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Factors affecting spatial pattern of groundwater hydrochemical variables and nitrate in agricultural region of Korea

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(Received: May 8, 2019; Revised accepted: May 18, 2019)

<https://doi.org/10.18814/epiugs/2019/019011>

The overuse of fertilizers for high yields in agricultural production increases the leaching problem particularly in areas affected by heavy monsoon precipitations in Republic of Korea. The extent of leaching depends on various factors such as rate of monsoon precipitation, agricultural management practices, land use, soil properties, topographic features, and hydrochemistry. We studied spatial variations based on temporal fluctuations of groundwater level below the ground surface, pH, electrical conductivity (EC), and nitrate (NO₃) from year 2011 to 2014 in 70 groundwater wells in agricultural region of Korea. We found that groundwater level deepening across the studied period in agricultural area. Low pH (5.50–6.50) gradually vanished in year 2014 and high pH (8.51–9.50) appeared in the same year. Variations in the EC values shifted from high range EC (>500 µS/cm) to lower range (<100 µS/cm) from 2011 to 2014. Similar to spatial pattern of groundwater EC and contrary to pH, the high values (>50 mg/L) for groundwater NO₃ which appeared in the start of the study period, got vanished at the end of the study period. These findings are attributed to improved agricultural practices and change in land use pattern from vegetables to orchards and ginseng fields which do not require excessive fertilizer application.

Introduction

Fertilizers are applied extensively to increase crop production in agricultural areas, which results in excessive NO₃ in soil and water systems. One of the most prevalent and harmful impacts of NO₃ polluted groundwater (used for drinking purpose) is to pose risks of human and ecological health (Schroder et al., 2004). Drinking water having concentrations >10 mg/L for NO₃ causes human health problems such as blue baby syndrome and cancer in adults specially in the digestive tract (Powelson et al., 2008; World Health Organization, 2011; Kim et al., 2015). The high concentration of NO₃ in groundwater is a severe problem in Korean agricultural areas (Lee et al., 2017). Monitoring groundwater contamination and identifying factors influenced

the NO₃ concentration in groundwater are important steps to reduce contamination and manage groundwater quality.

Several factors have been identified affecting NO₃ contamination in groundwater, including amount of applied fertilizer and accumulation of soil organic content, which can lead to high NO₃ concentration and thus have higher rate of nitrate leaching to groundwater (Sieling and Kage, 2006). In addition to that, use of animal manure, crop growing duration (Rankinen et al., 2007), soil properties (De Ruijter et al., 2007), rate of monsoonal precipitation (Salo and Turtola, 2006), and topographic features such as elevation and land slopes in complex terrain area (Wick et al., 2012) have been found to effect the degree of agricultural NO₃ leaching to groundwater. Other potential factors have been investigated as sustainable agriculture practices, land use and land cover change, and change in hydrochemical variables of groundwater in changing climate (Rankinen et al., 2007; Buczko et al., 2010; Wick et al., 2012). Shallow groundwater level, the upper borderline of the saturated zone shows variation depending on seasonal changes in precipitation events (Kurunc et al., 2016). Fluctuation in groundwater level is important at watershed-scale, because of its nearness to the surface, chemical transport, drainage, and soil salinity.

Groundwater monitoring is fundamental to provide information for sustainable water management. Sustainable land and water management measures are based on the results of groundwater hydrochemical variables which provide the changes in groundwater quality and quantity. In agricultural areas, monitoring wells are generally used to assess spatiotemporal changes in groundwater level and groundwater quality variables (Kaman et al., 2011). Groundwater monitoring responds gradually to changes in agricultural practices and greatly influenced by weather events (Lord et al., 2002). Other than monitoring of groundwater wells data analysis is important to provide useful information of hydrochemical variables in the area. A systematic variation in spatial pattern allow to predict the value of a particular variable at specified location. Geostatistics has been used to analyze the systematic variation of water variables in space and time (Kurunc et al., 2016). Geostatistical analysis includes three steps: descriptive statistics, semivariograms for spatial structure, and predicting unknown value for specified variable at unvisited location. Spatial relationship between hydrochemical properties of groundwater is important for a sustainable water management planning. Kriging is widely geostatistical techniques for making spatial interpolation on sufficient data (Corwin et al., 1997).

Haeon basin is one of the most active highland agricultural areas in Soyang watershed, Korea with limited information on spatiotemporal distribution of hydrochemical variables and factors influencing their distribution. Hence, investigating the spatial and temporal variations in groundwater hydrochemical variables and NO_3 related to fertilization and seasonal differences in agricultural area were the purposes of this study. From 2011 to 2014, a visible change in factors affecting NO_3 in groundwater was observed in land use, rate of monsoonal precipitation, and fertilizer use. The study was conducted to evaluate temporal variations in spatial pattern of groundwater level, pH, EC, and NO_3 concentrations in groundwater wells in the Haeon basin. The special concern of this study was groundwater NO_3 pollution and factors influencing high or low concentrations of NO_3 in groundwater. The main objectives were to;

- 1) Find temporal variations in spatial pattern of groundwater hydrochemical variables and NO_3
- 2) Determine the potential factors that are controlling and increasing the NO_3 concentration in groundwater

The results of this study will help to understand the change in groundwater quality of an agricultural area and reasons behind improved or deteriorated groundwater quality. Such results might have important implications for agricultural areas with similar natural conditions including hydrogeological properties, climate conditions, and land use.

