

***In situ* measurements and numerical simulation of nitrate leaching from farm soil**

N. YAMASHITA & S. SUGIO

Graduate School of System Engineering, Miyazaki University, 1-1 Gakuen-kibanadai-Nishi, Miyazaki 889-2192, Japan

e-mail: sugio@civil.miyazaki-u.ac.jp

Abstract The unconfined aquifers in the southern part of Kyushu Island in Japan are contaminated by $\text{NO}_3\text{-N}$. It is considered that the contamination is caused by seepage of rainwater with $\text{NO}_3\text{-N}$ dissolved from the barnyard manure on the farms. To investigate $\text{NO}_3\text{-N}$ leaching in the surface soil at an actual farm, *in situ* measurements of $\text{NO}_3\text{-N}$ concentrations in soil moisture within 3 m of the ground surface were carried out. Moreover, one-dimensional $\text{NO}_3\text{-N}$ leaching from soil was calculated numerically to simulate the observed leaching. From these results, it was clarified that $\text{NO}_3\text{-N}$ dissolved from the barnyard manure and the fertilizer was leaching in the surface soil at high concentrations.

INTRODUCTION

The southern part of Kyushu Island in Japan has over 2500 mm of annual precipitation and also consists of thick aquifers with high permeability. All drinking water resources in this part depend on the groundwater resources. However, groundwater contamination by $\text{NO}_3\text{-N}$ has been detected. According to previous investigations, one of the causes of this contamination was over-use of barnyard manure on the farms (Gillham & Webber, 1969; Yamashita & Sugio, 1999). The requirement for groundwater conservation in this part of the island is to understand $\text{NO}_3\text{-N}$ percolation from farms. Studies using numerical models were introduced in several fields but only some of them were validated against *in situ* measurement data. In this study, *in situ* measurements of $\text{NO}_3\text{-N}$ concentration were carried out, and these results were simulated approximately by means of numerical simulation.

$\text{NO}_3\text{-N}$ CONCENTRATIONS OF SOIL MOISTURE IN THE FARM

$\text{NO}_3\text{-N}$ concentrations of soil moisture within 3 m of the ground surface were measured at a farm in the study area. Porous cups were used to collect the soil moisture. They were arranged in two rows: a west row and an east row. In each row, porous cups were buried at depths of 25, 50, 75, 100, 150, 200, 250 and 300 cm in the soil as shown in Fig. 1. *In situ* measurements were started in April 1998 and forage crops were cultivated on the farm. Table 1 shows the application dates and the quantities as nitrogen of both the manure and the chemical fertilizer, and Fig. 2 shows daily rainfall. From the measurements, it was found that high concentrations of $\text{NO}_3\text{-N}$ were detected as shown in Fig. 3 and the maximum $\text{NO}_3\text{-N}$ concentration value

reached 280.8 mg l⁻¹ at 50 cm depth. The vertical distributions of NO₃-N concentration measured on 21 July and 11 September seem to have not changed (Fig. 3). This shows that NO₃-N leaching depends strongly on rainfall conditions.

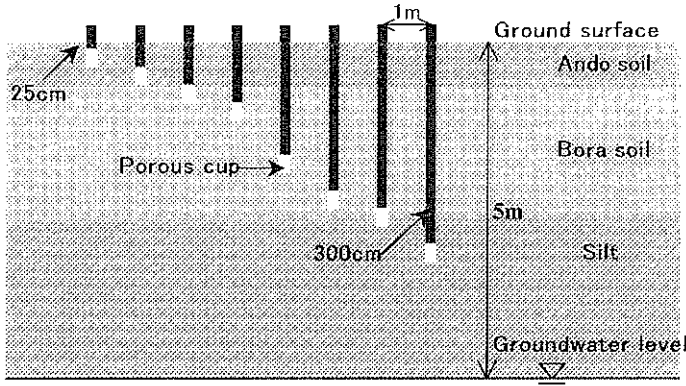


Fig. 1 Schematic diagram of soil moisture samplers.

