



# Performance evaluation of partially saturated vertical-flow constructed wetland with trickling filter and chemical precipitation for domestic and winery wastewaters treatment

Boram Kim, M Gautier, S Prost-Boucle, Pascal Molle, Philippe Michel, R Gourdon

## ► To cite this version:

Boram Kim, M Gautier, S Prost-Boucle, Pascal Molle, Philippe Michel, et al.. Performance evaluation of partially saturated vertical-flow constructed wetland with trickling filter and chemical precipitation for domestic and winery wastewaters treatment. *Ecological Engineering*, 2014, 71, pp.41-47. 10.1016/j.ecoleng.2014.07.045 . hal-01078368

**HAL Id: hal-01078368**

**<https://hal.science/hal-01078368>**

Submitted on 28 Oct 2014

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

**Performance evaluation of partially saturated vertical-flow constructed wetland with  
trickling filter and chemical precipitation for domestic and winery wastewaters  
treatment**

B. Kim<sup>a,b</sup>, M. Gautier<sup>a,\*</sup>, S. Prost-Boucle<sup>c</sup>, P. Molle<sup>c</sup>, P. Michel<sup>b</sup> and R. Gourdon<sup>a</sup>

<sup>a</sup> *University of Lyon, INSA Lyon, LGCIE, Team DEEP, 20 av. Albert Einstein, 69621  
Villeurbanne cedex, France*

<sup>b</sup> *SCIRPE, 5 Allée Alban Vistel, 69110 Sainte-Foy-Lès-Lyon, France*

<sup>c</sup> *IRSTEA, UR MALY, 5 rue de la Doua, 69100 Villeurbanne, France*

\* Corresponding author.

E-mail address: mathieu.gautier@insa-lyon.fr

Address: LGCIE Site Carnot

20 av. A. Einstein

F-69621 Villeurbanne Cedex

Tel.: +33(0)472438348

## Abstract

The use of Vertical Flow Constructed Wetlands (VFCW) is growing rapidly in Europe for domestic wastewater treatment in small communities. In order to improve denitrification and dephosphatation as compared to classical VFCW, the Azoé-NP<sup>®</sup> process has been developed. The process line consists of: a biological aerobic trickling filter as a primary treatment stage, ferric chloride (FeCl<sub>3</sub>) addition for phosphorus (P) treatment and two stages of partially saturated VFCW. A municipal wastewater treatment plant using Azoé-NP<sup>®</sup> process has been monitored during eight years through 44 campaigns of 24h time-proportional inlet-outlet sampling followed by analyses of TSS, BOD<sub>5</sub>, COD, TKN, NO<sub>3</sub>-N and TP concentrations. The results revealed good performances of the overall treatment. To better characterize the performance of each treatment step, five additional 24h monitoring campaigns were performed with samples taken from four different points along the treatment line. Results showed a good performance in dissolved carbon removal and nitrification by the trickling filter. The main part of the treatment was found to be done by filtration throughout the first filtration stage. Nitrate removal was achieved principally at the second filtration stage. Phosphorus migration through the first stage and its slight retention at the second stage was observed.

Keywords: vertical flow constructed wetland; trickling filter; winery wastewater; wastewater treatment; nitrification-denitrification; phosphorus retention

## 1. Introduction

The use of Vertical Flow Constructed Wetlands (VFCW) has been growing rapidly in Europe over the last decade for domestic wastewater treatment in small towns (Abou-Elela and Hellal, 2012; Molle et al., 2006; Torrens et al., 2009). The process has proven its efficiency in the treatment of organic matter and nitrogen from raw municipal wastewaters or after primary treatment (Brix and Arias, 2005; Kadlec et al., 2000). However, there still remain some fields for improvement, not only for total nitrogen removal by nitrification / denitrification but also for phosphorus retention (Brix et al., 2001). Indeed, the release of nitrates and phosphorus into sensitive aquatic ecosystems may promote eutrophication (Schindler, 1977; Tiessen, 2008), and therefore the concentration in the treated effluents must satisfy increasingly low regulatory limits.

The Azoé-NP<sup>®</sup> process has been developed by the French company SCIRPE to improve denitrification and P removal (EP1857419A1; PCT/EP2012/058119). The process line consists of: (i) a screening (3 mm mesh) ii) a biological aerobic trickling filter as a first biological treatment step (mainly for organic carbon removal), (ii) ferric chloride (FeCl<sub>3</sub>) addition for phosphorus precipitation and (iii) two stages of partially saturated VFCW. Since the organic load is partly transformed and removed by the trickling filter, the required surface of VFCW is reduced in the Azoé-NP<sup>®</sup> system to a maximum of 1.5 m<sup>2</sup> per Population-Equivalent (PE) as compared to 2 m<sup>2</sup> in the general operational recommendations in France for classical two-stage VFCWs (Molle et al., 2005). In addition, the trickling filter is passively aerated by its bottom part and therefore provides good condition to initiate nitrification.

The main treatment achievement occurs through two successive stages of partially saturated VFCW. Each filtration stage consists of two different zones: the upper zone which is not saturated, and the lower one which is saturated. These conditions allow good total nitrogen removal through sequential nitrification / denitrification. The depth of the saturated zone can be adjusted to optimize efficiency.

