Environment Conservation Journal 19 (3)17-30, 2018
ISSN 0972-3099 (Print) 2278-5124 (Online)
Abstracted and Indexed

Ion chromatography of major anions in the Neeru stream, Bhaderwah, J&K, India

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Received: 05.04.2017 **Revised: 21.05.2017** **Accepted: 28.06.2017**

Abstract

Water samples from 25 different stations along the Neeru stream and its tributaries / sub-tributaries has been collected and analysed for anions viz., Fluoride, Chloride, Nitrate, Phosphate and Sulphate for two years (Jan 2014- Dec 2015) by using Ion chromatography with 3.2 mM sodium carbonate and 1 mM sodium bicarbonate as mobile phase and 50 mM solution of sulphuric acid as suppressor. The observed values of Fluoride, Chloride, Nitrate, Phosphate and Sulphate were found to be in the range of BDL - 0.85, 0.11 - 9.88, 0.02 - 7.78, BDL - 0.39 and 0.26 - 9.56, respectively. Possible reasons for the fluctuations of these anions in the Neeru stream have also been discussed. All analysed anions have been found within WHO and BIS permissible limits.

Key words: Anions, Bhaderwah, Ion chromatography, Neeru stream, Water quality

Introduction

The chemical constitution and properties of surface water in a particular region is determined by natural processes such as precipitation rate, weathering processes, soil erosion and anthropogenic disturbances. However these may be polluted from point and non point sources such as urban, industrial and agricultural activities (Jarvie *et al*., 1998; Giridharan *et al*., 2009; Nouri *et al*., 2009 & 2011; Garizi *et al*., 2011). Quality of surface water, both lotic as well as lentic, has been continuously altered by various pollutants contributed by different anthropogenic activities (Varekar *et al*., 2015). Lotic water bodies, mostly originating from snow covered mountains and constituting a continuous source of water supply to the people living in mountainous watershed as well as downstream, play a significant role in moderating or dispersing municipal, industrial and agricultural waste waters in a watershed (Wang *et al*., 2007), however, in doing so these water bodies get polluted with different type of pollutants which makes their water unfit for drinking and other domestic purposes. Several studies has been conducted on the water quality of lotic water bodies with respect to anions and other parameters by various workers (Semwal and Jangwan, 2009; Garizi *et al*., 2011; Das *et al*., 2014; Dutta, 2014; Majumdar and Dutta, 2014; Baba *et al*., 2015; Bora and Goswami, 2015; Sharma *et al*., 2015 a, b & c; Bourasi *et al*., 2016; Dhamodharan *et al*., 2016; Khadse *et al*., 2016; Paudyal *et al*., 2016; Chandan *et al*., 2017; Rameez and Srivastava, 2017; Ebadati, 2017). However, Neeru stream, an important left bank glacial fed perennial tributary of the river Chenab, has not received due attention except for a stray study conducted by Shekhar (1990). The stream passes through various human inhabited places and also joined by various tributaries / sub-tributaries before its confluence with the River Chenab at Pul Doda. During its course of journey, it receives waste from municipal and agricultural area including biological waste from slaughters houses, which is seriously affecting its water quality. As it is facing impacts of increasing urbanization and other anthropogenic activities, thereby, monitoring of stream water quality becomes essential for present and future water quality management, for which the present study has been carried out.

Study area: Neeru stream is a glacial fed perennial left bank tributary of the river Chenab which originates from Kaplash Kund (4200 m a.s.l.) and

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equally contributed by Ashapati glacier. While draining the Neeru watershed it passes through Bhaderwah town and other densely populated villages and finally joins the river Chenab at Pul Doda (850 m a.s.l.) in the Bhaderwah tehsil of district Doda of Jammu and Kashmir (Figure 1). There is a considerable variation in altitude in the study area (850 m to 4200 m a.s.l.) which results in great climatic variation from snow laden mountains peaks in the upper reaches of Kaplash Kund and Ashapati glacier to almost dry subtropical climate at Pul Doda. This great climatic variation in the Neeru catchment also supports a rich diversity and density of both flora and fauna.

Sampling Stations: Water samples from 25 different stations (Table 1) have been collected for 2 years (Jan 2014- Dec 2015) on monthly basis from the main Neeru stream and its tributaries/ sub-tributaries for monitoring the water quality of the stream with respect to major anions. At the point of joining of a tributary with the main stream (Neeru stream), water samples has been collected from two stations - one located at 200 m before the confluence of a tributary with the main stream while the other located at 200 m after the confluence of a tributary with the main stream, to monitor the effect of water quality of that particular micro watershed on the water quality of the main stream. Geo-coordinates and altitude of different monitoring stations has been recorded using Garmin, Montana 680 (GPS). Samples were collected in the morning hours (9:00-11:00 am) in the polypropylene bottles and sealed on the spot for ion chromatographic analysis at IC laboratory in the Department of Environmental Sciences, University of Jammu, Jammu. Owing to the difficult terrain and large distance between stations, monthly sampling on 25 different stations was completed within 2-3 days.

Material and Methods

Chemicals and Reagents

Mobile Phase: Mobile phase was prepared using 3.2 mM (milli molar) sodium carbonate and 1 mM sodium bicarbonate in a volumetric flask of 1000ml.

Supressor: 50 mM solution of sulphuric acid was used as suppressor.

Standard: IC Multi-elements standard I for anions (F^- = 100 mg/l, Cl^- = 250 mg/l, NO_3^-, SO_4^{2-} = 500 mg/l and PO_4^{3-} = 1000 mg/l) manufactured by Merck (Millipore) were used for calibration of the IC system.

