Urban Streamwater Contamination and Self-purification in a Central-Eastern European City. Part I

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Similar to many other former communist European cities, Suceava city experiences environmental problems. In order to document some of the water-related problems, we conducted water sampling and analyses in 2011 and 2012 on the stream network of Suceava city area. The measured parameters included metal concentrations (total iron, total manganese, total chromium, Al\textsuperscript{3+}, total zinc, total copper), nutrient loads (PO\textsubscript{4}\textsuperscript{3-}, NO\textsubscript{2}- - N, NO\textsubscript{3}- - N) and other contaminants (S\textsuperscript{2-}, SO\textsubscript{4}\textsuperscript{2-}, Cl, Br, I) in order to assess water quality, contamination sources and control factors. An upstream-downstream catchment comparative analysis was performed for Suceava River and its tributaries in order to determine the spatial evolution of contaminants. The contamination level is generally lower for Suceava River than for its tributaries. We observed self-purification phenomena for some parameters (nutrients, heavy metals) that could be explained by the formation of iron oxy-hydroxides and the co-treatment of diversely contaminated waters.

Keywords: pollution, geochemistry, self-purification, co-treatment

Suceava city is a Romanian settlement with several environmental problems, some of which are linked to streamwater quality. As yet, there is no study dedicated to analyzing the spatial evolution of water quality of Suceava River within Suceava urban area and its urban tributaries. A limited number of chemical analyses were performed on water from Suceava River, sampled at Mihoveni and Tîrăuşi (Suceava city area) as part of environmental studies and published in the form of national environmental reports, whereas the published scientific papers on this topic are rather scant [1]. While few papers analyze the general water chemistry in this part of Romania [2-9], the international scientific literature comprises of numerous case studies concerning the urban water contaminants (such as Sörme and Lagerkvist [10]).

Within the Romanian urban network, Suceava city is considered a medium-large city with a population of approx. 114,000 inhabitants (2011), including those with temporary residence. During the communist era, this urban area was highly polluted due to its heavy industrial activity. Post-1989, the shift towards the capitalist economy in Romania gradually diminished the industrial activity of the city and resulted to an increase in the share of the tertiary sector within the local economy. In this regard, the economic evolution of the city is representative for an entire range of cities of similar size located in the central-eastern part of Europe which had previously been part of the communist economic systems and were endowed with highly polluting industries and are currently facing economic and environmental adaptation problems. Overall, environmental problems caused by urban polluted waters are common in this part of Europe, as shown, for example, by Vystavna et al. [11].

The objective of the present study is to show the measured urban water contaminants obtained in field analysis campaigns. During the period when the field analyses were performed, the urban wastewater treatment plant was officially unable to remove P and N from wastewater under the maximum admissible concentrations. Prior to the initiation of works for the modernization and extension of the urban water supply network and sewage system (in 2011), only 83% of the city population had access to drinking water from the urban water system, whereas just 81% of the population used the city sewerage network. Thus, at least 19% of the urban population contaminated the groundwater by using leaking septic tanks and cesspits.

Experimental part

The analyzed parameters in the field campaigns are pH, dissolved oxygen, electrical conductivity, total copper, total iron, total manganese, total chromium, Al\textsuperscript{3+}, SO\textsubscript{4}\textsuperscript{2-}, Cl, Br, I, orthophosphate, nitrite-N (NO\textsubscript{2}- - N) and nitrate-N (NO\textsubscript{3}- - N), measured in 2 different seasons, wet and dry. The analyses were conducted on a daily basis. We selected 12 sampling points such that streams could be analyzed in upstream-downstream pairs. Within this network, Suceava River is the main river and collects the tributaries under analysis from both sides (fig. 1).

