Korean J. Soil Sci. Fert. **42**(1), 37-45(2009)

Article

Regional-Scale Evaluation of Groundwater Susceptibility to Nitrate Contamination Based on Soil Survey Information

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Susceptibility assessment of groundwater contamination is a useful tool for many aspects of regional and local groundwater resources planning and management. It can be used to direct regulatory, monitoring, educational, and policy-making efforts to highly vulnerable areas. In this study, a semi process-based was proposed to evaluate relative susceptibilities to groundwater contamination by nitrate on a regional scale. Numerical simulation based on data from each soil series was done to model water flow within soil profiles that were related to groundwater contamination by nitrate. Relative vulnerability indices for each soil series were produced by manipulation of amount of leaching flux, amount of average water storage in a soil profile, and amount of average water storage change. These indices were designed to convey the trend of leaching flux and to maximize spatial resolution. The resulting vulnerability distribution map was used to locate highly vulnerable sites easily with an appropriate grouping the indices, and was then compared with those from groundwater nitrate concentrations monitored. An excellent agreement was obtained across nitrate concentrations from the highly vulnerable regions and those from the low to stable regions.

Key words: Water movement, Simulation, Soil series, Pedotransfer function, Geographical information system

Introduction

The groundwater, another important water resource, is generally in stable condition throughout the nation. The nation's total reserves of groundwater amount to 1.5 trillion m^3, which is 12 times the volume of annual precipitation (Yoo and Jung, 1999). An estimated 13.6 billion m3 of the total groundwater reserves can be tapped for use. It is estimated that 23 billion m^3 of water, 18% of the total amount of annual precipitation, flows into groundwater annually. At present, about 2.6 billion m^3 of groundwater, accounting for about 10% of total water in use, is used annually (Lee, 1994). The needs for development of groundwater resources is increasing because of limited supplying capacity and increased pollution of the surface water resources. Concomitant with the increased reliance on groundwater has come the need to protect groundwater resources from possible contaminations (Aller et al., 1987).

The control practices to protect groundwater resource may require enormous monitoring efforts and financial burden on regulators. Therefore, a system evaluating the relative vulnerability of areas to groundwater contamination from various sources of pollution can have actual value in assisting planners, managers, and administrators (Aller et al., 1987; NRC, 1993). It can be used to help direct resources and land-use activities to the appropriate areas including (1) prioritization of areas where groundwater protection is critical, (2) identifying areas where special attention, or protection efforts are warranted, (3) prioritization of areas for monitoring purposes, a denser monitoring system might be installed in areas where pollution potential is higher and land use suggests a potential source, and (4) efficient allocation of resources for clean-up and restoration efforts.

Often, nitrate is regarded as the most important pollutant in assessing groundwater quality. It is well known that high level of nitrate in drinking water can cause methemoglobinemia, chronic toxicity, and possible development of cancer from nitrosamines (Comly, 1945; Preussman and Stewart, 1984). Nitrate, beside its importance in water quality, has been also regarded as an indicator (surrogate contaminant) for vulnerability of groundwater to contamination by nonpoint sources (Cohen et al., 1984; Domagalski and Dubrovsky, 1992; Richards et al., 1996). It is reasonable to assume that if nitrate can migrate from the surface to groundwater, there

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Received : January 2. 2009 Accepted : February 6. 2009

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can be also viable pathways for pesticides and other pollutants. Nitrate can be leached very easily into groundwater compared to other pollutants because it is anionic and not adsorbed to soil matrix. Its movement into groundwater is mainly controlled by the water transport process (Viten and Smith, 1993).

