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Technological Parameters of Rotating Electrochemical and Electrobiological Disk Contactors Depending on the Effluent Quality Requirements

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Abstract: Soilless tomato cultivation wastewater, with typically low COD, high concentrations of phosphorus, and oxidized forms of nitrogen, may be effectively treated in a rotating electrochemical disk contactor (RECDC) and in a bioelectrochemical reactor (BER), such as a rotating electrobiological disk contactor (REBDC). The aim of this study was to determine the technological parameters of both reactors, i.e., electric current density (J) and hydraulic retention time (HRT), depending on the effluent quality requirements. The study was conducted with four one-stage RECDCs and with four one-stage REBDCs, at four hydraulic retention times, i.e., 4, 8, 12, and 24 h, and electric current densities of 0.63, 1.25, 2.50, 5.00, and 10.00 A/m^2. It was demonstrated that soilless tomato cultivation wastewater could be effectively treated in electrochemical and electrobiological disk contactors, and then discharged to sewage system facilities. In a RECDC, the highest denitrification (53.4%) and dephosphatation (99.8%) performance was achieved at J = 10.0 A/m^2 and HRT = 24 h. If the effluents are to be discharged to natural reservoirs, their effective treatment is only feasible in a REBDC. The bioelectrochemical disk contactor ensured over 90% dephosphatation effectiveness. At HRT = 24 h and all electric current densities studied, the concentrations of pollutants in the effluent met requirements set for industrial wastewater discharged into natural waters and the ground. By applying J = 2.5 A/m^2 and HRT = 24 h in the REBDC, it was possible to achieve a phosphorus concentration below 3.0 mg P/L and concentrations of ammonia nitrogen and nitrites lower than the permissible levels for treated industrial wastewater introduced to waters and to the ground. Given the nitrate concentration (exceeding 30 mg N/L), an external carbon source is recommended to aid a treatment process that uses a technological system with a REBDC. Technological schemes were proposed for wastewater treatment plants (WWTPs) with a RECDC and a REBDC, for discharging treated wastewater to natural waters, the ground, and sewage systems.

Keywords: soilless tomato cultivation wastewater; nutrient removal; bioelectrochemical reactor (BER); rotating electrochemical disk contactor (RECDC); rotating electrobiological disk contactor (REBDC); technological parameters

1. Introduction

The problem of good quality food delivery has been exacerbated by the growing global population as well as changes in lifestyles associated with the need to deliver fresh vegetables to consumers throughout the year. This, in turn, requires very efficient food production. The annual global tomato production is more than 188 million tons, which makes it the most popular vegetable across the world [1].

Cultivation of plants with soilless and hydroponic methods is conducted almost 100% in open fertilization mode which means that the nutrient medium flows through the plant cultivation system once, and then is removed from the installation [2]. In the case of "vertical farms" located not only in "smart cities", wastewater (drainage water with a high concentration of nutrients) is discharged into municipal sewage systems. Wastewater from soilless plant cultivation farms located in rural areas is usually discharged into the environment (the ground and natural waters) in an uncontrolled manner.

Considering tomato cultivation on mineral wool, the amounts of nitrogen and phosphorus may reach 23–245 kg N/ha and 2–54 kg P/ha per month, respectively, with production periods of 9–10 months. The concentrations of nutrients in wastewater from soilless cultivation of tomatoes may even reach 600 mg N/L for nitrate nitrogen and 370 mg P/L for total phosphorus, whereas its COD is below 50 mgO_2/L [3–5]. Such wastewater should not be discharged into a municipal sewage system or into the environment without an appropriate pretreatment or treatment, respectively.

Pursuant to the regulations in force in Poland regarding the discharge of industrial wastewater into municipal sewage systems [6,7], it is the owner of the system that determines the permissible values of pollution indicators in wastewater discharged by an industrial wastewater supplier Table 1. In the case of COD, BOD_5, TOC, and total phosphorus, the values of the indicators should be determined based on the permissible pollutant load of the treatment plant.

Table 1. The EU regulations for wastewater discharged into natural waters and the ground and Polish regulations regarding industrial wastewater discharged into municipal sewage systems.

Parameter	Maximum Permissible Values of Pollutants, for Biodegradable Industrial Wastewater Generated during the Production and Processing of Fruit and Vegetables, Discharged into the Water or Ground	Permissible Values of Pollution Indicators in Industrial Wastewater Introduced into Sewage Systems
COD [mg O_2/L]	125	*
Total nitrogen [mg N/L]	30	-
Ammonia nitrogen [mg N/L]	10	100 (1) 200 (2)
Nitrite [mg N/L]	1	10
Nitrate [mg N/L]	30	-
Total phosphorus [mg P/L]	3	*
Temperature [°C]	35	35
pH	6.5–9.0	6.5–9.5
Iron [mg/L]	10	10
Aluminium [mg/L]	3	10

* The values of the indicators should be determined on the basis of the permissible load of the treatment plant with the load of these pollutants. (1), Applies to wastewater discharged to treatment plants for agglomerations with a population equivalent < 5000; (2), applies to wastewater discharged to treatment plants for agglomerations with a population equivalent ≥ 5000.

In accordance with Polish and EU regulations [8], wastewater with such a high concentration of nutrients cannot be discharged into natural waters or the ground untreated. The requirements for the quality of wastewater generated during the production of tomatoes are presented in Table 1, which highlights the challenges faced by designers who decide about the choice of facilities used for their treatment.

Considering the current scale of the phenomenon and the expected intensive development of soilless forms of plant cultivation, including vertical farms in urbanized areas, there is a need to find a simple, easy-to-use, energy efficient [9], and at the same, highly effective technological solution for wastewater treatment. However, it is not possible to apply processes with heterotrophic microorganisms [3]. Wastewaters from soilless plant cultivation farms have limited carbon source to be utilized as electron donors for heterotrophic denitrification. In this case, an abundant carbon source or appropriate C:N ratio has been proven to be a key factor for achieving efficient removal of nitrate [10]. Biological, physical, and chemical methods have been applied for removing biogenic compounds from this type of wastewater [5].

Previous research and papers by the authors of this article have shown that wastewater with a low COD, for example, from soilless cultivation systems, with typically high concentrations of oxidized forms of nitrogen and phosphorus, may be effectively treated in electrochemical reactors, such as a rotating electrochemical disk contactor (RECDC), and in a bioelectrochemical reactor (BER), such as a rotating electrobiological disk contactor (REBDC) [11–13].

The passage of electric current through a REBDC with an aluminum anode [14] results in water electrolysis and the generation of gaseous hydrogen which is a source of energy for autotrophic bacteria that conduct hydrogenotrophic denitrification. These processes are accompanied by heterotrophic denitrification with organic matter present in wastewater. Carbon dioxide formed in this reaction serves as an additional source of inorganic carbon for autotrophic bacteria [15]. The aluminum anode also enables electrochemical reduction of nitrate, whereas phosphorus compounds are removed in the process of electrocoagulation. Nitrogen and phosphorus are also removed during the growth of biofilm biomass.

In a REBDC, some processes may be limited due to the small amount of carbon compounds in wastewater from soilless plant cultivation. The introduction of an external source of carbon may increase the contribution of the heterotrophic denitrification process in the removal of nitrate nitrogen. More intensive biomass growth also influences an increase in N and P removal. A REBDC with heterotrophic-autotrophic denitrification (HAD) may create conditions for synergistic interactions between autotrophic and heterotrophic bacteria [16]. In turn, in a rotating electrochemical disk contactor, the removal of nitrogen and phosphorus compounds is a result of electrochemical reduction of nitrate and electrocoagulation of phosphorus compounds. In a RECDC, the disks are not covered with a biofilm. During experiments, the disks are regularly cleansed to remove developing microorganisms on the biofilm.

The results of previous studies have shown that electrochemical and bioelectrochemical methods ensured a high degree of neutralization of pollutants, and were also characterized by a minimum amount of sludge as well as low investment and operating costs as compared with physicochemical processes such as ultrafiltration or ion exchange [17–19]. At the same time, they are characterized by ease of use as well as technological and technical reliability, which is important in agricultural facilities often located far from towns with a sewerage system and without access to technologically qualified staff [20].

