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Assessment of mountain water quality with special emphasis on nitrate contamination of Kullu Valley, Himachal Pradesh, India

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Abstract
Nitrate pollution of groundwater is a serious environmental issue in developing countries. The present study was aimed to evaluate the nitrate contamination levels in surface and groundwater samples collected from different developing areas in the Kullu valley of Himachal Pradesh and these samples were analyzed for nitrate in triplicates using an ion-selective electrode method. The mean concentrations of nitrate in surface and groundwater in the study areas ranged from a minimum of 3.3 mg/l to a maximum of 16 mg/l and 4.19 mg/l to 77 mg/l, respectively. The concentration of nitrate in groundwater at Akhara, Shamshi, and Pirdi in Kullu valley has exceeded the permissible limit of WHO (>50 mg/l) and may not be safe for drinking purposes. The average intake of nitrate through consumption of drinking water by local inhabitants was also found maximum for middle age (31-50 years) population i.e., 1.89 mg NO_3-N/day/kg body weight as compared to other age groups. Thus, the present study indicates that groundwater has been contaminated with nitrate which may be either resulting from human activities or natural. The frequent monitoring of nitrate contamination of groundwater in the developing areas of Kullu valley is suggested.

Keywords: nitrate, surface and ground water, permissible limit, consumption, Kullu Valley

Introduction

Approximately more than 2 billion people in the world are impacted by water scarcity, although United Nations declared 17 sustainable development goals in 2015, in which the 6th goal is "secure water availability for all" (Gupta and Vegelin 2016; Arikrishnan and Rajendran 2021) [6, 2]. There are several major factors such as different heavy metals, fluoride, nitrate and lot of emerging contaminants, etc. responsible for quality degradation of groundwater and surface water, due to rapid growth of population, urbanization, industrialization, transport sectors and different types of fertilizers and pesticides use in agriculture sectors, etc. Nitrate contamination of water and its irrigated plants has been identified as a growing concern not just in India, but also across the world such as Australia, China, the USA, Morocco, Bangladesh, and several other countries (Kim *et al*., 2018; Uddin *et al*., 2021) [10, 23]. Due to its higher capacity of solubilization and mobilization, excess nitrate can be released in groundwater and surface water badly affects the quality of water, nitrate enters the human body via both exogenous and endogenous mechanisms, causing a variety of diseases such as brain tumors, hypertension, goiter, stomach cancer, nasopharyngeal and thyroid abnormalities, etc. (Weigelhofer and Hein 2015; Singh *et al*., 2022) [24, 21]. Nitrate contamination in groundwater is closely related to the subsurface geological properties of subsurface permeability (Haghbin *et al*., 2021) [7]. Change in climatic conditions plays an important role in groundwater nitrate contamination due to alteration in dynamics of the hydrogeological system (Ortmeyer *et al*., 2021) [16]. In the case of surface water, India has 14 major rivers which participate in 85% of the total surface flow reported by Mutiyar *et al*. (2018) [13] which contributed to the entire earth in several aspects like environmental, economic, historical, and social, etc. The chemistry of surface water is impacted by various biogeochemical processes such as chemical weathering, atmospheric precipitation, and evapocrystalization, etc. (Pant *et al*., 2018; Li *et al*., 2021) [17, 11]. The maximum permissible limit of nitrate in drinking water has been set by WHO (WHO 1993) [25] and Indian standard (BIS) are 50 and 45 mg/L, respectively. Higher occurrence of nitrate in groundwater causes hazardous effects especially on newborn babies by "Blue baby syndrome" or methemoglobinemia disease i.e., below the 6 months age baby more susceptible, due to less availability of methemoglobin reduction enzymes in RBCs (Soomro *et al*., 2017; Shukla *et al*., 2021) [22, 20]. Nitrate enrichment in groundwater due to different point sources as well as non-point sources, which percolate down from the ground surface and solubilize in the aquifer zone. The major cause of higher concentrations of nitrate in a rapid-urbanized area might be an excessive amount of NOx generated by vehicular traffic, and excretory matter of different stakeholders like a cow, buffalo, and poultry, etc., improper management of sewage, and leakage problem of fecal matter from the septic tanks and different unorganized, irregular landfill sites (Patel *et al*., 2016; Karunanidhi *et al*., 2021) [18, 8].

