Nitrate-leaching and groundwater vulnerability mapping in North Bihar, India

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Abstract

Aquifer vulnerability assessment is crucial for studying the impact of increasing pollution load scenarios on the quantification of contaminant concentration with movement of plume for protecting groundwater resources. Only a few recent studies have focused on the performance evaluation of vulnerability assessment methods using process-based modeling approach. However, the moisture flow and pollutant transport through partially saturated zone plays a crucial role under varying hydrogeological conditions, which are generally ignored in index-based methods. Thus, the objective of this research is to evaluate the vulnerability of groundwater resources to nitrate in Samastipur, Darbhanga and Madhubani districts of Bihar State, India, using soil moisture flow and solute transport modeling. Richard's equation integrated with the classical advection dispersion equation is simulated using HYDRUS 1D by incorporating a constant head and atmospheric boundary conditions. The time taken to reach the nitrate peak concentration at groundwater table is considered to estimate vulnerability index (VI). Results have shown that high risk in terms of nitrate-leaching vulnerability in southern part of study area is dominated by Gangetic kankar in subsurface. Further, high pollution risk was reported in eastern north part of study area having alluvial deposition in subsurface. The main causes of high risk were due to the short depth of water table, little discharge and more hydraulic conductivity presence in the subsurface media. Moreover, comparatively low vulnerability was observed in area having clay capping of 2–4 m from surface. This research may help in better implementation of agricultural, soil–water conservation practices and urban/industrial infrastructure development in and around the study area.

Keywords Vadose zone · Groundwater resource · Pollution · Nitrate · Vulnerability · Clay capping

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Introduction

Pollution of natural water bodies such as lake, pond and rivers through application of agrochemicals in cultivated lands is major concern to fulfill safe drinking water demands of citizens of a country (Gupta and Sharma 2019; Gupta 2020a, b; Gupta and Yadav 2020; Gupta et al. 2020). Nitrate is one of the well-known agrochemicals used in irrigation practices, which possibly will leach into subsurface and ultimately reach underlying groundwater resources (Zhu et al. 2005; Siyal et al. 2012). High value of concentration of nitrate in drinking water may cause health hazardous, especially methemoglobinemia in infants (Spalding and Exner 1993; Fewtrell 2004).

Numerous studies have been published on pollution risk assessment using statistical/overlay/index-based modeling. Almasri and Kaluarachchi (2007) used GIS to characterize the spatial distribution of nitrogen-N causes and underlying groundwater pollution loadings. Similarly, De Paz

et al. (2009) incorporated GIS to present nitrogen-N leaching index along with farm and best management practices across a Mediterranean region. Chaudhuri et al. (2012) in their study highlighted the variability, both spatial and temporal, of nitrate in groundwater for Texas. Rahmati et al. (2015) investigated nitrate pollution from N fertilizers in Ghorveh–Dehgelan aquifer, Iran. GIS mapping was used in most of the studies related to estimation and forecasting of pollution source and load to water resources. In general, GIS maps of DRASTIC index are reported in many studies, which highlighted groundwater vulnerability of target areas (Evans and Myers 1990; Merchant 1994; Rahman 2008; Chitsazan and Akhtari, 2009; Pathak et al. 2009; Huan et al. 2012; Yin et al. 2013; Sener and Davraz 2013; Ghosh et al. 2015; Tiwari et al. 2016a, b, 2017; Neshat and Pradhan 2017; Mondal et al. 2017, 2018). In Indian context, some important studies by Tiwari et al. (2016a, b, 2017) have integrated GIS to assess groundwater quality and DRASTIC modeling, especially in mining areas. Most of these studies are performed using oversimplified methods without considering realistic site-specific conditions.

Pollution risk assessment using modeling approaches is an effective research framework for understanding the biogeochemical mechanisms of nitrate-N movement in subsurface and predicting its load through subsurface leaching on underlying groundwater resources (Enfield et al. 1982; Steenhuis et al. 1987; Dean et al. 1989; Gelhar et al. 1992; Kazakis and Voudouris 2015; Machiwal et al. 2018). Yadav and Junaid (2013) mapped groundwater vulnerability by utilizing soil water flow and modeling of solute transport at different locations in Doon valley. Kessavalou et al. (1996) investigated nitrate-N movement under irrigated corn in central Nebraska. Numerical simulations are providing a more accurate method for quantitative vulnerability mapping by solving moisture flow and solute transport governing equations and by incorporating realistic conditions. North Bihar region is highly affected by nitrate-N due to shallow groundwater table and intense use in agricultural practices (Rao et al. 2017). Thus, there is an urgent need to evaluate pollution risk due to nitrate to groundwater resources.

Index-based qualitative models, like DRASTIC, have been preferred over process-based models in the past, because the latter require a large amount of field data. However, mechanistic modeling approach is in practice and is getting more attention to simulate a complex subsurface process which improves the predictive capability and forecasts more accurate pollutant loads on underlying groundwater resources. Thus, an effort has been made to model nitrate leaching through partially saturated zone and its loading on groundwater resources at 19 sites fairly located in three districts of North Bihar. This study is novel as: (1) soil moisture flow and solute transport equations have been used to simulate nitrate leaching through site-specific lithology and

by incorporating realistic boundary conditions, (2) time taken to travel nitrate concentrations to underlying water table has been used to plot vulnerability map of area. The vulnerability map developed in this study will be helpful for implementation of pollution risk reduction strategies in the study area. This research may also be helpful to manage aquifer recharge and sustainable agricultural practices for better management of soil water resources.

