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Trace elements in Turkusowe Lake waters and bottom sediments (Wolin National Park, Poland)

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Abstract. Turkusowe Lake is a pit lake formed after the extraction of $CaCO_3$, located within the Wolin National Park (northwestern Poland). The aim of the study was to assess the potential contamination of water and bottom sediments with trace elements (TE) in relation to previous research, which indicated the impact of anthropogenic pressure. To achieve this, basic physico-chemical parameters of the water (pH, temperature, electrical conductivity, oxygen content) were measured, and the concentrations of TEs (Ag, As, Ba, Be, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb, Tl, V, Zn) in water and bottom sediments. The potential contamination of water by trace elements was evaluated using the trace metal evaluation index (TMEI), while sediment contamination was assessed using the geoaccumulation index (I_{geo}) and contamination factor (CF). The results of the TMEI and I_{geo} did not indicate contamination, whereas the CF pointed to low, initial contamination of the bottom sediments by Ni and V. It has been demonstrated that the lower layer of the lake becomes contaminated by TEs more quickly, in contrast to the upper layer, which undergoes faster eutrophication. Turkusowe Lake continues to demonstrate significant resistance to TEs contamination due to its alkaline pH, but regular monitoring is recommended.

Keywords: pit lake, trace elements (TEs), contamination of water and sediments, anthropogenic pressure, protected area, Wolin National Park.

Introduction

Turkusowe Lake is one of the most significant attractions in Wolin National Park, as highlighted by various authors (e.g. Rosa, 2011; Janeczko et al., 2013; Abramowicz & Woźniewicz, 2019; Żyto, 2019a, b; Zwoliński et al., 2024). This pit lake was formed after World War II when a chalk quarry was flooded. Until the end of the war, marl was extracted here for cement production at a now-defunct plant near the village of Wapnica (Poleszczuk et al., 2013). The lake was created without any organized reclamation strategy, filling naturally after dewatering systems were stopped. Alkaline pit lakes, like those formed after $CaCO_3$ extraction, are of particular scientific interest due to their rarity and notable resistance to eutrophication (e.g. Raczyńska & Kubiak, 2003; Poleszczuk et al., 2013, 2014). Turkusowe Lake's long-standing oligotrophic state and the resulting water clarity give it a distinctive turquoise color, which is the source of its name. This coloration is caused by the dispersal of sunlight reflected from the white Cretaceous chalk in the lake bottom and calcareous lake sediments (Zwoliński et al., 2024). The lake is currently protected as part of one of six strict conservation areas within Wolin National Park, specifically the Professor Władysław Szafer Strict Protection Area (Abramowicz & Woźniewicz, 2019).

Despite its general resistance to eutrophication, research by Poleszczuk et al. (2014) indicated that around 2010, Turkusowe Lake had reached a mesotrophic state due to increasing trophic levels. This change is attributed to intensified anthropogenic pressure (Poleszczuk et al., 2005, 2013, 2014), which persists despite the strict legal protection of the site (Abramowicz & Woźniewicz, 2019). Key sources of this pressure include the illegal discharge of municipal sewage from private properties through unauthorized pipelines from the village of Wapnica on the western shore (just outside the park's boundaries) and the discharge of sewage from the village of Lubin into the ground within the recharge zone of the springs that feed Turkusowe Lake (Poleszczuk et al., 2005, 2014). Additionally, the lake may have been exposed to atmospheric deposition (i.e. acid rain) resulting from the operations of the "Police" Chemical Plants near Szczecin and the "Dolna Odra" Power Station Complex near Gryfino (Poleszczuk & Jakuczun, 1996).

Studies conducted in other National Parks in Poland confirm that, despite their protected status, lakes can still experience anthropogenic pressure (e.g. Kurzyca et al., 2009; Walna & Siepak, 2012; Sojka et al., 2021, 2022, 2024). One of the main concerns is the introduction of trace elements (TEs) (Sojka et al., 2022). Research indicates that TEs can enter National Parks through several pathways, including contaminated rivers (Sojka et al., 2022), migration of polluted groundwater (Walna & Siepak, 2012), tourist activity (Memoli et al., 2019), and both wet and dry atmospheric deposition (Walna & Siepak, 1999; Korzeniowska & Krąż, 2020). Given the confirmed anthropogenic pressure on Turkusowe Lake, it became necessary to investigate whether its waters and bottom sediments have been contaminated by TEs and to update knowledge on the lake's ongoing degradation.

