

Original Research Article

Groundwater contamination with nitrate and human health risk assessment of North Eastern alluvial plains of Bihar

ABSTRACT

Groundwater is natural resources with high economic value and social significance. Its supply is almost half of all drinking water in the world and plays a key role in food production. Consuming water containing high nitrate concentration have almost immediate effect on infant and could cause the risk of various diseases one of which is Methemoglobinemia in which blood lacks the ability to carry sufficient oxygen to the individual body cells. As difference in nitrate concentration in water, made it important to study the undesirable effect of nitrate. In rural areas, groundwater contamination by nitrate is a problem related to the excessive use of chemical fertilizers by local farmers and to some extent, to effluents from domestic sewage systems. Shallow groundwater plays a vital role in water use and the yield of Maize. Nitrogen application significantly affects crop uptake and utilization of water from irrigation, but little is known about groundwater use. Farmers are applying nitrogen on an average 27.8 kg/ha in Kharif maize, which is about 131.72% more than the RDN of Kharif maize i.e. 120 kg N/ha. The mean value of N application by farmers ranges from 251-323 kg/ha. The Maximum rate of N application was observed in Khagaria (323 kg N/ha) followed by Madhepura (275.08 kg N/ha) and minimum in case of Saharsa district (251.16 kg N/ha). The application rate of nitrogenous fertilizer, varying from 109.25% to 169.16% over the RDN, resulting in NO_3^- leaching. The ground and surface water from 12 villages was collected and various water quality parameters were analysed. The nitrate in groundwater varied (1.87-6.19 mg/l) and surface water (1.87-3.84 mg/l) being maximum concentration of nitrate in Madhepura district. The present study on nitrate leaching in soil, its level of contamination in groundwater and human health risk assessment by chronic daily intake of nitrate and Hazard Quotient (H.Q) values in the study area of Khagaria, Saharsa, Madhepura and Supaul has been carried out in the eastern alluvial region of Bihar.

Key Words: Maize, NO_3^- N contamination, groundwater, Hazard Quotient, HHRA

Introduction

The eastern region of Bihar is popularly known as the 'maize hub' where the maize is cultivated in two lakh hectares area. Maize (*Zeamays L.*) is an important cereal crop after wheat. The importance of maize lies in its wide industrial uses. The demand for maize is growing globally due to its multiple uses for food, feed and industry sectors. Maize is grown throughout the year due to its photo-thermo insensitive character and highest genetic yield potential among the cereals. In Bihar, Maize is grown in almost all the districts of all the three agro-climatic zones of Bihar, but Zone-II is major maize producing area that comes under North-Eastern alluvial plains of Bihar, where summer corn, paddy, winter corn and winter wheat (*Triticum aestivum L.*) are the major crops grown. Notably, Katihar boasts the highest productivity at 6510 kg/ha, succeeded by Madhepura (5285 kg/ha), Saharsa (4636 kg/ha), Araria (4272 kg/ha), Supaul (4096 kg/ha), Vaishali (4067 kg/ha), and Muzaffarpur (3935 kg/ha). This zone is renowned for its Rabimaize production. The comprehensive data underscores the escalating trends in the area, production, and producti

vity of maize in Bihar, as elucidated by Ahmad et al. in 2017. The application rates of nitrogenous fertilizer in this area by local farmers often exceed crop requirements, resulting in high accumulation of nitrate (NO_3^-) in the soil. The impact of downstream nutrient export from agricultural lands continues to be of much more concern. Nitrate-nitrogen (NO_3^-) is troublesome as it leaches through the soil into groundwater. Permeable soils make the regions susceptible to groundwater pollution by NO_3^- . Nitrate has accumulated in soils, which is highly prone to leaching, which is directly threatening the quality of groundwater. The optimal management decisions for maize production involve crucial considerations of both the rate and timing of nitrogen (N) application, as emphasized by Davies et al. in 2020. In the realm of maize production, nitrogen and water stand out as pivotal factors. In the pursuit of elevated yields, there has been a tendency to apply excessive nitrogen fertilizer (ranging from 300 to 400 kg N ha⁻¹) within the current rotation system. This surpasses the crop's actual demands, which typically range between 100 to 150 kg N ha⁻¹. The nitrate-nitrogen, once accumulated, steadily moves downward with percolating water, eventually entering the groundwater. Consequently, the excessive use of nitrogen fertilizer and flood irrigation has led to pronounced N leaching as reported by Yadav, 1997 (amounting to 15–55% of applied N fertilizer) and an augmented risk of groundwater nitrate contamination, as highlighted by Sun et al. (2018). The application of nitrogen fertilizer is a common practice to achieve high yields. In India, the annual consumption of nitrogen fertilizer is approximately 27.23 million tons. Specifically, in Bihar, the consumption of urea accounts for 18.34%, slightly exceeding the nationwide figure of 17.5% (Year End Review-2020: Ministry of Chemicals & Fertilizers). Low efficiencies of nitrogen utilization were observed (30 to 40%) in regions where high nitrogen application rates are common (Ju et al., 2009). The abundance of nitrogen fertilizer leads to nitrate accumulation in the soil (Vitousek et al., 2009), in regions with double or triple cropping systems exist (Zhou et al., 2016). This nitrate accumulation in the soil becomes problematic when heavy rain or irrigation occurs, as contaminating drinking water sources. Groundwater contamination with NO_3^- is a vital concern, especially in these regions due to the intensive maize production. The widespread use of nitrogenous fertilizer is recognized as a significant contributor to nitrate pollution in groundwater (Huang et al., 2011). Nitrate, being highly soluble in water and poorly retained by soil, poses a risk of leaching into the subsoil and eventually reaching groundwater if not taken up by plants or denitrified to N_2O and N_2 (Majumdar and Gupta, 2000). Consuming water with elevated nitrate levels can lead to various health hazards for humans. Infants are generally more susceptible to nitrate, but adults may also experience adverse effects from consuming water rich in nitrates, such as thyroid dysfunction in children and pregnant women (Gatseva and Arginova, 2008). The concentration of NO_3^- in drinking water can reach critical levels, and established safety limits are set by regulatory bodies. According to the Bureau of Indian Standards (45 mg L⁻¹) and the World Health Organization (50 mg L⁻¹), the safe limit for nitrate in drinking water is defined. Both the World Health Organization and the European Community (Council of European Communities, 1980) recommend a limit of 50 mg NO_3^- L⁻¹ (11 mg NO_3^- L⁻¹) in potable water. The US Environmental Protection Agency (USEPA, 1995) and the Canadian Water Quality Branch (Water Quality Branch, 1995) have set a limit of 44 mg NO_3^- L⁻¹ (10 mg NO_3^-

