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Ecological assessment of Cr (VI) concentrations in the surface waters of Stara Zagora Region used in agriculture

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Abstract. *The aim of the present study was ecological monitoring of Cr (VI) concentrations in surface waters from Stara Zagora Region used in agriculture. The research goal was accomplished by: determining Cr (VI) concentrations in real surface water samples taken from 16 sampling points situated in four municipalities: Stara Zagora, Kazanlak, Chirpan and Gurkovo, by AAS (Atomic Absorption Spectrometry) during the period December, 2009 - November, 2010; comparative estimation of the experimental results from the standpoint of national/international surface water quality standards; statistical analyses of the data. The scientific investigations revealed that the II category standard (0.05 mg/L Cr (VI)) was exceeded by 2 % - 30 % in approximately 19 % of the samples analyzed (Tundzha River - bridge Yagoda Village, Radova River, Zetyovo Dam, Chirpan Dam). The registered Cr (VI) concentrations during November, 2010, in 31 % of the surface water bodies practically coincided with the above cited permissible limit.*

Keywords: Cr (VI), water bodies, ecological assessment, AAS, Stara Zagora Region

Introduction

The interest in inorganic chromium speciation originates from the widespread industrial use of this metal in the metallurgical, chemical and refractory brick industries. Major applications of hexavalent chromium compounds include metal plating, manufacture of pigments and dyes, corrosion inhibitors, chemical synthesis, refractory production, leather tanning, wood preservation, anodising aluminium in aircraft industries (Giusti and Barakat, 2005; Gowd and Govil, 2008; Vasilatos et al., 2008). Therefore, the quantity of inorganic Cr has been identified as an important water quality index (Yusof et al., 2007). Industrial releases of Cr (VI) compounds to surface water and soil can result in the transport and leaching of these substances into groundwater.

Chromium can exist in oxidation states ranging from -2 to +6, but only +3 and +6 are typically found within the range of pH and redox potential common in environmental systems (Losi et al., 1994; Cieslak-Golonka, 1996; Kotas and Stasicka, 2000; Krishnani and Ayyappan, 2006). Both Cr (III) and Cr (VI) have contrasting biological, geochemical and toxicological effects. Cr (III) is considered to be a trace element species essential for the proper functioning of mammals. It is able to maintain the metabolism of glucose, lipids and proteins, although in excess it can cause allergic skin reactions and cancer (Yusof et al., 2007; El-Sikaily et al., 2007; Miretzkya and Cirelli, 2010). Cr (III) has low solubility in water and readily precipitates as $Cr(OH)_3$ or mixed Cr-Fe hydroxides at pH values greater than 4. The oxidation potential for transforming Cr (III) into Cr (VI) is high, and the probability of transformation into a higher oxidation form in environmental conditions is reduced (Vasilatos et al., 2008).

The environmental impact of Cr (VI) is a controversial issue, critical to the protection of natural water resources (Vasilatos et al., 2008). Cr (VI) is the most toxic form, being carcinogenic and mutagenic to living organisms. In addition, it leads to liver damage, pulmonary congestion and causes skin irritation resulting in ulcer formation. Inhalation and retention of Cr (VI)-containing materials can cause the increased incidence of bronchogenic carcinoma. Skin contact of Cr (VI) compounds can lead to skin allergies and cancer (Yusof et al., 2007). It is assumed that hexavalent chromium is about 100-1000 times more toxic than trivalent chromium and due to the fact that it has limited hydroxide solubility, it is less mobile and less bioavailable (Yayıntas et al., 2007; Miretzkya and Cirelli, 2010). Cr (VI) has the ability to permeate biological membranes. Once inside the cell, it is reduced to trivalent chromium, either enzymatically or nonenzymatically - a process called "intracellular" reduction. A series of *in vitro* and *in vivo* studies have demonstrated that Cr (VI) induces an oxidative stress through enhanced production of reactive oxygen species (ROS) leading to genomic DNA damage and oxidative deterioration of lipids and proteins. A cascade of cellular events occur following Cr (VI)-induced oxidative stress including enhanced production of superoxide anion and hydroxyl radicals, increased lipid peroxidation and genomic DNA fragmentation, modulation of intracellular oxidized states, activation of protein kinase C, apoptotic cell death and altered gene expression (Bagchi et al., 2001).