Some recent techniques could be applied for the detection of the source of specific pollutants, stable isotopes have been successfully applied in search of their transformations of compounds in various ecosystems (Cao *et al*., 2018) [3]. For example, δ15N nitrate and δ18O nitrate for und the mechanism of the denitrification process and identification of sources of nitrate pollution in aquifers (Nikolenko *et al*., 2018) [14]. Zhai *et al*. (2017) [26] reviewed that health risks due to nitrate varies with age and sex for example-infants and males are more susceptible to nitrate contamination than adults and females in China. According to Zhang *et al*. (2021) [27], 7.83 % of water samples in China exceeded its national nitrate threshold level (45 mg/L). Levels of nitrate exceeded 90 mg/L in the Mudan River (Linkou County), Haihe river (Beijing), and Yangtze River estuary (Shanghai), indicating severe pollution. In the eastern region of Uttar Pradesh different work has been done like Madhav *et al*. (2018) [12] found 40 % of groundwater samples of Bhadohi have nitrate content above the permissible limit (>50 mg/l) which may have harmful effects for the human health. Ahamad *et al*. (2018) [1] also studied in the Varanasi area that 80% of the total groundwater samples contaminate with nitrate which exceeded the acceptable limit. Very few data on nitrate contamination of ground and surface water from the Indian Himalayan Region and Kullu valley of Himachal Pradesh, in particular, are available. Therefore, the present study was carried out to investigate the levels of nitrate contamination in surface and groundwater in developing areas of Kullu valley in Kullu district, Himachal Pradesh, and their dietary consumption rate by the local inhabitants.

Material and Methods

Study Area

The study area covers different rural, peri-urban, and urban areas of Kullu valley. Kullu has a 2.57 lakhs population and 5500 sq. km, area (Census 2011) [4]. The altitude of sampling sites ranged between 1100-2200 m AMSL (Above mean sea level). The main sources of drinking water in the study area are hand pumps, supply or tap water and for irrigational purposes use river water and spring water.

Sampling and Analysis

The samples of water were collected in pre-acid-washed polyethylene bottles of 100 ml capacity in triplicate at each site from two different sources i.e., ground and surface water sources. The water was filled into a bottle at the interval of 30 sec. Before filling the water, the sources such as hand pump and tap water or natural water (Jaru) were kept open for five minutes. Similarly, the water samples were collected from the surface water system by immersing the bottle one by one in the river at intervals of 15 seconds. To inhibit microbial activity, 1mL of concentrated nitric acid was introduced to each bottle. The bottles were brought back to the laboratory and kept in the refrigerator at 4 ^{0}C till the analysis. All the samples of water were collected during August 2015. To eliminate undesirable soil particles and plant debris from the water samples, Whatman no. 1 filter paper was used. The different physicochemical parameters of water as pH, electrical conductivity (EC), and total dissolved solids (TDS) was measured directly using a calibrated multiparameter system (Eutech, model 3130). The same samples were used for the measurement of nitrate-nitrogen (NO_3-N) after mixing with 1M $(NH_4)_2SO_4$ in a 2:1 ratio. The samples were properly mixed and the concentration of NO_3-N was measured using an ion-selective glass electrode fitted in an ion analyzer (pH/ion 304, Germany). The glass electrode was kept in the samples for 30 sec and the reading was noted at constant variation (±2%).

Twenty-five individuals of each age group were interviewed randomly from the study area for the bodyweight and consumption rate of water daily standing. Daily intake of NO_3-N through drinking water was obtained by dividing the value obtained from multiplying the NO_3-N concentration in filtered drinking water with daily consumption of water to the bodyweight of the individual. The consumption/ intake or exposure of the local population to nitrate contamination was expressed as mg NO_3-N/day/kg body weight. The mean and standard error was also calculated using SPSS software to assess the extent of contamination levels of nitrates in both surface and groundwater. The correlations between nitrate concentrations in surface and groundwater were also determined by linear regression analysis.

