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Changes in the concentration of aluminum in water after the treatment process - experience from technological research

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ABSTRACT

Technological research was carried out to determine an effective treatment technology for surface water characterized by low alkalinity, high-temperature volatility and an occasionally high content of organic substance. Three large technical installations operating in parallel were analyzed: coagulation with sludge flotation, micro-sand ballasted coagulation with sedimentation, coagulation with sedimentation and sludge recirculation. The capacity of the installations ranged from 10 to 40 m^3/h. The critical treatment process for the analyzed water was coagulation in an acidic environment (6.5 < pH < 7.0) carried out in a system with rapid mixing, a flocculation chamber, preliminary separation of coagulation products and removal of residual suspended solids through filtration. The effectiveness of the treatment technology was assessed based on such parameters as the content of the organic substance and the iron and aluminum concentrations. The purpose of the research was to select the primary treatment technology for the surface water to be used ultimately in the energy sector. In addition to organic compounds, aluminum also turned out to be a critical parameter for deciding about the effectiveness of the primary water treatment. Therefore, particular attention was paid to this parameter - it was controlled in connection with other relevant technological indicators.

Keywords: Aluminum; Full scale technological investigation; Water treatment; Coagulation; Filtration

1. Introduction

The essential requirements for the quality of water used for energy purposes are set out in the relevant regulations and guidelines. Regardless of these requirements, each installation (power plant) has its individual characteristics and conditions (because of the construction and the manner of operation of the equipment, type, and parameters of boilers or types of cooling circuits). These determine individual criteria and the required parameters of the quality of water used. In recent years, based on modernized installations in Poland, it can be concluded that the requirements for water and steam quality are tightening.

In our case, individual criteria for treating water have been defined by the investor, taking into account the warranty requirements set out by the contractor for new power units. Among these criteria was also limited aluminum concentration. In a specific technological system for cooling the water of this power plant, the excess of aluminum in the circulating water could cause the formation of undesirable deposits in installations.

The research results presented in this article are original and relate to research carried out on a technical scale simultaneously on three different research installations.

Industrial (Power) plants often use their surface water intakes, requiring the use of extensive technological systems for their treatment. In such systems, classic water treatment methods such as coagulation, sedimentation, filtration, and

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sorption are usually only preliminary water preparations for further purposes. However, it is crucial for the effectiveness of further treatment in the more advanced technological processes such as the ion exchange or the membrane processes.

The basic unit process in the preliminary water preparation for the needs of the energy industry, due to the high demand for the surface water, is the coagulation process. An increase in the effectiveness of this process leads to the increased efficiency of further treatment stages and their better economic effect. To achieve the maximum effect of suspension removal, high doses of chemicals (coagulants and flocculants) are often used. As a result, the effectiveness of mineral and organic particle removal may be reduced and the content of residual aluminum in water may increase. Aluminum found in water is in its trivalent oxidation state. Depending on the water pH value, it may be present in the dissolved or colloidal forms or occur in organic-mineral combinations.

In the process of water treatment involving coagulation, the commonly used compounds include aluminum sulfates ($Al_2(SO_4)_3{\cdot}18H_2O$ or $Al_2(SO_4)_3{\cdot}14H_2O$ and aluminum chloride ($AlCl_3$), as well as, less frequently, sodium aluminate ($Na_2O{\cdot}Al_2O_3$). Currently, the use of polyaluminum chlorides with the general formula ($Al_x(OH)_yCl_{3x-y}$) and polymerized aluminum salts that are, aluminum polyhydroxychlorosulfates is gaining in popularity since the dosing of the coagulants supplied in liquid form is easier. Numerous studies have been devoted to detailed analysis of the forms of residual aluminum found in the water after the coagulation process using coagulants with varying degrees of polymerization [1–4].

Aluminum salts used for coagulation release Cl^- and/or SO_4^{2-} anions upon hydrolysis and react with the alkaline minerals of water, reducing its pH value after coagulation [5,6]. Study results show that when the coagulation process is executed using a coagulant with a low degree of polymerization, aluminum monomers can be converted into polymers regardless of the pH value. The value of pH determines the intensity of the process. This means that coagulants that originally contain considerable amounts of monomers can be more effective than the ones with a limited content of monomers [7–12]. When the abstracted water is characterized by low alkalinity and aluminum sulfate is used, it may be necessary to enhance the buffering capacity of water, for instance, by using calcium hydroxide, sodium hydroxide or calcium carbonate, which are capable of binding strong acids released upon the coagulant hydrolysis. The quality of water after technological processes using aluminum salt depends to a high extent on the alkalinity of the water. Coagulation may result in the lowering of the pH value and a resulting increase in the content of dissolved aluminum which is more difficult to capture in conventional treatment processes [13–18].

In the technological investigations which are the basis for this paper, aluminum sulfate was used as a coagulant. The purpose of the research was to select the primary treatment technology for surface water to be used as cooling water in power plants. The most important types of contamination to be removed from water were organic compounds. The best effects derived from removing organic compounds were obtained while maintaining the pH after coagulation at 6.5 to 7.0. However, during investigations, aluminum also turned out to be significant from the quality requirements of treated water [19]. This paper supplements the information presented in earlier publications concerning, inter alia, the effectiveness of reducing the content of organic matter in water [20–22].

2. Methods

The research took place over a two-year period, and the part of the research discussed in this article lasted 5 months. The research aimed to determine water treatment technology for water to supply a cooling water circuit for a large power plant working in Poland. The quality of treated water obtained as a result of pre-treatment processes, enabling its application in a cooling circuit, had to meet individual criteria set by the Investor and described in Table 1.

Raw water from a river to a maximum amount of 85 m³/h was delivered to the contact tank, where primary oxidation was carried out using chlorine dioxide added to the pipeline, before the static mixer. The dose of chlorine dioxide ranged from 0.8 to 1.25 mg/L. After preliminary oxidation, water was split into three independent technological lines where the following processes were carried out:

- Coagulation + flotation and filtration through the anthracite and quartz sand bed (capacity of 25 m³/h). Coagulation products were removed together with air bubbles and transported to the water surface. Coagulant dose was 3.0–7.8 mg Al/L (Fig. 1).
- Coagulation + sedimentation with sludge recirculation and filtration through the anthracite and quartz sand bed (capacity of 20 m³/h). Coagulation products were removed by gravity flow in a multi-stream settler. Prior to its delivery to the sludge section, sludge was first recirculated to the initial chamber. Coagulant dose was 2.4–5.0 mg Al/L (Fig. 2).
- Micro-sand ballasted coagulation + sedimentation and filtration through the anthracite and quartz sand bed (capacity of 40 m³/h). Removal of coagulation products was enhanced by micro-sand, which after sedimentation and hydrocyclone washing was returned to the process. Coagulant dose was 2.25–5.0 mg Al/L (Fig. 3).