NL^{-1}) as the maximum safe level in drinking water. These standards underscore the importance of monitoring and managing nitrate levels to safeguard water quality and human health. The nitrate content in groundwater can significantly influence the nitrogen flux in the soil when used for crop irrigation. Consequently, the prevalence of high nitrate levels in shallow groundwater may be influenced by the cycle of pumping and return flows in the underground water system (Buvaneshwari et al., 2017). In the north-

eastern alluvial plains of Bihar, where groundwater serves as the primary drinking water source for a majority of the population, it is crucial to investigate the potential health risks associated with excessive intake of such water. Therefore, maintaining nitrate levels below the maximum contaminant level is essential. The overconsumption of nitrate in drinking water poses serious health risks and toxicity in humans. A well-

documented example of nitrate toxicity is Methemoglobinemia, which affects infants and pregnant women (Sajil et al., 2014). Beyond infants, adults are also susceptible to gastric cancer, respiratory problems, headaches, fatigue, thyroid gland hypertrophy and multiple sclerosis (Tao and Xin, 2014; World Health Organization (WHO), 2011). Therefore, ensuring nitrate levels in drinking water remain below established safety thresholds is critical for safeguarding public health in these regions.

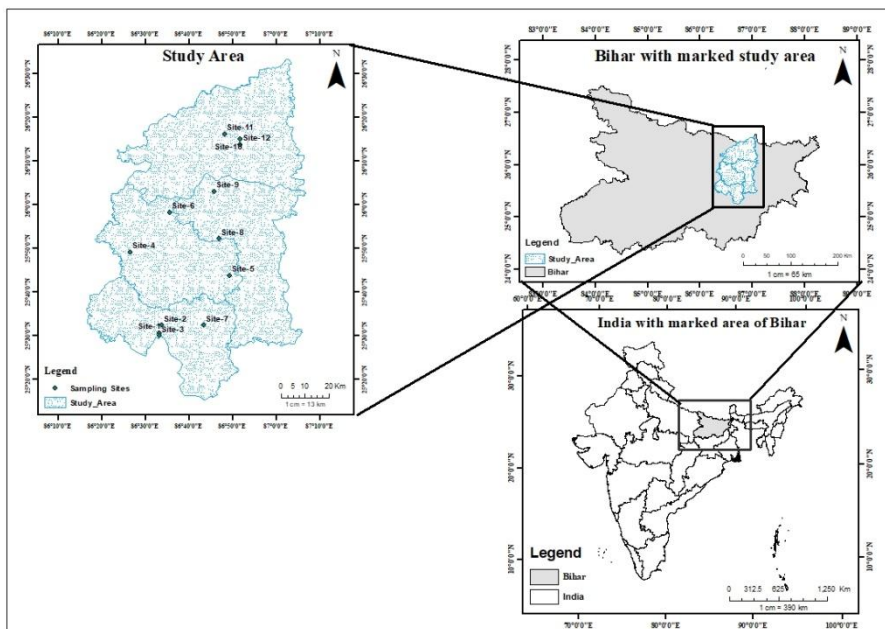
In addition to the use of fertilizer and irrigation method, crop, climatic factors like rain fall and soil properties such as soil texture, affect soil NO_3^-

accumulation and leaching (Li et al., 2016). The level of nitrate accumulation in soil has become a significant hazard to potable water since 90% of farming people of the area are frequently using this water for drinking purposes as well as for irrigation purposes also. The NO_3^- in groundwater has been enlisted as an emerging issue for groundwater safety and human health. In some areas, it has been reported significantly higher than the prescribed safe concentrations for drinking water. The study area has been surveyed and primary data has been collected through standard questionnaire developed by research team of the project and also some additional data used in this study is extracted from the literature that reported post-harvest soil NO_3^- concentrations in maize fields in North East alluvial plains of Bihar.

As per our best knowledge, no comprehensive study is yet undertaken by any previous research to explore the nitrate concentration and its possible health hazards in the NE alluvial plains, Bihar. Most of the earlier studies had identified N based fertilizers as a critical source of nitrate in groundwater. This study aimed to investigate the groundwater nitrate content in NE alluvial plains of Bihar and associated health risks in humans

beings. The load of nitrate in groundwater of this region may pose a serious threat to residents as they rely on groundwater for potable water sources. The survey was done on groundwater quality of the study region to estimate the overall concentration of nitrate in groundwater. Since a part of the state has a high human population, this area could be at risk due to nitrate contamination in local surface water and groundwater sources. Looking into these facts, the university decided to estimate the groundwater nitrate level in these areas and correlating the high nitrate content (than a safe limit as suggested by BIS) to possible human health risk using a human health risk assessment model as proposed by USEPA (1992, 1999, 2004).

Map 1



Map 2 :Details of sampling site

Materials and methods

Description of the study area

Bihar is situated in the eastern part of India in between latitudes $24^{\circ}20'10''\text{N}$ and $27^{\circ}31'15''\text{N}$ and longitudes $83^{\circ}19'50''\text{E}$ and $88^{\circ}17'40''\text{E}$. It is an entirely landlocked state, in a subtropical region of the temperate zone. Bihar lies between the humid West Bengal in the east and the subhumid Uttar Pradesh in the west, which provides a transitional position in respect of climate, economy and culture. It is bounded by Nepal in the north and by Jharkhand in the south. Geographically Bihar plain is divided into two unequal halves (North Bihar and South Bihar) by the river Ganges which flows through the middle from west to east. Bihar's land has an average elevation above sea level of 173 feet. As per agro-climatic zone it is divided into ACZ-I, II, IIIA and IIIB.

Water sampling depth and geographical location of sampling site

Among districts of Agro-climatic zone-II, four districts covering Khagaria, Saharsa, Madhepura and Supaul were selected. Total of 12 villages were selected (3 villages from each district) for water sampling. GPS based samples were collected for analysis. The groundwater levels in various districts of ACZ-II are recorded beyond 60 meter below ground level; this is due to the overexploitation. The groundwater level in Bihar has declined drastically in past few decades. According to an estimation net, the dynamic groundwater resources of the state are 29.19 BCM (Billion cubic meters), and the net groundwater left is 10.77 BCM. The geographical location of all sampling sites is presented in Table 1.

Water Sample collection, Sampling procedure and Water Analysis

The water samples were collected across 12 different sites in Seemanchal districts and neighboring Koshi river region, whose depths varied from 6 m to 18 m. The water samples were fetched from borewells located around cultivated lands. The fresh groundwater samples were collected in pre-cleaned sample bottles of 500 mL capacity. Each sample of collected bottles was tightly capped to avoid leakage and contamination during handling and transportation. The containers were adequately labelled by date, time, GPS coordinates etc. to recognize exact sampling point. All the collected samples were initially preserved in cold and transported to the laboratory where they were stored in the freezer at 4°C until used for final chemical analysis. Water quality parameters analysed in accordance to standard methods of (American Public Health Association (1998)) were pH, temperature, conductivity, total suspended solids (TSS), total dissolved solids (TDS) and nitrate (NO_3^-).

