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RESEARCH ON CORRELATION BETWEEN NITROGEN COMPOUNDS, IRON AND MANGANESE CONCENTRATIONS IN DRINKING WATER SUPPLY SYSTEMS

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Abstract. A wide range of material sources potentially contribute to contaminant loads in potable water. The ability of water supply systems to act as emission control barriers to tap water micro pollutants, thereby providing environmental benefits in addition to potable water savings, have not been fully explored. This paper investigates the sources, presence and potential fate of a selection of nitrogen micro pollutants in water supply systems. All of the investigated compounds are listed under the requirements to the quality of water intended for human consumption. Significant water quality changes are identified. A wide range of potential treatment trains are available for water treatment and reuse but treatment efficiency data for nitrogen substances is very limited. Nitrogen substances removal through water treatment is following to be predominantly due to ammonium ions, nitrate and nitrite limited concentrations requirement, with only minor contributions to the water supply network. The majority of conventional water treatment plants periodically supply water with nitrogen compounds residual to the potable water distribution system. Hence, it is important to ensure that other nitrogen sources control options (e.g. pipelines materials, and groundwater sources controls) for potable water supply continue to be pursued, in order that nitrogen compounds emissions from these sources are effectively reduced and/or phased out as required under the demands of the requirements to the quality of water intended for human consumption. The aim of this research was to estimate the correlation between nitrogen compounds, iron and manganese concentrations in drinking water supply pipelines in Vilnius (Lithuania) and Warsaw (Poland).

Keywords: nitrogen substances, ammonium, nitrate, nitrite, drinking water, water pollution.

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Introduction

Drinking water is prepared only from ground water sources in all Lithuanian settlements (Jaskelevicius, Lynikiene 2009). All around the EU countries open water sources (rivers, lakes, desalinated sea water) are used (Ranieri, Swietlik 2010). Drinking water safety and quality standards of Lithuania and Poland are based on European Directive 98/83/EC on the quality of water intended for human consumption and World Health Organization Guidelines on Drinking-water Quality (2004). The main problems of water quality decrease occur during water supply and stagnation in distributing networks (Agatemor, Okolo 2007; Nawrocki *et al.* 2010), when water quality indicators and toxic parameters were increased till unacceptable values (Agatemor, Okolo 2008; Venkatesan, Swaminathan 2009). Nitrogen and its substances are associated with significant water quality decrease to water users. The most toxic nitrogen substances (Gladwin *et al.* 2004), nitrite and nitrate, generally are found in shallow wells water. It is the result of nitrogen-based fertilizers usage in agriculture. During the nitrification process nitrifying bacteria oxidize ammonium ions and ammonia into nitrite and finally into nitrate. Nitrification process is very fast, so generally detectable compound is nitrate (final stage of nitrification process). Allowed concentrations are: for nitrite – 0.50 mg/L, nitrate – 50 mg/L, most harmful of these is the nitrite.

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Ammonium is described as indicating variable and its regulated concentration is 0.50 mg/L.

It is known that the microorganisms that occurred in raw water sources also have favourable conditions to survive in pipelines. Ammonium and nitrate that naturally exist in the groundwater create favourable conditions for anaerobic ammonium oxidizing bacteria and nitrite reducing bacteria (Berry, Raskin 2006). Therefore, the total growth of microorganisms can increase as well (Markku *et al.* 2004).

Microbiological nitrification process can cause interference of chemical and microbiological changes of water quality at water supply network (Lee *et al.* 2005; Lipponen *et al.* 2002). Certain material of pipelines (Neville 2001), dissolved oxygen concentration, admixtures of water and other factors (Lehtola *et al.* 2005; McNeill, Edwards 2002) create favourable conditions for vital activities of microorganisms (Bonadonna *et al.* 2009).

