Original Research Article

GroundwatercontaminationwithnitrateandhumanhealthriskassessmentofNorthEastalluvialplainsofBihar

ABSTRACT

Groundwaterisnaturalresourceswithhigheconomicvalueandsocialsignificance.Itsuppliesalmosthalfofalldrinkingwaterintheworldandplaysakeyroleinfoodproduction.ConsumingwatercontaininghighnitrateconcentrationhavealmostimmediateeffectonainfantandcouldcausetheriskofvariousdiseasesoneofwhichisMethemoglobinemiainwhichbloodlackstheabilitytocarrysufficientoxygentotheindividualbodycells.Asdifferenceinnitrateconcentrationinwater, madeitimportanttostudytheundesirableeffectofnitrate.Inruralareas,groundwatercontaminationbynitratesisaproblemrelatedtotheexcessiveuseofchemicalfertilizersbylocalfarmersandtosomeextent,toeffluentsfromdomesticsewagesystems.ShallowgroundwaterplaysavitalroleinwateruseandtheyieldofMaize.Nitrogenapplicationsignificantlyaffectscropuptakeandutilizationofwaterfromirrigation,butlittleisknownaboutgroundwateruse.Farmersareapplyingnitrogenonanaverage278kg/hainKharifmaize,whichisabout131.72%morethantheRDNofKharifmaizei.e.120KgN/ha.ThemeanvalueofNapplicationbyfarmersrangesfrom251-323kg/ha.TheMaximumrateofNapplicationwasobservedinKhagaria(323kgN/ha)followedbyMadhepura(275.08kgN/ha)andminimumincaseofSaharsadistrict(251.16kgN/ha).Theapplicationrateofnitrogenousfertilizer,varyingfrom109.25%to169.16%overtheRDN,resultinginNO_3^- leaching.Thegroundandsurfacewaterfrom12villageswascollectedandvariouswaterqualityparameterswereanalysed.Thenitrateingroundwatervaried(1.87-6.19mg/l)andsurfacewater(1.87–3.84mg/l)beingmaximumconcentrationofnitrateinMadhepuradistrict.Thepresentstudyonnitrateleachinginsoil,itslevelofcontaminationingroundwaterandhumanhealthriskassessmentbychronicdailyintakeofnitrateandHazardQuotient(H.Q)valuesinthestudyareaofKhagaria,Saharsa,MadhepuraandSupaulhasbeencarriedoutintheeasternalluvialregionofBihar.

***KeyWords*:**Maize,NO_3^-Ncontamination,groundwater,HazardQuotient,HHRA

Introduction

TheeasternregionofBiharispopularlyknownasthe‘maizehub’wherethemaizeiscultivatedintwolakhhectaresarea.~~Maize(*ZeamaysL.*)isanimportantcerealcropafterwheat.Theimportanceofmaizeliesinitswideindustrialuses.~~Thedemandformaizeisgrowinggloballyduetoitsmultipleusesforfood,feedandindustrysectors.~~Maizeisgrownthroughouttheyearduetoitsphoto-thermoinsensitivecharacterandhighestgeneticyieldpotentialamongthecereals.~~InBihar,Maizeisgrowninalmostallthedistrictsofallthethreeagro-climaticzonesofBihar, butZone-IIismajormaizeproducingareathatcomesunderNorth-EasternalluvialplainsofBihar,wheresummercorn,paddy,wintercornandwinterwheat(*Triticumaestivum*L.)arethemajorcropsgrown.Notably,Katiharboaststhehighestproductivityat6510kg/ha,succeededbyMadhepura(5285kg/ha),Saharsa(4636kg/ha),Araria(4272kg/ha),Supaul(4096kg/ha),Vaishali(4067kg/ha),andMuzaffarpur(3935kg/ha).ThiszoneisrenownedforitsRabimaizeprodu ction.Thecomprehensivedataunderscorestheescalatingtrendsinthearea,production,andproductivityofmaizeinBihar,aselucidatedbyAhmadetal.(~~in~~2017).

Theapplicationratesofnitrogenousfertilizerinthisareaby localfarmers'oftenexceedcroprequirements,resultinginhighaccumulationofnitrate(NO_3)inthesoil.Theimpactofdownstreamnutrientexportfromagriculturallandscontinuestobeofmuchmoreconcern.Nitrate-nitrogen(NO_3^- N)istroublesomeasitleachesthroughthesoilintogroundwater.PermeablesoilsmaketheregionsusceptibletogroundwaterpollutionbyNO_3^- N.Nitratethathasaccumulatedinsoilsishighlypronetoleaching,whichisdirectlythreateningthequalityofgroundwater.Theoptimalmanagementdecisionsformaizeproductioninvolvecrucialconsiderationsofboththerateandtimingofnitrogen(N)application,asemphasizedbyDaviesetal.(~~in~~2020).Intherealmofmaizeproduction,nitrogenandwaterstandoutaspivotalfactors.Inthepursuitofelevatedyields,therehasbeenatendencytoapplyexcessivenitrogenfertilizer(rangingfrom300to400kgNha^{-1})withinthecurrentrotationsystem.Thissurpassesthecrop'sactualdemands,whichtypicallyrangebetween100to150kgNha^{-1}.Thenitrate-nitrogen,onceaccumulated,steadilymovesdownwardwithpercolatingwater,eventuallyentersintothegroundwater.Consequently,theexcessiveuseofnitrogenfertilizerandfloodirrigationhasledtopronouncedNleaching as reported by Yadav, 1997(amountingto15–55%ofappliedNfertilizer)andanaugmentedriskofgroundwaternitratecontamination,ashighlightedbySunetal.(2018).Theapplicationofnitrogenfertilizersisacommonpracticetoachievehighyields.

