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Article

Removal of N and P in a Rotating Biological Contactor Plant: Case Study Agnita, Romania

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Abstract: The wastewater treatment plant of Agnita, Romania was designed with a rotational biological contactor system for a population of approximately 9500, but for environmental protection reasons it must comply with regulations concerning nitrogen and phosphorus designed for larger communities. In order to achieve the prescribed limits for these pollutants, we have used a 40% $FeCl_3$ solution, continuously added to the distributor, without changes in flow or equipment. Its use boosts the removal of ammonia nitrogen, and phosphorus, bringing them within the limits and with reasonable cost. To determine the ferric chloride to be used we considered, aside from the pollutant load, the water temperature, and introduced a new parameter: specific removal power that enabled us to optimize the volume of $FeCl_3$. A major contribution to nitrogen removal was achieved by the denitrification bacteria favored by the presence of ferric ions, which also precipitate phosphorus. The results of this study, performed since September 2021, enable us to continue to use this method and enlarge its application to other plants owned by the local operator.

Keywords: RBC; ammonia; nitrification; denitrification; continuous dripping; ferric chloride; specific removal power

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1. Introduction

To demonstrate the importance of clean water in the 21st century, for mankind and nature, is almost a tautology due to mankind's awareness and the huge number of norms, regulations, and papers on this theme [1], but efforts are still needed to keep waters clean. Regulators all over the world have issued norms and standards for water quality, and in Europe they are grouped under the umbrella of Council Directive 91/271/EEC [2] with its multiple amendments; in Romania it is implemented by the new Law of the Environment [3], also with multiple amendments and subsequent norms for wastewater treatment [4,5], which include permitted concentrations when using wastewater treatment plants (WWTPs) [6], as well as the disposal of the resulting sludge [7].

There are multiple ways of treating wastewater [1]. The core of each plant is the biological reactor, which mainly converts organic pollutants into harmless compounds, usually found in sewage sludge [8]. Each approach has its advantages and disadvantages in terms of cost and efficiency [9,10].

Among the possibilities for treating wastewater, rotating biological contactors (RBCs) are used mainly for communities with population equivalents (PEs) ranging from 100 to 10,000, but they can also be used for treating several million liters per day [11,12].

For communities larger than 1000 inhabitants modular RBCs are used [13], which consist of several modules of biodiscs that usually work in parallel.

The efficiency of primary pollutant removal, expressed as biological oxygen demand (BOD), chemical oxygen demand (COD), and total suspended solids (TSS), depends on the ability to provide oxygen to the active microorganisms (aeration). In RBCs, the oxygen transfer is achieved using cyclical air exposure and renewal of the air–water interfaces (biofilm), as well as by water trickling back into the tank [14].

Disc rotation mixes the liquid and in addition to oxygen provides the nutrients for the development of the microorganisms that form the biofilm as well as breaking the biofilm due to friction, thus resulting in sludge [15].

The advantages of RBCs compared to activated sludge (aeration) equipment are: small space requirement, simple monitoring and process control, low operational and maintenance costs, high biomass concentration, low excess sludge production, short hydraulic retention time, high oxygen transfer efficiency, no need for sludge recirculation, resistance to shock and toxic loads, and compact design [16].

Usually, RBCs are used in communities with less than 10,000 PE, so, at least in Europe, these WWTPs are not required to comply with regulations regarding the maximum permitted values for nitrogen and phosphorus. The removal of these two pollutants can easily be achieved in conventional activated sludge plants, where anoxic zones can be designed and operated [17], but in typical RBCs, achieving this is more complicated, although on the same disc nitrifying (aerobic) Betaproteobacteria *Nitrosomonas* and *Nitrospira* [18] can coexist with anaerobic ones at the contact zone with the water, closer to the disc surface [19,20]. Regarding the populations of anaerobic microorganisms, the literature presents many species and genera, depending on the type of wastewater and environmental conditions [21]: *Comamonas denitrificans* [22,23], *Acinetobacter* sp., *Bacillus* sp., *Diaphorobacter* sp., *Pseudomonas* sp., *Rhodococcus* sp., *Zobellella* sp. [24], *Tetrasphaera* (which besides denitrification is also a good phosphate remover) [25], *Riemeralla* sp., *Parabacterioides* sp., *Candidatus*, *Accumulibacter phosphatis* [26], *Thauera* sp., and *Paracoccus* sp. [27].

Among the best technologies used for nitrogen removal are: moving bed biofilm reactors (MBBRs), moving bed biofilm reactor-membrane bioreactors (MBBR-MBRs), moving bed membrane bioreactors (MB-MBRs) and the integrated fixed-film activated sludge (IFAS) process [28–31]. Recently, a novel technology—anaerobic membrane bioreactors (AnMBRs)—has gained momentum, with good results in nitrogen removal [32,33].

For the improvement of N removal in RBCs, special techniques and equipment were conceived, i.e., submerged discs [34,35], transformation into an electrochemical reactor with heterotrophic–autotrophic denitrification [36], Donnan dialysis [37], or the use of certain microorganisms, such as the mixotrophic *Paracoccus denitrificans* that can undertake simultaneous aerobic carbon oxidation, nitrification, and denitrification [38], or other autotrophic microorganisms [16]. Good results in denitrification within RBCs have been demonstrated by processes like anaerobic ammonium oxidation (AnAmmOx) coupled with nitrite reduction and denitrifying anaerobic methane oxidation (DAMO) [17].