This investigation presents comparison research on using two different water sources for the evaluation of nitrogen compounds diversity (Mazeikiene *et al.* 2010). Ground water from Vilnius (Lithuania) was chosen as it is natural and has no negative influence on water quality. Open sources water in Warsaw (Poland) was strongly affected by using physical and chemical water treatment methods. The research on comparison of water quality changes was carried out using water samples from both water distributing systems. The novelty of this research is related to all findings about the results showed that distribution networks in boreal region are potential environments for nitrifying bacteria, and in some conditions nitrite accumulation may exceed the acceptable concentration (Lipponen *et al.* 2002). Some additional water quality characteristics (ammoniumions, nitrates, iron and manganese concentrations) were absent from previous research.

The aim of this research was to estimate the correlation between nitrogen compounds, iron and manganese concentrations in drinking water supply systems of Vilnius (Lithuania) and Warsaw (Poland), in pipelines networks from different water sources (groundwater and open waters sources).

1. Materials and methods

In order to evaluate changes of nitrogen compounds in water supply network, Vilnius and Warsaw city's water supply systems were selected.

Vilnius is supplied with groundwater from water reservoir (at sampling point No. 1) then water is pumped to water supply system and is delivered to water users. During the research samples were taken from three sectors of Vilnius water supply network (Fig. 1).

Changes of nitrogen compounds were observed in pipelines which supply water from water reservoir located in Savanoriu Avenue. Water from the water reservoir is pumped into three different directions until it gets to sampling points. Three sectors of tested water supply network have the same distance between water reservoir and sampling points (Table 1). Explored part of Vilnius water network system composes cast iron and steel pipelines. Diameter of pipelines varies from 150 to 500 millimetres. Some previous research showed that cast iron and steel pipelines materials have very similar impact on water quality (Amosenkiene *et al.* 2009).

The water to Warsaw (Poland) users is supplied from 3 water treatment plants: Central, Praski and Polnocny (Fig. 2). They cover 50%, 20% and 30% of water users needs respectively. Almost all water is supplied from surface water sources taken by infiltration or bank uptakes from Vistula (Central and Praski) or from Zegrzynskie Lake (Polnocny).

At Central Water Treatment Plant water is collected in four water infiltration uptakes that are located along the Vistula River. Water is taken from under 6–13 m of the river bottom. It is collected as the water after infiltration processes. Then water is supplied to the water treatment plant and is prepared for drinking purposes (Table 1).

The water is collected by the same infiltration technology by three uptakes at Praski Water Treatment Plant, and then treated at the water treatment plant.

Surface water is taken from Zegrzynskie Lake constructed by a dam on the Bug-Narew River at the Polnocny Water Supply Plant. Collected water is treated using flotation process. This treatment method is based on introduction of microscopic air bubbles, which adhere to pollutants and float them to the water surface creating an easily removable blanket.

Water samples from part of Vilnius water network were taken monthly from four points, namely: at the users' taps (sampling points No. 2, 3, 4) and the water tank outlet (sampling point No. 1). Water samples from Warsaw water supply network were taken monthly from 9 points distributed between 3 water supply districts (Central, Praski and Polnocny). The analyses of samples were carried out at Drinking Water Laboratory of JSC "Vilniaus Vandenys" and the Laboratory of Ecotechnology-Water Center (Warsaw).

Water quality analyses were made and certain technological parameters were determined for the control and evaluation of water quality changes process using International Standard methods: total iron concentration, mg/l; manganese concentration, mg/l; ammonium ions concentration, mg/l; nitrate and nitrite concentrations, mg/l.

