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The Impact of the Watershed Use Changes on the Water Chemistry of the Shallow, Urban Lake—A Case Study of Lake Mielenko (Pomeranian Lakeland, Poland)

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Abstract: The research was carried out on the flow-through Lake Mielenko (7.8 ha; 1.9 m), which also acts as a stormwater receiver. In 2015, a disposal for road salts was created in the lake's catchment area. As a result of the inflow of salt-contaminated stormwater, there was a significant increase in the concentration of calcium (57 mg Ca/L), chloride (220 mg Cl/L) and electrolytic conductivity (790 µS/cm). Increased calcium concentrations in lake waters changed their hardness from low to medium-hard. The ecological effect of the change in hydrochemical conditions in Lake Mielenko is the *Potamogeton crispus* that grows abundantly in this reservoir, which prefer calcium-rich water. The overall aesthetics of the lake have deteriorated significantly, and the availability of water for recreation has also been limited.

Keywords: lake; contamination; chloride; electrolytical conductivity (EC)

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1. Introduction

Over 100 years of excessive human pressure on the environment has led to the degradation of many aquatic ecosystems, especially lakes [1,2]. It is well known that the lake water quality is related to the amount of supply and the chemistry (quality) of water flowing in from the catchment area [3,4]. In the natural environment, these parameters are determined by the nature of the bedrock and soil, the type of vegetation, and climatic conditions [5–7]. Therefore, human activity consisting of transforming the catchment area into agricultural, urban or industrialized areas, as well as the discharge of various types of wastewater into lake water, distort the natural chemistry of the water and rapidly accelerate the eutrophication process [8,9].

Apart from the risk of contamination of waters with an excess of nutrients, toxic substances, suspension, and acidification [10–15], there is a problem of salinity, which is associated with an increase in the concentration of mineral substances, including chlorides and EC [16,17]. Since the mid-twentieth century, salts such as NaCl salt; road salt being a mixture of NaCl and $CaCl_2$ and $K_2Fe(CN)_6$—potassium ferrocyanide added to prevent salt agglomeration; brine composed of $CaCl_2$ and $MgCl_2$ to eliminate winter slippery streets; and their use is constantly increasing [18]. In Poland, about 500,000 tons of road salt are used annually, and in the United States up to 12 million tons per year [18]. The salt used on the roads is washed away into lakes and rivers or into groundwater resources. Salt water infiltrating the ground can lead to salinization of drinking groundwater resources. It is a health problem for people on a sodium-restricted diet, and a taste problem for everyone else. When salt enters lakes and water courses, it can also harm aquatic plants and animals. The strong influx of sodium and chloride ions—which are formed when salt dissolves—will disrupt the ability of freshwater organisms to regulate the flow of fluids into their bodies. Changes in the salinity of a pond or lake can also affect water dynamics and lead to the formation of a monimolimnion and biologically dead zones above the bottom in deeper bodies of water. Road salt landfills are another problem. Road salt can be stored under a shelter or in the open air on a hardened, non-retractable base isolated from the inflow of moisture. Salt stored in the open air should be covered to protect against moisture precipitation [19]. Calcium chloride and magnesium chloride should be stored in packaging (foil bags or closed drums) placed in piles on a hardened floor and isolated from the inflow of moisture floor in a warehouse or under a shelter, or under cover in the case of outdoor storage. Mixtures of NaCl with $CaCl_2$ or $MgCl_2$, designed to combat winter slipperiness at temperatures below −7 °C, should be prepared immediately before loading onto spreaders [20].

Lake Mielenko is the flow-through water body. A small forest stream flows into it from the north-west, and the outflow leading to Karczemne Lake is on the north-eastern shore. Lake Mielenko also acts as a stormwater receiver. In 2015, a road salt disposal area was created in the lake's catchment area. The disposal has no secured ground and is not roofed. The situation dramatically worsened the quality of water in the reservoir due to excessively high salinity. The aim of the study was to present changes in the chemistry of the water of Lake Mielenko (on the example of selected ions determining the EC of water), which occurred in a short time (several years) as a result of increased anthropopressure in the catchment area of the lake and the impact of these changes on the ecological state of the water body.

2. Material and Methods

2.1. Study Site

Lake Mielenko is located in the Kashubian Lake District, which is the westernmost part of Pomerania [21]. The lake area is 7.8 ha (Table 1). It is a very shallow water body with a maximum depth of only 1.9 m and an average depth of 1.3 m (Figure 1).

Table 1. Morphometric data of Lake Mielenko.

Parameter	Value
Surface water table	7.8 ha
Water volume	102.9 thousand m^3
Maximum depth	1.9 m
Average depth	1.3 m
Relative depth	0.0068
Depth index	0.68
Maximum length	460 m
Maximum width	252 m
Elongation	1.8
Average width	170 m
Length of shoreline	1314 m
Shoreline development	1.3

Lake Mielenko basin, containing 102,900 thousand m^3 of water, is flat and relatively even. The value of the depth indicator—0.68—informs us that the bowl has the shape of a hemisphere.

The lake is poorly sunk into the ground, which is indicated by the low value of the relative depth—0.0068. The shoreline of the water body, 1314 m long, is poorly developed (K—1.3) (Table 1). The shores of the lake are relatively flat and in some places wet.

Lake Mielenko is a water body with extremely unfavorable natural conditions, which determine the rapid degradation of the lake in the event of excessive load of mineral and organic compounds flowing from the catchment area. The morphometric features of the lake favoring rapid degradation include a very low average depth and a high ratio of the active bottom surface from which nutrients are emitted to the volume of water in which primary production processes take place.