Regardless of the biological reactor type, a WWTP is a "captive customer" having almost no means to control the influent, which continuously changes day by day and even hour by hour. On the other hand, strictly regulated effluent parameters require a thorough operation of the plant, with reduced costs. Thus, the operators are supposed to maintain the plant in continuous function and, if possible, to improve its economic and environmental efficiency, and this is what we have tried at Agnita.

2. Materials and Methods

The town of Agnita is situated in the middle of Romania (Sibiu County), on the banks of the Hartibaciu River, at an altitude of 490 m (GPS coordinates 45°58′23″ N, 24°37′2″ E), with a population of 8732 inhabitants according to the 2011 census, a decrease from the 2002 census value of 10,894.

For environmental protection, and in accordance with European and Romanian laws, in 2010 the design and construction of a new WWTP started on behalf of the local operator, *Apa Tarnavei Mari* S.A. (English: Water of Tarnava Mare River Co.) in Medias. The plant was planned for 9500 PE, on the right bank of the Hartibaciu, at the southwestern town limits, and was commissioned in June 2014. From the very beginning, the WWTP was equipped with a supervisory control and data acquisition (SCADA) system that monitors the main parameters and acts accordingly.

The only major non-residential wastewater generators are a company manufacturing charging cables and solenoid systems, a producer of wood parquet flooring, and the city hospital. Their average cumulative outflow is 64.8 m^3/day (2000 m^3/month), which is less than 7% of the treated wastewater (data not shown). According to the reports, only the wastewater generated by the hospital sometimes exceeds the maximal permitted limits for BOD and nitrogen, but the situation is known, and it will be remediated.

The main parameters for the design and operation of the WWTP were:

- Daily average inflow: 2607 m^3 (dry weather);
- Daily maximal inflow: 2971 m^3 (35 L/s);
- Q max during rain: 540 m^3/h (150 L/s);

For planning the multi-annual inflow, concentration and load values were considered. Their mean values [39] are shown in Table 1.

Table 1. Means of the multi-annual inflow concentration and load values.

	Inflow Concentration		Inflow Load	
BOD	220	mg/L	573	kg/day
COD	440	mg/L	1147	kg/day
TSS	256	mg/L	667	kg/day
TKN	45	mg/L	118	kg/day
Total P	9	mg/L	24	kg/day

The schematic flow of this WWTP is presented in Figure 1.

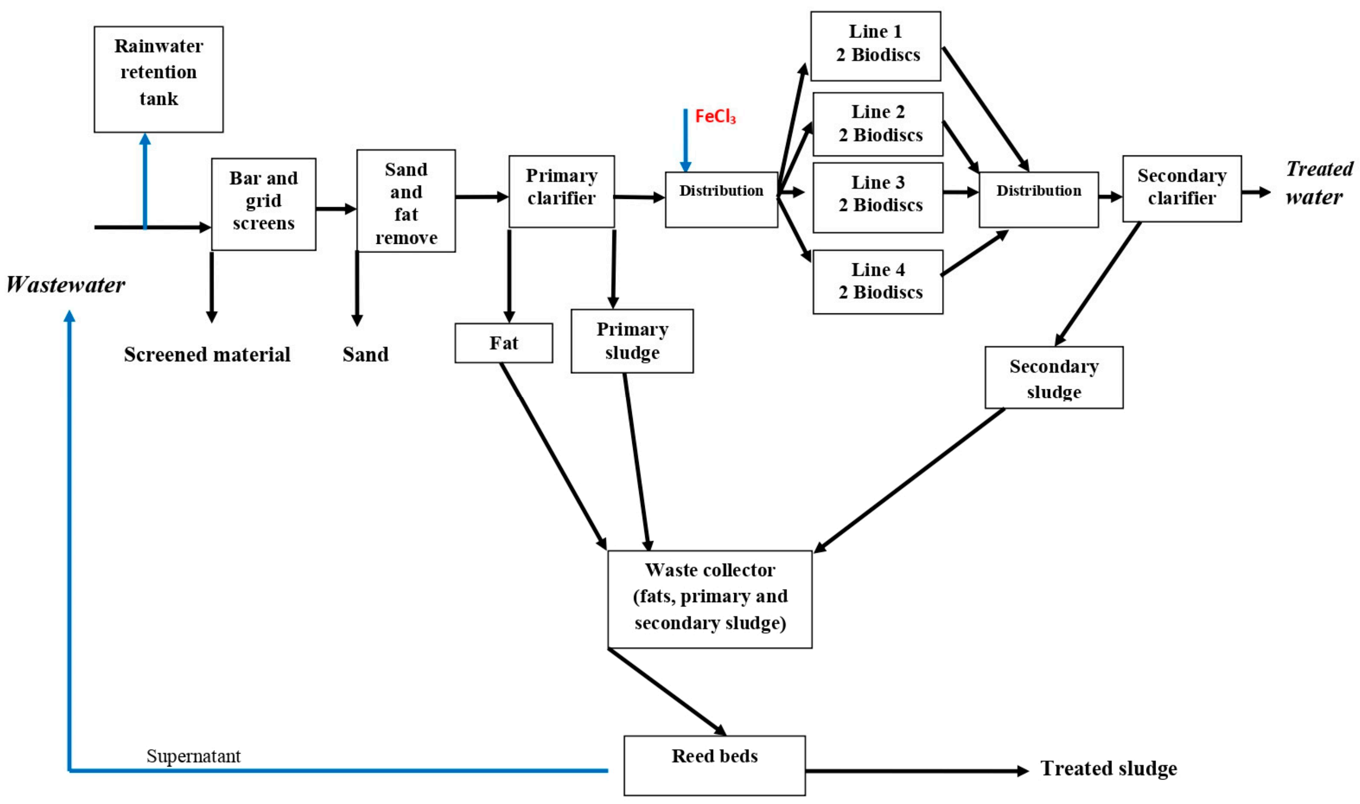


Figure 1. Flowchart of the Agnita WWTP.

