



Magnetic water treatment in a wastewater treatment plant: Part I - sorption and magnetic particles

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ABSTRACT

The application of magnetic sorption to treat wastewaters is nowadays seen as a potential industrial method. In this work we apply magnetite particles to remediate real wastewater samples, with several contaminants competing for the same active sorption center at the same time. We also apply our studies at three different sampling points of a Wastewater Treatment Plant. In general terms, magnetite particles have shown a very good behaviour concerning the reduction of detergents and COD, while phosphates and total nitrogen, and the majority of heavy metals are high to moderately removed. The influence of the type of wastewater (i.e., sampling point) has also shown to be important especially for high concentration of contaminants.

1. Introduction

Wastewater treatment is an important problem that is not yet completely solved. In fact, the increasing world scarcity in water supply and the continuous increase in world population leads to an increase concern on reducing wastewater volumes and on its reusability (Boretti and Rosa, 2019; Angelakis and Snyder, 2015; Zarei et al., 2020; de Aquim et al., 2019). There is also a surge of emergent pollutants, including antibiotics, acidic pharmaceuticals and novel pesticides (Taheran et al., 2018; EPA, 2020; Peña-Gusmán et al., 2019; Castelo-Grande, 2010a, 2010b, 2010c). Besides, pollutants emitting limits are becoming more stringent (Castelo-Grande et al., 2018). All this urges an improvement on the current methods used for wastewater treatment, as some of them are simply not efficient anymore or become too expensive to be applied. Furthermore, a typical wastewater treatment process generates activated sludge, which is still an environmental complicate

sub-product to handle. (Zhang et al., 2018; Wagas et al., 2020; Augusto et al., 2019; Castelo-Grande et al., 2008).

Selecting the best methods and materials for the treatment of contaminated water is a difficult task in which many factors must be considered, the 4 main factors to look at are: efficiency, reuse of particles, environmental safety and low cost (Zhang and Fang, 2010; Oller et al., 2011).

Many methods have been proposed in the last years as possible alternatives for remediation of wastewaters. These are the cases of advanced oxidation, electrocoagulation, microalgae use, etc. (Rajasulochana and Preethy, 2016; Miklos et al., 2018; Wollmann et al., 2019; Islam, 2019; Salehi, 2019; Castelo-Grande et al., 2010). Nonetheless, many of them are not efficient enough and several of them also generate wastes.

One emerging method that has been proposed and studied is based on the application of magnetism and magnetic particles (Chibowski and

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Szczes, 2018; Almeida et al., 2020; Hammad et al., 2020; Tang et al., 2019; Sherlala et al., 2019). In fact, all materials are influenced to a greater or lesser extent by a magnetic field, and some of them are even very prone to it or have higher magnetic properties, such as iron, nickel or cobalt. Magnetism involves the phenomenon of application of forces, either attractive or repulsive, on other materials (Augusto et al., 2007a, b,c,d; Augusto et al., 2004). When micro and nano magnetic particles are used, the effect of the drastic reduction in size offers several advantages and new capabilities to these particles, for example, by presenting a larger effective surface (in contact with water) with respect to the volume occupied by the particle (NANO, 2020; Buzea et al., 2007). These magnetic particles may be applied either bare naked, or with some kind of coating and functionalization (to make them more specific) (Gao, 2019; Baresel et al. 2019; Hassan et al. 2020; Castelo-Grande et al., 2015). The main advantage of applying magnetic particulate systems resides on using their sorption, tagging or reaction properties, while they are retained, recovered or deflected (at the same time or at the end of the experiment) by the application of magnetic fields (Augusto et al. 2002, 2017). These particles have the potential to be fully recovered, regenerated and reused. An important application is to use these particles for environmental remediation specially in systems with continuous contact, like Magnetic Stabilized and Fluidized Beds (Castelo-Grande et al., 2010b,c; 2015) allowing higher throughputs to be used.