Figure 1. Lake Mielenko's location in Europe and Poland. Available online: https://www.google.pl/maps/@52.0666816,18.7171332,6z?hl=en (accessed on 1 July 2022).

A negative feature of the studied water body is also the lack of thermal stratification (polymictic lake) in the summer and the high value of the quotient of the lake volume (i.e., the mass of water into which contaminants from the outside enter) and the length of the contact line of the reservoir with the surrounding area (shoreline). Lake Mielenko is a flow-through water body. A small forest stream flows into it from the north-west, and the outflow leading to Lake Karczemne is on the north-eastern shore.

The total catchment area of Lake Mielenko covers an area of 3.82 km^2, and is dominated by forest complexes and wetlands. In the direct catchment area (0.22 km^2), forests predominate (65%), followed by meadows and pastures (35%).

2.2. Water Sample Collection and Analysis

The research of Lake Mielenko was carried out in 2013, 2019, 2020, and 2021. Samples were taken once a month, using a Ruttner sampler, at the points situated above the deepest part of lakes (1 m below water mirror) (Figure 1). In addition, samples of rain collector outlet (Ø 500 mm, a separator) were taken at one-month intervals during the period January to December in 2013, 2019, 2020, and 2021.

Flow velocity measurements were performed by using a VALEPORT (801 model) (Valeport Limited St. Peter's Quay, Totnes, Devon TQ95EW UK) electromagnetic flowmeter and determining the water discharge by using Harlacher's method [22].

The scope of the water analysis included: PO_4^{-3} (with the use of ammonium molybdate—$(NH_4)_6Mo_7O_{24}$ * $4H_2O$ and $SnCl_2$ as indicator λ = 690 nm—colorimetrically with the NANOCOLOR spectrophotometer) (GmbH&Co. KG, Düren, Germany), NO_3^-, Fe, Mn (Merck SQ118) (KGaA, Darmstadt, Germany), Ca (titration method with EDTA solution—$C_{10}H_{14}N_2O_8Na_2$ and $C_{20}H_{12}N_3O_7SNa$ as indicator), Cl (argentometric method with $AgNO_3$ solution and K_2CrO_4 as indicator), pH, and conductivity—EC (YSI 6600V2)

(Yellow Springs, OH, USA). Every analysis was performed in triplicate. The coefficient of variation (CV) for the repeated analysis was 2% [23].

The results of anions were PO_4^{-3}, NO_3^-, elements: Ca, Cl, Fe, Mn, and parameters: pH and EC were statistically analyzed (one–way ANOVA, p = 0.05, Tukey's HSD) using a Statistica 13.3 software package(Tibco Software, Palo Alto, CA, USA) [24]. An alternative hypothesis tested was the presence of significant differences in mean annual values of PO_4^{-3}, NO_3^-, Ca, Cl, Fe, Mn, pH, and EC between 2013 (inflow of stormwater) and 2019, 2020, and 2021 (after the establishment of a road salts disposal area).

3. Results and Discussion

In 2015, a road salts disposal area was created. The salts used are NaCl salt; road salt being a mixture of NaCl and $CaCl_2$ and $K_2Fe(CN)_6$—potassium ferrocyanide added to prevent salt agglomeration; and brine composed of $CaCl_2$ and $MgCl_2$. The inflow of storm water from the road salts disposal to Lake Mielenko caused a radical change in the water chemistry of this small and shallow water body.

The analysis of the obtained research results showed significant differences between the average annual loads of selected ions, which were introduced into the waters of Lake Mielenko before the change of the catchment development (2013), and the loads introduced after the creation of a road salts disposal area (Table 2).

Nitrate nitrogen loads decreased. The loads of phosphates, as well as calcium, iron and manganese fluctuated (increased or decreased), while the amount of introduced chlorides increased by more than thirty times. The loads depended mainly on the amount of rainwater, but also on the concentrations of individual ions that were recorded in the storm water. Noteworthy are the values of EC, which reached the value of 5452 µS/cm (Table 2).

3.1. Changes in Chemical Parameters

Statistical analysis showed statistically significant differences in the content of NO_3, PO_4, Ca, Cl and Fe ions in the waters of Lake Mielenko between 2013 (storm water inflow) and 2019, 2020, and 2021 (after creation of a road salts disposal) (Table 3). There were no statistically significant differences in manganese concentrations (Table 3).

Table 2. Characteristics of the selected parameters of rainwater flowing into Lake Mielenko.

Variables		Flow (L/s)	EC (µS/cm)	pH	NO_3 (mg/L)	PO_4 (mg/L)	Ca (mg/L)	Cl (mg/L)	Fe (mg/L)	Mn (mg/L)
2013	Mean	0.783	461	7.80	0.947	0.057	44.2	36	1.718	1.020
	SD	0.651	17	0.095	0.349	0.023	3.4	1	0.398	0.125
	Maximum	1.932	485	7.92	1.240	0.078	40.2	38	2.150	1.200
	Minimum	0.185	439	7.65	0.300	0.031	40.5	35	1.050	0.900
	AnLo (kg/y)	-	-	-	**23.4**	**1.4**	**1091.4**	**884.0**	**42.5**	**25.2**
2019	Mean	0.303	2264	8.52	0.641	0.142	83.3	532	0.880	1.050
	SD	0.055	272	0.025	0.132	0.014	3.8	39	0.010	0.010
	Maximum	0.360	2470	8.54	0.794	0.152	86.0	574	0.890	1.060
	Minimum	0.250	1956	8.49	0.564	0.125	79.0	495	0.870	1.040
	AnLo (kg/y)	-	-	-	**6.1**	**1.4**	**795.9**	**5081.0**	**8.4**	**10.0**
2020	Mean	0.910	3669	7.95	0.341	0.089	80.7	124	1.229	1.171
	SD	0.908	995	0.376	0.101	0.032	41.0	588	0.273	0.384
	Maximum	2.300	5452	8.31	0.508	0.141	156.1	2000	1.780	2.000
	Minimum	0.080	2371	7.17	0.150	0.051	37.0	570	0.920	0.700
	AnLo (kg/y)	-	-	-	**9.8**	**2.6**	**2315.9**	**35,585.0**	**35.3**	**33.6**
2021	Mean	1.662	2146	7.70	0.177	0.033	68.2	574	0.685	0.840
	SD	1.781	1591	0.092	0.116	0.026	28.4	479	0.626	0.606
	Maximum	3.600	3978	7.76	0.320	0.062	102.8	1125	1.540	1.400
	Minimum	0.150	768	7.57	0.059	0.009	42.8	154	0.400	0.140
	AnLo (kg/y)	-	-	-	**9.3**	**1.8**	**3574.6**	**30,111.2**	**35.9**	**44.0**

