Effects of Rainfall Pattern Change on Nitrate Leaching from Agricultural Land

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1. INTRODUCTION

Climate change which will take place near future would result in changes in precipitation patterns, onset of snowmelt and runoff. These changes in timings of available water may affect volume and frequency of water input over farmland. The rate of nitrate leaching from agricultural land is controlled by many factors including precipitation and porous medium properties and farming practices such as irrigation and fertilizer application. Laboratory and small-scale field experiments have given fundamental insights into micro to small scale mechanisms of nitrate leaching. Catchment studies, have pointed out importance of preferential flow and subsurface storm flow which were resulted from larger scale subsurface heterogeneity in nitrate discharge into surface water (*e.g.* Soulsby, *et al.*, 2003, Schilling and Zhang, 2004). In order to access subsurface heterogeneity effects on nitrate transport, larger scale field or laboratory experiments are required. Large lysimeter experiments allow us to assess effects of such subsurface heterogeneity on nitrate transport without sacrificing accuracy in quantification of fluxes and mechanisms. Owens *et. al.* (1995) obtained accurate nitrate mass balance using large undisturbed lysimeter and proposed strategy for reducing nitrate leaching from agricultural land to groundwater. Schoen *et al.*, (1999) performed tracer experiments using a large lysimeter packed with undisturbed soil and found an occurrence of preferential flow during nitrate leaching.

In this study, combination effects of rainfall input patterns and subsurface heterogeneity on nitrate leaching in the vadoze zone was investigated by means of laboratory tracer experiments using a weighing lysimeter equipped with an artificial rainfall generator. Following nitrate tracer application to the surface, rainfalls of constant amount and constant intervals were repetitively applied over the weighing lysimeter packed with either a homogeneous or a heterogeneous porous medium. The movements of water and solutes were monitored. Then, the experiments were repeated using different rainfall amount per event. In total, 12 cases of experiments (4 different rainfall patterns over 3 different porous media) were performed.

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2. METHODLOGY

The weighing lysimeter has a dimension of 3.0m in length, 1.0m in width and 0.6m in depth. The groundwater table was maintained at 0.55m below the surface by a mariot tank connected to the lysimeter during all experiments conducted in this study. Air flow over the lysimeter was controlled to have constant conditions that are the velocity of 1.5m/sec, the temperature of 20℃ and the relative humidity of 50%. The soil water was allowed to evaporate into air during all experiments. Evaporation rate was monitored by the weighing the lysimeter. Figure 1 shows the photo of the weighing lysimeter fitted underneath the wind tunnel. Table 1 summarizes the experimental conditions and the number of rainfall applied in each case.

The first set of experiments was conducted over the lysimeter packed with homogeneous fine sand (average diameter =0.14mm). Tracer solution which contains 1000mg/l of chloride and 1000mg/l of nitrate was applied as 2 mm of rain prior to every set of experiment. Artificial rainfall of 4mm was applied every 24 hours over the lysimeter for 16 times. It took approximately 10minutes to apply a single rainfall event over the lysimeter.

Water movement in the lysimeter was monitored by pressure transducers (Daiki Rika Kogyo Ltd.) connected to tensiometers placed at 0.05m, 0.1m, 0.15m, 0.2m, 0.3m, 0.45m and 0.5m below the surface in the lysimeter for every one minute. The lysimeter as well as mariot tank were weighed every one minutes to measure water loss from the system. The drainage from the lysimeter was collected in a plastic bottle and was measured manually once a day when the drainage was observed. The tracer movement was monitored by taking 2 ml to 10 ml of soil water samples using suction samplers (Sankei Rika Ltd.) placed at 0.05m, 0.1m, 0.15m, 0.2m 0.3m 0.4m 0.5m 0.6m below the surface in the lysimeter. Then the water samples were analyzed for chloride and nitrate using ion chromatography (Shimazu Ltd., PIA-1000). The set of experiments was repeated by changing rainfall amount per event to 6.8 mm, 13.0 mm and 26.0 mm. Each rainfall was repetitively applied every 24 hours until total rainfall exceeded at least 52mm.

The sources of nitrate almost always located on the ground surface. Thus, it is inevitable for nitrate to become in contact with soil before infiltrating to the groundwater. The top sand layer of 0.2m was replaced by field soil and the whole series of experiments were repeated over the two-layered medium.

