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Scientific Paper

# Removal of phosphorus and nitrogen from swine manure using a natural coagulant

*Remoção de fósforo e nitrogênio de água residual de suinocultura utilizando coagulante natural*

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## ABSTRACT

Due to a significant amount of nutrients in its composition, swine manure (SM) is commonly used to complement or replace commercial agricultural fertilizers. However, an improper application in crops could cause negative environmental impacts, especially due to the excessive supply of nutrients in its composition. This paper aimed to evaluate the removal of total Kjedahl nitrogen (TKN) and total phosphorus (TP) from SM biodigested by the coagulation/flocculation (CF) process with organic polymers. The SM samples were collected in two distinct seasonal periods (rainy and dry seasons), with CF tests in two stages. During the first stage, slow mixing times (SMTs) of 15, 20, and 25 min were tested, and in the second stage, the coagulant (0 – 1.75 g $L^{-1}$) and flocculant dosage (0 – 0.0025 g $L^{-1}$) effects were evaluated. The results show that the optimal SMT was 15 min in both the seasons. In the rainy season, the highest phosphorus and nitrogen removals were 80 and 27%, respectively, while in the dry season, the highest phosphorus and nitrogen removals were 70 and 25%, respectively. The finding shows that the use of flocculants did not improve the removal of TP and TKN when compared with the control conditions. It is concluded that the CF process can be used to remove TP and TKN from SM, with a possible concentration of them in the generated sludge.


**Keywords:** coagulation; flocculation; nutrient removal; swine manure.


## RESUMO

Por conter uma quantidade significativa de nutrientes em sua composição, a água residual de suinocultura (ARS) é comumente utilizada para complementar ou substituir fertilizantes agrícolas comerciais. No entanto, a aplicação inadequada nas culturas pode causar impactos ambientais negativos, especialmente devido ao aporte excessivo de nutrientes presentes em sua composição. Neste estudo, o objetivo foi avaliar a remoção de nitrogênio total Kjedahl (NTK) e fósforo total (FT) da ARS biodigerida pelo processo de coagulação/floculação com polímeros orgânicos. As amostras de ARS foram coletadas em dois períodos sazonais distintos (estações de chuva e seca), com testes de coagulação/floculação (CF) em duas etapas. Na primeira etapa, foram testados tempos de mistura lenta (TML) de 15, 20 e 25 minutos, e na segunda etapa, foram avaliados os efeitos da dosagem do coagulante (0 – 1,75 g $L^{-1}$) e do floculante (0 – 0,0025 g $L^{-1}$). Os resultados indicaram que o TML ótimo foi de 15 minutos em ambas as estações. Na estação chuvosa, as maiores remoções de fósforo e nitrogênio foram de 80 e 27%, respectivamente, enquanto que na estação seca, as maiores remoções de fósforo e nitrogênio foram de 70 e 25%, respectivamente. Verificou-se que a utilização do floculante não trouxe melhorias nas remoções de FT e NTK. Conclui-se que o processo de CF pode ser empregado na remoção de FT e NTK da ARS, com possível concentração desses no lodo gerado.


**Palavras-chave:** coagulação; floculação; remoção de nutrientes; resíduos suínos.

## INTRODUCTION

In the search for solutions to increase productivity and reduce agricultural production costs, one of the techniques employed is the replacement of chemical fertilizers with swine manure (SM) (PEREIRA *et al*., 2016). The main advantages of using SM are its large availability (SOUSA *et al*., 2022), the possibility of using organic matter (LOPES *et al*., 2021), the recycling of essential nutrients for plants (SCHEID *et al*., 2020; SILVA *et al*., 2012), and the improvements in the physical and biological properties of the soil (ANTONELI *et al*., 2019; DAS *et al*., 2017).

Although SM presents soluble elements that are nutrients for plants, their high concentration as well as their high value of electrical conductivity (EC) (MORES *et al*., 2016) and the presence of microorganisms can cause unwanted impacts on the environment (SOUZA *et al*., 2020; FONGARO *et al*., 2014). Direct and unrestricted application of SM to the soil can negatively impact soil and groundwater with

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phosphorus (P), potassium (K), calcium (Ca), and magnesium (Mg) (SANTOS *et al.*, 2021; SILVA *et al.*, 2015) and total nitrogen (PEDROSA *et al.*, 2018). In addition, the presence of solids can interfere with the infiltration capacity of soils (SEPASKHAH; SOKOOT, 2010). In studies of SM application in lysimeters (KESSLER *et al.*, 2014), high concentration values were found for the elements copper (Cu), iron (Fe), manganese (Mn), nitrate ($NO_3^-$), nitrite ($NO_2^-$), sodium (Na), and zinc (Zn) in the leached material, indicating the pollution potential of SM.