Nitrate Exposure and Human Health Risk Assessment (HHRA)

The US Environmental Protection Agency (USEPA) proposed an HHRA model for the identification of hazard and exposure. The HHRA can be estimated by calculating possible adverse impacts of given contaminant over a specific period. The HHRA is computed using the values of present concentration of a contaminant in groundwater and its exposure duration to humans. The excess intake of nitrate through drinking water can cause serious health hazards in human beings. The amount of nitrate in the human body depends on its actual concentration in water and the intensity of drinking $\text{day}^{-1} \text{kg}^{-1}$ of body weight. To estimate the health hazards of high nitrate dose in drinking water, the USEPA model was adopted which was implemented in four different steps, namely, hazard identification, dose response assessment, exposure assessment and risk characterization (Li and Qian 2011; Anornu *et al.* 2017). Some studies suggest that ingestion and dermal contact as the leading pathways of nitrate exposure

nhumans(WuandSun2016;Chenet.al.2017)butingestionseemstohaveevengreaterriskthanderma lcontact.TheexposureofnitratethroughingestionwithdrinkingwateriscalculatedbyfollowingEq.(i)(Zhouetal.2016a).

$$CDI = \frac{C \times IR \times EF \times ED}{ABW \times AET} \dots\dots\dots(i)$$

whereCDI(chronicdailyintake)istheingestiondosefromdrinkingwater(mg/kg/day),Cisth e concentrationofnitrateestimatedingroundwatersamples(mgL⁻¹),IRistheaveragedailyingestionrateofdrinkingwater(L/day)andthevaluesofIR(2L/dayforadult(male&female),0.78L/dayforchildrenand0.3L/dayforinfants)wereusedforthismodelastakenfro mpublishedliterature;EFistheexposurefrequencies(365days/year),EDistheexposeduration(st andardexposedurationinliteratureissuggested40yearsforadult(maleandfemale),12yearsforchi ldrenand<1forinfants),ABWisheaveragebodyweight(65kgformale,55kgforfemale,20kgforchil drenand8kgforinfants);andAETistheaverageexposuretime(days)whichis14,600daysformaleand femaleand4380daysforchildrenand365daysforinfants(YadavandGupta,2022).Thepresentstudy focusesonthenon- carcinogenichealthriskofnitratemainlyestimatedbythehazardquotient(HQ_{nitrate})values,whichis e stimatedthroughfollowingEq.(ii)(USEPA1999;Suetal.2017):

$$HQ \text{ nitrate} = \frac{CDI}{RfD} \dots\dots\dots(ii)$$

WhereRfDisreferencedose,RfDindicatesreferenceofNO₃⁻(1.6mg/kg/d)wereobtaine dfromthedatabaseofIntegratedRiskInformationSystem(IRIS)andUSEPA(2001).Thecalculation ofhazardquotientvalue,HQ_{nitrate}>1isreferredaspotentiallyknowntocausehealthrisksandvaluesof HQ_{nitrate}<1indicateshatitisanacceptablelimitofnon- carcinogenicriskinindividualsduetoingestionofNitratecontaminatedgroundwater.

Table 1. Parameters and their values used for HRA computation (USA EPA)

Parameter	Description	Male	Female	Children	Infants
IR	Ingestion Rate (L/day)	2	2	0.78	0.3
ED	Exposure Duration (years)	40	40	12	≤1
EF	Exposure Frequency (days/year)	365	365	365	365
ABW	Average Body Weight (kg)	65	55	20	8
AET	Average Exposure Time (days)	14600	14600	4380	365
RfD	Reference Dose (mg/kg/d)	1.6	1.6	1.6	1.6

Spatial mapping of sampling site and vulnerable hazard zones

GIS-

based interpolation technique was used to represent the spatial variation of health risk distribution of nitrate intake among adults and children across the study area. All the maps were prepared using ArcGIS software.

Result and discussion

Soil Analysis/Bulk soil nitrate-nitrogen measurements

Soil characterisation

The soil in ACZ-II is comprised of alluvial deposition of Koshi river basin occupied by clays and loam type of soil which contains kankar, clay, sand particles, gravels, pebbles, sandstone, etc. The characteristics of soil affect the infiltration, percolation, and groundwater recharge capacity of the region. Higher recharge rate as higher groundwater contamination potential from surface contaminants. The soil at Khagaria was a loamysand complex (Sandy, mixed) and received flood irrigation during the growing season. Soils at Saharsa, Madhepura and Supaul were Fine-loamy, and clay loam (Fine-loamy, mixed) respectively. The topography of the area is flat, with pH of 8.4 in the top soil and organic matter ranging from 2.18 to 3.6 g kg⁻¹ within the profile depth of 0 to 120 cm. The soil textural characteristics (Percent soil fraction) and bulk density of study area was recorded (Fig. 1, 2, 3 and 4). The soil texture and bulk density is varying depthwise in different districts. The soil textural class of Khagaria district varies from Loam (0-60 cm), Clay loam (60-75 cm), Sandy loam (75-90 cm), Loamysand (90-105 cm) and Sandy loam (105-120) under varying depth. While bulk density varies from 1.27-1.79 (g/cm³). The soil textural class of Madhepura district varies from Loam (0-30 cm), Sandy clay loam (30-90 cm), Sandy loam (90-120). The bulk density varies from 1.18-1.58 (g/cm³). The soil textural class of selected village in Saharsa district varies from Loam (0-45 cm), Sandy clay loam (45-75 cm), Sandy loam (75-120 cm). The bulk density varies from 1.25-1.52 (g/cm³). The soil textural class of selected village in Supaul district varies from Loam (0-45 cm), Sandy clay loam (45-60 cm), Sandy loam (60-120 cm). The bulk density varies from 1.18-1.50 (g/cm³). After the analysis of soil, it has led to its categorization for understanding of the study area. The major soils identified include loam to silt loam, found in plain upland; loam to loamy clay, were obtained in deep waterlogged areas; clay loam, loam to silt loam, were specifically found in mid upland to lowland regions; and sandy, sandy clay, and sandy loam, which were obtained in areas within the Kosi Embankment.