Sewage water is pumped into the WWTP and then it flows gravitationally until it is discharged in the Hartibaciu River.

The screen, the sand and fat remover, and the primary clarifier act as mixers and buffers, hence the load (concentration × flow) of the water entering the RBCs is quite constant over time.

The $FeCl_3$ solution is added before distribution; it then enters the biodiscs, which have the main characteristics presented in Table 2.

Table 2. Biodisc characteristics.

Characteristic	Value	Characteristic	Value
Type of RBC	Modular	Disc diameter (mm)	3000
Number of lines	4	Disc thickness (mm)	1.5
Line volume (m^3)	58.87	Distance between discs (mm)	20
Number of modules per line	2	Disc material	HDPE
Number of biodisc sets per module	2	Rotation speed (min^{-1})	2
Number of individual discs per set	103 (each of 10 sectors)	Water retention time (hours)	2.17

The rotation speed was set after multiple tests at 2 min^{-1}, less than in other cases [40].

Considering the multi-annual data, the minimal water temperature during the plant operation was 10 °C, which during the years of operation was reached only in very few and short periods. In the RBC biofilm, the metabolic processes are less temperature-dependent than in activated sludge, one reason being the polycarbonate casing that provides some thermal insulation. The RBCs of Agnita are presented in Figure 2.

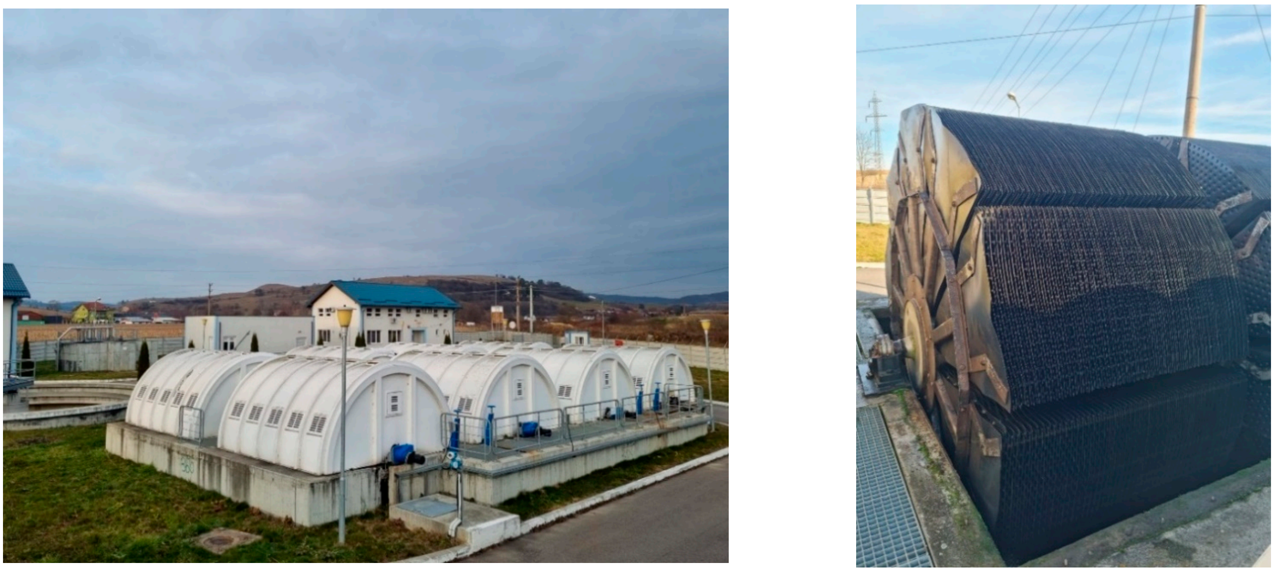


Figure 2. The RBCs of Agnita: the modules (**left**), and discs (**right**).

From the secondary clarifier, the treated water is discharged into the Hartibaciu River, while the resulting wet sludge is pumped onto the sludge-reed (genus *Phragmites*) beds, a simple and cost-effective dehydration process (air-drying) [41]. Reeds have an active growth and use the sludge nutrients and water; the resulting biomass is harvested every year and used for composting.

Analysis

The analyses performed in the WWTP are the usual ones. The standards in force according to the requirements and the authorization issued by the Olt River Catchment Administration No. 43 of 13 May 2020 (and those before it) for determination of the main parameters, the equipment, and the method as well as the maximal permitted limits (MPLs) (as daily averages) are presented in Table 3. All values are the mean of three determinations.

Table 3. Parameters and their analytical methods used in the Agnita WWTP.