Table 1
Quality of treated water

Parameters and content	Unit	Requirements for treated water
pH	–	6.5 < pH ≤ 8.5
Total hardness	mval/L	≤3.2
Total alkalinity	mval/L	≤2.0
Total suspended solids	mg/L	≤2.0
Total iron	mg Fe/L	≤0.05
Total aluminum	mg Al/L	≤0.1
Chlorides	mg Cl/L	≤90
Sulfates	mg SO_4/L	≤112
TOC	mg C/L	≤4.0
COD $KMnO_4$	mg O_2/L	≤7.5

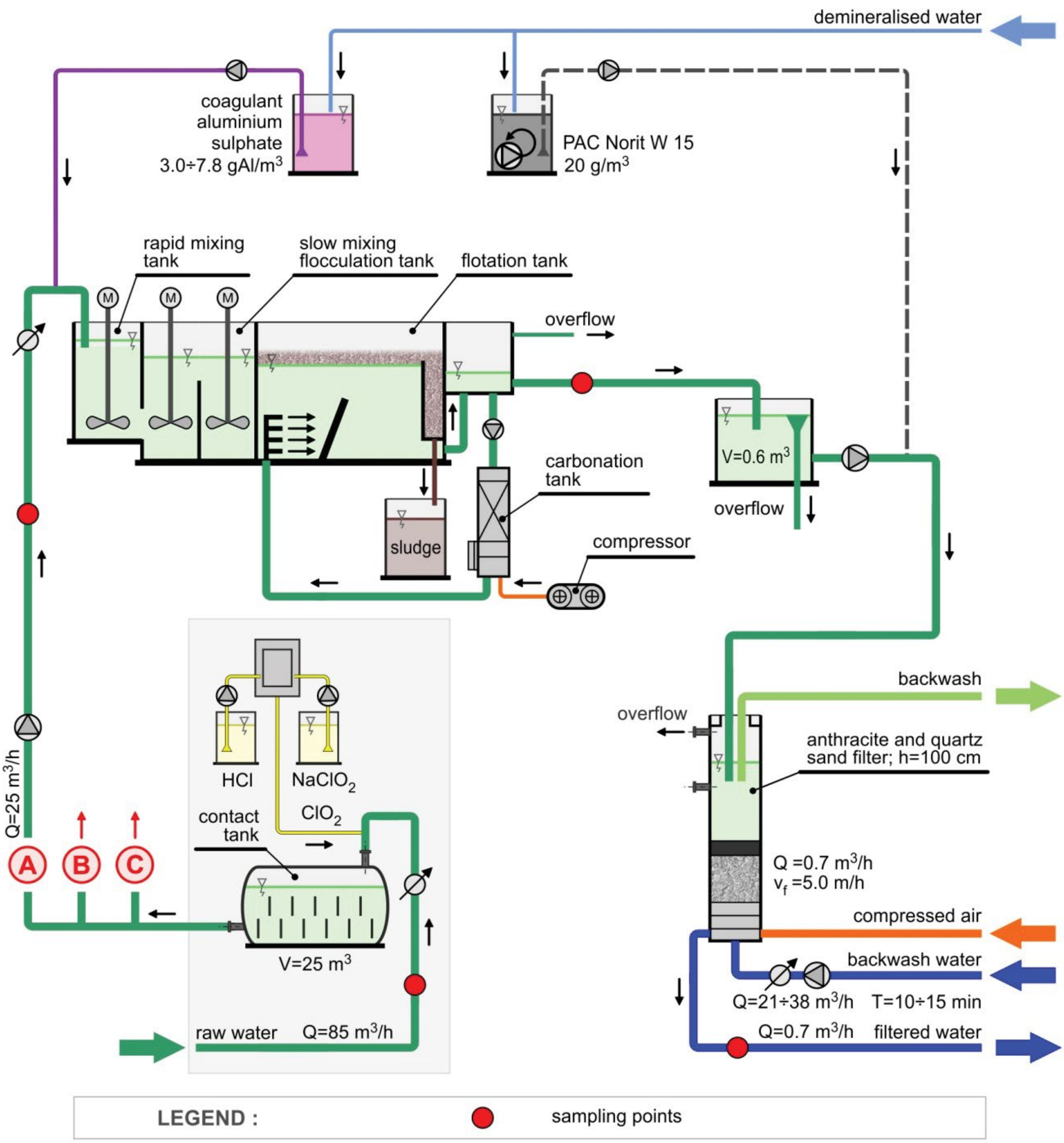

Fig. 1. Flow chart of the technological processes analyzed during the coagulation with sludge flotation and filtration.

Aluminum sulfate was used as a coagulant, individually and automatically dosed into each of the process lines. Coagulation was carried out in an acidic environment (6.5 < pH < 7.0). The acidic environment was achieved by coagulant dosing with the correction of soda carbonate. Because of the wide scope of raw water quality changes, the optimum dose of coagulant was determined via a once or twice a day jar test.

Subsequently, the water passed through a rapid filter with hydro-anthracite N and quartz sand bed. Filtration velocity was 5.0–7.5 m/h. The filter was filled with 300 mm of hydro-anthracite N (effective size de = 1.9 mm, uniformity coefficient < 1.5) and 700 mm of quartz sand (effective size de = 0.955 mm, uniformity coefficient = 1.28). To check the possibility of increasing the effectiveness of total organic carbon (TOC) removal, between 25 and 29 March

