Use of dewatered alum sludge as main substrate in treatment reed bed receiving agricultural wastewater: Long-term trial

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Abstract

This study aims to explore a novel application of dewatered alum sludge cakes (DASC) as the main medium in a single model reed bed to treat phosphorus-rich animal farm wastewater under "tidal flow" operation on a long term basis. It is expected that the cakes act as the carrier for developing biofilm and also serve as adsorbent to enhance phosphorus (P) immobilization. Results have demonstrated that average removal efficiencies of 73.3±15.9% for COD, 82.9±12.3% for BOD_5, 86.4±6.0% for RP (reactive P), 88.6±7.2% for SRP (soluble reactive P) and 77.6±17.5% for SS can be achieved during the two year's operation. More significantly, the "P-adsorption proportion" by DASC in the reed bed is 42% of the overall P removal. The remaining removal of P may be contributed by the trapping and filtration process of DASC. Therefore, the lifetime of the DASC in reed bed is reasonably longer than that determined from the batch isotherm test.

Keywords: Dewatered alum sludge, phosphorus, reed bed, tidal flow strategy, wastewater treatment

1. Introduction

It has been recognised and generally accepted that urbanization, industrial revolution, and deficient waste disposal practices have left a legacy of polluted sediments in water environment including rivers, estuaries, lakes and seas. Appropriate disposal of such resultant wastes in line with sustainable development remains a great challenge to engineers and scientists who are called upon to develop solutions that are technically, economically, and socially sound. Dewatered alum sludge cakes (DASC) refer to the by-product in water industry from potable water treatment process where aluminium sulphate is employed for the raw water flocculation. In most countries worldwide including Ireland, the DASC is regarded as waste and disposed off in landfill. Although a number of attempts have been made to beneficially reuse such “waste” as raw materials in civil and environmental engineering (Babatunde and Zhao, 2007), up till now, landfill remains the dominant option. However, it is noted that DASC differs significantly from other industrial wastes such as pharmaceutical industrial waste and petroleum industrial waste that may contain high hydrocarbons and toxic substances. DASC is derived from the residual of raw water which contains mainly turbidity, colour and humic substances with no toxic substances in most cases, except probably for the arsenic in some source waters in special circumstances. Two major features of the DASC are: (1) high content of aluminium (29.7 ±13.3% by mass (Babatunde and Zhao, 2007)) and (2) its locally, largely and easily available nature coupled with the fact that it is free of charge. These make it possible to reuse such kind of sludge as a valuable raw/resourceful material in wastewater treatment to enhance adsorption and chemical precipitation processes that remove various pollutants, especially phosphorus (P).

On the other hand, constructed wetlands (or commonly known as reed beds) have been used successfully worldwide as one of the most popular technologies to treat various types of wastewaters (Scholz, 2006; Healy et al., 2007; Vymazal, 2007, Wood et al., 2007; Babatunde et al., 2008). In particular, reed bed system is considered to be efficient and at the same time economically and environmentally attractive and sustainable. In the last several years, extensive studies at University College Dublin, Ireland have been undertaken to develop novel approaches for the purpose of enhancing pollutant removal in reed bed treatment systems. These include "tidal flow" operation strategy and DASC-based reed bed system. These innovative approaches have demonstrated the improved ability of reed bed systems to enhance oxygen transfer and a high immobilization capacity for P removal from the wastewater (Sun et al., 2005; Sun et al., 2006; Babatunde et al., 2007; Zhao et al., 2008).

Tidal flow operation strategy is a batch wise, fill-and-draw type operation (Green et al., 1998; Sun et al., 2005; Sun et al., 2006). When a periodic influent feeding and periodic discharge is applied, the matrices of the reed bed with wastewater enable the bed matrices to be fully submerged during the filling process. This provides maximum media-wastewater contact and avoids the problem of poor wastewater distribution often associated with conventional continuous-flow reed bed systems. Subsequent draining process then allows air to be drawn from the atmosphere into the bed matrices without mechanical input, thereby enhancing aeration and stimulating aerobic biological processes to decompose organic pollutants and ammoniacal-nitrogen in wastewater.