The most common and/or simple assessment of groundwater susceptibility to nitrate contamination is the parameter weighting method such as DRASTIC and SEEPAGE (Aller et al., 1987; USEPA, 1993; Evans et al., 1995). However, since these approaches rely on simple mathematical representations of expert opinion, and not on process representation or empirical data, there are arguments on whether the factors included in those methods are appropriate and whether the factors are the relevant ones for vulnerability assessment (Regan, 1990; Pettyjohn et al., 1991; Riggle and Schmidt, 1991). On the other hand, sophisticated process-based approaches require extensive model parameters to consider all the detailed geophysical processes of transport and transformation (Rundquist et al., 1991, Tomas, 1992; Navulur and Engel , 1996). Naturally, many of those parameters are not readily available and thus require laborious laboratory or field efforts to evaluate them.

The primary objective of this study was to develop a semi process-based evaluation methodology for groundwater susceptibility to nitrate contamination, which can be applied on a regional scale with minimum model parameters or at least with parameters available from existing soil survey data. To accomplish those goals, numerical simulation based on each soil series was done to model water flow within soil profiles, which were then regionally integrated to produce the geographical distribution of the relative potentials of groundwater load. Vulnerability indices deduced from the simulation results were then compared with monitored groundwater nitrate data to validate the proposed evaluation approach.

Materials and Methods

Study Site Jeju Island, the largest (73 from east to west 41 from north to south) and southernmost island in Korea was chosen as an ideal study site, because the water resource of the region depends predominantly on groundwater. Perennial surface water resources can hardly exist due to the highly water permeating nature of thesoils (Song, 1989). The volcanic ash soils are derived mainly from basalt, and partly from trachyte or trachybasalt (Song, 1989). The soils show relatively wide variation in their soil profile developments and physicochemical properties (ASI, 1976; Song, 1982). At present, 63 different soil series are identified and more than 50% of them are classified into Andisols according to the ICOMAND (International Committee on the Classification of Adisols for Soil Taxonomy, 1988).

Simulation The semi process-based approach deduced relative vulnerability indices for NO_3^- contamination via simulation of water transport based on each soil series (Fig. 1). To do this, several input parameters for water transport model had to be evaluated for each soil layer (183 soil layers total in the case of JejuIsland).

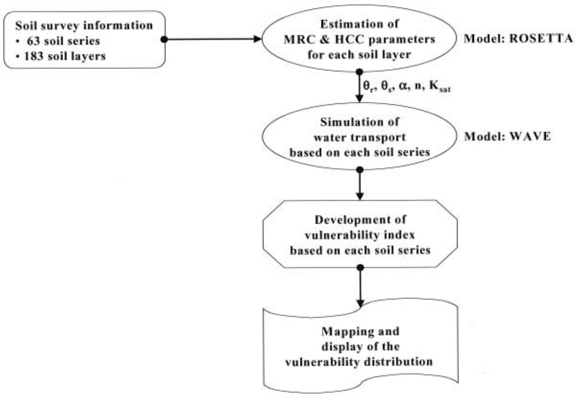


Fig. 1. Schematic of the proposed semi process-based approach for ground susceptibility.

These essential parameters, associated with MRC (moisture retention characteristic function) and HCC (hydraulic conductivity function), were evaluated using a pedo-transfer function ROSETTA (US Salinity Laboratory, USDA-ARS, 1999). ROSETTA was used to estimate unsaturated hydraulic properties from existing information and soil survey results from this study.

The evaluated MRC and HCC parameters for each soil layer were then input to a 1-D water transport model WAVE (Water and Agrochemicals in soil, crop and Vadose Environment, Institute for Land and Water Management, Belgium in 1994 Vanclooster et al., 1994).

