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RESEARCH ARTICLE

Internal phosphorus loading as the response to complete and then limited sustainable restoration of a shallow lake

Katarzyna Kowalczewska-Madura[1,*], Renata Dondajewska[1], Ryszard Gołdyn[1], Joanna Rosińska[2] and Stanisław Podsiadłowski[3]

[1] Department of Water Protection, Faculty of Biology, Adam Mickiewicz University, Umultowska 89, 61-614 Poznań, Poland
[2] Department of Environmental Medicine, Poznań University of Medical Sciences, Rokietnicka 8, 60-806 Poznań, Poland
[3] Institute of Biosystems Engineering, University of Life Sciences, Wojska Polskiego 50, 60-627 Poznań, Poland

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Abstract – The urban Swarzędzkie Lake, into which sewage had been diverted many years ago, was still characterised by low ecological status. Three restoration methods were used in order to improve the water quality, *i.e.* aeration of the waters overlying the bottom sediments, inactivation of phosphorus in the water column with iron sulphate and magnesium chloride and biomanipulation with pike stocking. The aim of the research was to define seasonal and spatial changes of phosphorus internal loading from bottom sediments and to compare this with previous years. We also considered changes in the process of P release when the restoration treatments were limited after 3 yr from 3 methods to 1 method. The highest phosphorus release from bottom sediments was found in the profundal zone, where in summer periods it would reach up to 29.2 $mgP\,m^{-2}\,day^{-1}$. The lowest P release was observed in the littoral zone, down to a depth of 3 m, where it did not exceed 10.0 $mgP\,m^{-2}\,day^{-1}$. 31% of the whole load of P released from the bottom sediments was originated from this zone. The research showed an increase of phosphorus release in the first years of restoration treatment and a systematic decrease at all stations in the following years.

Keywords: Lake / sustainable restoration / phosphorus release / bottom sediments

1 Introduction

One of the most serious global threats to inland aquatic ecosystems is the excessive eutrophication of lakes (Søndergaard *et al.*, 2007). Progressive eutrophication leads to changes in lake ecosystems as a result of significant external loading with nutrients (Gołdyn *et al.*, 2015; Klimaszyk *et al.*, 2015), but phosphorus internal loading from bottom sediments could also pose a major threat (Boström *et al.*, 1988; Golterman, 1995; Søndergaard *et al.*, 2001, 2002).

High internal loading of phosphorus from lake sediments is frequently reported as an important mechanism delaying lake recovery after a reduction of external loading and it can endure for up to 10–15 yr after such reduction (Søndergaard *et al.*, 2007). Thus, the high productivity of phytoplankton is maintained by intensive internal P loading for decades, even if external loading is reduced to a tolerable level (Horppila *et al.*, 2017).

Highly eutrophied lakes are characterised by intensive phytoplankton blooms in summer, oxygen depletion near the bottom sediments and high chlorophyll-a concentrations. According to the Water Framework Directive (WFD) all water bodies should achieve good ecological and chemical status (Directive, 2000). It is possible to apply a variety of methods to protect lake ecosystems. However, protection measures are not sufficient in many cases and lakes need additional restoration treatment (Dunalska *et al.*, 2015; Kostecki *et al.*, 2017). In the case of intensive internal loading it is vital to use phosphorus inactivation methods which decrease its release from the sediments. These methods consist of iron and/or aluminium treatment, as well as the use of a lanthanum modified clay named Phoslock (Cooke *et al.*, 1993; Søndergaard *et al.*, 2002; Klapper, 2003; Meis *et al.*, 2012). The principal claim made against such methods is the use of chemical compounds added to the ecosystem and the use of large amounts of chemicals which influence biological elements in the lake ecosystem. An effective solution to these problems is sustainable restoration which uses chemical compounds naturally present in the lake ecosystem like iron sulphate or magnesium chloride. These compounds are applied in precise doses that do not exceed their natural

*Corresponding author: madura@amu.edu.pl

occasional concentrations in lake ecosystems (Gołdyn *et al.*, 2014). An auxiliary technique also worth using is biomanipulation, which is based on removing excessive numbers of cyprinids (Cyprinidae) and a gradual stocking with predatory species of fish (Gołdyn and Mastyński, 1998; Kasprzak *et al.*, 2002; Mehner *et al.*, 2002; Jurajda *et al.*, 2016).

Additional use of sustainable aeration, *e.g.* a wind-driven aerator, enables the redox potential at the water–sediment interface to be increased, which in turn increases the sorption capacity of the bottom sediments (Søndergaard *et al.*, 2002; Podsiadłowski *et al.*, 2018).

Since sustainable lake restoration is a relatively recent technique, the processes occurring in bottom sediments that cause a reduction of phosphorus release to the water column are still to be fully elucidated (Kowalczewska-Madura *et al.*, 2017).

The aim of the experimental research was to determine changes in internal P loading as a result of a complete sustainable restoration (simultaneous use of three methods) as well as changes that occur under the influence of limitation of this restoration. The initial research was conducted prior to the restoration, the main research took place during the 3 yr of the restoration period and was continued 2 yr after phosphorus inactivation and biomanipulation had terminated. At the same time spatial and seasonal variability of internal P loading was analysed. The results were compared to those of the previous years (2001–2003).

2 Material and methods

2.1 Lake characteristics

Swarzędzkie Lake (Poland) is a natural water body of glacial origin. It is located in the northwestern part of the town of Swarzędz, near the City of Poznań (52°25′N, 17°04′E). The lake surface is 93.7 ha, maximum depth 7.2 m, mean depth 2.6 m and volume 2 000 000 m^3. The lake has 2 islands with a total area of 0.3 ha (Szyper *et al.*, 1994). It is elongated, narrowing from half its length towards the outlet. The wider part is deeper (7.2 m), while the narrower part is no more than 2 m deep. Before the restoration this shallower part of the lake was covered with floating leaved plants: *Nuphar lutea*, *Nymphaea alba*, pleustophytes: *Hydrocharis morsus-ranae*, *Lemna minor*, *Spirodela polyrhiza* and the submerged macrophyte *Ceratophyllum demersum* (Rosińska *et al.*, 2017b). Swarzędzkie Lake is not thermally stratified, because only epilimnion and metalimnion are formed in summer (Lampert and Sommer, 1999). The metalimnion is in contact with only 15% of the lake bottom. The remaining part is the so-called active bottom, which is located in the littoral zone within contact of the epilimnetic water layer. The Osgood index (mean depth over square root of a surface area) which predicts vertical phosphorus transport in lakes, calculated for Swarzędzkie Lake, was 2.9. When the index is less than 6 the lake may be considered polymictic, as it may be subjected to periodical wind mixing during heavy storm events (Mataraza and Cooke, 1997).

The total catchment area of the lake is 17 230 ha, but the direct catchment area adjacent to the shoreline is only 452 ha. The total catchment area contains mostly farmland (75.5%), forests (18%) and built-up areas (5.3%). The lake is supplied with water mainly by the River Cybina and partially by the Mielcuch Stream (Szyper *et al.*, 1994). Water discharge from the Cybina is characterised by small seasonal fluctuations because 3 other lakes located in the course of the river above Swarzędzkie Lake have a retention effect. Long-term mean discharge of the river is 0.675 $m^3\,s^{-1}$. The Mielcuch Stream flows initially in an open ditch, but in the town of Swarzędz it has been transformed into a rain collector, receiving stormwater and some sanitary sewage from illegal connections in the centre of town. The waters of this tributary are heavily contaminated by point, diffuse and dispersed sources of pollution. An important role is also played by runoff from farmland. The River Cybina is rich in nutrients originating from arable fields in the catchment and fish ponds situated in its valley (Kowalczewska-Madura, 2003). During this study total phosphorus concentrations in the River Cybina varied from 0.03 mgP L^{-1} to 0.64 mgP L^{-1} (mean 0.18 mgP L^{-1}) and they were much higher in the Mielcuch Stream and ranged from 0.23 mgP L^{-1} to 1.84 mgP L^{-1} (mean 0.73 mgP L^{-1}). Total nitrogen concentration in the Cybina reached 9.24 mgN L^{-1} (mean 3.99 mgN L^{-1}) and in the Mielcuch Stream 32.9 mgN L^{-1} (mean 9.99 mgN L^{-1}) (unpublished data).