AV—average value, SD—standard deviation, AnLo—annual loading, EC—electrical conductivity.

Table 3. Results of one-way ANOVA analyses (with Tukey HSD) for the investigated variables in Lake Mielenko (n = 384).

Variable	F Value	p Value	Years Which Differed Significantly from 2013 (Before Inflow of Stormwater with Pollution from the Road Salts Disposal)
NO_3	9.3925	<0.001	2021
PO_4	4.7270	<0.001	2020; 2021
Ca	9.0366	<0.001	2019, 2021
Cl	343.7683	<0.001	2019, 2020, 2021
Fe	7.3686	<0.001	2020, 2021
Mn	1.5363	<0.001	no statistically significant differences
Reaction	4.8187	<0.001	2020, 2021
EC	99.8546	<0.001	2019, 2020, 2021

In the selected research years, the concentrations of nitrate nitrogen in the waters of Lake Mielenko were subject to significant fluctuations and varied in the range from 0.000 to 0.332 mg N/L. During the growing season, the concentrations even dropped to the analytical zero. In 2013, during inflow of stormwater before changes of the use of catchment area an average concentration of NO_3 in the water was 0.163 ± 0.068 mg N/L (Figure 2). After the creation of a road salts disposal area, an average concentration of NO_3 in water decreased to 0.076 ± 0.036 mg N/L in 2021.

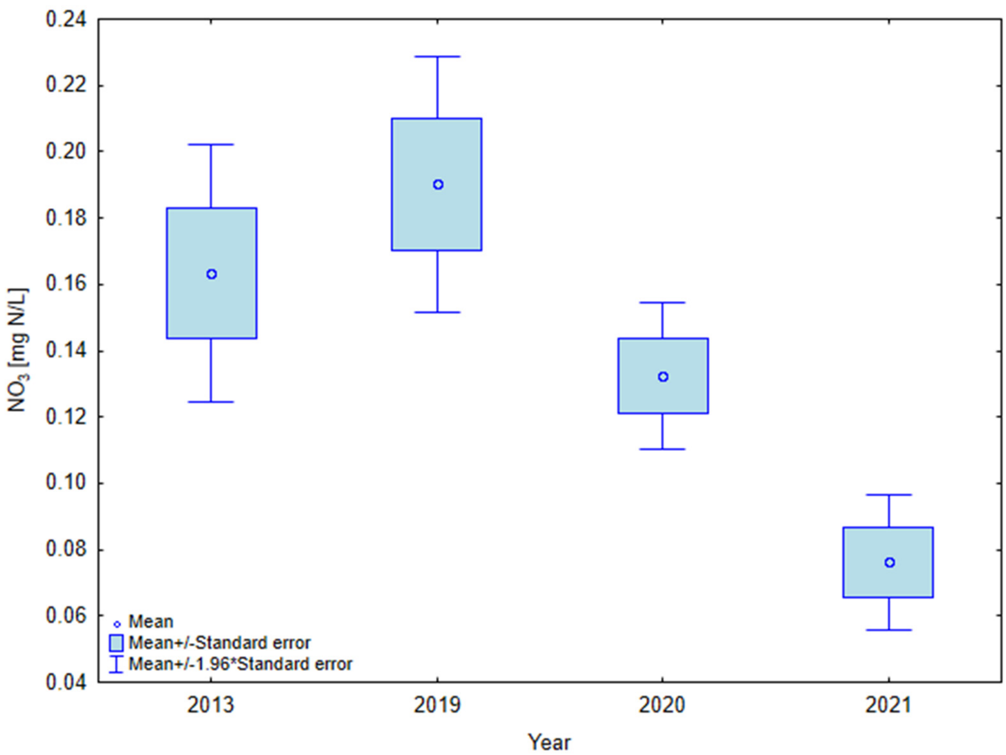

Figure 2. Average annual values of NO_3 content in the water of Mielenko Lake.

Nitrates are the product of the first stage of nitrification, which occurs mainly as a result of the activity of *Nitrosomonas europea* bacteria, while the oxidation of nitrites to nitrates is carried out mainly by bacteria of the genus *Nitrobacter winogradskyi* [25]. In addition to the presence of a sufficient amount of oxygen in the water, elevated temperature favors the action of nitrifying bacteria. However, they are very sensitive to the action of toxic substances. Very low concentrations of iron increase the activity of these bacteria, and manganese, even in low concentrations, is toxic to them [26,27]. Due to the polymictic nature of the water of Lake Mielenko and the low concentrations of Mn, the nitrification processes could take place in the lake waters with high intensity, with a high temperature of around 20 °C during the growing season. Hence, the concentration of nitrate nitrogen before the creation of a road salts disposal reached 0.17 mg N/L. After the disposal area was created, the nitrification processes were stopped. Factors that did not favor this process were the concentration of manganese in the water, but mainly the increase in the concentration of chloride, calcium and, at the same time, EC.

