Tran, D.A., Tsujimura, M., Huu, H.L., Dang, D.H., Le Vo, P., Doan, T.H., Thu Nguyen, T.T., Le, C.C., Bich Pham, T.T., Dang, T.D., Batdelger, O., **Nguyen, V.T.** (2021):
Groundwater quality evaluation and health risk assessment in coastal lowland areas of the Mekong Delta, Vietnam
Groundwater for Sustainable Development **15** , art. 100679

http://dx.doi.org/10.1016/j.gsd.2021.100679

Groundwater quality evaluation and health risk assessment in coastal lowland areas of the Mekong Delta, Vietnam

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Abstract

This study provides a comprehensive assessment of groundwater quality in the Mekong Delta (Vietnam) and their potential risks to human health. The dataset consists of 282 groundwater samples collected in dry and rainy seasons between 2017 and 2018. Multiple physio-chemical parameters, groundwater quality index (GWQI), and irrigation water quality indices (IWQI) were employed to evaluate the suitability of groundwater resources for drinking and irrigation purposes. Thereupon, the hazard quotient (HQ), and total hazard index (HI) were used to assess the non-carcinogenic risks to different demographic groups. GWQI indicates that groundwater samples are excellent (41.5%), good (40.4%), poor (17%), and very poor (1.1%) classes for drinking purpose. The total hazard index (HI) indicated that 15.2%, 8.5%, 7.4%, and 7.1% of samples express non-carcinogenic health threats to infants, females, children, and males. Irrigation water quality indices (IWQI), namely, EC, sodium adsorption ratio (SAR), residual sodium bicarbonate (RSBC), soluble sodium percent (SSP), permeability index (PI), and magnesium hazard (MH), reveal that 82.2%, 94%, 20.2%, 69.6%, 45.4% and 12.8% of samples have EC < 2,250 μS/cm, SAR < 26, RSBC <1.25, SSP <60%, PI > 75%, and MH < 50%, respectively, which are suitable for irrigation use. Wilcox diagram shows that 17.54 % of total groundwater samples exceed the permissible levels of salt-tolerant crops and solidity hazards in the study area. Furthermore, high concentrations of nitrate (>50 mg/L) and chloride (> 1,000 mg/L) were detected in the shallow, intermediate, and deep aquifers. GWQI and IWQ maps imply that groundwater in the South-western area is suitable, whereas groundwater in the central area is not recommended for any purpose. These findings provide useful insights into the vulnerable groundwater system of the coastal lowland area of the Mekong Delta related to human activities (agricultural practices, dam construction) and global climate change impacts. Accordingly, appropriate management strategies for water resources and mitigation solutions are imperatively needed to ensure the sustainability of the groundwater resource and the protection of public health in the Mekong Delta.

Keywords: Multi-groundwater Quality Assessment, Human Health Risk Assessment, Geochemistry, Coastal aquifers, Mekong Delta

1. Introduction

Groundwater resources play an essential role in human consumption, irrigation purpose, and industrial development (Adimalla, 2019; Velis et al., 2017). It has extremely become a vital freshwater resource in coastal lowland regions, where surface water is scarce due to seawater intrusion and pollution (Alcérreca-Huerta et al., 2019; Lu et al., 2020). Over the recent decades, the degradation of groundwater quality was mainly associated with intensive applications of toxic chemicals (e.g., inorganic contaminants, pesticides, fertilizers) posed by agricultural and rural development activities and excessive discharge of waste and wastewater without proper treatment (Chen et al., 2020; Habib et al., 2020; Hossain and Patra, 2020). As a result, the long-term use of polluted groundwater for drinking and irrigation purposes leads to severe human health effects such as (i) cardiovascular diseases (Bai et al., 2019; Chabukdhara et al., 2017; Muhammad et al., 2011; Tran and Nguyen, 2018; Ustaoğlu, 2020; Wen et al., 2019; Yang et al., 2020), (ii) cancers (Kaur et al., 2019; Mohammed Abdul et al., 2015; Mondal et al., 2008; Rasheed et al., 2016; Saha and Rahman, 2020) and also adverse impacts on the natural ecosystems (Bartzas et al., 2015; Szymczycha et al., 2020).

In addition to toxic chemicals, nitrate contamination and salt intrusion in coastal groundwater systems have increasingly become a global issue (Knoll et al., 2019; Pazhuparambil Jayarajan and Kuriachan, 2020; Torres-Martínez et al., 2021; Wu et al., 2019). High nitrate concentrations and salinity in groundwater are not only detrimental to human health but also to agricultural ecosystems (Bartzas et al., 2015; Egbi et al., 2020; Lu et al., 2020; Marghade et al., 2020; Zolekar et al., 2020). First, nitrogen pollution, especially nitrate, is considered a critical pollutant of groundwater resources (Adimalla and Qian, 2021; Egbi et al., 2020). It is directly associated with serious disease such as methemoglobinemia, colorectal cancer, thyroid disease, and neural tube defect (Ward et al., 2018). Long-term ingestion of high nitrate concentrations through groundwater is the leading cause of elevated non-carcinogenic risks in the Indian population, especially infants and children (Adimalla et al., 2019; Adimalla and Qian, 2021; Adimalla et al., 2020). Negative impacts of nitrate contamination on human health were also observed in several regions worldwide (Barakat et al., 2019b; Emenike et al., 2018; Ijumulana et al., 2020; Li et al., 2021; Naderi et al., 2020; Rezaei et al., 2019). Second, the increase in dissolved concentrations of major ions (salinity) became an issue of imminent concern (Rahaman et al., 2020). Global climate change (e.g., sea-level rise) and human activities (e.g., unsustainable groundwater withdrawal, dam construction along major rivers) are the major factors instigating seawater intrusion into both surface and groundwater systems (Alcérreca-Huerta et al., 2019; Telahigue et al., 2020; Zeynolabedin et al., 2020). Such an increase in the ionic compositions of freshwater has a major effect on the ecosystems (Bugica et al., 2020; Liang et al., 2020; Zhu et al., 2020) and also cause severe health problems (Mitchell et al., 2019; Rakib et al., 2019; Rakib et al., 2020; Shukla and Saxena, 2020; Wen et al., 2019). Meanwhile, long-term consumption of water containing salt and total hardness (Ca and Mg) concentrations, in excess, increases the risk of hypertension (Shammi et al., 2019; Vineis et al., 2011), coronary heart disease (Park and Kwock, 2015; Rahaman et al., 2020), and chronic kidney disease (Naser et al., 2017; Rahaman et al., 2020; Ravindra et al., 2019). On the other hand, irrigation water with high salinity and total hardness can deteriorate soil fertility and thus reduce crop yields (Korres et al., 2019; Parvin et al., 2019; Radanielson et al., 2018).

The deterioration of groundwater quality by different pollutants could severely impact the ecosystem and human health. This is a persistent issue that could eventually worsen because of increasing pressure on groundwater resources associated with growing populations and anthropogenic activities (e.g., intensive agriculture and industries). Therefore, groundwater quality and human health risk assessment are critical to protect the people relying on groundwater and ensure food safety (Adimalla et al., 2019; Kaur et al., 2020; Rabeiy, 2018; Rezaei et al., 2019; Shukla and Saxena, 2020; Singh et al., 2019; Singh et al., 2020; Wagh et al., 2020; Wen et al., 2019; Yin et al., 2020; Zhang et al., 2020; Zhang et al., 2019).

The Mekong Delta (MD) has been recognized as one of the most vulnerable regions to climate change and sea-level rise globally (Dang et al., 2018; Shrestha et al., 2016; Smajgl et al., 2015). Although water resources play a crucial role in socio-economic development in the Mekong Delta, water quality has been increasingly deteriorated because of intensive human activities and natural variations (Dao et al., 2020; Ha et al., 2019; Hoang and Bäumle, 2019; Hung Van et al., 2019; Thanh Giao et al., 2021; Thanh Nguyen, 2020; Thu Minh et al., 2020; Tran et al., 2020). It is especially applicable to the coastal regions because of severe water pollution and seawater intrusion affecting both surface and groundwater systems (Dang et al., 2020; Dao et al., 2020; Thanh Nguyen, 2020; Tran et al., 2021; Tran et al., 2020; Trung and Tri, 2014). Intensive agricultural development and rapid industrialization in combination with extreme drought events and seawater intrusion have resulted in severe degradation of surface and groundwater resources (Buschmann et al., 2008; Chea et al., 2016; Nguyen et al., 2020; Tran et al., 2020). Recent studies indicated that the long-term and unsuitable use of groundwater for drinking and irrigation purposes amplified human health problems and also threatened agricultural productions in the Mekong Delta (Braun et al., 2019; Le Luu, 2017; Merola et al., 2015; Reid et al., 2021; Toan et al., 2013; Tran et al., 2020). However, there are few studies providing a quantitative assessment of groundwater quality for drinking and irrigation purposes. Moreover, it is necessary to evaluate potential human health risks caused by long-term consumption of saline and nitrate-rich groundwater in coastal lowland regions of the Mekong Delta. Hence, this research aims to evaluate the links between groundwater quality and its suitability for use as irrigation and drinking water and potential human health risk. The specific objectives of this study are to (1) evaluate the suitability of groundwater for drinking purposes, (2) assess potential effects and health risks to different population groups due to high chloride and nitrate contaminations.