With respect to phosphorus removal, different physico-chemical processes are used in constructed wetland systems. Most subsurface flow (SSF) CW systems use filter media with high phosphorus sorption capacity (Molle et al., 2011; Vohla et al., 2011) but can be a problem when looking for a cheap and efficient solution on a long term. In the Azoé-NP<sup>®</sup> process however, phosphorus is precipitated by injection of FeCl<sub>3</sub> at the outflow of the trickling filter and removed by filtration through the partially saturated VFCW as highlighted by Kim et al (2013). The deposits, which progressively accumulate on the surface of VFCW, should be finally withdrawn every 10-15 years.

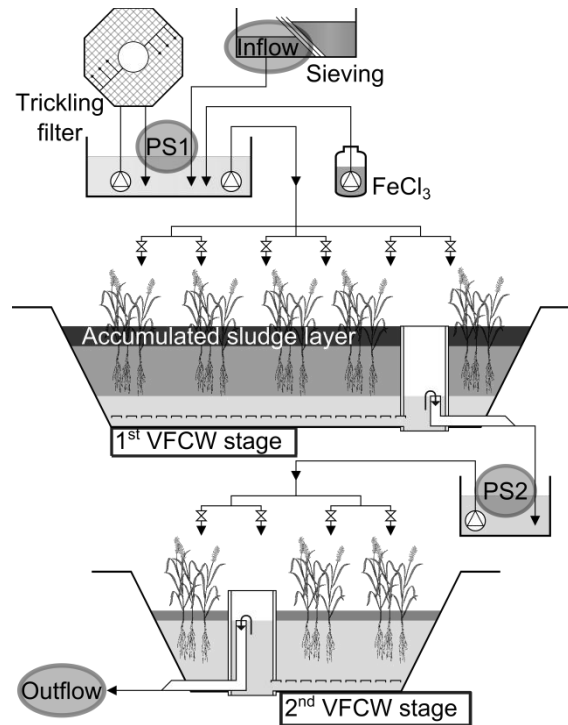
As the process is relatively new in France (10 years), the objectives of this study were to (1) estimate the overall efficiency of the Azoé-NP<sup>®</sup> treatment plant through a series of inflow/outflow 24h time-proportional sampling analyses done over eight years, (2) determine the respective efficiency of

each individual step of the process, based on specific surveys at each treatment step and (3) focus on nitrogen removal and phosphorus retention.

## 2. Materials and Methods

### 2.1. Description of the plant selected for the study

The study was conducted by monitoring a municipal wastewater treatment plant (WWTP) located in Vercia (Jura, France) between 2004 and 2012. This unit, schematically represented in Fig.1, is the first Azoé-NP<sup>®</sup> full-scale plant and has been in operation since 2004. It treats mostly domestic wastewater, but also receives wine-production effluent in autumn. Its maximum capacity is 1,100 PE. It was designed for a nominal volumetric flow of 70 m<sup>3</sup>.day<sup>-1</sup> and an organic load of 56 kg COD.day<sup>-1</sup>. During grape harvesting period however, the hydraulic and organic loads increased to 77 m<sup>3</sup>.day<sup>-1</sup> and 122 kg COD.day<sup>-1</sup> respectively for about one month.



**Fig. 1.** Simplified diagram of the Vercia WWPT showing the four sampling points on the process line: [Inflow] raw wastewater sieved to 3 mm, [PS1] effluent of the trickling filter with ferric chloride injection, [PS2] effluent of the 1<sup>st</sup> partially saturated VFCW and [Outflow] effluent of the 2<sup>nd</sup> partially saturated VFCW. PS stands for pumping station.

The trickling filter has a volume of 30 m<sup>3</sup> fully packed with corrugated plastic sheet packing (ordered) developing a specific surface of 100 m<sup>2</sup>.m<sup>3</sup>. Each VFCW has a depth of 0.8 m and a total surface of 600 m<sup>2</sup>. The filtration layer is made of a layer of fine gravels (d<sub>10</sub>: 1.6 mm; d<sub>60</sub>: 3.6 mm) in the first filter and sand (d<sub>10</sub>: 0.16 mm; d<sub>60</sub>: 1.3 mm) in the second one. The water-saturation level in each filter is adjustable by a control pipe in the outlet drainage system. Over the period of

investigation of this study, the level was maintained around half of the filter depth for the first stage and between 0.6-0.75 m from the bottom of the second filter. The first stage is divided into three hydraulically independent identical units of 200 m<sup>2</sup>, which are fed alternately to favour mineralization of sludge. Their bottom drainage systems communicate one with each other. The second stage is separated into two identical units of 300 m<sup>2</sup>. Each unit is fed over a period of 7 days and then rested for 14 days (for the first stage) or 7 days (for the second stage). Phosphorus removal is realized by injection of about 0.140 L of a 574 g/L ferric chloride solution per m<sup>3</sup> of wastewater, corresponding to a total iron concentration of 0.5 mmol.L<sup>-1</sup>.

## 2.2. Description of field monitoring campaigns

The plant was monitored since the date of its implantation in 2004 until year 2012. Different types of sampling campaigns were conducted as detailed below.

### 2.2.1. *Regular campaigns*

The regular campaigns were conducted to evaluate the overall inlet-outlet performance of the plant. A 24h time-proportional inflow/outflow sampling method was followed. A total of 44 regular campaigns (four to six campaigns per year) were made, including during autumn when wine-production effluents were treated by the plant.

