Nitrogen removal from water containing high nitrate nitrogen in a paddy field (wetland)

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Abstract Nowadays, it has become very common to find in Japan that nitrate nitrogen concentrations are very high in spring water and in well water where the land use of a watershed is agricultural. We have often observed around 50 mg/L of nitrate nitrogen in the spring water where we live. Crops produced in those fields are mainly vegetables such as celery, cabbage, lettuce, carrots, and so on. Green tea is also popular in Japan. In order to produce good quality green tea, farmers apply a great amount of nitrogen fertilizer. This amount can reach up to 1,000 kg/ha in some areas, although the average application amounts to 628 kg/ha in Japan. As a result, ground water that is rich in nitrate flows into the river, which results in a high nitrogen concentration in river water and ground water. Further, this causes a low pH in river water in some tributary rivers in Japan, though this kind of case is very rare. We knew from field tests that if water contained a high nitrogen concentration and was introduced into paddy fields, high nitrogen removal would be performed. This paper presents the outline and results of a system on how to remove nitrogen using paddy fields (wetlands). Further, this paper presents the evaluated results of the removal quantity at the watershed level.
Keywords Denitrification; ground water contamination; nitrogen removal; paddy field; wetland

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Introduction

Contamination issues of underground water from nitrate nitrogen are hidden worldwide. We have the same issues in Japan. Rice was the main product in the agricultural industry forty years ago in Japan. We have focused on how we could obtain a greater yield of rice for a long time. In order to minimize the nitrogen release from soil, researchers have been keenly studying how to prevent denitrification from taking place in paddy fields. However, nowadays we are studying earnestly how to remove nitrogen using denitrification in the paddy fields. Japan has a mountainous topography and low lands are commonly used for paddy fields where the irrigation water is secured. However, the over-production of rice started from the 1970s and a set aside policy was introduced for the paddy fields after that. Paddy fields, which have low production efficiency, have been abandoned. Our laboratory started a study on nitrogen removal in such abandoned paddy fields (Tabuchi *et al*., 1993, 1995, 1996, 2001a, 2001b; Kuroda *et al*., 2000), especially for topography as a terrace, as shown in Figure 1. We have insisted that nitrogen removal should be done as a topographical chain as shown in Figure 1 (Kuroda, 1998). The nitrogen fertilizer applied into the upland fields oxidized quickly under the aerobic condition in the soil of the fields, then it percolated through the soils into the ground water when there was rain (Nakasone *et al*., 2002). It causes the nitrate contamination of the river. Sometimes, it causes acidification of irrigation reservoirs. We found such an acidified reservoir in the Shizuoka Prefecture in Japan. Hence, nitrate nitrogen removal using paddy fields (wetland system) is very important in such areas.

Outline of surveyed area

Experimental fields in Ibaraki prefecture

Our laboratory has been continuing an experiment at set aside paddy fields in the Ibaraki Prefecture for ten years. The topography of this area is just as shown in Figure 1. Through

