

## MICROPOLLUTANTS IN WASTEWATER TREATMENT PLANTS OF RURAL AREAS

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### Abstract

The next challenge of wastewater treatment is to reliably remove micro-pollutants at the micro-gram per litre range in order to meet the environmental quality standards set the water framework directive. The present work assessed the efficiency of different types of secondary processes adapted for the treatment of wastewaters in rural areas (stabilization pond, vertical reed-bed filters, trickling filter and rotating biodisc associated with reed-bed filters). The removal of more than 100 priority substances and other relevant emerging pollutants were determined with mass balances at four full-scale domestic wastewater treatment plants. It is shown that the treatment processes for rural areas led to a significant reduction of the pollution loads, and the work indicates refractory micropollutants that could require additional treatment.

**Key-words:** emerging and priority micropollutants, wastewater treatment plants, rural areas

### Introduction

To reach the good status of the water bodies, the European Water Framework Directive (WFD, 2000/60/EC) targets a list of priority substances for which the emissions into the environment have to be reduced or stopped by 2015, and for which the concentrations in surface waters have to be below environmental quality standards. Some of these pollutants, are released by conventional wastewater treatment plants that are currently designed to remove macropollutants at the mg per liter range (carbon, nitrogen, phosphorus). The fate of these substances during biological treatment has been documented for municipal areas equipped with activated sludge process (Martin Ruel *et al.*, 2008; Miège *et al.*, 2009), biofilter or membrane bioreactor (Choubert *et al.*, 2011). For rural areas, constructed wetlands have been studied (Matamaros *et al.*, 2008; Verlicchi *et al.*, 2010).

The present work aims to assess, through on-site mass balance, the removal rates of more than 100 micropollutants (priority and emerging substances) for treatment processes adapted for rural areas that are not documented in literature, like stabilization pond and vertical reed-bed filters, trickling filter and rotating biodisc associated with reed-bed filters. A special care was taken to ensure the reliability of the sampling, analysis and calculation of removal rates.

## Methods

127 substances have been measured: 41 priority substances of the WFD, additional substances that could potentially be defined as priority substances in the following years (other pesticides and industrial substances), 5 hormones and 38 pharmaceuticals (antibiotics, hypolipemiant, bronchodilant, analgesic, antidepressant, betablockers). Thirteen chemical methods were developed for the analysis in dissolved and particulate phases of wastewater (except volatiles in raw samples) and in the particulate phase of sludge.

Four treatment plants of rural areas below 2 000 population equivalent (P.E.) were studied:

- 1 stabilization pond (3000 m<sup>2</sup>) associated with vertical reed-bed filter (RBF) of 100 m<sup>2</sup>/filter (300 P.E.);
- 1 rotating biological contactor (1468 discs, 8647 m<sup>2</sup>) associated with vertical RBF (1000 P.E.);
- 1 trickling filter associated with a vertical RBF (1000 P.E.);
- 1 vertical RBF (30 m<sup>2</sup>/filter, gravels) associated with horizontal RBF (60 m<sup>2</sup>/filter) (100 P.E.).

Mass balances were performed based with measurements on the influent and the effluent during 2 successive 24h-periods and dry weather conditions. Refrigerated samplers equipped with Teflon pipes and glass containers were used. Strict procedures of cleaning, sampling, and field blanks were carried out (US Geological Survey, 2002). Accurate calculation rules were applied to secure the data calculated for the removal yields of micro-pollutants (Choubert *et al.*, 2011).

## Results and discussion

The concentrations of micropollutants in raw wastewater have been quantified for 100 substances among the 127 substances that were analysed. Among them 44 were always quantified at concentrations above 0,1 µg/L, like Zn, Cd, Pb, Hg, Cu, Ni, nonylphenol and ethoxylates, di(2-ethylhexyl)phtalate, glyphosate and AMPA, estriol and pharmaceuticals (like propanolol, carbamazepine, ibuprofene, ketoprofene, aspirine, diclofenac, fluoxetine, theophylline). In treated effluents of processes of urban areas, 48 substances were still always quantified, with only 32 with concentrations above 0,1 µg/L, like di(2-ethylhexyl)phtalate, some metals (Cr, Ni, Cu, Zn, As, Pb), nonylphenol and ethoxylates, glyphosate and AMPA, some betablockers (propanolol, acebutolol, atenolol, sotalol), antibiotics (sulfamethoxazole, roxithromycine), caffeine, and other pharmaceuticals (ketoprofene, naproxene, diclofenac, theophylline). The substances never quantified in treated effluents of rural areas are COV, PBDE, PAH due to the lack of industrial activity and dry weather conditions of sampling.