InIndia,theannualconsumptionofnitrogenfertilizerisapproximately27.23milliontons.Specifically,inBihar,theconsumptionofureaaccountsfor18.34%,slightlyexceedingthenationwidefigureof17.5%(YearEndReview-2020:MinistryofChemicals&Fertilizers).Lowefficienciesofnitrogenutilization was observed(30to40%)inregionswherehighnitrogenapplicationratesarecommon(Juetal.,2009).Theabundanceofnitrogenfertilizerleadtonitrateaccumulationinthesoil(Vitouseketal.,2009),inregionswithdoubleortriplecropping system exist (Zhouetal.,2016).Thisnitrateaccumulationinthesoilbecomesproblematicwhenheavyrainfallorirrigationoccurs,ascontaminatingdrinkingwatersources.GroundwatercontaminationwithNO_3^- Nisavitalconcern,especiallyinthese regionsdue to theintensivemaizeproduction.Thewidespreaduseofnitrogenousfertilizerisrecognizedasasignificantcontributortonitratepollutioningroundwater(Huangetal.,2011).

Nitrate,beinghighlysolubleinwaterandpoorlyretainedbysoil,posesariskofleachingintothesubsoilandeventuallyreachinggroundwaterifnottakenupbyplantsordenitrifiedtoN_2OandN_2(MajumdarandGupta,2000).Consumingwaterwithelevatednitratelevelscanleadtovarioushealthhazardsforhumans.Infantsaregenerallymoresusceptibletonitrate,butadultsmayalsoexperienceadverseeffectsfromconsumingwaterrichinnitrates,suchasthyroiddysfunctionsinchildrenandpregnantwomen(GatsevaandArginova,2008).TheconcentrationofNO_3^- Nindrinkingwatercanreachcriticallevels,andestablishedsafetylimitsaresetbyregulatorybodies.AccordingtotheBureauofIndianStandards(45mgL^{-1})andtheWorldHealthOrganization(50mgL^{-1}),thesafelimitfornitrateindrinkingwaterisdefined.BoththeWorldHealthOrganizationandtheEuropeanCommunity(CouncilofEuropeanCommunities,1980)recommendalimitof50mgNO_3^-L^{-1}(11 mgNO_{3-}

NL^{-1})inpotablewater.TheUSEnvironmentalProtectionAgency(USEPA,1995)andtheCanadian WaterQualityBranch(WaterQualityBranch,1995)havesetalimitof44mgNO_3^- L^{-1}(10mgNO_3^- NL^{-1})asthemaximumsafelevelindrinkingwater.

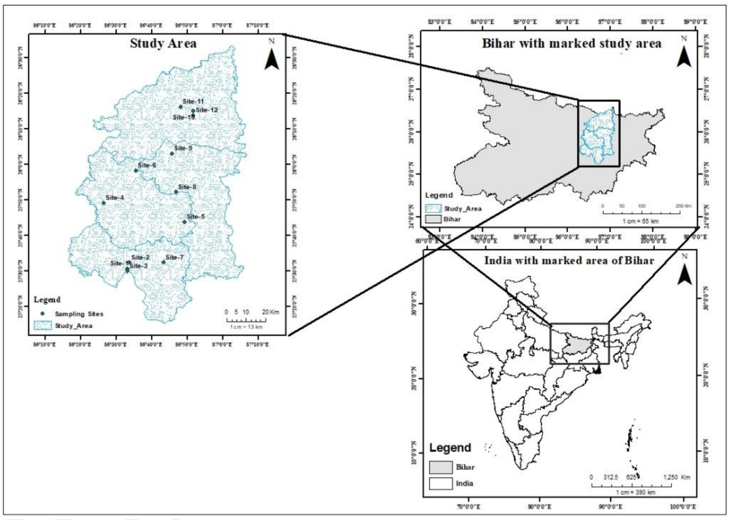
Thesestandardsunderscoretheimportanceofmonitoringandmanagingnitratelevelstosafe guardwaterqualityandhumanhealth.Thenitratecontentingroundwatercansignificantlyinfluencet henitrogenfluxinthesoilwhenusedforcropirrigation.Consequently,theprevalenceofhighnitratele velsinshallowgroundwatermaybeinfluencedbythecyclesofpumpingandreturnflowsintheunderg roundwatersystem(Buvaneshwarietal.,2017).Inthenorth-easternalluvialplainsofBihar,wheregroundwaterservesastheprimarydrinkingwatersourceforam ajorityofthepopulation,itiscrucialtoinvestigatethepotentialhealthrisksassociatedwithexcessivei ntakeofsuchwater.Therefore,maintainingnitratelevelsbelowthemaximumcontaminantlevelises sential.

