

Human Health Risk Assessment of Nitrate and Trace Metals Via Groundwater in Central Bangladesh

Rahman, M. M.^{1*}, Bodrud-Doza, M.², Muhib, M. I.³, Hossain, K. F. B.⁴, Sikder, MT.⁵, Shammi, M.¹, Akter, R.¹, Uddin, M. K.¹

1. Department of Environmental Sciences, Jahangirnagar University, Dhaka-1342, Bangladesh.
2. Climate Change Programme, BRAC, Dhaka 1212, Bangladesh
3. Department of Textile Engineering, City University, Bangladesh
4. Graduate School of Environmental Science, Hokkaido University, Japan
5. Department of Public Health and Informatics, Jahangirnagar University, Dhaka, Bangladesh

Received: 04.10.2019

Accepted: 18.02.2020

ABSTRACT: Groundwater plays a pivotal role as the largest potable water sources in Bangladesh. As agriculture is widely practiced in Bangladesh, potential nitrate (NO_3^-) pollution may occur. Besides, excess amount of arsenic (As) has already been found in groundwater in many parts of Bangladesh including the present study area. Thus, this study was conducted to assess the NO_3^- status along with some trace metals and associated human health risk in the Central Bangladesh. A total of 99 groundwater samples were analyzed to assess human health risk due to high level of NO_3^- and other trace elements i.e. arsenic (As), iron (Fe), and manganese (Mn). Concentration of NO_3^- was determined using column chromatography and inductively coupled plasma optical emission spectrometer (ICP-OES) was used to measure As, Fe and Mn concentrations. It was found that the mean concentration of NO_3^- 253.17 (mg/L) in the groundwater samples exceeds the recommended guideline value by the WHO (50 mg/L). Moreover, this study area also characterized with elevated concentration of As (19.44 $\mu\text{g/L}$), Fe (811.35 $\mu\text{g/L}$), and Mn (455.18 $\mu\text{g/L}$) in the groundwater. Non-carcinogenic human health risk was calculated by justifying HQ (Hazard Quotient) and HI (Hazard Index) and attributed potential conjunctive human health risks due to NO_3^- , As, Fe and Mn in the study area. Child (9.941) is more vulnerable than adult (7.810) considering non-carcinogenic human health risk. Moreover, high carcinogenic risk was found due to As contamination in the groundwater samples and children (1.94×10^{-3}) are more susceptible to carcinogenic risk compared to adults (9.2×10^{-4}).

Keywords: nitrate; arsenic; health risk assessment; hazard quotient; hazard index.

INTRODUCTION

Access to clean drinking water which is the number six of sustainable development goal (SDG) is becoming harder in densely populated countries like Bangladesh.

Groundwater in Bangladesh is the major drinking water sources which can be considered no longer safe due to its quality degradation by anthropogenic activities over the time. In coastal and south-central part of Bangladesh, salinity is a major

* Corresponding Author, Email: rahmanmm@juniv.edu

problem in the groundwater (Rahman et al., 2017), where in many other districts' groundwater is laden with different types of trace elements. The presence of trace elements such as arsenic (As), iron (Fe), manganese (Mn) and nitrate (NO_3^-) in ground water is predominantly influenced by natural and anthropogenic sources (Sharma et al., 2014, Rahman et al., 2016).

Nitrate is an essential plant nutrient (Messier et al., 2019). It is one of the main pollutants in agriculturally impacted groundwater systems (Wild et al., 2018) and another major environmental pollution stressor (Tavakoly et al., 2019) that has stimulated significant research interest (Huno et al., 2018). Agricultural land use affects groundwater quality from two points of view: salinization is one of the major concerns within irrigated areas, especially in arid regions (Nasrabadi and Maedeh, 2013). Leaching of nitrate from animal wastes and fertilizers at agricultural operations can result in nitrate contamination of groundwater, lakes, and streams (Bourke et al., 2019). The source of nitrate (NO_3^-) in groundwater includes surface leaching from wastewater and waste dump sites, animal excreta disposal, industrial effluents (Ahada and Suthar, 2018; Taufiq et al., 2019), population growth (Taufiq et al., 2019), poor sewerage, human excreta leakage from septic tanks (Ahamad et al., 2018), poorly maintained disposal of solid waste locally, N-based fertilizers from agricultural activities, wastewater irrigation and irrigation runoff (Ahada and Suthar, 2018; Ahamad et al., 2018).

Nitrate concentrations exceeding the 50 mg/L limit established for drinking water pose the human health at risk (Biddau et al., 2019). High nitrate concentrations were found in groundwater both within the wastewater treatment plant site and surrounding market garden farms (maximum of 99 mg/L and 78 mg/L nitrate as N, respectively) (Adebowale et al., 2019). Moreover, in many developing

countries nitrate concentrations up to 250 mg L^{-1} in groundwater have been reported (Ahada and Suthar, 2018; Ahamad et al., 2018; Marques Arsénio et al., 2018; Qasemi et al., 2018; Wagh et al., 2019; Zhang et al., 2018) due to the widespread use of latrines and septic tanks that allow for constant infiltration of its content into the soil and eventually to groundwater sources (Marques Arsénio et al., 2018) as well as intensive agriculture activities.

Groundwater hazard assessments involve many activities dealing with the impacts of pollution on groundwater, such as human health studies and environment modelling. Nitrate contamination is considered a hazard to human health, environment and ecosystem (Rizeei et al., 2018). Ingestion of nitrate can lead to the endogenous formation of N-nitroso-compounds, which are known human carcinogens (Messier et al., 2019). Long term consumption of excessive NO_3^- can create cancer risk on human body due to formation of nitrosamines (Pannala et al., 2003). It has also been known to create spontaneous absorptions, birth defects, respiratory tract infections and changes in immune system (Lohumi et al., 2004; Fewtrell et al., 2004; Greer and Shannon, 2005; Ward et al., 2005; Rachid et al., 2006; Ma et al., 2007). Subsequently, health risk assessment of these contaminants for predicting the health hazard is very important. According to United Greer and Shannon States Environment Protection Agency (USEPA), human health risk assessment (HHRA) is the process of estimating the nature and probability of adverse health effects in humans who may be susceptible to chemicals in contaminated environmental media, now or soon (Momot et al., 2005). Nitrate associated HHRA for both adult and children has been calculated in many studies (Ahada and Suthar, 2018; Li et al., 2019; Paladino et al., 2018; Wagh et al., 2019). Mostly four steps must be followed

to fulfill the health risk assessment e.g. identification of hazard, dose-response relationship, exposure assessment as well as risk characterization (Wu et al., 2010). Thus, human health risk assessment is an effective way to justify health risk levels posed by various contaminants (Bortey-Sam et al., 2015).