Study area

Three districts, namely Samastipur, Darbhanga and Madhubani of Bihar State, India (Fig. 1), was targeted for groundwater vulnerability assessment. The geographical area of Samastipur, Darbhanga, and Madhubani is about 2905, 2288.15 and 3440.64 sq. km., respectively. 1142–1290 mm average annual rainfall and evapotranspiration of 2–6 mm/day were reported in study area (Kumar et al. 2016). In summer, daily max. and min. temperature are about 36 °C and 24 °C, respectively. Lithology is dominating by sandy alluvium soil, generally in flood plains. Northern parts of the study area are in continuation with the Tarai zone of Nepal and slopping toward the river Ganga in the South. The major geomorphological drainages are Burhi Gandak, Kosi, Baya, Kamla, Kareh, Jhamwari and Balan in Samastipur and Baghmati, Kamla, Tiljuga in Darbhanga and Bhitahi Balan, Kosi Dhar, Dhaus Nadi, Kamla Balan in Madhubani. An intense application of nitrogen-N fertilizer can be observed during Rabi (October to April) and Kharif (June–October) sessions.

The shallow aquifers are unconfined, mainly dominated by sand, clay and silt, locally known as kankar, in this area. Alluvial disposition is main surface/vadose zone soil types in Madhubani District and alluvial deposition underlying by clay capping in Darbhanga District. Similarly, Samastipur District is also dominated by Gangetic alluvial soil underlying clay capping. The lithological is available up to 150 m below top surface level from borehole (CGWB 2018). Water table of the study area is represented in Fig. 2 for the different sessions, i.e., pre-monsoon, monsoon and post-monsoon for year 2016. This GIS map was prepared using depth of groundwater table of sampling sites. The pre-monsoon level generally varies from 2.7 to 15 m bgl, in which Madhubani area has very shallow aquifers. In 1998–1999, in the pre-monsoon, the groundwater was at 2.05–8 m bgl in Samastipur and very shallow, i.e., 2.05–4.71, in Madhubani area. The observed depth of water table was considered as bottom boundary of the numerical simulation domains.
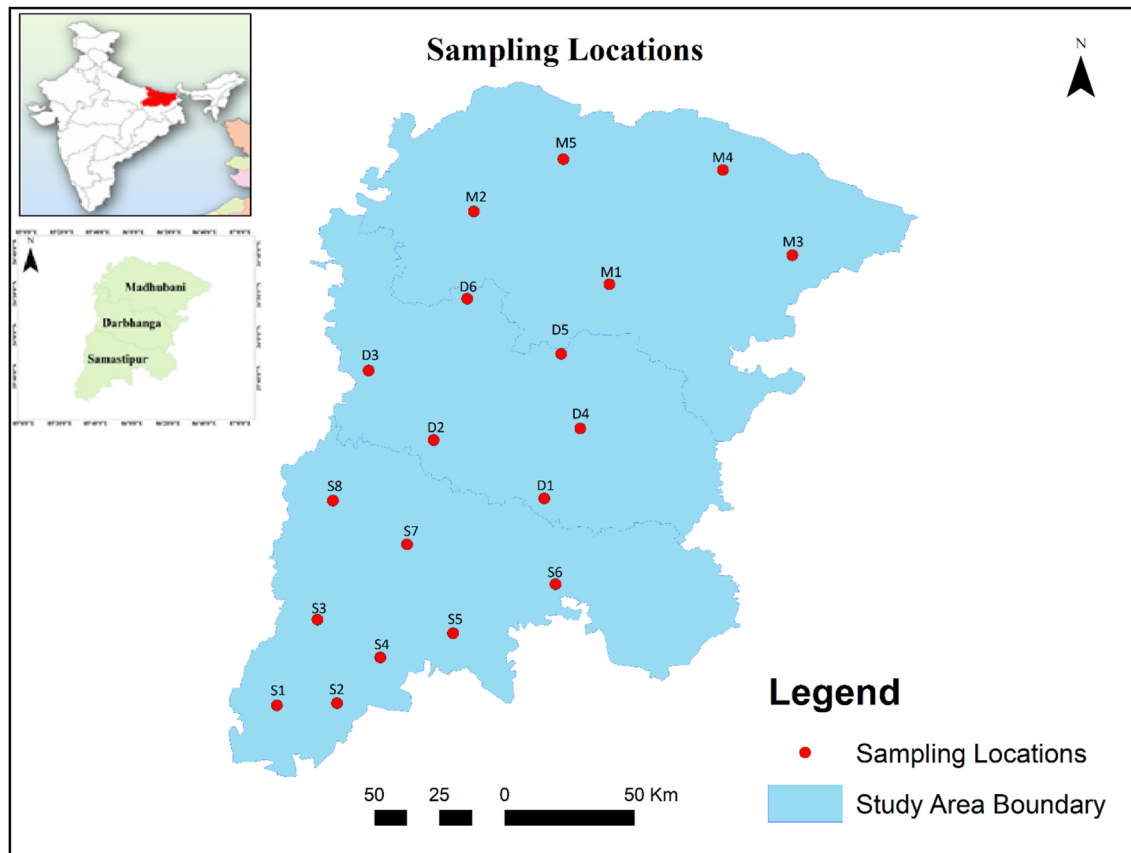
Fig. 1 The study area presenting locations of S1: Khanua; S2: Mohiuddinnagar; S3: Patori; S4: Vidhayapatinagar; S5: Dalsinghsarai; S6: Rosera; S7: Samastipur (rural); S8: Pusa; D1: Baheri; D2: Hayaghat; D3: Singhwara; D4: Benipur; D5: Manigachi; D6: Keotiranway; M1: Madhubani (rural); M2: Benipatti; M3: Phulparas; M4: Laukaha; M5: Jaynagar. S, D and M represent site location in Samastipur, Darbhanga and Madhubani districts, respectively (*Source*: Google Image; locations are interpolated using GIS techniques)

Methodology

Process-based aquifer vulnerability has been evaluated using soil moisture flow and solute transport modeling using HYDRUS 1D. Constant head and atmospheric boundary conditions were simulated for each 19 sites located in three districts, namely Samastipur, Darbhanga and Madhubani. In atmospheric boundary conditions, rainfall and evapotranspiration data were used along with land use and land pattern and lithological material distributions.