2.2 Restoration of Swarzędzkie Lake

For many years Swarzędzkie Lake has been subject to strong human impacts, namely the direct discharge of urban sewage and high external nutrient loads from non-point sources. Over 80% of the direct sewage load was diverted from the lake and directed to a sewage treatment plant in Poznań in 1991. After treatment the sewage is discharged outside the lake catchment, namely to the River Warta. Despite this, the water quality did not markedly improve. The lake remained hypereutrophic for the next 20 yr. It was still overloaded with nutrients released from bottom sediments and partly with those from the catchment area (Kowalczewska-Madura, 2003; Kowalczewska-Madura and Gołdyn, 2009). The lake was characterised by intensive cyanobacterial blooms, low water transparency and oxygen depletion in the deeper water layers (Stefaniak *et al.*, 2007). It was not used for recreation because of its low water quality (Kozak *et al.*, 2014).

Swarzędzkie Lake has been undergoing restoration since September 2011 on the basis of 3 methods: phosphorus inactivation with iron sulphate (PIX-112 preparation $Fe_2(SO_4)_3$) and magnesium chloride ($MgCl_2$), water aeration using a wind aerator and biomanipulation. The aim of the applied treatments was to decrease phosphorus concentration in the water column and oxygenation of the waters overlying bottom sediments and to decrease the internal phosphorus loading from the sediments. Phosphorus inactivation using small doses of iron sulphate and magnesium chloride (dosage: 200–300 kg $Fe_2(SO_4)_3$ and 200 kg $MgCl_2$ per lake) was performed 5–9 times per year in 2012–2014. Coagulants were dosed as a solution on the lake surface using special mobile equipment in order to precipitate phosphorus from the water and increase the phosphorus-binding capacity of the sediment (Søndergaard *et al.*, 2002; Gołdyn *et al.*, 2014). Aeration of the water layer above the bottom sediments with the use of a wind-driven aerator located in the deepest part of the lake was begun in autumn 2011 and continues to the present (Podsiadłowski *et al.*, 2018). Biomanipulation consisted of *ca.* 5% of cyprinid

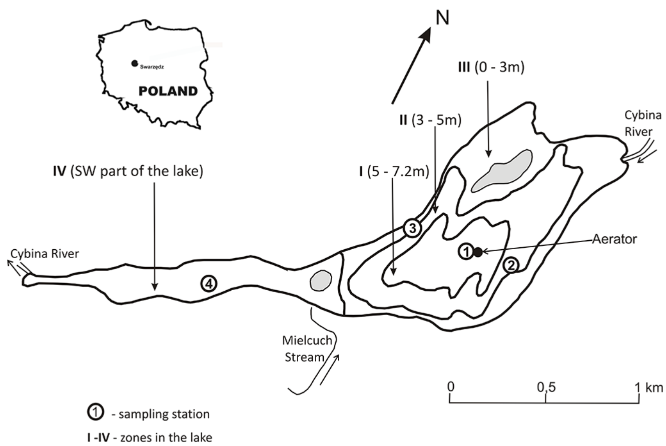

Fig. 1. Location of sampling stations and P release zones on a bathymetric map of Swarzędzkie Lake (according to Kowalczewska-Madura and Gołdyn 2009, modified).

removal and stocking of pike *Esox lucius* L. and pikeperch *Sander lucioperca* L. fry (Rosińska *et al.*, 2017b).

Unfortunately, from 2015, *i.e.* after 3 yr of restoration, phosphorus inactivation and biomanipulation were halted due to a lack of financial support.

2.3 Changes of water quality

Swarzędzkie Lake has been studied several times, mainly with respect to water quality, macrophytes and lake ecosystem functioning (Kowalczewska-Madura, 2003; Kowalczewska-Madura and Gołdyn, 2009, 2012; Kozak *et al.*, 2014, 2018; Rosińska and Gołdyn, 2015; Rosińska *et al.*, 2017a,b). Due to the intense restoration measures water quality had already begun to improve in the first year of restoration. The domination of cyanobacteria was inhibited and the total abundance of autotrophic organisms was reduced (Rosińska *et al.*, 2017a). After the cessation of two of the restoration processes (phosphorus inactivation and biomanipulation) the abundance of this group increased again in 2016 (Kozak *et al.*, 2018). The phytocoenotic richness of vegetation increased (from 9 to 12 plant communities), although the total area covered by macrophytes decreased by 12% during restoration efforts. The area dominated by hypereutrophic plant species (*C. demersum*, *H. morsus-ranae* and *Typha angustifolia*) decreased, although the former submerged plant typical of less eutrophicated water bodies (*Potamogeton lucens*) returned and the area dominated by some existing species (nymphaeids) increased (Rosińska *et al.*, 2017b). Chlorophyll-a concentration halved during restoration, with an average annual concentration of between 21.2 and 44.4 $\mu g\,L^{-1}$ (before restoration it reached 95.0 $\mu g\,L^{-1}$). The concentration of nitrogen and phosphorus slightly decreased, especially in the deep water layer. Secchi depth increased to 1.0 m and oxygenation improved, as the anaerobic period in the deep water layer shortened to 1 month. However, the mean oxygen content in the surface layer decreased to 8.6 $mgO_2\,L^{-1}$ during the process as a result of a decreased amount of phytoplankton. The range of pH was 6.5–8.1 in the depth profile at the deepest place of the lake, and was slightly lower compared to the period before the restoration (Rosińska *et al.*, 2017a, 2018).

2.4 Bottom sediments – methods

Bottom sediments from Swarzędzkie Lake were sampled every month at 4 research stations. The stations differed in depth, oxygenation and thermal conditions. Station 1 was located in the deepest part of the lake near the wind aerator, where oxygen depletion occurred in deep water in summer. The depth at station 2 was 4 m, it was situated near the marina, where low oxygen concentration near the sediments frequently occurred. The other stations were located in the littoral zone at a depth of 2 m. Station 3 was in the wider part of the lake near the forest catchment and station 4 in the centre of the shallower part of the lake (Fig. 1). Sediments at stations 1–3 were sampled from March 2011 to November 2016, and at station 4 from 2012 to 2016.

Taking into account the lake morphometry and character of the bottom sediments 4 zones differing in rates of P release were distinguished in the lake. Zone I denotes the bottom at a depth of 5–7.2 m in the northeastern part of the lake (area 118 720 m^2), in which sediments in summer are in contact with anoxic over-bottom waters. The release was approximated on the basis of data from station 1. Zone II was set at a depth of 3–5 m (area 236 060 m^2), based on the data from station 2, in which sediments are in contact with water periodically weakly oxygenated. Zone III was at a depth of 0–3 m (area 300 700 m^2) based on the data from station 3, characterised by the contact of sediments with well oxygenated epilimnetic waters. This zone could be affected by the main inflow of the River Cybina. Zone IV (area 138 520 m^2) was located in the shallow southwestern part of the lake (station 4). The sediments in this zone were enriched with allochthonous organic matter and nutrients discharged by the Mielcuch Stream (Fig. 1).

Bottom sediments were sampled using a Kajak tube sampler. The surface layer of the sediments was analysed. Sediment samples were dried and then incinerated at 550 °C. They were analysed for organic matter content (%) and total phosphorus (mgP g^{-1}DW). TP content was analysed using the molybdate method with ascorbic acid as a reducer (PN-EN ISO 6878:2006).

Phosphorus was fractionated in the collected sediment samples using the protocol proposed by Psenner *et al.* (1988) and Lewandowski *et al.* (2003). The fractions were loosely bound phosphorus (NH_4Cl-P), phosphorus bound with iron (BD-P), phosphorus bound with aluminium and organic matter (NaOH-P and NaOH-NRP, respectively), phosphorus bound with calcium and magnesium (HCl-P) and the residual phosphorus (Res-P), which was calculated as the difference between total P and the sum of the first 4 fractions.

The additional variables in bottom sediments were analysed only at stations 1 and 4 in 2012–2015, namely nitrogen, sulphates, iron, calcium and magnesium. Total nitrogen was determined using a TOC-L Shimadzu analyser with a TNM-L unit via catalytic thermal decomposition and the chemiluminescence method (Shimadzu, Japan). Determination of SO_4^{2-} was performed using the gravimetric method, and Fe (total iron) using atomic absorption spectrometry with flame atomisation

(F-AAS) (Shimadzu AA7000, Japan). Total water hardness and Ca^{2+} concentration were determined by the versenate method, while Mg^{2+} concentration was calculated from the difference between total hardness and the concentration of Ca^{2+} ions (Standard Methods, 1999).

Pore waters were separated by centrifugation for 1 h at 3000 revolutions per minute. Soluble reactive phosphorus (SRP) concentration was analysed in the supernatant with the molybdate method with ascorbic acid as a reducer, and total phosphorus (TP) with the same method, after mineralisation (PN-EN ISO 6878:2006). SRP and TP concentration were also analysed in the water layer above the sediments at 4 sampling stations in every season from 2011 to 2016.