Theoverconsumptionofnitrateindrinkingwaterposesserioushealthrisksandtoxicityinhu mans.Awell documentedexampleofnitratetoxicityisMethemoglobinemia,whichaffectsinfantsandpregnantw omen(Sajiletal.,2014).Beyondinfants,adultsarealsosusceptibletogastriccancer,respiratoryprobl ems,headaches,fatigue,thyroidglandhypertrophyandmultiplesclerosis(TaoandXin,2014;World HealthOrganization(WHO,2011).Therefore,ensuringnitratelevelsindrinkingwaterremainbelow establishedsafetythresholdsiscriticalforsafeguardingpublichealthintheseregions. Inadditiontotheuseoffertilizerandirrigationmethod,crop,climaticfactorslikerainfallandsoilprop ertiessuchassoiltexture,affectsoilNO_3^-accumulationandleaching(Li*etal*.,2016).

Thelevelofnitrateaccumulationinsoilshasbecomeasignificanthazardtopotablewatersinc e90%offarmingpeopleoftheareaarefrequentlyusingthiswaterfordrinkingpurposesaswellas for irrigation purposes also. TheNO_3ingroundwaterhasbeenenlistedasanemergingissueforgroundwatersafetyandhumanheal th.Insomeareas,ithasbeenreportedsignificantlyhigherthantheprescribedsafeconcentrationsfordr inkingwater.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 datausedinthisstudy is extracted from the literature thatreportedpost-harvestsoilNO_3concentrationsinmaizefieldsinNorthEastalluvialplainsofBihar.

Asperourbestknowledge,nocomprehensivestudyisyetundertakenbyanypreviousresearc hertoexplorethenitrateconcentrationanditspossiblehealthhazardsintheNEalluvialplains,Bihar. MostoftheearlierstudieshadidentifiedNbasedfertilizersasacriticalsourceofnitrateingroundwater .ThisstudyaimedtoinvestigatethegroundwaternitratecontentinNEalluvialplainsofBiharandasso ciatedhealthrisksinhumans beings.Theloadofnitrateingroundwaterofthisregionmayposeaseriousthreattoresidentsastheyrel yongroundwaterforpotablewatersources.Thesurveywasdoneongroundwaterqualityofthestudie dregiontoestimatetheoverallconcentrationofnitrateingroundwater.Sincepartofthestatehashighe rhumanpopulation,thisareacouldbeatriskduetonitratecontaminationinlocalsurfacewaterandgro undwatersources.Lookingintothesefacts,theuniversitydecidedtoestimatethegroundwaternitrate levelintheseareasandcorrelatingthehighnitratecontent(thansafelimitsassuggestedbyBIS)topossi blehumanhealthriskusingahumanhealthriskassessmentmodelasproposedbyUSEPA(1992,1999, 2004).

Map 1



Map 2 : Detailsofsamplingsite

Materialsandmethods

Descriptionofthestudyarea

BiharissituatedintheeasternpartofIndiainbetweenlatitudes24°20'10''Nand27°31'15''Nandlongitudes83°19'50''Eand88°17'40''E.Itisanentirelyland–lockedstate,inasubtropicalregionofthetemperatezone.BiharliesbetweenthehumidWestBengalintheeastandthesubhumidUttarPradeshinthewest,whichprovidesatransitionalpositioninrespectofclimate,economyandculture.ItisboundedbyNepalinthenorthandbyJharkhandinthesouth.GeographicallyBiharplainisdividedintotwounequalhalves(NorthBiharandSouthBihar)bytheriverGangeswhichflowsthroughthemiddlefromwesttoeast.Bihar'slandhasaverageelevationabovesealevelof173feet.Asperagro-climaticzoneitisdividedinACZ-I,II,IIIAandIIIB.

Watersampling depthandgeographicallocationofsamplingsite

AmongdistrictsofAgro-climaticzone-II,fourdistrictscoveringKhagaria,Saharsa,MadhepuraandSupaul were selected.Totalof12villageswereselected (3villagesfromeachdistrict) forwatersampling.GPSbasedsampleswerecollectedforanalysis.ThegroundwaterlevelsinvariousdistrictsofACZ-IIarerecordedbeyond60meterbelowgroundlevel;thisisduetotheoverexploitation.ThegroundwaterlevelinBiharhasdeclineddrasticallyinpastfewdecades.Accordingtoanestimationnet,thedynamicgroundwaterresourcesofthestateare29.19BCM(Billioncubicmeters),andthenetgroundwaterdraftis10.77BCM.ThegeographicallocationofallsamplingsiteispresentedinTable1.

WaterSamplecollection,SamplingprocedureandWater Analysis

TheWatersampleswerecollectedacross12differentsitesinSeemanchaldistrictsandneighbouringKoshiriverregion,whosedepthsvariedfrom6mto18m.Thewatersampleswerefetchedfrom borewells locatedaroundcultivatedlands.Thefreshgroundwatersampleswerecollectedinpre-cleanedsamplebottlesof500mLcapacity.Eachsampleofcollectedbottleswastightlycappedtoavoidleakageandcontaminationduringhandlingandtransportation.Thecontainerswereadequatelylabelledbydate,time,GPScoordinates etc.torecognizeexactsamplingpoint.Allthecollectedsampleswereinitiallypreservedincoldandtransportedtothelaboratorywheretheywerestoredinthefreezerat4°Cuntilusedforfinalchemicalanalysis.Waterqualityparametersanalysedinaccordancetostandardmethodsof (AmericanPublicHealthAssociation(1998))werepH,temperature,conductivity,totalsuspendedsolids(TSS), total dissolved solids (TDS) and nitrate(NO_3^-).