Many solute infiltration researches reported fast infiltration of nitrate from ground surface to depths via macropores generated by earth worms and plant roots. Omoti and Wilted (1979) reported that earth worm channels ranged from 2mm to 10mm in diameter and had an average density of 100 pores per square meters. They also reported that macropores played an important role in transporting nitrate to the depths in the field conditions. Non-connected 300 pores were artificially generated over the lysimeter (*ie.* 100 pores per square meters). The half of the pores were extended to 0.2m from the surface, while 75 pores extended to 0.3m and the remaining 75 pores extended to 0.6m from the surface, since macropores were densely distributed near ground surface in actual fields (Munyankusi, et al., 1994). Then the series of the experiments were repeated.

3. Water and nitrate movements in response to rainfall applications

3.1 Homogeneous medium

Concentration profiles of chloride and nitrate after 52mm of total rain by repeating different rainfall amount in the homogeneous medium were shown in Figure 2. These are the profiles observed 24hours after 13 times of 4.0mm rain, after 8 times of 6.8mm rain, after 4 times of 13.0mm rain and after 2 times of 26.0mm rain. Evaporation from the lysimeter was almost comparable or exceeded rainfall amount during 4mm rains. Thus, no significant solute downward movement was observed under these light rains. No apparent defference between chloride and nitrate profiles was observed.

Approximately 80% of the rain was lost to evaporation during 6.8mm rains, while almost half of the rain was lost to evaporation during 13.8mm rains with 24hour intervals. Though increase of water content in the vadoze zone was observed, no significant downward solute infiltration took place under these intermediate rains. The profiles of chloride and nitrate during these intermediate rains almost coincides each other in the homogeneous medium (Fig.2).

Apparent solute downward infiltration was observed only during the heaviest 26.0mm rains within our experimental conditions. The solute profiles after 52mm and 104mm of total rain by 26.0mm rainfalls are shown in Figure 3. The profiles of chloride and nitrate perfectly coincides each other, and piston-type infiltration was clearly observed.

In the homogeneous medium, chloride and nitrate profiles coincided to each other under all experimental conditions, indicating nitrate is behaving as a conservative tracer in the homogeneous medium. No degradation of nitrate was observed. Both solutes reached groundwater table by piston-flow generated by 26.0mm rains. On daily basis, approximately 90% of the rain was less than 25mm per day during the year of 2005 in Tokyo (calculated based on JMA, 2006). Thus, nitrate applied from the ground surface would not undergo infiltration by 90% of the time of rain if the porous medium were homogeneous.

3.2 Two-layered medium

Field soil was taken from a woodlot where vegetables used to be grown near the laboratory, and the top 0.2m of sand layer in the lysimeter was replaced by the soil. In actual fields, nitrogen usually enters subsurface system from the ground surface. Thus, it is inevitable for nitrate to become in contact with organic soil before it undergoes infiltration. The two-layered system represents actual field conditions better than the homogeneous medium, although it still is a highly idealized and artificially packed porous medium.

Evaporation exceeded rainfall amount during 4.0mm rains as was in the homogeneous medium. Consequently, no significant downward solute movement was observed under 4.0mm rains in the two-layered system.

Figure 4 shows the chloride and nitrate concentration profiles after 52mm and 104mm of total rain by 13.0mm rains. The profiles were obtained 96 hours (4 rainfalls) and 192 hours (8 rainfalls) after the tracer application. The nitrate concentration in the soil water was inherently high probably due to preceding land use which was vegetable field. Nitrate did not infiltrate beyond 0.3m below the surface during 13.0mm rainfalls. It was observed, however, that both nitrate and chloride concentrations increased in the soil during the first 4 rainfalls and then they started decrease, indicating solute infiltration followed by dilution due to the repeating rainfall applications.

The ratio of nitrate concentrations to chloride concentration should be constant if they behave in the same manner in the porous medium. The ratio of nitrate to chloride deceased from 2.5 to 2.2 at 0.15m below surface and 0.88 to 0.83 at 0.2m below the surface in the soil layer between 96 hours and 192 hours after the tracer application, whereas the nitrate to chloride ratios were constant or slightly increased in the sand layer below the soil. The decrease of the ratios suggests occurrence of nitrate degradation in the soil layer. Though average denitrification rate was very small, hot-spots where denitrification is partially high are often found in unsaturated zone (Parkin,1987). The soil layer inherently has heterogeneity in it and the anaerobic parts in the soil may have played as hot-spots of denitrification in our lysimeter.