The experiments on phosphorus release from bottom sediments were performed *ex situ* using intact sediment cores in tubes, sampled with a modified Kajak bottom sampler at 4 stations in every season. Sediments were sampled in 3 replicates in rigid plastic tubes (PMMA – polymethyl metacrylate) 6 cm in diameter. Each transparent tube contained the collected sediment sample (*ca.* 15 cm layer) and overlying water (*ca.* 25 cm). The cores were incubated in the laboratory under constant thermal conditions (similar to the temperature at the bottom of lake during sampling) in darkness. Depending on the oxygenation of the overlying water, the experiments were conducted under aerobic or anaerobic conditions. To maintain the anaerobic conditions the tubes were tightly capped with rubber stoppers. In experiments with aerobic conditions the tubes were simply left open to the atmosphere or artificially aerated. Water from above the sediments (50 mL) was sampled from every tube at definite intervals (1–3 days) over a period of 2 weeks. TP content was analysed in these samples using the molybdate method with ascorbic acid as a reducer, after mineralisation (PN-EN ISO 6878:2006). Water temperature, dissolved oxygen concentration, pH, conductivity and redox potential were measured in every tube before each water sampling (WTW Multi 350i meter).

Ex situ experiments were used to assess the amount of phosphorus released or accumulated in bottom sediments per unit of area and time ($mgP\,m^{-2}\,day^{-1}$). The experimental results from every tube were recalculated to 1 m^2 per day (in consideration of the overlying water volume in the tube and the sediment surface of the core). When the obtained value was positive, the sediments were released phosphorus, while a prevailing negative value indicated the accumulation of phosphorus in the bottom sediment. Next, mean values were calculated for the data from the 3 tubes used in each experiment at each research station (Kowalczewska-Madura *et al.*, 2015, 2017).

The annual internal TP loading for each zone (I–IV) was calculated by multiplying the daily mean TP release from laboratory experiments by the area of this zone and by the number of days per year (in $kgP\,a^{-1}$).

Statistical calculations were made with STATISTICA version 10.0 software. To confirm the significance of differences between the analysed sediment variables in time and space, non-parametric tests were used, *i.e.* Kruskal–Wallis (K–W) and Mann–Whitney (M–W) tests. The Pearson coefficient was used to determine relationships between variables. We used the value $\alpha = 0.05$ to assess the significance of all statistical tests.

3 Results

The bottom sediments of Swarzędzkie Lake were characterised by considerable variation in structure and composition. The sediment from stations 1 (7.2 m, profundal zone) and 2 (4 m) can be classified as dark-grey algal gyttja. They were characterised in summer by the odour of hydrogen sulphide. Sediment from station 3 (littoral) was sandy and from station 4 coarse detritus gyttja with macroscopic plant debris. The sampling stations also differed in the oxygenation of the water overlying the sediments. The highest concentrations of oxygen were found at station 3 (mean $5.18 \pm 2.7\,mgO_2\,L^{-1}$) and the lowest at station 1 (mean $3.5 \pm 2.9\,mgO_2\,L^{-1}$). At stations 2 and 4, they were $4.08 \pm 3.1\,mgO_2\,L^{-1}$ and $5.03 \pm 1.2\,mgO_2\,L^{-1}$, respectively. There were anaerobic conditions in summer at stations 1 and 2, and the oxygen concentration did not exceed $2\,mgO_2\,L^{-1}$.

The content of TP in the bottom sediments of Swarzędzkie Lake at the particular stations ranged from $0.31\,mgP\,g^{-1}DW$ to $2.63\,mgP\,g^{-1}DW$ in 2011–2016. The highest values were observed at station 4 located in the shallow southern part of the lake, while the lowest values were recorded at station 3, *i.e.* in the littoral zone of the main part of the lake (Fig. 2a). The average annual concentration of phosphorus at station 1 clearly decreased in the first year of restoration and rose to a value close to that of before the restoration in subsequent years. A similar variability was found at the other stations, although the differences were not so distinct. A statistically significant difference was found between the stations considered (K–W (5:241) = 30.088; $p < 0.001$) and the particular years of studies (K–W (3:241) = 98.341; $p < 0.001$).

The highest average content of organic matter in bottom sediments was recorded at station 4, *i.e.* in the shallow part of the lake, overgrown by macrophytes. The lowest values were found at station 3, in the littoral zone with sandy sediments. However, the mean values at stations 1 and 2 were similar to those at station 3 (Fig. 2b). They showed insignificant fluctuations in subsequent years of research. A minor decrease was found at station 4, but the differences between the years were not statistically significant (Fig. 2b).

The dominant fraction of TP in the bottom sediments was Res-P, phosphorus biologically almost unavailable, which occurred as insoluble mineral and organic compounds. Its mean content was close to 50% at all research stations. The total content of 3 fractions of the best biological availability, *i.e.* NH_4Cl-P, BD-P and NaOH-P, was low, especially at shallower stations 3 and 4. Higher values were found at stations 1 and 2, *i.e.* 15.2% and 12.0%, respectively. In the case of the fraction characterizing phosphorus bound with calcium (HCl-P), a higher mean share was found at stations 3 and 4 (approx. 21%), and lower in the profundal zone (13%) (Tab. 1). The K–W test indicated statistically significant differences between particular stations for all analysed fractions (K–W (3:225) = 96.99 for NH_4Cl-P; 88.41 for BD-P; 29.34 for NaOH-P; 32.26 for NaOH-NRP; 50.05 for HCl-P and 48.98 for Res-P; $p < 0.001$).

The variability of P fractions in the subsequent years of the study indicated that in the case of most of them, *i.e.* NH_4Cl-P, NaOH-P, NaOH-NRP and HCl-P, their share in TP initially decreased and then slightly increased (Fig. 3). In the case of P

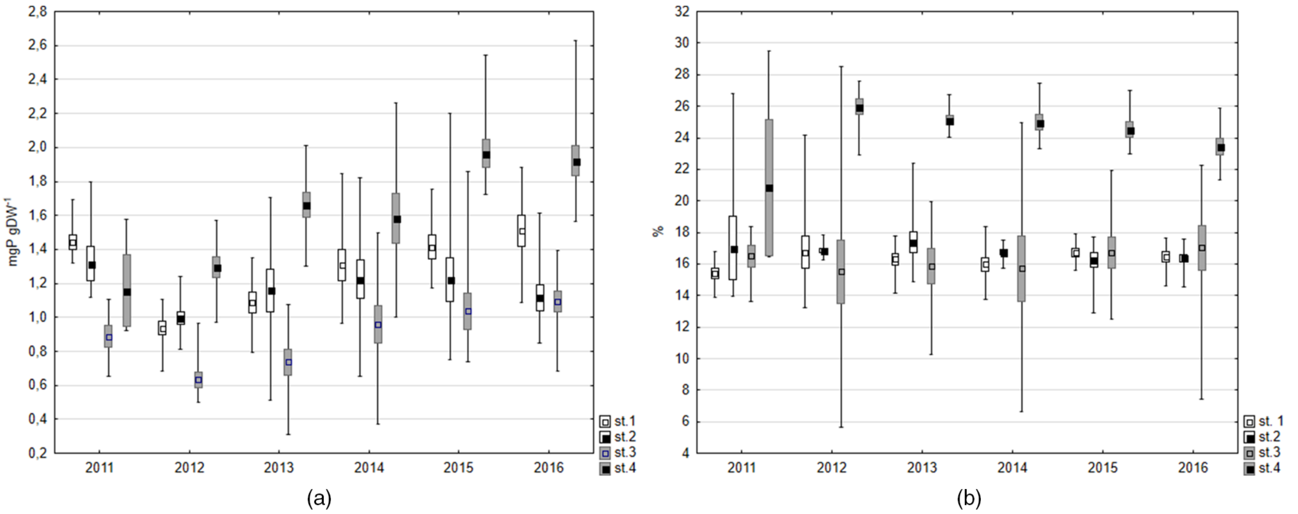

Fig. 2. Time and spatial variability of TP content (a) and organic matter concentration (b) in sediments of Swarzędzkie Lake in 2011–2016 (box – mean ± standard deviation, whiskers – minimum and maximum).

Table 1. Percentage share of extractable fractions of TP in bottom sediments of Swarzędzkie Lake at analysed stations in 2011–2016.