The objective of this study was to examine, in controlled laboratory experiments, the effectiveness of a model reed bed treatment system with DASC as main filter medium treating a P-rich wastewater. This study focused on the treatment efficiency in a long term operation of such kind of treatment reed bed, rather than the feasibility study, which has been reported previously (Zhao et al., 2008). Moreover, a real animal farm yard wastewater was used in this study to mimic actual application conditions. The results of this study provide insight into changes over time in treatment efficiency, BOD_5, P loading rates and the understanding of the operational lifetime regarding the saturation of the DSAC used. It is expected that the current study will serve as the basis to establish a DSAC–based reed bed system (say multi-stage system) for such real animal farm yard wastewater treatment.

2. Materials and Methods

2.1 Materials

The experimental DASC was collected from the mechanical dewatering unit of a water treatment plant located in South-west Dublin, Ireland. The plant uses aluminum sulphate for a reservoir water flocculation at a typical dose of 42-60 mg/l and thus the Al ion is the dominant component in this DASC. Specific characteristics of the DASC that are relevant to its use as a reed bed substrate and as a reed growth medium have been examined in detail (Babatunde et al., 2007). The DASC collected was air-dried and ground and sieved to the particle size less than 2 mm as the main substrate to be used in a laboratory scale reed bed system. The wastewater studied was collected periodically from the secondary holding tank of a local animal farm, which includes about 2000 livestock units of sheep, pigs, cattle and horses. The animal farm effluent is derived from all the activities on the farm and it undergoes some form of primary sedimentation before being pumped to the secondary holding tank. The collected wastewater was allowed for further settlement and the supernatant was stored at 4 ±1 ºC environment.

2.2 Model reed bed set-up and operation

The single model DASC-based reed bed under investigation was set up using a 145 mm (internal diameter) Pyrex column filled firstly with gravel to 10 cm as support layer followed by 35 cm in depth of prepared DASC (2.5 kg) as main substrate. *Phragmites australis* were planted on the top of the bed. The supernatant of the collected wastewater (with or without dilution with tap water) was used as the influent with concentrations of 213 ± 127 mg/l (COD), 110 ± 69 mg/l (BOD_5), 28 ± 15 mg/l (PO_4^{3-}-P), 72 ± 66 mg/l (SS) and 6.8 ± 0.4 (pH). The influent was loaded onto the reed bed via a peristaltic pump from a feed tank at a daily flow rate of 8L, which gives a hydraulic loading rate of 0.5 m^3/m^2.d. The treatment system was operated using the tidal flow strategy for over 730 days. The rhythmical filling and draining generated the tides and this was realised using peristaltic pumps which were controlled by a preset electronic timer.

2.3 Analysis

During the testing period, samples of influent and effluent from the model reed bed were collected and analysed periodically for BOD_5 (using Hach BODTrack apparatus), COD, SS, P (using Hach DR/2400 spectrophotometer) and pH (WTW, pH 325, Germany). The P analysis was based on the reaction of orthophosphate in the samples with molybdate in an acid medium to produce a mixed phosphate/molybdate complex. In particular, the P analysis was done in two parts: (1) Samples were directly reacted (without filtration) with the reagent and analysed to determine the reactive P (RP) and (2) Samples were filtered using a 0.45 μm membrane filter and the filtrate was analysed to determine the soluble reactive P (SRP). In order to monitor biofilm development onto the surface of DASC used in the reed bed, surface examination/observation of the fresh alum sludge particles (referred to as clean sludge) and the sludge particles that were used in the reed bed system (referred to as used sludge) were carried out under a scanning electron microscope (SEM) using a JEOL JSM 6400 Scanning Electron Microscope. In addition, measurement of molecular size distribution (MSD) of dissolved organic substances using a high-pressure size exclusion chromatography (HPSEC) for the samples of influent and effluent at 705th day was carried out for the purpose of providing insight into the pollutants removal inside the reed bed. HPSEC consists of a Waters 1515 isocratic pump, a Waters 2487 UV dual λ detector operated at 254 nm and a PL Aquagel-OH 40 (300×7.5 mm) column. Molecular weight standards were composed of sodium polystyrenesulfonates (35, 18, 8, 5.4, and 1.8K) and acetone.