Despite its important elements for agricultural production, the direct application of SM in the fields, without any sort of treatment, is not recommended. Thus, treatments for SM should be used with the objective of improving its physical, chemical, and microbiological aspects. Among the treatment options, anaerobic biodigestion stands out (MANYI-LOH *et al.*, 2013). Specifically, this translates to using the covered lagoon biodigester model, which consists of a closed chamber that prevents the contact of SM with atmospheric air and stores the biogas, favoring the sedimentation of solids and the degradation of a portion of the SM's organic matter (TÁPPARO *et al.*, 2021). Anaerobic biodigestion, in general, is well accepted from a general point of view for the reuse of SM in the soil (CÂNDIDO *et al.*, 2022; SOARES; FEIDEN; TAVARES, 2017), as the biodigestion process preserves all nutrients, making them readily available for plants (DORNELAS *et al.*, 2021). On the contrary, to meet the standards of releases into water bodies, the biodigested SM lacks a later stage (post-treatment).

Specifically for nutrient removal, there is a well-known and established treatment, namely, a struvite precipitation, that focuses on nutrients removal, especially P and N (MAVHUNGU *et al.*, 2021; LORICK *et al.*, 2020; WRIGLEY; WEBB; VENKITACHALM, 1992), but not works like a clarification treatment, in case of releases into water bodies. For this, among the processes that could be used as a post-treatment, coagulation/flocculation (CF) is widely used (ZHAO *et al.*, 2021; GÖKÇEK; ÖZDEMIR, 2020; IRFAN *et al.*, 2017; VERMA; DASH; BHUNIA, 2012), as it is considered an efficient and traditional method in the treatment and clarification of effluents. With CF, inorganic coagulants are normally used (based on aluminum and iron salts). Even though they are efficient (PAULA *et al.*, 2018), and of low acquisition cost (KEELEY *et al.*, 2016), these reagents generate non-biodegradable sludge, with low potential for agricultural use due to the presence of metals (TEH *et al.*, 2016).

Other studies demonstrate the potential for using natural coagulants (LEITE; HOFFMANN; DANIEL, 2019; MAURYA-DAVEREY, 2018; YUSOFF *et al.*, 2018) in separation processes. According to Teixeira *et al.* (2022), the use of natural coagulants, such as those based on tannin, is a technical alternative that provides benefits to the environment.

Tannins act in colloidal systems, neutralizing charges and forming bridges between these particles, flocs, and subsequent sedimentation. Due to their cationic characteristic, they tend to bond with negative charges present in the medium, which is interesting for SM. Furthermore, the process enables the formation of two products of interest: liquid (supernatant), which can be sent to receiving bodies, and sludge (semi-solid), eventually with fertilizing characteristics.

A large number of studies on CF treatment in swine effluents are presented in the literature (CHELME-AYALA *et al.*, 2011; RIAÑO; GARCÍA-GONZÁLEZ, 2015; GABRIEL *et al.*, 2019; EL BIED *et al.*, 2021). However, few reports on the application of CF as a post-treatment to biodigestion are described (LEE; CHANG, 2022). The objective of this paper was to evaluate the removal of total Kjeldhal nitrogen, TKN, and total phosphorus, TP, from biodigested SM using CF with organic polymers as a post-treatment. Different slow mixing times (SMT 15, 20, and 25 min), coagulant (0 – 1.75 g $L^{-1}$) and flocculant (0 – 0.0025 g $L^{-1}$) dosages, and the influence of seasonality (rainy and dry seasons) were studied bringing efforts to a new way of treating swine manure.

## MATERIALS AND METHODS

### Swine manure sampling location

SM samples were collected on a rural property, with a herd of approximately 50,000 pigs, located in the city of Vera, in northern Mato Grosso, Brazil. The animals are raised in an intensive system within collective pens featuring a concrete floor. The water level in the pens is carefully drained once a day, and the animals have unrestricted access to water through nipple drinkers. The drained water, which consists of raw swine manure, is directed to the biodigester using open channels, and then it flows through buried pipes.

The predominant climate in the region is *Aw* (dry winter), characterized by the presence of two well-defined seasons, the rainy season (October to April) and the dry season (May to September), with a low annual thermal amplitude (averages 24 and 27°C) and an average annual precipitation at around 1974 mm (SOUZA *et al.*, 2013), concentrated mainly in the rainy season (94%).

### Collection, preservation, and characterization of samples

The SM exits the biodigester (25-day hydraulic retention) and goes to a storage pond (2500 $m^2$ and 7500 $m^3$) through an underground pipe. Hence, the SM collection was carried out in the pond, in a location close to the SM inlet pipe in the pond. Single samples were collected at the edges of the storage pond, with the aid of a bucket-type sampler, in two different periods: one collection in the rainy season (January 17) and one collection in the dry season (June 17). The material was stored in 4 gallons of 20 L polyethylene (~80 L in each season), previously cleaned, and maintained by cooling. Sample collection and preservation followed the protocols established by the Standard Methods for Examination of Water and Wasterwater (APHA 2012).

The initial characterization of SM was performed through the parameters pH, EC, apparent color, turbidity, TKN, TP, total solids (TS), chemical oxygen demand (COD), and biochemical oxygen demand (BOD) (Table 1) and followed the standards established by the Standard Methods for Examination of Water and Wasterwater (APHA, 2012).