NitrateExposureandHumanHealthRiskAssessment(HHRA)

TheUSEnvironmentalProtectionAgency(USEPA)proposedanHHRAmodelfortheidentificationofhazardandexposure.TheHHRAcanbeestimatedbycalculatingpossibleadverseimpactsofgivencontaminantsoveraspecificperiod.TheHHRAiscomputedusingthevaluesofpresentconcentrationofacontaminantingroundwateranditsexposuredurationstohumans.Theexcessintakeofnitratethroughdrinkingwatercancauseserioushealthhazardsinhumanbeings.Theamountofnitrateinthehumanbodydependsonitsactualconcentrationinwaterandtheintensityofdrinkingday^{-1}kg^{-1}ofbodyweight.Toestimatethehealthhazardsofhighnitratedoseindrinkingwater,theUSEPAmodelwasadoptedwhichwasimplementedinfourdifferentsteps,namely,hazardidentification,doseresponseassessment,exposureassessmentandriskcharacterization(LiandQian2011;Anornu*etal*.2017).Somestudiessuggestthatingestionanddermalcontactastheleadingpathwaysofnitrateexposuresi nhumans(WuandSun2016;Chen*et.al*.2017)butingestionseemstohaveevengreaterriskthandermalcontact.TheexposureofnitratethroughingestionwithdrinkingwateriscalculatedbyfollowingEq.(i)(Zhou*etal*.2016a).

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

whereCDI(chronicdailyintake)istheingestiondosefromdrinkingwater(mg/kg/day),Cistheconcentrationofnitrateestimatedingroundwatersamples(mgL^{-1}),IRistheaveragedailyingestionrateofdrinkingwater(L/day)andthevaluesofIR(2L/dayforadult(male&female),0.78L/dayforchildrenand0.3L/dayforinfants)wereusedforthismodelastakenfrompublishedliterature;EFistheexposurefrequencies(365days/year),EDistheexposureduration(standardexposuredurationinliteratureissuggested40yearsforadult(maleandfemale),12yearsforchildrenand<1forinfants),ABWistheaveragebodyweight(65kgformale,55kgforfemale,20kgforchildrenand8kgforinfants;andAETistheaverageexposuretime(days)whichis14,600daysformaleandfemaleand4380daysforchildrenand365daysforinfants(YadavandGupta,2022).Thepresentstudy focusesonthenon-carcinogenichealthriskofnitratemainlyestimatedbythehazardquotient($HQ_{nitrate}$)values,whichisestimatedthroughfollowingEq.(ii)(USEPA1999;Su*etal*.2017):

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

WhereRfDisreferencedose,RfDindicatesthatreferenceofNO_3^-(1.6mg/kg/d)wereobtainedfromthedatabaseofIntegratedRiskInformationSystem(IRIS)andUSEPA(2001).Thecalculationofhazardquotientvalue,$HQ_{nitrate}$>1isreferredaspotentiallyknowntocausehealthrisksandvaluesof $HQ_{nitrate}$<1indicatesthatitisanacceptablelimitofnon-carcinogenicriskinindividualsduetoingestionofNitratecontaminatedgroundwater.

Table1.ParametersandtheirvaluesusedforHHRAcomputation(USAEPA)

Parameter	Description	Male	Female	Children	Infants
IR	IngestionRate(L/day)	2	2	0.78	0.3
ED	ExposureDuration(years)	40	40	12	≤1
EF	ExposureFrequency(days/year)	365	365	365	365
ABW	AverageBodyWeight(kg)	65	55	20	8
AET	AverageExposureTime(days)	14600	14600	4380	365
RfD	ReferenceDose(mg/kg/d)	1.6	1.6	1.6	1.6

Spatialmappingofsamplingsiteandvulnerablehazardzones

GIS-basedinterpolationtechniquewasusedtorepresentthespatialvariationofhealthriskdistributionofnitrateintakeamongadultsandchildrenacrossthestudyarea.AllthemapswerepreparedusingArcGISsoftware.

