Mass Balance of Selected Pharmaceuticals in an Austrian River Catchment Area: Estimation of the Different Source Contributions

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Abstract
The study refers to the assessment of the annual mass loadings of two pharmaceuticals (PhCs), sulfamethoxazole (SMX) and carbamazepine (CBZ), released in the river of an Austrian catchment area by wastewater treatment plant (WWTP) effluent, combined sewage overflows (CSOs), surface runoff, tile drainage and deep water (referring to the sludge- or manure-amended soil).

WWTP effluent and excess sludge loadings, based on PhC national human consumption, literature excretion rates, were modeled by Activity SimpleTreat. CSO loading was modelled by MoRE. PhC load in manure was estimated on the basis of the animal annual production and literature data of concentration. Surface runoff, tile drainage and deep water loadings were modelled integrating a literature “leachate” approach with the expected PhC phenomena of accumulation, degradation and erosion occurring in the soil.

The study develops a mass balance of the different pharmaceutical loading contributions to the receiving water body by means of STAN model and highlights the uncertainties associated to the selected values in the estimation. It emerges that manure amount applied on the soil is fundamental in defining the priority contributions among the different sources (WWTP effluent, CSOs, surface runoff, tile drainage and deep water).

Keywords: Pharmaceuticals, rural catchment area, Surface runoff, treated effluent, combined sewage overflows.

1. Introduction
The water quality of a river is strictly related to the releases and withdrawals along its course. In case of a rural catchment area the contributions are due to wastewater treatment plant effluent, combined sewage overflow (CSO), surface runoff, tile drainage and deep water. If in the area, treated sludge and/or manure are applied on the soil, the quality of surface runoff, tile drainage and deep water from arable land is affected of the residues of pollutants occurring in sludge and manure. Recent studies focused on the release of some of these contributions with regard to conventional pollutants and some contaminants of emerging concern. In this context, this study wants to compare the mass loads of two pharmaceuticals released by the different sources in a rural catchment area in Austria which can be considered representative of many other study cases.

It evaluates, on an annual basis, the mass balance of carbamazepine and sulfamethoxazole starting from their human and animal consumption to the release of their residues in surface water through wastewater treatment plant effluent, combined sewage overflows, arable land runoff, tile drainage and deep water.

2. Materials and Methods

2.1. Study area
It refers to an Austrian river catchment area of 404 km², 108,000 inhabitants with extended arable lands (51 %), forests (41 %), urban areas (5.5 %) and grasslands (2.5 %). It is schematically reported in Figure 1 with the corresponding release locations in each river stretch.

Figure 1. Schematics of the catchment area flow
2.1. Evaluation of the different contributions

Pharmaceutical human consumptions are taken from Clara et al. (2013) and Kirchner et al. (2017). Excretion factors are taken as the average values from different literature data (Lienert et al., 2007). PhC mass loads in the effluent and in the excess sludge were modelled by Activity SimpleTreat model (Franco et al., 2011) which considered the characteristics of the wastewater treatments in the catchment area; PhC concentrations of the anaerobically treated sludge (used for soil amendment) were taken for literature (Verlicchi and Zambello, 2015) and its amount was provided by the local WWTP management body. The load released via CSO was estimated as a percentage of the total influent load by means of the software MoRE (Fuchs et al., 2017). PhC concentrations in manure were taken from Bloem et al. (2017), the amount produced was based on real data. It was assumed that all the collected manure is spread on the soil.

PhC mass load released via surface runoff, tile drainage and deep water was evaluated following the approach described in Langdon et al. (2010) and also considering accumulated PhC in soil and its potential degradation (Albero et al., 2018; Li et al., 2013), sediment erosion during rain events, modelled by MoRE (Fuchs et al., 2017). Surface runoff, tile drainage and deep water flow rates were evaluated by MoRE.

Statistical analysis – Mass loads were processed by STAN (Cencic and Rechberger, 2008) and estimation of each contribution is given as average value and corresponding standard deviation. An uncertainty analysis was carried out for each parameter of the model following the approach by Laner et al. (2015).

3. Results

Two different scenarios were considered for the evaluation of annual SMX mass load contribution by each source corresponding to the maximum and minimum concentration of SMX in the manure according to literature data. As to CBZ, only one scenario was considered (it is not present in manure). For each of them, the study analyzed the substance flows taking into account the relationships among the input and the sources (WWTP, CSO, surface runoff, tile drainage and deep water) which may impact the receiving river, as well as the degradation during the transport. Table 1 summarizes the main results in terms of contributions of the different sources.

<table>
<thead>
<tr>
<th>Source</th>
<th>SMX (low)</th>
<th>SMX (high)</th>
<th>CBZ</th>
</tr>
</thead>
<tbody>
<tr>
<td>WWTP effl.</td>
<td>72.7</td>
<td>8.7</td>
<td>80</td>
</tr>
<tr>
<td>CSOs</td>
<td>17.3</td>
<td>3.1</td>
<td>20</td>
</tr>
<tr>
<td>Surface runoff</td>
<td>0.4</td>
<td>22.1</td>
<td>0.004</td>
</tr>
<tr>
<td>Tile drainage</td>
<td>0.4</td>
<td>22.6</td>
<td>0.004</td>
</tr>
<tr>
<td>Deep water</td>
<td>1.0</td>
<td>43.7</td>
<td>0.02</td>
</tr>
</tbody>
</table>

The results refer to two compounds which represent two different behaviour during the treatment (different removals) and after the application on soil: SMX may be degraded and also sorbed onto particle aggregates, CBZ is a very persistent compound. It emerges that in case of low SMX load applied on soil, the contribution partition is similar to the case of CBZ. Moreover, the contribution due to CSOs is of uppermost importance for SMX and CBZ in case of heavy rain events and land runoff+tile drainage+deep water could represent priority sources of impact on surface water body in case of soil amended with manure containing a high concentration of pharmaceuticals.

References


