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Groundwater Contamination with Nitrate and Human Health Risk Assessment of North East Alluvial Plains of Bihar, India

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Authors' contributions

This work was carried out in collaboration among all authors. Authors AK, SKM did the conceptualization. Authors AK, SKM, Sanjay Kumar Singh did the data curation. Authors Sanjay Kumar Singh, S.K. Sinha did the formal analysis. Authors AK, SKM, S.K. Sinha did the investigation. Authors AK, SKM, Sanjay Kumar Singh did the methodology. Author AK did the project administration. Authors SKM, S.K. Sinha did the resources. Authors LR, SKM did the software. Authors AK, Sanjay Kumar Singh did the supervision. Authors AK, SKM did the visualization. Author AK did the original draft. Authors AK, S.K. Sinha, KS did the review & editing. All authors read and approved the final manuscript.

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ABSTRACT

Groundwater is natural resources which supplies almost half of all drinking water in the world and plays a key role in food production. Consuming water containing high nitrate concentration have immediate effect on infant and could cause the risk of diseases 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 it. In rural areas, groundwater contamination is a problem related to the excessive use of chemical fertilizers by local farmers. 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 278 kg/ha in Kharif maize, which is about 131.72% more than the RDN of Kharif maize i.e. 120 KgN/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 NO3- leaching. The groundwater and surfacewater from 12 villages was collected and various quality parameters were analysed. The nitrate in ground water 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 ground water 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.

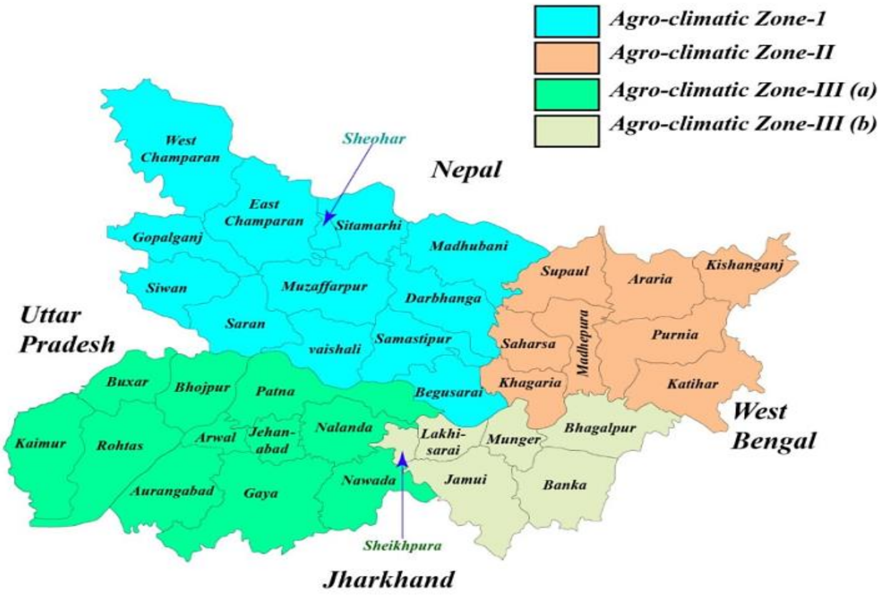
Keywords: Maize; NO3-N contamination; groundwater; hazard quotient; HHRA.

