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Spatial Distribution and Potential Health Risk Assessment of Fluoride and Nitrate Concentrations in Groundwater from Mbour-Fatick Area, Western Central Senegal

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Abstract

This study aims to delineate the spatial distribution of nitrate and fluoride in groundwater and to estimate the non-carcinogenic risks using the human health risk assessment model recommended by the United States Environmental Protection Agency (USEPA). Forty-two samples were collected from wells and boreholes and analyzed for nitrate, fluoride and other water quality parameters. Results of the study indicate that fluoride and nitrate concentrations vary respectively from 0.13 to 9.41 mg·L^{-1} and from 0.13 to 432.24 mg·L^{-1} with respective median values of 2.65 and 13.85. About 69% of groundwater samples exceed the allowable limit (1.5 mg·L^{-1}) of fluoride for drinking water. Spatial distribution of fluoride shows high concentrations in certain localities with values ranging from 6.74 mg·L^{-1} to 9.41 mg·L^{-1}. The spatial distribution of nitrate indicates that the majority of water samples (87.18%) have nitrate concentrations lower than the World Health Organization (WHO) standard guideline value of 50 mg·L^{-1}. Assessment of non-carcinogenic risks associated with intake of polluted groundwater in local populations indicates that 82.05% and 87.18% of groundwater samples have a THI > 1 in adults and children, respectively. However, the highest THI value (15.87) was recorded for children suggesting that children face greater non-carcinogenic risks than adults. The results of this study can be used as a support by the policymakers and practitioners to develop appropriate policies for effective and sustainable groundwater management and to monitor human health implications.

Keywords

Fluoride, Nitrate, Groundwater Quality, Human Health Risk, Western Central Senegal

1. Introduction

Groundwater constitutes a vital resource worldwide, essential and compulsory for the development of all living organisms. It plays a very important role in the development of many human activities, including agriculture, commerce, industry, and domestic activities. However, strong demographic growth with its corollary of anarchic land use, the development of numerous anthropogenic activities as well as the establishment of industries, located in large cities, lead to a progressive deterioration of groundwater quality. Thus, groundwater pollution has become a global problem, reported by numerous studies [1]-[6]. In several countries, the contamination of groundwater by nitrate and fluorine constitutes a major concern which leads to a limitation of their exploitation for various uses, particularly for human consumption. Thus, groundwater pollution by fluorine is a recurring problem encountered in many countries around the world, particularly in Africa [7]-[9] in India [2] [3] [10] [11] in China [12] [13] in Korea [14] in Iran [15]. Fluorine is the thirteenth most abundant natural element in the earth's crust and it is widely dispersed. It is the most reactive and the most electronegative of all elements and, therefore, almost never occurs in nature in the elementary state but is found in the form of mineral fluoride complexes [15] [16]. The fluoride in groundwater may be related to natural factors or anthropogenic activities. Higher concentrations of fluoride in groundwater may be linked to the weathering and dissolution of fluoride minerals in rocks such as fluorite, apatite, fluorapatite, hornblende, topaz, villianmite [2] [14] [17] [18] micas and pyroxenes [2] [10] [19]. Furthermore, as OH^- and F^- have similar ionic sizes, OH^- can replace F^- in fluoride bearing minerals such as biotite or apatite, resulting in higher fluoride concentrations in groundwater [10] [11]. However, low fluoride concentrations in drinking water constitute an advantage since they allow normal bone mineralization and the formation of dental enamel [5] [13] [14] [19] [20]. In contrast, very low concentrations ($<0.5\ mg \cdot L^{-1}$) of fluoride in groundwater can cause health risks to the populations with the appearance of dental caries [21]. The desirable fluoride concentration in drinking water is 0.6 - 1.2 $mg \cdot L^{-1}$ and at this level it prevents dental caries and promotes bone development [17]. The maximum permissible limit for fluoride and nitrate in drinking water is 1.5 $mg \cdot L^{-1}$ and 50 $mg \cdot L^{-1}$ respectively [22]. Beyond 1.5 $mg \cdot L^{-1}$ and 50 $mg \cdot L^{-1}$ in water for fluoride and nitrate, respectively, there is a health risk, especially to vulnerable populations (children, the elderly, and pregnant women). In several regions of the world, the permissible limit value of fluoride and nitrate for drinking water is often greatly exceeded. For example, [4]-[7] [11] [15] [20] [23]-[26] reported high fluoride concentrations in groundwater in several countries around the world, thus constituting a major public health risk. Indeed, the consumption of water with high fluoride content can lead to the appearance of diseases such as dental fluorosis, skeletal fluorosis and bone deformation in children and adults [13] [23] [27]. In Senegal, the problem of fluorosis affects certain regions, such as the regions of Kaolack, Diourbel, Fatick and a part of the Thies region [28]. In the Senegalese basin, [29] found fluoride contents varying between very low levels and a maximum value of 13 $mg \cdot L^{-1}$. This high concentration of fluoride is recorded in the Paleocene aquifer approximately 35 km northeast of Mbour. According to [30] [31] the aquifer formations facing the fluoride problem are those of the lower and middle Eocene, the Paleocene and the Maastrichtian. The highest concentrations of fluoride are recorded in the Paleocene aquifer.

Nitrate is one of the most frequently reported contaminants in groundwater. In natural waters, its content is low, generally below 10 $mg \cdot L^{-1}$ [3] [32]. However, high nitrate concentrations are often recorded in groundwater. Groundwater nitrate contamination can be related to several factors such as excessive use of nitrogen fertilizers in agricultural areas, poor sanitation in urban areas, agricultural runoff, sewage systems, human and animal wastes, leaking septic tanks, manure, feedlots, dairy and poultry farming [13] [20]. Over the past few decades, groundwater nitrate pollution has become a global problem, which has been widely reported by numerous previous studies [21] [33] [34]. Thus, in order to protect the population from health risks, the WHO has set a standard guideline value of 50 $mg \cdot L^{-1}$ for drinking water. Furthermore, nitrate is a nitrogen compound that is naturally present in many environments at moderate levels. In groundwater, the increase of nitrate contents can have a natural or anthropogenic origin. Natural origin of nitrate can be linked to the biological fixation of organic nitrogen in the soil, promoting the formation of nitrate through the nitrification process [35]. However, its anthropogenic origin can be associated with agriculture through the use of fertilizers in agricultural areas or with the lack of sanitation in urban areas, the use of septic tanks, domestic organic waste, sewage, leaking municipal sewers, human and animal waste, etc. [20]. Regular consumption of water with high nitrate concentrations can lead to certain diseases, such as methemoglobinemia, particularly observed in infants, and also to the development of stomach cancer [36]. The study area is characterized by strong demographic growth and the development of numerous sectors of activity such as agriculture, tourism, livestock farming, industry, etc., all of which lead to a considerable increase in water requirements. In the area, most of the population's water needs are covered by groundwater, which is the main resource due to the scarcity of surface water. Thus, the overexploitation of groundwater to meet the growing need for drinking water and for other sectors of activity is leading to a drop in groundwater levels and, above all, a gradual deterioration in water quality, exacerbating the problem of water availability. Water availability is a recurring problem in many countries in arid and semi-arid zones, and faces a number of constraints related to climate change. The current study was conducted as part of the assessment of groundwater quality in the Mbour Fatick area and aims to determine the spatial distribution of nitrate and fluoride content in groundwater and to estimate the health risk in adults and children associated to consumption of nitrate and fluoride-rich groundwater for local population.

2. Study Area Description

2.1. Location and Climate

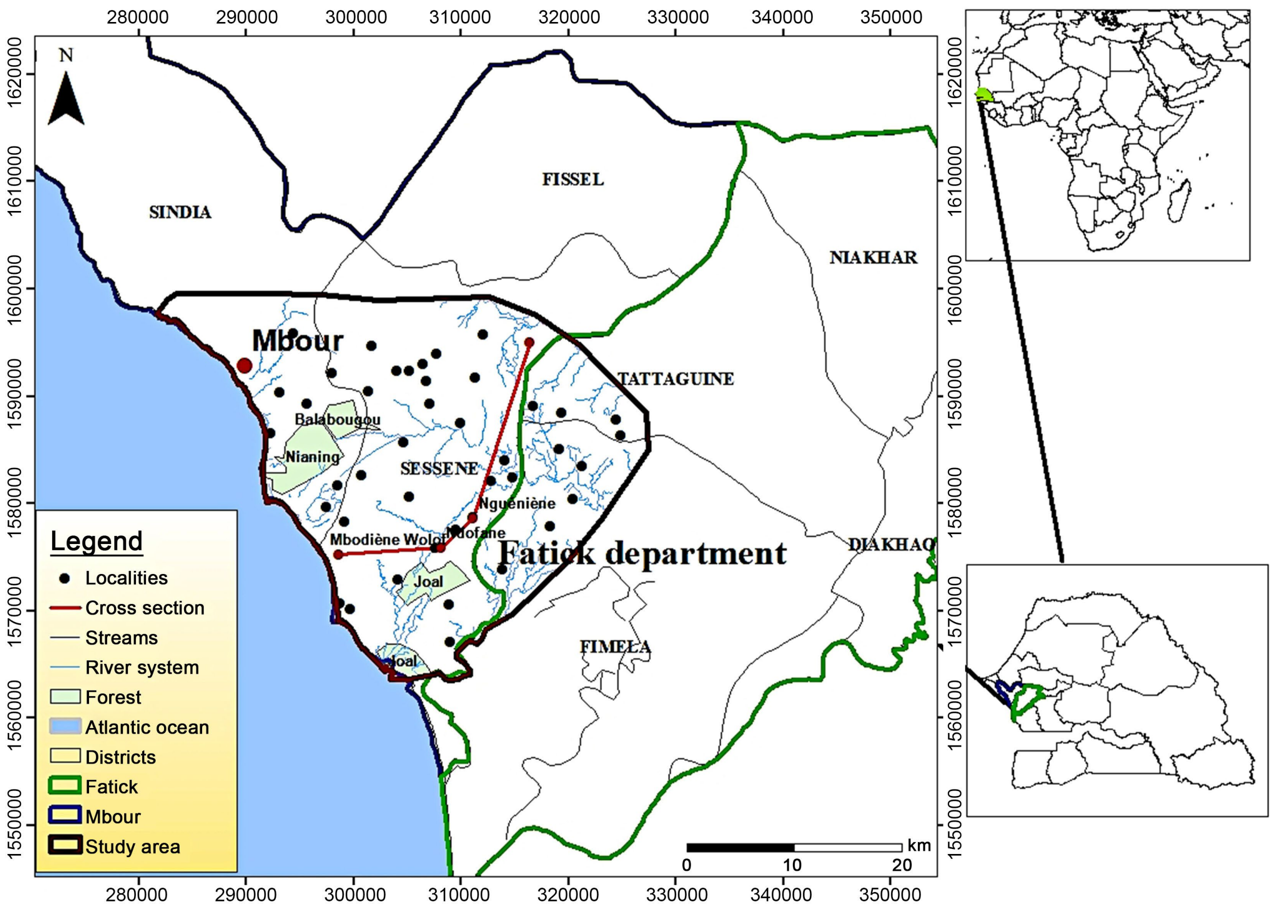

Figure 1. Geographical location of the study area.