The aim of this study was to determine the technological parameters of a RECDC and a REBDC, i.e., electric current density (J) and hydraulic retention time (HRT), depending on the quality requirements set for treated wastewater discharged to the environment (according to sewage effluent disposal consent) or to a municipal sewage system (according to owner's requirements). The presented graphs show phosphorus and nitrogen concentrations in treated wastewater and nutrient load removed in a RECDC and a REBDC depending on current density and HRT, which would be useful for technologists and designers. In turn, the delivered tables present the values of other pollution indicators for wastewater treated in both reactors.

Finally, we propose technological schemes for wastewater treatment plants with an electrochemical or electrobiological contactor developed based on the results of this study.

2. Materials and Methods

This study was conducted with four one-stage RECDCs and four one-stage REBDCs, at four hydraulic retention times, i.e., 4, 8, 12, and 24 h. The densities of electric current used for each hydraulic retention time were: 0.63, 1.25, 2.50, 5.00, and 10.00 A/m^2 [10,11]. Over the experimental period, the temperature at the laboratory was about 20.0 ± 1 °C. Wastewater was fed to the reactors with Minipuls 3 peristaltic pumps (Gilson, Middleton, WI, USA). An aluminum anode was mounted in the flow tank of each reactor. Disks made of stainless steel served as cathodes and both electrodes were connected to a laboratory power supply (HANTEK PPS2116A, Zunhua, China), which was a source of direct electric current for maintaining its desired intensity (Figure 1) [11].

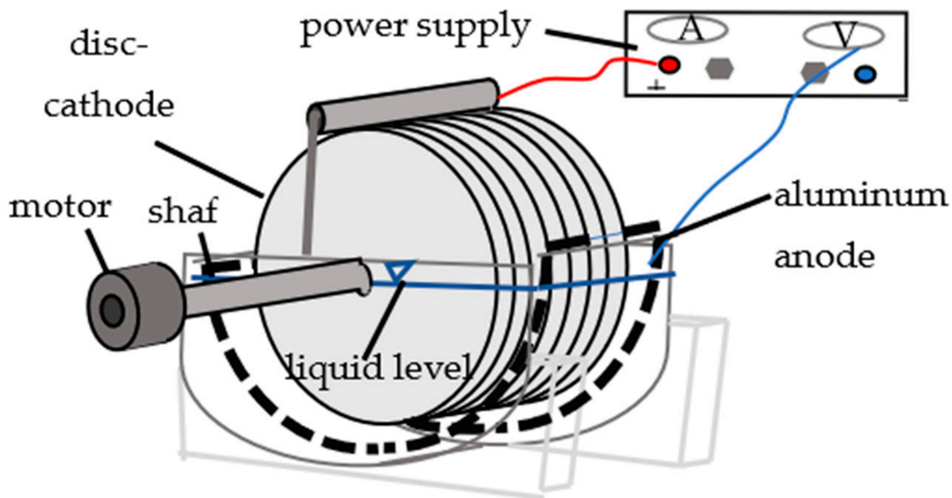

Figure 1. Scheme of the unit experimental model.

The synthetic wastewater used in this study had the composition characteristics of wastewater from soilless cultivation of tomatoes, and was adopted after Saxena and Bassi [4] as well as Mielcarek et al. [5]. To provide appropriate conditions for biofilm development, sodium acetate as a source of carbon was added to the wastewater treated in the REBDC. This produced a C:N ratio of 0.5, which ensured the growth of biofilm biomass and, simultaneously, allowed for autotrophic denitrification. The composition of the synthetic raw wastewater used in this study is presented in Table 2 [11,12].

Table 2. The composition of the synthetic wastewater.

Fertilizers	Sample Weight
Calcium nitrate	2.06 g/L
Potassium nitrate	0.89 g/L
Potassium monophosphate	0.353 g/L
Potassium sulphate	0.395 g/L
Magnesium sulfate	1.08 g/L
Iron chelate	0.023 g/L
Microelements	0.2 cm^3/L

The analytic control of processes began after adaptation of the reactors. Samples were collected for analyses in 24-hour intervals. Physicochemical analyses of 20 samples were conducted at each of the electric current densities and HRTs applied in both reactors.

The influent and the effluent were analyzed using the following indicators:

1. Organic matter expressed as the COD value, following the bichromate method;
2. Total nitrogen, using a total organic carbon analyzer TOC-L CPH/CPN with a TNM-L device (Shimadzu Corporation, Kyoto, Japan) and the method of oxidative combustion and chemiluminescence;
3. Ammonia nitrogen, nitrate nitrogen, and nitrite nitrogen, using a VWR UV-3100PC spectrophotometer (VWR International, Shanghai, China), with a colorimetric method;
4. Total phosphorus (±0.01 mgP/L), using a UV-Vis 5000 DR spectrophotometer (HACH Lange, Duesseldorf, Germany), with an HACH Lange LCK 348–350 method;
5. pH value (±0.01 pH) and temperature (±1 °C), using a CP-105 pH meter (Elmetron, Zabrze, Poland); redox potential (±1 mV), with a pH 211 m (Hanna Instruments, Eibar, Spain); and electrolytic conductivity (±0.01 mS/cm), using an HQ 440d multimeter (Hach Company, Loveland, CO, USA).

3. Results and Discussion

3.1. EU and Polish Regulations Regarding Industrial Wastewater Discharged into Sewage Systems, Natural Waters, or the Ground

The study results were presented in compliance with the principles of data presentation in scientific and technical literature, including parameters and correlations computed for rotating biological contactors (RBCs) [21]. The figures presented in this section depict, firstly, the correlations observed for the electrochemical contactor (RECDC), and then for the electrobiological contractor (REBDC) with respect to the nutrient load removed. Similarly, the figures show changes in the pollutant concentration in the effluent (total phosphorus, total nitrogen, and nitrates) upon the use of various electric current densities and hydraulic retention times. In the case of wastewater discharged from soilless cultivation installations to a municipal sewage system (Table 1), the permissible levels of pollutants were established based on the quantity and quality balance of municipal wastewater inflowing to a municipal wastewater treatment plant, the current capacity of the wastewater treatment plant, and the achieved efficiency of pollutant removal [7]. In addition, the sewage system operator (water and sewage company) required the industrial wastewater supplier (in this case, the supplier of soilless tomato cultivation wastewater) to use the best available technique (BAT) for its treatment that would ensure its discharge into the sewage network. The RECDC and the REBDC can be expected to meet the BAT requirements.

3.2. Quality of Wastewater Treated in the RECDC and in the REBDC

Based on the permissible wastewater flow and concentrations of nitrite nitrogen and total phosphorus in the wastewater from the wastewater network imposed by the water and sewage company, it is necessary to define the technological parameters of the RECDC and the REBDC that treat the wastewater before it is discharged into the sewage system. This can be achieved based on the correlations presented in Figures 2–5 and the data provided in Table 3. Therefore, it is possible to determine the operating conditions of the rotating electrochemical and electrobiological disk contactors, i.e., to select the electric current density and duration of the hydraulic retention time that will ensure the removal of the required phosphorus and nitrogen load in the RECDC (Figure 2) and the REBDC (Figure 3). In turn, based on the correlations depicted in Figures 4 and 5, it is possible to determine the phosphorus and nitrogen concentrations in the effluent achieved with the parameters established using Figures 2 and 3.

3.3. Removal Rate of Phosphorus and Nitrogen Loads per Time Unit

The study results demonstrated that the removal rate of phosphorus and nitrogen loads per time unit in the RECDC was affected primarily by HRT.

The highest daily phosphorus load removed was achieved at the shortest HRT tested (HRT = 4 h), whereas the lowest daily phosphorus load removed was achieved at HRT = 24 h (Figure 2a). The HRT extension contributed to a decrease in the daily load removed. In turn, an increase in electric current density led to an increase in pollutant load removed at all HRTs tested. In the case of HRT = 4 h, current density increases from 0.63 to 10.0 A/m^2 caused an over 20% increase in the phosphorus load removal rate, i.e., from 30.14 to 36.91 g P/m^2·d. The HRT extension to 24 h caused a significant decrease in the daily phosphorus removal performance to values of 5.46 and 5.94 g P/m^2·d, respectively. At HRTs of 8, 12, and 24 h, the changes in electric current densities had a negligible effect (below 5%) on the increase in the daily load removal rate (Figure 2a).

