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## **Impacts of Hydraulic Residence Time Prediction and Diurnal Loading Pattern on the Estimation of Drug Abuse in Urban Areas**

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### **ABSTRACT**

The measurement of illicit drugs and their human urinary metabolites in influents of municipal wastewater treatment plants (WWTPs) has been recently used to estimate prohibited drug consumption in catchment communities. In this study, a preliminary estimation of the consumption of cocaine (COC) in Lynetten catchment (Copenhagen, Denmark) was performed. The estimation was done from measured levels of the biomarker benzoylecgonine (BE), its major metabolite and COC by coupling hydrodynamics and biokinetics models. To assess the impact of hydraulic residence time (HRT) prediction on COC consumption estimates, we tested two methods. The conceptual hydrodynamic catchment model was developed to estimate HRT distribution in the catchment. Additionally, a simplified scaling equation was used to estimate mean HRT. A combination of hydrodynamics model and activated sludge model for xenobiotics (ASM-X) simulation results allowed to predict the extent of in-pipe removal of COC and BE in the sewer system. The preliminary assessment results showed that COC consumption rate when mean HRT is considered was 17% lower than HRT from conceptual model. The prediction of COC consumption rates when no diurnal variation of consumption was considered showed that this assumption could impose an error up to 40% as compared to considering 6-hour resolution sampling.

### **KEYWORDS**

Urban drainage, sewage epidemiology, illicit drug consumption, ASM-X

### **INTRODUCTION**

Over the past decade, sewage epidemiology has emerged as a promising technique to provide policy makers with improved knowledge about consumption and potential abuse of illicit drugs (Daughton, 2001; Zuccato et al., 2005). This approach is based on the analysis of urinary drug biomarkers (i.e. excreted parent drugs and/or their metabolite) in sewage to estimate drug use by specific populations. Sampling campaigns for biomarkers detection and quantification are generally performed in the influent of municipal wastewater treatment plants (WWTPs). A drawback of such an approach can be that the concentration of sewage biomarkers of illicit drugs detected can be increased or reduced as compared to the discharge points due to in-sewer bio-chemical fate processes, mainly biological transformation and sorption (Plósz et al., 2013). In this sense, a number of studies (e.g., van Nuijs et al., 2012) demonstrated that illicit drugs can have comparably low stability in raw sewage. Therefore, a good understanding of in-sewer processes occurring between the point of excretion and the sampling point is essential for sewage epidemiological studies.

Sewer systems can be considered as series of interconnecting plug-flow reactors where organic and inorganic chemicals, besides transport, undergo different physical and biochemical processes. Due to the lack of knowledge of in-sewer reaction rates and absence of proper measurements, the inclusion of biochemical processes, influencing the fate of trace chemicals in urban drainage models still remains challenging. The most accepted framework for modelling biochemical processes in wastewater is the Activated Sludge Model series (Henze et al., 2000). Huisman et al. (2003) used ASM3 to describe in-sewer biokinetics for suspended and biofilm sewer biomass combined with Saint-Venant (SV) equations to describe hydrodynamics of the system. However, in complex drainage systems, such detailed models show high computational requirements. Conceptual hydrological models are therefore commonly applied due to their lower data and simulation time requirements. For instance, Vanrolleghem et al. (2005) showed how complex networks can be successfully modelled using a relatively simple structure of tanks-in-series. Similar approach is used to model the micro pollutant fluxes and their fate within Integrated Urban Wastewater and Stormwater system (Vezzaro et al., 2014). The Lynetten catchment (Copenhagen, Denmark) was simulated by applying a simple conceptual model to predict the residence time for the period when monitoring campaign were carried out. To estimate the rate of drug consumption for a long period, using daily composite sample may impose large inaccuracy on the estimation. Furthermore, estimating consumption in short period (less than a day) does not seem to be possible only with average daily samples. Ort et al. (2014) investigated the error introduced in the estimation of annual mean COC load in a small town under different sampling scenarios.