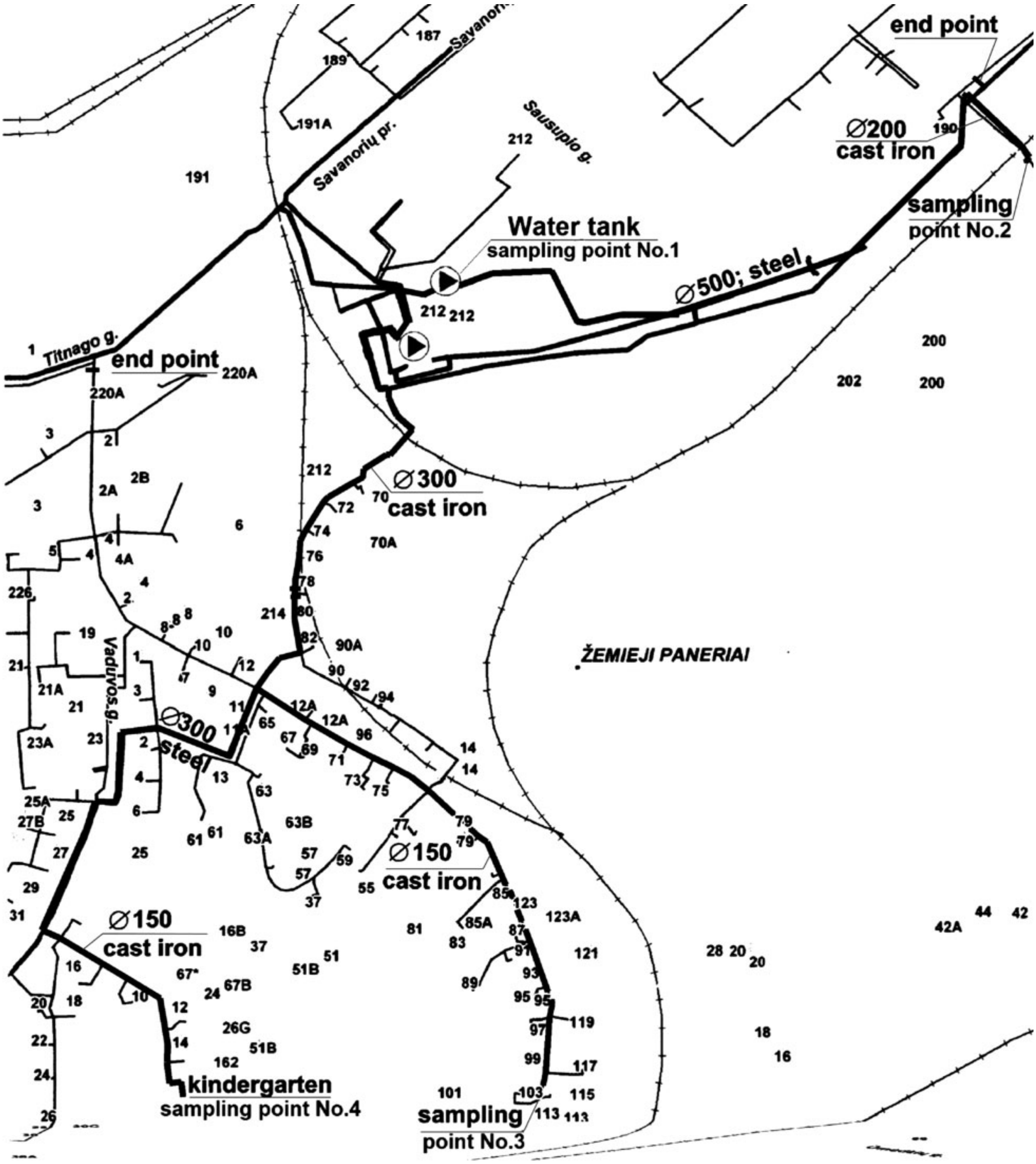

Fig. 1. Sampling scheme of Vilnius water supply network (Lithuania)

The results of the experimental investigation were statistically estimated from the registered analyses. The average concentration at the typical point was calculated on the basis of three measured values of one sample.

Collected water samples were then analyzed using International Standards (ISO) and/or European Norms (EN) approved methods for determination of water quality indicators and toxic parameters.

The quantitative results are presented as the arithmetic mean of six independent measurements ($x \pm SD$, $n = 6$). Significant differences ($p < 0.05$) were removed from the results estimations and the measurements were analyzed once again.