Resultanddiscussion

SoilAnalysis/Bulksoilnitrate-nitrogenmeasurements

Soilcharacterisation

The soil inACZ-IIiscomprisedofalluvialdepositionofKoshiriverbasinoccupiedbyclaysandtoloamtypeofsoilwhichcontainskankar,clay,sandparticles,gravels,pebbles,sandstone,etc.Thecharacteristicsofsoilaffecttheinfiltration,percolation,andgroundwaterrechargecapacityoftheregion.Higherrechargeratehashighergroundwatercontaminationpotentialfromsurfacecontaminants.ThesoilatKhagariawasaloamysandcomplex(Sandy,mixed)andreceivedfloodirrigationduringthegrowingseason.Soilsat Saharsa,MadhepuraandSupaulwereFine-loamy,andclayloam(Fine-loamy,mixed)respectively.Thetopographyoftheareaisflat,withpHof8.4inthetopsoilandorganic matterrangingfrom2.18to3.6gkg^{-1}withintheprofiledepthof0to120cm.Thesoiltexturalcharacteristics(Percentsoilfraction)andbulkdensityofstudyareawasrecorded (Fig.1,2,3 and 4).Thesoiltextureandbulkdensityisvaryingdepthwiseindifferentdistricts.ThesoiltexturalclassofKhagariadistrictvariesfromLoam(0-60cm),Clayloam(60-75cm),Sandyloam(75-90cm),Loamysand(90-105cm)andSandyloam(105-120)undervaryingdepth.Whilebulkdensityvariesfrom1.27-1.79(g/cm^3).ThesoiltexturalclassofMadhepuradistrictvariesfromLoam(0-30cm),Sandyclayloam(30-90cm),Sandyloam(90-120).Thebulkdensityvariesfrom1.18-1.58(g/cm^3).ThesoiltexturalclassofselectedvillageinSaharsadistrictvariesfromLoam(0-45cm),Sandyclayloam(45-75cm),Sandyloam(75-120cm).Thebulkdensityvariesfrom1.25-1.52(g/cm^3).ThesoiltexturalclassofselectedvillageinSupauldistrictvariesfromLoam(0-45cm),Sandyclayloam(45-60cm),Sandyloam(60-120cm).Thebulkdensityvariesfrom1.18-1.50(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.

Comment [JRS1]: Tables are made so that readers can easily understand the results of the soil analysis