Table 1 Application dates and quantities of manure and fertilizer nitrogen applied to the farm.

Date	1 March 1998	28 April 1998	28 April 1998	21 May 1998	7 August 1998
Activity in the farm	Manuring	Seeding	Fertilizing	Fertilizing	Harvest
Nitrogen applied (kg ha ⁻¹)	227	--	68	107	--

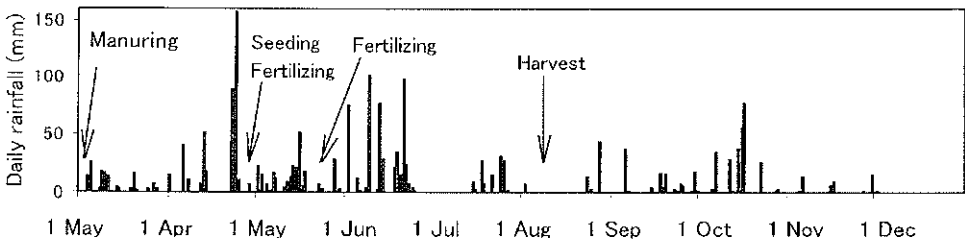


Fig. 2 Daily rainfall and agricultural activities at the farm.

PHYSICAL CHARACTERISTICS OF THE FARM SOILS

The surface soil at the observed farm consists of three kinds of volcanic soil containing plenty of micro-organisms and moisture. To evaluate the hydraulic parameters such as hydraulic conductivity, porosity and the unsaturated seepage characteristics, experimental measurements using undisturbed soil samples and *in situ* infiltration tests were carried out (Sugio & Okabayashi, 1994). Unsaturated seepage characteristics were expressed by the parameters of the van Genuchten equation (van Genuchten, 1980) as the following equations:

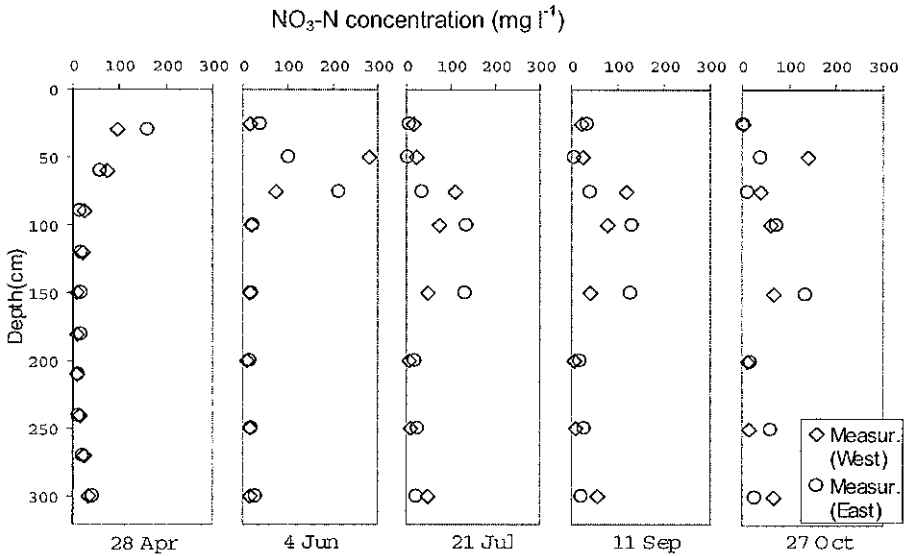


Fig. 3 Vertical distribution of NO₃-N concentration in the soil at the west side and the east side of the farm.

$$S_e = \left[1 / \left(1 + |\alpha \psi|^n \right) \right]^m, \quad k_u / k_s = S_e^{1/2} \left[1 - \left(1 - S_e^{1/m} \right)^m \right]^2, \quad m = 1 - 1/n \tag{1}$$

$$S_e = (\theta - \theta_r) / (\theta_s - \theta_r)$$

where ψ is capillary suction head, k_u is unsaturated hydraulic conductivity, k_s is saturated hydraulic conductivity, θ is volumetric water content, θ_s is saturated water content approximated as the value of porosity and θ_r is residual water content. Table 2 shows the parameters identified from their measurements or tests.