**Table 1** – Parameters and methods of analysis applied for initial characterization of swine manure.

| Parameter analyzed | Unit | Method |
|---|---|---|
| pH | – | Electrometric (4500 $H^+$ – B.) |
| EC | mS $cm^{-1}$ | Conductimetric (Instrumental measurements) |
| Apparent color | mg Pt-Co $L^{-1}$ | Spectrophotometric-single-wavelength (2120 – C.) |
| Turbidity | NTU | Nephelometric (2130 – B.) |
| TKN | mg $L^{-1}$ | Semi-micro-Kjeldahl (4500 $N_{org}$ – C.) |
| TP | mg $L^{-1}$ | Ascorbic acid (4500 P – E.) |
| TS | mg $L^{-1}$ | Gravimetric (2540 B.) |
| COD | mg $L^{-1}$ | Closed reflux, Colorimetric (5220 – D.) |
| BOD | mg $L^{-1}$ | 5-day BOD test (5210 – B.) |

EC: electrical conductivity; TKN: total Kjedahl nitrogen; TP: total phosphorus; TS: total solids; COD: chemical oxygen demand; BOD: biochemical oxygen demand.

### Post-treatment experimental tests applied to swine manure

To carry out the CF tests, all samples were subjected to sieving (1-mm opening) to remove larger solids. The CF tests were performed in a 1-L jar test, using an SM volume of 0.25 L. In each seasonal period (rainy and dry seasons), the tests performed evaluated speed mixing time (15 – 25 min) and dosage of the coagulant (0 – 1.75 g $L^{-1}$) and flocculant (0 – 0.0025 g $L^{-1}$) that provided a greater significant removal of TP and TKN, defining two treatment stages.

In the first stage of the tests, the optimal SMT was determined using the following experimental conditions: rapid mixing time (RMT) of 2 min, rapid mixing speed (RMS) of 120 rpm, SMT of 15, 20, and 25 min, with a slow mixing speed (SMS) of 20 rpm, and decanting time of 90 min in all conditions. In the second stage of the experiments, the best significant dosage of coagulants and flocculants were defined using 2 min RMT, 120 rpm RMS, optimal SMT (obtained in the first stage), 20 rpm SMT, and a settling time of 90 min in all conditions.

The coagulant used was the tannin-based cationic polymer, and the flocculant was an anionic polymer. In both stages, the coagulant dosages evaluated were 0.5, 0.75, 1.0, 1.25, 1.5, and 1.75 g $L^{-1}$, and the control experiment was performed without the coagulant. The flocculant dosages were 0.0000 (control), 0.0005, 0.0015, and 0.0025 g $L^{-1}$, which correspond to the values between 0.03 and 0.5% of the coagulant concentrations. The operational conditions were established based on other studies performed with tannin-based coagulants that demonstrated good results for other organic effluents (SCHNEIDER *et al.*, 2021; WOLF *et al.*, 2015).

### Experimental design and statistical analysis

In the first stage of the experiment, a completely randomized design (CRD) was used, in a 3 × 7 factorial scheme, with three times of slow mixing (SMT of 15, 20, and 25 min) and six doses of coagulant, and the control (0.5, 0.75, 1.0, 1.25, 1.5, 1.75, and 0.0 g $L^{-1}$), respectively, totaling 21 treatments, with three replications each. In the second stage of the experiment, a CRD was also used, in a 7×4 factorial scheme, with seven doses of coagulant (0.0, 0.5, 0.75, 1.0, 1.25, 1.5, and 1.75 g $L^{-1}$), and four doses of flocculant (0.0000, 0.0005, 0.0015, and 0.0025 g $L^{-1}$), totaling 28 treatments, in three replications each. The experimental design conditions used in the first and second steps of the SM treatment (T) in TKN and TP removal are described in Table 2.

**Table 2 –** Experimental design for first and second steps of the swine manure treatment evaluating slow mixing time and coagulant-flocculant dosages in total Kjeldahl nitrogen and total phosphorus removal.

| SMT (min) | Step 1 | | | | | | |
|---|---|---|---|---|---|---|---|
| | Coagulant (g $L^{-1}$) | | | | | | |
| | 0.00 | 0.50 | 0.75 | 1.0 | 1.25 | 1.50 | 1.75 |
| 15 | TF1 | TF2 | TF3 | TF4 | TF5 | TF6 | TF7 |
| 20 | TF8 | TF9 | TF10 | TF11 | TF12 | TF13 | TF14 |
| 25 | TF15 | TF16 | TF17 | TF18 | TF19 | TF20 | TF21 |
| Flocculant (g $L^{-1}$) | Step 2 | | | | | | |
| | Coagulant (g $L^{-1}$) | | | | | | |
| | 0.00 | 0.50 | 0.75 | 1.0 | 1.25 | 1.50 | 1.75 |
| 0.0000 | TS1 | TS2 | TS3 | TS4 | TS5 | TS6 | TS7 |
| 0.0005 | TS8 | TS9 | TS10 | TS11 | TS12 | TS13 | TS14 |
| 0.0015 | TS15 | TS16 | TS17 | TS18 | TS19 | TS20 | TS21 |
| 0.0025 | TS22 | TS23 | TS24 | TS25 | TS26 | TS27 | TS28 |

STM: slow mixing time.

