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# NITRATE LEACHING UNDER INTENSIVE FARMING AND SUCH INFLUENCING FACTORS AS TYPE OF FERTILIZER AND SOIL

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## ABSTRACT

Nitrate ( $\text{NO}_3$ ) leaching from agricultural area is related to ground and surface water contamination.  $\text{NO}_3$  leaching is greatly affected by types of fertilizer. Excessive N for crops derived from two chemical fertilizers (ammonium nitrogen and polyolefin-coated urea) caused substantial  $\text{NO}_3$  leaching, which was predicted by an OECD model that calculates the risk of groundwater contamination. Organic N from compost appeared to accumulate in topsoil over several years, but  $\text{NO}_3$  leaching reached the same level as that in the chemically fertilized in the following years. Since preferential flow in soil also influences  $\text{NO}_3$  leaching, soil types should be evaluated with leaching experiments and models. Preferential flow in clay monoliths was well described by a modified tanks-in-series model through the parameters  $n$  and  $f_m$ , that is, in terms of the degree of mixing of water in soil and the ratio between mobile regions and the maximum amount of water in a monolith, when freely drained. Further studies are necessary to establish the best management of agro-ecosystems to maintain agricultural productivity and the environmental quality.

## KEYWORDS

Coated urea; compost; leaching; monolith lysimeter; nitrate; preferential flow

## INTRODUCTION

$\text{NO}_3$  leaching from arable land, which causes contamination of groundwater, has become a matter of worldwide concern. In Japan,  $\text{NO}_3\text{-N}$  concentrations in 6.3% of the groundwater investigated in 1998 exceeded  $10 \text{ mg L}^{-1}$ , a Japanese standard for groundwater and agricultural land was reported to be a major source (Environment Agency, 1999).

Nitrogen is intensively applied to Japanese agricultural land as a major essential nutrient. Nitrogen forms in fertilizer are ammonium ( $\text{NH}_4$ ),  $\text{NO}_3$ , urea, etc. Compost made by livestock waste or sewage sludge is also regarded as N fertilizer and its adequate application to arable land is recommended for recycling valuable nutrient resources.  $\text{NH}_4$  is a product of mineralization of organic N.  $\text{NH}_4$  derived from inorganic and/or organic fertilizer is easily oxidized into  $\text{NO}_3$  by microbes in upland fields.  $\text{NO}_3$ , which is an anion, is not absorbed on to soil and is freely leached out of root zones, because soil has usually negative charge.

The climate in most of Japan belongs to the category of the Asian monsoon, in which averaged annual rainfall reaches 1800 mm (Kuwabara, 1989) and heavy rainfall events often occur with typhoons in summer. This relatively high precipitation may increase  $\text{NO}_3$  leaching from agricultural area. It is necessary to establish the best agricultural management to maintain both high yields/quality of crops and the environmental quality. The effects of different N fertilizers and soil types on  $\text{NO}_3$  leaching were discussed in the present paper.

## NITRATE LEACHING FROM DIFFERENT TYPES OF FERTILIZERS

Slow-release fertilizers, such as polyolefin-coated urea, have been used to save labor from frequent application of chemical fertilizer and to increase N recovery rates for crops in Japan (Saigusa *et al.*, 1993). Substitution of slow-release fertilizer for readily available conventional fertilizer suppressed N leaching in short-term studies (Sakata *et al.*, 1995; Matsumaru, 1997). Both of them noted that N derived from slow-release fertilizer remained in the topsoil at the end of the experiments, but its later leaching was not examined. Although  $\text{NO}_3\text{-N}$  concentration in subsurface flow under pastures treated with readily available fertilizer increased more rapidly than that with slow-release fertilizer,  $\text{NO}_3\text{-N}$  concentrations from both types of fertilizer treatment continued to increase even after 10 years (Owens *et al.*, 1992). These results indicate the importance of long-term research.

Excessive use of livestock manure is also related to groundwater contamination (Thomsen *et al.*, 1993; Adams *et al.*, 1994; Chang and Entz, 1996).  $\text{NO}_3$  leaching is affected by manure types with different amounts of available N (Beckwith *et al.*, 1998). Compost, which has a great advantage in handling, reducing offensive odors, and eliminating pathogens, is a major product of livestock waste in Japan. Because most livestock farmers do not have sufficient farm land for recycling waste, excessive waste must be transported to other farmers (Harada and Yamaguchi, 1998). Accordingly, livestock compost has been applied not only to pastures but also to vegetable fields, where fertilization is usually more intensive. Residual N from previously applied manure is mineralized and may be available for leaching in continuously managed fields (Angle *et al.*, 1993; Thomsen *et al.*, 1993; Bergstöm and Kirchmann, 1999).

