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OCCURRENCE AND REMOVAL OF PRIORITY POLLUTANTS BY LAMELLA CLARIFICATION AND BIOFILTRATION

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Abstract

This study investigates the occurrence of all priority substances (n = 41) listed in the Water Framework Directive and additional substances (n = 47) in raw sewage, as well as the removal performance of lamella clarification and biofiltration techniques. Once the efficiency of both types of techniques has been assessed for typical wastewater parameters, the differences in each technique's ability to remove pollutants becomes obvious; nevertheless, pollutant removal in quantitative terms still depends on the physico-chemical properties of the compounds used and operating conditions within the selected facility. For lamella clarification, the removal of organic chemicals was found to be primarily correlated with their sorption potential and, hence, strongly dependent upon $\log K_{ow}$ of the compound under study. Compounds with a strong hydrophobic character ($\log K_{ow} > 4.5$) are removed to a significant extent (approx. 85%), while hydrophilic compounds ($\log K_{ow} < 3.5$) are poorly removed (< 20%). For biofiltration, the removal of chemicals appears to be compound-dependent, although this outcome involves several mechanisms, namely: i) physical filtration of total suspended solids, ii) volatilisation, iii) adsorption on biomass, and iv) biotransformation of substances. Even if the complex processes within a biofilter system do not yield an accurate prediction of pollutant removal, two groups of chemicals can still be clearly identified: i) hydrophobic or volatile compounds, for which moderate to high removal rates are observed (from 50% to over 80%); and ii) hydrophilic, non-volatile and refractory compounds for which a low removal rate would be expected (< 20%).

Keywords

Lamella clarifier, biofiltration, priority pollutants, Water Framework Directive

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1 **1. Introduction**

2 The European Community's strategy to combat surface water pollution by means of
3 adopting a control policy was set forth in the European Water Framework Directive
4 2000/60/EC (WFD, Decision No. 2455/2001/EC). This directive offered the first list of 33
5 substances or groups of substances to be identified as action priorities at the Community level
6 and required EU Member States to ensure a "satisfactory chemical and biological status for
7 surface waters" by 2015. This proposal has therefore mandated a gradual reduction in
8 emissions, losses and discharges of all priority substances, along with a phase-out or cessation
9 of emissions, losses and discharges of priority hazardous substances over a 20-year period.

10 In pursuit of the WFD objectives, several steps have been taken to considerably reduce
11 stormwater pollution (storage tanks, stormwater treatment facilities, etc.) and improve the
12 efficiency of wastewater treatment technologies. On the one hand, these improvements have
13 led to a significant decrease of carbonaceous and nutrient pollution in receiving waters,
14 while on the other these improvements have played an important role in minimising the
15 release of xenobiotics into the aquatic environment. Regarding this point, among the various
16 wastewater technologies available, conventional activated sludge wastewater treatment plants
17 (AS-WWTPs) are the most well-documented. Numerous studies have already been conducted
18 on the fate of certain priority substances or emerging pollutants in AS-WWTPs (Fauser *et al.*,
19 2003; Jacobsen *et al.*, 2004; Katsoyiannis and Samara, 2004; Vogelsang *et al.*, 2006; Clara *et*
20 *al.*, 2005). Despite the contribution of this experimental work, data are currently lacking on
21 the removal of WFD priority pollutants using more compact wastewater treatment
22 technologies, which have now become widely implemented in modern WWTPs. Among such
23 technologies, lamella clarification and biofiltration (aerated or non-aerated biological filters)
24 are particularly attractive.

1 Lamella clarification uses a chemical additive followed by flocculation and a lamellar
2 clarifier. A coagulant (causing destabilisation of colloidal particles) and polymer (promoting
3 floc formation) are then injected into the influent wastewater prior to entering the flocculation
4 zone. Since this technique is operated more compactly and contains less exposed surface area,
5 lamella clarification is considered an essential process in wastewater treatment and is widely
6 applied to provide advanced primary treatment, in addition to producing a highly-clarified
7 effluent (Tchobanoglous *et al.*, 2003). This type of method can also serve to treat wet weather
8 flows or combined sewer overflows.

9 The biofiltration technique has been derived from drinking water production filters
10 and combines physical and biological purification processes using an immersed filtration
11 material (aerated or not, depending on the desired treatment), onto which the bacteria
12 populations ready for pollution abatement will settle. The benefits of these immersed
13 biological systems lie in their compactness (small site encumbrance), modularity (treatment
14 procedure can be adapted to match the wastewater flow the plant is able to accommodate) and
15 intensiveness (short hydraulic retention time). As a result of these advantages, biofilters have
16 become an alternative to activated sludge tanks and are perfectly suited for treatment plants
17 built in large urban areas, where real estate development pressures make available land
18 scarce.

19 Though both types of techniques are implemented in modern WWTPs and despite
20 them being highly recommended and increasingly popular for stormwater management, only
21 limited data are presently available on the removal of all WFD priority substances through
22 application of such techniques. Most studies in the literature focus on a group of substances or
23 on performance in removing carbonaceous and nutrient pollutants (Mendoza-Espinosa and
24 Stephenson, 1999; Chang *et al.*, 2004; Imasuen *et al.*, 2004; Rocher *et al.*, 2006 and 2008).

1 This study was therefore launched as part of the OPUR (Observatory of Urban
2 Pollutants in Paris) research programme, with the objective of examining the removal of all
3 priority pollutants and additional compounds by means of lamella clarification and
4 biofiltration. For this purpose, the *Seine Centre* WWTP, which is localized in the Parisian
5 suburban and combines both technologies, was selected during a total of three sampling
6 campaigns. A total of 88 substances (ranked into 13 groups of compounds) were determined
7 in raw sewage as well as at particular points within the WWTP. The distribution of pollutants
8 between the dissolved and sorbed phases of wastewater was also investigated.

9 **2. Materials and methods**

10 ***2.1. Wastewater treatment plant description***

11 The *Seine Centre* plant receives some 240,000 m³.d⁻¹ of wastewater. The treatment
12 process comprises: screening, grit removal, primary sedimentation using coagulant and
13 flocculant, followed by biofiltration units (Fig. 1). Raw sewage is initially pre-treated; this
14 stage includes screening and grit/oil removal, thus allowing the removal of coarse floating
15 solids, sand and some grease components. Following this pre-treatment, the wastewater
16 undergoes physical and chemical treatment, which traps a large amount of particles and
17 phosphorus. This step is performed by lamellar settling (use of 9 settling tanks - Densadeg®),
18 with the addition of both a coagulant (ferric chloride) and flocculant (anionic polymer).

19 After primary treatment, a biological treatment takes place over 3 stages of biofilters.
20 The first stage (24 Biofor®-type filters with biolite as the medium) is aerated and mainly
21 serves to remove carbonaceous pollution. The second stage (29 Biostyr®-type filters with
22 biostyrene as the medium) is also aerated and allows for the nitrification of ammoniacal
23 pollution. Lastly, the denitrification step occurs during stage 3 (12 Biofor®-type filters),
24 which is not aerated. Denitrification requires adding methanol, which acts as an exogenous

1 carbonaceous substrate. Once this biological treatment has been completed, effluents are
2 discharged into the Seine River.

