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Romain MAILLER *, Johnny GASPERI *, Vincent ROCHER ** and Ghassan CHEBBO *

* LEESU (UMR MA 102, Université Paris-Est, Agro ParisTech), 6-8 avenue Blaise Pascal, Champs-sur-Marne, 77455 Marne-la-Vallée Cedex 2, France. (E-mail: maillerr@leesu.enpc.fr; gasperi@u-pec.fr; chebbo@leesu.enpc.fr)
** SIAAP, Direction du Développement et de la Prospective, 82 avenue Kléber, 92700 Colombes, France. (E-mail: Vincent.Rocher@siaap.fr)

Abstract

My researches are divided in two principal parts. The first part concerns the fate of micropollutants in conventional wastewater treatment plants (WWTPs) composed by primary and biological treatments. Different studies have been held by OPUR research program on primary treatments, conventional activated sludge (CAS) and biofiltration (BF), the thesis will synthesize them and add data from measurement campaigns on industrial scale membrane bioreactor (MBR) unit. This will allow comparing existing units together and to increase knowledge on a very promising and innovative intensive process (MBR).

Second part of researches concerns tertiary treatments, which are imported from drinking water industry, to improve quality of treatment regarding priority and emerging pollutants, and will monitor pharmaceuticals, pesticides and priority substances in a powdered activated carbon process at semi industrial scale, placed after a BF unit. Some laboratory scale tests will be held to better understand fundamental mechanisms and parameters which are involved in such a process. The interest of this project is to test a very promising technology on a large scale WWTP, and to provide data on a large range of micropollutants removal by adsorption tertiary treatment, which doesn't create by products contrary to oxidation processes. In the context of regulations hardening and better understanding of micropollutants effects on environment and health, these new data are very relevant and interesting for scientists and water managers who wants to anticipate and not undergo regulations.

Keywords

Membrane bio reactor, activated carbon, biofiltration, conventional activated sludge, adsorption, biodegradation, priority substances, emerging substances, efficiency.

INTRODUCTION

The micropollutants fate in the environment has become an increasing topic of interest last decades, especially in heavily urbanized area. To struggle against water contamination, the European Community adopted a control policy strategy materialized by the European Water Framework Directive (WFD, Decision No. 2455/2001/EC) (EC 2001). This legislation requires Member States to achieve a good ecological and chemical status in surface waters by 2015. Concretely, 41 chemicals are defined as priority substances because they present a significant risk over the aquatic environment, and maximum thresholds in surface waters (Environmental Quality Standards, EQS) have been set for 33 of them. In parallel of WFD pollutants, a large number of molecules such as pesticides, personal care products, pharmaceuticals, flame retardants, etc. is detected in the environment and also represents a potential threat for it (Rogers 1996, Jørgensen et Halling-Sørensen 2000, Heberer 2002).

Furthermore, the fate of pollutants within the WWTPs is today well studied and WWTP effluents are generally considered as an important source of contamination for a long time, especially in the case of urban areas (Heberer 2002). This implies a large understanding of wastewater treatment processes as they play a crucial role in the origin of micropollutants.

WWTPs are classically composed by pre-treatment, primary and biological treatments, but persistence of some pollutants encourages wastewater managers to think about tertiary treatments. Primary treatments allow removal of total suspended solid (TSS) and particulate fraction of pollution thanks to settling which can be improved and forced by addition of coagulant ($FeCl_3$) and flocculant (polymer and/or microsand). This enhanced primary treatment is called physico-chemical settling (Gaïd 2008). Then, biological treatments allow removal of carbonaceous, nitrogenous and phosphorous pollution thanks to microorganisms' action by biodegradation, and are principally represented by activated sludge flocs technologies (CAS and MBR) and biofilm technologies (BF and moving bed bio reactor - MBBR). However, efficiency of MBR is not totally well known, especially on priority substances. Moreover, conventional primary and biological treatments don't allow a sufficient removal of certain categories of molecules like pesticides or pharmaceuticals and personal care products. That's why tertiary treatments, like activated carbon reactor or oxidative processes are now studied and developed to improve global treatment.

