НАУКИ О ЗЕМЛЕ

HYDROCHEMISTRY OF A SMALL RIVER IN A BIG CITY: RIVER HRAZDAN AND CITY OF YEREVAN, ARMENIA

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Abstract The article contains a study of River Hrazdan water quality within of Yerevan over a 6-year period (2012-2017) by some hydrochemical parameters: physico-chemical, of water salt contents, mineral nitrogen-containing substances, nitrates and phosphates, heavy metals Ag, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, Zn, Hg, As. The research included the determination of spatial (8 permanent stations) and seasonal (high- and low water) dynamics of hydrochemical parameters by a methods of principal component (PCA) and cluster analysis. A comparative analysis was done between data for 2002-2007 and 2012-2017, and geochemical series constructed. It has been established that the 5th class water quality limits protection and normal functioning of the river ecosystem, river water use in fish farming and irrigation.
Keywords: hydrochemical observations, urban river, water quality assessment.

Аннотация Изучено качество воды р.Раздан в черте города Ереван (Армения) за шесть лет (2012-2017гг.) по гидрохимическим показателям (физико-химические, показатели солевого состава воды, минеральные азотсодержащие вещества, нитраты и фосфаты, а также тяжелые металлы - Ag, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, Zn, Hg и As). Определены пространственная (8 стационарных пунктов) и сезонная (половодье и межень) динамика гидрохимических показателей методом кластерного анализа. Проведен сравнительный анализ с данными за 2012-2017гг. Построены геохимические ряды тяжелых металлов. Установлено, что пятый класс качества воды ограничивает защиту и нормальное функционирование экосистемы реки, использование воды для рыбоводства и орошения.
Ключевые слова: гидрохимические наблюдения, городская река, оценка качества воды.

1. Introduction

River Hrazdan is the longest left-bank tributary of River Araks in the transboundary basin of Rivers Kura-Araks (Armenia, Georgia, Azerbaijan, Iran, Turkey). Among the basin states, Armenia is the only country whose water resources form on her own territory, the rest of neighboring states being to a varying degree dependent on water delivery from Armenia's territory [9]. Major transboundary issues attracting the interest of all the basin states include complex water resource management, conservation of river ecosystems and safe water use [11].

The impact of manmade factors upon water quality is the strongest on a 36 km - long river section in Yerevan boundaries. Active development of a million-strong city has been accompanied by natural and ecological disturbances throughout the basin and intense point and diffuse pollution of water. Pollutants enter the river as a diffuse flow from polluted soils (erosion, surface runoff, drainage waters, atmospheric precipitation) and non-drainable surfaces (urban stormwater, the impact of construction activities, road transport, etc.). The major contributors to that are morphological features of the river and a mountain-basin relief of the city. Direct discharges include untreated household, fecal and industrial wastewaters of a centralized sewerage, stormwater canalization and so on [10].

The outcomes of Armenia's rivers water quality assessment reported in the literature mainly suggest collation between hydrochemical data and maximum allowable concentrations [3,7]. This particular research involved a modified approach to observation data processing and interpretation. Firstly, the given ecologo-hydrochemical investigations were done using a basin approach, that became possible due to implementation of ecological (background, regional) standards for 14 large river basins including the Hrazdan mean internal flow which were put into practice in 2011 by the decision of the Government of Armenia. The standards represent the average statistical concentrations of basic hydrochemical parameters for a decade-long period (5 quality classes and 6 purposes of water use) [6]. Secondly, water quality assessment was done through methods of statistical analysis, which help have a deeper vision of distribution of hydrochemical parameters in time and space and reveal associations and interrelationships. Thirdly, geochemical assessment of the river by heavy metal contents was provided [12]. Currently, the quality of river water is a matter of serious concern due to rapid increase in the population, urbanization, industrialization and deforestation. The available river water resources are getting depleted and being adversely affected both qualitatively and quantitatively.

These hydrochemical studies of in-Yerevan section of River Hrazdan were aimed at assessment of river water quality, analysis of spatial and temporal variations of hydrochemical parameters. The initial material consisted of the results of observations done by the Center for Ecological-Noosphere Studies (CENS) on 8 permanent stations in 2 hydrological seasons (low water and flood) for 2012-2017 and archival data for 2002-2017.

