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Original Research

Manganese Fractions in Waters of the Polyhumic Siemianówka Dam Reservoir

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Abstract

Hydrochemical studies were performed from May to December 2004 at four stations in the polymictic, polyhumic Siemianówka Dam Reservoir (SDR) located on the upper Narew River in northeastern Poland. The total manganese (TMn) concentrations in the reservoir waters varied from 118 $\mu g/dm^3$ to 638 $\mu g/dm^3$, and their increased values are caused by forming complexes with organic matter present in waters flowing from wetlands and forests. Until the moment when the maximum manganese concentration occurred in the reservoir, its upper part was characterized by the higher manganese concentration than the lower part. A long retention time of water delays the occurrence of total manganese maximum in the lower part in SDR. Maximal TMn concentrations were noted in the lower part of the reservoir in the end of summer, while minimal for the upper part in winter. The organic dissolved manganese fractions (ODMn), were the largest part of TMn, slightly less was the particular manganese (PMn) fraction, and the smallest part constituted the reactive dissolved manganese fraction (RDMn). Low concentrations of reactive manganese in winter are due to a low pH value and low concentrations of dissolved organic matter (DOC). Manganese sorption on colloids increases in summer and is accompanied by a pH increase during an intensive photosynthesis of algae and cyanoprocaryota.

Keywords: manganese, fraction, water, dam reservoir, DOC

Introduction

Chemical elements in various forms, phases and oxidation states occur in water. Manganese is an element that forms numerous compounds at various oxidation states (5 typical oxidation states). Kabata-Pendias [1] indicates that in a hypergenic environment as well as in soils, even 14 ionic forms of manganese – at the oxidation states from +II to + VII – can be present. The oxidation state of this metal depends mainly on the oxygen content in water, on reduction-oxidation conditions and on the pH value of water. In reduction conditions and when there is a lack of oxygen only ionic forms, at the lowest oxidation states, occur.

Biogeochemical studies concerning manganese are not numerous and they are mainly related to sediments or technological problems of water treatment. Other information on manganese in limnic and lotic ecosystems are dispersed in the references. Fractional structure of manganese in surface waters can constitute an important feature of chemical environment decisive for plants – such as algae, cyanoprocaryota and macrophytes – growth. Manganese is very important for growth of algae and cyanophyta. This trace element plays an important role in the photosynthesis process. It is called the special task element since it accumulates in enzymes. Manganese constitutes an important building element of such enzymes as: acidic peroxide dismutases, glycoside phosphatases, and active proteins in the photosystem II. This element also influences reduction of

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nitrates (V) in plants, hydrolysis of peptides and amides (peptidases) as well as urea (erginases) [2].

Part of manganese in surface waters originates from the catchment. A relief, climatic conditions, vegetation and soil cover types are also agents influencing the manganese concentration in soils [3]. On average, the manganese concentration is in the range from 100 to 1300 mg/kg [1]. Sandy soils are leaner than heavy ones formed on clays. However, organic soils are significantly richer in manganese as compared with mineral soils.

Manganese deposition in bottoms of lakes and rivers occurs mostly in the form of insoluble oxides, which – after a long time – gather in ferromanganese concretions. A total manganese concentration in bottoms is usually quite high, e.g. in the Rein River it equals 960 ppm and in the Odra River 770 ppm [4].

Chemical changes of manganese in water ecosystems are very complex, mainly depending on reduction-oxidation conditions. Due to this, manganese influences the performance of other metals (especially trace ones) since it (together with iron) causes their bond. The ferromanganese concretions mentioned above, containing active manganese forms, immobilize cobalt, copper and nickel. On the other hand, an increased concentration of manganese, at the second oxidation state, increases migration of calcium and magnesium [5]. Regardless of pH of the environment manganese often occurs as Mn^{2+}, but in oxidizing conditions this reactive form precipitates and sediments very fast down the reservoirs [1].

The aim of our study is to present the seasonal changes of the manganese fraction concentration in the lowland reservoir dam of a long retention time and of an increased concentration of organic compounds. Another aim of our paper is to determine the relationships between manganese fractions in water and other physicochemical parameters influencing bioaviability of this element for aquatic autotrophic plants.

