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Influence of artificial infiltration on the removal of surfactants from surface waters

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ABSTRACT

Surfactants present in natural surface waters negatively affect the self-purification processes of water due to foaming, oxygen deficiency and eutrophication of water reservoirs. Consumed with water, they adversely affect human health. The purpose of the research conducted in the period from 2019 to 2022 was to determine the effectiveness of removing anionic and non-ionic surfactants in the process of artificial infiltration carried out at surface water intakes. The research was carried out on an experimental field installation located at the surface water infiltration intake. The experimental installation consisted of three metering wells (piezometers) located on the way between the infiltration pond and the collecting well. Water samples collected from the pond, from piezometers and from the well allowed to assess the change in concentrations of selected surfactants during the flow of water through the ground from the pond to the well. The analysis of the content of non-ionic surfactants was carried out by the method of iodobismutane determination with the final measurement of bismuth-thiourea (BiAS-thio) absorbance. A simplified method for the determination of anionic surfactants using methylene blue (MBAS) was used to measure the concentration of anionic surfactants. The presented studies carried out during the pandemic in 2019–2022 show a significant increase in the concentration of non-ionic surfactants in water collected from the infiltration pond. The presented research results indicate that artificial infiltration can be treated as a robust and effective barrier eliminating surfactants from treated water before the treatment plant technology. The main effect of surfactant removal occurs in the first section of the ground flow. Interpretation of the results allowed to conclude that the sediment layer at the bottom of the infiltration pond plays a major role in the removal of surfactants. Biodegradation is the main process responsible for the removal of non-ionic and ionic surfactants in the infiltration process.

Keywords: Water intake; Artificial infiltration; Anionic surfactants; Non-ionic surfactants; MBAS; BiAS-thio

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1. Introduction

The use of surface-active substances based on natural ingredients is known since ancient times, while synthetic surfactants appeared in common use only in the middle of the 20th century. In the early 1940s, the widespread of synthetic surfactants has been increased by making industrial and household products for example pesticides, detergents, and cosmetics [1–3]. Nowadays we can observe the continuous increase of the demand for surfactants by both households and industry.

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It is now well established that once surfactants reach water resources they can affect terrestrial and aquatic organisms as well as humans [4,5]. The prolonged effect of anionic and non-ionic surfactants on aquatic bodies generally appears at concentrations higher than 0.1 mg/L as stated [6–10]. According to numerous research, the effects of surfactant on aquatic life are notable and exposure in case of water organisms, mostly fish, may accelerate serum transaminases and alkaline acid phosphatase activity [11]. Afterwards they cause unfavorable effects on human health by producing inhibitors to various enzymes in their body. Furthermore, the growth of algae and other microorganisms is affected by discharge of surface-active agents in water as stated by the study of Yuan et al. [4]. On the other hand, the metabolites of non-ionic surfactants, especially alkylphenols, are recognized as dangerous because of high resistance to biodegradation, toxicity and strong estrogenic action [12–17].

Surface-active agents are among the common examples of amphiphilic molecules; they are soluble in both organic and water solution, which allows them to reduce surface tension, and hence be used in the cleaning and laundry. The molecular structure of surfactants consists of two parts, namely hydrophilic and lipophilic groups that are bonded in a single molecule [4,18–23]. The biggest group of the produced surfactants are anionic compounds, which cover almost 50% of the total global production – they are used as foaming agents, emulsifiers, antistatic agents and dispersants whereas the non-ionic are mostly toxic [4]. The share of the non-ionic surfactants is approx. 30% and still rising.

Studies over the past two decades have provided important information on how every year Western Europe has become the greatest user of detergents and softener products with 4.2 and 1.2 million tons, respectively [24,25]. Moreover, previous researches have shown that in 2000, the annual production of surfactants worldwide was 7million tons [25], while over the past 6 y, there has been a dramatic increase of production by approx. two times as reported by Johnson et al. [26] and Chen et al. [27]. Numerous scientific reports confirm the presence of surfactant residues in surface and even groundwaters [13,28,29].

Many, especially large, water treatment plants work on the basis of a multi-barrier approach, which is able to ensure a high degree of operational safety in removing biological and chemical contaminants from water during both normal operation and emergency situations [30–32]. However, the most commonly used simple processes for surface water treatment are usually ineffective with respect to micropollutants, especially those belonging to the group of contaminants of emerging concern (CEC) [30,33,34]. An effective method of removing CEC from water is the adsorption process. It allows to achieve a high degree of removal of contaminants, but the effectiveness depends on the type of adsorbent and parameters such as pH and contact time [35–38].

To deal with surfactants removal, combined advanced treatment method (e.g., membrane filtration and advanced oxidation process) became a promising solution to micropollutants removal [39,40]. However, the operational cost of using both methods is high. Based on numerous scientific research, the combination or alternative treatment methods are a compromising solution [41–43].

Artificial infiltration is a promising alternative and effective method for improving some physical, chemical, and biological characteristics of surface water due to non-use of additional reagents; it is an underground process that uses soil media for treatment [44–47]. Artificial infiltration is a very effective and sustainable method of surface water treatment worldwide [48,49].

During the infiltration process, surface water enters aquifers through recharge, thus reducing or removing different physico-chemical parameters such as turbidity, colour, organic matters and microbiological parameters, as a result it became an efficient measure for turbidity removal as has proven by numerous researchers [31,32,48,50,51] and it was also confirmed that total and fecal coliforms were decreased or eliminated by infiltration [52,53]. The time of water passage through the ground causes water retention lasting a few or even a dozen weeks. The temperature of water is more stable throughout the year. The water quality is more averaged throughout the whole year. The technology based on infiltration processes is much simpler than that used in systems for typical surface water. It usually consists of only processes typical for groundwater treatment.

Different geochemical, biological, and hydrologic factors influence the efficiency of artificial infiltration. Seepage velocity of water and the concentration of pollutants should be taken into account during evaluation of the artificial infiltration development due to the huge difference between concentration of filtered water and their patterns [54]. The composition of surface water after infiltration becomes similar to the quality of groundwater. Penetrating into the ground, water has to pass the layer of sediment formed at the bottom of infiltration ponds, usually a few centimetres thick. The sediment is a natural mineral and biological membrane where mechanical filtration, ion exchange, adsorption and biological processes take place causing retention of most heterogeneous contaminants [48,55]. Then water passes through aeration and saturation zones. Biochemical process, ion exchange or adsorption occur in the aeration zone resulting in the removal of organic matter and micropollutants. Afterwards, the concentration of organics and oxygen are diminished, autotrophic bacteria provide the process of biodegradation and the carbon dioxide concentration is increased. The variety of treatment processes occurring during infiltration allow for the removal of different contaminants, including CEC.

2. Goal of the research

The aim of the research conducted in the period from 2019 to 2022 was to determine the effectiveness of the removal of anionic and non-ionic surfactants in the process of artificial infiltration carried out in surface water intakes.

The goal of the research and interpretation of the results was also to determine the nature of the processes that reduce the concentration of surfactants in water infiltrating through the ground.

Analysis of changes in the quality of the intake water in the context of the surfactant content would allow to determine whether artificial infiltration can be the first, reliable and effective barrier in the water treatment system.