The soil field water balance was be defined as

$$\Delta W = (P+I+U) - (R+E+D) \qquad (1)$$

where W stands for change in water content in the soil volume (mm), P is precipitation (mm), I is irrigation depth applied (mm), U is upward capillary flow in to the soil profile (mm), R is water depth lost by runoff (mm), E is actual evapotranspiration (mm), and D is percolation or drainage depth (mm). Generally, P and I are known system input, while U, R, E, D are unknown terms of the water balance. In order to quantify the unknown terms, the soil water flow equation Richards equation (Jury et al., 1991) has to be solved:

$$C(h)\frac{\partial h}{\partial \tau} = \frac{\partial}{\partial z}\left[K(\theta)\left(\frac{\partial h}{\partial z} + 1\right)\right] \qquad (2)$$

where C(h) is the differential water capacity, q is the volumetric water content ($m^3\ m^{-3}$), z is the vertical coordinate (cm) defined as positive upward, t is the time (day), $K(\theta)$ is the hydraulic conductivity function (cm/day), and h is the soil water pressure head (cm). Equation 2 is applicable for both unsaturated and saturated flow conditions. In the first case this equation is parabolic, whereas in the second case it reduces to an elliptic differential equation.

To solve this flow equation, the moisture retention (MRC $\theta(h)$) and hydraulic conductivity (HCC = $K(\theta)$ or K(h)) functions need to be specified. MRC was described by the power function model of van Genuchten (van Genuchten, 1980):

$$\theta(h) = \theta_r + \frac{\theta_s + \theta_r}{[1+(\alpha h)^n)^m]} \qquad (3)$$

where θ_s is the saturated volumetric soil water content, θ_r is the residual volumetric soil water content, α is the inverse of the air entry value (m^{-1}), and n, m are empirical shape parameters. The HCC function was represented by the theoretical hydraulic conductivity function of Mualem (1976) with the restriction of m = 1-1/n:

$$K(S_e) = K_{sat} \cdot S_e^{\lambda}\left[1-(1-S_e^{\frac{2}{m}})^m\right]^2 \qquad (4)$$

where S_e is degree of saturation (or reduced water content).

Boundary and initial conditions The upper boundary conditions about potential evaporation and precipitation were obtained from the Jeju and Seoguipo weather stations during 1998. Because WAVE can account maximally 50 mm precipitation for one day, precipitations over 50 mm were reduced to that value. And, the potential evapotranpiration rates were calculated by multiplying a factor 0.7 to the amount of water evaporation. To solve the water flow equation for n nodes of a soil profile, the flow at the bottom boundary needs to be quantified. Among the seven available bottom conditions in WAVE, the free drainage bottom condition was used in this study. When free drainage occurs, the flux through the bottom of the soil profile is always negative (downward) and equal to the hydraulic conductivity of the bottom compartment. According to this assumption, the pressure head at the bottom of the soil profile is constant with depth and the flow of water is only controlled by gravity. This assumption is valid for conditions of a deep groundwater table. In this case, a flux condition exists at the bottom of the soil profile. The initial moisture condition of each soil profile was determined through pre 1-year simulation at 33 kPa initial condition (presumed field capacity) for whole compartments. The final water profile on the last day of the pre simulation was used as the initial condition for main simulation.

Soil survey and sample analysis To obtain more detail information about Cheju soil properties and to get missed data in the existing detailed soil survey map (ASI, 1976) of Cheju Island, soils from 100 points across the island were allocated in 1999 and analyzed for their surface and subsurface physicochemcial properties. Soil texture was determined by pipet the method (Day, 1965; Green, 1981); organic matter content by rapid dichromate oxidation technique (Walkley, 1947); CEC by ammonium acetate (pH 7) displacement after washing method; pH in water using a glass electrode-calomel electorde pH meter (1:5 ratio); Available phosphorus by Mo-ascorbate colorization for phosphorus soluble in dilute acid-fluoride (Bray and Kurtz, 1945; Olsen and Sommer, 1982). Concentration of major exchangeable cations were measured by ammonium acetate method using an atomic absorption spectrometer (Shimadzu, AA-6051F). Phosphorus adsorption coefficent was determined by vanadomolybdate colorization after batch adsorption with 2.5% $(NH_4)_2HPO_4$ according to the standard recommendation method of ASI (1988).