The goals of the study were: (1) to examine the content of TEs in waters and bottom sediments, (2) to identify potential water and bottom sediment contamination of the lake using indexes, (3) to expand the knowledge about the processes responsible for shaping the geochemistry of the lake, (4) to expand knowledge on the progressive degradation of the lake and (5) to identify potential threats to the tourism potential of Turkusowe Lake.

Materials and methods

Reaserch area

Turkusowe Lake is situated south of the village of Wapnica, within Wolin National Park (Fig. 1), specifically in the Professor Władysław Szafer Special Protection Area (Abramowicz & Woźniewicz, 2019). Wolin National Park encompasses a large portion of Wolin Island, which is part of the Uznam and Wolin Islands mesoregion (Solon et al., 2018). Due to the tourist significance of Turkusowe Lake (Janeczko et al., 2013; Żyto, 2019a, b), its surroundings have been developed accordingly. The surrounding tourist infrastructure includes parking, dining facilities, a viewpoint, and essential amenities such as tourist maps, information boards, rain shelters, benches, tables, and a children's playground (Rosa, 2011). Additionally, the lake is located along two blue-marked tourist trails: a pedestrian trail no. 2 (Międzyzdroje-Wolin) and a cycling trail (Świnoujście-Wolin) (Żyto, 2019b).

Turkusowe Lake is a pit lake, formed approximately 80 years ago within a former $CaCO_3$ quarry after World War II (Poleszczuk et al., 2014). The lake's mining origins are reflected in its morphology. The shores are bordered by hills that rise up to 30 meters above sea level, with steep slopes that are prone to erosion due to mass movements (Włodarczyk et al., 2008). These high slopes, along with the trees growing on them, create shaded areas and shelter the lake from wind, leading to persistent chemical and thermal stratification of the water throughout most of the year (Poleszczuk et al., 2013). The lake covers an area of about 6.2 hectares, and its current depth is approximately 12.1 meters (based on own measurements), which indicates ongoing sedimentation and shallowing (Fig. 2; Włodarczyk et al., 2008). This process may result from water level fluctuations, mineral precipitation (mainly carbonates and phosphates) from the water column, and mass movements along the shores.

The lake is primarily fed by atmospheric precipitation, surface runoff, and groundwater inflows, some of which originate from numerous springs located at the base of the surrounding hills. Additionally, Turkusowe Lake receives water from the smaller "Upper