### 2.2.2. *Specific campaigns*

In addition to the regular monitoring campaigns, a total of 5 specific 24 h sampling campaigns were conducted where sampling was done at four different points of the treatment line to determine the efficiencies of each step of the treatment line. Three of them were done in 2007 (one in winter and two in autumn during wine production period) following a time-proportional sampling method. The last two specific campaigns were performed in 2011 and 2012 during wine production period, following a 24h flow-proportional sampling method to provide additional data to the study. The positions of the sampling points are shown in Fig. 1: (1) raw wastewater [Inflow], (2) effluent from the trickling filter after ferric chloride injection in 1<sup>st</sup> pumping station [PS1], (3) effluent from the first stage of partially saturated VFCW in 2<sup>nd</sup> pumping station [PS2] and (4) treated water at the outlet of the second filter [Outflow].

## 2.3. Analytical procedures

Refrigerated auto-samplers were used in all campaigns to keep the samples at a temperature around 4°C until they were transported to the laboratory within 24h after sampling. The following parameters were analysed following French standard methods (AFNOR, 2005): Total Suspended Solids (TSS), five-day Biochemical Oxygen Demand (BOD<sub>5</sub>), Chemical Oxygen Demand (COD),

Total Kjeldahl Nitrogen (TKN), Nitrate (NO<sub>3</sub>-N), Total Nitrogen (TN) and Total Phosphorus (TP) concentrations. The pH was measured using a portable instrument (ProfiLine pH 3110, WTW).

In the last two specific campaigns, samples were divided into two aliquots. The first aliquot was treated as indicated above, and the second aliquot of each sample was filtered through 0.45 µm syringe filter (Sartorius Minisart) to analyze for dissolved ions. Dissolved orthophosphate was determined by the colorimetric molybdenum blue method (Murphy and Riley, 1962). Total soluble iron was determined by ICP-AES (Ultima 2; Horiba JobinYvon SAS). Chloride, nitrate and sulphate concentrations were determined by ion chromatography using a Dionex Ion Pac® AS17C column preceded by an AG17 guard-column.

#### 2.4. Exploitation of data

The performance of the treatment with respect to each parameter was calculated in the form of a 24h “(input – output) / input” concentration of the considered parameter expressed in percentage. The 95% confidence interval was determined by  $\pm$  standard error of the mean (SEM), which was calculated as the standard deviation (SD) divided by the square root of the number of samples. One-way analysis of variance (ANOVA) with a probability value of  $p < 0.05$  was used to evaluate the effect of hydraulic and organic loads on the treatment performance for each parameter.

### 3. **Results and Discussion**

#### 3.1. Regular campaigns – overall efficiency

The average concentrations of the selected parameters measured in the influent and effluent of the Vercia plant during the whole set of campaigns (altogether 44 campaigns) are gathered in Table 1. The average conditions of operation in the considered campaigns were a hydraulic load (HL) of 82 m<sup>3</sup>.d<sup>-1</sup> (SD: 74) and organic load (OL) of 56 kg COD.d<sup>-1</sup> (SD: 42).

**Table 1.** Average values  $\pm$  SEM (number of campaigns) for selected parameters in inflow and outflow of the Vercia WWTP over eight years of operation (2004-2012)

Variable	Unit	Inflow	Outflow	Percent removal (%)
pH	-	7.86 $\pm$ 0.28 (39)	7.95 $\pm$ 0.19 (39)	Non applicable
TSS	mg.L <sup>-1</sup>	450 $\pm$ 104 (44)	3.4 $\pm$ 0.7 (44)	98.8 $\pm$ 0.5 (39)
BOD <sub>5</sub>	mg.L <sup>-1</sup>	286 $\pm$ 65 (43)	3.2 $\pm$ 0.6 (43)	97.9 $\pm$ 1.1 (38)
COD	mg.L <sup>-1</sup>	794 $\pm$ 179 (44)	25 $\pm$ 3 (44)	94.4 $\pm$ 2.5 (39)
TP	mg.L <sup>-1</sup>	9.5 $\pm$ 1.6 (40)	2.8 $\pm$ 0.4 (40)	59.6 $\pm$ 12 (35)
TN	mg.L <sup>-1</sup>	71 $\pm$ 10 (40)	17 $\pm$ 3 (39)	70.9 $\pm$ 6.2 (35)
TKN	mg.L <sup>-1</sup>	70 $\pm$ 9 (42)	1.6 $\pm$ 0.3 (43)	97.2 $\pm$ 1.0 (37)
NO <sub>3</sub> -N	mg.L <sup>-1</sup>	1.4 $\pm$ 1.4 (40)	15 $\pm$ 3 (40)	Non applicable