As in the case of phosphorus, the HRT was the main driver of the daily nitrogen load removal (Figure 2b). The highest daily removed load was noted at HRT = 4 h, and the lowest daily removed load was at HRT = 24 h. In the case of HRT = 4 h, an electric current increase from 0.63 to 10.0 A/m^2 doubled the daily removed load from 0.95 to 2.03 g N/m^2·d, while at HRT = 24 h, the respective values ranged from 0.36 to 0.87 g N/m^2·d. At the remaining HRTs tested, the increases were more noticeable than at HRT = 4 h. At the same time, the removal rates noted were substantially lower than those determined for phosphorus (from 5.46 to 36.91 g P/m^2·d). It should be remembered, however, that, in the electrochemical contactor, phosphorus compounds were removed in the process of highly effective electrocoagulation, whereas nitrogen compounds were removed during less effective electrochemical reduction of nitrates [22,23].

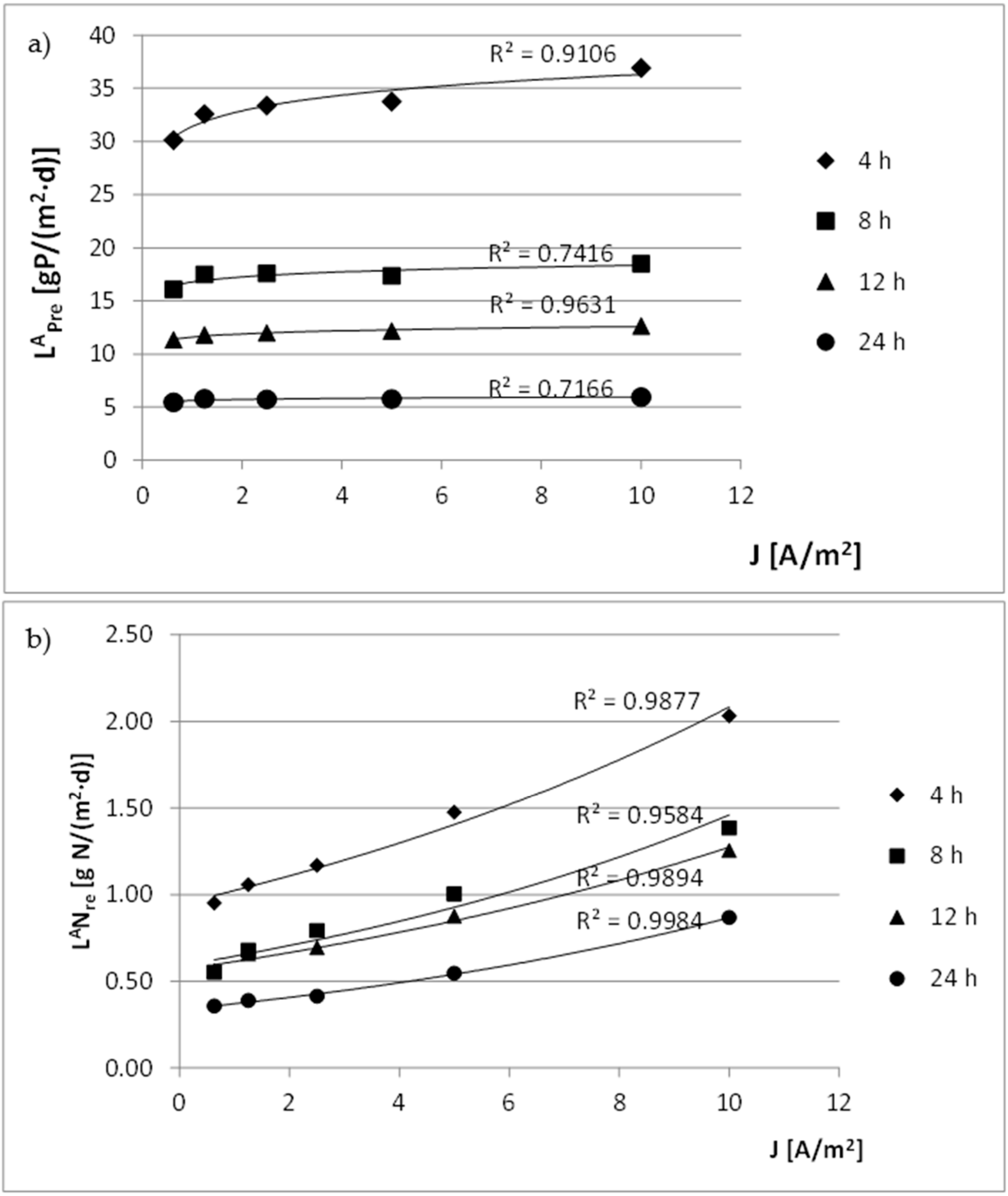

Figure 2. The influence of current density and HRT on the removed load (L^A) per day in the electrochemical rotating disc contactor (RECDC): (**a**) Phosphorus (L^AP_{re}); (**b**) total nitrogen (L^AN_{re}).

