A new method to estimate a ratio of nitrogen derived from chemical fertilizer to organic origin nitrogen in groundwater

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Abstract: A new separation of nitrogen index was proposed to estimate the nitrogen origin ratio of chemical fertilizer to organic sources in groundwater. It proved very useful for applying to heavy nitrate contamination in groundwater derived from livestock manure and fertilizer. SNI is defined as the equivalent ratio of SO_4^{2-} to a total nitrogen.

Key words: Nitrogen load, Nitrogen contamination, Livestock manure, Chemical fertilizer

Introduction

In Japan, the nitrogen contamination in groundwater has become a serious problem for sustainable drinking water supply. A new index is proposed for determining the source of nitrogen and for the estimation of the ratio of nitrogen of chemical fertilizer to organic origin. The index is an equivalent ratio of SO_4^{2-} to a total of nitrogen. The method was applied to a region of heavy nitrogen contamination where there are some different sources of nitrogen. The study area is one part of the slope of the Fugen volcano in the North West of Kyushu. The study area was composed of 29 % residential, 13 % paddy field, 26 % dry field, 1 % orchard, and 31 % forest of total area 61km^2. Beef and dairy cow (about 6100), pig (43000), chicken and broiler (about 1 million) were bred stock in the study area in 2001. Many livestock barns are scattered on the middle and upper parts of the slope. Groundwater samples were collected from wells for drinking water between September and October 2001 (28 samples) and June 2002 (13 samples).

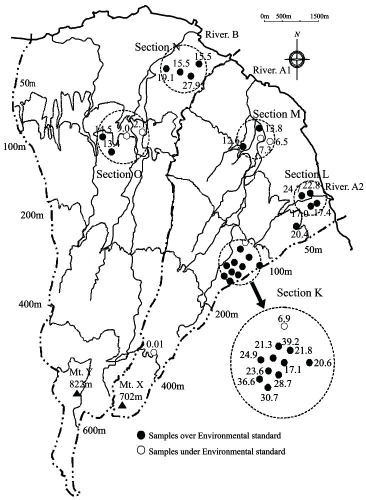

Figure1. Distribution of NO_3-N concentration

Results and discussion

Figure 1 illustrates the distribution of NO_3-N at 29 wells in the study area. NO_3-N concentrations of 32 samples from all 41 samples exceeded 10 mg/L (Japanese environmental quality standard). The average, lowest, and highest NO_3-N concentrations were 17.3 mg/L, 0.01 mg/L, and 39.2 mg/L respectively.

The theory about SNI

The Separation of Nitrogen Index SNI, is an equivalent ratio of SO_4^{2-} to a total of NH_4^+, NO_2^-, and NO_3^- shown as the following equation (1).

$$SNI\ (Separation\ of\ Nitrogen\ Index) = \frac{SO_4^{2-}\ (meq/L)}{NH_4^+ + NO_2^- + NO_3^-\ (meq/L)} \quad (1)$$

Scientific paper

The SNI value is low for water derived from livestock manure but is high for water derived from chemical fertilizer. It was reported that nitrogen and sulfur in the protein of livestock feed was 16 % and 1 % on average [1]. The ratio of sulfur to nitrogen in livestock manure is thought to be in agreement with that of livestock feed. SNI for the water derived from livestock manure is thought to be low. Ammonium sulfate ($(NH_4)_2SO_4$) has been usually used as fertilizer and it changes into nitrate in the soil zone as shown in the following equation (2) [2].

$$(NH_4)_2SO_4 + 4O_2 \rightarrow 2NO_3^- + SO_4^{2-} + 4H^+ + 2H_2O \qquad (2)$$

Therefore the SNI for water derived from chemical fertilizer is thought to be 1 or more because some nitrogen is absorbed by plants.

Verification of the Validity of SNI

Typical land use areas were studied in order to estimate SNI values for different land uses. Also, livestock wastewater and 4 supernatant waters obtained after dissolving 20 g of animal excrement in 100 g of water were used as representative samples originating from livestock manure. Groundwater samples (120 samples from each sites) were collected from a tea plantation region in Shizuoka, central Japan and used as representative water samples originating from chemical fertilizer. Tea growing uses a lot of chemical fertilizer, reaching up to approximately 11t/ha/year. Therefore, NO_3-N concentration in groundwater is increased by chemical fertilizer. Table.1 shows that SNI values for water derived from chemical fertilizer varies widely from 0.41 to 3.33 and is 1.39 on average. The livestock species and the waste disposal management are thought to greatly affect SNI for livestock manure. The SNI value of the water originating from livestock manure varies from 0.03 to 0.29 and is 0.1 on average. SNI values for livestock manure are low and SNI values for chemical fertilizer are high on the whole. Therefore, SNI is a clear indicator able to separate different nitrogen origins.