Study Area and Methods

The object under testing was the Siemianówka Dam Reservoir (SDR), which was filled with water for the first time in spring 1990. It is situated in northeastern Poland in the Upper Narew valley. The Narew River flowing from Belarus into Poland has a variable width of the valley, from 0.8 to 4 km, with the riverbed in the direction SE–NW. The SDR locations was prefered as the most advantageous one out of four locations considered initially as the potential places for building the dam on the Narew River valley [6].

The investigated reservoir bottom is covered by alluvial and peat deposits from the Holocene, while at larger depths are the Pleistocene sands of thickness of strata up to 20m, under which boulder clays are situated. The thickness of peat with sand layers deposited on the bottom approaches 1.7 m [7]. The valley gently slopes are covered solely by sandy-clay sediments.

Table 1. Siemianówka Dam Reservoir characteristics [7].

feature	water level	
	minimum	maximum
Total volume (mln m^3)	17.5	79.5
Usefull volume (mln m^3)	0	62
Area (km^2)	11.7	32.5
Depth max (m)	6.0	9.0
Depth average (m)	1.5	2.5
Hight of damming (m)	4.1	7.0
Water exchange time in year	1	2 – 3

The SDR is shallow, polymictic, with a large water capacity (Table 1). As far as its surface is concerned, it is the 12th largest reservoir in Poland. During the exploitation significant changes of the Reservoir surface occur, up to 60% of its area, and the average depth varies from 2.5 to 1m [7]. The deepest part of the reservoir is near the front of the dam. The reservoir is 70% supplied by the Narew River, while the remaining 30% is delivered by 5 small streams and waters pumped from drainage ditches from places located on lower levels and separated from the Reservoir by five side dams. Kołonka River and Pszczółka River directly enter the reservoir, while Cisówka River, Łuplanka River and Podrzeczka River via the intermediate pumping stations. Basin of the Narew River in the profile of the dam face covers an area of 1050 km^2, while the average annual flow discharge about 5.8 m^3/s. The average annual discharge calculated − from the period 1964-1988 – for the Kołonka and Pszczółka Rivers are 1.16 m^3/s and 0.15 m^3/s, respectively [8].

Hydrochemical investigations were performed from May to December 2004 at four stations on reservoir. At one of them water samples were taken from depths of: 0.5, 2 and 4 meters using a bathometr, while at the remaining three stations the samples were taken from the surface layer. Temperature, electrolytic conductivity and water pH values were measured in situ. Oxygen concentrations and water oxygen saturation were measured by oxygen sonde in situ too.

Laboratory analyzes were performed according to methods described by Hermanowicz et al. [9]. Bicarbonates were analyzed by the acid-base titration method, the total hardness and calcium by complexometric titration, while the amount of magnesium was calculated from the difference between the total hardness and calcium concentration. Remaining ions, it means: total iron, nitrates (V), sulphates (VI), chlorides, silicates and ammonium ions were determined by spectrophotometric methods with the application of procedures and reagents recommended by the Riedel de Häen Company. Chlorophyll "a" concentration was determined in the laboratory by the spectrophotometric method after hot extraction by 90% ethanol. In addition, 3 fractions of phosphorus: TP, DP and SRP were detected by the spectrometric molybdenium method, while two remaining fractions were calculated (PP and ODP). The dissolved organic carbon content (DOC) was determined in the analyzer of organic carbon TOC – 5050A (Shimadzu prod.) according to the procedure developed by Zieliński and Górniak [10]. Concentrations of 3 manganese fractions, it means: total manganese (TMn), dissolved (DMn) and reactive manganese (RDMn) were determined by the spectrophotometric formaldoxime method with their own modifications [11]. Reactive manganese (RDMn) was analyzed by added standard method. Concentrations of the first two fractions of manganese were assayed in water samples undergoing the UV mineralization with concentrated sulphuric acid (VI) and 30% hydrogen peroxide. Total manganese (TMn) was determined in unfiltered water, after mineralization, while dissolved manganese (DMn) in water filtered by the filter paper "GF/C" of a pore diameter 0.45 μm, also after mineralization. Reactive dissolved manganese (RDMn) was determined directly in water filtered by the filter paper "GF/C" but without mineralization. Other manganese fractions, in particular manganese (PMn) and organic dissolved manganese (ODMn), were calculated – analogically as in the case of phosphorus – in the following way:

ODMn = DMn – RDMn, PMn = TMn – DMn

Results and Discussion

Average concentrations of manganese fractions in the Siemianówka Dam Reservoir in 2004 were as follows: TMn – 367.9 μg/dm^3, DMn – 268.1 μg/dm^3, RDMn – 66.8 μg/dm^3, PMn – 91.4 μg/dm^3, and ODMn – 181.7 μg/dm^3. The lowest concentrations of total manganese (TMn) occurred in early winter – 118.6 μg/dm^3 and were 5 times lower than the maximum value detected in the second half of August (Fig. 1A). Concentrations of total manganese were slightly decreasing from the upper part, via the middle, to the lower part of the reservoir. However, the average difference being only 19.4 μg/dm^3 was not significant statistically. High manganese concentrations in the reservoir can be explained by the type of river basin consisting of peatlands and forests and by manganese-rich bottom deposits. Polymictic water in the ice-free period provides constant supply of manganese and other substances from bottom deposits.

The dissolved manganese fraction had the lowest concentration in May (not in December as in the case of total manganese), while the highest value was in the second half of August and was 6 times higher than the lowest concentration of this fraction (Fig. 1B). During the major time of study, the highest concentration of dissolved manganese was detected in the lower part of the reservoir. Dissolved manganese constituted 79% of TMn and varied slightly more in concentrations than the fraction of total manganese. However, statistically significant differences in variations of dissolved manganese in the specific parts of the reservoir were not found.

The range of reactive manganese concentration was from 9.7 μg/dm^3 (minimum) to 142.3 μg/dm^3 (maximum) and usually was the lowest out of all analyzed manganese fractions (Fig. 1C). Minimal concentrations were found in May, while maximal in August, thus similarly to the dissolved manganese fraction. The highest concentration was measured in the upper part of the reservoir during the majority of the season (Fig. 2). The reactive manganese concentration increases slowly up to August and then successively falls down. In the case of total manganese its violent increase occurs in May-June followed by the stagnation period and abrupt fall at the turn of August and September.

Regardless of the season, ODMn constituted the highest share in TMn concentration. Spring time was characterised by the high percentage of PMn and ODMn, 41% and 47% of TMn − respectively. This was related to the spring surface flow, which carried into the reservoir significant amounts of manganese complexed with organic and inorganic materials. During the summer time the participation of the dissolved organic fraction (ODMn) increased at the expense of the particular manganese fraction (PMn), participation of which decreased by 50% in comparison to the spring period and obtained the minimum value of 19%. The dissolved organic manganese fraction the highest participation in TMn achieved in summer (60%), when there was a large spatial variation of this ion concentration (on average 3.5 times). The smallest variations of the dissolved organic manganese fraction occurred in the spring-winter period. The highest percentage of the reactive manganese fraction (RDMn) was during winter – 30%, (this period was characterised by a small variation coefficient (V = 20%)), while the lowest was during spring – 10%. Summer and autumn were characterized by small variability of the RDMn participation in TMn. The most stable, when the variability in the participation in TMn in the reservoir is taken into account, was the particular manganese fraction (Fig. 4). Differentiation of the participation of the reactive fraction gradually decreased in the direction of the dam face, reaching in its vicinity the variability coefficient 3 times lower than in the upper part of the Reservoir (91.9%).

Maximum concentration values of nearly all fractions (with the exception of PMn) in the upper part of the reservoir occurred in the first half of August, while in the lower part in the first half of September. This was caused by an intensive growth of cyanoprocaryota in the reservoir [26] in this time of the year. The Siemianówka Reservoir delays the time of the occurrence of the maximum manganese concentration in waters of the Narew River below the SDR. This delay time between the upper and the lower part of the reservoir depends on hydrologic conditions. Until the time when the maximum manganese concentration occurred in the reservoir its upper part had a higher manganese concentration than the lower part. However, after reaching the maximum, the manganese concentration in the lower part of the reservoir was higher than in the upper part. It was probably related to the summer denudation of manganese in the catchment and transferring of the free charge into the reservoir.