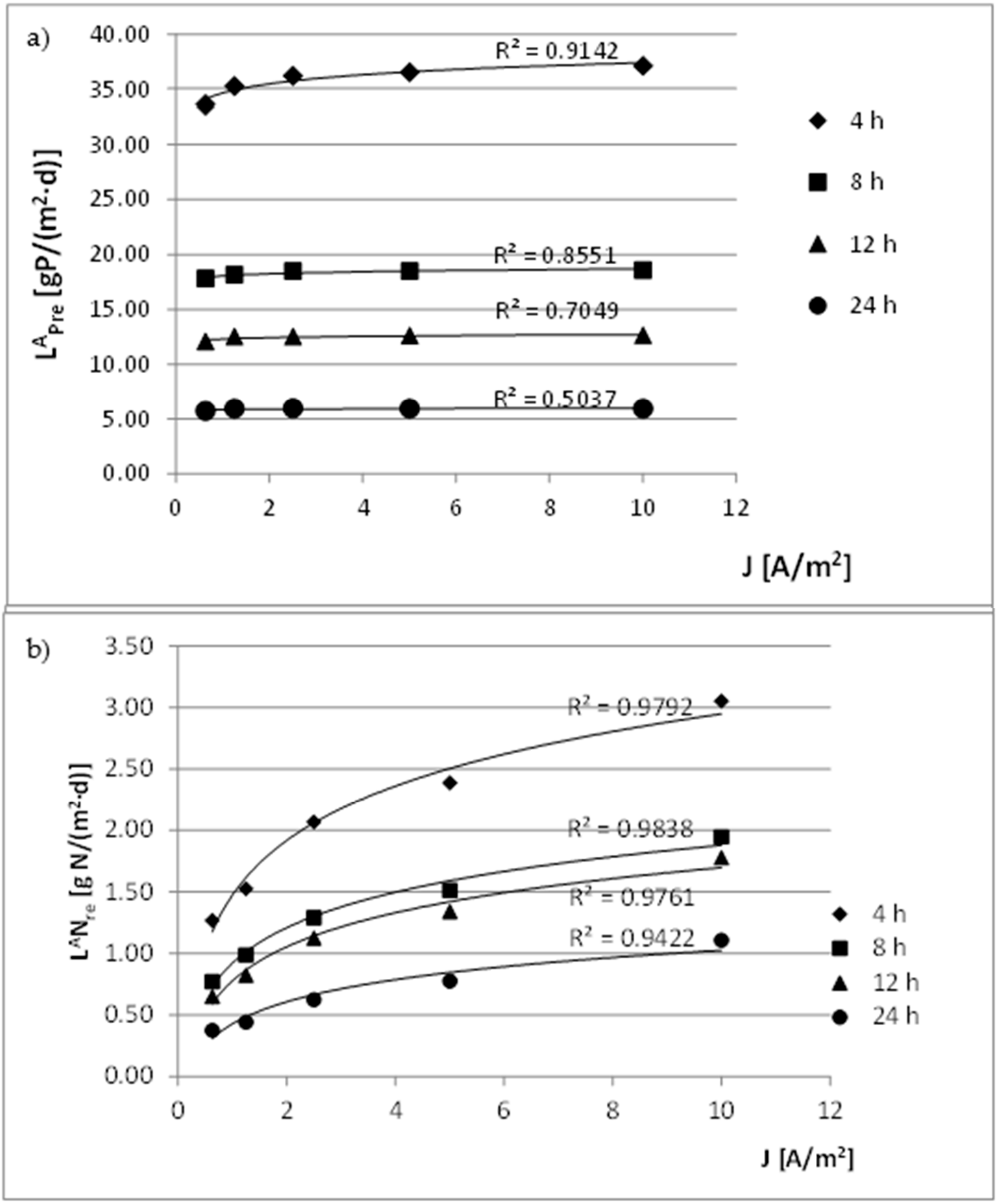

Figure 3. The influence of current density and HRT on the removed load per day (L^A) in the electrobiological rotating disc contactor (REBDC): (**a**) Phosphorus (L^AP_{re}); (**b**) total nitrogen (L^AN_{re}).

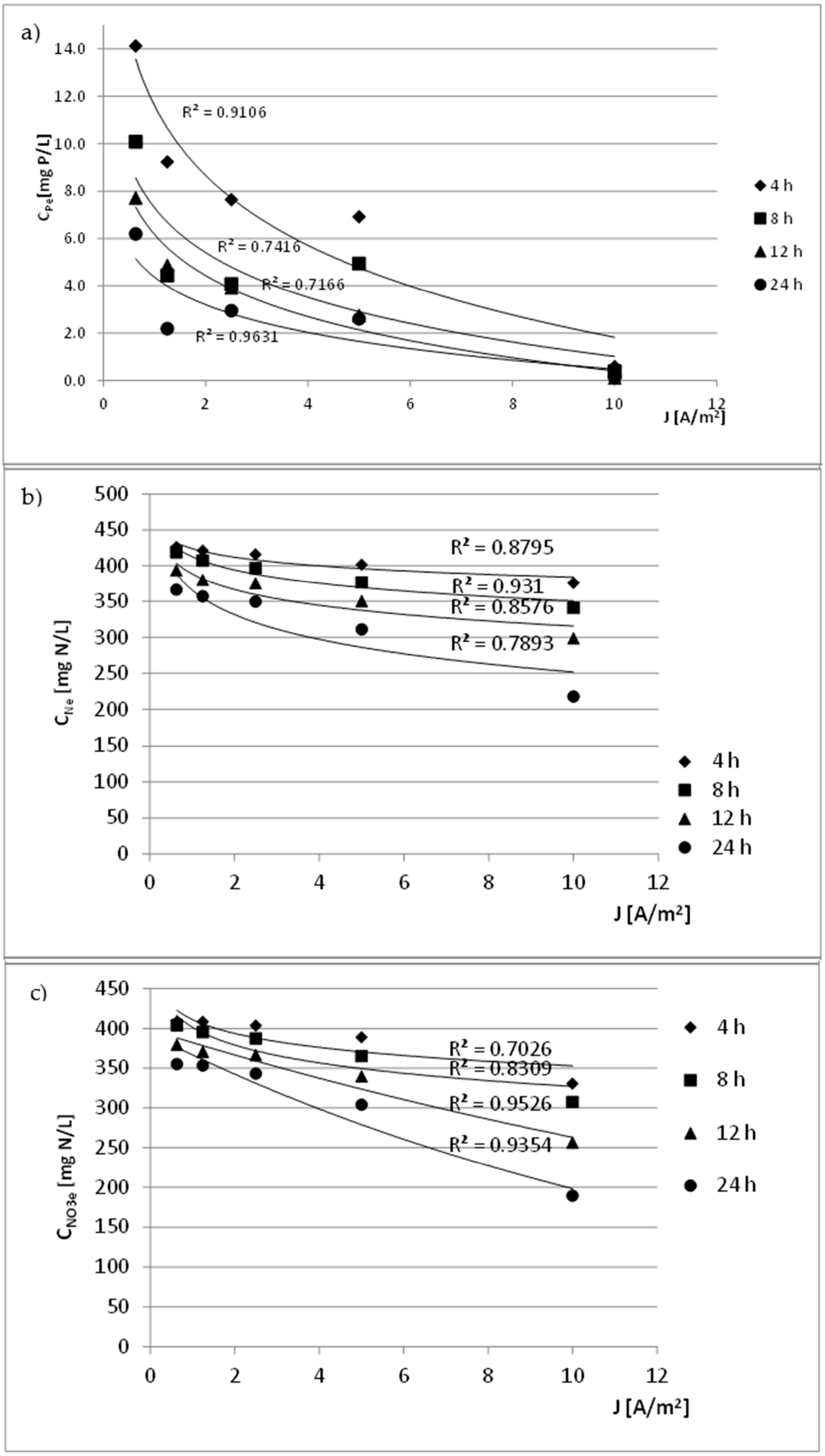

Figure 4. The influence of current density and HRT on the concentration of pollutants in the treated wastewater in the electrochemical rotating disc contactor (RECDC): (**a**) Phosphorus (C_{Pe}); (**b**) total nitrogen (C_{Ne}); (**c**) nitrates (C_{NO3e}).