In environmental systems, Cr (VI) exists as oxyanions such as chromate (CrO_4^{2-}), bichromate ($HCrO_4^-$) and dichromate ($Cr_2O_7^{2-}$) and therefore has high solubility in water and is far more mobile than the trivalent chromium. High Cr (VI) concentrations in surface water bodies are indicators of anthropogenic pollution (Vasilatos et al., 2008) and require the application and implementation of proper treatment methods and/or other quality control strategies, especially when these waters are intended for irrigation or animal watering (Mishra et al., 1995).

The aim of the present study was ecological assessment of Cr (VI) concentrations in surface waters from Stara Zagora Region

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used in agriculture.

Material and methods

A total of 128 surface water samples, taken from 16 sampling points situated in four municipalities, Stara Zagora, Kazanlak, Chirpan and Gurkovo, of Stara Zagora Region, were collected and analyzed during the period December, 2009 - November, 2010 (Table 1). All water samples were collected and preserved according to the standard methods for the examination of water and wastewater (American Public Health Association, 2008). The concentrations of Cr (VI) were determined by atomic absorption spectrometry (AAS) ISO 8288 on AAnalyst 800 (*Perkin Elmer)* Atomic Absorption Spectrometer. All investigations were performed in triplicate.

The ecological assessment of the surface water quality was done on the basis of national and international surface water quality standards (Regulation No. 7/8.08.1986; USEPA, 1996; WHO, 1997), and statistical analyses of the data.

The statistical significance of the results was tested on the basis of the Standard Deviation (SD) values calculated by the Student's t-test.

Results and discussion

The temporal and spatial trends of natural water quality fluctuations depend on a number of factors such as: physical-geographical conditions, soil content, the hydro-physical, hydro-chemical and hydro-biological processes occurring in the water bodies (Kostadinova, 2006). The assessment of the anthropogenic impact on the quality of natural water bodies implies the application of the ecosystematic approach (Kostadinova et al., 2006). In the present study, the spatial and temporal variations of the experimentally determined Cr (VI) concentrations in the investigated 128 surface water samples taken from 8 rivers and 3 dams during the period December, 2009 - November, 2010, were traced out.

The analyses of the downstream of Tundzha River quality regarding Cr (VI) contents, including assessment of the influence of its tributaries flowing through the investigated area, is essential and imperative, considering that it is one of the biggest rivers in Bulgaria with about 50 tributaries and the most significant tributary of Maritsa River, flowing into it on the Turkish territory near Edirne, and one of the main water resources for drinking, domestic purposes and irrigation (Barakova et al., 2004). Thus, the concentrations of the toxic metal in four sampling points from the river itself (s.p. 206, 210, 211, 214) and its five tributaries (s.p. 207, 208, 209, 212, 213) passing through the territory of Kazanlak and Gurkovo Municipalities were determined in the present study (Figure 1). The ecological monitoring established elevated Cr (VI) contents in the surface water samples from s.p. 211 (Tundzha River - bridge Yagoda Village) in November 2010 and in s.p. 212 (Radova River) in December 2009 and July 2010. The II category standard for Cr (VI) contents in surface waters, regulated by the Bulgarian legislation, (0.05 mg/L Cr (VI)) in these water bodies was exceeded by 14 % - 30 % (Regulation No. 7/8.08.1986). Besides, the presented concentration curves are characterized with analogous mode - with clearly outlined peak concentrations in s.p. 210, 211 and 212.

To compare and assess the relative influence of the feeders waters on the quality of Tundzha River, Cr (VI) contents was

Table 1. A list of the investigated surface water sampling points in Stara Zagora Region