Materials and Methods

Study Area

The study area (Haeon basin) is located in Yanggu County of Gangwon Province and extends from $128^\circ 5' - 128^\circ 11' \text{E}$ in latitudes and $38^\circ 15' - 38^\circ 20' \text{N}$ in longitudes (Fig. 1a). This area covers approximately 64 km^2 of which 40% of the area is used for agriculture (rice paddy fields, vegetables, and fruits) (Kim et al., 2015). Remaining 2% is used for residential and 58% of area is covered with forested mountains surrounding the flat agricultural area. The geology of area in

Figure 1. (a) Location of the Haeon Basin in Republic of Korea, (b) Georeferenced locations of monitored wells and land use in the study area.

center is accredited to weathering of meta-sedimentary rocks and Jurassic granites and outer area comprised of meta-sedimentary rocks including mica schists, biotite-feldspar gneiss and quartzite (Lee et al., 2013). The base land contains alluvial deposits 5–10 m thick in area close to the relatively large (more than 2 m) streams in the flat area (Kaown et al., 2007). The mountains surrounding the agricultural area have elevation up to 1,320 m (above the sea level) with average slope of 20° in surrounding ridges and average slope of 5° in the center agricultural area (Kettering et al., 2012). The study area is in Humid Continental climate zone with mean annual temperature of 8.7°C which goes down to -27°C in January and goes up to 33°C in July (Ko et al., 2014; Lim et al., 2018). The average annual rainfall is 1,200 mm with 50% falling during the season of summer monsoon (Kim et al., 2018; Lee et al., 2018).

Groundwater is the major source of irrigation water and domestic water supply in the study area. There are some small streams that went dry during dry season, join to three large streams Dosol, Mandae, and Seonghwang (10–15 m wide) at the center of study area. Eventually these streams leave the study area at the eastern side and congregate with the Soyang River which is the major source of drinking water supply in the Seoul metropolitan city (Lee et al., 2013). The number of groundwater wells in study area increased due to increased agricultural activities.

Land Use

Approximately 58% (around 38 km²) of the study area is covered with forests, especially on the high elevation lots of the bowl-shaped Haeon basin (Kim et al., 2013). The highland forest predominately composed of 30 to 40 year old mixed deciduous forest with major species; Daimyo oak (*Quercus dentata*), Korean ash (*Fraxinus rhynchophylla*), and Mongolian oak (*Quercus mongolica*) (Shope et al., 2014). The remaining 2% is used for residential purpose and 38% of the study area principally in the lower elevation portion is used for agriculture conquered by rice paddy, vegetable fields, and orchards on intermediate elevation (Kim et al., 2015). Rice paddy fields are mostly located on lower elevation with flattened area, vegetable fields are located between paddy rice field orchards, and orchard are located on intermediate to high elevation before forests (Fig. 1b). Among agricultural crops in the study area rice prevails maximum (31.5%–51.5%, mean = 38.7%) of agricultural land followed by vegetable fields and orchards (Lee, et al., 2019). The main crops in vegetable field radish and cabbage have growing season starting before monsoon season and ends after monsoon. Rice crop has similar growing season. The area covered by vegetable fields got decreased during study period from 2011 to 2014 with an increase in orchard area (Lee et al., 2019). After 2010, the subsidies for the application of chemical fertilizer have been reduced by the government, but increased for the use of organic fertilizers. This has resulted in increasing use of organic fertilizers, while a decrease in use of synthetic fertilizers in the Haeon basin. According to an estimation the annual rate of organic fertilizer application in the Haeon basin has increased from 95 kg N ha⁻¹ in 2011 to 134 kg N ha⁻¹ in 2012 and so on in the next years (Yanggu County Office, 2014).

Hydrochemical Data of Groundwater

There were 70 groundwater wells observed and georeferenced with

geographical positioning system (GPS) device during first trip to the field of study. The monitored well location data was transferred to computer for processing in ArcGIS (version 10.4) to build digital map of study area (Figs. 1a and 1b). Groundwater samples were collected twice a year, once in dry season (September–May) and other in wet season (June–August) in year 2011, 2012, 2013, and 2014. Groundwater level was measured in all monitored wells using water level meter (Solinst model 101 P7) (Yun et al., 2017). The container used for groundwater sampling was first rinsed with the water sample and then analyzed for pH and EC using portable multi meter probe (HORIBA D-74 and D-75). One groundwater sample was taken from each well in a plastic bottle and 2 mL toluene was added to stop microbial activities. Each plastic bottle was labeled and stored in ice-box during transportation to laboratory and all groundwater samples were stored at <4°C before analysis. A Whatman-42 filter paper was used to filter the sample before NO₃ analysis in Ion Chromatograph (model DX-120, Dionex Corporation).

Geostatistical Analysis

There are three main stages involved in geostatistical analysis for this study. First stage consists of exploratory data analysis including the descriptive statistics. Second stage involves geostatistical modeling to measure the spatial structure of water quality parameters and fitted to theoretical models. Third stage is followed by two steps; spatial prediction of missing values and mapping of overall spatial structure based on data and predicted values.