The above assumptions are also confirmed by the high negative correlation coefficient between NO_3 and chloride concentrations and between NO_3 and EC. A similar situation occurs in wastewater treatment processes, where an increase in chloride concentrations causes a decrease in the effectiveness of wastewater treatment through nitrification and denitrification processes [28].

During the research period, phosphate concentrations varied from 0.020 to 0.065 mg P/L. In its occurrence, seasonal variability was found, which consisted of a decrease in concentrations during the period of increased photosynthesis. Before changes of the use of catchment area (2013) mean quantity of phosphates in water of lake was 0.021 ± 0.008 mg P/L (Figure 3). In 2021, the mean values of PO_4 decreased to c.a. 0.013 ± 0.014 mg P/L.

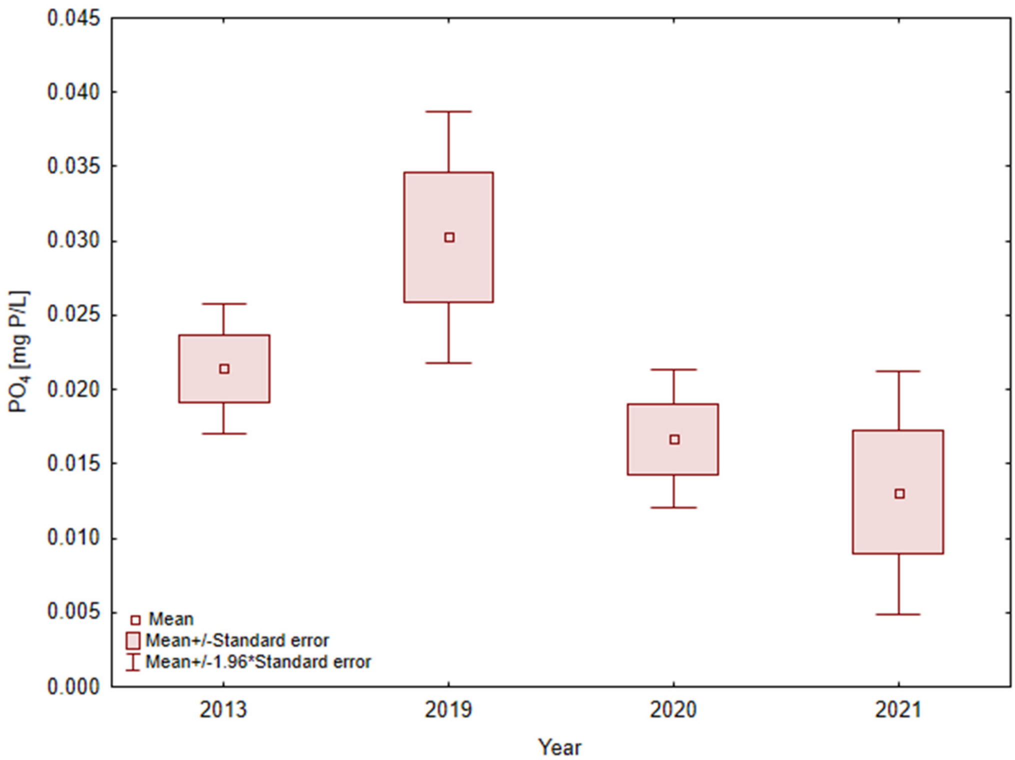

Figure 3. Average annual values of PO_4 content in the water of Lake Mielenko.

In surface waters, phosphorus occurs in the form of organic and inorganic suspension, dissolved minerals (mainly P–PO_4), and dissolved organic material [29,30]. The research of Lake Mielenko carried out in 2019 showed a very clear increase in the concentration of phosphates in the water, and in the following years a decrease in its amount. Based on the analysis of the waters flowing in through the rain collector, it was found that in 2019 the concentrations of phosphates in these waters were approximately three times higher than before the creation of the road salts disposal area. However, the change in the management of the catchment area did not affect the amount of phosphate in storm water. This situation can be explained by the air quality in the area. According to Berthold et al. [31] and Amos at al. [32], the content of phosphorus in precipitation is constantly increasing. It is reported that phosphate concentrations in rain are so high (almost 1.5 mg PO_4/L) that they exceed the amounts discharged into surface runoff waters. The reason for this is probably the illegal burning of rubbish, plastics, or tires, which is confirmed by field observations.

Calcium concentrations in the water of Lake Mielenko ranged from 32.8 to 57.1 mg Ca/L. Seasonal changes in the occurrence of this metal consisted of a clear decrease in concentration in the summer months, in the period of increased primary production. In 2013, when the storm water inflowed without contaminants from the road salts disposal area, the average calcium content in the lake water was 37.7 ± 1.1 mg Ca/L, while in 2021 this value was 45.1 ± 6.5 mg Ca/L (Figure 4).