$\text{NO}_3$  leaching was studied in an Andisol treated with 4 N fertilizers (AN: ammonium N at  $400 \text{ kg N ha}^{-1}\text{y}^{-1}$ , CU: polyolefin-coated urea at  $400 \text{ kg N ha}^{-1}\text{y}^{-1}$ , SC: swine compost at a rate of  $800 \text{ kg N ha}^{-1}\text{y}^{-1}$ , or NF: no fertilizer) for 6 years (Maeda *et al.*, 2001a). The application rate of SC was determined on the assumption that 50% of N in SC is available for crops (Harada, 1997). Andisols (volcanic ash soils) cover more than half of the upland fields in Japan. Sweet corn was grown in summer, followed by Chinese cabbage or cabbage in autumn each year.

$\text{NO}_3\text{-N}$  concentrations in soil water at 1-m depth rose markedly in the summer of the second year and then fluctuated between  $30$  to  $60 \text{ mg L}^{-1}$  in the CU and AN plots (Fig. 1).  $\text{NO}_3\text{-N}$  concentration in the SC plot began to increase in the fourth year, reaching the same level as that in the CU and AN plots in the beginning of the sixth year. In the NF plot,  $\text{NO}_3\text{-N}$  concentration was about  $10 \text{ mg L}^{-1}$  for the first 4 years and decreased to  $5 \text{ mg L}^{-1}$  toward the end to the experiment.

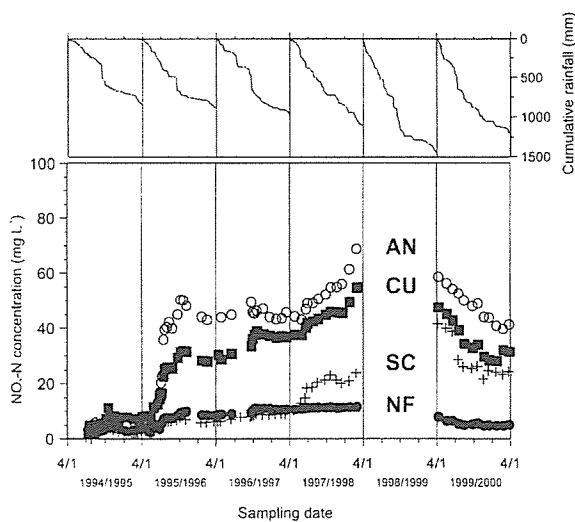


Fig. 1.  $\text{NO}_3\text{-N}$  concentration at 1-m depth under 4 different N treatments and cumulative precipitation during the experimental periods. AN, CU, SC, and NF represent ammonium nitrogen, coated urea, swine compost, and no fertilizer, respectively.

The highest  $\text{NO}_3\text{-N}$  concentrations were observed in the spring of 1998 and the lowest in the winter of

1999/2000 in the AN and CU plots during the experimental period after the summer of the second year (1995/1996) when the influence of fertilization clearly appeared. The highest precipitation, recorded in 1998/1999, may have caused the lower NO<sub>3</sub>-N concentration in the following year of 1999/2000, because more humid conditions contribute to water dilution of NO<sub>3</sub>-N and denitrification.

To evaluate the NO<sub>3</sub>-N concentration in soil water at 1-m depth, a simple equation suggested by OECD was used (OECD, 1999). OECD authorized the following method calculating the risk of water contamination by nitrogen, based on N and water balance on the soil surface. This model is used for comparing the risk among countries or local areas in a country (OECD, 1999).

$$PNC = \frac{PNP}{EW} \quad (1)$$

where *PNC* (mg L<sup>-1</sup>) is the potential NO<sub>3</sub>-N concentration, *PNP* (mg ha<sup>-1</sup>) the potential NO<sub>3</sub> present, and *EW* (L ha<sup>-1</sup>) excess water. N surplus in topsoil can be used as *PNP* at a preliminary step (OECD, 1999). *PNP* was obtained by subtracting the mean N uptake from the mean N application in the present study.

