Spatio-temporal Analysis of the Water Quality of the Ozana River

GHEORGHE ROMANESCU[1]*, MARINA IOSUB[1], ION SANDU[2], IONUT MINEA[1], ANDREI ENEA[1], DAN DASCALITA[3,4], OANA-ELENA HAPCIUC[1]

[1]Alexandru Ioan Cuza University of Iasi, Faculty of Geography and Geology, Department of Geography, 20A Carol I Blvd., 700505, Iasi, Romania

[2]Alexandru Ioan Cuza University of Iasi, ARHEOINVEST – Interdisciplinary Platform, Laboratory of Scientific Investigation & Conservation, 22 Carol I Blvd., G Building, 700506, Iasi, Romania

[3] Siret Water Basin Administration, Bacau, 1 Cuza Voda Str., 600274, Bacau, Romania

[4] Vasile Alecsandri University of Bacau, 158 Marasesti Str., 600115, Bacau, Romania

The assessment of the chemistry and water quality of the Ozana river is necessary from two perspectives: firstly, the fact that the groundwater of its lower basin is tapped and supplied to four cities from eastern Romania, namely Roman, Pascani, Targu Frumos and Iasi, which sum up a total of 450000 inhabitants; secondly, the lower basin is classified as vulnerable to nitrite pollution (according to the Siret river basin management plan, established based on the regulations stipulated in the 60/2000/CE Framework Directive). The water quality index (calculated for each parameter, according to the multi-annual mean and its respective weight) has clearly emphasized the impact of the use of nitrites and phosphorus (90%). The water quality, according to the index, is very good in Boboie°ti (-16), and good to poor in Dumbrava (48).

Keywords: physicochemical parameters, quality classes, Ozana river, pollution, water quality index

The analysis of river waters represents a major concern for the competent institutions, considering both their use in the urban supply network, as well as the risk of potential ecosystem damage from poor quality waters [1]. The Ozana river, through its catchment, becomes an important subject in the study of water chemistry and quality. Chemical parameters are subject to variations along the course of a river, being influenced by natural factors (lithology, slope, climate) or anthropic agents (input from wastewater treatment, industrial discharge, the use of fertilizers, the lack of septic tanks meant to prevent the seepage of organic matter etc.) [2, 3].

The purpose of the present study lies in the descriptive statistical analysis of 14 physicochemical parameters of the water samples collected each month/semester from two monitoring sections (Boboie°ti and Dumbrava) on the Ozana river. The monitoring period was 11 years (2004-2014). Within the study, the spatio-temporal variations of the physicochemical parameters were identified, along with the similarities and differences at the two stations and the evolution of water quality. The areas and companies regarded as pollution sources were monitored. For the multi-annual qualitative evaluation, a weighted arithmetic water quality index was used [4]. The main potential pollution sources on the Ozana river are represented by the petroleum-derived products of S.C. Mihoc S.R.L., located in the Pipirig commune, and by the Tg. Neamþ urban agglomeration [5, 6]. The secondary sources are represented by the smaller, riverine urban settlements, wood-processing centers or factories, and tourist accommodation facilities within the Ozana river basin.

The influence of the physicochemical parameters over water quality was determined using the weighted arithmetic water quality index [4, 7-13]. This index is suitable for the analysis of the water of the Ozana river due to, on the one hand, the ability to incorporate a variable number of chemical and biological parameters into the equation, and, on the other, the fact that equations with a low number of indicators yield results which do not reveal the true quality of the water. Although the weighted arithmetic water quality index is complex, the results are easy to interpret and facilitate its use by the institutions in charge of decision making.

Study area

The Ozana river basin is located in the NE part of the Eastern Carpathians, extending across three major landform units: the Stâni°oara Mountains, the Moldavian Subcarpathians, and the Moldavian Plateau. The basin, which spreads across a W-E direction, is framed by the 47°08’19" and 47°18’15" northern latitude parallels, and by the 25°55’35" and 26°33’45" eastern longitude meridians (fig. 1). The Ozana has its source in the flysch mountainous area, at an altitude of ~ 1520 m. It is 59 km long, and it gathers its waters from a 410 km^2 watershed. It is one of the right-side tributaries of the Moldova River, discharging downstream of the Timi°e°ti locality [14, 15]. Its Eastern Carpathian hydrological regime is characterized, in spring, by high water levels, with a delayed apex during the month of May [16]. Geologically, the Ozana river floodplain is composed of clastic rock deposits (gravel, boulders, sand and clay), with an average thickness of 12 m. The permeable, unconsolidated rock deposits facilitate water seepage and emphasize a “river drying” effect close to the river mouth [17]. The Ozana river crosses 3 communes (Pipirig, Vânãtori-Neamþ and Timi°e°ti) and Tg. Neamþ City, riverine settlements with a total population of 42500 inhabitants.