Figure 5 shows the average water distribution and estimated chloride and nitrate distribution after 104mm of total rains for three different rainfall cases. During 6.8mm rains, it was observed that more than 80 % of water was lost to evaporation and 100% of the chloride remain in the porous medium, while 1.6 % of nitrate was estimated to undergo denitrification. Approximately 50% of water was lost to evaporation under 13.0mm rain cases. Slight loss of mass was estimated for nitrate whereas 100% of the chloride remained in the porous medium. During 26.0mm rainfall case, approximately one third of the water was lost to evaporation and another one third was lost to discharge. The distribution of chloride and nitrate are very similar and no evidence of denitrification was observed. The residence time of the solute was 17 days for the 6.8mm rain case, 9 days for the 13.0 mm rain case and 5 days for the 26.0 mm rain case. Short residence time did not allow nitrate to undergo denitrification during 26 mm rains.

In the two-layered system, water and solute infiltrated into groundwater table only by the strongest 26.0mm rains, as was in the homogeneous medium. However, denitrification was observed in the soil layer under light rains. High concentration of nitrate remained in the soil layer generating large dispersion of the solute as front or center of mass infiltrate into deeper zone by intermediate to strong rains.

3.3 Macroporous medium

Macropores which are relatively large continuous openings in porous media are distributed ubiquitously over the field and densely near surface since they are created mainly by earthworms and plant roots (Munyankusi *et al.*, 1994). Onset of macropore flow depends on rainfall intensities, while rate of macropore flow depends on total rainfall amount (Sugita *et al.*, 2005). Rainfall pattern effects on nitrate transport in a macroporous medium were examined using the fine sand medium with artificially generated macropores.

Soil water near surface drained or evaporated quickly after the rain, so that water samples shallower than 0.2m were unable to obtain under light to intermediate rain conditions in the macroporous medium. Concentration profiles of chloride and nitrate after 52mm and 104mm of total rain by 13.0mm rains are shown in Figure 6. Considerable increase in both chloride and nitrate concentrations was observed up to 0.3m below the surface, indicating fast transport of solutes via macropores followed by quick infiltration of solute from macropores to matrix. Dilution is estimated to be the main mechanism of the concentration decrease observed.

The concentration profiles of chloride and nitrate under 26.0 mm rains are shown in Figure 7. Double peaks are clearly observed in both profiles, suggesting fast macropore flow followed by matrix flow, which resulted in large spreading of the solute.

In the macro porous medium, fast nitrate infiltration under intermediate rains and piston-flow with double peaks under strong rains are observed due to the presence of macropores . No effect of macropores was found during light 4mm rains.

4. Conclusions

Nitrate movement observed in response to repeating rains in the homogeneous and heterogeneous porous media are summarized in Table 2. No infiltration was observed under 4.0mm rains in all porous media due to water loss to evaporation processes. Significant amount of nitrate infiltrated into the groundwater only by piston-flow caused by heavy (26mm) rainfalls in the homogeneous and heterogeneous media.

Double peaks were observed in a macroporous medium under strong rainfalls, indicating that nitrate was transported by preferential and matrix flows. Trace of the nitrate reached groundwater under intermediate (13mm) rainfalls in a macroporous medium due to preferential flow, which made broad vertical distribution of nitrate. In a layered medium, which has soil cover over the sand, the high nitrate concentration remained in the topsoil under all rainfall conditions, resulting in large dispersion of the solute under intermediate to heavy rains. Heterogeneity in the porous media is contributing to large spreading of nitrate in these cases.

The nitrate movement perfectly coincided with that of chloride in a homogeneous sand medium. Nitrate/chloride ratio in the topsoil of the two-layered medium, however, decreased with time under light rains, suggesting presence of degradation processes. A part of soil may be serving as spots for denitrification in the lysimeter when the solute residence time is long.