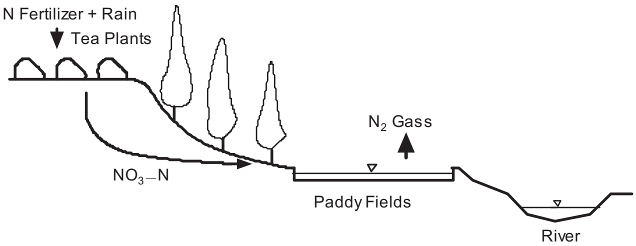

Figure 1 The outline of the topographical chain of the watershed

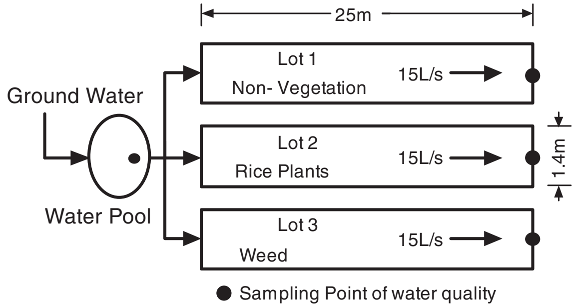

Figure 2 Outline of experimental lots in Ibaraki prefecture

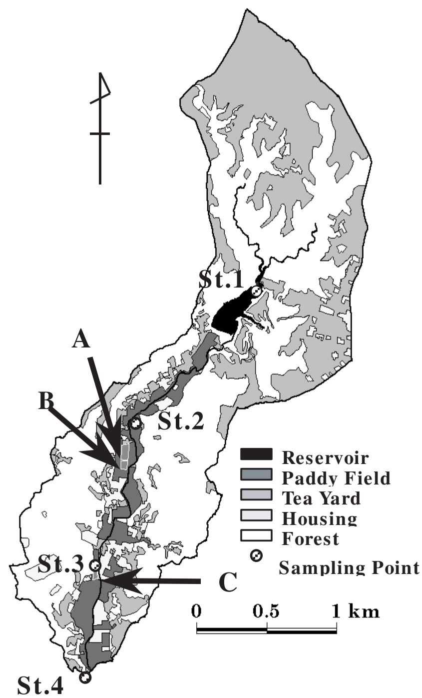

Figure 3 Land use of watershed in Shizuoka prefecture

the years, some kinds of fruit such as watermelon and melon, and vegetables such as lettuce and cabbage were produced on the upland fields. These crops require high amounts of nitrogen fertilizer. For example, the inorganic nitrogen application amount on average is 284 kg-N/ha for lettuce, 338 kg-N/ha for cabbage, and 226 kg-N/ha for watermelon in Japan. The lowland used to be used for the paddy fields in ancient times in Japan. Irrigation water used for these paddy fields was originally spring water from a terrace. As a result, nowadays this water contains a high nitrate nitrogen concentration with an average of 20 mg/L throughout the year. Fortunately, a great amount of nitrate nitrogen was removed by the denitrification taking place in the paddy fields during the irrigation period. The T-N concentration downstream of the river was around 4 mg/L. We made three experimental strips of 25 m × 1.4 m in the paddy field. We introduced spring water from the terrace into the water pool for the irrigation water. We delivered this water into the three experimental paddy fields through a weir in order to measure the irrigation quantity. An outline of the experimental paddy fields is shown in Figure 2.

Experimental fields in Shizuoka prefecture

The test site where we performed the survey is shown in Figure 3. We have three experimental paddy fields at this location. Also, this location has a very special land use compared with other areas of Japan. Almost three kinds of land use are in this area. The land use on the terrace is for tea fields, the land use on the low land is only for paddy fields, and between the two land uses there are sloped forests. We had found a very special irrigation reservoir that is acidified from the over use of nitrogen fertilizer (Nakasone *et al.*, 2002). The nitrate nitrogen concentration was very high at around 33 mg/L since denitrification was not taking place because of the lack of organic matter in the water. The reason why denitrification was not taking place in this reservoir was that organic matter was not produced in it because of a low pH. Sometimes the pH drops down to 5.0, hence, the phytoplankton cannot be produced. As mentioned above, the average application of nitrogen fertilizer is 628 kg/ha in Japan. Farmers are trying to produce good quality tea, hence, they apply more nitrogen fertilizer. It is said that the application amount reaches to around 1,000 kg/ha in this area. Unfortunately, this idea leads farmers into a vicious cycle because the roots of the tea plants are damaged by the low pH although a low pH of 3.5 is rather favorable for tea plants. However, a great amount of nitrogen fertilizer is leaching into the ground water, and then flows out into the stream.