The aim of the study was establishing the correlation of surfactant removal with other water quality parameters.

The additional goal of presented research was to refer to the impact of the pandemic on the concentration of surfactants in natural waters.

3. Materials and methods, experimental installation

The research was carried out on an experimental field installation located at an infiltration intake of surface water. The intake is located at Warta River in Poland.

The water collected in the intake mostly comes from infiltration ponds (70%). The riverbank infiltration supplies the intake with 20%. The rest 10% come from groundwater reservoir [56].

The experimental installation consists of 5 measuring points including: infiltration pond, three metering wells (piezometers) – marked as PP-1, PP-2 and PP-3 and the deep well S-48. The cross-section of the infiltration path is shown in Fig. 1 [45]. The metering points form a line perpendicular to the edge of the pond and to the line of the collecting wells. Landform and the forest influenced the exact position of the PP1, PP2 and PP3 metering wells thus the distances between the piezometers are not equal. The depth of piezometers is correlated with the infiltration water level in the ground.

The distance between the pond's bank and each of the measuring points is given in Table 1.

The water samples were collected monthly since January 2019 until September 2022. The chemical analyses of water samples were carried out in the laboratory of the Institute of Environmental Engineering and Building Installations and the Institute of Chemistry and Technical Electrochemistry at the Poznań University of Technology, Poland.

3.1. *Simplified MBAS method procedure to determine anionic surfactants*

In order to conduct the determination, 250 mL of the filtered sample, which contained anionic surfactants, was placed in a 500 mL separating funnel, then 5 mL of an acid

Table 1
Distance of measuring points from the bank of the pond

Sampling points (Fig. 1)	Distance from pond's bank (m)
PP-1	18.00
PP-2	45.50
PP-3	62.90
S-48 well	85.90

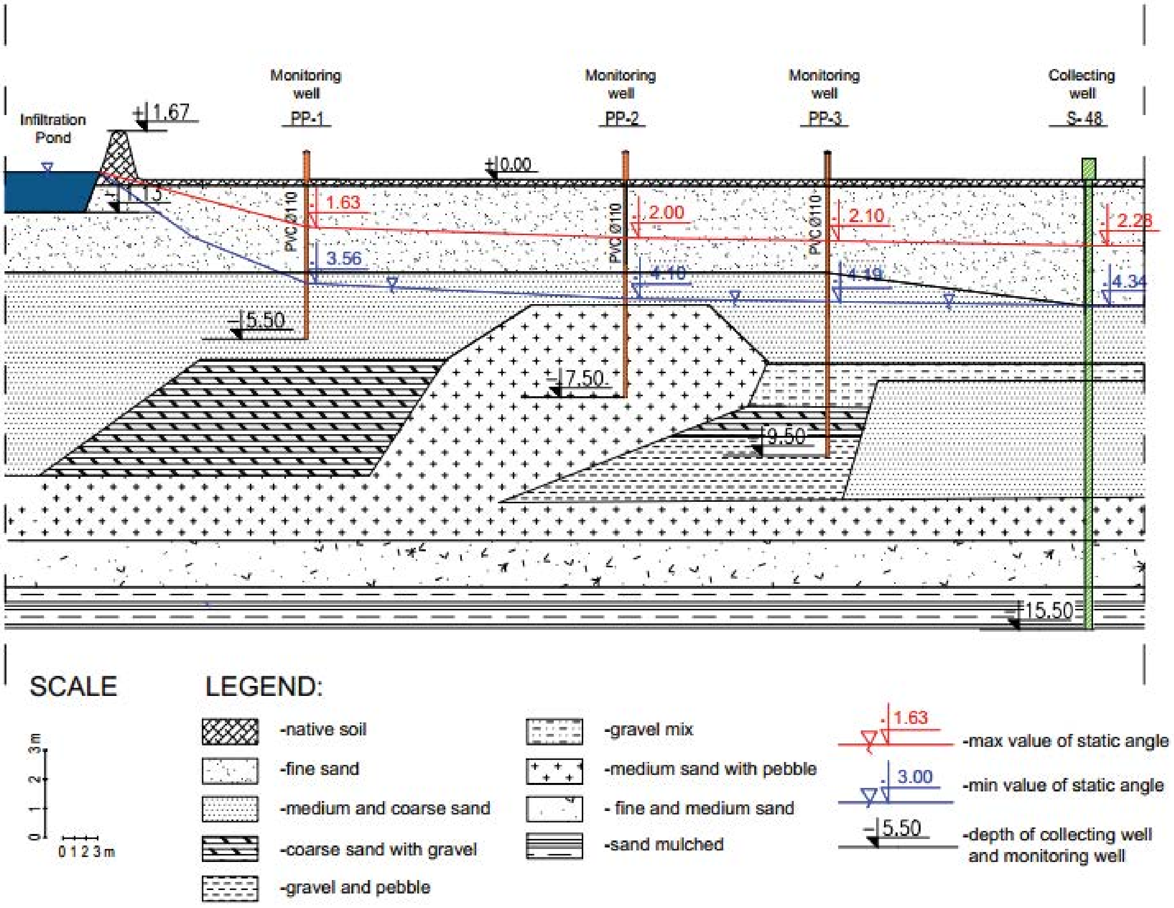

Fig. 1. Cross-section of the infiltration path [45].

solution of methylene blue (0.65 g of methylene blue and 6.5 mL of sulfuric acid with a density of 1.84 g/mL, made up to 1 L with demineralized water was added) and 10 mL of chloroform [57,58]. The funnel was shaken for 5 min and the phases were allowed to separate. The lower chloroform layer was filtered through filter paper into a glass measuring cuvette. Absorbance was measured using a spectrophotometer at wavelength λ_{max} = 652 nm with respect to chloroform. The LOD method has a detection limit of 1.38 µg in a sample and a LOQ limit of quantification of 4.13 µg in a sample. The results were reported relative to the reference anionic surfactant sodium dodecylbenzene sulfonate. The general scheme of the method is shown in Fig. 2.

The precision of determinations of the simplified MBAS method was determined for model samples (at the level of 10 and 50 µg of sodium dodecylbenzenesulfonate in the sample) and environmental samples for water from the Warta River. The results are listed in Tables 2–4. The relative standard deviation – RSD value included in tables is expressed by Eq. (1):

$$\text{RSD} = \left[\frac{\text{SD}}{x}\right] \times 100\% \tag{1}$$

where: SD – standard deviation and x – mean.