The samples were collected using polyethylene bottles and were analyzed within 6 h from sampling. The three weeks of sampling were conducted in 2011 and 2012: 16-22 March 2011, 31 October-6 November 2011 and 14-20 March 2012. The sampling points analyzed during the first

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The second week of sampling, as these were the most contaminated ones according to the results yielded by the previous week of analysis. In the first 2 weeks, the following parameters were analyzed: total copper, total iron, total manganese, total chromium, S\textsubscript{2}-, SO\textsubscript{4}\textsuperscript{2-}, Br\textsuperscript{-}, I\textsuperscript{-} and Cl\textsuperscript{-}. The pH, electrical conductivity and dissolved oxygen were measured daily during the second week, but in the first week only pH was measured, on 21 March 2011. The third week of analyses occurred in late spring, during the wet season, with new types of analysis, for measuring the nutrient load (PO\textsubscript{4}\textsuperscript{3-}, NO\textsubscript{2}--N and NO\textsubscript{3}--N) and the detection of other metals (Zn\textsuperscript{2+} and Al\textsuperscript{3+}). During the latter week, the old types of analyses were all performed only on SRD, while total copper, SO\textsubscript{4}\textsuperscript{2-} and S\textsubscript{2}- were repeated for all sampling points. pH and electrical conductivity were also measured during the third week.

The samples were collected at the depth of 0.1 m and at 0.1 m distance from the river bank. There is a very good mixing of waters on Suceava River tributaries because they are shallow (0.1-0.3 m average depth) and narrow (a maximum width of 2 m). In turn, Suceava River is considerably wider, but its average depth is only 0.5 m and is characterized by good mixing of water due to the numerous meanders (natural and anthropogenic) and bridge hydraulic jumps. Downstream of the Wastewater Treatment Plant (WTP) the sampling point was located just 1 km downstream due to the good water mixing (river water + wastewater). In the first 2 weeks, the sampling was performed simultaneously in all sampling points at 5 p.m., whereas in the third week the sampling time was at 2 p.m.

The contaminant analysis was carried out using the Hach DR2800 spectrophotometer with reagents according to the Standard Methods for the Examination of Water and Wastewater (20th ed.). Our chemical measurements used unfiltered samples (the sum of particulate and dissolved parameter concentrations). The reagents had the following detection ranges (mg/L): 0.01-3 Zn\textsuperscript{2+}, 0.008-0.8 Al\textsuperscript{3+}, 0.04-5 total copper, 0.1-20 total manganese, 0.02-3 total iron, 0.01-0.7 total chromium, 0.02-2 Cl\textsuperscript{-}, 0.05-4.5 Br\textsuperscript{-}, 0.07-7 I\textsuperscript{-}, 0.02-2.5 PO\textsubscript{4}\textsuperscript{3-}, 0.1-10 NO\textsubscript{2}--N, N, 0.002-0.3 NO\textsubscript{3}--N, N, 2-70 SO\textsubscript{4}\textsuperscript{2-} and 0.005-0.8 S\textsubscript{2}-. The dilution method was used when necessary. The precision of our measurements was (mg/L): 0.013 Zn\textsuperscript{2+}, 0.008 Al\textsuperscript{3+}, 0.04 total copper, 0.1 total manganese, 0.023 total iron, 0.005 total chromium, 0.02 Cl\textsuperscript{-}, 0.05 Br\textsuperscript{-}, 0.07 I\textsuperscript{-}, 0.02 PO\textsubscript{4}\textsuperscript{3-}, 0.1 NO\textsubscript{2}--N, N, 0.002 NO\textsubscript{3}--N, N, 0.04 SO\textsubscript{4}\textsuperscript{2-} and 0.005 S\textsubscript{2}-. The electrical conductivity, pH and dissolved oxygen were measured at 22°C using the Hach Lange multiparameter HQ40D. The average streamwater temperature in the sampling weeks varied between 2-4°C in spring and autumn 2011 and was 11°C in spring 2012.

The Romanian Waters National Administration provided BOD\textsubscript{5} data for Suceava River, obtained in a laboratory meeting the SR EN I.S.O./CEI 17025:2005 requirements.