3. Results

Fig. 1 illustrates the progressive treatment performance of the reed bed. From the results, the initial removal of the carbonaceous substrates averaged 70% for BOD_5 and 56% for COD although this would mainly be attributable to filtration due to the lack of biological activities in the newly setup reed bed matrix. However, with the gradual establishment of a dynamic biological system through intense activities of the reeds and microorganisms in the alum sludge matrix, improved removal of over 80% for both BOD_5 and COD was observed after 100 day's operation. Additionally, enhanced oxygen supply due to the tidal flow operation strategy led to over 90% BOD_5 removal by the 260th day with a short period of exception as shown in Fig. 1. The calculated value of the theoretical oxygen supply rate by the tidal flow operation strategy used in the system is $137.2 g/m^2.d$ (Babatunde, 2007). The average removal of 73.3±15.9% for COD, 82.9±12.3% for BOD_5 was maintained during the entire experimental period. It can be seen from Fig. 1 that the COD removal exhibited some considerable fluctuations, and this may be caused by integrated removal of enhanced adsorption, filtration and biological degradation of pollutants in terms of COD. Relatively, BOD_5 removal was solely from the biological degradation. The relative low removal of BOD_5 and COD of the reed bed in the later stage of the operation period can be adduced to operational reasons as the laboratory was relocated during that period and this affected the routine operation.

[Fig. 1 here]

Fig. 2 provides evidence of a good correlation between BOD_5 loading and its removal (in g/m^2.d). Increased BOD_5 loading resulted in higher BOD_5 removal in terms of g/m^2.d of the reed bed, thus indicating the intensive activities of the microorganisms inside the reed bed. Data of pH monitoring were not presented since there were no specific characteristics related to the pH except for the trend of effluent pH being lower than that of influent due to the hydrolysis of DASC in the bed (Yang et al., 2006).

[Fig. 2 here]

Removals of both RP and SRP were significant in the reed bed throughout the experiments. The average removal efficiencies of 86.4 ±6.0 % for RP and 88.6 ±7.2 % for SRP, respectively, were achieved under the P-loading of 13.5 ±7.2 g-PO_4^{3-}/m^2 d for RP and 8.2 ±3.2 g-PO_4^{3-}/m^2 d for SRP, respectively. Mass balance estimation shows that about 35.8 mg-PO_4^{3-}/g-sludge was adsorbed by DASC in the reed bed during the operation. Interestingly, the DASC in the bed has not been saturated and a P removal efficiency of 70 % was still obtained, although an obvious decline of removal efficiency is observed from Fig. 3. In-depth study has revealed that the main P-adsorption mechanism is ligand exchange reactions (Yang et al., 2006). This highlights the obvious advantage of the use of the DASC in enhancing P removal in the system.

[Fig. 3 here]

Behaviour of SS removal in the reed bed is shown in Fig. 4. Although an average SS removal of 77.6 ±17.5% was obtained, and the DASC has been demonstrated as a potential medium in reed bed for trapping and filtering SS, it has to be stressed that there was ample fluctuation in the SS removal, and therefore, necessary care should be taken to further study this issue.

During the reed bed operation period, serious problems, such as ponding, did not occur, but concerns of bed clogging in relation to proper arrangement of DASC in the reed and the pre-treatment of the raw wastewater still warrant further investigation.