For primary treatments, primary settling (PS) and physico-chemical lamellar settling (PCLS) are commonly used in wastewater treatment. While PS was initially and widely employed in WWTPs, PCLS is more and more frequent since this technique operates more compactly and allows a wider flexibility of configuration and use. To our knowledge, some papers exist on one or another technology (Alexander *et al.* 2012, Choubert *et al.* 2011) but there is no study comparing both technologies and examining the impact of coagulant and flocculant on the pollutant removal at the scale of industrial units.

Concerning biological treatments, common biological units have already been well documented like conventional activated sludge (Clara *et al.* 2005, Ruel *et al.* 2010, Joss *et al.* 2005, Katsoyiannis et Samara 2005). Moreover, some studies have compared conventional activated sludge process with membrane bio-reactor (Sipma *et al.* 2010, González *et al.* 2007, Bernhard *et al.* 2006, De Wever *et al.* 2007), but only a few have compared conventional treatments with biofiltration (Choubert *et al.* 2011, Joss et Maurer 2006). Furthermore, global comparison of CAS, BF and MBR about priority and emerging substances is still missing in literature, especially at industrial scale.

BF is a fix bed technique consisting of the development of a specific biofilm on a filtration material. Thus, it combines a physical retention of particles and a biological treatment of dissolved molecules by microorganisms. Its compactness (small footprint), modularity (ability to adapt operating parameters to match the wastewater flow) and intensiveness (short hydraulic retention time) allow this technology to develop worldwide since 80s, particularly in urbanized area, where it is the most suitable. Despite these strengths, BF remains very poorly studied regarding efficiency over priority and emerging pollutants.

MBR is a free developing biomass technology where microorganisms develop on flocs like CAS. It is based on coupling of biodegradation from biomass and highly efficient retention from membrane. Thus, it allows a total retention of biomass inside reactor (Weiss et Reemtsma 2008) resulting in a higher sludge retention time and sludge concentration which improve microorganisms activity (Joss *et al.* 2008). In fact, specialized bacteria can develop in MBR and increase diversity as all sludge is kept inside, contrary to CAS, and can eliminate hardly biodegradable matter (Rosenberger *et al.* 2002). Furthermore, quality of effluents is very stable, and kinetics of reaction quicker allowing weaker hydraulic retention time.

Tertiary treatments are developed because some molecules are persistent to primary and biological treatments like pesticides and a large number of pharmaceuticals and personal care products, and wastewater managers want to anticipate hardening of regulations. Different kinds of technologies already exist and are mainly exported from drinking water industry. First kind of technology is the oxidative process like ozonation or photooxidation, but a major problem is the formation of oxidation by products like nitrosamine or bromates. The other process family is the adsorption technologies principally represented by adsorption onto activated carbon. Some papers have already shown its efficiency over persistent micropollutants (Margot *et al.* 2011, Boehler *et al.* 2012).

In the framework of the OPUR research program, different studies were carried out on primary and biological treatments. PCLS and BF were first studied by (Gasperi *et al.* 2010) for priority pollutants and by (Gilbert *et al.* 2012) for alkylphenols and polybromodiphenylethers (PBDEs). The same methodology was also applied for PS and CAS. More recently, triclosan, triclocarban and parabens were also studied for both processes (Geara-Matta 2012).

In this context, the principal goal of the thesis is to continue this work to have a precise idea of the fate of micropollutants during wastewater treatments, at industrial scale. This will be done in two ways: i) synthetizing former OPUR studies about primary and biological treatments and ii) realizing measurement campaigns on intensive and innovative treatments: MBR and tertiary processes. This will allow to realize a large and precise comparison of the three main biological processes currently existing, and to have a clear idea of potential of activated carbon tertiary treatment to remove emerging pollutants.
Another part of the thesis is the study of micropollutants fate in sewage sludge, as an important part of these molecules is removed by sorption onto sludge during the different steps of water treatment. Real contamination of sludge and evolution of it during sludge management is not so well understood but is necessary to have a global view on the micropollutants in WWTPs issue. However, this part of the thesis is not presented in details here.