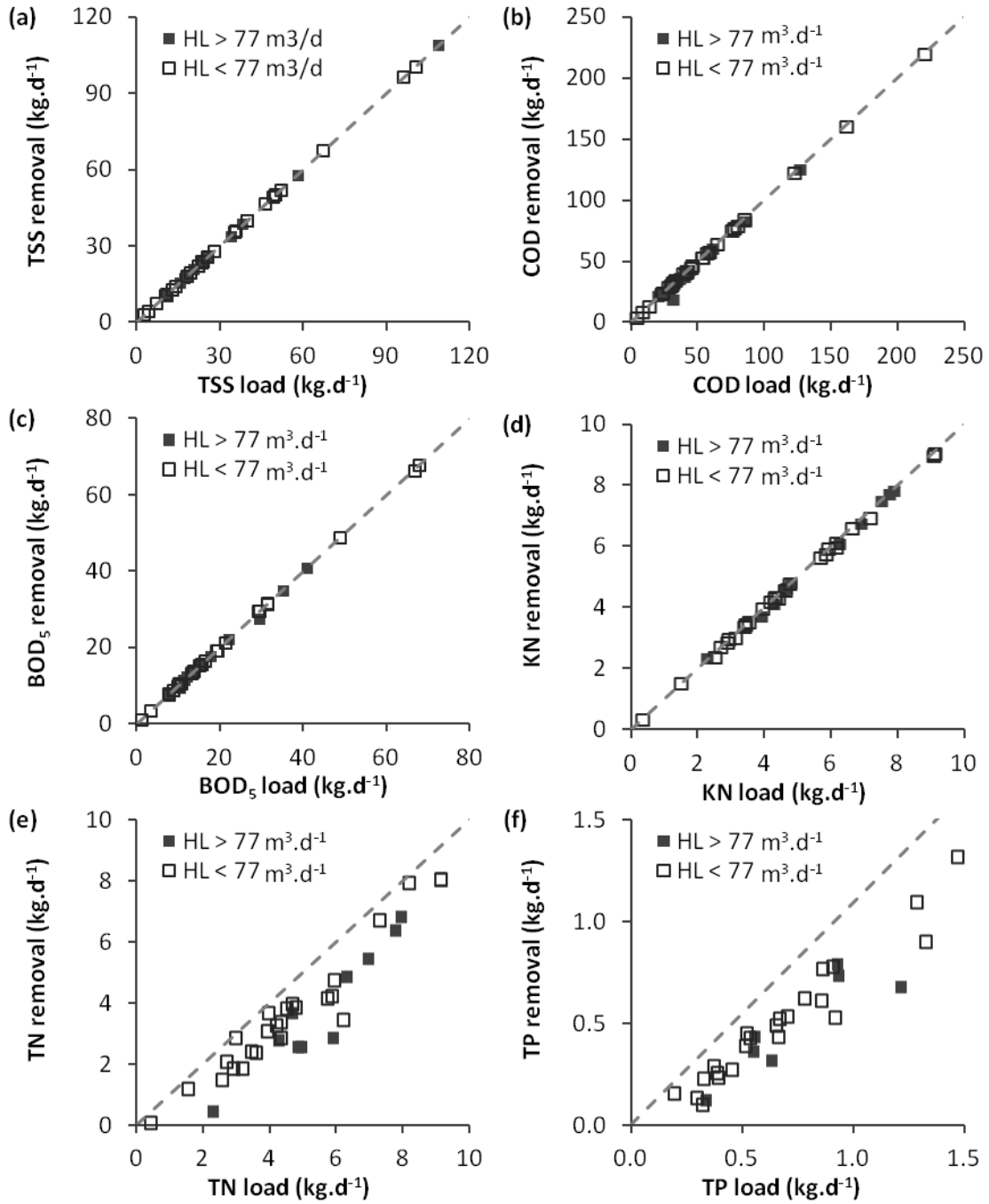
The results given in Table 1 revealed a very good efficiency in carbon and Kjeldhal nitrogen removals. The SS removal efficiency (99%) was excellent as well as BOD<sub>5</sub> (98%) and COD (94%) removals with satisfactory outflow concentrations. TKN percent removal (97%) ranges in the upper values of the performances reported for two-stage classical French VFCW monitored with the same methodologies (average of 85%; Molle et al., 2005). This good performance may probably be explained by the activity of the biological trickling filter which is known to be efficient for nitrification (Almstrand et al., 2011; Persson et al., 2002).

Furthermore, the concentration of total nitrogen (TN) in the treated effluent (17 mg.L<sup>-1</sup>) and its percent removal (71%) indicated that the plant was efficient in total nitrogen removal. This efficiency was probably related to the denitrification phenomenon favoured in the saturated zone of VFCW stages. However, differences between TN and TKN outlet concentrations revealed that denitrification was not 100 % efficient. It would be therefore of high interest to analyse further the parameters influencing TN removal performances.

Regarding phosphorus, the percent removal was slightly lower than for the other parameters, and the outflow concentration was more variable with an average of 2.8 mg.L<sup>-1</sup> (SD: 1.6). Regarding the dosage of iron-based coagulants, a Fe:P molar ratio higher than 1.5:1 is usually recommended (Caravelli et al., 2010; Szabó et al., 2008; Thistleton et al., 2002). In Azoé-NP<sup>®</sup> process, the ferric chloride injection is proportional to flow therefore, the Fe:P molar ratio was below 1.5:1 for almost half of the campaigns due to inlet P concentration variation. This situation, as well as other factors discussed in section 3.3, may explain in part the observed variability of phosphorus removal.

Fig. 2 shows for various parameters the effect of the load on the percent removal of whole treatment line. For each parameter, all the data points obtained from the different campaigns are represented in the graph, including during autumn when wine-production effluents were treated. The nominal hydraulic load of Vercia plant was 77 m<sup>3</sup>.day<sup>-1</sup> during wine-production period. This nominal value was used as a criterion to determine whether the plant was being operated over hydraulic load or not. Fig. 2 shows that TSS, TKN, COD and BOD<sub>5</sub> percent removals were close to 100%, indicating that in the ranges of this study, neither hydraulic nor organic loads influenced the performance of the plant with respect to these parameters. For the other 2 parameters monitored (TN and TP), the dispersion of the data points was higher. However, one-way ANOVA showed that the percent removals were also not significantly affected by the hydraulic and organic loads ( $p > 0.1$  in all cases). These observations also showed that the introduction of wine-production effluent into the plant did not affect its performance, underlining the robustness of the system.





**Fig. 2.** Performance of the treatment as a function of the hydraulic load (HL) and pollutant loads for various parameters: (a) total suspended solid (TSS), (b) chemical oxygen demand (COD), (c) biological oxygen demand (BOD<sub>5</sub>), (d) Kjeldahl nitrogen (TKN), (e) total nitrogen (TN), total phosphorus (TP). The dotted line shows the 100% removal rate.