Exploratory Statistics

Exploratory statistics of mean, median, minimum, maximum, standard deviation of mean, coefficient of skewness, coefficient of kurtosis, and coefficient of variation were calculated for groundwater level below the ground surface, pH, EC, and NO₃. The distributions of data were examined by Kolmogorov–Smirnov test with significant P-value (<0.05). Parametric statistical approach was used for normally distributed data and nonparametric approach was used where data distribution is not normal.

Spatial Structure Identification

Spatial structures of groundwater hydrochemical variables and NO₃ were assessed using geostatistical modeling technique, to generate mathematical models for spatial correlation structure (Sheikhy et al., 2013). A variogram is one of the rudimentary geostatistical tool that measures the spatial variability of different parameters between two points. A semivariogram is exactly same as variogram except the denominator in the equation, the number 2 is excluded. Semivariogram reveals the spatial correlation as a function of distance between locations (Fentanes et al., 2018). A mathematical expression is given as:

$$\gamma(h) = \frac{1}{2} \sum_{i=1}^{n(h)} \frac{[v(x_i) - v(x_i + h)]^2}{n(h)} \quad (1)$$

where $n(h)$ is number of data pairs at h distance, $v(x_i)$ is value at location x_i , $v(x_i + h)$ is value at location $x_i + h$.

There are three representative components generally considered for

semivariogram; nugget is the semi-variance at 0 distance measured as nugget variance, range is the distance (m) which is spatially correlated, and sill is the semi-variance at remoteness above the range, measured as structural variance sill. We used software GS+ to apply experimental semivariogram modeling on data. The best set of semivariogram including nugget, sill, and range components were generated by changing the active lag distance and lag class distance interval. Model fitting was done by changing model type, until minimum residual sum of squares (RSS) and maximum coefficient of determination (r^2) were achieved. Anisotropic semivariograms were calculated at n degrees (0° , 45° , 90° and 135°) (Isaaks and Srivastava, 1989). The fitted variogram model was used to predict the parameter value at un-sampled locations.

Prediction and Mapping of Spatial Pattern

Geostatistical interpolation is a powerful technique to estimate and present the spatial pattern from point data (Belkhirri and Narany, 2015). The application of geostatistics is required when, spatial structure looks more similar for sampling points near to each other than sampling points far apart from each other. Kriging is an appropriate interpolation method which demonstrates the likelihood of interpolation of the approximation error of the variables where initial data is lacking. ArcGIS was used to conduct kriging interpolation and mapping of kriging predicted values for the groundwater hydrochemical variables and NO_3 . Expression of kriging estimations is given as Kumar (2007):

$$\hat{Z}(S_0) = \sum_{i=1}^N \lambda_i Z(S_i) \quad (2)$$

where $Z(S_i)$ is measured value at the i location, λ_i is the unknown weight of measured value at i location, S_0 is predicted location, and N is number of measured values.

Kriging estimates presented with 95% confidence intervals and digitally mapped for predicted values of nitrate and important water quality parameters. Maximum 14 and minimum 7 neighboring values of study area were used in kriging predictions. Digital kriging map was generated for groundwater level using class intervals of 0–5, 6–10, 11–20, and 21–30 m and maps were used for evaluating the presence of water quality variables. The kriging prediction map of groundwater pH was built with class intervals of 5.50–6.50, 6.51–7.50, 7.51–8.50, and 8.51–9.50. Digital map kriging prediction of groundwater EC was generated using class intervals of <100.0, 100.0–250.0, 250.1–500.0, and >500 $\mu\text{S}/\text{cm}$. The kriging prediction map of groundwater nitrate was built with class intervals of <10.00, 10.01–25.00, 25.01–50.00, and >50.01 mg/L. After generating maps, the areal coverage of each class was calculated in km^2 along with its percentage to observe spatiotemporal variations.

Results

Hydrochemical Characteristics

The hydrochemical results of groundwater samples showed in Table 1, indicate descriptive statistics for the groundwater variables collected in rainy and dry seasons from year 2011 to 2014. Groundwater hydro-

chemical variables were measured in 70 monitoring wells located in the study area. Some monitoring wells were inaccessible for sampling in different seasons, except the first rainy season of sample collection. The groundwater level was highly variable and indicate lowering of groundwater level across the studied years. Mean for groundwater level fluctuated between seasons while median indicated deep groundwater level during dry seasons of years except for the dry season of 2014, when median of rainy season was higher than dry season and showing deep groundwater level. Data with high maximum values become strongly right skewed with high kurtosis and high variance values. This suggests high values (deep groundwater level) took place in the season that changed the median value from the mean value. Maximum values for groundwater level are generally more important than mean or median values since those values do not indicate the mutilation caused by high values in the study area. The minimum values of groundwater level fluctuated, while maximum values got increased and results in deepening of groundwater level across the studied period.

The pH is important variable which controls most of the biochemical processes in groundwater. Table 1 indicates the mean of groundwater pH increased gradually from 6.55 to 7.82 during the studied period, but slightly decreased in fourth season (dry season, 2012) and then sharply increased until the dry season of 2014. Variance, strong positive skewness, and high kurtosis values followed the high maximum values of pH that were mostly observed in rainy season of the years, except in 2011 when high maximum value occurred in dry season. The maximum pH value increased from 6.94 in the dry season of 2011 to 9.50 in dry season of 2014, while minimum values do not show noticeable fluctuations.