Relatively large number of forms of organic phosphorus and seston nitrogen were observed in the Siemianówka Dam Reservoir [26], which indicates intensive phytoplankton growth. Massive water blooming, which happened at

Table 2. Manganese fractions (µg/dm^3) differentiation in profile of the Siemianówka Dam Reservoir in 2004.

fractions	0.5 m	2 m	4 m	average
TMn	353.1 ± 186.3	390.7±205.8	353.0 ± 210.2	365.6 ± 89.0
DMn	258.4 ± 160.1	256.2 ± 160.6	268.6 ± 172.6	261.1 ± 151.2
RDMn	78.3 ± 51.0	77.0 ± 47.8	68.3 ± 46.6	74.5 ± 47.8
PMn	94.7 ± 101.9	134.5 ± 108.5	84.4 ± 85.1	104.5 ± 84.7
ODMn	180.1 ± 114.4	179.3 ± 127.1	200.3 ± 148.6	186.6 ± 114.1

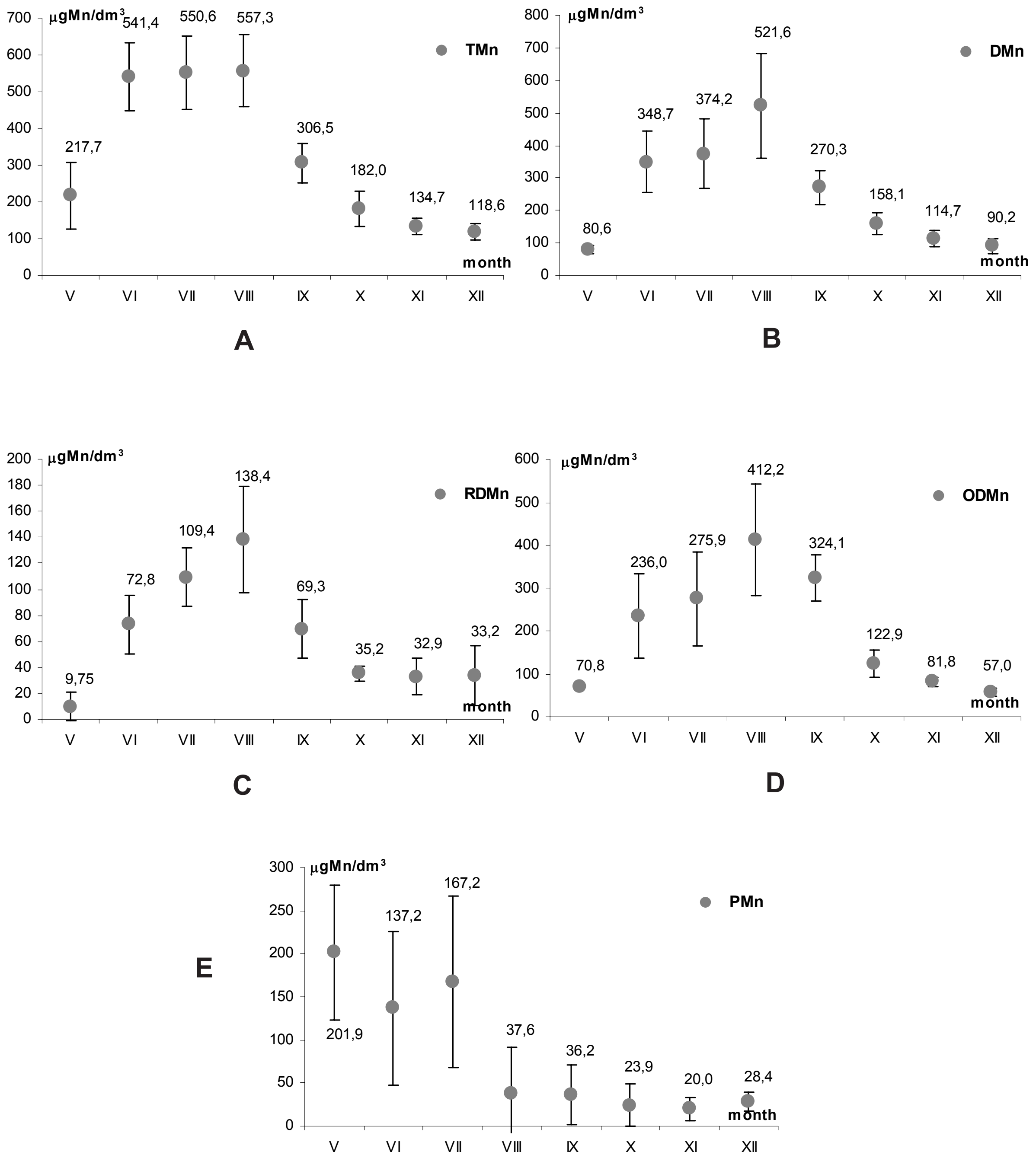

Fig. 1. Manganese fractions concentrations in the Siemianówka Dam Reservior in 2004.
A – TMn, B – DMn, C – RDMn, D – ODMn, E – PMn