3 ***2.2. Experimental procedure***

4 During this study, 3 sampling points, corresponding to raw sewage (RS), decanted
5 effluents (DE) and final effluents (FE), were considered (Fig. 1). In 2008, three sampling
6 campaigns were carried out (March, September and December). At each site, 24-h composite
7 samples were collected using automatic refrigerated samplers (at 4°C). To avoid problems
8 associated with sample contamination and/or pollutant adsorption during sampling, the
9 samplers were equipped with glass bottles and Teflon® pipes. In accordance with the
10 constraint to analyse 88 substances on the particulate material (i.e. from 0.2 to 2.0 g of
11 particles were required), large volumes had to be collected (10 l for RS, 30 l for DE and FE),
12 and the sampling campaigns had to be performed on three consecutive days. Each day, 3 or 4
13 groups of compounds were analysed. To avoid any modification of pollutant distribution
14 between the dissolved and particulate phases, samples were filtered as soon as possible on a
15 0.45-µm filter. After filtration, the dissolved phase was quickly sent to the laboratory for
16 analysis. As for the particulate material, filters were first lyophilised and then sent to the
17 laboratory.

18 Based on the WFD list, 41 individual substances were initially considered. Depending
19 on the chemical group, 1 to 13 additional substances were also included (Table 1), thereby
20 accounting for 88 compounds. Except for the metals, halogenated volatile organic compounds
21 (HVOCs) and BTEX (benzene, toluene, ethylbenzene and xylenes), which were analysed on
22 the total fraction, the dissolved and particulate fractions were assessed for each individual
23 compound. Analysis work was performed at the IPL-Bretagne laboratory, certified by
24 France's Environment Ministry (via the COFRAC accreditation committee).

1 **3. Results and discussion**

2 ***3.1. Priority pollutants in raw sewage***

3 Of the 88 molecules targeted, 51 (including 18 substances listed in the WFD) were
4 detected in RS. All non-detected compounds along with their limits of detection (LOD) have
5 been listed in the Supplementary Material section (Table 1). Among the pollutants detected,
6 12 substances (7 pesticides, mono- and di-butyl tin, polychlorobiphenyls 183 and 153, and
7 ethylbenzene) were highlighted since they could all be detected once during the three
8 campaigns performed, with concentrations for these elements lying at or close to the LOD.

9 In all, 39 substances were thus selected to study chemical removal using the lamella
10 clarification method. Total concentrations, as well as their respective LOD, are reported in
11 Fig. 2. The array of molecules detected and corresponding concentration ranges are typically
12 representative of data found in the literature on wastewater (Gasperi *et al.*, 2008a in France,
13 or Rule *et al.*, 2006a and b in the United Kingdom). As regards contaminant levels, three
14 groups of chemicals can be distinguished.

15 The first group includes metals and di(2-ethylhexyl) phthalate (DEHP); it displays the
16 highest concentrations (10-300 $\mu\text{g.l}^{-1}$). Metal concentrations lie between 180 and 260 $\mu\text{g.l}^{-1}$
17 for Zn, between 70 and 90 $\mu\text{g.l}^{-1}$ for Cu, and 13-15 $\mu\text{g.l}^{-1}$ for Pb. Like for DEHP, the
18 concentration is bound within the 10-25 $\mu\text{g.l}^{-1}$ range. The presence of these substances stems
19 from their extensive use both throughout industry and in household products. Metals are
20 currently used as chemical additives in a wide variety of consumer products or input into a
21 number of metal finishing processes (Rule *et al.*, 2006a and b). Similarly, DEHP is widely
22 used as an additive in plastics (Wams, 1987).

23 The second group (6 volatile compounds, nonylphenol - NP, and amino methyl
24 phosphonic acid - AMPA) contains total concentrations in the 1.0-6.0 $\mu\text{g.l}^{-1}$ range. Due to
25 their excellent degreasing properties, the HVOCs and BTEX presence can certainly be related

1 to their widespread use as a degreasing agent and solvent. For example, tetrachloroethylene is
2 introduced primarily for dry cleaning and metal degreasing purposes yet can also be found in
3 numerous household products (CEPA, 1993). The presence of NP ($1.0\text{-}1.7\ \mu\text{g.l}^{-1}$) results
4 mainly from the degradation of alkylphenol polyethoxylates, which are extensively used as
5 non-ionic surfactants in many industrial, commercial and laboratory detergents and industrial
6 processes (Ying *et al.*, 2002); this presence might also be correlated with the direct
7 application of alkylphenols as plasticisers in plastics. Lastly, AMPA has been observed in RS
8 ($< 0.03\text{-}1.5\ \mu\text{g.l}^{-1}$). This compound, generally considered to be a primary degradation product
9 of glyphosate (Rueppel *et al.*, 1977), was more frequently detected in RS and with higher
10 concentrations than glyphosate ($< 0.03\text{-}0.08\ \mu\text{g.l}^{-1}$). According to Skark *et al.* (1998), AMPA
11 may also be formed by the degradation of phosphonic acids in detergents.

12 The final group includes 26 substances and yields the lowest total concentrations
13 (between 0.005 and $0.5\ \mu\text{g.l}^{-1}$). Polycyclic aromatic hydrocarbons (PAHs), a number of
14 pesticides (diuron and, to a lesser extent, chlorpyrifos, isoproturon, metaldehyde and atrazine)
15 and tributyl tin (TBT) have been observed. These substances are frequently detected in
16 wastewater, as previously cited by Gasperi *et al.* (2007 and 2008a) for Paris wastewater or by
17 Thomaidis *et al.* (2006) for TBT in Greek wastewater. Of the 14 pesticides investigated, only
18 diuron was actually detected in all samples, with concentrations ranging from 0.03 to 0.16
19 $\mu\text{g.l}^{-1}$. This marked presence in Paris wastewater is attributed to the city's extensive use, given
20 that this pesticide accounts for a significant proportion of the total contribution from public
21 works agencies (Blanchoud *et al.*, 2007). Moreover, the presence of TBT can likely be
22 explained by its use as one of the main active ingredients in biocides for controlling a broad
23 spectrum of organisms (wood treatment, antifungal action in textiles, and industrial water
24 systems).

1 Since this section only discusses total concentration values, more detailed information
2 on the partitioning of chemicals is available in the Supplementary Material section (Table 2).

3 ***3.2. Pollutant removal during wastewater treatment***

4 This study is aimed at examining the removal of priority pollutants by means of
5 clarification and biofiltration. Prior to assessing pollutant removal levels, some precise
6 knowledge is required for removing conventional pollutants. Consequently, the efficiency of
7 both techniques was examined as an initial step for typical wastewater parameters, such as
8 total suspended solids (TSS), chemical and biochemical oxygen demand (COD and BOD),
9 ammonium (NH_4^+), total Kjeldahl nitrogen (TKN), total phosphorus (P_{tot}) and
10 orthophosphates (PO_4^{3-}). Data obtained in 2008 at the *Seine Centre* plant were processed; all
11 analyses were conducted on 24-h composite samples at similar points in the plant.

