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Main inorganic ions and electric conductivity of polluted urban streams

Главные неорганические ионы и электропроводность загрязненных водотоков

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Abstract. Concentrations of main inorganic ions were determined in waters of city streams affected by domestic and industrial wastewaters – the Okhta and the Murinsky creek in St. Petersburg. It was shown that pollution of streams led to increasing of molar fraction for sodium ion and chlorides, decreasing of molar fraction of magnesium, calcium, sulfates and hydrocarbonates. These changes took place while electric conductivity of waters was increasing. Positive correlation coefficients (from 0.35 to 0.63) were found between molar fraction of sodium and parameters connected with wastewater pollutants: total nitrogen (TN), ratio of TN to TOC (total organic carbon), ratio of protein-like fluorescence (excitation at 230 or 270 nm, emission at 350 nm) to humic-like fluorescence (excitation at 230 or 270 nm, emission at 420 nm). The obtained results show that electric conductivity can be applied as an express-method for environmental monitoring in the studied streams. Additional use of ions molar fractions for such purpose is also possible.

Аннотация. Определены концентрации главных неорганических ионов в воде реки Охты и Муринского ручья – водотоков Санкт-Петербурга, загрязненных бытовыми и промышленными сточными водами. Показано, что загрязнение приводит к увеличению мольной доли ионов натрия и хлоридов, снижению мольной доли ионов магния, кальция, сульфатов и гидрокарбонатов, при этом электропроводность воды возрастает. Положительные коэффициенты корреляции (от 0,35 до 0,63) получены между мольной долей натрия и параметрами, связанными с загрязнением сточными водами: концентрацией общего азота (TN), отношением TN к концентрации общего органического углерода, отношением интенсивности флуоресценции белкового типа (длина волны возбуждения 230 или 270 нм, регистрации 350 нм) к интенсивности флуоресценции гуминового типа (длина волны возбуждения 230 или 270 нм, регистрации 420 нм). Полученные результаты показывают, что электропроводность может применяться в качестве экспресс-метода для мониторинга окружающей среды в исследуемых водотоках. Также возможно использование мольных долей ионов в качество дополнительного параметра.

Introduction

Environmental monitoring plays essential role in environmental management and civil engineering. Results of monitoring enhance knowledge of ecosystems and social impacts on them; provide rationale for setting standards and making engineering decisions for protection and rehabilitation of natural systems [1]. Water quality monitoring has a special importance giving information about influence of wastewater effluents on drinking water sources.

Rivers of big cities usually are affected by untreated domestic and industrial wastewaters that can be discharged on a regular basis or during emergency episodes. Even if amount of direct discharges is gradually decreased due to efforts of municipality and wastewater treatment companies, there is still possibility of pipes misconnection or illicit discharge of wastewaters by individuals or organizations. Such cases took place in St.Petersburg, where in nowadays 98 % of city wastewaters are accepted by canalization and transported to wastewater treatment plants [2]. Significant pollution of rivers in St.Petersburg was sometimes noticed by citizens due to unpleasant smelt, oil sheen, and presence of dead fish. Several such accidents happened in river Okhta [3, 4]. Pollutants in storm waters from industrial and residential areas and filtrates from unauthorized landfills also make worse water quality in rivers of St.Petersburg [5–7].

Rapid detection of pollutants is important in several cases. It helps to find the sources of unwanted substances and prevent inflow of impurities from them in future due to administrative means and engineering measures. For this purpose, express-methods of water quality monitoring are needed. In natural waters with low mineralization measurement of specific electric conductivity (EC) could be a useful express-method of water quality control.

EC has been investigated as marker of contamination from wastewaters discharges [8–12]. It is considered to be useful for screening of the pollution level [9, 11]. For rivers of the North-Western part of Russia natural background concentrations of inorganic salts are small or moderate [13]. This makes possible registration of pollution in rivers by increasing of EC in water samples.

For the Okhta waters cations of calcium and anions of hydrocarbonates are contained in majority [13, 14]. According to the results of monitoring (at observation point Novoye Devyatkino at the boundary of St.Petersburg) sum of concentrations for the dissolved ions in the Okhta waters was in the range from 60 to 140 mg/L [13]. Concentration of ions (as dissolved solids) in untreated domestic wastewaters is usually higher: about 200–1000 mg/L [15, 16]. This data give basis for applying EC as a parameter for water quality monitoring in polluted urban rivers.

