

## Removal of pharmaceuticals in a vertical flow constructed wetland for raw wastewater treatment in subarctic climate

Klapp, R.\* , Hedström, A.\* , Molle, P.\*\* , Herrmann, I.\*

\*Department of Civil, Environmental and Natural Resources Engineering, Luleå University of Technology, 97187 Luleå, Sweden

\*\*INRAE, UR REVERSAAL, 5 Rue de la Doua, 69100 Villeurbanne, Villeurbanne, France

### Highlights:

- A pilot-scale French two-stage vertical-flow constructed wetland treating domestic wastewater in subarctic climate was investigated regarding removal of pharmaceuticals.
- Ibuprofen was removed by 86 %, caffeine by 98 %, metoprolol by 27 % and diclofenac by 20 %.
- The removal was highest during the warmer season and lowest in spring, not during the coldest part of the year.

Keywords: micropollutants; cold climate; nature-based treatment

## INTRODUCTION

Vertical flow constructed wetlands treating raw domestic wastewater have been acknowledged for their robust treatment performance and low resource demand, and have been implemented in many French communities (Molle et al., 2005), with the advantage that they also treat the sludge on-site. The concept has thereafter been developed to intensified wetlands where the treatment efficiency is pushed further by e.g. forced aeration and additional treatment steps. In cold climate regions, however, wetlands have not yet been comprehensively studied although they could be sustainable options for wastewater treatment, for example in Sweden where local wastewater treatment is required in many small communities and where the collection and treatment of the sludge from those systems is resource demanding. The surface area requirements of wetlands would also not be problematic in rural areas of Sweden, as space is available. One aspect of the treatment performance is the ability to treat micropollutants such as pharmaceuticals, which are commonly found in domestic wastewater. The original French wetland has not yet been studied with regard to the removal of pharmaceuticals, especially not in cold climate, but wetlands with vertical flow and multiple stages generally perform better in removing pharmaceuticals than other types of wetlands (Ilyas and van Hullebusch, 2020; Sossalla et al., 2021), mainly due to aerobic biodegradation. Therefore, in this study, we investigated a pilot-scale French wetland in subarctic climate, with special focus on the removal of pharmaceuticals.

## METHODOLOGY

The studied pilot-scale wetland (12 pe) was set up in a small village in northern Sweden in 2021 and has been running continuously since then, except for a 5-month shut down during the first winter due to freezing of piping. The climate is subarctic with cold, long, and snowy winters and short and mild summers. The wetland is composed of two stages of filter media (gravel and sand) planted with *Phragmites australis*, and three batching and collection tanks, one before the first stage (A) with raw wastewater, one between the stages (B) with wastewater treated in the first stage of the wetland, and one after the second stage (C), with the final effluent. The wetland was intermittently fed with untreated wastewater, diverted from the influent of the municipal conventional wastewater treatment plant serving the village, with the sludge from the wastewater being deposited on the surface of the first stage. The water samples were collected monthly over the course of one year in 2023, using 24-hour flow-proportional sampling with automatic samplers in the collection tanks and tipping buckets for flow monitoring, so that representative samplings were obtained. The samples were analysed for 99 pharmaceuticals using LC-MS/MS, selected based on a list of priority substances from the Swedish EPA. In addition, the samples were analysed on common wastewater characteristics, including biochemical oxygen demand (BOD), total phosphorus and nitrogen. Air temperature continuous measurements and water temperature grab sampling at the time of the flow proportional sampling were also performed.

## RESULTS AND CONCLUSIONS

About 15 of the analysed substances were detected both in the influent and effluents and 20 substances were only detected in one of the two. This could be due to measurement uncertainties or due to the substance being removed in the wetland. The total concentration of pharmaceuticals in A, the untreated wastewater, were 86–581 µg/L. The total reduction in concentration ranged between 41–75 % in B, and by 68–96 % in C, with the highest reduction during the warmer months. It should however be taken into account that individual substances can be easily degradable and therefore have a big impact on the total reduction. Surprisingly, the lowest reduction both in B and C occurred in April, i.e. not during the coldest winter months, possibly due to meltwater infiltrating the sewer in spring slowing down the biochemical processes in the wetland. This is supported by low BOD concentrations during this time indicating dilution, although the influent water temperature was not lower than the previous months. Individual substances detected in both influent and effluent samples, ibuprofen, caffeine and metoprolol, are considered priority micropollutants (Gros et al., 2017) and are shown in Figure 1, along with commonly studied diclofenac. Ibuprofen and caffeine were removed by 86 and 98 %, on average, respectively in C, which is similar to removal in conventional wastewater treatment plants (Clara et al., 2005). The removal of metoprolol was less effective and unregular with on average 27 % removal in C. Diclofenac was removed by on average 20 % in C. In conclusion, the French wetland removed many easily degradable pharmaceuticals in cold climate but struggled with the more persistent substances. The ability of the wetland to remove pharmaceuticals varied with the season, with the better removal occurring during the warmer summer season and the least removal occurring in spring.