The data from the experimental research was statistically calculated from the registered analyses. The concentration of substances (ammonium ions, nitrate, nitrite, total iron, manganese) was measured

Table 1. Data of analysed water supply networks

	Vilnius (Lithuania)			Warsaw (Poland)		
Sampling point	No. 2	No. 3	No. 4	A	B	C
Location	Sausupio str.	Umedziu str.	Vaduvos str.	Szolc Rogozinskiego str., Nowoursynowska str., Kabacki Dukt str., Krasinskiego str.	Palestynska str.	Madalinskiego str., Kramarska str., Gierczak str., Sonaty str.
Sector of water network	Water tank-S.P. No. 2	Water tank-S.P. No. 3	Water tank-S.P. No. 4	Central Water Supply System A	Polnocny Water Supply System B	Praski Water Supply System C
Water treatment technologies	Groundwater is taken from wells field, no treatment applied, pumped to water tank and distributed to water users.			Surface Vistula water infiltration, aeration, coagulation, sedimentation and stabilization, filtration through sand and activated carbon filters, chlorination-disinfection.	Surface Vistula water infiltration, aeration, filtration through fast sand filters and chlorination-disinfection	Surface Zegrzynski reservoir water treatment based on flotation.

S.P. – Sampling point

11 times in raw water, in taps of users and in the water storage reservoirs. The average concentration at typical points was estimated following the formula, where concentration of substances at typical points, probability at the occurrence of concentration, number of days, number of different values of the concentration were evaluated.

The average concentrations of substances, mentioned above, at the characteristic point, were also calculated. The standard statistical estimation error of the arithmetic mean was approximately 11%.

2. Results and discussion

The values of the quality of water samples that were taken at the characteristic points of the pipeline of Vilnius and Warsaw water supply networks are presented in Table 2.

Nitrogen was defined to be measured in different forms, because of denitrification occurrence. Total iron concentration was chosen to be measured because it makes influence on denitrification process as nitrate reducer (Gerke *et al.* 2008). Manganese concentrations in water samples were measured as they are micronutrients needed for effective metabolism of denitrifying bacteria.

Concentrations of measured chemical compounds were analyzed in order to find dependencies between nitrogen substances and other chemical elements such as total iron and manganese (Cerrato *et al.* 2006). Also, there was reliable dependency found between total iron and ammonium ions concentrations.

The results of the present research showed the dependency between nitrogen compounds and total iron concentrations at the different sampling points of Vilnius water supply network (Lithuania). There was no dependency defined between nitrogen compounds concentrations and total iron or manganese in Warsaw water network (Poland).

Some graphics with the results from Vilnius water network are presented in Figures 3, 4, 5 and 6.

Dependency between ammonium ions and total iron concentrations was established as third order polynomial regression type ($R^2 = 0.77$):

$$C_{Fe_{total}} = -24.08 \cdot C^3_{NH_4} + 33.303 \cdot C^2_{NH_4} - 6.987 C_{NH_4} + 0.913. \quad (1)$$

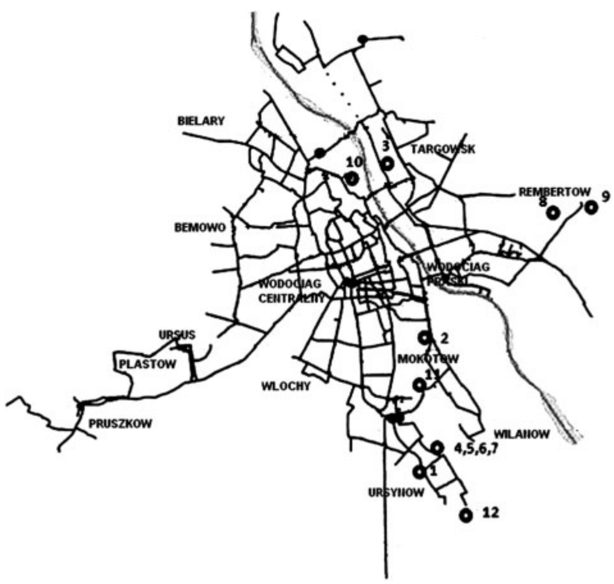

Fig. 2. Sampling scheme of Warsaw water supply network (Poland)

Table 2. The results of water quality at the characteristic point in the pipeline of water supply systems