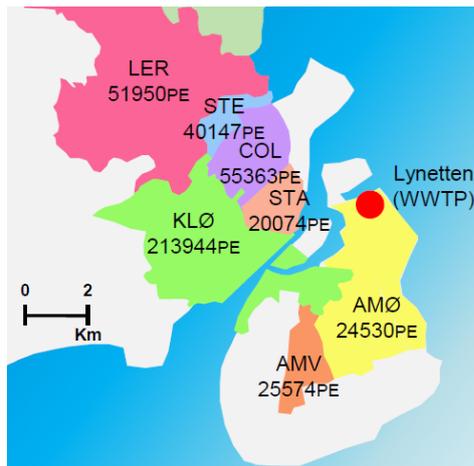
The objectives of the present study are: (i) to develop and evaluate a hydraulic conceptual model of the Lynetten sewer catchment area under dry- and wet-weather flow conditions using WEST<sup>®</sup>; (ii) preliminary prediction of the rate of consumption ( $\text{g d}^{-1} 1000\text{PE}^{-1}$ ) of cocaine (COC) in the Lynetten catchment based on the measured COC biomarker concentrations (COC and BE) in the local WWTP influent; (iii) to assess the impact of hydraulic residence time prediction on model-based back-calculation substance abuse; (iv) to assess the uncertainty introduced by omitting diurnal chemical loading variation in back-calculated consumption estimation.

## **MATERIALS AND METHODS**

### **Description of study area**

The catchment area assessed in this study is located in the city centre of Copenhagen (Denmark) and it discharges to the Lynetten WWTP. The plant has a design capacity of 750000 PE with an average daily wastewater inflow of  $175000 \text{ m}^3\text{d}^{-1}$ . The treated effluent is discharged in the Øresund strait. Lynetten catchment covers a reduced area of 7600 ha with an estimated population of 531000 inhabitants (last census: 2009)

Fig. 1 presents schematic layout of the eight sub-catchments used to describe the system along with the estimated population: Colloseum (COL), Strandvænget (STR), Lersøe (LER), Amager Vest (AMV), AmagerØst (AMØ), Kløvermarken (KLO), St. Annæ Plads (STA), Skovshoved (SKO). While the majority of the flow in the catchment is gravity driven, the inlet to the Lynetten WWTP is characterized by three pressurized pipes. In this study, due to lack of data, the SKO sub-catchment is not included.



**Figure 1.** Considered sub-catchments of the Lynetten catchment area with the respective number of inhabitants.

### Catchment model development

Runoff generation in the catchment is simulated by a three cascade reservoir approach. The average dry weather flow (DWF) for each sub catchment was calculated based on population density  $p$  (PE km<sup>-2</sup>), mean water consumption,  $\bar{Q}_{PE}$  (LPE<sup>-1</sup>d<sup>-1</sup>) and diurnal pattern of DWF based on the hourly contribution factor  $\delta_{DWF}(t)$  (where  $\bar{\delta}_{DWF}(t)=1$ ):

$$Q_{DWF}(t) = \delta_{DWF}(t) \cdot \bar{Q}_{PE} \cdot p \cdot A \quad (1)$$

where  $A$  is the catchment area in km<sup>2</sup>,  $\delta_{DWF}(t)$  was derived from the average WWTP influent DWF pattern measured over several days during dry weather,  $\bar{Q}_{PE}$  was estimated to be 215 L PE<sup>-1</sup>d<sup>-1</sup>. Data for rainfall in *mm* during rain events were retrieved from the rain gauges belonging to the Danish Water Pollution Committee network and operated by the Danish Meteorological Institute (Jørgensen et al., 1998). Runoff transportation time through each sub catchment was taken from the Lynetten catchment model in WaterAspect (Vezzaro. et al., 2013). Data for flow rates (measured at three main pumping stations) is obtained during a sampling campaign from (6/3/2013) until (14/3/2013), with the frequency of 2 minute.

A conceptual sewer model of Lynetten catchment was implemented in WEST<sup>®</sup> 2012 (DHI, Denmark). This conceptual model was calibrated against virtual data obtained from a hydrodynamic model of the catchment implemented in MIKEURBAN<sup>®</sup> (DHI, Denmark).

### Drug abuse back calculation

Values of the excreted average biomarker loads in Lynetten catchment was estimated using the in-sewer hydraulic retention time distribution obtained using the conceptual catchment model developed in WEST<sup>®</sup>. Average daily COC consumption was assessed using the back-calculated loads of COC and BE at the average discharge point and excretion ratios.