Results and discussions

The chemical analyses show a lower average contamination in the dry season (i.e. measurements made in November 2011) as compared to the wet season (measurements made in March and May) (figs. 2-4). The generally lower contaminant concentrations in the water of Suceava River compared to the total average values also indicate that the former receives more contaminated waters from its urban tributaries. It is the case of PO\textsubscript{4}\textsuperscript{3-} (0.23 mg/L – Suceava, 0.58 mg/L – total average values), NO\textsubscript{2}--N (0.026 mg/L – Suceava and 0.083 mg/L – total average values), NO\textsubscript{3}--N (0.4 mg/L – Suceava and 0.73 mg/L – total average values) and Zn\textsuperscript{2+} (0.17 mg/L – Suceava and 0.176 mg/L – total average values). The contaminants which generally have higher concentrations on Suceava River compared to its tributaries are Al\textsuperscript{3+} (0.073 mg/L – Suceava, 0.048 mg/L – total average), S\textsubscript{2} (43 µg/L – Suceava, 24 µg/L – total average value in March; 20 µg/L – Suceava and 19.31 µg/L – total average value in November; 25.8 µg/L – Suceava and 17.2 µg/L – total average value in May) and total iron (0.42 mg/L – Suceava, 0.27 mg/L – total average value in March; 0.17 mg/L – Suceava and 0.14 mg/L – total average value in November).
Only in March, Cl-, Br- and I- concentrations are higher on the Suceava River (0.04, 0.13 and 0.2 mg/L, respectively) than on its tributaries (0.03, 0.1 and 0.16 mg/L, respectively).

The total streams average value for Dissolved Oxygen (DO) is 8.33 mg/L. The average Electrical Conductivity (EC) was highest in November (731.05 µS/cm) and lowest in May (552.1 µS/cm). The greater EC of SRD, downstream of the WTP of Suceava city, compared with SRU, indicates an increased concentration of contaminants. This phenomenon is also due to the analyzed Suceava River tributaries which have EC values much higher than those of SRD. The streams located within the study area have a slightly alkaline average, as the majority of them have pH
values above 7: March - 7.1, November - 7.87, May - 7.97. 
\( pH \) is lowest at CCD and SRD because CCD receives 
untreated urban wastewaters, while SRD receives the 
waters of CCD and WTP.

The average \( pH \) values on Suceava River were 7 (March), 
7.8 (November) and 7.96 (May). The electrical conductivity 
was 675 \( \mu S/cm \) in November, and 406.76 \( \mu S/cm \) in May. 
Dissolved oxygen was 8.7 mg/L. These values, compared 
with the total averages per study area, suggest that the 
tributaries are more contaminated.

Catchment analyses were undertaken in order to 
determine the spatial evolution of contaminants. A 
comparative analysis of values recorded in upstream and 
downstream sampling points in a given catchment (figs. 
5-8) was performed. In order to have comparable average 
values for the sampling points undergoing comparison, we 
used only wet season measurements. Some points were 
not measured in both months of the wet season, i.e. March 
and May, therefore in these cases it is not possible to use a 
March and May average. Consequently, the existing values

Fig. 4. Temporal evolution of contaminants in May 2012

Fig. 5. Upstream-downstream contaminants evolution in Suceava River (A-SRU, B-SRB, C-SRD)
were used for the comparison with the March-May averages of the other sampling points.

One can observe that, generally, there is a downstream increase in the concentration of contaminants. There are some parameters that oscillate or even record a concentration reduction downstream. For the majority of our sampling points it's hard to estimate if the oscillations or reductions are transient events or persisting self-purification effects due to various mechanisms.

The using of three sampling points (instead of 2) on Suceava River allowed for the observation that, the nutrient contamination (PO₄³⁻, NO₃⁻ - N, NO₂⁻ - N) decreases as the river crosses the urban area and increases subsequently until point C (SRD), albeit it does not reach the values previous to entering the city. Apparently, this is a paradoxical behaviour, especially because we already known that Suceava River collects tributaries with higher
contamination within the built-up area, as well as, during the measuring weeks, partially treated waters from the urban WTP. The latter source (WTP) was known to be unable to remove the high amount of phosphorus from the wastewater in the years of water sampling.