Ultrapure water: Ultrapure water was prepared using Millipore (DirectQ-3 with pump).

Instrument: Metrohm Ion Chromatograph system (model 850 professional) integrated with Compact Autosampler (model 863) through a computer system has been used for analysis.

Analysis

a) **Sample preparation:** Samples were prepared using 1:10 dilution factor (i.e., one ml of sample was raised to 10 ml using ultrapure water) to avoid ionic saturation of the column.
b) **Sample loading:** Properly marked sample vials were arranged in the Autosampler for analysis. Information about the sample identification and position was entered in the determination series in the workplace of the MagIC Net software.
c) **Sample analysis:** Calibration of the system was done using IC Multi-elements standard solution . 0.1 ml of stock standard solution was raised to 10 ml to get concentrations of F^- = 1 ppm, Cl^- = 2.5 ppm, NO_3^-, SO_4^{2-} = 5 ppm and PO_4^{3-} = 10 ppm. Similarly standards of higher concentration were made for calibration of the IC system. Flow rate of 0.7 mL/min and Pressure of 12.26 MPa was maintained in the Metrosep A Supp 5 – 250/4.0 column. 20 µl loop was used for sample injection and 30 min of recording time was set for each sample.
d) **Reprocessing of Results:** After scheduled recording time of 30 minutes results were obtained in the Database of IC software. The results were reprocessed using 1 ppm standard to phase out any errors in the retention time and percentage window for analyte under consideration.

Results and Discussion

Average monthly concentration of Fluoride, Chloride, Nitrate, Phosphate and Sulphate for two years (Jan 2014- Dec 2015) in the main stream and tributaries/ sub-tributaries has been presented in the Tables 2a & b, 3a & b, 4a & b, 5a & b and 6a & b, respectively.

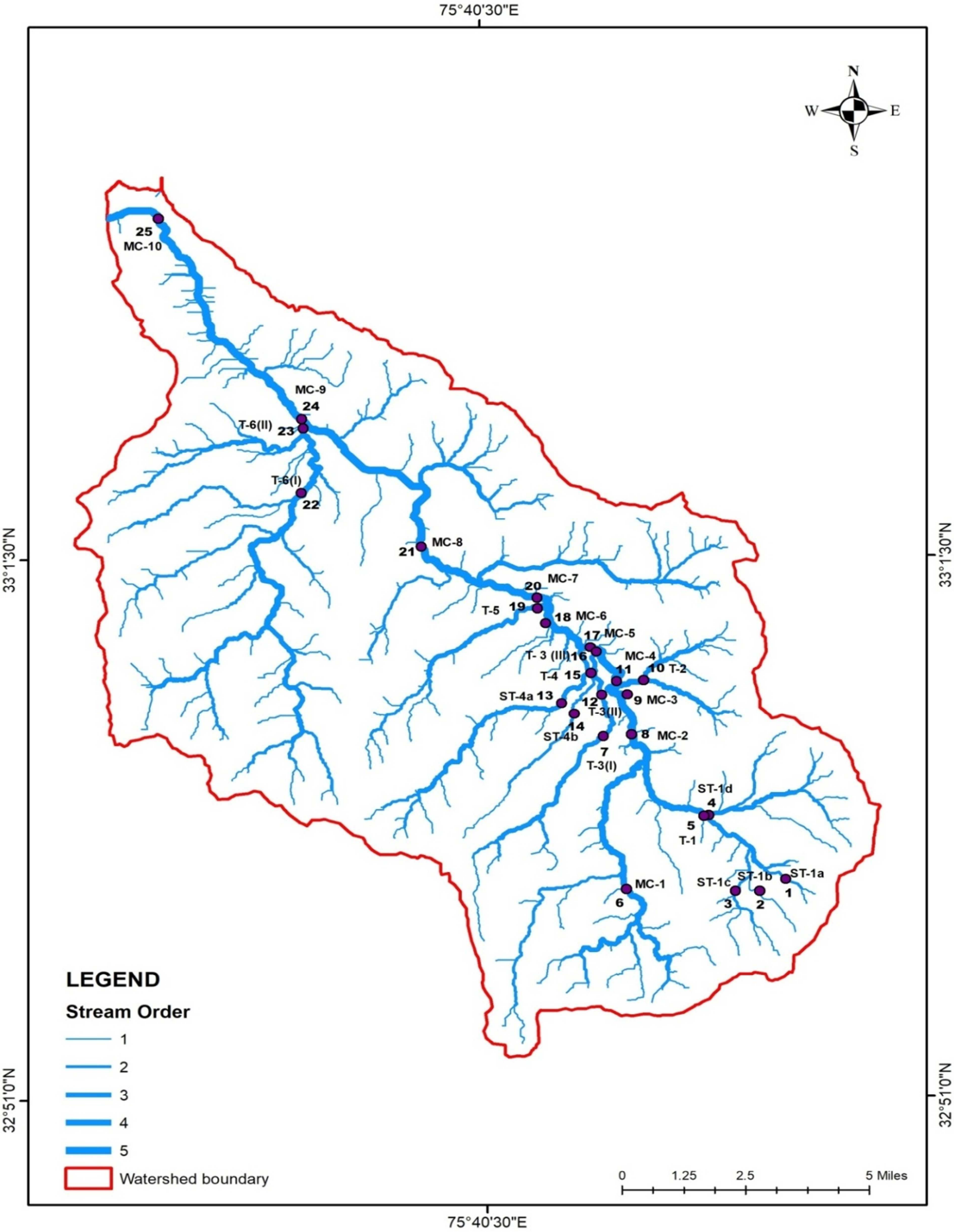

Figure 1: Map showing different sampling stations and stream order in the Neeru watershed.

