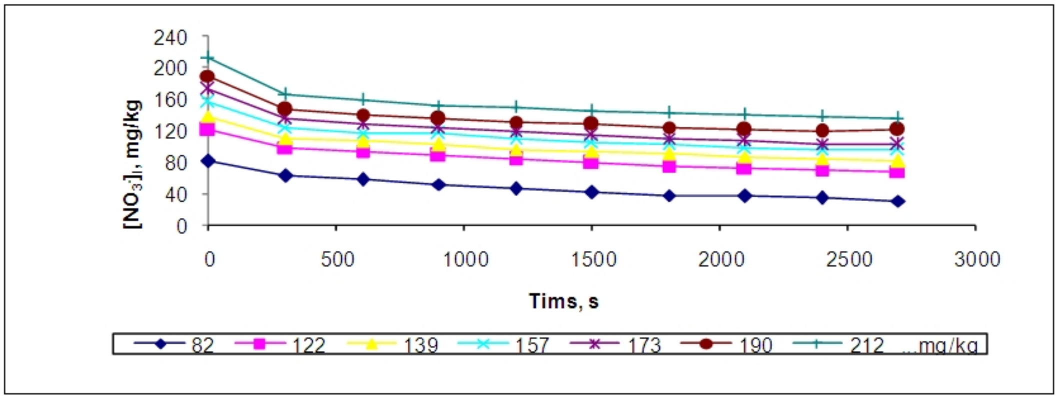


Fig. 1. Initial leaching rate profiles for NO_3^- leaching at different $[NO_3^-]_i$ at 30ºC during urea application

3.4 Dependence of LR_{obs} on $[Ion]_i$

It can be seen from the Tables 3 that on increasing $[Ion]_i$, LR_{obs} values are increases. The log-log plots of $[Ion]_i$ vs LR_{obs} are found to be second order for NO_2^- leaching and fractional order of greater than one for nitrate leaching as given in Table 4. The leaching rates fitted the following rate laws (equations 3 and 4).

$$\text{For Nitrite } LR_{obs} = k_1 [NO_2^-]_i^{\,n_1} \qquad (3)$$
$$\text{For Nitrate } LR_{obs} = k_2 [NO_3^-]_i^{\,n_2} \qquad (4)$$

The values of k and n were determined from log- log plots between rate and concentration terms of the rate laws

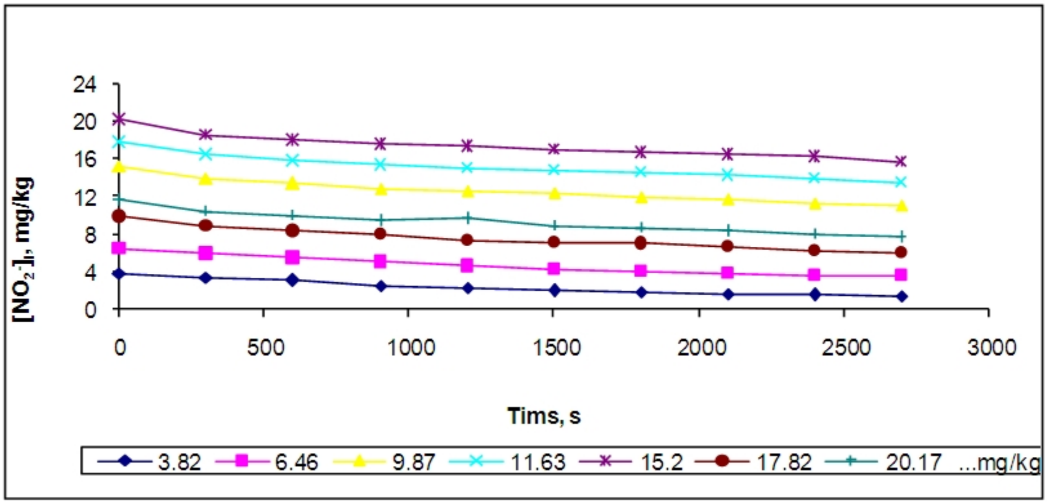


Fig. 2. Initial leaching rate profiles for NO_2^- leaching at different $[NO_2^-]_i$ at 30ºC during urea application