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

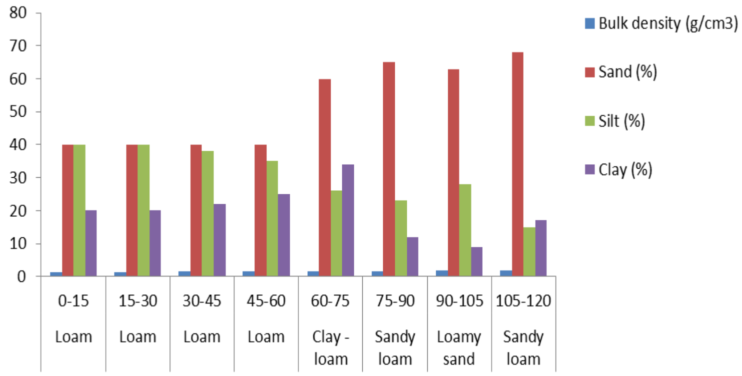


Figure1. Depth-wise soil textural class of Khagaria District

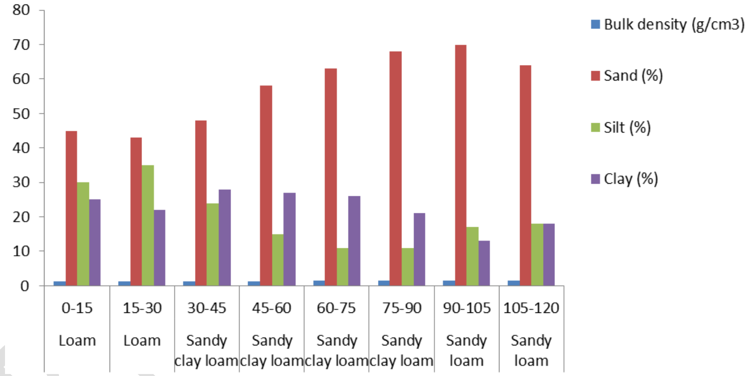


Figure2. Depth-wise soil textural class of Madhepura District

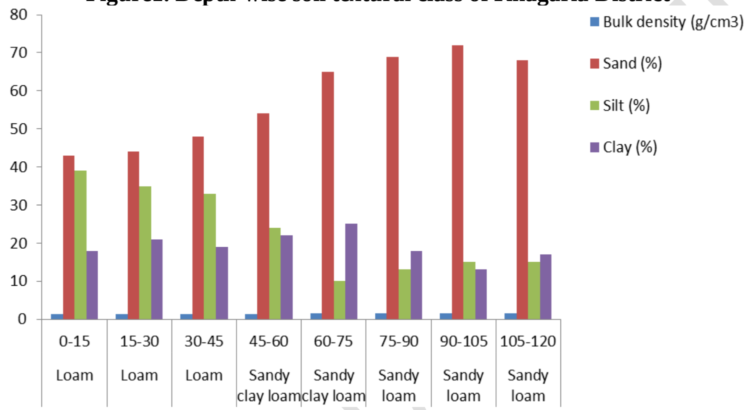


Figure3. Depth-wise soiltexturalclass of Saharsa District

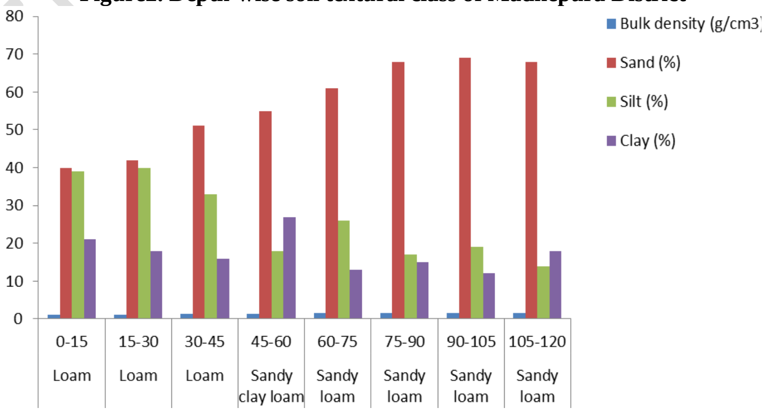


Figure4. Depth-wise soiltexturalclass of Supaul District

VerticaldistributionofnitrateandleachingPercentage

Nitrateleachinginsoil

Total96soilsamplesfromdifferentsoildepthsattheintervalof15cmdepth,sampledup-to105-120cmdepthhavebeencollectedfrom12selectedsites(villages)offourdistricts.Theverticaldistributionofnitrateindicatesaccumulationofnitrateinsoilanditvariedfrom26.73to42.95kg/ha(105-120cmdepth).Theoverallleachingofnitraterangesfrom9.43–12.50%withanaveragevalueof11.02%overapplieddoseofNitrogen.ThehighestleachingwasrecordedinKhagaria(42.95kgN/ha)andminimuminSaharsadistrict(26.73kgN/ha).ThepreliminaryresultindicatedthatoveruseofnitrogenfertilizerhascausedNleachinginsoil.

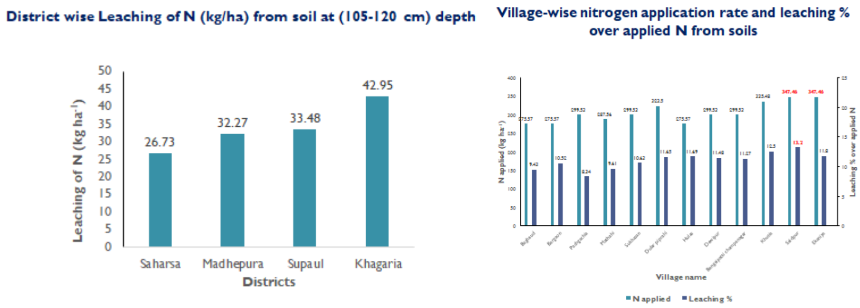


Fig.5:NitrogenapplicationrateandleachingofNitratefromsoilinACZ-II

GeographicallocationofsamplingsiteandWater-depth

ThewatersampleswerecollectedtoinvestigatetheconcentrationofNO$_3$indrinkingwaterfromanintensivelycultivatedbeltofmaizefromtheAgro-climaticzone-IIoftheBihar.

Table.2: Thecoordinates of watersamplingsitesinAgro-climaticzone–IIoftheBihar

SiteNo.	Villagename	Villagestypes	Sampling Depth(m)	NameofDistricts	Coordinates	
					Latitude	Longitude
Site-1	Khutia	CCS	6.09	Khagaria	25.51°N	86.55°E
Site-2	Saidpur	Non-CCS	18.28	Khagaria	25.54°N	86.56°E
Site-3	Ekaniya	Non-CCS	12.19	Khagaria	25.50°N	86.55°E
Site-4	Baghaud	CCS	12.19	Saharsha	25.82°N	86.44°E
Site-5	Bangaon	Non-CCS	15.24	Saharsha	25.73°N	86.82°E
Site-6	Pachgachia	Non-CCS	9.14	Saharsha	25.97°N	86.59°E
Site-7	Mathahi	CCS	9.14	Madhepura	25.54°N	86.72°E
Site-8	Sukhasan	Non-CCS	7.26	Madhepura	25.87°N,	86.78°E
Site-9	Dularpiprahi	Non-CCS	9.14	Madhepura	26.05°N	86.76°E
Site-10	Hulas	CCS	15.24	Supaul	26.25°N,	86.86°E
Site-11	Dewipur	Non-CCS	18.28	Supaul	26.27°N	86.80°E
Site-12	Bengaipattichampanagar	Non-CCS	13.71	Supaul	26.23°N,	86.86°E

*MSL:meansealevel

NitrogenuptakebyMaize

ThegrainyieldofRabimaizeisvaryingfrom6873–9915(kg/ha)inzonewithhighestvalueintheKhagariadistrictandbeinglowestintheSaharsadistrict.Thenitrogencontentingrainrangesfrom1.49to1.63%withmeanvalueof1.54%.Thenitrogenuptakeingrainvaryingfrom105.84to161.62(kgN/ha)withthemeanvalueof123.34(kgN/ha).Similarlythestoveryieldofmaizerangesfrom5413-

7746withanaveragevalueof6266.25kg/ha.Thenitrogen%instoverrangesfrom0.63to0.66%withmeanvalueof0.65%.Thenitrogenuptakeinstovervaryingfrom35.18to50.42(kgN/ha)withthemeanvalueof40.70(kgN/ha).Themaizestoverincludesstalk,leaves,cobsandhusks.Thetotalplantuptakeofnitrogenrangesfrom141.02to212.04kgN/hawithanaveragevalueof164.04kgN/ha.

Table:3.NitrogenuptakeinMaizegrainandStoverinstudiedvillages(Mean of 3 villages)

DistrictsName	**Grain**			**Stover**			**TotalUptake** (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**

*****weightafteradjustmentof12-13%moistureingrain(adjustedgrainyield)

Table:4.MeanN-fertilization;cropN-removal,LeachingofnitrateandcalculatedN-Surplus.