HHRA study of trace elements present in the groundwater is very limited numbers in the central Bangladesh. Previously, Ahmed et al. (2018) reported that the groundwater in Sylhet, south-eastern part of Bangladesh exceeds the allowable limits of Fe, Mn and As concentrations. Arsenic is considered as a dangerous contaminant with adverse effects on human health. It is also highly restricted by international environmental standards (Mehrdadi, et al., 2019). However, relatively higher concentration of Fe and Mn were found in deep water samples and reverse trend was found in case of As. Rahman et al. (2017) reported HHRA of groundwater used in Gopalganj district (Rahman et al., 2017), Joseph et al. on arsenic exposure for Bangladeshi adults (Joseph et al., 2015). Rahman et al. (2016) reported the status of As contamination in Singair Upazila, Manikganj District and geogenic factors were found to be the major sources of As in the groundwater in that area (Rahman et al., 2016). A study in Iran shows that, the concentration of arsenic increases from upstream areas towards the downstream estuarine zone with a substantial rise in the central part (Nasrabadi, et. al., 2015; Nasrabadi, et. al., 2013). Considering the above-mentioned factors, we hypothesized that the level of contaminants existing in the groundwater such as NO_3^- and trace metals might be higher than acceptable level and could be major risk factor for human health. Therefore, the aim of this study was to determine the level of NO_3^- along with As, Fe and Mn in groundwater and to evaluate the health risk associated with exposure to these contaminants *via*

oral ingestion route. This research may be the first attempt for assessing the health risk associated with NO_3^- and trace metals in the groundwater of central Bangladesh.

MATERIALS AND METHODS

The study area of Singair, Manikganj district is located in the central part of Bangladesh. Geographically it lies between 23.8167° N and 90.1500° E longitudes (Fig. 1). The total area of the district is 217.38 km^2 with the population of 231,628 (BBS 2011). The study area is mainly surrounded by Kaliganga and Dhaleshwari river floodplain. The Dhaleshwari is the main river, which is bounded with the north and east portion of the study area and Kaliganga River flows in the southern part. The study area lies in the tropical monsoon climatic zone, which is subjected to tropical climate and is characterized by the moderate to high temperature (34.5°C), heavy rainfall and often with excessive humidity. Maximum rainfall occurs during May to October in summer. Climate is one of the most important factors for the occurrence and movement of groundwater.

Topographically, the study area is flat. The geology of the study area can be attributed to Quaternary alluvial sequence, which is a part of Ganges – Jamuna flood plain. The north and western part of the area is covered by the Pleistocene sediments, which are known as Madhupur clay. In general, the stratigraphic sequence is fining upward sequence i.e. from gravels to cobbles, pebbles, sand and finally silts and clays; micas are abundance in whole sequence in Pliocene- Pleistocene-Holocene time (Islam et al. 2018).

Total 99 groundwater samples were collected from different depth depending on the tube-well bore depth (m) in the central Bangladesh (Figure 1). Sampling time was August 2011 and analyses were performed within 1 month from the sampling time. Before taking the sample, the tube-well was pumped for around 10 minutes. All the

collected samples were stored in 250 ml polypropylene plastic bottles with (for trace metal)/without (NO_3^-) acidification by 2 drops of concentrated HNO_3 to prevent any precipitations. The inductively coupled plasma (ICP-OES) optical emission spectrometer was used (SII NanoTechnology inc.) for the determination of As, Fe, and Mn as depicted elsewhere (Rahman et al., 2016).

In the trace metal analysis the quality control was maintained in all cases through strictly following the instruction manuals and the method precision was more than 95% in confidence interval. The methods

were recalibrated after running 15 samples and all analyses were performed at least in triplicate to ensure data precision. Overall data reproducibility for nitrate was within $\pm 10\%$ error levels. Nitrate (NO_3^-) was determined by chromatography method (DX-120, Dionex, USA) using the columns "Ion Pac AS12A" (Dionex, Abbreviation of state, USA). Samples were filtered with glass fiber filters with $0.45 \mu\text{m}$ pore. All the samples were analyzed triplicate for ensuring reproducibility and statistical validity. Statistical analysis was done by MS-excel 2007.

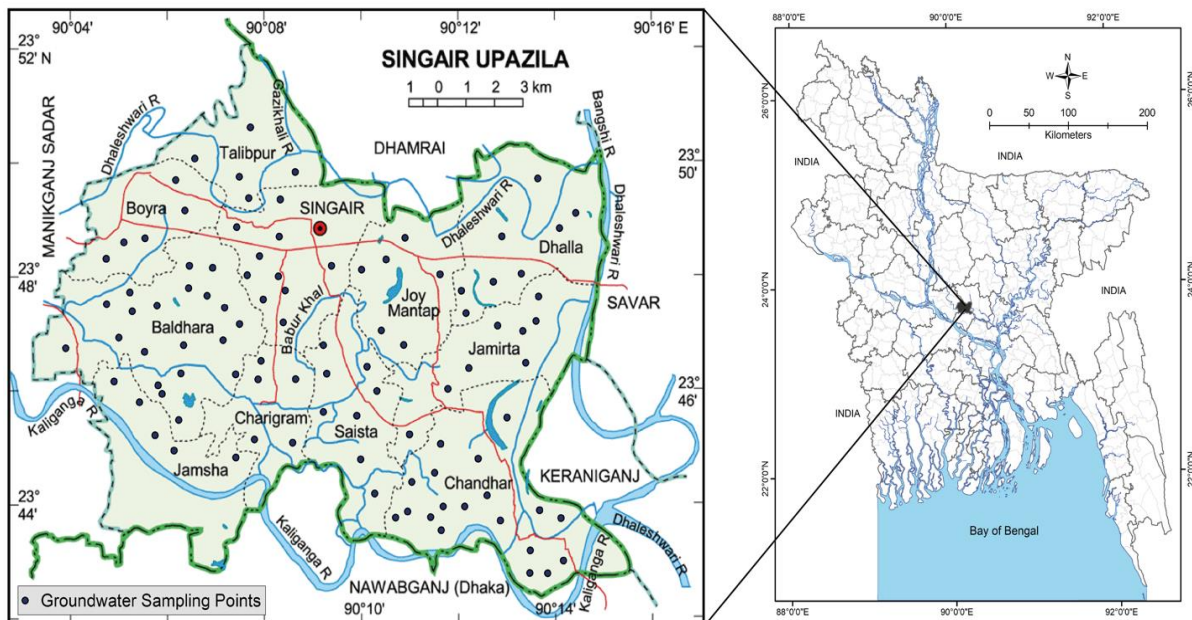


Fig. 1. Map of the study area, singair, Manikganj.