3.2. BiAS-thio procedure for the determination of non-ionic surfactants

The content of non-ionic surfactants in surface waters is generally not high, therefore SPE columns were used to isolate and concentrate them. A 200 mL sample of filtered surface water was passed through an SPE column (Supelco Discovery DSC-18 LT, 6 mL), then the adsorbed non-ionic surfactants were eluted from the bed with 8 mL of methanol, which was evaporated in quartz beakers at 70°C. The modified BiAS-thio (bismuth active substances) method was used for determination of non-ionic surfactants. The method is based on the formation of an oxyethylate precipitate using the Dragendorff's reagent which, after washing with glacial acetic acid, dissolves in an acidic thiourea solution and is subjected to spectrophotometric determination of bismuth as an equivalent of non-ionic surfactants. Approximately 1 mL of the solution including non-ionic surfactants and 1 mL of the modified Dragendorff reagent

Table 2
Results of the determination of anionic surfactants precision by the simplified MBAS method for a model sample containing 10 µg of sodium dodecylbenzenesulfonate in the sample

Lp.	Absorbance	Content in the sample (µg)
1	0.19126	9.98
2	0.19165	10.00
3	0.19221	10.03
4	0.20026	10.45
5	0.22195	11.58
Average		10.52
Standard deviation		0.74
Relative standard deviation of the RSD		7.04%

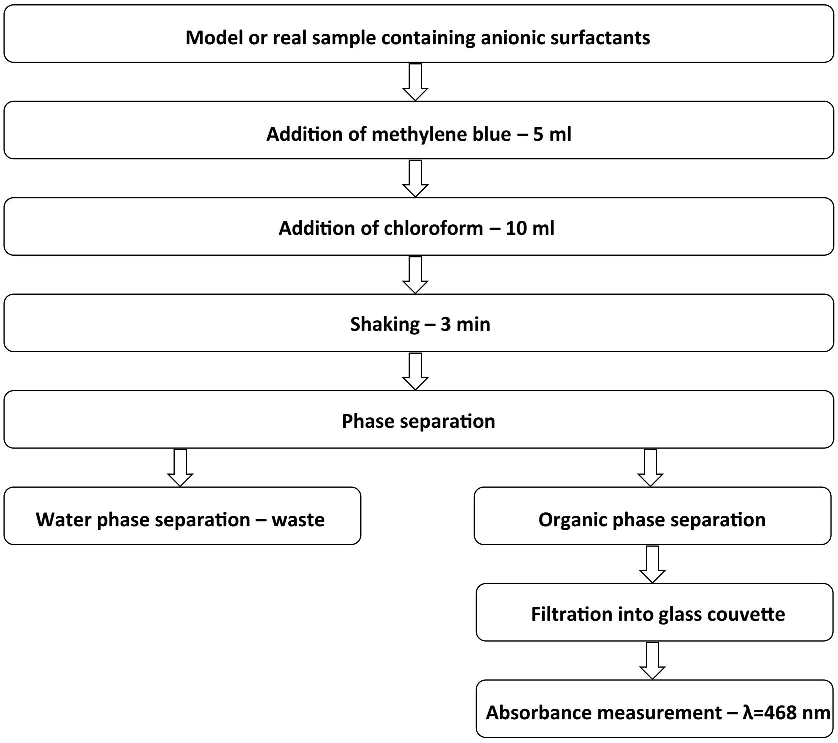

Fig. 2. Scheme of the simplified MBAS method for the determination of anionic surfactants.

were introduced into centrifuge tubes. Next, the tubes were centrifuged (type 352 MPW, 12,000 rpm for 5 min). After precipitation and separation of the oxyethylate precipitate using the modified Dragendorff reagent, the solution was decanted, and the orange precipitate was rinsed 3 times using 1 mL of glacial acetic acid in order to remove the residual Dragendorff reagent. The precipitate was dissolved in 2 mL of the dissolving-complexing solution (15% solution of thiourea in 1 M HNO_3) and placed in 1 cm glass cuvette. Absorbance was measured for λ_{max} = 468 nm against demineralized water. The results were calculated using a non-ionic surfactant model – Triton X-100 [47–50]. The LOD method has a limit of detection of 1.54 μg per sample and the LOQ limit of quantification is 4.59 μg per sample. The general scheme of the method is shown in Fig. 3.

The precision of the determination of the BiAS-thio method was determined for model samples (at the level of 10 and 50 μg of Triton X-100 in a sample) and environmental samples for water from the Warta River). The results are listed in Tables 5–7.

The effect of the infiltration process on organic contaminants removal was evaluated based on the following parameters: chemical oxygen demand with potassium permanganate (COD-$KMnO_4$) and total organic carbon (TOC). The TOC analyses were done using the Multi N/C® 3100, Analytik Jena, Germany apparatus and MultiWin software. The other water quality parameters were analysed in accordance with Standard Methods.

During the research, the retention time of infiltrating water in the ground was evaluated. The basis for determining the retention time were cyclic temperature measurements of water at measurement points. Temperature of water was measured every 2 weeks.

4. Results, interpretation and discussion

During the research period from 2019 until 2022 the raw water quality in the infiltration pond was constantly monitored. The water samples were collected every month. The values of selected water quality parameters including surfactants are presented in Table 8 as averages and the range (minimum and maximum).

During the entire research period from January 2019 until September 2022, analytical series for surfactant analysis were carried out. The obtained concentration values of anionic and non-ionic surface-active agents are listed in Table 9.

The values of chemical oxygen demand with potassium permanganate used as chemical oxidant (COD-$KMnO_4$) and the dissolved oxygen concentration are presented in Table 10. The values COD-$KMnO_4$ indicate the concentration of organic compounds in the collected water samples. Results presented in tables are the averages values calculated from two independent analyses.

Table 3
Results of the determination of anionic surfactants precision by the simplified MBAS method for a model sample containing 50 μg of sodium dodecylbenzenesulfonate in the sample

Lp.	Absorbance	Content in the sample (μg)
1	0.95354	49.77
2	0.97052	50.65
3	0.9788	51.09
4	0.99071	51.71
5	1.0052	52.46
Average		51.48
Standard deviation		0.79
Relative standard deviation of the RSD		1.53%

Table 5
Precision results of the determination of non-ionic surfactants by the BiAS-thio method for a model sample containing 10 μg of Triton X-100 in the sample

Lp.	Absorbance	Content in the sample (μg)
1	0.12288	9.78
2	0.12545	9.99
3	0.12745	10.15
4	0.13069	10.41
5	0.13248	10.55
Average		10.27
Standard deviation		0.25
Relative standard deviation of the RSD		2.45%

Table 4
Results of the precision of determination of anionic surfactants by the simplified MBAS method for environmental samples (water from the Warta River)

Lp.	Sample volume (mL)	Absorbance	Absorbance after blanking	Content in the sample (μg)	Concentration (μg/L)
1	250	0.15644	0.13799	7.20	28.81
2	250	0.15854	0.14009	7.31	29.25
3	250	0.16223	0.14378	7.50	30.02
4	250	0.16562	0.14717	7.68	30.72
5	250	0.17673	0.15828	8.26	33.04
Average				30.76	
Standard deviation				1.64	
Relative standard deviation of the RSD				5.33%	