Vilnius (Lithuania)						Warsaw (Poland)					
	Concentration of chemical compounds						Concentration of chemical compounds				
Sampling Point	NH_4, mg/l	NO_3, mg/l	NO_2, mg/l	Fe, mg/l	Mn, mg/l	Sampling point	NH_4, mg/l	NO_3, mg/l	NO_2, mg/l	Fe, mg/l	Mn, mg/l
No.1	0.18	0.30	0.03	0.69	0.17	A	0.07	10.60	0.002	<0.02	0.036
No.2	0.17	0.10	0.02	0.24	0.18	B	<0.01	11.12	<0.005	0.02	0.017
No.3	0.20	1.50	0.03	0.67	0.30	C	0.02	11.00	<0.005	0.01	0.030
No.1	0.22	1.50	0.09	0.69	0.07	A	0.11	11.20	0.004	<0.02	0.039
No.2	0.23	0.80	0.06	0.38	0.14	B	0.01	9.45	<0.005	<0.01	0.020
No.4	0.12	0.30	0.07	0.50	0.15	C	<0.01	12.00	<0.005	<0.01	0.030
No.3	0.11	0.30	0.08	0.52	0.24	A	0.06	13.70	0.002	<0.02	0.025
No.1	0.18	0.40	0.04	0.94	0.37	B	0.01	11.81	0.007	<0.01	0.025
No.2	0.10	0.50	0.03	0.34	0.19	C	<0.01	13.80	<0.005	0.07	0.030
No.4	0.04	0.70	0.03	0.75	0.49	A	<0.01	9.45	<0.002	<0.02	0.024
No.3	0.06	0.50	0.03	0.52	0.29	B	<0.01	9.68	<0.005	<0.01	0.025
No.1	0.22	0.60	0.03	0.71	0.21	C	0.01	9.30	<0.005	<0.01	0.030
No.2	0.18	0.80	0.02	0.42	0.16	A	<0.01	7.44	0.002	<0.02	0.020
No.4	0.14	0.50	0.03	0.49	0.26	B	0.01	6.79	<0.005	<0.01	0.010
No.4	0.10	1.00	0.04	0.82	0.29	C	0.03	8.70	0.005	0.02	0.030
No.3	0.11	0.50	0.03	0.68	0.29	A	0.01	6.27	<0.002	<0.02	0.029
No.1	0.31	0.30	0.03	1.23	0.39	B	<0.01	3.73	<0.005	0.02	0.014
No.2	0.08	0.20	0.03	0.41	0.23	C	0.02	6.40	<0.005	0.02	0.030
No.4	0.11	0.40	0.03	0.56	0.36	A	0.01	6.32	0.002	<0.02	0.026
No.3	0.18	0.70	0.04	2.75	0.60	B	<0.01	2.60	<0.005	<0.01	0.011
No.1	0.25	1.00	0.03	0.97	0.43	C	0.03	6.90	<0.005	<0.01	0.030
No.2	0.09	1.10	0.03	0.50	0.25	A	<0.01	5.94	0.002	<0.02	0.035
No.4	0.21	0.70	0.04	2.73	0.65	B	<0.01	2.71	<0.005	0.03	0.008
No.4	0.37*	1.10*	0.47*	4.29*	13.56*	C	0.02	6.10	<0.005	0.01	0.030
No.3	0.10	1.30	0.03	0.73	0.31	A	0.01	6.02	<0.002	<0.02	0.044
No.1	0.30	0.80	0.03	1.14	0.44	B	<0.01	7.04	<0.005	0.02	0.007
No.2	0.11	0.80	0.03	0.38	0.26	C	0.02	6.80	<0.005	0.02	0.020
No.4	0.14	1.10	0.04	1.80	0.53	A	<0.01	6.72	0.002	<0.02	0.025
No.4	0.08	1.00	0.03	2.01	0.45	B	<0.06	5.20	<0.041	0.03	0.007
No.3	0.10	1.30	0.03	0.72	0.31	C	<0.01	7.60	<0.005	<0.01	0.030

*- samples after water network brake down

Dependency between nitrate and total iron concentrations was established as third order polynomial regression type ($R^2 = 0.76$):

$$C_{Fe_{total}} = -0.135 \cdot C_{NO_3}^3 - 0.292 \cdot C_{NO_3}^2 + 1.045 \cdot C_{NO_3} + 0.208. \quad (2)$$

There is some evidence that ground water containing Fe^{2+} normally contains little or no nitrates (Rivett *et al.* 2008). Srinivasan *et al.* (2008) demonstrated that Fe^{2+} acts to promote denitrification process, in which Fe^{2+} reduces nitrate to nitrite, and then it is regenerated by the oxidation of organic carbon. Nitrite can then be reduced to gaseous nitrogen compounds in absence of organic compounds by the further oxidation of iron (Rivett *et al.* 2008). Mn^{2+} is also potential electron donor for autotrophic denitrification processes.