2. Study Area

The study is conducted in Soc Trang, a coastal province located in the Southeast of the Mekong Delta (Vietnam), covering approximately 3,310 km^2 with a flat and low elevation, ranging from 0.5 to 2.5 m (Figure 1). The study area is known for its tropical monsoon climate system with two distinct seasons: the dry season starts from May to November and the rainy season lasts from December to April of the following year. The average annual rainfall is about 1,770 mm with high seasonality: about 85% of the annual rainfall occurs during the rainy season. In the study area, land use/land cover is dominated by agricultural land (84.8% of total area), followed by residential land (5.2%) and forested land (4.40%) (Tran et al., 2021). Additionally, Soc Trang province has various soil types, including sandy, alluvial, salt, and acid sulphate soils (Kawahigashi et al., 2008). Economically, most of the local population depends primarily on agricultural activities, which contribute to 42% of the total provincial GDP. Therefore, the demand for freshwater resources in this province is incredibly high, approximately 25 million cubic meters per year (field survey data, data not shown).

The study site has a dense surficial drainage network with a density of 2.21 km/km^2 (Tran et al., 2021). The drainage network directly connects the Hau River (the Bassac River) to the Northeast and the East Sea to the Southeast. Therefore, the surface and subsurface flow dynamic in the area is driven by meteorological forces and flow regimes of the Hau River streamflow and tidal regimes of the east Sea (Tran et al., 2020; Tran et al., 2019). Together, sands, fluvial deposits, and gravels with various grain sizes dominate sedimentary deposits (Tran et al., 2020; Tran et al., 2019) also influence on the groundwater systems of the study area. The aquifer system consists of seven hydrogeological units, including Holocene (qh), Upper Pleistocene (qp_3), Middle Pleistocene (qp_{23}), Lower Pleistocene (qp_1), Middle Pliocene (n_{22}), Lower Pliocene (n_{21}), and Upper Miocene (n_{13}) (Tran et al., 2020; Wagner et al., 2012). Each hydrogeological unit has the upper and lower layers with different lithological characteristics. The top layer consists of silt, clay or mixed silt clay with low water yield (Wagner et al., 2012). In contrast, the lower part is relatively permeable, consisting of fine to coarse sand, gravel, and pebbles (Hoang and Bäumle, 2019; Tran et al., 2020). Groundwater in the Pleistocene aquifers and Upper Miocene are the primary supply source of drinking water because of its high yield and good quality compared to other aquifers (Tran et al., 2018). However, long-term excessive groundwater extraction has caused an increase in chloride and nitrate concentrations in the aquifer system (Tran et al., 2020). Indeed, Soc Trang frequently faces extreme seawater intrusion events causing severe water scarcity, especially in the dry season.

[Figure 1]

3. Materials and Methods

3.1. Groundwater sampling and chemical analysis

A total of 282 groundwater samples were collected from 145 wells in the dry and rainy season between 2017 and 2018 in Soc Trang (Figure 1). The wells are either household tube wells of Soc Trang Water Supply Company and Center for Rural Water Supply and Sanitation and boreholes from the national groundwater monitoring system. The sampling depths varied from 3.5 m to 485 m below ground surface to cover all aquifer layers. Physical and chemical parameters such as pH, electrical conductivity (EC), dissolved oxygen (DO), and total dissolved solids (TDS) were measured directly at the field by using Hanna Instruments portable meters. Each groundwater sample was collected after 15-20 minutes of purging until pH, EC, and DO values were stabilised. All groundwater samples were stored in 100 mL polyethylene bottles, kept at dark and 4^{o}C. Subsequently, all groundwater samples were filtered through 0.2-µm cellulose ester filters.

Physio-chemical properties and compositions of groundwater were analysed according to the standard procedures of the American Public Health Association (E.W. Rice, 2017) and were described in detail in a previous publication (Tran et al., 2020). Briefly, chemical analyses were performed at the Hydrology and Water Environment Laboratory, University of Tsukuba, Japan. Major cations (Na^{+}, K^{+}, Ca^{2+} and Mg^{2+}) were measured by ICP-OES, Perkin Elmer Optima 7300. Main anions (Cl^{-}, NO_3^{-}, and SO_4^{2-}) were analysed using Ion Chromatography (Shimadzu, Japan). Bicarbonate (HCO_3^{-}) was determined by the titration method. Finally, ion balance was calculated for each sample; the error was below 5%, justifying the accuracy of the chemical analyses.

3.2. Drinking groundwater quality index

Groundwater quality index (GWQI) is widely used to assess general groundwater quality by converting multiple groundwater quality parameters into a single value (Adimalla and Qian, 2019; Barakat et al., 2019a; Ghouili et al., 2018; Zhai et al., 2017). Therefore, the suitability of the groundwater for use as drinking water could be determined based on GWGI-derived water quality status (Table 1). To determine GWQI (Eq.1), each parameter was assigned with a specific weight value (W_i) depending on its potential to affect groundwater quality for drinking purposes. The overall GWQI was calculated linearly by adding the quality rating (Q_i) to the unit weight (W_i).

$$GWQI = \frac{\sum Q_i \times W_i}{\sum W_i} \quad (1)$$

$$where\ Q_i = \frac{(C_o - C_i)}{(S_i - C_i)} \times 100 \quad (2)$$

$$W_i = \frac{K}{S_i} \quad (3)$$

$$K = \frac{1}{\sum 1/S_i} \quad (4)$$

Where Q_i is a water quality rating for the i^{th} parameter (Eq.2), W_i is a unit weight for the i^{th} parameter (Eq.3), S_i is a standard value of the i^{th} parameter, C_o is an observed value of the n^{th} parameter, C_i is an ideal value of i^{th} parameter, and K is a constant for proportionality (Eq.4). Here, the main parameters used for GWQI calculations are provided in Table 2.

[Table 1]

[Table 2]

3.3. Irrigation water quality indices (IWQI)

Natural groundwater often has high concentrations of dissolved ions because of geochemical reactions (e.g., mineral dissolution, ion exchange). Therefore, using groundwater for irrigation could adversely affect the physiochemical conditions of soils and crops (Adimalla and Qian, 2019). To evaluate the suitability of groundwater for irrigation, there are several indices, including sodium adsorption ratio (SAR), residual sodium bicarbonate (RSBC); soluble sodium percent (SSP), permeability index (PI), and magnesium hardness (MH) (Anim-Gyampo et al., 2019; Sabarathinam et al., 2020). The relations used to estimate these indices are listed from Eq. 5 to Eq. 9, respectively. According to these calculations, we classified groundwater samples as per their suitability for irrigation.

$$\text{Sodium adsorption ratio (SAR)} = \frac{Na^{+}}{\sqrt{\frac{Ca^{2+} + Mg^{2+}}{2}}} \quad \text{(Richards, 1954)} \quad (5)$$

Residual sodium bicarbonate (RSBC) $= HCO_3^- - Ca^{2+}$ (Gupta, 1983) (6)

Soluble Sodium Percent (SSP) (Todd, 1980) (7)

$$\text{SSP} = \left(\frac{Na^+ + K^+}{Na^+ + Ca^{2+} + Mg^{2+}}\right) \times 100$$

Permeability index (PI) $= \dfrac{Na^+ + \sqrt{HCO_3^-}}{(Na^+ + K^+ + Ca^{2+} + Mg^{2+})} \times 100$ (Doneen, 1964) (8)

Magnesium hazard (MH) $= \dfrac{Mg^{2+}}{Ca^{2+} + Mg^{2+}} \times 100$ (Szaboles, 1964) (9)

3.4. Health risk assessment

Human health problems are often related to long-term consumption of unsafe water sources caused by natural and anthropogenic contaminants in surface and subsurface environments. Therefore, assessing the potential health risks caused by substandard groundwater quality requires intensive investigation of water supply quality. However, water quality intimately depends on the hydrogeographic and hydrogeochemical conditions of each specific geographical location. Therefore, the study conducted a health risk assessment (HRA) to determine the potential impacts of contaminated water on human health. Accordingly, we calculated the chronic daily intake (CDI) by intake and dermal contact of two contaminants, nitrate (>50 mg/L) and chloride (> 1,000 mg/L), as they represent the chemicals of highest concern in drinking water for non-carcinogenic risk assessments (Adimalla, 2020; Pazhuparambil Jayarajan and Kuriachan, 2020; Saleh et al., 2019). The exposures of such pollutants to humans are calculated using Eq.10 and Eq.11 (Ali and Chidambaram, 2021; Proshad et al., 2020).

$$CDI_{oral} = \frac{C_i \times IR \times EF \times ED}{BW \times AT} \quad (10)$$

$$CDI_{dermal} = \frac{C_i \times K_p \times IR \times ET \times EF \times ED \times CF \times SA}{BW \times AT} \quad (11)$$

CDI_{oral} and CDI_{demal} are exposure doses through ingestion and dermal absorption, respectively in mg/kg/day. C_i is the concentration of chloride and nitrate in groundwater (mg/L). IR is the ingestion rate of water per day. EF is exposure frequency in the number of days per year. ED is exposure duration (years). BW is average body weight (kg). AT is averaging time (days). K_P represents a dermal permeability coefficient in water (cm/h). ET is the exposure time (h/day). CF is the unit conversion factor (L/cm^3). SA is skin exposure that was calculated by using Eq.12 (Barakat et al., 2019b), where H is the average height of the population. To consider various physiological characteristics and health conditions of the populations, we considered in this study four subpopulations, which correspond to adult males, adult females, children, and infants. The selected parameters for each sub-population were presented in **Table 3**.

$$SA = 239 \times H^{0.417} \times BW^{0.517} \tag{12}$$

[Table 3]

The calculated CDI allows the non-carcinogenic hazard quotient (HQ) determination through oral and dermal pathways using Eq.13 and Eq.14. The total hazard index (HI) was calculated as the sum of the hazardous quotient (Eq.15). These calculations directly reflect the risks that the exposed population may develop serious health problems under specific conditions of exposures (e.g., concentrations of the pollutants, exposure duration).

$$HQ^{i}_{oral} = \frac{CDI_{oral}}{RfD_{oral}} \tag{13}$$

$$HQ^{i}_{dermal} = \frac{CDI_{dermal}}{RfD_{dermal}} \tag{14}$$

$$HI^{i} = \sum_{i=1}^{N} HQ^{i} \tag{15}$$

In these calculations, i reflects the water quality indicators (i.e., chloride and nitrate). RfD_{oral} and RfD_{dermal} are the reference dose for non-carcinogenic risks through the oral intake and dermal absorption pathways. It is important to note that HQ_{oral} and HQ_{dermal} depend significantly on the selected parameters used in Eq. 10 and Eq. 11, respectively (Karunanidhi et al., 2020). Also, in order to accurately assess the overall health risks associated with ingestion and dermal pathways, it is necessary to take into consideration as much as possible the solutes in water sources that are deemed harmful. In this light, chloride and nitrate parameters were selected to assess. Chloride reference dose (78.68 mg/kg/day) was used for both oral and dermal pathways (USEPA 2018) whilst nitrate reference dose of 1.6 and 0.8 mg/kg/day was applied for oral and dermal contact, respectively (ECHA, 2020; USEPA, 2018).