In situ observations and data collections

An effort has been made to observe in situ groundwater nitrate concentration by performing sampling at each site (as shown in Fig. 1). Groundwater samples were collected from shallow hand pumps, and its locations were recorded with a global position system. Nitrate concentrations were analyzed using spectrophotometer at a wavelength of 410 nm. Nitrate analysis method was adopted and calibrated

in spectrophotometer before samples analysis. Subsurface soil distributions were considered using lithological maps which were collected from local authorities and validated using available information of Central Ground Water Board (CGWB). Further, the water table depth data were downloaded from Water Resource Information System of India (WRIS). Similarly, rainfall and evapotranspiration rate was acquired from Indian Meteorological Department (IMD).

Modeling approach

Richard's equation (Eq. 1), which is resultant after integration of Darcy's law with the mass balance equation, is used to simulate the moisture flow:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial z}\left[K(h)\left(\frac{\partial h}{\partial z} + 1\right)\right] - S_W \tag{1}$$

where θ represents the water content [–], t shows the duration [T], h represents the water head in the soil [L], z indicates the vertical coordinate [L] and S_W is a sink term [T^{-1}].
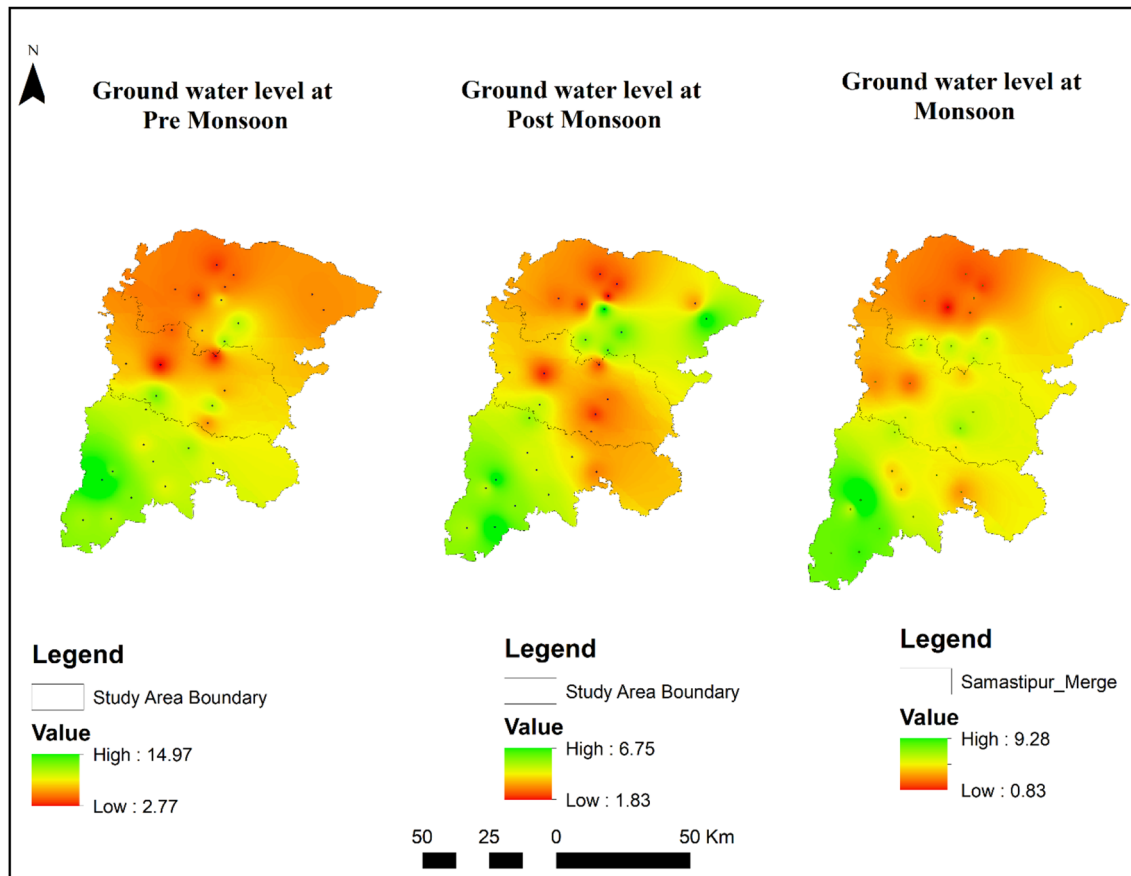
Fig. 2 Groundwater table level in the study area for the period of the pre-monsoon, monsoon and post-monsoon for year 2016 (values in meter)

$K(h)$ in Eq. (1) represents the partially saturated hydraulic conductivity in LT^{-1}.

The relationships between moisture content and hydraulic conductivity (Mualem 1976; van Genuchten 1980) are used to estimate saturated hydraulic conductivity (Eqs. 2a, 2b):

$$K(h) = K_s S_e^l \left[1 - \left(1 - S_e^{1/m} \right)^m \right]^2 \tag{2a}$$

$$S_e = \frac{\theta(h) - \theta_r}{\theta_s - \theta_r} = \left[1 + |\alpha h|^n \right]^{-m} \tag{2b}$$

where K_s represents the saturated hydraulic conductivity measured in LT^{-1}, S_e which is unitless is termed as effective water saturation ($0 \leq S_e \leq 1$), θ_s and θ_r (–) indicate saturated

and residual water content, respectively, whereas parameters α (L^{-1}), m (–) and n (–) are obtained by curve fitting which is reliant on type of soil parameter $m = (1 - 1/n)$ and l designates the tortuosity coefficient which is estimated to have a value of 0.5 after analyzing different soils (Mualem 1976). Likewise, to simulate nitrate-N movement in subsurface, the advection–dispersion equation (Eq. 3) is used:

$$\frac{\partial \theta C}{\partial t} = \frac{\partial}{\partial x_i} \left(\theta D_{ij} \frac{\partial C}{\partial x_j} \right) - \frac{\partial v_i C}{\partial x_i} - S_c \tag{3}$$

where C denotes the nitrate-N con. in $M\ L^{-3}$, v shows the water velocity measured in L/T, D_{ij} displays the coefficient hydrodynamic dispersion measured in L^2/T ($i, j = 1, 2, 3$) and S_c represents the sink term in $ML^{-3}\ T^{-1}$. Simulation mesh