Risk assessment is the process of estimating the occurrence probability of any given magnitude of adverse health effects over a specified time period (Bortey-Sam et al., 2015). The health risk assessment of each element is based on evaluation of the risk level and is expressed in terms of a carcinogenic or non-carcinogenic health risk (USEPA, 2009). Slope factor (SF) for carcinogen risk characterization and the reference dose (RfD) for non-carcinogen risk characterization were also evaluated (Lim et al., 2008). Oral ingestion pathway was

considered for the study (Ning et al., 2011). Below equation was adopted to calculate the chronic daily intake for oral and dermal adsorption pathways (Giri and Singh, 2015; Wongsasuluk et al., 2013; USEPA, 1989; USEPA, 1993; ECETOC, 2001).

$$CDI_{Oral} = \frac{(CW \times IR \times EF \times ED)}{(BW \times AT)} \quad (1)$$

Here, CDI = Chronic daily intake ($\mu\text{g}/\text{Kg}/\text{day}$) CW = Concentration of trace metal in water ($\mu\text{g}/\text{L}$), IR = Ingestion rate (L/day; 2.2 for adult 1 for Child), EF =

Exposure frequency (Days/year, 365), ED = Exposure duration [Year; for oral = 70 for Adult, 10 for Child], BW = Body weight (Kg; 70 kg for Adult, 25 kg for Child), AT = Average time (Days; 25,550 for Adult, 3650 for Child) (USEPA, 2009; USEPA, 1993; ECETOC, 2001; USEPA, 2001; USEPA, 1999; Weyer et al., 2001; Kavcar et al., 2009).

The health risk due to groundwater consumption was justified on the basis of chronic (noncarcinogenic) and carcinogenic effects. The noncarcinogenic risk was calculated as Hazard quotient (HQ) by the following equation-

$$HQ = \frac{CDI}{RfD} \quad (2)$$

Where HQ is hazard quotient (unitless) and RfD ($\mu\text{g}/\text{Kg}/\text{day}$) originates from risk-based concentration table (USEPA, 1993).

For elemental risk assessment, the individual HQs are combined to form Hazard Index (HI). If the value of HQ and HI exceeds 1, there could be potential noncarcinogenic effects on health while HI less than 1 indicates the no risk of health effects (ECETOC, 2001; USEPA, 2001).

$$HI = HQ_1 + HQ_2 + \dots + HQ_n \quad (3)$$

The carcinogenic risk was measured from the calculation of CDI_{oral} ($\text{mg}/\text{kg}/\text{day}$) and Slope Factor (SF) ($\text{mg}/\text{kg}/\text{day}$)⁻¹ and a range of characterization has been provided in Table 1 chronic and cancer risk assessment (Kundu et al., 2008).

Table 1. Scales for chronic and carcinogenic risk assessment (USEPA, 1999; Bortey-Sam et al., 2015).

Risk level	HQ or HI	Chronic risk	Calculated cases of cancer occurrence	Cancer risk
1	< 0.1	Negligible	< 1 per 1000,000 inhabitants (10^{-6})	Very low
2	$\geq 0.1 < 1$	Low	> 1 per 1000,000 inhabitants (10^{-6}) < 1 per 100,000 inhabitants (10^{-5})	Low
3	$\geq 1 < 4$	Medium	> 1 per 100,000 inhabitants (10^{-5}) < 1 per 10,000 inhabitants (10^{-4})	Medium
4	≥ 4	High	> 1 per 10,000 inhabitants (10^{-4}) < 1 per 1000 inhabitants (10^{-3})	High
5			> 1 per 1000 inhabitants (10^{-3})	Very high

Table 2. Descriptive Statistical Analysis of the groundwater quality data (n=99).

Parameters	Minimum	Maximum	Mean	Std. Deviation	Coefficient of variance	Bangladesh Standard (1997)	Indian standards (2012)		WHO (2011)
							Acceptable Limit	Permissible Limit	
Well Depth (m)	10.67	82.3	24.322	11.825	48.617				
As ($\mu\text{g}/\text{L}$)	BDL	113	19.444	24.756	127.319	50	10	50	10
Fe ($\mu\text{g}/\text{L}$)	BDL	19296	811.35	2747.7	338.651	300-1000	300	No relaxation	No proposed value
Mn ($\mu\text{g}/\text{L}$)	54	2716	455.18	394.12	86.585	100	100	300	500
NO_3^- (mg/L)	BDL	708.11	253.18	168.8	66.674	10	45	No relaxation	50

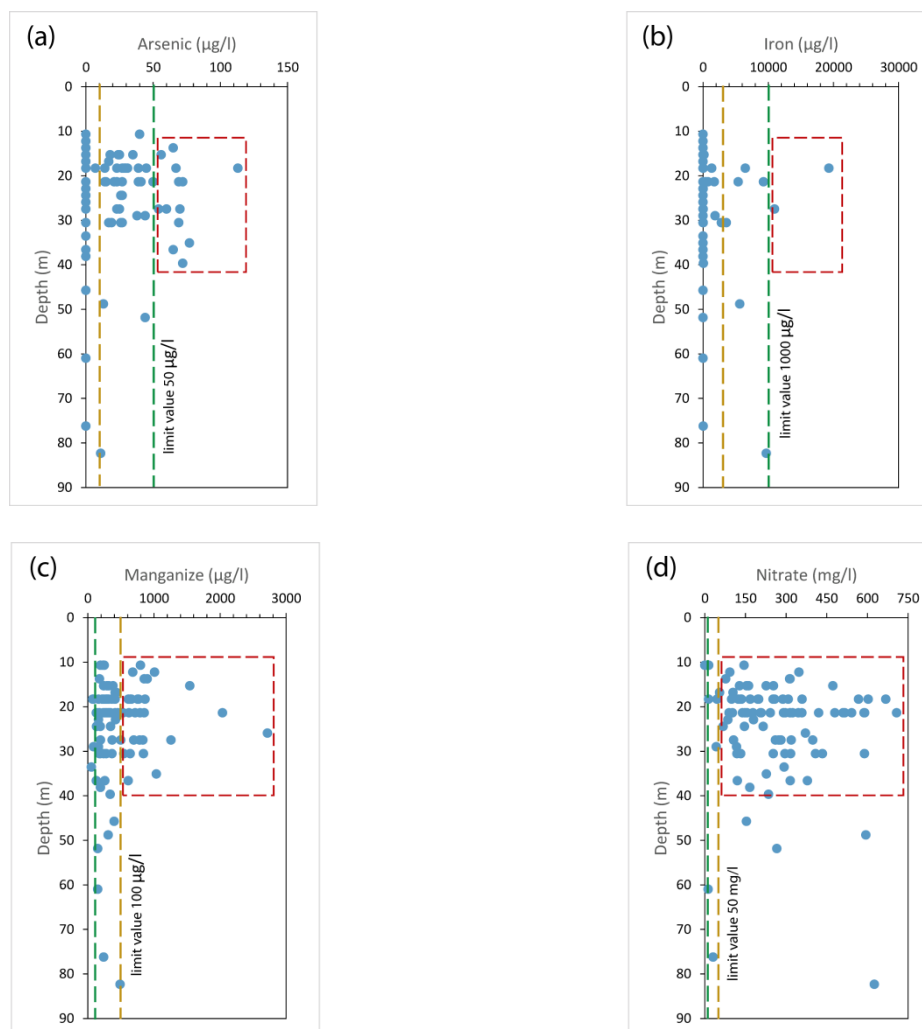


Fig. 2. Depth wise water analysis. Dotted line representing recommended/limit value and the red dotted box refers the clusters of sampling points exceeding limit value for each of the parameters limited to 40 m depth.