1. INTRODUCTION

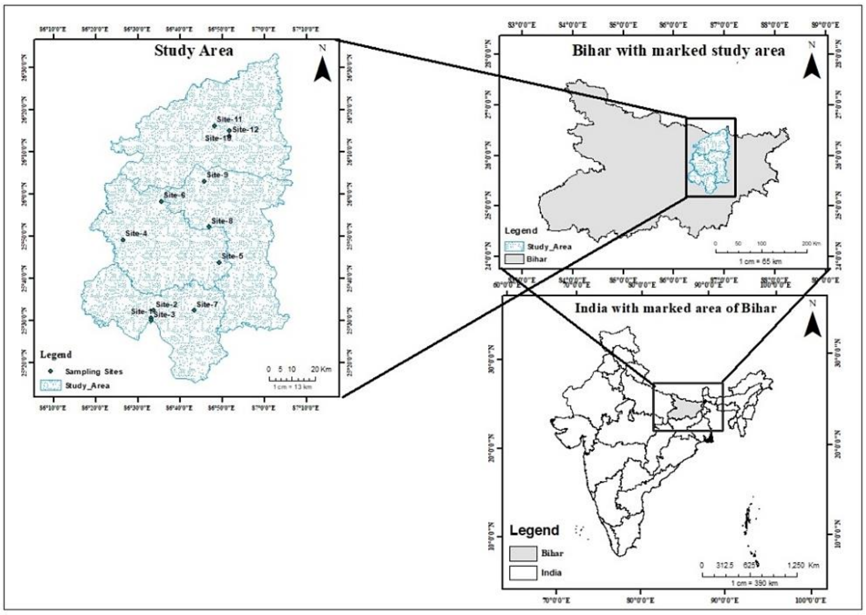
“The eastern region of Bihar is popularly known as the ‘maize hub’ where the maize is cultivated in two lakh hectares area. The demand for maize is growing globally due to its multiple uses for food, feed and industry sectors. In Bihar, Maize is grown in almost all the districts of all the three agro-climatic 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 Rabi maize production. The comprehensive data underscores the escalating trends in the area, production, and productivity of maize in Bihar” Ahmad et al. [1]. “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^- N) is troublesome as it leaches through the soil into groundwater. Permeable soils make the region susceptible to groundwater pollution by NO_3^-N. Nitrate that has accumulated in soils 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”, Davies et al. [2]. 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 enters into the groundwater. Consequently, the excessive use of nitrogen fertilizer and flood irrigation has led to pronounced N leaching as reported by Yadav, [3] (amounting to 15–55% of applied N fertilizer) and an augmented risk of groundwater nitrate contamination, as highlighted by Sun et al. [4]. The application of nitrogen fertilizers 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 was observed

(30 to 40%) in regions where high nitrogen application rates are common [5]. The abundance of nitrogen fertilizer lead to nitrate accumulation in the soil [6], in regions with double or triple cropping system exist [7]. This nitrate accumulation in the soil becomes problematic when heavy rainfall or irrigation occurs, as contaminating drinking water sources. Groundwater contamination with NO_3^- N 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 [8]. 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 [9]. “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 dysfunctions in children and pregnant women” [10]. “The concentration of NO_3^- N 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 recommend a limit of 50 mg NO_3^- L^{-1} (11 mg NO_{3-} N L^{-1}) in potable water. The US Environmental Protection Agency and the Canadian Water Quality Branch [11] have set a limit of 44 mg NO_3^- L^{-1} (10 mg NO_3^- N L^{-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 cycles of pumping and return flows in the underground water system” [12]. “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” [13,14-17]. “Beyond infants, adults are also susceptible to gastric cancer, respiratory problems, headaches, fatigue, thyroid gland hypertrophy and multiple sclerosis” [18-22]; World Health Organization [23]. “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 rainfall and soil properties such as soil texture, affect soil NO_3^- accumulation and leaching” [24-26]. The level of nitrate accumulation in soils 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 [27-31]. 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 researcher 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”. [32] 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 studied region to estimate the overall concentration of nitrate in groundwater. Since part of the state has higher human population, this area could be at risk due to nitrate contamination in local surface water and groundwater sources”. [32] Looking into these facts, the university decided to estimate the groundwater nitrate level in these areas and correlating the high nitrate content (than safe limits as suggested by BIS) to possible human health risk using a human health risk assessment model as proposed by USEPA [33].



Map 1. Agro-climatic zones of Bihar



Map 2. Details of sampling site

2. MATERIALS AND METHODS

2.1 Description of the Study Area

Bihar is situated in the eastern part of India in between latitudes 24°20'10"N and 27°31'15"N and longitudes 83°19'50"E and 88°17'40"E. It is an entirely land–locked state, in a subtropical region of the temperate zone. Bihar lies between the humid West Bengal in the east and the sub humid 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 average elevation above sea level of 173 feet. As per agro-climatic zone it is divided in ACZ-I, II, IIIA and IIIB.