2. Materials and methods

River Hrazdan originates from high-altitude Lake Sevan. The Upper Hrazdan flows south through a mountain valley towards Yerevan, the Lower crosses the Ararat Valley, and finally empties into River Araks on the Armenian-Turkish border [12]. The Hrazdan belongs to those mountain watercourses which are fed by diverse sources: underground, rain water, melting snow. The water regime is characterized by phases of spring high water into the early summer, fall and winter low water. The watershed area 2565 km^2, the length being 146 km. Main components of water budget include river runoff 733; atmospheric precipitation 1572; evaporation 876 million m^3 year^{-1}. The river network of the Hrazdan basin consists of 541 rivers with a total length of 1262 km and network density 0.49 km^2 km-^{1} [9,10]. According to data for 1956-1967, in natural (background) conditions, water at river head is more mineralized than that in its lower reaches because of a dominant impact of Lake Sevan. The latter is characterized by increased mineralization values and a clear excess of magnesium contents vs. calcium ions. Down the river water mineralization values are decreasing under the impact of one of River Hrazdan tributaries - the Marmarik and remains approximately constant before entering the Yerevan area [3].

River Hrazdan as an economic resource and a leisure facility serves an exclusively essential role to Armenia; its basin is the most densely populated region that comprises 7 towns and more than 30 rural settlements. Even at the source the Hrazdan has a significant anthropogenic component because of deterioration of Lake Sevan water quality and natural runoff control [1]. In the limits of the Yerevan the manmade influence increases. A part of untreated urban surface rainwater and flood runoff enters the network of urban stormwater collectors having a poor sanitary and technical condition, and partially drains directly into the river via open systems (road ditches, canals, road flumes). The quality of river water also largely depends on that of irrigation water used by agricultural lands and home gardens located close to the river. Another contributor to river water pollution are partially treated wastewaters of a centralized sewerage system: Yerevan wastewaters treatment plant (WWTP) accepts municipal and industrial runoffs from Yerevan and towns of Charentsavan, Abovian and Byureghavan [13]. The WWTP provides mechanical treatment alone, reconstruction to resume biological treatment being postponed steadily. According to data for 2017, percent of decrease in BOD_5 of incoming and outcoming waters approximates to 6%, ranging 1.3 to 20% for the rest of parameters, whereas according to the requirements of the EU Directive, the reduction should be 90% for suspended substance and 75% for BOD_5 [2].

Before entering Yerevan (permanent station Hrw-1) the river is impacted by a set of manmade factors (Fig.1) and namely a cascade of hydroelectric power plants