4. Results and Discussion

4.1. Groundwater quality for drinking purposes

4.1.1. Physio-chemical characteristics

EC is a fundamental parameter reflecting the ionic concentrations or concentrations of dissolved salts in a water sample. In this study, groundwater has a wide range of variation in EC values, ranging from 117 to 18,500 µS/cm (average of

1,755 ± 2,250 µS/cm, Table 4). EC values show a heterogeneous spatial distribution (Figure S1a). Most of the high saline groundwater samples are found in shallow wells (depth < 40 m, n = 19), excepting a small number of samples (n=4) in deep wells (> 40m) (Figure S1b). Such a spatial heterogeneity of EC can be attributed to several factors such as minerals dissolution, seawater intrusion, and human impacts (Han and Currell, 2018; Hoang and Bäumle, 2019; Tran et al., 2020).

[Table 4]

The range of pH variations could indicate the origin of groundwater and geochemical processes during water movement from recharge throughout porous media to discharge areas. Groundwater in the study area has a relatively wide variation in pH, ranging from 6.1 to 9.3 (average of 7.3±0.5, Table 4). Alkaline groundwater samples (pH>7.5) were observed in inland to coastal areas (southern bound direction, Figure S1c). These high pH values are commonly found in deep aquifers (depth > 200m, Figure S1d) and are associated with the influence of the bicarbonate/carbonate system (Tran et al., 2020). Relative to the drinking water standard (pH=6.0-8.5), only three groundwater samples out of 282 exceeded the maximum permissible limit of 8.5 for drinking water. However, 15 samples have pH values below the minimum acceptable limit of 6.0. This issue was attributed to the process of iron oxidation occurring in the groundwater of the Mekong Delta (Ha et al., 2019).

Total dissolved solids (TDS) consist of major dissolved compounds in groundwater such as sodium, potassium, calcium, magnesium, chloride, sulphates, nitrate, and bicarbonate. For this study, groundwater TDS was in the range of 82-12,950 mg/L (average of 1,228 ±1,576 mg/L, Table 4). According to the WHO drinking water standard (WHO, 2017), acceptable drinking water quality should have TDS values < 500 mg/L and is unpalatable if TDS >1,500 mg/L. Accordingly, 31.6% of the groundwater samples are safe for drinking while at least 19.5% are unpalatable. High TDS (>1,500 mg/L) was observed in both inland and coastal areas (Figure S1e), especially in deep wells (Figure S1f), indicating influences of groundwater salinization (Tran et al., 2020). It was estimated that approximately 35% of groundwater samples exceed the maximum concentration of TDS (1,000 mg/L) based on the National Technical Regulation on Domestic Water Quality (QCVN 01-1:2018/BYT) as shown in Table 4.

Overall, concentrations of major elements were in the following order of dominance: $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ for cations and $Cl^- > HCO_3^- > SO_4^{2-} > NO_3^-$ for anions. Sodium and Cl^- showed higher concentrations than other ions, and their Na^+/Cl^- ratios approximated 0.86 (characteristic of seawater composition), suggesting seawater intrusion into the shallow aquifer system of the study area (Xiong et al., 2020). Although a certain amount of sodium daily intake is essential to ensure human health, exceeding sodium intake causes adverse health risks, especially hypertension and nausea diseases (Park and Kwock, 2015; Rahaman et al., 2020). Therefore, sodium is one of the most important parameters for human health risk assessment. In this study, sodium concentrations varied from 17 mg/L to 8,536 mg/L (average of 297 ± 683 mg/L) while potassium concentrations ranged from 0.5 to 279 mg/L (average of 19± 26 mg/L) (Table 4). High sodium and potassium concentrations were mainly observed in shallow aquifers of coastal areas (Figure S1i, S1k) (Figure S1j, S1l), indicating that seawater intrusion during the Holocene period may play an important factor in the accumulation of these ions (Meyer et al., 2019). Accordingly, more than 30% and 43% of groundwater samples in the study area exceed the maximum limit of sodium and potassium, respectively.

Calcium and magnesium are also important elements for human health; however, excessive uptake of these ions leads to several health problems, especially kidney stones and hypertension (Cormick and Belizán, 2019; Ndii et al., 2020). Concentrations of calcium varied from 0.1 to 927 mg/L (average of 45±82 mg/L), while magnesium concentrations ranged from 0.03 mg/L to 693 mg/L (average of 46 ±81 mg/L, Table 4). According to WHO and QCVN 01-1:2018/BYT, 2% and 4% of total groundwater samples in this study area exceeded the maximum calcium and magnesium concentrations, respectively. Total hardness (TH) reflects the hardness of water caused by calcium and magnesium (Adimalla and Qian, 2019). However, the variation of TH concentrations corresponds to different classes of water hardness. For instance, TH values < 75 indicate soft water, while TH in the ranges of 75-150, 151-300, and >300 mg/L are related to moderate, hard, and very hard water, respectively (Adimalla and Qian, 2019). The TH value of groundwater in this area varied from 13 mg/L to 3,080 mg/L (average of 280 ± 377 mg/L). Only 5.7% of groundwater samples in this area exceeded the maximum permissible limit of 500 mg/L for drinking purposes (WHO, 2017). High TH concentration (>300 mg/L) in groundwater was observed in some locations in inland and coastal areas (Figure S1g) in shallow aquifers. Meanwhile, deeper aquifers show relatively low TH (< 150 mg/L) (Figure S1h). We have previously demonstrated that the precipitation of minerals (e.g., calcite) could be responsible for decreasing soluble Ca/Mg in those deep aquifers (Tran et al. 2020).

Besides, chloride concentrations in this area ranged from 2 mg/L to 15,883 mg/L (average of 413 ± 1,312 mg/L) (Table 4). High chloride concentration (> 1,000 mg/L) was found not only in coastal areas but also in inland areas far from the East Sea (Figure 2a) as well as both shallow and deep wells (Figure 2b). This observation implies the complexity of past and presents seawater intrusion processes in this area (Hoang and Bäumle, 2019; Pham et al., 2019;

Tran et al., 2020). Approximately 28% and 23% of groundwater samples in the study area exceeded chloride concentration compared to the maximum concentration of 600 mg/L and 300 mg/L as regulated by WHO and QCVN 01-1:2018/BYT (Table 4), respectively. This result, coupled with spatial heterogeneity of salinity at the aquifer system, may exacerbate groundwater quality and put high risks to the water supply system in this study area.

Finally, more than 70% of groundwater samples have relatively high bicarbonate concentrations (>200 mg/L), while only 4% of groundwater samples exceed 200 mg/L of sulphates. This could be associated with pyrite oxidation leading to an accumulation of sulphate in groundwater (Ha et al., 2019).

[Figure 2]

Nitrate is among a few representative compounds for anthropogenic impacts on the aquatic environment (Egbi et al., 2020; Singh et al., 2019). Indeed, high nitrate concentrations in groundwater are mainly related to domestic waste, agricultural fertilizers, industrial waste, and wastewater (Egbi et al., 2020). For several decades, nitrate contamination has been a global problem, deteriorating groundwater quality and causing severe health problems in many countries worldwide (Abdelwaheb et al., 2019; Adimalla and Qian, 2019; Njuguna et al., 2020; Wagh et al., 2020). Nitrate concentrations in groundwater samples ranged from 0.1 mg/L to 260 mg/L with an average of 9±26 mg/L (Table 4). Relatively high nitrate concentrations (>10 mg/L) were observed in the urban area of Soc Trang city in an aquaculture field along the main river and on the coastline (My Xuyen and Vinh Chau, Figure 3a), which may be attributed to anthropogenic emissions. Additionally, high nitrate concentration (> 20 mg/L) was also found in many deep wells (depth > 40 m, n = 16) (Figure 3b). The contamination of nitrate in these deep wells may be associated with leaking artificial nitrate sources from landfills, urban and agricultural areas via unprotected wells (Bourke et al., 2019; Lockhart et al., 2013; Pastén-Zapata et al., 2014; Tran et al., 2020).

In summary, this assessment of groundwater quality in this area revealed that saline water and nitrate contaminations are significant issues exceeding the WHO drinking water standards. As groundwater is the primary source of drinking water in coastal regions of the Mekong Delta, the results suggested that long-term consumption of this groundwater could lead to severe human health problems for the local population. Therefore, assessing human health risks related to high nitrate and chloride concentration in the water supply system in this area is an imperative need.

