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Elimination of pharmaceutical residues in biologically pre-treated hospital wastewater using advanced UV irradiation technology: A comparative assessment

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ABSTRACT

UV irradiation technology as a membrane bioreactor (MBR) post-treatment was investigated and assessed. Both UV low pressure (LP) and medium pressure (MP) lamps were examined. The technology was installed in a pilot plant treating hospital wastewater to provide the study with adequate field data.

The effect of the UV irradiation was enhanced with varying dosages of H_2O_2 to establish an advanced oxidation process (AOP). The efficiency of the pharmaceutical removal process was assessed by examining 14 micropollutants (antibiotics, analgesics, anticonvulsants, beta-blockers, cytostatics and X-ray contrast media) which are typically released by hospitals and detected with liquid chromatography coupled tandem mass spectrometry (LC–MS/MS).

While the MBR treatment generally showed only a low degradation capacity for persistent pharmaceuticals, much better degradation was obtained by applying UV irradiation and H_2O_2 as AOP. The "conventional" cost-benefit analysis of the different technology options taking into account both electrical energy consumption and pharmaceutical removal efficiency, revealed clearly better performance of low pressure UV lamps as AOP. However, a holistic comparison between the different scenarios was carried out by evaluating their environmental impacts using the life cycle assessment (LCA) methodology. Decisive advantages were highlighted to include this approach in the decision making process.

1. Introduction

Pharmaceuticals and their metabolites are excreted by humans through faeces and urine. Hospitals are considered as point sources within the urban wastewater system [1,2] and they significantly influence the load of certain pharmaceuticals being transported to municipal wastewater treatment plants (WWTPs). Since standard treatment plants (STP) are not designed to remove pharmaceutical residues from wastewater, elevated concentrations of these contaminants have been detected in receiving waters downstream WWTPs [3]. Recent studies have revealed that pharmaceuticals persist in the water cycle [1,4,5].

In-house studies found elevated levels of relevant trace-pollutants at both the inlets and outlets of two investigated STPs [6]. The observed removal rate for Carbamazepine and Diclofenac was almost negligible. This resulted in a chronic ambient concentration of around 400 ng L^{-1} of both pharmaceuticals in the receiving river. With respect to the environmental impact of pharmaceutical residues, they generally show low acute toxicity to aquatic organisms but a number of pharmaceuticals are of concern to ecosystem health due to chronic effects [7–9].

This study focuses on the treatment of wastewater with high levels of pharmaceuticals from point sources. A pilot membrane bioreactor (MBR) was installed at a hospital containing around 360 beds. The MBR was continuously operated with a permeate flow of 2 m^3 d^{-1}. It served as pre-treatment to obtain a suitable wastewater quality for the subsequent advanced treatment using ultraviolet (UV) irradiation. So far, only a few and incoherent data can be found about UV irradiation as an advanced treatment technology. Also, current research is focussing at MBR permeate treatment with ozone, since satisfying results were obtained [10]. However, with regards to operation and maintenance UV technology is considered to have decisive advantages.

The removal efficiency of different MBR and UV treatment process setups were evaluated for pharmaceuticals which are part of the medication groups of analgesics, antibiotics, anticonvulsants, beta-blockers, cytostatics and X-ray contrast media. Additionally, the effects of a low and a medium pressure UV lamp on the removal of the pharmaceutical substances were examined. The former was used with a fixed power of 0.25 kW while the

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latter was investigated with an adjustable power between 2 kW and 10 kW. Furthermore, the effect on the pharmaceutical degradation by UV irradiation in combination with hydrogen peroxide (H_2O_2) as advanced oxidation process was also determined. The pharmaceutical degradation kinetic of each operational/experimental UV setup was modelled and compared. The main objective of the investigation was to assess the feasibility of eliminating pharmaceuticals from hospital wastewater in a cost-effective and environmentally sound way and to obtain field data that could serve as important design information for these technologies. To this end, LCA was applied as one of today's most consensual methodologies for environmental assessment of products and processes. In this context, for each treatment scenario, the environmental impact generated by the infrastructure and the resources used, such as electrical energy and H_2O_2, was directly compared to the reduced environmental impact caused by the treatment efficiency in terms of macro- and micropollutants. It is worth pointing out that the pilot study could directly feed the LCA with first-hand data. This makes the present work particularly coherent and closes the gap in previous investigations [11,12].

2. Experimental

The efficiency of the MBR has been determined by a seven and a five-day measurement campaign in which typical wastewater compounds and pharmaceuticals were analysed, both observing the influent and the effluent of the MBR treatment. Subsequently, several batch experiments were conducted to evaluate the UV treatment.