RESULTS

The average depth wise results of 99 ground water samples for As, Fe, Mn and NO_3^- are provided in Table 1. The mean concentrations of As, Fe, Mn and NO_3^- were also compared in Table 2 with the BDWS (1997), Indian Standards (2012) and WHO (2011). Statistical analysis from Table 2 represented that mean concentration of NO_3^- (253.175 mg/L) exceeded the standard concentration of Bangladesh Drinking Water Standard (BDWS 1997), India (2012) and WHO (2011). Again, the mean concentration of Mn (455.181 µg/L) was found to be greater than BDWS (1997) and Indian Standards

(2012) but lesser than WHO (2011). Though the mean concentration for Fe (811.253 µg/L) was within the range of BDWS (1997), it crossed the parameters standardized by India (2012) and WHO (2011). The mean concentration of As (19.444 µg/L) did not exceed the limit for BDWS (1997) and permissible limit of Indian Standards (2012) but break the concentration limit given by acceptable limit of Indian Standards (2012) and WHO (2011).

Elemental depth wise distribution was also illustrated in terms of depth (m) vs. elemental concentrations (µg/L) [Figure 2(a-d)]. Figure 2d suggested that

approximately 87.89% of groundwater samples exceeded all the standard limit values of NO₃⁻ in the depth range between 10m-40m. Figure 2c, 2a and 2b also

showed that approximately 21.21% groundwater sample for Mn, 13.13% for As and 2.02% for Fe crossed the standard limit values in the same depth range.

Table 3. Summary of HQ and HI of As, Fe, Mn and NO₃⁻ and Carcinogenic risk of Arsenic for oral ingestion in groundwater samples (average of 99 groundwater samples)

Health Risk	Inhabitants	HQ for As	HQ for Fe	HQ for Mn	HQ for NO ₃ ⁻	Hazard index (HI)	Non-Carcinogenic Risk
Non-carcinogenic risk	Adult	2.037	0.085	0.715	4.973	7.810	High
	Child	2.593	0.108	0.910	6.329	9.941	High
Carcinogenic risk (CR) of As							Carcinogenic Risk
Carcinogenic risk	Adult					9.2x10 ⁻⁴	High
	Child					1.17x10 ⁻³	Very High

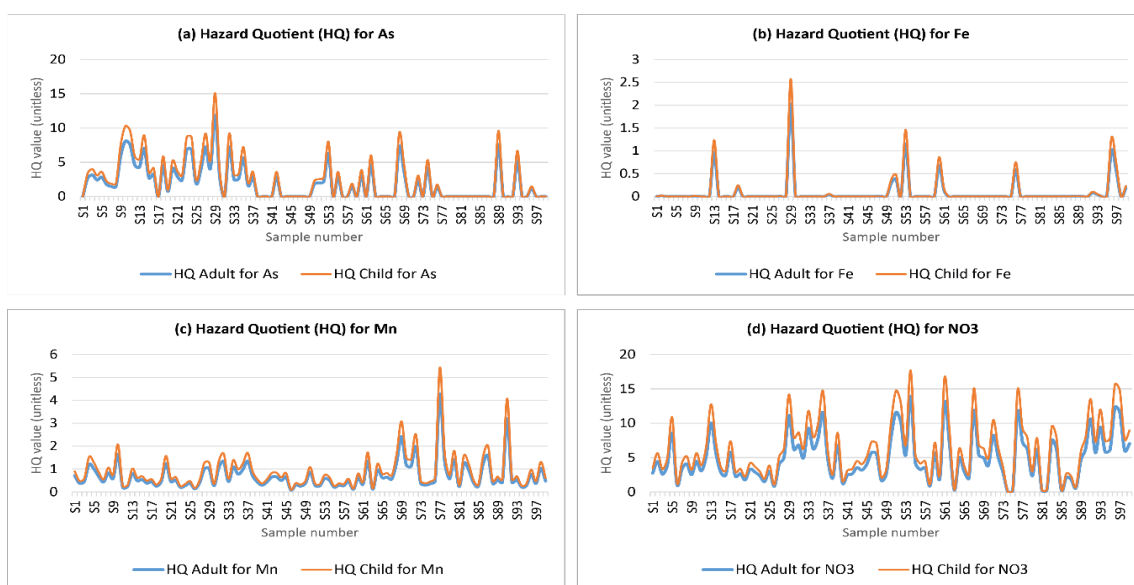


Fig. 3. Hazard Quotient (HQ) oral ingestion pathway for As, Fe, Mn and NO₃⁻. Water sample identification number and HQ are plotted in X and Y axis respectively.

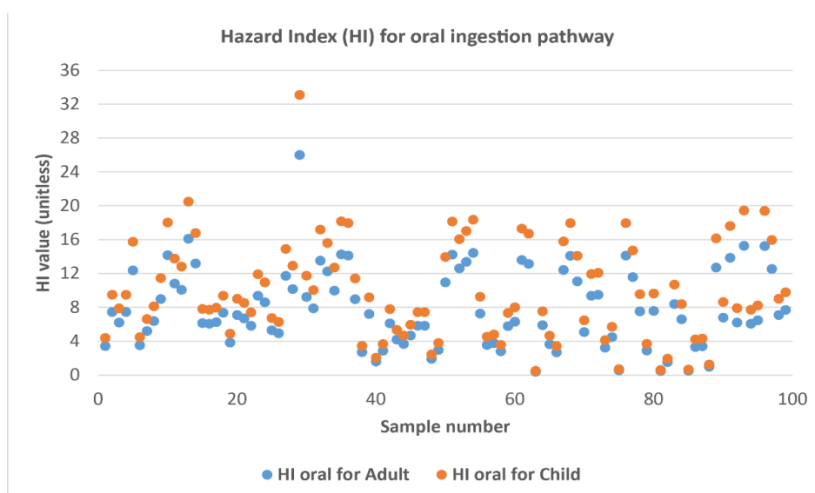


Fig. 4. Hazard Index (HI) for oral ingestion pathway for children and adults in the study area.

Above described table 2 and table 3 already have shown that the groundwater of this area is polluted with nitrates and trace metals. To evaluate the impact on human health related to this pollution, establishing Hazard Quotient (HQ) with related Hazard index (HI) is an important tool to calculate non-carcinogenic health risk. In this study, HQ and HI of As, Fe, Mn and NO₃ for 99 ground water samples are illustrated in Figure 2 and 3. Considering average of these elements, both oral as well as dermal adsorption risks were identified in terms of non-carcinogenic risk measurement for both child and adult generation (Table 3). Oral ingestion pathway for adult and child showed high HQ level in As, Fe, Mn and NO₃⁻. Considering oral ingestion of As, Fe, Mn and NO₃⁻, HQ values for adults were 2.037, 0.085, 0.715 and 4.973; whereas, HQ values for child were 4.321, 0.180, 1.517 and 10.549, respectively. The following order was found in respect of HQ for adults in oral ingestion NO₃>As>Mn>Fe. For child the order was NO₃>As>Mn>Fe in oral ingestion. HI calculation also done for adult and child. For adult, about 75.76% samples showed high non-carcinogenic risk while

19.19% samples showed medium non-carcinogenic risk. For child, about 83.84% samples showed high non-carcinogenic risk while 11.11% samples showed medium non-carcinogenic risk. Low non-carcinogenic health effect (5.05% samples) was shown in both adult and child. Average hazard index result for adult and child also placed in Table 3. Results showed that both adult (HI=7.810) and child (HI=9.941) showed high non-carcinogenic risk effect.