Figure(s): Depth-wise soil textural class (Percent soil fraction) of studied districts in North East Alluvial Plains of Bihar

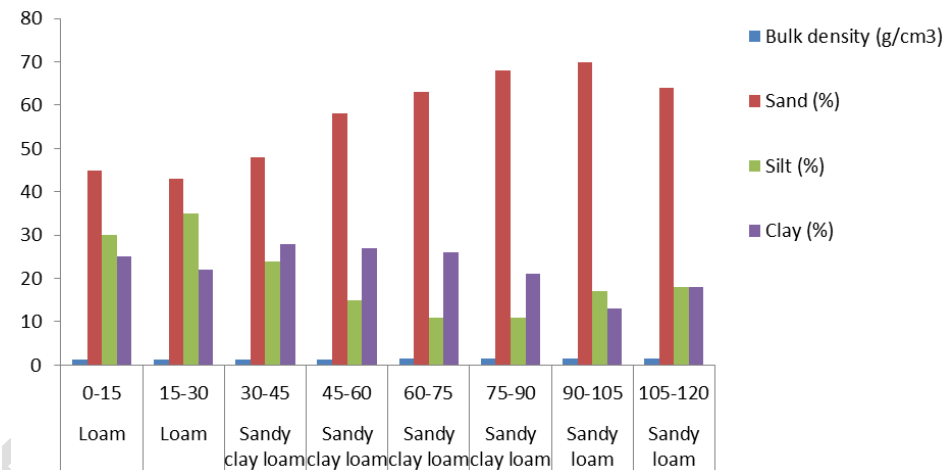
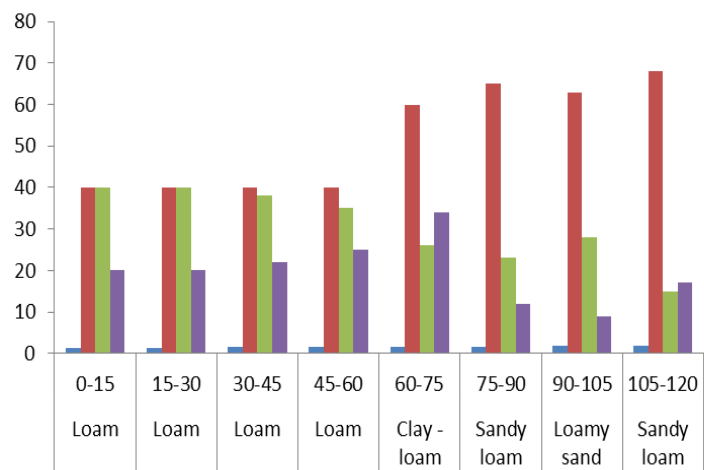


Figure1. Depth-wise soil textural class of Khagaria District

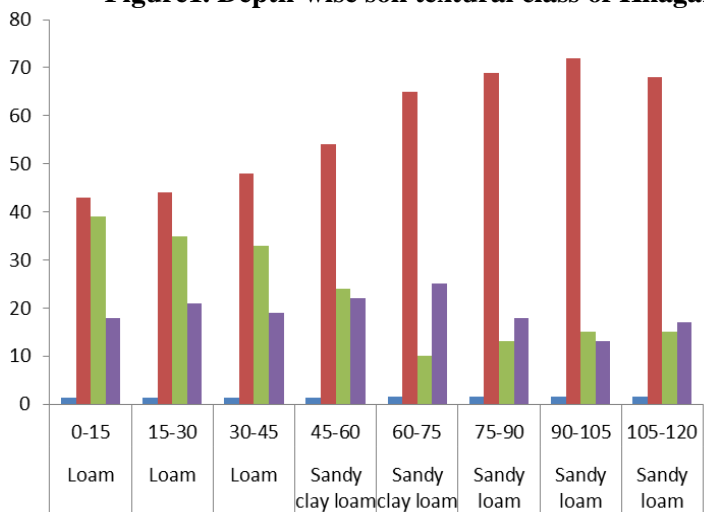


Figure2. Depth-wise soil textural class of Madhepura District

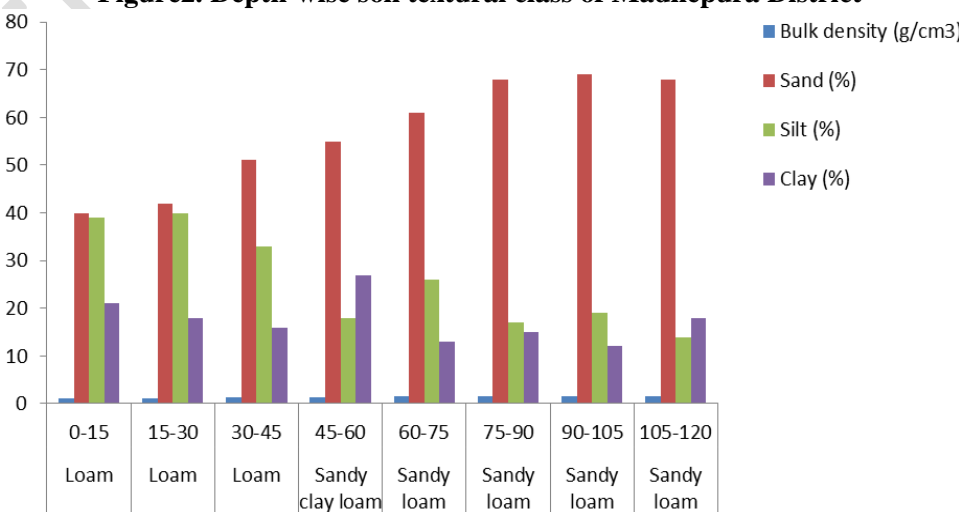


Figure3. Depth-wise soil textural class of Saharsa District

Figure4. Depth-wise soil textural class of Supaul District

7746 with an average value of 6266.25 kg/ha. The nitrogen % in stover ranges from 0.63 to 0.66% with mean value of 0.65%. The nitrogen uptake in stover varies from 35.18 to 50.42 (kg N/ha) with the mean value of 40.70 (kg N/ha). The maize stover includes stalk, leaves, cobs and husks. The total plant uptake of nitrogen ranges from 141.02 to 212.04 kg N/ha with an average value of 164.04 kg N/ha.

Table:3. Nitrogen uptake in Maize grain and Stover in studied villages (Mean of 3 villages)

Districts Name	Grain			Stover			Total Uptake (KgN/ha)
	Yield* (Kg/ha)	N (%)	N-Uptake (KgN/ha)	Yield (Kg/ha)	N (%)	N-Uptake (KgN/ha)	
Saharsha	6873	1.54	105.84	5413	0.65	35.18	141.02
Madhepura	7522	1.49	112.07	6017	0.63	37.91	149.98
Supaul	7538	1.51	113.82	5889	0.66	39.30	153.12
Khagaria	9915	1.63	161.62	7746	0.64	50.42	212.04

*weight after adjustment of 12-13% moisture in grain (adjusted grain yield)

Table:4. Mean N-fertilization; crop N-removal, Leaching of nitrate and calculated N-Surplus.