For the removal rates (R) of micropollutants, the number of calculated values are displayed on Fig. 1 for different treatment processes set-up in the four treatment plants studied.

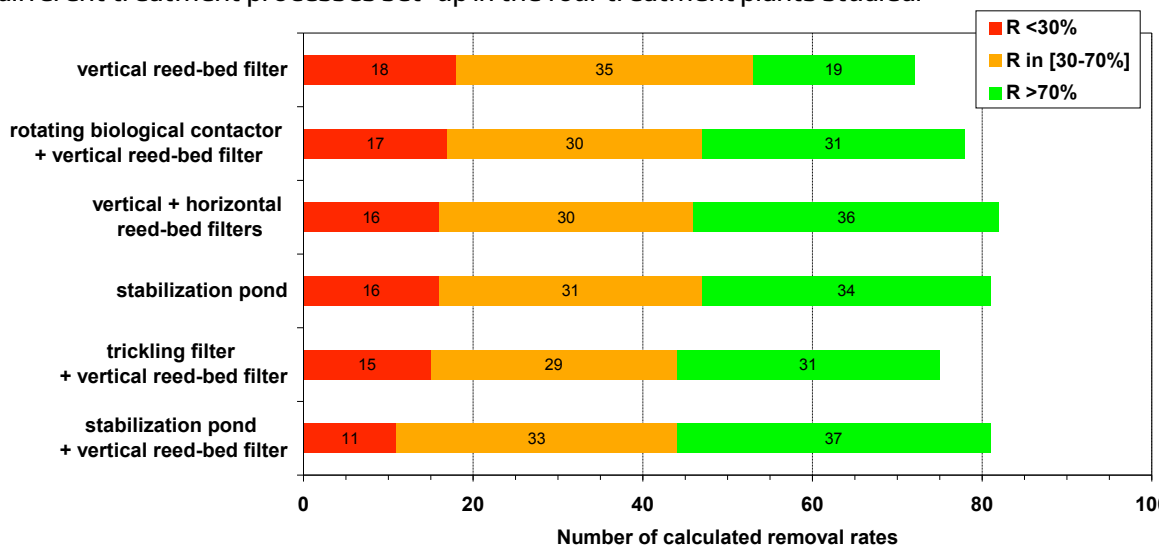


Figure 1. Number of removal rates for different process of treatment.

Removal rates could not be calculated for 35% of the analysed substances. Removal rates could be calculated for 70 to 81 substances among the 127 analysed substances:

- 30% are removed with R >70%, in particular 4-tert-butyl-, nonyl-, octyl-phenol and ethoxylates, Hg, Pb, Zn, Cu, Cr, trichloromethane, di(2-ethylhexyl)phtalate, dichlorophnols, triclosan, mono di-butyltin. But also hormones (estrone, 17 $\alpha$ -estradiol, 17 $\beta$ -estradiol, estriol) and pharmaceuticals (fluoxetine, doxepine, atenolol, nadolol, acebutolol, amitriptyline, ketoprofene, naproxene, ibuprofene, aspirine, paracetamol); 25% are removed with R between 30 and 70% ;
- 10% are not removed by the treatment plants (Cd, Ni, atrazine, diuron, trichlorobenzène, simazine, Sb, Co, As, Ti, benzothiazole, tributyl phosphate, glyphosate and AMPA, acide nonylphenoxyacetique, diclofenac, sotalol, propranolol, metoprolol, sulfamethoxazole, carbamazepine, nordiazepam).

The vertical reed-bed filter with gravels, that performed a partial nitrification and released a treated effluent with TSS around 60 mg/, had a lower number of removal rates R >70%, and a higher number in both classes (R <30% and R in 30–70%). Nevertheless, the association with a horizontal filter provides similar removal rates with the other treatment processes.

## Conclusions

From on-site mass balance applied to more than 100 micro-pollutants at four wastewater treatment plant of rural areas (6 process types), the removal rates were assessed. More than half of substances are quantified at low concentrations or not detected. Assessed biological processes of rural areas have shown comparable removal rates, with more than 30% removal for 85% of the substances quantified. They can therefore lead to a significant reduction of pollution loads, but complementary

treatments are required for refractory substances and also to bring concentration levels of accessible micro-pollutants close to their limits of quantification. The full paper will contain the data of concentrations, removal rates and released fluxes per P.E. for all the substances.

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