Calcium is an element that determines water hardness [33,34]. After the creation of the road salts disposal area, a significant increase in calcium concentration was noted in the waters of Lake Mielenko. As mentioned before, calcium salts ($CaCl_2$) are a component of the mixture used for sprinkling roads and streets. Calcium chloride absorbs moisture quickly, which makes it easier for sodium chloride to start its melting process, for which it needs some heat and moisture. It is recommended to use the following mixtures of NaCl with $CaCl_2$ in a weight ratio: 4:1 (80% NaCl + 20% $CaCl_2$); 3:1 (75% NaCl + 25% $CaCl_2$) or 2:1 (67% NaCl + 33% $CaCl_2$). According to Kolada et al. [35] the calcium content in lakes does not exceed 75 mg Ca/L, while Marszelewski [36] reported that the average calcium concentration in municipal reservoirs is about 60 mg Ca/L. Calcium concentrations currently recorded in the waters of Lake Mielenko indicate that it is an anthropogenically polluted reservoir. Increased calcium concentrations in lake waters changed their hardness from low to medium-hard according to the Dojlido classification [33].

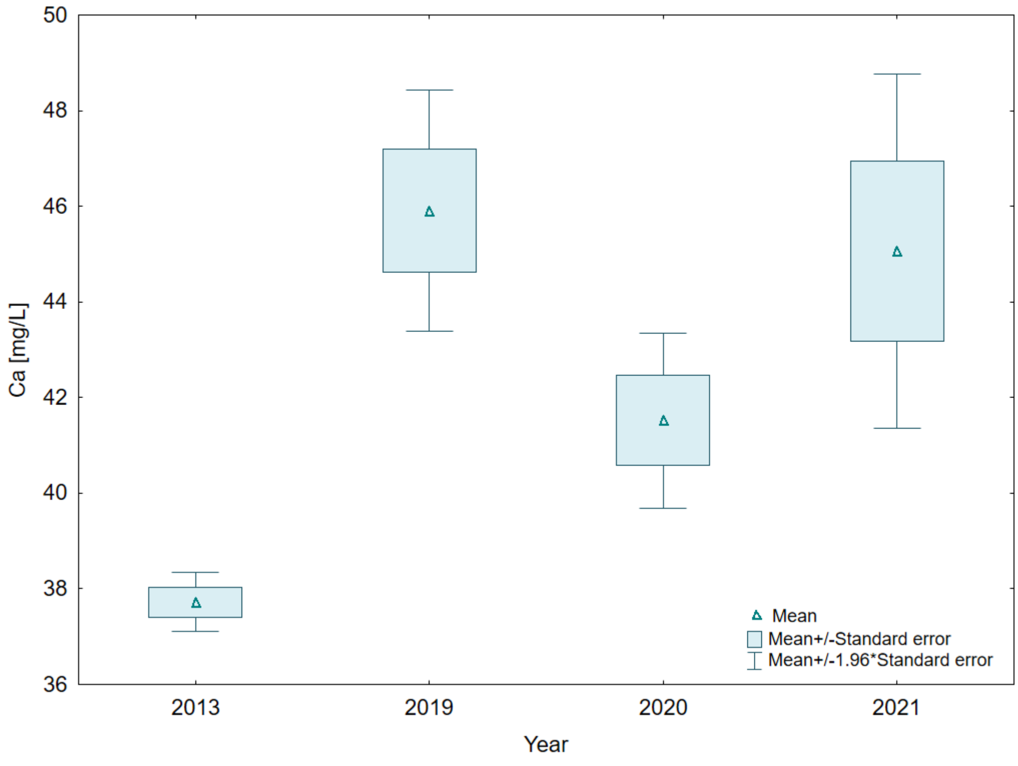

Figure 4. Average annual values of Ca content in the water of Mielenko Lake.

In 2013, when only stormwater was discharged into Lake Mielenko, chloride concentrations varied within a very small range, from 33 to 37 mg Cl/L. After the creation of a road salts disposal area, chloride concentrations in water increased dramatically and ranged from 128 to 221 mg Cl/L. In 2013, the average concentration of Cl in the water was 35 ± 1.6 mg Cl/L (Figure 5). After the creation of the road salts disposal area, the average concentration of Cl in the water increased to 179 ± 10.1 mg Cl/L in 2021.

Chlorides are ions that are considered when determining the degree of contamination of water with minerals. Chlorides are not transformed in soil and water, nor are they absorbed by soil material or bottom sediments, and therefore remain fully soluble in water and soil solution. However, they are taken up by plants and easily washed from the soil, also with surface runoff, which makes them a natural factor indicating the movement of water and its substances, and at the same time an excellent indicator of anthropogenic pollution of waters. Chlorine in the form of chlorides constitutes 0.045% of the mass of the Earth's crust (lithosphere), from which it is released in the process of weathering rock-forming minerals and then washed with surface waters to the seas and oceans. The chloride concentration in inland surface waters is about 9 mg Cl/L [37]. In Lake Mielenko, chlorides concentrations did not exceed 40 mg Cl/L; however, they were still quite high, which is a characteristic feature of municipal reservoirs, which are exposed to surface runoff of salt that defrosts roads and streets in winter. After the creation of a road salts disposal area in the catchment area of Lake Mielenko, chloride concentrations in water increased almost fivefold and periodically exceeded 180 mg Cl/L. On the other hand, rainwater from the landfill, which is discharged to the lake through the bank collector, contained on average over 500 mg Cl/L. The disposal stores, among others, salts such as NaCl, $CaCl_2$, and $MgCl_2$. The rain rinses these substances and then the excess chlorides are discharged into the waters of the analyzed reservoir through the stormwater drainage system.