The effect of such treatments on the removal of the parameters TKN and TP was tested by means of an analysis of variance (ANOVA) at 5% of significance. When differences were verified, average tests were performed (Tukey or Scot-Knott). In both stages, the statistical program used was Sisvar (FERREIRA, 2011).

## RESULTS AND DISCUSSION

The physical and chemical characterization of the biodigested SM showed that the quality requirements for the release of this effluent into receiving water bodies were not met (BRASIL, 2005) due to the presence of high values of color, turbidity, organic matter, and TP. The analyzed parameters showed higher mean values in the dry period, except for the EC parameter (Table 3).

According to Santos *et al.* (2016), biodigesters operating stably produce effluents with neutral or slightly alkaline pH values. This corresponds to the pH values found in the SM.

The reported EC values indicate a high concentration of ions (Na, Mg, Ca, and K mainly), which is expected in effluents. Halder *et al.* (2017) evaluated 66 samples of swine effluents and reported average EC values of 4.10 – 39.7 mS $cm^{-1}$, which is the range of values in which those obtained in this study fall.

The color demonstrated high measured values (greater than 7000 mg Pt-Co $L^{-1}$), which is a characteristic of effluents and waters with high levels of organic matter (SANTOS *et al.*, 2018), as is the case of SM. In relation to turbidity, there was an increase of 2.7 times in the value in the dry season in relation to the rainy season. It was observed that this parameter showed a similar relationship with the concentration of TS in both samples (turbidity/ST = 3.1 (rainy) and 3.3 (dry)). The concentration of solids proportionally influenced the turbidity value of the sample.

In terms of nutrients, for TKN, in both seasons, and for TP, in the rainy season, the quantified values are low when compared with the values obtained by Pedrosa *et al.* (2018), who reported values of 308.7 and 150.29 mg $L^{-1}$ of TKN and TP, respectively, at neutral pH values (pH 6.85) after the SM went through the biodigester. At neutral-alkaline pH, there is precipitation of phosphorus (MAVHUNGU *et al.*, 2021), and the balance of the reaction of the nitrogenous fraction tends toward the formation of ammonium ions (ACHILLEOS; ROBERTS; WILLIAMS, 2022). However, the difference in values obtained by Pedrosa *et al.* (2018) and those obtained in this work does not explain the chemical route of the processes that occurred, indicating that biological factors or biochemical changes may have contributed to the difference.

**Table 3 –** Physicochemical characterization of SM in the rainy and dry seasons.

| Parameters analyzed | Unit | Rainy season | Dry season |
|---|---|---|---|
| pH | – | 7.89 ± 0.02 | 8.08 ± 0.16 |
| EC | mS $cm^{-1}$ | 8.74 ± 0.25 | 8.35 ± 0.19 |
| Apparent color | mg Pt-Co $L^{-1}$ | 7100 ± 14.14 | 8600 ± 14.14 |
| Turbidity | NTU | 787 ± 64.4 | 2100 ± 21.2 |
| TKN | mg $L^{-1}$ | 7.5 ± 0.1 | 10.50 ± 0.2 |
| TP | mg $L^{-1}$ | 37.16 ± 1.01 | 179.36 ± 1.12 |
| TS | mg $L^{-1}$ | 254 ± 0.00 | 642 ± 0.00 |
| COD | mg $L^{-1}$ | 2167 ± 19.1 | 3537 ± 2.0 |
| BOD | mg $L^{-1}$ | 206.7 ± 4.2 | 305.8 ± 5.5 |

EC: electrical conductivity; TKN: total Kjedahl nitrogen; TP: total phosphorus; TS: total solids; COD: chemical oxygen demand; BOD: biochemical oxygen demand.

The values of BOD and COD indicate the presence of organic matter in the SM, with recalcitrant characteristics, i.e., it has a low value for the BOD/COD ratio (value < 0.3). González and Varela (2016) reported that wastewater from pigs and cattle has highly variable BOD/COD ratios ranging from 0.02 to 0.66.

The ratio between the average values of each parameter, in both the rainy and dry seasons (Figure 1), varies generally, not showing equal relationships between them (variations between 0.96 and 4.8). These results indicate that, apparently, they do not have the same source of variation, being dependent, in addition to climate (season), on animal management, diet, water consumption, and other factors, resulting in higher average values in the dry season. Sarto *et al.* (2019) reported differences between the characteristics of swine effluents between seasonal seasons, indicating that the differences are dependent on the generation of the effluent itself.

## Post-treatment with coagulation/flocculation – Step 1

The results of the ANOVA based on the data obtained from the application of CF in the digested SM showed that there were significant effects ($p \leq 0.05$) of the interaction between SMT and dosage in the removal of TP and TKN. The TP and TKN removal values in the rainy and dry seasons are shown in Figures 2 and 3, respectively. In general, it was observed that the removals of TP and TKN were quite variable according to treatments and seasons.