MATERIAL AND METHODS

Comparison of conventional primary and biological treatments

To compare the three main biological treatments (CAS, BF and MBR), different measurement campaigns were decided. First, CAS and BF WWTPs were monitored to be able to compare these two technologies. Then, MBR WWTP will be monitored in the same way to have a better understanding of fate of micropollutants with this process, and to compare the three biological units.

Biofiltration WWTP vs. conventional activated sludge WWTP

WWTP description and sampling points
Two WWTPs were studied (Figure 1) upstream (Seine Amont) and downstream (Seine Centre) Paris. Both are supervised by the Parisian public sanitation service (SIAAP).
The Seine Amont plant receives 600 000 m^3 of wastewater per day. Wastewater is first pre-treated (screening and grit/oil removal), and then settled by primary settling tanks to remove a

large amount of particles. An extended aeration activated sludge unit (biological reactor combined with a secondary settling tank), composed by three compartments, allows the carbon and nitrogen removal. This configuration of activated sludge unit belongs to the most efficient existing one as it operates at very low load (< 0.32 $kgDBO_5/(m^3.j)$, (Gaïd 2008)). The first zone operates in anaerobic conditions implying a phosphor release by bacteria; the second one operates in anoxic conditions to remove phosphates and realize the denitrification; the third step operates in aerobic conditions and allows the carbon removal and the nitrification. Finally, the effluent undergoes a tertiary treatment by clariflocculation to complete particles and orthophosphates removal.
Seine Centre receives 240 000 m^3 of wastewater per day and its design consists of a pre-treatment (screening, grit/oil), a lamellar settler (performed by settling tanks - Densadeg®) with coagulant (ferric chloride) and flocculant (anionic polymer) injection, and a three stages biofiltration unit. The first stage (Biofor® - type filters with biolite as the medium) realizes the carbon removal in aerated conditions, the second one (Biostyr® - type filters with biostyrene as the medium) realizes a nitrification step in aerated conditions and the third one (Biofor® - type filters) consists of a denitrification step in anoxic conditions. This three stages biofiltration configuration (downstream denitrification) is the most efficient one over nutrients, as showed by (Rocher *et al.* 2012). Once treatments are achieved, both effluents are discharged into the Seine River.

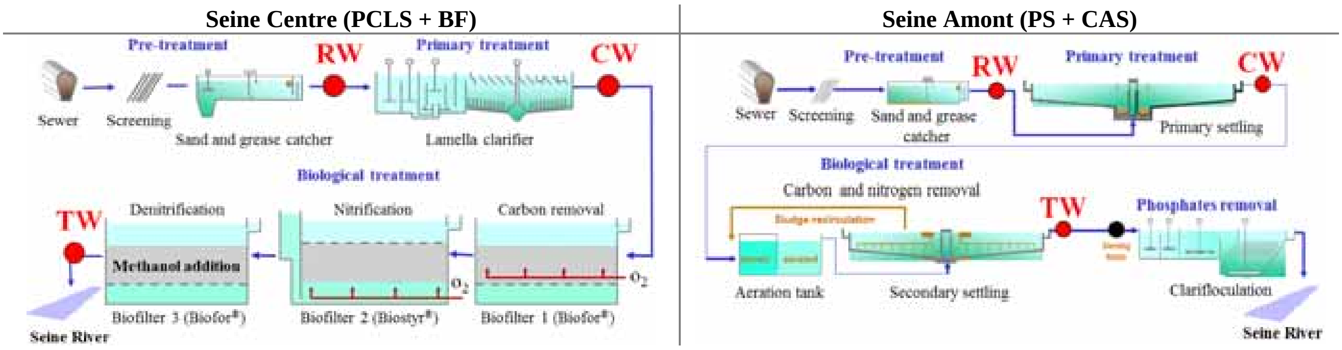

Figure 1. Layouts of Seine Centre and Seine Amont WWTPs

Sampling points defined for both plants are at the same stage of the treatment process: raw water (RW) represents the pre-treated water, clarified water (CW) represents the effluent of primary treatment and treated water (TW) represents the effluent of biological treatment. Considering the quantity of particles required for analyze (from 0.2 to 2.0 g), large volumes of water were collected (10 L for RW, 30 L for DW and TW) using automatic refrigerated samplers (at 4°C) equipped with glass bottles and Teflon® pipes to avoid any contamination. These samples were 24 h composite samples to obtain representative measures.