Table 1: Details of the different sampling stations at Neeru stream and its tributaries/ sub-tributaries.

S. No.	Station Name	Station Code	Impact Category	Geo-Coordinates	Altitude (in meters)
1.	**Thanalla I**	**ST-1a**	Baseline Station	32^0 55' 13.3" N 75^0 46' 46.6" E	2240
2.	**Thanalla II**	**ST-1b**	Baseline Station	32^0 54' 59.8" N 75^0 46' 13.5" E	2184
3.	**Thanalla III**	**ST-1c**	Baseline Station	32^0 54' 59.9" N 75^0 45' 43.4" E	2156
4.	**Bheja I**	**ST-1d**	Trend or Impact Station	32^0 56' 28.5" N 75^0 45' 10.5" E	1823
5.	**Bhaja II**	**T-1**	Trend or Impact Station	32^0 56' 27.6" N 75^0 45' 04.7" E	1815
6.	**Thanthera**	**MC-1**	Baseline Station	32^0 55' 03.0" N 75^0 43' 26.5" E	2163
7.	**Puneja**	**T-3(I)**	Trend or Impact Station	32^0 58' 01.2" N 75^0 42' 59.4" E	1733
8.	**Dareja**	**MC-2**	Trend or Impact Station	32^0 58' 03.2" N 75^0 43' 34.4" E	1683
9.	**Gupt Ganga**	**MC-3**	Trend or Impact Station	32^0 58' 49.4" N 75^0 42' 57.4" E	1638
10.	**Atalgarh**	**T-2**	Trend or Impact Station	32^0 58' 39.7" N 75^0 42' 07.1" E	1682
11.	**Renda**	**MC-4**	Trend or Impact Station	32^0 58' 27.5" N 75^0 42' 22.6" E	1700
12.	**Dharampura**	**T-3(II)**	Trend or Impact Station	32^0 59' 15.1" N 75^0 42' 44.0" E	1572
13.	**Launcher Morh**	**ST-4a**	Trend or Impact Station	32^0 58' 49.5" N 75^0 43' 29.2" E	1628
14.	**Hallayan**	**ST-4b**	Trend or Impact Station	32^0 59' 06.6" N 75^0 43' 50.1" E	1636
15.	**College link Road**	**T-4**	Trend or Impact Station	32^0 59' 05.2" N 75^0 43' 16.2" E	1553
16.	**Sarol Bagh I**	**T-3(III)**	Trend or Impact Station	33^0 00' 13.3" N 75^0 41' 47.6" E	1467
17.	**Sarol Bagh II**	**MC-5**	Trend or Impact Station	33^0 00' 43.1" N 75^0 41' 37.2" E	1423
18.	**Domail**	**MC-6**	Trend or Impact Station	33^0 00' 30.8" N 75^0 41' 37.9" E	1480
19.	**Hanga *Nallah***	**T-5**	Trend or Impact Station	32^0 59' 40.3" N 75^0 42' 51.0" E	1521
20.	**Amira Nagar**	**MC-7**	Trend or Impact Station	32^0 59' 45.3" N 75^0 42' 43.2" E	1510
21.	**Dhrudu**	**MC-8**	Trend or Impact Station	33^0 01' 43.4" N 75^0 39' 12.3" E	1334
22.	**Mallothi**	**T-6(I)**	Trend or Impact Station	33^0 02' 46.9" N 75^0 36' 42.3" E	1357
23.	**Bhalla I**	**T-6(II)**	Trend or Impact Station	33^0 04' 02.3" N 75^0 36' 45.1" E	1202
24.	**Bhalla II**	**MC-9**	Trend or Impact Station	33^0 04' 12.9" N 75^0 36' 43.3" E	1185
25.	**Galgander**	**MC-10**	Trend or Impact Station	33^0 08' 07.5" N 75^0 33' 44.4" E	863

MC= Main Stream, T= Tributary, ST= Sub Tributary

Table 2a: Average monthly fluoride concentration at different stations along Neeru stream for two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
MC-1 (S6)	BDL	BDL	BDL	0.07	0.11	0.04	0.13	0.03	0.06	0.13	0.02	BDL
Impact/trend stations												
MC-2 (S8)	BDL	BDL	0.11	0.24	0.83	0.38	0.18	0.17	0.07	0.06	BDL	BDL
MC-3 (S9)	BDL	0.06	0.03	0.19	0.48	0.19	0.14	0.10	0.06	0.05	BDL	BDL
MC-4 (S11)	BDL	BDL	0.09	0.08	0.17	0.08	BDL	BDL	BDL	0.06	BDL	0.16
MC-5 (S17)	BDL	BDL	0.04	0.09	0.21	0.12	0.10	0.10	0.09	BDL	BDL	BDL
MC-6 (S18)	BDL	BDL	0.09	0.08	0.17	0.08	BDL	BDL	BDL	0.06	BDL	0.16
MC-7 (S20)	BDL	0.05	0.06	0.16	0.85	0.46	0.14	0.11	0.11	0.13	0.06	BDL
MC-8 (S21)	BDL	BDL	0.05	0.08	0.36	0.15	0.13	0.11	0.10	0.07	0.05	BDL
MC-9 (S24)	BDL	BDL	0.09	0.13	0.14	0.15	0.12	0.10	0.09	0.07	0.03	BDL
MC-10(S25)	BDL	BDL	BDL	0.11	0.18	0.16	0.12	0.11	0.09	0.05	BDL	BDL