[Fig. 4 here]

4. Discussion

4.1 BOD_5 and COD removal

The goal of this study lies in the long term examination of DASC which is expected to act as a carrier for developing biofilm and also serve as adsorbent to enhance P immobilization from a real wastewater treatment. As a biofilm carrier, the DASC has successfully demonstrated its role. The significant removals of both BOD_5 and COD (see Fig. 1) have provided the vital evidence of biofilm development and intensive biological activities inside the reed bed. In addition, both the measurements of MSD and SEM have demonstrated the development of biofilm in the reed bed and the role of DASC as the biofilm carrier although the MSD data and the SEM image are not shown. MSD measurement provides the evidence which differs from COD and BOD_5 measurement to illustrate the molecular size distribution of dissolved organic pollutants before and after the reed bed treatment. The result support that the reed bed degraded/broke the organics from large molecular sizes into smaller ones via mainly biological activities inside the bed medium. SEM observation of clear and used sludge samples showed an obvious difference in morphology. The used sludge was fully covered by thick slimes, which showed fluffy and transparent characteristics. While clear sludge showed a crystal lattice structure surface. The striking change of the surface characteristics may provide the evidence of biofilm development onto the sludge surface, indicating the growth and considerable activities of the micoorganisms although the SEM here is a qualitative tool to describe the biofilm formation.

The influent and effluent of COD and BOD_5 during the testing period are illustrated in Fig. 5. Continuous reduction of COD and specially BOD_5 provides a definite indication of biological degradation of the organics from the wastewater. It should be noted that except for the biological process inside the reed bed, a multi-function of the bed which involves adsorption, filtration, precipitation processes is existed in parallelism. The removal of COD and BOD_5 is the results of the integrated functions inside the bed. Therefore, it is suggested that the monitoring of such DASC-based reed bed treatment system should use both COD and BOD_5 as benchmarks to reveal the degree of treatment efficiency, rather than using one (either COD or BOD_5) as index which has been frequently employed in wastewater treatment practice. In addition, considering the much strong animal farm wastewater in practice, it should be pointed out that a single reed bed is impossible to satisfy with the treatment standard. A multi-stage reed bed system is thus suggested and further tests should be conducted. However, it is out of the scope of the current study.

[Fig. 5 here]

4.2 Phosphorus removal and the lifetime of DASC in reed bed

During the monitoring period, the DASC exhibited good ability as adsorbent to achieve high P immobilization (see Fig. 3). Both RP and SRP were monitored in this study and there was a good correlation between them as illustrated in Fig. 6, suggesting that either RP or SRP can be used as sole index for P removal in practice for such kind of reed bed treatment system. Fig. 7 shows the RP and SRP removal (in $g/m^2.d$) with the RP loading rate applied to the reed bed studied. It is noted that although the RP loading rates varied 6.2-32.2 g PO_4^{3-}-$P/m^2.d$, the reed bed system could remove P efficiently even as the loading rates increased, as indicated by a linear relationship between P loading and removal. This suggests a good potential and high P adsorption ability of the DASC. From the linear regression analysis (see Fig. 7), it is interesting to note that the slopes of the two linear regressions are obviously different. If the removal of SRP refers mainly to the adsorption and biological assimilation of the micoorganisms growth, then the gap between the two lines could be considered as an indication of the sludge's 'other' functions of physical trapping, filtration and sedimentation in the bed. Obviously, adsorption plays 42% (0.38/0.90) role in P removal based on the data of the current study. Thus it is reasonable to state that the maximum adsorption capacity obtained experimentally from batch isotherms tests cannot be used as a guide to the actual lifetime of such reed bed in real operation, although attempts have been made to do so in literature with little or no success (Arias and Brix, 2005; Dong et al., 2005; Park and Polprasert, 2008; Zhao et al., 2008). It is understood that the batch isotherm tests is solely designed for the determination of adsorption, eliminating filtration, sedimentation and biological functions of the material tested. Therefore, the lifetime of the DASC regarding its P adsorption saturation in reed bed could be much longer than that determined based on batch isotherm test.