[Figure 3]

4.1.2. Groundwater quality index (GWQI)

Groundwater quality index (GWQI) allows assessment of groundwater quality, and it was classified into five categories corresponding to different levels of the suitability of the groundwater for use as drinking water (Table 1). In this study, GWQI values ranged from 8.5 to 284.7 (average of 63.8 ± 40.6 Table 6). Accordingly, groundwater quality was classified into excellent (41.5%), good (40.4%), poor (17%), and very poor (1.1%) for drinking purposes. It was noted that GWQI has a heterogeneous spatial distribution in the study area. For example, poor (GWQI =101 - 200) and unusable groundwater quality (GWQI =201- 300) were found in the north-western regions and along rivers to the East Sea (Figure 4). It was attributed to the fact that an ongoing and intensive extraction of groundwater in these areas may further reduce the availability of groundwater resources (Tran et al., 2021; Tran et al., 2020). Therefore, alternative solutions should be adapted, and pumping activities should be minimized in the North-western and coastal areas of Soc Trang province.

[Figure 4]

4.2. Irrigation water quality

Groundwater quality assessment also plays a vital role in sustainable agricultural development in the region (Hoang and Bäumle, 2019; Tran et al., 2020). Hence, we employed a combination of indices (EC, SAR, RSBC, SSP, PI, and MH) as shown in Table 5 and Figure 5, and Wilcox's graphic (Richards, 1954) (Figure 6) was used to evaluate the suitability of the groundwater source for irrigation purposes. It is widely accepted that high sodium concentrations lead to the formation of sodic soil while exceeding salinity causes soil salinization; both are detrimental to plant development (Kurunc et al., 2020; Machado and Serralheiro, 2017). In addition, high concentrations of sodium and carbonate in irrigation water inhibit plant growth and subsequently reduce crops yield (Kurunc et al., 2020; Phogat et al., 2020). Therefore, electrical conductivity and sodium concentrations are simple and crucial indicators in classifying irrigation water (Kurunc et al., 2020; Taloor et al., 2020).

According to the classification of irrigation water (Wilcox, 1948), we categorized the groundwater samples into four classes based on their electrical conductivity (Table 5). These classes reflect their suitability to be used for irrigation, as excellent, good, permissible, and unsuitable; 1.4%, 33.3%, 47.5%, and 17.7% of our groundwater samples fall into these categories, respectively (Tables 5). In other words, almost one in every five groundwater samples of this study area is too saline that could negatively influence the practices of sustainable agricultural development. More importantly, SAR is a valuable index for the contribution of Na^+ relative to the sum of divalent cations (Richards, 1954). Higher SAR reflects the situation where a high proportion of Na^+, relative to Ca^{2+} and Mg^{2+}, could saturate the cation exchange complexes in soil and thus decrease soil permeability (Kurunc et al., 2020; Taloor et al., 2020). In this area, SAR values of groundwater varied from 0.3 to 51.8 with an average of 6.8 ± 9.2. High SAR values (> 26, or unsuitable groundwater for irrigation) correspond to 6% of the total dataset. In other words, this groundwater of substandard quality, if used for agricultural purpose, may damage soil structure, and form sodic soils.

Moreover, another indicator of long-term exacerbates soil health risks is the SSP index, representing the relative contribution of alkali metals (Na^+ and K^+) to the total cation charges (Eq. 7). SSP values ranged widely between 14.3% and 98.3%, with an average of 58.5 ± 22.7%. It was estimated that 40% of groundwater samples were doubtful and unsuitable for irrigation due to the potential impacts of Na^+ and K^+ on the soil structure. Additionally, it is also essential to identify the chemical speciation of these alkali metals in groundwater to correctly identify their impacts on arable soils. Carbonate is a crucial ligand in groundwater; high carbonate concentrations in groundwater often result in precipitation of Ca^{2+} and Mg^{2+}. However, the residual carbonate could form complexes with sodium that significantly decrease soil permeability and refer to the RSBC index. Accordingly, RSBC values varied from -39.9 to 12.5 with an average of 3.1 ± 5.0, in which nearly 69% of groundwater samples were unsuitable for irrigation based on the RSBC indices. The estimation of permeability index (PI %) values (Eq. 8) showed that 54.6% of total groundwater samples in the study area are inappropriate for irrigation because of low permeability.

In summary, most of the groundwater aquifers in this area are unsuitable for irrigation purpose. This is mainly due to the alkali metals (Na^+ and K^+) and their dissolved chemical complexes (carbonate). More noticeable, the alkali earth metals (Ca^{2+}, Mg^{2+}) are an immediate concern as 87.2% of groundwater samples in the study area have high MH values (> 50%). Integration of EC and SAR as the two significant factors associated with the poor quality of the groundwater was presented in the Wilcox diagram (Figure 6). This approach allowed an overall classification of groundwater quality as excellent, good, permissible, and unsuitable (Richards, 1954). The result shows that 101 samples (35.4%) fall under the excellent and good water classes for irrigation while 49 samples (17.5%) fall under the unsuitable water class sodium for irrigation. All these indexes pointed out that unsuitable groundwater is observed in the inland (Soc Trang city and My Tu district) and coastal areas (Tran De and Vinh Chau districts) (Figure 5), indicating the need for alternatives to the usage of groundwater for irrigation in these specific locations. Therefore, these results call for imminent actions to protect groundwater resources for the agricultural sector, which play a crucial socio-economic factor in the Mekong Delta coastal area.

[Table 5]

[Figure 5]

[Figure 6]

4.3. Health risk assessment

Long-term consumption of groundwater containing high nitrate and salt concentrations causes serious human health problems (Rahaman et al., 2020; Rakib et al., 2019). Therefore, non-carcinogenic risks of nitrate and chloride concentration in drinking water are of primary concern to many coastal regions across the world where most of the population depends on groundwater as a primary water source, especially in developing countries (Adimalla, 2019; Chakraborty et al., 2019; Rahaman et al., 2020; Rakib et al., 2019; Talukder et al., 2016), including Vietnam (Le et al., 2020; Tran et al., 2020). With this regard, the USEPA health risk assessment method was used to evaluate the non-carcinogenic effects of nitrate and chloride on adults (males, females), children, and infants. The total hazard indices for those groups are calculated based on input parameters integrating nitrate and chloride concentration in each sample. The scales for chronic and non-carcinogenic risk assessment were classified into four categories, including negligible ($HI < 0.1$), low ($0.1 < HI < 1.0$), medium ($1.0 \leq HI < 4.0$), and high ($HI \geq 4.0$) risks levels (Bortey-Sam et al., 2015; Egbi et al., 2020). In general, most groundwater samples (> 85%) in this study have $HI < 1.0$, indicating groundwater use in the study area is safe for human consumption (Figure 7). However, approximately 15% of groundwater samples may cause a severe health risk for consumers.

Health Risk Assessment for Adults

The HI values for adult males ranged from 0.01 to 15.0 with an average of 0.4 ± 1.3 (Table 6). Accordingly, approximately 93% of groundwater samples are below the male's actual safety limit, suggesting that groundwater resources are safe to use (Figure 7). However, around 7% of groundwater samples have their $HI > 1$ for males, indicating that they are probably exposed to the risks induced by nitrate contamination and saline water. Similarly, the HI value of adult females varied from 0.01 to 17.7 (average of 0.5 ± 1.5), with 91% of groundwater samples under the acceptable level.

The distribution of HI values among the four categories of risks for male and female individuals has a similar trend (Figure 7). However, the maximum HI value for the female group was 17.7, which was higher than the value of 15.0 for the male group. In other words, adult males show a stronger anti-risk ability against nitrate contamination and high salinity in groundwater. This result also indicated that human health risks for adult females are more significant than for adult males in the study area; this observation agrees with previous studies (Egbi et al., 2020; Zhai et al., 2017). For the health risk assessment approach, gender differences constitute an essential factor affecting the accuracy of health risk assessment results because of obvious differences in physiology and resistance to pollutants' interferences between adult males and females (Zhai et al., 2017).

The spatial distributions of non-carcinogenic index HI values for males (Figure 8a) and females (Figure 8b) indicated that high-risk areas (HI >1) were located mainly in the southern inland and estuarine regions. This is important to the local authorities as the inception of policies and mitigation solutions should be focused on these areas to protect residents from health risks caused by consuming unsuitable groundwater.

Health Risk Assessment for Children and Infants

Children and infants are highly vulnerable groups to water contamination because of the lower resistance to pollutants' negative impacts when compared to adults (Zhai et al., 2017). Consequently, the HI values for children and infants were higher than that of adults with maximum values of 20.0 and 29.9, respectively (Table 6). The infant group has the highest HI values ranging from 0.01 to 20.0, in which 87 % of the study area had HI values <1 for infants, i.e., no severe effects on the most sensitive population group. More specifically, infants' health risks were classified into negligible (13%), low (72%), medium (12%), and high risk (3%) (Figure 8d). Similarly, HI values of children indicated that about 91% of the groundwater samples have an acceptable level of water quality, e.g., negligible category (24%) and low-risk category (67%). On the other hand, approximately 10 % of the children population is exposed to significant risks: medium (7%), and high (2%) (Figure 8c).

In short, the health risks of different population groups in the study area were relatively high. The highest nitrate and saline concentrations were found in the inland and coastal areas (Figures 2, 3). Consequently, the population in these regions are exposed to higher risks when compared to the rest of the province. In general, the average HI indices for different populations in the study area vary widely with the order of infants > adult females > children > adult males.

[Table 6]

[Figure 7]

[Figure 8]

5. Conclusion

This study provides the first comprehensive assessment into the suitability of groundwater quality for drinking and irrigation purposes and human health risks in Soc Trang province, one of the most vulnerable coastal regions facing water scarcity in the Mekong Delta, Vietnam. The following main findings from the present study are summarized below:

(1) High nitrate (>50 mg/L) and chloride (>1,000 mg/L) concentrations were found not only in shallow but also in deep aquifers with the maximum values of 260 mg/L and 16,000 mg/L, respectively.

(2) Integrating all datasets into the general groundwater quality index confirmed that 18% of groundwater samples were poor and very poor for drinking.