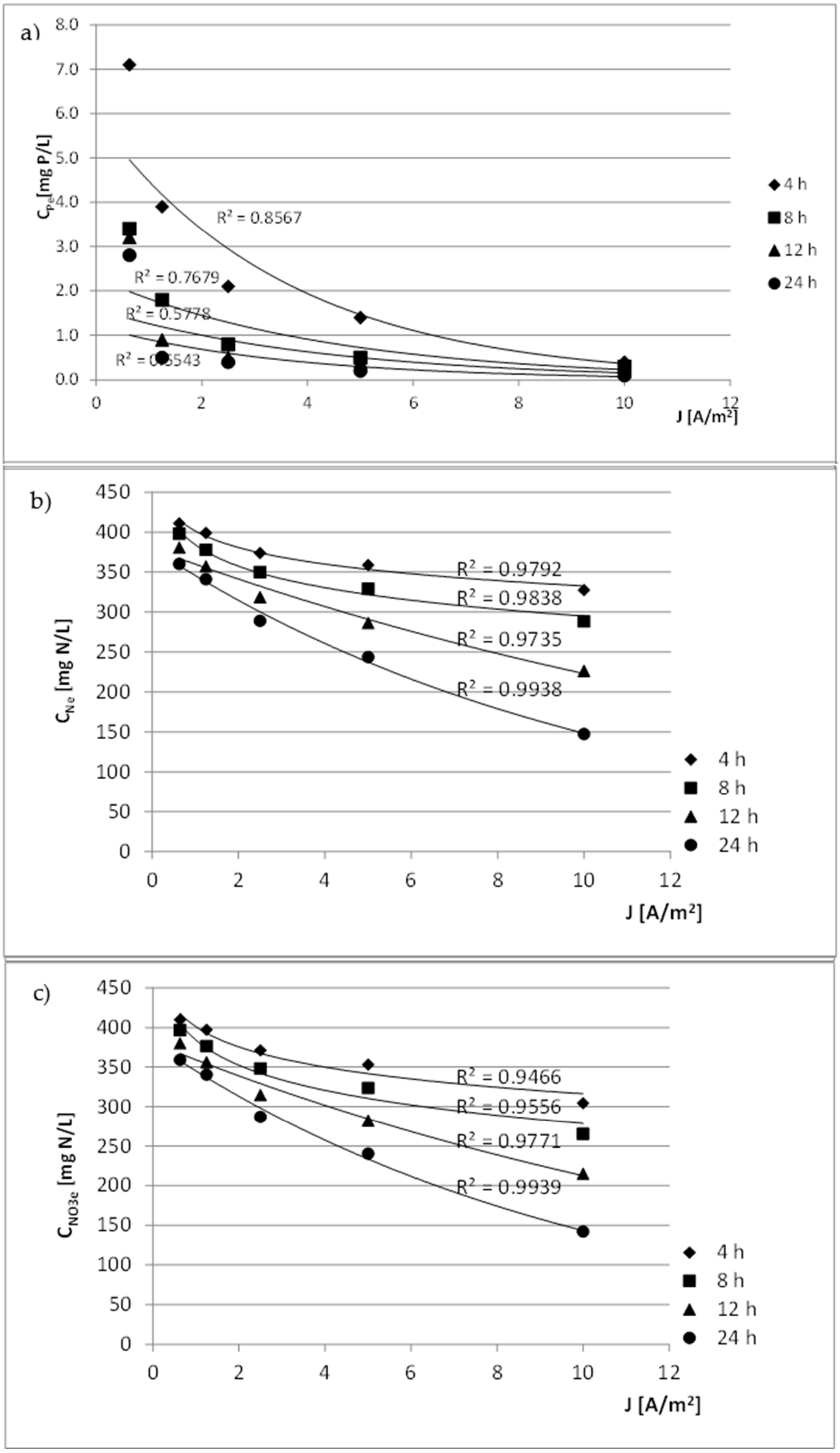

Figure 5. The influence of current density and HRT on the concentration of pollutants in the treated wastewater in the electrobiological rotating disc contactor (REBDC): (**a**) Phosphorus (C_{Pe}); (**b**) total nitrogen (C_{Ne}); (**c**) nitrates (C_{NO3e}).

Table 3. Quality of wastewater treated in the RECDC and in the REBDC (mean values).

Parameter	Synthetic Wastewater Inflow	HRT	RECDC					REBDC				
			Electric Current Density [A/m^2]					Electric Current Density [A/m^2]				
			0.63	**1.25**	**2.50**	**5.00**	**10.00**	**0.63**	**1.25**	**2.50**	**5.00**	**10.00**
COD [mg O_2/L]	260 * 45 **	4 h	28.6	27.8	27.2	25.8	22.0	30.3	34.6	30.0	41.8	56.3
		8 h	25.7	25.3	24.4	23.0	21.0	27.6	33.9	27.5	27.3	55.6
		12 h	22.4	21.9	21.0	20.1	19.3	26.1	26.3	27.3	24.8	32.7
		24 h	18.2	18.7	17.1	16.0	14.5	19.7	21.7	24.5	24.3	28.2
Total nitrogen [mg N/L]	470	4 h	426.0	421.0	415.9	401.5	375.6	411.1	399.2	373.8	358.9	327.8
		8 h	418.9	407.1	396.5	376.6	341.0	398.3	378.0	350.0	329.3	288.6
		12 h	393.4	380.1	375.3	350.3	298.5	380.7	357.4	316.0	286.6	226.6
		24 h	366.4	357.1	349.7	311.2	217.7	360.3	341.3	288.3	243.8	147.7
Nitrate [mg N/L]	453	4 h	408.4	408.2	403.5	389.0	330.8	409.9	397.1	371.3	353.2	304.7
		8 h	403.9	395.4	387.2	365.0	307.6	396.9	376.6	348.3	323.5	265.3
		12 h	379.4	370.5	366.5	339.8	256.6	380.0	356.2	314.6	282.5	215.1
		24 h	355.4	353.5	343.3	304.3	189.4	359.2	340.6	287.2	240.6	142.2
Nitrite [mg N/L]	0.015	4 h	0.4	0.2	0.1	0.1	30.8	1.0	1.6	2.2	5.2	21.5
		8 h	1.2	0.3	0.1	0.1	23.0	0.8	0.8	1.1	4.7	20.9
		12 h	1.9	0.4	0.1	0.1	35.5	0.3	0.6	0.9	3.0	10.4
		24 h	6.1	1.1	0.1	0.1	27.9	0.2	0.2	0.6	2.2	5.2
Ammonia nitrogen [mg N/L]	18.0	4 h	17.2	12.7	12.3	12.4	14.0	0.2	0.5	0.3	0.6	1.6
		8 h	13.8	11.4	9.2	11.5	10.4	0.6	0.6	0.5	1.1	2.3
		12 h	12.0	9.3	8.8	10.5	6.5	0.4	0.6	0.5	1.0	1.1
		24 h	4.9	2.6	6.4	6.8	0.4	0.9	0.5	0.5	0.9	0.3
Total phosphorus [mg P/L]	74.0	4 h	4.1	9.2	7.7	6.9	0.6	7.1	3.9	2.1	1.4	0.4
		8 h	10.1	4.5	4.1	4.9	0.4	3.4	1.8	0.8	0.5	0.3
		12 h	7.7	4.9	3.9	2.7	0.1	3.2	0.9	0.5	0.4	0.2
		24 h	6.2	2.2	2.9	2.6	0.1	2.8	0.5	0.4	0.2	0.1
Temperature [°C]	19.5	4 h	19.8	19.8	22.4	26.3	39.6	19.3	19.6	22.3	24.8	37.8
		8 h	19.8	20.0	22.5	28.6	40.0	19.5	19.6	22.5	25.7	39.3
		12 h	19.9	20.0	23.6	30.1	48.8	19.8	19.9	22.7	28.4	47.8
		24 h	20.0	20.1	24.2	34.2	50.2	19.9	20.0	23.8	33.6	50.0
pH	6.98	4 h	7.97	7.87	7.85	7.84	8.12	8.40	8.43	8.46	8.28	8.22
		8 h	8.05	7.97	7.97	7.90	8.57	8.48	8.41	8.55	8.25	8.85
		12 h	8.06	8.05	7.93	7.90	8.75	8.58	8.50	8.54	8.35	8.41
		24 h	8.07	8.05	8.07	8.06	9.25	8.42	8.06	8.38	8.75	8.75
Iron [mg/L]	-	4 h	0.2	0.2	0.2	0.3	0.3	0.2	0.2	0.2	0.3	0.3
		8 h	0.2	0.2	0.2	0.3	0.3	0.2	0.2	0.2	0.3	0.3
		12 h	0.2	0.3	0.3	0.3	0.3	0.2	0.2	0.2	0.3	0.3
		24 h	0.3	0.3	0.3	0.3	0.3	0.3	0.2	0.3	0.3	0.3
Aluminium [mg/L]	-	4 h	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.1	0.1	0.2
		8 h	0.0	0.0	0.0	0.1	0.2	0.0	0.0	0.1	0.2	0.2
		12 h	0.0	0.0	0.1	0.2	0.3	0.0	0.0	0.1	0.2	0.3
		24 h	0.0	0.1	0.1	0.3	0.3	0.0	0.0	0.2	0.3	0.3

* wastewater with sodium acetate; ** wastewater without sodium acetate.

In the bioelectrochemical contactor, the daily load of removed phosphorus ($L^A P_{re}$) was higher than that in the electrochemical reactor (Figure 3a). At HRT = 4 h, it reached 33.56 and 37.08 g P/m^2·d for electric current densities of 0.63 and 10.0 A/m^2, respectively. At HRT = 24 h, the respective values were 5.72 and 5.92 g P/m^2·d, which confirmed the results obtained in the RECDC, where HRT extension led to a decrease in the daily removal rate. It was also found that electric current density increases at a given HRT contributed to the neutralization of a difference noted between both reactors in the daily amount of removed phosphorus load. These observations were consistent with the results of our previous investigation [23] and with findings reported by Lagum [24], and this agreement could be due to the fact that the electrochemical coagulation of phosphorus observed in bioelectrochemical reactors was accompanied by the removal of this element induced by biofilm biomass growth.