Cancer risk on adult and child due to As for both oral ingestion pathway was illustrated in Figure 4 and 5 and the calculated result placed in Table 3. Considering oral ingestion pathway, high (9.2×10^{-4}) and very high (1.94×10^{-3}) carcinogenic risk was justified for adult and child respectively. Out of 99 samples for adult, 39.39% samples exhibited very high carcinogenic risk and 12.12% samples showed high carcinogenic risk. Moreover, for children, 42.42% samples possessed very high carcinogenic risk while 9.09% samples showed high carcinogenic risk. For both adult and children about 48.48% samples seemed to be not harmful.

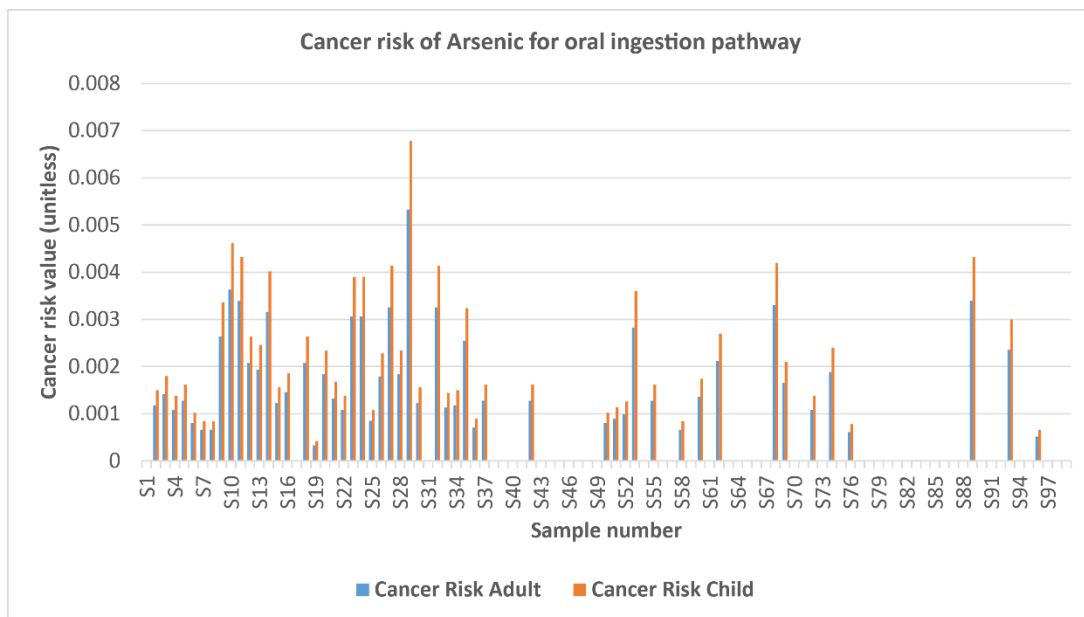


Fig. 5. Cancer Risk (HI) for oral ingestion pathway for children and adults in the study area.

DISCUSSION

In groundwater management, the hazard should be assessed before any action can be taken, particularly for groundwater pollution and water quality. Thus, pollution due to the presence of nitrate poses considerable hazard to drinking water, and excessive nutrient loads deteriorate the ecosystem (Rizeei et al., 2018). The possible health hazards of high nitrate intake were estimated using USEPA human health risk assessment (HHRA) model for both adult and children. Results of this study suggested the chronic daily intake (CDI) in the ranges of 1.09–5.65 and 2.56–13.20 in adult and children population of this region, respectively. The hazard quotient (HQ_{nitrate}) value was > 1 in most sampling locations ranging 1.09–5.65 for the adult and 2.56–13.20 for children population of Malwa (Ahada and Suthar, 2018). Consumption of high NO_3^- containing water may pose serious health hazard especially in children (< 5 years) (Ahada and Suthar, 2018). In general, shallow unconfined aquifer is more vulnerable to NO_3^- contamination compared to the deep confined aquifer because denitrification partly occurs in deep anoxic aquifer and this led attenuation of NO_3^- pollution in groundwater flowing (Taufiq et al., 2019). The availability and reactivity of electron donors control the prevalent redox conditions in aquifers and past nitrate contamination of groundwater can be ameliorated if denitrification occurs (Wild et al., 2018).

Water quality parameters reflect the level of contamination in water resources. In this study, the quality of groundwater in terms of nitrate (NO_3^-), arsenic (As), iron (Fe) and manganese (Mn) varied drastically among different sampling sites in central Bangladesh. The results have shown that the mean value of NO_3^- (253.175 $\mu\text{g/L}$) exceeded Bangladesh standard (BDWS, 1997), Indian Standards (2012) and WHO standard (2011) (Table 2). Both the geogenic and anthropogenic sources can be the contributor of elevated level of NO_3^- in

groundwater of the study area e.g. application of nitrogenous fertilizer or manure slurries in agriculture and aquaculture, poor soil profile, and irrigation mechanisms. Relatively little of the NO_3^- found in natural waters is of mineral origin, most coming from organic and inorganic sources, the former including waste discharges and the latter comprising chiefly artificial fertilizers. The higher occurrence of NO_3^- in shallow depth groundwater was likely due to anthropogenic activities e.g. excessive use of agrochemicals and unmanaged irrigation system that could cause microbial mineralization in the groundwater (Sharma et al., 2014). Alongside, anthropogenic activities like sewage effluents, unsewered sanitation, and unplanned disposal of solid waste in densely populated areas may also leached NO_3^- into groundwater. In Maputo, the capital of Mozambique, nitrate concentrations above 250 mg L^{-1} in groundwater have been reported. This happens due to the widespread use of latrines and septic tanks that allow for constant infiltration of its content into the soil and eventually to groundwater sources (Marques Arsénio et al., 2018). In addition, manure represents one of the main nitrate sources in groundwater from agriculture, the other being synthetic fertilizers (Martinelli et al., 2018).

Nitrogenous fertilizers rapidly convert into NO_3^- in soils, are highly soluble and hence easily leachable to deep soil layers and ultimately enter into shallow aquifers (Kundu et al., 2008a). For instance, sandy or sandy clay soil with high coarse texture have high water filtration rate and possibly contributing in nitrate leaching to underground waters (Suthara et al., 2014). Moreover, extensive agricultural practices in the study area may contribute in NO_3^- leaching in aquifers of this area (Zhai et al., 2017). Similar NO_3^- concentration increase over the acceptable limit was reported by Majumder et al. (2008) in shallow groundwater of central-west region of

Bangladesh. This excess NO_3^- contamination in drinking water can cause increased cancer risk (Suthara et al., 2009; Shukla and Saxena, 2018) as well as various health risks such as methemoglobinemia, diabetes, etc. on humans and to some extent on livestock populations as well (Shukla and Saxena, 2018).