The study area is located in west-central Senegal and includes part of the department of Mbour (Thies region) and part of the department of Fatick (Fatick Region). It covers an area of approximately 971 km^2 and is limited to the North by the districts of Notto and Fissel to the East by those of Niakhar and Diakhao, to the Southeast by the "bolongs" which constitute part of the tributaries of the Saloum River and to the West by the Atlantic Ocean (**Figure 1**). It has a tropical climate with an alternation of two seasons: a short rainy season which extends from July to October and a long dry season which extends from November to June. Rainfall generally shows great temporal and spatial variability. The heaviest rains are generally recorded in August and September. Average annual rainfall during the periods 1991-2020 and 1990-2020 in Mbour and Fatick, respectively was 561 mm and 595 mm [37]. The minimum annual temperature is recorded in December with a value of 20˚C, while the maximum annual temperature is recorded in May with a value of 40˚C. Relative humidity is higher during the rainy season, with values ranging from 60% to 96% [38]. Evaporation is highly variable in the area, with average annual values of 1600 mm per year and 2200 mm per year in Mbour and Fatick, respectively.

2.2. Geological and Hydrogeological Settings

The study area belongs to the Senegal-Mauritania sedimentary basin, which covers a large part of Senegal, with the exception of its eastern part. Lithological descriptions of the formations in the study area indicate the presence of detrital soils with a predominance of clay and sand, which constitute the geological layers of the Terminal Continental and the Quaternary. The Eocene, due to its strong erosion in the area, is represented only by its lower terms where it consists of several levels:

- ✓ A level formed of clayey limestones, marls and clays, phosphatized or silicified, encountered at contact with the Paleocene [39];
- ✓ A clayey or marly unit with some very frequent intercalations of limestone in the upper part;
- ✓ A horizon consisting of alternating limestone and marly limestone found mainly in the Ngazobil area [29] (**Figure 2**). The base of the Lower Eocene consists of grey marl and clay with flint intercalation [40]. Phosphate levels are encountered in the Lower and Middle Eocene [30].

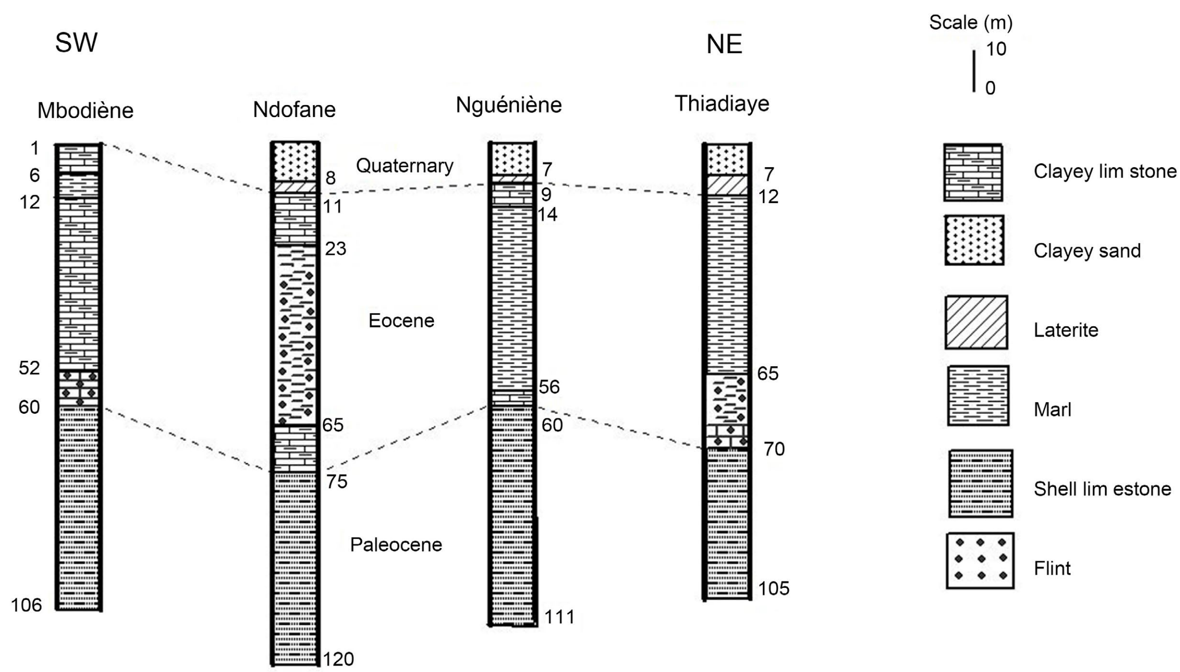

Figure 2. Lithology and stratigraphic correlations across the study area [40] modified.

The Paleocene, which outcrops around the town of Mbour, sinks towards the East and Southeast under Eocene, Continental Terminal and Quaternary formations. It is characterized by homogeneous facies of limestone and marly limestone, often shell-bearing [41]. In the middle and upper part of the Paleocene, numerous levels of sandstone limestone, varying in fineness and rich in shell debris, are constantly encountered [39]. Throughout the region, the base of the Paleocene is composed of hard limestone and grey marly limestone, which may be sandstone [42]. The Paleocene-Eocene boundary is marked by the sudden disappearance of microfauna and macrofauna, and the appearance of facies that are very poor in fossils and more typically marine [39]. The transition from Paleocene limestones to Lower Eocene marls often occurs through the deposition of a marl-clayey flint level, particularly in the northeastern part of the area [43]. In the Thiadiaye and Fatick areas, phosphate deposits mark the roof of Paleocene limestones [41]. Hydraulically, the study area is characterized by the presence of several types of aquifers belonging either to the shallow aquifer system or to the deep system. The superficial aquifer system is made up of Mio-Pliocene, Quaternary and upper Ypresian aquifers. The latter is formed by marly limestone, while the Mio-Pliocene and Quaternary aquifers are formed by clayey sand. The aquifers of the superficial system are mainly exploited by traditional wells to satisfy domestic water needs and for market gardening. The Eocene limestone aquifer is distinct or associated with the Miocene-Quaternary aquifer. The deep system, essentially tapped by boreholes and modern wells, is made up of the deep aquifer of the Paleocene and the Maastrichtian. The Paleocene aquifer is relatively thin and lies on the sandstone-clay sediments of the Maastrichtian. It consists of limestone, argillaceous limestone and marl with flint, glauconite and phosphate [40]. Its bottom is mainly made of marlstone, which, as a result of a change of facies, is replaced by shell limestone in the west, and by marlstone in the Northwest, East and South [44]. The Paleocene aquifer is currently the most exploited due to the depletion or salinization in some areas of the superficial aquifers. The hydraulic conductivities of the Paleocene aquifer are highly variable and range from 6.6×10^{-6} to 2.0×10^{-2} $m \cdot s^{-1}$, while the storages coefficients range from 1×10^{-4} to 7×10^{-2}. In the Mio-Plio-quaternary aquifer, the hydraulic conductivities are also variable, with mean values of hydraulic conductivities and effective porosity of about 1.5×10^{-4} $m \cdot s^{-1}$ and 20% respectively [38] [45] [46].

3. Materials and Methods

3.1. Groundwater Samples Collection and Analysis

In September 2019, a sampling campaign was carried out in the study area, during which 42 groundwater samples were collected from boreholes and wells. The position of the sampling points, in the study area, was determined in the field using Garmin GPS (Global Position System) (**Figure 3**). At each sampling point, two groundwater samples were collected in polyethylene bottles. One of the bottles was acidified with HNO_3^- for cations and traces metals analysis and another was kept unacidified for anions analysis. However, before sampling, polyethylene bottles were rinsed two to three times with the water to be sampled. After sampling, the groundwater samples were labeled, stored in an ice box and transported to the laboratory for chemical analysis. Physical parameters such as Temperature, Electrical conductivity (EC) and pH were measured directly in the field using a portable multi-parameter (WTW-multi 350i). The water samples were analysed at the Chrono Environment Laboratory of the UFR Sciences and Techniques at the University of Bourgogne Franche Comté, Besancon, France. In the laboratory, collected groundwater samples were filtrated using cellulose nitrate membrane (0.22 µm pore size). However, before filtration, bicarbonate concentrations were measured by titration. Anions analysis was performed using a Dionex-100 ion chromatography whereas cations and metals of the water samples were determined using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). The analytical accuracy for the measurements of ions was determined by computing the charge balance error (CBE) which was generally within ±5%.