12 3.2.1 Lamella clarifier

13 *Conventional wastewater parameters*

14 The removal rates (in %) of conventional wastewater parameters (TSS, total and
15 dissolved COD - COD_{tot} and COD_s, BOD, TKN, P_{tot}) with clarification are illustrated in Fig.
16 3 (the box plot illustration indicates the median, d25 and d75 percentile removal rates). In
17 addition, median concentrations \pm standard deviations (SD) are shown for RS and DE.

18 First of all, Fig. 3 displays the high efficiency of lamella clarifier for TSS and total
19 COD pollution. For these elements, median removal rates were evaluated at 86% for TSS and
20 63% for COD_{tot}, giving rise to DE concentrations of $34 \pm 10 \text{ mg.l}^{-1}$ and $166 \pm 31 \text{ mg.l}^{-1}$,
21 respectively. No seasonal variations or temperature influence was remarked, and the decanted
22 effluent concentrations appeared not to be influenced by the quality of raw sewage entering
23 the facility. This trend has resulted primarily from adapting clarification operating conditions
24 in accordance with the quality of influent, by adjusting the ferric chloride (25 and 40 g.m^{-3})

1 and polymer doses (0.3 and 0.45 g.m⁻³). BOD was also removed to a great extent by
2 clarification (63% removal rate, with a concentration in DE at 69 ± 15 mg.l⁻¹). This high
3 removal rate is correlated with the elimination of particles and colloids. The dissolved organic
4 pollution removal is rather slight, yet nevertheless able to reach a 20% median (Fig. 3).

5 For nitrogen, only a small portion is removed by clarification (40 mg N.l⁻¹ in RS vs. 35
6 mg N.l⁻¹ in DE). This limited elimination rate is related to the removal of organic nitrogen
7 (mainly associated with particles), whereas NH₄⁺ is predominant in wastewater (approx. 80%
8 of total nitrogen) and principally in dissolved form (Boari *et al.*, 1997).

9 Furthermore, phosphorus pollution removal is considerable, as demonstrated by the
10 removal rates for PO₄³⁻ and P_{tot} (81% and 75%, respectively). DE concentrations equal
11 roughly 1.4 ± 0.3 mg P.l⁻¹ for P_{tot} and 0.5 ± 0.2 mg P.l⁻¹ for PO₄³⁻. The high phosphorus
12 pollution elimination rate is tied to particle sedimentation and, to a greater extent, to PO₄³⁻
13 precipitation with the ferric ion (FePO₄).

14 During the three sampling campaigns, median influent flow was about 240,000 m³.d⁻¹.
15 The removal rates for conventional wastewater parameters with lamella separator were close
16 to those obtained using conventional and typical operating modes (87% ± 2% for TSS, 63% ±
17 4% for COD_{tot}, 64% ± 3% for BOD, 75% ± 6% for P_{tot}, and 81% ± 10% for PO₄³⁻).

18 *Priority substances*

19 Table 2 summarises, for all molecules detected, the observed removal rates with
20 lamella separator. According to these results, the pollutants studied can be classified as: i)
21 "poorly removed" compounds, when the elimination rate is below 20%; ii) "weakly removed"
22 compounds, when this rate ranges between 20% and 50%; iii) "moderately removed", with an
23 elimination rate of 50% to 80%; and lastly iv) "efficiently removed", i.e. a rate above 80%.

1 •*Experimental results*

2 According to the previously defined criteria, metals are moderately removed by
3 clarification (> 33% for Pb and between 50% and 80% for Zn and Cu). Total metal
4 concentrations decreased markedly between RW and DE, thus confirming the strong impact
5 of lamella clarifier and suggesting that the removal of metals is likely to be strongly
6 correlated with the affinity of these metals for particles (Buzier *et al.*, 2006). As reported by
7 Gasperi *et al.* (2008b) for Paris wastewater, 40%-80% of Zn and 75%-95% of Pb and Cu are
8 associated with particles.

9 Depending on the physico-chemical properties of the pollutant, removal rates for
10 organic pollutants vary significantly. HVOCs and BTEX are only weakly removed by
11 clarification (< 20%), while heavy molecular weight PAHs (4-6 rings, HMW PAHs) are
12 efficiently removed (> 80%). The differences observed are in fact tied to the hydrophobicity
13 of the compounds, which may be reflected by the octanol - water coefficient (K_{ow}). As
14 suggested by Rogers (1996) and Byrns (2001), hydrophobic compounds ($\log K_{ow} > 4$) are
15 associated mainly with particles and hence efficiently removed by means of decantation. On
16 the other hand, hydrophilic compounds ($\log K_{ow} < 4$) are weakly removed since they
17 preferentially occur during the dissolved phase. This trend is clearly highlighted in Fig. 4. For
18 hydrophilic compounds ($\log K_{ow} < 4$), removal rates vary considerably yet still lie below
19 20%. In this study, 18 substances with a strong hydrophilic character are involved (6
20 pesticides, 6 HVOCs and BTEX, 3 light molecular weight PAHs (LMW PAHs), and 3
21 individual substances). For hydrophobic compounds ($\log K_{ow} > 4$), three groups are
22 distinguishable: i) Group 1 ($4.0 < \log K_{ow} < 4.5$), with removal rates between 20% and 50%;
23 Group 2 ($4.9 < \log K_{ow} < 7.2$), with removal rates of 50% to 80%; and iii) Group 3 ($5.6 < \log$
24 $K_{ow} < 6.7$), with removal rates exceeding 80%.

1 •*Theoretical approach*

2 Three main mechanisms require special attention in order to predict the removal rates
3 of pollutants during wastewater treatment, i.e.: i) sorption, ii) volatilisation, and iii)
4 biodegradation (Byrns, 2001). For high-rate clarification, it can reasonably be assumed that
5 sorption prevails while both volatilisation (no stripping) and biodegradation (short hydraulic
6 retention time and no high biomass concentration) processes exert only a minor influence.
7 The removal of chemicals as part of the clarification technique is therefore predominantly
8 linked to their sorption potential on primary settled sludge. According to Karickhoff (1984),
9 the adsorption of hydrophobic non-polar organic compounds is commonly described as a
10 linear function of both the K_{ow} of the solute and the organic carbon content of the adsorbent
11 (f_{oc} in $g\ C.g^{-1}$); moreover, the tendency to accumulate in sewage sludge solids can be assessed
12 using both parameters. The phase partitioning of a given pollutant between dissolved and
13 sorbed phases can thus be computed using the eq. 1 adapted from Byrns (2001).

$$FDW = 100 \times \frac{1}{(1 + 6.3 \times 10^7 \times F_{OC} \times K_{ow} \times [TSS]_{RS})}$$

Eq 1
adapted from Byrns (2001)

$$FSS = 100 - FDW$$

14 with: FDW = Fraction of chemical dissolved in water
15 FSS = Fraction of chemical sorbed on solids
16 $F_{OC} = 0.7\ g\ C.g^{-1}$ (Tchobanoglous *et al.*, 2003)
17 $[TSS]_{RS}$ = Total suspended solid concentration in raw sewage

18 The fraction of chemical dissolved in water (FDW) is shown in Fig. 5 as a function of
19 $\log K_{ow}$ (black squares). Experimental partitioning, when available, is also indicated on this
20 figure (white squares). Once the fraction of pollutant sorbed on the solid (FSS) had been
21 evaluated, the theoretical removal was calculated according to the TSS removal rate. Fig. 5
22 then compares experimental (median value \pm SD) with theoretical removal rates.