Although, NO_3^- itself is not a direct toxicant but is a health hazard because of its conversion to nitrite. Nitrate itself is not harmful, but in human gastrointestinal tract it can be endogenously reduced to toxic nitrite through nitrosation in the stomach with amines and amides to form various types of N-nitroso compounds (NOCs) [(Qasemi et al., 2018) and the references there in]. The results of this study thus indicate that groundwater from the 10 m to 40 m depth in the study area is severely polluted with NO_3^- . Furthermore, the HQ and HI value of NO_3^- implicated for potential non-carcinogenic health risk in the study area both for adult and for children (Table 3 and Figure 3). Dellavalle et al. (2014) suggested that high dietary nitrate intake expected to have higher exposure to endogenously formed NOCs and increases risk of colorectal cancer. Moreover, our result has been shown that children health risk due to increased NO_3^- concentration in drinking water is higher than that of adults. Similar results were reported by Zhai et al. (2017) in China, Iran (Qasemi et al., 2018), and India (Wagh et al., 2019)

The study area is also characterized with elevated levels of trace metals (As, Fe, and Mn) in the groundwater. The mean concentration of all three metals is exceeds the limit of WHO standards (2011). The major potential sources of these trace metals are geogenic (Rahman et al., 2016; Das et al., 2009). The mean value of arsenic (19.44 $\mu\text{g/L}$) lies within the range of BDWS (1997) standard and permissible limit of Indian Standards (2012) but exceeded the concentration limit given by

acceptable limit of WHO standard (2011). This occurrence of As, Mn and Fe in the shallow depth water might be attributed to the geogenic formation of the Bengal Basin (Rahman et al., 2016) as well as from anthropogenic activities (Kundu et al. 2008). Arsenic in groundwater is associated with skin damage, increased risk of cancer, and problems with circulatory system (Bodek et al., 1988). This study shows that carcinogenic risk of arsenic for oral ingestion from groundwater sample is high for child than the adults. Similar high carcinogenic risk for adult and child were also found by Rahman et al. (2017) in ground water samples of Gopalganj of Bangladesh, which is located southern side of our sampling area. Moreover, Chen et al. (2011) reported that exposure to arsenic in drinking water is adversely associated with mortality from heart disease. Skin lesions caused by As contaminated drinking water have already been reported among adults and children in Bangladesh (Das et al., 2009). However, Sohel et al. (2009) stated that increased risks at low level exposure of As (50–149 $\mu\text{g/L}$) were observed for death due to cancers, cardiovascular disease, and infectious diseases in Bangladesh. However, the HQ for Fe (children; 0.108) and Mn (children; 0.910) are relatively very lower compared to NO_3^- and As in the study area. Though the mean concentration of Fe and Mn is quite high but the calculated HQ is within the safe limit for both adult and children in the study area. Thus the study shows that the shallow aquifer groundwater of central is polluted by NO_3^- and As with substantial human health risk.

CONCLUSION

The outcome of the study depicts the human health risk of NO_3^- and As by analyzing 99 ground water samples from the Central Bangladesh. The elevated occurrence of NO_3^- and As is predominant within the 10 to 40 m depth zone of tubewell. Furthermore,

HQ values of NO_3^- and As are the dominant contributors in HI for the non-carcinogenic health risk for both adults and children. Additionally, carcinogenic risk due to As is also high in the study area. However, the Mn and Fe possess lower HI compared to NO_3^- and As. Finally, the shallow tube-well is not safe for drinking water collection in the study area. However, further study warrants considering wider area with spatial and temporal variability to predict health risk more precisely including the human level direct detection of the NO_3^- and As. Moreover, it is immensely important to regulate the use of nitrogen complex fertilizer, cattle manure, household waste management, landfill leachate, septic tanks for proper groundwater management practices to prevent the associated risks to human health.

ACKNOWLEDGEMENTS

Authors are grateful to Dr. Masahiro Maruo and Dr. Tanvir Ahmed from the Department of Ecosystem Studies, the University of Shiga Prefecture, Japan for analytical supports.

GRANT SUPPORT DETAILS

The present research did not receive any financial support.

CONFLICTS OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

REFERENCES

Adebowale, T., Surapaneni, A., Faulkner, D., McCance, W., Wang, S. and Currell, M. (2019). Delineation of contaminant sources and

denitrification using isotopes of nitrate near a wastewater treatment plant in peri-urban settings. *Sci. Total Environ.* 2701-2711. doi:10.1016/j.scitotenv.2018.10.146

Ahada, C.P.S. and Suthar, S. (2018). Groundwater nitrate contamination and associated human health risk assessment in southern districts of Punjab, India. *Environ. Sci. Pollut. Res. Int.* 25, 25336-25347. doi:10.1007/s11356-018-2581-2

Ahamad, A., Madhav, S., Singh, P., Pandey, J. and Khan, A.H. (2018). Assessment of groundwater quality with special emphasis on nitrate contamination in parts of Varanasi City, Uttar Pradesh, India. *Appl. Water Sci.* 8(4). doi:10.1007/s13201-018-0759-x

Biddau, R., Cidu, R., Da Pelo, S., Carletti, A., Ghiglieri, G. and Pittalis, D. (2019). Source and fate of nitrate in contaminated groundwater systems: Assessing spatial and temporal variations by hydrogeochemistry and multiple stable isotope tools. *Sci. Total Environ.*, 647, 1121-1136. doi:10.1016/j.scitotenv.2018.08.007

Bodek, I., Lyman, W. J., Reehl, W. F. and Rosenblatt, D. H. (1988). *Environmental Inorganic Chemistry: Properties, Processes and Estimation Methods*, Pergamon Press, Elmsford, NY, USA.

Bortey-Sam, N., Nakayama, S.M., Ikenaka, Y., Akoto, O., Baidoo, E., Mizukawa, H. and Ishizuka, M. (2015). Health risk assessment of heavy metals and metalloid in drinking water from communities near gold mines in Tarkwa, Ghana. *Environ. Monit. Assess.* 187, 397. DOI 10.1007/s10661-015-4630-3

Chen, Y., Graziano, J.H., Parvez, F., Liu, M., Slavkovich, V., Kalra, T., Argos, M., Islam, T., Ahmed, A., Rakibuz-Zaman, M., Hasan, R., Sarwar, G., Levy, D., Geen, A.V. and Ahsan, H. (2011). Arsenic exposure from drinking water and mortality from cardiovascular disease in Bangladesh: prospective cohort study. *BMJ*, 342, d2431

Das, B., Rahman, M.M., Nayak, B., Pal, A., Chowdhury, U.K., Mukherjee, S.C., Saha, K.C., Pati, S., Quamruzzaman, Q. and Chakraborti, D. (2009). Groundwater arsenic contamination, its health effects and approach for mitigation in West Bengal, India and Bangladesh. *Water Qual. Expo. Health* 1, 5–21. DOI.10.1007/s12403-008-0002-3

Dellavalle, C.T., Xiao, Q., Yang, G., Shu, X.O., Aschebrook-Kilfoy, B., Zheng, W., Lan, L.H., Ji, B.T., Rothman, N., Chow, W.H., Gao, Y.T. and Ward, M.H. (2014). Dietary nitrate and nitrite intake and risk of colorectal cancer in the Shanghai Women's Health Study. *Int. J. Cancer.* 134(12), 2917-2926. Doi: 10.1002/ijc.28612.