Table 2 Unsaturated seepage parameters.

	Ando soil	Bora soil	Silt
k_s (cm h ⁻¹)	1.12×10^{-3}	4.86×10^{-3}	1.05×10^{-5}
θ_s	0.66	0.65	0.80
θ_r	0.36	0.50	0.66
α	0.0222	0.0258	0.0222
n	2.8	3.2	2.8

NUMERICAL SIMULATION OF NO₃-N LEACHING

To understand NO₃-N percolation in the farm, one-dimensional unsaturated seepage flow of rainwater was simulated first. The governing equation for the unsaturated seepage flow is expressed by:

$$C_w(\psi) \partial \psi / \partial t = \nabla(k_u(\psi) \nabla \psi - k_u(\psi)) \tag{2}$$

where C_w is specific moisture capacity. As the boundary condition, the observed daily rainfall was applied at the soil surface and the groundwater table was assumed to be fixed at 5 m in depth. Evapotranspiration processes were not considered.

The leaching processes of $\text{NO}_3\text{-N}$ were simulated by NITRAT, which is one of the useful numerical models for simulating multispecies transport in saturated groundwater flow (Kinzelbach & Schäfer, 1991). NITRAT was applied to the individual field because the volumetric water content of the surface soil of the farm can be approximated as saturated due to the weather characteristic of the study area. This model describes heterotrophic processes, which take into account the interaction of oxygen, nitrate, organic carbon and bacteria. The governing equations are:

$$\partial \text{NO}_{3,w} / \partial t = -\nabla(\text{UNO}_{3,w}) + \nabla(D\nabla\text{NO}_{3,w}) - \beta(\text{NO}_{3,w} - \text{NO}_{3,bio}) \quad (3)$$

$$\partial \text{O}_{2,w} / \partial t = -\nabla(\text{UO}_{2,w}) + \nabla(D\nabla\text{O}_{2,w}) - \beta(\text{O}_{2,w} - \text{O}_{2,bio}) \quad (4)$$

$$\partial \text{C}_{org,w} / \partial t = -\nabla(\text{UC}_{org,w}) + \nabla(D\nabla\text{C}_{org,w}) - \beta(\text{C}_{org,w} - \text{C}_{org,bio}) \quad (5)$$

$$\partial \text{NO}_{3,bio} / \partial t = -\gamma_{den} / Y_{den} - \gamma_{aer} / Y_{aer} + \beta(\text{NO}_{3,w} - \text{NO}_{3,bio}) \quad (6)$$

$$\partial \text{O}_{2,bio} / \partial t = -\gamma_{aer} / Y_{aer} + \beta(\text{O}_{2,w} - \text{O}_{2,bio}) \quad (7)$$

$$\partial \text{C}_{org,bio} / \partial t = -\gamma_{den} / Y_{den} - \gamma_{aer} / Y_{aer} + f_{use} \gamma_{dec} + \beta(\text{C}_{org,w} - \text{C}_{org,bio}) \quad (8)$$

$$\partial (X_{aer} + X_{den}) / \partial t = \gamma_{aer} + \gamma_{den} - \gamma_{dec} \quad (9)$$

where NO_3 , O_2 , C_{org} and X are the concentrations of nitrate-nitrogen, oxygen, organic carbon and bacteria, respectively; the subscripts w , bio , aer and den indicate pore water, biophase, aerobic and denitrifying, respectively; U is the seepage velocity, D ($= a_L U$) is the dispersion coefficient, a_L is the dispersion constant, β is the exchange coefficient between pore water and biophase, Y is the yield coefficient, f is utilizable portion of dead bacteria, and γ is the growth rate of bacteria which is also expressed by the following equations:

$$\gamma_{aer} = \mu_{max}^{aer} [1 - F(\text{O}_2)] \frac{\text{C}_{org}}{(K_c^{aer} + \text{C}_{org})} \frac{\text{NO}_3}{(K_{NO_3}^{aer} + \text{NO}_3)} \frac{\text{O}_2}{(K_{O_2}^{aer} + \text{O}_2)} X_{aer} \quad (10)$$

$$\gamma_{den} = \mu_{max}^{den} F(\text{O}_2) \frac{\text{C}_{org}}{(K_c^{den} + \text{C}_{org})} \frac{\text{NO}_3}{(K_{NO_3}^{den} + \text{NO}_3)} X_{den} \quad (11)$$

$$\gamma_{dec} = \lambda (X_{aer} + X_{den}) \quad (12)$$

where μ_{max} is maximum growth rate of bacteria, K_i is half-velocity concentration for species i , λ is the first-order decay coefficient, $F(\text{O}_2)$ is oxygen dependent weight.

The initial concentrations of each species and the boundary condition of organic carbon and oxygen at the ground surface are set as shown in Table 3 and the values of biological parameters used in NITRAT are also set as shown in Table 4. These parameters were identified from the results of column experiments using Ando soils (Waner & Gujer, 1984; Sugio, 1997). In the calculation of $\text{NO}_3\text{-N}$ leaching, the mean value of seepage velocities calculated every 10 days at five distributed intervals (the depths 35, 35, 50, 90 and 90 cm, respectively) were applied to the convection term of the governing equations.

The boundary condition of $\text{NO}_3\text{-N}$ at the soil surface must coincide with the actual application dates and the quantities applied. In the Ando soil developed at the top of the

Table 3 Initial and boundary values applied to NITRAT.

	NO_3	O_2	C_{org}
Initial values ($mg\ l^{-1}$)	20	0.5	10
Boundary values at the ground surface ($mg\ l^{-1}$)	–	10	10

Table 4 Biological parameters used in NITRAT.

Parameters	Values	Parameters	Values	Parameters	Values
β	10.0	X ($mg\ l^{-1}$)	0.3	K_C^{aer} ($mg\ l^{-1}$)	0.76
Y_{den}	0.4	a_L (m)	0.01	K_C^{den} ($mg\ l^{-1}$)	0.5
Y_{aer}	0.09	$K_{NO_3}^{aer}$ ($mg\ l^{-1}$)	7.0	μ_{max}^{aer} ($mg\ l^{-1}$)	0.64
f_{use}	0.9	$K_{NO_3}^{den}$ ($mg\ l^{-1}$)	7.0	μ_{max}^{den} ($mg\ l^{-1}$)	0.3
λ (day^{-1})	0.2	$K_{O_2}^{aer}$ ($mg\ l^{-1}$)	0.6		

farm, the nitrogen in NH_4 -N and NO_2 -N are oxidized quickly by biological nitrification and are changed to NO_3 -N in the soil; the NO_3 -N is then leached under the ground with the seepage of rainwater (Sugio & Imamura, 1991). Therefore, boundary concentrations at the soil surface were applied as follows: the rainwater dissolves barnyard manure NO_3 -N at a concentration of $300\ mg\ l^{-1}$, and the chemical fertilizer at $600\ mg\ l^{-1}$, and the forage crops absorb 50% of NO_3 -N after seeding. Based on these assumptions, the dissolution periods of barnyard manure and fertilizer are evaluated as shown in Table 5.

Table 5 Dissolution periods of manure and fertilizer, and accumulated rainfall.

Manuring or fertilizing date	1 March 1998	28 April 1998	21 May 1998
Dissolution periods (day)	70 (up to 5 May)	15 (up to 12 May)	20 (up to 9 June)
Accumulated rainfall (mm)	619.0	113.3	178.3

SIMULATED RESULTS

Figure 4 shows the measured and simulated vertical distribution of NO_3 -N concentration in the farm. Although the unsaturated percolation of NO_3 -N was simulated approximately by NITRAT when applying the mean value of the unsaturated seepage velocity, the simulated results agreed well with the measured results. According to the simulated results, two concentration peaks are generated on 28 April after manuring on 3 March. Rainfall occurring between the manuring date and the middle of April leads to the lower peak, and heavy rainfall at the end of April leads to the upper peak (Fig. 2). The number of concentration peaks changes from three on 4 June to one on 27 October with leaching from soil below ground level. Therefore, it is clear that some of the concentration peaks are generated in the surface zone after manuring combined with the influence of a rainfall event, and that these peaks are balanced by leaching below the surface.