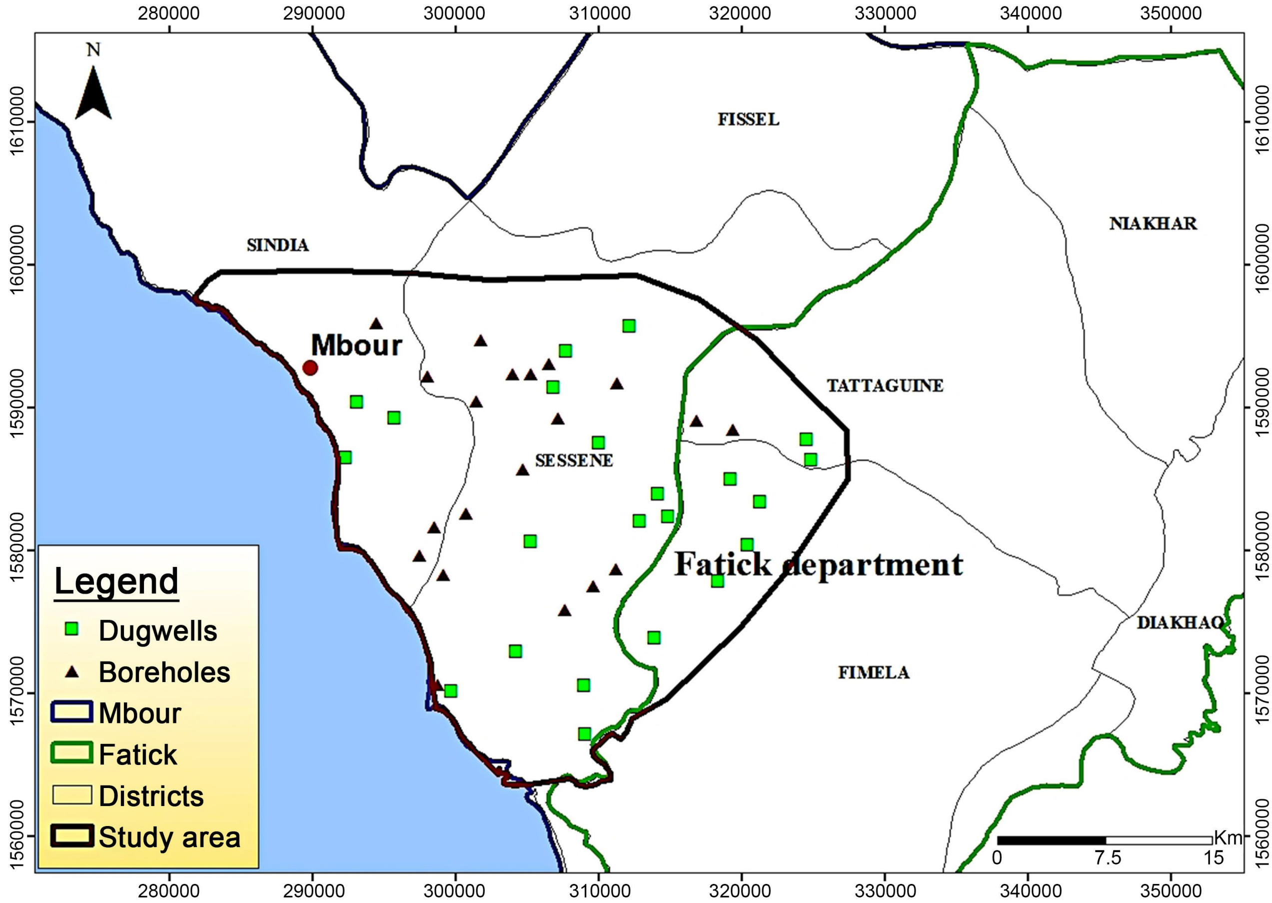

Figure 3. Locations of the sampling sites in the study area.

3.2. Health Risk Assessment (HRA)

In recent decades, groundwater pollution has become a worldwide problem. It is particularly acute in urban areas with strong population growth and in rural areas with intensive human activities (agriculture, livestock farming, etc.). Water consumption and long-term exposure to water containing high concentrations of nitrate and fluoride are considered by many studies [1] [20] [34] [47] as risk factors that can lead to the onset of certain diseases. Thus, assessing the health risks of drinking water with high nitrate and fluoride concentrations is an important and essential step in understanding the deterioration of water quality and the probability of adverse effects on public health. It also makes it possible to take effective protection measures for water resources and to guarantee a sustainable supply of drinking water. Human can be exposed to pollutants through several pathways such as direct ingestion (drinking), breathing, washing and cleaning (dermal contact) [33] [34] [48]. However, drinking water intake and dermal contact are considered as the main exposure pathways by which contaminants enter the human body. However, [49] [50] showed that the health risks of contaminated groundwater through dermal contact are low and therefore, negligible compared to the health risks associated with oral drinking water intake of contaminated groundwater. Thus, in this study, only oral drinking water intake was considered for the assessment of the health risks of local residents. Based on the models proposed by [51], two parameters F^- and NO_3^- in groundwater of the study area were used to evaluate the potential human health risk of the groundwater drinking pathways. The parameters and values used to calculate the health risk quotients are presented in **Table 1**. The exposure dose through intake can be computed by Equation (1) [1] [34] [47] [51].

Table 1. Parameters and values used for the computation of Health Hazard Quotients.

Parameters	Adults	Children	References
Ingestion rate (IR, $L \cdot Day^{-1}$)	2.5	0.78	[20] [47] [52]
Exposure duration (ED, years)	30	12	[1] [27] [49]
Exposure frequency (EF, days per year)	365	365	[49] [52]
Average body weight (BW, kg)	65	15	[20]
Average exposure time (AT, days)	10950	4380	[1] [27] [49]
Concentration of pollutant in groundwater ($mg \cdot L^{-1}$)	Fluoride (0.13 to 9.41) Nitrate (0.13 to 432.24)		Present study

$$\mathrm{CDI} = \frac{\mathrm{C} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \quad (1)$$

where CDI is the chronic daily intake *i.e.* the exposure dose through intake of drinking water ($mg \cdot kg^{-1} \cdot day^{-1}$); C is the concentration of the contaminant in drinking water ($mg \cdot L^{-1}$); IR represents the ingestion rate of water ($L \cdot Day^{-1}$) and in this study the IR values for adults and children are 2.5 $L \cdot Day^{-1}$ and 0.78 $L \cdot Day^{-1}$ respectively [20] [47] [52]; EF is the exposure frequency (day per year; EF = 365 days per year for both adults and children); ED is the exposure duration (year); BW and AT are respectively the average body weight (kg) and the average exposure time (days). For this study, the values of ED obtained from the literature review are 30 years and 12 years for adults and children respectively. The BW is considered as 65 kg and 15 kg respectively for adults and children. AT values are 10,950 and 4380 days for adults and children respectively.

The reference dosage is used as a measure of non-carcinogenic chronic hazard. If exposure doses to the pollutant exceed the reference dose, toxic effects are likely to occur. The non-carcinogenic effect of single element can be expressed as hazard quotient (HQ) which is calculated using Equation (2):

$$\mathrm{HQ} = \frac{\mathrm{CDI}}{\mathrm{RfD}} \tag{2}$$

where RfD represents the reference dose of a specific contaminant for non-carcinogenic health risk. For this study the reference dose for chronic oral exposure of fluoride and nitrate are 0.06 $mg \cdot kg^{-1} \cdot day^{-1}$ and 1.6 $mg \cdot kg^{-1} \cdot day^{-1}$ respectively [20] [47] [52]. HQ values > 1 are potentially recognized as posing risks to public health. Therefore, when the value of HQ > 1, the non-carcinogenic risk exceeds the acceptable level, indicating high potential health risk for human and is unacceptable for adults and children, while HQ < 1 indicates an acceptable level of non-carcinogenic risk for individual drinking water. The total hazard index (THI), which represents the cumulative non-carcinogenic risk is computed using Equation (3)

$$\mathrm{THI} = \sum_{i=1}^{n} \mathrm{HQi} \tag{3}$$

Based on [52], the maximum allowable threshold for non-carcinogenic THI is 1. If THI values exceed 1, it is considered as an intolerable risk of adverse non-carcinogenic effects on health, while THI < 1 suggests that the non-carcinogenic risk is within the acceptable limit. Spatial distribution maps of fluoride and nitrate content, as well as total hazard index (THI) values, were then generated with the spatial analysis module of ArcGIS software version 10.3, using the inverse-square interpolation technique.

4. Results and Discussions

4.1. Hydrochemical Characteristics of Groundwater

Chemical results of groundwater samples analysis are presented in **Table 2**. **Table 3** shows the descriptive statistics analysis with maximum, minimum, mean, median and standard deviation of the physicochemical parameter of groundwater samples in the study area. As seen from **Table 2**, the pH values of groundwater samples in the study area ranged from 7.1 to 8.2 with a median value of 7.6, demonstrating neutral to slightly alkaline nature of groundwater. Groundwater samples of the study area show pH values close to neutrality, which are characteristic of a carbonate system, suggesting that dissolution of carbonate rocks is an important factor in the variation of pH values. According to the World Health Organization [22] the permissible limits of pH for drinking water lie between 6.5 and 8.5. pH values lower than 6.5 are considered too acidic for human consumption and can cause health problems such as acidosis while pH values higher than 8.5 are considered to be too alkaline for human consumption [53]. In this study, all the measured values of pH in groundwater are well within the acceptable limit of 6.5 to 8.5 recommended by WHO [22] for drinking water. The electrical conductivity provides information on the overall amount of dissolved salts and reflects the efficiency with which the water conducts an electrical current. The measured electrical conductivity of groundwater in the study area ranged from 166.80 to 8880 $\mu S \cdot cm^{-1}$ with a median value of 1065 $\mu S \cdot cm^{-1}$. About 31% of groundwater samples exceed the acceptable limit of drinking water of 1500 $\mu S \cdot cm^{-1}$ in the study area. The total dissolved solid (TDS) is generally measured or computed to evaluate the degree of groundwater quality. The computed values of TDS in the study area ranged from 111.76 to 5949.6 $mg \cdot L^{-1}$ with a median value of 713.6 $mg \cdot L^{-1}$. Out of the 42 samples collected, 35 (83%) had TDS values exceeding the acceptable limit value of 500 $mg \cdot L^{-1}$ prescribed by the WHO. Furthermore, according to [54] classification, the majority of groundwater samples (67%) belong to the freshwater category [37]. The concentration of cation in groundwater samples shows that Na^+ and Ca^{2+} are the dominant cations followed by Mg^{2+} and K^+ in the abundance order of $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ (**Figure 4**). The concentrations of Na^+, which is the most abundant cation, ranged between 8.1 and 1106.1 $mg \cdot L^{-1}$ with a median value of 72.2 $mg \cdot L^{-1}$. In the study area, only 6 water samples (14%) have sodium contents above the acceptable limit for drinking water. The concentrations of calcium, magnesium and potassium vary respectively from 17.8 to 562.1 $mg \cdot L^{-1}$, 2.7 to 168.4 $mg \cdot L^{-1}$ and 0.3 to 10.7 $mg \cdot L^{-1}$ with respective median values of 71.3, 43.9 and 2.3 $mg \cdot L^{-1}$. Potassium shows the lowest concentrations in groundwater samples with values which are all below the acceptable limit for drinking water. The abundance of anions, based on the median value of groundwater is in the order of $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^- > F^-$ (**Figure 4**). HCO_3^- which is the most abundant anion in groundwater samples of the study shows high spatial variability. Its concentrations ranged from 9.4 to 541.4 $mg \cdot L^{-1}$ with a median value of 344.6 $mg \cdot L^{-1}$. High bicarbonate concentrations are recorded in 4 villages (Mbassis, Ngueniene Serere, Ngazobil and Diolofira Serere) where the bicarbonate content exceeds the maximum permissible limit for drinking water (**Table 2**). High concentrations of bicarbonate in groundwater can be related to the dissolution of carbonate minerals contained in the limestone and marl-limestone rocks that constitute the main geological formations in the study area. They may also result from the degradation of organic matter in the soil. Chloride occurs naturally in all types of water. Weathering of halite and evaporates are considered as the major lithogenic source of chloride [55]. For this study, the measured concentration of chloride varies from 11.51 to 2626.48 $mg \cdot L^{-1}$ with a median value of 132.50 $mg \cdot L^{-1}$. In the study area, fifteen (15) groundwater samples exceed the maximum allowable limit of 250 $mg \cdot L^{-1}$ for drinking water (**Table 3**). The high chloride contents can be derived from agricultural activities, wastewater in the inhabited area but also from the dissolution of salts accumulated on the surface by rainwater. In water, sulfate generally derived from the dissolution of gypsum or other sulfate bearing minerals. The concentration of sulfate in groundwater samples of the study area ranged from 1.2 to 527.5 $mg \cdot L^{-1}$ with a median of 30.4 $mg \cdot L^{-1}$. Three groundwater samples gathered in the villages of Ngueniene, Ngueniene Serere and Nianing have sulfate concentrations above the acceptable limit of 250 mg·L^{-1} for drinking water (Table 3). Chloride, sulfate and bicarbonate show wide spatial variability in the area, with respective standard deviation values of 436, 113 and 152, suggesting that their concentrations in groundwater are affected by various factors, both natural and anthropogenic.