Municipality	Sampling Point (s.p.)	Code	Coordinates
Stara Zagora	Bedechka River before Zagorka Lake	201	N 42.27049^0 E 25.37937^0
	Bedechka River after Zagorka Lake	202	N 42.32740^0 E 25.33805^0
	Sazliika River - St. Zagora Mineral Baths	204	N 42.26914^0 E 25.29015^0
	Tundzha River (bridge Yagoda Village)	211	N 42.32740^0 E 25.33805^0
	Sazliika River (after Galabovo City) (Gurkovo Municipality)	205	-
Chirpan	Chirpan Dam	215	N 42.09753^0 E 25.18842^0
	Zetjovo Dam	216	N 42.09112^0 E 25.21541^0
	Malak Yurt Dam	217	N 42.19494^0 E 25.18706^0
Kazanlak	Tundzha River (before Koprinka Dam)	206	N 42.41215^0 E 25.18863^0
	Gabrovnitsa River (Dolno Sahrane Village)	207	N 42.38983^0 E 25.16554^0
	Leshnitsa River (after Dunavtsi Village)	208	N 42.38960^0 E 25.16479^0
	Eninska River (after Enina Village)	209	N 42.40004^0 E 25.24538^0
	Tundzha River (after Buzovgrad Village)	210	N 42.34484^0 E 25.24920^0
Gurkovo	Radova River	212	N 42.39392^0 E 25.46381^0
	Lazova River	213	N 42.41709^0 E 25.47040^0
	Tundzha River (Zhrebchevo Dam mouth)	214	N 42.38333^0 E 25.49350^0