Results and Discussion

The selected Physico-chemical properties of surface water (rivers and springs) and groundwater (filtered and non-filtered borewell water) are given in Table 1. The pH of the tested water samples was found below the values (7.0-8.5) set by WHO reported in Divya and Belagali (2012) [5] except spring water i.e., 8.77. The average concentration of nitrate, EC, and TDS in tested water samples was further recorded lower than the values (50 mg/l, 2500 m mhos/cm, and 500 mg/l, respectively) set by WHO for the drinking purposes (Table 1).

Table 1: Nitrate contamination and selected physicochemical properties of ground and surface water in Kullu valley, Kullu

Parameters	Water sources			
	Groundwater		Surface water	
	Non-filtered	Filtered	Rivers	Springs
N	13	5	15	5
NO_3-N (mg/l)	29.07±2.92	27.07±0.25	6.14±1.20	16.03±1.20
pH	8.46±0.04	8.44±0.02	8.27±0.05	8.77±0.03
Temperature (^{0}C)	25.24±0.09	24.87±0.03	25.63±0.06	25.92±0.04
EC (mhos/cm)	571.13±29.68	355.00±7.89	203.15±17.97	668.89±42.25
TDS (ppm)	284.38±14.59	178.67±3.86	99.36±9.15	336.44±22.15

Values are mean ± SE of N samples

The maximum concentration of nitrate in groundwater samples was recorded at Akhara and in surface water at Nogujode. Similarly, the minimum concentration of nitrate in the ground and surface water was recorded at Nogujode and Bajaura, respectively. The low and high nitrate contamination, respectively in the ground and surface water at the above sites may be due to the massive use of nitrogen-based fertilizers in agriculture and their maximum runoff to surface water and low leaching to the groundwater. The nitrate concentration in ground and surface water samples was grouped into three classes based on their concentrations as per classification made by Obeidat *et al*. (2008) [15] and the results are presented in Figure 1.

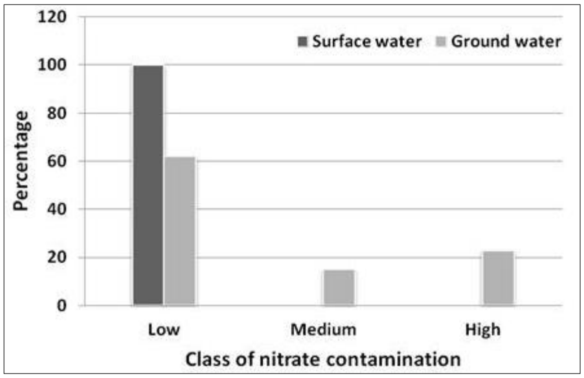

Fig 1: Percentage of nitrate contaminated study sites under different low (>20mg/l), medium (20-50 mg/l) and high (>50 mg/l) classes

These classes are as: low (<20mg/l), medium (20-50 mg/l) and moderate (>50 mg/l). The nitrate concentrations in the high class exceed the recommendation for drinking water set by WHO (1993) [25]. Study sites such as Akhara, Shamshi, and Pirdi (77.04 mg/l, 67.09 mg/l, and 63.09 mg/l, respectively) were categorized into the high concentration class. The spatial patterns of mean nitrate contamination levels in ground and surface water collected during August 2015 from different areas of Kullu valley were ranged from minimum to maximum as 4.19 mg/l to 77.7 mg/l and 3.3 mg/l to 16mg/l, respectively (Figures 2 and 3).

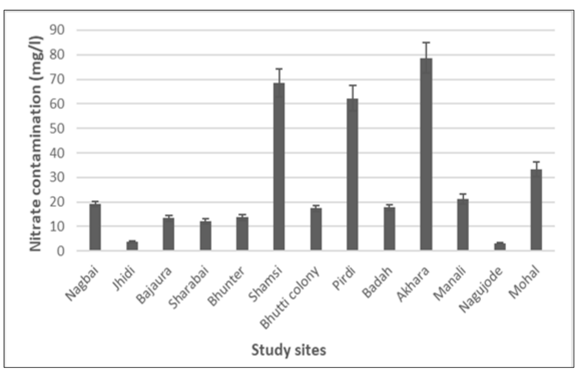

Fig 2: Spatial variation in nitrate contamination levels of groundwater collected from different locations of Kullu valley during August 2015