[Fig. 6 here]

[Fig. 7 here]

4.3 The proactive nature of the DASC-based reed bed treatment system

The success of this innovative reed bed has demonstrated the beneficial and sustainable reuse of DASC (a hitherto designated waste product) as a raw material in wastewater treatment engineering. Regarding its large scale application in practice, some important issues such as possible clogging of the bed and release of some substances from the DASC still need to be studied and properly addressed. Unlike other waste by-products derived from various industries, DASC remains an inescapable by-product from the current drinking water treatment process/technology. The continuous supply of such by-product is certain for now in water treatment industry and it is free of charge. For example, only in Ireland, a double-fold increase in alum sludge generation has been forecast by the end of the next decade, from a current estimate of 15,000–18,000 t/Pa of the DASC (Yang et al., 2006). Application of DASC in reed bed system will further reduce the capital invest of the reed bed treatment system and make such treatment technology more attractive. From a previous study by Zhao et al. (2007) using the same DASC as used in the current study, the P adsorption capacity of the DASC determined using batch isotherm tests would imply that the lifetime of the DASC used in the reed bed for domestic wastewater treatment could be 4-17 years based on the estimate described by Xu et al., (2006). By considering the "adsorption proportion" of 42% in overall P removal derived from the current study, the lifetime of the DASC can be 9-40 years. Even in the case of high P effluent of animal farm wastewater treatment tested in this study, the lifetime of DASC can be estimated as 2.5-3.7 years. In most cases of the practical use of reed bed system, a multi-stage treatment system is usually applied. Therefore, the lifetime is reasonably expected to be longer. However, as pointed out by Xu et al., (2006), such an estimate of the lifetime of a reed bed should only be used as a guide because the P sorption capacity is influenced by many factors. The most examined factors are particle size, pH, initial P concentration and temperature etc (Ippolito et al., 2003; Dayton and Basta, 2005; Yang et al., 2006; Zhao et al., 2007; Korkusuz et al., 2007). For this reason, the P adsorption capacity could vary in magnitude (Korkusuz et al., 2007). Nevertheless, DASC-based reed bed has clearly demonstrated its advantage and is expected to be a promising idea in constructed wetland technology for wastewater treatment.

It should be pointed out that two major issues related to the potential application of such DASC-based reed bed have not been fully studied. One is the clogging tendency of such sludge reed bed since the SS trapped in the bed increases the average water retention time and thus reduces the effective area available for water flow. Reduced hydraulic conductivity and the infiltration rate lead to ponding, which represents a serious threat to the normal operation for the treatment function. Although obvious bed clogging did not occur in this study, the scatter of the SS removal efficiency (see Fig. 4) indicates that SS removal should be specially studied to avoid bed clogging. One suggestion could be made that the use of the DASC-based reed bed as a stand-alone treatment system would not be recommended and pre-treatment to remove SS would be necessary and vital to result in the successful operation. The other issue is the possible release of some substances from DASC including polymer residual to the effluent from DASC since polymer is normally added as sludge conditioner to enhance its dewaterability. Polymer residual in the DASC and its potential release when the sludge is reused should be studied since the toxicity of degraded polymer in the environment remains an unknown health risk from the long term point of view (Majam and Thompson, 2006).

5. Conclusions

Two years experiment was conducted in a model DASC-based reed bed aimed to exploring the feasibility and effectiveness regarding P-rich wastewater treatment. The novel use of DASC as the main reed bed substrate is a promising idea in constructed wetland technology. The average removal efficiencies of 73.3 ±15.9% for COD, 82.9 ±12.3% for BOD_5, 86.4 ±6.0% for RP, 88.6 ±7.2% for SRP and 77.6 ±17.5% for SS were achieved during the entire operation period. Results revealed that the “P-adsorption proportion” by DASC in the reed bed is 42% of overall P removal. Therefore, the lifetime of the DASC is reasonably longer than that determined from batch isotherm test. Regarding the large scale application of such reed bed system, further studies on bed clogging tendency and possible release of some substances inside the DASC are recommended.