Fractions	Station	Mean	Minimum	Maximum	SD
NH_4Cl-P	1	7.60	2.92	13.53	2.66
	2	5.38	1.01	11.04	2.29
	3	3.35	0.58	10.91	2.09
	4	3.96	0.78	13.51	2.35
BD-P	1	4.21	1.01	9.81	2.07
	2	3.08	0.57	9.55	2.32
	3	1.67	0.21	7.53	1.49
	4	2.19	0.24	9.78	1.22
NaOH-P	1	3.41	0.06	10.33	2.14
	2	3.55	0.41	10.12	2.25
	3	2.81	0.12	10.71	2.07
	4	3.52	0.19	16.85	2.70
NaOH-NRP	1	17.21	5.39	38.06	7.31
	2	18.83	6.02	46.03	8.74
	3	17.55	3.91	52.74	9.62
	4	18.13	2.42	37.14	8.07
HCl-P	1	13.28	3.54	28.77	6.23
	2	19.91	5.86	41.13	8.26
	3	21.05	3.98	47.15	9.45
	4	20.61	6.69	43.65	8.25
Res-P	1	54.29	22.62	84.77	13.87
	2	49.25	21.22	81.59	15.52
	3	53.57	9.25	86.67	15.95
	4	51.57	10.13	86.75	16.66

bound with iron (BD-P) the decrease was very distinct and a slight increase was observed only in recent years, while the share of the Res-P fraction increased almost throughout the whole study period (Fig. 3). Statistical analysis (K–W test) indicated significant differences between the analysed years in the case of all fractions (for $p \leq 0.05$).

The mean content of the remaining variables in the bottom sediments showed that, with the exception of calcium, they reached higher values at shallow station 4 than at station 1. Statistically significant differences between the stations were noted only for nitrogen and iron (M-W non-parametric test for nitrogen: -2.63, $p = 0.008$ and for iron: -2.78, $p = 0.005$).

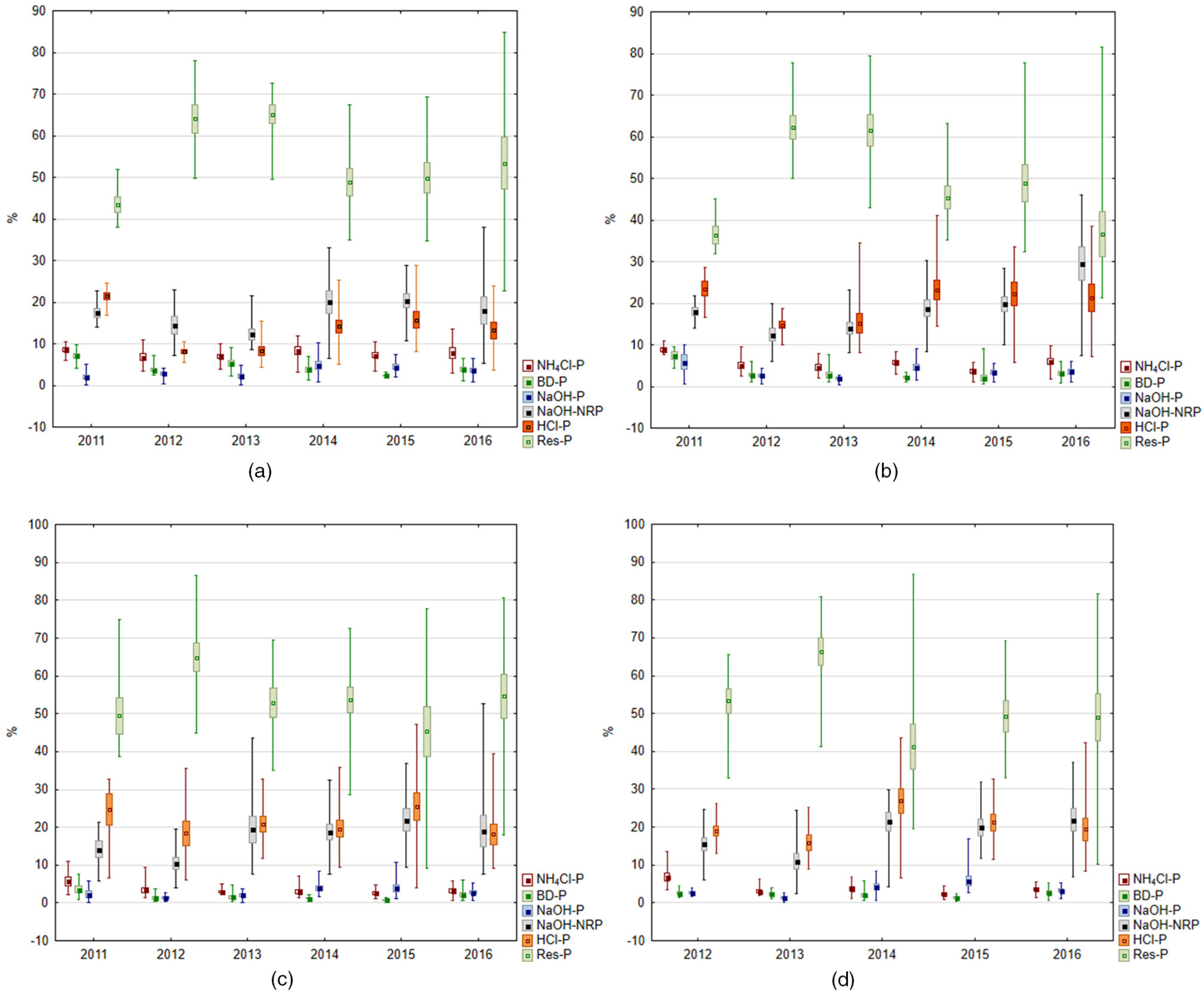

Fig. 3. Changes of percentage share of extractable fractions of TP in bottom sediments of Swarzędzkie Lake in 2011–2016 (a – station 1, b – station 2, c – station 3, d – station 4) (box – mean ± standard deviation, whiskers – minimum and maximum).

The content of sulphates, iron and magnesium first increased and then decreased in the subsequent years of the study, especially in 2015, while the concentration of nitrogen fluctuated (Fig. 4). Statistically significant differences (K–W test) occurred between the successive years of the study in the case of most of the analysed parameters (except for calcium) (Fig. 4).

The concentration of SRP and TP in interstitial water varied within a wide scope from 0.006 mgP L^{-1} to 5.82 mgP L^{-1} for SRP and from 0.14 mgP L^{-1} to 6.79 mgP L^{-1} for TP. The highest values were noted in summer and autumn in the profundal zone (station 1), and the lowest in the littoral zone (stations 3 and 4). The concentration of both forms of phosphorus at stations 2, 3 and 4 decreased in the first 4 yr of the study and then slightly increased (Fig. 5a and b). No significant changes were observed in the case of station 1. Statistically significant differences were found between the analysed stations: K–W (3:232) = 109; $p < 0.001$ and K–W (3:232) = 116; $p < 0.001$ for SRP and TP, respectively. Such differences were also found in all data from particular years of the study: K–W (5:232) = 27; $p < 0.001$and K–W (5:232) = 24; $p < 0.001$ for SRP and TP, respectively.

In 2011–2016 the mean concentration of SRP and TP in the waters overlying the sediments of Swarzędzkie Lake reached the highest values at station 1 located in the profundal zone. Its maximum value was up to 1.08 mgP L^{-1} for SRP and up to 1.13 mgP L^{-1} for TP. The highest values were noted in summer. Both forms of phosphorus suddenly increased in the first year of restoration and decreased in the following 3 yr, then slightly increased again in 2016. The mean concentrations of both forms of phosphorus at the other stations were rather similar and did not exceed 0.04 mgP L^{-1} and 0.11 mgP L^{-1} for SRP and TP, respectively (Fig. 5c and d). Statistically significant differences were found between particular years of the study: K–W (5:85) = 11.14, $p = 0.0486$ and K–W (5:85) = 16.16, $p = 0.0064$ for SRP and TP, respectively.