Table 2b: Average monthly fluoride concentration at different stations on tributaries/ sub-tributaries for two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
ST-1a (S1)	BDL	BDL	BDL	0.07	0.07	0.14	0.39	0.10	0.16	0.09	0.06	BDL
ST-1b (S2)	BDL	BDL	BDL	0.08	0.03	0.04	0.03	0.05	0.03	BDL	BDL	BDL
ST-1c (S3)	BDL	BDL	BDL	0.02	0.17	0.20	0.04	0.02	BDL	BDL	BDL	BDL
Impact/trend stations												
ST-1d (S4)	BDL	BDL	BDL	0.05	0.69	0.12	0.03	0.06	0.03	BDL	BDL	BDL
T-1 (S5)	BDL	0.04	0.08	0.14	0.21	0.22	0.17	0.14	0.20	0.06	BDL	BDL
T-2 (S10)	BDL	0.04	0.10	0.16	0.57	0.19	0.1	0.15	0.10	0.04	0.02	BDL
T-3(I) (S7)	0.06	0.02	0.12	0.3	0.62	0.15	0.05	0.07	0.08	0.13	0.09	BDL
T-3(II) (S12)	BDL	0.05	0.17	0.21	0.76	0.21	0.15	0.06	0.13	0.09	BDL	BDL
T-3 (III)(S16)	BDL	BDL	0.08	0.09	0.19	0.11	0.09	0.07	0.04	BDL	BDL	BDL
ST-4a (S13)	BDL	0.14	0.34	0.84	0.20	0.15	0.11	0.07	0.03	BDL	BDL	BDL
ST-4b (S14)	0.06	0.04	0.08	0.37	0.64	0.13	0.13	0.11	0.12	0.14	0.06	0.04
T-4 (S15)	BDL	BDL	0.12	0.17	0.31	0.30	0.15	0.16	0.10	0.06	0.03	BDL
T-5 (S19)	BDL	0.04	0.07	0.13	0.21	0.20	0.13	0.12	0.07	0.06	BDL	BDL
T-6(I) (S22)	BDL	BDL	0.06	0.13	0.20	0.18	0.17	0.12	0.11	0.09	0.03	BDL
T-6(II) (S23)	BDL	0.05	0.09	0.12	0.16	0.14	0.13	0.13	0.12	0.10	0.09	0.03

BDL- Below Detection Limits

Table 3a: Average monthly chloride concentration at different stations along Neeru stream for two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
MC-1 (S6)	1.49	1.34	1.22	0.67	0.48	0.51	0.54	0.81	0.92	0.97	1.15	1.37
Impact/trend stations												
MC-2 (S8)	2.29	1.87	1.33	1.08	0.80	1.00	1.11	1.23	1.64	1.81	1.91	2.21
MC-3 (S9)	2.93	2.35	1.71	1.46	1.23	1.54	1.79	1.91	2.14	2.28	2.38	2.89
MC-4 (S11)	3.44	3.05	2.64	1.36	1.07	1.27	1.55	1.96	2.21	2.44	2.62	3.15
MC-5 (S17)	5.65	4.35	2.27	1.64	1.32	1.78	2.29	3.66	3.96	4.37	5.37	6.19
MC-6 (S18)	3.74	3.05	2.54	1.72	1.36	1.47	1.55	1.96	2.11	2.24	2.42	3.35
MC-7 (S20)	4.75	3.95	3.02	1.35	1.04	1.38	2.17	2.78	3.17	3.54	3.81	4.56
MC-8 (S21)	4.99	3.54	3.14	1.88	1.53	1.74	2.33	3.31	4.35	4.55	5.39	5.45
MC-9 (S24)	4.01	4.26	3.73	1.57	1.21	1.48	2.65	3.32	4.05	4.17	4.28	4.59
MC-10 (S25)	5.45	4.53	4.11	2.13	1.23	1.77	2.32	3.28	4.34	4.66	4.84	5.06

Table 3b: Average monthly chloride concentration at different stations on tributaries/ sub-tributaries for two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
ST-1a (S1)	1.65	1.46	1.37	0.73	0.35	0.27	0.21	0.98	1.19	1.29	1.45	1.61
ST-1b (S2)	0.75	0.54	0.43	0.19	0.11	0.21	0.26	0.37	0.58	0.61	0.73	0.76
ST-1c (S3)	1.25	1.13	0.91	0.35	0.31	0.47	0.61	0.64	0.69	0.73	0.9	1.16
Impact/trend stations												
ST-1d (S4)	2.45	2.13	1.86	0.83	0.6	0.77	0.97	1.02	1.49	1.95	2.19	2.55
T-1 (S5)	2.06	1.67	1.52	0.94	0.72	0.85	1.01	1.24	1.29	1.37	1.93	2.04
T-2 (S10)	3.61	3.16	2.99	1.57	1.83	2.07	2.51	2.54	2.75	2.83	3.07	3.71
T-3(I) (S7)	2.82	2.74	2.57	1.35	0.82	1.13	1.55	2.04	2.23	2.32	2.52	2.68
T-3(II) (S12)	9.08	9.31	7.58	2.11	1.70	2.25	3.66	4.32	5.64	6.41	7.43	9.59
T- 3 (III) (S16)	9.18	9.11	7.6	2.65	2.42	3.33	3.87	7.54	9.4	9.08	9.88	8.95
ST-4a (S13)	1.36	1.09	0.84	0.65	0.55	0.59	0.78	0.94	1.02	1.14	1.28	1.44
ST-4b (S14)	2.99	2.14	1.67	1.24	1.42	1.65	2.23	2.50	2.87	2.75	2.5	2.67
T-4 (S15)	7.41	7.33	6.88	2.76	1.83	2.44	4.43	4.94	6.42	6.97	7.93	7.70
T-5 (S19)	4.34	3.31	2.59	1.33	1.13	1.41	1.89	2.21	2.32	3.02	3.18	3.63
T-6(I) (S22)	2.93	2.31	1.89	1.22	0.95	1.21	1.83	1.98	2.42	2.43	2.23	2.68
T-6(II) (S23)	3.12	2.55	2.12	1.47	1.18	1.43	2.44	2.72	2.53	2.47	3.01	3.08