District	**N-fertilization (kg/ha)**	**Totalplant 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
#calculatedasdifferencebetweenNfertilizationandNremoval				

Levelofnitrateinwater

Thegroundandsurfacewaterfrombothcost of cultivation scheme (CCS)andNon-CCSvillagehasbeencollectedandvariouswaterqualityparameterswereanalysed.Thedepthofshallowgroundwatervariedfrom20-60feet.Thenitratelevelingroundwatervaried(3.42-5.27mg/l)andsurfacewater(4.61-5.72mg/l)beingmaximumconcentrationofnitrateinMadhepuradistrict.Thewatersamplehasbeenalsocollectedfromdeepdepthofgroundwaterup-todepthof200-400feetdepthandusedasreferencewatersamplewherenitratelevelvariedfrom0.86to1.05mg/l.

NitrateExposureandHumanHealthRiskAssessment

Groundwaterqualityhasbeensteadilydecliningforrecentdecadesasaresultofnumerouspollutionsourcessuchfertilisersand chemicals.Theingestionofcontaminatedgroundwatercanadverselyaffectthehealthofhumansthroughvarietiesofexposuresincludingdirectingestion,dermalcontact,washing,etc.(USEPA2001).SpatialmapofnitrateconcentrationwasmadeusingGISsoftware(ArcGIS10.7.1)showninfig.2.Nitrateconcentrationinsurfacewater(rangedfrom4.16mg/Lto6.78mg/L)andgroundwatersamples(rangedfrom3.19mg/Lto6.14mg/L)to108mg/L,hasbeenshownintable.8,Regularexposuretonitrate,oneoftheprimarycontaminantsingroundwaterreservoirs,canhaveanegativeimpactonhealthandincreasetheriskofbluebabysyndrome,particularlyincommunitieswithsmallchildren.Hencehealthriskassessmentofnitratehasbeencarriedout.TheChronicDailyIntake(CDI)valuesformale,female,children,andinfantsrangesfrom0.1280to0.2086;0.1512to0.2465;0.1622to0.2644and0.1560to0.2542,respectivelyfornitratecontaminatedsurfacewater(Table.8),whilethisvaluesforallfourgroupofpeoplesrangesfrom0.0981to0.1889;0.1160to0.2232;0.1244to0.2394and0.1196to0.2300respect ivelyfornitratecontaminatedgroundwater(Table.9).SimilarlytheHazardquotient(HQ)valuesfor male,female,children,andinfantsrangesfrom0.0800to0.1303;0.0945to0.1540;0.1014to0.1653and0.0975to0.1589,respectivelyforsurfacewater(table.8),while0.0613to0.1181;0.0725to0.1395; 0.0777to0.1497and0.0740to0.1439,respectivelyforgroundwater(Table.9)intake.HQvaluemore than1indicateshighrisk.ThefindingofdatashowedthatallHQvaluewaslessthan1ofallsamplesinall fourgroups,howeverthedataofHQvaluereachingtowardsunit,soitisgoodtimetobecautionsformaintainthelevelofnitratecontaminationinthestudyareabyadoptingthecertainmitigationoptionsassuggestedintheendofthismanuscript.

Table5:WaterqualityparametersofselectedsiteslocatedinACZ-IIindryseason

Districts	Villagename	Surfacewater				Groundwater				
		pH	Conductivity (µS/cm)	TDS (mg/L)	NO_3^- (mg/L)	SamplingDepth(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
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 (µS/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

TABLE7Nitrateconcentrationinsurfacewaterandtheir ChronicDailyIntake(CDI)andHazardQuotient(HQ)forfourgroups

Location	Latitude	Longitude	NO_3^- (mg/l)	CDI(mg/kg/day) $CDI=\frac{C \times IR \times EF \times ED}{ABW \times AET}$				HQValues $HQ_{nitrate}$=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

TABLE8.NitrateconcentrationinGroundwaterandtheirChronicDailyIntake(CDI)andHazardQuotient(HQ)forfourgroups

Location	Latitude	Longitude	NO_3^- (mg/l)	CDI(mg/kg/day) **CDI=$\frac{C \times IR \times EF \times ED}{ABW \times AET}$**				HQValues **$HQ_{nitrate}$=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:HQRANGEOFSAMPLESFORFOURGROUP

Human	RangeofHQ	Healthrisk	No.ofsamples
Male	>1	Highrisk	0
	<1	Norisk	12
Female	>1	Highrisk	0
	<1	Norisk	12
Children	>1	Highrisk	0
	<1	Norisk	12
Infant	>1	Highrisk	0
	<1	Norisk	12