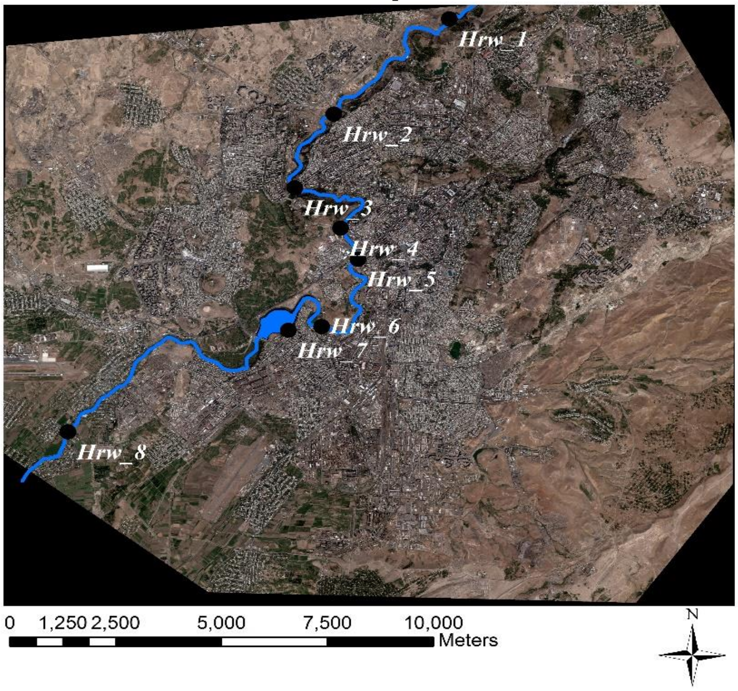

Fig.1 A scheme of River Hrazdan water sampling stations within Yerevan

and a network of irrigation canals, the Hrazdan and Marmarik water reservoirs, untreated wastewater of towns of Sevan, Hrazdan and Tsaghkadzor, point and diffuse pollution of towns of Charentsavan, Abovian, Byureghavan and over 20 rural communities. Hrw-2, Hrw-3, Hrw-4 Hrw-5, Hrw-6 located in different parts of Yerevan and experience the impact of irregular surface (stormwater and snowmelt) runoff with different intensity from urban areas and uncontrolled point discharges of industrial and household waters. Before Hw-7 the river receives more polluted waters of its tributary – River Getar after which the river runoff is regulated by the Yerevan water reservoir.

Water samples were collected in spring high and summer low water periods in 2012 through 2017. The QA/QC plan for field works was implemented through the analysis of duplicates and transport, equipment and transfer blanks. Water was sampled from central portions of River Hrazdan into new 0.5-1.5L plastic bottles. As a preservative agent concentrated nitric acid was used for heavy metals and sulphuric acid (pH ≤ 2 max.) for phosphates. The samples were transported to the lab within 1-2 hours [8]. Field measurements of physico-chemical parameters were done with Horiba and HACH SensION analysers and employing the following methods: pH - ISO-10523; electro conductivity (EC) - ISO-7888; turbidity - ISO-7027; DO - ISO-5813. Other parameters were determined in the Central Analytical Laboratory CENS (accreditation certificate ISO/IEC17025-2005) by the following methods: HCO_3^- и CO_3^{2-} - ISO-9963-1; Cl^- - ISO-9297; SO_4^{2-}- ISO-9280; Ca^{2+} и Mg^{2+} - ISO-6059; Na^+ и K^+ - ISO-9964-3; NH_4^+ - ISO-7150-1; NO_2^- - ISO-6777; NO_3^- - ISO-7890-3; PO_4^{3-} - ISO-6878; Hg - ISO-5666; As - ISO 17378-2:2014; Ag, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, Zn - ISO 15586:2003 (Perkin Elmer AAnalyst 800). The accuracy of analytical work is verified by reference solution and a repeated analysis of a water sample (n=3).

Pretreatment of analytical results included determination of water salt contents, construction of trilinear diagrams, calculation of statistical parameters. When developing a data set for statistical treatment, very high and very low values were not removed since a data set is heterogeneous by its content because of a strong manmade pressure from the watershed Based on hydrochemical profiling we applied data reduction techniques as principal component analysis to evaluate the importance of various water quality parameters and determine the most meaningful parameters responsible for the spatial and temporal variations in river water quality [5]. Spatial and temporal differences in chemical composition of River Hrazdan were determined through a cluster analysis by the Ward's method [4]. River Hrazdan basin was mapped and sampling sites plotted using ArcGIS software, diagrams of water salt content were constructed with help of GV charts, statistical data treatment – by SPSS Statistics 12. Water quality assessment was done through collation between obtained data and regional ecological standards for River Hrazdan basin. River water is classified into 5 quality classes: I-high, II- good, III- moderate, IV-poor, V-bad [6]. In order to assess ecological situation, we calculated coefficient of concentration of heavy metals against a background level – HW-1, a conventional entrance into the city - HW-1 [12].