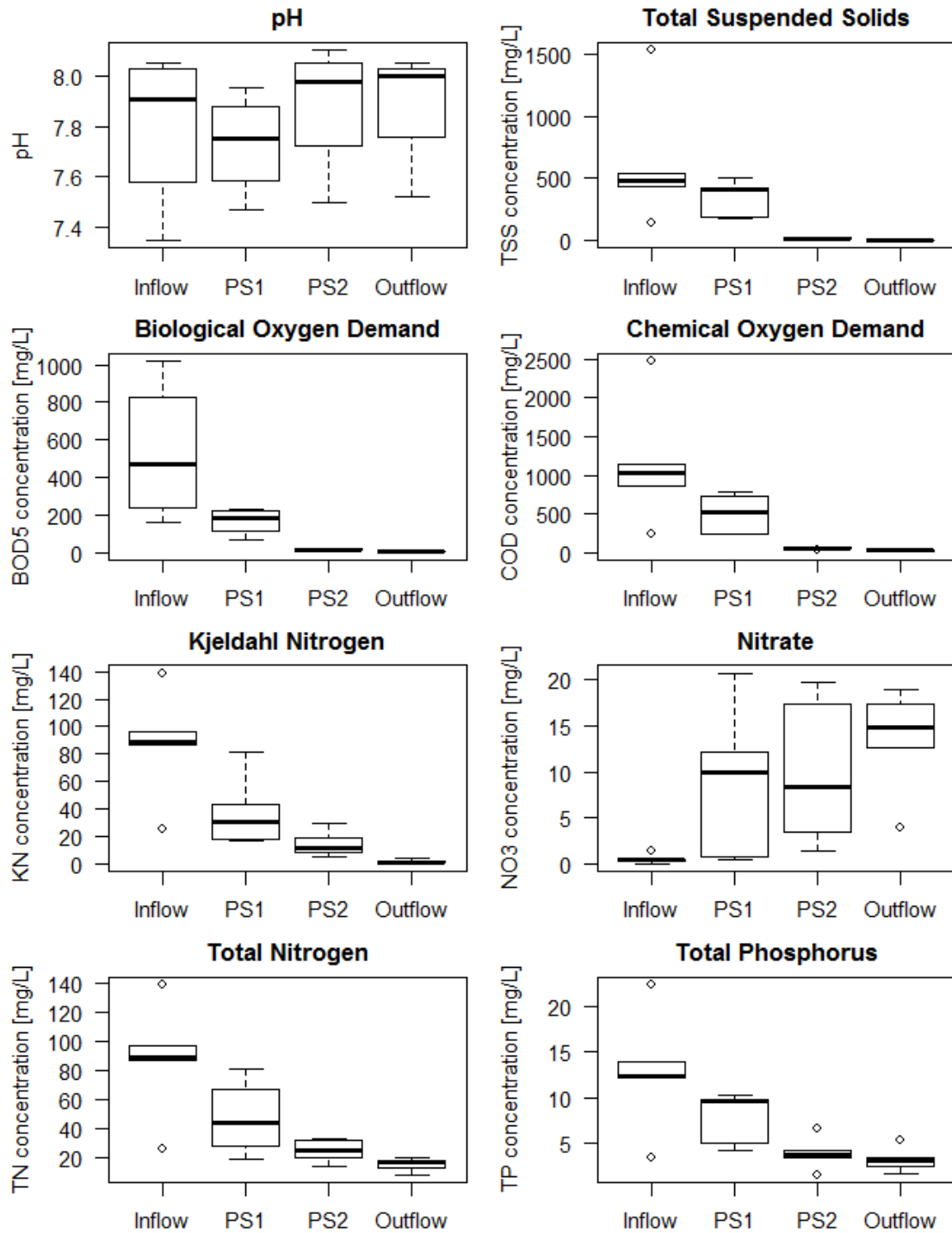
### 3.2. Specific campaigns

#### 3.2.1 Performance of each treatment step

The operational conditions of the plant during these specific campaigns were as follows: mean hydraulic and organic loads of 86 m<sup>3</sup>.d<sup>-1</sup> (SD: 56) and 77 kg COD.d<sup>-1</sup> (SD: 49) respectively.

The results obtained for the parameters monitored at the four sampling points (Inflow, PS1, PS2 and Outflow) along the treatment line are shown in Fig. 3. They are presented by box plot which indicate the median, quartile, minimum and maximum values of measured parameters at the different sampling points indicated in Fig.1. It can be seen firstly that no significant variation of the pH was observed along the treatment line. The primary treatment step (trickling filter) achieved percent removal of about 49% of BOD<sub>5</sub>, 40% of COD, 52% of TKN and 26% of TP. These results are in agreement with the fact that the main objective of the trickling filter is to degrade dissolved organic pollutant. The observed percent removal for TKN (52%) was probably due to the nitrification of organic nitrogen occurring in the trickling filter. The removal of TP observed at this step suggested that phosphorus was partially retained within the trickling filter, most probably in the form of microbial biomass due to the growth of the biofilm in the trickling filter. Excess biomass was probably detached from the trickling filter in the long term, but in an irregular manner which could not be completely taken into account by the monitoring protocol .