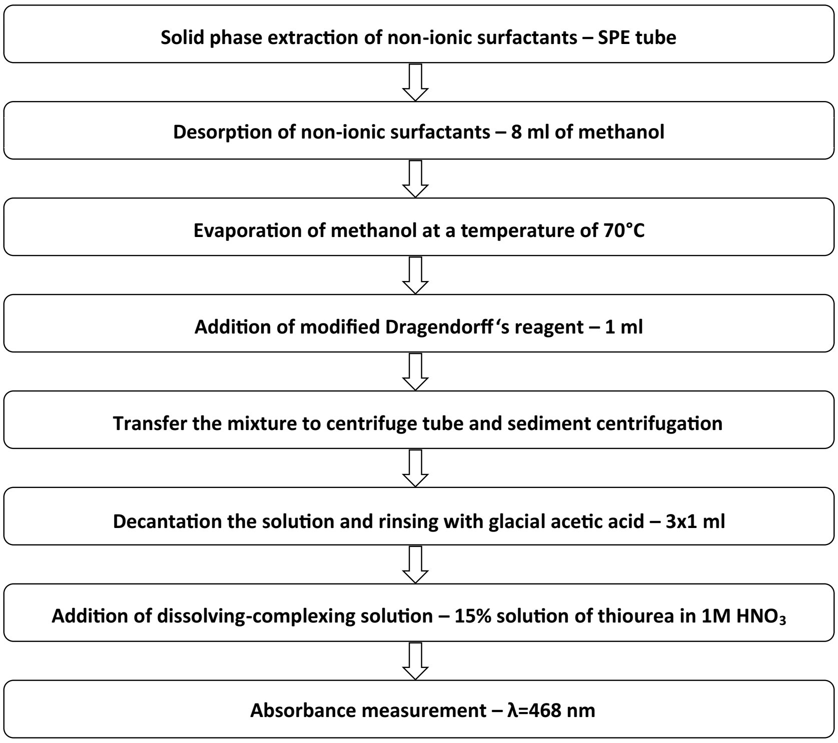

Fig. 3. Scheme of the BiAS-thio method for the determination of non-ionic surfactants from surface water samples.

Table 6
Precision results of the determination of non-ionic surfactants by the BiAS-thio method for a model sample containing 50 μg of Triton X-100 in the sample

Lp.	Absorbance	Content in the sample (μg)
1	0.59352	47.25
2	0.61718	49.14
3	0.62682	49.91
4	0.62982	50.14
5	0.64768	51.57
Average		50.19
Standard deviation		1.01
Relative standard deviation of the RSD		2.02%

The interpretation of the research results presented in this study included:

- determination of the retention time in the soil by the method of cyclic measurements of the temperature of infiltrating water,
- assessment of the effects of artificial infiltration in terms of anionic and non-ionic surfactants removal,
- biodegradation vs. adsorption classification evaluated using the Eberhardt, Madsen, Sontheimer (EMS) method.

4.1. Infiltration retention time

Analysis of changes of water temperature in soil throughout the infiltration route allowed to determine the infiltration time between the measuring points. Water which infiltrated in the soil flows in time determined by

Table 7
Precision results of non-ionic surfactant determinations for an environmental sample (water from the Warta River)

Lp.	Sample volume (mL)	Absorbance	Absorbance after blanking	Content in the sample (μg)	Concentration (μg/L)
1	200	0.49437	0.45970	36.60	183.00
2	200	0.49546	0.46079	36.69	183.44
3	200	0.49736	0.46269	36.84	184.19
4	200	0.50138	0.46671	37.16	185.79
5	200	0.50354	0.46887	37.33	186.65
Average				185.02	
Standard deviation				1.47	
Relative standard deviation of the RSD				0.79%	

Table 8
Water quality of infiltration pond during the research period

Parameters	Units	Maximum	Minimum	Average
pH	–	8.97	7.15	8.05
Orthophosphates	mg·P/L	1.10	0.00	0.16
Nitrate	mg·NO_3/L	6.57	0.00	0.85
Nitrite	mg·NO_2/L	0.65	0.02	0.24
Ammonium	mg·NH_4/L	1.04	0.07	0.42
Total organic carbon	mg/L	11.69	3.92	7.50
Turbidity	NTU	19.10	0.67	4.07
COD-$KMnO_4$	mg·O_2/L	10.70	5.70	8.15
Colour	mg·Pt/L	62.08	20.72	37.05

Table 9
Anionic (AS) and non-ionic (NS) surfactants' concentration (μg/L) in all sampling points

Series No.	Date	Pond		PP-1		PP-2		PP-3		S-48	
		AS	NS	AS	NS	AS	NS	AS	NS	AS	NS
1	18.01.2019	25.58	36.17	15.18	16.82	12.42	14.96	8.42	4.85	13.61	13.13
2	01.03.2019	43.50	100.47	10.36	98.33	17.20	91.97	14.23	83.8	11.44	92.83
3	15.03.2019	39.51	28.50	26.46	9.88	26.1	6.91	22.89	8.04	18.00	7.7
4	09.04.2019	57.91	50.65	27.07	34.21	36.7	31.13	35.49	27.51	30.17	21.81
5	16.04.2019	66.20	53.09	25.95	17.04	29.02	12.08	25.19	8.88	x	x
6	14.05.2019	64.57	50.04	28.03	1.40	28.85	11.56	31.04	4.09	35.37	2.48
7	05.06.2019	49.90	50.18	25.56	4.90	30.00	5.01	30.67	7.08	29.24	8.64
8	09.07.2019	34.20	50.29	14.30	8.46	20.59	3.80	20.71	5.27	20.06	16.58
9	27.08.2019	44.04	87.90	13.55	12.13	19.52	13.78	17.98	11.40	27.21	26.24
10	23.09.2019	54.83	57.36	20.96	15.31	22.50	20.24	24.42	17.37	27.49	21.83
11	28.10.2019	24.71	78.71	17.29	7.14	17.40	7.64	15.07	7.51	17.83	10.71
12	26.11.2019	26.98	58.05	14.19	25.36	16.36	22.01	16.89	17.36	17.75	34.81
13	16.12.2019	34.21	66.70	14.90	12.93	11.73	13.53	17.34	15.08	19.91	23.13
14	08.01.2020	33.00	55.20	12.73	30.43	18.36	16.85	22.49	31.26	23.37	31.83
15	04.03.2020	39.12	73.05	16.63	44.18	20.53	26.92	19.19	25.91	21.34	53.02
16	26.05.2021	35.29	116.22	21.43	50.64	19.99	38.22	18.55	32.52	17.40	32.33
17	21.06.2021	35.63	96.23	17.73	42.75	15.84	39.89	15.09	30.80	15.59	36.58
18	27.07.2021	39.37	80.09	19.18	51.12	20.47	31.35	18.42	30.34	10.19	26.84
19	24.01.2022	x	x	17.54	55.02	16.35	45.21	15.30	36.16	14.35	45.95
20	07.03.2022	32.17	119.28	18.78	51.21	17.33	42.44	15.50	35.60	14.66	31.74
21	04.04.2022	64.68	191.70	38.64	112.1	35.44	90.43	35.72	79.88	35.70	88.86
22	16.05.2022	29.22	149.88	19.78	66.52	18.99	65.32	18.09	52.19	17.65	57.58
23	21.06.2022	39.22	117.38	22.87	59.33	19.74	53.48	25.56	50.61	21.61	51.28
24	22.07.2022	29.19	102.73	19.59	49.63	17.37	47.29	15.30	42.46	17.06	45.02
25	12.09.2022	44.27	148.13	24.86	99.97	20.67	86.21	20.04	69.12	22.48	63.46

waves with the same temperature. The temperature was measured during the experiment at measuring points every 1–2 weeks. The graph that presents the temperature of the water measured at the measuring points is shown in Fig. 4. The graphs of temperature changes in time at each measurement point are sinusoidal. The distances between the curves' maxima (summer) and minima (winter) correspond to the times of travel of the wave of the same temperature and the same times of water flow between the measuring points. The distances of the measuring points from the bank of the pond and the average time when the water overcomes this distance are presented in Table 11.