Pollutants and analytical procedures

A total of 104 pollutants were monitored. Depending on the substance, two methodologies were deployed. First, a large panel of 81 priority and emerging pollutants (Zgheib *et al.* 2008) was selected (Table 1) to be screened during three campaigns in 2008 (March, September, December) for PCLS + BF and 2010 (July, November, December) for PS + CAS. These analyzes were carried out by an external laboratory (IPL Bretagne).

Additional analyzes were carried out on 23 other molecules of interest. Five campaigns were performed for alkylphenols and PBDEs in 2010 and three for biocides (triclosan and triclocarbon) and parabens in late 2010 - early 2011. Whatever the period considered and as confirmed by the similar removals of conventional wastewater parameters, the operating conditions and performances on both plants were similar.

Particulate and dissolved concentrations were individually measured for each sample and for all pollutants except for metals and BTEX/HVOCs analyzed on total fraction.

The screening compounds were analyzed by IPL-Bretagne, a French certified laboratory (COFRAC). For metals, samples were digested during 2 h using a concentrated nitric-chlorhydric acid mixture. For organic pollutants, the dissolved compounds were extracted by liquid-liquid extractions (hexane or dichloromethane) for most of them or by solid phase extraction - SPE (polystyrene/divinylbenzene-copolymer cartridges) for pesticides. After lyophilization, particulate matter was extracted by assisted solid extractions (acetonitrile/water for pesticides, hexane/dichloromethane for the rest of organic compounds). Different analytical procedures were used depending on the molecules (Table 1).

Additional analyzes were performed at LEESU laboratory following internal analytical protocols for alkylphenols and PBDEs (Gilbert *et al.* 2012), and for biocides (triclosan and triclocarban) and parabens (Geara-Matta 2012).

Table 1. Groups of pollutants and analytical methods employed during biological treatments campaigns

Groupsa	Totalb	nc	Standards	Methodsd	Phasee
Phenols	2	3	ISO 18857-1	GC-MSMS	P + D
BTEX	5 (1)	3	NF EN ISO 11423-1	GC-MS	T
Chloroalkanes	1 (1)	3	IPL Internal method	GC-ECD	P + D
Chlorobenzenes	5 (3)	3	EN ISO 6468	GC-MS	P + D
HVOCs	7 (4)	3	NF EN ISO 10301 + 6468	GC-MS	T
PAHs	16 (8)	3	ISO 17993	HPLC-Fluo	P + D
Metals	8 (4)	3	NF EN ISO 11885 + 1483	ICP and AAS	T + D
Organotins	3 (3)	3	NF EN ISO 17353	GC-MS	P + D
PCBs	8	3	NF EN ISO 6468	GC-MSMS	P + D
Phtalates	1 (1)	3	Internal method	GC-MS	P + D
Pesticides	25 (12)	3	NF EN ISO 11369 + IPL Internal method	GC-MS UPLC-MSMS	P + D
Screening	81 (37)				
Alkylphenols	6 (2)	5	Internal method (Gilbert et al. 2012)	UPLC-MSMS	P + D
PBDEs	9 (6)	5	Internal method (Gilbert et al. 2012)	GC-MS	P + D
Biocides	2	3	Internal method (Geara-Matta 2012)	UPLC-MSMS	P + D
Parabens	6	3	Internal method (Geara-Matta 2012)	UPLC-MSMS	P + D
Additional analyses	23 (8)				