Correlations of EC with concentrations of total nitrogen (TN) and fluorimetric parameters that are characteristic of wastewater pollution were found in previous studies [17, 18]. The goal of the actual work was to clarify roles of major inorganic ions in formation of EC in urban river Okhta and its polluted tributary. To achieve the goal the following tasks were set:

– measurement of EC and concentrations of main ions (sodium, potassium, magnesium, calcium, chlorides, sulfates, hydrocarbonates) in waters upstream and downstream outlets of wastewaters;

– revealing connection between ions concentrations and other markers of wastewater pollution, such as TN and fluorimetric parameters of water.

Materials and Methods

Water Samples. Water samples were taken from the Okhta and its tributary the Murinsky creek. The Okhta is considered to be the most polluted river in St.Petersburg. Discharge of wastewater into river Okhta had contributed for about 20 % of its flow rate. The Murinsky creek takes domestic wastewaters from high-rise residential buildings (district Grazhdanka in St. Petersburg); inflow of wastewaters had contributed for about 60 % of its flow rate [19, 20]. Scheme of sampling points location is given in [18].

Series of water samples were collected from July 2013 to March 2014. Three series of samples for each water object were taken, 8–15 samples from the Murinsky creek and 7–17 samples from the Okhta in each series. The samples of each series were collected during the same day and transported to the laboratory. Bottles with water were stored in a fridge at +4...+6°C for several days in vertical position while suspended matter was sedimenting. The upper part of water was analyzed.

Chemical Analysis. EC was measured in the laboratory at the day of sampling by conductometer “HI 8713” (HANNA Instruments) at 20 °C (measurement error ± 5 %). Other parameters were determined in the supernatant after storage of samples. Concentrations of total organic carbon (TOC), inorganic carbon (IC) and TN were determined by analyzer “TOC L vpn-TNM” (Shimadzu, Japan) (measurement error ± 10 %). For the calculations it was supposed that all measured IC was present in the form of hydrocarbonates.

Concentrations of major inorganic cations (potassium, sodium, magnesium, calcium) and anions (chlorides, sulfates) were measured by capillary electrophoresis method on “Capel-103R” (Lumex, Russia) with measurement error ± 15 %.

Fluorescence spectra of water samples were obtained on analyzer “RF 5301 PC” (Shimadzu, Japan) at excitation wavelengths 230 and 270 nm, emission wavelength from 200 to 650 nm. The wavelengths were chosen in the previous investigations [21, 22]. Detailed process of fluorimetric analysis is described in [18]. Ratio of protein-like to humic-like fluorescence was used as a marker of pollution with organic matter from wastewaters [18].

Results and Discussion

Concentrations of ions varied one order of magnitude from time and place during the period of present study. Obvious increasing of ions concentrations and EC was observed downstream places where wastewater discharges were supposed to be or were clearly seen. For example, in waters of the Okhta 2–3 fold increasing of EC (compared to the values in the upstream samples) was observed downstream settlement Enkolovo not far from the boundary of St.Petersburg. Peak of EC (1.4–5 fold increasing) was observed in samples of the Okhta downstream inflow of the Murinsky creek; after that place conductivity decreased due to mixing and dilution with other waters of the Okhta and its other tributaries. As an example data for samples collected in September 2013 are shown at Figures 1 and 2. In Table 1 ranges of values are given for three groups of sampling points (represented as in [18], M notes for the Murinsky creek, O – for the Okhta). Samples of the first group were taken from unpolluted parts of streams that were far from direct canalization outlets. Samples of the second group were taken from possibly polluted or slightly polluted parts of streams, samples of the third group–from definitely polluted parts.

Table 1. Ranges of water parameters. Concentrations are given in mkmol/L, electric conductivity – in mkSm/cm

Samples	$C(K^+)$	$C(Na^+)$	$C(Mg^{2+})$	$C(Ca^{2+})$	$C(Cl^-)$	$C(SO_4^{2-})$	IC	EC
M9p–M10p	50–110	210–390	250–470	610–1065	220–280	310–420	430–2500	110–320
M1–M5	10–160	60–8100	120–790	500–2630	860–15600	200–590	1100–4160	235–1970
M6–M13	20–440	335–4130	150–485	715–1700	2400–7000	430–870	1900–4460	540–1160
O1–O4	15–25	120–200	50–100	90–230	60–160	10–85	220–580	40–75
O5–O9	30–90	250–820	70–205	225–670	350–910	70–290	550–1550	120–250
O10–O17	60–165	480–2520	110–340	340–1200	590–4000	195–690	340–3250	230–725

Seasonal variation of EC and ions concentrations were significant; the values at the same sampling points in the river and the creek differed by a factor of 10 or less. This could be explained by changing of water precipitation and evaporation depending on weather conditions. The data correspond with the figures from observation station Novoye Devyatkino according to which concentrations of main ions changed not more than one order of magnitude during periods of low and high water flow rate [13].