Bio- and physico-chemical processes that determine the fate of COC and BE in sewer pipes were considered to occur from the sampling point at WWTP inlet until a theoretical discharge point in the catchment. The Activated Sludge Model for Xenobiotic trace chemicals, ASM-X (Plósz et al., 2013) was used to describe biotransformation and sorption processes for COC and BE. A number of studies (e.g., van Nuijs et al., 2012) reported that, in raw and preclarified sewage, COC can biologically transform to BE. Therefore, ASM-X takes into account that COC can only be biotransformed and that BE can be both biotransformed and formed from COC:

$$\frac{dC_{COC}}{dt} = - \frac{k_{Bio,COC}}{1 + k_{D,COC} \cdot X_{SS}} C_{COC} \cdot X_{SS} \quad (2)$$

$$\frac{dC_{BE}}{dt} = \left( \frac{k_{Bio,COC}}{1+k_{D,COC} \cdot X_{SS}} \cdot \frac{M_{BE}}{M_{COC}} \cdot C_{COC} - \frac{k_{Bio,BE}}{1+k_{D,BE} \cdot X_{SS}} \cdot C_{BE} \right) \cdot X_{SS} \quad (3)$$

where  $k_{Bio,COC}$  and  $k_{Bio,BE}$  are the biotransformation coefficients ( $L \text{ gTSS}^{-1} \text{ d}^{-1}$ ) of COC and BE, respectively.  $X_{SS}$  denotes the concentration ( $\text{g L}^{-1}$ ) of TSS in raw sewage.  $M_{COC}$  and  $M_{BE}$  the molar mass ( $\text{g mol}^{-1}$ ) of COC and BE, respectively. The sorption coefficients,  $k_{D,COC}$  and  $k_{D,BE}$  ( $L \text{ gTSS}^{-1}$ ), are retrieved from Plósz et al. (2013). Considering experimental evidences by Plósz et al. (2013), no differences were considered for fate processes to occur under aerobic and anaerobic conditions. Values of consumption estimates were referenced by data obtained using a structurally different model, which were employed to predict average in-sewer HRT, developed by Plósz et al. (2013) based on the Bretting empirical formula:

$$\tau^* = \left( \frac{Q}{Q_{Average}} \right)^{-0.29} \tau_{Average} \quad (4)$$

where  $Q$  is the current/actual flow.  $\tau_{Average}$  is the mean hydraulic residence time under normal dry weather conditions and  $Q_{Average}$  is the corresponding mean dry weather flow.

The excreted concentration of BE,  $C_{BE}(t=0)$ , in the whole catchment and in the sub-catchments was calculated solving Eqs. 2 and 3, based on measured COC and BE concentrations,  $C_{COC}(t=\tau)$  and  $C_{BE}(t=\tau)$ , at Lynetten WWTP inlet (Ort et al., 2014). Daily COC consumption,  $r_{COC}$  ( $\text{g d}^{-1} 1000\text{PE}^{-1}$ ), was then estimated accounting for human metabolism (Eq. 5):

$$r_{COC} = C_{BE}(t=0) \cdot Q \cdot \frac{R}{E_{BE} \cdot P}, \quad (5)$$

where  $E_{BE}$  represents the mass of excreted drug (here BE) per mass of drug consumed (COC). Additionally,  $Q$  denotes the total flow per sampling time ( $\text{m}^3$ ) and  $P$  is the number of inhabitants in the catchment. The COC administration pattern in Denmark is reported to be mostly (85.8%) via nasal insufflation (EMCDDA, 2011), hence the  $E_{BE}$  value of 0.31 was assumed (Khan and Nicell, 2011).

The distribution of drug in sewer network is based on the assumption that there is an even probability of drug consumption in the catchment area. This simplification makes the calculation easier to estimate the average rate of COC consumption in the catchment.