## ACKNOWLEDGEMENTS

The financial support of Formas, a Swedish Research Council for Sustainable Development, grant number 2019-01438, is gratefully acknowledged. The authors thank Erika Broström and Håkan Talludde from LUMIRE for their help with establishing and operating the pilot wetland as well as Peter Rosander and Mashreki Sami (Luleå University of Technology) for their help with problem-solving and sampling.

## REFERENCES

- Clara, M., Strenn, B., Gans, O., Martinez, E., Kreuzinger, N., Kroiss, H., 2005. Removal of selected pharmaceuticals, fragrances and endocrine disrupting compounds in a membrane bioreactor and conventional wastewater treatment plants. *Water Res.* 39, 4797–4807. <https://doi.org/10.1016/j.watres.2005.09.015>
- Gros, M., Blum, K.M., Jernstedt, H., Renman, G., Rodríguez-Mozaz, S., Haglund, P., Andersson, P.L., Wiberg, K., Ahrens, L., 2017. Screening and prioritization of micropollutants in wastewaters from on-site sewage treatment facilities. *J. Hazard. Mater.* 328, 37–45. <https://doi.org/10.1016/j.jhazmat.2016.12.055>
- Ilyas, H., van Hullebusch, E.D., 2020. Performance comparison of different types of constructed wetlands for the removal of pharmaceuticals and their transformation products: a review. *Environ. Sci. Pollut. Res.* 27, 14342–14364. <https://doi.org/10.1007/s11356-020-08165-w>
- Molle, P., Liénard, A., Boutin, C., Merlin, G., Iwema, A., 2005. How to treat raw sewage with constructed wetlands: An overview of the French systems. *Water Sci. Technol.* 51, 11–21. <https://doi.org/10.2166/wst.2005.0277>
- Sossalla, N.A., Nivala, J., Reemtsma, T., Schlichting, R., König, M., Forquet, N., van Afferden, M., Müller, R.A., Escher, B.I., 2021. Removal of micropollutants and biological effects by conventional and intensified constructed wetlands treating municipal wastewater. *Water Res.* 201. <https://doi.org/10.1016/j.watres.2021.117349>

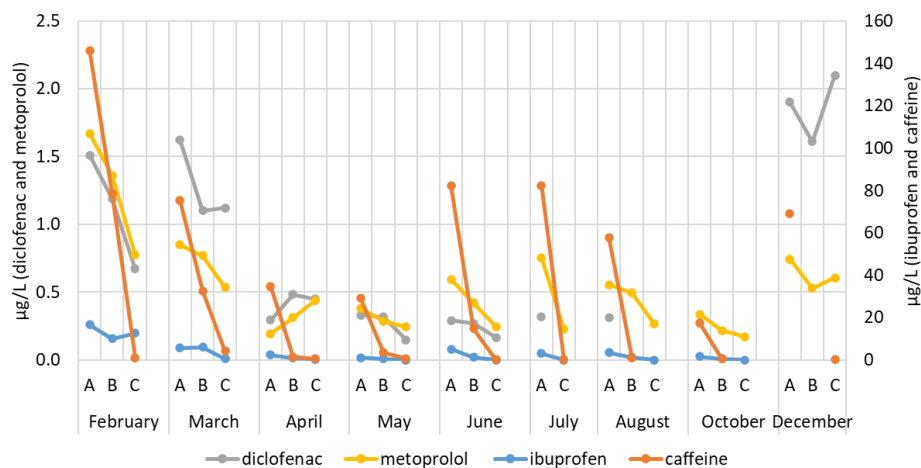


Figure 1. Concentrations of four pharmaceuticals measured in the three collection tanks (A, B and C) for one year. Concentrations of ibuprofen were below detection limits in C on several occasions and were replaced with half the detection limit as a conservative estimation.