Table 2. Analytical results of groundwater samples from the study area.

Samples ID	Localities	pH	E C	TDS	F^-	Cl^-	NO_3^-	SO_4^{2-}	HCO_3^-	Ca^{2+}	K^+	Mg^{2+}	Na^+
B1	Keur Balla lo	7.70	910.00	609.70	0.47	277.52	0.23	25.45	37.96	43.86	7.80	15.67	122.93
B2	Samane	7.40	1066.00	714.22	2.21	259.52	2.07	14.90	145.98	79.25	1.59	42.70	63.96
B3	Mbourok Cisse	7.40	833.00	558.11	0.20	184.77	191.35	10.80	44.24	102.26	5.18	10.42	38.51
B4	Bacoumbel	7.20	1156.00	774.52	5.50	115.26	15.49	47.68	435.26	67.82	2.48	68.65	73.26
B5	Soussane	7.43	928.00	621.76	3.99	72.10	0.35	53.36	375.29	66.21	3.52	49.61	54.07
B6	Ndollor	7.42	1001.00	670.67	3.63	79.70	10.49	46.57	411.43	70.88	5.78	48.27	69.27
B7	Ndiouck Fissel	7.70	736.00	493.12	2.16	45.94	11.60	10.34	343.97	68.15	5.17	34.26	32.02
B8	Ndioudiouf	7.50	1089.00	729.63	3.40	41.14	0.23	7.11	351.42	54.74	1.53	41.70	30.95
B9	Pethemakha	8.05	999.00	669.33	5.91	92.23	1.86	47.20	399.19	51.91	2.72	55.77	81.95
B10	Mbassis	7.75	1165.00	780.55	3.95	68.81	n.a	32.62	541.42	45.51	3.78	59.55	112.40
B11	Ngenienne	8.00	2670.00	1788.90	4.98	452.10	148.61	290.67	400.67	137.07	9.01	89.65	319.93
B12	Ngenienne Serere	7.56	2900.00	1943.00	8.56	427.97	0.21	527.49	502.02	96.50	9.01	138.08	361.63
B13	Mbodiene Nord	7.67	1761.00	1179.87	5.33	265.74	0.17	163.35	456.07	85.90	5.05	91.47	163.04
B14	Ndofane	7.35	881.00	590.27	1.48	57.94	17.20	36.96	374.84	102.12	0.32	12.10	71.70
B15	Ngazobil	7.62	4500.00	3015.00	4.87	1107.16	0.89	246.25	512.45	122.30	7.89	59.98	787.27
B16	Roff_K Seck	7.60	1963.00	1315.21	2.79	433.70	40.07	82.69	256.25	151.55	3.09	61.70	148.10
B17	Roff	7.33	1246.00	834.82	2.51	186.40	12.95	51.00	345.14	123.88	0.96	43.73	64.51
B18	Ndiemane	7.51	2390.00	1601.30	2.42	464.60	5.63	202.40	416.43	206.96	2.10	82.00	174.17
B19	Balabougou	7.40	2080.00	1393.60	5.52	454.87	n.a	54.94	378.12	76.75	3.95	87.79	221.50
B20	Diolofira oualof	7.38	878.00	588.26	4.16	68.37	0.36	20.97	393.50	66.01	1.81	47.74	48.32
DW 1	Diolofira Serere	7.56	1089.00	729.63	6.74	88.25	n.a	7.68	518.53	52.90	0.99	76.17	69.35
DW 2	Keur Yerim	7.58	968.00	648.56	0.20	132.84	0.13	2.84	336.90	102.41	1.93	15.87	72.67
DW 3	Sessene	7.56	2140.00	1433.80	1.94	516.96	1.56	24.99	293.23	219.95	0.69	64.76	91.31
DW 4	Diokhar Ngolem	7.46	872.00	584.24	1.39	62.86	18.46	6.81	404.65	81.37	0.70	43.99	36.01
DW 5	Ngohe Ndofongor	7.79	565.00	378.55	1.94	27.46	10.76	18.71	257.27	47.94	1.01	12.95	55.10
DW 6	Ngohe Pofine	7.70	625.00	418.75	3.10	47.19	19.82	29.89	232.58	69.37	0.49	10.12	49.24
DW 7	Ndiagamba	8.02	566.00	379.22	5.73	41.56	14.75	30.89	204.40	45.73	0.31	24.68	35.93
DW 8	Pombane	7.68	1064.00	712.88	8.47	132.15	0.31	55.83	336.55	34.93	2.53	62.37	96.53
DW 9	Foua 1	7.68	871.00	583.57	5.47	80.07	0.33	22.28	352.33	50.62	2.71	50.08	54.53
DW 10	Foua 2	7.95	1175.00	787.25	9.41	149.44	2.75	41.73	382.14	43.08	2.42	66.53	103.96
DW 11	Ndiol Khokhane	7.55	2030.00	1360.10	0.40	456.67	19.40	67.03	262.75	215.37	1.94	13.90	165.53
DW 12	Boyar Niodior	7.40	1258.00	842.86	1.20	267.66	16.23	21.77	175.00	115.83	8.44	29.83	66.34
DW 13	Ndianda	7.42	1963.00	1315.21	0.20	244.06	342.10	92.16	268.19	209.69	4.02	14.30	165.59
DW 14	Soudiane Thieleme	7.14	599.00	401.33	0.18	95.04	28.11	55.20	52.49	71.71	1.56	8.12	20.70
DW 15	Bagana Serere	7.58	2720.00	1822.40	0.15	787.59	36.18	29.84	66.53	222.40	10.73	20.08	265.46

Continued

DW 16	Fadial	7.58	166.80	111.76	0.13	11.51	17.35	1.19	48.79	17.76	0.65	2.68	8.10
DW 17	Joal Caritas	7.70	517.00	346.39	2.82	26.80	44.47	15.61	172.78	69.08	2.13	5.58	22.10
DW 18	Nianing	7.26	8880.00	5949.60	2.10	2626.48	432.24	432.85	9.39	562.13	0.99	168.37	1106.10
DW 19	Sidibougou	7.33	1696.00	1136.32	0.40	499.28	5.09	4.27	18.89	99.03	2.84	12.74	195.72
DW 20	Gagnabougou	7.39	958.00	641.86	0.92	188.97	55.08	13.05	112.57	69.96	1.47	10.61	93.90
DW 21	Aga Biaram	7.60	968.00	648.56	3.81	91.56	0.75	17.95	404.21	63.75	1.98	49.71	64.74
DW 22	Guedj Ngo Diagne	8.15	920.00	616.40	2.28	11.81	37.75	9.30	437.55	55.98	0.50	36.29	69.34

B: Borehole; DW: Dug Wells; n.a: Not analyzed. All parameters are expressed in $mg \cdot L^{-1}$ except for pH and EC ($\mu S \cdot cm^{-1}$).

Table 3. Descriptive statistics of groundwater chemistry in the study area.

Water quality parameters	Min.	Max.	Mean	Median	Standard deviation	WHO standard	Total number of samples exceeding allowable limits
pH (–)	7.14	8.15	7.57	7.56	0.23	6.5 - 8.5	0
EC ($\mu S \cdot cm^{-1}$)	166.8	8880	1518.16	1065	1421.32	1500	13
TDS ($mg \cdot L^{-1}$)	111.8	5949.6	1017.17	713.55	952.28	500	35
Ca^{2+} ($mg \cdot L^{-1}$)	17.76	562.13	103.35	71.29	90.15	75	20
Mg^{2+} ($mg \cdot L^{-1}$)	2.68	168.37	46.20	43.86	35.38	50	16
Na^{+} ($mg \cdot L^{-1}$)	8.10	1106.10	141.61	72.19	201.53	200	6
K^{+} ($mg \cdot L^{-1}$)	0.31	10.73	3.19	2.28	2.73	12	0
Cl^{-} ($mg \cdot L^{-1}$)	11.51	2626.48	279.57	132.50	435.74	250	15
NO_3^- ($mg \cdot L^{-1}$)	0.13	432.24	44.51	13.85	88.86	50	5
SO_4^{2-} ($mg \cdot L^{-1}$)	1.19	527.49	70.82	30.39	112.64	250	3
F^{-} ($mg \cdot L^{-1}$)	0.13	9.41	3.17	2.65	2.48	1.5	29
HCO_3^- ($mg \cdot L^{-1}$)	9.39	541.42	296.89	344.55	152.44	500	4

