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Oncological ward wastewater treatment by membrane bioreactor: Acclimation feasibility and pharmaceuticals removal performances

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ABSTRACT

Discharges of care, analyses and research activities from hospital wards are the source of the specificity of hospital effluents because they contain, among others, drug residues, detergents and disinfectants. Even if hospitals represent a small fraction of the total drug load discharged into the environment, below 10% for drugs, the characterization of this specific effluent shows that global pollution is 2–3 times more concentrated than urban wastewater. Moreover this ratio increases to 150 times for some micropollutants. Activated sludge acclimation in 2 membrane bioreactor (MBR) configurations (external and external submerged) to effluents from an oncological ward will be studied monitoring the performances on conventional pollution parameters (chemical oxygen demand, ammonium, total suspended solids etc.). The performances of drug degradation are compared with the data of the literature and with degradation tests in batch reactor with no acclimated biomass from a municipal wastewater treatment plant. The results are achieved for effluents with a high concentration of drug molecules, up to 6.82 mg L^{-1} for ifosfamide. The treatment allows the development of enhanced purification efficiencies on drug molecules and confirms the choice of a MBR process to treat this effluent, although the simultaneous presence of the various compounds leads to a complex biological response. Indeed, 5-FU was eliminated almost systematically over 90%. Sulfamethoxazole and codeine can be significantly eliminated biologically, respectively to 79 and 95%. IF and CP removal in the reactor appeared more moderate since it does not exceed 40% but membrane fouling led to higher removals of both molecules.

1. Introduction

The problem of drug residues in the environment is global and affects all segments as pharmaceuticals have been detected in surface water and groundwater, wastewater, soil and sewage sludge [20,7,47]. The presence of drug residues in the environment and the subsequent transmission are considered as a negligible risk for human health [12]. However, these studies have some limitations since the therapeutic dose does not play the role of toxicological reference value and the effects of mixtures are not considered. In view of the reality of environmental contamination and the first proven effects it is necessary to develop innovative treatment methods in order to propose effective solutions [39]. However, the very important flow of wastewater to be treated represents a major brake to the optimization of the existing Waste Water Treatment Plant (WWTP) to address this pollution and to spread with an energy-efficient aspect of advanced technologies [39]. On the contrary, the directly on-site treatment of effluents potentially heavily loaded with drug residues could represent a sensible compromise. The implementation of reduced treatment units would indeed limit investment costs and treat effluents more concentrated in drug residues. Hospital WasteWaters (HWW) come from all the hospital activities and can be qualitatively divided into three categories [15,17]: (i) domestic discharges, (ii) industrial discharges (laundry, boiler rooms…) (iii) discharges of care, analyses and research activities. The latter category is the source of the specificity of hospital effluents because they contain, among others, drug residues, detergents and disinfectants. Biochemical characterization [61] shows that HWW conventional pollution is 2–3 times more concentrated than Urban Waste Water (UWW) and the variability of concentrations is very important according to hospitals. The patient related pollution is therefore about twice more than the inhabitant equivalent pollution. Microbiological characterization [18,27] shows that bacteria in HWW are more resistant to antibiotics than those in UWW. Micropollutant pollution average concentrations in HWW are from 2 to 150 times higher than in UWW depending on micropollutants [62]. The highest ratios primarily involve antibiotics and analgesics. Orias and Perrodin [45] point out their great complexity

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Nomenclature

List of symbols

HWW	hospital wastewater
MWW	municipal wastewater
COD	chemical oxygen demand
COD_S	supernatant chemical oxygen demand
COD_P	Permeate chemical oxygen demand
MLVSS	mixed liquor volatile suspended solids
F/M	food to microorganisms ratio
TSS	total Suspended Solids
eMBR	external membrane bioreactor
sMBRe	external submerged membrane bioreactor
WWTP	wastewater treatment plant
RR	retention rate
MIC	minimum Inhibitory Concentration
MWCO	molecular weight cut off

with significant qualitative and quantitative changes indicating that the HWW ecotoxicity strongly depends on the individual case. Hospitals represent a small fraction of the total drug load, below 10% for most drugs and even less than 3% for some of them, whereas they are generally considered as a strategic objective to reduce emissions [62,52,39]. However, the contribution of hospitals in UWW varies considerably according to drugs. [62] note that between 16 and 67% of 12 analyzed drugs out of 73 in UWW come from HWW. Santos et al. [52] indicate in their study that analgesics, antibiotics and non-steroidal anti-inflammatory drugs are the 3 major classes brought by hospitals in UWW with a rate of up to 50%.

Some activities of hospitals produce very specific pollutants that could be interesting to treat directly by implementing small treatment units directly on-site [37,28,58,69,33,3]. [38] Orias and Perrodin [45] also note that little attention was paid so far to antineoplastic drugs during studies about HWW ecotoxicological risks despite “highest concentration measured in waste water/predicted non-effect concentration” high ratios (244,000 for fluorouracil).