A mixing ratio of nitrogen derived from chemical fertilizer to total nitrogen was calculated from SNI values of the sampled groundwater using representative SNI values and is represented by the following formulas (3), (4). SNI values of each sample are between the representative SNI values of livestock manure (SNI_l) and chemical fertilizer (SNI_f).

$$SNI_s = SNI_l \times R + SNI_f \times (1 - R) \qquad (3)$$

$$SNI_s = 0.14 \times R + 1.39 \times (1 - R) \qquad (4)$$

Where R is mixing ratio of the representative chemical fertilizer and SNI_l depends on the waste disposal technique in the study area. Table.2 shows the number of livestock bred in the study area. Annual nitrogen load generation was calculated by multiplying the animal number by the nitrogen load generation of each species of livestock per animal per year. Also, each nitrogen load shown in Table-2 was calculated by multiplying each nitrogen load generation by a ratio of discharge that was 0.4 in the case of open stock feces and 0.65 in the case of disposal management of livestock wastewater including urine[3]. Then the proportion of each nitrogen source in the total nitrogen load was calculated. A mean value for SNI_l of 1.39 was calculated after multiplying each livestock SNI by the calculated discharge ratio and summing. SNI_f was the average value from 120 groundwater samples collected from the tea plantation. In this regard, mixing ratio of chemical fertilizer is taken as 0 or 1 if the calculated SNI value is under 0 or over 1.

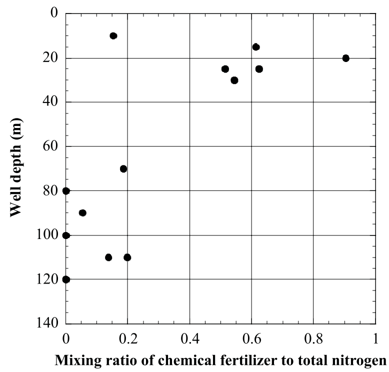

Figure 2. Relationship between the mixing ratio of chemical fertilizer to total nitrogen and well depth

Table 1. SNI values of typical contaminated

Type	Sample		NO_3^-N (mg/L)	SO_4^{2-} (meq/L)	$NO_2^-+NO_3^-+NM_4^+$ (meq/L)	SNI
Livestock manure	Feces of beef cow		ND	0.02	0.36	0.04
	Feces of dairy cow		ND	0.07	2.00	0.03
	Feces of pig		ND	0.51	7.21	0.07
	Feces of chicken		ND	1.49	5.15	0.29
	Livestock vastewater		ND	7.52	150	0.05
Livestock manure	Groundwater samples of tea plantation	Average	20.2	2.78	1.83	1.49
		Mininum	0.5	0.14	0.24	0.41
		Maximum	80.5	8.16	6.89	3.33

Table 2. Number and nitrogen load of each

Species	Cow		Pig		Chicken	Broiler	Total
Number	6142	43009	1050548	48000	1147699		
Excretory type	Feces	Urine	Feces	Urine	Feces+Urine	Feces+Urine	-
Daily nitrogen bad generation per livestock (gN/animal/day)	150	50	15	25	0.2	1.4	-
Annual nitrogen bad generation (tN/day)	336	112	235	392	767	24.4	1868
Annual nitrogen bad (tN/year)	135	73	94	255	307	10	873

Table 3. Measured results for every 50m altitude in Section K

Symbol	A	B	X	NLf	NL1	NL	NL1/NL
Altitude	Paddy field (ha)	Farm land (ha)	Livestock bam (animals)	Nitrogen bad from chemical fertilizer (tN/Year)	Nitrogen bad from livestock manure (tN/Year)	Total nitrogen load (tN/Year)	The ratio of livestock manure to total nitrogen (%)
0 m- 50 m	148	281	4	47.2	16.7	64.0	26.2
50 m-100 m	43	228	0	33.7	0.0	33.7	0.0
100 m-150 m	15	185	4	26.2	16.7	42.9	39.0
150 m-200 m	2	102	5	14.0	20.9	35.0	59.8
200 m-250 m	0	34	5	4.6	20.9	25.6	81.9
250 m-300 m	0	23	2	3.1	8.4	11.5	72.7
over 300 m	0	3	0	0.4	0.0	0.4	0.0