4.2. Assessment of the effects of artificial infiltration in terms of anionic and non-ionic surfactants removal

The artificial infiltration creates the treatment conditions similar to slow sand filtration. During the passage of

Table 10
COD-$KMnO_4$ values and oxygen concentrations in all sampling points at the time of research

Date	Pond		PP-1		PP-2		PP-3		S-48	
	COD (mg/L)	Oxygen (mg/L)	COD (mg/L)	Oxygen (mg/L)	COD (mg/L)	Oxygen (mg/L)	COD (mg/L)	Oxygen (mg/L)	COD (mg/L)	Oxygen (mg/L)
15.01.2019	6.8	12	5	4.5	5.1	4.2	4.8	1.2	4.1	1
3.02.2019	7.41	10.4	5.21	6.6	5.56	4.4	4.05	2.4	4.98	1.6
19.03.2019	8.8	12	5.5	4.4	5	6	4.5	2	4.7	0.8
16.04.2019	8.2	8.2	5.2	5.2	5	5	5	5	–	1
5.06.2019	8.6	6.2	4.8	1.4	4.6	1	6.1	0.6	4.5	0.8
9.07.2019	8.35	4.8	8.9	1	5.25	0.8	5.15	0.8	5.6	1
27.08.2019	10.1	7.1	4.45	1.56	4.95	0.6	5.5	1.6	5.2	1.2
23.09.2019	8.55	12.6	4.1	1.7	5.1	1.5	5.15	1.4	4.95	1.2
28.10.2019	9.7	5.2	2.6	1.2	4.5	1	4.4	1.1	12	0.8
26.11.2019	5.8	9.5	3.4	1.2	3.7	0.6	4.45	0.7	5.2	1.1
16.12.2019	6.2	12	3.2	1.4	3.5	0.8	3.8	0.8	4.7	1.6
8.01.2020	5.7	13	2.8	1.8	2.8	1.1	3.4	1	3.9	1
4.03.2020	8.5	14	2.5	1	2.6	0.6	2.9	0.2	3.7	1
26.05.2021	9.9	7.6	5.1	2.3	5.5	4	6.6	2.8	5.8	1.4
21.06.2021	10.7	6.4	5.9	1	5.4	1.2	5.8	0.8	5.5	3
27.07.2021	10.0	8.2	5.2	0.6	4.9	1	5.4	1.2	5.3	0.6
24.01.2022	–	–	6.1	–	5.3	–	5.5	–	5.2	–
7.03.2022	–	10.6	–	4	–	4.4	–	4.6	–	0.8
4.04.2022	–	9.4	–	2.7	–	5.5	–	3.1	–	1.4
16.05.2022	10.6	10.2	6.7	1.98	5.7	3.66	6.1	2.13	5.4	3.3
21.06.2022	8.4	3.17	6.2	0.34	5.75	0.34	5.6	0.1	5.8	2.25
12.07.2022	9.3	4.3	7.15	0.32	6.8	0.53	6.6	0.63	6.3	0.22
12.09.2022	8.3	–	5.2	–	3	–	4.3	–	3.7	–

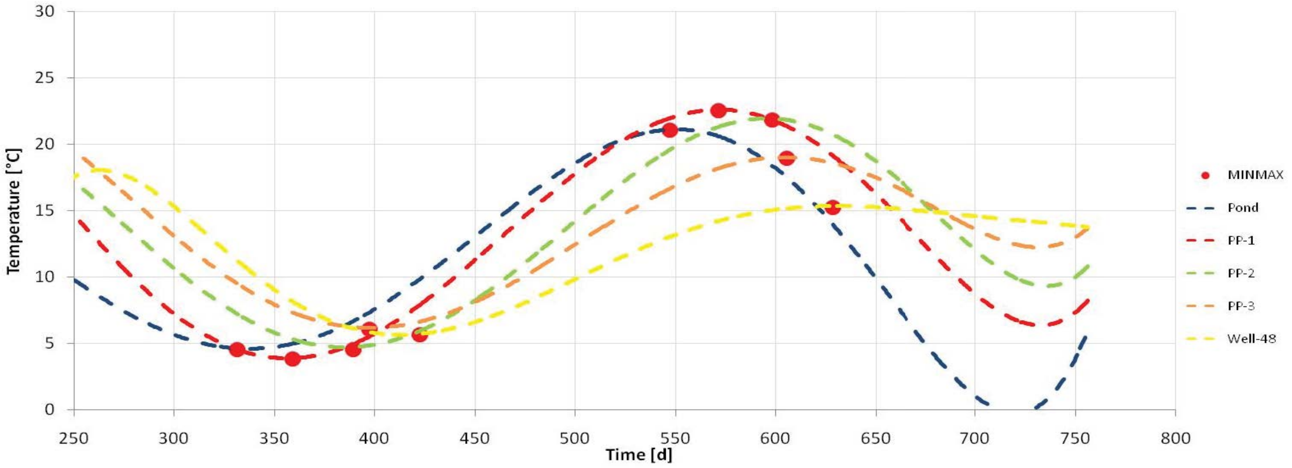

Fig. 4. Temperature changes in the sampling points during the research period from October 2018 to March 2020 [55].

water, the biological, physical and chemical processes occur in the ground. The main role is played by biodegradation, filtration, adsorption and ion exchange [45,55,59,61–63].

The surfactant concentration results presented in Table 10 were collected over a long period of time from January 2019 to September 2022 corresponding with the pond operation cycles. At that time, the Covid-19 pandemic began. Safety rules applied in water supply systems in Poland caused interruptions in water sampling from the field installation. Stricter hygiene requirements at the intake meant that no one from outside the waterworks could stay at the intake area and therefore sampling and chemical analyzes could not be performed and regular monthly water sampling at the water intake was not possible. This resulted in the division of all set of results (January 2019 to September 2022) into the following 3 periods of time creating 3 measurement series listed below in which samples for analysis were taken almost regularly:

- Serie 1: 15.01.2019 - 4.03.2020 - before pandemic,
- Serie 2: 26.05.2021 - 27.07.2021 - beginning the pandemic,
- Serie 3: 24.01.2022 - 12.9.2022 - late phase of pandemic.

The average concentrations of surfactants in the infiltration pond for selected series are presented in Fig. 5.

The values presented in the graph indicate that there was a significant increase in the concentration of non-ionic surfactants in the infiltration pond during the pandemic. The infiltration pond is supplied directly from the Warta River, therefore, an increase in the concentration in the pond indicates an increase of surfactants in the natural water supplying the infiltration intake.