23 For compounds offering the possibility of comparison, a good correlation between
24 prediction, experimental FDW and removal rates can be observed. Below a $\log K_{ow}$ of around
25 2, FDW is predominant and, consequently, the compounds are weakly removed ($< 20\%$). In

1 contrast, the compounds with a log K_{ow} of above 5 are preferentially associated with particles
2 and hence efficiently removed ($80\% \pm 10\%$), except for DEHP which displays a slightly
3 lower removal rate (70% vs. 86%). For compounds with a log K_{ow} between 4 and 5, FDW
4 ranges from 12% to 51%, meaning that removal rates can vary significantly (20%-80%).

5 The good correlation between experimental and theoretical results justifies the
6 empirical predictive approach for easily determining the evolution of pollutants during
7 primary treatment. The above procedure provides a suitable description of sorption behaviour
8 and therefore furnishes important information on the evolution of a wide array of non-polar
9 organic compounds during lamella clarification. Theoretical approaches are critical to
10 wastewater management since the set-up and execution of experimental campaigns is
11 complex and expensive. Model generalities for screening purposes can be extended to polar
12 organics and trace metals through the use of alternative formulations to describe the relevant
13 partitioning phenomena. Although a number of empirical methods can be implemented to
14 predict the extent of partitioning, the K_d (distribution coefficient) for polar or charged
15 compounds in aquatic systems can be evaluated using laboratory tests, as performed by
16 Ternes *et al.* (2004) for pharmaceuticals and musk fragrances.

17 3.2.2. Biofiltration

18 *Conventional wastewater parameters*

19 The removal rates for typical wastewater parameters with biofiltration are displayed in
20 Fig. 6; in addition, median concentrations \pm SD have been indicated for DE and FE.

21 Results reveal that a significant portion of TSS are treated when settled effluents flow
22 through the biofiltration units ($89\% \pm 5\%$). TSS concentrations in the final effluents were
23 typically found in the 3-7 mg.l^{-1} range. According to Rocher *et al.* (2006), the first stage

1 performs the physical filtration of particles, while the subsequent biofiltration units play a
2 minor role in particle filtration.

3 By combining the physical and biological purification processes, biofiltration can
4 effectively treat carbonaceous, nitrogenous and phosphorous pollution. For carbonaceous
5 pollution, the 3-stage biofilter system removes over 92% and 83% of BOD and COD loads,
6 respectively. Like for TSS, BOD is mainly consumed during the first stage (Rocher *et al.*,
7 2006): concentrations do not exceed $10 \text{ mg O}_2 \cdot \text{l}^{-1}$ (with the median being $5.9 \text{ mg O}_2 \cdot \text{l}^{-1}$). In
8 spite of the significant removal of COD loads, a residual organic pollution, mainly in
9 dissolved form, is commonly observed in discharged effluents at a level of about $30 \text{ mg O}_2 \cdot \text{l}^{-1}$.
10 For the nitrogenous pollution, removal rates of roughly 99% are observed for NH_4^+ (FE
11 concentrations at $0.4 \text{ mg N} \cdot \text{l}^{-1}$). Water transit through the biofiltration units is accompanied by
12 the biological oxidation of ammonia-containing pollutants. The nitrification process
13 intensifies during the second stage, when over 95% of ammoniacal pollution is oxidised (0.7
14 $\text{ kg NO}_3^- / \text{m}^3 \text{ media} \cdot \text{d}^{-1}$, with an aeration of approx. $100 \text{ Nm}^3 / \text{applied kg of NH}_4^+$). Within the
15 stage 3, nitric nitrogen formed upon the oxidation of ammonia-containing pollutants is
16 reduced to atmospheric nitrogen. The efficiency of nitrification and denitrification stages
17 minimizes the discharges of nitrogen inputs ($\text{NH}_4^+ / \text{NO}_3^-$) in aquatic systems. Moreover,
18 biofiltration allows for the efficient removal of phosphorous pollution (73% for P_{tot} and 58%
19 for PO_4^{3-}), leading to FE concentrations of 0.4 and $0.2 \text{ mg P} \cdot \text{l}^{-1}$, respectively. The high level
20 of phosphorous abatement is related to the assimilation of phosphorus by bacteria.

21 During the three campaigns conducted, the biofiltration units operated at nominal
22 flow, and the removal rates of typical wastewater parameters were close to those obtained
23 with conventional operating procedures ($87\% \pm 7\%$ for TSS, $84\% \pm 3\%$ for CODt, $92\% \pm 3\%$
24 for BOD, $96\% \pm 1\%$ for TKN, and $99\% \pm 1\%$ for NH_4^+).

1 *Priority substances*

2 Table 3 lists the removal rates of priority substances using biofiltration. The removals
3 for 23 substances remaining in decanted effluents are described. Detailed information on
4 dissolved and sorbed chemicals in DE is available in the Supplementary Material section
5 (Table 3). As previously stated, the pollutants can be classified as: i) poorly removed (<
6 20%), ii) weakly removed (20-50%), iii) moderately removed (50-80%), and iv) efficiently
7 removed (> 80%).

8 Depending on the compound, strong differences appeared regarding removal
9 efficiency with biofiltration. On the whole, all pesticides (diuron, atrazine, isoproturon,
10 metaldehyde and desethylatrazine) were poorly removed (< 20%), while all volatile
11 compounds (1 HVOC, 4 BTEX and 3 LMW PAHs) were at least half removed. For other
12 pollutants, removal rates varied from 20% to 80% (Table 3). Observed differences are due to
13 differing processes occurring in the biofilter system. In theory, four mechanisms are involved
14 herein: i) physical filtration, ii) volatilisation, iii) adsorption, and iv) biotransformation of
15 substances. Although the exact apportioning of each mechanism cannot be assessed, it
16 nevertheless remains possible for each chemical to be identified with a predominant
17 mechanism.

18 *i) Physical filtration.* As previously mentioned, biofiltration units act as a physical
19 filter, removing roughly $89\% \pm 5\%$ of TSS. This retention rate leads to a reduction in
20 particulate pollutants. High removal rates are also observed for most of the hydrophobic
21 compounds; this finding appears to be positively correlated with TSS removal. The highest
22 particulate fraction removal rates were observed for NP ($90\% \pm 4\%$), followed by
23 fluoranthene ($85\% \pm 2\%$), phenanthrene ($83\% \pm 2\%$) and pyrene ($82\% \pm 1\%$).