Regarding the 1<sup>st</sup> partially saturated VFCW, high percent removals were observed for TSS, BOD<sub>5</sub> and COD: 94%, 87% and 85% respectively. The high TSS removal indicated that the performance of filtration through the first filter was good. Percent removals for TKN, TN and TP were found to be lower: around 57%, 39% and 49% respectively. The analyses done in the outlet effluent from the first filter showed however fairly low concentrations for most parameters (Fig. 3).

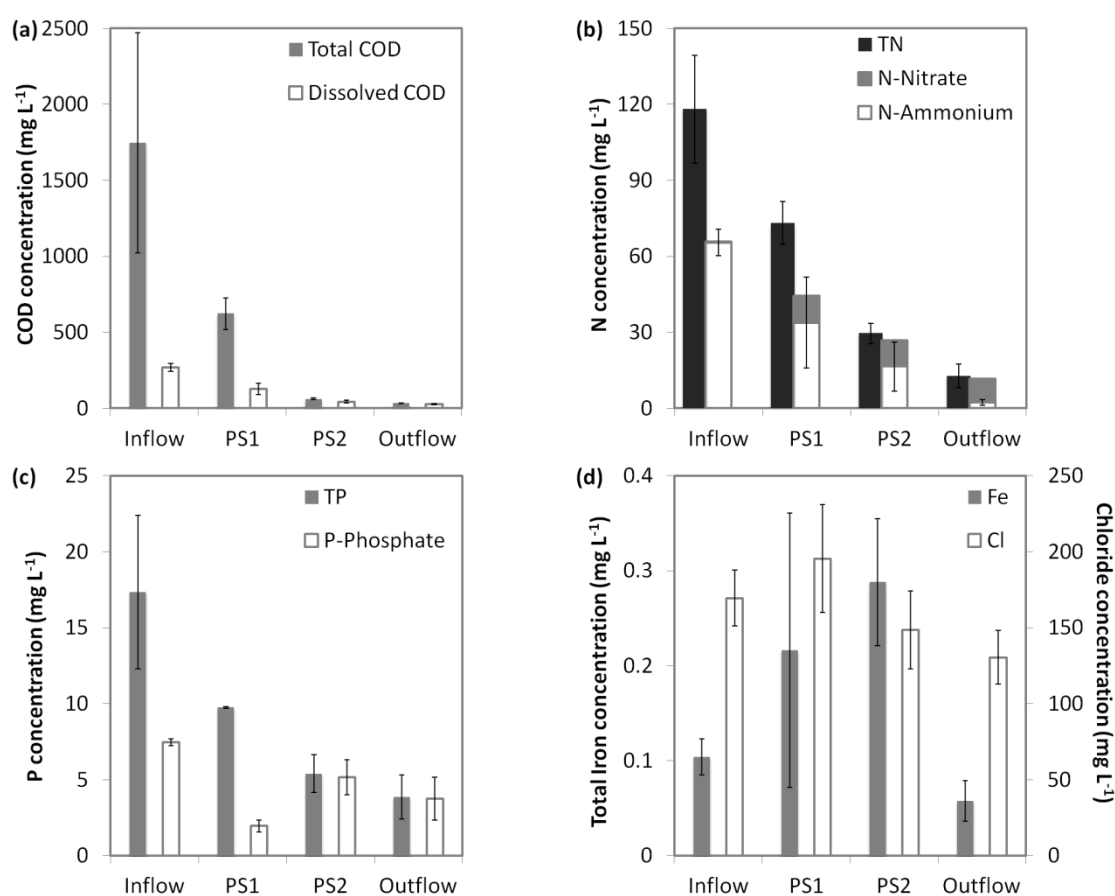


**Fig. 3.** Pollutant concentrations as measured in field campaigns at four sampling points on the treatment line.

Finally, the 2<sup>nd</sup> filter achieved logically a lower TSS percent removal than the 1<sup>st</sup> filter (28% vs. 94%) but excellent outlet concentration of TSS (maximum 9 mg.L<sup>-1</sup>). Considering the low concentrations of most parameters in the inlet of the second filter, this filtration stage exhibited fairly good performances with 52% removal of BOD<sub>5</sub>, 61% of COD, 86% of TKN, 32% of TN and 14% of TP.

### 3.2.2 Detailed investigation of nitrogen and phosphorus removal

Fig. 4 shows the concentrations of various ions in solution in the samples taken at each point along the treatment line. According to this figure, the major part of the COD at inflow was present in particulates above 0.45  $\mu\text{m}$ . The trickling filter reduced both forms of COD. This observation may seem surprising since trickling filters are rather considered to transform soluble COD into particulate COD (in the form of microbial biomass). However, the particulate COD present in the suspended solid of the influent may also be physically retained and subsequently hydrolyzed into soluble COD in the trickling filter. In addition, part of the feed COD is also mineralized into  $\text{CO}_2$  in the trickling filter, thereby contributing to the significant percent removal observed. After the 1<sup>st</sup> stage of VFCW, almost all COD was found to be in the  $< 0.45 \mu\text{m}$  fraction due to good TSS removal by this stage.



**Fig. 4.** Average concentrations measured in the last 2 “specific” field campaigns at four sampling points on the treatment line: (a) total and dissolved chemical oxygen demand (COD), (b) total nitrogen (TN),  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$ , (c) total phosphorus (TP) and P-phosphate ( $\text{PO}_4\text{-P}$ ); (d) total dissolved iron ( $\text{Fe}^{2+}$  or  $\text{Fe}^{3+}$ ) and chloride ( $\text{Cl}^-$ ).