The distribution of EC is indicator of overall contaminants present in the study area. In Table 1, the mean EC value gradually decreased from 604 to 80 $\mu\text{S}/\text{cm}$ with a slight rise in fourth season (dry season, 2012). Extremely right skewed, variance, and kurtosis followed the similar pattern as of EC maximum values that decreased from 1,298 to 109 $\mu\text{S}/\text{cm}$ during the studied period. Among different seasons, EC found to be higher in rainy season, except in year 2012 when it was higher in the dry season. The outlook of groundwater EC distribution suggests a gradual drop in value irrespective of seasonal variations.

Table 1 shows, the mean values for groundwater NO_3 decreased gradually during the study period except in the second season (dry season, 2011). The NO_3 values followed a similar decreasing pattern to EC value and opposite to pH and groundwater level increasing values, across the study period. Among seasonal variations, the maximum higher values of groundwater NO_3 were observed in rainy seasons which decreased from 88.24 to 19.10 mg/L. Skewness, kurtosis, and variance followed the similar decreasing pattern as of maximum NO_3 value. Very high variance particularly for groundwater EC and NO_3 are demonstrating large variability of the considered variables in the study area.

Spatial Structure of Groundwater Hydrochemical Variables and NO_3

Spatial structure of groundwater level varied across the eight studied seasons had a temporal stability in structure. Fig. 2 demonstrated the theoretical model and related parameters for the spatial structure during rainy and dry season of the studied years. The proportion of nugget variance and sill $C_0/(C_0+C)$ increased unexpectedly in both

Table 1. Overall descriptive statistics of hydrochemical data collected in rainy and dry season from 2011 to 2014

Parameter	GWL (m)	pH	EC ($\mu\text{S}/\text{cm}$)	NO_3 (mg/L)	GWL (m)	pH	EC ($\mu\text{S}/\text{cm}$)	NO_3 (mg/L)
2011	Rainy season (June–August)				Dry season (September–May)			
n (n.m.)	70 (0)				65 (5)			
Mean	3.18	6.55	604	34.40	2.80	6.69	272	38.80
Median	2.21	6.53	453	33.64	2.35	6.59	272	38.90
Min	0.69	6.17	297	1.96	1.1	6.34	265	6.60
Max	17.24	6.94	1298	88.24	10.45	7.18	282	70.81
SD	2.84	0.31	391.65	32.17	1.83	0.33	7.08	26.57
S	3.24	0.04	1.20	0.52	2.54	0.72	0.45	-0.02
K	13.25	-1.82	-0.09	-0.93	8.25	-0.78	-1.25	0.71
V	8.06	0.09	1.53	1.03	3.35	0.11	50.2	706.1
2012								
n (n.m.)	67 (3)				66 (4)			
Mean	2.91	6.80	212	33.00	3.18	6.69	248	29.40
Median	2.50	6.85	216	31.71	2.73	6.52	254	33.15
Min	0.72	5.79	188	8.69	1.23	6.11	235	6.67
Max	10.10	7.88	234	66.81	15.4	7.20	256	47.44
SD	1.87	6.80	17.14	19.60	2.65	0.45	9.57	17.32
S	1.92	0.18	-0.11	0.22	4.08	-0.01	-0.8	-0.20
K	5.21	-0.34	-1.80	-1.37	17.21	-1.80	-2.05	-2.15
V	3.51	0.32	293.86	384.31	7.06	0.2	91.7	300.23
2013								
n (n.m.)	62 (8)				60 (10)			
Mean	4.22	6.98	179	23.26	4.23	7.16	161	14.92
Median	3.15	6.90	180	21.50	3.17	7.02	159	13.94
Min	1.03	6.05	176	8.97	1.20	6.70	150	0.11
Max	13.28	8.14	182	39.69	19.82	7.71	175	33.88
SD	3.52	0.49	2.44	10.09	3.89	0.34	7.95	11.57
S	1.88	0.45	-0.50	0.12	3.27	0.42	0.30	0.30
K	2.57	0.42	-1.40	-1	12.25	-1.29	-1.19	-0.71
V	12.39	0.24	5.98	101.88	15.15	0.11	63.24	133.88
2014								
n (n.m.)	66 (4)				65 (5)			
Mean	5.17	7.50	125	12.87	4.08	7.82	80	9.46
Median	3.36	7.48	128	11.35	3.21	7.88	84	9.00
Min	1.25	6.11	99	0.20	1.41	6.77	56	0.93
Max	29.5	9.50	143	34.92	12.67	8.65	109	19.10
SD	5.9	0.94	13.63	11.65	3.01	0.55	13.27	6.31
S	3.39	0.93	-0.35	0.54	2.07	-0.71	-0.54	0.46
K	13.09	1.42	-1.19	-0.83	4.66	0.82	-0.01	-0.54
V	34.91	0.89	185.93	135.82	9.08	0.30	176.27	39.91

n: observation number; n.m.: Not measured; Min: minimum; Max: maximum; SD: standard deviation; S: skewness; K: kurtosis; V: variance; GWL: ground water level below ground surface; EC: electrical conductivity.