- ECETOC (European Center for Ecotoxicology of Chemical) 2001. Aquatic Toxicity of Mixtures. Technical Report. 80, Brussels.
- Fewtrell, L. (2004). Drinking-water nitrate, methemoglobinemia, and global burden of disease: A discussion. *Environ. Health Perspect.* 112, 1371–1374.
- Giri, S. and Singh, A.K. (2015). Human health risk assessment via drinking water pathway due to metal contamination in the ground water of Subarnarekha river basin, India. *Environ. Monit. Assess.* 187, 63.
- Greer, F.R. and Shannon, M (2005). Infant methemoglobinemia: The role of dietary nitrate in food and water. *Paediatrics.*, 116, 784–786.
- Huno, S.K.M., Rene, E.R., van Hullebusch, E.D. and Annachhatre, A.P. (2018). Nitrate removal from groundwater: a review of natural and engineered processes. *J. Water Supply Res. T.* 67(8), 885-902. doi:10.2166/aqua.2018.194
- Islam, M., Bashar, K., Ahmed, N., Rasul, M. G., Hossain, S. and Sarker, M.M. (2018). Hydrogeologic Characteristics and Groundwater Potentiality of Lower Aquifer of Singair Upazila, Manikganj District, Bangladesh. *Journal of Bangladesh Academy of Sciences*, 42(1), 25-40. <https://doi.org/10.3329/jbas.v42i1.37830>
- Joseph, T., Dubey, B. and McBean, E. A. (2015). A critical review of arsenic exposures for Bangladeshi adults. *Science of The Total Environment*, 527-528, 540–551. doi:10.1016/j.scitotenv.2015.05.035
- Kavcar, P., Sofuoglu, A. and Sofuohlu, S.C. (2009). A health risk assessment for exposure to trace metals via drinking water ingestion pathway. *Int. J. Hyg. Environ. Health* 212, 216-227.
- Kundu, M.C. and Mandal, B. (2008a). Agriculture activities influence nitrate and fluoride contamination in drinking groundwater of an intensively cultivated district in India, *Water air Soil Pollut.* doi:10.1007/s1270-008-9842-5
- Kundu, M.C., Mandal, B. and Sarkar, D. (2008b). Assessment of the potential hazardous of nitrate contamination in surface and groundwater in a heavily fertilized and intensively cultivated district of India. *Environ. Monit. Assess.* 146, 183–189
- Li, P., He, X. and Guo, W. (2019). Spatial groundwater quality and potential health risks due to nitrate ingestion through drinking water: A case study in Yan'an City on the Loess Plateau of northwest China. *Hum. Ecol. Risk Assess.* 1-21. doi:10.1080/10807039.2018.1553612
- Li, P., Wu, J., Qian, H., Lyu, X. and Liu. H. (2013). Origin and assessment of groundwater pollution and associated health risk: a case study in an industrial park, northwest China. *Environ. Geochem. Health.* 36, 693-712.
- Lim, H.S., Lee, J.S., Chon, H.T. and Sager, M. (2008). Heavy metal contamination and health risk assessment in the vicinity of the abandoned Songcheon Au–Ag mine in Korea. *J. Geochem. Explor.* 96, 223–230.
- Lohumi, N., Gosain, S., Jain, A., Gupta, V.K. and Verma, K.K. (2004). Determination of nitrate in environmental water samples by conversion into nitrophenols and solid phase extraction-spectrophotometry, liquid chromatography or gas chromatography–mass spectrometry, *Anal. Chim. Acta* 505, 231–237.
- Ma, H.W., Hung, M.L. and Chen, P.C. (2007). A systemic health risk assessment for chromium cycle in Taiwan. *Environ. Int.* 33, 206-218.
- Majumder, R.K., Hasnat, M.A., Hossain, S., Ikeue, K. and Machida, M. (2008). An exploration of nitrate concentrations in groundwater aquifers of central-west region of Bangladesh. *J. Hazard. Mater.* 159, 536-543
- Marques Arsénio, A., Câmara Salim, I., Hu, M., Pedro Matsinhe, N., Scheidegger, R. and Rietveld, L. (2018). Mitigation potential of sanitation infrastructure on groundwater contamination by nitrate in Maputo. *Sustainability*, 10, 858. doi:10.3390/su10030858
- Martinelli, G., Dadomo, A., De Luca, D. A., Mazzola, M., Lasagna, M., Pennisi, M. and Saccon, P. (2018). Nitrate sources, accumulation and reduction in groundwater from Northern Italy: Insights provided by a nitrate and boron isotopic database. *Appl. Geochem.* 91, 23-35. doi:10.1016/j.apgeochem.2018.01.011.
- Mehrdadi, N., Nabi Bidhendi, G. R., Nasrabadi, T., Hoveidi, H., Amjadi, M. and Shojae, M. A. (2009). Monitoring the arsenic concentration in groundwater resources, case study: Ghezel ozan water basin, Kurdistan, Iran. *Asian journal of chemistry*, 21(1), 446-450.
- Messier, K.P., Wheeler, D.C., Flory, A.R., Jones, R.R., Patel, D., Nolan, B.T. and Ward, M.H. (2019). Modeling groundwater nitrate exposure in private wells of North Carolina for the agricultural health study. *Sci Total Environ.* 655, 512-519. doi:10.1016/j.scitotenv.2018.11.022
- Momot, O. and Synzynys, B. (2005). Toxic aluminium and heavy metals in ground wate of middle Russis: health risk assessment. *Int. J. Environ. Res. Pub. Health* 2, 214-218.
- Nasrabadi, T. and Maedeh, P. A. (2014). Groundwater quality assessment in southern parts

