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Agriculture and Environment

Manganese levels in water, sediment and algae from waterbodies with high anthropogenic impact

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Abstract. *A survey and assessment of manganese (Mn) levels was carried out on the chain water – sediment – algae from 6 monitoring points, situated at three rivers and a dam with high anthropogenic impact in Stara Zagora region, South Bulgaria. International references of ISO and BSS for sampling and sample preparation of water, sediment and algae analysis were used. Manganese concentration in the collected samples was determined by atomic absorption spectrometry (AAS). It was found that despite the anthropogenic pressures on the studied waterbodies Mn content in water from all investigated waterbodies does not exceed the maximum permissible concentration, according to national Regulation No 7/1986. Mn accumulates in high levels in sediment and algae from all surveyed monitored waterbodies. The highest Mn concentrations in sediment were measured in Sazliyska River (714.5 mg/kg) and Bedechka River (799.6 mg/kg). With the highest levels of Mn were distinguished algae delivered by Yagoda Village (663.8 mg/kg), Jrebchevo Dam (476.0 mg/kg) and Sazliyka River (411, 5 mg/kg). The estimated ratios between Mn concentrations in sediment and water have shown that the accumulation of this metal in the sediment is from 1407 (Jrebchevo Dam) to 15466 (Tundzha River, Banya Village) times more than in the water. By the sediment/algae ratio it is found that Mn is accumulated from 0.5 (Jrebchevo Dam) to 2.4 times (Bedechka River) more in sediment compared to algae. The data from algae/water ratio show that Mn is accumulated from 1301 (Tundzha River at Jrebchevo Dam) to 19565 (Tundzha River at Banya Village) times more in the algae compared to the water. This fact suggests the mechanism of accumulation of Mn in the sediment and algae, probably different from simple diffusion. The obtained results indicate that sediment and algae can serve as good indicators of pollution by Mn. They can also be used for purification of water from that metal.*

Keywords: water, sediment, algae, manganese, assessment

Abbreviations: BSS – Bulgarian State Standard, MP – Monitoring Point, MPC – Maximum Permissible Concentration, WWTP – waste water treatment plant

Introduction

Environmental pollution is one of the global problems of our contemporary world. As a result of this changes infringing the natural development of ecosystems has been registered. One of the main causes of distortion of the natural equilibrium is heavy metal pollution. The groups of elements with a mass density greater than 4.5 g/cm3 are called heavy metals. They tend to form simple cations in water solutions and are important contaminants of aquatic environments worldwide (Sevcikova et al., 2011). Many studies have already reported that most metals are moderately soluble in water, depending on their chemical state. One of the key factors determining the solubility of metals in water is its hardness. The greater the degree of the hardness, the lower the degree of solubility of most metals. Salts actually bind metals, making them insoluble. Most commonly these bound forms of metals end up sinking to the substrate and cannot be easily absorbed by aquatic organisms from the water. (Rai et al., 1981; Everall et al., 1989; Rios-Arana et al., 2004). In small quantity some of them are absolutely necessary for the life and reproduction of all living organisms, but over certain levels they make disorders on the ecological balance and diversity of hydrobionts (Farombi et al*.,* 2007; Vosyliene and Jankaite, 2006; Ashraj, 2005). Because heavy metals haven't got the ability to decompose, they have accumulated in sediment and aquabionts.

The water mass current density creates conditions for sedimentation (i.e., streambed) of the hauled precipitating suspended particles. Sedimentation ability depends on the flow speed and particle size. Suspended sediments adsorb pollutants such as heavy metals and reduce their concentration in the water column (Douben and Koeman, 1989). Monitoring water and sediment quality is one way of assessing waterbody conditions and the effectiveness of regulatory and management efforts.

Heavy metals in sediment can pass along through the food chain, starting with benthic bacteria, algae, and benthic invertebrate organisms. Metals transferred through aquatic food chains and webs to fish, humans and other animals are of more environmental concern to human health (Farkas et al., 2001; Chen et al., 2000a). Heavy metals have a low threshold for toxicity and a wide range of disabilities. The metals serving as microelements in living organisms usually occur in trace amounts that are precisely defined for each species. Both their deficiency and excess badly affect living organisms (Szyczewski et al., 2009).