Table 1 Input constraints in simulation of soil water flow and solute transport

Types of soil	Bulk density (g/cm³)	Dispersivity (cm)	θ_s	θ_r	α	n	Ks (cm/day)
Alluvial sandy	1.54	2	0.06	0.43	0.142	2.6	685.2
Loamy	1.40	10	0.12	0.38	0.055	1.4	35.4
Clayey	1.28	50	0.087	0.40	0.320	1.3	12.5

was developed using HYDRUS 1D (Šejna and Šimůnek 2007) incorporating site-specific lithology data. The Galerkin finite elements method and Crank–Nicholson iterative were used to simulate process by considering model input constraints as listed in Table 1. Initial and boundary conditions were selected as in the simulation study by Kumari et al. (2019). Influx at a specific site was retained constant, and influx at different sites was as per site-specific conditions, while daily rainfall along with evapotranspiration was used to top boundary in order to characterize the authentic flow of soil at the top surface. A constant flux (500 m^3/area) of nitrate-N mixed with irrigation water of 30 mg/L was taken at the top boundary. Details of GIS and map preparations can be found from Kumari et al. (2019). Figure 3 presents the boundary condition and material distributions used to simulate nitrate movement at each site.

Results and discussion

Soil water flow and nitrate leaching have been modeled numerically to evaluate aquifer vulnerability for 19 target sites located in three districts, namely Samastipur, Darbhanga and Madhubani of North Bihar, India. Land use and land cover analysis has been discussed first followed by nitrate concentration distribution in vadose zone. Nitrate leaching on water table at different target sites under constant and atmospheric head has been highlighted to discuss the effect of boundary conditions. Finally, vulnerability map is prepared based on time taken by solute to reach groundwater table and discussed in the last subsection.

Land use and land cover (LULC) analysis

Map for analyzing the LULC was obtained from supervised classification of the Landsat OLI satellite data (11 January, 2018) interpreted through FCC (false color composite) having band combination of 5–4–3. The signature of land cover classes was obtained by taking help from high-resolution Google Earth Data and visual interpretation of the FCC. The Landsat 8 OLI data were taken from USGS Earth Explorer (www.earthexplorer.com). The obtained map was classified in five types of LULC and shown in Fig. 4. The calculated LULC area is listed in Table 2. The LULC map shows that the Samastipur District is an agricultural-dominated area of 1711.46 km^2, while only 96.72 km^2 is covered with urban buildups, whereas Madhubani District is dominated by trees/forest followed by agricultural lands. Darbhanga District was also dominated by agricultural land and has maximum urban buildup among all districts.

Nitrate profile in vadose zone

Due to extensive use of fertilizers (mostly ammonium nitrate) in the study areas, nitrate leaching is predominant in the study area. Figure 5 illustrates that the nitrate-N concentration is customarily great near the top soils (0–0.5 m depth) due to incorporation of 100 mg/L NO_3-N concentration as upper surface flux. NO_3-N concentration sharply reduced to 50–70 mg/L at a depth of about 2–4 m from surface. The simulated nitrate concentration in the vadose zone for the observation sites (mentioned in Fig. 1) is depicted in Fig. 5. Mostly, near surface, reduction of nitrate is reported in area (S2–S7) having clay capping at a depth of 2–4 m. The nitrate-N progressively amplified in the deeper region of the soil–water system and extended to a background values after reaching a minimum at the groundwater table. The key cause for high nitrate concentration with depth was due to relatively very shallow aquifer, kankar and alluvial types matrix and high soil moisture level due to high groundwater fluctuations. Khanua (S1) and Mohiuddinnagar (S2) belonged to Gangetic kankar region provide more interconnected pathway to leach nitrate to underlying groundwater resources. Vertical nitrate concentrations in Madhubani