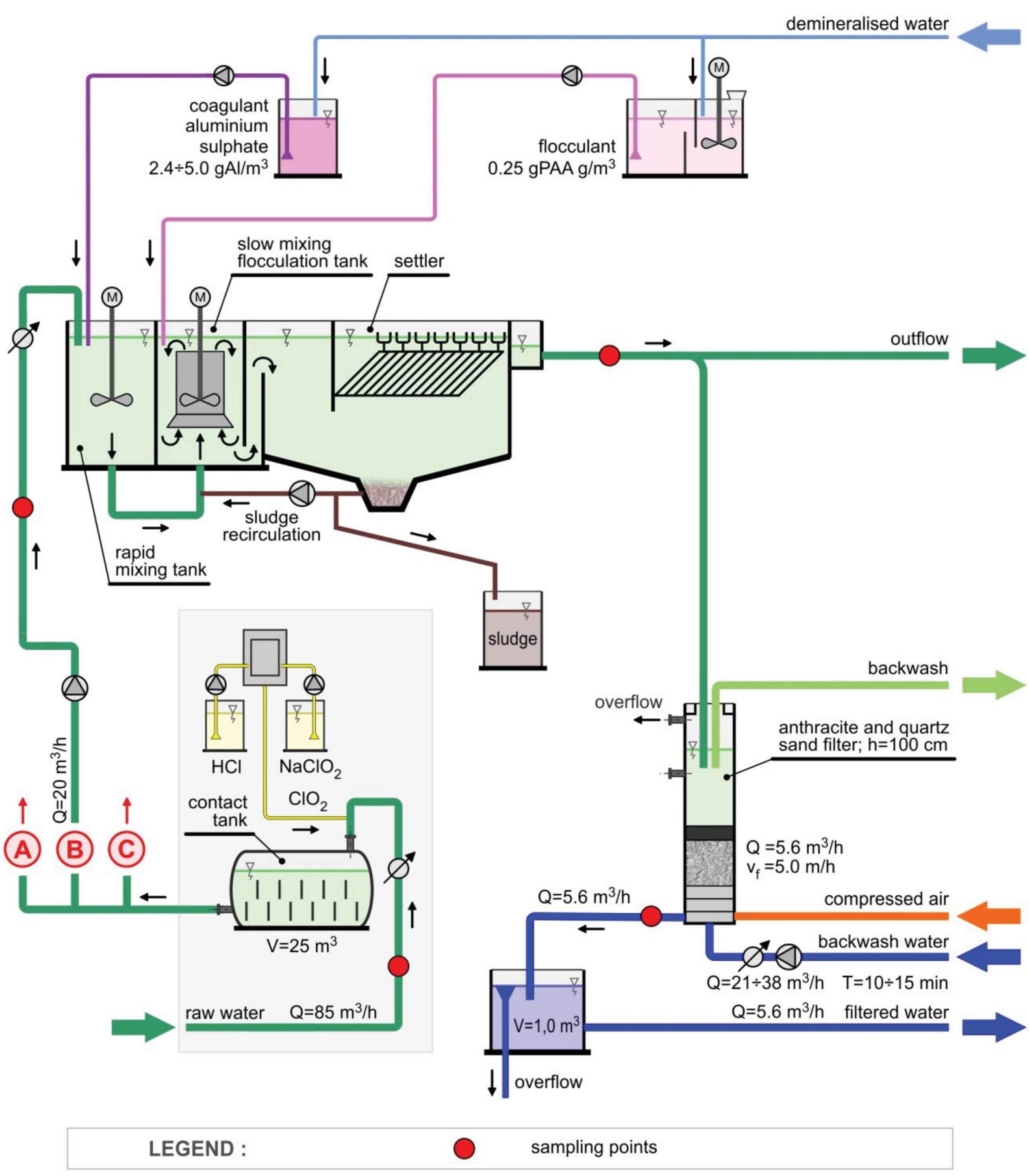

Fig. 2. Flow chart of the technological processes analyzed during the coagulation, sedimentation with sludge recirculation and filtration.

2013, 20 mg C/L powder activated carbon (PAC) NORIT W 15 (Brenntag Company) (Iodine number 1200, Molasses number 200, Methylene blue adsorption 22 g/100 g; total surface area (Brunauer–Emmett–Teller method) 1,150 m^2/g, particle size D50 = 15 μm) was added to the system before the filters. The hydraulic retention time for PAC before it entered the sand bed was about 10 min.

In the case of experimentation with sludge sedimentation, a flocculant was also used in addition to a coagulant. In the system with sludge flotation, flocculant was not used. A detailed description of the pilot station is presented in earlier publications [21,22].

Samples for analysis were taken 3 times daily at 7.00, 14.00 and 20.00. For the physicochemical analyzes, samples

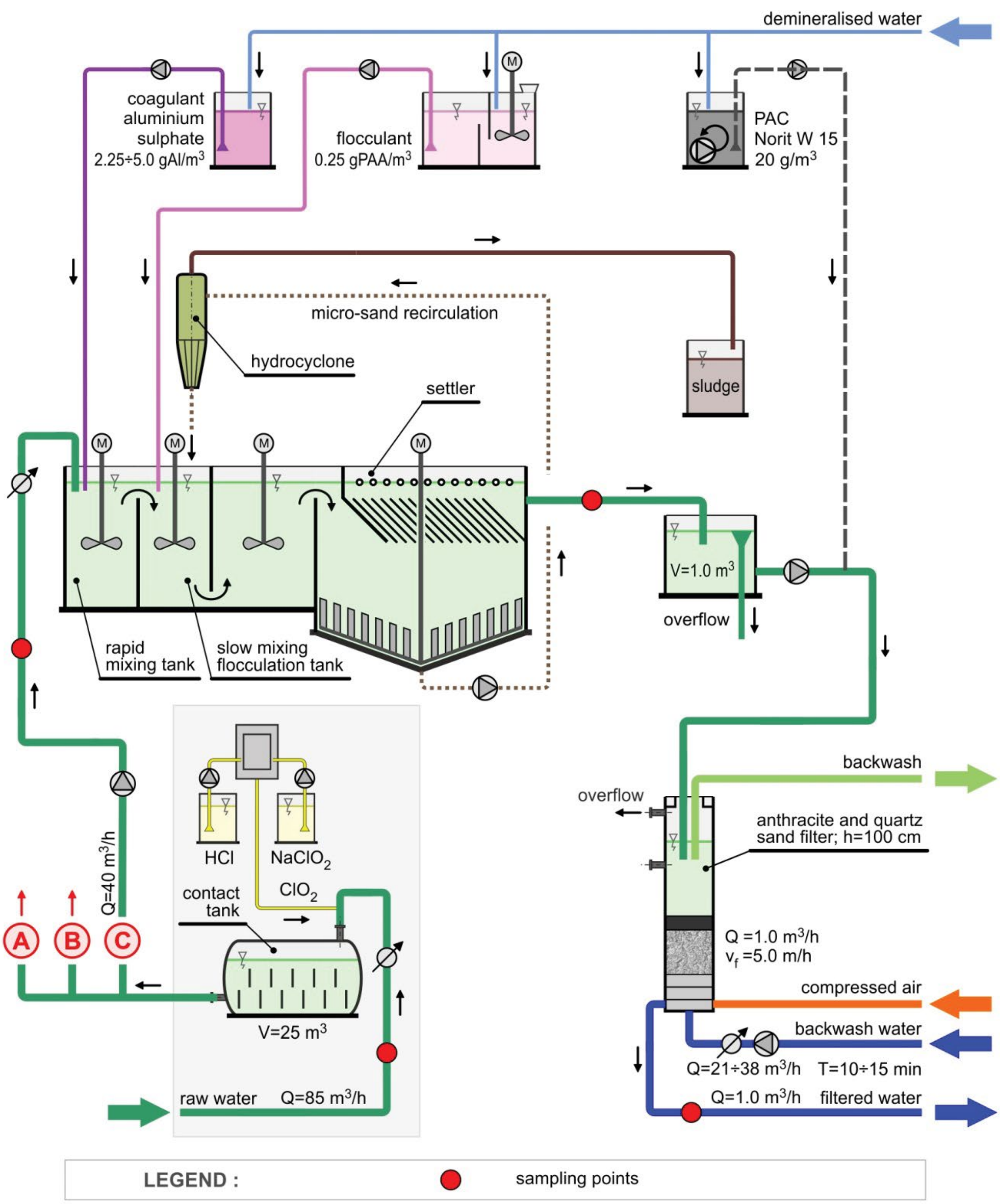

Fig. 3. Flow chart of the technological processes analyzed during the micro-sand ballasted coagulation with sedimentation and filtration.

were collected from the raw water, the water after coagulation and water after filtration. In total, physical and chemical analyzes of 36 water samples were performed daily. They were carried out by certified laboratories (Table 2). The analytical methodology complied with the Standard Method [23]

Due to the fact that the aluminum remaining in the water occurs mainly in the form of small particles, which due to their size (1–5 μm) do not affect the turbidity of the water [4] in the conducted research, instead of turbidity, the silt density index (SDI) was determined.