24 *ii) Volatilisation.* During both the 1st and 2nd biofilter system stages, the physical
25 forces due to injecting diffused air induce atmospheric transfer (called air stripping). Given

1 the use of air injection, volatile organic compounds can be volatilised. According to Rogers
2 (1996), compounds with a Henry's constant (K_H) above 10^{-4} atm.m⁻³.mol⁻¹ and a $K_H / \log K_{OW}$
3 ratio exceeding 10^{-9} exhibit a high potential to volatilise. In this study, 8 compounds (toluene,
4 xylenes, dichloromethane, chloroform, tri- and tetra-chloroethylene, naphthalene and
5 phenanthrene) feature $K_H > 10^{-4}$ atm.m⁻³.mol⁻¹. All these compounds are either moderately
6 (50-80%) or efficiently (> 80%) removed. As regards BTEX and HVOCs, tetrachloroethylene
7 has the highest removal rate (82% ± 4%), followed by trichloroethylene (76% ± 8%), toluene
8 (71% ± 4%) and dichloromethane (70% ± 14%), while the lowest rate for volatile compounds
9 was recorded for chloroform (39% ± 20%). The fact that tri- and tetra-chloroethylene are
10 more widely removed can be correlated with their higher K_H value (1.81 and 1.04 x 10⁻²
11 atm.m⁻³.mol⁻¹). For naphthalene and phenanthrene, the removal rates were also substantial, as
12 shown by the major reduction in dissolved concentrations (89% ± 6% and 82% ± 6%,
13 respectively).

14 *iii) Adsorption on biomass.* Adsorption can also occur within the biofilter system. The
15 adsorption process corresponds to electrostatic interactions of positively-charged groups of
16 chemicals with the negatively-charged surface of micro-organisms and, consequently,
17 concerns ionic species. In this study, the adsorption process has been clearly highlighted for
18 Zn and TBT, whose losses amount to about 30% and 20%, respectively.

19 *iv) Biotransformation of substances.* Like for dissolved organic matter (with a removal
20 rate of 79%), chemicals can be degraded by biomass during biofiltration treatment. According
21 to Byrns (2001), the complete mineralisation of xenobiotic compounds in treatment systems is
22 rare, and the term biotransformation would more accurately describe potential changes to the
23 composition and molecular structure of such a compound. Depending on their
24 biodegradability, the three following pollutant types are distinguished: i) easily
25 biodegradable, ii) moderately biodegradable, and iii) resistant to biodegradation or refractory.

1 Given the short hydraulic retention time in the biofilter system (approx. 90 min for all three
2 stages), it can reasonably be assumed that only the easily or moderately biodegradable
3 compounds can be biodegraded.

4 For example, biodegradation process can explain the differences of removal between
5 volatile compounds exhibiting a quite similar volatilization potential ($2.7 \times 10^{-4} < K_H < 6.3 \times$
6 $10^{-3} \text{ atm.m}^{-3}.\text{mol}^{-1}$). Indeed, a more important removal is observed for readily biodegradable
7 compounds such as naphthalene and phenanthrene in comparison to moderately degradable
8 compounds (dichloromethane and chloroform). This observation is in accordance with study
9 led by McNally et al. (1998), who demonstrated that LMW PAHs prove to be readily
10 biodegradable under both aerobic and denitrifying conditions.

11 Similarly, biodegradation process can be responsible of a decrease of dissolved
12 concentration for no volatile compounds ($K_H < 10^{-4} \text{ atm.m}^{-3}.\text{mol}^{-1}$). For example, a reduction
13 of dissolved concentrations was observed for DEHP (from 20 to 60 % depending on the
14 sampling campaign) and nonylphenols (10 - 60 %) which are recognized as easily
15 biodegradable compounds (Ying et al., 2002, Fauser et al., 2003). A slighter losses have even
16 been observed for fluoranthene (0 - 30 %) and fluorene (55 - 83 %).

17 The removal of chemicals in a biofiltration system is compound-dependent and dues
18 to several mechanisms. The multiplicity and complexity of processes involved in chemical
19 removal do not allow for a simple generalisation of results obtained. Though an accurate
20 prediction of pollutant removal is not possible from their physico-chemical properties alone,
21 two groups of chemicals with differing biofiltration-induced removal rates can still be clearly
22 identified: i) hydrophobic or volatile compounds, for which moderate to high removal rates
23 are observed; and ii) hydrophilic compounds, which are neither volatile nor easily
24 biodegradable and hence expected to be poorly removed. High removal rates were indeed
25 recorded for hydrophobic compounds (mainly in the particulate fraction) or easily volatilised

1 compounds following air injection. Total removal ranged from 50% to over 80% for
2 compounds with $K_H > 1 \times 10^{-4} \text{ atm.m}^{-3}.\text{mol}^{-1}$ and between 50% and 80% for hydrophobic
3 compounds ($\log K_{ow} > 4$) due to TSS filtration. On the other hand, poor removal was
4 observed for pesticides. This low rate ($< 20\%$) with biofiltration is tied to the low
5 hydrophobicity ($\log K_{ow} < 2.6$), combined with both the low volatilisation potential ($1.53 \times$
6 $10^{-9} < K_H < 3.46 \times 10^{-5} \text{ atm.m}^{-3}.\text{mol}^{-1}$) and low biodegradability of such compounds.

7 3.2.3. Concentrations in final effluents

8 To provide a complete overview of biofiltration performance, the quality of
9 discharged effluents was also investigated. Out of the 39 pollutants initially detected in RS,
10 20 compounds including 10 substances listed in the WFD exhibited concentrations above the
11 discharged effluent detection limit (Fig. 7). More detailed information on the dissolved and
12 particulate concentrations is available in the Supplementary Material section (Table 4). These
13 higher concentrations were found for Zn ($40\text{-}50 \mu\text{g.l}^{-1}$) and DEHP ($2.1\text{-}5.8 \mu\text{g.l}^{-1}$), followed
14 by chloroform ($1.2 \mu\text{g.l}^{-1}$), tetrachloroethylene ($0.4\text{-}1.2 \mu\text{g.l}^{-1}$) and NP ($0.1\text{-}0.3 \mu\text{g.l}^{-1}$). High
15 levels of DEHP and, to a lesser extent of NP can pose an environmental threat since these
16 compounds are recognised as endocrine disrupters and have been found to persist as
17 pollutants in the environment, in addition to being blamed for causing developmental
18 disorders and/or fertility problems (Jobling *et al.*, 2002; Ying *et al.*, 2002). For the other
19 compounds, concentrations lie within the ng.l^{-1} range and typically vary from 0.005 to 0.3
20 $\mu\text{g.l}^{-1}$. In final effluents, 8 pesticides were present with concentrations generally ranging
21 between 0.03 and $1.23 \mu\text{g.l}^{-1}$. Given their poor removal during clarification and biofiltration
22 treatments, the effluents revealed concentration ranges quite similar to those observed in raw
23 sewage, except for AMPA. Of the three campaigns carried out, AMPA was detected twice
24 with higher concentrations in final effluents than in RS or DE. This increase could reflect

1 local AMPA production relative to the degradation of glyphosate, but more likely to the
2 degradation of some detergent components (Koplin *et al.*, 2006; Skark *et al.*, 1998).