2.2 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 ground water draft is 10.77 BCM. The geographical location of all sampling site is presented in Table 1.

2.3 Water Sample Collection, Sampling Procedure and Water Analysis

The Water samples were collected across 12 different sites in Seemanchal districts and neighbouring Koshi river region, whose depths varied from 6 m to 18 m. The water samples were fetched from bore wells 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 [34]) were pH, temperature, conductivity, total suspended solids (TSS), total dissolved solids (TDS) and nitrate (NO_3^-).

2.4 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 contaminants over a specific period. The HHRA is computed using the values of present concentration of a contaminant in groundwater and its exposure durations 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 day^{-1} 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” [35,36]. Some studies suggest that ingestion and dermal contact as the leading pathways of nitrate exposures in humans [37,38] but ingestion seems to have even greater risk than dermal contact. The exposure of nitrate through ingestion with drinking water is calculated by following Eq. (i) [39].

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

where CDI (chronic daily intake) is the ingestion dose from drinking water (mg/kg/day), C is the concentration of nitrate estimated in groundwater samples (mg L^{-1}), IR is the average daily ingestion rate of drinking water (L/day) and the values of IR (2 L/day for adult (male & female), 0.78 L/day for children and 0.3 L/day for infants) were used for this model as taken from published literature; EF is the exposure frequencies (365 days/year), ED is the exposure duration (standard exposure duration in literature is suggested 40 years for adult (male and female), 12 years for children and < 1 for infants), ABW is the average body weight (65 kg for male, 55 kg for female, 20 kg for children and 8 kg for infants; and AET is the average exposure time (days) which is 14,600 days for male and female and 4380 days for children and 365 days for infants [40]. The present study focuses on the non-carcinogenic health risk of nitrate mainly estimated by the hazard quotient (HQ_{nitrate}) values, which is estimated through following Eq. (ii) [41]:

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

Where RfD is reference dose, RfD indicates that reference of NO_3^- (1.6 mg/kg/d) were obtained from the database of Integrated Risk Information System (IRIS) and USEPA [42]. The calculation of hazard quotient value, $HQ_{\text{nitrate}} > 1$ is referred as potentially known to cause health risks and values of $HQ_{\text{nitrate}} < 1$ indicates that it is an acceptable limit of non-carcinogenic risk in individuals due to ingestion of Nitrate contaminated groundwater.

Table 1. Parameters and their values used for HHRA computation (USAEPA)

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

2.5 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 Arc GIS software.

3. RESULTS AND DISCUSSION

3.1 Soil Analysis/ Bulk Soil Nitrate-Nitrogen Measurements

3.1.1 Soil characterisation

The soil in ACZ-II is comprised of alluvial deposition of Koshi river basin occupied by clay sand to 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 has higher groundwater contamination potential from surface contaminants. The soil at Khagaria was a loamy sand 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 depth wise 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), Loamy sand (90-105 cm) and Sandy loam (105-120) under varying depth. While bulk density varies from 1.27-1.79 (g/cm^3). 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/cm^3). 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/cm^3). 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/cm^3). 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.

3.2 Vertical Distribution of Nitrate and Leaching Percentage

3.2.1 Nitrate leaching in soil

Total 96 soil samples from different soil depths at the interval of 15 cm depth, sampled up-to 105-120 cm depth have been collected from 12 selected sites (villages) of four districts. The vertical distribution of nitrate indicates accumulation of nitrate in soil and it varied from 26.73 to 42.95 kg/ha (105-120 cm depth). The overall leaching of nitrate ranges from 9.43 – 12.50 % with an average value of 11.02 % over applied dose of Nitrogen. The highest leaching was recorded in Khagaria (42.95 kg N/ha) and minimum in Saharsa district (26.73 kg N/ha). The preliminary result indicated that overuse of nitrogen fertilizer has caused N leaching in soil.

3.2.2 Geographical location of sampling site and water-depth

The water samples were collected to investigate the concentration of NO_3 in drinking water from an intensively cultivated belt of maize from the Agro-climatic zone-II of the Bihar.