Table 4a: Average monthly nitrate concentration at different stations along Neeru stream for two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
MC-1 (S6)	0.60	1.25	1.32	5.10	2.87	2.42	1.99	1.81	1.41	1.09	0.84	0.56
Impact/trend stations												
MC-2 (S8)	0.77	1.49	1.73	1.88	4.18	3.69	1.37	1.25	0.85	0.18	0.08	0.04
MC-3 (S9)	1.67	2.17	2.69	2.95	5.65	2.67	2.25	1.66	1.38	0.47	0.31	0.28
MC-4 (S11)	0.05	0.94	2.79	3.70	5.07	3.31	1.46	1.42	1.35	0.7	0.4	0.36
MC-5 (S17)	0.44	0.62	1.66	3.54	3.6	1.22	1.16	0.9	0.89	0.67	0.48	0.22
MC-6 (S18)	0.05	0.94	2.79	3.70	5.07	3.31	1.46	1.42	1.35	0.7	0.4	0.36
MC-7 (S20)	0.64	0.78	2.04	4.05	7.33	6.68	3.55	3.48	3.19	1.97	1.4	0.78
MC-8 (S21)	0.03	0.76	1.33	1.92	2.69	1.61	1.19	0.17	0.14	0.11	0.07	0.06
MC-9 (S24)	1.13	1.28	4.34	6.59	7.01	5.42	2.27	2.1	1.95	1.13	0.78	0.70
MC-10 (S25)	0.53	1.04	1.50	2.42	3.96	3.58	1.28	0.96	0.79	0.69	0.48	0.39

Table 4b: Average monthly nitrate concentration at different stations on tributaries/ sub-tributaries two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
ST-1a (S1)	1.86	2.35	2.60	2.85	3.46	5.88	5.47	6.1	1.66	2.00	1.45	2.24
ST-1b (S2)	1.98	2.41	2.63	4.01	2.22	3.35	2.09	2.00	1.76	1.40	1.32	0.85
ST-1c (S3)	1.99	3.02	3.53	3.53	2.82	3.03	2.21	1.85	1.54	1.38	1.30	1.17
Impact/trend stations												
ST-1d (S4)	0.20	1.46	2.09	2.72	1.88	3.72	2.21	1.23	1.18	1.28	1.41	1.26
T-1 (S5)	1.52	2.56	3.08	3.65	4.28	0.35	0.41	0.3	0.22	0.68	0.95	1.22
T-2 (S10)	1.60	1.74	1.81	1.88	5.45	3.94	1.67	1.64	0.56	0.51	0.38	0.18
T-3(I) (S7)	0.02	0.13	2.10	1.84	1.75	1.46	1.34	1.25	0.81	0.13	0.12	0.10
T-3(II) (S12)	7.78	6.32	6.02	4.43	2.40	2.80	3.36	3.83	4.50	6.10	6.15	7.10
T- 3 (III)(S16)	6.66	5.39	4.10	3.21	2.73	3.23	4.53	4.45	5.05	6.06	6.61	7.56
ST-4a (S13)	0.73	1.38	4.07	3.22	1.44	2.39	0.57	1.14	0.93	0.71	0.15	0.10
ST-4b (S14)	0.57	0.80	1.72	1.91	2.02	1.81	1.77	1.12	0.55	0.31	0.28	0.20
T-4 (S15)	7.4	6.15	4.66	3.90	3.15	4.12	4.40	5.07	5.21	6.06	6.39	7.56
T-5 (S19)	0.59	0.73	2.14	2.51	2.91	1.95	0.73	0.48	0.27	0.15	0.13	0.11
T-6(I) (S22)	0.13	0.20	1.68	1.88	3.46	3.41	1.76	1.43	0.48	0.13	0.05	0.05
T-6(II) (S23)	0.71	1.30	1.39	2.44	2.52	2.07	0.99	0.72	0.60	0.56	0.40	0.29

Table 5a: Average monthly phosphate concentration at different stations along Neeru stream for two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
MC-1 (S6)	BDL	BDL	BDL	BDL	0.05	0.24	0.19	0.09	BDL	BDL	BDL	BD
Impact/trend stations												
MC-2 (S8)	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
MC-3 (S9)	0.02	0.05	0.07	0.13	0.15	0.12	0.11	0.1	0.1	0.09	0.08	0.04
MC-4 (S11)	0.02	0.05	0.07	0.12	0.17	0.11	0.10	0.09	0.07	0.06	0.05	0.03
MC-5 (S17)	BDL	BDL	BDL	0.05	0.13	0.04	BDL	BDL	BD	BDL	BDL	BDL
MC-6 (S18)	0.02	0.05	0.07	0.12	0.17	0.11	0.1	0.09	0.07	0.06	0.05	0.03
MC-7 (S20)	0.06	0.09	0.12	0.17	0.25	0.17	0.15	0.12	0.08	0.07	0.06	0.05
MC-8 (S21)	BDL	BDL	BDL	0.04	0.09	0.08	0.03	BDL	BDL	BDL	BDL	BDL
MC-9 (S24)	BDL	BDL	0.04	0.06	0.13	0.10	0.09	0.08	0.07	0.06	0.05	BDL
MC-10 (S25)	BDL	BDL	BDL	BDL	0.09	0.07	BDL	BDL	BDL	BDL	BDL	BDL