Research by Abo-Rady (1980) and Mortimer (1985) showed that aquatic macrophytes and certain algae could be used as bio-indicator organisms to evaluate the presence of selected heavy metals in aquatic ecosystems. Macroalgae have been used

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extensively to measure heavy metal pollution in freshwater and marine environments throughout the world (Whitton, 1984; Maeda and Sakaguchi, 1990; Haritonidis and Malea, 1999; Conti and Cecchetti, 2003; Kamala-Kannan et al., 2008; Stengel et al., 2004). They are used as bio-indicators to eco-assessment through their distribution, size, longevity, presence at pollution sites, ability to accumulate metals to a satisfactory degree and ease of identification (Whitton, 1984; Conti and Cecchetti, 2003; Stengel, 2004; Kamala-Kannan et al., 2008).

In this context, the comparative study of the contents of these elements in water, sediment and algae is of significant importance for assessing the condition of waterbodies and its impact on aquatic organisms. The element manganese (Mn) plays an essential role in the body of hydrobionts. Over certain concentration, however, this metal has toxic effect on hydroecosystems. Determination of this heavy metal in water, sediment and algae permits evaluation of the degree of anthropogenic impact in concrete areas. A suitable object for the study of this problem are waterbodies of Central Southern Bulgaria, which are subjected to strong anthropogenic impact. In this part of the country there are large settlements (Stara Zagora, Kazanlak) and many industrial enterprises (including for production of weapons and ammunition). Moreover, the largest energy complex in the country with 4 thermal power plants is located in this area. The agriculture and many other environmental activities are very intensive, too (Christov, 2008; Georgieva, 2012; Mihaylova et al., 2012).

The aim of this study was to investigate and assess the content of the element manganese (Mn) in water, sediment and algae from waterbodies with high anthropogenic pressure.

Material and methods

Study area. Object of the study were four waterbodies from Stara Zagora region, South Bulgaria – three rivers (Tundzha River - upper reaches of the river basin, Bedechka River and Sazliyska River) and a dam – Jrebchevo Dam (Figure 1). Tundzha River is the second longest river in Bulgaria and it is important for the economy of the country – agriculture, industry, energy and other activities (RBMP, 2010). The river is subjected to strong anthropogenic impact as it passes through big settlements, industrial enterprises, farms and areas with intensive agriculture of which wastewater is discharged into it and creates conditions for deterioration of the water quality. The other two rivers (Bedechka River and Sazliyska River) have only local significance. In them wastewater from many anthropogenic activities is discharged and their waters are used primarily for irrigation of crops. Jrebchevo Dam has capacity of 400 mln.m^3 water, which is used for irrigation, electricity generation and pisciculture.

Monitoring points. 6 monitoring points were set for screening purposes in accordance with Regulation No 5/2007 and Regulation No 13/2007, as follows:

- Monitoring Point 1 (MP1) – Bedechka River, Stara Zagora Municipality (N42.27049° E25.37937°);
- Monitoring Point 2 (MP2) – Sazliyska River, Stara Zagora Municipality (N42.26914° E25.29015°);
- Monitoring Point 3 (MP3) – Tundzha River at Yagoda village, Maglizh Municipality (N42.32740° E25.3380°);
- Monitoring Point 4 (MP4) – Tundzha River at Jrebchevo Dam, Nikolaevo Municipality (N42.38333° E25.49350°);
- Monitoring Point 5 (MP5) – Jrebchevo Dam, Nova Zagora Municipality (N42.35346° E25.57020°);
- Monitoring Point 6 (MP6) – Tundzha River, Banya Village, Nova Zagora Municipality (N42.36243° E25.59466°).

Investigated indicators. In water, sediment and alges samples, collected from the MP of the studied waterbodies, the element Mn was investigated.

Samples collected. Water: Seventy-two water samples were collected from MPs of the studied waterbodies each month during one year (from November 23, 2009 to November 23, 2010) in