3 **4. Conclusion**

4 This study has investigated the occurrence of priority substances in raw sewage as
5 well as the removal of a wide range of contaminants by means of both lamella separator and
6 biofiltration techniques. 88 substances, ranked into 13 groups of compounds, were determined
7 in raw sewage and at particular points in the WWTP.

8 This paper first confirmed that a broad array of contaminants is present in raw sewage.
9 Of the 88 molecules investigated, 39 substances (18 of which appeared on the WFD list) were
10 detected. On the whole, metal concentrations ranged from 15 to 260 $\mu\text{g.l}^{-1}$, while other
11 organic pollutants were found to lie in the $\mu\text{g.l}^{-1}$ range, with the exception of DEHP (13.5-
12 24.7 $\mu\text{g.l}^{-1}$) and volatile organic compounds (0.8-4.8 $\mu\text{g.l}^{-1}$). The presence of most of these
13 chemicals has resulted from their extensive use in a wide variety of consumer products and
14 household appliances.

15 The performance of both techniques, as regards the removal of conventional pollutants
16 and priority substances, was then assessed. The differing capability of each technique to
17 remove pollutants was obvious; nonetheless, pollutant removal was still quantitatively
18 dependent upon the physico-chemical properties of the chemicals and operating conditions
19 within the facility.

20 By operating at low coagulant and flocculant doses (25-40 g $\text{FeCl}_3.\text{m}^{-3}$ and 0.30-0.45 g
21 polymer. m^{-3}), lamella clarification can effectively clarify wastewater, thus achieving total
22 solid and organic load reductions of 86% and 65% respectively for COD. The adjustment of
23 coagulant and polymer doses, according to RS quality entering the facility, enables
24 maintaining this performance level. As for the chemicals, lamella clarifier can also effectively
25 retain most of those tracked. Backed by the theoretical approach, the removal of organic

1 chemicals is highly dependent on the log K_{ow} of the compound under consideration.
2 Compounds with a strong hydrophobic character are removed to a significant extent (similar
3 to TSS). This removal mechanism operates through TSS sedimentation and through sorption
4 to sludge particles with subsequent transfer to sludge processing systems. Conversely,
5 hydrophilic compounds (18 compounds with log K_{ow} < 4 out of the 39 detected in raw
6 sewage) were poorly removed (< 20%).

7 By combining physical and biological purification processes, biofiltration can
8 effectively treat both carbonaceous and nitrogenous pollution. Concerning the removals of
9 priority pollutants, this study has also demonstrated that chemical removal in biofiltration
10 system is compound-dependent and derived from several mechanisms, including: i) physical
11 filtration of suspended solids, ii) volatilisation or air stripping, iii) adsorption on biomass, and
12 iv) biotransformation of substances. Of the 23 substances detected in decanted effluents, 12
13 were removed at over 50% and 3 over 80%, while 5 pesticides were poorly removed (< 20%).
14 While the combination of complex processes within biofilter systems does not allow an
15 accurate prediction of pollutant evolution, two groups of chemicals are nonetheless clearly
16 distinguished for their removal rates with biofiltration: i) hydrophobic or volatile compounds,
17 for which moderate to high rates are observed; and ii) hydrophilic compounds, which are
18 neither volatile nor easily biodegradable and hence expected to be poorly removed.

19 By examining the occurrence and removal of a broad set of pollutants by lamella
20 clarification and biofiltration, this study has provided relevant information on both
21 wastewater treatment technologies and their ability to remove contaminants. Considering that
22 both techniques are currently implemented in WWTPs, the data generated for a predetermined
23 number of hazardous substances may be used in the future to identify other techniques of
24 potential significance and/or deserving of implementation within the Water Framework

1 Directive. Such additional knowledge is even more necessary given that the two techniques
2 are also recommended and have become increasingly popular for stormwater management.

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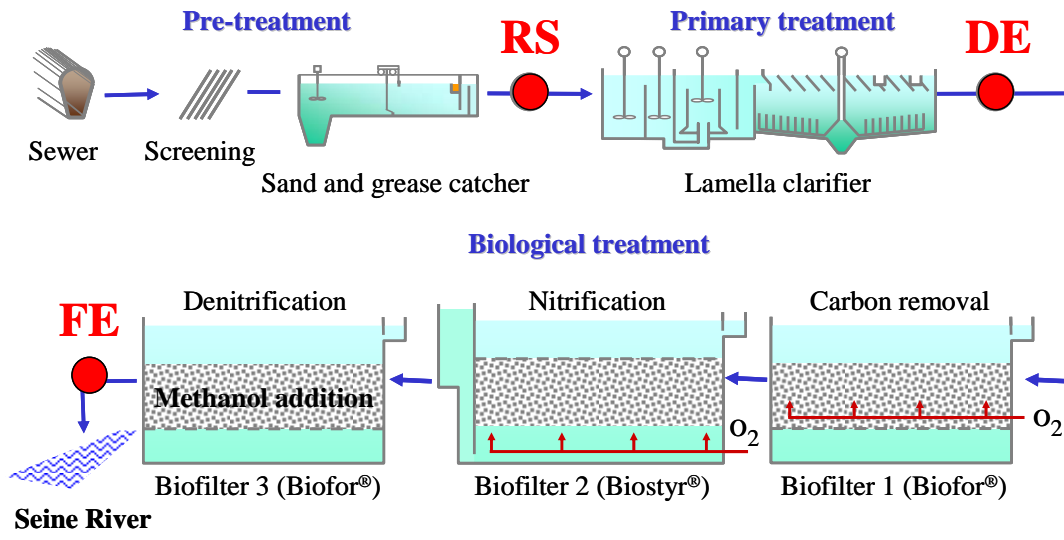
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1 **Figure caption**