Nitrate leaches only under extreme rainfall events both in homogeneous and heterogeneous media, and considerable degradation under light rains in the soil layer and large spreading under intermediate rains in the two-layered and macroporous media are observed. On daily basis, 52.8% of rains that Tokyo received in 2005 was less than 5mm per day, and approximately 10% of rains exceeded 26mm (calculated based on JMA, 2006). These indicate that nitrate does not infiltrate at all by more than 50% of the rains and hardly infiltrates by 90% of rains in Tokyo under current climate conditions. However, the climate change which may result in higher frequencies of heavier rains could significantly increase nitrate leaching and dispersion and decrease chance of nitrate degradation in the subsurface system near future. It was also found that nitrate movement changes in responses to rainfall pattern change more sensitively in heterogeneous media than in a homogeneous medium.

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Table 1 Number of rainfall events repeated in each experimental condition

Rainfall amount per event	Number of rainfall event repeated
4.0mm	16
6.8mm	16
13.0mm	8
26.0mm	5

Table 2 Summary of nitrate profile properties observed at the end of each experimental case. (Depths of the concentration peak and front are in meters below surface. ○:degradation observed in soil, ×:degradation not observed)

Rainfall Amount	Profile properties	Homogeneous medium	Two-layered medium	Macroporous medium
4.0mm	depth of peak (m)	0.1	0.1	0.1
	depth of front (m)	0.1	0.1	0.1
	degradation	×	○	×
6.8mm	depth of peak (m)	0.1	0.1	0.1
	depth of front (m)	0.1	0.1	0.2
	degradation	×	○	×
13.0mm	depth of peak (m)	0.1	0.1	0.1
	depth of front (m)	0.3	0.1	0.3
	degradation	×	○	×
26.0mm	depth of peak (m)	0.3	0.3	0.2, 0.4
	depth of front (m)	0.5	0.5	0.5
	degradation	×	×	×