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Figure caption:

Figure 1. Removal efficiencies of COD and BOD_5 throughout the experimental period

Figure 2. Correlation between BOD_5 loading and its removal

Figure 3. Removal efficiencies of RP and SRP throughout the experimental period

Figure 4. Removal efficiencies of SS throughout the experimental period

Figure 5. Variations of COD and BOD_5 throughout the experimental period

Figure 6. Correlation between RP and SRP removals

Figure 7. Correlation between RP loading and the removals ($g/m^2.d$) of RP and SRP

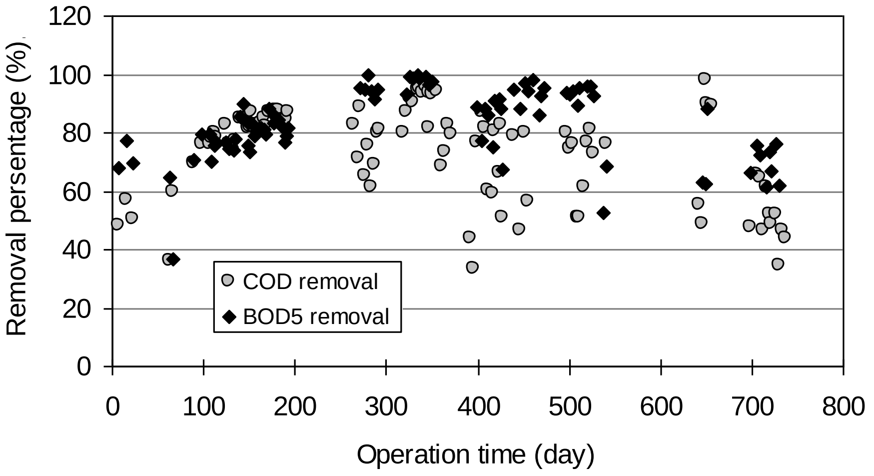
Removal persentage (%)
0
20
40
60
80
100
120
COD removal
BOD5 removal
0
100
200
300
400
500
600
700
800
Operation time (day)

Fig. 1

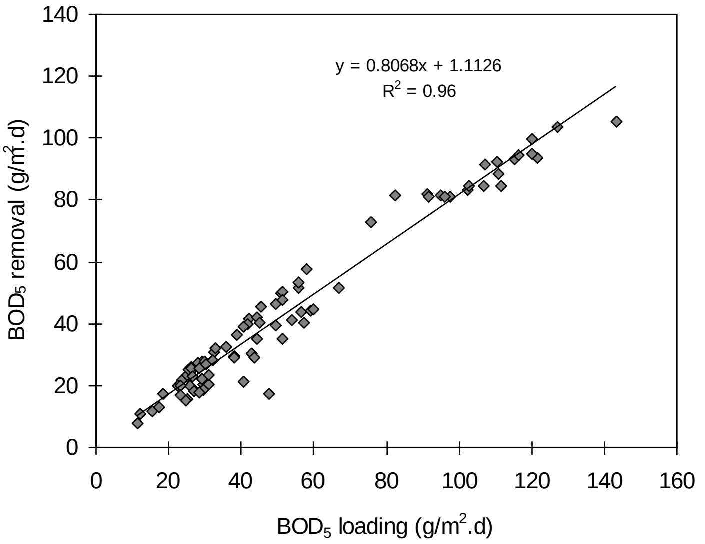
y = 0.8068x + 1.1126
R^2 = 0.96
BOD$_5$ removal (g/m^2.d)
0
20
40
60
80
100
120
140
0
20
40
60
80
100
120
140
160
BOD$_5$ loading (g/m^2.d)

Fig. 2

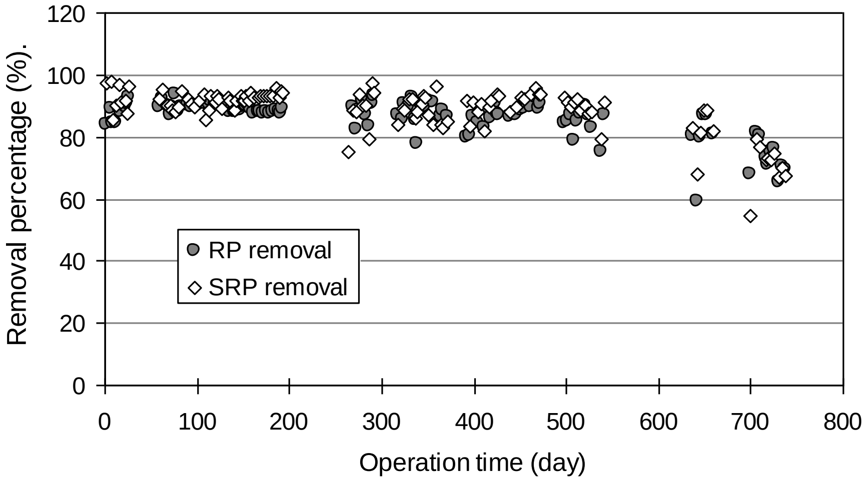
Removal percentage (%).
120
100
80
60
40
20
0
0
100
200
300
400
500
600
700
800
Operation time (day)
● RP removal
◇ SRP removal