Dependency between manganese and nitrite concentrations was established as third order polynomial regression type ($R^2 = 0.86$):

$$C_{NO_2} = 0.888 \cdot C_{Mn}^3 - 1.06 \cdot C_{Mn}^2 + 0.412 \cdot C_{Mn} - 0.021. \quad (3)$$

Denitrifying bacteria obtain energy for metabolism and growth from the oxidation of reduced iron and manganese (Sarin *et al.* 2004). Their metabolic requirements for nitrogen can be met by available NH_4^+ or organic nitrogen in the environment, or from the direct assimilation of nitrate (Rivett *et al.* 2008). Manganese is used as micronutrient for effective metabolism.

The causality of obtained dependencies is in the case Fe^{3+} is precipitated as ferric oxide or oxyhydroxide (Rivett *et al.* 2008). This precipitation reaction releases H^+ ions into solution, balancing some of their

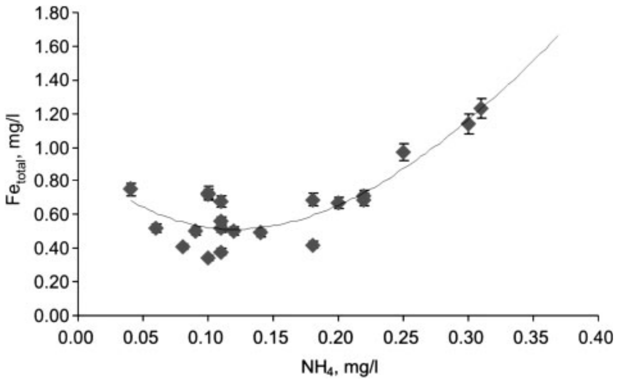

Fig. 3. Dependency between ammonium ions and total iron concentrations in Vilnius network sampling points

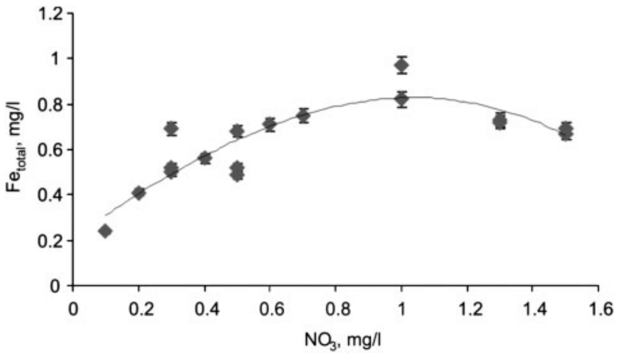

Fig. 4. Dependency between nitrate and total iron concentrations in Vilnius network sampling points

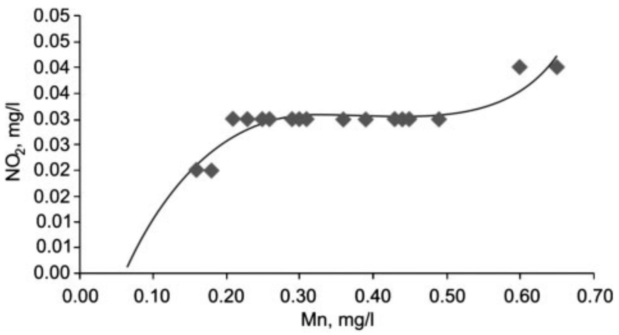

Fig. 5. Dependency between manganese and nitrite concentrations in Vilnius network sampling points