Table 5b: Average monthly phosphate concentration at different stations on tributaries/ sub-tributaries for two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
ST-1a (S1)	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
ST-1b (S2)	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
ST-1c (S3)	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Impact/trend stations												
ST-1d (S4)	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
T-1 (S5)	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
T-2 (S10)	0.02	0.06	0.12	0.18	0.39	0.17	0.16	0.15	0.11	0.07	0.04	0.03
T-3(I) (S7)	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
T-3(II) (S12)	0.06	0.08	0.11	0.17	0.25	0.16	0.14	0.12	0.11	0.09	0.08	0.05
T-3 (III)(S16)	0.09	0.07	0.09	0.11	0.15	0.1	0.08	0.07	0.06	0.09	0.07	0.09
ST-4a (S13)	BDL	BDL	0.05	0.15	0.17	0.11	0.05	BDL	BDL	BDL	BDL	BDL
ST-4b (S14)	0.03	0.06	0.13	0.15	0.28	0.16	0.11	0.1	0.09	0.07	0.03	0.03
T-4 (S15)	0.17	0.12	0.15	0.12	0.35	0.25	0.21	0.14	0.13	0.12	0.14	0.12
T-5 (S19)	BDL	BDL	BDL	0.09	0.12	0.06	BDL	BDL	BDL	BDL	BDL	BDL
T-6(I) (S22)	BDL	BDL	BDL	0.02	0.05	0.09	0.06	BDL	BDL	BDL	BDL	BDL
T-6(II) (S23)	BDL	BDL	0.04	0.09	0.14	0.11	0.09	0.08	BDL	BDL	BDL	BDL

BDL- Below Detection Limits

Table 6a: Average monthly sulphate concentration at different stations along Neeru stream for two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
MC-1 (S6)	3.69	1.93	1.63	1.53	0.73	1.13	2.15	2.29	2.47	2.79	2.93	3.59
Impact/trend stations												
MC-2 (S8)	4.48	2.07	2.03	1.12	0.85	1.25	1.64	1.88	3.39	4.02	4.23	4.72
MC-3 (S9)	2.95	2.12	1.79	1.76	0.84	2.17	3.38	4.78	4.22	2.24	1.91	1.46
MC-4 (S11)	4.59	4.51	2.88	1.83	1.28	1.91	3.48	3.85	3.84	3.24	3.43	3.80
MC-5 (S17)	2.93	2.64	2.47	1.58	1.39	1.86	3.06	4.08	3.62	3.46	3.21	2.65
MC-6 (S18)	4.59	4.51	2.88	1.83	1.28	1.91	3.48	3.85	3.84	3.24	3.43	3.80
MC-7 (S20)	4.91	4.35	4.09	1.94	1.27	1.67	2.8	4.19	3.84	3.03	2.87	2.39
MC-8 (S21)	5.57	5.23	4.18	2.28	1.18	1.75	3.91	6.00	5.33	5.07	4.96	3.80
MC-9 (S24)	3.86	3.17	2.87	1.87	1.74	2.26	2.65	4.08	3.51	3.46	2.88	2.71
MC-10 (S25)	3.78	3.05	2.77	1.94	1.34	1.54	3.26	4.66	3.74	3.57	3.5	3.89

Table 6b: Average monthly sulphate concentration at different stations on tributaries/ sub-tributaries for two years (Jan 2014 to Dec 2015).

Station / Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Baseline stations												
ST-1a (S1)	2.79	2.34	2.12	1.89	1.33	2.09	3.23	5.27	3.37	3.68	3.5	2.37
ST-1b (S2)	2.17	1.71	1.48	1.09	0.9	0.26	0.68	1.14	1.9	2.63	1.66	2.02
ST-1c (S3)	2.37	1.81	1.53	1.13	1.14	1.08	1.14	1.57	1.83	2.39	2.55	2.68
Impact/trend stations												
ST-1d (S4)	9.56	7.83	6.96	6.1	4.85	4.79	6.05	6.18	7.61	7.67	8.74	9.18
T-1 (S5)	7.38	6.04	5.36	3.33	2.06	2.66	4.01	4.17	5.86	6.65	6.53	8.64
T-2 (S10)	4.30	4.07	3.93	2.78	2.39	2.76	3.4	4.48	3.99	3.29	3.03	3.22
T-3(I) (S7)	3.07	2.34	2.14	1.55	0.78	0.82	0.97	1.73	1.98	2.1	2.46	3.02
T-3(II) (S12)	3.06	2.32	2.16	1.13	0.83	1.01	2.66	3.72	2.66	2.45	2.36	1.23
T- 3 (III)(S16)	2.04	2.58	2.19	1.72	1.11	1.33	2.37	3.36	2.81	1.84	1.82	1.66
ST-4a (S13)	2.83	3.51	1.87	0.86	1.45	2.18	3.45	4.37	3.17	2.81	3.01	3.17
ST-4b (S14)	2.61	2.09	1.89	1.73	1.29	1.65	2.62	3.69	2.99	2.67	2.59	2.46
T-4 (S15)	2.06	2.01	1.96	1.56	1.24	1.27	1.56	2.54	2.22	2.09	1.88	1.67
T-5 (S19)	2.27	2.31	1.33	1.6	1.01	1.19	2.13	2.88	2.06	1.56	1.68	1.57
T-6(I) (S22)	1.48	1.52	1.25	1.2	0.82	0.88	1.59	1.66	1.4	1.34	1.24	1.76
T-6(II) (S23)	2.66	2.13	1.64	1.35	1.15	1.18	1.7	2.96	2.82	2.73	2.7	1.98

Table 7: WHO and BIS drinking water acceptable and permissible limits for analysed anions.