### Wastewater sampling and analysis

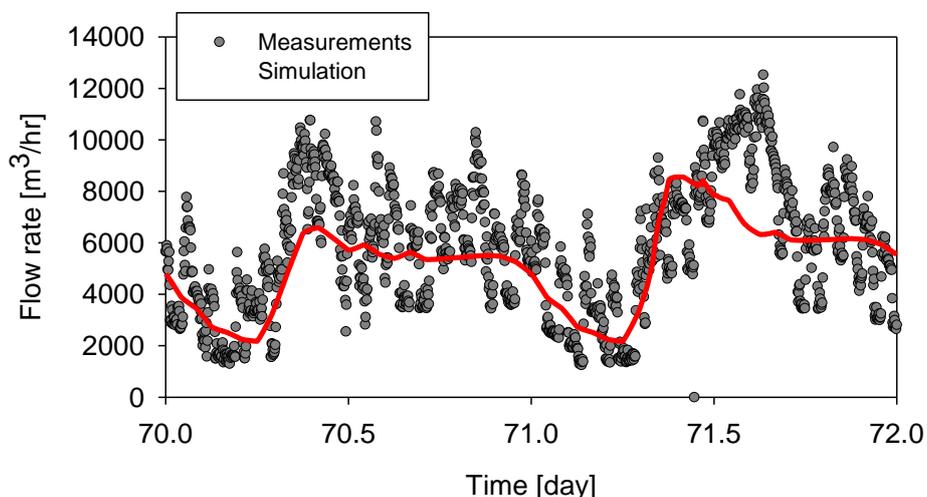
COC and BE concentrations,  $C_{COC}(t=\tau)$  and  $C_{BE}(t=\tau)$ , were measured in daily volume-proportional samples collected at Lynetten WWTP in the upstream of primary sedimentation from 6<sup>th</sup> to 14<sup>th</sup> March 2013. Sample clean-up was performed via preliminary filtration (0.7  $\mu\text{m}$ ) and solid phase extraction (SPE) on Oasis MCX cartridges (150 cc, 6 cc, Waters, Milford, USA). Sample analysis was performed via Liquid Chromatography High Resolution Mass Spectrometry (LC-HRMS). Further details on sample preparation and analysis are presented elsewhere (Ort et al., 2014).

## RESULT AND DISCUSSION

### Model evaluation

The conceptual model is calibrated against data for three rain events (June 24<sup>th</sup>–26<sup>th</sup>, 2012) with the average precipitation of 13.1 mm/day and the average run-off time of 3.6 hours among the sub-catchments. For validation, Fig. 2 shows the simulated flow rate at the influent of Lynetten WWTP for an interval (from 12/03/2013 day 70 to 13/03/2013 day 71) within the period assessed in this study. As DWF is generated in WEST<sup>®</sup> by using fixed daily flow

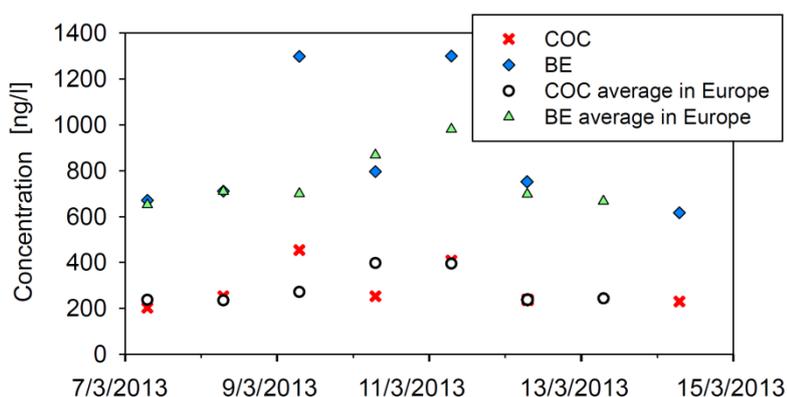
patterns, it is clear that the model cannot represent the random daily variations seen in the measured data. During the eight days of sampling campaign, wastewater flow was mainly governed by dry weather flow and 1.8 mm as an average precipitation was recorded on day 71.



**Figure 2.** Simulation result generated by WEST<sup>®</sup> and measurement data for the Lynetten WWTP influent. Dry weather flow condition for day 70 followed by rain event in day 71.

### Drug back-calculation

The levels of COC and BE a samples taken after the primary sedimentation of Lynetten WWTP are shown in Fig. 3. Data shows similar biomarker concentration patterns in Copenhagen to the averaged measured values in European countries during this period. COC measured on Friday and Sunday samples (referred to the 24-h from 07:00 to 07:00 of the following day) are approximately 2-fold increased as compared to other days in this period. The lowest concentration was measured for the last day (13/03/2013), possibly due to rain dilution.



**Figure 3.** Values of COC and BE concentration in Lynetten WWTP influent (Ort et al., 2014). Data are corresponding to day of sample collection. Measurement for 12/03/2013 in Lynetten WWTP is not available.