Methodology

In the Ozana river basin, there are two sampling sites for the assessment of water chemistry and quality: Boboie°ti (located in the mountainous area, downstream

* email: geluromanescu@yahoo.com

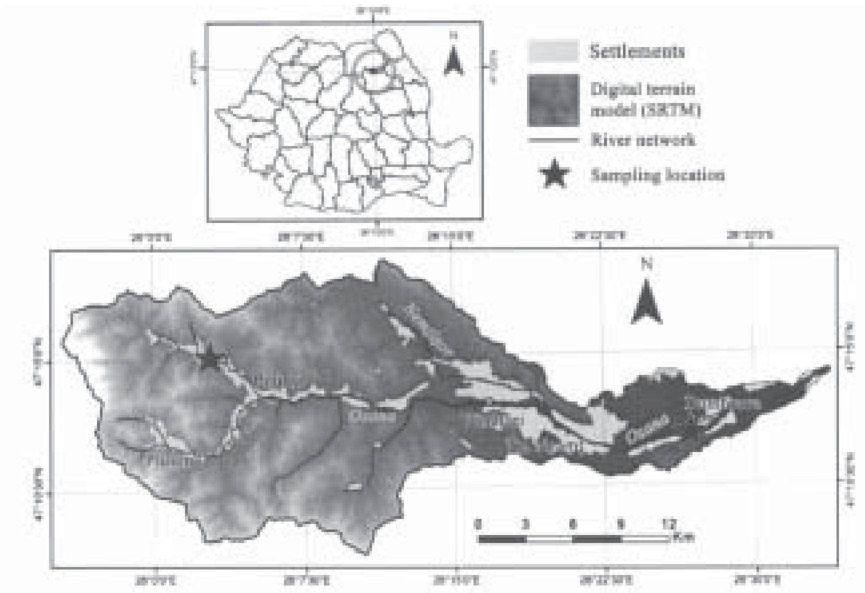

Fig. 1 Location of the Ozana river basin and the sampling sites

of the confluence of the Neamþului cel Mic and Neamþului cel Mare rivers), and Dumbrava (located 11 km upstream of the confluence of the Ozana and Moldova rivers) (fig. 1). Water samples were collected from the two sections so as to determine indicators of the following groups: oxygen consumers, nutrients, toxic substances, in agreement with the monitoring system manual developed by the specialists of the Siret Water Basin Administration (Bacău). The data used were obtained from the measurements carried out in the monitoring sections over a period of 11 years (2004-2014). In order to highlight the chemical characteristics and to evaluate the water quality of the Ozana river, a series of methods, which imply the evaluation of the following descriptive statistical parameters: central tendency indicators, scatter indicators and shape distribution indicators, were employed.

The time span during which the study was carried out is relatively short. Nevertheless, the comparative evaluation of the statistical parameters can highlight several changes in water chemistry in the Ozana floodplain, especially those induced by anthropic activities. The water quality assessment was performed according to the provisions of Order 161/2006 and Water Bodies Law 107/1996, as well as all subsequent changes and additions. The water chemistry was evaluated with respect to the impact of hazardous substances (organic micro-pollutants, heavy metal ions etc.) over water quality. In order to spatially interpret the evolution of water quality, a table summarizing the physicochemical parameters was devised, through which the differences in their respective values, as recorded at the upstream and downstream stations, were highlighted. Thus, the impact which the riverine population has over water quality could be monitored.

In order to achieve a more synthetic interpretation of water quality, the weighted arithmetic water quality index was used [4]. Its calculation is based on the annual mean of the parameters analyzed, leading to a single value characterizing the degree of water pollution.

Results and discussions

Oxygen indicators

Dissolved oxygen displays high values, as well as a seasonal variation, at both stations. A significant increase was noticed during the cold season, and a decrease during the warm one, when the minimum values are registered (fig. 2). The Bravais-Pearson linear correlation between dissolved oxygen and temperature has a value of -0.7, which represents a strong negative correlation. Therefore, the values are inversely proportional: the higher the temperature, the lower the dissolved oxygen level. The seasonal differences for this parameter can be explained by the fact that, for all elements (C into CO_2, HCO_3 into CO_3, N into NO_3, S into SO_4 etc.) [18-20], maximum oxidation occurs in a greater proportion during the warm season, as well as when there is increased algal growth [21, 22].