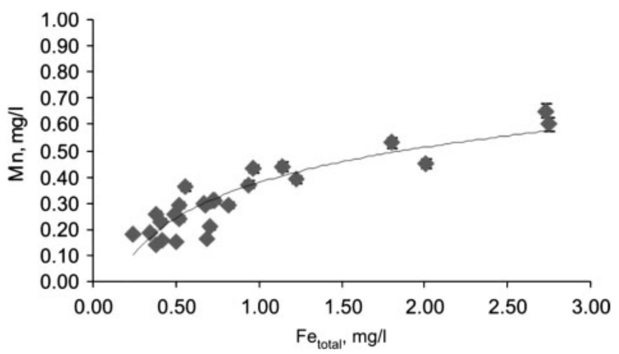

Fig. 6. Dependency between total iron and manganese concentrations in Vilnius network sampling points

Table 3. Dependency between nitrogen compounds and metals in Warsaw water network (Poland)

R^2	NH_4	NO_3	NO_2	Fe_{total}	Mn
NH_4	–	–	–	0.25	–
NO_3	–	–	–	0.52	–
NO_2	–	–	–	–	0.21
Fe_{total}	0.25	0.52	–	–	0.78
Mn	–	–	0.21	0.78	–

consumption by the denitrification reactions and Mn^{2+} is also potential electron donor for authotrophic or abiotic denitrification reactions (Rivett *et al.* 2008). The dissimilatory nitrate reduction to ammonium reaction occurs under much the same conditions as denitrification (Rivett *et al.* 2008).

Dependency between total iron and manganese concentrations was established as second order polynomial regression type ($R^2 = 0.84$):

$$C_{Mn} = -0.042 \cdot C_{Fe_{total}}^2 + 0.304 \cdot C_{Fe_{total}} + 0.095. \quad (4)$$

As it was mentioned, no dependency between different compounds was defined in water network of Warsaw (Poland). Estimated dependencies are below significant level and unreliable (Table 3). The causality of obtained results is in inauspicious circumstances e.g. low concentrations of nitrogen compounds, iron and manganese.

The obtained results also showed that the potential nitrogen substances increasing induced by the distribution of groundwater could be reduced if: nitrogen compounds concentrations were extremely low (Warsaw water supply) at the outlet of the water treatment plant; water treatment implementation should reduce the levels of the nitrates and nitrites of the treated supplied water. These differences from Lithuanian case can be explained as follows. Drinking water in Warsaw is prepared from surface water sources. Water treatment technologies are strongly influenced by different chemical compounds that can completely change initial water composition (Langmark *et al.* 2005). Lithuanian ground water is used for drinking purposes and do not require complicated water treatment technologies. In this particular case the explored part of water supply system in Vilnius supplies ground water without treatment.

Different water quality changes were obtained using research results from groundwater and surface water sources of Vilnius and Warsaw respectively.

Conclusions

1. Results showed that groundwater (Vilnius water supply) was extremely susceptible to different nitrogen compounds concentration changes in the

pipelines, the mostly significant range of nitrate from 0.1 to 1.5 mg/L, in case ammonium concentrations from 0.09 to 0.25 mg/L. The occurrence of ammonium and nitrates in drinking water samples correlated positively ($R^2 = 0.76$ and $R^2 = 0.76$ respectively) with total iron concentration and negatively with the content of different nitrogen compounds (ammonium ions, nitrite, nitrate).

2. Significant increase data were obtained for manganese compounds in drinking water supplied to Vilnius, Lithuania. Nitrite concentration was slightly increasing during the research with positive correlation ($R^2 = 0.86$) on high manganese concentrations because of occurring micronutrient decomposition processes.

3. Comparing ammonium ions concentrations from two water sources (groundwater from Vilnius and surface water from Warsaw), it was noticed that ammonium ions concentration from Vilnius water supply was decreased to the maximum of 64%; the most significant maximum decrease of ammonium compounds from Warsaw water supply was nearly 91%.

4. Obtained results showed that different concentrations of nitrate up to ten times higher in Warsaw water supply comparing with Vilnius are found in the water supply of different origin (open water sources and groundwater respectively).

5. The obtained characteristics of iron and ammonium ions dependency can be useful for the estimation, project design, and the equipment start up procedure, renovation and maintenance of existing water supply systems.

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