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# Substance flow analysis as a tool for mitigating the impact of pharmaceuticals on the aquatic system

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ABSTRACT

Pharmaceuticals constitute an important environmental issue for receiving waters. A holistic approach, taking into consideration the sources of these compounds (hospitals, domestic use), discharges (wastewater effluent, combined sewer overflows) and related risks to the environment, is therefore needed to develop the best protection strategy.

The substance flow analysis (SFA) approach, applied, for example, to the city of Lausanne, Switzerland, is an ideal tool to tackle these issues. Four substances were considered: one antibiotic (ciprofloxacin), an analgesic (diclofenac), and two anti-epileptics (carbamazepine and gabapentin). Consumption data for the main hospital of the city (916 beds) and for the population were available. Micropollutant concentrations were measured at different points of the system: wastewater inlet and outlet (WWTP), combined sewer overflows (CSO) and in the receiving waters (Vidy Bay, Lake Geneva). Measured and predicted concentrations were in agreement, except for diclofenac, for which analytical uncertainties were expected. Seven different scenarios were considered (supplementary treatment at the WWTP, at the hospital or at both places, etc.). Based on the results obtained, the supplementary treatment at the WWTP decreases the load of pharmaceuticals reaching surface water by a factor between 2 and 27, depending on the compound and on the technique. The treatment at the hospitals only influences the amount of ciprofloxacin reaching the environment and decreases the release by one third. The contribution of CSO to surface water pollution is low compared to that of the WWTP for the selected compounds. Regarding the risk for the receiving waters, ciprofloxacin was found to be the most problematic compound, with a risk quotient far above 1. In this particular case, a treatment at the WWTP is not sufficient to reduce the risk, and additional measures at the CSO or at the hospital should be considered. SFA is an ideal tool for developing the best strategy for pharmaceutical elimination, but its application depends on data availability and local conditions.



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## 1. Introduction

The presence of pharmaceuticals in surface water around urban areas was highlighted by several researchers during the last decade (Alder et al., 2001; Kolpin et al., 2002; Schowanek and Webb, 2002; Bendz et al., 2005; Zuccato et al., 2006; Van De Steene et al., 2012). Indeed, pharmaceuticals consumed by humans are excreted in urine and faeces as parent compounds or metabolites and enter the sewage system; they usually reach the wastewater treatment plant (WWTP), where they are partially removed before being discharged into surface water (Kümmerer, 2008). Many of these substances are of major concern regarding their possible long-term impacts on both humans and the aquatic environment (Fent et al., 2006; Jones et al., 2005; Kostich and Lazorchak, 2007; Dussault et al., 2008). Scientists and engineers therefore came to the conclusion that reducing pharmaceuticals and, more generally, micropollutant discharge from urban areas is of high priority (Kümmerer, 2008; Williams, 2005; Ternes and Joss, 2006).

In recent years, an increasing number of research has been undertaken to investigate different technical solutions to remove pharmaceuticals in the WWTP (Ternes and Joss, 2006; Miège et al., 2008). Common investigated technologies include ozonation combined with sand filters (see for example Nakada et al., 2007; Kim et al., 2008) or activated carbon (see for example Roswell et al., 2009) combined or not combined with nanofiltration (Kazner et al., 2008) or ultrafiltration.

However, wastewater discharge is not the only way for pharmaceuticals to reach surface water. During rain periods, mixtures of stormwater and wastewater are also rejected in the aquatic system through combined sewer overflows (CSO). These episodic discharges are rarely taken into account despite the fact that they may contribute to an important aquatic contamination, and can sometimes present a risk for the aquatic organisms (Even et al., 2007; Weyrauch et al., 2010; Phillips et al., 2012). Therefore, the management of pharmaceuticals in urban areas should take into account all emission sources to the aquatic system, and should also include the impacts of these discharges on the receiving environments. A holistic view of emissions and risk for the aquatic environment is thus needed for the mitigation of pharmaceutical impacts.

In this paper, we propose such an approach based on substance flow analysis (SFA). Briefly, SFA describes and quantifies the material flows through a defined system, and has been originally proposed for regional analysis by Baccini and Brunner (1991). Later the SFA was enhanced with modelling concepts to allow simulations on the basis of current system knowledge (Baccini and Bader, 1996; Bader and Scheidegger, 2012). Such an approach was successfully used for phosphorous management in a large city (Huang et al., 2007). Furthermore, different studies in Switzerland and in Austria demonstrated its potential as a basis for river basin pollution control with respect to nutrients (Lampert and Brunner, 1999; Sarikaya et al., 1999; Somlyódy et al., 1999). A SFA was also tested and validated for heavy metals (Chèvre et al., 2011) for the same region of interest presented in this paper. This latter study focused on stormwater discharges carrying organic and inorganic substances to the aquatic environment mainly during rain events (Burton and Pitt, 2002; Rossi et al., 2004). The SFA, as proposed in this last study, allows modelling with little data and profound system knowledge. Measurements, literature values and estimations or plausible reasoning can be used as input data.

The overall goal of this paper is to use SFA as a tool for the implementation of strategies aimed at reducing the risk of pharmaceutical discharges in the aquatic environment. We will base our investigations on the city of Lausanne, whose CSOs, stormwater and WWTP ends up in the Bay of a large lake, Lake Geneva. This lake serves as a recreational area and is an important source of drinking water for more than 600,000 habitants (www.cipel.org). Two sources of pharmaceuticals will be considered, namely urban and hospital consumption. Discharges into the lake can occur via WWTP and CSOs. Finally, the environmental risk for the aquatic life in the Bay will be used as an indicator to evaluate the current state and to select the best strategy.

## 2. Material and methods

### 2.1. *Substance flow analysis*

The analysis of the contribution of the different discharges to surface water was performed using a mathematical substance flow analysis (Bader and Scheidegger, 2012). Such an analysis is based on the principle of mass balance: a substance enters a closed system and may be transported or transformed in the system, and may also leave the system. In our case, the system is defined by the sewer catchment and the receiving water, the bay of Vidy in Lake Geneva.

The SFA proposed in this study is carried out in four steps (Bader and Scheidegger, 2012):

1. System analysis: this step defines the temporal and spatial boundaries and the indicator substance(s). Based on an acquired understanding of the system, the relevant balance for volumes ($m^3$) and flows (kg) of the system are identified.
2. Model approach: the relationships within the system are formulated as mathematical equations, in which the variables describe the flows and stock change rates of the system. A set of parameters is used to describe the current knowledge of the system's behaviour.
3. Data collection and calibration: the input data for the parameters is acquired from all available sources, including measurement results as well as grey literature completed with estimations, discussions with experts and specific primary data assessment. Based on these datasets, the model is calibrated.
4. Simulation and assessment: with the compiled dataset, the current state (status quo) of the substance flows within the system is simulated. A Monte Carlo simulation allows the uncertainty of the model results to be evaluated based on the uncertainties of the parameters. The plausibility of the simulations is checked by comparing these with data from primary measurements and other studies. Furthermore, possible mitigation measures (scenarios) are simulated and evaluated for their effectiveness.