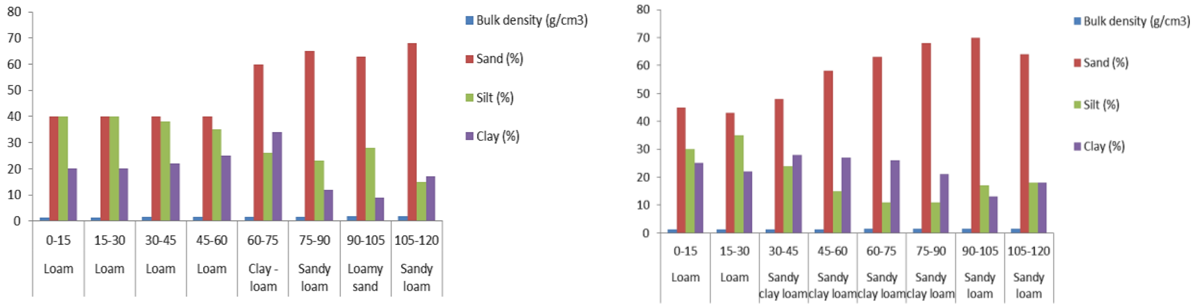


Fig. 1a. Khagaria district

Fig. 1b. Madhepura district

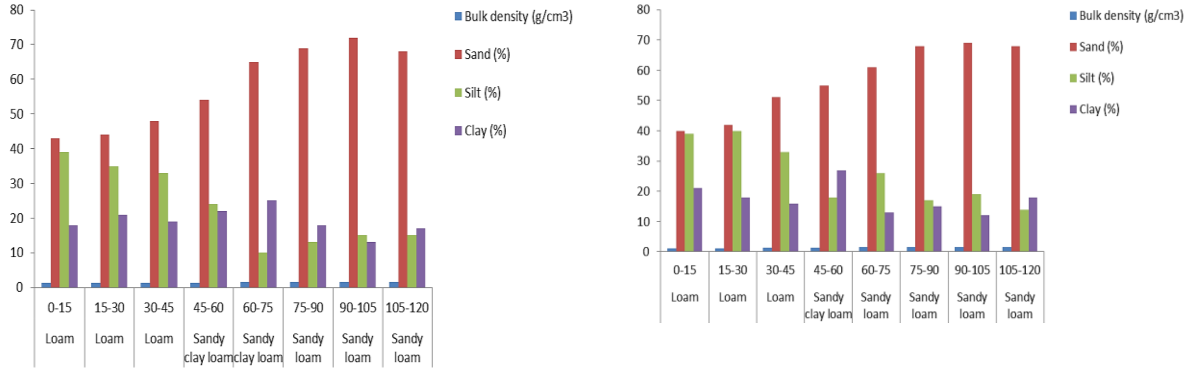


Fig. 1c. Saharsa district

Fig. 1d. Supaul district

Fig. 1. Depth-wise soil textural class (Percent soil fraction) of studied districts in north east alluvial plains of Bihar

3.2.3 Nitrogen uptake by maize

The grain yield of Rabi maize is varying from 6873 – 9915 (kg/ha) in zone with highest value in the Khagaria district and being lowest in the Saharsa district. The nitrogen content in grain ranges from 1.49 to 1.63 % with mean value of 1.54%. The nitrogen uptake in grain varying from 105.84 to 161.62 (kg N/ha) with the mean value of 123.34 (kg N/ha). Similarly the stover yield of maize ranges from 5413 -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 varying 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.

3.2.4 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 ground water varied from 20 - 60 feet. The nitrate level in ground water 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 ground water 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.