SDI is a measurement of the fouling potential of the suspended solids. It does not measure the quantity of particular matter since the size and shape vary. Turbidity is a measurement of the amount of suspended solids. They are not the same and there is no direct correlation between them. In practical terms however, the membranes show very little fouling when the feed water has a turbidity of <1 NTU.

Correspondingly, the membranes show very low fouling at a feed SDI of less than 5. The SDI test is used to predict and then prevent the particulate fouling on the membrane surface. Other names for it are the Kolloid-Index or the Fouling-Index. The test is defined in the ASTM Standard D4189, the American Standard for Testing Material.

This standard measures the time required to filter a fixed volume of water through a standard 0.45 μm pore size microfiltration membrane with a constant given pressure of 30 psi (2.07 bar). The difference between the initial time and the time of the second measurement, normally after 15 min (after silt-built up), represents the SDI value (https://www.lenntech.com/sdi.htm#ixzz64QAHb65i).

The flow charts of the technological processes analyzed during the three large technical installations operating in parallel are presented in Figs. 1–3.

3. Results and discussion

During the study period, the surface water captured was characterized by aluminum content in the range of 0.041–0.274 mg Al/L. The content of organic compounds was high: TOC from 6.3 to 18.4 mg C/L, chemical oxygen demand (COD) $KMnO_4$ from 4.43 to 12.35 mg O2/L. Selected parameters of raw water are presented in Table 3. Parameters relevant for testing (e.g. pH) were determined more often than others. On the other hand, less important parameters (e.g. total hardness) were measured with a lower frequency. After comparing the quality of raw water and the requirements for treated water (Table 3), key parameters determining the effectiveness of water treatment were determined. They were TOC, total iron, total aluminum, total suspension and COD $KMnO_4$.

Changes in the concentration of residual aluminum found in the water after the treatment processes are presented in Figs. 4, 6 and 8. The comparison of the aluminum content in water after the application of the investigated treatment lines shows significantly higher values in the case of processes with sludge flotation. According to the calculated average values throughout the entire study period presented in Tables 4–6, after flotation, the concentration of aluminum was nearly 50% higher than the values recorded for other processes. Incidentally, in some cases, the concentrations determined after filtration were even higher than 0.1 mg Al/L. Similar but less frequent excess values were also found in the water after other technological lines. Worth noting is the fact that an increase in the filtration rate to 7.5 m/h had no significant impact on the concentration of residual aluminum (Table 7). Fig. 5 shows no correlation between a pH value ranging from 6.5 to 7.1 and the concentration of residual aluminum. Likewise, as demonstrated in Fig. 6, reduced coagulant doses did not influence aluminum concentrations in the treated water. Fig. 7 shows no correlation between a dose of coagulant ranging from 2.25 to 7.8 mg Al/L and the concentration of the residual aluminum.

In view of the foregoing, it has been concluded that it is an effect of the pumps used to supply water to the filters where the flocs found in the water after coagulation are subject to dispersion. This conclusion is additionally supported by higher concentrations of residual aluminum after flotation during which no flocculant was used. The lower 'hydraulic

Table 2
Analytical test method

Parameter	Test method
pH	PN-90/C-045540/01
Total hardness	PN ISO 6059:1999
Total alkalinity	PN-EN ISO 9963:2001
Total suspended solids	PN-EN 872:2007+Ap1:2007
Total iron	PN-EN ISO 11885:2009
Total aluminum	PN-EN ISO 11885:2009
Chlorides	PN-EN ISO 10304-1:2009+AC:2012
Sulfates	PN-EN ISO 10304-1:2009+AC:2012
TOC	PN-EN 1484:1999
COD KMnO4	PN-EN ISO 8467:2001
SDI	ASTM Standard D4189

Table 3
Quality of raw water and requirements set for treated water

Parameters and content	Number of analysis	Raw water				Requirements for treated water[a]
		Min.	Max.	Average	Standard deviation	
pH	109	7.28	7.89	7.47	0.16	6.5 < pH ≤ 8.5
Total hardness, mval/L	17	2.31	3.09	2.54	0.20	≤3.2
Total alkalinity, mval/L	48	1.35	1.80	1.65	0.10	≤2.0
Total suspended solids, mg/L	53	2.8	39.0	12.29	11.69	≤2.0
Total iron, mg Fe/L	53	0.51	1.36	0.78	0.18	≤0.05
Total aluminum, mg Al/L	53	0.04	0.27	0.11	0.06	≤0.1
Chlorides, mg Cl/L	53	28	40	31.07	2.73	≤90
Sulfates, mg SO_4/L	53	50	75	59.52	9.74	≤112
TOC, mg C/L	53	6.30	18.40	9.04	2.93	≤4.0
COD $KMnO_4$, mg O_2/L	53	4.43	12.35	6.88	1.85	≤7.5

[a] requirements for the make-up water for cooling the circuit provided by the investor.