For the oxygen demand, an obvious difference exists between the two stations, based on the fact that an increase in nutrients occurs (fig. 3, table 1) as a result of a significant amount of organic matter from agricultural lands, poorly-rigged domestic sewage, and the Tg. Neamþ water treatment station, which is ranked as a major pollution source (colloidal suspensions, organic substances, ammonia, nitrogen, sulfides) [6]. The lack of a developed economic infrastructure in the upper river basin is reflected in the low values of the oxygen indicators (biochemical and chemical oxygen demand) in the control section at Boboie°ti. Downstream, given the increase in

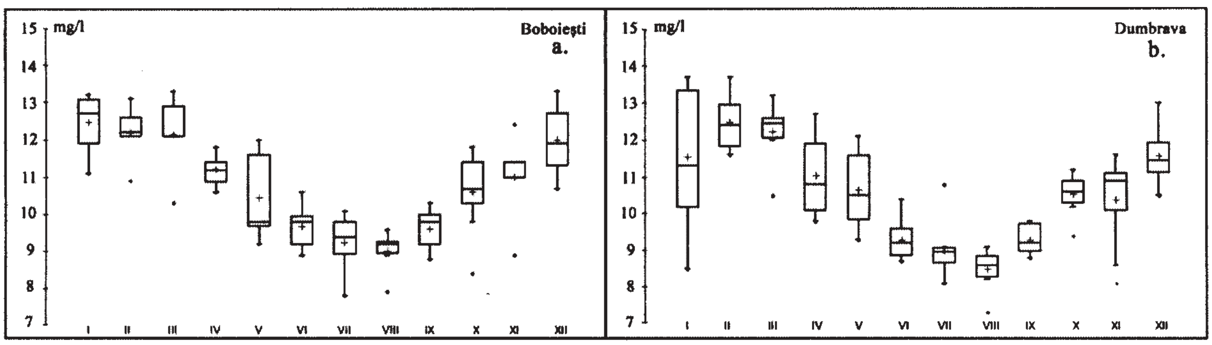

Fig. 2 Dissolved oxygen – Boboie°ti (a) and Dumbrava (b) – for the 2004-2014 period

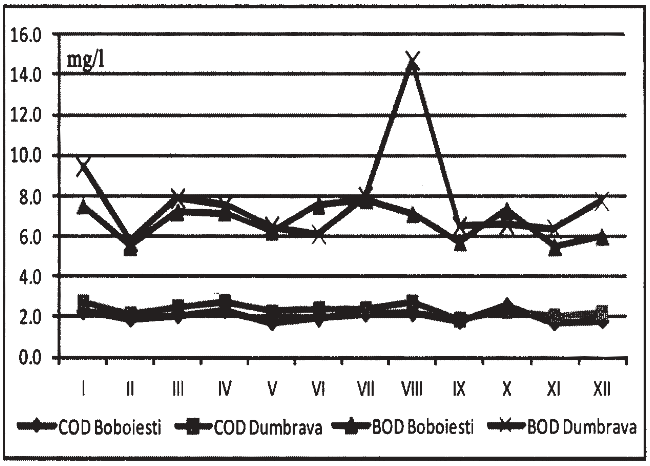

Fig. 3 COD and BOD values – Boboie°ti and Dumbrava (2004-2014 multi-annual mean)