In the bioelectrochemical reactor (REBDC), the daily amount of the removed nitrogen load (L^AN_{re}) was higher than that in the electrochemical reactor (RECDC) (Figure 3b), because nitrates were removed in the REBDC upon hydrogenotrophic and heterotrophic denitrification as well as electrochemical reduction, while in the RECDC reactor, nitrogen was removed primarily during electrochemical reduction of nitrates [11,18,25,26]. At HRT = 4 h, the nitrogen removal performance was the highest and reached 1.27 and 3.09 g N/m^2·d at current densities of 0.63 and 10.0 A/m^2, respectively. At HRT = 24 h, the nitrogen removal performance was the lowest, reaching 0.38 and 1.11 g P/m^2·d, respectively. These results confirmed findings from the experiment with the RECDC, where the HRT extension contributed to a decrease in the daily amount of the removed nitrogen load. At higher electric current densities, the differences between both reactors in daily nitrogen load removed were aggravated. At longer hydraulic retention times, the differences between the reactors successively diminished.

3.4. Correlations between Electric Current Density and HRT and Pollutant Concentrations

Table 1 presents the highest permissible concentrations of pollutants in wastewater from soilless plant cultivation discharged into natural waters or the ground and to sewage systems. Figure 4 shows correlations between operating parameters (electric current density and HRT) and pollutant concentrations in the effluent treated in the electrochemical contactor. At all HRTs tested, an increase in electric current density resulted in decreased effluent phosphorus concentration. A similar tendency was observed while changing HRT. Its extension was found to improve the effluent's quality. The lowest phosphorus concentrations were at HRT = 24 h and ranged from 6.2 to 0.14 mg P/L at current densities increasing from 0.63 to 10.0 A/m^2. At HRT = 24 h, the pollutant concentrations in the effluent failed to meet the requirements set for the industrial wastewater discharged into natural waters or the ground (3.0 mg P/L, Table 1) only at the lowest current density tested (0.63 A/m^2). At HRT = 12 h, the concentration of phosphorus was lower than 3.0 mg P/L only at current densities of 5.0 and 10.0 A/m^2. At all HRTs and J = 10.0 A/m^2, the concentration of phosphorus in the effluent was below 1.0 mg P/L, which means it could be discharged into natural waters and the ground.

Figure 4b,c presents concentrations of total nitrogen achieved at various current densities and HRTs, and reveals similar tendencies as those observed in Figure 4a, i.e., increased current density and extended HRT decreased nitrogen concentrations in the effluent. At HRT = 24 h and J = 10.0 A/m^2, the concentration of total nitrogen was at 217.7 mg N/L, which means that the effluent treated in the electrochemical reactor could not be discharged into natural waters or the ground as it failed to meet the requirements set for industrial wastewater (Table 1).

The concentration of phosphorus in the effluent from the bioelectrochemical reactor (REBDC) was lower than in that from the electrochemical reactor (Figure 5a). At HRT = 24 h, the lowest phosphorus concentrations in the effluent from the RECDC reached 2.80 and 0.10 mg P/L at electric current densities of 0.63 and 10.0 A/m^2, respectively. At HRT = 24 h and all current densities tested, the effluent concentration met the requirements set for industrial wastewater discharged into natural waters and the ground (3.0 mg P/L). At HRT = 12 h, the phosphorus concentration exceeded 3.0 mg P/L only at J = 0.63 A/m^2. At current densities of 2.5, 5.0, and 10.0 A/m^2 and all HRTs tested, the effluent met the requirements set for industrial wastewater discharged into natural waters and the ground. At all HRTs and J = 10.0 A/m^2, the phosphorus concentration in the effluent was lower than 1.0 mg P/L, which made the effluent suitable for discharge into natural waters and the ground [8].

The concentrations of nitrogen and nitrates in the effluent discharged from the REBCD (Figure 5) were lower than the respective values achieved in the experiment with the RECDC. The lowest values were determined at HRT = 24 h and J = 10.0 A/m^2, when the concentration of total nitrogen reached 147.7 mg N/L, which means that the wastewater treated in the bioelectrochemical reactor could be discharged into natural waters and the ground as it met the requirements set for industrial wastewater (Table 1). Nevertheless, it needs to be remembered that the external carbon source (sodium acetate) used in the present study was a very low dose, to reach a C:N ratio of 0.5 and to ensure the growth of biofilm biomass, and simultaneously allow for autotrophic denitrification [10]. Increasing the carbon dose has a direct impact on the effectiveness of denitrification and nitrogen removal [27,28].

The study results presented in Figures 4 and 5 demonstrate that the lowest pollutant concentrations in the effluent were obtained in both types of reactors (electrochemical and bioelectrochemical) at longer hydraulic retention times and higher current densities.

Maintaining the appropriate HRT and J in the bioelectrochemical reactors ensures the conditions required for the development of hydrogenotrophic bacteria, such as an optimal concentration of electron donors, beneficial pH, and optimal duration of the reaction. However, an HRT that is too long leads to denitrification inhibition due to the accumulation of nitrites [29,30], whereas current density that is too high contributes to a substantially exceeded saturation constant of hydrogen and to its excess remaining in the biofilm, which adversely affects denitrification [29,31,32]. In their study, Zhang et al. [33] proved that lower current intensities were more suitable for denitrification in bioelectrochemical reactors and that high intensities impaired this process. Their observation was confirmed in the present study conducted with electrochemical and bioelectrochemical contactors (Table 3). At the highest electric current density tested, i.e., J = 10.0 A/m^2, the concentration of nitrites in the effluent from the RECDC exceeded 20.00 mg N/L regardless of HRT. Therefore, if wastewater is planned to be discharged into a municipal sewage system, the electrochemical contactor should operate at lower current densities to meet the requirements (Table 1). In turn, the concentration of ammonia nitrogen, determined at all HRTs and current densities tested, did not exceed 100 mg N/L, which made the effluent suitable to be discharged into a municipal sewage system.

Hao et al. [34] demonstrated a strong effect of HRT on nitrogen removal and nitrite nitrogen accumulation in a bioelectrochemical reactor. At short HRTs, for example, 5 h, they achieved 66.0% and 54.6% removal effectiveness of nitrates and total nitrogen, respectively. The concentrations of NO_{3^-}-N and NO_{2^-}-N in the effluent from the reactor were at 10.0 mg N/L and 3.5–4.0 mg N/L, respectively. The rate of denitrification was observed to increase along with extending hydraulic retention times and reached the maximal value at HRT = 10 h. Successive HRT extension (over 10 h) led to a decreased rate of denitrification. The longer HRTs (i.e., 10–12 h) contributed to the lesser accumulation of nitrite nitrogen in the effluent (below 1 mg N/L). When the environment is poor in organic carbon (i.e., at C:N = 1.5), the activities of both heterotrophic and autotrophic bacteria may prove to be important. The autotrophic bacteria need more time to remove nitrogen due to their relatively low growth rate. Therefore, a longer HRT can facilitate nitrogen removal by autotrophic denitrifying bacteria and result in less accumulation of nitrite nitrogen. When organic carbon concentration is relatively low, the continuous mode of reactor operation with an HRT that is too long may lead to the extension of the endogenous phase of heterotrophic bacteria respiration. This, in turn, would reduce the number of heterotrophs and, by this means, decrease denitrification effectiveness. Extension of the HRT can improve nitrogen removal performance and reduce nitrite nitrogen accumulation. Nevertheless, an excessively long HRT is not recommended due to the interaction of heterotrophic and autotrophic mechanisms of denitrification [34]. This was also the case in the REBDC, where the concentration of nitrites exceeded 10.00 mg N/L only at the highest electric current density tested and three HRT values. At the other J and HRT values studied, the quality of the effluent met the requirements set for wastewater discharged into municipal sewage systems. The requirements set for wastewater discharged into natural waters and the ground were met by the effluents from the REBDC treated at J = 0.63 A/m^2; the effluents treated at J = 1.25 A/m^2 and HRTs of 8, 12, and 24 h; and the effluents treated at J = 2.50 A/m^2 and HRTs of 12 and 24 h. In turn, the concentration of ammonia nitrogen was below 3.00 mg N/L, at all J and HRT values.

In the present study, the extension of hydraulic retention time at all electric current densities tested led to increased effectiveness of nitrogen compounds removal. The results obtained corroborated earlier findings reported by Hao et al. [34] that the longer hydraulic retention time facilitating denitrifying bacteria development contributed to the less accumulation of nitrite nitrogen (Table 1).