As a result, *PNC*s were 38, 44, and 109 mg L<sup>-1</sup> for the CU, AN, and SC plots, respectively. The *PNC*s satisfactorily predicted NO<sub>3</sub>-N concentration in the AN and CU plots (Fig. 1). In contrast, the calculated *PNC* in the SC plot was substantially overestimated, presumably because little organic N from SC was available for NO<sub>3</sub> leaching during the experimental period. In other words, excessive N for crops from AN and CU would cause NO<sub>3</sub> leaching in due time. On the other hand, organic N from SC appeared to accumulate in topsoil during the first several years; yet the accumulation of the organic N may cause late N mineralization in soil, resulting in an increase in NO<sub>3</sub>-N concentration after the fourth year of the experiments and later (Fig. 1).

To examine NO<sub>3</sub> fate in deeper soil near groundwater, NO<sub>3</sub> distribution in deeper soil was examined in the AN plot after 6-year continuous fertilizer application (Maeda *et al.* 2001b). NO<sub>3</sub>-N concentration in soil water declined with depth (Table 1). NO<sub>3</sub>-N concentrations in deeper than 3 m were almost equal to that in groundwater (Table 1). Groundwater table fluctuated between 120 cm and 400 cm during the experimental period. On the sampling date, the groundwater table existed around 3.6 m below the ground. The groundwater table rose to within 2-m depth a few times in summers with heavy rainfall. Probably, rising groundwater washed away NO<sub>3</sub> in the soil solution and resulted in decrease in NO<sub>3</sub> concentration in the deeper layers.

**Table 1.** NO<sub>3</sub>-N and Cl concentrations, mole ratios of NO<sub>3</sub>-N to Cl, and δ<sup>15</sup>N values in soil water sampled from different soil depths and groundwater in the ammonium nitrogen (AN) plot

	NO <sub>3</sub> -N	Cl	NO <sub>3</sub> -N/Cl	δ <sup>15</sup> N
cm	(mg L <sup>-1</sup> )	(mg L <sup>-1</sup> )	(mol / mol)	(‰)
100	40.8 ± 8.9	65.7 ± 7.6	1.6	2.2 ± 0.6
200	46.5 ± 7.5	56.4 ± 9.0	2.1	-0.7 ± 0.6
250	9.6 ± 4.0	28.9 ± 3.1	0.8	-0.5 <sup>†</sup>
300	3.8 ± 0.6	23.1 ± 0.8	0.4	8.9 <sup>†</sup>
350	3.2 ± 0.1	23.1 ± 0.7	0.4	7.6 <sup>†</sup>
400	3.2 ± 0.0	22.4 ± 1.5	0.4	7.6 <sup>†</sup>
430	3.5 ± 0.6	22.7 ± 1.5	0.4	9.1 ± 0.6
groundwater	3.9 ± 1.0	23.7 ± 0.6	0.4	6.8 ± 0.6

All water samples were obtained on 18 April 2000. N=2-5. †: No replicate because of insufficient amount of water.

Cl concentrations decreased with depth as well as NO<sub>3</sub>-N but the rate of the decline for NO<sub>3</sub>-N was larger than that for Cl (Table 1). The δ<sup>15</sup>N for soil water deeper than 3 meters also showed higher values than that at 1-m depth (Table 1). δ<sup>15</sup>N value (‰) is expressed as the following equation:

$$\delta^{15}N = \left( \frac{R_{sample}}{R_{standard}} - 1 \right) \times 1000 \quad (2)$$

where *R* is the ratio of <sup>15</sup>N/<sup>14</sup>N and the standard is atmospheric N<sub>2</sub>. δ<sup>15</sup>N value of the substrate becomes

higher through microbial reaction because the heavy isotope of N,  $^{15}\text{N}$ , becomes relatively enriched in the residual fraction of the substrate. Namely,  $\delta^{15}\text{N}$  increases through reactions such as denitrification (Yoneyama, 1987). Böttcher *et al.* (1990) found decreasing  $\text{NO}_3$  concentrations with groundwater depth correspond to the increase in  $\delta^{15}\text{N}$  values, which reveals the significance of denitrification in the decrease of  $\text{NO}_3$  concentration. Hence, anaerobic conditions resulting from rising groundwater may have caused denitrification in the present study.