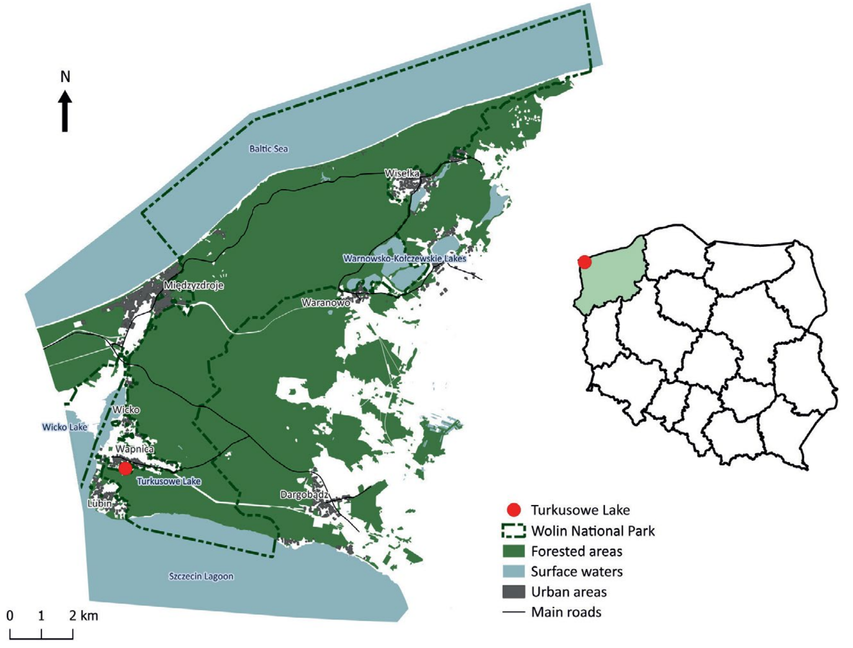

Fig. 1. Location of Turkusowe Lake

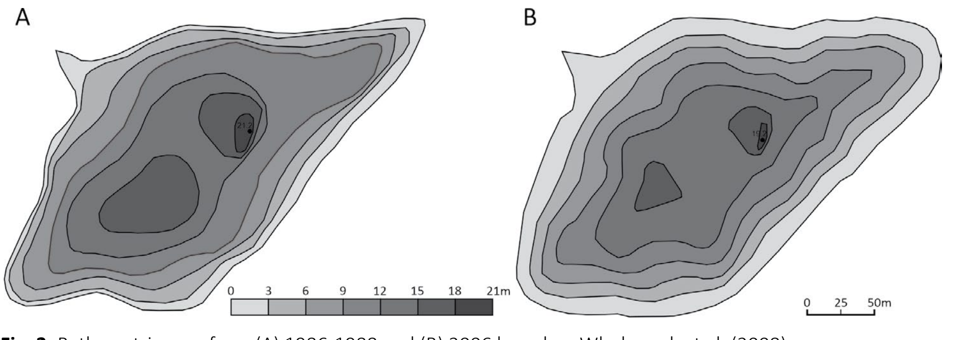

Fig. 2. Bathymetric map from (A) 1986-1988 and (B) 2006 based on Włodarczyk et al. (2008)

Turkusowe Lake" (Poleszczuk et al., 2013). Groundwater contaminated by municipal sewage from the villages of Lubin and Wapnica is the main source of pollution in the Turkusowe Lake catchment area (Poleszczuk et al., 2005, 2014). This contamination has caused the lake to shift from an oligotrophic to a mesotrophic state (Poleszczuk et al., 2014). Excess water from Turkusowe Lake drains through the soil layers on the northwest side, eventually flowing towards Wicko Lake (Poleszczuk et al., 2005).

Sample collection and preparation

Water and bottom sediment sampling took place on 10.08.2024. A total of 12 water samples were collected: 1 surface sample (10 cm below the water table) and additional samples at every 1-meter depth in a vertical profile. The USB 50015 sampler (UWITEC GmbH, Mondsee, Austria) was used for this purpose. The samples were placed in 250 ml Nalgene® polyethylene (HDPE) bottles and preserved *in situ* with 60% HNO_3 Ultrapur® (to pH <2). After collection, the samples were transported to the chemical laboratory in a car cooler at a temperature of about 4 ± 2.5°C. An Ekman sediment grab was used to collect the bottom sediments, and the surface layer of the sediment was placed in a ziplock bag. To prepare the bottom sediments for analysis, they were dried at 105°C using drying oven (Digitheat, J.P. Selecta, Spain), and TEs were extracted using 1:4 HCl (Sojka et al., 2019; Śniady et al., 2024). Additionally, during *in situ* sampling, basic physico-chemical parameters of the water were measured: pH, temperature, conductivity (SPC) and dissolved oxygen (DO). The ProDSS Multiparameter Digital Water Quality Meter by YSI (Ohio, USA) was used for this purpose.

Chemical analysis

The determination of TEs (Ag, As, Ba, Be, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb, Tl, V, Zn) in water and bottom sediments was performed using inductively coupled plasma mass spectrometry (ICP-QQQ-MS), model 8800 Triple Quad, from Agilent Technologies (Japan). The analyses were conducted in two gas modes: O_2 at a flow rate of 0.3 mL/min (30%) and He at a flow rate of 5 mL/min. All parameters were manually optimized to achieve the best signal intensity and stability. A 1% HNO_3 solution (Ultrapur, Merck, Darmstadt, Germany) was used as the continuing calibration blank (CCB) sample. Calibration solutions were prepared by appropriately diluting multi-element calibration standards. The MassHunter software for ICP-QQQ-MS (Agilent Technologies, Japan) was used to control the instrument and process the data. The analytical procedures and instrumental operating parameters used are detailed in the works of Walna and Siepak (2012), Siepak and Sojka (2017), Sojka et al. (2021), and Sojka et al. (2024).

Reagents

During the ICP-QQQ-MS analysis, multi-element standard solutions from VHG Labs (Manchester, USA) with a base concentration of 100 µg/mL were used. Ultrapure reagents and deionized water with a resistivity of 18.2 MΩ·cm, purified using the Direct-Q® 3 Ultrapure Water System (Millipore, France), were employed. To verify the accuracy of the obtained results, certified reference materials 1643f (National Institute of Standards and Technology, USA), SPS-SW2 (Spectrapure Standards As, Oslo, Norway) and CRM No. LGC 6187 for River sediments (Manchester, England) were used.