- of Tehran plain, Iran. *Environmental earth sciences*, 71(5), 2077-2086.
- Nasrabadi, T., Maedeh, P. A., Sirdari, Z. Z., Bidabadi, N. S., Solgi, S. and Tajik, M. (2015). Analyzing the quantitative risk and hazard of different waterborne arsenic exposures: case study of Haraz River, Iran. *Environmental earth sciences*, 74(1), 521-532.
- Ning, L., Ni, T., Xia, J., Dai, M., He, C. and Lu, G. (2011). Non-carcinogenic risks induced by metals in drinking source water of Jiangsu Province, China. *Environ. Monit. Assess.* 177, 449-456. DOI 10.1007/s10661-010-1646-6.
- Paladino, O., Seyedsalehi, M. and Massabo, M. (2018). Probabilistic risk assessment of nitrate groundwater contamination from greenhouses in Albenga plain (Liguria, Italy) using lysimeters. *Sci. Total Environ.* 634, 427-438. doi:10.1016/j.scitotenv.2018.03.320
- Pannala, A.S., Mani, A.R., Spencer, J.P.E., Skinner, V., Bruckdorfer, K.R., Moore, K.P. and Rice-Evans, C.A. (2003). The effect of dietary nitrate on salivary, plasma, and urinary nitrate metabolism in humans. *Free Radic. Biol. Med.* 34, 576-584.
- Qasemi, M., Afsharnia, M., Farhang, M., Bakhshizadeh, A., Allahdadi, M. and Zarei, A. (2018). Health risk assessment of nitrate exposure in groundwater of rural areas of Gonabad and Bajestan, Iran. *Environ. Earth Sci.* 77(15). doi:10.1007/s12665-018-7732-8
- Rachid, A., Christophe, M., Marc, B., Laure, O., Sylvie, T. and Paul, P. (2006). Methemoglobinemia by cerium nitrate poisoning, *Burns* 32, 1060-1061.
- Rahman, M., Islam, M., Bodrud-Doza, M., Muhib, M., Zahid, A., Shammi, M. and Kurasaki, M. (2017). Spatio-temporal assessment of groundwater quality and human health risk: a case study in Gopalganj, Bangladesh. *Expo Health*. doi:10.1007/s12403-017-0253-y
- Rahman, M.M., Sultana, R., Shammi, M., Bikash, J., Ahmed, T., Maruo, M., Kurasaki, M. and Uddin, M.K. (2016). Assessment of the status of groundwater arsenic at SingairUpazila, Manikganj, Bangladesh; Exploring the correlation with the other metals and ions. *Expo. Health* 8, 217-225.
- Rizeei, H. M., Azeez, O. S., Pradhan, B. and Khamees, H. H. (2018). Assessment of groundwater nitrate contamination hazard in a semi-arid region by using integrated parametric IPNOA and data-driven logistic regression models. *Environ. Monit. Assess.* 190(11), 633. doi:10.1007/s10661-018-7013-8
- Shukla, S. and Saxena, A. (2018). Global status of nitrate contamination in groundwater: its occurrence, health impacts, and mitigation measures. 1-21. doi:10.1007/978-3-319-58538-3_20-1
- Sohel, N., Persson, L.A., Rahman, M., Streatfield, P.K., Yunus, M., Ekstro'm, E.C. and Vahter, M. (2009). Arsenic in Drinking Water and Adult Mortality A Population-based Cohort Study in Rural Bangladesh. *Epidemiology*, 20, 824-830
- Suthara, S., Bishnoib, P., Singh, S., Mutiyara, P.K., Nema, A.K. and Patil. N.S. (2009). Nitrate contamination in groundwater of some rural areas of Rajasthan, India. *J. Hazard. Mater.* 171, 189-199
- Taufiq, A., Effendi, A. J., Iskandar, I., Hosono, T. and Hutasoit, L. M. (2019). Controlling factors and driving mechanisms of nitrate contamination in groundwater system of Bandung Basin, Indonesia, deduced by combined use of stable isotope ratios, CFC age dating, and socioeconomic parameters. *Water Res.*, 148, 292-305. doi:10.1016/j.watres.2018.10.049
- Tavakoly, A.A., Habets, F., Saleh, F., Yang, Z.-L., Bourgeois, C. and Maidment, D.R. (2019). An integrated framework to model nitrate contaminants with interactions of agriculture, groundwater, and surface water at regional scales: The STICS-EauDyssée coupled models applied over the Seine River Basin. *J. Hydrol.* 568, 943-958. doi:10.1016/j.jhydrol.2018.11.061
- USEPA (US Environmental Protection Agency) (1989). Risk Assessment Guidance for Superfund Volume I Human Health Evaluation Manual (Part A). United States Environmental Protection Agency, Washington, D.C.
- USEPA (US Environmental Protection Agency) (1993). Risk assessment guidance for superfund (RAGS), volume I: human health evaluation manual (part E) interim. United States Environmental Protection Agency, Washington, D.C.
- USEPA (US Environmental Protection Agency) (1999). A risk assessment-multiway exposure spreadsheet calculation tool. United States Environmental Protection Agency. Washington, D.C.
- USEPA (US Environmental Protection Agency) (2001). Baseline human health risk assessment, Vasquez Boulevard and 1-70 superfund site, Denver CO.
- USEPA (US Environmental Protection Agency) (2009). National primary/secondary and drinking water regulations. Washington, D.C.

Wagh, V. M., Panaskar, D. B., Mukate, S. V., Aamalawar, M. L. and Laxman Sahu, U. (2019). Nitrate associated health risks from groundwater of Kadava River Basin Nashik, Maharashtra, India. *Hum. Ecol. Risk Assess.* 1-19. doi:10.1080/10807039.2018.1528861

Ward, M.H., DeKok, T.M., Levallois, P., Brender, J., Gulis, G., Nolan, B.T. and Derslice, J.V. (2005). Workgroup report: Drinking-water nitrate and health—Recent findings and research needs, *Environ. Health Perspect.* 113, 1607–1614.

Weyer, P.J., Cerhan, J.R., Kross, B.C., Hallberg, G.R., Kantamneni, J., Breuer, G., Jones, M.P., Zheng, W. and Lynch, C.F. (2001). Municipal drinking water nitrate level and cancer risk in older women: the Iowa women's health study. *Epidemiology* 11, 3.

Wild, L. M., Mayer, B. and Einsiedl, F. (2018). Decadal delays in groundwater recovery from nitrate contamination caused by low O₂ reduction rates. *Water Resour. Res.* doi:10.1029/2018wr023396

Wongsasuluk, P., Chotpantarat, S., Siriwong, W., and Robson, M. (2013). Heavy metal contamination and human health risk assessment in drinking water from shallow groundwater wells in an agricultural area in UbonRatchathani province, Thailand. *Environ. Geochem. Health* 36, 169–182. DOI.10.1007/s10653-013-9537-8

Wu, B., Zhang, Y., Zhang, X., and Cheng, S. (2010). Health risk from exposure of organic pollutants through drinking water consumption in Nanjing, China. *Bull. Environ. Contam. Toxicol.* 84, 46-50.

Zhai, Y., Zhao, X., Teng, Y., Li, X., Zhang, J., Wu, J., and Zuo, R. (2017). Groundwater nitrate pollution and human health risk assessment by using HHRA model in an agricultural area, NE China. *Ecotoxicol. Environ. Saf.* 137, 130-142

Zhang, Y., Wu, J., and Xu, B. (2018). Human health risk assessment of groundwater nitrogen pollution in Jinghui canal irrigation area of the loess region, northwest China. *Environ. Earth Sci.* 77(7). doi:10.1007/s12665-018-7456-9