Due to the gentle nature of magnetism, the applications of magnetic methods have reached a broad spectrum of scientific areas. This is the case, for example, of biomedical applications (Liu et al., 2007; Luong et al., 2014; Wierucka and Biziuk, 2014; Castelo-Grande et al., 2010; Augusto et al., 2005). Concerning environmental applications they are useful, for example, in the case of (Liu, 2006) microbial detection and monitoring processes (accelerating the process), for contaminant chemical degradation (Fu et al., 2014; Tratnyek and Johnson, 2006; Hernández et al., 2020; Augusto et al., 2020 a, b; Alvaro et al., 2007), or to remove contaminants by sorption (Singh et al., 2011).

Concerning the removal of contaminants by sorption, the last decade has brought an intensive research in this area, leading to the publication of several works (Mehta et al., 2015; Zhang et al., 2020; Rai et al., 2018; Yang et al., 2020; Zhu et al., 2019; Augusto et al., 2019; Estevez et al., 2008; Castelo-Grande et al., 2020b). Nonetheless, scale-up and real applications of magnetic sorbers at industrial levels are very reduced, due mainly to their high-cost, low-throughputs, low existent knowledge about the influence of the main operating and design variables (due to the low number of studies done concerning it), low applicability when many contaminants are present at the same time and compete for the same active center, and the few number of environment-friendly regeneration and reusing methods developed so far (Augusto et al., 2019; Augusto et al., 2020a, 2020b).

We have been trying to direct our research work trying to solve the current issues that magnetic techniques and methods present when research with real samples and applications in real plants are in order (Augusto et al. 2019, 2020b). In the work we develop and present here, we study the applicability of magnetite particles in the sorption of contaminants/nutrients present in real wastewater. Furthermore, we have developed a method that is capable to: use low-cost particles and treat multicontaminated effluents (real wastewaters containing several contaminants/nutrients that are sorbed at the same time by the particles used). As it is not clear in which point of the treatment plant magnetic processes may be introduced with greater efficiency (either alone or in conjugation with other existent and currently applied techniques), an analysis of the process of treatment of wastewater at three different points of the wastewater treatment plant (WWTP) is described.

2. Materials and methods

2.1. Reagents

In this work we used 25–63 μm particles of magnetite, obtained by

crushing natural magnetite and by chemical coprecipitation (Augusto et al., 2019; Augusto et al., 2020 a) - Fig. 1. Characterization of the particles is detailed in Augusto et al. (2019).

Hach-Langue kits were used for analysis of the environmental parameters.

The wastewater samples were always characterized each time they were applied. Section 2.4 shows examples of such characterization.

2.2. Experimental methodology

2.2.1. Sampling locations

In Fig. 2 is represented a schematic diagram of the WWTP of Salamanca, Spain, from where the samples used in this study were extracted. The three extraction points are identified: Sampling Point 1 (Sample 1 – Stream 1) – Entrance; Sampling Point 2 (Sample 2 – Stream 2) – Exit of the first settling tank; Sampling Point 3 (Sample 3 – Stream 3) – Liquid flow exiting the activated sludge centrifugation.

2.2.2. Methodology

2.2.2.1. Sample analysis. All samples were analyzed, at the beginning and after predetermined times, by using Hach-Langue kits and the respective spectrophotometer, as detailed in Table 1 (Augusto et al., 2019), to determine their content in organic matter (COD), nitrates, nitrites, ammonium, total nitrogen, phosphates, chlorides and sulfates. In what concerns their content on heavy metals, the analytical services of the University of Salamanca were used. Before analyzing some of the samples, for example concerning heavy metals, a pre-centrifugation and filtration procedure had to be made in order to avoid possible solids interference.

2.2.2.2. Sorption experiments. 50 mL of the sample are inserted together with 1 g of magnetic particles in an appropriate vessel. The vessel is closed to avoid the possible development of organisms and the uptake of external oxygen and is placed on an orbital shaker, which promotes the contact of the particles with the wastewater sample. After a specified time, the vessel is removed from the shaker and magnetic sedimentation/separation is used to separate the particles from the remaining wastewater sample. This part of the procedure is carried out with precaution in order to get an efficient separation between particles and liquid. The next step is the measurement of the different contaminants for each water sample already treated, and this is done as pointed out in the previous section.

2.3. Instrumentation

For the sample analysis: Spectrophotometer (Hach-Lange DR3900),



Fig. 1. Magnetite particles used.