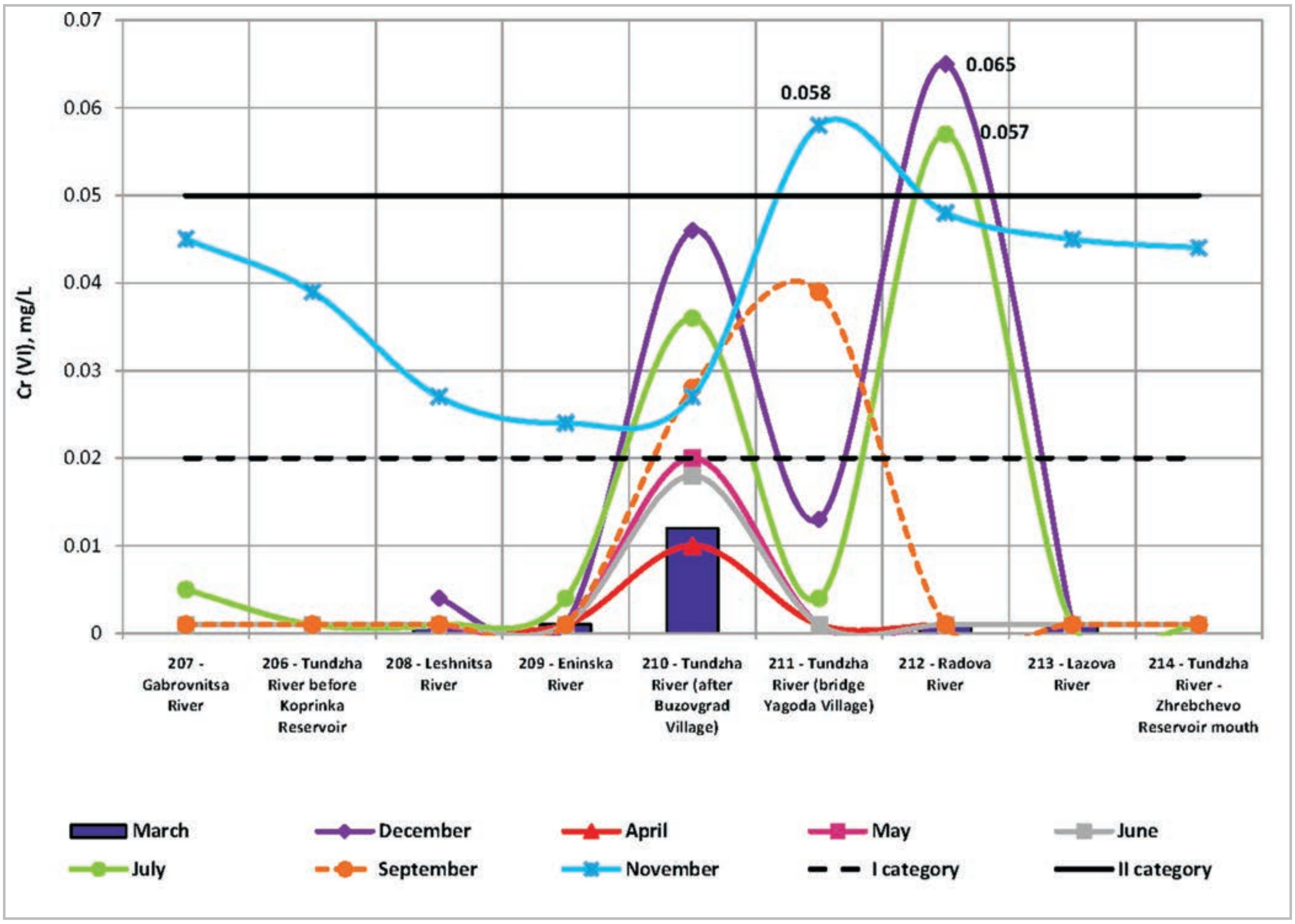

Figure 1. Monthly variations of Cr (VI) concentrations in the surface water samples from the downstream of Tundzha River and five of its feeders

presented in an area plot on monthly bases (Figure 2). It should be outlined that the data displayed was selected to represent logically the measured metal concentrations in the feeder and in the sampling point in the river itself, immediately after its influx. Consequently, the greater Cr (VI) loading of Tundzha River waters in June and September, 2010, could not be attributed to the tributaries waters. However, the registered peaks for both water types in July, 2010, and especially in November, 2010, almost coincided. Thus, the assumption of Cr (VI) mass transfer from the feeders waters into the river seemed logical. Accordingly, these elevated heavy metal concentrations in the upstream and downstream waters of the river could be due to uncontrolled discharge of industrial wastewaters from the wood-processing, machine building and metal plating enterprises situated in the studied area. The latter was sustained by the observations of Kostadinova et al. (2007), which characterized the wastewaters from the town of Kazanlak as irregularly allocated according to their pollution extent, quality indices, temporal and spatial distribution, and established a few pollution cases based on individual quality parameters.

The surface water samples taken from points 201 and 202 (Table 1) were representative for the quality of one of the biggest rivers passing through the territory of Stara Zagora Municipality: Bedechka River - crossing the eastern part of the district center - the city of Stara Zagora, characterized by concentrated industrial activities. The ecological assessment of Bedechka River and Zagorka Lake waters quality, conducted during the period 2004-2005, proved unconformities with the permissible standards only according to the parameters nitrites and nitrates (Kostadinova, 2006). Sampling points 204 and 205 were selected as representatives of the water quality of Sazliika River (Table 1). The measured Cr (VI) concentrations in these surface water bodies are presented in Figure 3. Obviously, the highest levels detected in November, 2010, in s.p. 201, 202 and 204 were only 4 % lower than the II category limit, i.e. practically coinciding with the standard. The mode of the corresponding concentration curves seemed analogous with that observed for Tundzha River (s.p. 211), namely characterized by sharp concentration increase during September and November, 2010.

The experimental concentration curves for Cr (VI) contents in Chirpan (s.p. 215), Zetyovo (s.p. 216) and Malak Yurt (s.p. 217) dams, Chirpan Municipality (Figure 4), also demonstrated elevated heavy metal levels during September and November, 2010. The sharply outlined steep concentration leap from values below the I category standard (0.02 mg/L) to levels exceeding the II category standard (0.05 mg/L) was disturbing (Regulation No. 7/8.08.1986). The highest measured Cr (VI) concentrations during both cited months in s.p. 216 were 0.051 mg/L and 0.052 mg/L, respectively. Regarding the temporal variations of Cr (VI) concentrations in the surface waters of Stara Zagora Region, an analogous outlined tendency of elevated chromium contents during September and November in approximately 60 % of the investigated water samples was observed. The assumption of accidental spills and pollution events that had probably occurred in some of the sampling sites during both months could be accepted as a probable pollution