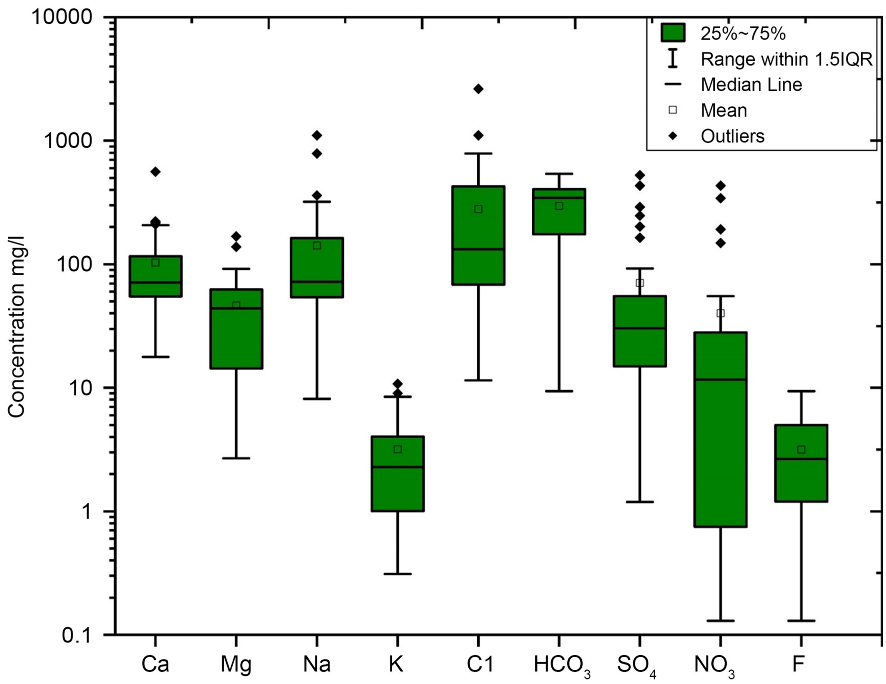

Figure 4. Box plot of groundwater samples chemical composition.

4.2. Spatial Distribution of Fluoride

In this study, fluoride concentrations ranged from 0.13 to 9.41 mg·L^{-1} with a median value of 2.65 mg·L^{-1}. The spatial distribution of fluoride levels shows that approximately 31% of groundwater samples have fluoride concentrations lower than the maximum permissible limit of 1.5 mg·L^{-1} set by the WHO [22]. These samples with low fluoride concentration are recorded in the North in municipality of Sindia and in the South in the municipalities of Sessene and Fimela. However, in the study area, the majority of groundwater samples (69%) have fluoride contents higher than the WHO standard (1.5 mg·L^{-1}), indicating water unsuitable for human consumption. Groundwater samples with high fluoride content are distributed throughout the study area, particularly in the localities of Sessene and Tattaguine (**Figure 5**). Fluoride is an essential microelement for human health. Lower fluoride values (<1 mg/L) are, in fact, considered to have beneficial effects on teeth by reducing dental decay [15]. However, high or low levels of fluoride mainly in drinking water, have been found to cause adverse health effects, including dental and skeletal fluorosis [15]. High concentrations of fluoride in groundwater constitute a major public health problem in many countries around the world, particularly in arid and semi-arid zones where rural populations are most often affected by fluorosis due to their dependence on groundwater. Depending on the effects of fluoride on human health, fluoride concentrations can be classified into several categories. According to [6], risks of dental decay occur when the fluoride concentration in water is less than 0.5 mg·L^{-1}. Fluoride concentrations between 0.6 and 1.5 mg·L^{-1} are considered beneficial for human health. However, fluoride levels in water between 1.6 and 2 mg·L^{-1} lead to the appearance of dental fluorosis and levels between 2.1 and 3 mg·L^{-1} promote the development of dental and skeletal fluorosis and finally, fluoride levels above 3 mg·L^{-1} lead to skeletal fluorosis. Based on this classification, 21% of groundwater samples have fluoride contents less than 0.5 mg·L^{-1}, 10% have concentrations between 0.5 and 1.5 mg·L^{-1}, 5% have fluoride contents between 1.5 and 2 mg·L^{-1}, 19% have contents between 2 and 3 mg·L^{-1} and finally the majority of samples (45%) have contents greater than 3 mg·L^{-1} (**Figure 5** and **Figure 6**). In the study area, the highest concentrations are found in the localities of Foua 2 (9.41 mg·L^{-1}), Pombane (8.47 mg·L^{-1}), Diolofira Serere (6.74 mg·L^{-1}) and Ngenienne Serere (8.56 mg·L^{-1}); while the lowest concentrations are recorded in the localities of Keur Balla lo (0.47 mg·L^{-1}), Mbourok Cisse (0.20 mg·L^{-1}), Keur Yérim (0.20 mg·L^{-1}), Ndiol Khokhane (0.40 mg·L^{-1}), Ndianda (0.20 mg·L^{-1}), Soudiane Thieleme (0.18 mg·L^{-1}), Bagana Serere (0.15 mg·L^{-1}), Fadial (0.13 mg·L^{-1}) and Sidibougou (0.40 mg·L^{-1}) (**Figure 7**).

4.3. Spatial Distribution of Nitrate

In the current study, nitrate concentrations vary from 0.13 mg·L^{-1} to 432.24 mg·L^{-1} with a median value of 13.85 mg·L^{-1}. The spatial distribution map of nitrate concentrations is generated to facilitate the identification of good quality water and, above all, to facilitate decision-making and effective groundwater management. Figure 8 shows that the majority (87.18%) of sampled sites have nitrate concentrations lower than the WHO drinking water standard of 50 mg·L^{-1}, indicating that the water is fit for human consumption. However, 5 groundwater samples (12.82%) have nitrate concentrations above the WHO standard. The highest concentrations were recorded in the localities of Mbourok Cisse, Ngenienne, Nianing and Ndianda, with nitrate concentrations of 191.35 mg·L^{-1}, 148.61 mg·L^{-1}, 432.24 mg·L^{-1} and 342.1 mg·L^{-1} respectively. According to [20] [47] water containing nitrate concentrations below 45 mg·L^{-1} poses a low health risk to populations, while water with nitrate levels between 46 and 100 mg·L^{-1} poses a high risk, and water with nitrate levels above 100 mg·L^{-1} poses a very high health risk. Based on this classification, out of 39 groundwater samples, 34 (87.18%) have nitrate levels below the WHO standard, indicating suitable water for human consumption and exhibiting a low health risk. However, only one groundwater sample (2.56%) and four groundwater samples (10.26%) have high (50 - 100 mg·L^{-1}) and very high (>100 mg·L^{-1}) nitrate concentrations respectively, indicating unsuitable water for drinking purposes and showing high and very high health risks respectively (Figure 9).

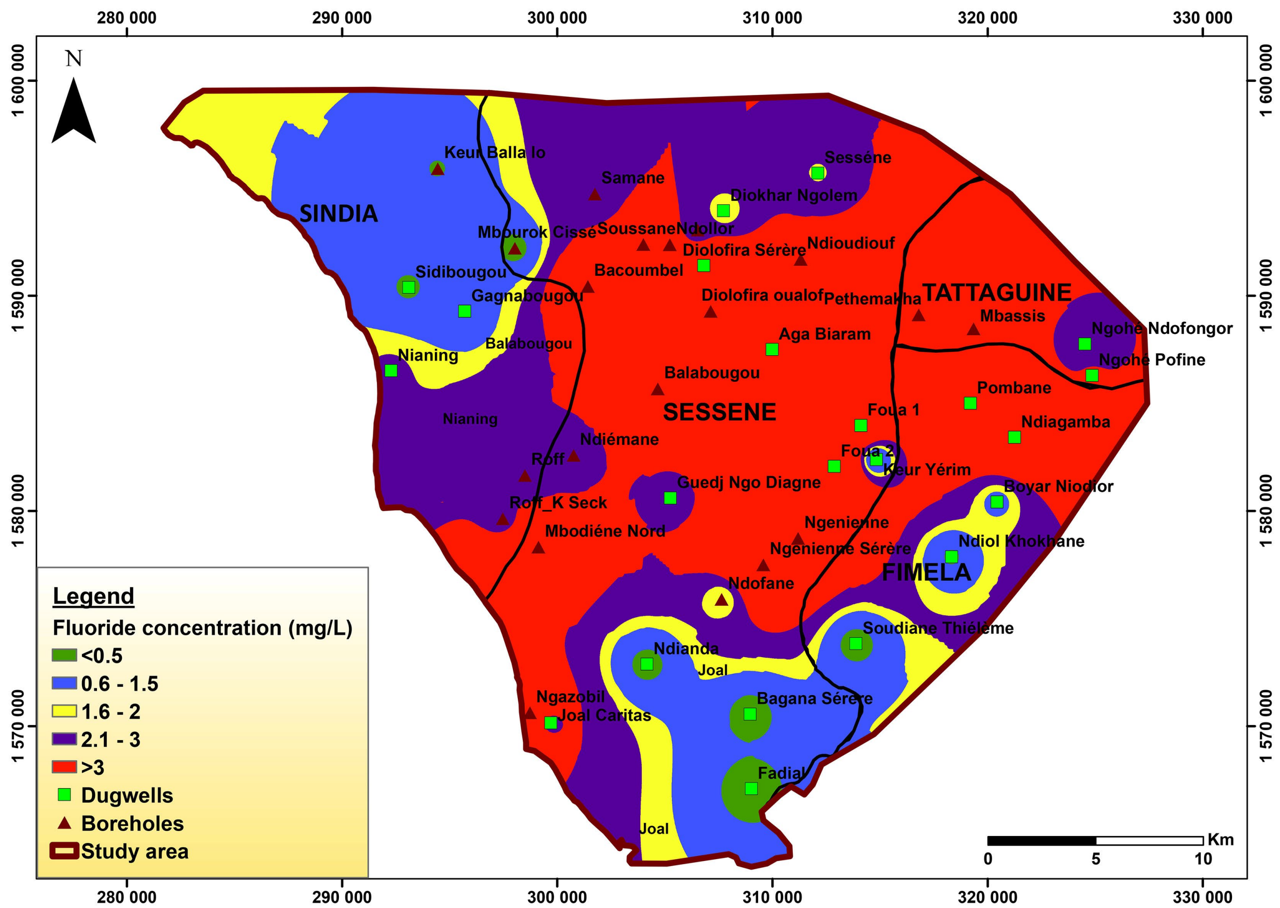

Figure 5. Spatial distribution of fluoride and its classification of health risk.