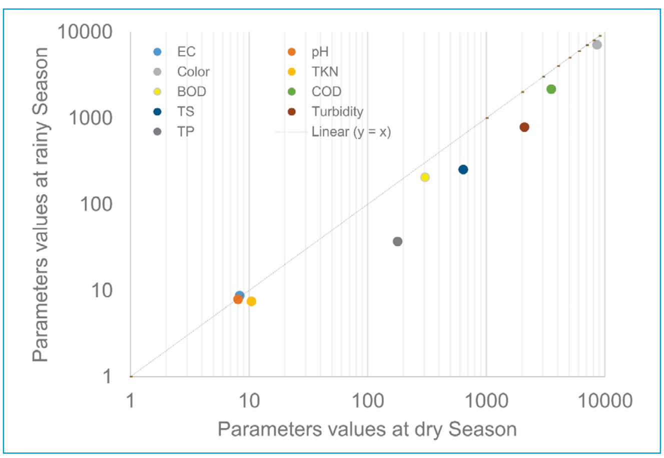


**Figure 1** – Relationship between the collections in the rainy and dry seasons (axes in logarithmic scale).

As for the variations of SMT on TP in the rainy season (Figure 2A), statistically equal effects were verified for 15, 20, and 25 min at dose values of 1.25 and 1.50 g $L^{-1}$, and removal values were greater than 55%. In the dosages of 0.50 – 1.00 g $L^{-1}$, the time of 15 min resulted in a higher removal percentage (greater than 79%).

TKN removal results for SMT variations (Figure 2B) during the rainy season showed statistically equal effects for 15, 20, and 25 min at the dosages of 0.00, 0.75, and 1.75 g $L^{-1}$, with removal values lower than 30%. A nitrogen removal rate above 50% was observed at the dosage of 1.00 g $L^{-1}$ in SMT 25 min, 45% in the dosage of 1.50 g $L^{-1}$ in SMT 15 min, and above 35% in the dosages of 0.5 and 1.25 g $L^{-1}$ in SMT 25 min.

As for the dry season (Figure 3A), the results of TP removal for the SMT variations within each dosage showed that the highest TP removals (> 68%) were achieved when SMT was equal to 15 min, except for the dosage of 1.50 and 1.75 g $L^{-1}$ (57.5 and 55.9%, respectively). Removals were statistically equal at doses of 1.00 and 1.25 g $L^{-1}$, greater than 68%, regardless of the tested SMT. Removal of TP was above 68% at dosages from 0.50 to 1.25 g $L^{-1}$ in SMT 15 min, 1.0 to 1.75 g $L^{-1}$ in SMT 20 min, and 0.75, 1.0, 1.25, and 1.75 g $L^{-1}$ in SMT 25 min.

We encountered a removal of TKN (Figure 3B) above 24% in the dosages of 0.75, 1.00, and 1.75 g $L^{-1}$, regardless of the tested SMT. For the dosage of 1.50 g $L^{-1}$, the SMT of 15 and 20 min allowed greater removals (> 24%) and did not differ from one another. As for the dosage of 0.5 g $L^{-1}$, the 20-min SMT resulted in a greater removal (25.5%).

Overall, upon reaching the ideal dosage, we verified that values referring to nutrient removal either decreased or stabilized. The reason for this may be the restabilization of organic particles, indicating a process controlled by charge neutralization (BOLTO, 1995). Michael-Kordatou *et al.* (2015) indicated that increasing the dosage of the coagulant promotes an increase in positive charges in the effluent, favoring the dissociation of the particles instead of their agglutination. The excess coagulant is adsorbed by the colloidal particles present in the SM, leading to the reversion of the colloidal charges from negative to positive, and these colloids are restabilized and begin to repel each other (IBAHIM; YASER, 2019).

Most of the phosphorus found in crude SM is in organic form (MONTALVO *et al.*, 2020), but after anaerobic treatment, which is the case with the SM in this study, part of the phosphorus is transformed into soluble phosphates, such as the