(3) Nearly 18% of total samples were unsuitable for irrigation purposes because of high salinity and solidity hazard. Long-term use of inappropriate groundwater for irrigation may cause high risks of deteriorating soil health and the formation of sodic soils. Therefore, this study draws more attention to actions and mitigation solutions to decrease the pressure on the available water resources in the impacted areas.

(4) Human health risks were coherent among different population groups and locations, in which the highest risks level of contamination was observed for infants (15.2%) and followed by adult females (8.5%), children (7.4%), and adult males (7.1%).

These findings have been able to elucidate insights into the current water quality of the groundwater resources of the Mekong Delta lowland region. We demonstrated that the current water quality was of vital concern as a significant proportion of wells were not suitable for drinking and irrigation purposes and could promote long-term non-carcinogenic health effects on the local populations. The situation will likely deteriorate with both the ever-intensifying anthropogenic activities and the rise of sea level. Collectively, these two pressures would foster seawater intrusion in this multi-layered aquifer system and amplify the negative impacts of substandard groundwater resources. Finally, appropriate management strategies and mitigation solutions should be put in place to ensure groundwater sustainability and protect public health in this critical area.

Acknowledgements

We'd like to express our thankfulness to HCMUT for supporting time for which Dr. Phu Le Vo can join along with us in this study.

Conflicts of Interest

The authors declare no conflict of interest.

Funding

This study was not supported by any funding.

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Revised manuscript (clean version)

Groundwater quality evaluation and health risk assessment in coastal lowland areas of the Mekong Delta, Vietnam

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Abstract

This study provides a comprehensive assessment of groundwater quality in the Mekong Delta (Vietnam) and their potential risks to human health. The dataset consists of 282 groundwater samples collected in dry and rainy seasons between 2017 and 2018. Multiple physio-chemical parameters, groundwater quality index (GWQI), and irrigation water quality indices (IWQI) were employed to evaluate the suitability of groundwater resources for drinking and irrigation purposes. Thereupon, the hazard quotient (HQ), and total hazard index (HI) were used to assess the non-carcinogenic risks to different demographic groups. GWQI indicates that groundwater samples are excellent (41.5%), good (40.4%), poor (17%), and very poor (1.1%) classes for drinking purpose. The total hazard index (HI) indicated that 15.2%, 8.5%, 7.4%, and 7.1% of samples express non-carcinogenic health threats to infants, females, children, and males. Irrigation water quality indices (IWQI), namely, EC, sodium adsorption ratio (SAR), residual sodium bicarbonate (RSBC), soluble sodium percent (SSP), permeability index (PI), and magnesium hazard (MH), reveal that 82.2%, 94%, 20.2%, 69.6%, 45.4% and 12.8% of samples have EC < 2,250 µS/cm, SAR < 26, RSBC <1.25, SSP <60%, PI > 75%, and MH < 50%, respectively, which are suitable for irrigation use. Wilcox diagram shows that 17.54 % of total groundwater samples exceed the permissible levels of salt-tolerant crops and solidity hazards in the study area. Furthermore, high concentrations of nitrate (>50 mg/L) and chloride (> 1,000 mg/L) were detected in the shallow, intermediate, and deep aquifers. GWQI and IWQ maps imply that groundwater in the South-western area is suitable, whereas groundwater in the central area is not recommended for any purpose. These findings provide useful insights into the vulnerable groundwater system of the coastal lowland area of the Mekong Delta related to human activities (agricultural practices, dam construction) and global climate change impacts. Accordingly, appropriate management strategies for water resources and mitigation solutions are imperatively needed to ensure the sustainability of the groundwater resource and the protection of public health in the Mekong Delta.

Keywords: Multi-groundwater Quality Assessment, Human Health Risk Assessment, Geochemistry, Coastal aquifers, Mekong Delta

1. Introduction

Groundwater resources play an essential role in human consumption, irrigation purpose, and industrial development (Adimalla, 2019; Velis et al., 2017). It has extremely become a vital freshwater resource in coastal lowland regions, where surface water is scarce due to seawater intrusion and pollution (Alcérreca-Huerta et al., 2019; Lu et al., 2020). Over the recent decades, the degradation of groundwater quality was mainly associated with intensive applications of toxic chemicals (e.g., inorganic contaminants, pesticides, fertilizers) posed by agricultural and rural development activities and excessive discharge of waste and wastewater without proper treatment (Chen et al., 2020; Habib et al., 2020; Hossain and Patra, 2020). As a result, the long-term use of polluted groundwater for drinking and irrigation purposes leads to severe human health effects such as (i) cardiovascular diseases (Bai et al., 2019; Chabukdhara et al., 2017; Muhammad et al., 2011; Tran and Nguyen, 2018; Ustaoğlu, 2020; Wen et al., 2019; Yang et al., 2020), (ii) cancers (Kaur et al., 2019; Mohammed Abdul et al., 2015; Mondal et al., 2008; Rasheed et al., 2016; Saha and Rahman, 2020) and also adverse impacts on the natural ecosystems (Bartzas et al., 2015; Szymczycha et al., 2020).

In addition to toxic chemicals, nitrate contamination and salt intrusion in coastal groundwater systems have increasingly become a global issue (Knoll et al., 2019; Pazhuparambil Jayarajan and Kuriachan, 2020; Torres-Martínez et al., 2021; Wu et al., 2019). High nitrate concentrations and salinity in groundwater are not only detrimental to human health but also to agricultural ecosystems (Bartzas et al., 2015; Egbi et al., 2020; Lu et al., 2020; Marghade et al., 2020; Zolekar et al., 2020). First, nitrogen pollution, especially nitrate, is considered a critical pollutant of groundwater resources (Adimalla and Qian, 2021; Egbi et al., 2020). It is directly associated with serious disease such as methemoglobinemia, colorectal cancer, thyroid disease, and neural tube defect (Ward et al., 2018). Long-term ingestion of high nitrate concentrations through groundwater is the leading cause of elevated non-carcinogenic risks in the Indian population, especially infants and children (Adimalla et al., 2019; Adimalla and Qian, 2021; Adimalla et al., 2020). Negative impacts of nitrate contamination on human health were also observed in several regions worldwide (Barakat et al., 2019b; Emenike et al., 2018; Ijumulana et al., 2020; Li et al., 2021; Naderi et al., 2020; Rezaei et al., 2019). Second, the increase in dissolved concentrations of major ions (salinity) became an issue of imminent concern (Rahaman et al., 2020). Global climate change (e.g., sea-level rise) and human activities (e.g., unsustainable groundwater withdrawal, dam construction along major rivers) are the major factors instigating seawater intrusion into both surface and groundwater systems (Alcérreca-Huerta et al., 2019; Telahigue et al., 2020; Zeynolabedin et al., 2020). Such an increase in the ionic compositions of freshwater has a major effect on the ecosystems (Bugica et al., 2020; Liang et al., 2020; Zhu et al., 2020) and also cause severe health problems (Mitchell et al., 2019; Rakib et al., 2019; Rakib et al., 2020; Shukla and Saxena, 2020; Wen et al., 2019). Meanwhile, long-term consumption of water containing salt and total hardness (Ca and Mg) concentrations, in excess, increases the risk of hypertension (Shammi et al., 2019; Vineis et al., 2011), coronary heart disease (Park and Kwock, 2015; Rahaman et al., 2020), and chronic kidney disease (Naser et al., 2017; Rahaman et al., 2020; Ravindra et al., 2019). On the other hand, irrigation water with high salinity and total hardness can deteriorate soil fertility and thus reduce crop yields (Korres et al., 2019; Parvin et al., 2019; Radanielson et al., 2018).

The deterioration of groundwater quality by different pollutants could severely impact the ecosystem and human health. This is a persistent issue that could eventually worsen because of increasing pressure on groundwater resources associated with growing populations and anthropogenic activities (e.g., intensive agriculture and industries). Therefore, groundwater quality and human health risk assessment are critical to protect the people relying on groundwater and ensure food safety (Adimalla et al., 2019; Kaur et al., 2020; Rabeiy, 2018; Rezaei et al., 2019; Shukla and Saxena, 2020; Singh et al., 2019; Singh et al., 2020; Wagh et al., 2020; Wen et al., 2019; Yin et al., 2020; Zhang et al., 2020; Zhang et al., 2019).

The Mekong Delta (MD) has been recognized as one of the most vulnerable regions to climate change and sea-level rise globally (Dang et al., 2018; Shrestha et al., 2016; Smajgl et al., 2015). Although water resources play a crucial role in socio-economic development in the Mekong Delta, water quality has been increasingly deteriorated because of intensive human activities and natural variations (Dao et al., 2020; Ha et al., 2019; Hoang and Bäumle, 2019; Hung Van et al., 2019; Thanh Giao et al., 2021; Thanh Nguyen, 2020; Thu Minh et al., 2020; Tran et al., 2020). It is especially applicable to the coastal regions because of severe water pollution and seawater intrusion affecting both surface and groundwater systems (Dang et al., 2020; Dao et al., 2020; Thanh Nguyen, 2020; Tran et al., 2021; Tran et al., 2020; Trung and Tri, 2014). Intensive agricultural development and rapid industrialization in combination with extreme drought events and seawater intrusion have resulted in severe degradation of surface and groundwater resources (Buschmann et al., 2008; Chea et al., 2016; Nguyen et al., 2020; Tran et al., 2020). Recent studies indicated that the long-term and unsuitable use of groundwater for drinking and irrigation purposes amplified human health problems and also threatened agricultural productions in the Mekong Delta (Braun et al., 2019; Le Luu, 2017; Merola et al., 2015; Reid et al., 2021; Toan et al., 2013; Tran et al., 2020). However, there are few studies providing a quantitative assessment of groundwater quality for drinking and irrigation purposes. Moreover, it is necessary to evaluate potential human health risks caused by long-term consumption of saline and nitrate-rich groundwater in coastal lowland regions of the Mekong Delta. Hence, this research aims to evaluate the links between groundwater quality and its suitability for use as irrigation and drinking water and potential human health risk. The specific objectives of this study are to (1) evaluate the suitability of groundwater for drinking purposes, (2) assess potential effects and health risks to different population groups due to high chloride and nitrate contaminations.