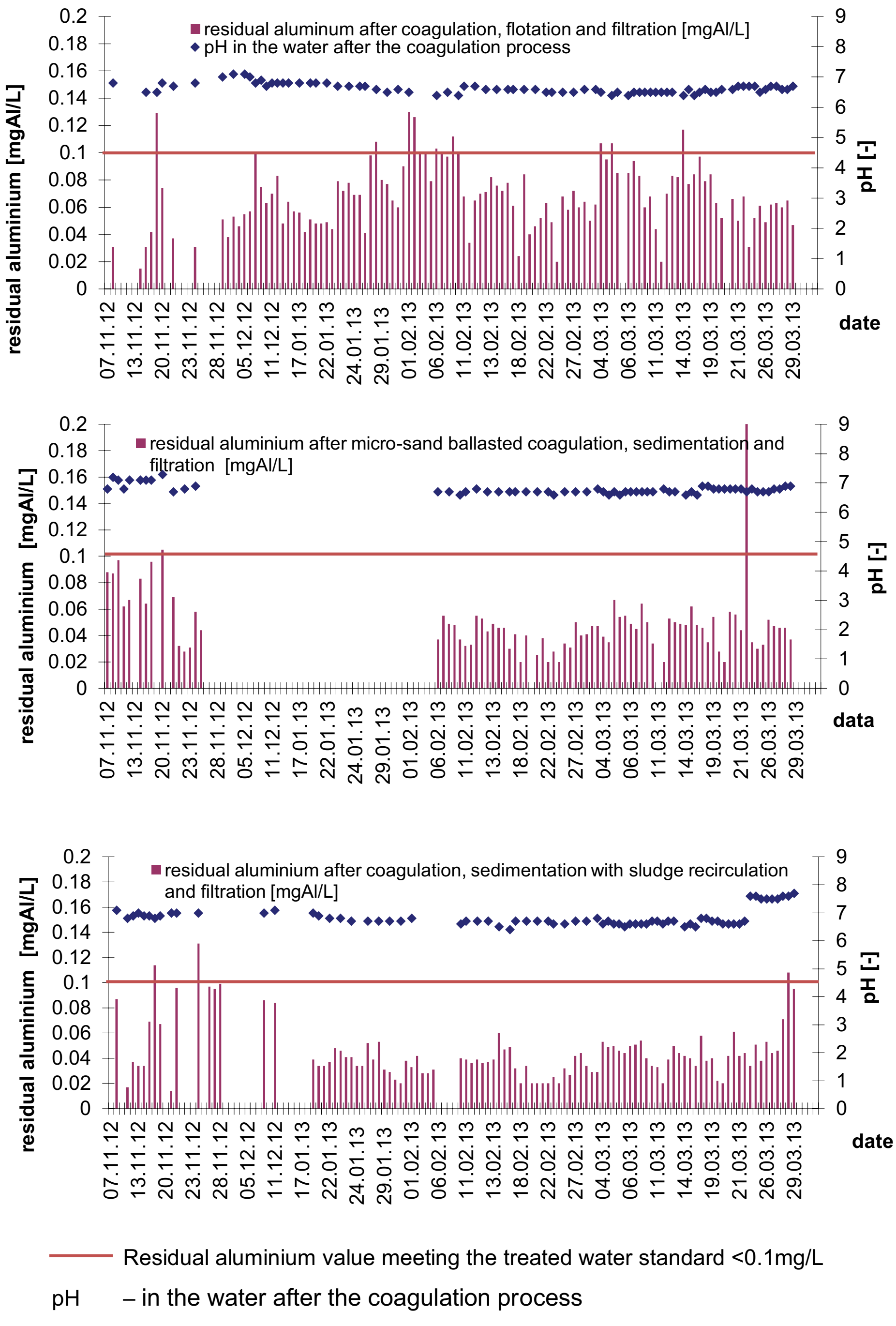

Fig. 4. Residual aluminum in water after coagulation, flotation or sedimentation and filtration depending on the pH.

resistance' of flocs increased the degree of their dispersion by the water pumps. It needs to be added that in a case where there is a gravitational flow of water, the phenomenon would most likely not occur [16].

Residual aluminum is present in water mainly in the form of small particles that do not have a significant impact on water turbidity [8]. In the research, the concentration of suspended solids past the filters was not determined and the turbidity parameter being conditional on the degree of dispersion would not provide information about the suspension mass. Therefore, a decision was made to measure the SDI. Fig. 8 presents the impact of the residual aluminum density on the value of SDI, which is crucial for deciding whether membrane methods can be used in the secondary treatment of water. A review of Fig. 9 shows that the concentration of residual aluminum in the range of determination found in the three coagulation systems did not influence the SDI value.

Fig. 5 shows the correlation between the concentration of residual aluminum in the water and the pH value. Analysis of the results showed no relationship between the concentration

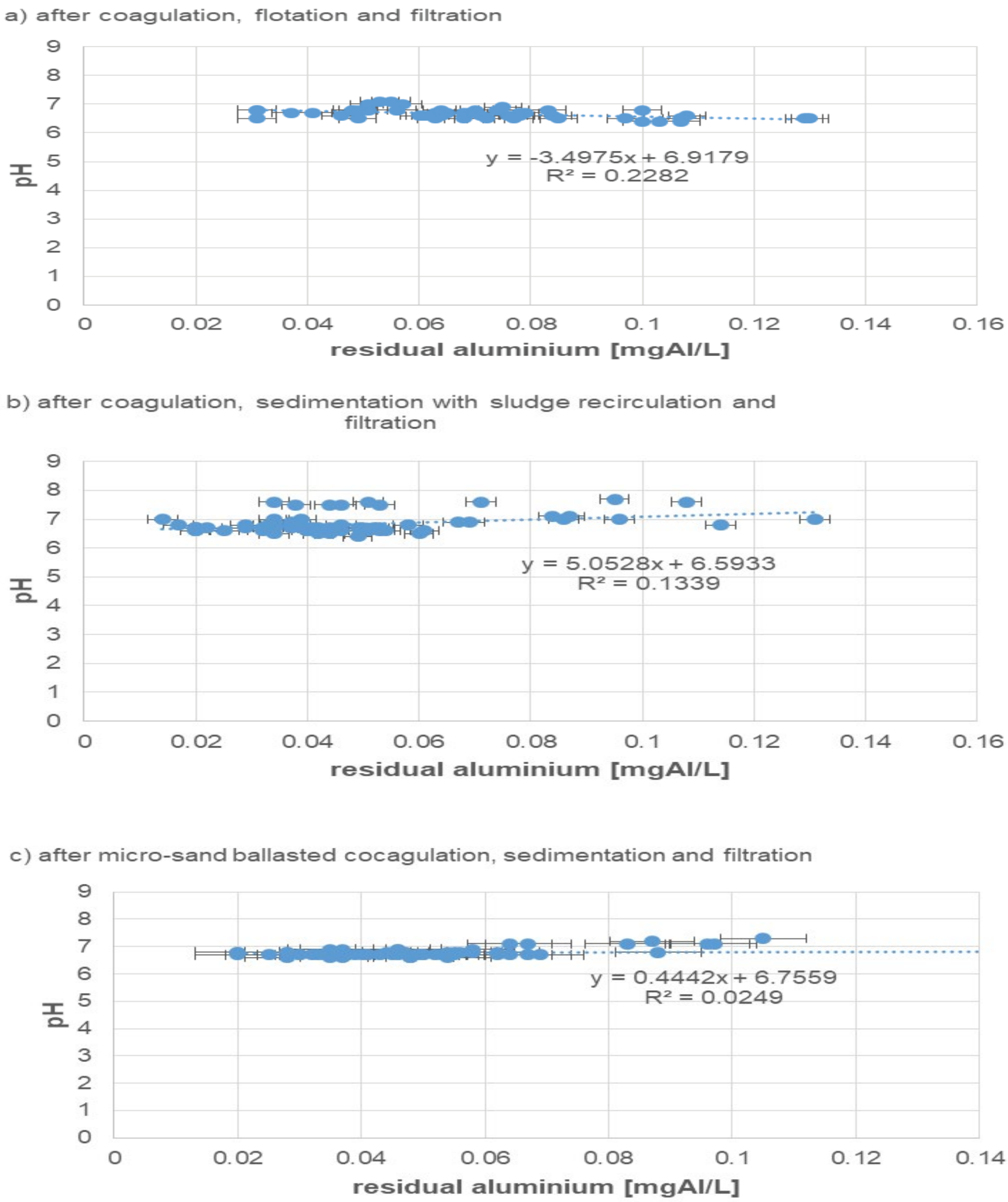

Fig. 5. Relationship between the concentration of the residual aluminum and the pH in the water after the coagulation, flotation or sedimentation, and filtration processes.

of residual aluminum in water and the pH of the water after processing. It should be noted that the operation of the process of coagulation in an acid medium (6.5 < pH < 7.0) was deemed as highly effective for removing organic compounds from the water, as presented in previous publications [20–22].

The coagulation process is a typical process commonly used for surface water treatment. Various aluminum coagulants selected based on preliminary tests are used in this process [6]. In the described studies, aluminum sulfate was selected as the optimal water coagulation agent [19] based on jar tests and short-term flow tests. When using this non-hydrolyzed coagulant, pH has a significant impact on the intensity of changes in aluminum forms [1,13].