a Groups: BTEX = benzene, toluene, ethylbenzene and xylenes, HVOCs = halogenated volatile organic compounds, PAHs = polycyclic aromatic hydrocarbons, PBDE = polybromodiphenylethers, PCB = polychlorobiphenyls.
b Number of substances listed in the WFD is in bracket.
c Number of campaigns.
d Analytical methods: ICP = inductively coupled plasma, AAS = atomic absorption spectrometry, GC = gas chromatography, GC-ECD = GC with electron capture detector, GC-MS = GC with mass spectrometer, GC-MSMS = GC with tandem mass spectrometer, HPLC-Fluo = high pressure liquid chromatography with fluorescent detector, UPLC-MSMS = ultra performance liquid chromatography with tandem mass spectrometer,
e Phase considered with D = dissolved, P = particulate, T = total.

MBR WWTP

WWTP description and sampling points

A MBR unit will be monitored at Seine Morée WWTP in order to compare the three main biological processes. Seine Morée is a new WWTP supervised by SIAAP which will be operational in late 2013, and placed in the north-east of Paris. This WWTP is designed to treat 50 000 m^3 of wastewater from 200 000 inhabitants. Wastewater is first pre-treated (screening and grit/oil removal), and then settled by primary settling tanks (no chemicals) to remove particles. The biological treatment is realized by a MBR unit composed by a 39 500 m^3 aeration tank and an ultrafiltration (membrane) separation unit to separate water from activated sludge flocs.

Finally, treated water is discharged into La Morée River which flows into Seine River through Saint-Denis Canal (downstream Paris).
Measurement campaigns are planned in 2014 and will follow the same strategy as biofiltration and CAS WWTPs campaigns. Thereby, raw water, clarified water and treated water concentrations will be measured in both dissolved and particulate phases on 24 h composite samples.

Pollutants and analytical procedures
List of pollutants and analytical procedures from biofiltration and CAS screenings will be applied to the MBR process of Seine Morée (Table 1). A potential enlargement of the list of molecules is possible involving another laboratory for pharmaceuticals but is not decided yet.

Efficiency of tertiary treatments over micropollutants

Tertiary treatments can principally use adsorption or oxidation mechanisms. Ozonation is the most common oxidative technology while activated carbon treatment is the most promising one for adsorption. Choice was made to study adsorption process as it doesn't create by products which can be toxic and it seems to allow similar performances on pesticides and pharmaceuticals products despite its simplicity (Margot *et al.* 2011).

<u>WWTP description and sampling points</u>

Concerning the tertiary treatments part, an activated carbon process will be monitored. This unit, called CarboPlus®, consists in a reactor where activated carbon (powdered or micro granular) is contacted with biologically treated water (Figure 2). Water passes through activated carbon bed upstream with a hydraulic retention time of about 15 min, and separation of adsorbent and water is ensured by coagulation/flocculation with a control of bed height (no specific separation unit). A high quantity of activated carbon stays in the reactor to form a fluidized bed thanks to coagulant and flocculant addition, but a certain amount is always renewed to respect a certain dose injected per m^3 of water treated (crucial parameter) and ensure a continuous input of fresh and unsaturated activated carbon.

This industrial prototype is installed in Seine Centre WWTP, whose layout is given in Figure 1, and is feeded with treated water thanks to a pumping unit. Campaigns start in middle of 2013.