The verification of SNI using local measurement

Section K was selected as a model section to evaluate the suitability of estimating the source of nitrogen using SNI because the sampling was better than other sections. Fig.2 shows the relationship between the mixing ratio of chemical fertilizer to total nitrogen against well depth in Section K. The groundwater was classified into shallow groundwater (from 10 to 30 m) and deep groundwater (from 70 to 120 m). Although the mixing ratio for shallow groundwater is high (0.56 on average), the mixing ratio for deep groundwater is low (0.06 on average). These results show that shallow groundwater was mostly contaminated by chemical fertilizer more than livestock manure with 4:6 being the mean ratio of livestock to chemical fertilizer. Deep groundwater was contaminated more by livestock manure with a ratio of 9:1.

The land use area of paddy fields, orchards, and farmland in Section K was measured for every 50 m altitude from data issued by the National Land and Transportation Ministry. The number and locations of livestock barns were scanned from a land-use map of 1:10000 scale for every 50m altitude. The nitrogen loads from livestock manure and chemical fertilizer were calculated from land use for every 50 m altitude Section K, for the area between the east river and the west river of Section K, using the formulae (5), (6), (7).

$$\textit{Nitrogen load from chemical fertilizer } NL_{fi} = \sum (A_i \times N_p + B_i \times N_a)$$

$$\textit{Nitrogen load from livestock waste } NL_{li} = \sum X_i \times N_l \times \alpha$$

$$\textit{Total nitrogen load } NL_i = NL_{fi} + NL_{li}$$

i : Interbal altitude, α : Ratio of discharge = 0.4
N_p : Amount of nitrogen fertilizer appied into paddy field = 60 (kg/ha/year)
N_a : Amount of average nitrogen fertilizer appied into farmland = 136.5 (kg/ha/year)
N_l : Nitrogen load generation per livestock barn = 4186.7 (kg/piece/ year)
A_i : Paddy field(ha)
B_i : Farmland(ha)
X_i : Livestock barn(animals)

Table 3 shows the calculated results for every 50 m altitude. The nitrogen load derived from chemical fertilizer decreased with the decrease of farmland area from the upper slope to the lower slope. The nitrogen load derived from livestock manure did not greatly increase with altitude.
Consequently, the ratio of the nitrogen load from livestock manure to the total nitrogen load increased with altitude. According to Table 3, the main land use of Section K from 100 to 150 m in altitude is farmland and the shallow groundwater around Section K is mostly influenced by chemical fertilizer. At the upper slope of Section K deep groundwater is influenced by livestock manure. Therefore each nitrogen origin determined by SNI analysis was in agreement with the actual land use although the land use derived for deep groundwater by SNI is not directly in agreement with the surface land use at the deep groundwater sampling point. SNI was however a useful indicator for determining the ratio of nitrogen source to total nitrogen

CONCLUSIONS

The study area is one part of the slope of the Fugen volcano in the North West of Kyushu. High NO_3-N concentration wells are distributed over the whole of the slope and even wells of depth over 100 m have NO_3-N over 30 mg/L. A new index (SNI) was proposed to determine the source of nitrogen in groundwater. SNI is defined as the equivalent ratio of SO_4^{2-} to a total of NO_2^-, NO_3^-, and NH_4^+. SNI values of typical land use areas were measured in order to estimate actual SNI values. SNI values for livestock manure are low and SNI values for chemical fertilizer are high. A mixing ratio of nitrogen derived from chemical fertilizer to total nitrogen was calculated from the SNI values of the sampled groundwater using representative SNI values for fertilizer and livestock waste. The mixing ratio was calculated for heavily nitrate-contaminated groundwater. Each nitrogen origin determined by SNI analysis was in agreement with the actual land use. Therefore SNI analysis is a useful indicator for determining the ratio of nitrogen source to total nitrogen.

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