The presence of drug residues and their metabolites in the environment involves that they are not completely eliminated by conventional wastewater treatment plants (WWTP) with activated sludge. The sludge age appears to be the key parameter to eliminate micropollutants by activated sludge processes [8,43]. The almost total biomass retention in the membrane bioreactor (MBR) process helps to work on a sludge age much higher than in WWTPs with conventional activated sludge. Sipma et al [55] compared the performances of conventional WWTPs with the performances of membrane bioreactors: only three molecules (solatol, famotidine, hydrochlorotiazide) out of the 30 selected drugs were better eliminated by conventional WWTPs than by MBRs. The reduction of the 27 other molecules was either similar or better by MBR, although it should be noted that some molecules remain poorly removed. For poorly biodegradable, polar and persistent micropollutants such as most of the pharmaceutical molecules, several studies indicate that better eliminations are obtained with MBR [13,4,49,50,32].

It is noteworthy that for identical Sludge Residence Time (SRT) similar reductions were achieved by MBR and conventional WWTPs on various micropollutants confirming the major role of SRT in the reduction of micropollutants Clara et al., 2004. Therefore, the major advantage of MBR seems to be the capability to define SRT independently from Hydraulic Residence Time (HRT). Tambosi et al. [59] confirmed the role of SRT obtaining a better removal of 6 pharmaceutical molecules by increasing the SRT from 15 to 30 days in a MBR. As a consequence, a high SRT maintains a metabolic potential when a particular substrate is no longer in the effluent. MBR thus has a “memory effect” and is therefore more flexible during important fluctuations in concentrations [13].

Few studies have been devoted to the treatment of effluents from a specific hospital department by MBR. In addition, most of the studies on specific anticancer drugs have been carried out with synthetic effluents [14,54] composed of few pharmaceuticals. The originality of this study is to test the treatment feasibility of a real effluent directly on site of an oncological ward by MBR. The biomass acclimation to effluents from a department of oncology is thus studied following the MBR performances on conventional pollution parameters (COD, SOUR, Ammonium, etc.) as well as the evolution of Total Suspended Solids (TSS) in the bioreactor. The acclimation to oncological ward effluent will be compared for two different MBR configurations: external membrane (eMBR) and external submerged membranes (sMBRe). The results are achieved for effluents with very heavy drug concentrations.

2. Materials & methods

2.1. Compounds, follow-up and analysis

The 3 most consumed anticancer drugs in the unit of the department of oncology of the hospital La Timone (Marseille, France) belong to the 7 anticancer treatments listed by the AFFSA (French Food Safety Agency): ifosfamide (IF), fluorouracile (5-FU), cyclophosphamide (CP). The codeine painkiller (CD) and the sulfamethoxazole antibiotic (SM) were added to the list of medicines taken for the project. According to their physical and chemical properties the major anticancer drugs seem mostly persistent with a high mobility in water [33,68]. This is true for 5-FU, CP and IF but also for CD and SM. The very low values of the water-octanol partition coefficient K_{ow} of the 5 selected drugs indicate that these molecules are slightly hydrophobic and strongly polar. The fluorouracil analyses are carried out in the laboratory of pharmacology and toxic kinetics from the hospital la Timone in Marseille (France). The cyclophosphamide, ifosfamide, codeine, and sulfamethoxazole analyses were conducted by the Ianesco laboratory (Institut d’Analyses et d’Essais en Chimie de l’Ouest) from Poitiers. The protocol for dosing 5-FU in blood plasma was successfully used for dosing in hospital effluents and treated waters. 5-FU analysis is performed via HPLC-UV. The quantification limit is 5 $\mu g\ L^{-1}$. The four other molecules i.e. cyclophosphamide, ifosfamide, sulfamethoxazole, and codeine are analyzed simultaneously via liquid chromatography together with a mass spectrometry (LC/MS-MS). The quantification limit of the analysis is 2.5 $\mu g\ L^{-1}$. Raw waters are settled then filtered on a filter with a porosity of 0.45 μm before analysis. The elimination of the coarser solids is not a priori a problem of underestimation of the concentration of drugs in HWW since the selected drugs are excreted only through urine and are hydrophilic thereby neglecting the sorption on the TSS of HWW.

2.2. Hospital wastewaters

The studied effluents come from oncology ward at the hospital La Timone (Marseille, France). The pipe collects wastewater (sink, shower, toilets) of 6 rooms without being diluted by the other activities of the ward. Oncological ward wastewater (HWW) is batch sampled and gathered in a buffer vessel. Pretreatments consist of a saniflo macerator system (Plus Silence, SFA, France) and a grid with a 0.5 mm cutoff. The sampling is performed in the morning in order to have an effluent loaded enough to ensure a minimum food to microorganism ratio (F/M) of 0.07 $kg_{COD}\ kg_{VSS}^{-1}\ d^{-1}$ corresponding to the operating boundary conditions chosen regularly for urban effluents [23]. The related F/M ratio always lies between 0.07 and 0.14 $kg_{COD}\ kg_{VSS}^{-1}\ d^{-1}$. The BOD_5/COD ratio shows that the effluent is biodegradable because it is relatively steady between 29 and 48%. These 160 days long campaign has