Fig. 3 Modeling approach used to simulate soil moisture and nitrate movement in partially saturated zone to evaluate aquifer vulnerability
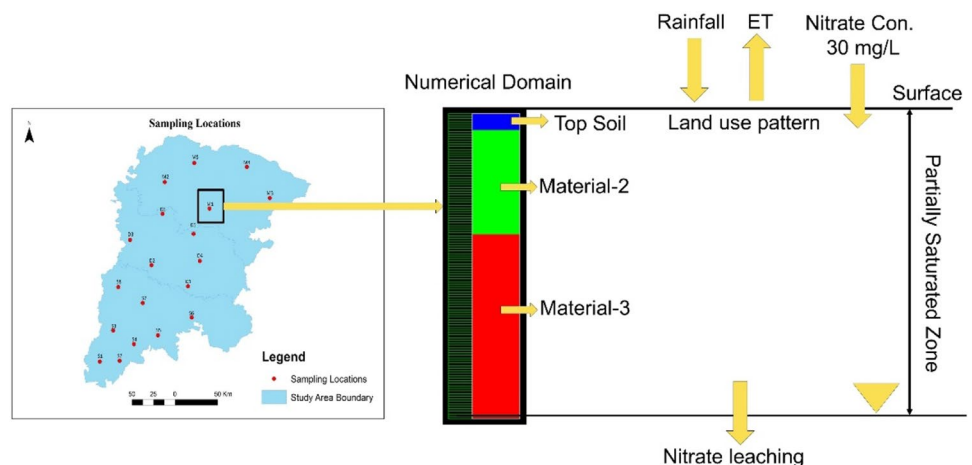
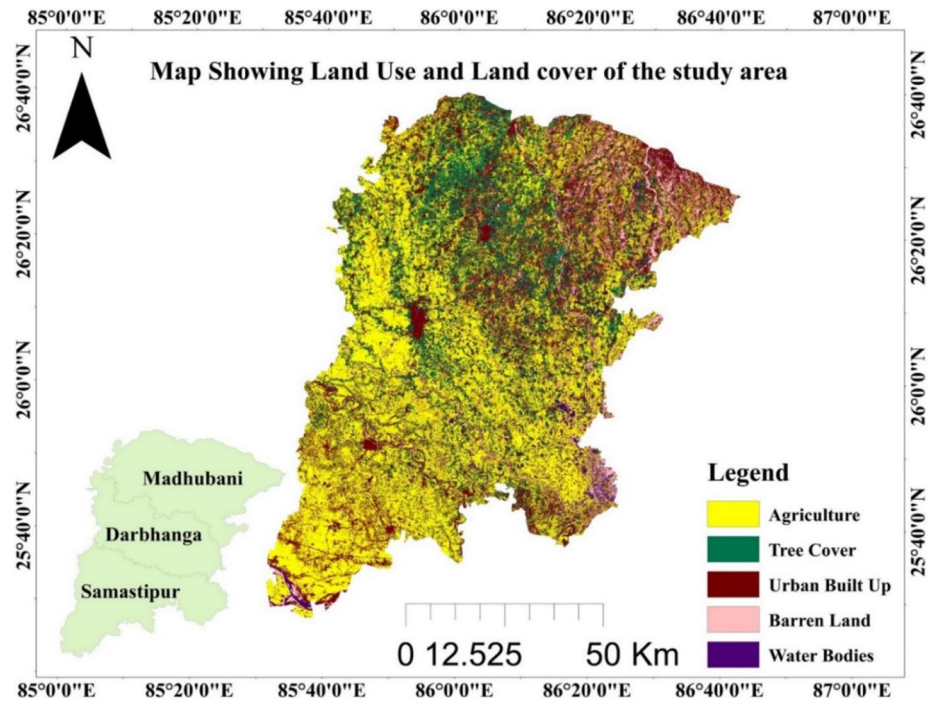**Fig. 4** Land use and land cover analysis for the study area

Table 2 Classification of LULC of study area

LULC type	Samastipur area (sq.km)	Darbhanga area (sq.km)	Madhubani area (sq.km)
Agriculture	1711.46	1169.96	982.59
Barren land	238.74	76.78	857.87
Tree cover	577.56	451.84	1047.03
Urban built-up	96.72	421.07	301.43
Waterbodies	279.92	168.49	251.72

District have also shown similar trends of concentration profile due to relatively similar lithological (alluvial) deposition in subsurface. Likewise, location D3, i.e., Singhwara District, also shows similar vertical nitrate concentration to Madhubani District due to Kamala Balan deposition of sandy lithologs. In Manigachi area of Darbhanga District, sharp reduced concentration was observed due to clay capping at a depth of 2–4 m from surface. Nitrate concentration at bottom observation node of simulation domains was well matched to in situ observed concentrations. Observed nitrate concentration validates the simulated concentrations and has shown good accuracy in estimation of groundwater vulnerability. In few sites, the observed nitrate concentrations show high concentrations; this might be due to nitrate movement through fracture zones.