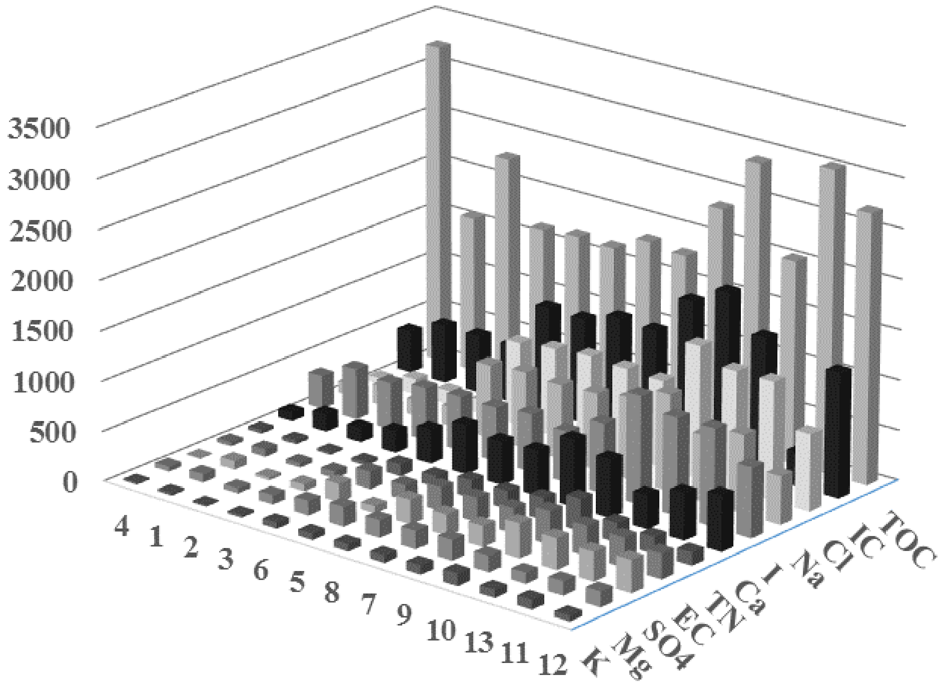

Figure 1. Parameters of water from river Okhta.
Concentrations of ions, TN, TOC and IC are given in mkmol/L, EC in mkSm/cm,
I refers for fluorimetric parameter I 230,350/420 (see explanation in the text),
which value was multiplied by 1000.

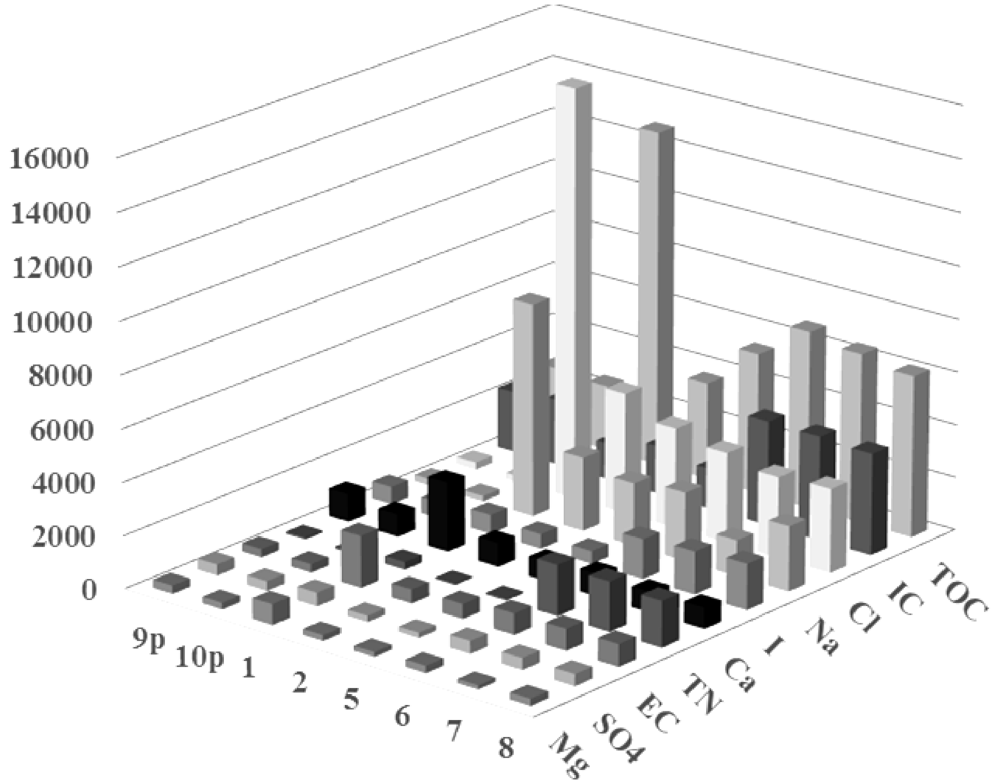

Figure 2. Parameters of water from the Murinsky creek.
Concentrations of ions, TN, TOC and IC are given in mkmol/L, EC in mkSm/cm,
I refers for fluorimetric parameter (I 230,350/420 (see explanation in the text),
which value was multiplied by 1000.