the turn of August and September in 2004, caused an increase of the dissolved organic compounds concentration (DOC) in water. During the 2004 vegetation season, the chlorophyll "a" concentration in the reservoir was nearly 500 μg/dm^3, but it was lower than the record one measured in 2000 [26]. Concentration of the reactive manganese (RDMn) and the chlorophyll "a" concentration are significantly correlated (Fig. 5B).

Another parameter which influences the manganese concentration is phosphorus. In natural conditions manganese does not occur dissolved in water in significant amounts since it precipitates fast and turns out into an insoluble form [3]. High phosphorus (phosphates (V)) concentrations cause precipitation of manganese (II) in a form of manganese (II) phosphate (V). Due to a decreased concentration of one of the soluble forms of those elements their liberation from the insoluble compounds occurs, which is documented by the statistically significant correlation between TMn and TP and $r^2 = 0.57$, $p<0.001$ (Fig. 5A). At higher concentrations of phosphorus (of the order of 200 μg/dm^3) total manganese concentration decreases, which means that the process analogical to the one between reactive manganese and phosphates (V) takes place. This process probably constitutes the defence of ecosystems against an excessive increase of manganese concentration. Extreme manganese concentrations in waters might be the cause of chlorosis in plants or permanent disturbances in animal organisms (haemolysis of erythrocytes, changes in the DNA chain structure) [12, 13]. The manganese concentration (and other ions) in water depends not only on the season of the year and the presence of other substances or conditions, but also on the depth of water layer. The highest concentration of dissolved manganese was found in the surface layer while reactive dissolved manganese was found near the bottom of the SDR (Table 2).

From the very beginning the reservoir was filled with fertile waters, due to which an intensive growth of phytoplankton occurred [14] and corresponding to it low water transparency was observed [15]. In natural eutrophic lakes as well as in the reservoir under study high contents of organic matter in water were detected. The periodical water deoxygenation in the lower part of the reservoir (near the front of the dam) was observed. On the other hand, the Siemianówka Reservoir has advantageous morphologic conditions, which should influence permanent water oxygenation. The large water area in proportion to the total water mass and small average depth facilitate water mixing and in consequence the oxygen penetration from the atmosphere into water – during the major part of the year [8]. Due to the polymicticity of the investigated reservoir its waters are fast warming at high summer temperatures and fast cooling at low autumn temperatures. In consequence there are diurnal variations of the dissolved oxygen concentration in water, from a deep oxygen deficit to a significant supersaturation [16]. At the total saturation with oxygen (e.g. in winter) the reactive manganese fraction (RDMn) dominated in the reservoir water, while at the low concentration of oxygen – the organic dissolved fraction (ODMn). An intensive growth of microorganisms of the summer time causing inflow and decomposition of organic matter with significant oxygen consumption leads to an increase of the organic dissolved fraction (ODMn) (Fig. 5C). On the other side, wind actions reduces periodic water hypoxia corresponding to changes of dissolved manganese. When, after the period of hypoxia, an increase of oxygen in water occurs, Mn (II) changes into Mn (III) and (IV), which results as precipitation of manganese oxides and hydroxides. In effect the dominating fractions in SDR water are PMn and ODMn. With water temperature decreasing in late autumn and winter, the microbiological decomposition of organic matter slows down, thus decreasing the demand for Mn^{2+} of phytoplankton. Therefore, the RDMn fraction starts to dominate other manganese fractions. Periodic oxygen deficits in the SDR waters under the ice cover liberate additionally reactive manganese from precipitates such as e.g. MnO_2 or Mn_2O_3 remaining from the summer time on the surface of sediments. Galicka studies [17], who found the maximum concentration of reactive manganese (150 μg/dm^3) in the Sulejowski Reservoir in winter periods, confirm our findings. It should be mentioned that the manganese content in bottom sediments of the Sulejowski Dam Reservoir varied from 0.004-0.83 mg/g of dry matter [17] – in 1988-1990, while the bottom depostis in 2004 in the Siemianówka Reservoir contained much higher amounts, varied from 0.01-1.04 mg/g of dry matter [18].