Parameter	Analytical Standard or Method	Equipment	Maximal Permitted Limits mg/L
pH	SR ISO 10523/2012, PO-01	WTW pH/Conductivity Multimeter model 330i with SENTIX® 41 electrode	6.5–8.5
COD	SR ISO 6060/1996	Velp eco 16 thermoreactor Merck Spectroquant® Multy Spectrophotometer	125.0
BOD	SR EN ISO 5815-1/2020 Method WTW 997,230 OxiTop, PO-07	WTW incubator model TS 606/2-i WTW OxiTop® bottles	25.0
NH_4^+	SR ISO 7150-1/2001	WTW PhotoLab S6 Spectrophotometer	Not yet
Total N	SR EN 25,663:2000 Method WTW Ntot TC LR 251995, PO-09	WTW Thermoreactor CR 2200, Merck Spectroquant® Multy Spectrophotometer	15.0
Total P	SR EN ISO 6878/2008	WTW Thermoreactor CR 2200, Merck Spectroquant® Multy Spectrophotometer	Not yet
TSS	SR EN 872/2009	Classical filtration equipment	35.0

The morphological cluster analysis and identification of aerobic and anaerobic microorganisms were performed with an Olympus BX40 microscope (Olympus Corporation, Shinjuku City, Tokyo) with a digital camera and phase contrast, as prescribed by Eikelboom [42]. Both fresh samples and those after Gram staining were observed in a clear field, with 100× magnification [43]. Cluster dimensions and filament thickness were measured using Micro Image 4.0 software.

It is noteworthy that the Olt River Catchment Administration imposed a limit for total nitrogen, although this is not required to be determined by the laws in force; however they argued that the number of inhabitants is close to 10,000, so they applied the precautionary principles for environmental protection. Moreover, in 2022 a national census took place and there is the possibility the population could exceed 10,000 inhabitants, thus driving the town and the WWTP into another category where total phosphorous must be below 2 mg/L [44] and the minimal elimination efficiency, according to the aforementioned norm, has to be 80% for total phosphorus and 70–80% for total nitrogen.

The WWTP operators take samples every day at 8 in the morning from both the influent and the effluent and perform the required analyses, the inflow results being used for setting the operational parameters and the outflow results being reported for the previous day.

Sampling points are placed before the grids and after the secondary settler, where the water flow is measured as well. For operational reasons, there are sampling points after each piece of equipment.

In time, we observed that there are good correlations between the loads of influent ammonia nitrogen ($N\text{-}NH_4^+$), total nitrogen (total N), and total phosphorus as presented in Supplementary Material S1. Because of this observation, for operational reasons we have decided to calculate the volume of 40% $FeCl_3$ solution to be dripped during the next 24 h considering the ($N\text{-}NH_4^+$) load in the influent, as it is much simpler and cheaper.

The operator has the possibility of varying the flow of the respective peristaltic pump in case outstanding inflows are presented, exceeding the storage capacity of the primary settler, and thus increasing the flow in the RBC as well. By entering the inflow and the concentration into an Excel form, the total load (for all pollutants, total nitrogen and phosphorus included) as well as the elimination power required for their removal can be calculated.

3. Results and Discussion

The first two years (June 2014–December 2016) were dedicated to testing and fixing the WWTP, to adjust to real work conditions. After this period, noting that the total nitrogen reduction was low and the concentrations were above permitted levels, in January 2017 we decided to add 40% $FeCl_3$ solution, taking into account both the literature [45,46] and the good results we already had at the conventional (with activated sludge) WWTP in Medias [47,48] where, besides coagulation, $FeCl_3$ had been used for phosphorus removal, too. Other reasons we used $FeCl_3$ were because ferric ions, in small amounts, are beneficial for bacterial metabolism [49], as a coagulant, increase the agglomeration and deposition of sludge, and last but not least, because the resulting sewage sludge was to be landfilled on a reed bed, and we did not want the addition of Al in it.

The in- and outflow values allowed us to calculate both the pollutant removal, as the difference between the initial and final loads, and the removal efficiency (*RE*) with the classical (Equation (1)):

$$RE = \frac{Load_{in} - Load_{out}}{Load_{in}} \times 100\,(\%) \tag{1}$$

where:

$Load_{in}$ = inflow amount of a certain pollutant (COD, BOD, TSS, total N, or Total P) for a certain time period (kg); and

$Load_{out}$ = outflow amount of the same pollutant for the same period (kg).

The data for the years 2015–2021 are presented in Supplementary Material S2; the removal efficiency is shown in Figure 3.

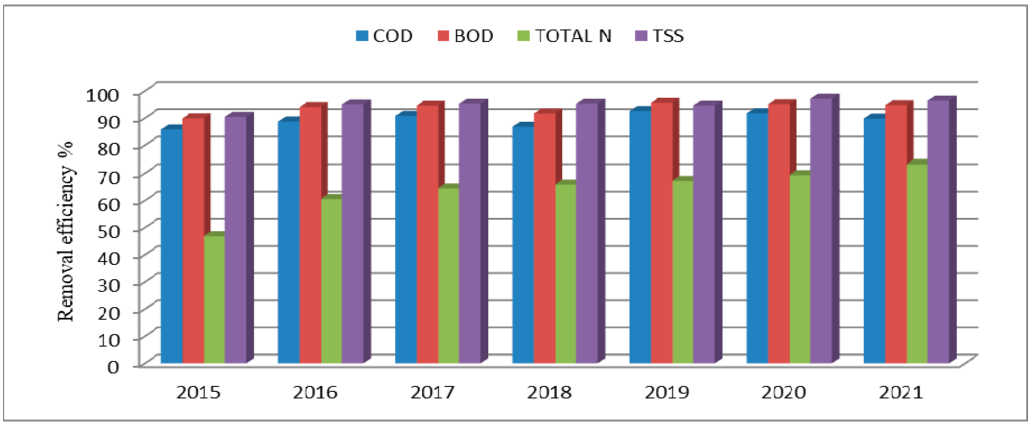


Figure 3. Removal efficiency for the main pollutants during the period 2015–2021.