Correlation coefficients r between concentrations of ions and other water parameters related to pollutants from wastewaters were calculated (Tables 2, 3).

Table 2. Correlation coefficients for water samples of the Murinsky creek (n = 32). Concentrations are given in mkmol/L, electric conductivity – in mkSm/cm

Parameter1 / Parameter2	$C(K^+)$	$C(Na^+)$	$C(Mg^{2+})$	$C(Ca^{2+})$	$C(Cl^-)$	$C(SO_4^{2-})$	IC
EC	0.65	0.93	0.70	0.88	0.95	0.60	0.62
TN	0.46	0.20	0.01	0.03	0.09	0.36	0.65
TN/TOC	0.55	0.22	0.04	0.10	0.12	0.52	0.77

Table 3. Correlation coefficients for water samples of the Okhta (n = 30). Concentrations are given in mkmol/L, electric conductivity – in mkSm/cm

Parameter1 / Parameter2	$C(K^+)$	$C(Na^+)$	$C(Mg^{2+})$	$C(Ca^{2+})$	$C(Cl^-)$	$C(SO_4^{2-})$	IC
EC	0.94	0.95	0.86	0.91	0.96	0.91	0.91
TN	0.93	0.80	0.74	0.79	0.78	0.76	0.81
TN/TOC	0.78	0.65	0.69	0.64	0.59	0.54	0.68

The water parameters were chosen according to the following considerations. Increased amounts of organic and inorganic nitrogen compounds are usually present in domestic wastewaters [15, 20]; they are summarized in parameter TN. Presence of organic matter (as TOC) cannot be used to distinguish between natural and wastewaters in the studied streams [18] because of high background content of natural organic matter, such as humic substances [16, 18]. Instead of it, molar ratio of TN to TOC (TN/TOC) is used to distinguish between nitrogen in natural organic matter (in streams) from nitrogen added with wastewaters. It is known that TN/TOC increases in human-disturbed streams and rivers compared to the unpolluted water objects [23].

Data from tables 2 and 3 show that all the studied ions have significant role in increasing of water conductivity in the Okhta and its tributary: correlation coefficients between ions concentration and EC were from 0.60 to 0.95. The data does not allow selecting a certain ion that is more characteristic for pollution than others.

Correlation coefficients with TN or TN/TOC reveal some difference among ions: r was positive in all cases, the highest values were for potassium-ion among cations (from 0.46 to 0.93) and IC among anions (from 0.65 to 0.81). However, in the Okhta waters rather high correlation coefficients were found also for the rest of studied ions (from 0.54 to 0.80). This can be explained by difference in wastewater content, because the Murinsky creek accepts mainly domestic wastewater and the Okhta accepts wastewaters from industrial enterprises. Properties of soils, natural water sources and variation of flow rates in catchment basins also may produce effect.

In order to compare relative concentrations of ions molar fraction of cation or anion among studied cations or anions were calculated according to the formula (1):

$$R_i = (C_i \cdot 100\%) / (C_{sum}) \quad (1)$$

where C_i – molar concentration of cation or anion (mol/L), C_{sum} – sum of molar concentrations for studied cations (K^+, Na^+, Mg^{2+}, Ca^{2+}) or anions (Cl^-, SO_4^{2-}, HCO_3^-), correspondingly.

Data for molar fractions in tables 4 and 5 show difference between ions in unpolluted and polluted waters. Growth of EC is followed by increasing of molar fractions of one cation and one anion: sodium (r is 0.65 for the Murinsky creek, 0.61 for the Okhta) and chlorides (r is 0.54 for the Murinsky creek, 0.70 for the Okhta). Molar fractions of potassium has small positive or negative correlation coefficients with EC (r = 0.12 for the Murinsky creek and r = –0.06 for the Okhta). Other studied ions have negative correlation coefficients between their molar fractions and conductivity (r varies from –0.03 to –0.72). These results correspond to the information that sodium-ions, chlorides and hydrocarbonates in general prevail over other ions in domestic wastewater [24].