Effect of boundary conditions

Constant head and atmospheric boundary conditions were simulated to understand effects of soil–atmospheric processes on fate of contaminant concentration. Such processes enhance nitrification of urea which usually happens more rapidly in high pH soils and hydrolyzes within 4 days due to the presence of soil moisture level (Stuart et al. 2011). The measured concentrations of nitrate of all target sites are represented in Fig. 6. The groundwater load of nitrate concentrations was reported more in case of atmospheric boundary, while high nitrate concentration was observed at Warisnagar (84.36 mg/L) under constant boundary conditions due to less residual time in sandy domain. Upward movement of groundwater table reduces soil column length by improving moisture level and residual time in domain. Such conditions significantly affect the hydrolysis of urea and ultimately nitrate leaching. Thus, shallow groundwater table was one of the most influencing factors for high concentration of nitrate in atmospheric boundary condition. Furthermore, the Gangetic kankar around Mohiuddinnagar site (Fig. 1) has shown high concentrations, even in this area groundwater table located in comparatively more deep. High nitrate concentration was also might be due to agriculture-dominated surface conditions (Fig. 6). The upper location of the study area, i.e., Madhubani District, also showed very high nitrate concentrations due to alluvial lithologs and shallow groundwater table. In Balan area (around Lokaha, Lokahi, Phulparas), nitrate moves rapidly in the sandy soil, which took very
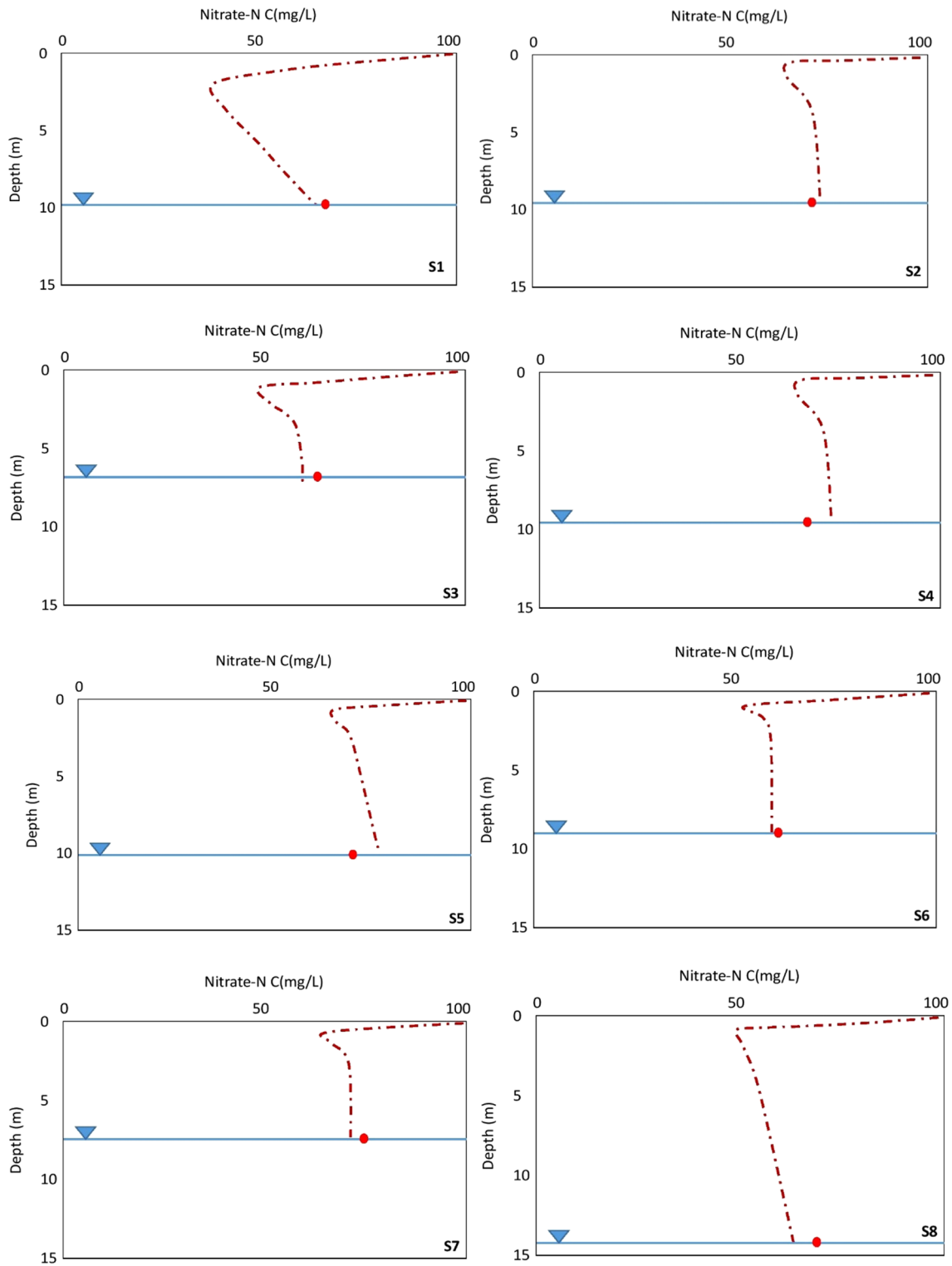
Fig. 5 Nitrate-N concentration in vadose zone simulated using HYDRUS 1D. Groundwater table locations of respective locations are represented by solid line and measured nitrate-N concentration by red dots
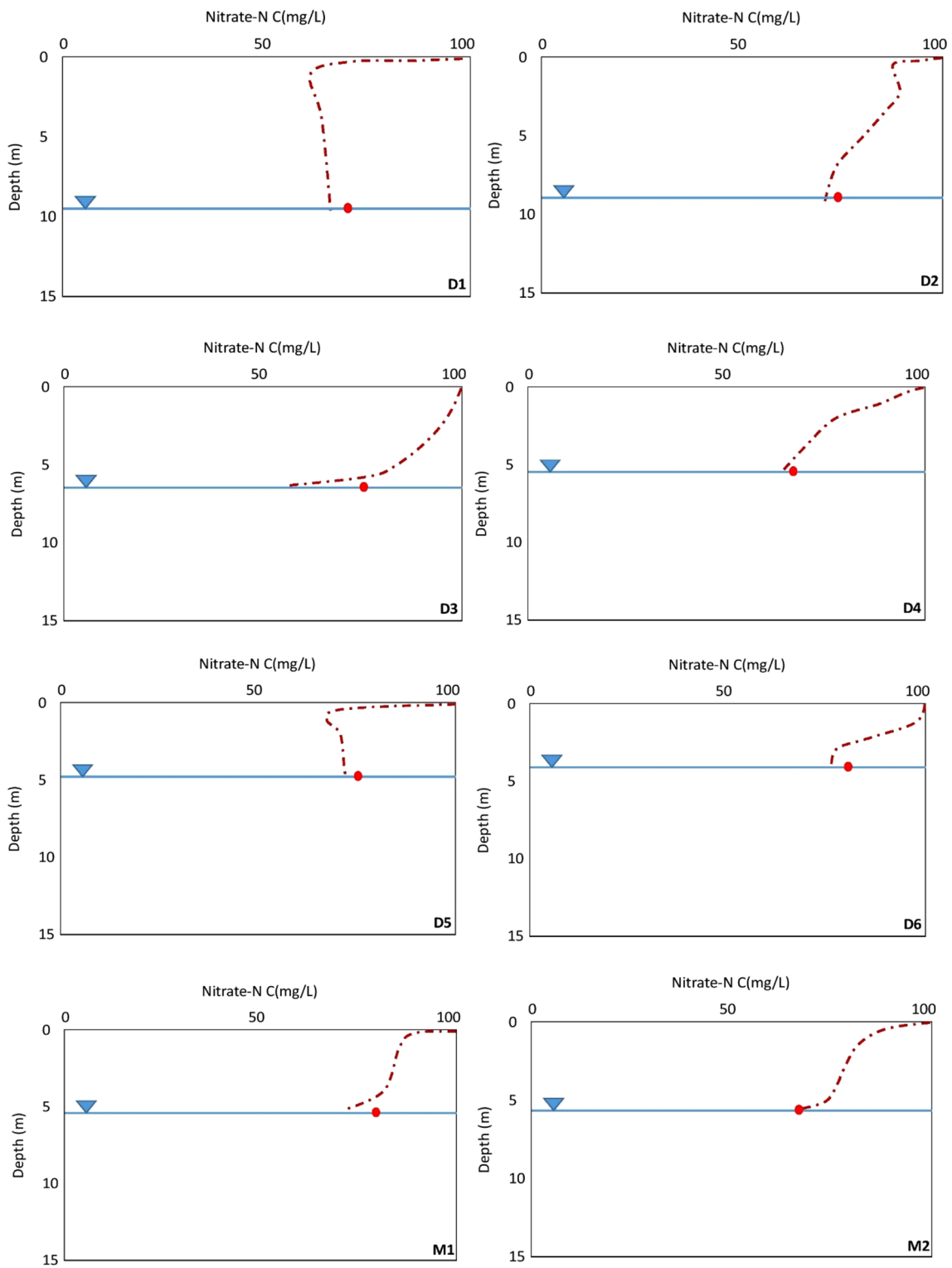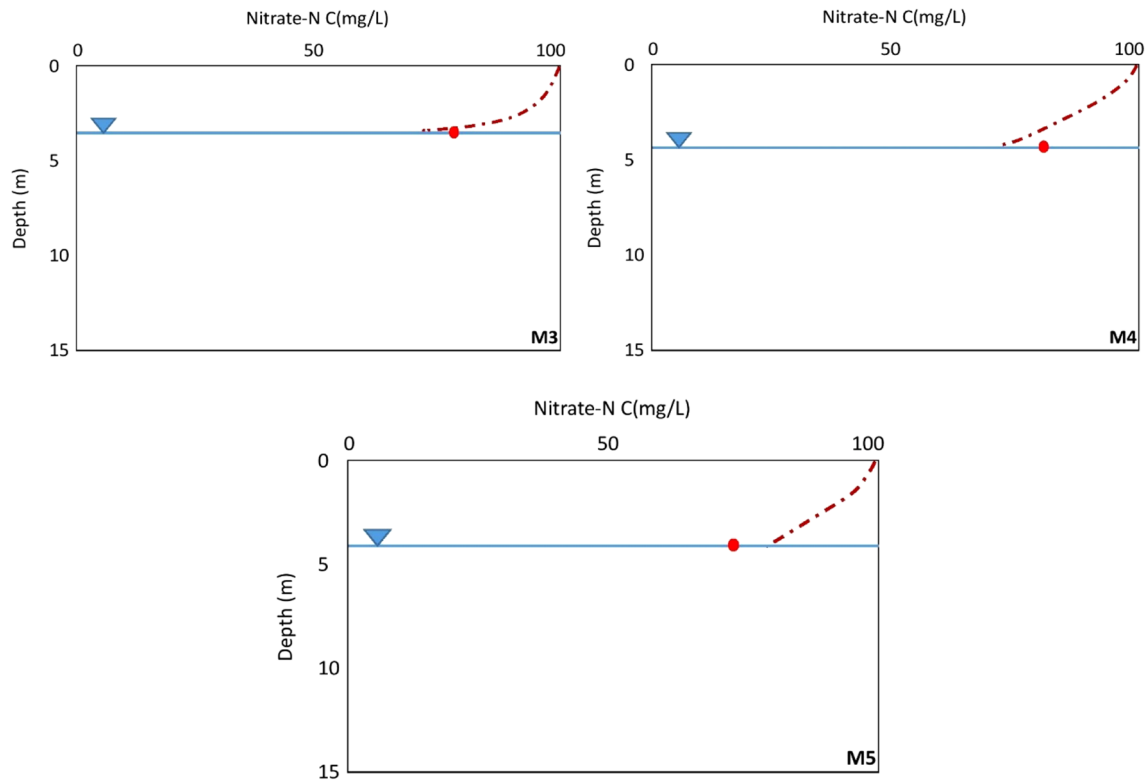
Fig. 5 (continued)