S. No.	Anion Species	WHO, 1999		BIS, 2012	
		Acceptable	Permissible	Acceptable	Permissible
1.	Fluoride (mg/l)	*	*	1.0	1.5
2.	Chloride (mg/l)	250	1000	250	1000
3.	Nitrate (mg/l)	45	100	45	No relaxation
4.	Sulphate (mg/l)	200	400	200	400
5.	Phosphate (mg/l)	0.1	1.0	*	*

* Limits not provided by the concerned agency.

Table 8a: Correlation analysis of various analysed anions at main stream stations.

Anions	Fluoride	Chloride	Nitrate	Phosphate	Sulphate
Fluoride	1				
Chloride	-0.84	1			
Nitrate	0.89*	-0.93	1		
Phosphate	0.93*	-0.95	0.90*	1	
Sulphate	-0.82	0.82*	-0.91	-0.82	1

*Statistically significant positive relationships, $p < 0.05$.

Table 8b: Correlation analysis of various analysed anions at stations on tributaries/ sub-tributaries.

Anions	Fluoride	Chloride	Nitrate	Phosphate	Sulphate
Fluoride	1				
Chloride	-0.92	1			
Nitrate	0.63*	-0.66	1		
Phosphate	0.98*	-0.92	0.70*	1	
Sulphate	-0.87	0.88*	-0.79	-0.91	1

*Statistically significant positive relationships, $p < 0.05$.

Acceptable and permissible limits of various analysed anions as per WHO and BIS have been listed in the Table 7. Correlation between various anions at sampling stations located on main stream and tributaries / sub-tributaries has been presented in the Tables 8a and 8b, respectively. Perusal of Tables 2 a & b revealed that the average monthly value of Fluoride, an important parameter to access the water quality for drinking purpose, in main stream ranges from a maximum of 0.85 mg/l in the month of May (MC-7) to a minimum of BDL at different stations during different months whereas in tributaries / sub-tributaries, average monthly fluoride concentration ranged from a minimum value of BDL at different stations during different months to a maximum of 0.84 mg/l in the month of April (ST-4a). Concentrations of Fluoride present in most natural waters are small, generally less than 1.0 mg/l (Hem, 1985). The comparatively higher concentration of fluoride in tributaries / sub-tributaries T-3 I, T-3 II and ST-4a, can be attributed to the geological contribution by micro-watersheds drained by these tributaries / sub-tributaries. Karthikeyan *et al*., (2010) has also attributed presence of Fluoride in underground water to geological deposits and geochemistry of the location. Along the longitudinal profile of the Neeru stream, fluoride values showed decreasing trend from upstream to the downstream areas. Though, little bit higher concentration of fluoride has been detected at some stations (MC-7, T-3 I and ST-4a) it has remained within the permissible limits of 1.5 mg/l (BIS, 2012) (Table 7) in the Neeru stream. However, Fluoride concentrations exceeded the maximum permissible limits of 1.5 mg/l WHO (2004) in about 22.7% of the drinking water samples collected from Doda district by Central Ground Water Board (2010), which results in the prevalence of the fluorosis in some of the region of District Doda (Arya *et al*., 2013).

Chloride is a major component which maintains the cation-anion balance of ecosystem. Concentration of Chloride ranged between minimum average monthly values of 0.11 mg/l in the month of May (ST-1b) to a maximum value of 9.88 mg/l recorded in the month of November (T-3 III). The value of chlorides in the Neeru stream has been observed to be comparatively lower than some of the other Indian rivers where the concentration of chloride has been found to range between 198.57 - 1200.00 mg/l (Vaishali and Punita, 2013). However, similar lower values of Chloride in the River Bhagirathi, River Torsa and River Teesta and stream Relli has been observed by Gautam (1990), Bhadra *et al.*, (2003) and Acharjee (2013), respectively. Thresh *et al.*, (1949) and Sharma *et al.*, (2015a) suggested Chloride concentration as indication of organic waste primarily of animal origin. In the present investigation, slightly higher values of Chloride in colder periods (October to March) have been recorded at sampling stations (T-3II, T-3III and T-4) affected by sewage and municipal waste of the Bhaderwah town. Along the longitudinal profile of the Neeru stream, Chloride did not show much fluctuation. In general, concentration of Chloride remained within the acceptable limits of 250 mg/l prescribed by WHO (1999) and BIS (2012).