By inserting the calibrated parameters in Eqs. 2 and 3, and solving the equations for initial concentration, the load for COC and BE is back-calculated ( $t=0$ ). By converting the excreted COC and BE to consumed drug based on excretion ratio values ( $E_{BE}$ ), rate of drug consumption can be calculated using Eq. 5. Table 1 shows the results for such back calculation using different calculation methods: (1) HRT calculated based on the conceptual

catchment model in WEST<sup>®</sup>, (2) mean HRT obtained from Eq. 4 using average HRT of 7.84 hours found from the conceptual catchment model. Results are shown in Table 1 for all days covered by sampling campaign including dry weekdays, dry weekends and one wet weekday.

**Table 1.** Back calculation data for rate of COC consumption based on measured biomarkers BE and COC.  $r_{COC}$  (Eq. 5) is calculated based on the conceptual catchment model (method 1). (\*) indicates that the calculation is done based on mean HRT (method 2) in which  $\tau^*$  is obtained from Eq. 4. Deviation is the difference between  $r_{COC}^*$  and  $r_{COC}$ . Dates are indicating the day of sample collection.

Date	Flow condition	$r_{COC}$ (g d <sup>-1</sup> 1000PE <sup>-1</sup> )	Measured WWTP influent flowrate (L/s)	$\tau^*$ (h)	$r_{COC}^*$ (g d <sup>-1</sup> 1000PE <sup>-1</sup> )	Deviation (%)
07-03-2013	Dry weekday	0.70	1573	7.774	0.641	-8.46
08-03-2013	Dry weekday	0.72	1595	7.744	0.670	-6.51
09-03-2013	Dry weekday	1.07	1528	7.841	1.173	9.36
10-03-2013	Dry weekend	0.82	1477	7.918	0.704	-14.15
11-03-2013	Dry weekend	1.35	1456	7.951	1.135	-15.67
12-03-2013	Dry weekday	0.77	1519	7.854	0.678	-11.38
14-03-2013	Wet weekday	0.78	1678	7.631	0.607	-22.04

The calculation reveals that COC to BE ratio from the point of measurement to excretion point has increased almost 47% in both back calculation methods. This is due to biotransformation of COC to BE, which leads to the possibility of a net production of BE in the sewer system (Plósz et al., 2013).

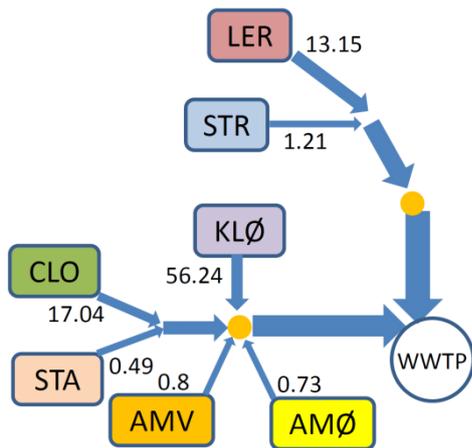
It should be noted that the accuracy of Eq. 4 depends highly on good estimation of mean residence time for the entire catchment. Table 1 indicates that the simplified model can give reasonable prediction under dry-weather flow conditions (~22% and 15% underestimation of the rate of COC consumption under wet-weather and weekends, respectively).

Furthermore, mass of COC use can be estimated as a daily average value using the catchment model.

The catchment model can also be used to back calculate the mass of COC released to the sewer from each sub-catchment. Fig. 4 shows such estimation when total mass load of COC in the catchment is 93.78 g d<sup>-1</sup>, as the average mass load during dry weekdays of sampling campaign. However, this is not the primary purpose in this study as more measurements are required upstream of the catchment to evaluate the spatial distribution of biomarker load in the sub-catchment.