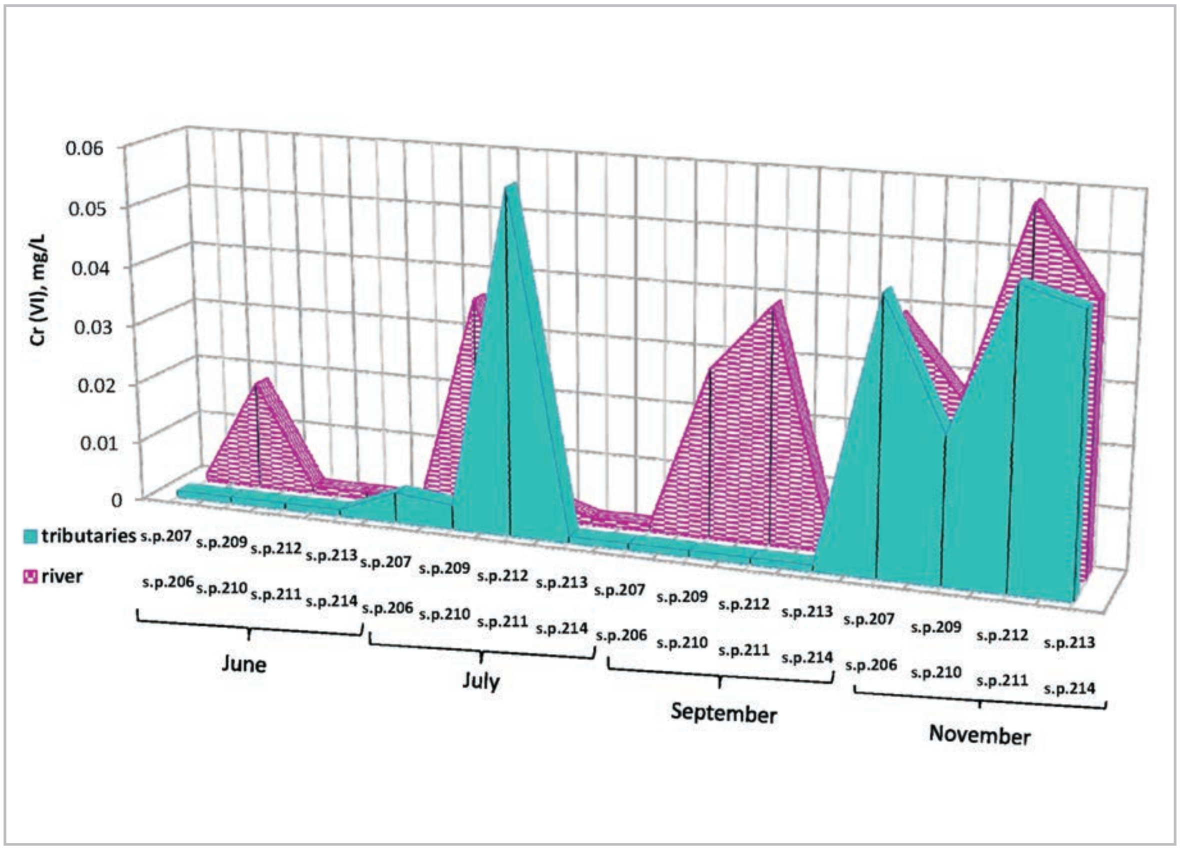

Figure 2. Area plot of Cr (VI) contents in the surface waters of Tundzha River and its feeders during the period June, 2010 - November, 2010