3.5. Proposals of Technological Systems for the Treatment of Wastewater from Soilless Tomato Cultivation

The regulations pertaining to the quality of treated wastewater discharged into the natural environment and a sewage network also included requirements related to COD levels (Table 1). If the effluent is to be discharged into natural waters or the ground, its COD should not exceed 125 mg O_2/L. In the present study, the COD did not exceed 60 mg O_2/L in any of the effluents. Its value was higher in the wastewater treated in the REBDC, which was due to the external carbon source fed to the reactor. In this reactor type, the COD decreased along with extended HRT, which was consistent with the literature data [35]. Because the reported concentrations of organic compounds were very low, the COD value of the effluent should not hamper its discharge to a municipal sewage system [7].

The requirements pertaining to treated wastewater also include concentrations of iron and aluminum ions. Electric dissolution of the aluminum anode and the dissolution of contactor case and disks made of steel result in the formation of aluminum and iron ions, respectively, in both the electrochemical and bioelectrochemical contactors [36]. The Fe and Al concentrations in the effluents from both contactors were lower than 0.5 mg/L (Table 3), which means that the presence of these ions would not hamper effluent discharge to both natural receivers and municipal sewage system (Table 1).

The pH values of the effluents from both reactors met the requirement of pH values falling within the range of 6.6–9.5 at all J and HRT values tested. In the case of temperature, it was higher than the permissible value of 35 °C in both contactors upon the flow of electric current with a density of J = 10.0 A/m^2.

The study results presented in this manuscript demonstrate that soilless tomato cultivation wastewater can be treated in both electrochemical and electrobiological disk contactors, and afterwards can be discharged to municipal sewage facilities. Regardless of the intended effluent receiver (natural waters, the ground, or a sewage network), the technological system of the proposed installation should also include a reservoir, which also functions as an equalizing tank (with an agitator), a pump system (delivering wastewater evenly to the contactors 24 h a day), a one-stage electrobiological disk contactor, and a secondary sedimentation tank (Figure 6). Due to the characteristics of the generated wastewater, there is no need to use devices for mechanical wastewater treatment (screens, grit chambers, and sedimentation tank). The system should be supplemented by devices for sludge collection, thickening, and dehydration. Given the low organic matter concentration in the sludge, there is no need to provide sludge stabilization devices in the system. This should be important in relation to vertical farming located in cities.

The study results presented in Figures 2 and 3 establish the operating conditions of the electrobiological disk contactor (REBDC), i.e., adjusting the J and HRT to enable achieving the earlier specified performance of the removal of the required pollutant load. In turn, correlations shown in Figures 4 and 5 establish the technological parameters of the REBDC depending on the required quality of the effluent, including concentrations of phosphorus, nitrogen, and nitrates. The remaining data required for signing a contract with a network operator regarding effluent discharge into a sewage system can be determined from the data presented in Table 3.

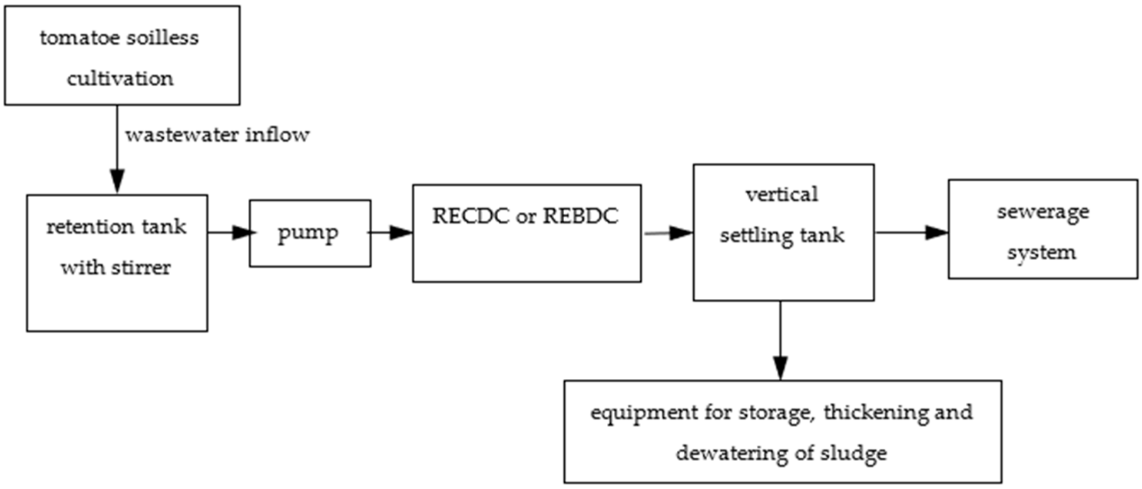

Figure 6. Scheme of a technological system with electrochemical or electrobiological rotating disc contactors for treating wastewater discharged to sewage systems.

In compliance with the EU and Polish legal regulations [37], the concentrations of nitrogen and nitrates in treated wastewater from soilless tomato cultivation discharged to the natural environment should not exceed 30 mg N/ L. The present study demonstrated that, in the case of the REBDC, it was feasible to obtain 150 mg N/L nitrate nitrogen concentration in the effluent at the highest current density, i.e., J = 10.0 A/m^2, and the longest HRT = 24 h tested. The respective value recorded in the electrochemical reactor reached 210 mg N/L, which meant that the RECDC was not a viable solution to obtain effluent suitable to be discharged into the natural environment.

In turn, the discharge of soilless tomato cultivation wastewater treated in the REBDC directly into natural waters or the ground would be feasible upon the use of a solution enabling nitrate nitrogen concentration reduction below 30 mg N/L. While searching for a solution involving the use of the electrobiological disk contactor, an assumption should be made that, at HRT = 4 h and all electric current densities tested, the concentration of nitrite nitrogen in the effluent will exceed the permissible level (1.0 mg N/L). Due to the possibility of nitrite nitrogen presence in the effluent, wastewater cannot be treated at electric current densities of J = 5.0 A/m^2 and J = 10.0 A/m^2. At J = 2.5 A/m^2, the HRT cannot be shorter than 12 h, whereas at J = 0.63 A/m^2 and J = 1.25 A/m^2, the HRT cannot be shorter than 8 h. To ensure the permissible nitrate nitrogen concentration in the effluent, the selected treatment technology should envisage the supply of an external carbon source to the disk contactor, providing carbon concentrations exceeding C:N = 0.5 (used in the present study). These carbon sources may include commercial chemical agents, for example, acetic, propionic, or citric acids. Our previous investigations have shown the feasibility of affecting nitrate concentration in the effluent from the biofilm reactors by adjusting the type and dose of the organic substrate [16,27,38]. In that instance, the technological system of the wastewater treatment plant should additionally provide tanks for storing acids, solution tanks, and a feeding system (Figure 7).

A more viable solution considering the environmental protection perspective and the idea of a circular economy would be to support the technological system with an installation for the production of volatile fatty acids (VFAs) from tomato stem and leaf biomass (Figure 8). It would be necessary to include devices for grinding and homogenizing plant residues and a reactor for prepared biomass fermentation. The system should include a tank for storing the digestate containing VFAs and a feeding installation. In both cases, higher suspended solid loads in the effluent and a much greater proportion of organic matter in the sludge must be taken into consideration. Therefore, apart from sludge dehydrating devices, additional devices for its stabilization should be designed. An alternative to sludge management may be its disposal to a municipal wastewater treatment plant. This would, however, require constructing a sludge retention reservoir or a thickener. The thickener option would result in a lower frequency of sludge disposal and, consequently, lower transport costs.