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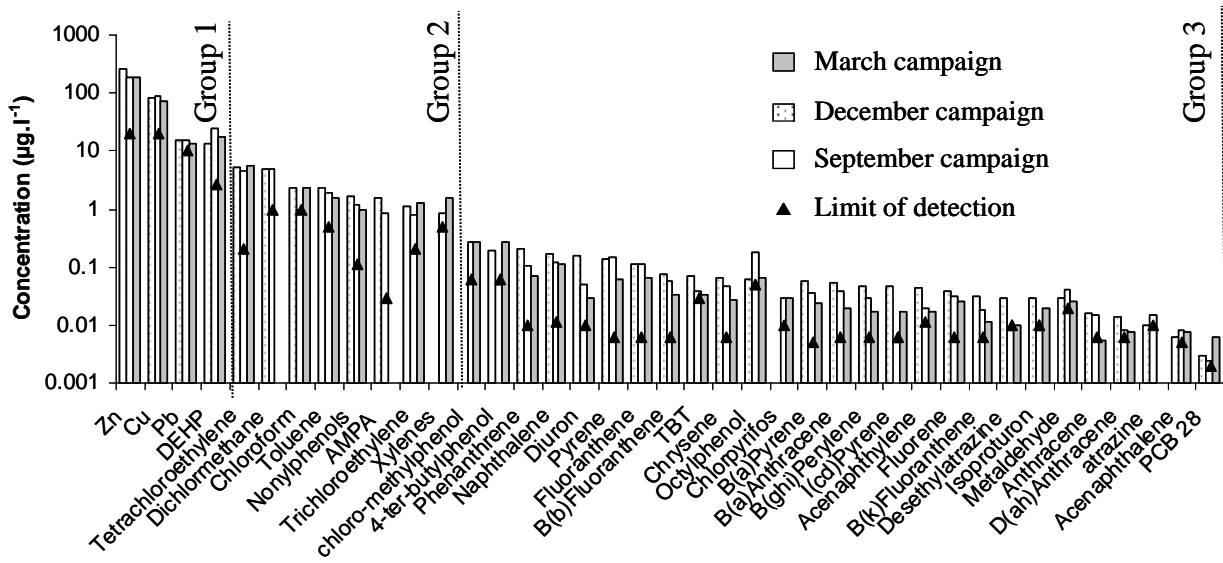


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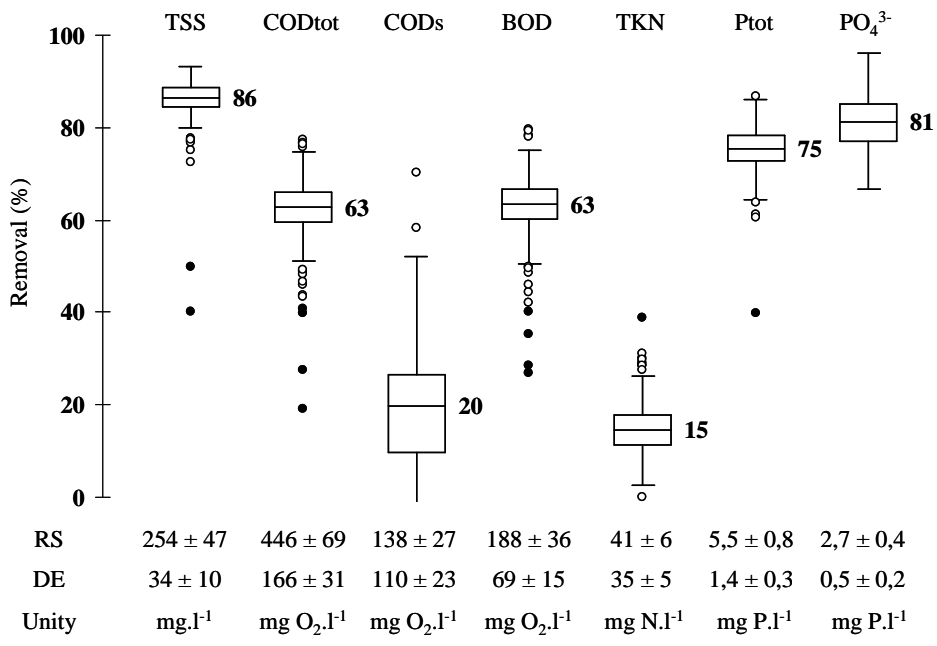
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Fig. 1 - Wastewater treatment plant studied



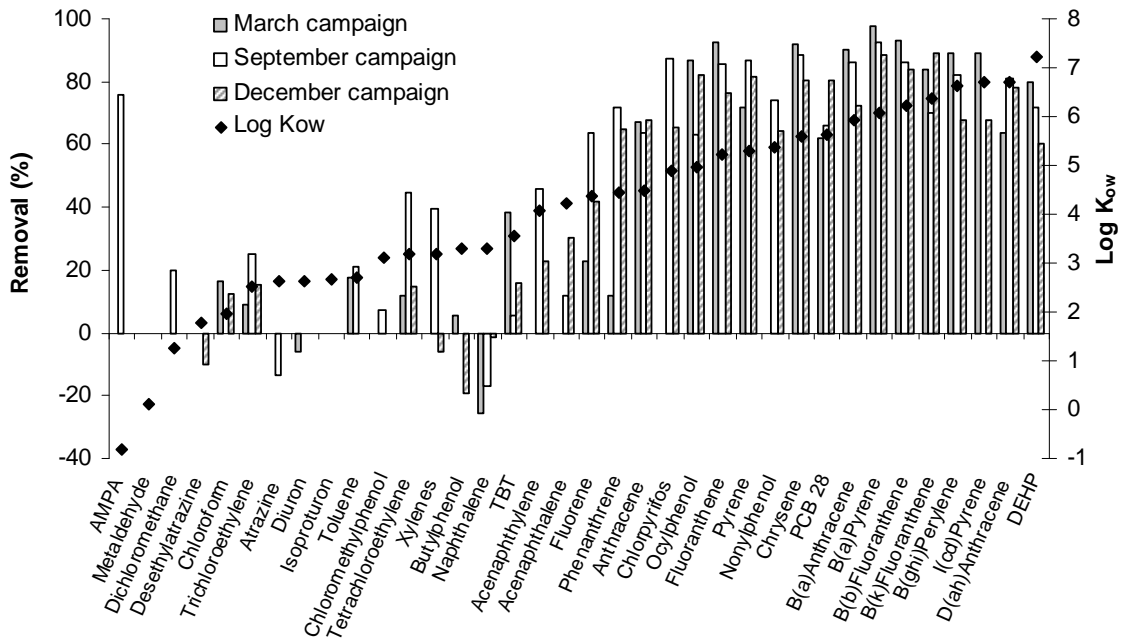
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Fig. 2 - RS concentrations ($\mu\text{g.l}^{-1}$) and limits of detection



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Fig. 3 - Removal of carbonaceous, nitrogenous and phosphorous pollutions by lamella separator