Nitrate and ammonium, the forms of nitrogen, are very important elements in water fertility. The monthly variation in the Nitrate content in main stream and tributaries / sub-tributaries does not exhibit much variation and its value varies between a minimum of 0.02 mg/l (January) to a maximum of 7.78 mg/l (January) at different stations (Tables 4, a & b). The maximum concentration of ammonium (100.59 mg/l) has been recorded in the month of May and minimum (0.19 mg/l) in the month of January. In Snow fed water bodies like Neeru stream, higher concentrations of nitrates / ammonium during the warmer months can be attributed to melting of snow during spring /summer season which brings the nutrient rich water to the stream. The low value of nitrates / ammonium during colder months might be due to absence of melting of snow. Wetzel (2001) reported the higher content of nitrate in snow as compared to rain and also that rise of nitrates during summers is due to melting of snow. Also the higher values of Nitrate / ammonium during warmer months can be attributed to increase in the process of nitrification at higher temperature that convert organic matter to nitrates and nitrites (Kumar and Singh, 2002; Sivakumar and Jaganathan, 2002). The transport of nitrate from the watershed areas with the runoff water during early summers also contribute towards the highest concentrations of nitrate as has also been recorded in the present study during early summer period in which the study area experience the local rains. This view has also been supported by the findings of Jha and Barat (2003) and Acharjee (2013). Along the longitudinal profile of the Neeru stream nitrates / ammonium did not show much fluctuation, however higher concentrations have been recorded in the middle section of the stream where it receives the tributaries draining the inhabited / agricultural areas. The observed values of Nitrate were found to be much below the acceptable limit of 45mg/l (WHO, 1999; BIS, 2012) (Table 7).

The main supply of Phosphate in natural waters comes from the weathering of phosphorus bearing rocks, leaching of soils of the catchment area by rain, cattle dung and night soil (Jhingran, 1982). From the perusal of the Tables 5 a & b, it has been observed that Phosphate in main stream / tributaries / sub-tributaries ranged between minimum values of BDL in all baseline stations and some trend or impact stations to a maximum of 0.39 mg/l in the month of May at T-2 station. Low values of Phosphate have been reported from various Himalayan rivers (Chakraborty, 1998; Dobriyal *et al.*, 1993; Barat and Jha, 2002 and Thapa *et al.*, 2010). The release of Phosphate from sediments might also contribute to the enrichment of Phosphate during early summer. The increased solar radiations also encourage the biological degradation of organic matter and subsequent release of more Phosphate in early summer season (Kumar and Singh, 2002). Weathering of phosphorus bearing rocks, leaching of soils rich in Phosphorus from catchment area by rains also contribute for higher phosphate in the spring seasons. Chakraborty *et al.*, (1959) has also reported higher Phosphate value during spring due to washings received from catchment areas. Low phosphate level during winter may be due to its sedimentation in the form of ferric complexes in the soil, low Calcium level in the water and low water temperature as has also been reported by Seena yya (1971) and Khan and Siddiqui (1974). Along the longitudinal profile of the Neeru stream, Phosphate did not show much fluctuation, however higher concentrations have been recorded in the middle section of the stream where it receives the tributaries draining the inhabited /agricultural areas. The amount of Phosphate found in the present study is mostly within the permissible limits prescribed by World Health Organization (1999) i.e., 1 mg/l (Table 7). Sulphur, an important component in protein metabolism (Pandey *et al*., 2000), is also ecologically important for the growth of plants as its short supply may inhibit the development of plankton. In the present investigation, the monthly variations in the sulphates contents varied from a minimum of 0.26 mg/l in the month of June to a maximum value of 9.56 mg/l in the month of January (Tables 6, a and b). Similar monthly variation has also been observed by Baurasi *et al*. (2016) and Patel *et al*. (2016). Along the longitudinal profile of the river, Sulphates did not show much fluctuation and its concentrations were found to be much below the acceptable limits of 200 mg/l as prescribed by WHO (1999) and BIS (2012).

Correlation analysis revealed significant positive correlation between Fluoride and Nitrate, r(10) = 0.89, 0.63; Fluoride and Phosphate, r (10) = 0.93, 0.98; Chloride and Sulphate, r (10) = 0.82, 0.88 and Nitrate and Phosphate, r (10) = 0.90, 0.70 while significant negative correlation has been observed between Fluoride and Chloride, r (10) = -0.84, -0.92; Fluoride and Sulphate, r (10) = -0.82, -0.87; Chloride and Nitrate, r (10) = -0.93, -0.66; Chloride and Phosphate, r (10) = -0.95, -0.92; Nitrate and Sulphate, r (10) = -0.91, -0.79 and Phosphate and Sulphate, r (10) = -0.82, -0.91 among all the stations located on the main stream as well as on the tributaries / sub-tributaries at $p < 0.05$, respectively (Table 8a & 8b). Significant positive correlation between pair of anions shows their similar source or chemical process (Sekabira *et al*., 2010; Gandhi *et al*., 2017).

Conclusion

Analysed anions viz., Fluoride, Chloride, Nitrate, Phosphate and Sulphate were found to be within the permissible limits as prescribed by WHO (1999) and BIS (2012). Slightly higher concentrations of Fluoride are contributed by sub-tributaries, ST-4a & 4b, during peak flow months which may be attributed to the increased runoff and geological composition of the micro-watersheds drained by these sub-tributaries. Higher concentrations of Chloride, Nitrate and Phosphate as compared to the rest of the watershed has been observed at stations T-3(II), T-3(III) and T-4, present in the highly urbanised areas (Bhaderwah town) can be attributed to municipal solid waste, sewage, agricultural runoff and biological waste from slaughter houses which directly or indirectly find their way in these tributaries. Sulphate in higher concentration has been observed at station ST-1d which may be due to the increased deforestation and resulting accelerated soil erosion in the micro-watershed drained by the sub-tributary. Therefore, it can be concluded that rapidly growing population, increased urbanisation and anthropogenic activities in the Neeru watershed are negatively affecting the water quality of the Neeru Stream.

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