Ex situ experiments on internal loading showed that at station 1, located in the profundal zone, phosphorus release from bottom sediments to the overlying water dominated during the whole period of the study. Its highest intensity was noted in summer of the first 4 yr (Fig. 6). The release was lower and more equal in particular seasons during the later years of the study. The lowest values were observed in spring and

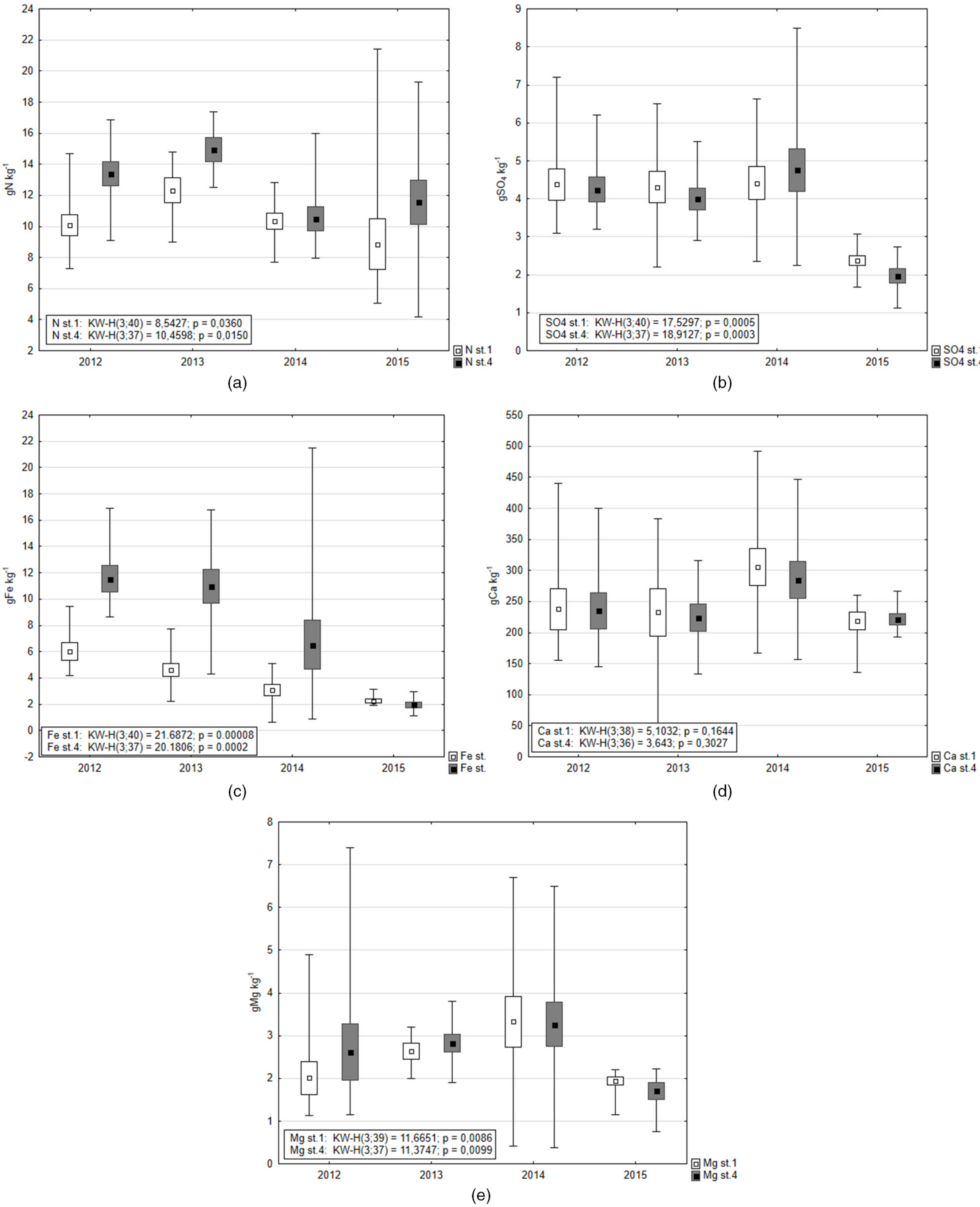

Fig. 4. Changes of nitrogen (a), sulphur (b), iron (c), calcium (d) and magnesium (e) concentration (mean values) in bottom sediments of Swarzędzkie Lake in 2012–2015 (box – mean ± standard deviation, whiskers – minimum and maximum).

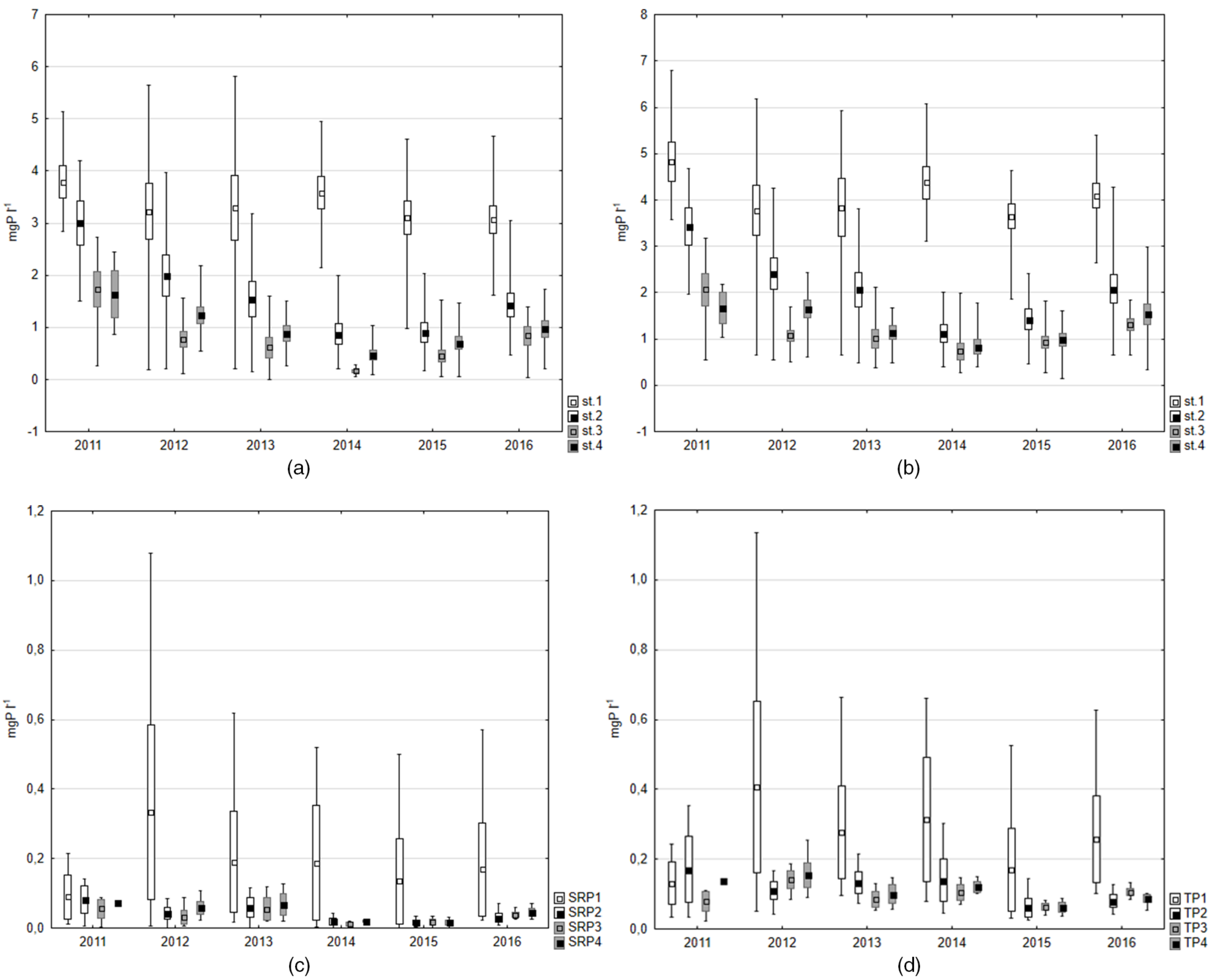

Fig. 5. Time and spatial variability of SRP and TP concentrations in pore waters of bottom sediments (a and b) and in over-bottom water (c and d) of Swarzędzkie Lake (box – mean ± standard deviation, whiskers – minimum and maximum).

winter, even in the first period of research. The release of phosphorus from bottom sediments also dominated at station 2 located at a water depth of 4 m. The highest values were found in summer with a maximum in 2013. Slightly lower values were observed in autumn. However, a slight dominance of accumulation of phosphorus in bottom sediments was found at the same station in winter and spring 2013. Phosphorus release in summer 2014–2015 decreased and in 2016 it rose slightly again. At station 3, located in the littoral zone of the main part of the lake, in the colder periods of 2011–2013 phosphorus absorption from the over-bottom water to the sediments was noted. Phosphorus release was predominant in summer of the following years, but was lower than at the other stations. The pattern of seasonal variability at station 4 located in the narrow and shallower part of the lake was similar to station 3. Phosphorus release from the sediments to the water column was predominant mainly in summer, reaching its highest value in 2013 (Fig. 6). Statistical analysis indicated significant differences between the analysed stations (K–W (3:85) = 18.31, $p < 0.001$).

The release of phosphorus from bottom sediments predominated at all stations in summer during the restoration treatments. It increased twice in the profundal zone (station 1) in 2012–2013 in comparison to 2011, and it decreased significantly in the following years. A similar variability was also observed at station 2. The internal loading from the littoral zone of the lake slightly declined in the subsequent years of restoration (Fig. 6).