It is important to note that one of the causes of groundwater contamination is over application of barnyard manure in the study area. An attempt was made to estimate the

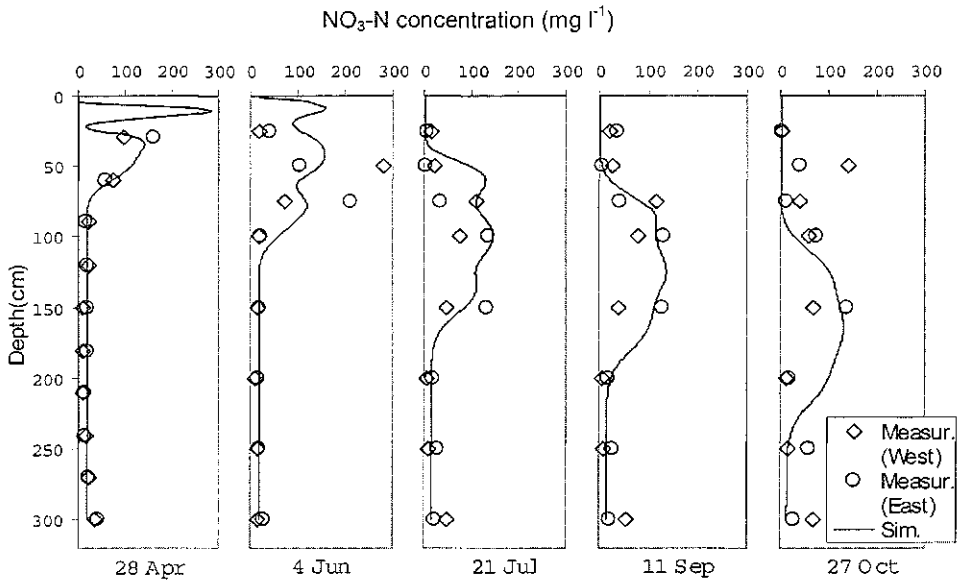


Fig. 4 Comparison of simulated results of NO₃-N concentration with measured results at the farm.

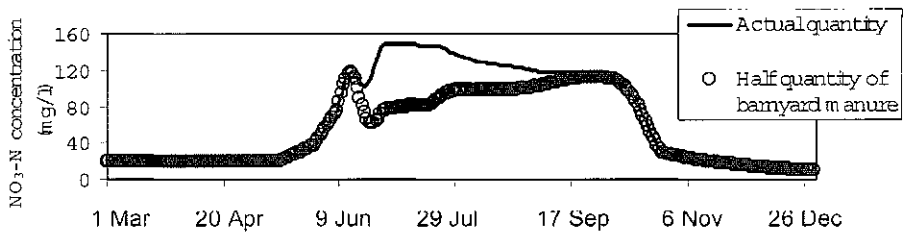


Fig. 5 Simulated NO₃-N concentration at 100 cm depth in half quantity manuring case.

effect of reduced applications of barnyard manure by means of simulation. Figure 5 shows a result of the daily variation of NO₃-N concentration at a depth of 100 cm. The solid line shows NO₃-N concentration in the case of the actual quantity of the barnyard manure and the fertilizer. The circle marks show it in the case of the same sources but with only half the quantity of barnyard manure. The results for the reduced application case show a lower peak compared to the actual case. The lower peak is considered to be caused by reduction of the dissolution periods of manure and by separation to dissolution periods of fertilizer. The result indicates that NO₃-N concentration in the soil can be reduced by considering appropriate dates and quantities for manuring.

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