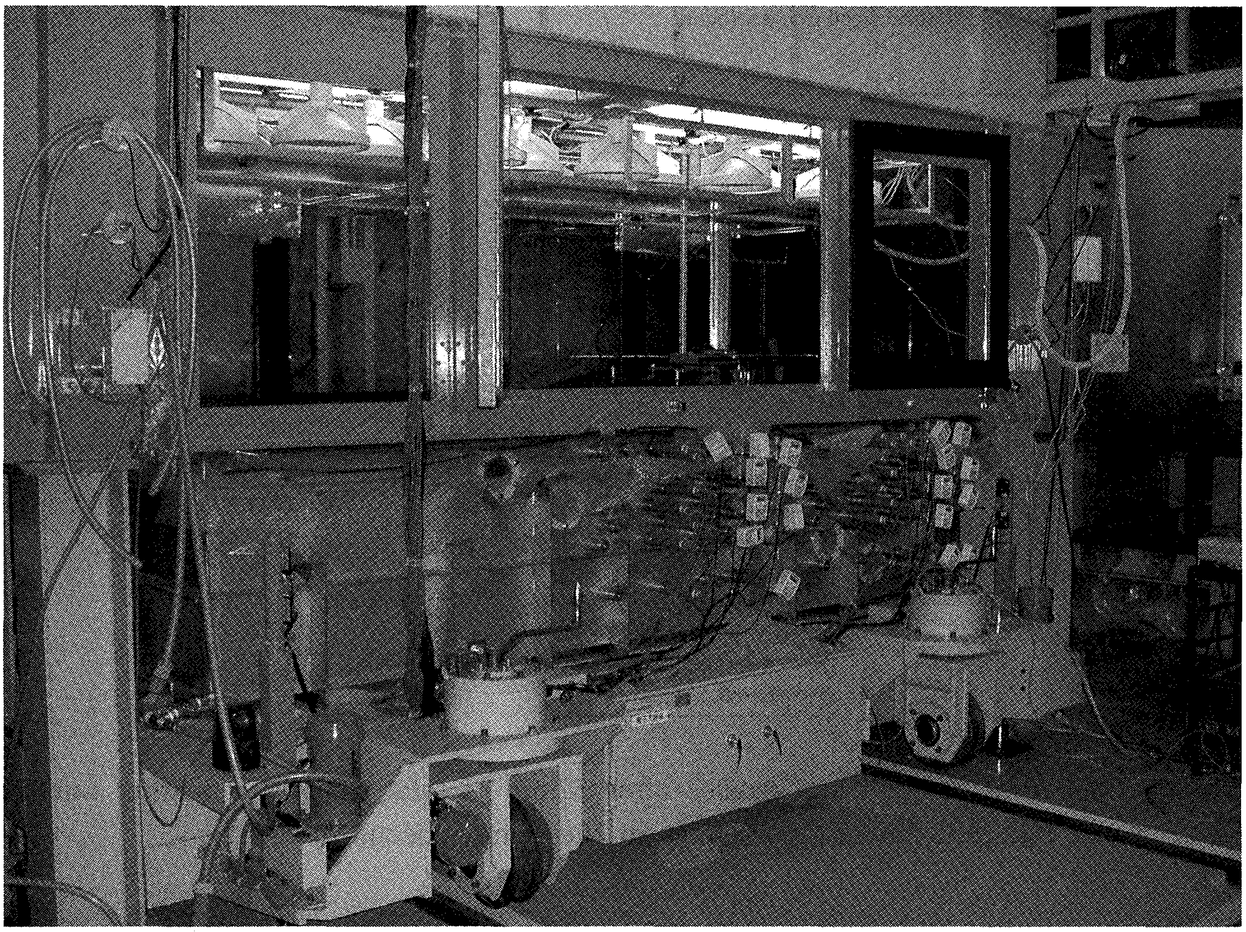

Figure 1. The weighing lysimeter fitted underneath the wind tunnel

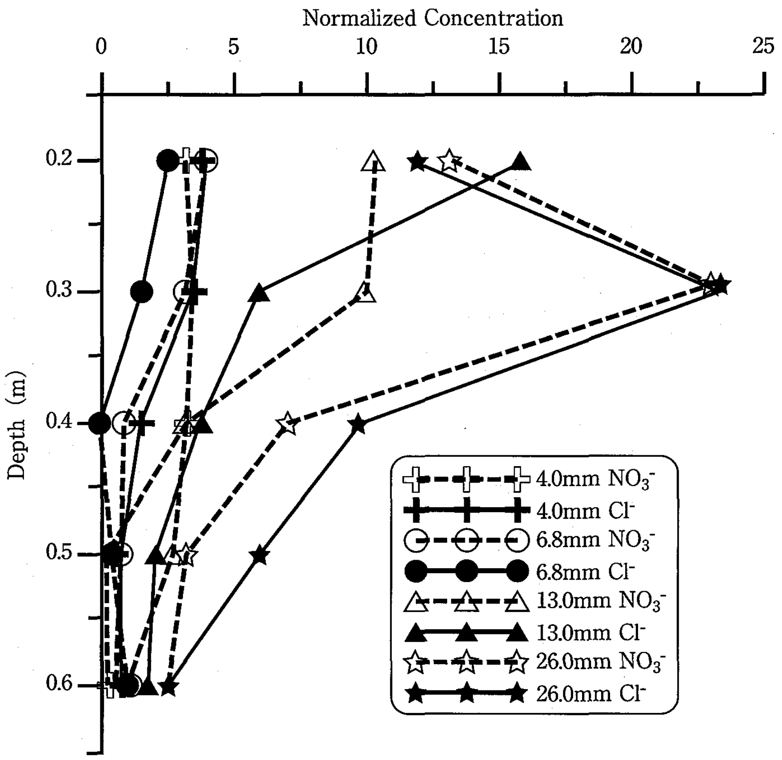

Figure 2. Chloride and nitrate profiles after 52mm of total rain by repeating rainfall of different amount per event in the homogeneous medium.