Böttcher *et al.* (1990) attempted to investigate the contribution of denitrification to the decrease in  $\text{NO}_3\text{-N}$  concentration by use of the following Rayleigh equation (Mariotti *et al.*, 1981):

$$\delta^{15}\text{N} = \delta^{15}\text{N}_{ini} + \varepsilon \ln f_{\text{NO}_3\text{-N}} \quad (3)$$

where  $\delta N_{ini}$  is the initial value of  $\delta^{15}\text{N}$  for the substrate (‰),  $\varepsilon$  an enrichment factor (‰), and  $f_{\text{NO}_3\text{-N}}$  the fraction of unreacted residual substrate, which is obtained to divide substrate concentration by initial substrate concentration. The enrichment factor  $\varepsilon$  in groundwater was -16‰ (Böttcher *et al.*, 1990), although this factor in the previous studies ranged from -7.8 to -30‰ (Arai and Tase, 1992). Assuming that  $\varepsilon$  is -16‰ and  $\text{NO}_3\text{-N}$  at 2-m depth ( $47 \text{ mg L}^{-1}$ ,  $\delta^{15}\text{N}_{ini} = -0.7\text{‰}$ ) moved to that at 3-m depth ( $\delta^{15}\text{N} = -8.9\text{‰}$ ),  $\text{NO}_3\text{-N}$  concentration at 3-m depth will be expected to be  $26 \text{ mg L}^{-1}$ , which is by far greater than the measured value of  $3.8 \text{ mg L}^{-1}$ , indicating the contribution of denitrification was probably small in this study.

## SOIL STRUCTURE AND NITRATE LEACHING

$\text{NO}_3$  leaching is affected by preferential water flow in soil. Preferential flow refers to macropore flow, fingering, and funneled flow (Steenhuis *et al.* 1995). Macropores, which reflect soil structure, root decay, wormholes, etc., constitute preferred flow pathways for infiltrating water in soil. Fingering occurs as a result of wetting front instability such as change in hydraulic conductivity with depth and compression of air ahead of the wetting front. Funneled flow takes place when sloping geological layers cause pore water to flow laterally, accumulating at a low region.

To evaluate  $\text{NO}_3$  leaching in soils where preferential flow may occur, undisturbed soil should be used and exposed to natural field conditions. A new drilling method for collection of soil monoliths up to 100 cm depth was developed in Sweden (Persson and Bergström, 1991) and modified by my group in Japan to attach it on any tractors (Photo 1). The drill consists of a steel cylinder with four mounted cutting teeth at the bottom, into which a PVC tube (286 mm i.d.) is inserted. The drill was attached on the rear of a tractor. The hydraulic power of the tractor was used for pushing, lifting, and rotating the drill.

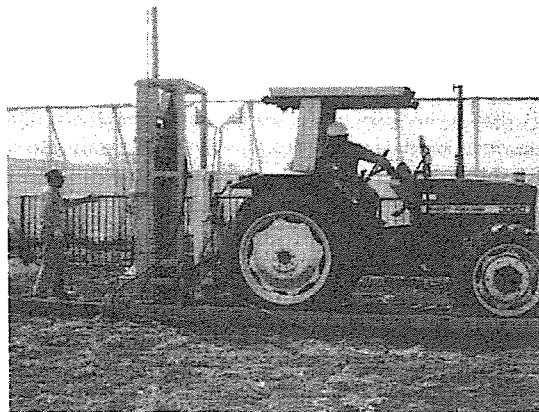
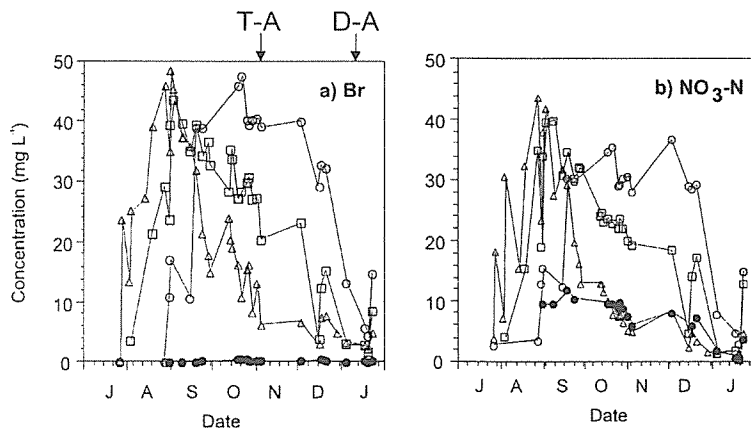


Photo 1. A device, which uses hydraulic system of a tractor, for collecting soil monoliths.