Experimental results and discussion

Results in Ibaraki prefecture

We ran this experiment from 1991 to 1999. Thereafter, we have been studying the relation between denitrification and organic matter as a hydrogen supplier in our laboratory. The experiment had been performed setting up three strips as shown in Figure 2. Previously, we have presented the precise experimental results (Tabuchi *et al.*, 2001a). Therefore, we present here three examples among them such as water temperature, NO_3-N concentration, and removal rate of NO_3-N. The experimental results are located in the references (Tabuchi *et al.*, 2001a, 2001b). As shown in Figures 4 and 6, there are four seasons in Japan, hence, the water temperature goes down to less than 10°C during the winter and the removal rate also goes down to less than 20%. These values of the parameters did not vary greatly between the vegetation and non-vegetation or between the kinds of plant (Kuroda *et al.*, 2000). Therefore, we present here the case of the rice planted strip. The irrigation water used here is spring water from the upland fields. The water temperature of the spring water does not drop to under 11°C even in the winter. However, the air temperature often goes below 0°C in the winter, hence, the irrigated water becomes around 1°C in the winter. It is well known that the efficiency of denitrification decreases according to a decrease of water temperature. This is clearly shown in Figures 5 and 6. Nitrate nitrogen concentrations of the spring water are around 20 mg/L as shown in Figure 5. There is a decrease in nitrate nitrogen concentrations of about 40% although there are big fluctuations as shown in Figure 6. As shown in Figure 7, the annual average removal rate was 0.43 $gm^{-2}d^{-1}$ in the beginning. After that, this value gradually decreased until 1995, then the value became stable. We did not apply any fertilizer during the experiment. Also, the paddy fields and each strip were always flooded. Therefore, the fertilizer, except nitrogen, has been decreasing gradually from the soils up to 1995, since plants used it. We assume that the nutrient supply became stable after 1995 since the nutrient input from the irrigation water and the nutrient output into the plant uptake balanced.

Many researchers have presented ways on how to construct wetland systems (Moshiri, 1993). None presented an equation except Tabuchi (2001), who calculated a removal rate. It has been said that the research on quantity was merely luck. Many researchers focused only on the removal concentrations of NO_3-N. This value changes according to the influent concentration of nitrate nitrogen. Other factors that affect the removal rate are water temperature, vegetation, and water depth. One point in which the paddy field is different from wetland systems would be the water depth. Usually, the water depth stays between 5 cm and 15 cm in paddy fields. This water depth determines the hydraulic retention time, which in turn decides the removal quantity. Tabuchi (2001) presented an empirical equation considering the important factors affecting removal quantity as follows.

$$R_o = \alpha X \quad (1)$$

Here, R_o is the removal rate ($gm^{-2}d^{-1}$); α is a coefficient which changes according to the water temperature and the kind of soil. α has a relation with the water temperature and it is determined from the experiment of Ibaraki Prefecture as shown in Figure 1.

$$\alpha = 0.000011T^2 + 0.005 \quad (T>10°C) \quad (2)$$

$$\alpha = 0.0006T \quad (T<10°C) \quad (3)$$

X (nitrogen concentration) decreases with time and is expressed as follows:

$$X = X_o \exp(-\alpha t/H) \quad (4)$$

Here, X_o is the initial condition; t is time (d); H is water depth (m). If irrigation is discharged