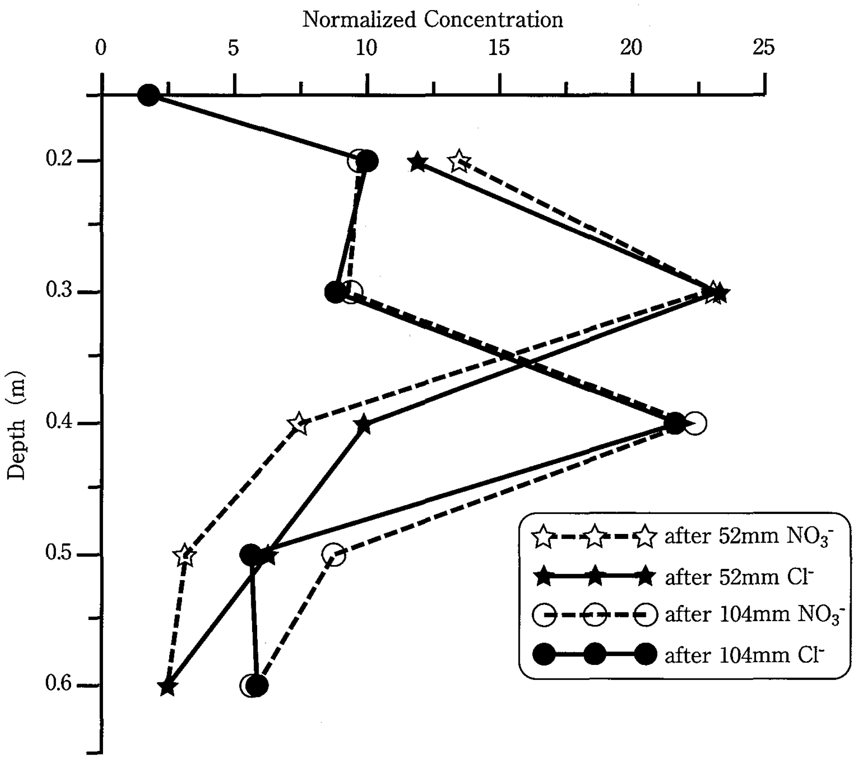

Figure 3. Chloride and nitrate profiles after 52 mm and 104 mm of total rain by repeating 26.0 mm rainfalls in the homogeneous medium.

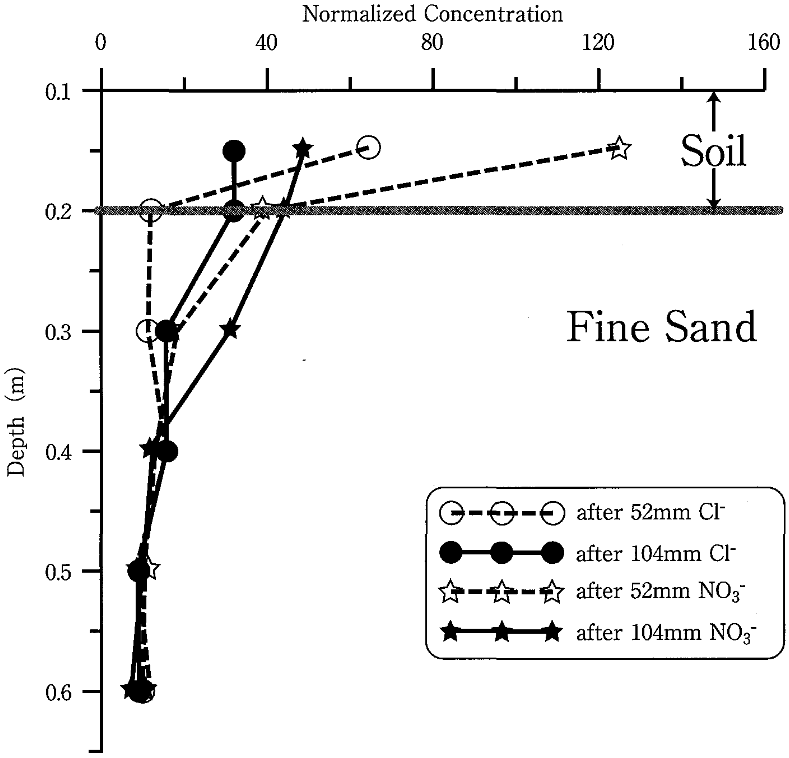

Figure 4. Chloride and nitrate profiles after 52mm and 104mm of total rain by repeating 13.0 mm rainfalls in the two-layered system.