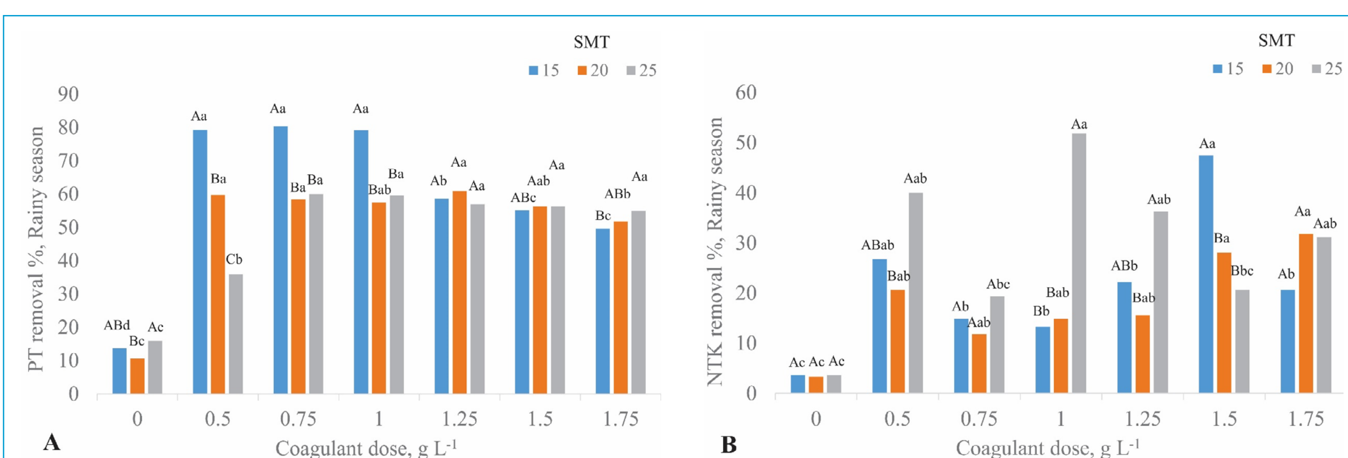


**Figure 2** – Mean values of total phosphorus and total Kjeldahl nitrogen removal (%) in the rainy season. Equal uppercase letters in each coagulant dose, in all SMT (comparison of all colors, within each coagulant dose), and lowercase letters in each slow mixing time, in all coagulant dose (comparison of each color, among all coagulant dosages), indicate that the values do not differ from each other, at 5% of significance.

phosphate group ($PO_4^{3-}$) (METCALF; EDDY, 1991). Thus, the coagulant, with a cationic character, possibly acted basically in the agglutination and removal of this negatively charged phosphate.

High phosphorus removals (79%) were observed by Kunz, Steinmetz e Bortoli (2010), who studied the removal of phosphorus from crude SM in a dissolved air flotation (DAF) system, using synthetic tannin and a polymer-based coagulant. On the contrary, Riaño and García-González (2014), when treating swine effluents (post-sieving) by CF with synthetic polymers, found phosphorus removals of 87.4%. Lee and Chang (2022), when evaluating the CF of SM post-anaerobic digester + anoxic reactor, found high removals of total nitrogen (99%) and TP (95%), in tests where the pH value was greater than 11 and where the flocculant was non-ionic.

The differences in TP removal between the rainy and dry seasons are, so it seemed, less (average values of 60%). However, it is noteworthy that, in the dry season, most of the TP removal took place without the use of coagulants for the 15-min SMT. But for 20 and 25 SMT, removals increased by at least 6% for doses higher than 0.75 g $L^{-1}$.

The CF process acts mainly in colloidal systems and, probably, the largest portion of TP was linked to particulate solids that sedimented even without the presence of the coagulant. This physical action dominated the process, even when the coagulant was used. Such a result would indicate that CF does not need to be used in the dry period. Nevertheless, we reinforce that the use of the coagulant increased TP removal and, therefore, eventually increased the TP concentration in the decanted solid (sludge).

Analyzing the results for the TKN, we noticed that the removals ranged from 30 to 50% in the rainy season to approximately 25% in the dry season. Zordan, Saléh e Medonça (2008) indicated that part of the nitrogen present in the SM is mainly in the form of ammonium ion ($NH_4^+$), which has a positive charge. Considering that the coagulant used in this study is cationic, it does not act in the destabilization of the ammonium ion. It is noteworthy that the study was carried out in the quantification of TKN, which quantifies the organic and ammoniacal fractions simultaneously. On the contrary, Achilleos, Roberts e Williams (2022) have drawn attention to the uncontrolled precipitation of struvite ($Mg:NH4:PO_4^{2-}$) in the neutral-alkaline pH range (pH 5.9 to 8.4). This occurrence arises from the presence of different ions in the organic effluent, leading to competitive reactions, and the rapid kinetics of phosphorus precipitation. Such conditions could explain the variations in removal efficiency observed during different seasons. However, it is also possible that biological and biochemical processes, involving urea consumption, have contributed to the observed values. To gain a deeper understanding, further investigation into the chemical, biological, and biochemical routes facilitated by the present microorganisms is necessary.

One should note that, even though TKN removal is not high, there is still TKN removal from the SM, reinforcing the importance of using the CF process. Doses of 0.75 g $L^{-1}$ have already provided an increase of at least 10% in TKN removal compared with non-use.

With the results of the first step, the optimal SMT was established, considering the shortest mixing time and using the least amount of coagulant to achieve the desired removal effect for TP and TKN. Thus, we opted that, in both the rainy and dry seasons, the most suitable conditions for TP and TKN removals occurred in the 15-min SMT.

## Step 2

The ANOVA showed that there were significant effects ($p \leq 0.05$) in the removal of TP and TKN with the treatments tested (dosages of coagulant and flocculant) in the rainy and dry seasons.

The results for TP removal in the rainy season (Figure 4A) show that removals in the order of 80% took place without the addition of a flocculant, at coagulant doses between 0.50 and 1.00 g $L^{-1}$. With the addition of the flocculant, TP removals were also observed in all dosages (15 to ~45%), including tests without a coagulant (~15%). However, removals were, in general, lower (from 20 to 50%) than removals with the coagulant alone. For the dry season (Figure 5A), the highest TP removals (approximately 70%) occurred at doses between 0.5 and 1.25 g $L^{-1}$ without the use of a flocculant. Removal differences between tests, with and without a flocculant, are less evident in the dry season (differences from 5 to 25%), with most of the highest removals observed (65 – 70%) when not using the flocculant.

Despite this, we noticed that, in general, the use of the flocculant reduced phosphorus removal efficiency, that is, there were lower removal efficiencies when the flocculant was added in both seasons.