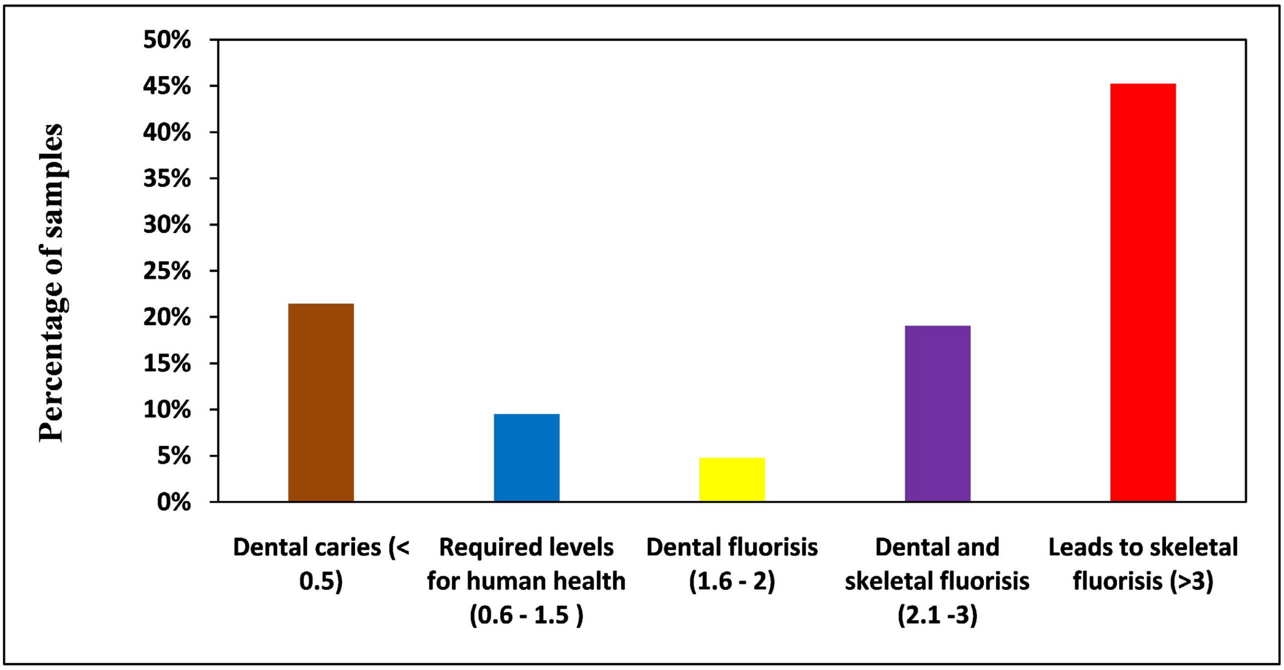

Figure 6. Health risk effect associated with fluoride ingestion in individuals.

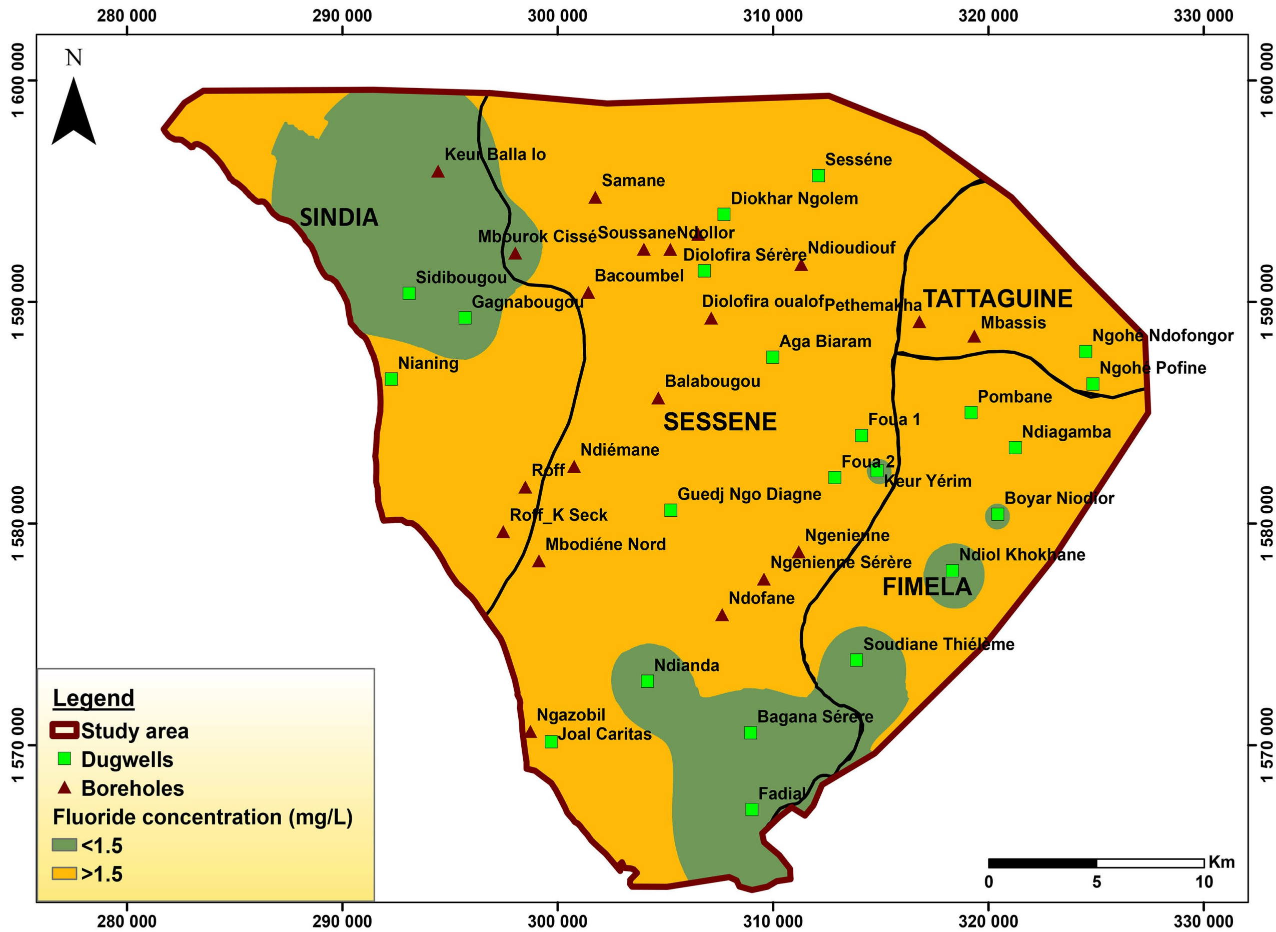

Figure 7. Spatial distribution map of fluoride content in the study area.

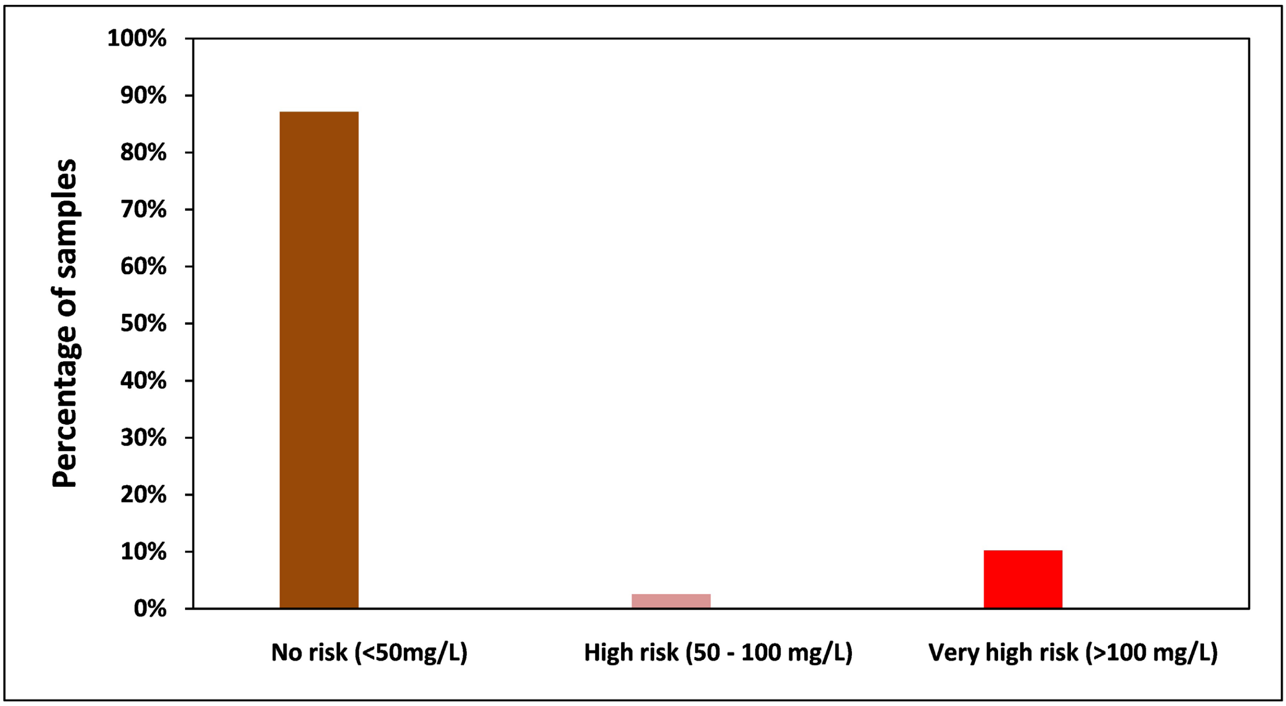

Figure 8. Percentage distribution of nitrate concentration in groundwater for risk evaluation in the study area.