Figure 2. Semivariogram for groundwater level below ground surface, pH, EC, and NO_3 in rainy and dry seasons from 2011 to 2014. Red line represents sample variance and black line shows variogram model. C_0 : Nugget variance; $C_0 + C$: Structural variance sill; A_0 : Range; r^2 : Coefficient of determination; RSS : Residual sum of squares; n.a.: Not available.

seasons of year 2013. This indicates that spatial heterogeneity in the spatial structure of groundwater level, caused by random aspects was minimal. Increase in nugget variance is followed by increased range value of the theoretical models. Table 2 indicated the spatial structure of groundwater level falls in class of strong spatial dependence during both seasons of the studied years.

Groundwater pH showed high values of proportion $C_0/(C_0+C)$ during dry season of 2011, dry season of 2013, and rainy season of 2014 with pure nugget effect (Fig. 2). The large range of theoretical models followed by high nugget variance, which is indicator of extent of range cannot be measured with present sampling paradigm or due to some experimental errors. The spatial dependence of groundwater pH structure showed similar pattern of high nugget variance with weak spatial dependence (Table 2).

Spatial structure of EC was weakly spatially dependent in rainy season of 2012 and dry season of 2013, while in other seasons it showed strong spatial dependence (Table 2). During some seasons, strong spatial autocorrelation in the study area is result of pairing made between values in different locations and the values at highest value locations. The spatial structure of EC will follow wider range of classified values. Fig. 2 is showing high range values where pure nugget effect is observed.

Contrary to other variables, spatial structure of NO_3 varied highly across the studied period as showed in Fig. 2, differed in theoretical models (Exponential, Gaussian, Linear, and Spherical) and related parameters. Table 2 shows that groundwater NO_3 was weakly to partially to strongly spatially dependent. Highest proportions $C_0/(C_0+C)$ found in dry season of year 2011 and 2014, specifically in dry season of 2011 indicated presence of short range dissimilarity that could not be measured by the existing sampling paradigm. This nugget effect can be attributed to experimental error, that it was unable to adopt any of the theoretical model and falls in weakly spatially dependent class. Other's proportion $C_0/(C_0+C)$ values are indicating partial to strong spatial dependence in dry season of 2012, which gone through same experimental procedures as the values in weakly spatially dependent class. These long range variations in spatial structure of groundwater

NO_3 might be an indication of interactions between groundwater nitrogen and pH. A higher pH value >7 can enhance denitrification processes in anaerobic to semi-anaerobic conditions (Glass and Silverstein, 1998).

Spatial Pattern of Groundwater Hydrochemical Variables and NO_3

Maps based on the kriging interpolation of the groundwater level for the ranges 0–5, 6–10, 11–20, and 21–30 m of depths showed that the inconsistency of groundwater level was intermediate and its spatial pattern was in the form of intermediate sized patches (Fig. 3). The variability in groundwater level increased over the years 2011–2014, with deeper water level values. The most extreme changes arisen at some locations in north-western part of the study area, although some locations with shallow groundwater level in the middle portion of the study area remained relatively unchanged. The variation in the areal coverage of groundwater level ranges across the rainy and dry seasons during the studied time period was inconsistent, but generally giving an outlook of shallow groundwater levels during the dry season. This outlook is attributed to differences in use of irrigation water and cropping season. In addition, the first and third year were different from the second and fourth years in climate variable like monsoon precipitation and temperature.

Spatial pattern of pH was highly different in the first year having wide patches from the last year values with small patches (Fig. 4). The wide patches represents small variations in the pH value, while the small patches of pattern are showing large variation in pH values. Areal coverage for pH 5.50–6.50 gradually vanished in the last year and areal coverage of pH 8.51–9.50 appeared in the last year which revealed an increasing trend of pH value across the studied period. Change in groundwater pH have influence on groundwater NO_3 and EC value. Groundwater chemical composition is based on its pH value along with other climatic factors. For example, in this case, increased pH in the study area from low to high, will result in increase of denitrification process to minimize NO_3 concentrations.

The kriging map of groundwater EC is showing drastic changes in

Table 2. Spatial dependence of groundwater level below ground surface, pH, EC, and NO_3 in wet and dry seasons from 2011 to 2014

Parameter	GWL (m)	pH	EC ($\mu\text{S}/\text{cm}$)	NO_3 (mg/L)	GWL (m)	pH	EC ($\mu\text{S}/\text{cm}$)	NO_3 (mg/L)
2011	Rainy season (June–August)				Dry season (September–May)			
Proportion ($C_0/[C_0+C]$)	0.12	2.90	2.97	17	0.03	25	0.11	n.a.
Spatial dependence	Strong	Strong	Strong	Partial	Strong	Partial	Strong	Weak
2012								
Proportion ($C_0/[C_0+C]$)	0.006	2.90	100	18	0.09	2.38	0.04	0.68
Spatial dependence	Strong	Strong	Weak	Partial	Strong	Strong	Strong	Strong
2013								
Proportion ($C_0/[C_0+C]$)	3.60	4	0.12	30	5.60	100	100	15
Spatial dependence	Strong	Strong	Strong	Partial	Strong	Weak	Weak	Partial
2014								
Proportion ($C_0/[C_0+C]$)	0.25	100	8.55	91	0.11	0.28	0.05	100
Spatial dependence	Strong	Weak	Strong	Partial	Strong	Strong	Strong	Weak

C_0 : Nugget variance; C_0+C : Structural variance sill; n.a.: Not available.