3.3 Nitrate Exposure and Human Health Risk Assessment

Groundwater quality has been steadily declining in recent decades as a result of numerous pollution sources such fertilisers and chemicals. The ingestion of contaminated groundwater can adversely affect the health of humans through varieties of exposures including direct ingestion, dermal contact, washing, etc. [42]. 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,

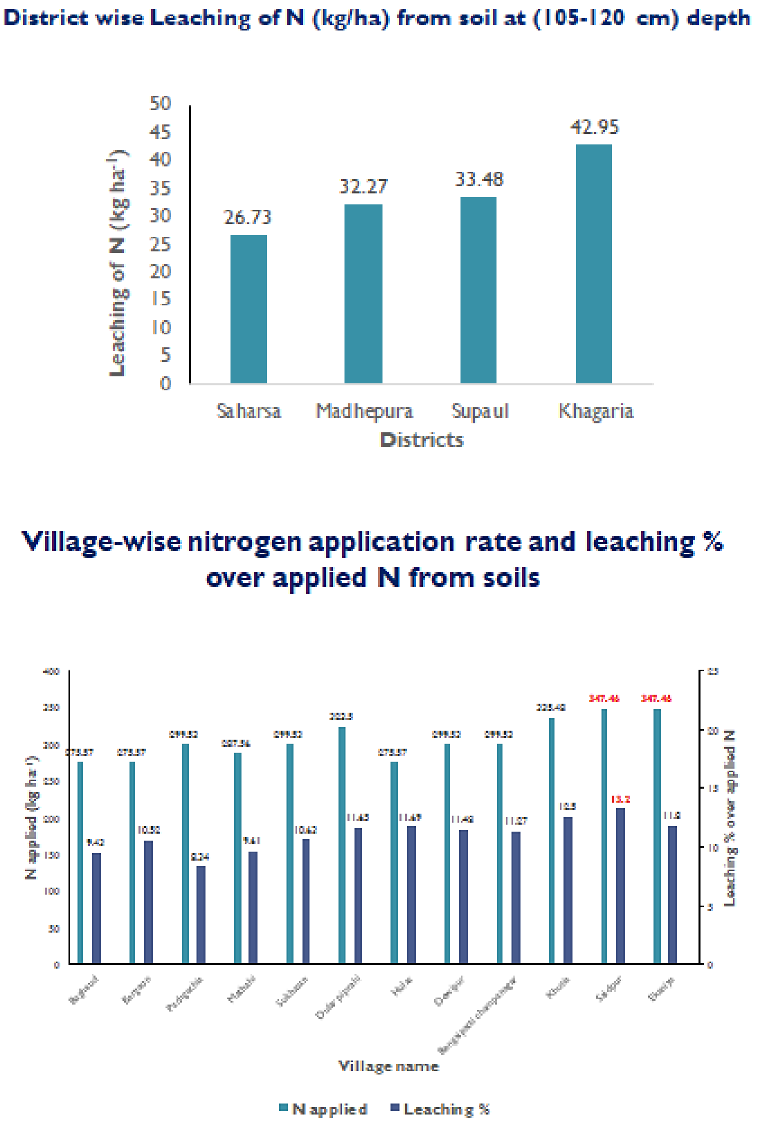


Fig. 2. Nitrogen application rate and leaching of Nitrate from soil in ACZ-II

Table 2. The coordinates of water sampling sites in Agro-climatic zone –II of the Bihar

Site No.	Village name	Villages types	Sampling Depth (m)	Name of Districts	Coordinates	
					Latitude	Longitude
Site-1	Khutia	CCS	6.09	Khagaria	25.51ºN	86.55º E
Site-2	Saidpur	Non-CCS	18.28	Khagaria	25.54ºN	86.56ºE
Site-3	Ekaniya	Non-CCS	12.19	Khagaria	25.50ºN	86.55ºE
Site-4	Baghaud	CCS	12.19	Saharsha	25.82º N	86.44º E
Site-5	Bangaon	Non-CCS	15.24	Saharsha	25.73º N	86.82º E
Site-6	Pachgachia	Non-CCS	9.14	Saharsha	25.97º N	86.59º E
Site-7	Mathahi	CCS	9.14	Madhepura	25.54º N	86.72º E
Site-8	Sukhasan	Non-CCS	7.26	Madhepura	25.87º N,	86.78º E
Site-9	Dular piprahi	Non-CCS	9.14	Madhepura	26.05º N	86.76º E
Site-10	Hulas	CCS	15.24	Supaul	26.25º N,	86.86º E
Site-11	Dewipur	Non-CCS	18.28	Supaul	26.27º N	86.80º E
Site-12	Bengaipatti champanagar	Non-CCS	13.71	Supaul	26.23º N,	86.86º E