2. Study Area

The study is conducted in Soc Trang, a coastal province located in the Southeast of the Mekong Delta (Vietnam), covering approximately 3,310 km^2 with a flat and low elevation, ranging from 0.5 to 2.5 m (Figure 1). The study area is known for its tropical monsoon climate system with two distinct seasons: the dry season starts from May to November and the rainy season lasts from December to April of the following year. The average annual rainfall is about 1,770 mm with high seasonality: about 85% of the annual rainfall occurs during the rainy season. In the study area, land use/land cover is dominated by agricultural land (84.8% of total area), followed by residential land (5.2%) and forested land (4.40%) (Tran et al., 2021). Additionally, Soc Trang province has various soil types, including sandy, alluvial, salt, and acid sulphate soils (Kawahigashi et al., 2008). Economically, most of the local population depends primarily on agricultural activities, which contribute to 42% of the total provincial GDP. Therefore, the demand for freshwater resources in this province is incredibly high, approximately 25 million cubic meters per year (field survey data, data not shown).

The study site has a dense surficial drainage network with a density of 2.21 km/km^2 (Tran et al., 2021). The drainage network directly connects the Hau River (the Bassac River) to the Northeast and the East Sea to the Southeast. Therefore, the surface and subsurface flow dynamic in the area is driven by meteorological forces and flow regimes of the Hau River streamflow and tidal regimes of the east Sea (Tran et al., 2020; Tran et al., 2019). Together, sands, fluvial deposits, and gravels with various grain sizes dominate sedimentary deposits (Tran et al., 2020; Tran et al., 2019) also influence on the groundwater systems of the study area. The aquifer system consists of seven hydrogeological units, including Holocene (qh), Upper Pleistocene (qp_3), Middle Pleistocene (qp_{23}), Lower Pleistocene (qp_1), Middle Pliocene (n_{22}), Lower Pliocene (n_{21}), and Upper Miocene (n_{13}) (Tran et al., 2020; Wagner et al., 2012). Each hydrogeological unit has the upper and lower layers with different lithological characteristics. The top layer consists of silt, clay or mixed silt clay with low water yield (Wagner et al., 2012). In contrast, the lower part is relatively permeable, consisting of fine to coarse sand, gravel, and pebbles (Hoang and Bäumle, 2019; Tran et al., 2020). Groundwater in the Pleistocene aquifers and Upper Miocene are the primary supply source of drinking water because of its high yield and good quality compared to other aquifers (Tran et al., 2018). However, long-term excessive groundwater extraction has caused an increase in chloride and nitrate concentrations in the aquifer system (Tran et al., 2020). Indeed, Soc Trang frequently faces extreme seawater intrusion events causing severe water scarcity, especially in the dry season.

[Figure 1]

3. Materials and Methods

3.1. Groundwater sampling and chemical analysis

A total of 282 groundwater samples were collected from 145 wells in the dry and rainy season between 2017 and 2018 in Soc Trang (Figure 1). The wells are either household tube wells of Soc Trang Water Supply Company and Center for Rural Water Supply and Sanitation and boreholes from the national groundwater monitoring system. The sampling depths varied from 3.5 m to 485 m below ground surface to cover all aquifer layers. Physical and chemical parameters such as pH, electrical conductivity (EC), dissolved oxygen (DO), and total dissolved solids (TDS) were measured directly at the field by using Hanna Instruments portable meters. Each groundwater sample was collected after 15-20 minutes of purging until pH, EC, and DO values were stabilised. All groundwater samples were stored in 100 mL polyethylene bottles, kept at dark and 4^{o}C. Subsequently, all groundwater samples were filtered through 0.2-µm cellulose ester filters.

Physio-chemical properties and compositions of groundwater were analysed according to the standard procedures of the American Public Health Association (E.W. Rice, 2017) and were described in detail in a previous publication (Tran et al., 2020). Briefly, chemical analyses were performed at the Hydrology and Water Environment Laboratory, University of Tsukuba, Japan. Major cations (Na^+, K^+, Ca^{2+} and Mg^{2+}) were measured by ICP-OES, Perkin Elmer Optima 7300. Main anions (Cl^-, NO_3^-, and SO_4^{2-}) were analysed using Ion Chromatography (Shimadzu, Japan). Bicarbonate (HCO_3^-) was determined by the titration method. Finally, ion balance was calculated for each sample; the error was below 5%, justifying the accuracy of the chemical analyses.

3.2. Drinking groundwater quality index

Groundwater quality index (GWQI) is widely used to assess general groundwater quality by converting multiple groundwater quality parameters into a single value (Adimalla and Qian, 2019; Barakat et al., 2019a; Ghouili et al., 2018; Zhai et al., 2017). Therefore, the suitability of the groundwater for use as drinking water could be determined based on GWGI-derived water quality status (Table 1). To determine GWQI (Eq.1), each parameter was assigned with a specific weight value (W_i) depending on its potential to affect groundwater quality for drinking purposes. The overall GWQI was calculated linearly by adding the quality rating (Q_i) to the unit weight (W_i).

$$GWQI = \frac{\sum Q_i \times W_i}{\sum W_i} \qquad (1)$$

$$where\ Q_i = \frac{(C_o - C_i)}{(S_i - C_i)} \times 100 \qquad (2)$$

$$W_i = \frac{K}{S_i} \qquad (3)$$

$$K = \frac{1}{\sum 1/S_i} \qquad (4)$$

Where Q_i is a water quality rating for the i^{th} parameter (Eq.2), W_i is a unit weight for the i^{th} parameter (Eq.3), S_i is a standard value of the i^{th} parameter, C_o is an observed value of the n^{th} parameter, C_i is an ideal value of i^{th} parameter, and K is a constant for proportionality (Eq.4). Here, the main parameters used for GWQI calculations are provided in Table 2.

[Table 1]

[Table 2]

3.3. Irrigation water quality indices (IWQI)

Natural groundwater often has high concentrations of dissolved ions because of geochemical reactions (e.g., mineral dissolution, ion exchange). Therefore, using groundwater for irrigation could adversely affect the physiochemical conditions of soils and crops (Adimalla and Qian, 2019). To evaluate the suitability of groundwater for irrigation, there are several indices, including sodium adsorption ratio (SAR), residual sodium bicarbonate (RSBC); soluble sodium percent (SSP), permeability index (PI), and magnesium hardness (MH) (Anim-Gyampo et al., 2019; Sabarathinam et al., 2020). The relations used to estimate these indices are listed from Eq. 5 to Eq. 9, respectively. According to these calculations, we classified groundwater samples as per their suitability for irrigation.

$$\text{Sodium adsorption ratio (SAR)} = \frac{Na^+}{\sqrt{\frac{Ca^{2+} + Mg^{2+}}{2}}} \qquad \text{(Richards, 1954)} \qquad (5)$$

Residual sodium bicarbonate (RSBC) $= HCO_3^- - Ca^{2+}$ (Gupta, 1983) (6)

Soluble Sodium Percent (SSP) (Todd, 1980) (7)

$$\text{SSP} = \left(\frac{Na^+ + K^+}{Na^+ + Ca^{2+} + Mg^{2+}}\right) \times 100$$

$$\text{Permeability index (PI)} = \frac{Na^+ + \sqrt{HCO_3^-}}{(Na^+ + K^+ + Ca^{2+} + Mg^{2+})} \times 100 \quad \text{(Doneen, 1964)} \quad (8)$$

$$\text{Magnesium hazard (MH)} = \frac{Mg^{2+}}{Ca^{2+} + Mg^{2+}} \times 100 \quad \text{(Szaboles, 1964)} \quad (9)$$

3.4. Health risk assessment

Human health problems are often related to long-term consumption of unsafe water sources caused by natural and anthropogenic contaminants in surface and subsurface environments. Therefore, assessing the potential health risks caused by substandard groundwater quality requires intensive investigation of water supply quality. However, water quality intimately depends on the hydrogeographic and hydrogeochemical conditions of each specific geographical location. Therefore, the study conducted a health risk assessment (HRA) to determine the potential impacts of contaminated water on human health. Accordingly, we calculated the chronic daily intake (CDI) by intake and dermal contact of two contaminants, nitrate (>50 mg/L) and chloride (> 1,000 mg/L), as they represent the chemicals of highest concern in drinking water for non-carcinogenic risk assessments (Adimalla, 2020; Pazhuparambil Jayarajan and Kuriachan, 2020; Saleh et al., 2019). The exposures of such pollutants to humans are calculated using Eq.10 and Eq.11 (Ali and Chidambaram, 2021; Proshad et al., 2020).

$$CDI_{oral} = \frac{C_i \times IR \times EF \times ED}{BW \times AT} \quad (10)$$

$$CDI_{dermal} = \frac{C_i \times K_p \times IR \times ET \times EF \times ED \times CF \times SA}{BW \times AT} \quad (11)$$

CDI_{oral} and CDI_{demal} are exposure doses through ingestion and dermal absorption, respectively in mg/kg/day. C_i is the concentration of chloride and nitrate in groundwater (mg/L). IR is the ingestion rate of water per day. EF is exposure frequency in the number of days per year. ED is exposure duration (years). BW is average body weight (kg). AT is averaging time (days). K_P represents a dermal permeability coefficient in water (cm/h). ET is the exposure time (h/day). CF is the unit conversion factor (L/cm^3). SA is skin exposure that was calculated by using Eq.12 (Barakat et al., 2019b), where H is the average height of the population. To consider various physiological characteristics and health conditions of the populations, we considered in this study four subpopulations, which correspond to adult males, adult females, children, and infants. The selected parameters for each sub-population were presented in **Table 3**.