The increase in the concentration of surfactants was and is caused by the increased consumption of cleaning agents in order to maintain hygiene related to the prevention of Covid-19. Anionic and non-ionic surfactants are toxic to some strains of bacteria and algae and are therefore used as an active ingredient in disinfecting preparations. These conclusions are consistent with the data regarding the increase in the production of surfactants in Poland and in Europe [60].

The presented data indicate a much greater increase in the concentration of non-ionic surfactants compared to anionic ones. This phenomenon can be explained by a greater increase in the production of cleaning agents with non-ionic detergents or better biodegradability of anionic detergents, which are much more effectively removed at wastewater treatment plants.

Table 11
Distance of measuring points from the bank of the pond and retention time

Sampling points (Fig. 1)	Distance from pond bank (m)	Retention time (d)
PP-1	18.00	26.0
PP-2	45.50	54.5
PP-3	62.90	62.0
S-48 well	85.90	86.0

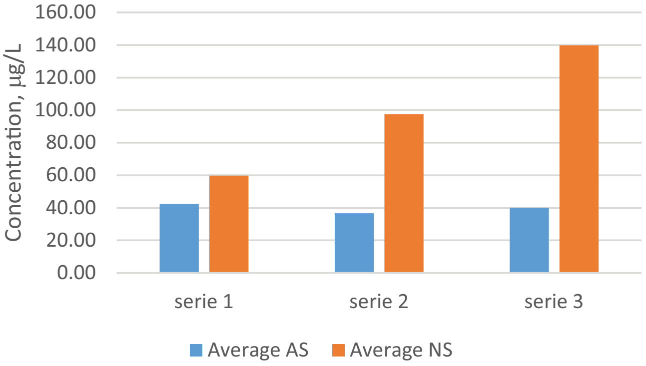

Fig. 5. Average concentrations of surfactants in infiltration pond during the research time from January 2019 to September 2022.

The surfactants present in the water in the infiltration pond at the intake are subject to the infiltration process during which biological, chemical and physical processes take place, causing their concentration to decrease as the water flows through the ground.

The relationships between the surfactants concentrations and the infiltration distance for each sampling date are presented in Figs. 6 and 7. The values shown in these graphs take into account the flow time of water in the ground.

The average concentrations of the tested surfactants determined in all 3-research series at given measuring points are presented in Fig. 8.

It is visible that the most effective removal of both anionic and non-ionic surfactants occurred in the first part of the infiltration path between the pond and the monitoring well PP-1 (Figs. 6–8). To quantify the results of infiltration with respect to the reduction of surfactant content, the removal efficiency of anionic and non-ionic surfactants was calculated. The results presented in Table 12 show the pond water temperature and total efficiency of surfactants removal for a given sampling data achieved in the whole distance (from pond to the well S-48, named E-total). The efficiency was calculated according to Eq. (2):

$$E_{\text{total}} = \frac{C_{\text{average pond}} - C_{\text{average S48}}}{C_{\text{average pond}}} \times 100\% \tag{2}$$

where $c_{\text{average pond}}$ – average concentration of surfactant in water from the infiltration pond for each series, $c_{\text{average S48}}$ – average concentration of surfactant in water from well S-48 for each series.

The effectiveness of anionic and non-ionic surfactants removal for the whole distance (E-total) from the pond to the collecting well are presented in Table 12. The relationship of E-total value and pond water temperature during all research is presented in Fig. 9. The total effect of removal of anionic and non-ionic surfactants is correlated with temperature. The total removal of analysed surfactants increases with the increase of water temperature. This correlation indicates the possible significant role of biodegradation in the removal of the surfactants from the infiltrating water.

The first phase of water purification in the process of artificial infiltration occurs during the flow of water through sediment layer formed at the bottom of infiltration ponds. This layer is usually characterized by a thickness of a few centimetres. The sediment is a natural mineral and biological membrane at which filtration, ion exchange, adsorption and biological processes occur and the majority of contaminants are separated [33,56]. The values presented in Table 8 as well as Figs. 6–8 indicate that the first part of the infiltration distance is crucial for removal of surfactants. The role of the first distance from the pond to the PP-1 well is more visible in case of anionic surfactants than non-ionic surfactants. Then, water passes through aeration and saturation zones where further biodegradation and adsorption

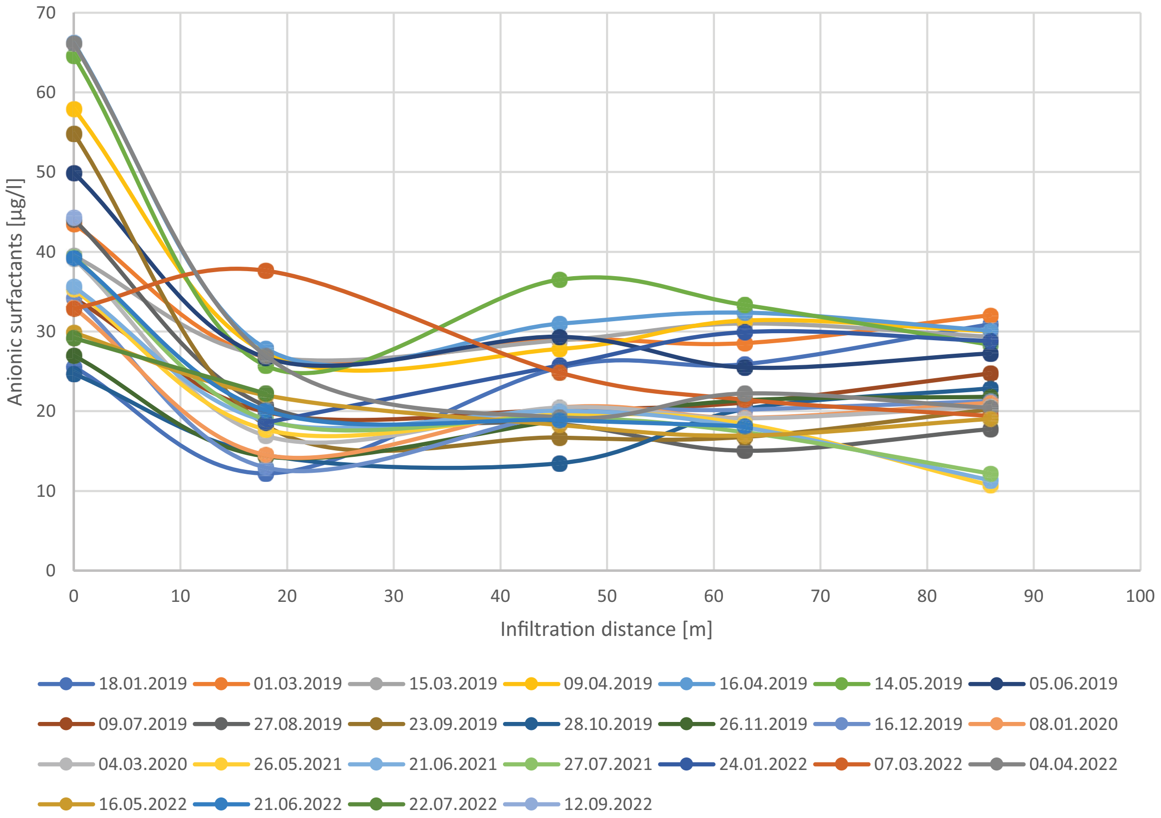

Fig. 6. Relationship between anionic surfactants concentration and infiltration distance.