Assessment of trace element contamination

To assess the potential contamination of the Turkusowe Lake waters, the obtained results were compared with the standards for specific and priority substances as outlined in the Regulation of the Minister of Infrastructure (2021). The Trace Metal Evaluation Index (TMEI) was employed for this purpose. It is defined as the ratio of the monitored trace element concentration ($TM_{Conc.}$) to its maximum permissible concentration (TM_{MPC}) (Zakir et al., 2020). This index is calculated using the following formula:

$$TMEI = \sum_{i=1}^{n} \frac{TM_{Conc.}}{TM_{MPC}}$$

To assess water contamination by TE, a threshold value of 1.0 was used. Values above 1.0 indicate a poor condition of the lake, while values below 1.0 suggest a good condition (Zakir et al., 2020).

For evaluating sediment contamination by TE, the Geoaccumulation Index (I_{geo}) and the Contamination Factor (CF) was employed (Müller, 1981; Sojka et al., 2019). The I_{geo} classifies sediments into one of seven categories, ranging from 0 (uncontaminated) to 6 (extremely contaminated), while the CF classifies sediments into one of four categories, from 1 (uncontaminated) to 4 (heavily contaminated). They are calculated using the following formulas:

$$I_{geo} = \log_2 \left(\frac{C_i}{1,5 \times B_i} \right)$$

$$CF = \frac{C_i}{B_i}$$

where C_i represents the concentration of the specific TE in the sediment [mg/kg], and B_i refers to the geochemical background level. The adopted geochemical background values are as follows: Ag – 1 mg/kg, As – 5 mg/kg, Ba – 16 mg/kg, Be – 0.5 mg/kg, Cd – 0.5 mg/kg, Co – 1 mg/kg, Cr – 2 mg/kg, Cu – 3 mg/kg, Ni – 2 mg/kg, Pb – 7 mg/kg, V – 2 mg/kg, Zn – 29 mg/kg (Pasieczna, 2012), and Mo – 0.63 mg/kg, Sb – 0.27 mg/kg, Tl – 0.12 mg/kg (Salminen et al., 2005).

Results

Basic physico-chemical parameters

The results of the physico-chemical water parameters measured in the vertical profile are presented in Figure 3. The highest recorded temperature, 22.4 °C, was observed at the water surface. The lake water exhibits isothermy down to approximately 5 meters, with the temperature remaining just above 22 °C. Below this depth, the temperature decreas-

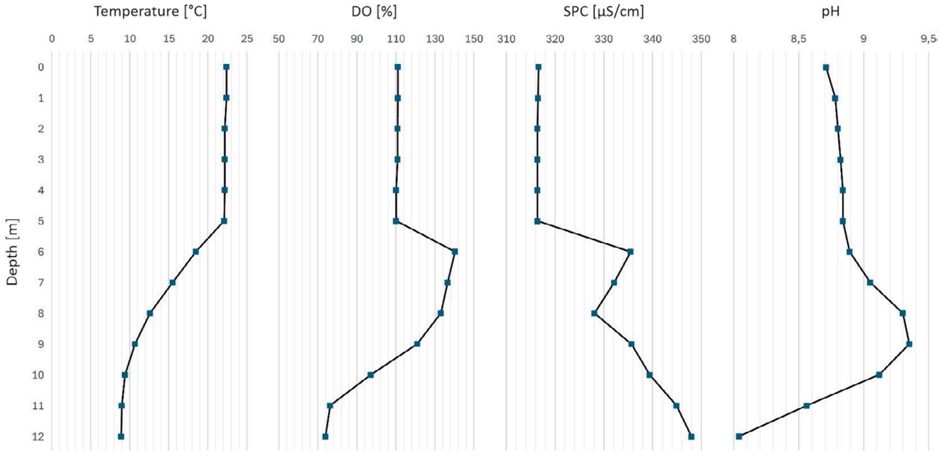

Fig. 3. Variability of basic physico-chemical parameters with depth: temperature, dissolved oxygen (DO), conductivity (SPC) and pH

es significantly, stabilizing at 9 °C at a depth of 11 meters. The lowest temperature, 8.9°C, was recorded at the lake bottom (Fig. 3).