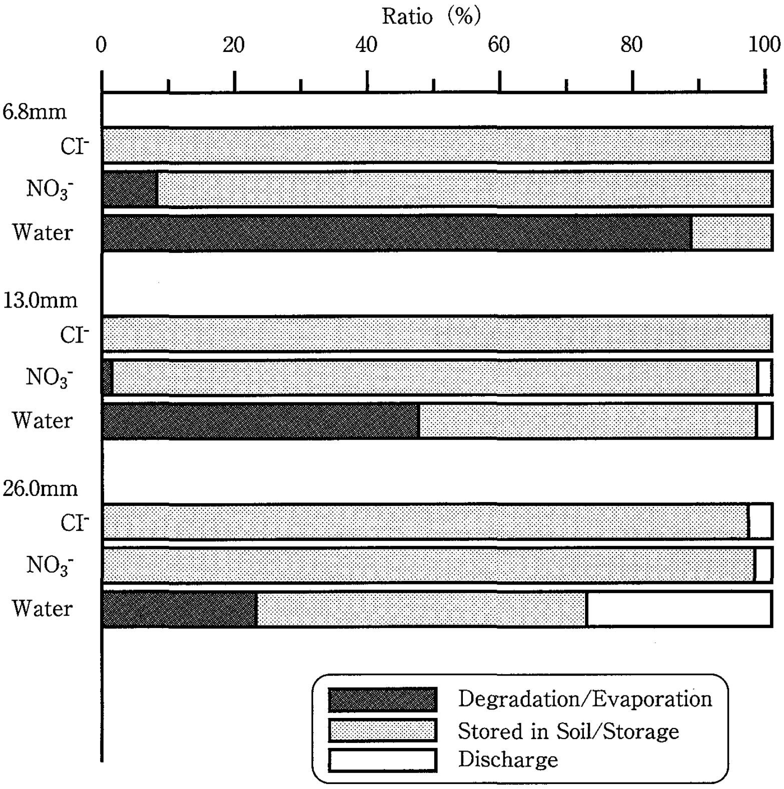

Figure 5. Average water and solutes distributions after 104mm of total rain.

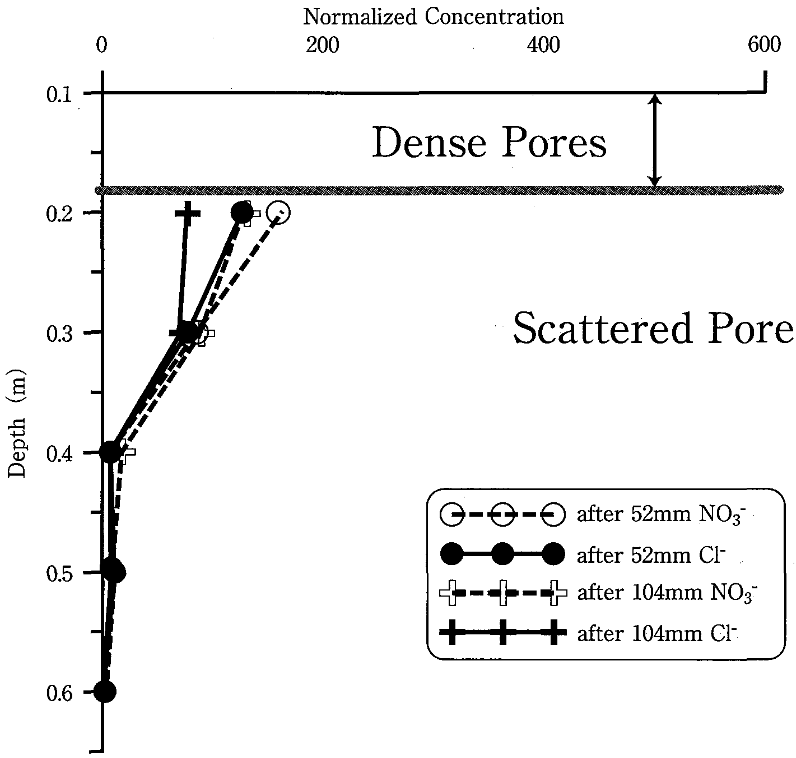

Figure 6. Chloride and nitrate profiles after 52mm and 104 mm of total rains by repeating 13.0 mm rainfalls in the macroporous medium.