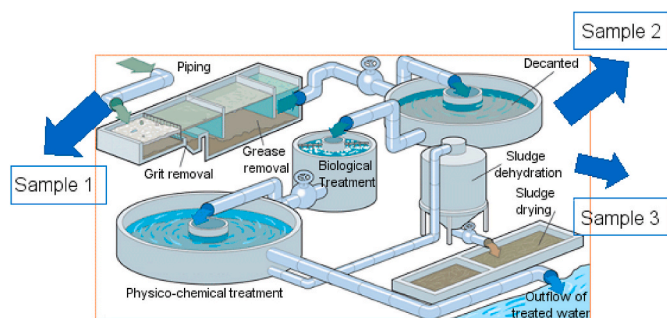


Fig. 2. Schematic diagram of the WWTP of Salamanca, Spain. Extraction points: 1 – Entrance; 2 – Exit of the first settling tank; 3 – Exit of the liquid flow of the activated sludge centrifugation.

Table 1
Environmental parameters, applicable methods and used kits.

Parameter	Method	Kit type
COD	Dichromates (ISO 15705)	LCK 514 100–2000 mg/L O ₂
Nitrates	2,6-Dimetilphenol (EN 38405 D-2)	LCK339 0.23–13.5 mg/L NO ₃ ⁻ N
Nitrites	Diazotization (EN ISO 26777)	LCK341 0.015–0.6 mg/L NO ₂ ⁻ N
Ammonium	Indophenol Blue (ISO 7150–1, DIN 38406 E5-1, UNI 11669:2017)	LCK303 2–47 mg/L NH ₄ ⁺ N
Total nitrogen	Koroleff digestion (peroxodisulfate) and photometric detection with 2,6-dimetilphenol	LCK238 5–40 mg/L NT
Phosphates	Phosphomolybdenum blue (EN ISO 6878) with digestion for total P	LCK349 ortophosphate/ total Phosphate 0.005–1.5 mg/L PO ₄ ³⁻ - P
Chlorides	Iron (III)-thiocyanate with 2 measure ranges using the same kit	LCK311 1–70 mg/L Cl ⁻ and 70–1000 mg/L Cl ⁻
Sulfates	Barium sulfate	LCK153 40–150 mg/L SO ₄ ²⁻

digestor (Hach-Lange LT200); fridge (TEKA Cl3 350).

For sorption experiments: Oven (Argolab G-TCF-120); analytical balance (Sartorius Cubis MSE225S-100-DA); orbital shaker (ELMI Sky Line Shaker DOS-20 M); magnetic separation system (home-made).

General instrumentation: Glass material (Erlenmeyers, beakers, burettes, vials, etc.); Buckner filter device (Nahita 300 mL), among other non-specific instrumentation.

2.4. Initial conditions

Table 2 presents typical wastewater composition at the three sampling sites.

3. Results and discussion

This section is divided into two major sections: the first where we present and discuss the results obtained by the application of magnetic sorption to the wastewater samples collected at each sampling point, and the second where these results are compared and discussed.

3.1. Streams

3.1.1. Sampling point 1

Table 3 presents the results obtained for the application of the sorption process, using the magnetite particles, to samples of wastewater collected at the entrance of the WWTP (Sample 1).

The results presented in Table 3 are depicted in a graphical form, in Supplementary material.

3.1.1.1. Discussion of results for sampling point 1.

Magnetite particles are

Table 2

Typical composition of the wastewater samples collected at the three different sampling sites.

Contaminant	Wastewater origin		
	Sample 1 - Entrance	Sample 2 – Set. Tank	Sample 3 - Centrifuge
Sulfate (mg/L)	44	44.1	45.3
Phosphates (mg/L)	7.8	5.8	83.3
Nitrates (mg/L)	0.775	0.553	4.8
Nitrites (mg/L)	0.077	0.043	9
Total Nitrogen (mg/L)	60.2	50.8	911
COD (mg O ₂ /L)	590	370	556
Detergents (mg/L)	30	12.05	7.05
Cr (ppb)	7.1	7.3	–
Co (ppb)	0.9	0.3	–
Ni (ppb)	5.2	3.8	–
Cu (ppb)	3.3	3.1	–
Zn (ppb)	59.5	47.2	–
As (ppb)	3.8	3.7	–
Cd (ppb)	0	0	–
Hg (ppb)	0.1	0.2	–
Pb (ppb)	0.2	0.4	–