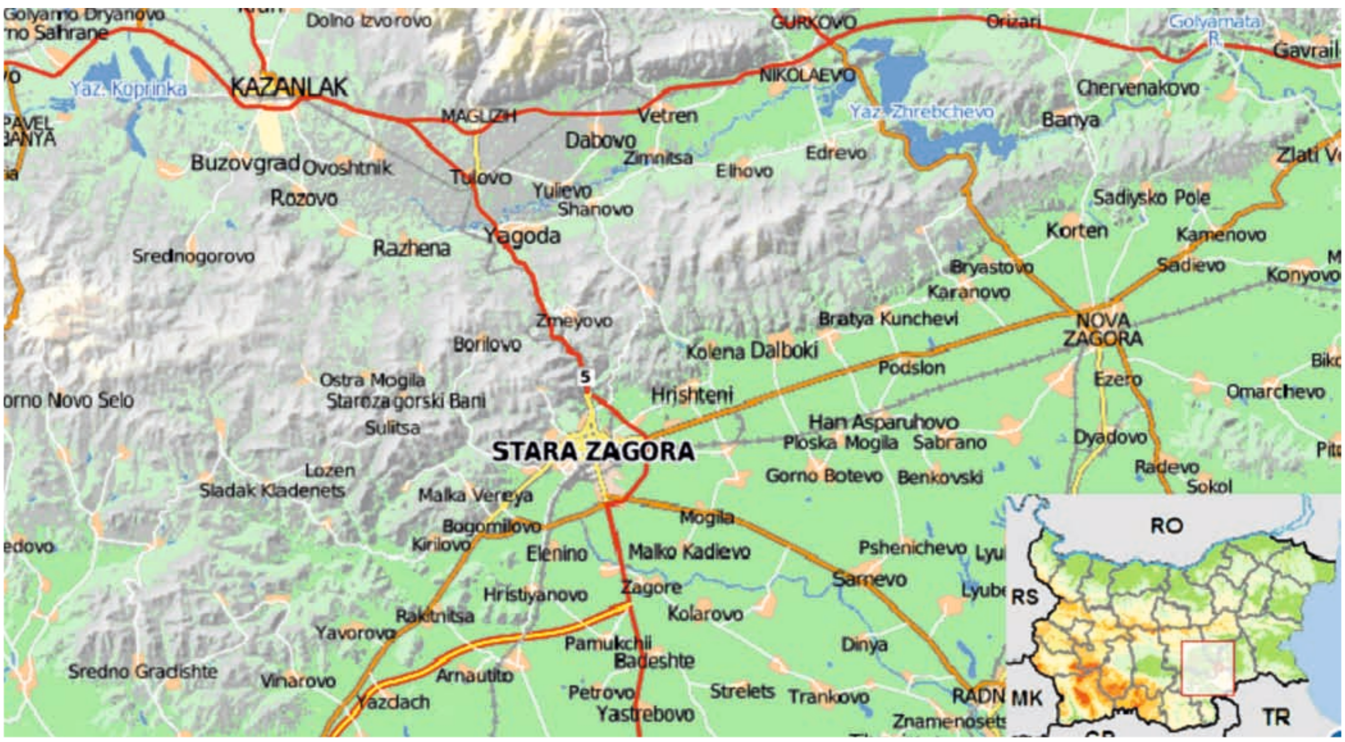

Figure 1. Map of Stara Zagora region

accordance with the requirements of BS EN ISO 5667 – 1/2007. The water samples were stored in accordance with BS EN ISO 5667 – 3/2006.

Sediment: Six sediment samples, collected from the studied waterbodies were prepared, archived, stored and analyzed for the period from May to December 2010.

Algae: A total of six algae samples, collected from the studied waterbodies were prepared, archived, stored and analyzed for the period from May to December 2010.

Methods for analysis. Water, sediment and algae samples were analyzed in the laboratories of the Research Scientific Center for Environment at the Faculty of Agriculture, Trakia University, Stara Zagora.

The content of the studied heavy metal Mn in water, sediment and algae samples was determined on atomic absorption spectrophotometer (AAS) "A Analyst 800" - Perkin Elmer. Analyses for manganese in surface water samples were performed in graphite tube or flame (depending on the concentration of these elements), at a definite wavelength and water preservation in advance of the samples with 5 cm^3 k.HNO_3 of a sample (ISO 8288, BS EN ISO 5667-3/2006). The contents of Mn in water samples were measured in mg/L.

The delivered specimens of sediment, algae and aquatic plants are lyophilized to constant weight. The whole amount of dried sediment was ground and weeds out repeatedly to fine powder. So an average representative sample of not less than 20 grams was received. Decomposition with concentrated hydrochloric and nitric acids of sediment samples was carried out in accordance with ISO Standard 11466. It was followed by filtration and dilution of samples to 50 ml with distilled water.

The test samples of algae were prepared for the analysis by wet combustion in a microwave oven Perkin Elmer Multiwave 3000. The extracts were extended up to 25 mL with distilled water. The final metal concentrations in the acid solutions were amended of AAS in accordance with BSS ISO 11047. The concentrations of the investigated elements of sediment and algae were expressed as mg/kg dry weight.

The instrument was periodically calibrated with standard chemical solutions prepared from commercially available chemicals (Merck, Germany). An air-acetylene flame and hollow cathode lamp for all samples were used. Calibration curves were prepared using dilutions of stock solutions. The samples (water, sediment and algae) were registered three times and the mean values were calculated.