3. *Results and discussion*

A statistical analysis of experimental data for 6 years (Tab.1) has indicated that the water temperature range is 8.7-28.3^{0}C, pH - 7.36-9.25; EC - 312-1314 μS/sm; turbidity – 0-356 NTU; DO – 7.26-19.92 mg L^{-1}; TDS – 225-706 mg L^{-1}; Cl^- - 36.9-156.0 mg L^{-1}; SO_4^{2-} – 32.5 -142.0 mg L^{-1}; hardness - 2.0-7.9 mmol L^{-1}; NH_4^+ - 0.1-12.0 mg L^{-1}; NO_2^- – 0.01-0.79 mg L^{-1}; NO_3^- - 1.1-69.3 mg L^{-1}; PO_4^{3-} 0.08-5.92 mg L^{-1}; Cr – 0.39 - 4.39 μg L^{-1}; Cu - 0.75-28.5 μg L^{-1}; Mn - 1.2-92.0 μg L^{-1}; Mo – 0.05-10.0 μg L^{-1}; Zn – 2.5-85.02 μg L^{-1}. Significant data were too scarce to provide a statistical analysis of Ag with detection limit <0.1 and maximal concentration equal to 10,7; Cd - <0.02 and 11.73; Co - <0.05 and 12.73; Ni - <0.03 and 5.12; Pb - <0.03 and 5.95 μg L^{-1} respectively. Hg and As concentrations in all the samples did not exceed 0.6 and 0.8 μg L^{-1} respectively.

Separate study parameters exhibit a wide range of values against the average, lower leveling of study values and a variation coefficient value exceeding 33% (NH_4^+ - 194.7; turbidity - 173.8; NO_2^- - 133.6; PO_4^{3-} - 104.7; Zn - 80.5; Mn - 75.8%). Extreme concentrations of hydrochemical parameters trigger dramatic deviations from normal distribution. These depend on climatic changes, soil and rock composition, hydrodynamic processes, hydrological regime, flow velocity, river sinuosity, and so on. Besides, a dominant role is given to polluting substances which enter the city from the watershed basin. Resolving the issue concerning the contribution of natural and manmade factors to water quality formation based solely on a simple collation between obtained results and standards can lead to incorrect conclusions. So, to identify the causes of water quality deterioration different methodological approaches are employed in general and in this research in particular.

Table 1.

Statistical hydrochemical parameters for 2012-2017

Parameters	Unit	N	Min	Max	Mean	STD	CV
Temp	^{0}C	64	8.70	28.30	16.64	4.73	28.41
pH	-	80	7.36	9.25	7.97	0.38	4.71
EC	μS/sm	80	312.0	1314.0	770.5	168.0	21.8
Turb	NTU	80	0.0	356.0	25.8	44.8	173.8
DO	mg L^{-1}	72	7.26	19.92	11.38	2.99	26.25
TDS	mg L^{-1}	80	225.1	706.0	491.9	108.0	22.0
HCO_3^-	mg L^{-1}	80	80.5	348.9	218.0	53.0	24.3
CO_3^-	mg L^{-1}	80	0.0	24.0	2.8	5.8	210.3
SO_4^{2-}	mg L^{-1}	80	12.50	142.00	74.62	29.62	39.70
Cl^-	mg L^{-1}	80	36.87	156.00	105.11	25.27	24.04
TH	mmol L^{-1}	80	2.00	7.90	4.73	1.16	24.42
NH_4^+	mg L^{-1}	80	0.10	12.00	0.82	1.59	194.68
NO_2^-	mg L^{-1}	80	0.01	0.79	0.11	0.15	133.61

NO_3^-	mg L^{-1}	80	1.12	69.26	17.73	14.70	82.93
PO_4^{3-}	mg L^{-1}	80	0.08	5.92	0.76	0.80	104.68
Cr	μg L^{-1}	66	0.39	4.39	1.58	0.91	57.63
Cu	μg L^{-1}	80	0.75	28.50	8.46	4.75	56.16
Mn	μg L^{-1}	77	1.20	92.00	17.21	13.05	75.84
Mo	μg L^{-1}	62	0.05	10.00	4.06	2.39	58.84
Zn	μg L^{-1}	80	2.50	85.05	19.62	15.79	80.46

In the first instance water salt content and changes in basic ions in different hydrological years were assessed. In most of the studied samples anions are dominated by hydrocarbonate- and chloride-ions, cations-by calcium- and magnesium ions. On the whole, River Hrazdan water both at flood and low water has a hydrocarbonate-calcium composition with the exception of Hw-1 (entrance to the city) in which and natural conditions alike cations in some samples are dominated by magnesium ions. Besides, the increasing share of %-equivalent magnesium in water ionic composition at the entrance is observable both during high and low water. In the city boundaries the share of chloride- and sodium-ions increases downstream in both hydrological seasons.