Geographical visualization Using the simulation results, several appropriate vulnerability indices was examined. Finally, a vulnerability distribution map was produced from those relative vulnerability indices using ArcView®(ESRI, Redlands, CA, USA). The base raster map of soil series distribution was provided by KRIHS

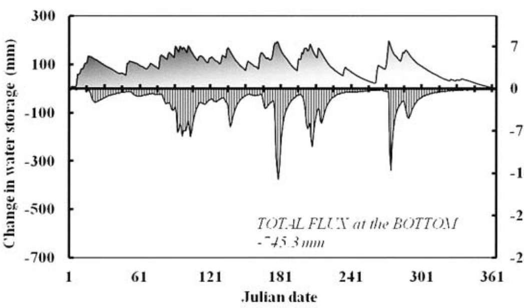
(Korea Research Institute for Human Settlements).

Statistical analysis The vulnerability evaluation results were compared to the statistical analysis of 2020 monitored data from 99 wells in 12 subregions of Jeju Island during 1994 to 1997 by the JIHE (Jeju Institute of Health and Environment). To estimate probabilities of contamination, the frequency with which threshold concentrations of nitrate are exceeded in groups of groundwater measurements was calculated. Three thresholds in mg L^{-1} nitrate as nitrogen, was chosen: 3, 5, and 10 mg L^{-1}. These exceeding probabilities were evaluated based on the 12 subregions.

Results and Discussion

Downward water flux The total amount of downward flux through bottom boundary was calculated along with daily change of water storage in the profile. Typical annual variations in downward flux are shown in Fig. 2. In case of Daejeong series (Fig. 2a), total amount of bottom flux calculated was 745.3 mm, approximately half of the total annual precipitation. During early period of the year, the water supply from precipitation was mainly used to raise the water storage within the profile without net downward flux beneath the bottom layer. Achieving enough water storage to drive downward flux, significant amount of water migrates through the bottom layer. Occasional precipitation stimulates this process and leads some reduction of water storage. Soil evaporation explains the rest parts of water storage reduction. Generally, it can be said that the more water storage occurs, the more downward flux is made.

(a) DAEJEONG



(b) DAEHEUL

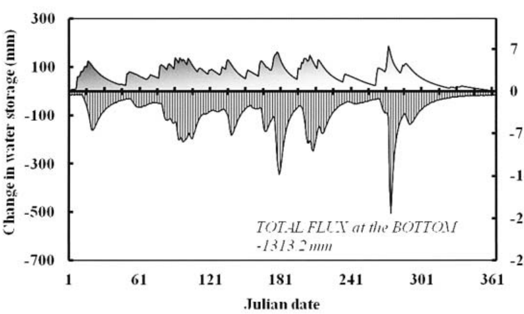
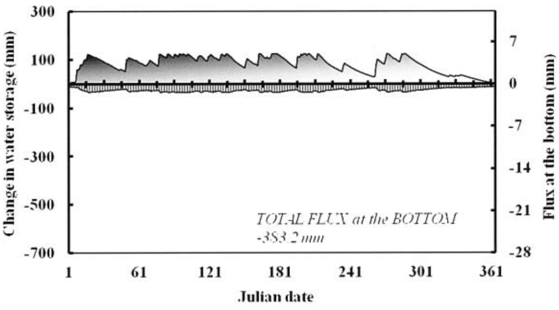


Fig. 2. Typical annual variations in downward bottom flux and water storage change in the soil profile: (a) Daejeong series and (b) Daeheul series.

On the other hand, the amount of total annual bottom flux of Daeheul series (Fig. 2b) was approximately 1.7 times higher than that of Daejeong series. This large difference can be explained by their differences in hydraulic properties and water storage capacity. Although the complex and inter-related process cannot be explained by a simple manner, Daeheul series had relatively high water conducting properties: higher n (of MRC) and saturated hydraulic conductivities, smaller air entry value, and more soil layers with such well conducting properties. In this point of view, if it is assumed that an equal amount of a nitrate source was discharged, and that the transformation kinetics is equal for these two soil series, the Daeheul series will exert more nitrate loading upon groundwater. Generally, it can be said that the more downward bottom flux exists, the higher the susceptibility of that soil series will be.