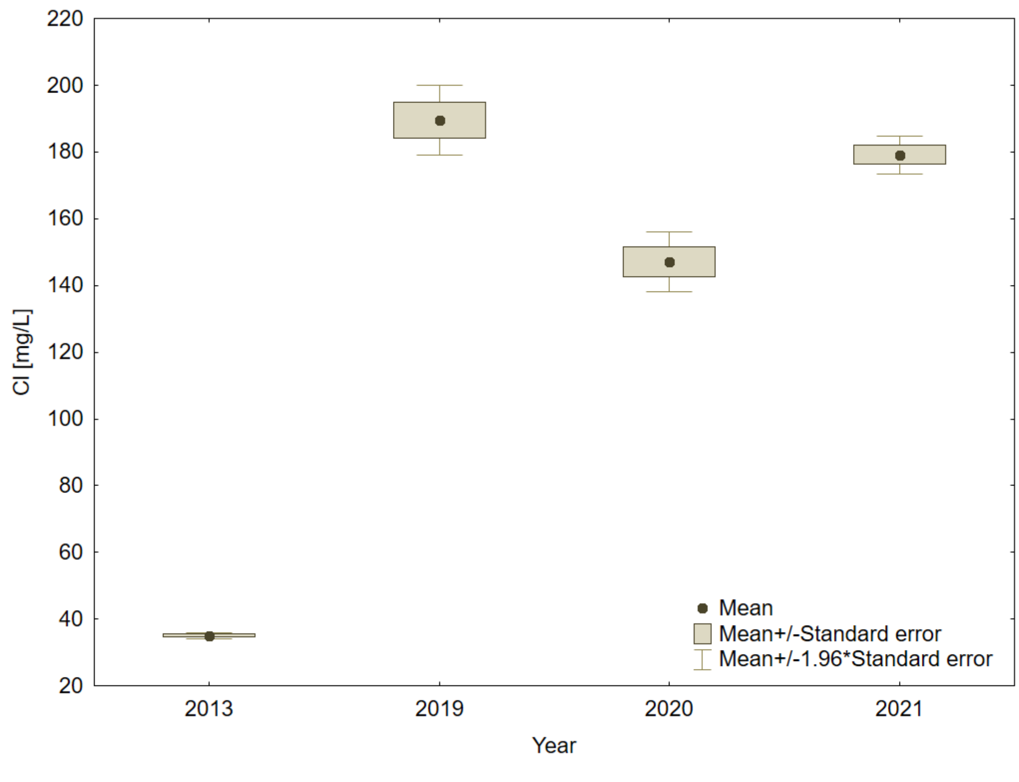

Figure 5. Average annual values of Cl content in the water of Mielenko Lake.

The iron content in the waters of Lake Mielenko varied in the range from 0.13 to 0.78 mg Fe/L. In the last research years, clearly higher concentrations occurred in summer, in July. Before changes of the use of the catchment area (2013), the mean quantity of iron in the water of the lake was 0.455 $\pm$ 0.032 mg Fe/L (Figure 6). In 2021, the mean values of Fe decreased to c.a. 0.240 $\pm$ 0.120 mg Fe/L.

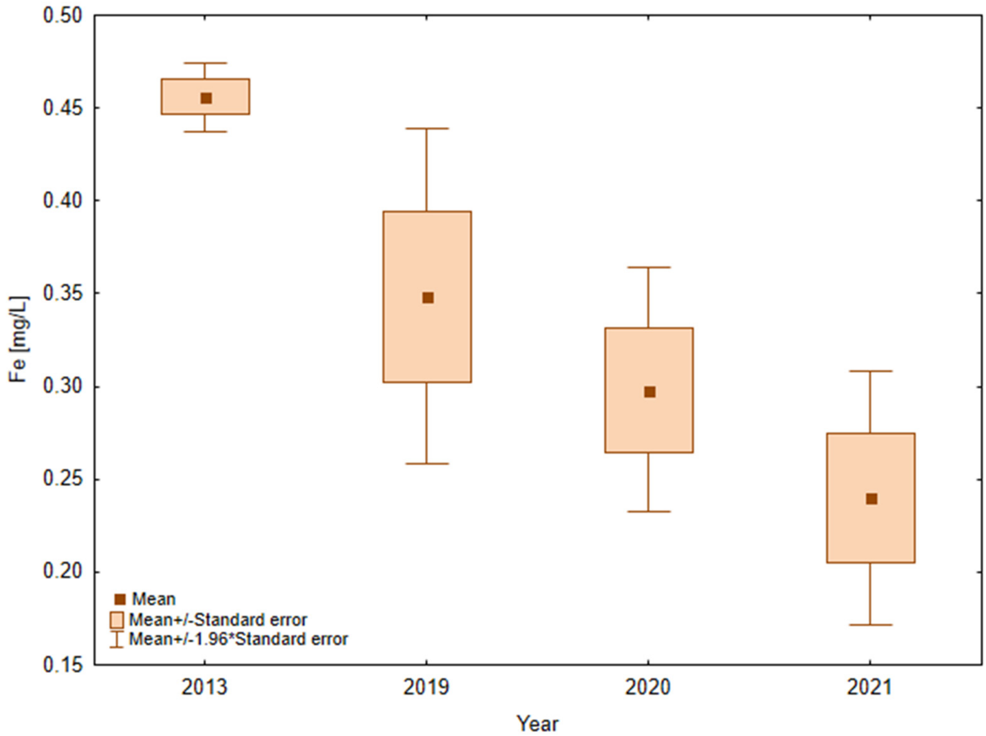

Figure 6. Average annual values of Fe content in the water of Lake Mielenko.

In the water of Lake Mielenko, manganese concentrations fluctuated in the range of 0.14 to 0.74 mg Mn/L. After the creation of a road salts disposal area, manganese concentrations in the water did not change significantly. In 2013, the during inflow of stormwater before changes of the use of the catchment area, the average concentration of Mn in the water was 0.35 ± 0.07 mg Mn/L (Figure 7). In 2021, the average concentration of Mn in the water was 0.30 ± 0.15 mg Mn/L.