The CF study with a focus on TKN showed that TKN removals were sometimes greater, sometimes similar, or slightly lower, in the tests without the flocculant, varying according to the dosages of coagulant and flocculant applied, in both seasons (Figures 4B and 5B). The highest TKN removal in the rainy season was observed at 1.50 g $L^{-1}$, without the flocculant (approximately 50% removal). For the same season, at dosages of 0.75 and 0.0025 g $L^{-1}$ of coagulant and flocculant, the removal of TKN

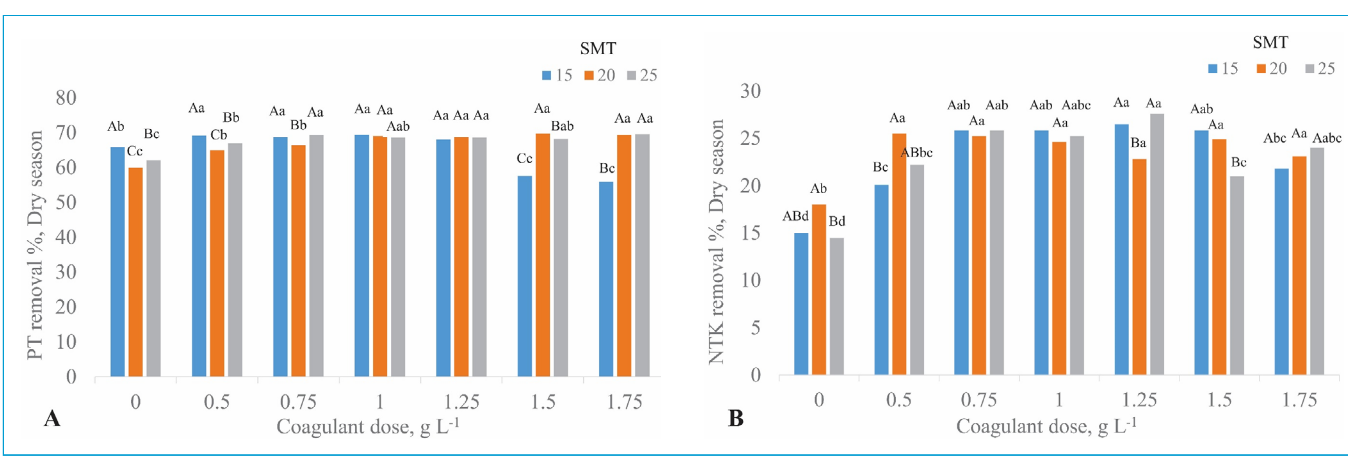


**Figure 3 –** Mean values of total phosphorus and total Kjedahl nitrogen removal (%) in the dry season. Equal uppercase letters in each coagulant dose, in all slow mixing times (comparison of all colors, within each coagulant dose), and lowercase letters in each slow mixing time, in all coagulant dose (comparison of each color, among all coagulant dosages), indicate that the values do not differ from each other, at 5% of significance.

approached 40%. On the contrary, the highest removals of TKN in the dry period were 25% at a coagulant dosage between 0.75 and 1.50 g $L^{-1}$, without the flocculant, and 30% at dosages 1.0 and 0.0005 g $L^{-1}$ of coagulant and flocculant, respectively.

The anionic flocculant, even at low dosages (from 0.03 to 0.5% of the coagulant dosage), in general, in addition to not improving TP and TKN removals, promoted the destabilization of the system, with a reduction in removals when compared with the process without using it. Eventually, for TKN, in the dosages of 0.5 g $L^{-1}$ (rain) and 0.75 g $L^{-1}$ (dry) of the coagulant, the flocculant (0.0025 and 0.0005 g $L^{-1}$) contributed to the increase in the removal of this nutrient. Therefore, it was observed that the incorporation of the flocculant brought eventual gains in the removal of TP and TKN. The presence of the flocculant basically did not cause the effect of reducing the dosage of the coagulant, being of less importance in the process of treating SM.

The issue of the non-functioning flocculant in the treatment performed may be due to its anionic character. When inserting the flocculant, there is an increase in negatively charged particles, and these unbalance the system loads, causing the restabilization (disagglutination) of part of the TKN.

Thus, regarding the removal of TP and TKN nutrients, considering the best dosages as those that demand the least amount of products with the maximum removal, it was defined that the nutrient of greatest interest to be removed from the liquid phase is TP. Therefore, the use of a flocculant can be discarded, and the best coagulant dosages were 0.5 g $L^{-1}$ in the rainy season (80 and 27% of TP and TKN removal) and 0.75 g $L^{-1}$ in the dry season (70 and 25% of removal of TP and TKN) (highlighted in blue in Figures 4 and 5).

As a preliminary test, it was demonstrated that CF can be a tool to be adopted in the removal of nutrients from the SM, employed as a nutrient recovery technology (MONTALVO *et al.*, 2020; LEE; CHANG, 2022), enabling the transfer of nutrients to the sludge with the aid of the organic coagulant.

## CONCLUSION

The CF process possesses the ability to remove phosphorus and nitrogen nutrients from the SM. Removals vary according to coagulant dosages and seasonality. The use of flocculants is not recommended for the removal of nitrogen and phosphorus from the SM.