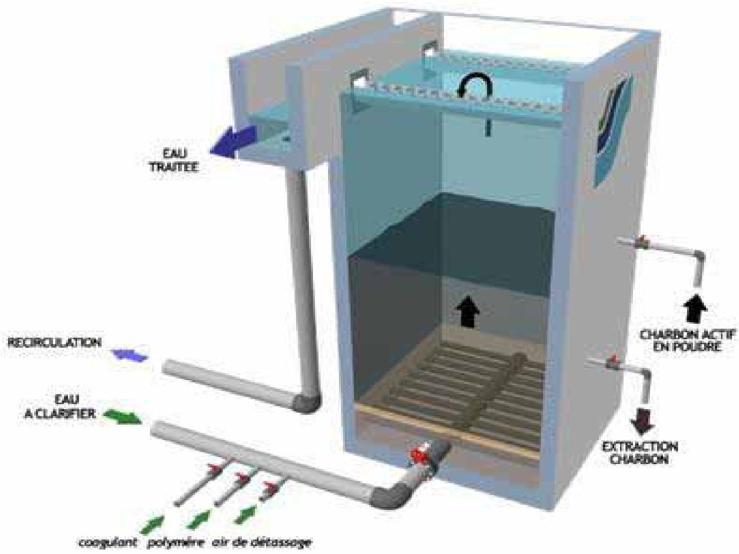

Figure 2. Layout of CarboPlus® process

Pollutants and analytical procedures

A strategy of intensive (every two weeks) and large (135 molecules) screening campaigns has been built to get a very large range of information, by measuring inlet and outlet concentrations. The list of molecules of interest have been determined regarding regulations (priority substances), and data in literature (inefficiency of conventional treatments, level found in environment and biologically treated water) (Table 2). Thus, pharmaceuticals, hormones, contrast agents, pesticides, endocrine disrupting compounds, perfluorinated molecules, personal care products and priority substances will be measured in partnership with SCA laboratory in Lyon. Two campaigns per month during 20 months from May 2013 will be realized for a total of 35 campaigns and 70 samples.

Table 2. List of molecules for tertiary treatment screenings

DCE (except pesticides)	Pharmaceuticals	Hormones	Pesticides	Other compounds
Nonylphenol	Ketoprofen	Ethinylestradiol	Alachlore	Zn, Cu
4-nonylphenol	Naproxen	Androstenedione	Atrazine	Li, V, Sb, B,
Octylphenol	Trimethoprim	Testosterone	Chlorfenvinphos	Rb, Co, As,
Para-ter-octylphenol	Bezafibrate	Progesterone	Diuron	Mo, Ba, Se,
Pentachlorophenol	Paracetamol	Estrone	Isoproturon	U, Ti, Fe, Cr,
Simazine	Metronidazole	Estradiol (α and β)	Trifluraline	Sn, Al, Ag
DEHP	Sulfamethoxazole	Levonorgestrel	HCH alpha	Iohexol
PFOA	Diclofenac	Norethindrone	Hexachlorobenzene	Iopromide
PFOS	Lorazepam	Gestodene	HCH beta	Iopramidol
Bisphenol A	Oxazepam	Estriol	HCH gamma	Musk xylene
1,2,4-Trichlorobenzene	Roxithromycine		HCH delta	
Hexachlorobutadiene	Fenofibrate		Chlorpyrifos	
Naphtalene	Ofloxacine		Aldrine	
Acenaphtylene	Atenolol		Isodrine	
Acenaphtene	Carbamazepine		Endosulfan alpha	
Fluorene	Ciprofloxacine		DDE pp	
Phenanthrene	Propranolol		Dieldrin	
Anthracene	Econazole		Endrin	
Fluoranthene	Furosemide		DDD pp	
Pyrene	Ibuprofen		DDT op	
Benz[a]anthracene	Acide salicylique		Endosulfan beta	
Chrysene	Fluvoxamine		DDT pp	
Benzo[b]fluoranthene	Tetracycline		Dicofol	
Benzo[k]fluoranthene	Oxytetracycline		Terbutryne	
Benzo[a]pyrene	Chlortetracycline		Mecoprop	
Indeno[1,2,3-cd]pyrene	Erythromycine		Aclonifen	
Dibenz[a,h]anthracene	Tylosine tartrate		Bifenox	
Benzo[ghi]perylene	Enrofloxacine		Bentazone	
Pb, Cd, Hg, Ni	Danofloxacine		Glyphosate	
	Difloxacine			
	Marbofloxacine			
	Orbifloxacine			
	Norfloxacine			
	Narasin			
	Monensin			
	Dicyclanile			
	Ampicilline			
	Penicilline G			
	Sulfadimerazine			
	Sulfanilamide			
	Sulfabenzamide			
	Sulfadiazine			
	Sulfameter			
	Sulfathiazole			
	Sulfadimethoxine			

In addition to prototype tests, laboratory scale tests will be held to determine the better powdered activated carbon between 5 models pre-selected, and to characterize it (size and partitioning of pores, specific surface area, etc.). Moreover, laboratory tests will allow understanding the influence of process parameters on removal, like hydraulic retention time, dose of coagulant, dose of activated carbon, etc. Same experiences will be done on a new type of micro granular activated carbon.