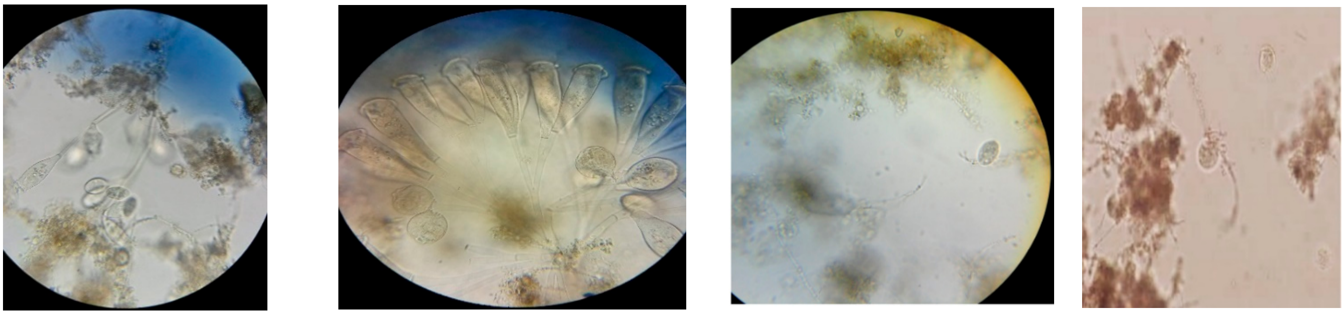
A good removal efficiency may be seen for COD, BOD, and TSS, and increased efficiency of total nitrogen after the addition of $FeCl_3$ (January 2017). The addition of $FeCl_3$ was beneficial for the removal of phosphorus, which gets closer to the permitted limits, as seen in Table 4.

Table 4. Phosphorus removal after the use of $FeCl_3$.

Year	Inflow m^3	Total P, mg/L		Elimination Efficiency, %
		Influent	Effluent	
2016	419,271	5.83	3.52	39.62
2017	351,662	5.76	3.38	41.32
2018	382,011	4.48	2.09	53.35
2019	335,185	5.54	2.7	51.26
2020	253,793	6.65	2.72	59.10
2021	307,925	6.67	3.03	53.46

It may be seen that the addition of $FeCl_3$ improves phosphorus removal, although the effluent concentration is above the MPL. Preliminary tests (data not shown) have proven that the supplementation with 40–50% of the amount of $FeCl_3$ will reduce the effluent concentration below the prescribed values for a WWTP serving more than 10,000 PE, but with higher costs, and so it was decided not to use additional $FeCl_3$.

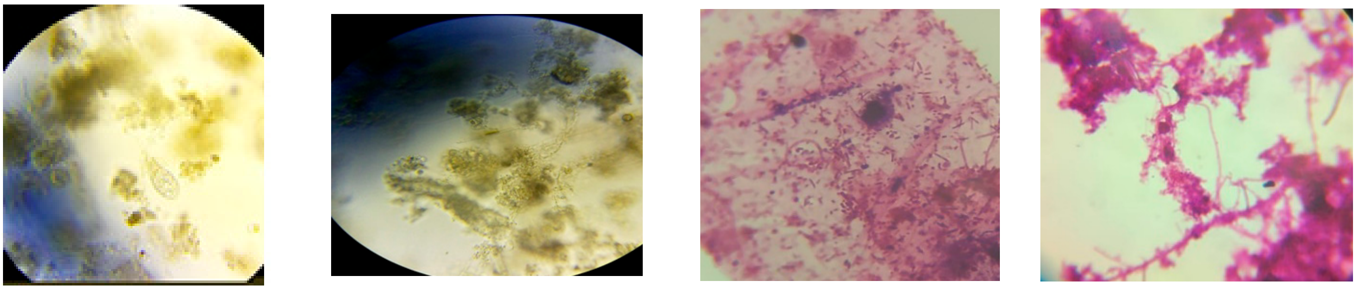
When performing the microscopic determination of the existing bacterial colonies within the RBC at the laboratory of the Apa Tarnavei Mari Co. in Medias, we found predominantly aerobic microorganisms, responsible for the transformation of ammonia nitrogen into nitrates, as seen in Figure 4.



Vorticella sp. *Carchesium* sp. *Aspidisca* sp.

Figure 4. Microscopic image of aerobic microorganisms (magnification 100×).

After the first two years of its commissioning, we also found anaerobic microorganisms in the RBC that transform nitrates into gaseous nitrogen, thus reducing the amount of total nitrogen in the effluent, as seen in Figure 5.



Paramecium sp. *Peranema* sp. *Acinetobacter* sp.

Figure 5. Microscopic image of anaerobic microorganisms (magnification 100×).

Their presence is beneficial for the WWTP because they provide the release of nitrogen into the air, thus allowing us to achieve the prescribed values for total nitrogen.