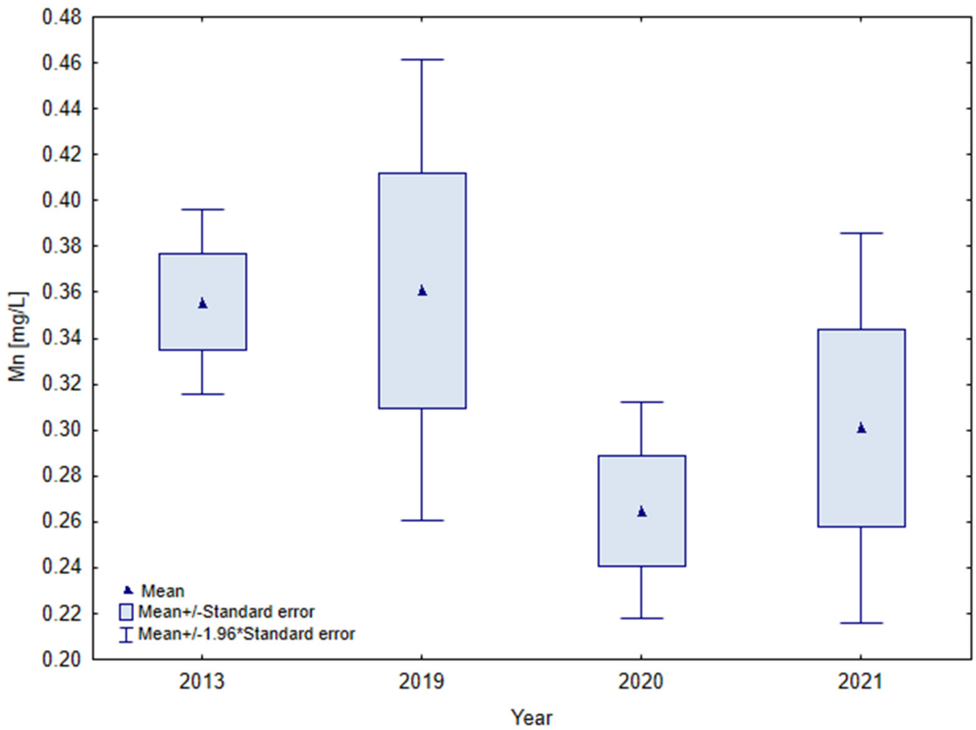

Figure 7. Average annual values of Mn content in the water of Lake Mielenko.

The changes in manganese and iron content were also analyzed while examining the changes in the chemistry of the waters of Lake Mielenko after the disposal was created. In the case of manganese, no significant differences in concentrations were found, and the amount of iron decreased. Lake Mielenko is periodically supplied with the waters of a natural watercourse that flows out of the fen and flows through forest areas. Such waters are characterized by a high abundance of iron compounds, especially chelates, i.e., iron complexes with humic substances [38]. Due to the fact that in recent years there have been periods of drought in the Kashubian Lake District, the inflow of water through the watercourse appeared sporadically, which resulted in a decrease in the iron content in the lake. The inflow of highly saline waters decreased the mean value of the reaction in the reservoir; however, it still remains at a slightly alkaline level.

3.2. Changes in Physical Parameters

The pH of the water of Lake Mielenko ranged from 6.71 to 9.81 pH. Before the creation of the road salts disposal area, the average pH value was 8.18 ± 0.45 pH, while in 2021, a few years after the change of the catchment development, the average pH was 7.95 ± 0.24 pH (Figure 8).

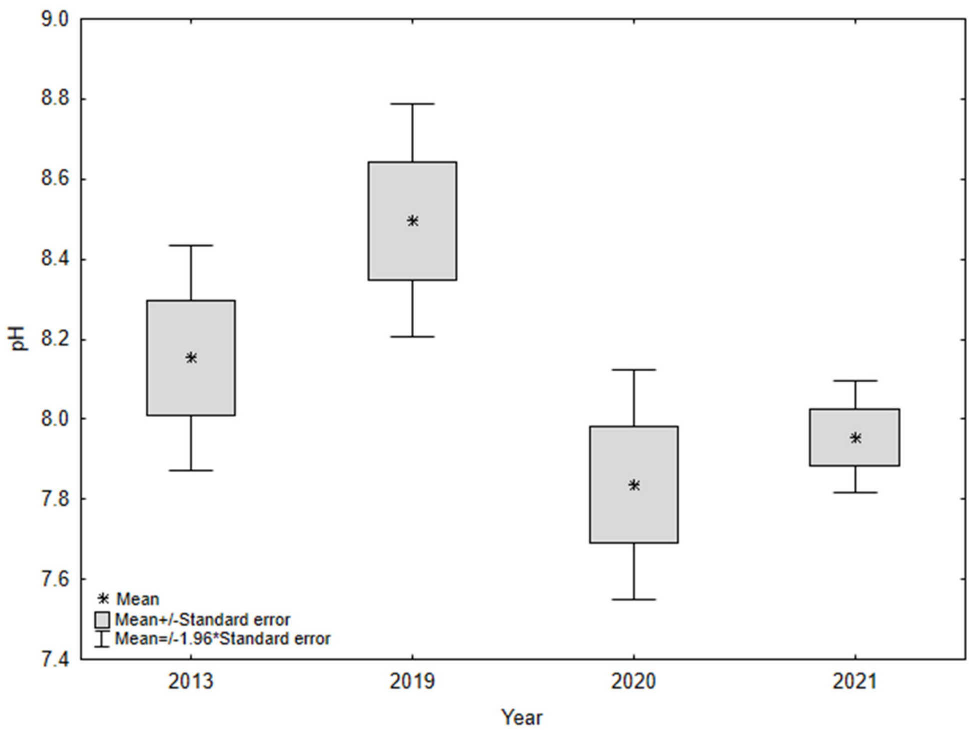

Figure 8. Average annual values of pH in the water of Lake Mielenko.