highly effective in reducing detergents (92.3%) and the efficiency reaches more than 90% after 6 h; after only 10 min, detergents content has been reduced into less than 60% of their initial value; all these values prove the great easiness that magnetite particles show on sorbing detergents. Organic matter (COD content) is also very effectively sorbed, and thus its content highly reduced; in fact, starting from a concentration of almost 600 mg O₂/L we are able to reduce it into about 100 mg O₂/L (82.0%) after 16 h (into less than 200 mg O₂/L after only 30 min). Phosphates are moderately sorbed (56.3% efficiency). Total Nitrogen (41.5%) and Nitrates (48.7%) are also moderately sorbed, while nitrites behave erratically; this erratic behaviour is related to sorption and desorption cycles that appear and to transformations between the composed forms of nitrogen (nitrites, nitrates, ammonia, etc.) (Augusto et al., 2019). Sulfates are not efficiently sorbed by magnetite. Concerning heavy metals, magnetite particles are moderately effective on sorbing (and thus reducing the content of) Cr and Zn, and slight effective on reducing the content of As, Co and Ni; in what concerns Cu, Hg and Pb, the behaviour is erratic and the results inconclusive.

3.1.2. Sampling point 2

Table 4 presents the results obtained of the sorption process, using the magnetite particles, for the samples of wastewater collected at the exit of the 1st Settling Tank of the WWTP (Sample 2).

The results presented in Table 4 are depicted in a graphical form, in Supplementary material.

3.1.2.1. Discussion of results for sampling point 2.

Magnetite particles are quite effective in reducing detergents content (74.3%) and organic matter (COD content) - starting from a concentration of almost 400 mg O₂/L we are able to reduce it into about 100 mg O₂/L (72.2%) after 16 h. Phosphates are moderately sorbed (47.4% efficiency). Total nitrogen (33.1%) is also sufficiently sorbed. Nitrates and nitrites behave erratically; this erratic behaviour again has its origin on the sorption-desorption cycles that occur and to the transformations cycle of Nitrogen between its composed species (nitrites, nitrates, ammonia, etc.), as already pointed in Sample 1 analysis. Sulfates are not sorbed by magnetite particles. In what concerns heavy metals, magnetite particles are very effective in sorbing Pb, Co and Zn, sufficiently effective in sorbing Cr and As; and have an erratic behaviour in what concerns Cu, Ni and Hg, leading to inconclusive results in these cases.

3.1.3. Sampling point 3

Table 5 presents the results obtained of the sorption process, using

Table 3
Results of the magnetic sorption process for the different contaminants, for different contact times and the maximum reduction value obtained (%) – Sampling Point 1 - # (°) Identifies elements were the efficiency varies incoherently with time, and (*) means low/not measured values.

Time (min)	Sulfate (mg/L)	Phosphates (mg P/L)	Nitrates (mg N/L)	Nitrites (mg N/L)	Total Nitrogen (mg/L)	COD (mg O ₂ /L)	Detergents (mg/L)	Cr (ppb)	Co (ppb)	Ni (ppb)	Cu (ppb)	Zn (ppb)	As (ppb)	Cd (ppb)	Hg (ppb)	Pb (ppb)
0	44.0	7.80	0.775	0.077	60.2	590	30.0	7.1	0.9	5.2	3.3	59.5	3.8	*	0.1	0.2
5	43.4	4.96	0.499	0.104	48.4	232	23.7	5.10	0.7	4.3	*	13.5	2.6	*	0.1	0.1
10	42.0	4.80	0.398	0.040	43.8	232	11.6	5.60	0.8	4.8	*	15.6	3.0	*	*	*
30	52.4	5.05	0.550	0.060	41.9	198	15.0	6.00	0.9	4.6	*	13.1	2.6	*	0.2	0.2
60	56.9	4.64	0.443	0.043	40.2	183	12.0	1.90	0.8	4.5	2.1	35.0	3.0	*	*	0.1
360	48.5	3.70	0.499	0.023	39.4	135	3.15	4.80	0.8	4.8	2.3	35.0	3.2	*	0.4	*
480	45.6	3.87	0.494	0.057	38.6	124	3.20	3.90	0.8	5.0	4.5	34.9	3.1	*	0.2	*
720	50.0	3.50	0.515	0.120	35.9	125	3.15	3.90	0.8	4.7	2.1	32.5	3.2	*	*	*
960	44.2	3.41	0.406	0.078	35.1	106	2.30	7.90	0.6	5.4	7.2	28.4	3.4	*	0.1	*
% of reduction	4.55 [#]	56.3	48.7	70.1	41.7	82.0	92.3	73.2	33.3	17.3	36.4 [#]	78.0	31.6	*	0	50.0 [#]