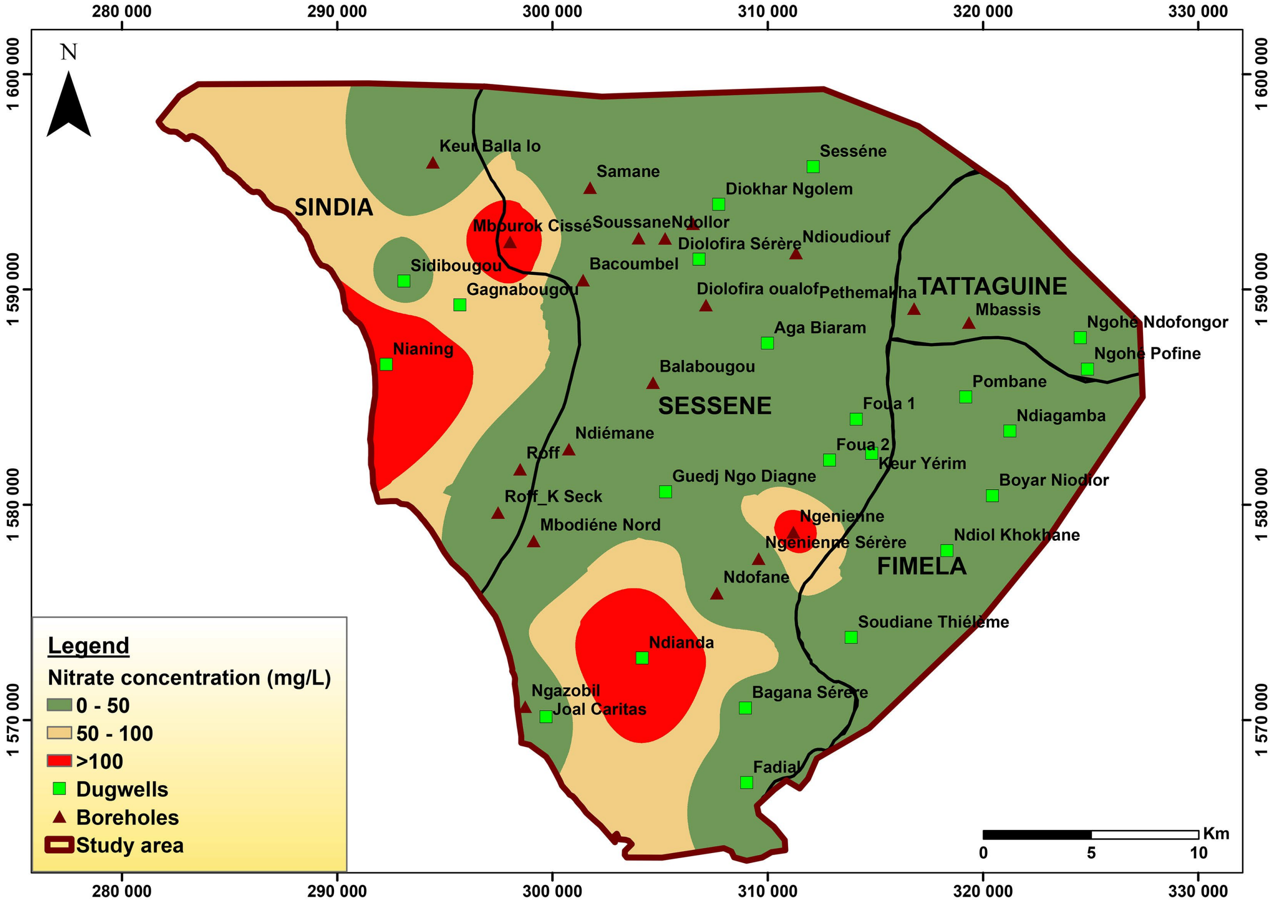

Figure 9. Spatial distribution map of nitrate content in the study area.

4.4. Health Risk Assessment

Health risk assessment is an important and essential step in estimating the adverse effects of pollutants on human health. Consuming polluted water can lead to serious risks for human health if contaminated water is drunk over a long period without any prior treatment [1] [21] [26] [27]. In this study, the potential risks to population health due to exposure to nitrate and fluoride in groundwater were assessed in each sampled site by calculating the hazard quotient (HQ) and the Total Health Index (THI) for both adults and children. The results of the HQ and THI of non-carcinogenic risks for adults and children are presented in **Table 4**. The computed hazard quotient values of fluoride and nitrate for adults through drinking water intake ranged respectively from 0.08 to 6.03 and from 0.003 to 10.39 with a respective average value of 1.92 and 0.96. For children, the hazard quotient values for fluoride and nitrate in the study area vary respectively from 0.11 to 8.16 and from 0.004 to 14.05 with a respective average value of 2.60 and 1.30. In adults, approximately 66.67% and 15.38% of groundwater samples respectively have fluoride and nitrate hazard quotient greater than the admissible limit (HQ = 1) indicating that the non-carcinogenic health risk related to fluoride, in adults, is much higher than for nitrate. In children, 74.36% and 23.08% of groundwater samples respectively have fluoride and nitrate hazard quotient above the permissible limit suggesting that the non-carcinogenic health risk related to fluoride is also much higher than for nitrate. Thus, the results of this study indicate that the non-carcinogenic risk of nitrate is lower than that of fluoride, suggesting that fluoride contributes much more than nitrate to the non-carcinogenic health risk in adults and children in the study area. Furthermore, the total health index, which is the sum of the hazard quotient for fluoride and nitrate, was computed for adults and children. The total health index values for adults ranged from 0.13 to 11.74 with an average value of 2.88 while for children the total health index values vary from 0.18 to 15.87 with an average value of 3.90. In adults, approximately 82.05% of sample points have a THI > 1 while in children, 87.18% of water samples have a THI > 1 suggesting that the majority of samples can induce non-carcinogenic health risk in adults as well as in children (**Figure 10** and **Figure 11**). Thus, the results of the current study reveal that children are more prone to non-carcinogenic risk through ingestion of contaminated drinking water than adults in the study area. Similar results have been found by many other previous studies carried out in various parts of the world. For example, [20] performed an assessment of human health risk based on the occurrence and geochemical mechanisms of fluoride and nitrate in groundwater in the rock-dominated semi-arid region of Telegana State in India and indicate that children face higher non-carcinogenic health risk than adults. [1] also assessed groundwater contamination by fluoride and nitrate and the associated health risk of rural residents in a semi-arid region of northwest China. Their study shows that infants and children are more exposed to non-carcinogenic health risks than adults (men and women). Additionally, [21] assessed the spatial distribution, exposure and potential health risks from consumption of nitrate-contaminated water in the semi-arid Peddavagu region of central Telangana (South India) and found that human exposure to nitrate-contaminated water was above the critical limit of non-carcinogenic risk and that children were more exposed than adults (men and women) in the region. Similarly, [47] from a perspective of sustainable groundwater management, assess their quality and the health risks associated with fluoride and nitrate in a semi-arid region of southern India. Their results reveal that infants are more exposed to non-carcinogenic risks than adults and children.

Table 4. Hazard quotient and total hazard index of the population (children and adults) in the study area.

Sampling sites	RfD		Children			Adults		
	NO_3^-	F^-	HQ_{NO3}	HQ_F	THI	HQ_{NO3}	HQ_F	THI
Keur Balla lo	1.6	0.06	0.007	0.407	0.415	0.006	0.301	0.307
Samane	1.6	0.06	0.067	1.915	1.983	0.050	1.417	1.466
Mbourok Cisse	1.6	0.06	6.219	0.173	6.392	4.600	0.128	4.728
Bacoumbel	1.6	0.06	0.503	4.767	5.270	0.372	3.526	3.898
Soussane	1.6	0.06	0.011	3.458	3.469	0.008	2.558	2.566
Ndollor	1.6	0.06	0.341	3.146	3.487	0.252	2.327	2.579
Ndiouck Fissel	1.6	0.06	0.377	1.872	2.249	0.279	1.385	1.663
Ndioudiouf	1.6	0.06	0.007	2.947	2.954	0.006	2.179	2.185
Pethemakha	1.6	0.06	0.060	5.122	5.182	0.045	3.788	3.833
Ngenienne	1.6	0.06	4.830	4.316	9.146	3.572	3.192	6.765
Ngenienne Serere	1.6	0.06	0.007	7.419	7.425	0.005	5.487	5.492
Mbodiene Nord	1.6	0.06	0.006	4.619	4.625	0.004	3.417	3.421
Ndofane	1.6	0.06	0.559	1.283	1.842	0.413	0.949	1.362
Ngazobil	1.6	0.06	0.029	4.221	4.250	0.021	3.122	3.143
Roff_K Seck	1.6	0.06	1.302	2.418	3.720	0.963	1.788	2.752
Roff	1.6	0.06	0.421	2.175	2.596	0.311	1.609	1.920
Ndiemane	1.6	0.06	0.183	2.097	2.280	0.135	1.551	1.687
Diolofira oualof	1.6	0.06	0.012	3.605	3.617	0.009	2.667	2.675
Keur Yerim	1.6	0.06	0.004	0.173	0.178	0.003	0.128	0.131
Sessene	1.6	0.06	0.051	1.681	1.732	0.038	1.244	1.281
Diokhar Ngolem	1.6	0.06	0.600	1.205	1.805	0.444	0.891	1.335
Ngohe Ndofongor	1.6	0.06	0.350	1.681	2.031	0.259	1.244	1.502
Ngohe Pofine	1.6	0.06	0.644	2.687	3.331	0.476	1.987	2.464
Ndiagamba	1.6	0.06	0.479	4.966	5.445	0.355	3.673	4.028
Pombane	1.6	0.06	0.010	7.341	7.351	0.007	5.429	5.437

Continued

Foua 1	1.6	0.06	0.011	4.741	4.751	0.008	3.506	3.514
Foua 2	1.6	0.06	0.089	8.155	8.245	0.066	6.032	6.098
Ndiol Khokhane	1.6	0.06	0.631	0.347	0.977	0.466	0.256	0.723
Boyar Niodior	1.6	0.06	0.527	1.040	1.567	0.390	0.769	1.159
Ndianda	1.6	0.06	11.118	0.173	11.292	8.224	0.128	8.352
Soudiane Thieleme	1.6	0.06	0.914	0.156	1.070	0.676	0.115	0.791
Bagana Serere	1.6	0.06	1.176	0.130	1.306	0.870	0.096	0.966
Fadial	1.6	0.06	0.564	0.113	0.677	0.417	0.083	0.500
Joal Caritas	1.6	0.06	1.445	2.444	3.889	1.069	1.808	2.877
Nianing	1.6	0.06	14.048	1.820	15.868	10.390	1.346	11.737
Sidibougou	1.6	0.06	0.165	0.347	0.512	0.122	0.256	0.379
Gagnabougou	1.6	0.06	1.790	0.797	2.587	1.324	0.590	1.914
Aga Biaram	1.6	0.06	0.024	3.302	3.326	0.018	2.442	2.460
Guedj Ngo Diagne	1.6	0.06	1.227	1.976	3.203	0.907	1.462	2.369