Figure 3. Spatial pattern of groundwater level below ground surface across different seasons from 2011 to 2014. Areal surface coverage is given in km^2 along with percentage (%) in the study area.

Figure 4. Spatial pattern of groundwater pH in different seasons for the period of 2011–2014. Areal surface coverage is given in km^2 along with percentage (%) in the study area.

Figure 5. Pattern of spatial variations in groundwater EC ($\mu\text{S}/\text{cm}$) during different seasons from 2011 to 2014. Areal surface coverage is given in km^2 along with percentage (%) in the study area.

its spatial pattern in the study area (Fig. 5). Variations in the EC values remained low within a season, but shifted from high range EC ($>500 \mu\text{S}/\text{cm}$) to lower range ($<100 \mu\text{S}/\text{cm}$) from 2011 to 2014. The United States Salinity Laboratory categorized groundwater based on EC values as; excellent ($<25 \mu\text{S}/\text{cm}$), good ($25\text{--}75 \mu\text{S}/\text{cm}$), fair ($75\text{--}225 \mu\text{S}/\text{cm}$), and poor ($>225 \mu\text{S}/\text{cm}$) (Kottureshwara et al., 2014). Results of this study exhibited that poor groundwater disappeared after 2012, and good to excellent groundwater appeared in 2014. This trend is attributed to increasing pH values across the period of study and intra seasonal variations are accredited to varied monsoon precipitations and over use of groundwater which concentrated the groundwater EC values.

Seasonal changes in spatial pattern of groundwater NO_3 from 2011 to 2014 are shown in Fig. 6. The areal coverage of different ranges for groundwater NO_3 varied differently across the study period. In the first year wider range resulted in small patches of groundwater NO_3 , while in the last year small variation in groundwater NO_3 formed large patches of pattern. Similar to spatial pattern of groundwater EC and contrary to pH, the high values ($>50 \text{ mg}/\text{L}$) for groundwater NO_3 which appeared in the start of study period, got vanished at the end of study period. These high NO_3 values are hazardous for groundwater aquifer, so monitoring of such location is of environmental importance. The locations with high NO_3 values appeared at different loca-

tions during different seasons and years, indicating that the sources of high values were different every time.

Discussion

Factors Affecting Groundwater NO_3 Concentrations

Haeon basin is one of the active agricultural highland area in the Soyang watershed, a water resource for downgradient Seoul metropolitan population (Lee et al., 2019). In such regions, high concentration of groundwater and surface water NO_3 is a problem due to use of inorganic and organic fertilizers. There are number of natural and anthropogenic factors that influence high NO_3 in groundwater including fertilizer use, different land use, rate of precipitation during rainy season, and groundwater recharge enhancing NO_3 leaching to groundwater (Jang et al., 2017). Some factors control the NO_3 in groundwater including pH, denitrifying bacteria, abundance of dissolved oxygen, geological setting that prohibits NO_3 leaching to groundwater, area topography, and the amount of precipitation (Ruidisch et al., 2013; Seo et al., 2014).

Figure 6. Spatial variation of groundwater NO_3 (mg/L) in rainy and dry seasons from 2011 to 2014. Areal surface coverage is given in km^2 along with percentage (%) in the study area.

Agriculture Activity and Fertilizer use

Groundwater NO_3 pollution is majorly attributed to excessive fertilizer use for high yields in agriculture productions. Haeon basin is an intensive agricultural area with multiple land uses have been involved in overuse of fertilizers. In the Haeon Catchment fertilizer application contributed to >90% of the NO_3 inputs and a minor contribution from direct precipitation as compared to annual N-fertilizer use in the agricultural part of Haeon basin (Kettering et al., 2012; Suarez et al., 2019). We found groundwater in wells located under vegetable fields had the highest NO_3 concentrations (Fig. 6). The elevated groundwater NO_3 concentrations are affected by fertilizer leaching from agricultural fields. Particularly in areas with high precipitation rates during monsoon and coarse-textured soils having the potential of NO_3 leaching is high (Zotarelli et al., 2007). In study area, topsoil is categorized as a coarse soil texture, which overlies by finer textured subsoil and an underlying dense bedrock material. These situations enriched by the application of sandy soil and fertilizers before growing season (Ruidisch et al., 2013).