Leaching rates of N were measured in 500-mm deep monolith lysimeters containing a heavy clay soil (Maeda and Bergström, 2000). Nitrogen was applied at a rate of  $100 \text{ kg ha}^{-1}$  ( $50 \text{ kg NO}_3\text{-N ha}^{-1}$  and  $50 \text{ kg NH}_4\text{-N ha}^{-1}$ ). Bromide was applied at a rate of  $100 \text{ kg ha}^{-1}$  to provide information on water movement

through the profiles. Three precipitation regimes were used in triplicate: (i) N-C and N-A, which were exposed to natural precipitation during the period 22 July 1998 to 25 January 1999; (ii) D-A, which was exposed to ‘double’ natural precipitation from 22 July to 9 November 1998, and then natural precipitation only until 25 January 1999; and (iii) T-A, which was exposed to ‘triple’ natural precipitation from 22 July to 9 November 1998, and then natural precipitation until 25 January 1999.

At the end of October for T-A and in the middle of January for D-A, the cumulative amounts of leachate reached a level, which was equivalent to the amount of water (218 mm) that was present in each monolith after being freely drained. The N-C and N-A monoliths did not reach this water saturation level during the period (i.e., it was only 68 % of the field capacity moisture content at the end of the experiment). If the water entering the soil moved downwards as matrix flow, the Br peak concentration would appear when the accumulated leachate reached an amount of water corresponding to the water content at field capacity in a monolith. However, in all treatments, the Br peaks eluted before the accumulated leachate reached this level, which is a clear indication of preferential flow (Fig. 2). Nitrogen leaching occurred in the form of  $\text{NO}_3\text{-N}$ , which was also largely displaced through preferential flow (Fig. 2). Concentrations of  $\text{NO}_3\text{-N}$  in leachate in the treatments which received the element often exceeded  $10 \text{ mg L}^{-1}$ .  $\text{NH}_4$  leaching accounted for only a few percent of the total input of  $\text{NH}_4\text{-N}$ .



**Fig. 2.** Concentrations of a) Br and b)  $\text{NO}_3\text{-N}$  in leachate through clay soil monolith lysimeters exposed to natural precipitation (N-A, treated with chemicals:  $\circ$ : control (N-C, not treated:  $\bullet$ ), ‘double’ natural precipitation in summer (D-A, treated with chemicals:  $\square$ ) or ‘triple’ natural precipitation in summer (T-A, treated with chemicals:  $\Delta$ ). Arrows indicate dates when the cumulative amounts of leachate reached a level equivalent to the amount of water that was present in each monolith after being freely drained.

Although many studies have suggested the contribution of preferential flow occurring in soil on rapid leaching of solutes by examining dyed flow paths, few quantification approaches have been discussed by use of a non-reactive tracer. In my study (Maeda and Bergstöm, 2000), a modified version of the ‘tank-in-series model’ was used to describe a distribution function of non-reactive tracer (Br) travel time and compare the results with experimental data obtained in monolith lysimeters. The theory of the model is based on the tanks-in-series model (Fogler, 1992). Distribution of travel time (residence time) of a non-reactive tracer is determined by injection of the compound into the feedstream entering the reactor, and then the number ( $n$  (-)) of equally sized completely mixed flow tanks in series is obtained as a parameter. The parameter  $n$  shows the degree of mixing of flow in the reactor, and varies from one for ideal completely mixed flow, to infinity for ideal plug flow. The following assumptions were made for soil systems: (i) when water content in soil exceeds the maximum amount of water held in a freely drained monolith, infiltration occurs and its rate is equal in all tanks; (ii) soil has stagnant flow regions and their ratio to the maximum amount of water in a monolith, when freely drained, does not change during the experiment; and (iii) rainfall does not contain the studied tracer. After some mathematical steps, the distribution function of travel time ( $E(\tau)$ ) was obtained as the following equation:

$$E(\tau) = \frac{C_n V}{M} = \frac{\left(\frac{n}{f_m}\right) \left(\frac{n}{f_m} \tau\right)^{n-1}}{\Gamma(n)} e^{-\frac{n}{f_m} \tau} \quad (4)$$

where  $C_n$  ( $\text{mg L}^{-1}$ ) is the tracer concentration in leachate,  $f_m$  (-) is the ratio between water in mobile regions and the maximum water content in a freely drained monolith,  $M$  ( $\text{mg m}^{-2}$ ) is the mass of the tracer applied on a lysimeter surface,  $V$  (mm) is the maximum water content in the freely drained monolith,  $\Gamma$  is called a gamma function, and normalized time ( $\tau$  (-)) is defined as the cumulative leachate divided by the field capacity water content. When determining parameter values, the required experimental period was made shorter by using a peak coordinate of distribution of non-reactive tracer travel time, instead of the moment analysis.