Soil water storage As can be seen from Fig. 3a, Ido series produced very small bottom flux throughout the simulation period: only 383.2 mm per year. During the whole simulation period, most of precipitation was stored within the soil profile and only small quantities of bottom fluxes were made. However, the trend of bottom flux seems relatively constant in contrast to the water storage change within a soil profile. The last horizon of Ido series has a very low hydraulic conductivity, thus mainly controlled water transport. In addition, the relatively large water storing capacity of the profile continuously supplied enough water to the least permeable layer even in the long dry period.

However, the leaching pattern and water storage pattern of Jocheon series (Fig. 3b) were quite different from those of Ido series (Fig. 3a). The downward bottom fluxes were produced very rapidly and very dramatically according to the precipitation events. In the period of dry days, the water storage within a soil profile returned rapidly to the initial dry condition. Variation of water storage was also small. This can be easily explained by the high leaching potential as well as relatively small water storage capacity (i.e., shallow soil depth) of the soil series. Leaching pattern like this implies more frequent leaching fluxes and larger amount of downward bottom fluxes.



(b) JOCHEON

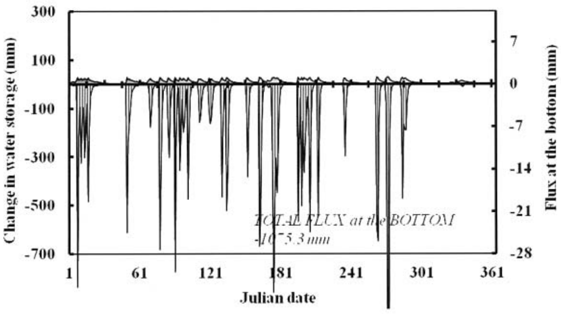
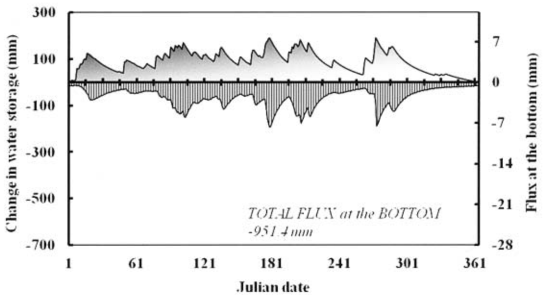


Fig. 3. Comparison between soil water storage capacity and leaching potential: (a) Ido series and (b) Jocheon series.

Leaching potential Figure 4 compares the relative contributions of downward flux and soil storage change to susceptibility evaluation. Yongdang series (Fig. 4a) produced larger amount of downward bottom flux compared to Gujwa series (Fig. 4b). However, the leaching pattern and water storage pattern of the Gujwa series imply that it is more vulnerable to contamination than the Yongdang series. Although the total amount of bottom flux was relatively smaller than that of the Yongdang series, if a contamination source is introduced at any time of simulation, the Gujwa series will transport that contaminant very rapidly downward to groundwater. In other words, the contaminant introduced in the Yongdang series can have more chances to be retarded in the soil profile. Such retardation can include simple dilution into large soil water body, detoxification (in case of nitrate, denitrification and immobilization by microorganism or higher plants). However, in case of Gujwa series, those attenuation processes have less chance to occur because of relatively short residence time in the soil profile.