When analyzing their distribution, we found (as expected) more nitrifying microorganisms on the external disc surface, while the denitrifying ones were present in the water and near the disc surface.

To determine the efficiency of the major treatment steps for the removal of nitrogen, we measured the values of ammonium and total nitrogen (mean of determinations made every other month) between September 2021 to August 2022 (the period considered in this paper), which are presented in Table 5.

This determination showed that the nitrogen removal takes place mainly in the RBC, but it continues in the secondary settler as well.

Starting in January 2017, the required volume of ferric chloride was divided into three equal portions, all manually added at 8 a.m., 4 p.m., and midnight. This proactive attitude provided the good results presented in Figure 3 and detailed in Supplementary Materials S1 and S2.

In June 2021, we decided not to manually add the coagulant, but automatically, using a dosing system built in-house. The results reflected better compliance with the permitted discharge limits and keeping the amount of $FeCl_3$ within reasonable limits.

It is also noteworthy that since June 2021, the average concentration of $FeCl_3$ in the water was 37.0357 ± 10.098 μM, ten times less than the 400 μM stated by Kim et al. [50] to be toxic for microorganisms (complete data in Supplementary Material S1), thus preventing any "ferric shock".

The good results after one year of application of this method (September 2021–August 2022), presented in detail in Supplementary Material S1, are very consistent with those of Hassard et al. [16], Ni et al. [17], and Mizyed [20], and are comparable with those of Waquas et al. [51] and Cvetkovic et al. [52].

As expected, a reverse dependency may be seen between the volume of added $FeCl_3$ and water temperature (Figure 6). In the same figure we have also represented the specific removal power for nitrogen, which has a similar rise as the water temperature variation. This is normal because bacterial activity is stimulated by temperature increase [12] and we have already proven this in an activated sludge reactor [45].

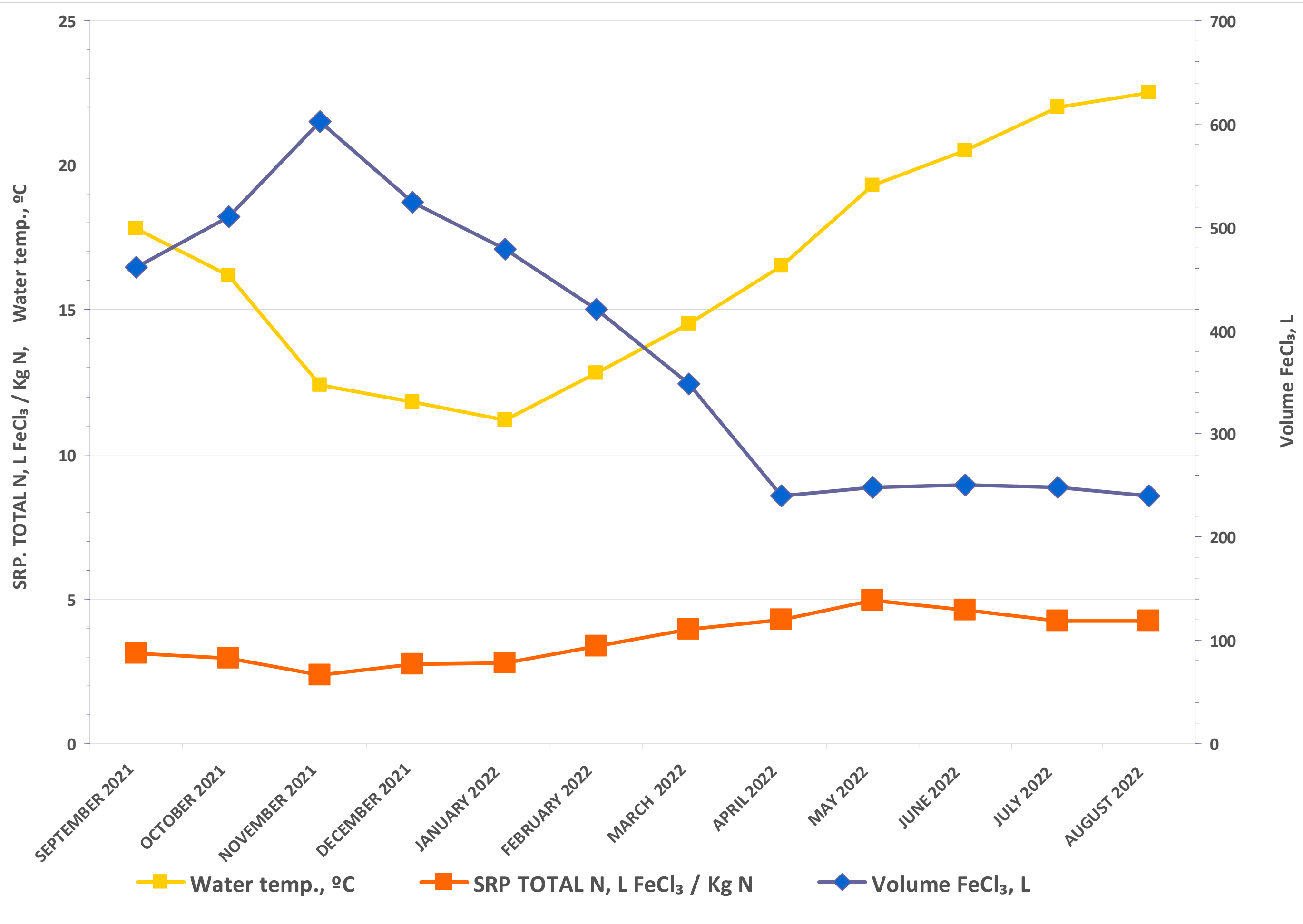


Figure 6. Seasonal variation in water temperature, SRP, and $FeCl_3$ consumption for the last 12 months (September 2021–August 2022).