** MSL: mean sea level*

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

Districts Name	Grain			Stover			Total Uptake (Kg N/ ha)
	Yield* (Kg/ha)	N (%)	N-Uptake (Kg N/ ha)	Yield (Kg/ha)	N (%)	N-Uptake (Kg N/ 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 (kg N/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

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 this values for all four group of peoples ranges from 0.0981 to 0.1889; 0.1160 to 0.2232; 0.1244 to 0.2394 and 0.1196 to 0.2300 respectively for nitrate contaminated ground water (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 ground water (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 cautions 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.

3.3.1 Groundwater contamination with NO_3^-N and other parameters

The groundwater samples were analysed for important characteristics (pH, conductivity, TDS and nitrate contamination level), which indicates 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 (µS/cm), 298 - 509 (mg/L), and 4.16 to 6.78 mg/L, respectively (Table 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 area ranged between 4.16 to 6.78 mg/L, which in the safer side but still it is high time to be sincere 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 root system of the inter crops. “There were significant a spatial variation in groundwater concentration in this area indicates 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,

Table 5. Water quality parameters of selected sites located in ACZ-II in dry season

Districts	**Village name**	**Surface water**					**Ground water**			
		pH	**Conductivity (µS/cm)**	**TDS (mg/L)**	**NO_3^- (mg/L)**	**Sampling Depth (ft)**	**pH**	**Conductivity (µS/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
Mean value		**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
Mean value		**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
	Dular piprahi	7.25	735.4	451	5.21	30	6.91	979.8	199	5.89
Mean value		**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
	Bengaipatti champanagar	7.19	826.5	368	4.93	45	7.67	949.6	152	4.16
Mean value		**7.35**	**719.83**	**328.33**	**4.61**	**-**	**7.60**	**1067.93**	**184**	**4.81**

Table 6. Water quality parameters of reference point located in ACZ-II in dry season

Districts	**Reference point**	**Reference water sampling depth (ft)**	**pH**	**Conductivity (µS/cm)**	**TDS (mg/L)**	**NO_3^- (mg/L)**
* Khagaria	Khutia	400	7.01	750	263	1.05
* Saharsa	Kali temple	200	7.18	732	218	0.92
*Madhepura	Singheswar temple	300	7.29	719	213	0.98
* Supaul	Ram janki Math	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) CDI = $\frac{C \times IR \times EF \times ED}{ABW \times AET}$				HQ Values 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) CDI = $\frac{C \times IR \times EF \times ED}{ABW \times AET}$				HQ Values 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. HQ range 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

depth of water table, aquifer media, etc". [12,43,44]. "The cropping patterns significantly affect the consumption of fertilizers, as reports suggest that wheat yields require high N-based fertilizers" [45]. "The geology of this area is characterized by alluvial plains formed by the fertile sediments, deposited by the Koshi Rivers that favours 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 fertilizer since the last 20 years could have enriched the local soils and groundwater with NO_3 contents. The fertilizer consumption in the state 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 tends to leach quicker the surface contaminants to the groundwater". [32] Groundwater NO_3^-N 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 applications 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^-N would be needed to dilute contaminated groundwater. The pronounced increase in NO_3^-N concentrations in August 2022 was accompanied by an elevation of the groundwater table (data not sown). Not only there was less travel distance for NO_3^-N in the topsoil to leach to the groundwater, but also NO_3^-N present in the soil readily dissolved in the groundwater. Indeed, groundwater table depth was significantly 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.

4. 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 render 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 majority of the rural populations in this area. Thus, consumption of such NO_3 contaminated water may pose serious health hazards in residents as NO_3 is enlisted as a non-carcinogenic chronic toxicant for humans. The carcinogenic and non-carcinogenic health risk as estimated through HQ nitrate showed values < 1 in all of 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 to the safe limit. Therefore it is time to be cautions to refrain away from use of heavy dose 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 result of present finding.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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