Table 4. The Vales of rate constant (k), order (n) and correlation parameters (r^2 and SEE) for NO_2^- and NO_3^- leaching during area application

Parameters	NO_2^-	NO_3^-
10^2k_1	0.696	5.62
n_1	2.00	1.60
r^2	0.983	0.957
SEE	0.078	0.051

Unit of k for NO_2^- = $mg^{-1}\ kg^{-1}\ s^{-1}$
Unit of k for NO_3^- = $mg^{-.60}\ kg^{-.60}\ s^{-1}$

3.5 Effect of Ca Levels of Extractant Water

Ca^{2+} level of extractant was varied from 500 to 2000 mg/l at fixed urea added concentration i.e. 2.40 g/kg of agriculture soil. Solubility of Ca^{2+} level increased by changing $CaCO_3$ into $CaCl_2$ by adding dilute HCl into required amount of $CaCO_3$. Figs. 3 and 4 shows variation in LR_{obs} with Ca levels of extractant H_2O.

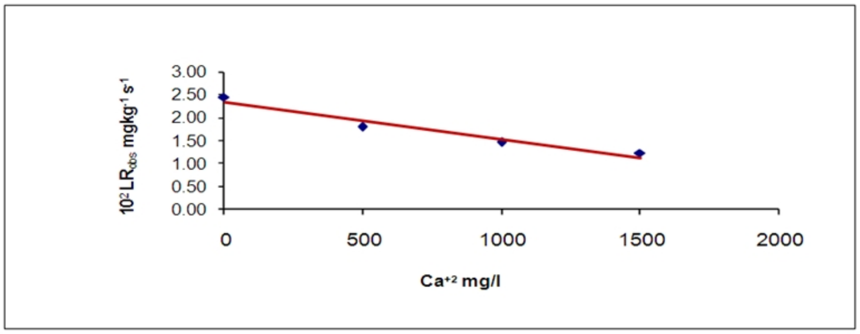


Fig. 3. Change in LR_{obs} with increase in Ca^{+2} concentration of extractant H_2O. For nitrite leaching $[NH_2CONH_2]_{ad}$ = 2.40 mg/kg, Temperature = 30ºC

$[NO_2^-]_i$ was found to decrease from 9.87 to 8.06 mg/kg and LR_{obs} for nitrite from 2.45 x 10^{-2} to 1.23 x 10^{-2} $mgkg^{-1}$ s^{-1} where as $[NO_3^-]_i$ was found to decrease from 139 to 99 mg/kg and LR_{obs} for nitrate from 6.95 to 3.30 mg kg^{-1} s^{-1} on increasing Ca levels of extractant water. Addition of Ca ions in the form of $CaCl_2$ in extracting water has retarded the initial rate of nitrate leaching, If irrigation water is hard, it may affect nitrate and nitrite leaching in two possible ways it may change the i) ion exchange capacity of soil [20] (ii) downward movement of nitrate and nitrite ions will be coupled with the hydrated Ca^{2+} ions. As the size of hydrated K^+ ion is smaller in comparison to Ca^{2+} hydrated ions, the rate of leaching of KNO_3 and KNO_2 is retarded in the presence of excessive Ca^{2+}ions.

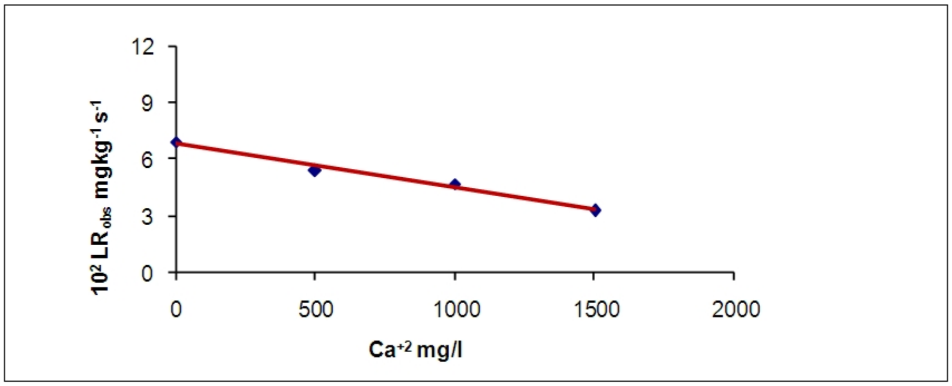


Fig. 4. Change in LR_{obs} with increase in Ca^{+2} concentration of extractant H_2O. For nitrate leaching $[NH_2CONH_2]_{ad}$ = 2.40 mg/kg, Temperature = 30ºC