Dissolved oxygen (DO) levels are relatively high. From the water surface to a depth of about 5 meters, the DO concentration remains stable at around 111%, then slightly decreases to 110%. Below this depth, the DO concentration increases sharply, peaking at 140% at 6 meters. In the deeper layers, oxygen levels gradually decline, with the lowest concentration of 73.9% recorded at the lake bottom (Fig. 3).

Electrical conductivity at the water surface is 316.6 µS/cm. This value remains relatively constant down to a depth of 5 meters, where the lowest conductivity, 316.4 µS/cm, was observed. Below 5 meters, conductivity increases, ranging between 328.1 µS/cm and a maximum of 348 µS/cm near the lake bottom (Fig. 3).

The pH of Turkusowe Lake is another critical physico-chemical parameter. The surface pH is 8.71 and increases gradually with depth, reaching a peak of 9.35 at 9 meters. Below this depth, pH levels drop significantly, reaching 8.04 at the lake bottom (Fig. 3).

Trace elements content

In general, the concentration of TEs in the waters of Turkusowe Lake increased in the following order: Tl<Ag<Co<Cd<Be<Cr<V<Sb<Ni<Mo<Pb<As<Cu<Zn<Ba. The lowest concentrations were observed for Tl at all depths (ranging from 0.002 to 0.017 µg/L), while the highest concentrations were found for Ba (ranging from 4.18 to 7.73 µg/L). Most of the analyzed TEs showed a slight decrease in concentration from the surface to a depth of approximately 5 meters. At a depth of 6 meters, an increase in the concentration of most TEs was observed (Fig. 4), which correlates with changes in the physico-chemical parameters of the water. This was followed by a decrease in TE concentrations down to a depth of 9 meters, and then a further increase near the bottom. All TEs, except Ag, reached their highest concentrations at the bottom of the lake.

The TE content in the bottom sediments of Turkusowe Lake can be arranged in ascending order as follows: Sb<Mo<Tl<Be<Cd<Ag<Co<As<Cr<Ba<Cu<Ni<V<Pb<Zn. The lowest concentration, 0.01 mg/kg, was recorded for Sb, whereas the highest, 8.54 mg/kg, was observed for Zn. The concentrations of individual TEs in the bottom sediment are

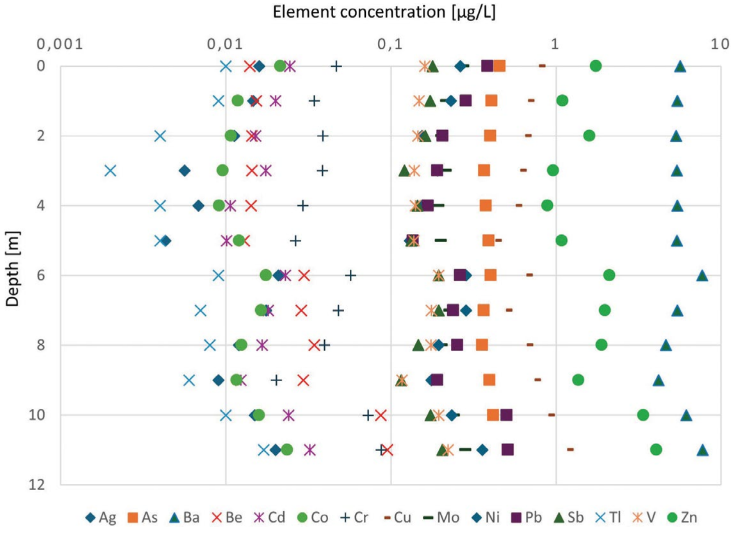

Fig. 4. Variability of TE concentrations with depth

Table 1. Trace element (TE) concentrations in bottom sediments, Geoaccumulation index (I_{geo}) results and Contamination Factor (CF) results

Parameter	Value [mg/kg]	I_{geo}	I_{geo} contamination class	CF	CF contamination class
Ag	0.07	-4.41		0.07	
As	0.83	-3.18		0.17	
Ba	0.92	-4.71		0.06	
Be	0.04	-4.19		0.08	
Cd	0.04	-4.08		0.09	uncontaminated
Co	0.75	-0.99		0.75	
Cr	0.85	-1.82		0.42	
Cu	1.21	-1.89	uncontaminated	0.40	
Mo	0.02	-5.96		0.02	
Ni	2.10	-0.51		1.05	low
Pb	5.91	-0.83		0.84	
Sb	0.01	-5.13		0.04	uncontaminated
Tl	0.04	-2.24		0.32	
V	2.13	-0.50		1.06	low
Zn	8.54	-2.35		0.29	uncontaminated

presented in Table 1. In comparison to the water studies, it should be noted that, proportionally relative to other elements, the concentrations of Pb, Ni, V, and Cr are higher.