District	N-fertilization (kg/ha)	Total plant Uptake (kgN/ha)	NO ₃ N Leaching (kg/ha)	Calculated N-Surplus#
Saharsha	283.48	141.02	26.73	142.46
Madhepura	303.61	149.98	32.27	153.63
Supaul	291.63	153.12	33.48	138.51
Khagaria	343.63	212.04	42.95	131.59

#calculated as difference between N fertilization and N removal

Level of nitrate in water

The ground and surface water from both cost of cultivation scheme (CCS) and Non-CCS village has been collected and various water quality parameters were analysed. The depth of shallow groundwater varied from 20-60 feet. The nitrate level in groundwater varied (3.42-5.27 mg/l) and surface water (4.61-5.72 mg/l) being maximum concentration of nitrate in Madhepura district. The water sample has been also collected from deep depth of groundwater up-to depth of 200-400 feet depth and used as reference water sample where nitrate level varied from 0.86 to 1.05 mg/l.

Nitrate Exposure and Human Health Risk Assessment

Groundwater quality has been steadily declining in recent decades as a result of numerous pollution sources such as fertilisers and chemicals. The ingestion of contaminated groundwater can adversely affect the health of humans through various types of exposures including direct ingestion, dermal contact, washing, etc. (USEPA 2001). Spatial map of nitrate concentration was made using GIS software (ArcGIS 10.7.1) shown in fig. 2. Nitrate concentration in surface water (ranged from 4.16 mg/L to 6.78 mg/L) and groundwater samples (ranged from 3.19 mg/L to 6.14 mg/L) to 108 mg/L, has been shown in table. 8. Regular exposure to nitrate, one of the primary contaminants in groundwater reservoirs, can have a negative impact on health and increase the risk of blue baby syndrome, particularly in communities with small children. Hence health risk assessment of nitrate has been carried out. The Chronic Daily Intake (CDI) values for male, female, children, and infants ranges from 0.1280 to 0.2086; 0.1512 to 0.2465; 0.1622 to 0.2644 and 0.1560 to 0.2542, respectively for nitrate contaminated surface water (Table. 8), while these values for all four groups of people ranges from 0.0981 to 0.1889; 0.1160 to 0.2232; 0.1244 to 0.2394 and 0.1196 to 0.2300 respect

ively for nitrate contaminated groundwater (Table.9). Similarly the Hazard quotient (HQ) values for male, female, children, and infants ranges from 0.0800 to 0.1303; 0.0945 to 0.1540; 0.1014 to 0.1653 and 0.0975 to 0.1589, respectively for surface water (table.8), while 0.0613 to 0.1181; 0.0725 to 0.1395; 0.0777 to 0.1497 and 0.0740 to 0.1439, respectively for groundwater (Table.9) intake. HQ value more than 1 indicates high risk. The finding of data showed that all HQ value was less than 1 of all samples in all four groups, however the data of HQ value reaching towards unit, so it is good time to be cautious for maintain the level of nitrate contamination in the study area by adopting the certain mitigation options as suggested in the end of this manuscript.

UNDER PEER REVIEW

Table5: WaterqualityparametersofselectedsiteslocatedinACZ-IIindryseason

Districts	Villagename	Surfacewater				Groundwater				
		pH	Conductivity (μ S/cm)	TDS (mg/L)	NO ₃ ⁻ (mg/L)	SamplingDepth(ft)	pH	Conductivity (μ S/cm)	TDS (mg/L)	NO ₃ ⁻ (mg/L)
Khagaria	Khutia	7.12	652.1	434	4.98	20	7.65	782.31	138	3.19
	Saidpur	7.13	735.3	413	5.85	60	8.13	832.6	123	3.32
	Ekaniya	7.29	749.5	387	5.79	40	7.91	618.7	168	3.74
Meanvalue		7.21	712.3	411.33	5.54	-	7.89	744.54	143	3.42
Saharsa	Baghaud	7.91	589.4	417	4.19	40	7.62	612.5	132	4.87
	Bargaon	7.59	653.2	399	5.11	50	7.57	748.3	124	5.51
	Pachgachia	7.63	776.5	383	5.04	30	7.19	707.5	145	5.11
Meanvalue		7.71	673.03	399.67	4.78	-	7.46	689.43	134	5.16
Madhepura	Mathahi	7.28	753.6	453	5.18	30	7.52	978.7	224	4.79
	Sukhasan	7.62	769.7	509	6.78	25	7.73	1019.5	264	5.12
	Dularpiprahi	7.25	735.4	451	5.21	30	6.91	979.8	199	5.89
Meanvalue		7.38	752.9	471	5.72	-	7.39	992.66	229	5.27
Supaul	Hulas	6.94	717.7	319	4.73	50	7.39	1275.4	247	4.12
	Dewipur	7.93	615.3	298	4.16	60	7.74	978.8	153	6.14
	Bengaipattichampanagar	7.19	826.5	368	4.93	45	7.67	949.6	152	4.16
Meanvalue		7.35	719.83	328.33	4.61	-	7.60	1067.93	184	4.81

Table6: waterqualityparametersofreferencepointlocatedinACZ-IIindryseason

Districts	Referencepoint	Referencewatersamplingdepth(ft)	pH	Conductivity (μ S/cm)	TDS (mg/L)	NO ₃ ⁻ (mg/L)
*Khagaria	Khutia	400	7.01	750	263	1.05
*Saharsa	Kalitemple	200	7.18	732	218	0.92
*Madhepura	Singheswartemple	300	7.29	719	213	0.98
*Supaul	RamjankiMath	250	7.50	675	98	0.86

TABLE 7 Nitrate concentration in surface water and their Chronic Daily Intake (CDI) and Hazard Quotient (HQ) for four groups