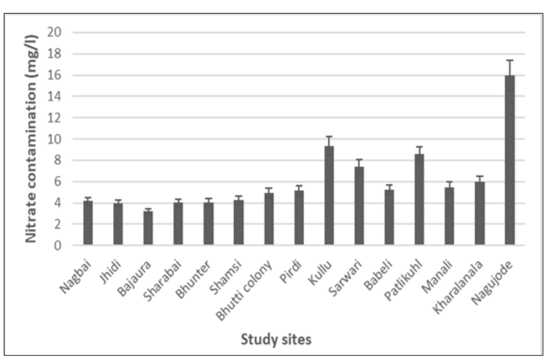

Fig 3: Spatial variation in nitrate contamination levels of surface water collected from different locations of Kullu valley during August 2015

The high concentration of nitrate in groundwater at Akhara, Shamshi, and Pirdi may be ascribed to leaching of nitrate from natural vegetation, heavy and dense population as well as leaching of nitrate-nitrogen from the septic tanks or drainages and high depth of borewells, respectively. The high-class water samples are likewise unsafe for human consumption. The study further suggests that these sites need high attention for further in-depth studies and policy intervention.

The medium class involves samples with high nitrate concentrations enough to indicate the influence of anthropogenic activities.

In the present study, 15% of the groundwater sampling sites are felled into this class. The concentrations of nitrate in surface water collected from all the sites were fallen into the low class, whereas 62% of the groundwater sampling sites were under the low class. This result indicated that water samples from these sites had a low risk to both human and environmental health. The regression analysis conducted between TDS and nitrate concentration in surface and groundwater, as well as nitrate concentration between surface and groundwater, showed significant and positive relationships (Table 2).

Table 2: Relationships between nitrate concentration and TDS in surface and groundwater, and nitrate concentration of surface and groundwater

Parameters	Regression equation	R^2	Significance levels
TDS × SW NO_3-N	TDS=6.49*SWNO_3-N + 59.49	0.10	P<0.05
TDS × GW NO_3-N	TDS=2.95*GWNO_3-N+198.70	0.35	P<0.05
GWNO_3-N × SWNO_3-N	GWNO_3-N=0.71*SWNO_3-N+ 24.03	0.29	P<0.05

GW: Groundwater, SW: Surface water

The highest degree of relationship between TDS and nitrate concentration of groundwater (R^2=0.35; p<0.05) was found which shows that TDS concentration in water influenced the nitrate contamination levels. Divya and Belagali (2012) [5] have found from their research that negatively charged nitrate ions may quickly take up positively charged basic cations like calcium, magnesium, sodium, and potassium to sustain electric charges on soil particles that will eventually be leached into either groundwater or surface water. The positive relationship between NO_3-N of surface and groundwater further indicated that NO_3-N from surface water can reach groundwater of their vicinity through leaching (Kawagoshi *et al*., 2019; Pla *et al*., 2019) [9, 19].

Table 3: Exposure of local population of different age groups to nitrate contamination through drinking water

Age group (Year)	Dietary intake of Nitrate (mg NO_3-N /day/kg body weight)
15-30	1.20
31-50	1.89
51-70	1.18

Table 3 showed the results of exposing the local population of three distinct age groups to nitrate pollution through the use of filtered water. The results showed that the exposure of the population of middle age (31-50 year) group to nitrate contamination was maximum, followed by low (15-30 year) and high (51-70 years) age group. This may be ascribed to more requirement of water during their physical or mental works as compared to other age groups.

Conclusions

The results of this study demonstrated that nitrate pollution levels in groundwater were greater than in surface water. The results further showed that groundwater contamination at Akhara, Shamshi, and Pirdi areas was exceeded the permissible limit of WHO and not safe for drinking purposes. The present study suggests that nitrate contamination of groundwater in Kullu valley should be regularly monitored with special attention to the above three areas of inhabitation.

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