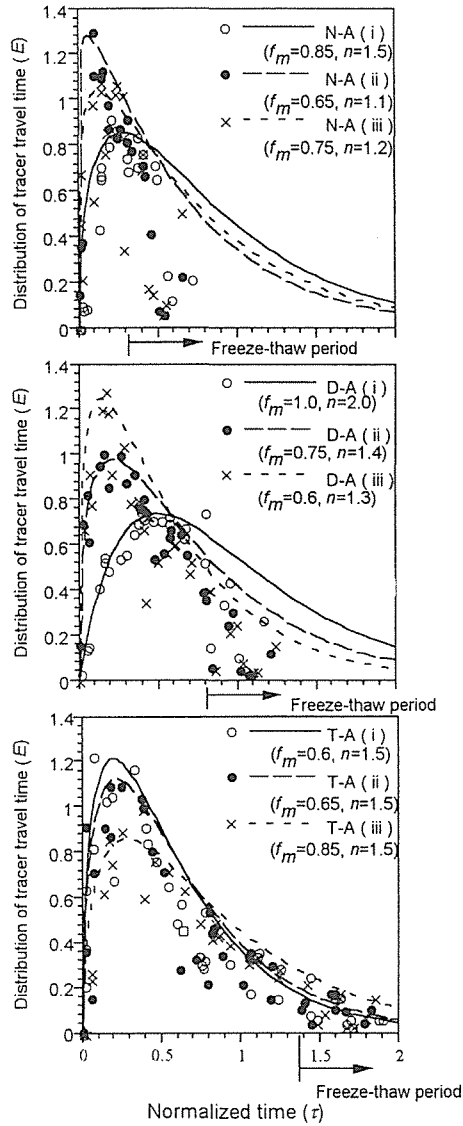


Fig. 3. Measured and simulated distributions of tracer travel time ( $E(\tau)$ ) vs. normalized time ( $\tau$ ): (i), (ii), and (iii) represent the three respective replicates. Monolith lysimeters were exposed to natural precipitation (N-A), 'double' natural precipitation in summer (D-A), or 'triple' natural precipitation in summer (T-A).

Preferential flow is caused mainly by two mechanisms. One is the existence of stagnant regions (dead space). This mechanism can be expressed in terms of the parameter  $f_m$ . The other is due to uneven water-flow rates. Some regions with rapid flow rates, such as macropores, and matrix flow regions will result in a large variability of flow rates. This condition may be expressed in terms of the parameter  $n$ .

Experimental results under 3 different water regimes are plotted in Fig. 3, together with fitting curves for different values on the parameters  $f_m$  and  $n$  used in the model. Generally, experimental data was well predicted by the modified tanks-in-series model. The values of  $f_m$  ranged from 0.6 to 1 and those of  $n$  from 1.1 to 2.0. These results suggest that the heavy clay soil used in this study might have 0-40% of stagnant regions in the profile. If the degree of mixing and stagnant regions were smaller without preferential flow, the peak of  $E(\tau)$  would appear at  $\tau = 1$ . The peaks of  $E(\tau)$  were, however, observed far earlier than  $\tau = 1$  in all treatments (Fig. 3). In other words, the mixing conditions in the monoliths were pronounced. This high degree of mixing suggests that matrix flow, and different kinds of preferential flow, occurred simultaneously in this clay soil.

During later periods of the experiment, measured values of tracer travel time were lower than simulated values in all monoliths (Fig. 3). These periods coincide with freeze-thaw periods. The flow conditions during such periods seemed to be quite different from those occurring during the summer months. Low concentrations of Br (low distributions of normalized tracer travel time) were seen in the first discharge after a long period of snow accumulation, which is similar to an observation by Bergström and Jarvis (1993). They explained this in terms of redistribution of the tracer to smaller unfrozen pores on freezing and that, upon melting, percolating water would be practically free from the tracer.

Overall, this approach can be a helpful tool when chemical leaching through soils with different preferential flow process are compared. Combining the model for a non-reactive tracer with chemical reaction models (including sorption, transformation, and biodegradation), will then contribute to give us a complete picture of chemical leaching.

## CLOSING REMARKS

Monitoring data on  $\text{NO}_3$  leaching under complex environmental conditions should be better understood for prediction or management of  $\text{NO}_3$  leaching. The author introduced the approaches, for this purpose, based on simple mathematics. Although precise prediction for N fate in the environment is required in some cases, the risk evaluation of agricultural management for  $\text{NO}_3$  leaching will also provide enough information for political decision or regulation.

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