Assessment of the Mn levels in the investigated components. Ecological assessment of the quality of surface water from the studied waterbodies was carried out on the basis of limit value of Mn concentration, stipulated in Regulation No 7/1986. Quality assessment of Mn content in sediment and algae samples was not done, because there are no standards for this element in the Bulgarian legislation. To this end, a comparative analysis of the data was made between the manganese content in the sediment and algae from the studied waterbodies.

Statistical analysis. Statistical processing of the results was computed by the program STATISTICA using ANOVA test.

Results and discussion

The modern idea of a comprehensive monitoring of the hydroecosystems includes tracking of certain parameters both in water and in hydrobionts inhabiting it (Atanasov et al., 2012). Postponed sediments in freshwater ecosystems adsorb heavy metals (including Mn) and decrease their concentrations in water. In this connection it is absolutely necessary to determine the quantity of the studied heavy metal in water first, and then in sediment and algae.

The composition of the sediment (mud) is extremely indicative of the history of a particular waterbody. Postponed in time debris could illustrate even temporary pollution with short and infrequent peak concentrations. In this respect the existence and concentration of a specific pollutant "seal" the past of a water hydro ecosystem. Moreover, the mud is very suitable for habitation of many organisms, giving rise to the food chain, which affects higher hydrobionts.

Our results about concentrations of the heavy metal Mn in water, sediment and algae from the investigated waterbodies are shown in Figures 2, 3 and 4.