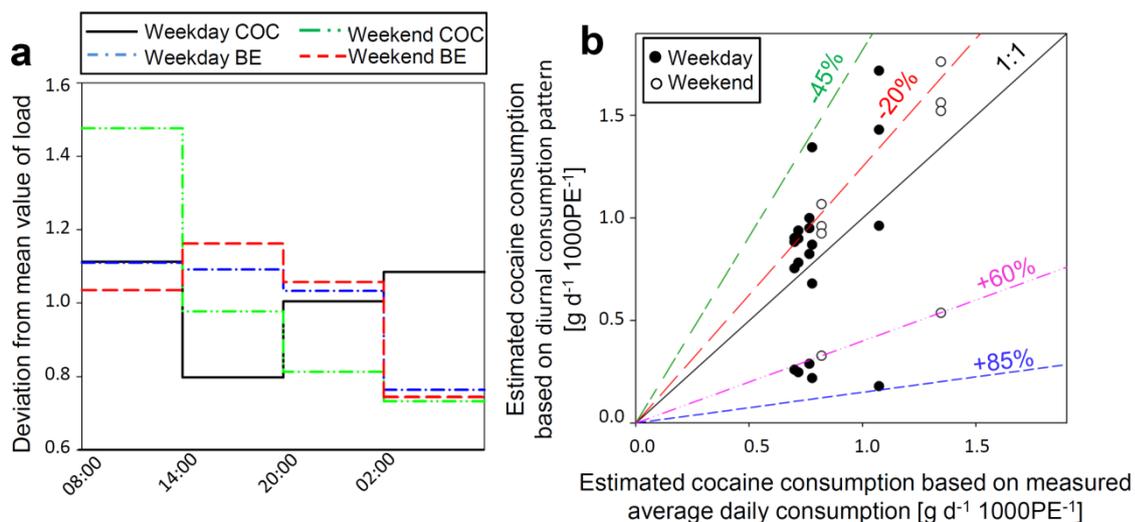
Back calculation of drug consumption based on daily composite sample may cause over or under estimation of real average daily consumption. In this study, the effect of diurnal consumption pattern for COC and BE on back calculation is assumed to follow the same consumption pattern for these drugs as in Reid et al. (2011), who reported diurnal variation of COC and BE with temporal resolution of 6 hours for 4 weeks in Oslo. The distribution is done based on the load of COC and BE received at the WWTP. By implementing the load-based distribution pattern (6-h resolution) reported by Reid et al. (2011), the effects of flow rate variations and drug level variations at WWTP on the back-calculated abuse rate is assessed.

Fig. 5a reports the deviation of biomarker loads from mean value for each 6-h period (i.e. the ratio between the load in each 6-h period and the daily average load) for COC and BE during weekdays and weekends.



**Figure 4.** Estimation of the total mass of COC use by summing up the back calculated mass of consumption [ $\text{g}\cdot\text{d}^{-1}$ ] for each sub-catchment.  $93.78 \text{ g}\cdot\text{d}^{-1}$  COC is considered as the average value during dry weekdays of sampling campaign.

In this approach  $r_{\text{COC}}$  can be estimated at higher resolution compared to when only daily average concentration is considered. A comparison of the two approaches is reported in Fig. 5b. Deviation in  $r_{\text{COC}}$  estimation using 6-h resolution measured concentrations ranged from approximately -45% to +85% when only one daily concentration was considered. Overall, these results suggest that sampling at higher than daily frequency is required for more accurate estimation of drug use rate.



**Figure 5.** Deviation from mean value of daily COC load for each 6-h period. Reported values are based on a 30-d sampling campaign in VEAS WWTP (Oslo, Norway; Reid et al. 2011)(a). Comparison of  $r_{\text{COC}}$  prediction in Lynetten catchment considering (i) 24-h average concentration (ii) diurnal (6-h average) variation in the occurrence of biomarkers. Continuous and dashed lines represent the deviation in  $r_{\text{COC}}$  predictions (b).

## CONCLUSIONS

A sewer model for the catchment area of Lynetten is developed in WEST to predict the hydraulic residence time distribution in the sewer network. ASM-X is used to simulate biotransformation and sorption of chemicals, and thereby back calculate the rate of consumption of COC based on its metabolite BE and COC.

The results for back calculation show that by using mean HRT, the consumption rate of COC deviates up to 22% compared to the consumption rate when HRT from calibrated conceptual catchment model is used. However, this is a comparably low uncertainty. Although the focus

is on dry weather flow, the catchment model can also perform well in wet weather flow conditions. Incorporating diurnal variation of drug use suggest that considering constant drug use pattern can lead to significant underestimation(45%) or overestimation(85%) of the rate of drug consumption.

## FUTURE PERSPECTIVES

In this study, consumption of the illicit drug COC in a large catchment area was estimated with the aid of hydrodynamic models (describing in-sewer transport), fate models (describing in-sewer biochemical processes) and analytical measurements. This can be considered as a preliminary estimation as a number of simplifying assumptions were considered such as even probability distribution of COC consumption in the catchment area. Our ongoing research is focusing on reducing uncertainty in the model-based prediction notably by assessing the distribution of biomarker load upstream to sampling point and by further developing ASM-X.

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