Location	Latitude	Longitude	NO ₃ ⁻ (mg/l)	CDI (mg/kg/day)				HQ Values			
				$CDI = \frac{C \times IR \times EF \times ED}{ABW \times AET}$				$HQ_{nitrate} = CDI / RfD$			
				Male	Female	Children	Infants	Male	Female	Children	Infants
Site-1	25.51°N	86.55°E	4.98	0.1532	0.1810	0.1942	0.1868	0.0958	0.1132	0.1214	0.1167
Site-2	25.54°N	86.56°E	5.85	0.1800	0.2127	0.2282	0.2193	0.1125	0.1329	0.1426	0.1371
Site-3	25.50°N	86.55°E	5.79	0.1781	0.2105	0.2258	0.2171	0.1113	0.1315	0.1411	0.1357
Site-4	25.82°N	86.44°E	4.19	0.1289	0.1523	0.1631	0.1571	0.0805	0.0952	0.1021	0.0982
Site-5	25.73°N	86.82°E	5.11	0.1572	0.1858	0.1993	0.9163	0.0982	0.1161	0.1246	0.0573
Site-6	25.97°N	86.59°E	5.04	0.1550	0.1832	0.1966	0.1890	0.0969	0.1146	0.1229	0.1181
Site-7	25.54°N	86.72°E	5.18	0.1593	0.1883	0.2020	0.1942	0.0996	0.1177	0.1263	0.1214
Site-8	25.87°N	86.78°E	6.78	0.2086	0.2465	0.2644	0.2542	0.1303	0.1540	0.1653	0.1589
Site-9	26.05°N	86.76°E	5.21	0.1603	0.1894	0.2032	0.1954	0.1009	0.1184	0.1269	0.1221
Site-10	26.25°N	86.86°E	4.73	0.1455	0.172	0.1845	0.1740	0.0909	0.1075	0.1152	0.1108
Site-11	26.27°N	86.80°E	4.16	0.1280	0.1512	0.1622	0.1560	0.0800	0.0945	0.1014	0.0975
Site-12	26.23°N	86.86°E	4.93	0.1516	0.1792	0.1923	0.1849	0.0947	0.1120	0.1201	0.1155

TABLE 8. Nitrate concentration in Groundwater and their Chronic Daily Intake (CDI) and Hazard Quotient (HQ) for four groups

Location	Latitude	Longitude	NO ₃ ⁻ (mg/l)	CDI(mg/kg/day)				HQ Values			
				$CDI = \frac{C \times IR \times EF \times ED}{ABW \times AET}$				$HQ_{nitrate} = CDI/RfD$			
				Male	Female	Children	Infants	Male	Female	Children	Infants
Site-1	25.51°N	86.55°E	3.19	0.0981	0.1160	0.1244	0.1196	0.0613	0.0725	0.0777	0.0740
Site-2	25.54°N	86.56°E	3.32	0.1021	0.1207	0.1295	0.1245	0.0638	0.0754	0.0809	0.0770
Site-3	25.50°N	86.55°E	3.74	0.1150	0.1360	0.1459	0.1402	0.0718	0.0850	0.0912	0.0870
Site-4	25.82°N	86.44°E	4.17	0.1498	0.1770	0.1899	0.1826	0.0936	0.1106	0.1187	0.1141
Site-5	25.73°N	86.82°E	5.51	0.1695	0.2003	0.2148	0.2066	0.1059	0.1252	0.1343	0.1290
Site-6	25.97°N	86.59°E	5.11	0.1572	0.1858	0.1993	0.1916	0.0983	0.1161	0.1245	0.1197
Site-7	25.54°N	86.72°E	4.79	0.1473	0.1741	0.1868	0.1796	0.0921	0.1088	0.1166	0.1192
Site-8	25.87°N	86.78°E	5.12	0.1575	0.1861	0.1996	0.1920	0.0984	0.1163	0.1248	0.1200
Site-9	26.05°N	86.76°E	5.89	0.1812	0.2141	0.2297	0.2208	0.1133	0.1138	0.1435	0.1380
Site-10	26.25°N	86.86°E	4.12	0.1267	0.1498	0.1606	0.1545	0.0792	0.0936	0.1004	0.0960
Site-11	26.27°N	86.80°E	6.14	0.1889	0.2232	0.2394	0.2300	0.1181	0.1395	0.1497	0.1439
Site-12	26.23°N	86.86°E	4.16	0.1280	0.1512	0.1622	0.1560	0.0800	0.0940	0.1014	0.0970

Table:9:HQRANGE OF SAMPLES FOR FOUR GROUP

Human	Range of HQ	Health risk	No. of samples
Male	>1	High risk	0
	<1	No risk	12
Female	>1	High risk	0
	<1	No risk	12
Children	>1	High risk	0
	<1	No risk	12
Infant	>1	High risk	0
	<1	No risk	12

Groundwater contamination with NO_3^- and other parameters

The groundwater samples were analysed for important characteristics (pH, conductivity, TDS and nitrate contamination level), which indicate surface leaching of contaminants to shallow aquifers. The results of pH, conductivity, TDS and nitrate contamination level were in the ranges of 6.94–7.93, 589.4–826.5 ($\mu\text{S}/\text{cm}$), 298–509 (mg/L), and 4.16 to 6.78 mg/L , respectively (Tables 6). These parameters showed some significant spatial variations among sampling sites. The average concentration of NO_3^- at various sampling locations in shallow aquifers were 4.98, 5.85, 5.79, 4.19, 5.11, 5.04, 5.18, 6.78, 5.21, 4.73, 4.16 and 4.93 mg/L at site-1, site-2, site-3, site-4, site-5, site-6, site-7, site-8, site-9, site-10, site-11, and site-12, respectively (Table 6). The maximum values at site-8 (6.78 mg/L), was lower than the BIS limit. The NO_3^- in this study are arranged between 4.16 to 6.78 mg/L , which in the safer side but still it is high time to be serious regarding lowering of the nitrate level in the area. High intensive double or triple cropping system of the area has mostly utilized the leached nitrate through ramified roots system of the intercrops. There were significant spatial variation in groundwater concentration in this area indicate significant deviation from the site mean values. The content in the majority of sites was significantly lower than the prescribed safe limit by WHO and BIS. The difference in NO_3^- content at various sampling locations may be attributed to the seasonal precipitation pattern, groundwater recharge rate, evapotranspiration process, etc. Other factors responsible for spatial variations in NO_3^- contaminations include soil particle size, soil water holding capacity, rainfall intensity, depth of water table, aquifer media, etc. (Buvaneshwari et al. 2017; Nakagawa et al. 2017; Reet al. 2017). The cropping pattern significantly affect the consumption of fertilizers, as reports suggest that wheat yields require high N-based fertilizers (Huang 2013). The geology of this area is characterized by alluvial plains formed by fertile sediments, deposited by the Koshi River that favour the extensive agriculture practices in this region. Extensive use of synthetic fertilizers to produce more yields can have negative impacts on groundwater quality but still it is in the safer side. Several reports suggest that in Bihar, the fertilizer consumption rate (per hectare) is highest (245.25 kg) in 2019–20 closely followed by Puducherry (244.77) in spite of its small size, than any other states in the country. The excessive use of fertilizers since the last 20 years could have enriched the local soils and groundwater with NO_3^- contents. The fertilizer consumption in the states since the last 2 decades has been increased drastically. Soils of the region are of sandy nature with high porosity and low water holding capacity that tend to leach quicker the surface contaminants to the groundwater. Groundwater NO_3^- concentrations were consistently around or less than 10 mg/L from the beginning of the experiment, but then gradually increased. This clearly shows that NO_3^- leaching was disproportional to the application rates. The results suggest that during the rainy season groundwater may not be suitable for drinking purpose. Recharging groundwater with water containing lower concentration of NO_3^- would be needed to dilute contaminated groundwater. The pronounced increase in NO_3^- concentrations in August 2022 was accompanied by an elevation of the groundwater table (data not shown). Not only there was less travel distance for NO_3^- in the topsoil to leach to the groundwater, but also NO_3^- present in the soil readily dissolved in the groundwater. Indeed, groundwater table depth was significant

antly correlated with groundwater NO_3^-

N. On the contrary, many scientists found that water table depth was significantly positively correlated with average groundwater NO_3^- N.