Fig. 3

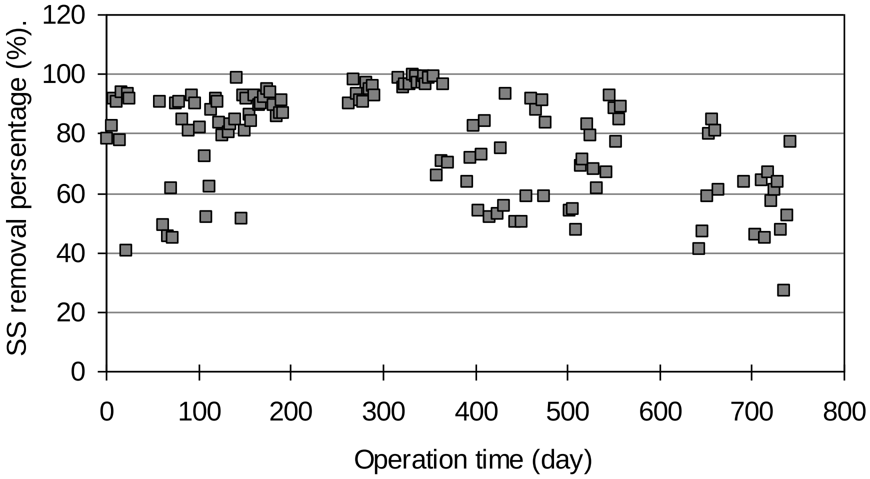
SS removal persentage (%).
120
100
80
60
40
20
0
0
100
200
300
400
500
600
700
800
Operation time (day)

Fig. 4

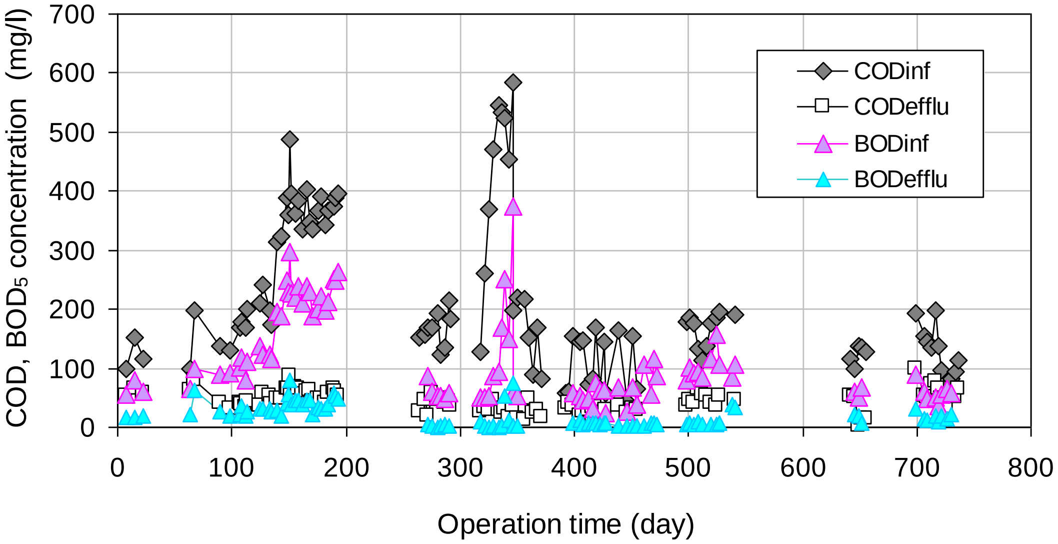
COD, BOD₅ concentration (mg/l)
0
100
200
300
400
500
600
700
0
100
200
300
400
500
600
700
800
Operation time (day)
CODinf
CODefflu
BODinf
BODefflu