In the case of surface water treatment, it is necessary to use varying doses of coagulant. The experiments described in other publications [6,16,17] show that in cases where there is a high content of organic impurities, it is advisable to maintain a reduced pH (6.8–6.9). Both of these relationships indicate the possibility of unstable conditions during the coagulation process of low-alkalinity water and the use

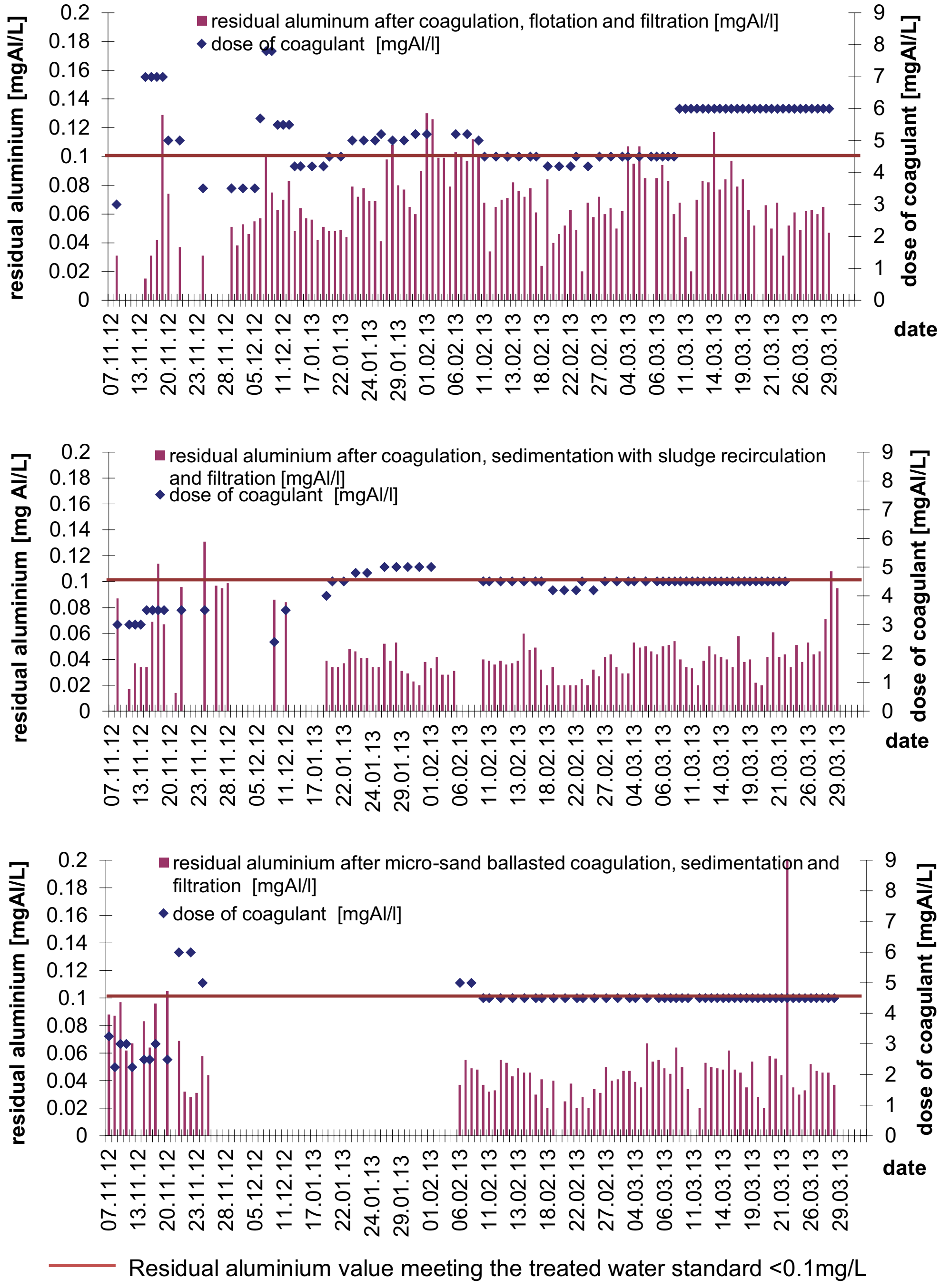

Fig. 6. Residual aluminum in water after coagulation, flotation or sedimentation and filtration depending on the coagulant dose.

of non-hydrolyzed coagulant. [19]. The possibility of these conditions, in conjunction with the variability of dissolved/colloidal aluminum forms, creates the risk of elevated aluminum concentrations in treated water.

The obtained test results indicate the stable quality of treated water with a particular regard to aluminum concentration. The concentration of aluminum in treated water under changing process conditions remained constant for each of the three tested technological lines. In the case of coagulation with the flotation of the sludge, slightly worse efficiency was obtained, which may be a reason for not using flocculant. This is in line with the conclusions of other studies [14,15].

4. Conclusions

The technological research showed 50% higher residual aluminum concentrations in water after the process of coagulation with sludge flotation than other installations. This probably results from the floc structure. In the case of sludge