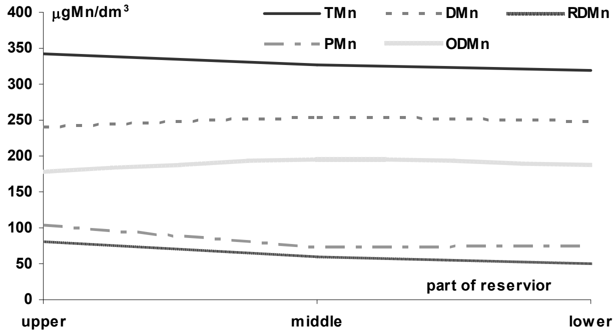

Fig. 2. Space differention of manganese fractions in Siemianówka Dam Reservior waters in 2004.

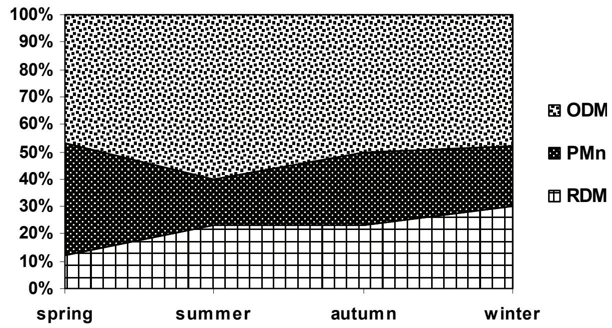

Fig. 3. Seasonal changes in manganese fractions in water of Siemianówka Dam Reservoir in 2004.

Table 3. Manganese concentrations in different water types.

Reservoir	[µg/dm^3]	manganese fractions	source
George lake	1500–2200	disolved total manganese	[21]
Lumsden lake	300–1700		
Plastik lake	1400–2200		
Crosson lake	300–500		
Harp lake	1750–3300		
Blue Chalk lake	250–2500		
Red Chalk lake	750–1300		
Biwa lake	1.5–240	reactive dissolved manganese	[22]
Aha lake	0–3000		[23]
Pine reservoir	400–1400		[24]
Adriatic Sea	0–175		[25]

In the Polish dam reservoirs (e.g. the Dobczycki Reservoir) TMn concentration range was 10-2200 µg/dm^3 [19], while in the Zegrzyński Reservoir it was only from 0 to 400 µg/dm^3 in 1981-88 [20]. Thus, it can be stated that – as far as the manganese concentrations are concerned – the reservoir under testing does not differ much from other reservoir dams and other types of analyzed limnic waters (Table 3).

In general, one can state that the Siemianówka Dam Reservoir caused an increase of higher TMn concentrations in the lower part than in the upper part, which is the effect of a significant high autochthonic, organic matter production. These organics are forming various coordination compounds with manganese causing it to exist in non-reactive forms. A significant percentage of the reactive manganese fraction (RDMn) in winter is the effect of water conditions (low pH value, small amount of DOC). The increased manganese concentration observed in summer is the result of washing this element out from bottom deposits due to which a part of washed out ions is coordinated by organic matter (ODMn), while the rest occurs in a dissolved form (RDMn). The observed increase of the reactive fraction concentration – after water passing via the reservoir - is the result of diminishing organic matter contents in water, which means that complexes are not formed and manganese is present in unbonded form. An increased manganese adsorption on colloidal particles is caused by an increased pH of water, which in turn is caused by the consumption of carbon dioxide (IV) in photosynthesis processes and in effect leads to the decrease of the reactive manganese concentration. Sedimentation of manganese particles on the reservoir bottom, being an effect of turbulent movements causes decreasing of the particular manganese concentration.