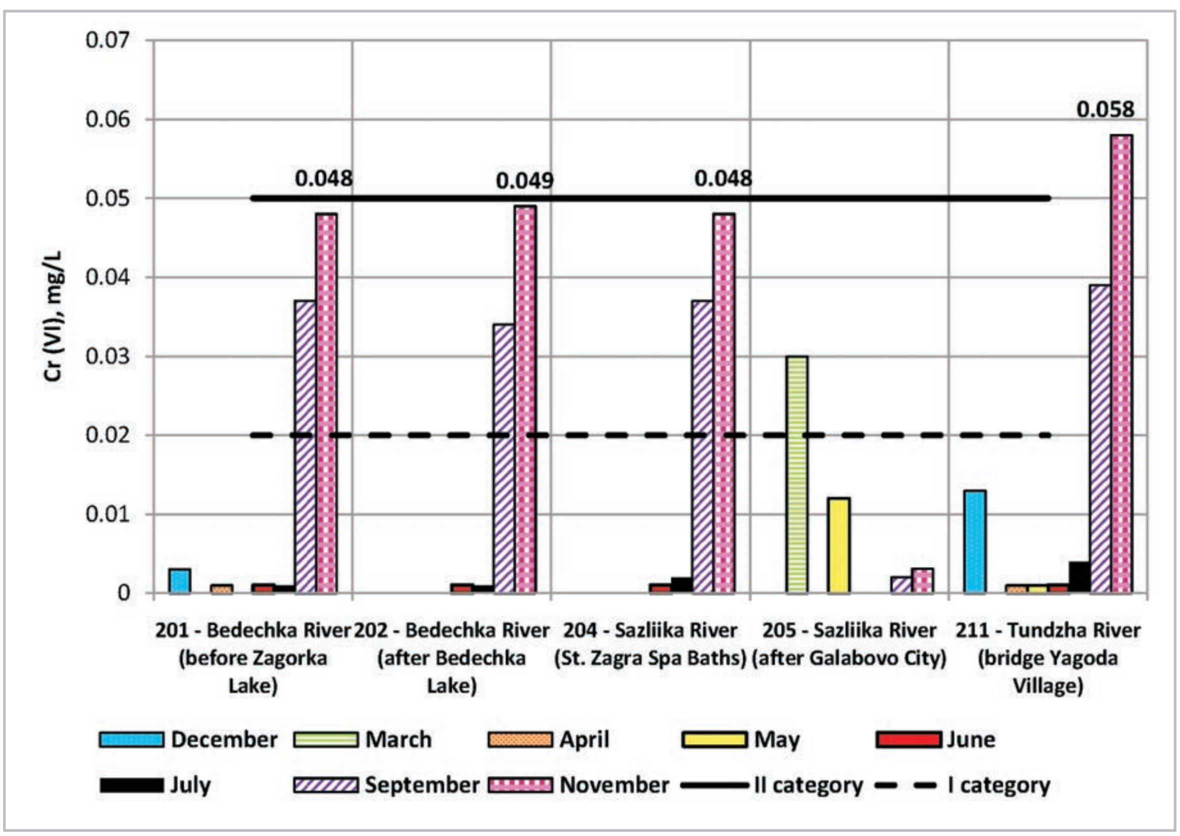

Figure 3. Chromiun (VI) concentrations in the surface waters of Stara Zagora Municipality during the monitored period