Correlation between molar fractions and TN was positive for monovalent cations sodium and potassium (see table 4 and 5, r from 0.16 to 0.60). Negative values of r were observed for divalent cations calcium and magnesium (r varied from –0.36 to –0.77), sulfates (r from –0.04 to –0.16). For molar fractions of other ions r had positive or negative values in different streams.

Table 4. Correlation coefficients for water samples of the Murinsky creek (n = 32)

Parameter1 / Parameter2	$R(K^+)$	$R(Na^+)$	$R(Mg^{2+})$	$R(Ca^{2+})$	$R(Cl^-)$	$R(SO_4^{2-})$	$R(HCO_3^-)$
EC	0.12	0.65	-0.66	-0.61	0.54	-0.57	-0.42
TN	0.57	0.35	-0.36	-0.38	-0.15	-0.18	0.26
TN/TOC	0.60	0.36	-0.40	-0.38	-0.14	-0.22	0.27
I 230,350/420	0.65	0.39	-0.46	-0.40	0.02	-0.31	0.12
I 270,350/420	0.51	0.35	-0.41	-0.35	-0.13	-0.26	0.29

Table 5. Correlation coefficients for water samples of the Okhta (n = 30)

Parameter1 / Parameter2	$R(K^+)$	$R(Na^+)$	$R(Mg^{2+})$	$R(Ca^{2+})$	$R(Cl^-)$	$R(SO_4^{2-})$	$R(HCO_3^-)$
EC	-0.06	0.61	-0.72	-0.39	0.70	-0.03	-0.60
TN	0.16	0.60	-0.77	-0.39	0.66	-0.04	-0.56
TN/TOC	0.075	0.63	-0.67	-0.47	0.63	-0.14	-0.50
I 230,350/420	-0.02	0.55	-0.53	-0.42	0.62	-0.12	-0.50
I 270,350/420	-0.21	0.44	-0.32	-0.35	0.46	-0.21	-0.32

The results were also compared with fluorimetric parameters of water (Tables 3 and 4). We used ratio of intensities of two fluorescence types: protein-like fluorescence (that is common for wastewaters) and humic-like fluorescence (that is common both for natural unpolluted waters and wastewaters) [25]. Such ratio increases when domestic wastewaters are discharged to river waters [18, 25]. In Tables 4–5 these ratios are denoted with "Iex, em1/em2", where "ex" – excitation wavelength (230 or 270 nm), "em1" and "em2" – emission wavelengths, "1" notes for protein-like fluorescence (at 350 nm), "2" – for humic-like fluorescence (at 420 nm). Ratios with the chosen wavelengths were most informative in revealing pollution for both studied streams as it was shown in [18, 25].

It can be seen from Tables 4 and 5 that character of correlation of ion molar fraction with fluorimetric parameters is close to correlation with TN, TN/TOC. In most cases correlation coefficients have the same sign (positive or negative). In both studied streams fluorimetric parameter increases together with growth of $R(Na^+)$ and reduction of $R(Mg^{2+})$ and $R(Ca^{2+})$.

Summary

Waters of urban river Okhta and its polluted tributary the Murinsky creek were studied. Concentrations of main inorganic ions K^+, Na^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-} and inorganic carbon were determined together with EC, TN, TOC and fluorimetric parameters of water samples.

Concentrations of all studied ions and EC increased as a result of pollution with wastewaters. The ions concentrations rose together with growth of EC. Correlation coefficients between ion concentration and EC were from 0.62 to 0.96 and it was impossible to select any ion specific for pollution. High positive values of correlation coefficients (0.54–0.70) were found between EC and molar fractions of several ions: sodium and chloride. Correlation coefficients between EC and molar fractions of bivalent cations, sulfates and hydrocarbonates were negative. These data suggest that pollution of river and creek with wastewaters together with general growth of ions concentrations increase also molar fractions for sodium and chlorides and reduce molar fraction for bivalent cations and sulfates. Such effect is a result of difference in ion relative concentration in polluted and unpolluted waters.

Positive correlation coefficients (from 0.35 to 0.63) were found between molar fraction of sodium and parameters connected with pollutants derived from organic matter degradation: TN, TN/TOC, ratio of protein-like fluorescence (excitation at 230 or 270 nm, emission at 350 nm) to humic-like fluorescence (excitation at 230 or 270 nm, emission at 420 nm).

The obtained results support the idea of wastewaters` leading role in EC changing for the studied streams. In general EC can be applied as an express-method for environmental monitoring in the Okhta and the Murinsky creek. Other parameters such as molar fractions of sodium and chlorides can also be informative in revealing pollution with wastewaters.

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