Table 1
DESCRIPTIVE STATISTICS FOR THE MAIN HYDRO-CHEMICAL PARAMETERS ANALYZED

Station	Parameter	min.	max.	med.	std. dev.*	VC**
Boboieşti	Dissolved oxygen (mg/L)	7.8	13.3	11.03	0.304	0.050
Dumbrava		7.3	13.7	10.76	0.914	0.085
Boboieşti	Oxygen saturation (%)	75.68	107.24	94.72	2.213	0.023
Dumbrava		66.93	122.17	95.97	3.899	0.041
Boboieşti	Biochemical oxygen demand (mg/L)	0.7	4.5	2.25	0.570	0.253
Dumbrava		1	6.4	2.50	0.509	0.203
Boboieşti	Chemical oxygen demand (potassium dichromate method) (mg/L)	0.3	5.57	2.08	0.257	0.124
Dumbrava		0.61	32.51	2.63	1.025	0.390
Boboieşti	Chemical oxygen demand (potassium permanganate method) (mg/L)	2.8	15	7.66	2.777	0.363
Dumbrava		2.2	47.5	8.51	2.673	0.314
Boboieşti	Ammonium (mg/L)	0	1.556	0.037	0.047	1.287
Dumbrava		0	3.446	0.113	0.105	0.931
Boboieşti	Nitrite (mg/L)	0	0.02	0.005	0.003	0.550
Dumbrava		0	0.09	0.015	0.004	0.297
Boboieşti	Nitrate (mg/L)	0.023	1.987	0.636	0.205	0.322
Dumbrava		0.007	8.733	1.574	0.353	0.225
Boboieşti	Phosphorus (mg/L)	0.003	0.050	0.015	0.007	0.444
Dumbrava		0.003	0.166	0.039	0.029	0.740
Boboieşti	Friable residue (mg/L)	139.5	343.5	243.324	85.742	0.352
Dumbrava		208	425.6	284.287	18.151	0.064
Boboieşti	Chlorides (mg/L)	5.3	27.3	13.072	3.077	0.235
Dumbrava		9.9	57.9	19.400	2.750	0.142
Boboieşti	Sulfates (mg/L)	12.6	38.4	23.396	2.475	0.106
Dumbrava		16.3	67.2	29.108	4.012	0.138
Boboieşti	Calcium (mg/L)	32.9	84.8	62.297	7.179	0.115
Dumbrava		44.900	109.3	77.846	4.128	0.053
Boboieşti	Iron (mg/L)	0.01	0.31	0.088	0.038	0.434
Dumbrava		0	0.27	0.089	0.035	0.393

*std. dev. – standard deviation;
**VC – variation coefficient.

anthropic impact through additional wastewater input in the rural areas, particularly in that surrounding Tg. Neamt City, the values of the oxygen indicators, especially those of the biochemical oxygen demand, increase, reaching values of up to 47.5 mg/L. The occurrence frequency of such values is, however, quite low, less than 10%. In both cases, there are no major seasonal changes to be considered. In the case of the biochemical oxygen demand, the highest value is recorded in the month of August.

Nutrients

The nutrients present in the Ozana river are the following: ammonium, nitrite, nitrate and phosphorus. They have relatively low toxicity, but are, nevertheless, regarded as indicators of anthropic pollution. An increase in values occurs between the Boboie°ti and Dumbrava sections (table 1). The highest variation in ammonium is registered in the Boboie°ti section, and it originates from agricultural production [23].

The variation coefficient is also high in the vicinity of Dumbrava, but due to climatic conditions. The fertilizers used during the previous warm season have low values in the months of May through August, and are correlated with the occurrence of floods (2004, 2008, 2010, 2011, 2013), because of which the topographic surface was washed by waters carrying ammonium [24, 25].

Nitrates register lower values at Boboie°ti, and higher values at Dumbrava (fig. 4a). The increase in nitrate

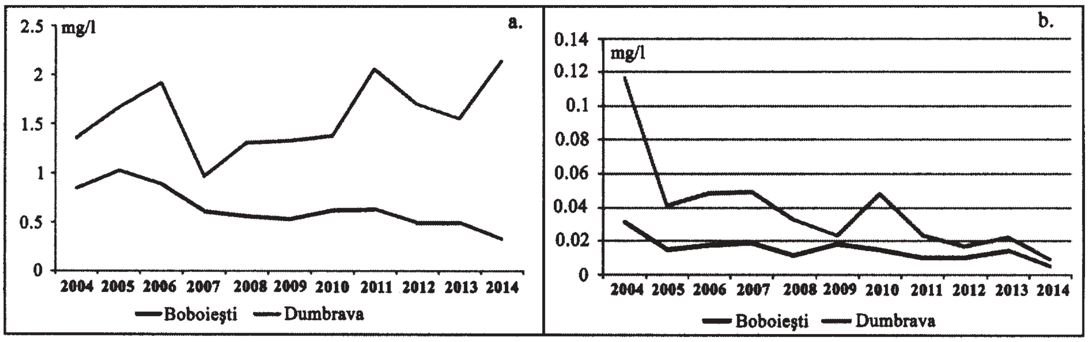

Fig. 4. Mean annual amount of nitrates (a) and phosphorous (b) at Boboiesti and Dumbrava