(a) YONGDANG



(b) GUJWA

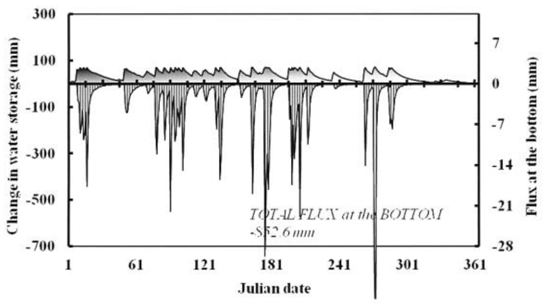


Fig. 4. Response of different soil series to precipitation events across the year: (a) Yongdang series and (b) Gujwa series.

Susceptibility Index Therefore, simple cumulative downward bottom flux would not be a reasonable susceptibility index to nitrate contamination. Rather, additional information on the leaching pattern and the change of water storage within the soil profile need to be considered. In Table 1, some simulation results for the 63 soil series in Jeju province are listed. The results include the annual total amount of downward bottom flux, the average change of the soil water storage, and the average water storage within a soil profile. In addition to the amount of bottom flux, the average change of the soil water storage can reveal some aspect of water holding and storage capacities of a soil series. Therefore the ratio of the two terms can be used for evaluation of relative susceptibilities (Index Ⅰ in Table 1).

The Index Ⅰ can differentiate reasonably the relative susceptibilities in the case of Yongdang and Gujwa series (Fig. 4). If a less water storage change can bring about a similar amount of bottom flux, the Index Ⅰ can produce a higher susceptibility number. However, this index cannot differentiate water storage change produced by soil series with thick and shallow soil depth. Another choice is using the ratio of the amount of bottom flux to the average water storage amount maintained throughout the simulation period. This is the Index Ⅱ in Table 1. This index can overcome the defect of the Index Ⅰ. However, the variation of this index for the whole 63 soil series was not enough to differentiate the relative susceptibilities from each other. Therefore, a new index

Table 1. Typical simulation results and evaluated susceptibility indices.

Soil series code	Average soil water storage change (mm) : A	Average soil water storage (mm) : B	Annual total bottom flux (mm) : C	Index Ⅰ = -C/A	Index Ⅱ = -C/B	Index Ⅲ = Index Ⅰ
1026	82	518	-383	5	1	3
1052	104	565	-695	7	1	8
1011	98	442	-684	7	2	11
1001	88	346	-783	9	2	20
1056	81	418	-926	11	2	25
1062	65	510	-1003	15	2	3
1022	70	378	-1036	15	3	40
1040	66	295	-946	14	3	46
1045	67	249	-946	14	4	54
1002	62	401	-1224	20	3	61
1025	58	415	-1336	23	3	74
1004	71	292	-1313	18	4	83
1033	51	192	-960	19	5	94
1017	52	160	-884	17	6	95

(Index Ⅲ) was made by multiplying Index Ⅰ by Index Ⅱ. The Index Ⅲ was thought to be enough to consider all the intended purposes while developing the previous indices including retardation, dilution, detoxification, and attenuation. This index can overcome the mimic limitation lied in the Index Ⅱ. Also, this index was intended to convey the trend of leaching flux, and to maximize spatial resolution.

Susceptibility categories It can be said that the higher Index Ⅲ, the greater the relative pollution potential. This index was further divided into five categories: stable, low vulnerability, moderate vulnerability, high vulnerability, and very high vulnerability (Fig. 5). This classification does not have a physical meaning. Rather it is an arbitrary grouping of the relative indices to locate highly vulnerable sites more conveniently. The vulnerability categories were chosen based on a review of the Index Ⅲ distribution. Generally, the groupings of indices were made in accordance with the breakthrough points. The stable category was below 10; low vulnerability, 10 to 30; moderate vulnerability, 30 to 50; high vulnerability, 50 to 100; and very high vulnerability, over 100. The 63 soil series were classified into these five categories according to their relative indices.

