Substance flow analysis as a tool for urban water management

ABSTRACT

Human activity results in the production of a wide range of pollutants that can enter the water cycle through stormwater or wastewater. Among others, heavy metals are still detected in high concentrations around urban areas and their impact on aquatic organisms is of major concern. In this study, we propose to use a substance flow analysis as a tool for heavy metals management in urban areas. We illustrate the approach with the case of copper in Lausanne, Switzerland. The results show that around 1,500 kg of copper enter the aquatic compartment yearly. This amount contributes to sediment enrichment, which may pose a long-term risk for benthic organisms. The major sources of copper in receiving surface water are roofs and catenaries of trolleybuses. They represent 75% of the total input of copper into the urban water system. Actions to reduce copper pollution should therefore focus on these sources. Substance flow analysis also highlights that copper enters surface water mainly during rain events, i.e., without passing through any treatment procedure. A reduction in pollution could also be achieved by improving stormwater management. In conclusion, the study showed that substance flow analysis is a very effective tool for sustainable urban water management.

Key words | heavy metals, sustainable water management, substance flow analysis, urban water cycle

INTRODUCTION

The presence of pollutants in water around urban areas has been highlighted by several researchers in the last few decades (Dewhurst et al. 2003; Bester et al. 2008). Indeed, it is now recognised that human activity is a source of many chemicals: pesticides, pharmaceuticals, heavy metals, etc. that can enter the water cycle through stormwater (Burton & Pitt 2002) or wastewater (Ternes & Joss 2006). Many of these substances are of major concern regarding their possible long-term impacts on both humans and the environment (Kümmerer 2001; Williams 2005). They can also re-enter the city via drinking water, which may pose long-term human health problems. It is therefore both vital and urgent to acquire tools that facilitate sustainable urban water management in terms of chemicals.

Recently, Huang et al. (2007) proposed using substance flow analysis (SFA) for phosphorus management in a large city. This kind of analysis is an extension of material flow analysis (MFA), originally developed in the 1930s in the economic sector (Leontief 1936) and later adapted to regional investigations by Baccini & Brunner (1991). Studies in the Bünz valley, Switzerland, and in the Danube Basin, Austria, demonstrated its potential as a basis for river basin pollution control regarding nutrients (Lampert & Brunneer 1993; Sarikaya et al. 1999; Somlyódy et al. 1999; Zessner & Kroiss 1999). In the last decade the approach was further developed to allow modeling with little data, but more profound system knowledge (mathematical material flow analysis; MMFA; Baccini & Bader 1996). Measurements, literature values and estimations/plausible reasoning were used as input data. Since then, MMFA has been applied in many studies in various fields such as energy (Bader et al. 2007) or substance behaviour at a national level (Kwonpongsagoon et al. 2007; Eriksson et al. 2008; Morf et al. 2008). However, very few studies have used MMFA/SFA to model chemicals...
in the urban water cycle (Seelsaen et al. 2007; Jonsson et al. 2008; Månsson et al. 2008).

Basically, a MMFA/SFA describes and quantifies the material flows through a defined system. 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 can be the urban water defined by the sewer catchment and the receiving water, which also serve as recreational areas and as a source of drinking water. To be used as a management tool, SFA should be coupled with water system quality criteria (Baccini & Brunner 1999). These values express the maximum concentration which is tolerable for a given substance in order to protect both humans and the environment. Having this limit in mind, one can detect the most problematic flows and take action to diminish them.

In this study, we applied SFA to copper in the city of Lausanne, Switzerland. Lausanne is a city of around 130,000 inhabitants situated in the south-west of Switzerland on Lake Geneva. The lake is a place of recreation (swimming, sailing) as well as an important source of drinking water and of fishing activities. It is thus a valuable ecosystem for biodiversity. It is therefore crucial to monitor the water quality and to ensure sustainable water protection and management. Among other chemicals, copper has been detected in high concentrations in sediment from the Vidy Bay near Lausanne (Wildi et al. 2004) and in water of the same area during rain events (Rossi 1998). Copper is also suspected to induce toxicity of sediments, which may affect the species living in the Bay (Wildi et al. 2004). In this study, we will estimate the SFA for copper and validate it with the large number of measurement data available for the catchment.

**METHODOLOGY**

The SFA proposed in this study is carried out in six iterative steps: (i) the system analysis, (ii) the description of the equations, (iii) the data acquisition and calibration, (iv) the current simulation, (v) the sensitivity analysis, and (vi) the simulation of the scenarios. More information can be found in Schaffner et al. (2009).

**System analysis and model approach**

The SFA is applied to the city of Lausanne, which is around 42 km² with 130,000 inhabitants. The system encloses the sewer catchment as well as the receiving water directly near the city (Vidy Bay). The Vidy Bay is around 1.32 km² (Poté et al. 2008) and the volume of water is estimated to be 1.32 × 10⁷ m³ (average depth of 10 m). The system itself is described in Figure 1. A one-year period was considered for the analysis.