GroundwatercontaminationwithNO$_3^-$Nandotherparameters

Thegroundwatersampleswereanalysedforimportantcharacteristics(pH,conductivity,TDSandnitratecontaminationlevel),whichindicatessurfaceleachingofcontaminantstoshallowaquifers.TheresultsofpH,conductivity,TDSandnitratecontaminationlevelwereintherangesof6.94-7.93,589.4–826.5(µS/cm),298-509(mg/L),and4.16to6.78mg/L,respectively(Tables6).Theseparametersshowedsomesignificantspatialvariationsamongsamplingsites.TheaverageconcentrationofNO$_3^-$ atvarioussamplinglocationsinshallowaquiferswere4.98,5.85,5.79,4.19,5.11,5.04,5.18,6.78,5.21,4.73,4.16and4.93mg/Latsite-1,site-2,site-3,site-4,site-5,site-6,site-7,site-8,site-9,site-10,site-11,andsite-12,respectively(Table6).Themaximumvaluesatsite-8(6.78mg/L),waslowerthantheBISlimit.TheNO$_3^-$ inthisstudyarearangedbetween4.16to6.78mg/L,whichinthesafersidebutstillitishightimetobesincereregardingloweringofthenitratelevelinthearea.Highintensivedoubleortriplecroppingsystemoftheareahasmostlyutilizedtheleachednitratethroughramifiedrootsystemoftheintercrops.Therewere significantaspatialvariationingroundwaterconcentrationinthisareaindicatessignificantdeviationfromthesitemeanvalues.Thecontentinthemajorityofsiteswassignificantlylowerthantheprescribedsafelimitby WHOandBIS.ThedifferenceinNO$_3^-$ contentatvarioussamplinglocationsmaybeattributedtotheseasonalprecipitationpattern,groundwaterrechargerate,evapotranspirationprocess,etc.OtherfactorsresponsibleforspatialvariationsinNO$_3^-$ contaminationsincludesoilparticlesize,soilwaterholdingcapacity,rainfallintensity,depthofwatertable,aquifermedia,etc.(Buvaneshwarietal.2017;Nakagawaetal.2017;Reetal.2017).Thecroppingpatternssignificantlyaffecttheconsumptionoffertilizers,asreportssuggestthatwheatyieldsrequirehighN-basedfertilizers(Huang2013).Thegeologyofthisareaischaracterizedbyalluvialplainsformedbythefertilesediments,depositedbytheKoshiRiversthatfavourstheextensiveagriculturepracticesinthisregion.Extensiveuseofsyntheticfertilizerstoproducemoreyieldscanhavenegativeimpactsongroundwaterqualitybutstillitisinthesaferside.SeveralreportssuggestthatinBihar,thefertilizerconsumptionrate(perhectare)ishighest(245.25kg)in2019-20closelyfollowedbyPuducherry(244.77)inspiteofitssmallsize,thananyotherstatesinthecountry.Theexcessiveuseoffertilizersincethelast20yearscouldhaveenrichedthelocalsoilsandgroundwaterwithNO$_3$contents.Thefertilizerconsumptioninthestatesincethelast2decadeshasbeenincreaseddrastically.Soilsoftheregionareofsandynaturewithhighporosityandlowwaterholdingcapacitythattendstoleachquickerthesurfacecontaminantstothegroundwater.GroundwaterNO$_3^-$ Nconcentrationswereconsistentlyaroundorlessthan10mg/Lfromthebeginningoftheexperiment,butthengraduallyincreased.ThisclearlyshowsthatNO$_3$leachingwasdisproportionaltotheapplicationsrates.Theresultssuggestthatduringtherainyseasongroundwatermaynotbesuitablefordrinkingpurpose.RechargingroundwaterwithwatercontaininglowerconcentrationofNO$_3^-$ Nwouldbeneededtodilutecontaminatedgroundwater.ThepronouncedincreaseinNO$_3^-$ NconcentrationsinAugust2022wasaccompaniedbyanelevationofthegroundwatertable(datanotsown).NotonlytherewaslesstraveldistanceforNO$_3^-$ Ninthetopsoiltoleachtothegroundwater,butalsoNO$_3^-$ Npresentinthesoilreadilydissolvedinthegroundwater.Indeed,groundwatertabledepthwassignific antlycorrelatedwithgroundwaterNO_3^-N.Onthecontrary,manyscientistsfoundthatwatertabledepthwassignificantlypositivelycorrelated withaveragegroundwaterNO_3^-N.

CONCLUSIONS

Comment [JRS2]: Good

ThisstudyrevealedgroundwaterNO_3contaminationintheareasofNEalluvialplainsofBihar - an area known for its high population density and extensive maize cultivation.ThecombinationofwatermovementthroughthesoilprofileduringtherainyseasontogetherwithhighresidualNO_3^-NfromNfertilizationandshallowgroundwatertablerendersoilsvulnerabletoexcessivenitrateleaching.Thestudysuggeststhesurfaceleachingasaprimesourceofnitratecontaminationingroundwater, which istheonlysourceofpotablewaterformajorityoftheruralpopulationsinthisarea.Thus,consumptionofsuchNO_3contaminatedwatermayposeserioushealthhazardsinresidentsasNO_3isenlistedasanon-carcinogenicchronictoxicantforhumans.Thecarcinogenicandnon-carcinogenichealthriskasestimatedthroughHQnitrateshowedvalues<1inallofsamplingsites,suggestingalowriskofthenon-carcinogenicorcarcinogeniceffectofexcessintakeofNO_3throughthewater.Sampledwaterfromdifferentlocationsshowednitratecontaminationwhichisjustapproachingtothesafelimit.Thereforeitistimetobecautionstorefrainawayfromuseofheavydoseofnitrogenousfertilizerbythefarmersofthestudyarea.FurtherstudiesonactualrecordsofNO_3toxicityinresidentsisneededtovalidatetheresultofpresentfinding.

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Comment [JRS3]: Complete newly added or replaced references, maximum of the last 7-5 years consisting of 15% book and 85% journal

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