Considering that ferric chloride boosts the removal of ammonia nitrogen, total nitrogen, and phosphorus, we introduced a new parameter, specific removal power (*SRP*), i.e., the volume of 40% $FeCl_3$ required for the elimination of 1 kg of the N or P, defined (Equation (2)) as:

$$SRP = \frac{V_{FeCl_3}}{M_x} \quad (2)$$

where:

V_{FeCl_3} = the volume of 40% ferric chloride solution (m^3);

x = the parameter considered for determination of the SRP (ammonia nitrogen, total N or total P); and

M_x = the total inflow load (kg) for this parameter for a certain time period.

Table 5. Removal efficiency of the major equipment.

Parameter Sampling Place	2021		2022	
	NH_4^+ mg/L	Total N mg/L	NH_4^+ mg/L	Total N mg/L
Influent	45.35	47	40.54	53.09
After primary settler	33.2	39	35.22	46.08
Removal efficiency—primary settler	26.79	17.02	13.12	13.20
After biodiscs	5.01	13.50	3.80	15.00
Removal efficiency biodiscs	**84.91**	**65.38**	**89.21**	**67.45**
After secondary settler	4.16	10.79	2.38	11.75
Removal efficiency—secondary settler	16.97	20.07	37.37	21.67
Total removal efficiency	**90.83**	**77.04**	**94.13**	**77.87**

A strong relationship between water temperature and bacterial activity, expressed by nitrogen removal when adding $FeCl_3$, was also observed (Figure 6, right OY axis).

In addition, we noticed that there are similarities between the shapes of the SRP curves (Figure 7).

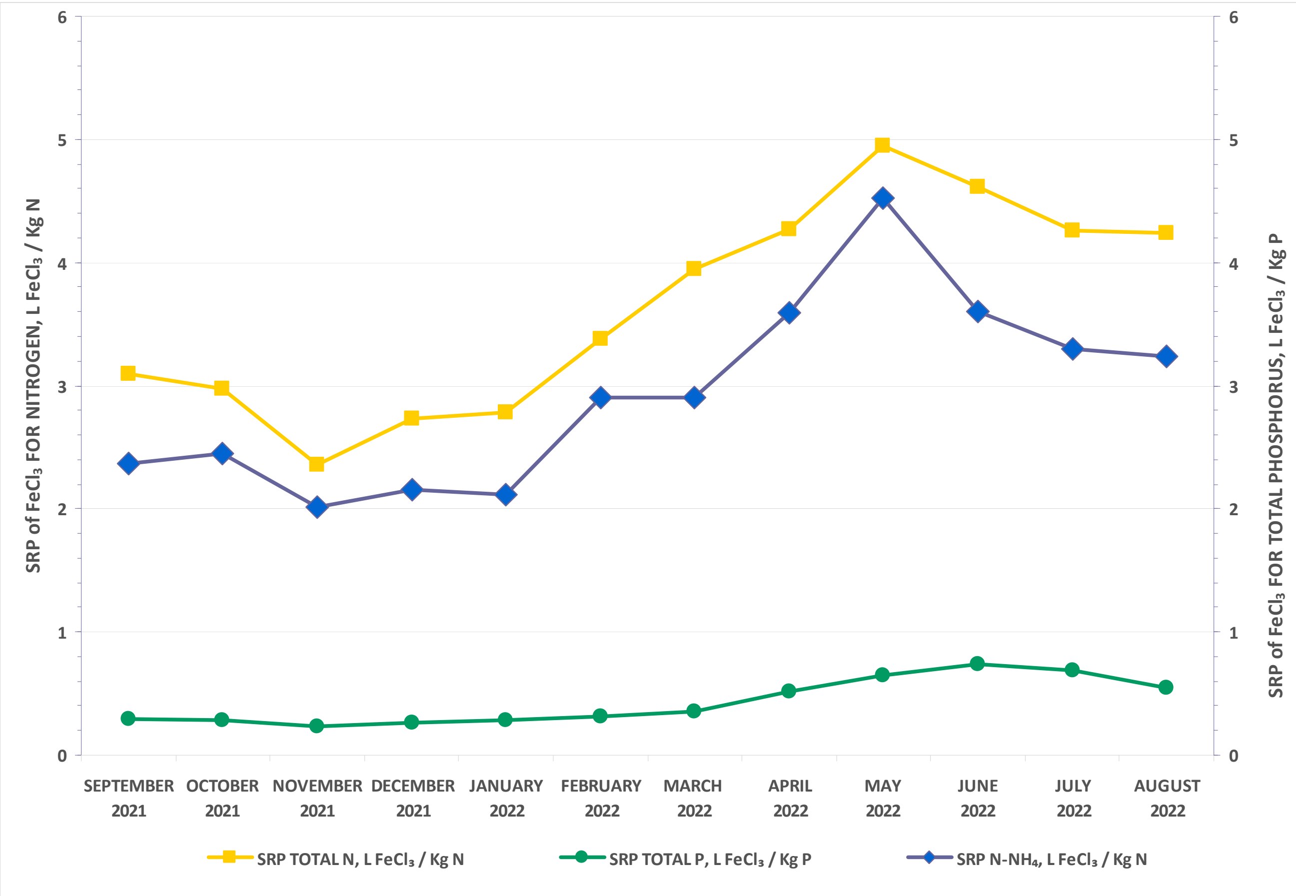


Figure 7. SRPs for total nitrogen, ammonia nitrogen, and phosphorus.