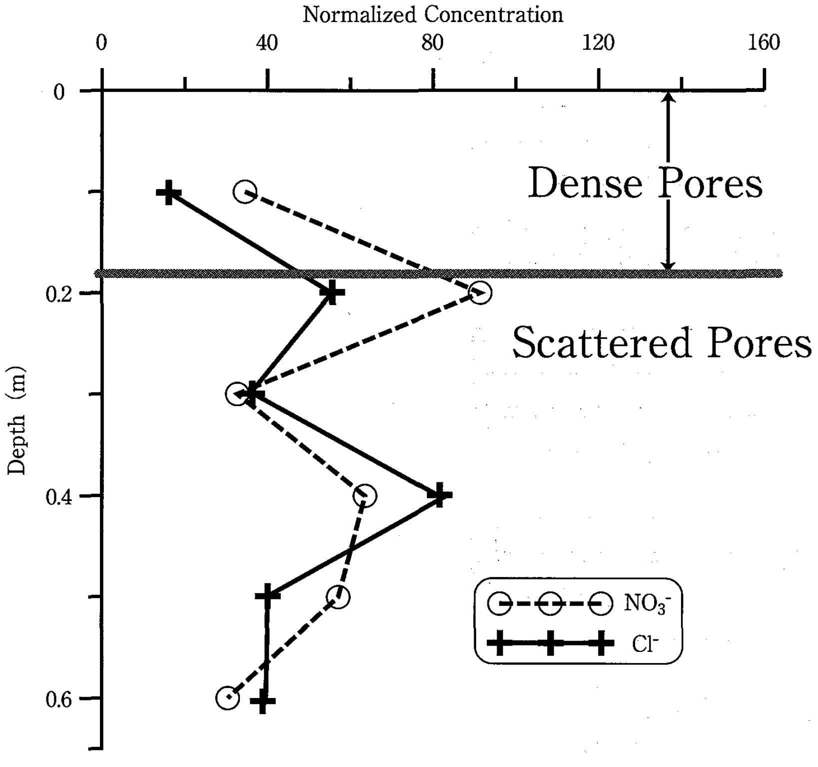

Figure 7. Chloride and nitrate profiles after 104 mm of total rain by repeating 26.0 mm rainfalls in the macroporous medium.

〔抄　録〕

降水パターンが硝酸態窒素の溶脱におよぼす影響

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近い将来，地球温暖化による気候変動により降水量の時空間分布が大きく偏ることが予測されている。偏った降水分布は，浸透する降水とともに地表面から地下水面へ移動する物質移動にも影響を与えると推測される。我が国で最も多く検出される地下水汚染物質である硝酸態窒素の地中での動きに降水パターンが与える影響を明らかにすることを目的として，大型計量ライシメータを用いた室内実験をおこなった。細砂を充填した均質媒体，実際の野外条件に近い土壌層を細砂の上に有する2層構造，および細砂中にマクロポアを有する不均質媒体に4 mm, 6.8mm, 13.6mm, 26mmの降水をそれぞれ24時間間隔でくり返し与え，地中における水およびトレーサとして地表面から与えた硝酸態窒素の動きを観測した。

硝酸態窒素は豪雨に相当する26mm降水をくり返し与えたときのみに均質・不均質媒体中でともに地下水面へ大量に溶脱した。その他の降水時には均質媒体内では水と硝酸態窒素は同じ動きを呈したが，2層構造をもつ不均質媒体では4 mm降水条件下のみに表層土壌内における硝酸態窒素の分解による濃度低下が観測された。一方，マクロポアを有する媒体内では6.8mmと13mmの中程度の降水条件下で一部の硝酸態窒素が表層に残存する一方マクロポア流による早い浸透が生じ，地中における硝酸態窒素の非常に大きな広がりが認められた。これらにより将来，気候変動により豪雨頻度が増加するとより多くの硝酸態窒素の溶脱および大きな分散が生じることが示唆された。

Rainfall distribution should change near future according to climate change expected to take place following global warming. Severer uneven distribution of rainfall with respect to time and space is estimated. Uneven distribution of water input may affect solute transport from ground surface to groundwater table, which causes groundwater contamination. Nitrate is the most common pollutants found in groundwater in Japan as well as in many other countries. In this study, laboratory experiments were conducted to investigate the effects of rainfall properties on movement of nitrate in porous media.

Various artificial rainfalls of constant amount and a constant interval was repetitively applied over a weighing lysimeter packed with either a homogeneous or a heterogeneous porous medium. Movements of nitrate, chloride and water in the lysimeter were monitored.

Nitrate leaches only under extreme rainfall events, while considerable degradation under light rains in the soil layer and large spreading under intermediate rains in the heterogeneous media are observed. It was suggested that amount of nitrate leaching may significantly increase due to the climate change which will increase frequencies of heavy rain.

Key words: lysimeter, nitrate, heterogeneous porous medium, climate change