Factor analysis of the data set outputs a four-component model with 88.59% (low water) and 83.59% (high water) of the total variance explained (Tab.2).

Table 2.

Variables and factor loadings after Varimax rotation for high and low water season

Variable	High water		Low water	
	Factor 1	Factor 2	Factor 1	Factor 2
Temp	0.580993**	-0.402312	-0.526937**	0.730360**
pH	-0.039639	0.680949**	***0.792923****	0.081794
EC	0.662214**	0.008692	***0.917246****	0.251394
Turb	***0.884653****	0.104940	-0.080659	***0.817460****
DO	-0.575204**	0.489424*	0.665309**	-0.640022**
TDS	***0.927506****	0.276364	***0.790308****	0.547234**
HCO_3^-	0.595791**	0.322199*	***0.873718****	0.137152
CO_3^-	0.217817	-0.022166	0.712312**	-0.464251*
SO_4^{2-}	0.723668**	-0.539465**	0.121567	***0.845302****
Cl^-	0.401225*	0.626793**	0.486604*	-0.066983
Ca^{2+}	0.631997**	0.590841**	-0.180745	0.613435**
Mg^{2+}	-0.303712*	-0.544195**	0.639377**	-0.451062*
Na^+	***0.933738****	0.159944	0.570519**	0.551925**
TH	0.394550*	0.019998	***0.782272****	-0.209147
NH_4^+	***0.946633****	0.022119	0.084189	***0.883675****
NO_2^-	***0.805574****	-0.164263	0.138924	***0.920314****
NO_3^-	-0.334097*	-0.504525**	-0.435742*	-0.101169
PO_4^-	***0.954649****	-0.077292	0.471321*	0.695991**
Cr	0.129850	***-0.758195****	***-0.840082****	-0.143898
Cu	***0.787583****	-0.570452**	0.071858	0.674403**
Mn	***0.809418****	-0.022458	0.075740	0.697791**
Mo	-0.556722**	-0.651151**	***-0.955156****	0.042122
Zn	0.122528	***-0.933202****	-0.614377**	0.406771*
Eigenvalue	9.542970	4.895601	8.167527	7.114307
% of total variance	55.32	28.38	47.35	41.24
Cumulative %	55.32	83.59	47.35	88.59

- ✓ *Factor Loadings (Varimax raw) (Date-Razdan) Extraction: Principal components (Marked loadings are >,750000);*
- ✓ *values in italics indicate statistically significant scores *0.30–0.50 (weak); **0.50–0.75 (moderate); ***>0.75 (strong loading).*

The first component (F1) describes 55.32% (high water) and 47.35% (low water) of the total variance. The first component in high water describes strong loadings of PO_4^{3-}, NH_4^+, TDS, Na^+, turbidity, NO_2^-, Mn and Cu; moderate loadings – temperature, EC, DO (inverse relation), HCO_3^-, SO_4^{2-}, Ca^{2+}, Mo, weak loading – Cl^-, Mg^{2+} and NO_3^-. The first component in low water was strongly correlated of pH, Ec, TDS, HCO_3^-, TH, Cr, Mo; moderately - temperature, DO, CO_3^{2-} , Na^+, Mg^{2+}, Zn (inverse relation); weak – of Cl^-, PO_4^{3-}, NO_3^-

. Factor 2 (F2) in low water shows approximately 41.24 and high water - 28.38 % of the total variance and during high water the strong loadings includes Cr, Zn; moderate – pH, Cl^-, Ca^{2+}, SO_4^{2-}, Mg^{2+}, NO^{3-}, Cu, Mo (inverse relation); weak – DO, HCO_3^-. In low water were strongly correlated of turbidity, SO_4^{2-}, NH_4^+, NO_2^-; moderate - temperature, DO, TDS, Ca^{2+}, Na^+, PO_4^{3-}, Cu, Mn; weak - CO_3^{2-}, Mg^{2+} (inverse relation), Zn.