S²⁻ contamination on Suceava River is the highest among the analyzed streams and is caused by the massive input from WTP. At the WTP outlet and in the lower Cetății Creek the rotten egg odour is persistent and indicative of decaying organic matter. It is noteworthy that at SRD there is a negative correlation between NO₂⁻ - N, NO₃⁻ - N and PO₄³⁻ with total iron (there is probably a nutrient adsorption by iron oxy-hydroxides, which is frequent in iron-rich rivers [12]), while total iron has a positive correlation with S²⁻ (table 1).

![Fig. 8. Upstream-downstream contaminants evolution in Dragomirna River (A-DRU, B-DRD)](http://www.revistadechimie.ro)

It is possible that the sulfides from WTP and Cetății Creek, locally abundant at SRD, form iron sulfides when iron is abundant. Suceava River has water rich in iron [13]. The iron sulfides precipitate and remove iron and sulfide from water into riverbed sediments. Total iron self-purification between A and B is 21.6%, whereas between B and C the self-purification is 34.8% (calculus based on measured concentrations).

The measurements of PO₄³⁻, NO₂⁻ - N and NO₃⁻ - N on Suceava River show a 43.4, 50.9 and, respectively, 31.4% self-purification between A and B, whereas between A and C the self-purification values are 27.4, 5.3 and 28.6%, respectively. These values could be an indicator of strong constant or transient self-purification effect between A and B; the self-purification mechanisms persist between B and

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C, but the heavy contaminated waters received from the Cetății Creek and WTP raise the concentrations of contaminants from B.

The values of total chromium, Al$^{3+}$ and total zinc increase in Suceava River from A to C, while the values of total copper and total iron have an inverse behaviour. Though all Suceava River tributaries have average values of total copper equal or 2-fold higher than the correspondent values of Suceava River, total copper concentrations strongly decrease after the river collects the WTP output, in point C. In the streamwater, copper and zinc are known to be often adsorbed onto iron oxy-hydroxides [14]. The copper and zinc can be organically bound to dissolved organic carbon, with copper having greater affinity than zinc for association to natural and synthetic organic ligands [15]. BOD5 and permanganate index during 2006-2011 were higher in T than in M (BOD5 average values: 2.14 mg at M, 3.49 mg at T; permanganate index average values: 4.1 mg at M, 5.6 mg at T). Even if there are numerous hydraulic jumps on Suceava River (fig. 1), dissolved oxygen is lower in T (9.8 mg/L) than in M (10.8 mg/L), demonstrating the great oxygen demand for chemical and biological processes. The high input of organic matter from wastewaters creates a high input of organic carbon that will cause total copper self-purification; total zinc self-purification was not observed on Suceava River. In Suceava River, total copper self-purification between A and B is 2.4%, whereas between B and C the self-purification is 37.3%. The sudden increase in total copper self-purification can be explained by the quasi-punctual downstream input of organic matter by Cetății Creek and WTP.

The effect of heavy metals on self-purification capacity of Suceava River was verified using the measured inhibitory effect of known concentrations on BOD [16-21]. Suceava River heavy metals concentrations were not high enough to affect self-purification and on very rare cases a 20% BOD reduction was possible due to the concentrations of some metals according to only some references.

Conclusions

The lower concentration of contaminants in Suceava River than in its tributaries indicates that Suceava city is an active source of water pollution. The same comparison also indicates that the urban pollution is greater than the agricultural one. Nutrient and heavy metals removal was observed, which could be explained by the formation of iron oxy-hydroxides due to the iron-rich waters and by the co-treatment generated by the untreated and partly treated wastewater received by Cetății Creek and Suceava River. The output of various urban activities has opposite action on contaminant concentration. The streamwater self-purification found in Suceava River is useful as it indicates that the urban impact on natural water in the Central-Eastern Europe is not as worse as previously thought.

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