The mean annual values of internal loading rose at station 1 from 7.7 mgP m^{-2} day^{-1} in 2011 to 13.8 mgP m^{-2} day^{-1} in the first year of restoration (2012) and fell in the following years, reaching 3.2 mgP m^{-2} day^{-1} in the final year of the study (Fig. 7). A similar variability of the process could be observed at station 2. At the same time, only insignificant changes of P release from bottom sediments were noted in the littoral zone (stations 3 and 4) in the successive years of the study (Fig. 7). As a result of the restoration treatment, P release from bottom sediments reached similar values at all stations in the final year of the study. The K–W test did not indicate statistically significant differences between particular years of the study at particular stations. Phosphorus release predominated over its accumulation in 2001–2003. The mean annual value was then 12.53 mgP m^{-2} day^{-1} and 10.5 mgP m^{-2} day^{-1} for stations 1 and 2, respectively (Fig. 7), so it was almost twice as high as in

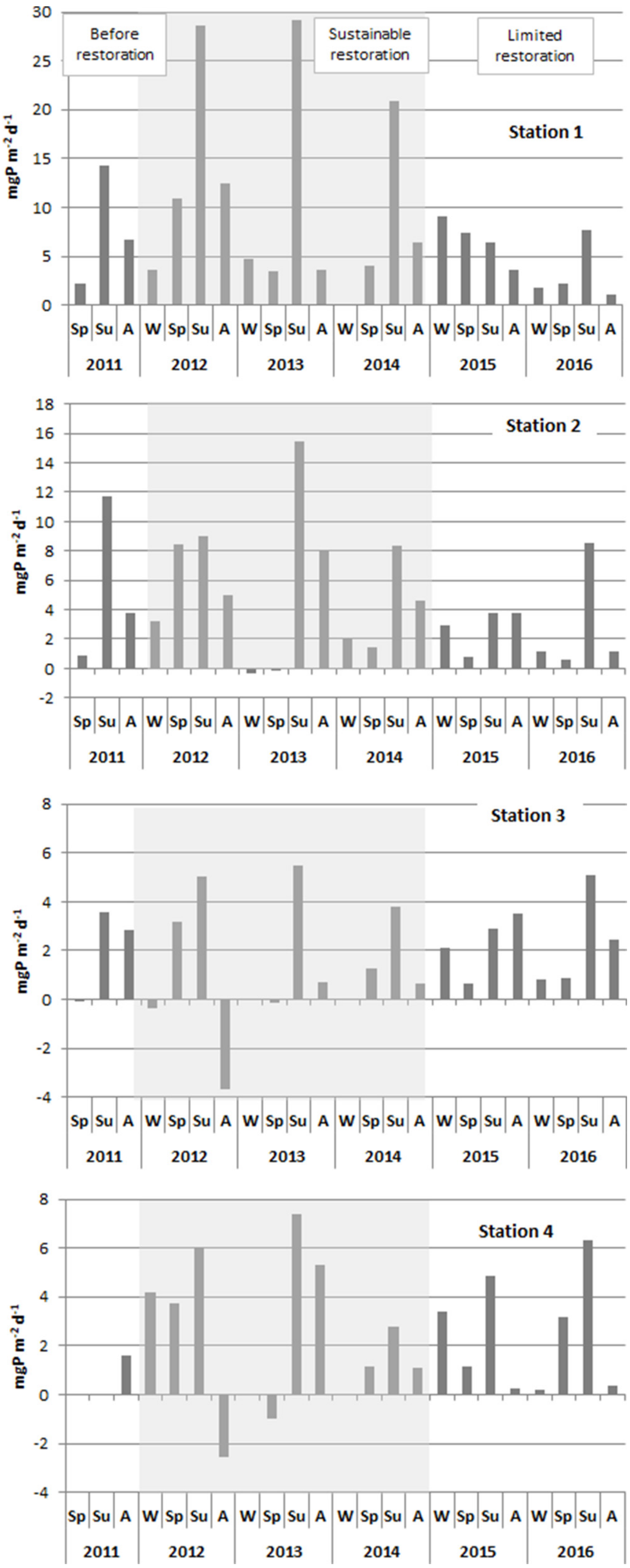

Fig. 6. Changes of domination of phosphorus release (positive value) or its accumulation (negative value) in bottom sediments of Swarzędzkie Lake at four stations in 2011–2016.

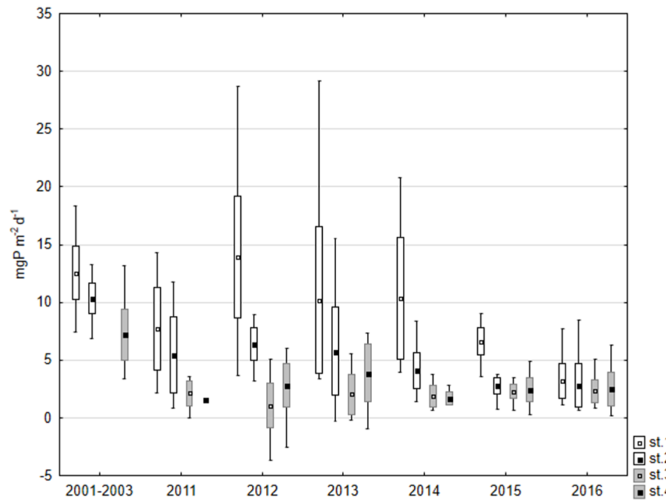

Fig. 7. Mean phosphorus release from bottom sediments of Swarzędzkie Lake at four stations in 2011–2016 and in 2001–2003 (box – mean ± standard deviation, whiskers – minimum and maximum).

2011 (before the restoration) (Kowalczewska-Madura and Gołdyn, 2009). No statistically significant difference was found between the years 2001–2003 and 2011 (K–W (1:19) = 2.86, $p = 0.09$).

A statistically significant correlation between phosphorus internal loading and the concentration of SRP and TP in water overlying the sediments was found in Swarzędzkie Lake at station 1 (for SRP: $r = 0.783$, $p < 0.001$ and for TP: $r = 0.784$, $p < 0.001$) and at station 2 ($r = 0.499$, $p = 0.015$). A similar significant relationship between the intensity of P release from the bottom sediments and the concentration of SRP and TP in the interstitial waters was recorded ($r = 0.516$ and $r = 0.466$, respectively, at $p < 0.05$). Furthermore, there was found to be a relationship between P release and the concentration of the two most mobile fractions in the sediments, *i.e.* NH_4Cl-P and BD-P ($r = 0.310$ and $r = 0.267$, respectively, at $p < 0.05$).

The annual phosphorus loading from the bottom sediments to the overlying water was defined for each zone of Swarzędzkie Lake designated in relation to the depth of water. The highest values were noted in zones I and II, located at depths of between 3 and 7.2 m in the main part of the lake, which together constituted 69% of the total internal loading of phosphorus as a mean from the years 2012–2016. Lower phosphorus loading was definitely observed from the shallow zones, located in the littoral of the lake. It reached its lowest value in the elongated south-west zone, and a slightly higher one in the shallow littoral zone of the main part of the lake (Tab. 2).

The annual phosphorus load from the bottom sediments of Swarzędzkie Lake in zone I increased significantly in 2012 by 45%. It dropped in the following years and the value noted in 2016 was 58% lower than in the first year of the study. A similar variability was observed in zone II in the subsequent years but in a smaller range. The phosphorus load rose by 15% in the first year of restoration, and decreased by 48% in the last year of restoration in comparison to 2011. Some slight changes could also be observed in zones III and IV located in the littoral

Table 2. Annual loading of phosphorus (kgP a^{-1}) from bottom sediments in four zones in Swarzędzkie Lake in 2011–2016.