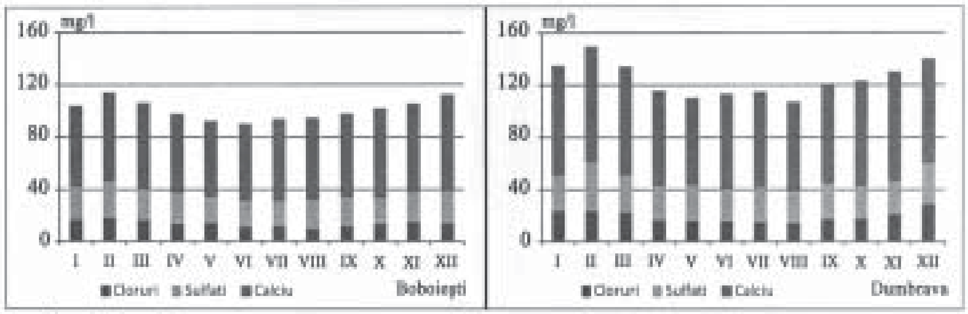

Fig. 5 Monthly multi-annual mean of the salinity in the Boboie°ti and Dumbrava sections

concentration is due to the use of nitrogen-based chemical fertilizers in the preceding seasons [26]. In recent years, the usage of manure has increased. This manure is stored in households or onto the river floodplain, being, therefore, exposed to pluvial runoff or floods. The villages located in the floodplain, and the high permeability of the fluvial deposits, lead to a rapid increase in the amount of nitrates present in the water [27].

Phosphorous pollution is attributed most often to human settlements and the precarious storage of human and animal waste from agricultural areas [28-30]. A sudden decrease in the phosphorous level occurs, caused by the introduction of regulations concerning the existence of septic tanks and the restriction of rural leakage (fig. 4b). The nutrients do not display seasonal fluctuation.

Salinity

The water chemistry of a river is determined, mainly, by the geological conditions under which the respective river unfolds its hydrographic basin [31]. For the Ozana river, the geological conditions, determined by the presence of friable sedimentary deposits, lead to high values of the fixed residue (high erosion capacity): 343.5 mg/L at Boboie°ti, and 425 mg/L at Dumbrava. The maximum chloride content increases from 27.3 mg/L at Boboie°ti to 57.9 mg/L at Dumbrava. The maximum sulphate content increases from 38.4 mg/L at Boboie°ti to 62.7 mg/L at Dumbrava (table 1). The frequency of the maximum values is much higher during the cold season, but they also exhibit a high percentage in autumn and, sparsely, in spring.

The salinity in the two sections registers visible seasonal variations. The high values are correlated with the cold season (December-February), being recorded during the lowest flow rate of the year (fig. 5). This type of salinity concentration distribution is globally specific, differences being noticed between the wet and dry seasons in all the climate zones [32-41].

Iron

Iron is present as a result of both natural causes (geological strata) and anthropic factors (mining and used waters). The values are low, with peaks of 0.310mg/L at Boboiesti and 0.270mg/L at Dumbrava. This ranks the water as having 1st class quality.

Water quality

For the assessment of water quality, data from 2004-2014 were used. The water quality index was calculated based on 14 parameters, grouped into 4 categories: oxygen indicators, nutrients, salinity and pollutants [42-46]. For the present study, only the physicochemical parameters were considered, along with the weight of each parameter.

The weighted arithmetic water quality index is obtained using the formula:

$$WQ_i = {\sum Q_i W_i}/{\sum W_i}$$

where Q_i is calculated for each parameter using the following equation:

$$Q_i = 100[(V_i - V_o)/(S_i - V_o)]$$

and W_i is calculated using the following formula:

$$W_i = K/S_i$$

where: $K = 1/\sum(1/S_i)$;

Q_i = the quality rating scale;

W_i = the weight unit;

V_i = the estimated concentration of the parameter in the water;

V_o = the ideal value of the parameter (V_o = 0 (except for pH = 7.0 and DO = 14.6 mg/L));

S_i = the recommended standard value of the parameter

K = the proportionality constant.

For each parameter, the analysis on an annual level was attempted, but the lack of data consistency led to the impossibility of obtaining a relevant result. Therefore, an evolutionary trend at parameter level could not be emphasized. Consequently, the water quality index was calculated for the multi-annual means of each parameter, so that the result would allow for the water of the Ozana river to be placed into a specific quality class, depending on the section where the sampling was performed.