The World Health Organization recommended a maximum NO_3 concentration of 50 mg/L in drinking water, while Republic of Korea is even stricter with the maximum concentration of 44.3 mg/L for NO_3 and 10 mg/L for $\text{NO}_3\text{-N}$ in drinking water (Min et al., 2002; Choi

et al., 2007; Ruidisch et al., 2013). The Korean government reduced the subsidies for the use of synthetic fertilizers while increased for organic fertilizers, after year 2010. As a result, a decrease in synthetic fertilizers use from 179 to 145 $\text{kg N ha}^{-1} \text{yr}^{-1}$ was observed during the study period (Kim et al., 2015). On the other hand, an increasing use of organic fertilizers from 80 to 129 $\text{kg N ha}^{-1} \text{yr}^{-1}$ was observed during the study period (Yanggu County Office, 2014; Kim et al., 2015). The amount of applied fertilizers was different for different crops grown in the study area (Table 3). The fertilizer application to fields started with the start of crops, and their growing season was approximately same as the monsoon season in the study area. Effect of applied fertil-

Table 3. Annual use of fertilizers in main crops of the Haeon Basin (Yanggu County Office, 2014)

Crop	Fertilizer use ($\text{kg N ha}^{-1} \text{yr}^{-1}$)	Fertilizing frequency per year
Chinese cabbage	238	Two
White radish	225	Three
Potato	137	One
Soybeans	30	One
Rice	90	Two

izer on groundwater quality starts with leaching of NO_3 from soil to groundwater, which is influenced by precipitation in unsaturated zone.

Seasonal Variations

High seasonal variation of groundwater NO_3 were accredited to management practices i.e. time and amount of applied fertilizers, irrigation time and amount of water applied, amount of precipitation. The excessive fertilization of crop fields resulted in elevated groundwater NO_3 concentrations owing to leaching losses from crop fields into the groundwater, enforced by monsoonal precipitation (Bartsch et al., 2013, 2014). The accessibility of NO_3 can be managed by farmers with their choice of fertilizer application rates, but NO_3 leaching is highly dependent on the amount and period of monsoon precipitation (Lee et al., 2014). Variations in precipitation during monsoon season can cause risks in terms of water quality and alterations in hydrological and biogeochemical drivers of NO_3 leaching to groundwater, from the applied fertilizers on soil surface (Arnhold et al., 2014; Suarez et al., 2019). The precipitation in the monsoon season fluctuated particularly in the four years of this study (Lee et al., 2019). An estimated rate of NO_3 leaching was >50% and up to 75% initiated from fertilized upland fields in the study area (Kim et al., 2015). In addition to by leaching during precipitation, slope of the area may contribute in reduced groundwater NO_3 concentrations at higher slopes occupied by forest with absence of fertilizers. An intermediate terrain area comprised of highly fertilized vegetable fields showed high NO_3 concentrations in groundwater. Relatively flat area of rice paddies have lower NO_3 in groundwater which is attributed to presence of denitrifying bacteria in abundance (Seo et al., 2014; Kim et al., 2015). The variation in seasonal precipitation may not have direct effect on groundwater NO_3 but, it can influence the factors controlling NO_3 in groundwater (Shope, 2016).

Land Use

Land use and seasonal variations are interdependent and of equal importance for the groundwater quality in areas under agricultural activities (Bartsch et al., 2013; Ali and Reineking, 2016). Different land uses have different effects on groundwater NO_3 as shrub and grasslands have lowest groundwater NO_3 values, whereas vegetable fields have higher or highly variable NO_3 concentrations in groundwater (Pasten-Zapata et al., 2014). The change in trend from annual to perennial crops assists the reduction of NO_3 surplus with spatially limited applications in the future (Ruidisch et al., 2013).

Application of fertilizers to agricultural area caused >90% of NO_3 surplus in the Haeon basin. Leaching and surface runoff were found as the main loss pathways for surplus NO_3 (Kettering et al., 2012; Shope and Maharjan, 2015). Among cultivated crops in the study area, radish indicated the highest uptake efficacy (43–45%) for NO_3 , whereas rice showed the lowest uptake efficacy (24–30%). The observed contribution to NO_3 surplus in agricultural land use showed an order as; cabbage < potato < soybean < radish, while ginseng and orchards did not play a substantial role (Ruidisch et al., 2013; Lee et al., 2019). Since 2011, increased perennial crops like ginseng and fruit orchards assumed to have a lower groundwater NO_3 due to lower fertilizer inputs, lower leaching, and higher surface runoff at high slopes (Lee et al., 2019).

Varying Hydrochemistry

The biogeochemical and hydrological processes in the groundwater are based on the chemistry of groundwater. Fig. 7 showed an increase in groundwater level value (groundwater deepening) across the studied period. Deepening of the groundwater level from 2011 to 2014, is attributed to a gradual increase in groundwater abstraction majorly for agricultural activities in Haeon basin, which resulted in observable drop of average groundwater level (Raza et al., 2019). Decreasing groundwater NO_3 in this study, found to have negative correlation with the increasing groundwater level from the soil surface, because fertilizer application mostly effect the shallow groundwater (Pociene and Pocius, 2005). Lowered groundwater level has less chances of having high NO_3 concentration due to anthropogenic activities in active agricultural areas.

An increase of groundwater pH is important since denitrifying bacteria are able to decontaminate groundwater at higher pH values (Seo et al., 2014). A microbial analysis supported high occurrences of denitrifying bacteria in groundwater of the paddy rice fields at relatively low elevation area in Haeon basin (Kim et al., 2015). Fig. 7 showed an increasing trend of pH value across the studied period. This situation can lead to decrease in groundwater NO_3 concentration, as a result of denitrification in shallow groundwater under the rice fields. A drastic change in pH from 5.5 to 9.5 in 2014 is attributed to precipitation as source in shallow groundwater during rainy season in the studied area (Lee et al., 2014). Another anthropogenic source of elevated groundwater pH was found as, corrode pipes of groundwater wells in both shallow and deep groundwater levels (Ann-Chatrin et al., 2017). Interactions between groundwater pH and NO_3 would be additional reason for the NO_3 variation in its spatial pattern. The spatial distribution maps indicate a link between two variables as the areas with elevated pH value have lower NO_3 value in groundwater.