Regarding nitrogen, results showed significant proportion of organic N in the influent ( $\text{N}_{\text{organic}} = \text{NH}_4\text{-N} - \text{NO}_3\text{-N}$ ) which was progressively removed along the treatment line. The majority of  $\text{N-NH}_4$  present in the raw wastewater influent was nitrified within the trickling filter and the 1<sup>st</sup> filter (84%). After the 1<sup>st</sup> stage, nitrogen was predominantly present in the  $< 0.45 \mu\text{m}$  fraction, both in

the forms of nitrates and ammonia. Denitrification occurred within the anoxic (saturated) zone of the first VFCW stage, thereby reducing TN concentration. In the second VFCW stage, both ammonium and nitrate removals were observed. Ammonium removal in the oxic zone can be easily explained by the nitrification process. In the anoxic zones, nitrates would then be removed by denitrification. When both ammonia and nitrates are present together, the anammox process (ANAerobicAMMoniumOxidation) may occur under anoxic conditions. This process was already observed in horizontal flow constructed wetlands and partially saturated vertical constructed wetlands by other authors (Dong and Sun, 2007; Tao and Wang, 2009).

Dissolved phosphorus concentration decreased in the first pumping station (PS1), due to the precipitation with ferric chloride. No significant increase of the total dissolved iron concentration was observed in PS1 after ferric chloride injection which confirmed the efficiency of ferric chloride addition for phosphorus removal in wastewater treatment (Caravelli et al., 2010; De Gregorio et al., 2011) and also revealed that the dosage of  $\text{FeCl}_3$  was not in excess. Chloride concentration increased in PS1 (due to  $\text{FeCl}_3$  injection) and then slightly decreased through the two stages of VFCW.

On the other hand, dissolved phosphorus concentration was found to increase from PS1 to PS2 (i.e. through the first VFCW stage). This observation may be firstly explained by the physical migration of phosphorus associated to particles and colloids (such as bacterial cells) within the first filter as already observed by Kim et al. (2013). This phenomenon may induce the solubilization of some phosphorus species depending on the local pH and Eh conditions, notably the release of phosphorus bound to reductible iron species at the bottom of the first filter (Hupfer and Lewandowski, 2008; Mortimer, 1941). Another possible explanation would be in the release of soluble phosphorus species from the sludge layer into the aqueous solution and their subsequent convective transfer through the porous media. Indeed, Kim et al. (2013) previously reported a phosphorus content as high as 2.7 % w/w of dry mass in the sludge. The mechanisms may be based on chemical reactions according to their equilibrium constants. Microbial activities may also contribute either directly or indirectly, for example through the hydrolysis and mineralization of organic compounds, or via oxidation-reduction reactions on iron, sulfur and other elements.

Meanwhile, part of the dissolved phosphorus found in PS2 was subsequently removed by the second stage of VFCW. Provided the residence time of the effluent in the filter was higher than the time elapsed between the sampling operations at PS1 and PS2 (24h), the observed decrease in soluble phosphorus concentration suggested that phosphorus was retained within the second filter. This removal may be explained on the one hand by the retention by filtration of the phosphorus-bearing particles, and on the other hand by the possible sorption or reprecipitation of the P species in solution due to the oxygenation of the effluent during its injection at the surface of the filter. Kim et al. (2013) already pointed out that the P contents in the top layer of the 2<sup>nd</sup> filter was higher than the maximum P adsorption capacity of the utilised filter medium (sand). This observation is consistent with the

suggested mechanisms of retention by filtration of P-bearing particles, and sorption or precipitation of initially dissolved P species).

#### **4. Conclusions**

The results shown in this study were obtained from a series of 44 campaigns conducted between 2004 and 2012 to evaluate the performance of a municipal partially saturated VFCWs designed and operated according to the Azoé-NP<sup>®</sup> system for the treatment of the municipal wastewater of the small city of Vercia (Jura, France). The field data thus obtained showed firstly that the overall rates of COD and organic nitrogen removals were quite stable and very good (never below 94%). COD removal and nitrification were found to be achieved by the aerobic biological trickling filter and the first stage of partially saturated VFCW. Denitrification was mostly achieved in the flooded zone of the partially saturated VFCW without requiring the injection of additional carbon source. COD, nitrogen and phosphorus were present predominantly under dissolved forms after the 1<sup>st</sup> VFCW stage underlining its very good filtration efficiency.

Regarding more specifically the retention of phosphorus, the efficiency of ferric chloride to precipitate dissolved phosphorus species was confirmed. However, phosphorus was not quantitatively retained within the sludge deposit layer accumulated at the surface of the first VFCW stage and additional P removal by the second stage was observed. The phosphorus retention/release processes within whole system are controlled by complex reactions and equilibria which are dependent upon the bio-physico-chemical conditions. Further investigations are therefore needed to evaluate the effects of these factors within the deposit layer and the porous filter medium on the retention of phosphorus. Further studies are underway in our group on the subject.

#### **Acknowledgements**

The authors would like to thank J. Colin and J-J. Prely for the data they kindly provided and their cooperation in field monitoring, the colleagues of Irstea Lyon for monitoring equipments and N. Dumont and D. Lebouil of LGCIE INSA Lyon for chemical analysis (ICP-AES and ion chromatography). They are also grateful to SCIRPE and CIFRE (Conventions Industrielles de Formation par la REcherche) for funding this research project and to reviewers for their contribution to improvement of this manuscript.