$$SA = 239 \times H^{0.417} \times BW^{0.517} \quad (12)$$

[Table 3]

The calculated CDI allows the non-carcinogenic hazard quotient (HQ) determination through oral and dermal pathways using Eq.13 and Eq.14. The total hazard index (HI) was calculated as the sum of the hazardous quotient (Eq.15). These calculations directly reflect the risks that the exposed population may develop serious health problems under specific conditions of exposures (e.g., concentrations of the pollutants, exposure duration).

$$HQ^{i}_{oral} = \frac{CDI_{oral}}{RfD_{oral}} \quad (13)$$

$$HQ^{i}_{dermal} = \frac{CDI_{dermal}}{RfD_{dermal}} \quad (14)$$

$$HI^{i} = \sum_{i=1}^{N} HQ^{i} \quad (15)$$

In these calculations, i reflects the water quality indicators (i.e., chloride and nitrate). RfD_{oral} and RfD_{dermal} are the reference dose for non-carcinogenic risks through the oral intake and dermal absorption pathways. It is important to note that HQ_{oral} and HQ_{dermal} depend significantly on the selected parameters used in Eq. 10 and Eq. 11, respectively (Karunanidhi et al., 2020). Also, in order to accurately assess the overall health risks associated with ingestion and dermal pathways, it is necessary to take into consideration as much as possible the solutes in water sources that are deemed harmful. In this light, chloride and nitrate parameters were selected to assess. Chloride reference dose (78.68 mg/kg/day) was used for both oral and dermal pathways (USEPA 2018) whilst nitrate reference dose of 1.6 and 0.8 mg/kg/day was applied for oral and dermal contact, respectively (ECHA, 2020; USEPA, 2018).

4. Results and Discussion

4.1. Groundwater quality for drinking purposes

4.1.1. Physio-chemical characteristics

EC is a fundamental parameter reflecting the ionic concentrations or concentrations of dissolved salts in a water sample. In this study, groundwater has a wide range of variation in EC values, ranging from 117 to 18,500 µS/cm (average of

1,755 ± 2,250 µS/cm, Table 4). EC values show a heterogeneous spatial distribution (Figure S1a). Most of the high saline groundwater samples are found in shallow wells (depth < 40 m, n = 19), excepting a small number of samples (n=4) in deep wells (> 40m) (Figure S1b). Such a spatial heterogeneity of EC can be attributed to several factors such as minerals dissolution, seawater intrusion, and human impacts (Han and Currell, 2018; Hoang and Bäumle, 2019; Tran et al., 2020).

[Table 4]

The range of pH variations could indicate the origin of groundwater and geochemical processes during water movement from recharge throughout porous media to discharge areas. Groundwater in the study area has a relatively wide variation in pH, ranging from 6.1 to 9.3 (average of 7.3±0.5, Table 4). Alkaline groundwater samples (pH>7.5) were observed in inland to coastal areas (southern bound direction, Figure S1c). These high pH values are commonly found in deep aquifers (depth > 200m, Figure S1d) and are associated with the influence of the bicarbonate/carbonate system (Tran et al., 2020). Relative to the drinking water standard (pH=6.0-8.5), only three groundwater samples out of 282 exceeded the maximum permissible limit of 8.5 for drinking water. However, 15 samples have pH values below the minimum acceptable limit of 6.0. This issue was attributed to the process of iron oxidation occurring in the groundwater of the Mekong Delta (Ha et al., 2019).

Total dissolved solids (TDS) consist of major dissolved compounds in groundwater such as sodium, potassium, calcium, magnesium, chloride, sulphates, nitrate, and bicarbonate. For this study, groundwater TDS was in the range of 82-12,950 mg/L (average of 1,228 ±1,576 mg/L, Table 4). According to the WHO drinking water standard (WHO, 2017), acceptable drinking water quality should have TDS values < 500 mg/L and is unpalatable if TDS >1,500 mg/L. Accordingly, 31.6% of the groundwater samples are safe for drinking while at least 19.5% are unpalatable. High TDS (>1,500 mg/L) was observed in both inland and coastal areas (Figure S1e), especially in deep wells (Figure S1f), indicating influences of groundwater salinization (Tran et al., 2020). It was estimated that approximately 35% of groundwater samples exceed the maximum concentration of TDS (1,000 mg/L) based on the National Technical Regulation on Domestic Water Quality (QCVN 01-1:2018/BYT) as shown in Table 4.

Overall, concentrations of major elements were in the following order of dominance: $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ for cations and $Cl^- > HCO_3^- > SO_4^{2-} > NO_3^-$ for anions. Sodium and Cl^- showed higher concentrations than other ions, and their Na^+/Cl^- ratios approximated 0.86 (characteristic of seawater composition), suggesting seawater intrusion into the shallow aquifer system of the study area (Xiong et al., 2020). Although a certain amount of sodium daily intake is essential to ensure human health, exceeding sodium intake causes adverse health risks, especially hypertension and nausea diseases (Park and Kwock, 2015; Rahaman et al., 2020). Therefore, sodium is one of the most important parameters for human health risk assessment. In this study, sodium concentrations varied from 17 mg/L to 8,536 mg/L (average of 297 ± 683 mg/L) while potassium concentrations ranged from 0.5 to 279 mg/L (average of 19± 26 mg/L) (Table 4). High sodium and potassium concentrations were mainly observed in shallow aquifers of coastal areas (Figure S1i, S1k) (Figure S1j, S1l), indicating that seawater intrusion during the Holocene period may play an important factor in the accumulation of these ions (Meyer et al., 2019). Accordingly, more than 30% and 43% of groundwater samples in the study area exceed the maximum limit of sodium and potassium, respectively.

Calcium and magnesium are also important elements for human health; however, excessive uptake of these ions leads to several health problems, especially kidney stones and hypertension (Cormick and Belizán, 2019; Ndii et al., 2020). Concentrations of calcium varied from 0.1 to 927 mg/L (average of 45±82 mg/L), while magnesium concentrations ranged from 0.03 mg/L to 693 mg/L (average of 46 ±81 mg/L, Table 4). According to WHO and QCVN 01-1:2018/BYT, 2% and 4% of total groundwater samples in this study area exceeded the maximum calcium and magnesium concentrations, respectively. Total hardness (TH) reflects the hardness of water caused by calcium and magnesium (Adimalla and Qian, 2019). However, the variation of TH concentrations corresponds to different classes of water hardness. For instance, TH values < 75 indicate soft water, while TH in the ranges of 75-150, 151-300, and >300 mg/L are related to moderate, hard, and very hard water, respectively (Adimalla and Qian, 2019). The TH value of groundwater in this area varied from 13 mg/L to 3,080 mg/L (average of 280 ± 377 mg/L). Only 5.7% of groundwater samples in this area exceeded the maximum permissible limit of 500 mg/L for drinking purposes (WHO, 2017). High TH concentration (>300 mg/L) in groundwater was observed in some locations in inland and coastal areas (Figure S1g) in shallow aquifers. Meanwhile, deeper aquifers show relatively low TH (< 150 mg/L) (Figure S1h). We have previously demonstrated that the precipitation of minerals (e.g., calcite) could be responsible for decreasing soluble Ca/Mg in those deep aquifers (Tran et al. 2020).

Besides, chloride concentrations in this area ranged from 2 mg/L to 15,883 mg/L (average of 413 ± 1,312 mg/L) (Table 4). High chloride concentration (> 1,000 mg/L) was found not only in coastal areas but also in inland areas far from the East Sea (Figure 2a) as well as both shallow and deep wells (Figure 2b). This observation implies the complexity of past and presents seawater intrusion processes in this area (Hoang and Bäumle, 2019; Pham et al., 2019;

Tran et al., 2020). Approximately 28% and 23% of groundwater samples in the study area exceeded chloride concentration compared to the maximum concentration of 600 mg/L and 300 mg/L as regulated by WHO and QCVN 01-1:2018/BYT (Table 4), respectively. This result, coupled with spatial heterogeneity of salinity at the aquifer system, may exacerbate groundwater quality and put high risks to the water supply system in this study area.

Finally, more than 70% of groundwater samples have relatively high bicarbonate concentrations (>200 mg/L), while only 4% of groundwater samples exceed 200 mg/L of sulphates. This could be associated with pyrite oxidation leading to an accumulation of sulphate in groundwater (Ha et al., 2019).

[Figure 2]

Nitrate is among a few representative compounds for anthropogenic impacts on the aquatic environment (Egbi et al., 2020; Singh et al., 2019). Indeed, high nitrate concentrations in groundwater are mainly related to domestic waste, agricultural fertilizers, industrial waste, and wastewater (Egbi et al., 2020). For several decades, nitrate contamination has been a global problem, deteriorating groundwater quality and causing severe health problems in many countries worldwide (Abdelwaheb et al., 2019; Adimalla and Qian, 2019; Njuguna et al., 2020; Wagh et al., 2020). Nitrate concentrations in groundwater samples ranged from 0.1 mg/L to 260 mg/L with an average of 9±26 mg/L (Table 4). Relatively high nitrate concentrations (>10 mg/L) were observed in the urban area of Soc Trang city in an aquaculture field along the main river and on the coastline (My Xuyen and Vinh Chau, Figure 3a), which may be attributed to anthropogenic emissions. Additionally, high nitrate concentration (> 20 mg/L) was also found in many deep wells (depth > 40 m, n = 16) (Figure 3b). The contamination of nitrate in these deep wells may be associated with leaking artificial nitrate sources from landfills, urban and agricultural areas via unprotected wells (Bourke et al., 2019; Lockhart et al., 2013; Pastén-Zapata et al., 2014; Tran et al., 2020).