After calculating the weight for each parameter (W_i), it was observed that 90% of the impact over water quality is due to the concentration of nitrates and phosphorous (Table 2). The reason for this exaggerated weight is given by the existence of a maximum allowed concentration close to

	W_i Boboieşti	Q_i Boboieşti	Q_i*W_i Boboieşti	W_i Dumbrava	Q_i Dumbrava	Q_i*W_i Dumbrava
Dissolved oxygen	0.00229	-93	-0.2129	0.00229	-78	-0.1785
Oxygen saturation	0.00023	-22.7	-0.0052	0.00023	-33.05	-0.0076
Biochemical oxygen demand	0.00320	-46	-0.1474	0.00320	-31	-0.0993
Chemical oxygen demand (potassium dichromate method) (mg/L)	0.00160	-59.2	-0.0949	0.00160	-47.2	-0.0756
Chemical oxygen demand (potassium permanganate method) (mg/L)	0.00064	-21.0667	-0.0135	0.00064	-14.8667	-0.0095
Ammonium	0.02003	-90	-1.8025	0.02003	-77.5	-1.5521
Nitrites	0.53407	-25	-13.3518	0.53407	25	13.3518
Nitrates	0.00534	-14	-0.0748	0.00534	27.5	0.1469
Total phosphorous	0.40055	8	3.2044	0.40055	100	40.0555
Friable residue	0.00002	-110.8	-0.0024	0.00002	-87.68	-0.0019
Chlorides	0.00032	-44.92	-0.0144	0.00032	-22	-0.0070
Sulfates	0.00013	-61.8167	-0.0083	0.00013	-53.3333	-0.0071
Calcium	0.00016	28.74	0.0046	0.00016	56.18	0.0090
Iron	0.03204	-110	-3.5249	0.03204	-105	-3.3647

Table 2
THE COMPONENTS OF THE FORMULA FOR THE WEIGHTED ARITHMETIC WATER QUALITY INDEX

the value of 0 (0.01 mg/L – nitrites; 0.015 mg/L phosphorous), which means that the reporting of these two elements in the water is associated, most likely, with pollution, and implicitly, a decrease in quality.

The allowed quality and inadequate quality thresholds range between 0 and 100. The low values designate high quality, while the high values are associated with a very low quality class. In the current case (where specific values for water quality in Romania were used), negative values were also recorded, highlighting the existence of a very good quality class. High, positive values are displayed by the parameters linked to nutrients (nitrates, nitrites and total phosphorous), indicating the presence of a pollution source upstream both of the sampling sites. The value of the water quality index is -16 at Boboie°ti, placing it directly into the very good quality class. The value of this index is 48.3 at Dumbrava, which ranks the quality of the water as being at the limit between good and poor.

Conclusions

The topic discussed, as well as its association with the study area, are currently of great importance, considering the existence of a water catchment system for a regionally important city (Tg. Neamt), but also of pollution sources deriving mainly from agricultural activities. The present study focused on 14 physicochemical parameters, which were analyzed in the water samples collected from two sections (Boboie°ti and Dumbrava). These parameters were grouped into 4 categories: oxygen indicators, nutrients, salinity and iron.

There is close correlation between the values of the parameters related to oxygen demand and seasons, of which the most prominent is the warm season, with a low chemical oxygen demand and a high biochemical oxygen demand. As far as the nutrients are concerned, a tendency toward reduction in quantity is noticeable. Exempt from this reduction are the nitrates, which display a growth tendency in the Dumbrava section between 2007 and 2010. Salinity values are dependent on seasonal variations of the flow, but also on the years with periods of drought. A pollution indicator is the iron content, whose maximum value is 0.21-0.31 mg/L (it does not, thus, represent a pollution risk).

In order to integrate all the available parameters into a unitary form, the water quality index was calculated for both sampling sections. The water of the Ozana river is associated with the excellent quality class in the Boboie°ti section, and the good to poor quality class in the Dumbrava section. The water quality index also revealed the fact that the parameters which have the most powerful impact on water quality are nitrites and phosphorous. Apart from the natural influence of the seasonal cycles, the anthropic impact is also emphasized, through the input of used waters (rural and urban environments), chemical fertilizers and human and animal waste.

Acknowledgments: The present work was supported by the POSDRU 159/1.5/133391 strategic grant, as part of the "Doctoral and post-doctoral programs of excellence for highly qualified human resources training for research in the field of Life Sciences, Environment and Earth Science" project, co-financed by the European Social Fund within the Sectorial Operational Program Human Resources Development 2007-2013.

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Manuscript received: 16.08.2015