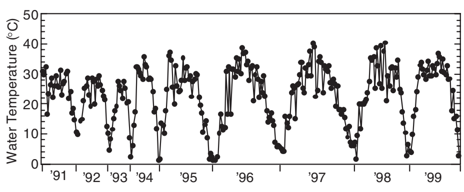

Figure 4 Fluctuation of water temperature at rice planted strip

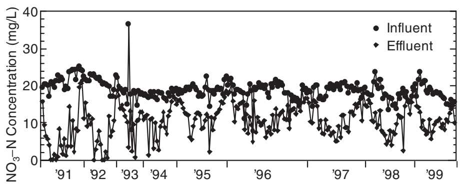

Figure 5 NO_3-N concentration between influent and effluent

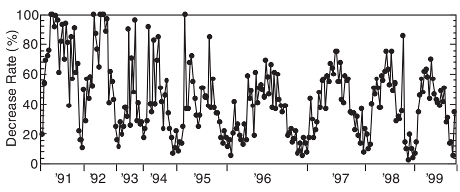

Figure 6 NO_3-N decrease rate between influent and effluent

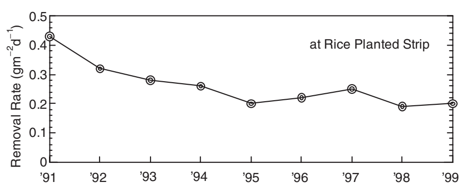

Figure 7 Annual average removal rate

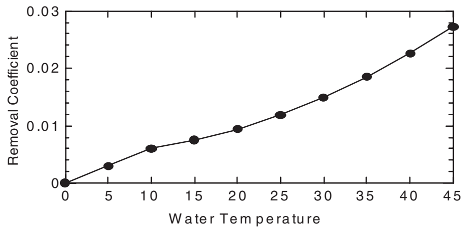

Figure 8 Values of removal coefficient

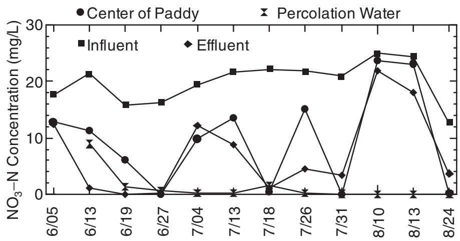

Figure 9 NO_3-N decrease at paddy field A

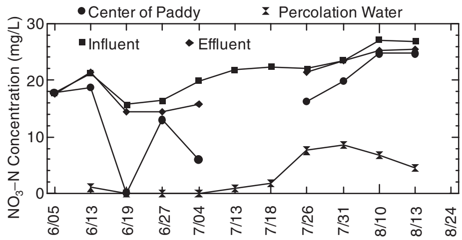

Figure 10 NO_3-N decrease at paddy field B

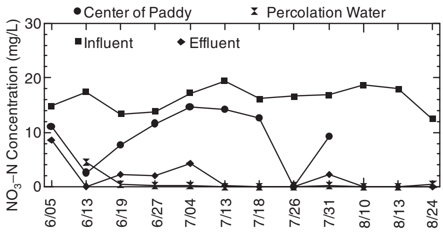

Figure 11 NO_3-N decrease at paddy field C

to a strip Q (m^3/d) and the area of a strip is A (m^2), q is $q = Q/A$ (m/d). Then the running time t between the inlet and outlet of the strip is expressed as follows:

$$t = AH/Q = H/q \tag{5}$$

Substituting Eq. (5) into (4), we get Eq. (6).

$$X = X_o \exp(-\alpha/q) \tag{6}$$

Then

$$R_o = \alpha X_o \exp(-\alpha/q) \tag{7}$$

Further, assuming that the nitrate nitrogen decrease rate is β_o (%), β_o is expressed like Eq. (8).

$$\beta_o = ((X_o - X)/X_o) \times 100 \tag{8}$$

Substituting Eq. (6) into (8), we get Eq. (9).

$$\beta_o = (1 - \exp(-\alpha/q)) \times 100 \tag{9}$$

Using Eqs (2), (3), and (9), we can calculate β_o only from the water temperature and q. The value of q was kept constant and the value was 0.037 m/d in this experiment. The values of a are presented in Figure 8. From nine years of experimentation the average water temperature, nitrate nitrogen removal rate, and NO_3-N decrease rate were obtained as shown in Table 1.