Finally, coupling of full scale and laboratory scale results with literature will enable to understand fundamental mechanisms involved in the adsorption of micropollutants by activated carbon.

PRIMARY RESULTS

Synthesis and analysis of former results from OPUR program have been done and an article is currently written. This paper compares two primary treatments and then two biological units (CAS and BF) in terms of micropollutants removal. The principal results and conclusion from this study are given below.
Comparing the two primary treatments (PS vs. PCLS), it is clear regarding our results that coagulation/flocculation represents a real gain in terms of micropollutants removal. This gain occurs mainly over particulate pollutants by the way of TSS removal, even if a slight improvement seems to be possible for some groups of soluble pollutants, removed with the colloids. Despite its existence, this effect is not obvious and clear because of the high variability of results, especially with PS process.
Jar tests and laboratory tests are maybe requested to really demonstrate the impact of coagulant and flocculant. The distribution of pollutants and the dissolved and colloidal fractions have also to be better studied.
BF appears to be able to remove most of micropollutants as efficiently as conventional activated sludge in percentage despite higher compactness and intensiveness of treatment. Removals are quite stable for both units with a maximal variation of 20% most of the time. However, the variability seems slightly weaker with the CAS treatment than with the BF treatment. All the molecules are eliminated from moderately to efficiently (20-80%, or > 80%), except pesticides which are not removed by both units, what is in good accordance with the study of (Ruel *et al.* 2012). Results for biocides and parabens are particularly interesting as variability is very low and removal rate very high with both units for them (> 70-80% for biocides and > 90% for parabens). Yet, some pollutants are slightly better removed by CAS (alkylphenols, metals, 4-chloro-3-methylphenol and PBDEs) due to better biodegradation and/or sorption. In fact, a higher biodegradation could be expected in CAS, regarding its higher hydraulic retention time (20-30 h for CAS vs. 45-60 min for BF), and this groups of pollutants could be more sorbed on activated sludge flocs than on biofilm due to their physico-chemical properties (Mahendran *et al.* 2012).
Considering the global processes, a comparison of quantity removed per quantity of total nitrogen (TN) removed has been done. With this innovative method, both configurations seem globally as efficient but removals between primary and secondary treatments vary. Finally, both processes are comparable for most of molecules at equivalent nitrogen removal as majority of points are distributed along y = x straight line. However some compounds are better removed by one or another system, with more molecules better removed by PS + CAS. These differences can be explained by two phenomena: the dependence of efficiency on influent concentration and the removal mechanisms (biodegradation and sorption). Indeed, removal depends on influent concentration as all molecules with comparable concentrations in raw water are removed comparably in quantity.
This representation is relevant as existing WWTPs are designed to treat nitrogen, so the knowing of efficiency over this parameter could allow estimating efficiency over micropollutants. Furthermore, nitrogen is a reference for wastewater managers in terms of WWTPs performances.
Finally, in the water discharged, most of compounds are not detected or just promptly. In particular, many compounds detected in raw water are never detected in treated water, showing

the positive effect of conventional wastewater treatments on many micropollutants. Despite that, some environmentally harmful molecules are still present at problematic levels, like metals, pesticides, DEHP or chloroalkanes, because of their high influent concentration or the weakness of treatments on them (pesticides). This issue incites to think about improvement of existing installations and/or addition of a tertiary level of treatment to complete their elimination. Thereby, the thesis is entirely relevant regarding this observation.

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