## References

- Abou-Elela, S.I. and Hellal, M.S., 2012. Municipal wastewater treatment using vertical flow constructed wetlands planted with *Canna*, *Phragmites* and *Cyperus*. *Ecol. Eng.* 47, 209-213.
- AFNOR, 2005. Recueil Normes et Réglementation Environnement., Qualité de l'eau, Vol 1 (p552), Vol 2 (p502).
- Almstrand, R., Lykmark, P., Sorensson, F. and Hermansson, M., 2011. Nitrification potential and population dynamics of nitrifying bacterial biofilms in response to controlled shifts of ammonium concentrations in wastewater trickling filters. *Bioresour.Technol.* 102 (17), 7.
- Brix, H. and Arias, C.A., 2005. The use of vertical flow constructed wetlands for on-site treatment of domestic wastewater: New Danish guidelines. *Ecol. Eng.* 25 (5), 491-500.
- Brix, H., Arias, C.A. and del Bubba, M., 2001. Media selection for sustainable phosphorus removal in subsurface flow constructed wetlands. *Water Sci. Technol.* 44 (11-12), 47-54.
- Caravelli, A.H., Contreras, E.M. and Zaritzky, N.E., 2010. Phosphorous removal in batch systems using ferric chloride in the presence of activated sludges. *J. Hazard. Mater.* 177 (1-3), 199-208.
- De Gregorio, C., Caravelli, A.H. and Zaritzky, N.E., 2011. Application of biological indices and a mathematical model for the detection of metal coagulant overload in a laboratory scale activated sludge reactor with phosphate simultaneous precipitation. *Chemical Engineering Journal.* 172 (1), 52-60.
- Dong, Z. and Sun, T., 2007. A potential new process for improving nitrogen removal in constructed wetlands—Promoting coexistence of partial-nitrification and ANAMMOX. *Ecol. Eng.* 31 (2), 69-78.
- Hupfer, M. and Lewandowski, J., 2008. Oxygen Controls the Phosphorus Release from Lake Sediments – a Long-Lasting Paradigm in Limnology. *International Review of Hydrobiology.* 93 (4-5), 415-432.
- Jetten, M.S.M., Wagner, M., Fuerst, J., van Loosdrecht, M., Kuenen, G. and Strous, M., 2001. Microbiology and application of the anaerobic ammonium oxidation ('anammox') process. *Current Opinion in Biotechnology* 12 (3), 283-288.
- Kadlec, R., Knight, R., Vymazal, J., Brix, H., Cooper, P. and Haberl, R., 2000. *Constructed Wetlands for Pollution Control*. IWA Publishing.
- Kim, B., Gautier, M., Michel, P. and Gourdon, R., 2013. Physical–chemical characterization of sludge and granular materials from a vertical flow constructed wetland for municipal wastewater treatment. *Water Sci. Technol.* 68 (10).
- Molle, P., Liénard, A., Boutin, C., Merlin, G. and Iwema, A., 2005. How to treat raw sewage with constructed wetlands: an overview of the French systems. *Water Sci. Technol.* 51 (9), 11-21.
- Molle, P., Liénard, A., Grasmick, A. and Iwema, A., 2006. Effect of reeds and feeding operations on hydraulic behaviour of vertical flow constructed wetlands under hydraulic overloads. *Water Res.* 40 (3), 606-612.
- Molle, P., Martin, S., Esser, D., Besnault, S., Morlay, C. and Harouiya, N., 2011. Phosphorus removal by the use of apatite in constructed wetlands: Design recommendations. *Water Practice & Technology* 6 (3).
- Mortimer, C.H., 1941. The Exchange of Dissolved Substances Between Mud and Water in Lakes. *J. Ecol.* 29 (2), 280-329.
- Mortimer, C.H., 1942. The Exchange of Dissolved Substances between Mud and Water in Lakes. *J. Ecol.* 30 (1), 147-201.
- Murphy, J. and Riley, J.P., 1962. A modified single solution method for the determination of phosphate in natural waters. *Anal.Chim.Acta.* 27 (0), 31-36.
- Persson, F., Wik, T., Sörensson, F. and Hermansson, M., 2002. Distribution and activity of ammonia oxidizing bacteria in a large full-scale trickling filter. *Water Res.* 36 (6), 1439-1448.
- Schindler, D.W., 1977. Evolution of Phosphorus Limitation in Lakes. *Science.* 195 (4275), 260-262.

- Szabó, A., Takács, I., Murthy, S., Daigger, G.T., Licskó, I. and Smith, S., 2008. Significance of Design and Operational Variables in Chemical Phosphorus Removal. *Water Environ. Res.* 80 (5), 407-416.
- Tao, W. and Wang, J., 2009. Effects of vegetation, limestone and aeration on nitrification, anammox and denitrification in wetland treatment systems. *Ecol. Eng.* 35 (5), 836-842.
- Thistleton, J., Berry, T.A., Pearce, P. and Parsons, S.A., 2002. Mechanisms of Chemical Phosphorus Removal II: Iron (III) Salts. *Process Safety and Environmental Protection.* 80 (5), 265-269.
- Tiessen, H., 2008. Phosphorus in the global environment, in: White, P.J. and Hammond, J.P., *The Ecophysiology of Plant-Phosphorus Interactions.* Springer Netherlands, pp. 1-7.
- Torrens, A., Molle, P., Boutin, C. and Salgot, M., 2009. Impact of design and operation variables on the performance of vertical-flow constructed wetlands and intermittent sand filters treating pond effluent. *Water Res.* 43 (7), 1851-1858.
- Vohla, C., Kõiv, M., Bavor, H.J., Chazarenc, F. and Mander, Ü., 2011. Filter materials for phosphorus removal from wastewater in treatment wetlands—A review. *Ecol. Eng.* 37 (1), 70-89.