CONCLUSIONS

This study revealed groundwater NO_3^- contamination in the areas of NE alluvial plains of Bihar - an area known for its high population density and extensive maize cultivation. The combination of water movement through the soil profile during the rainy season together with high residual NO_3^-

N from N fertilization and shallow groundwater table renders soils vulnerable to excessive nitrate leaching. The study suggests the surface leaching as a prime source of nitrate contamination in groundwater, which

is the only source of potable water for a majority of the rural populations in this area. Thus, consumption of such NO_3^- contaminated water may pose a serious health hazard to residents as NO_3^- is listed as a non-

carcinogenic chronic toxicant for humans. The carcinogenic and non-carcinogenic health risk as estimated through HQ nitrates showed values < 1 in all of the sampling sites, suggesting a low risk of the non-

carcinogenic or carcinogenic effect of excess intake of NO_3^- through the water. Sampled water from different locations showed nitrate contamination which is just approaching the safe limit. Therefore, it is time to be cautious to refrain from use of heavy doses of nitrogenous fertilizer by the farmers of the study area. Further studies on actual records of NO_3^- toxicity in residents is needed to validate the results of the present finding.

REFERENCES

- Alkemade, J.R.M., Van Grinsven, J.J.M., Wiertz, J. and Kros, J. (1998). Towards integrated national modelling with particular reference to the environmental effects of nutrients. 1st International Nitrogen Conference: 101–105.
- American Public Health Association APHA (1998): Standard methods for the examination of water and wastewater, 20th edition, APHA, AWWA, WEF, Washington DC.
- Anornu G, Gibrilla A and Adomako D (2017) Tracking nitrate sources in groundwater and associated health risk for rural communities in the white Volta River basin of Ghana using isotopic approach. *Sci Total Environ* 603-604: 687-698.
- Blake, G.R. and Hartge, K.H. 1986. Bulk density. In Klute, A. (ed.) *Methods of Soil Analysis. Part 1. Agron. Monogr. 9. 2nd ed.* ASA and SSSA, Madison, WI. pp. 363–375. Cambardella,
- Buvaneshwari S, Riotte J, Sekhar M, Kumar MSM, Sharma AK, Duprey JL, Andry S, Giriraja PR, Praveen Kumar Reddy Y, Moger H, Purand P, Braun JJ, Ruiz L (2017) Groundwater resource vulnerability and spatial variability of nitrate contamination: insights from high density tubewell monitoring in a hard rock aquifer. *Sci Total Environ* 579: 838–847.
- Chen J, Wu H, Qian H, and Gao Y (2017) Assessing nitrate and fluoride contaminants in drinking water and their health risk of rural residents living in a semiarid region of Northwest China. *Expo Health* 9: 183-195.
- Council of European Communities. (1980). Relating to the quality of water intended for human consumption. *Official J. Eur. Commun. (L229): 11–29.*

- Fang, Q.; Ma, L.; Yu, Q.; Malone, R. W.; Saseendran, S. A.; Ahuja, L. R. (2008) Modeling nitrogen and water management effects in a wheat-maize double-cropping system. *J. Environ. Qual.*, **37**, 1466–1479.
- Gatseva PD and Argirova MD (2008) High-nitrate levels in drinking water may be a risk factor for thyroid dysfunction in children and pregnant women living in rural Bulgarian areas. *Int J Hyg Environ Health* **211**: 555–559.
- Hu, C.; Saseendran, S. A.; Green, T. R.; Ma, L.; Li, X.; Ahuja, L. R. (2006) Evaluating nitrogen and water management in a double-cropping system using RZWQM. *Vadose Zone J.*, **5**, 493–505.
- Huang G (2013) Characterization of nitrate contamination in an arid region of China. *J Environ Prot* **4**(07): 46–52
- Huang J, Xu J, Liu X, Liu J, Wang L (2011) Spatial distribution pattern analysis of ground water nitrate nitrogen pollution in Shandong intensive farming regions of China using neural network method. *Math Comput Model* **54**(3): 995–1004.
- Joosten, L. T. A., Buijze, S. T. and Jansen, D. M. (1998). Nitrate in sources of drinking water?: Dutch drinking water companies aim at prevention. *Environ. Pollut.* (Supl. 1), **102**: 489–492.
- Ju, X. T., Xing, G. X., Chen, X. P., Zhang, S. L., Zhang, L. J., Liu, X. J., Cui, Z. L., Yin, B., Christie, P., Zhu, Z. L., Zhang, F. S., (2009) Reducing environmental risk by improving N management in intensive Chinese agricultural systems. *Proc. Natl. Acad. Sci. U. S. A.* **106**, 3041–3046.
- Keeney, D. R. and Nelson, D. W. 1982. Nitrogen-inorganic forms. In Page, A. L. (ed.) *Methods of Soil Analysis. Part 2. SSSA Monogr.* **9**. 2nd ed. ASA, Madison, WI. pp. 643–698.
- Lapworth DJ, Krishan G, MacDonald AM, Rao MS (2017) Groundwater quality in the alluvial aquifer system of Northwest India: new evidence of the extent of anthropogenic and geogenic contamination. *Sci Total Environ* **599**: 1433–1444.
- Li P and Qian H (2011) Human health risk assessment for chemical pollutants in drinking water source in Shizuishan City, Northwest China. *Iranian J Environ Health Sci Eng* **8**(1): 41–48.
- Li, Y., Liu, H. J., Huang, G. H., Zhang, R. H., Yang, H. Y (2016). Nitrate nitrogen accumulation and leaching pattern in a winter wheat: summer maize cropping field in the North China Plain. *Environ. Earth Sci.* **75**, 1–12.
- Liu HB, Li ZH, Zhang YG (2006) Nitrate contamination of groundwater and its affecting factors in rural areas of Beijing plain. *Acta Pedol Sin* **43**(3): 413.
- Majumdar D and Gupta N (2000) Nitrate pollution of groundwater and associated human health disorders. *Ind J Environ Health* **42**(1): 28–39.
- Minet EP, Goodhue R, Meier-Augenstein W, Kalin RM, Fenton O, Richards KG, Coxon CE (2017) Combining stable isotopes with contamination indicators: a method for improved investigation of nitrate sources and dynamics in aquifers with mixed nitrogen inputs. *Wat Res* **124**: 85–96.
- Nakagawa K, Amano H, Takao Y, Hosono T, Berndtsson R (2017) On the use of coprostanol to identify source of nitrate pollution in groundwater. *J Hydrol* **550**: 663–668.
- Parkinson, R. J. (1993). In *Nitrate: Processes Patterns Management* (eds Tim, P. B., Louise, H. A., Stephen, T. T., Wiley, Chichester, UK, pp. 321–339.
- Rao, E. V. S. P. and Puttanna, K. (2000). Nitrates, agriculture and environment, *Current Science*, Vol. 79, NO. 9