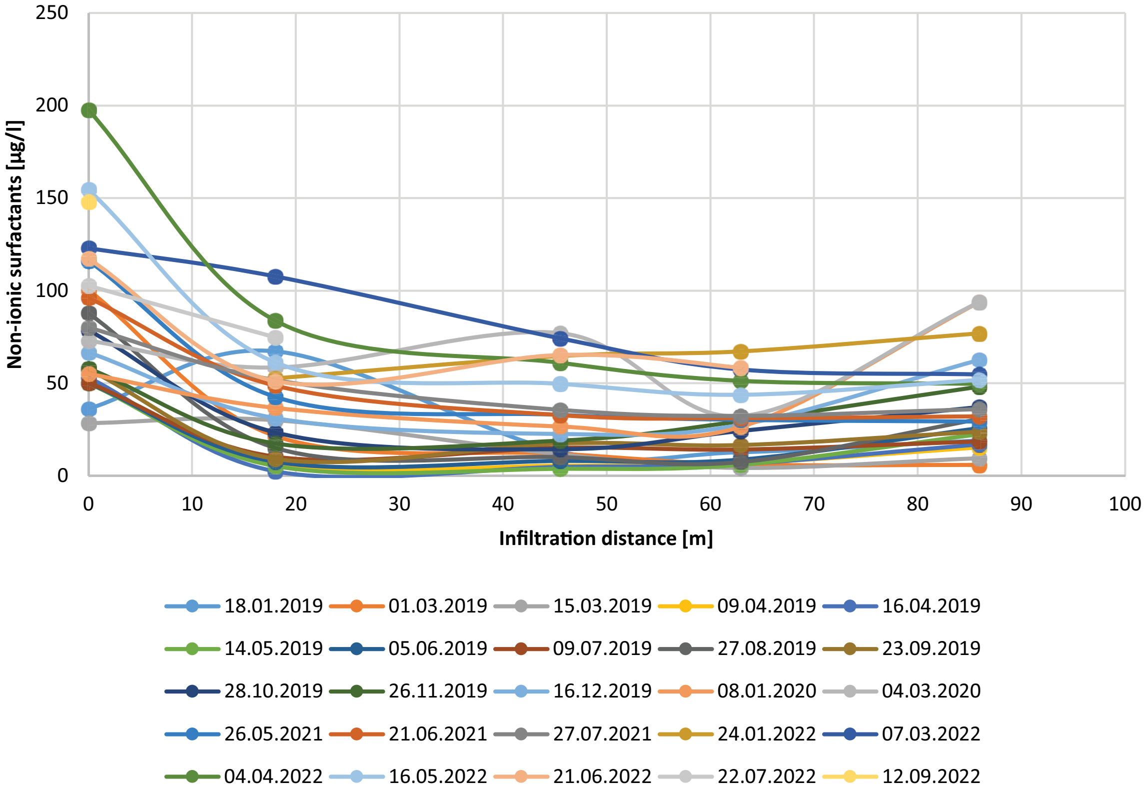

Fig. 7. Relationship between non-ionic surfactants concentration and infiltration distance.

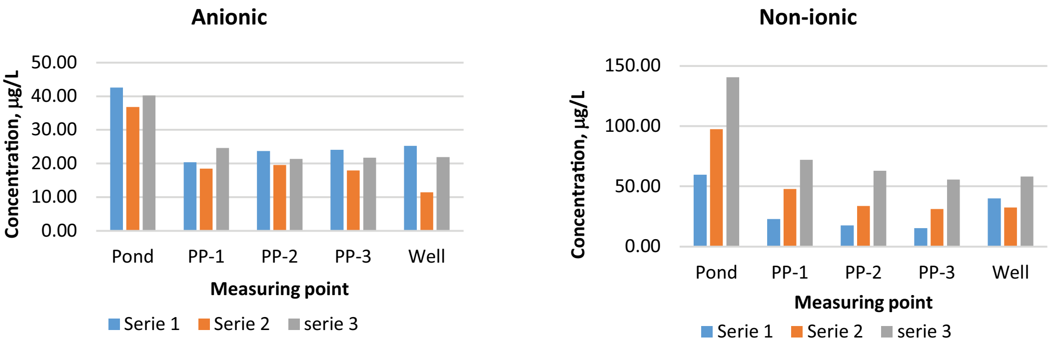

Fig. 8. Average concentrations of anionic and non-ionic surfactants determined in research series at given measuring point.

Table 12
Durfactant removal efficiency

Data	Temperature at the pond (°C)	Anionic E-total (%)	Non-ionic E-total (%)
18.01.2019	1.0	0.00	47.35
01.03.2019	4.0	26.38	94.19
15.03.2019	6.0	25.71	66.40
09.04.2019	11.9	48.13	69.57
16.04.2019	10.5	54.50	68.02
14.05.2019	12.4	56.08	55.05
05.06.2019	24.8	45.41	48.68
09.07.2019	18.0	27.69	62.91
27.08.2019	23.1	59.66	65.12
23.09.2019	14.3	63.14	58.36
28.10.2019	11.5	7.47	52.83
26.11.2019	5.5	19.17	17.13
16.12.2019	10.5	37.81	6.25
08.01.2020	2.2	36.14	0.00
04.03.2020	4.9	47.38	0.00
26.05.2021	14.4	69.56	74.71
21.06.2021	26.2	68.18	66.59
27.07.2021	21.2	69.10	55.09
07.03.2022	3.2	40.99	55.46
04.04.2022	5.5	69.09	74.88
16.05.2022	18.7	38.79	66.51

occurs. The aerobic environment favors further aerobic biodegradation of the organic matter. On the other hand, organic matter and other microelements may be adsorbed in the soil. Reducing the content of oxygen and organic substances in the infiltrating water allows autotrophic bacteria to carry out the biodegradation process and the concentration of carbon dioxide increases.

The above-mentioned processes which occur during the infiltration allow for the removal of different even emerging contaminants, including surfactants and pharmaceuticals [45,59,61]. The infiltration route might be then considered as a type of biological and chemical reactor. In the case of the described field installation, this reactor can be divided into sections corresponding to the water flow path to the subsequent measurement points. The field installation divides the infiltration route into 4 sections (reactors): pond-PP1; PP1-PP2; PP2-PP3, PP3-well. To determine whether sorption or biodegradation processes prevail in a given reactor, the EMS test was used. In this test, the coefficient S is calculated as a quotient of the decrease in the value of the chemical oxygen demand in the infiltrating water in the reactor - ΔCOD-KMnO$_4$ and the decrease in the concentration of dissolved oxygen – ΔO_2 on the same infiltration route. The value of the S coefficient indicates the dominance of one of the processes: adsorption or biodegradation as listed in Table 13 [61].