These similarities will allow us to consider ammonia nitrogen as a control parameter for the future development of an automated continuous dripping system.

When plotting the specific removal powers and water temperature, the results are shown in Figure 8.

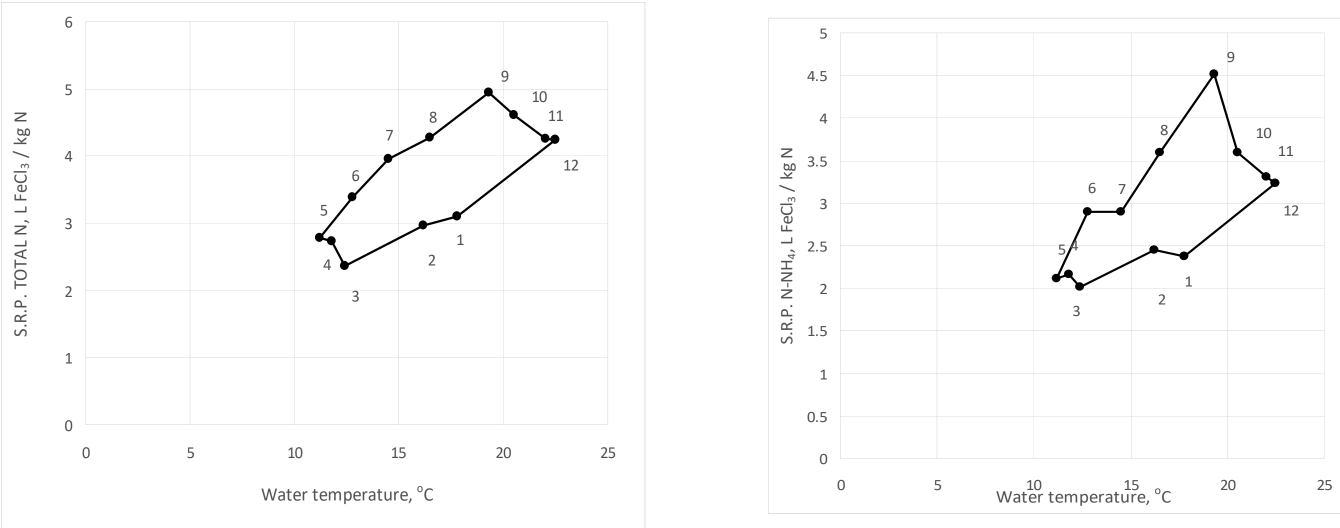


Figure 8. Correlations between water temperature and specific removal power for: **Left**: total nitrogen; **Right**: ammonia nitrogen.

When using this concept, the resulting diagrams (Figure 8) more explicitly show the pollutant removal after $FeCl_3$ addition correlated with the inflow load and temperature.

Furthermore, Figure 8 shows that the specific removal power for the studied parameters has a similar shape, and the diagrams can give valuable information to the plant operators.

Knowing the temperature and the inflow parameters (volume and pollutant concentration), the total daily load is calculated, and the operator can easily determine the volume of 40% $FeCl_3$ solution to be used for the next 24 h. This is performed manually at this moment, taking into account the diagram in Figure 8 and the season. For instance, if the water temperature is 15 °C in autumn, the SRP is 2.8 L 40% ferric chloride for 1 kg of total nitrogen, but in spring, for the same temperature, the SRP is 4 L/kg total N (i.e., for the same nitrogen load a smaller volume of $FeCl_3$ is required).

Although fast, this approach is not the best one, and in the future (2024) we intend to upgrade the WWTP with a system that will continuously measure the inflow load of ammonium and will automatically determine the $FeCl_3$ volume, considering the proportionality between the ammonia and total nitrogen, as presented in Supplementary Material S1 (in our case, on average, the total nitrogen is 1.24 times more than ammonia nitrogen).

4. Conclusions

Although the existing regulations do not oblige WWTPs operating for communities with less than 10,000 PE to comply with discharge limits for nitrogen and phosphorus, environmental protection and common sense require their removal at affordable cost.

Our method, new in the literature, is the addition of 40% $FeCl_3$ solution, without changing the design and operating parameters of our WWTP. The presence of ferric ions in water boosts the development of the microbiota responsible for both nitrification and denitrification, as well as phosphorus removal, discharging cleaner water.

For the quantification of the required $FeCl_3$ volume, we introduced a new parameter, the specific removal power, which is easy to calculate and very helpful in determining the volume of ferric chloride at different seasonal temperatures.

This method is simple to use, and the good results at Agnita have encouraged us to extend it to other wastewater treatment plants operated by the Apa Tarnavei Mari Co.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w14223670/s1, Supplementary Material S1 N P Operational Data 2021–2022; Supplementary Material S2 Main Operational Parameters 2015–2021.

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