Fig. 5 (continued)

less time to reach underlying groundwater at 2.7–15 m bgl. The area having clay capping was reported more safe than sandy- and kankar-dominated lithologs. These results might improve by considering heterogeneity and fractures of subsurface.

Aquifer vulnerability map

Figure 7 represents a classified GIS map of the estimated vulnerability index for the study area. Vulnerability to groundwater pollution at different sites was estimated using the time taken for nitrate-N to reach the underlying groundwater (Yadav and Junaid 2013). Agricultural land use-dominated area and kankar lithologs near Ganga plain show very high vulnerability index. Similarly, eastern north part having alluvial soils in Balan area has also shown very high vulnerability to pollution. This was due to very shallow groundwater resource and sandy depositions. The lesser depths of water table, little discharge and more hydraulic conductivity of subsurface media was mainly contributing to the high risk of nitrate load to underlying groundwater resources. The area having clay capping below 2–4 m from surface shows comparatively low vulnerability to contaminations, especially western part of Darbhanga District. High water table fluctuation also affects the time required to reach nitrate to groundwater. Consideration of subsurface heterogeneity and fracture will improve these results effectively (Harter et al. 2005).

Conclusion

Shallow groundwater of study area is contaminated with nitrate-N concentration mostly as a consequence of nitrate-N release after irrigated agricultural fields. This study was based on the implementation of HYDRUS 1D module to simulate moisture flow and nitrate-N movement in a site-specific lithology of three districts of North Bihar.
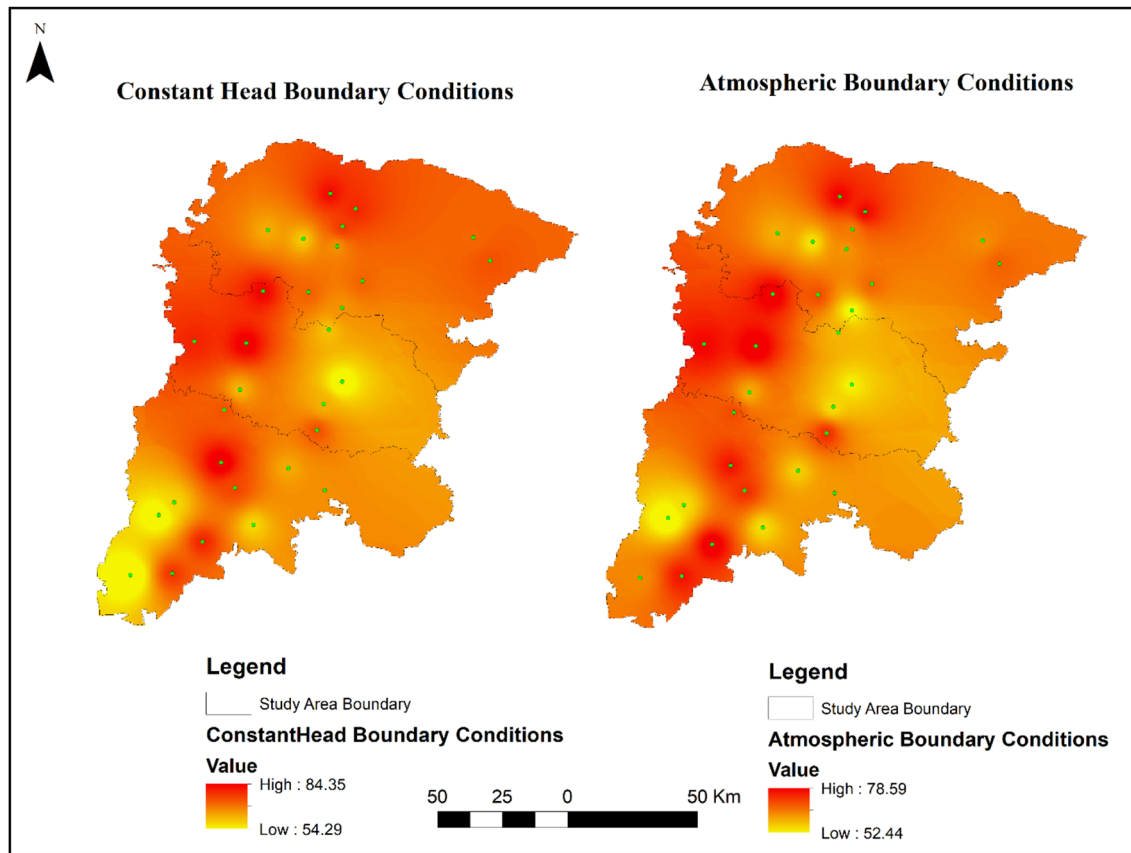
Fig. 6 Nitrate-N load to underlying groundwater under constant head and atmospheric boundary conditions (unit of nitrate concentration is mg/L)

Vulnerability map was prepared based of the time required to reach the nitrate-N to groundwater table. High vulnerability in the southern part of this study area is dominated by Gangetic kankar in subsurface litho logs. Further, high vulnerability is perceived in eastern north portion of the study area, i.e., Balan area (sandy river bed) having alluvial deposition in subsurface. A comparatively low vulnerability was observed in area having clay capping of 2–4 m from surface. Clay capping increases time of residence of nitrate-N and time to reach groundwater, which ultimately resulted in low vulnerability index. This research is limited to accurate consideration of lithological heterogeneity including consideration of fractures and thus preferential flow. One can improve results of this study by considering HYDRUS 3D modeling by incorporating dual-porosity approach. Furthermore, nitrate attenuation in vadose zone was not considered in this study, which can be considered in

Fig. 7 Classified GIS map presenting groundwater vulnerability
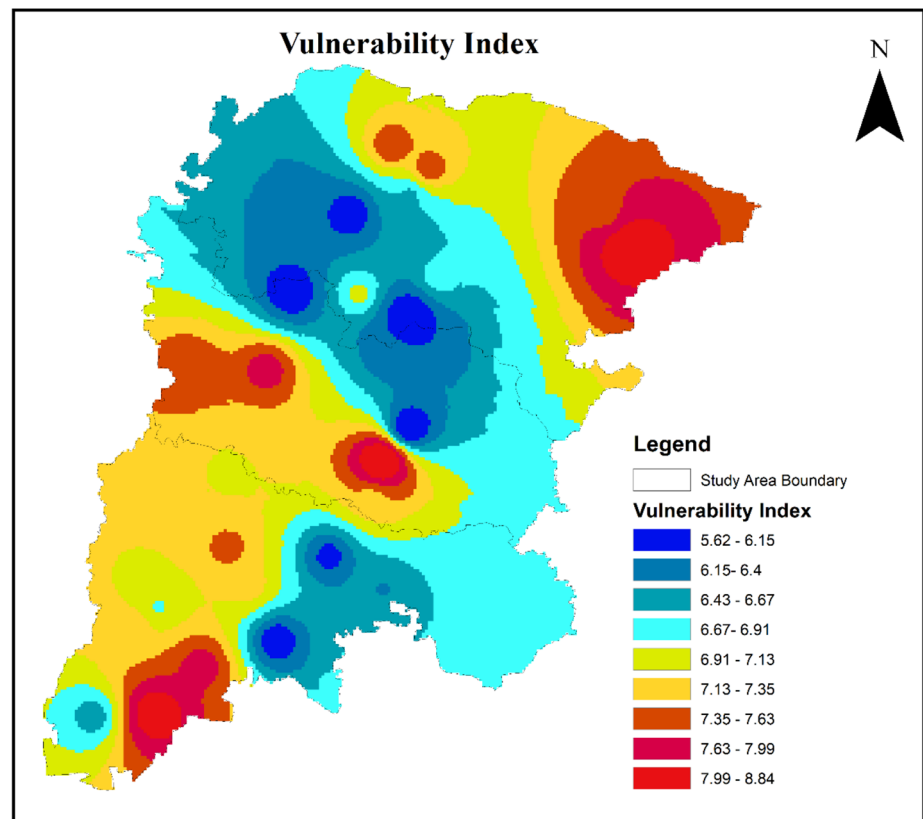
future studies. Solute injection test will be more helpful to predict nitrate movement in vadose zone and subsequently in groundwater zone. Maximum length of nitrate plume under varying groundwater flow rates is a potential topic for future research. This research may help to implement the agricultural, soil–water conservation practices and urban/industrial infrastructure development in and around the study area.

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