Results in Shizuoka prefecture

The experiment that was performed in Ibaraki Prefecture was just at an experimental level, however the one that was performed in the Shizuoka Prefecture was at watershed level. The Ministry of Agriculture and Fishery hired three paddy fields; A, B, and C in Shizuoka which are shown in Figure 3. The area of A is 41.8a, the area of B is 12.0a, and the area of C is 30.3a. The Tanno river runs in the watershed and we observed the water quality at four points that are shown in Figure 3 with the mark of ○. The experimental results for these three paddy fields are shown in Figures 9, 10 and 11. These figures show that denitrification was taking place in all of the paddy fields. The NO_3-N concentration decrease fluctuates greatly since the water requirement rate was changing. Water management of these paddy fields was performed by each of the farmers who own the paddy fields. The fluctuation occurred since the hydraulic retention time in the actual paddy fields would differ according to the water management. The values of the removal rate and the removal percentages are shown in Table 2. Compared with the removal rate achieved in Ibaraki Prefecture, the values obtained in Shizuoka were slightly larger than Ibaraki Prefecture. We assume that the reason was because the three experimental strips in Ibaraki were always flooded for ten years and percolation was rare. On the contrary, there was percolation in the paddy field of Shizuoka and the soil was dried during the winter. Therefore, the soil condition became aerobic in the winter. Then the organic matter in the soil would decompose in the spring. This organic matter would be used as a hydrogen supplier for denitrification that would be taking place during the irrigation period. The values of the removal rate in Table 2 are almost the same as the starting values in Figure 8, which means that there was an accumulation of organic matter in the soil of Ibaraki Prefecture at first. This organic matter was used for denitrification in the early stage of the experiment, hence, the removal rate gradually went down. Therefore, the data taken in Ibaraki Prefecture would be applied to the wetland.

There are some paddy fields alongside the Tanno river as shown in Figure 3. Also, there are several small weirs in the Tanno river and the irrigation water is taken from the river. Therefore, the water quality will recover by means of recycling water use. The Ministry of Agriculture and Fishery (2002) observed the water quality from four points of the Tanno river. We present the observed results for pH, NO_3-N, and T-P in Figures 12, 13 and 14 respectively. The pH values did not fluctuate throughout the year, however the pH values of sampling station 1 were very low. This is because of the over application of nitrogen fertilizer, which the authors have already reported (Nakasone *et al.*, 2002). This low pH neutralized quickly at the Tanno irrigation reservoir as shown in Figure 3. The NO_3-N concentrations decreased gradually from the upstream portion to the downstream, especially during the irrigation period. This fact also apparently shows that nitrogen removal was taking place in the paddy fields of the watershed. Further, Figure 11 shows that there is self-purification for nitrogen. The T-P fluctuated at station 3 and 4 during the irrigation period. The reason why T-P concentrations became high only during the irrigation period is unknown, however there are two assumptions concerning it. One is the dissolution from the paddy field since the soil condition became anaerobic during the

Table 1 Average values for nine years

	Unit	Lot 1	Lot 2	Lot 3
Average day time temp.	°C	18.3	22.7	23.9
Removal quantity	$gm^{-2}d^{-1}$	0.30	0.27	0.27
Decrease rate	%	49.7	43.4	44.4

Table 2 Nitrogen removal rate ($gm^{-2}d^{-1}$)

	Load ($gm^{-2}d^{-1}$)	Removal ($gm^{-2}d^{-1}$)	Percent (%)
Paddy Field A	0.42	0.37	89
Paddy Field B	0.57	0.46	80
Paddy Field C	0.34	0.33	96

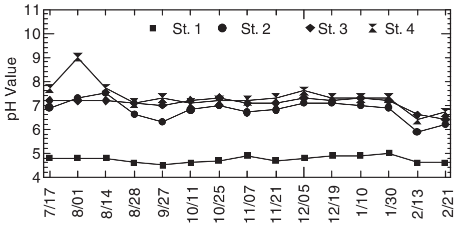

Figure 12 pH values from station 1 to station 4 in 2000

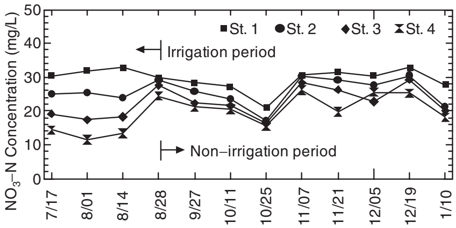

Figure 13 NO_3-N concentrations from St. 1 to St. 4

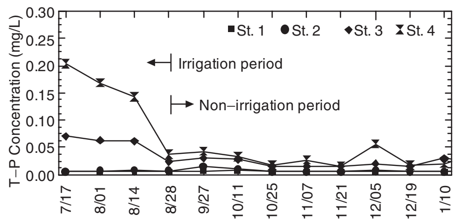

Figure 14 Change of T-P concentrations from St. 1 to St. 4

irrigation period. The Ministry of Agriculture and Fishery checked the ORP in the soil water below 40 cm from the soil surface and the vales of ORP were around –150 mV. The other assumption is a runoff from the big chicken coop factory.