Table 4
Results of the magnetic sorption process for the different contaminants/nutrients, for different contact times and the maximum reduction value obtained (%) – Sampling Point 2 - (#) Identifies elements were the efficiency varies with high incoherence with time, and (*) means low/not measured values.

Time (min)	Sulfate (mg/L)	Phosphates (mg P/L)	Nitrates (mg N/L)	Nitrites (mg N/L)	Total Nitrogen (mg/L)	COD (mg O ₂ /L)	Detergents (mg/L)	Cr (ppb)	Co (ppb)	Ni (ppb)	Cu (ppb)	Zn (ppb)	As (ppb)	Cd (ppb)	Hg (ppb)	Pb (ppb)	Time (min)
0	44.1	5.80	0.553	0.043	50.8	370	12.1	7.3	0.3	3.8	3.1	47.2	3.7	0.2	0.4	0	0
5	53.1	4.72	0.814	0.053	48.2	206	13.1	5.5	*	3.3	1.4	17.9	3.3	*	0.2	0.4	5
10	44.3	4.59	0.611	0.092	46.6	217	14.1	6.4	0.1	3.3	0.7	23.8	3.3	*	0.4	10	10
30	53.5	4.47	0.429	0.035	41.9	194	10.1	7.4	0.1	3.1	*	6.30	2.2	*	0.3	0.2	20
60	52.5	4.24	0.421	0.037	40.2	180	6.25	6.2	*	3.2	0.4	31.7	3.2	*	0.1	0.1	30
360	53.1	3.60	0.703	0.091	40.0	144	4.45	12.4	0.1	2.2	*	13.1	2.9	*	0.2	0.1	45
480	50.2	3.12	0.456	0.054	39.2	123	4.40	4.1	0.1	3.4	4.3	44.4	3.1	*	*	*	60
720	46.8	3.19	0.533	0.137	35.1	119	3.40	3.7	*	3.6	3.6	44.3	2.9	*	0.3	0.2	360
960	48.9	3.05	0.435	0.114	34.0	103	3.10	4.9	*	4.5	3.5	38.7	2.8	*	0.3	0.1	720
% of reduction	0	47.4	23.9	18.6 [#]	33.1	72.2	74.3	49.3	66.7	42.1	87.1	72.3	40.5	*	50.0	75.0	960

Table 5

Results of the magnetic sorption process for the different contaminants/nutrients for different application times and the maximum reduction value obtained (%) – Sampling Point 3.

Time (min)	Sulfate (mg/L)	Phosphates (mg P/L)	Nitrates (mg N/L)	Nitrites (mg N/L)	Total Nitrogen (mg/L)	COD (mg O ₂ /L)	Detergents (mg/L)
0	45.3	83.3	4.80	9.00	911	556	7.05
5	48.4	58.4	5.52	2.59	931	442	4.75
10	42.7	55.9	3.47	1.22	877	377	7.05
30	49.1	44.3	3.17	2.02	589	432	6.30
60	46.3	43.5	3.55	2.72	765	428	1.15
360	40.1	50.9	4.71	2.54	767	397	3.00
480	42.9	49.3	4.77	2.39	748	353	2.50
720	43.1	45.8	4.79	2.25	586	353	4.80
960	45.2	39.5	3.33	2.69	494	394	*
% of reduction	11.5	52.6	34.0	86.4	45.8	36.5	83.7

the magnetite particles, for samples collected at the exit of the centrifuge used to dry the activated sludge at the WWTP (sampling point 3).