Fig. 5

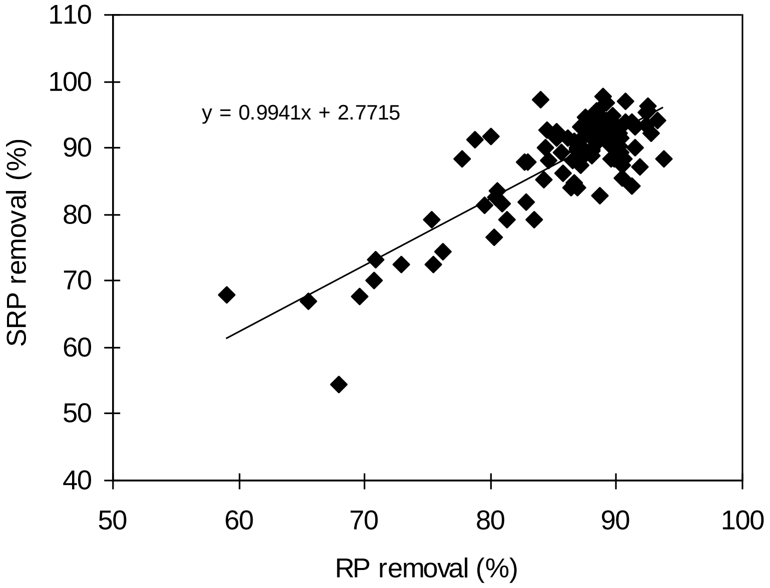
y = 0.9941x + 2.7715
SRP removal (%)
40
50
60
70
80
90
100
110
50
60
70
80
90
100
RP removal (%)

Fig. 6

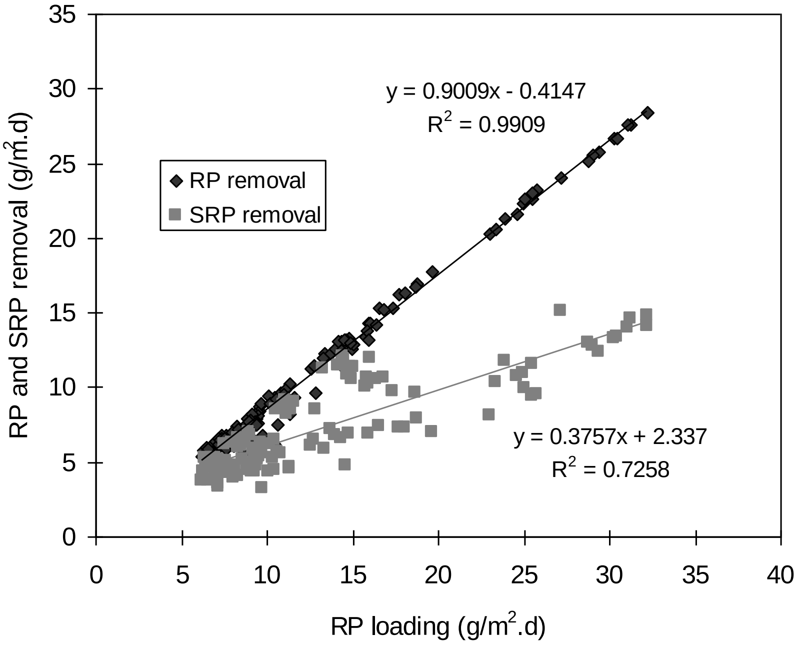
y = 0.9009x - 0.4147
R^2 = 0.9909
RP removal
SRP removal
y = 0.3757x + 2.337
R^2 = 0.7258
RP and SRP removal (g/m^2.d)
RP loading (g/m^2.d)
0
5
10
15
20
25
30
35
40

Fig. 7