3.6 Effect of Na levels of Extractant Water

To witness the role of Na content of irrigation water on leaching of nitrate and nitrite Na levels of extractant was varied from 50 to 250 mg/l by adding appropriate amount of NaCl in deionized water, salt NaCl is chosen because (i) high solubility of the salt in H_2O (ii) the column soil is saturated with Cl^- ions and are leached without affecting leaching rates of NO_2^- and NO_3^-. On increasing Na content [21] from 50 to 250 mg/l of extractant water, $[NO_2^-]_i$ was found to increase 1.30 to 2.17 mg/kg where as $[NO_3^-]_i$ increased from 9.87 to 12.52 mg/kg, LR_{obs} for NO_2^- leaching were increased 2.45 x 10^{-2} to 4.16 x 10^{-2}, mg kg^{-1} s^{-1} where as for nitrate leaching from 6.95 to 9.01 mg kg^{-1} s^{-1}. Figs. 5 and 6 shows variation of LR_{obs} with Na levels of extractant H_2O.

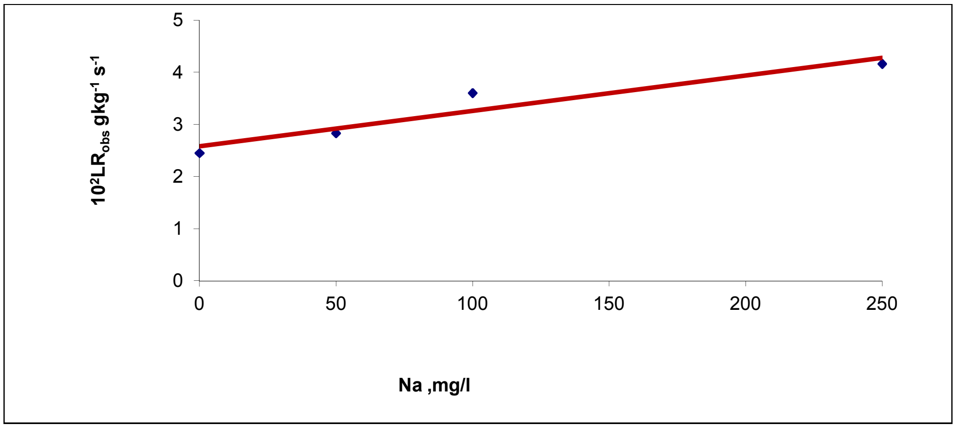


Fig. 5. Change in LR_{obs} with increase in Na levels of extractant $H_2O$for nitrite leaching $[NH_2CONH_2]_{ad}$ = 2.40 mg/kg, Temperature = 30ºC