Evaluation of nitrogen removal quantity in the Shizuoka prefecture

Firstly, we considered the annual water balance of the watershed. The annual rainfall of the area was 2,009 mm in 2000 and the runoff coefficient of the watershed was 0.56 from our survey. Then the annual runoff was 1,125 mm. Assuming the total nitrogen application is 1,000 kg-N/ha and the up-take of tea plants is 324 kg-N/ha, the residue would be 676 kg-N/ha (Nakasone *et al.*, 2000). The areas of the watershed are shown in Table 3. Hence, the annual average nitrogen concentration would be calculated as 22.2 mg/L. The quantity of runoff nitrogen per day would be 314.8 kg-N/d and the reduction quantity in the paddy fields would be 149.8 kg-N/d (4.15 kg/d × 36.1 ha = 149.8 kg/d). Therefore, the average nitrogen concentration during the irrigation period at St. 4 should be 11.4 mg/L since the average nitrogen concentration during the non-irrigation period would be 22.2 mg/L. The average concentration observed by the Ministry of Agriculture and Fishery (2002) at St. 1 was 28.9 mg/L, but our observation was 33.4 mg/L at St. 1. Our calculation (Nakasone *et al.*, 2000) was 35.8 mg/L at St. 1. This is due to the high percentage of tea yards in the upper portion of the watershed as shown in Figure 3. The average observed concentration at station 4 was 13.5 mg/L during the irrigation period and was 21.9 mg/L during the non-irrigation period. Hence, our assumption was fairly correct. There are several small weirs in the Tanno river, then effluent water from the upper portion of the paddy field is used again for irrigation water in the downstream portion of the watershed. This may lead to a further decrease of nitrogen. We have tremendous paddy fields in Japan, hence, they have been playing important roles to reduce the nitrogen concentration of the rivers. We should need to focus more on the role of the paddy field these days.

Table 3 The areas of the each land use

Total area	Paddy field	Tea field	Forest	Other
479.9 ha (100%)	36.1 ha (8%)	171.8 ha (36%)	264.3 ha (55%)	24.4 ha (5%)

Conclusions

We performed this experiment in two places, Ibaraki Prefecture and Shizuoka Prefecture. We tested the removal rate from the difference of vegetation in the Ibaraki Prefecture. This experimental site had always been flooded for the past ten years and the water level was kept around 15 cm. The main aim performed in the Shizuoka Prefecture was evaluating the nitrogen removal by means of a paddy field at the watershed level. From these experiments performed in the two regions, we have come to the following conclusions.

1. The experimental results obtained from Ibaraki Prefecture could be applied not only to nitrogen removal in paddy fields but also to wetland systems all over the world.
2. The removal rate is calculated from Eq. (7) and it depends on the influent concentration X_o, removal coefficient α, and q. A decrease percentage is calculated from Eq. (9) and it depends only on the removal coefficient α and q.
3. The average removal rate was about 0.4 ($gm^{-2}d^{-1}$) for an ordinary paddy field and 0.3 ($gm^{-2}d^{-1}$) for the flooded wetland. This is because organic matter such as stems and roots decompose in the non-irrigation period. Therefore, denitirification is taking place vigorously during the irrigation period.
4. From the calculated water balance and assumed application amount of nitrogen fertilizer, the average T-N concentration during the non-irrigation period has to be 22.2 mg/L at an exit point (St. 4) of the watershed. The concentration has to decrease to 11.4 mg/L during the irrigation period by calculation due to denitrification in a paddy field. The average observed value was 13.7 mg/L during the irrigation period and was 21.9 mg/L during non-irrigation. Hence, it is possible to evaluate the removal amount at the watershed level.

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