On the whole, in high water the strongest loads are those of PO_4^{3-}, NH_4^+ and TDS, in low water – EC and Mo. Strong loads by water salt indices characterize both natural processes of water quality formation in the result of chemical weathering of rocks and ore minerals and the entering of dissolved and suspended substances with a diffuse runoff from urban impermeable areas and point pollution sources including discharges from sewerage waters. Strong loads of PO_4^{3-}, NH_4^+ and Zn during snowmelt and rain and Mo in dry seasons are unambiguously caused by manmade factors. Phosphates, ammonium and nitrites (indices of recent fecal contamination) enter with irrigation return waters and fecal wastewater. During high water it is a diffuse runoff from River Hrazdan watershed, in low water – point pollution of undertreated wastewaters of centralized sewerage which contain household and industrial wastewaters. Summarizing the results of factor analysis one should mention among 29 studied hydrochemical parameters for characterizing river water quality 6 indices are indicatory: phosphates, ammonium, nitrites, EC (or mineralization), Zn and Mo.

Composition of Dendrogram clusters between sampling stations isolated by a set of hydrochemical parameters differs in different hydrological seasons (Fig.2).

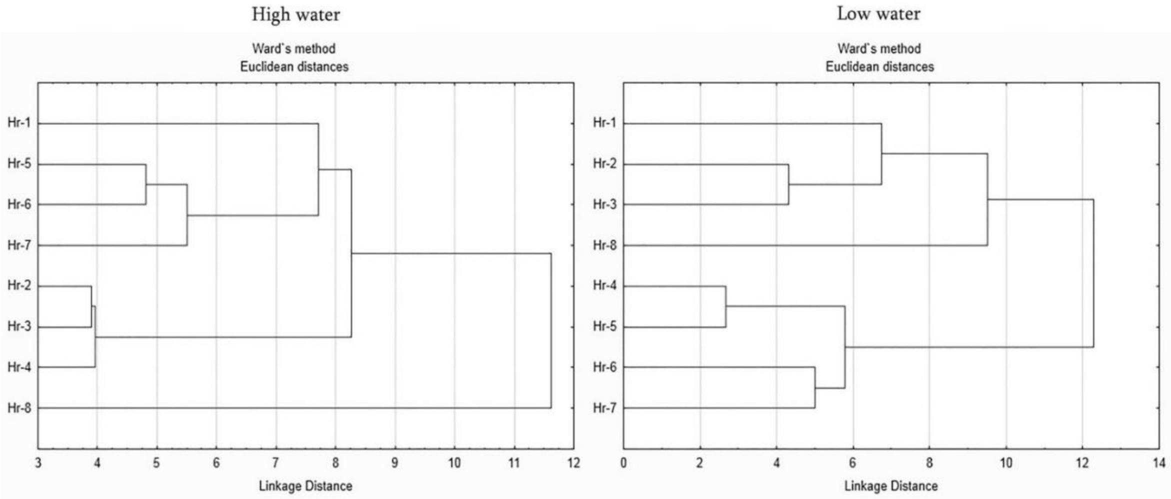

Fig.2. Dendrogram of a cluster analysis of a set of hydrochemical data on River Hrazdan by sampling stations

During high water individual groups form stations Hw-1, Hw-5, Hw-6, Hw-7 and Hw-2, Hw-3 and Hw-4; during low water the first cluster includes stations Hw-1, Hw-2, Hw-3 and Hw-8, the second - Hw-4, Hw-5, Hw-6 and Hw-7. The closest relations are established between Hw-2 - Hw-3 - Hw-4 (high water) and Hw-4 - Hw-5 (low water). Hw-8 is anomalous in high water season and is not involved in any of the isolated clusters. The results visualize heterogeneity of distribution of hydrochemical parameters per sampling stations, which is determined by mountain-basin type of Yerevan relief and urban infrastructure: the character of site development, presence of manufacturing enterprises and so on. Hw-8 – a place of entry of partially treated (mechanical treatment) wastewaters of Yerevan WWTP – differs from other sampling stations.