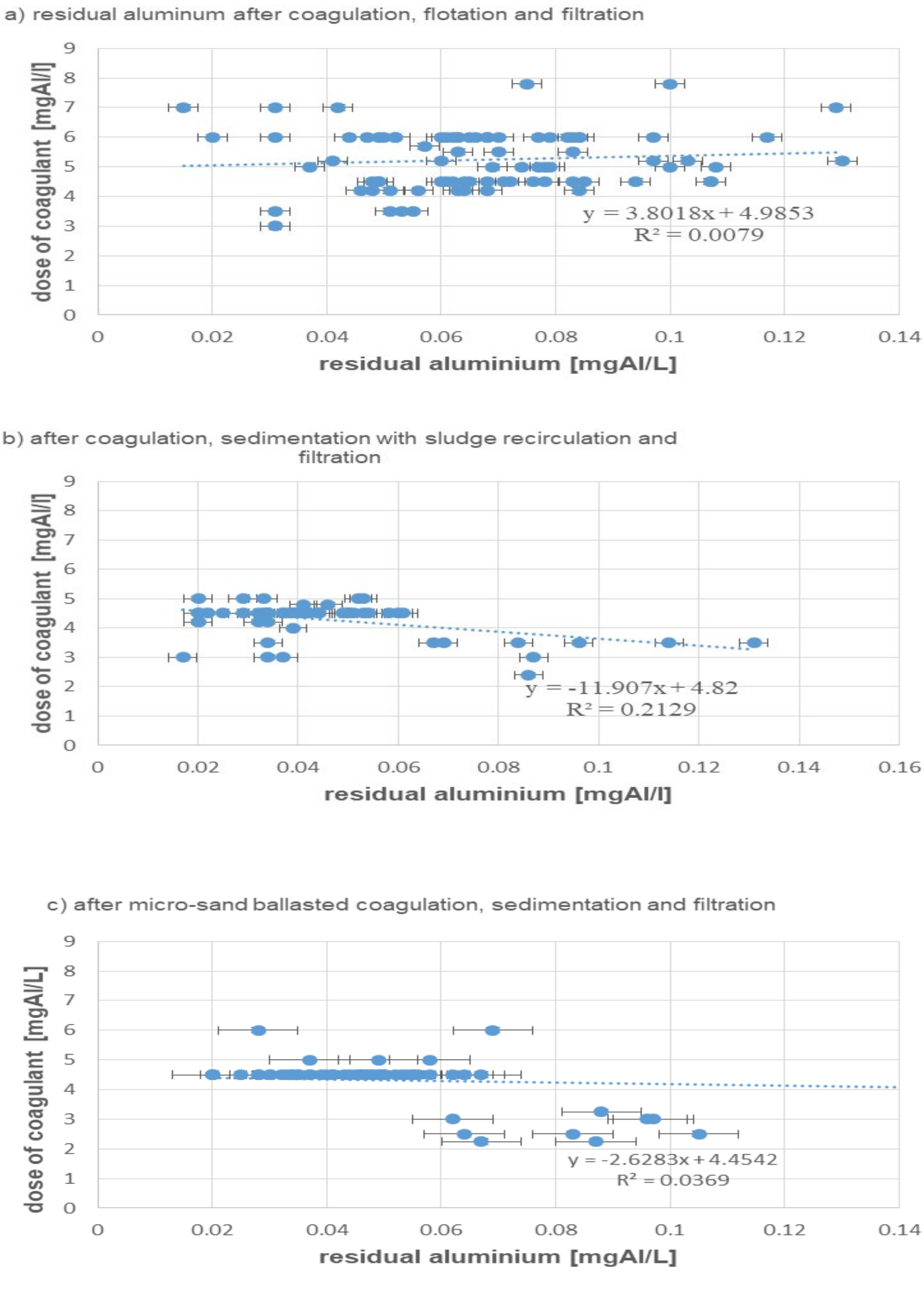

Fig. 7. Relationship between the concentration of the residual aluminum and the dose of coagulant in the water after the coagulation, flotation or sedimentation, and filtration processes.

flotation, no flocculant was used. This means that during the process of water being pumped onto rapid filters, flocs could have been dispersed and, as a result, the filter was less effective at removing smaller suspended particles containing aluminum. Therefore, it has been recommended that in the final technological process a gravity flow should be used to supply water past the flotation process to the filters.

The research showed that the pH value of the coagulation process or a coagulant dose had no impact on the residual aluminum concentrations found in the water after the coagulation and filtration processes. The studies have shown that despite different doses of coagulant, all the analyzed technological systems achieved the average concentration of aluminum remaining in the water in accordance with the requirements, which are

- 0.07 +/– 0.025 mg Al/L in the water after coagulation, flotation and filtration

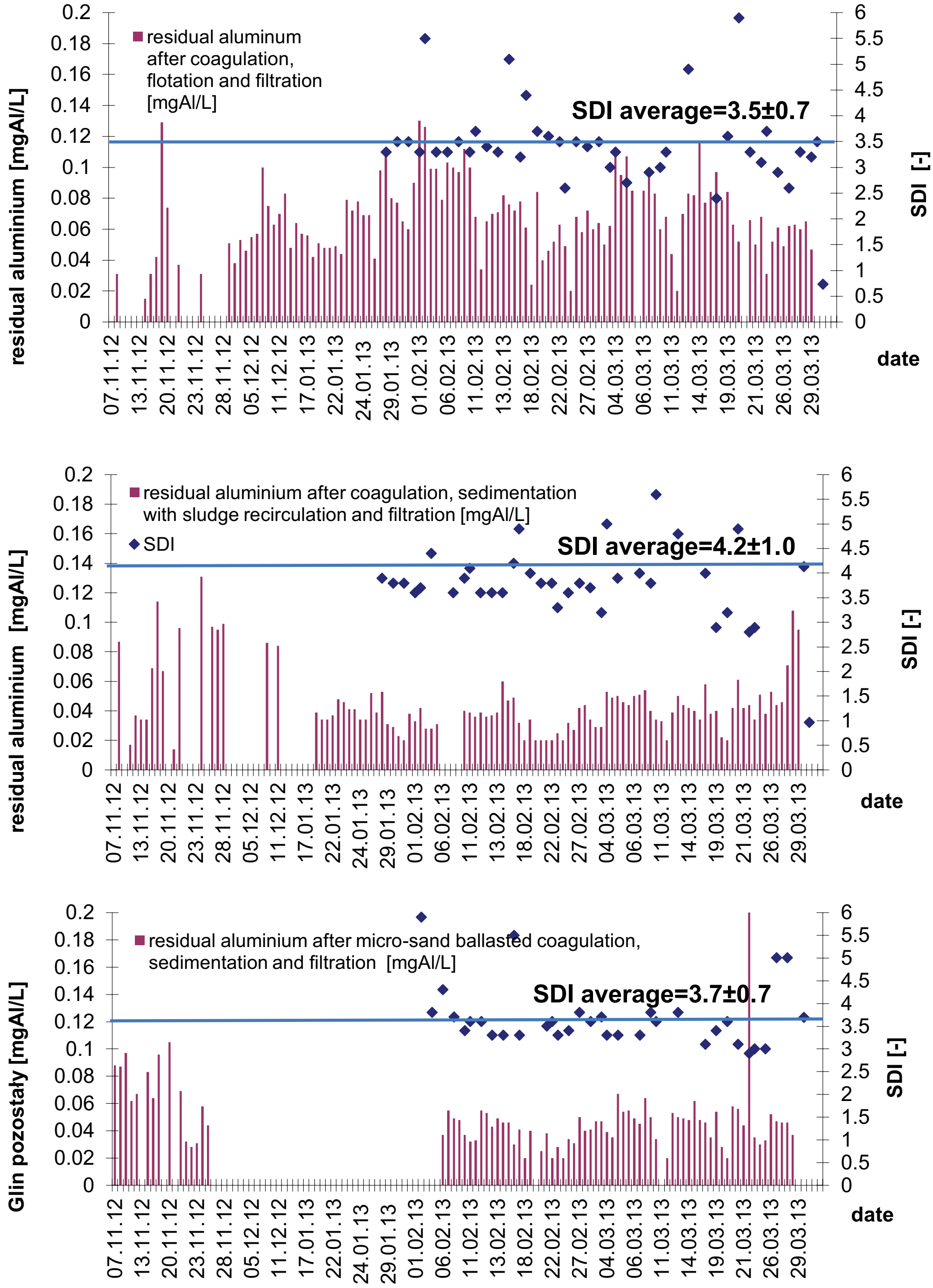

Fig. 8. Changes in SDI and residual aluminum in water after coagulation, sedimentation or flotation and filtration with an average SDI value.

- 0.046 +/– 0.025 mg Al/L in the water after coagulation, sedimentation with sludge recirculation and filtration
- 0.045 +/– 0.025 mg Al/L in the water after micro-sand ballasted coagulation, sedimentation and filtration

The treated water in the analyzed technological processes has different levels of residual aluminum. Despite this, the value of the SDI index in all cases is <5.0, which means the possibility of using membrane processes at further stages of treatment.