Coagulant dosages of 0.5 and 0.75 g $L^{-1}$ demonstrate the highest removals of TP and TKN from the SM in the rainy and dry seasons, respectively. CF with an organic coagulant can be used as an agent that removes nutrients from the SM. Understanding that this process agglutinates the elements which then leave the liquid mass by gravitational sedimentation, there is the formation of sludge enriched in nutrients. This phenomenon makes way for new papers and further investigation.

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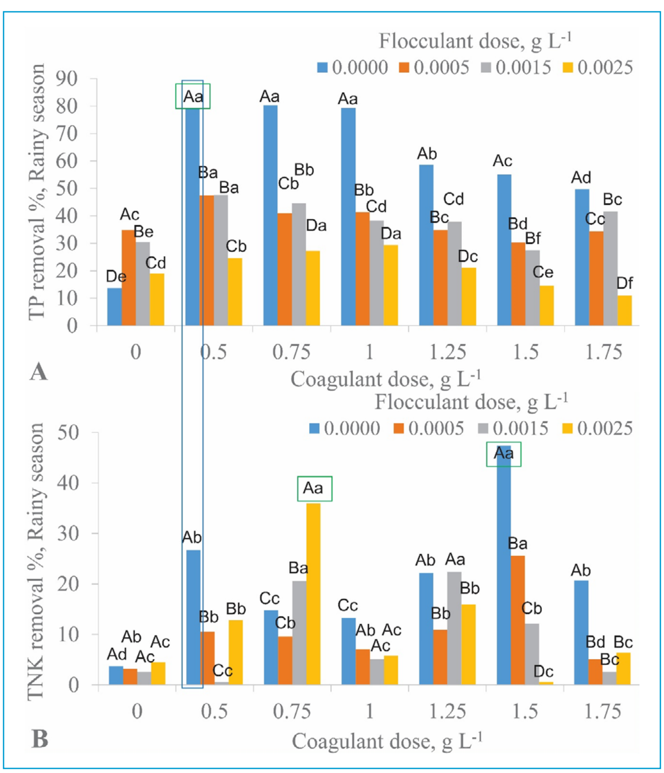


**Figure 4** – Mean values of nutrient removal as a function of coagulant and flocculant dosages. Total phosphorus (A) and total Kjedahl nitrogen (B) in the rainy season. Equal uppercase letters in each coagulant dosage, in all flocculant dosages (comparison of all colors, within each coagulant dosage), and lowercase letters in each flocculant dosage, in all coagulant dosages (comparison of each color, among all coagulant dosages), indicate that the values do not differ from each other, at 5% significance.

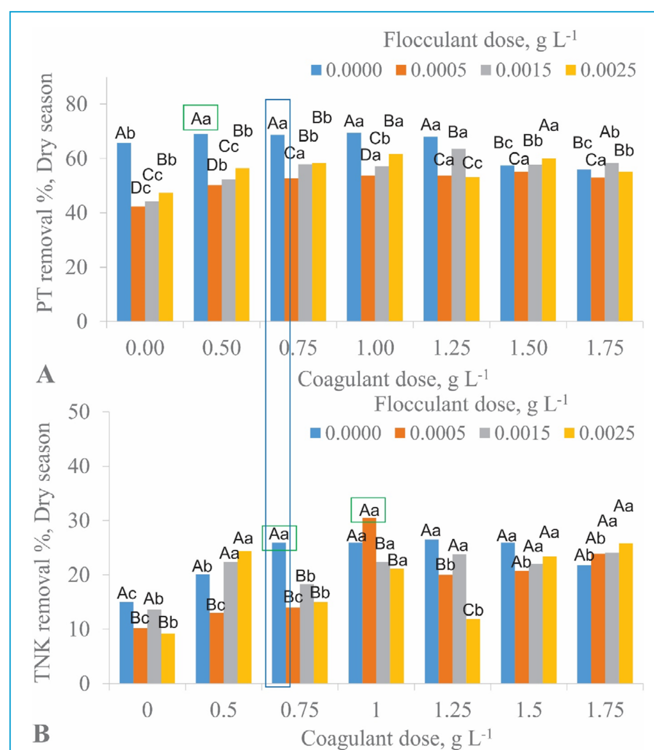


**Figure 5** – Mean values of nutrient removal as a function of coagulant and flocculant dosages. Total phosphorus (A) and total Kjedahl nitrogen (B) in the dry season. Equal uppercase letters in each coagulant dosage, in all flocculant dosages (comparison of all colors, within each coagulant dosage), and lowercase letters in each flocculant dosage, in all coagulant dosages (comparison of each color, among all coagulant dosages), indicate that the values do not differ from each other, at 5% significance.

## AUTHORS' CONTRIBUTIONS

Schneider, R.M. and Roveri, M.C.B.: Conceptualization, Methodology and Project administration. Paixão, G.C.: Investigation, Data curation and Writing – original draft. Paixão, G.C. and Amaral, A.G.: Formal Analysis. Schneider, R.M., Amaral, A.G., Boina, R.F. and Roveri, M.C.B.: Visualization, Writing – review & editing.

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