A comparative characteristic of 6-year observation data in 2012-2017 vs. 2002-2007 the mean value of hydrogen parameter and TDS, HCO_3^-, Cl^-, Ca^{2+} concentrations were not practically changed, CO_3^{2-}, SO_4^{2-}, Na^+, TH, NH_4^+, NO_2^-, PO_4^{3-} - were decreased; NO_3^-, Cr, Cu, Mn, Mo, Zn – increased. It is should however be mentioned that mean concentrations of heavy metals have significantly increased over the last five years: Cr and Cu - by 4.5, Zn – 3.7; Mo – 2.7 times.

With a purpose of studying those metals in detail respective geochemical series of heavy metals for 2017 in relation to the background station Hw-1 – a conventional "entrance" to the city, were constructed. According to generated data, a dominant heavy metal in a high water period is Mn, in low water – Zn. By the aggregate coefficient geochemical series are arranges are follows:

$$Zn_{(15.34)} – Cu_{(9.11)} – Cr_{(8.02)} – Mo_{(6.71)} – Mn_{(6.62)}$$

In period of snowmelt and shower rain (high water) concentrations of heavy metals on the background river station are higher than in the rest of stations except Hw-6 and Hw-8. An opposite pattern is observed in respect of a low water period this being evidence to pollutants transfer from watershed into the river just within the city. The value of aggregate coefficient of heavy metals is the highest for Hw-8, in the surroundings of which the Yerevan WWTP discharges untreated wastewaters which hold industrial waters as well. However, an insignificant difference between absolute values of aggregate coefficients points to the absence of large sources of heavy metal discharge into River Hrazdan.

In order to derive a set of acceptable values of hydrochemical parameters regional (ecological) standards for River Hrazdan, too, were used when assessing water quality. The following per-component tendency of changes in river water quality for 2017 is observable.

5th "bad" class: pH (Hw-7 high water), EC (Hw-1 high water; Hw-1 by Hw-3 low water); NH4+ (Hw-8 high water and low water); NO2- (Hw-7 and Hw-8 high water; Hw-8 low water); PO43- (Hw-1 and Hw-8 high water; Hw-1 by Hw-8 low water).

4th "poor" class: Cl- (Hw-1- high water); NO2- (Hw-6 – high water; Hw-1, Hw-7 – low water), NO3- (Hw-1, Hw-5 and Hw-6 – high water; Hw-1 by Hw-6 low water); PO43- (Hw-6 – high water), Mn (Hw-4 and Hw-8 –high water; Hw-8 – low water.

The rest of indices have the 3rd moderate, 2nd good and 1st high status. Even a single poor water quality parameter is sufficient to change for the worse the entire river basin water quality, so one may conclude that on the whole River Hrazdan water quality belongs to the 5th - bad class. The 5th class quality limits conservation of watercourses (River Hrazdan), protection of water ecosystems and salmonids farming (1st and 2nd class); normal functioning of water ecosystems and minnow farming (1st, 2nd, 3rd class); irrigation (1st, 2nd, 3rd, 4th class).

Conclusion

Spatial and temporal organization of River Hrazdan by water quality assessment data and using different methodological approaches suggests that pollution has the dominant role in formation of water quality in urban area. It is manmade factors such as a change in natural hydrological regime, runoff regulation, time diversity and irregularity of transportation of pollutants from diffuse and point pollution sources etc., which determine the anomalous character of distribution of hydrochemical parameters by river water sampling stations in different phases of hydrological regime. Representative parameters of pollution are ammonium, nitrites and phosphates (the 5th "bad" class quality), the most polluted places being stations located next to River Hrazdan confluence with its tributary River Getar and wastewaters of the Yerevan WWTP. According to long-term researches, over the last five years mean concentrations of practically all studied heavy metals have increased, dominant metals being Mn in high and Zn – in low water periods. The 5th class quality water limits its suitability for conservation of the river, the normal functioning of its ecosystem and water use in fish farming and irrigation.

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