The EC of the water of Lake Mielenko ranged from 382 to 794 µS/cm. In 2013, the average value of the reaction was 427 ± 27.8 µS/cm, while in 2021, a few years after the change of the catchment area management, the mean value of the EC was 754 ± 27.5 pH (Figure 9).

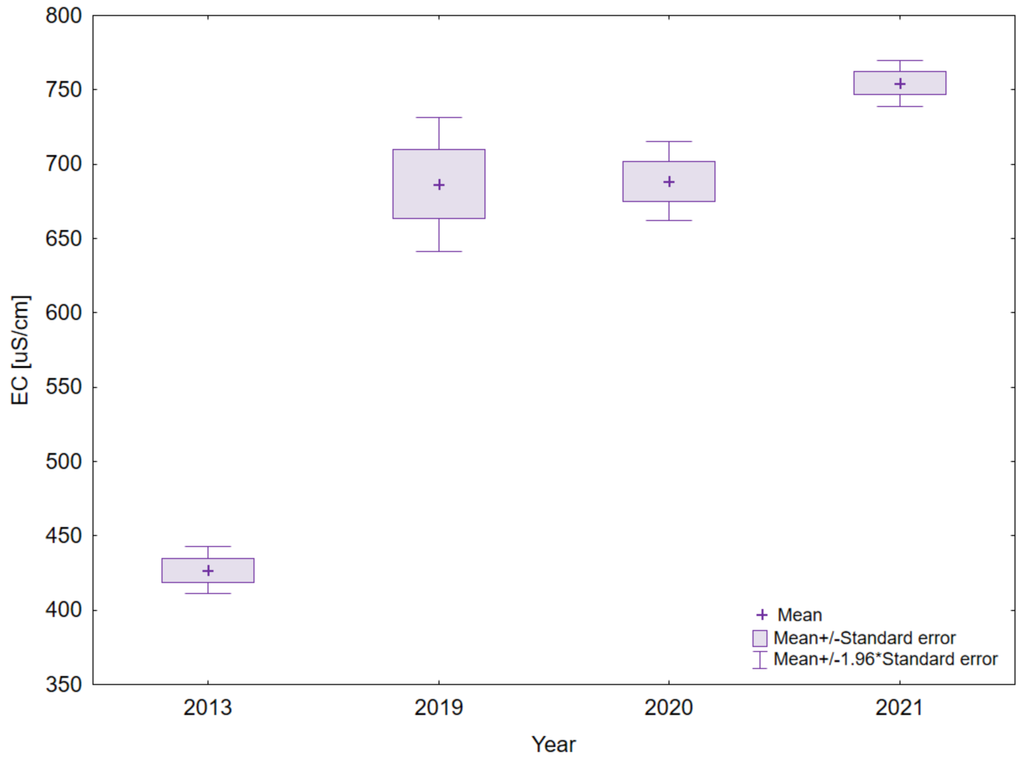

Figure 9. Average annual values of EC in the water of Lake Mielenko.

The EC determines the total content of ions dissolved in water [33], and it is thus an indirect indicator of the total mineralization of water [39]. There is a close relationship between the concentration of dissolved inorganic matter such as calcium, magnesium, and chlorides, and the EC. Therefore, the determination of the EC value is an important indicator for assessing the trophy of the reservoir. The studies by Jankowski and Rzętała (1997) [40] proved that high EC values are characteristic of lakes that are supplied with highly polluted waters for a long time. The EC of the waters of Lake Mielenko almost doubled and remained at the level of about 750 µS/cm, which is typical for lakes contaminated with mineral substances.

4. Conclusions

The use of sodium chloride, calcium chloride, magnesium chloride, and other chemicals as road slippage and de-icing agents has been common practice since the mid-20th century, and their use has been increasing dramatically. The example of Lake Mielenko clearly shows that the creation of unsecured disposal of road salts in the catchment area of water body is a serious source of pollution (Figure 10). Salts get into the waters as a result of infiltration and as a result of the inflow of stormwater. The negative effects are not only changes in water properties, but also ecological effects. The ecological effect of the change in hydrochemical conditions in Lake Mielenko is the *Potamogeton crispus* that grows abundantly in this water body, which prefer calcium-rich water. This plant has leaves with a wavy edge, resembling seaweed, often reddish or brown in color. The *Potamogeton crispus* is an annual plant that reproduces by shedding. In late autumn, young plants germinate from the seeds at the bottom. They bloom at the turn of May and June. In summer, this plant begins to die. In the period from May to July, practically the entire area of Lake Mielenko, whose average depth is only 1.3 m, is covered by *Potamogeton crispus*. The overall aesthetics of the lake have deteriorated significantly, and the availability of water for recreation has also been limited. The example of Lake Mielenko clearly shows how excessive human pressure can destroy a natural water reservoir in a short time.

Figure 10. Location of a material repository for the elimination of slippery roads in the catchment area of Lake Mielenko. Available online: https://www.geoportal.gov.pl/ (accessed on 15 June 2022).

In order to avoid situations such as that in Lake Mielenko, the so-called best management practices (BMP) should be implemented, i.e., special infiltration structures (swales, storm water basins, and bioretention cells) designed to reduce the load of mineral substances flowing into the waters.

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