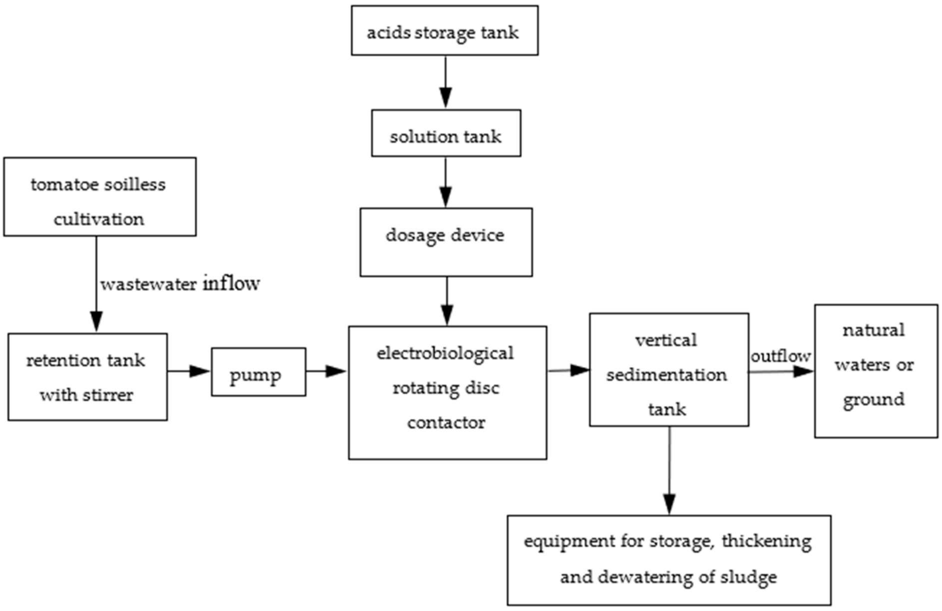

Figure 7. Scheme of a technological system with an electrobiological rotating disc contactor with external carbon source for treating wastewater discharged into natural waters or the ground.

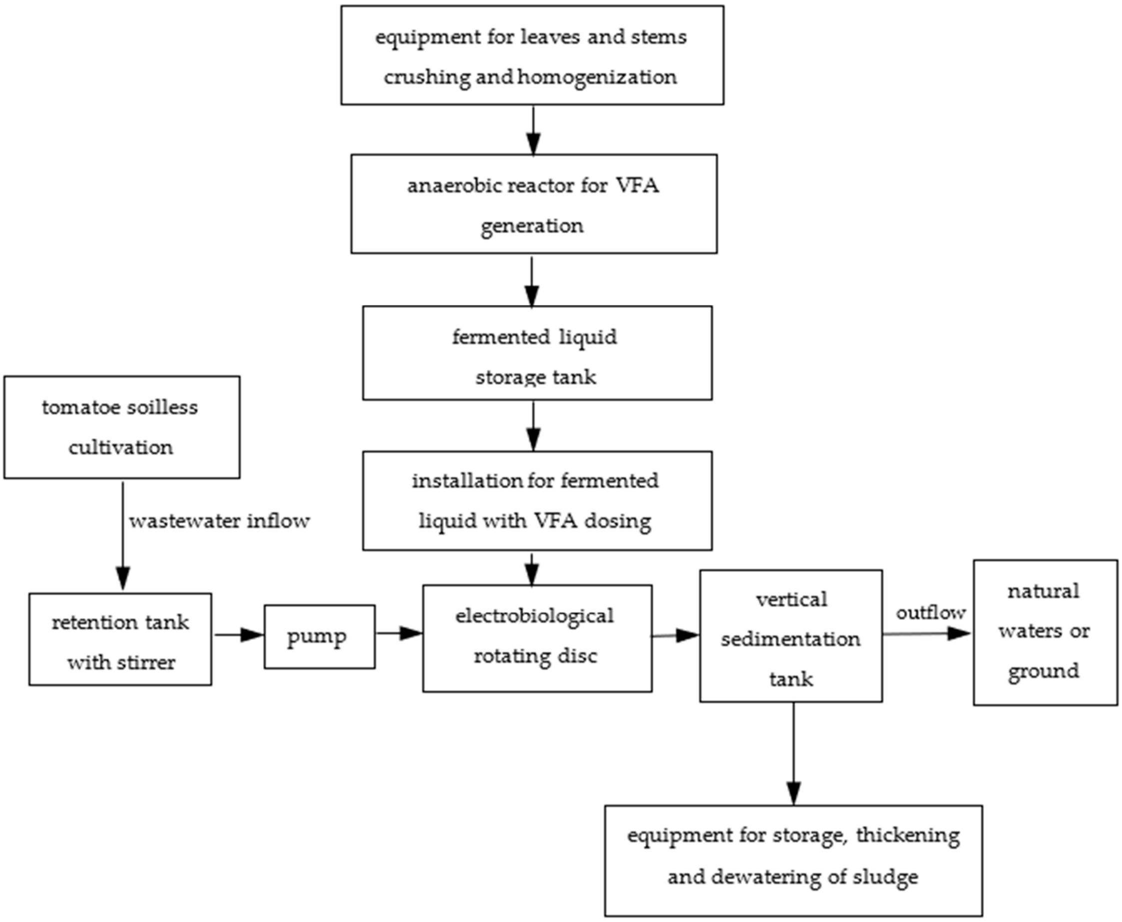

Figure 8. Scheme of a technological system with an electrobiological rotating disc contactor with equipment for VFA generation from leaves and stems crushing, for treating wastewater discharged into natural waters or the ground.

The discharge of treated soilless tomato cultivation wastewater from a REBDC into natural waters or the ground would be feasible only upon the application of technological treatments ensuring nitrate nitrogen concentration reduction below 30 mg N/L. In order to ensure nitrate concentrations in the effluent at an acceptable level, an external carbon source should be supplied to the REBDC in a technological system for a wastewater treatment plant. Nevertheless, the solution, assuming an additional carbon source fed to the REBDC to be used by heterotrophic denitrifying bacteria, requires further in-depth technological research in laboratory conditions or on-site during the start-up of an installation.

4. Conclusions

The results presented in this manuscript demonstrate that soilless tomato cultivation wastewater can be treated in both electrochemical and electrobiological disk contactors, and then discharged into a sewage system.

The effectiveness of removal of nitrogen and phosphorus compounds in a RECDC depended on the electric current density and hydraulic retention time. Phosphorus removal performance exceeded 80% regardless of current density and HRT. The highest effectivenesses of denitrification (53.4%) and dephosphatation (99.8%) were achieved at $J = 10.0$ A/m^2 and HRT = 24 h.

The study results showed over 90% dephosphatation effectiveness in the REBDC at all electric current densities and HRTs tested. At HRT = 24 h and all current densities, the concentrations of pollutants in the effluent met the requirements set for industrial wastewater discharged into naturals waters and the ground (3.0 mg P/L).

The discharge of treated soilless tomato cultivation wastewater from a REBDC into natural waters or the ground would be feasible only upon the application of technological treatments ensuring nitrate nitrogen concentration reduction below 30 mg N/L. While basing the operation of the technological system on a REBDC, caution should be exercised regarding the limitations established during the study and related to the quality of the effluent, namely to the permissible concentrations of nitrites (below 1.0 mg N/L) and ammonia nitrogen (below 10.0 mg N/L), and the effluent temperature (below 35 °C). For this reason, the best technological solution will be $J = 2.5$ A/m^2 and HRT = 24 h. These parameters resulted in achieving a high effectiveness of nitrogen compound removal; low concentrations of phosphorus compounds, ammonia nitrogen, and nitrate in the effluent; and additionally, did not lead to an effluent temperature increase above permissible values.

In the manuscript, we formulate technological assumptions for the treatment of wastewater from soilless tomato cultivation. In the case of discharging the effluent into a sewage system, the technological system should consist of a retention tank equipped with an agitator, a pumping system, a one-stage electrobiological disk contactor, and a secondary sedimentation tank. The system can be supported with devices for the collection, thickening, and dewatering of sludge.

If the effluent is to be discharged into natural waters or the ground, a solution should be designed in the technological system of the treatment plant to reduce the concentration of nitrate nitrogen to a level below 30 mg N/L, which is possible by introducing an external carbon source (e.g., VFAs and methanol) in the REBDC. The facility should include acid storage tanks, solution tanks, and a feeding system. An alternative solution could be the production of volatile fatty acids from the biomass of tomato stems and leaves. The system should be extended with devices for sludge stabilization.

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