	Before restoration	Sustainable restoration			Limited restoration	
		Aerator, P inactivation and biomanipulation			Aerator	
Zone	2011	2012	2013	2014	2015	2016
I	335.0	601.9	442.4	449.8	287.3	139.4
II	470.4	551.4	498.0	352.4	241.3	246.4
III	232.7	116.4	221.7	207.4	251.3	253.7
IV	82.4	144.2	197.2	84.4	122.9	127.0
Total	1120.5	1413.9	1359.3	1094.1	902.8	766.5

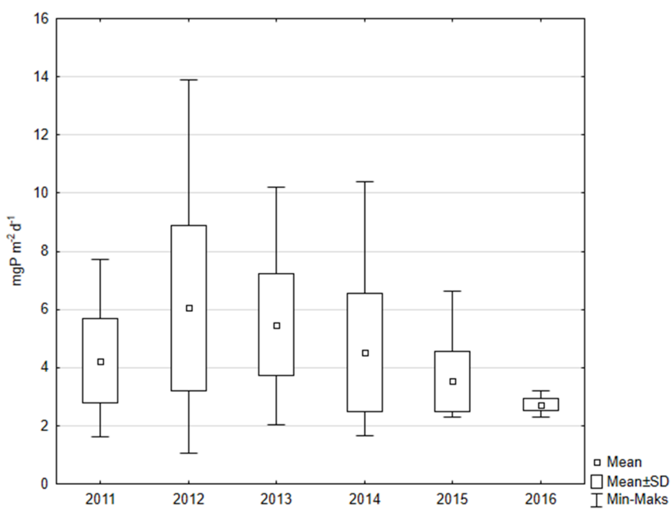

Fig. 8. Mean values of phosphorus release from bottom sediments of Swarzędzkie Lake in 2011–2016 (box – mean ± standard deviation, whiskers – minimum and maximum).

zone of the lake. Fluctuations of phosphorus loading were noted in the following years in both cases, but finally it increased in comparison to 2011 by 9% in zone III and 54% in zone IV (Tab. 2).

The analysis of the variability of mean phosphorus release in the particular years of restoration indicated that it increased by 26% until 2012 and was followed by a slow decline in subsequent years, up to 32% in the most recent year of the study in comparison to 2011, *i.e.* before the restoration (Fig. 8).

4 Discussion

Internal phosphorus loading from the bottom sediments of Swarzędzkie Lake before the restoration was typical of hypereutrophic lakes (Kleeberg and Kozerski, 1997; Søndergaard *et al.*, 2001; Horppila *et al.*, 2017). In 2001–2003, it was found that at all stations during all seasons the phosphorus release from bottom sediments to the water column predominated its accumulation and reached its maximum values in summer or autumn (Kowalczewska-Madura and Gołdyn, 2009) being almost twofold higher than in 2011. However, despite this reduction, internal phosphorus loading was still too excessive, causing cyanobacterial water blooms, which is why the restoration was undertaken.

The research conducted in 2011–2016 defined the seasonal and spatial variability of phosphorus release in zones differing in water depth, oxygenation of waters overlying the sediments (especially in summer) and the overgrowth of macrophytes. Moreover, the analysis of the multiannual changes in this process indicated its distinct variability, both in particular zones and in the following years of restoration treatments.

The greatest phosphorus release from bottom sediments was observed in summer in the bottom zone located around the deepest place of the lake (station 1). It was mainly caused by oxygen depletion in the over-bottom waters up to a depth of 4 m in summer. The concentration of oxygen is one of the most important factors that may influence the intensity of phosphorus exchange in the sediment–water interface. The lack of oxygen leads to a reduction of iron and other metals and consequently to the release of soluble phosphate ions into the water column (Søndergaard *et al.*, 2002). Phosphorus release in anoxic conditions is several times higher than under aerobic conditions (Boström *et al.*, 1982; Ishikawa and Nishimura, 1989; Kleeberg and Dudel, 1997; Golterman, 2004). This was also confirmed by research conducted in this lake in previous years and in other lakes in the Wielkopolska Region: Uzarzewskie Lake, Strzeszyńskie Lake, Sławskie Lake or Rusałka Reservoir (Kowalczewska-Madura and Gołdyn, 2009; Kowalczewska-Madura *et al.*, 2008, 2010a,b, 2011, 2015, 2017). The high rates of phosphorus release from the bottom sediments in this zone were also demonstrated by the high concentrations of this element in the interstitial waters, especially in summer and autumn; phosphorus concentration in pore waters is commonly regarded as an indicator of the intensity of its transport across the sediment–water interface (Kentzer, 2001; Komatsu *et al.*, 2006). This relationship was also found in previous research of this lake (Kowalczewska-Madura and Gołdyn, 2012). It is important that formerly there had been intensive primary production of phytoplankton in the lake; therefore, there was significant sedimentation to the bottom sediments (Kozak *et al.*, 2014). As stated by Katsev and Dittrich (2013) in Lake Sempach, intensive phosphorus release followed a higher sedimentation of organic matter. As a result of the intensive decomposition of organic matter, an oxygen shortage could be observed, which caused a significant phosphorus release from the bottom sediments. The released phosphorus remained in the over-bottom waters until the mixing period in the lake. In the Lower Havel River the seasonal course of P gross release was mainly driven by redox independent organic matter mineralisation of diagenetically young surface sediment (Grüneberg *et al.*, 2015). Moreover, the factor which determined significant phosphorus release was the amount of the 3 most mobile phosphorus fractions (NH_4Cl-P, BD-P and NaOH-P), whose total mean value equalled 15.2%.

High phosphorus release from bottom sediments was also observed in the zone located at a depth of 3–5 m (station 2). Significant decomposition of organic matter occurred in this area in summer, which caused a drop in oxygen concentration in the overlying water (Rosińska *et al.*, 2018) and phosphorus release from the bottom sediments. The intensity of the process was slightly lower than in the deepest place in the lake. When the oxygen conditions improved in other seasons and the water temperature dropped, a decline of phosphorus internal loading and even slight phosphorus accumulation in bottom sediments could be observed. Similar seasonal changes had been observed in a former study of this lake in the period 2001–2003 (Kowalczewska-Madura and Gołdyn, 2009). The percentage of phosphorus fractions of the highest biological availability (NH_4Cl-P, BD-P and NaOH-P), approximately 12.0%, determined the intensity of phosphorus release from the bottom sediments. Comparison of the mean annual values of phosphorus loading from this zone to the water column with such loading from the deepest zone in the lake shows that they were similar, and in particular years (2011 and 2013) even higher than in zone I. This was due to the fact that zone II was twice as large as zone I.

In the shallow zone III (station 3), the process of phosphorus release from the bottom sediments could also be observed, although the waters overlying sediments were well oxygenated. It was characterised by significantly lower intensity than in the other zones of the lake. The process of phosphorus release from bottom sediments to the water column is a common one, which also occurs in aerobic conditions (Boström *et al.*, 1982; Kleeberg and Dudel, 1997; Jiang *et al.*, 2006). According to Hupfer and Lewandowski (2008) the temporal existence of a thin oxidised sediment surface layer could only affect fluctuation of the temporary P pool at the sediment surface but not long-term P retention. P release was low in zone III and its highest intensity occurred in summer. When the water temperature over the sediments was high, short-term changes in the sediment–water interface were possible, *e.g.* a decrease in oxygen concentration. In shallow lakes, rapid, short-term changes of aerobic conditions in the above lying water, short-lived thermal stratification and mixing caused by turbulence are important (Søndergaard *et al.*, 2001), and may cause a gradient of phosphate concentrations in the sediment–water interface and may also increase phosphorus release. In colder seasons, however, the accumulation of phosphorus in the bottom sediments predominated.

A similar variability in the process could be found in the bottom sediments of the shallow, elongated zone with an abundance of macrophytes (zone IV with station 4). The amount of phosphorus loading in this zone was slightly higher than in the littoral zone III, situated in the main part of the lake. Possibly, this was a result of the intensive decomposition of organic matter whose content in the sediments was much higher than in the previous zone due to the development of macrophytes (Rosińska and Gołdyn, 2015). In addition, the high concentration of phosphorus in the water of the Mielcuch Stream flowing into this zone also affected phosphorus content. The phosphorus release in the shallow part of the lake covered with macrophytes could also be a result of the rather high pH (up to 8.6) due to photosynthesis present in this zone in summer (Rosińska *et al.*, 2018). An intensive production of phytoplankton or macrophytes in eutrophic lakes contributes to pH increase in water, and, therefore, also in the surface layer of the sediments, which leads to phosphorus release from bottom sediments, mainly phosphorus bound with iron (Boström *et al.*, 1988). A minor phosphorus release from the shallow zones of Swarzędzkie Lake could be caused by the lower participation of the phosphorus fraction of the highest biological availability, which in this case did not exceed 10%. The low intensity of the internal loading process is also indicated by the low concentration of phosphorus in the interstitial waters of the sediments in this zone.