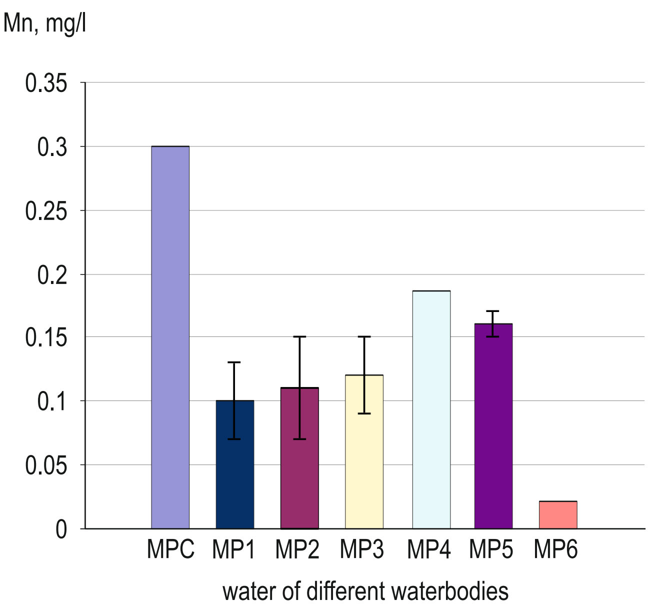

Figure. 2. Mn content in water from different waterbodies

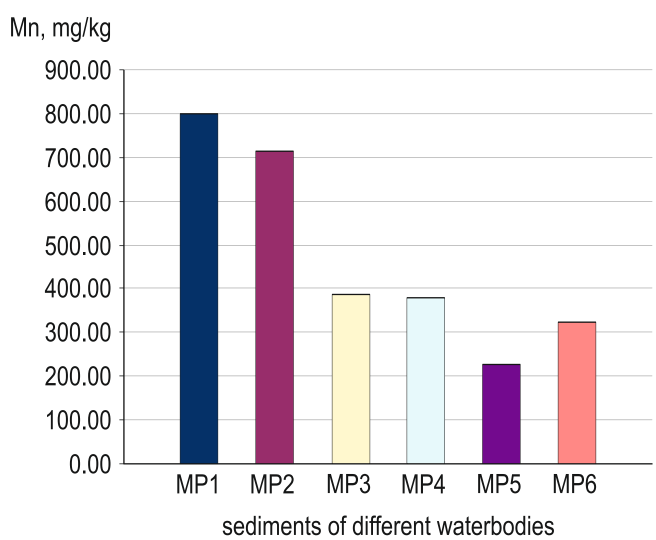

Figure 3. Mn content in sediment from different waterbodies

Mn levels ranged from 0.021 mg/l in water from MP6 to 0.186 mg/l in water from MP4 (Figure 2). The highest concentrations of Mn in MP4 can be explained by the fact that up to this MP on Tundzha River wastewater from different sources – WWTP of settlements, industrial enterprises, agricultural activities and probably by mining in the region of Tvarditsa town flow into it. All this leads to an increase of the concentration of this element in water up to high values. After it runs into the dam, the water is diluted and stays for a long time in it. Because of that, part of the Mn is absorbed by sediment and algae and thus its concentration in water decreases (MP5). When water runs out of the dam (MP6) it contains less Mn, as it is largely self-purified. Jrebchevo Dam serves as a "settler" of heavy metals (and possibly other pollutants), in this way purifying the Tundzha River. Intermediate significance had the values of Mn in MP1 (Bedechka River), MP2 (Sazliyska River) and MP3 (Tundzha River at Ygoda village). These results give reason to believe that anthropogenic impacts in these points (1, 2, 3 and 6) are less than in MP4 and MP5. Ecological assessment showed that water from all MPs meet the requirements for Mn content for second category water (in that category are all investigated waterbodies) according to Regulation No 7/1986. The values established were much lower than the maximum permissible concentration – 0.3 mg/l.

Mn content in the sediment varied from 225 mg/kg in Jrebchevo Dam (MP5) to 800 mg/kg in Bedechka River (MP1) (Figure 3). Data analysis showed that by this indicator MPs can be divided into two groups – MPs with high concentrations (MP1 and MP2) and MPs with low concentrations (MP3, MP4, MP5 and MP6). This suggests the existence of similar processes of absorption of Mn by sediment, specific to these two groups of waterbodies. We assume that the reasons for accumulation of large amounts of Mn in the sediment of the rivers Bedechka (MP1) and Sazliyska (MP2) are as follows: both rivers have little water flow, low flow velocity, which referred to the relatively large area occupied by sediment allows for complete absorption of Mn by the water. Future research will show whether it is reasonably surmised.

Comparing the data of Mn content in the water and sediment in the monitored points reveals a controversial trend. In MPs where Mn levels in water are relatively low (MP1, MP2 and MP6) in the sediment of these points the levels are higher. In two of MPs (MP4 and MP5) the content of Mn in water corresponds to that in the sediment, i.e. the level of Mn is relatively higher than in the sediment. In MP3 a clear relationship between the content of Mn in the water and sediment is not observed. The results do not give reason to draw sweeping conclusions about the processes of absorption of Mn by water into the sediment of waterbodies. It is necessary to receive further information in this regard. Ecological assessment of sediment on Mn content cannot be done because in the world and the country there is no standard for this indicator. In Bulgarian standard – Regulation No 3 of 10 August 2008 for limit values of content of harmful substances in the soils that element also lacks.

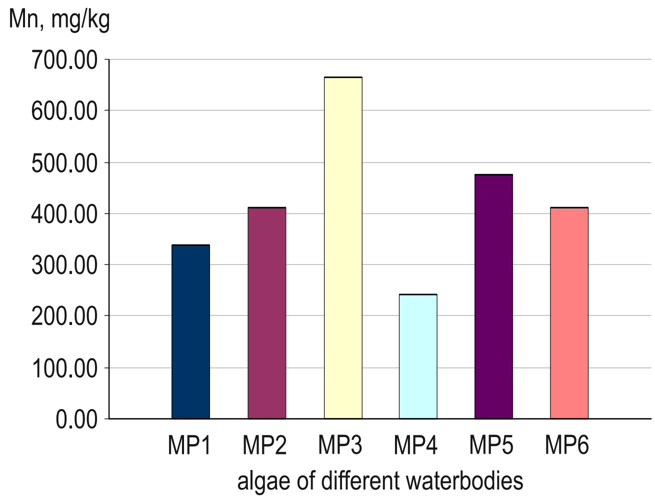

Figure 4. Manganese content in algae from different waterbodies

The exported data on Figure 4 show the highest value of Mn in samples of Tundzha River, Yagoda Village (MP3 – 664 mg/kg). Algae delivered by Tundzha River, Jrebchevo Dam (MP4) contain the lowest concentrations of this heavy metal (242 mg/kg). The results reveal significant variation on this indicator between different waterbodies. This suggests an impact of specific factors for the deposition of Mn in algae for each water body.

Comparing the results for Mn content in algae and sediment from the investigated waterbodies revealed different trends of change. The content of Mn was greater in sediment (mud) compared to that in algae at MP1, MP2 and MP4 (see Figures 3 and 4). In the remaining MPs (3, 5, and 6) an inverse relationship was observed – higher content of Mn in algae compared with that in sediment. These results give no reason to seek a direct relationship between the content of Mn in sediment and algae. A research in this direction has to continue.