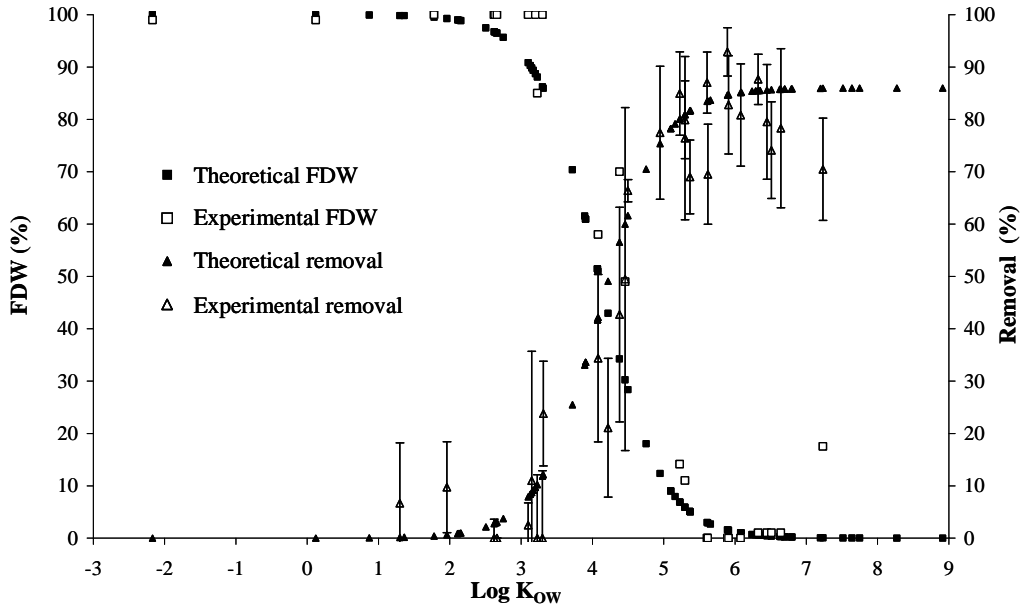


Most K_{ow} values were originally extracted from the ScorePP database
 (Source Control Options for Reducing Emissions of Priority Pollutants, <http://www.scorepp.eu>).

Fig. 4 - Removals of pollutants with lamellar clarifier, according to log K_{ow}

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Fig. 5 - Comparison between experimental and theoretical removals by lamellar clarifier

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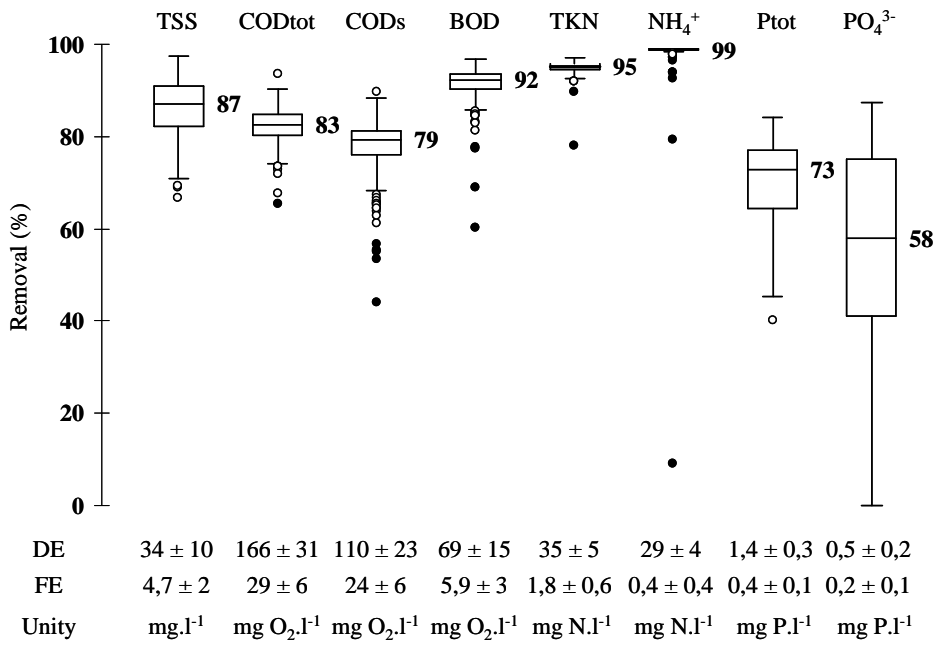


Fig. 6 - Removal of carbonaceous, nitrogenous and phosphorous pollutions by biofiltration

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1 **Table caption**

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Table 1 - Priority pollutants and analytical methods

Groups ¹	Total ²	Standards	Methods ³	Phase ⁴
Alkylphenols	5 (2)	ISO 18857-1	GC-MSMS	P + D
BTEX	5 (1)	NF EN ISO 11423-1	GC-MS	T
Chloroalkanes	1 (1)	Internal method	CG-ECD	P + D
Chlorobenzenes	5 (3)	EN ISO 6468	GC-MS	P + D
Chlorophenols	1 (1)	NF EN 12673 et ISO 6468	GC-MSMS	P + D
HVOCs	7 (4)	NF EN ISO 10301 + 6468	GC-MS	T
PAHs	16 (8)	ISO 17993	HPLC-Fluo	P + D
Metals	8 (4)	NF EN ISO 11885 + 1483	ICP and AAS	T + D
Organotins	3 (3)	NF EN ISO 17353	GC-MS	P + D
PBDEs	3 (1)	ISO 22032	CG-ECD	P + D
PCBs	8	NF EN ISO 6468	GC-MS-MS	P + D
Pesticides	25 (12)	NF EN ISO 11369+ Internal method	GC-MS UPLC-MSMS	P + D
Phthalates	1 (1)	Internal method	GC-MS	P + D

(1) Compound groups: BTEX = benzene, toluene, ethylbenzene and xylenes, HVOC = halogenated volatile organic compounds, PAHs = polycyclic aromatic hydrocarbons, PBDE = polybromodiphenylethers, PCB = polychlorobiphenyls.

(2) The substance in brackets is listed in the WFD.

(3) 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 gas chromatography with tandem mass spectrometer, HPLC-fluo = High pressure liquid chromatography with fluorescent detector, UPLC-MSMS = ultra performance liquid chromatography with tandem mass spectrometer.

(4) Phase considered with D = dissolved, P = particulate, T = Total

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Table 2 - Removals of pollutants by lamellar clarifier

n	< 20 %	20 – 50 %	50 – 80 %	> 80 %	
n = 3	tributyl tin		Cu		
	toluene		Zn	benzo(a)pyrene	
	chloroform		chlorpyrifos	benzo(b)fluoranthene	
	trichloroethylene	Pb*	DEHP	e	
	tetrachloroethylene	fluorene	nonylphenols	benzo(k)fluoranthene	
	diuron	phenanthrene	para-tert-octylphenol	e	
	metaldehyde	e	anthracene	benzo(ghi)perylene	
	naphthalene		dibenz(ah)anthracene	fluoranthene	
	acenaphthylene		e	benzo(a)anthracene	
	acenaphthene		pyrene	chrysene	
			PCB 28		
	n = 2	xylenes			
		dichloromethane			
atrazine					
desethylatrazine		-	-	indeno(cd)pyrene	
isoproturon					
AMPA					
chloromethylphenol					
butylphenol					
Total	18	3	10	8	

* Given the Pb detection limit, the removal rate exceeds 30%.

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Table 3 - Removals of pollutants by biofiltration

n	< 20 %	20 – 50 %	50 – 80 %	> 80 %
n = 3	diuron	tributyl tin DEHP	toluene trichloroethylene nonylphenols fluoranthene fluorene pyrene	tetrachloroethylen e naphthalene phenanthrene
n = 2	atrazine isoproturon metaldehyde desethylatrazin e	Zn	chloroform dichloromethane chloromethylpheno l 4-ter-butyl phenol benzo(a)pyrene xylenes	-
Total	5	3	12	3

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