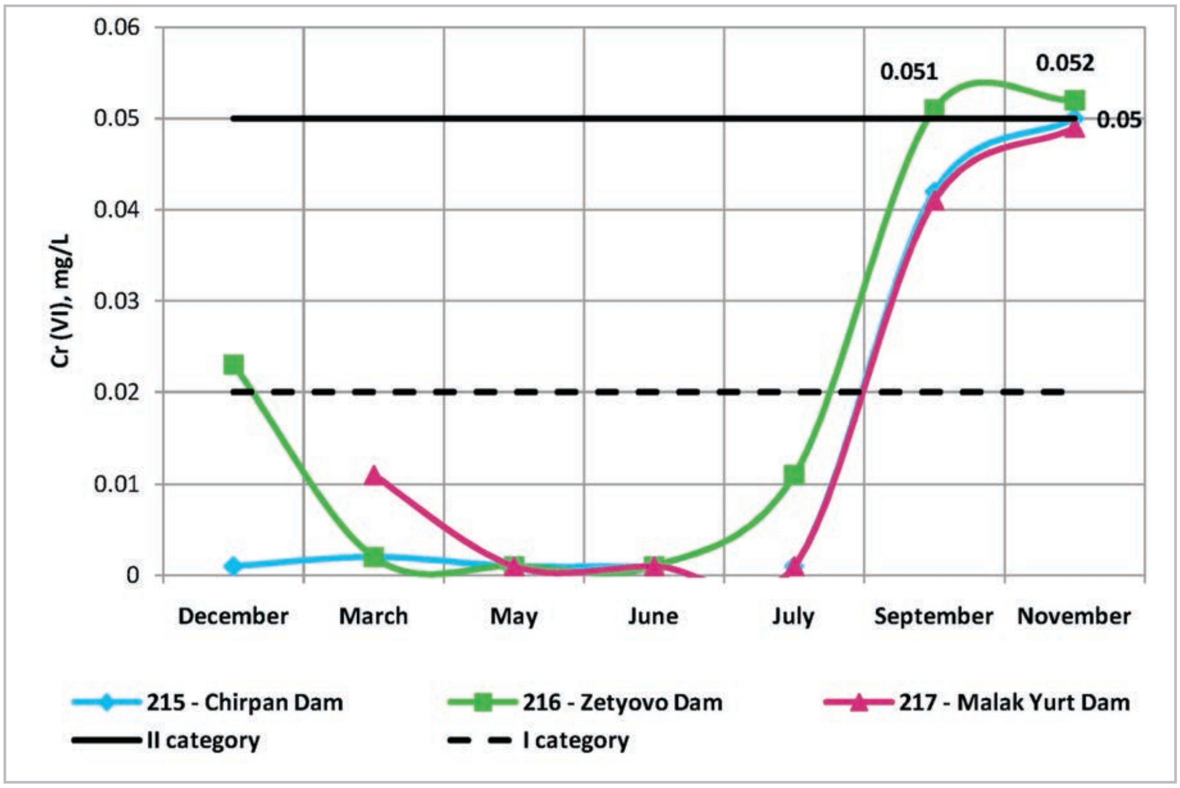

Figure 4. Chromium (VI) concentrations in the dam waters of Chirpan Municipality

Table 2. Maximum, minimum and mean annual Cr (VI) concentrations and SD values

Municipality	s.p.	C*, mg/L	C_{max}, mg/L	C_{min}, mg/L	SD
Stara Zagora	201	0.013	0.048	0.001	0.008
	202	0.021	0.049	0.001	0.012
	204	0.022	0.048	0.001	0.012
	211	0.017	0.058	0.001	0.009
Chirpan	215	0.014	0.050	0.001	0.008
	216	0.020	0.052	0.001	0.009
	217	0.017	0.049	0.001	0.009
Kazanlak	206	0.011	0.039	0.001	0.010
	207	0.013	0.045	0.001	0.011
	208	0.005	0.027	0.001	0.003
	209	0.004	0.024	0.001	0.003
	210	0.030	0.046	0.012	0.007
Gurkovo	212	0.022	0.065	0.001	0.006
	213	0.007	0.045	0.001	0.010
	214	0.012	0.044	0.001	0.011

C* - mean annual concentration; C_{max} - maximum concentration; C_{min} - minimum concentration resource to some extent. However, the explanation of the observed uniform Cr (VI) concentration curves modes and registered peaks should be sought at a more global scale. Thus, we propose that chromium contents variations in most of the examined surface water samples is mediated by the seasonal variations of the biological activity in the water bodies. Probably, Cr (VI) was reduced to Cr (III) during the periods of high activity, namely the spring and summer months, and the reduced form was removed via adsorption on particles or sediments. Later on, Cr (III) could be released during the remineralisation of the particles and returned in this form to the upper water layers, where it was either oxidised to Cr (VI), or stabilised by the organic compounds, remaining as Cr (III). The latter assumptions were supported by the investigations and observations of Connelly et al. (2006) based on seasonal changes in dissolved chromium speciation in the marine waters of Sargasso Sea.

The values of the maximum (C_{max}), minimum (C_{min}) and mean annual concentrations (C^*) in all examined surface water bodies measured during the entire monitored period are presented in Table 2. The SD values for all investigated sampling points were calculated and presented in Table 2. The range of their variation, from 0.003 to 0.012, proved the statistical significance of the experimental results. A clear tendency, with regard to the spatial distribution of Cr (VI) in the surface waters of the four municipalities, could not be outlined both because of the commensurable detected heavy metal levels in most of the examined water bodies and the narrow SD range. It is obvious only that the surface waters from Eninska River (s.p. 208) and Tundzha River after Buzovgrad Village (s.p. 209), Kazanlak Municipality are characterized by the lowest C^* and C_{max} Cr (VI) concentrations and the lowest SD values.

Conclusion

The one-year ecological monitoring of Cr (VI) concentrations revealed that the II category standard (0.05 mg/L Cr (VI)) was exceeded by 2 % - 30 % in approximately 19 % of the samples analyzed: Tundzha River - bridge Yagoda Village, Radova River, Zetyovo Dam, Chirpan Dam. The registered Cr (VI) concentrations in November, 2010, in 31 % of the surface water bodies practically coincided with the above cited permissible limit. The surface waters in Kazanlak Municipality are characterized by the lowest Cr (VI) contents. The generated database could be used to form an integrated Cr (VI) management strategy, as recommended by most up-to-date trends of environmental research in the field of agricultural land use planning aimed at water pollution control.

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