- Re V, Sacchi E, Kammoun S, Tringali C, Trabelsi R, Zouari K, Daniele S (2017) Integrated socio-hydrogeological approach to tackle nitrate contamination in groundwater resources. The case of Grombali basin (Tunisia). *Sci Total Environ* 593:664–676.
- Sajil KPJ, Jegathambal P, James EJ (2014) Chemometric evaluation of nitrate contamination in the groundwater of a hard rock area in Dharapuram, South India. *Appl Water Sci* 4(4):397–405.
- Salo, T., Turtola, E., (2006) Nitrogen balance as an indicator of nitrogen leaching in Finland. *Agric. Ecosyst. Environ.* 113, 98–107.
- Stigter T, Almeida P, Dill AC, Ribeiro L (2005) Influence of irrigation on groundwater nitrate concentrations in areas considered to have low vulnerability to contamination, groundwater and land development: IAH paper on hydrogeology, pp. 69–85.
- Su H, Kang W, Xu Y, Wang J (2017) Evaluation of groundwater quality and health risks from contamination in the north edge of the Loess Plateau, Yulin City, Northwest China. *Environ Earth Sci* 76: 467.
- Sun, M.; Huo, Z.; Zheng, Y.; Dai, X.; Feng, S.; Mao, X. (2018) Quantifying long-term responses of crop yield and nitrate leaching in an intensive farmland using a agro-eco-environmental model. *Sci. Total Environ.*, **613–614**, 1003–1012.
- Suthar S, Bishnoi P, Singh S, Mutiyar PK, Nema AK, Patil NS (2009) Nitrate contamination in groundwater of some rural areas of Rajasthan, India. *J Haz Mat* 171(1): 189–199.
- Tao, T., Xin, K., (2014) Public health: a sustainable plan for China's drinking water. *Nature* 511, 527–528.
- USEPA (1995) Drinking-water regulations and health advisories. office of water, USEPA, Washington, DC. 11 pp.
- USEPA (1992) EPA's approach for assessing the risks associated with chronic exposure to carcinogens. Jan 1992. <http://www.epa.gov/iris/carcino.htm>.
- USEPA (2001) Risk assessment guidance for superfund: process for conducting probabilistic risk assessment (volume III—part a, 540-R-502-002)
- USEPA (2004) The incidence and severity of sediment contamination in surface waters of the United States, National Sediment Quality Survey. EPA 823-R-04-007, second ed. U.S. Environmental Protection Agency, Office of Water: Washington, DC.
- USEPA. (1999) Guidelines for Carcinogen Risk Assessment Review draft. NCEA-F-0644, Jul 1999. <http://www.epa.gov/cancer/guidelines/draft-guidelines-carcinogen-ra-1999.htm>
- Vitousek, P.M., Naylor, R., Crews, T., David, M.B., Drinkwater, L.E., Holland, E., Johnes, P.J., Katzenberger, J., Martinelli, L.A., Matson, P.A., Nziguheba, G., Ojima, D., Palm, C.A., Robertson, G.P., Sanchez, P.A., Townsend, A.R., Zhang, F.S., (2009) Nutrient imbalances in agricultural development. *Science* 324, 1519–1520.
- Wang S, Zheng W, Currell M, Yang Y, Zhao H, Lv M (2017) Relationship between land-use and sources and fate of nitrate in groundwater in a typical recharge area of the North China plain. *Sci Total Environ* 609:607–620.
- Water Quality Branch. (1995) Canadian water quality guidelines. Inland Waters Directorate, Ottawa, Ont., Canada.
- Wick, K., Heumesser, C., Schmid, E., (2012) Groundwater nitrate contamination: factors and indicators. *J. Environ. Manage.* 111, 178–186.

- WorldHealthOrganization(WHO),(2011)Watersafetyandquality.http://www.who.int/water_sanitation_health/water-quality/en/.
- WuJ,andSunZ(2016)Evaluationofshallowgroundwatercontaminationandassociatedhumanhealthriskinanalluvialplainimpactedbyagriculturalandindustrialactivities,mid-WestChina.ExpoHealth8(3):311-329.
- YadavSKandGuptaA(2022)HumanHealthRiskAssessmentforNitrateintakeinLucknow,India.InternationalJournalforResearchinAppliedScience&EngineeringTechnology(IJRASET),10(10):1555-1559.
- Yadav,S.N.,(1997)Formulationandestimationofnitrate-nitrogenleachingfromcorncultivation.J.Environ.Qual.26,808–814.
- ZhangG,JiaoY,LeeDJ(2015)Alab-scaleanoxic/oxicbioelectrochemicalreactorforleachate treatments.BioresourTechnol186:97–105
- Zhang,X.;Chen,S.;Sun,H.;Shao,L.;Wang,Y.(2011)ChangesinevapotranspirationoverirrigatedwinterwheatandmaizeinNorthChinaPlainoverthreedecades.Agric.WaterManagement.98,1097–1104.
- ZhouY,WeiA,LiJ,YanLandLiJ(2016a)GroundwaterqualityevaluationandhealthriskassessmentintheYinchuanregion,NorthwestChina.ExpoHealth,8:443-456.
- Zhou,J.Y.,Gu,B.J.,Schlesinger,W.H.,Ju,X.T.,(2016)SignificantaccumulationofnitrateinChinesesemi-humidcroplands.Sci.Rep.6,1–8.
- DaviesB,CoulterJA,PagliariPH.(2020)Timingandrateofnitrogenfertilizationinfluencemaizeyieldandnitrogenuseefficiency.PLoSOne;15(5):e0233674.doi:10.1371/journal.pone.0233674.PMID:32469984;PMCID:PMC7259653.
- Ahmad,Nasim,Sinha,DK,SinghKM,andMishraRR(2017)GrowthPerformanceandResourceUseEfficiencyofMaizeinBihar:EconomicPerspectives.JournalofAgriSearch4(1):71-75.