Despite the large diversity of data for Mn content in water, sediment and algae it is necessary to determine Mn content ratio between these three components for a water body (Table 1). The data shows that Mn is accumulated from 1407 (MP5) to 15466 (MP-6) times more in the sediment compared to the water. The estimated ratios between Mn concentrations in algae and water have shown

Table 1. Mn content ratio between sediment/water, sediment/algae and algae/water of investigated waterbodies

Monitoring Points	Coefficient of Mn concentration (sediment/water)	Coefficient of Mn concentration (sediment/algae)	Coefficient of Mn concentration (algae/water)
MP1	7996	2.4	3395
MP2	6495	1.7	3741
MP3	3234	0.6	5531
MP4	2032	1.6	1301
MP5	1407	0.5	2975
MP6	15466	0.8	19565

that the accumulation of this metal in the algae is from 1301 (MP-4) to 19565 (MP-6) times more than the water. This fact suggests available mechanisms, probably different by simple diffusion of Mn between water-sediment and water-algae. Much different were the values for the ratio between the Mn content in sediment and algae. By the sediment/algae ratio it was found that Mn is accumulated from 0.5 (MP-5) to 2.4 (MP-1) times more in sediment compared to algae.

The analysis of results on the chain water – sediment – algae (Figures 2, 3 and 4) indicates multiple lower levels of Mn in water samples compared to sediment and algae. Despite the differences in values between Mn content in sediment and algae, and the two components Mn values are relatively comparable and of similar meaning, i.e. both sediment and algae have the ability to accumulate large amounts of manganese from water. All this gives reason to indicate that sediment and algae can serve as good indicators of pollution by Mn, because these two components have similar ability to accumulate that metal from water. They can also be used for purification of water from that metal.

In particular, very recent data in many studies have shown that algae and aquatic plants are autotrophic organisms located at the base of the food chain in each hydroecosystem (Strezov and Nonova, 2009). These organisms, extract their necessary organic and inorganic substances from the sediment of the waterbody, accumulate them in the body and in this way a relatively accurate picture of potential pollution for a certain past period of time can be given. In this aspect after studies of soil content of heavy metals in the studied points it is reasonable to make such specimens of algae and aquatic plants. In Europe many studies have been conducted using algae as bioindicators of heavy metal contamination (Conti and Cecchetti, 2003). The species of *Enteromorpha* and *Cladophora* are known to grow in freshwaters and marine habitats (Marsden and De Wreede, 2000; Storelli et al., 2001; Daka, 2005; Zbikowski et al., 2007; Brodie et al, 2007) and they are used all over the world as indicators of heavy metal pollution in both habitats (Whitton, 1984; Sawidis et al., 2001; Villares et al., 2002; Topcuoglu et al., 2003; Gosavi et al., 2004; Stengel et al., 2004; Al-Homaidan, 2007; Strezov and Nonova, 2009).

Conclusion

The highest levels of heavy metal Mn were found in the water of hydroecosystems with the highest anthropogenic impact: Tundzha River; Jrebchevo Dam (MP4); Jrebchevo Dam (MP5) and Tundzha River, after Jrebchevo Dam (MP6). But in all water samples the maximum permissible concentration (MPC) for second category waterbodies was not reached, to which group the investigated waterbodies belong. The highest Mn concentrations in sediment were measured in Sazliyska River (MP2) and Bedechka River (MP1). Deposits accumulated in time could illustrate even temporary pollution with short and rare peak concentrations, sealing the past of the water hydro-ecosystem. In this connection, sediment can serve as a good indicator of pollution of a particular water body. It is clear that sediment plays a role of a soil topping food chain in aquatic ecosystems. Algae extract Mn from the sediment of the water body, accumulate it and thereby is a relatively accurate picture of potential pollution for a particular past period of time can be given. As a result of this fact after study of the primer similar ones were made in samples of algae. With the highest levels of Mn algae delivered by Tundzha River, Yagoda village (MP3); Jrebchevo Dam (MP5) and Sazliyka River (MP2) have been distinguished. The accumulation of Mn in the sediment is from 1407 (MP5) to 15466 (MP6) times more than in water. This element is accumulated from 0.5 (MP5) to 2.4 (MP1) times more in sediment compared to algae and from 1301 (MP4) to 19565 (MP6) times more in algae compared to water. The results indicate that sediment and algae can serve as good indicators of pollution by Mn, because both components have similar ability to accumulate that metal from water. They can also be used for purification of water from that metal.

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