Two zones release the most phosphorus: zone I, the deepest place in the lake (mean 33.5%), and zone II between 3 and 5 m (mean 35.4%), while the littoral zone is also a source of phosphorus – approximately 18.5% and 12.6% from zones III and IV, respectively. According to James and Barko (1991) the littoral sediments may deliver on average 26% of the phosphorus load in the summer period. Shallow lakes usually have extensive littoral areas, so the littoral sediments play an important role in P cycling (Andersen and Ring, 1999). 96% of the internal phosphorus load in the hypereutrophic kettle shaped Uzarzewskie Lake was delivered from the profundal zone and only 4% from the littoral zone. The littoral sediments delivered 18% of the P load in the case of Strzeszyńskie Lake with an elongated littoral zone, although it was a mesoeutrophic lake (Kowalczewska-Madura *et al.*, 2015). Research conducted in eutrophic Rusałka Lake indicated a much greater 86% share of phosphorus loading in the littoral zone, mainly due to its size, amounting to 92% of the bottom (Kowalczewska-Madura *et al.*, 2010b, 2011).

The changes in the internal loading of phosphorus in particular years of restoration were unexpected, especially its significant increase at the beginning of this process. This increase, observed especially in the profundal zone (stations 1 and 2), could be caused by the aerator used for the oxygenation of waters overlying the sediments, which probably led to the multiplication of heterotrophic bacteria that increased the mineralisation of organic matter. The mineralisation process of sinking organic matter already begins in the water column (Lampert and Sommer, 1999). In a hypereutrophic lake this process occurs under anaerobic conditions, but it is not very intense (Kentzer, 2001). The oxygen provided by the aerator must significantly intensify this process, as a result of which an additional SRP load is released into the water. The intensity of the internal loading process fell in the following years as the sedimentation of fresh organic matter decreased due to the limitation of phytoplankton growth, presented by Rosińska *et al.* (2017a). Similar changes in the intensity of phosphorus release from deep water sediments were found in Uzarzewskie Lake (Kowalczewska-Madura *et al.*, 2008) and in Rusałka Reservoir (Kowalczewska-Madura *et al.*, 2011), although no aerator for deep water oxygenation was used there. It is probable that increased phytoplankton sedimentation due to the iron treatment was more important in increasing phosphorus release in Swarzędzkie Lake than oxidation by the aerator.

The shallow south-west part of the lake, highly overgrown by macrophytes, functioned slightly differently. The phosphorus internal loading slightly increased there in 2011–2013, especially in summer, and it fluctuated only a little over the following years. Submerged macrophytes, which may occur in great numbers in shallow lakes, have both a negative and positive impact on the phosphorus cycle (Frodge *et al.*, 1991; Kuczyńska-Kippen, 2001). Oxygen released from the roots may increase redox-sensitive phosphorus sorption. Nevertheless, in the case of a high abundance of macrophytes on the bottom, which decrease the oxygen concentration in the water overlying the sediment (Kuczyńska-Kippen and Klimaszyk, 2007), phosphorus release may increase (Stephen *et al.*, 1997; Søndergaard *et al.*, 2001). On the other hand, the thick macrophytes growing in this part of the lake produce a large amount of organic matter which, after decay, is deposited in the bottom sediments, increasing the amount of TP there in the successive years of the study. It is not surprising that the sediments from this zone contained the highest organic matter content, stimulating internal phosphorus loading (Golterman, 2004). Pollutants (especially high nutrient concentrations) in the Mielcuch Stream, which enters this part of the lake, may also have an effect on P release from bottom sediments in this zone.

According to Kleeberg *et al.* (2013) iron treatment can increase the redox-dependent sorption of phosphorus which contributes to reducing the trophic state. P release is not relevant for the supply of the epilimnion, since during mixing periods most P is coprecipitated by the concurrently accumulated Fe in the hypolimnion. Besides external and internal P sources to be precipitated in a stoichiometric Fe:P ratio of 5, additional Fe equivalents of 25% are required for sedimentary organic carbon and to bind soluble sulphides. The Fe:P ratio in the sediments of Swarzędzkie Lake in 2012–2015 decreased from 10.99 to 0.48 at station 1 and from 14.6 to 0.44 at station 4. According to Jensen *et al.* (1992) the higher the Fe:P ratio in surface sediment, the lower the P release rate. If the ratio Fe:P is above 15 (by weight) it may be possible to control internal P loading by keeping the surface sediment oxidised. They argue that since the Fe:P ratio is easy to measure it may be a useful tool in the management of shallow lakes. They found that release of SRP was negatively correlated with the surface sediment Fe:P ratio. The capability of aerobic sediments to buffer phosphate concentration correlated with the Fe:P ratio, while the maximum adsorption capacity correlated with total iron. Thus, according to Jensen *et al.* (1992) the Fe:P ratio may provide a measure of free sorption sites for orthophosphate ions on iron hydroxyoxide surfaces. Concentration of Fe in bottom sediments in the case of Swarzędzkie Lake systematically decreased in the following years of restoration treatment, while the phosphorus content gradually increased. The decrease of iron content in bottom sediments most likely resulted from its release from sediments during periodic oxygen depletion in the bottom sediments in summer and lower iron dosage in 2013–2014 as well as the total exclusion of dosing in 2015–2016. This resulted in a decreasing Fe:P ratio. Nevertheless, the internal loading of phosphorus markedly fell in the following years. It seems that both the concentration of iron in the sediment and the ratio of Fe:P is not as important for the release of phosphorus from the bottom sediment as the phosphorus content in the individual fractions. As we have shown, the increase of phosphorus in sediments was not related to iron content, since the BD-P fraction did not increase, but it was connected with phosphorus contained in organic matter (NaOH-NRP) and bound to calcium and magnesium (HCl-P). The increase of organic matter was associated with a more intense sedimentation of organic suspensions as a result of iron treatment and zooplankton grazing (Pociecha and Wilk-Woźniak, 2008; Jurajda *et al.*, 2016). The increase of HCl-P was due to the use of magnesium chloride in the restoration process, probably resulting in the formation of struvite (insoluble magnesium ammonium phosphate $MgNH_4PO_4 \cdot 6H_2O$). In this way, the sorption capacity of small amounts of iron in the sediments was sufficient to prevent the release of mobile phosphorus into the water column. It follows that low doses of iron but with the addition of low doses of magnesium chloride used in sustainable restoration can be as effective as the very high doses of iron. However, as the chemical phosphorus bonding process was slow and the sedimentation of organic matter was most intense at the beginning of the restoration (2012), this resulted in the release of part of the mobile phosphorus generated by the mineralisation of this fresh organic matter to the water column. This confirms that an important part of the released P could originate from the mineralisation of settling fresh organic matter. Fortunately, this increased loading did not cause a water bloom (Rosińska *et al.*, 2017a) as the repeated phosphorus inactivation treatments led to its reprecipitation to the sediments. The doses of chemicals (especially PIX) used in the case of sustainable restoration in Swarzędzkie Lake were low (on average $3\,kg\,ha^{-1}$) in comparison to the doses used in the restoration of other lakes (Kleeberg *et al.*, 2012, 2013).

It was surprising that in the final year of the study the mean values of phosphorus internal loading were almost the same at all stations, contrary to the previous years when they distinctly differed. This indicates that the release of phosphorus from the deep bottom sediments has been considerably reduced by the sustainable restoration to the level found in the oxygenated bottom of the littoral zone. However, in our opinion this is a state of unstable equilibrium, maintained by the conducted restoration. This is confirmed by the observed changes when 2 out of 3 restoration treatments were discontinued. The amount of sulphates, iron, calcium and magnesium in the sediments started to decrease. Also, the concentration of phosphorus in the interstitial and the waters overlying the sediments increased. Internal loading reached higher values in the summer period of 2016 at all stations than in the same period of the previous year. Moreover, when the inactivation of phosphorus stopped, the P loading from the littoral zone began to increase. This suggests that no stabilisation of the ecosystem has yet been reached, which would allow for the completion of restoration treatments. A similar reaction to the inactivation of phosphorus was observed in Uzarzewskie Lake (Kowalczewska-Madura *et al.*, 2017).

5 Conclusions

We may conclude, therefore, that the sustainable restoration treatment of Swarzędzkie Lake contributed to a distinct decrease of phosphorus internal loading, especially from the sediments in the deeper part of the lake. It was variable over time because its increase at the beginning of restoration, especially in summer periods, was probably due to the increased sedimentation of organic matter and its intensive mineralisation associated with oxygen delivery by the aerator.

A significant decrease of internal loading was observed in subsequent years of restoration up to the final year of the study because the sustainable restoration (pulverizing aerator, the use of iron sulphate and magnesium chloride) increased the bottom sediment sorption complex and intensified chemical phosphorus bonding. It is necessary to continue the sustainable restoration for a longer period in order to maintain the trend of reducing internal phosphorus loading and thus improve water quality in the lake. Limiting it to only one method (deep water aeration) after 3 yr of phosphorus inactivation will lead in the near future to an increase of internal phosphorus loading and the return of cyanobacterial water blooms.

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