The results presented in Table 5 are depicted in a graphical form, in Supplementary material.

3.1.3.1. Discussion of results for the sampling point 3. Magnetite particles are very effective in reducing detergents (83.7%), although with some erratic behaviour - the efficiency reaches more than 90% after only 60 min, but then it decreases. Nitrites are also very effectively sorbed, and thus its content highly reduced (86.4%); in fact, we are able to reduce its content to 86.4% after only 10 min. Phosphates (52.6%) and total nitrogen (45.8%) are moderately sorbed. Nitrates and organic matter (measured by COD) are sufficiently sorbed. Sulfates are not sorbed properly by magnetite. No heavy metals analysis was able to be done for this sampling point due to the high level of suspended solids, that impeded the analysis even after centrifugation and filtration steps were performed.

3.2. Comparison

In this section we compare the results obtained by the application of magnetic sorption to the three different sampling streams tested, and also to leachates (the latter results were presented previously in (Augusto et al., 2019)), concerning the maximum removal efficiency. In Figs. 3–10 such comparison is presented for all contaminants/nutrients. In supplementary material the comparison of the variation of the concentration of contaminants/nutrients with time is presented.

Magnetite particles show a great affinity with detergents, and thus the high reduction in its content is common for all the sampling points.

Phosphates are moderately to highly sorbed by the particles for all sampling points, except for the leachates, where the concentration and competition for active centers is very high between contaminants.

Organic matter (measured by COD), is highly sorbed when the initial concentration is not very high. For the cases of high concentrated initial samples (centrifuge and leachates) the reduction efficiency drops to

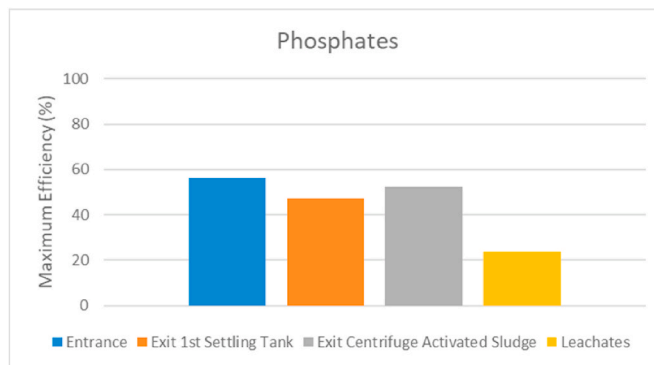


Fig. 4. Maximum removal efficiency for phosphates - Comparison.

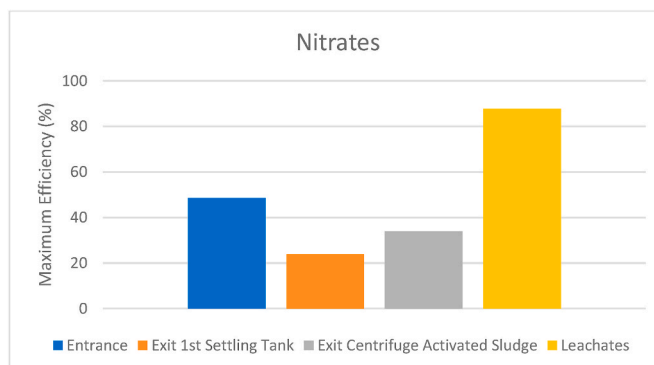


Fig. 5. Maximum removal efficiency for nitrates – Comparison.

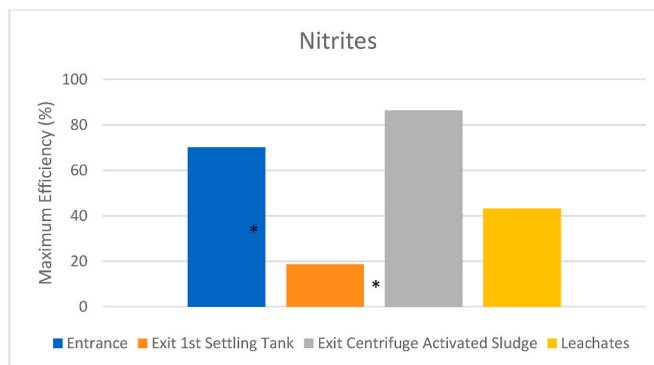


Fig. 6. Maximum removal efficiency for nitrites – Comparison.