Contamination assessment

The conducted analysis of water pollution in Turkusowe Lake revealed that the water does not exceed the standards established in the Regulation of the Minister of Infrastructure (2021) for specific and priority substances. The TMEI index used reflects the trends observed in the variability of TE concentrations (Fig. 4). A decrease in TMEI values is visible from the surface down to 5 meters (Tab. 2). Subsequently, the TMEI value increases at 6 meters and then gradually decreases again down to a depth of 9 meters. The highest TMEI concentrations are reached near the bottom of Turkusowe Lake. It should be noted that, despite the varying TMEI values in the vertical profile, the lower part of the profile generally shows poorer water quality compared to the upper part.

The evaluation of sediment contamination in Turkusowe Lake using I_{geo} indicated an absence of contamination (Tab. 1), with I_{geo} values ranging from -5.96 (Mo) to -0.5 (V). The CF analysis produced largely similar results for most elements (Ag, As, Ba, Be, Cd, Co, Cr, Cu, Mo, Pb, Sb, Tl, Zn), with CF values ranging from 0.02 (Mo) to 0.84 (Pb). However, CF identified low contamination for Ni and V, with values slightly exceeding the threshold for Class II ($1<CF<3$).

Table 2. Trace metals evaluation index (TMEI) results for water

Depth [m]	0	1	2	3	4	5	6	7	8	9	10	11
TMEI	0.20	0.18	0.17	0.14	0.15	0.14	0.22	0.21	0.19	0.16	0.30	0.35

Discussion

Wolin National Park is particularly valuable due to its biogeographical and geodiversity features (Czyryca et al., 2021; Zwoliński et al., 2024), with Turkusowe Lake being one of its most important tourist attractions (Rosa, 2011; Janeczko et al., 2013; Żyto, 2019a, b). Despite its strict protection status (Abramowicz & Woźniewicz, 2019), Turkusowe Lake is subject to ongoing anthropogenic pressure of varying intensity (Poleszczuk & Jakuczun, 1996; Poleszczuk et al., 2005, 2013, 2014), with one of the most significant issues being the inflow of water contaminated with municipal sewage from the villages of Wapnica and Lubin (Poleszczuk et al., 2005, 2014). This is likely a result of poor management of the areas surrounding the National Park. The inflow of sewage and sewage-contaminated water into lakes leads to their degradation through intensified eutrophication, contamination with inorganic substances, and disruption of the delicate balance of freshwater ecosystems (Bhat & Qayoom, 2021; Bamaniya et al., 2023). However, alkaline lakes are generally considered highly resistant to eutrophication (e.g. Raczyńska & Kubiak, 2003; Poleszczuk et al., 2013, 2014; Marszelewski et al., 2017). This resistance is primarily due to the strong ability of $CaCO_3$, abundant in these lakes, to bind (chemisorb) phosphorus compounds (Flower et al., 2022). TEs introduced along with other pollutants in alkaline water environments are typically bound as oxides, hydroxides, or sulfates, which then settle in the sediment (Takeno, 2005; Linnik et al., 2023). This is confirmed by the results of the study, which indicate low concentrations of the studied TEs in the waters of Turkusowe Lake.

The precipitation of introduced trace elements TEs into the bottom sediments likely accounts for the exceptionally low TMEI values observed for Turkusowe Lake. The assessment of sediment contamination using the widely recognized I_{geo} method did not reveal any significant contamination. However, the simpler CF calculation method (Sojka et al., 2019; Śniady et al., 2024) indicated initial contamination of the sediments with Ni and V (Table 1). The elevated Ni concentrations may be associated with the inflow of sewage-contaminated water into the lake (Gupta, 2020; Bhat & Qayoom, 2021), while the presence of V could be linked to the characteristics of the underlying bedrock (Wright & Belitz, 2010) or the deposition of industrial dust within Wolin National Park (Slukovskii, 2023). Additionally, note the shift in dominant metals between the bottom sediment and water. The increased concentrations of Pb and Cr in the bottom sediments, along with elevated levels of Ni and V, suggest that these elements may originate from anthropogenic sources (Gupta, 2020).