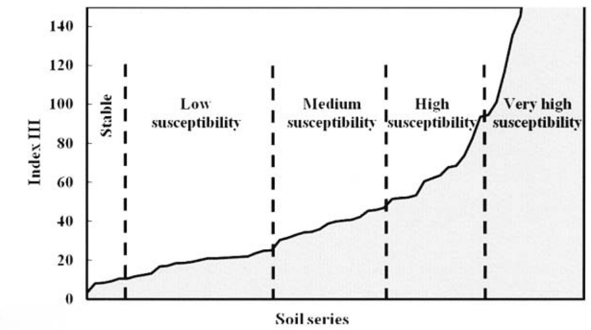


Fig. 5. Category classification of the relative susceptibility index for groundwater contamination by nitrate.

Spatial distribution of susceptibility The susceptibility distribution map of each soil series is shown in Fig. 6. Very high vulnerability areas were located primarily in the northeastern and western areas of the island: Jocheon, Gujwa, Hanrim, and Hankyeong subregions, as were some parts of eastern areas: Seongsan and Pyoseon subregions. The contribution of Gujwa, Gimyeong, Haweon, Gueom, and Jochoeon series led to this highest vulnerability. However, contribution and occupied areas of other soil series in the class Ⅴ were relatively smaller than those of the 5 main soil series.

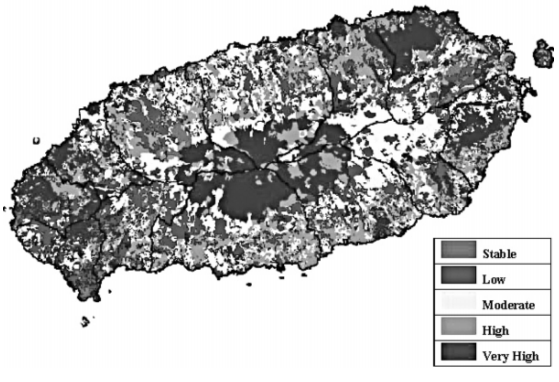


Fig. 6. Spatial distribution of the relative groundwater susceptibility to nitrate contamination.

The highly ranked lands were generally located in the same region as the very high vulnerability areas. The contribution of Ora, Jungeom, Ara, Sara, Wimi series led to the high vulnerability. However, their distributions extended to northern mid-moutaninous areas to some degree and were widely identified along the southern coastal line. Relatively high vulnerabilities were estimated along the southern coastal areas. Although any clearly identifiable area was not found, they were widely distributed along the coastal line.

The central region of Cheju Island showed very high vulnerability to nitrate contamination. Soils in that region around the main mountain Halla are coarse in texture and shallow in depth. Because of these low water holding capacities, very high vulnerabilities were estimated in these areas. The Noro, Heugag, Jeogag, Gunsan, Tosan series were located primarily in the region. The nortern-central region around Jeju city, however, showed relatively low vulnerability. The proposed process-based method was thought to give enough spatial resolution. Through the classification of vulnerability categories, it was possible to locate highly vulnerable areas easily. Interestingly, the southwestern areas had both stable and very high vulnerable soils. They were not locally confined, rather distributed randomly. It is thought that special attention for contamination is needed in that region with regard to the overall vulnerability. Although, some areas show relative resistance to contamination, nitrate transport through locally distributed vulnerable area can bring about significant contamination.

Validation with monitored data The susceptibility evaluation results were also compared to the statistical analysis of 2020 nitrate data from 99 wells monitored by JIHE from 1994 to 1997. Table 2 shows standard statics of the monitored nitrate concentration in groundwater. The average nitrate concentrations of the 12 subregions revealed that Hankyeong, Hanrim, Jocheon, and Andeog subregions were most contaminated. However, the Cheju area showed relatively low concentration. This generally coincides with the results of the vulnerability evaluation. From the Cheju area, higher concentrations of groundwater nitrate were measured to the western and eastern directions. Southern areas also showed relatively high concentrations. The highly contaminated sites also showed high maximum concentrations and large variations (standard deviation) in the monitored data. It is thought that the large variation of the nitrate concentration partly reflects the inherent susceptibility of soils situated on those areas in response to precipitation events. Yoon and Park (1994) have reported the concentration increases of groundwater nitrate from 1983 to 1993 to identify local degradation of groundwater quality during the past 10 years. In the case of Jocheon area, nitrate concentration in groundwater was 0.47 mg/L in 1983 and increased to 5.0 mg/L in 1993, approximately 10 times higher. However, in the case of Cheju area, only 1.5 times increase was observed at the same period. General coincidence was found between the evaluated susceptibility distribution and their report.