This study exhibited that area with good EC in groundwater (<100 $\mu\text{S}/\text{cm}$) appeared in 2014 and area with high groundwater EC (>500 $\mu\text{S}/\text{cm}$) disappeared after the first year. Unnecessary use of fertilizer and irrigation water in agricultural fields are the major factors provoking EC in the groundwater. Presence of chemical species cause high EC and their sources should be identified for mitigation of further harm to groundwater. Under intensive agricultural production, substantial amount of salts can move beyond the root zone, deteriorating below groundwater. Plants uptake good or pure water and leave salts that percolate into groundwater (Chaudhuri and Ale, 2014). In this study, overall spatial and temporal distribution trend of groundwater EC and NO_3 is similar (Fig. 7).

Measures to Reduce Groundwater Nitrate Concentrations

The pattern with high NO_3 concentrations in groundwater vanished toward the end of the studied period. Those high concentration patterns should be monitored specifically to avoid a NO_3 risk on groundwater aquifers and surface water. Difference in appearance and coverage of high NO_3 concentration suggesting that the sources of high concentrations were anthropogenic i.e. fertilizer application to agricultural fields.

Numerous measures can be applied to reduce NO_3 concentration in groundwater. Soil properties including its water holding capacity and

Figure 7. Temporal trend of groundwater hydrochemical variables and NO₃ in agricultural Haeon Basin.

hydraulic conductivity should be considered before fertilizer application and irrigation, in order to minimize NO₃ leaching below the root zone (Ren et al., 2010). Sprinkler irrigation method results in a decreased NO₃ leaching as compared to flooded or furrow irrigation method, because water is more uniformly distributed on the soil surface. Rate of water application should be slightly lower than the soil infiltration rate to avoid rapid transport of NO₃ to groundwater. Fertilizer application should be in smaller amount but more frequently, particularly during the rainy season, to reduce fertilizer movement to the groundwater. Also, the irrigation water and fertilization amount should be adjusted according to the amount of precipitation during rainy and crop growing season, to avoid NO₃ leaching to groundwater. Changing the cropping pattern from crops without plastic or organic mulching to crops with mulching cover would reduce NO₃ leaching to groundwater.

The vegetable crops on high elevation fertilized more which results in high NO₃ in groundwater. Increasing area for ginseng and orchard land use have reduced the groundwater NO₃, but vegetable crops require some best management practices to minimize NO₃ in groundwater. A trend of using plastic mulching cover in vegetable fields resulted in 26% of reduction in NO₃ leaching to groundwater, irrespective of applied fertilizer amount and elevation of the area (Berger et al., 2013; Nguyen et al., 2014). All sustainable management practices including split and ridge fertilizer application with plastic mulching cover can reduce the rate of NO₃ leached to groundwater.

Conclusions and Required Work

Variation in temporal trend and spatial distribution of important groundwater hydrochemical variables and NO₃ is important for planning sustainable management practices to protect groundwater quality in the Haeon basin. The results indicated groundwater high NO₃ concentrations under the vegetable field land use at relatively higher elevation. Anthropogenic factor elevating the groundwater NO₃ is the excessive

use of fertilizers which is influenced by natural and anthropogenic factors including seasonal variation, slope of area, land use, and change in hydrochemical parameters. Areas with high EC, pH, and NO₃ in groundwater should be monitored appropriately and implement mitigation measures to control factors that are elevating the groundwater contamination. The results and mitigation measures in this study can be applied to another highland area at catchment scale or with heavy agricultural activities.

A switch from high fertilizer demanding vegetable fields to less fertilizer demanding ginseng and fruit orchards is part of sustainable agriculture practice along with split application and plastic mulching cover. In present and future, there is concern to investigate hydrochemical variables and NO₃ in groundwater to draw an appropriate trend. Such trend identification can provide quantitative change of groundwater quality either improved or deteriorated. All considered measures can reasonably reduce the amount of NO₃ leached to groundwater, which is of environmental importance. In economic perspective, measures can reduce the net farm productivity like use of plastic mulch and limited fertilization. There should be more practice to find sustainable agriculture measures that are environment friendly with same economic benefits. Groundwater quality can be improved by implementing the mitigation measures, but it is problematic to decide whether adopted measure is sustainable, if we consider the surface water quality as well. Increasing precipitation amount and reduced NO₃ leaching to groundwater can result in increased NO₃ leaching to stream and river water in the area. There is a need to investigate the effects of measures for improved groundwater quality on the surface water quality of the Haeon basin and in the downgradient area.

Acknowledgements

This work was supported by the National Research Foundation of Korea (NRF) grant, funded by the Korean government (MSIT) (NRF-2015R1A4A1041105) and Basic Science Research Program through

the National Research Foundation of Korea (NRF) funded by the Ministry of Education (NRF-2018R1D1A1B07047200).

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