The classification of processes at each infiltration distance according to the EMS test is presented in Table 14. The values of the S coefficient, ΔCOD-KMnO$_4$ and ΔO_2 presented in the table show that the biodegradation process

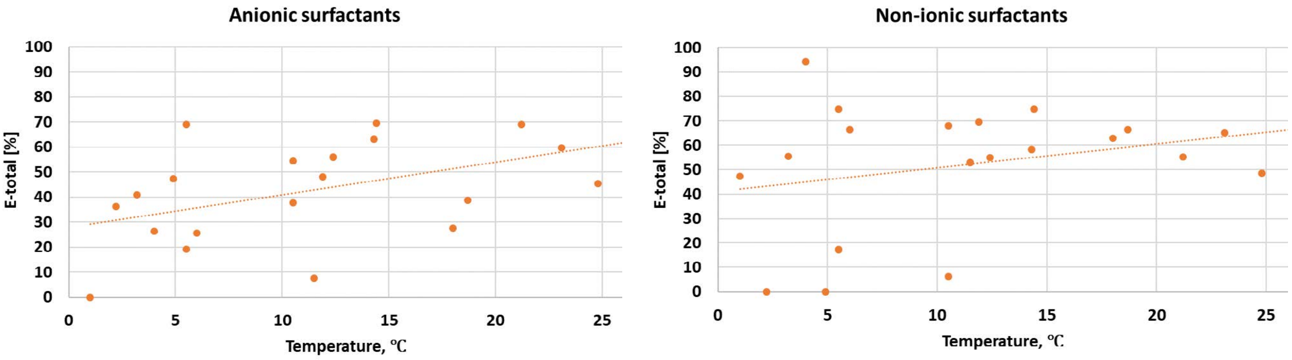

Fig. 9. Correlation between the removal of anionic and non-ionic surfactants and temperature.

Table 13
Classification of processes according to the EMS test

S = 1	Adsorption and biodegradation happen with identical intensiveness
S < 1	Biodegradation dominates
S > 1	Adsorption dominates
S = 0, ΔCOD-KMnO$_4$ = 0, $\Delta O_2 > 0$	Sorption and biodegradation processes stopped
S undetermined, ΔCOD-KMnO$_4$ > 0, $\Delta O_2 = 0$	Sorption present, biodegradation absent
ΔCOD-KMnO$_4$ = 0, $\Delta O_2 = 0$	Sorption and biodegradation absent

Table 14
Classification of processes at each infiltration distance according to the EMS test

Infiltration distance	ΔCOD-KMnO$_4$	ΔO_2	S	Classification
Pond-PP1	6.7	3.5	0.52	Biodegradation dominates
PP1-PP2	0	0.2	0	Sorption and biodegradation processes stopped
PP2-PP3	0.7	0	Undetermined	Sorption present, biodegradation absent
PP3-Well	0.3	0	Undetermined	Sorption present, biodegradation absent

dominates over adsorption in the first section of infiltration from the pond to PP1. This most likely occurs due to the greater oxygenation of the water. Under anaerobic conditions, biodegradation processes are limited. In the following sections the biodegradation does not occur and the process of adsorption dominates. The adsorption process is an effective method of removing CEC from water but its performance depends on the type of adsorbent and parameters such as temperature, pH and contact time [64,65]. The undoubted advantage is that adsorption is a physical process, which does not promote the formation of by-products that may be more toxic compared to the parent compounds [66–68].

It is noteworthy that the total removal efficiency of both types of surfactants in the infiltration process is comparable. It should be noted, however, that the concentration of non-ionic surfactants in the pond increased significantly during the pandemic and the efficiency of their removal remained at a comparable level (series 2 and 3). This indicates good conditions for the biodegradation of non-ionic surfactants in the infiltration process.

It should be noted that before the pandemic period anionic surfactants usually dominated in surface waters in comparison to non-ionic surfactants. In the first series of studies, the concentrations of both surfactants in the pond were comparable. Production of surfactants is steadily increasing, and production of non-ionic surfactants has begun to outperform anionic production especially within the pandemic period. Our studies also confirm this and show the dominant content of non-ionic surfactants (140 µg/L on the average) compared to the average concentration of anionic surfactants (43 µg/L in infiltration pond).

The majority of the commonly utilised anionic surfactants are relatively easily removed during pretreatment processes simultaneously with other pollutants. They are also readily biodegradable under aerobic conditions, resulting in low concentrations in the effluent from the wastewater treatment plant [69]. However, non-ionic surfactants, mainly oxyethylates, are only partially biodegradable by shortening their oxyethylene chain, which allows for the formation of more stable and toxic products. These metabolites are characterised by significant activity which disrupts the hormonal balance due to their similarity to natural estrogens [17,70]. The fact also draws attention to the need for further analyzes to determine the possible formation of intermediate biodegradation products of the tested surfactants.

5. Conclusions

Increasing concentrations of surfactants in natural waters (surface and groundwaters) encourage research and development of methods for their removal [13,45,71]. Evaluation of the effectiveness of different methods for removing surfactants from water taken in for human consumption is important because of the insufficient removal of surfactants by commonly used wastewater treatment systems and formation of biodegradation metabolites, which are often much more hazardous for the environment than the parent pollutants. Usually, they are also more resistant to further biodegradation [70].

Presented studies carried out during the pandemic in 2020–2022 show a significant increase in the concentration of non-ionic surfactants in water taken from the infiltration pond. The research results presented here indicate that artificial infiltration can be treated as a robust and effective barrier for elimination of surfactants from treated water before the treatment plant technology. The interpretation of results allowed to establish the following conclusions:

- the development of the production of surfactants results in increased levels of such compounds in natural waters; this was visible especially during the pandemic in 2020–2022, when the concentration of non-ionic surfactants more than doubled,
- interpretation of the test results allowed to determine that the effectiveness of removal of anionic surfactants in the process of artificial infiltration ranged from 41% to 69% and in case of non-ionic surfactants from 33% to 67%; the total removal efficiency of both types of surfactants in the infiltration process is comparable,

- the results of the study of changes in the concentration of surfactants in relation to the length of the infiltration path indicate that the first sector of infiltration path is the most important; the major treatment effect occurs on the way from the pond to the first metering point – the PP-1 well; this can be associated with greater water oxygenation at the initial stage of infiltration, explained by the significance of the sediment layer of at the bottom of infiltration pond and because of greater oxygen content in the water at the initial stage of infiltration, further passage improves the quality of infiltrated water to a lower extent with respect to surfactants removal,
- the processes evaluation by EMS test showed that the processes of biodegradation prevail over the processes of adsorption at the first section of infiltration path from the pond to the metering well PP1; in further sectors of the infiltration route conditions for adsorption prevail and biodegradation processes stop; this means that in the processes of water purification, the initial high efficiency is associated with the presence of biological decomposition processes of organic pollutants in the first sector of the infiltration path. The effect of surfactants removal increases with the temperature, which is typical for the biodegradation processes.

The monitoring of micropollutants, including surfactants, in the environment and protecting against their migration to soil and water is very important from the point of view of water production [30,71]. Another concern is the proper monitoring of produced metabolites. The products of biodegradation may be toxic due to significant biological activity which disrupts the hormonal balance of organisms. The qualitative and quantitative characterisation of metabolites is a goal for future studies.

Acknowledgments

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