Conclusion

Susceptibiltiy assessment of groundwater contamination is a useful tool for many aspects of regional and local groundwater resources planning and management. It can be used to direct regulatory,

Table 2. Statistical analysis of the monitored groundwater nitrate concentrations.

Area	Min.	Max.	Average	Std. Dev.	No. Samples	No. Wells
Aeweol	0.1	23.2	4.76	0.32	233	10
Andeog	0.1	38.0	8.82	0.91	126	6
Cheju	ND[†]	10.8	1.86	0.11	275	16
Daejeong	0.1	39.5	6.18	0.83	82	4
Gujwa	0.5	8.50	2.16	0.11	157	6
Hangyeong	0.5	28.6	7.12	0.38	223	10
Hanrim	0.1	18.4	8.45	0.61	81	5
Jocheon	ND	23.7	8.76	0.36	261	10
Namweon	ND	35.2	4.75	0.53	166	10
Pyoseon	0.1	17.2	3.15	0.29	115	6
Seogui	0.1	25.6	4.28	0.45	191	10
Seongsan	0.2	8.70	1.61	0.14	110	6

[†] ND: Not detected.

monitoring, educational, and policy-making efforts to highly vulnerable areas where they are most needed for the protection of groundwater quality. The semi process-based methodology proposed in this study provided enough potential usage to achieve those purposes. An excellent agreement was obtained across nitrate concentrations from the highly vulnerable regions and those from the low to stable regions. It is also highlighted that the proposed approach can give sufficient spatial resolution of susceptibility distribution. The proposed methodology primarily concerns the contribution of soil media to groundwater quality. Thus, prospective studies concerning geo-hydrological contribution are highly warranted.

Acknowledgements

Appreciation is greatly expressed to Korea Research Institute for Human Settlements and Jeju Institute of Health and Environment for providing GIS raster map and monitored groundwater data.

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토양정보를 이용한 광역 지하수의 질산태 질소 오염 민감도 분포 분석

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비점오염제어에 있어서 상대적으로 지하수오염의 가능성이 높은 지역을 선별하고 주된 오염원을 파악하는 것은 그 기여도가 대단히 크다. 이 연구는 제주도를 연구지역으로 하였으며, 현재 가장 중요한 오염물질 중의 하나인 질산태 질소를 대상오염물질로 하였다. 기존 및 실제 조사에 의한 토양정보를 토대로 pedotransfer function과 수분이동 모델을 이용하여 토양통별로 산출된 질산태질소오염 민감도 지수를 통해 민감도분포도를 작성하였으며(semi process-based 법), 이를 실제 모니터링된 2,020개의 수질자료와 비교하였고 제안된 semi process-based 법은 기존의 전문가의 의견과 판단에 의존하거나 방대한 파라메터를 요구하는 기존방법들의 단점을 극복하고 충분한 지형적 해상력을 제공하였을 뿐만 아니라, 알려진 질산태 질소 모니터링 자료와도 대부분 잘 부합하였다. 본 연구를 통해 얻어진 결과들은 오염민감성이 높은 지역을 우선 선정하여 주의와 관리대책을 세우는 동시에 주요한 오염원을 파악함으로써 지하수자원을 합리적이고 효율적으로 보호하는데 있어서 유용한 수단이 될 것으로 판단되었다.