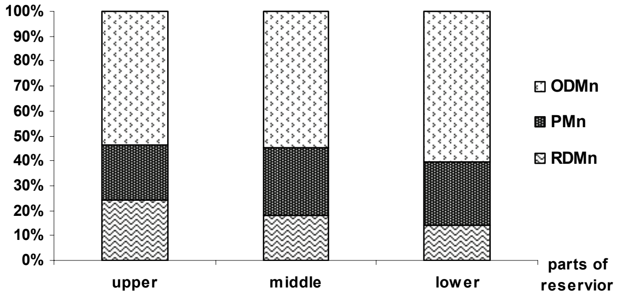

Fig. 4. Manganese fraction differentations in Siemianówka Dam Reservoir in 2004.

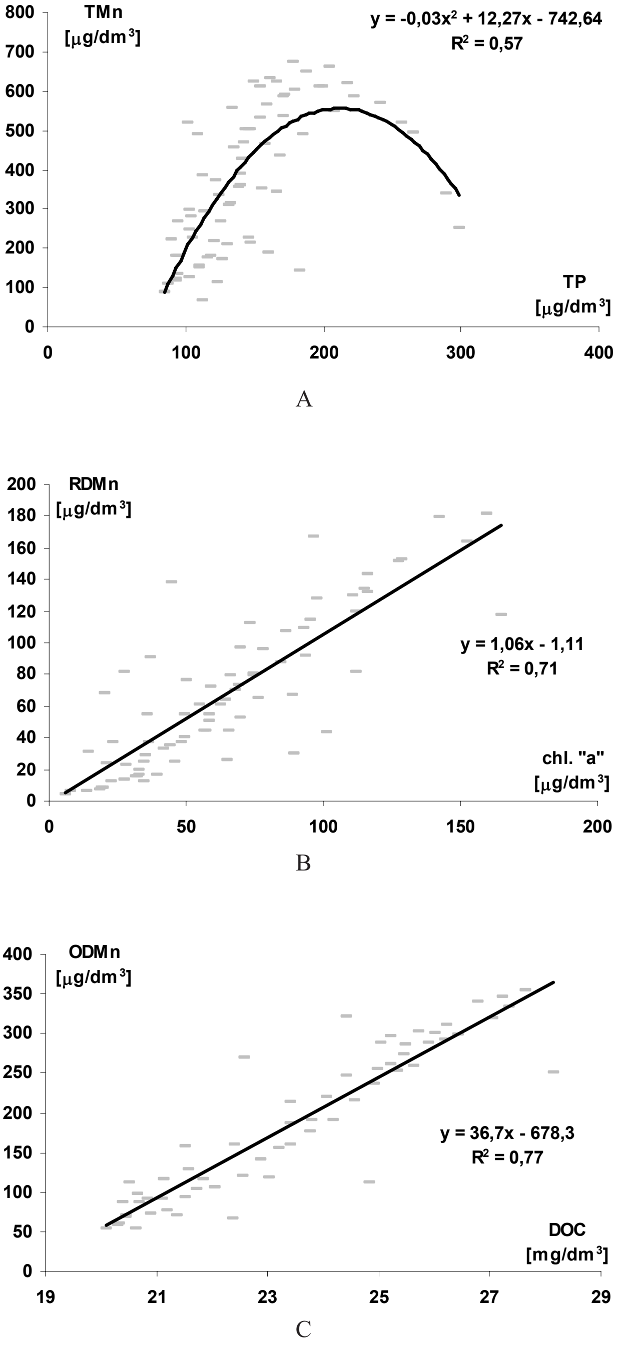

Fig. 5. Correlations between manganese fractions and chemical parameters of water in Siemianówka Dam Reservoir in 2004.

Conclusions

1. The range of total manganese concentration in the Siemianówka Dam Reservoir in 2004 was within the range 119-638 µg/dm^3, similarly to other artificial and natural lakes in Poland.
2. The largest part of total manganese constituted the organic dissolved manganese fractions (ODMn), slightly smaller part the particular manganese fractions (PMn), and the smallest: the reactive dissolved fractions (RDMn). Domination of ODMn over other manganese fractions was constant. Only during spring did the PMn fraction dominate RDMn, while the participation of this last one gradually increased from spring to winter.
3. The manganese resources in the SDR depends on the spring supply by the Narew River basin in the particular and organic fractions, as well as on the internal resuspension processes from plant detritus.
4. Low RDMn concentrations in winter are the effect of lowering the DOC concentrations and lowering the pH value, while in summer significant amounts of the reactive manganese fractions undergo sedimentation on the reservoir bottom.

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