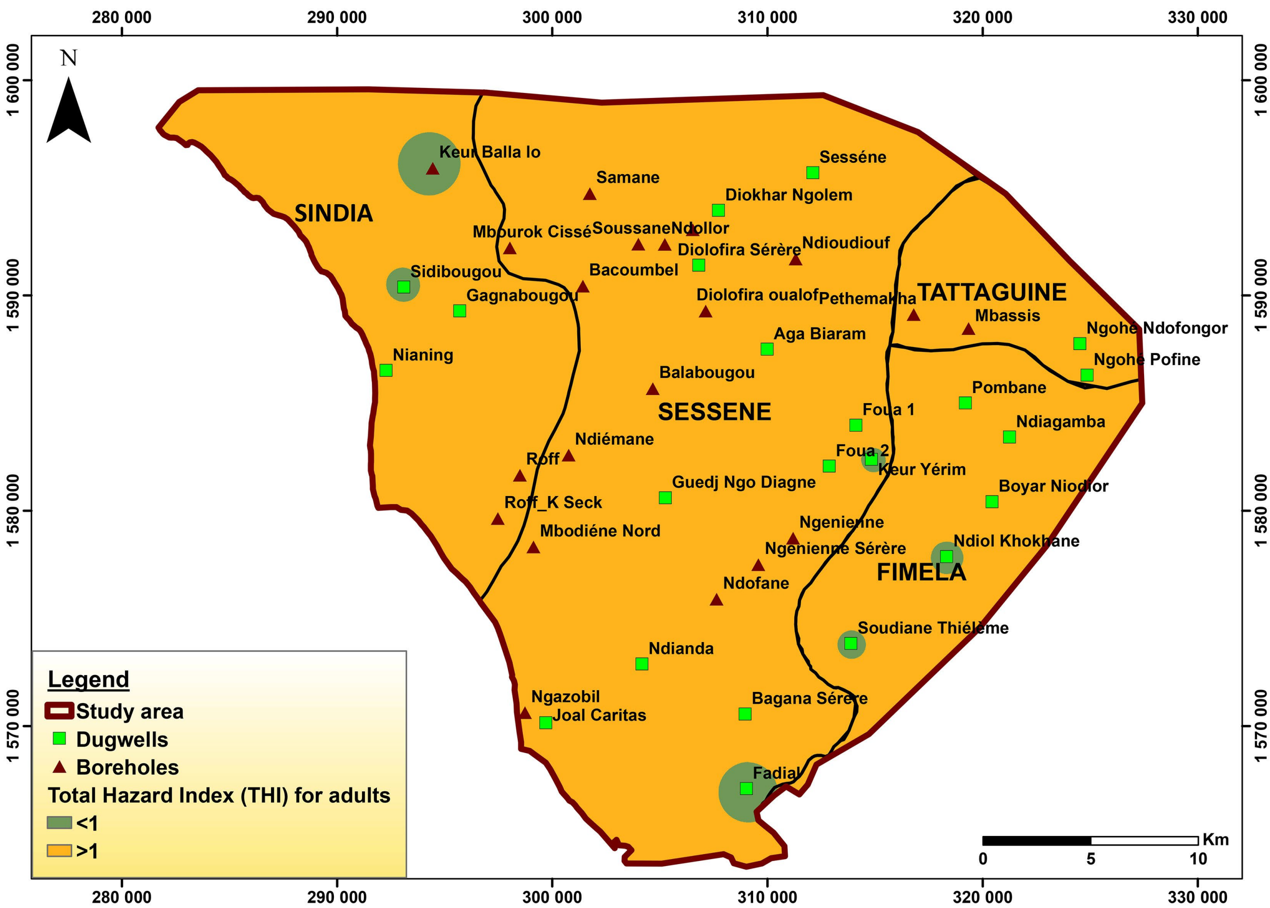

Figure 10. Non-carcinogenic THI map for adults in the study area.

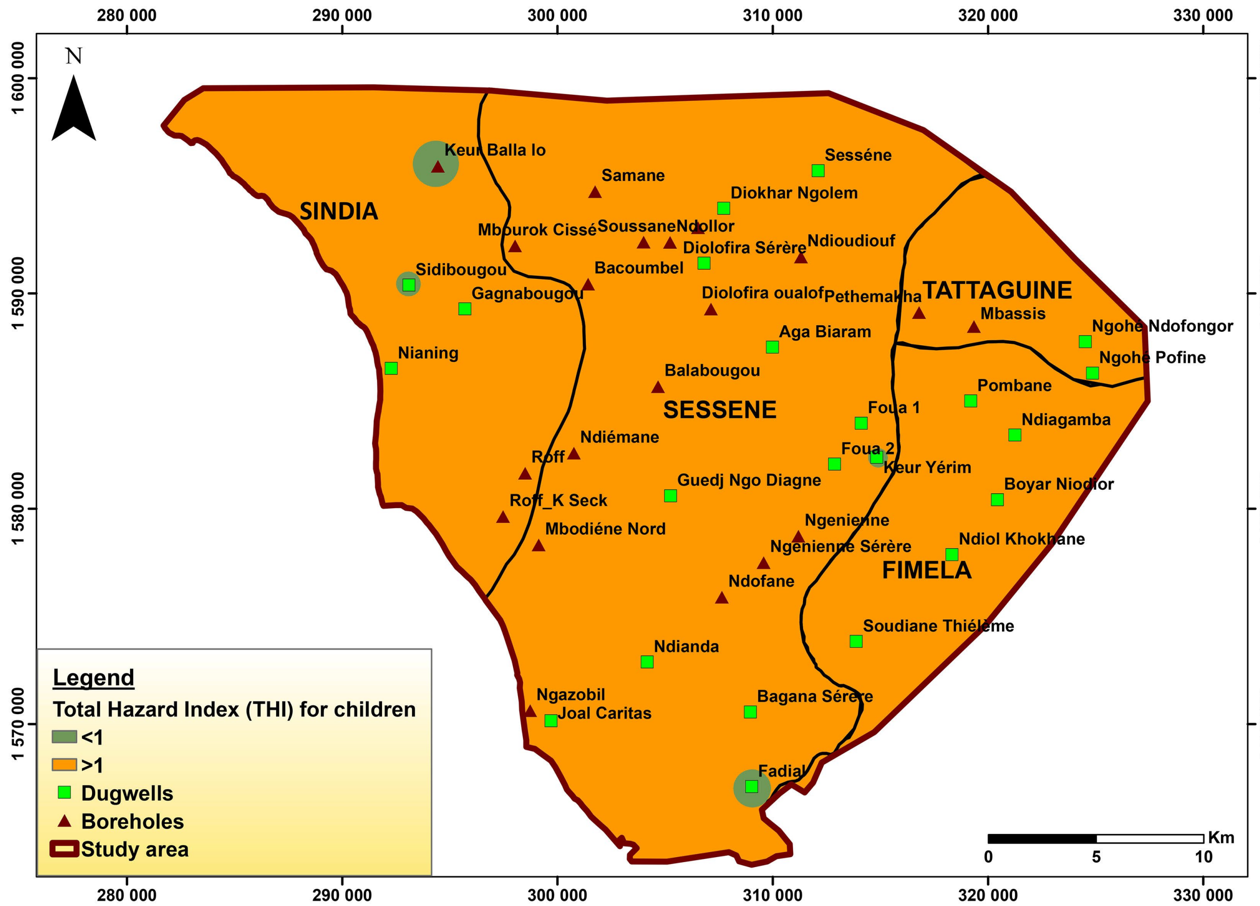

Figure 11. Non-carcinogenic THI map for children in the study area.

5. Conclusions

Groundwater is an important resource for drinking water supply and irrigation in the study area. In this study, groundwater quality and spatial distribution of nitrate and fluoride levels were investigated. Furthermore, health risks assessment of local populations, due to exposure to nitrate and fluoride through ingestion of contaminated drinking water was also performed for both children and adults. The results of this study lead to the following conclusions:

1) In the study area, groundwater is predominantly neutral to slightly alkaline with a median pH value of 7.6. All groundwater samples have pH values that are well within the permissible limits for drinking water. The results of chemical analyze indicate that the order of ion dominance is $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ for cations and $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^- > F^-$ for anions. In all the groundwater samples, concentrations of Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, SO_4^{2-}, HCO_3^- are, beyond half of the samples, below the permissible limits for drinking water.

2) Fluoride and nitrate concentrations in groundwater ranged from 0.13 to 9.41 $mg \cdot L^{-1}$ and 0.13 to 432.24 $mg \cdot L^{-1}$, respectively, with median values of 2.65 and 13.85 $mg \cdot L^{-1}$, respectively. Compared to WHO guideline values, the majority of groundwater sampling points (69%), for fluoride, are unsuitable for human

consumption. However, for nitrate, about 12.82% of groundwater samples have concentrations above the WHO permissible limit of 50 mg·L^{-1}. The spatial distribution of fluoride shows that fluoride concentrations, higher than the maximum admissible limit, are distributed over the entire study area. However, the highest concentrations are recorded in the localities of Foua 2 (9.41 mg·L^{-1}), Pombane (8.47 mg·L^{-1}), Diolofira Serere (6.74 mg·L^{-1}) and Ngenienne Serere (8.56 mg·L^{-1}); while the lowest concentrations are found in the localities of Keur Balla lo (0.47 mg·L^{-1}), Mbourok Cisse (0.20 mg·L^{-1}), Keur Yérim (0.20 mg·L^{-1}), Ndiol Khokhane (0.40 mg/L^{-1}), Ndianda (0.20 mg·L^{-1}), Soudiane Thiélème (0.18 mg·L^{-1}), Bagana Serere (0.15 mg·L^{-1}), Fadial (0.13 mg·L^{-1}) and Sidibougou (0.40 mg·L^{-1}). For nitrate, the spatial distribution indicates that the majority of sampled points (87.18%) have nitrate concentrations lower than the WHO standard guideline value. However, five localities have nitrate levels higher than the WHO standard. The highest nitrate concentrations are recorded in the villages of Mbourok Cisse, Ngenienne, Nianing and Ndianda with nitrate contents of 191.35 mg·L^{-1}, 148.61 mg·L^{-1}, 432.24 mg·L^{-1} and 342.1 mg·L^{-1} respectively.

3) Assessment of non-carcinogenic risks associated with contaminated drinking groundwater in local populations shows that THI values for adults ranged from 0.13 to 11.74 with an average value of 2.88, while for children, THI values ranged from 0.18 to 15.87 with an average value of 3.90. About 82.05% of sampled points have THI > 1 in adults, while in children, 87.18% of water samples have THI > 1 suggesting that local populations are exposed to non-carcinogenic health risks. Furthermore, the results indicate that children face greater non-carcinogenic risks, than adults through the ingestion of contaminated water,

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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