In summary, this assessment of groundwater quality in this area revealed that saline water and nitrate contaminations are significant issues exceeding the WHO drinking water standards. As groundwater is the primary source of drinking water in coastal regions of the Mekong Delta, the results suggested that long-term consumption of this groundwater could lead to severe human health problems for the local population. Therefore, assessing human health risks related to high nitrate and chloride concentration in the water supply system in this area is an imperative need.

[Figure 3]

4.1.2. Groundwater quality index (GWQI)

Groundwater quality index (GWQI) allows assessment of groundwater quality, and it was classified into five categories corresponding to different levels of the suitability of the groundwater for use as drinking water (Table 1). In this study, GWQI values ranged from 8.5 to 284.7 (average of 63.8 ± 40.6 Table 6). Accordingly, groundwater quality was classified into excellent (41.5%), good (40.4%), poor (17%), and very poor (1.1%) for drinking purposes. It was noted that GWQI has a heterogeneous spatial distribution in the study area. For example, poor (GWQI =101 - 200) and unusable groundwater quality (GWQI =201- 300) were found in the north-western regions and along rivers to the East Sea (Figure 4). It was attributed to the fact that an ongoing and intensive extraction of groundwater in these areas may further reduce the availability of groundwater resources (Tran et al., 2021; Tran et al., 2020). Therefore, alternative solutions should be adapted, and pumping activities should be minimized in the North-western and coastal areas of Soc Trang province.

[Figure 4]

4.2. Irrigation water quality

Groundwater quality assessment also plays a vital role in sustainable agricultural development in the region (Hoang and Bäumle, 2019; Tran et al., 2020). Hence, we employed a combination of indices (EC, SAR, RSBC, SSP, PI, and MH) as shown in Table 5 and Figure 5, and Wilcox's graphic (Richards, 1954) (Figure 6) was used to evaluate the suitability of the groundwater source for irrigation purposes. It is widely accepted that high sodium concentrations lead to the formation of sodic soil while exceeding salinity causes soil salinization; both are detrimental to plant development (Kurunc et al., 2020; Machado and Serralheiro, 2017). In addition, high concentrations of sodium and carbonate in irrigation water inhibit plant growth and subsequently reduce crops yield (Kurunc et al., 2020; Phogat et al., 2020). Therefore, electrical conductivity and sodium concentrations are simple and crucial indicators in classifying irrigation water (Kurunc et al., 2020; Taloor et al., 2020).

According to the classification of irrigation water (Wilcox, 1948), we categorized the groundwater samples into four classes based on their electrical conductivity (Table 5). These classes reflect their suitability to be used for irrigation, as excellent, good, permissible, and unsuitable; 1.4%, 33.3%, 47.5%, and 17.7% of our groundwater samples fall into these categories, respectively (Tables 5). In other words, almost one in every five groundwater samples of this study area is too saline that could negatively influence the practices of sustainable agricultural development. More importantly, SAR is a valuable index for the contribution of Na^+ relative to the sum of divalent cations (Richards, 1954). Higher SAR reflects the situation where a high proportion of Na^+, relative to Ca^{2+} and Mg^{2+}, could saturate the cation exchange complexes in soil and thus decrease soil permeability (Kurunc et al., 2020; Taloor et al., 2020). In this area, SAR values of groundwater varied from 0.3 to 51.8 with an average of 6.8 ± 9.2. High SAR values (> 26, or unsuitable groundwater for irrigation) correspond to 6% of the total dataset. In other words, this groundwater of substandard quality, if used for agricultural purpose, may damage soil structure, and form sodic soils.

Moreover, another indicator of long-term exacerbates soil health risks is the SSP index, representing the relative contribution of alkali metals (Na^+ and K^+) to the total cation charges (Eq. 7). SSP values ranged widely between 14.3% and 98.3%, with an average of 58.5 ± 22.7%. It was estimated that 40% of groundwater samples were doubtful and unsuitable for irrigation due to the potential impacts of Na^+ and K^+ on the soil structure. Additionally, it is also essential to identify the chemical speciation of these alkali metals in groundwater to correctly identify their impacts on arable soils. Carbonate is a crucial ligand in groundwater; high carbonate concentrations in groundwater often result in precipitation of Ca^{2+} and Mg^{2+}. However, the residual carbonate could form complexes with sodium that significantly decrease soil permeability and refer to the RSBC index. Accordingly, RSBC values varied from -39.9 to 12.5 with an average of 3.1 ± 5.0, in which nearly 69% of groundwater samples were unsuitable for irrigation based on the RSBC indices. The estimation of permeability index (PI %) values (Eq. 8) showed that 54.6% of total groundwater samples in the study area are inappropriate for irrigation because of low permeability.

In summary, most of the groundwater aquifers in this area are unsuitable for irrigation purpose. This is mainly due to the alkali metals (Na^+ and K^+) and their dissolved chemical complexes (carbonate). More noticeable, the alkali earth metals (Ca^{2+}, Mg^{2+}) are an immediate concern as 87.2% of groundwater samples in the study area have high MH values (> 50%). Integration of EC and SAR as the two significant factors associated with the poor quality of the groundwater was presented in the Wilcox diagram (Figure 6). This approach allowed an overall classification of groundwater quality as excellent, good, permissible, and unsuitable (Richards, 1954). The result shows that 101 samples (35.4%) fall under the excellent and good water classes for irrigation while 49 samples (17.5%) fall under the unsuitable water class sodium for irrigation. All these indexes pointed out that unsuitable groundwater is observed in the inland (Soc Trang city and My Tu district) and coastal areas (Tran De and Vinh Chau districts) (Figure 5), indicating the need for alternatives to the usage of groundwater for irrigation in these specific locations. Therefore, these results call for imminent actions to protect groundwater resources for the agricultural sector, which play a crucial socio-economic factor in the Mekong Delta coastal area.

[Table 5]

[Figure 5]

[Figure 6]

4.3. Health risk assessment

Long-term consumption of groundwater containing high nitrate and salt concentrations causes serious human health problems (Rahaman et al., 2020; Rakib et al., 2019). Therefore, non-carcinogenic risks of nitrate and chloride concentration in drinking water are of primary concern to many coastal regions across the world where most of the population depends on groundwater as a primary water source, especially in developing countries (Adimalla, 2019; Chakraborty et al., 2019; Rahaman et al., 2020; Rakib et al., 2019; Talukder et al., 2016), including Vietnam (Le et al., 2020; Tran et al., 2020). With this regard, the USEPA health risk assessment method was used to evaluate the non-carcinogenic effects of nitrate and chloride on adults (males, females), children, and infants. The total hazard indices for those groups are calculated based on input parameters integrating nitrate and chloride concentration in each sample. The scales for chronic and non-carcinogenic risk assessment were classified into four categories, including negligible ($HI < 0.1$), low ($0.1 < HI < 1.0$), medium ($1.0 \leq HI < 4.0$), and high ($HI \geq 4.0$) risks levels (Bortey-Sam et al., 2015; Egbi et al., 2020). In general, most groundwater samples (> 85%) in this study have $HI < 1.0$, indicating groundwater use in the study area is safe for human consumption (Figure 7). However, approximately 15% of groundwater samples may cause a severe health risk for consumers.

Health Risk Assessment for Adults

The HI values for adult males ranged from 0.01 to 15.0 with an average of 0.4 ± 1.3 (Table 6). Accordingly, approximately 93% of groundwater samples are below the male's actual safety limit, suggesting that groundwater resources are safe to use (Figure 7). However, around 7% of groundwater samples have their $HI > 1$ for males, indicating that they are probably exposed to the risks induced by nitrate contamination and saline water. Similarly, the HI value of adult females varied from 0.01 to 17.7 (average of 0.5 ± 1.5), with 91% of groundwater samples under the acceptable level.

The distribution of HI values among the four categories of risks for male and female individuals has a similar trend (Figure 7). However, the maximum HI value for the female group was 17.7, which was higher than the value of 15.0 for the male group. In other words, adult males show a stronger anti-risk ability against nitrate contamination and high salinity in groundwater. This result also indicated that human health risks for adult females are more significant than for adult males in the study area; this observation agrees with previous studies (Egbi et al., 2020; Zhai et al., 2017). For the health risk assessment approach, gender differences constitute an essential factor affecting the accuracy of health risk assessment results because of obvious differences in physiology and resistance to pollutants' interferences between adult males and females (Zhai et al., 2017).

The spatial distributions of non-carcinogenic index HI values for males (Figure 8a) and females (Figure 8b) indicated that high-risk areas (HI >1) were located mainly in the southern inland and estuarine regions. This is important to the local authorities as the inception of policies and mitigation solutions should be focused on these areas to protect residents from health risks caused by consuming unsuitable groundwater.

Health Risk Assessment for Children and Infants

Children and infants are highly vulnerable groups to water contamination because of the lower resistance to pollutants' negative impacts when compared to adults (Zhai et al., 2017). Consequently, the HI values for children and infants were higher than that of adults with maximum values of 20.0 and 29.9, respectively (Table 6). The infant group has the highest HI values ranging from 0.01 to 20.0, in which 87 % of the study area had HI values <1 for infants, i.e., no severe effects on the most sensitive population group. More specifically, infants' health risks were classified into negligible (13%), low (72%), medium (12%), and high risk (3%) (Figure 8d). Similarly, HI values of children indicated that about 91% of the groundwater samples have an acceptable level of water quality, e.g., negligible category (24%) and low-risk category (67%). On the other hand, approximately 10 % of the children population is exposed to significant risks: medium (7%), and high (2%) (Figure 8c).

In short, the health risks of different population groups in the study area were relatively high. The highest nitrate and saline concentrations were found in the inland and coastal areas (Figures 2, 3). Consequently, the population in these regions are exposed to higher risks when compared to the rest of the province. In general, the average HI indices for different populations in the study area vary widely with the order of infants > adult females > children > adult males.

[Table 6]

[Figure 7]