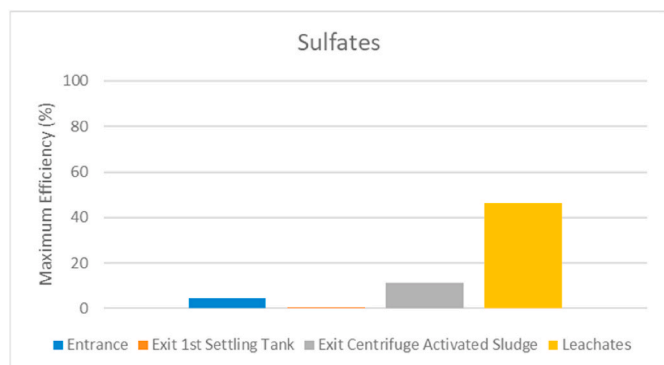


Fig. 3. Maximum removal efficiency for sulfates - Comparison.

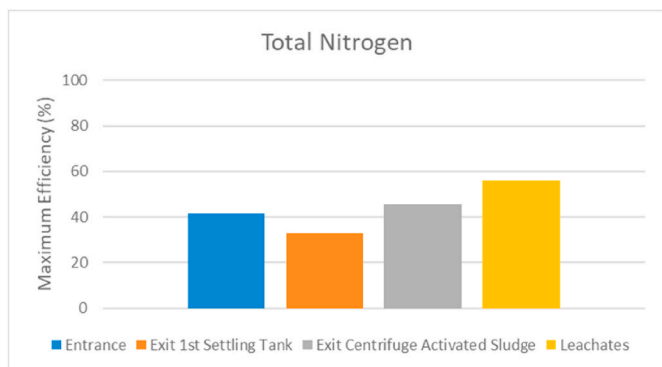


Fig. 7. Maximum removal efficiency for total nitrogen - Comparison.

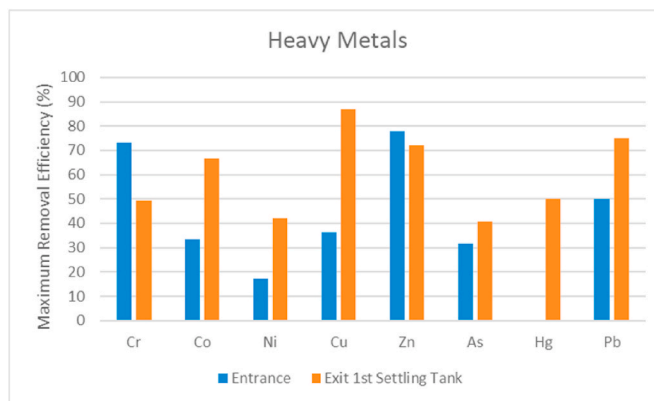


Fig. 10. Maximum removal efficiency of heavy metals - Comparison.

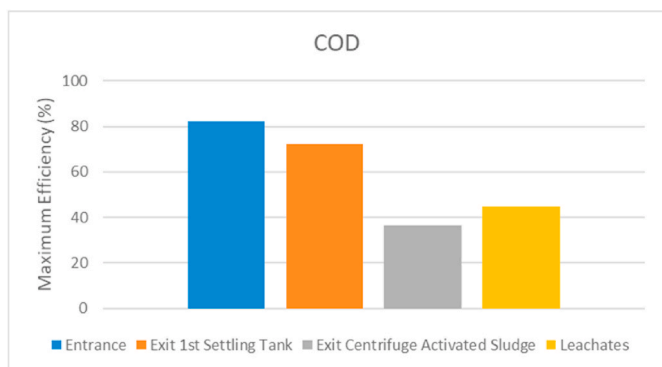


Fig. 8. Maximum removal efficiency organic matter (measured by COD)- Comparison.