Maintaining the good condition of alkaline lakes is generally linked to their pH buffering capacity. Despite the low concentrations of TEs, the lake's good ecological status may persist only until the buffering capacity is exhausted, at which point a pH drop is likely. This decrease in pH could result in the release of phosphorus bound in the sediment (Marszelewski et al., 2017) as well as TEs (Linnik et al., 2023), which would likely accelerate the eutrophication and degradation of Turkusowe Lake.

Additionally, the conducted studies, both of the basic physico-chemical parameters (Fig. 3) and the TE concentrations (Fig. 4), indicate the presence of stratification. Poleszczuk et al. (2014) noted that the waters of Turkusowe Lake rarely mix completely, with pronounced stratification observable throughout the year. This stratification is likely due to the lake's limited exposure to wind, resulting from the surrounding high slopes (Włodarczyk et al., 2008) and dense tree cover. It should be noted that the physico-chemical parameters remain relatively stable down to a depth of 5 meters, but below this depth, significant changes occur. In particular, there is an increase in dissolved oxygen at 6 meters and a decrease in pH near the bottom (Fig. 3). The increase in oxygen may be related to the presence of a thermocline, which hinders the mixing of waters with different densities, as well as possible microbial activity or photosynthesis leading to oxygen production (Spietz et al., 2015), enabled by the lake's mesotrophic status (Poleszczuk et al., 2013; 2014). The drop in pH near the bottom is likely the result of interactions between the calcareous sediment and the water. Generally, under conditions of high oxygenation, the dissolution of $CaCO_3$ should lead to an increase in pH (Morse & Arvidson, 2002). However, the exceptionally weak water mixing observed by Poleszczuk et al. (2013) could lead to localized CO_2 accumulation in the lower layers, a consequence of the lake's mesotrophic state (Poleszczuk et al., 2014). This localized CO_2 buildup could cause a decrease in pH due to the dissolution of $CaCO_3$ in CO_2-saturated water (Morse & Arvidson, 2002).

Due to the confirmed stratification, Poleszczuk et al. (2013, 2014) concluded that the upper layer of the lake would likely undergo faster eutrophication. However, the conducted analyses indicate that this does not apply to degradation by TEs, which accumulate in the lower layer of Turkusowe Lake. Additionally, it should be emphasized that the bottom sediments of Turkusowe Lake may become contaminated by TEs faster than the water, due to the precipitation of TEs into the sediments. This is supported by studies conducted in Bory Tucholskie National Park, which show significantly stronger anthropogenic pressure in the bottom sediments (Sojka et al., 2021, 2024) compared to the water (Sojka et al., 2020).

Monitoring the ongoing degradation of Turkusowe Lake and implementing preventive measures is crucial due to its unique biotic and abiotic environment (Wolender & Zych, 2006; Poleszczuk et al., 2013, 2014; König, 2017). Progressive eutrophication of the lake could lead to reduced water clarity, resulting in the loss of its distinctive turquoise color, which is a major tourist attraction and an important protected abiotic feature (Abramowicz & Woźniewicz, 2019). Additionally, the area surrounding Turkusowe Lake is home to many rare plant species (König, 2017), as well as ground beetles (Coleoptera: *Carabidae*) (Wolender & Zych, 2006). The introduction of TEs into the lake's waters could reduce biodiversity in the area and pose a threat to species sensitive to pollution. The failure to implement appropriate environmental protection measures for Turkusowe Lake could therefore pose a significant threat both to the environment and to the tourist appeal of this part of Wolin National Park.

Conclusion

The conducted studies, which involved examining the basic physicochemical parameters and TEs in the water and sediments of Turkusowe Lake, led to the following conclusions:

- the progressive eutrophication of Turkusowe Lake has not led to significant contamination by TEs,
- the measured trace element concentrations in the water did not exceed the limits set by the Regulation of the Minister of Infrastructure (2021), and the TMEI values ranged from 0.14 to 0.35,
- the calculated I_{geo} indicated no contamination, while the CF revealed initial contamination by Ni and V, which may be linked to anthropogenic pressure,
- the lower layer of the lake becomes contaminated by TEs more quickly, in contrast to the upper layer, which undergoes faster eutrophication,
- the bottom sediments of the lake are subject to faster contamination by TEs than the lake's waters due to their precipitation in response to the alkaline pH,
- regular monitoring of Turkusowe Lake is recommended to prevent the degradation of this highly important tourist site in Wolin National Park.

Unless otherwise stated, the sources of tables and figures are the authors', on the basis of their own research

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