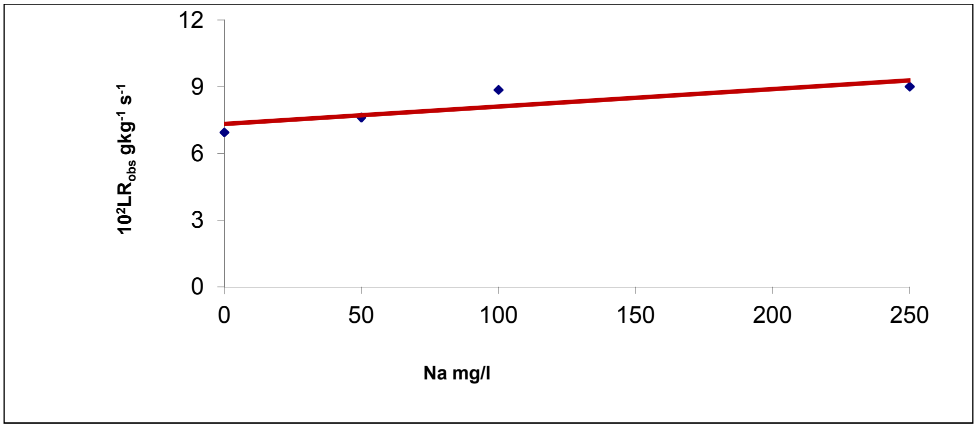


Fig. 6. Change in LR_{obs} with increase in Na levels of extractant H_2O for nitrate leaching $[NH_2CONH_2]_{ad}$ = 2.40 mg/kg,Temperature = 30ºC

On increasing the Na –content of percolating water, the rate of leaching of nitrate as well as total leachable content are found to increased. Na is the highly exchangeable cation in the soil systems; the high level of Na in irrigation water will mobilize adsorbed nitrate and nitrite by converting it into highly soluble sodium salt this increasing its leaching [22]. There is a decrease in LR_{obs} with increase in Ca $^{2+}$ level of extracting water and in case of Na^+, LR increased, this may be due to Na-Ca exchange reaction in soil [23].

3.7 Application to the Kintic Models

The data for the leaching of nitrite and nitrate on adding urea in alkaline Indian agriculture soil were used to find the best fit model, describing the kinetics of leaching. Seven kinetic model applied in our experimental conditions are.

Kinetic Models	Equations	References
1. Zero order	$C_0-C_t = a-bt$	Amachar & Baker [24]
2. First order	$\ln (C_0-C_t) = a-bt$	Jardine & Spark [25]
3. Second order	$1/C_t = 1/C_0 + kt$	Jardine &zelazny [26]
4. Third order	$1/C_t^2 = 1/C_0^2 + kt$	Chien &Mc Clellon [27]
5. Parabolic Diffusion	$C_t = a-bt^{1/2}$	Hodge & Johnson [28]
6. Elovich Equation	$C_t = a- b\ln t$	Hodges & Joneson [28]
7. Two constant rate equation	$C_t = a-b \ln t$	Jopeny & young [29]

All the models were tested with least square regression analysis, values of stastical

Parameters were calculated. Out of seven kinetic models only second and third order kinetic model are suggested to represent leaching kinetics in our experimental conditions.

4. DISCUSSION AND CONCLUSION

Results of this study shows that urea-soil interaction generates nitrate and nitrite although the conversion of added urea –N into NO_3^- -N and NO_2^- -N is only $<$ 1.61% and $<$ 0.17% respectively. Factors such as ammonia volatilization, urea leaching, microbial population, pH, electrical conductivity (EC), water content etc. determine nitrification & denitrification rates. Dry soil used for the work seems to have lower microbial population which retards % conversion of applied urea into nitrate and nitrite significantly. An increase in microbial population in wet soil can greatly increase NO_3^- -N and NO_2^- -N generation in soils.

Initial rate method successfully describe initial rate of leaching for both the ions. Rate of leaching of nitrite is quite lower in general than nitrate on equal added urea in identical columns obviously because of generation of nitrite in lower concentrations than nitrate. An order of 2 in case of nitrite and 1.6 in case of nitrate were calculated which describe the dependence of $[Ion]_i$ on LR_{obs}, which suggest more than one pathway involved in $[Ion]_i$ generation as well as controlling leaching.

Effect of irrigation water quality is also found to have significant effect on leaching rate of both the ions. Increase ion Ca level has decreased LR_{obs} as well as $[Ion]_i$ while increase in Na level has increased both the parameters. Out of seven kinetic model applied, only second and third order kinetic models are suggested to represent leaching kinetics in our experimental conditions. Present work is a pioneer investigation on nitrate and nitrite leaching in agriculture field soil of Kota region where urea is applied.

This study concludes that urea in the agriculture fields generate nitrate and nitrite in leachable amounts and factors such as ionic concentration, soil macropores, water content, temperature Ca and Na levels of irrigation water affect the leaching rates.

COMPETING INTERESTS

The authors declare that they have no competing interests.

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