2.1. MBR treatment

The pilot plant (MBR and UV treatment) is housed in a container and is located at the hospital Centre Hospitalier Emil Mayrisch (CHEM) in Esch-sur-Alzette, Luxembourg. The MBR treats in continuous operation about 1% of the diurnal hospital sewage. Technical details are given by Venditti et al. [13]. Before pharmaceutical measurement, the MBR treatment efficiency was assessed using the removal of suspended solids, organic carbon, nitrogen and phosphorus. Two-hour composite samples, with a 3 min sampling frequency, were taken from the influent and effluent of the MBR and stored in glass vessels at 4 °C. The maximum storage time did not exceed 72 h before the samples were analysed in the laboratory. Five days composite samples were prepared, from the 2 h composites for pharmaceutical analysis, to eliminate weekly fluctuations.

2.2. UV treatment

Two different UV lamps (IBL Umwelt- und Biotechnik GmbH, Heidelberg, Germany) were applied in the framework of this study. A medium pressure (MP) lamp with an adjustable power of 2–10 kW and a low pressure (LP) UV lamp with a fixed power of 0.25 kW (see Table 1). Besides the composition of the noble gases, their filling pressure influences the UV spectrum significantly. Consequently, the LP lamp offers two energy emission peaks at UV light wavelengths of 254 nm and 185 nm while the MP lamp has a polychromatic emission along the UV spectrum. Furthermore, the latter offers (besides direct photolysis) photochemical oxidation process, i.e. in situ production of hydrogen and hydroxyl radicals that are formed within the vacuum UV (VUV) spectrum (100–200 nm) [14].

For each test with the low and medium pressure UV reactor 1 m^3 of MBR permeate collected in a buffer tank served as influent. Tests have always been conducted by operating just one UV lamp. Before the water enters the reactor, H_2O_2 can be added to provide (besides the photochemical oxidation process) advanced oxidation processes. The hereby formed hydroxyl (OH) radicals are considered to react non-selectively in oxidizing organic material and therefore enhance the elimination of pharmaceuticals. The resulting photochemical oxidation is much faster than direct photolysis, i.e. when no H_2O_2 reactant is used [14,15].

In the comprehensive monitoring program several UV operation modes have been investigated. The operation modes were based on four different process conditions assumed to have significant effects on the degradation efficiency of pharmaceuticals and on the operation expenses:

i) electrical energy needed to reduce the content of pharmaceuticals to a specific concentration level
ii) power variation of the MP UV lamp
iii) difference between the MP and LP UV lamp
iv) dosage of H_2O_2

The variation of the MP UV lamp power (ii) was chosen to investigate potential effects of changes in the UV spectrum when a different lamp power is applied. For each of the observed scenarios one cubic meter of pre-treated hospital sewage (permeate) was recirculated in the UV reactor until a total electrical energy input of 10 kWh was obtained. Samples were taken at specific intervals as a function of the electrical energy input (e.g. at 0, 1, 2, 4, 8, and 10 kWh m^{-3}). The interval depended on the applied lamp power and contact time, which was not longer than 91 s (5 h operation) for the MP lamp and 1013 s (40 h operation) for the LP lamp. This sampling scheme has been used to show the degradation kinetics of the pharmaceuticals and the interaction with electrical energy input. The aim was to achieve a reasonable comparison of the different operational setups and to produce valuable results for possible full scale applications.

2.3. Analysis

Grab samples of 500 ml were directly taken from the 1 m^3 buffer tank prior to the UV treatment of the wastewater (MBR permeate) and throughout the experiments depending on the different UV treatment scenarios described before.

Pharmaceuticals were chosen considering those known to be excreted in the highest amount in the hospital and with the highest eco-toxicity: antibiotics (Acetyl-Sulfamethoxazole CAS-Reg. 21312-10-7, Ciprofloxacin CAS-Reg. 85721-33-1, Clarithromycin CAS-Reg. 81103-11-9, Erythromycin CAS-Reg. 114-07-8 and Sulfamethoxazole CAS-Reg. 723-46-6), analgesics (Diclofenac CAS-Reg. 15307-86-5, Lidocaine CAS-Reg. 137-58-6 and Naproxen CAS-Reg. 22204-53-1), anticonvulsant (Carbamazepine CAS-Reg. 298-46-4), betablocker (Atenolol CAS-Reg. 29122-68-7), cytostatics (Cyclophosphamide CAS-Reg. 50-18-0 and Ifosfamide CAS-Reg. 3778-73-2), X-ray contrast media (Iodixanol CAS-Reg. 92339-11-2 and Iohexol CAS-Reg. 66108-95-0).The analyses of the pharmaceuticals were performed in two steps: enrichment by solid phase extraction (SPE) and analysis of the SPE extracts by LC–MS/MS. The analytical method found in the literature [16] was adapted to the specific compounds: acid (pH 3) for ciprofloxacin and X-ray media with OASIS reversed-phase sorbent Hydrophilic Lipophilic Balanced (HLB) and resin-based sorbent ENV+ cartridges respectively; neutral (pH 7) with OASIS HLB cartridges for all the other compounds.

2.4. Life cycle assessment

The chosen functional unit (FU) is the treatment of 1 m^3 of MBR permeates in order to compare the different treatment scenarios. Accordingly, data concerning the operation and infrastructure of the treatment processes were collected from the experiments