An additional conclusion after the research was the confirmation of the possibility of changing process parameters such as pH and coagulant dose without affecting the deterioration of water quality in the aluminum range. This fact is important for practical, stable exploitation of future water treatment plants.

Table 4
Quality of water after coagulation, flotation and filtration

Parameters and content	Number of analysis	Water after coagulation, flotation and filtration			
		Min.	Max.	Average	Standard deviation
pH	83	6.4	7.1	6.67	0.17
Total suspended solids, mg/L	37	2.0	6.5	2.22	0.43
Total iron, mg Fe/L	75	0.00	0.014	0.0055	0.002
Total aluminum, mg Al/L	108	0.02	0.13	0.07	0.025
TOC, mg C/L	97	3.40	5.80	4.50	0.67
COD $KMnO_4$, mg O_2/L	20	0.9	3.28	2.29	0.70

Table 5
Quality of water after coagulation, sedimentation with sludge recirculation and filtration

Parameters and content	Number of analysis	Water after coagulation, sedimentation with sludge recirculation and filtration			
		Min.	Max.	Average	Standard deviation
pH	76	6.4	7.1	6.77	0.16
Total suspended solids, mg/L	39	4.4	23.0	4.58	3.85
Total iron, mg Fe/L	77	0.00	0.02	0.065	0.003
Total aluminum, mg Al/L	99	0.01	0.13	0.046	0.024
TOC, mg C/L	93	4.0	7.10	4.70	0.62
COD $KMnO_4$, mg O_2/L	20	1.86	3.83	2.52	0.57

Table 6
Quality of water after micro-sand ballasted coagulation, sedimentation and filtration

Parameters and content	Number of analysis	Water after micro-sand ballasted coagulation, sedimentation and filtration			
		Min.	Max.	Average	Standard deviation
pH	62	6.6	7.3	6.80	0.19
Total suspended solids, mg/L	27	4.2	9.2	4.2	1.84
Total iron, mg Fe/L	66	0.00	0.017	0.006	0.003
Total aluminum, mg Al/L	78	0.02	0.449	0.045	0.021
TOC, mg C/L	74	3.80	6.30	4.60	0.53
COD $KMnO_4$, mg O_2/L	18	1.77	3.64	2.54	0.60

Table 7
Aluminum concentration in water after technological processes depending on the filtration velocity

Number of analysis	Al (mg Al/L) after coagulation, flotation and filtration		Al (mg Al/L) after coagulation, sedimentation with sludge recirculation and filtration		Al (mg Al/L) after micro-sand ballasted coagulation, sedimentation and filtration	
Results obtained from the analysis of samples for filtration velocity 5.0 m/h (mg Al/L)						
79	Average	0.069	Average	0.048	Average	0.049
	Max.	0.130	Max.	0.131	Max.	0.105
	Min.	0.015	Min.	0.014	Min.	0.020
Results obtained from the analysis of samples for filtration velocity 7.5 m/h (mg Al/L)						
36	Average	0.071	Average	0.043	Average	0.044
	Max.	0.117	Max.	0.058	Max.	0.449
	Min.	0.020	Min.	0.020	Min.	0.020

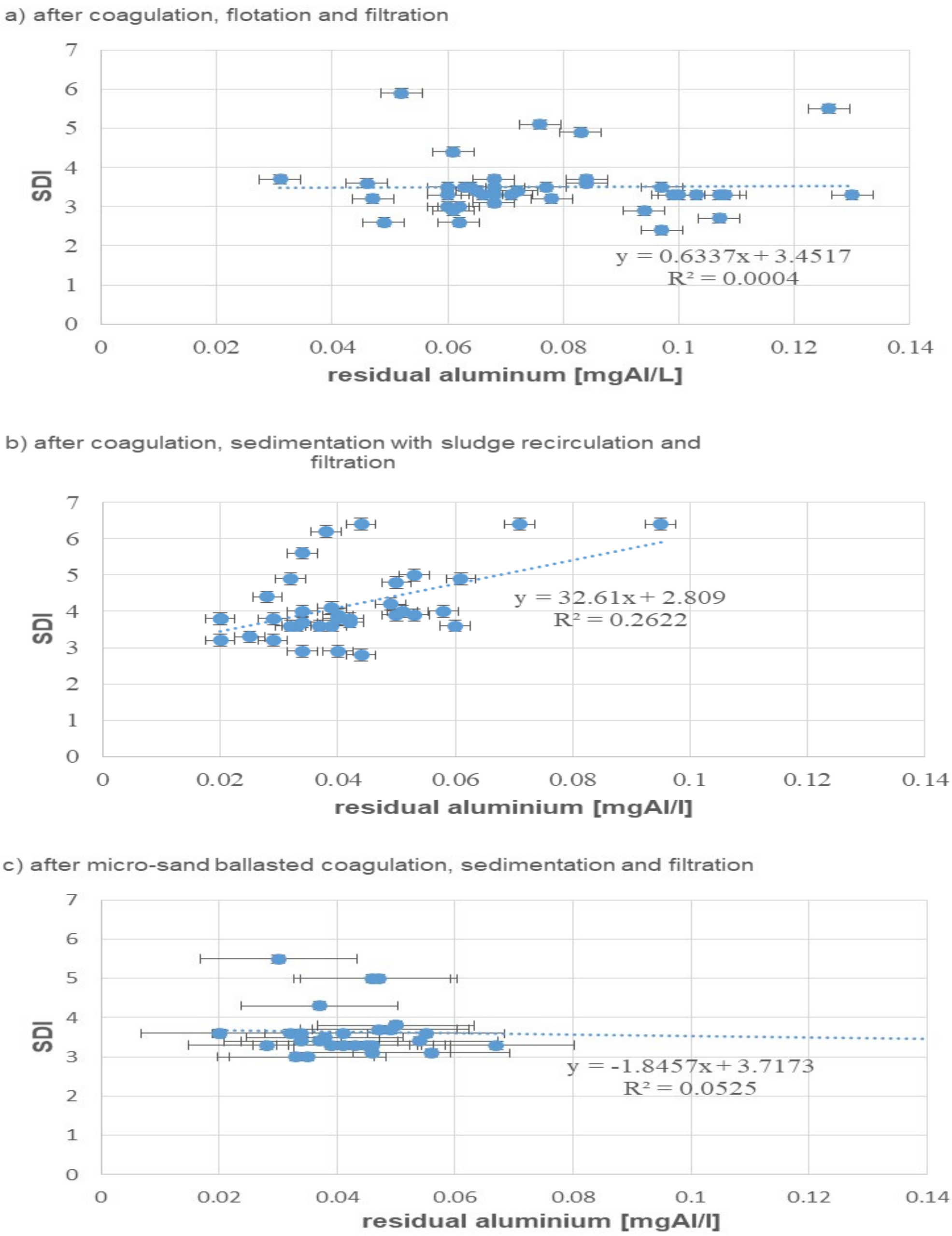

Fig. 9. Relationship between the concentration of the residual aluminum and the SDI in water after the coagulation, sedimentation or flotation, and filtration processes.

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