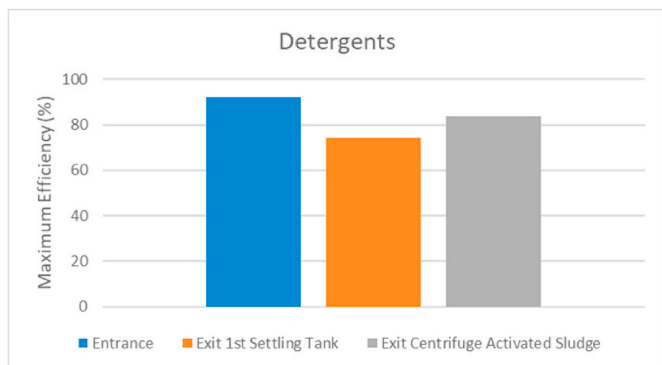


Fig. 9. Maximum removal efficiency for detergents - Comparison.

moderate levels. The reason behind this difference may reside in the saturation of active sorption centers at high organic matter concentrations (high COD).

Sulfates are always badly sorbed, except in the case of leachates where efficiency of removal is moderate.

Total nitrogen is moderately sorbed for all sampling points and leachates. Related content of nitrates is better sorbed in leachates and moderately to sufficiently in the remaining cases. Behaviour of nitrites sorption is usually erratic, except for the case of the centrifuge sampling point (high levels of removal are reached) and leachates (moderate levels of removal). However, these values must be seen as non-constant and with a low repeatability nature in practice, due to the permanent cycle of transformation between nitrogen-based species.

Concerning heavy metals, the analysis was only able to be done on Sample 1 and Sample 2, as Sample 3 and the Leachates showed a high

concentration on suspended solids, that interfered with measurements even after centrifugation and filtration steps. When magnetite is applied to samples coming from sampling point 2, sorption results are always at the same level, or superior, than when applied to samples coming from sampling point 1, except for the case of Cr. This is probably due to the lower competitiveness that exists in the case of samples 2 for active centers as the majority of the other components (organic matter - COD, Total Nitrogen, etc.) present lower concentrations, and the initial heavy metals content is almost the same at both sampling points.

For the interpretation of the results it is important to notice that in this work, and for the first time, as far as we know, sorption experiments are made with real sampling from several collecting points of a WWTP, measuring the sorption of several contaminants at the same time, that compete for the same active sorption centers. This increases a lot the complexity of the system and affects the results. In fact, all media tested is a “live media” that changes with time due to the complex equilibria that form and to the constant changes in several of the components, parameters and concentrations (Augusto et al., 2019). Therefore, for several components there exist a sorption-desorption process caused by the different conditions that keep changing in the media; this includes the effect of the solid content that also sorbs and desorbs substances, parallel to the magnetic sorption (Augusto et al., 2019). Therefore, it is not strange to see in some cases a fluctuation on the contaminants/nutrients concentration, even reaching, in extreme cases, concentrations higher than initial values. This usually happens when elements have a low affinity for being sorbed by the magnetite, while compounds with strong affinity usually do not show this erratic behaviour. In the particular case of nitrates, nitrites and total nitrogen, the erratic behaviour may be intensified due to the constant transformation cycle of nitrogen (which includes also ammonia).

4. Conclusions

Magnetite particles have shown a good efficiency on the removal of the large majority of contaminants under analysis, even in the cases of Heavy Metals. The influence of the presence of all contaminants competing for the same sorption active center at the same time, decreases in some cases the total efficiency (as compared to other literature), but seems to be negligible in other cases (like Detergents and COD). The application of magnetite sorption to the wastewater coming from sampling points were the contaminants content is lower, present equal or higher removal efficiencies, than the ones coming from sampling points where contaminants content is higher, as expected. Based on the analysis presented in this work, Wastewater Treatment Plants may decide the best contaminants to remove by this technique and where is the best place to insert the magnetic sorption in the overall process flow of the Plant.

Credit author statement

Teresa Castelo-Grande: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Supervision, Project administration, Funding acquisition; Paulo A Augusto: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Supervision, Project administration, Funding acquisition; Javier Rico: Investigation; Jorge Marcos: Investigation; Roberto Iglesias: Investigation; Lorenzo Hernández: Formal analysis, Investigation; Domingos Barbosa: Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Project administration, Funding acquisition

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2020.111872>.

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