We considered thirteen inputs of copper (Table 1). The input through drinking water (I₁) reaches the wastewater drainage system directly. The inputs through roof (I₂) and house-side (I₃) runoff, through road runoff-collecting car brakes (I₄), tire (I₅) abrasion as well as motor oil residues (I₆) and dry deposition (I₁₂), through particles from catenaries from trolleybuses (I₇) and through rainwater (I₁₃) reaching the stormwater drainage system. The inputs through trains, i.e., particles from catenary (I₈), brake (I₉) and wheel (I₁₀) abrasion, reach the train drainage system before going partly to wastewater and partly directly to surface water. Finally, inputs through boats (antifouling treatments, I₁₁) directly enter surface water.

The main compartments of the system were identified as the drainage systems (for wastewater, for stormwater and for train runoff), the three combined sewer overflows on the wastewater sewer system, the wastewater treatment plant (WWTP), the sludge collector, the surface water and the sediment. Note that the first combined sewer overflow (CSO₁) integrates all the overflows occurring in the urban catchment before the WWTP. The two other CSOs (CSO₂ and CSO₃) are situated at the entry of the WWTP and directly after the first clarifier.

For this first approximation and based on current system knowledge, a linear input-output function is applied to describe the system mathematically. Such models are characterised by given input and input-output relations, namely the flows, between the different compartments. These flows are related to the total inputs of the corresponding compartments by transfer coefficients (k₁ to k₁₄). We considered no output of the system; copper ends in sludge or in sediment. There is therefore an enrichment of the compartment sludge and sediment with time.

**Input data and transfer coefficient**

The **input data** as well as their sources are summarized in Table 1. All input data were collected from previous studies (Rossi 1998; Rossi et al. 2008), official websites or estimated (for more information see Guignard 2008). For all the data, we considered a one-year period of input, mostly representative of the 10 last years. Note that we considered the total copper entering the system, i.e., we did not differentiate between adsorbed and dissolved copper.
The transfer coefficients are also estimated from previous studies or official websites (Table 2; for more information see Guignard 2008).

### Uncertainties of input data and transfer coefficients

The uncertainties of the input data were calculated or fixed based on literature dependent on the available information. For input through roofs ($I_2$), the standard deviation was evaluated to 47% and for input through brakes ($I_4$), to 49%. Both these values were calculated with the minimal and maximal values found in the literature (Table 1). The standard deviation linked to the input through tires ($I_5$) was very high (150%) due to the wide range of data found in the literature (Table 1). For all the other parameters, the standard deviations were estimated to be 50%, according to Lindblom et al. (2007). The probability density distribution is assumed to be lognormal for all input flows and truncated lognormal for the transfer coefficients (Schaffner et al. 2009).

### Table 1 | Inputs of Cu [kg/year] into the urban water cycle in Lausanne. The mean values used in the substance flow analysis are given in fourth column, followed by the standard deviation and its probability density distribution assumed for each input

<table>
<thead>
<tr>
<th>Input loads</th>
<th>Mean value</th>
<th>Standard deviation</th>
<th>Density</th>
</tr>
</thead>
<tbody>
<tr>
<td>$I_1$: Drinking water</td>
<td>19.3 kg/year</td>
<td>±50%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_2$: Roofs</td>
<td>837 kg/year</td>
<td>±47%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_3$: House sides</td>
<td>56 kg/year</td>
<td>±50%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_4$: Brakes</td>
<td>137 kg/year</td>
<td>±73%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_5$: Tires</td>
<td>7.1 kg/year</td>
<td>±150%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_6$: Motor oil</td>
<td>0.15 kg/an</td>
<td>±50%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_7$: Trolleybus</td>
<td>670 kg/year</td>
<td>±50%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_8$: Catenaries</td>
<td>190 kg/year</td>
<td>±50%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_9$: Brakes</td>
<td>48.5 kg/year</td>
<td>±50%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_{10}$: Wheels</td>
<td>1.8 kg/year</td>
<td>±50%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_{11}$: Boats</td>
<td>42.3 kg/year</td>
<td>±50%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_{12}$: Dry deposition</td>
<td>30.7 kg/year</td>
<td>±49%</td>
<td>lognormal</td>
</tr>
<tr>
<td>$I_{13}$: Rain</td>
<td>18.5 kg/year</td>
<td>±50%</td>
<td>lognormal</td>
</tr>
</tbody>
</table>
Table 2 | Transfer coefficients [%] through the different compartments. The standard deviation and its probability density distribution is given in the two last columns for each transfer coefficient

<table>
<thead>
<tr>
<th>Transfer coefficient</th>
<th>Standard deviation</th>
<th>Density</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_1 = 1 - k_2$: stormwater joining the combined sewage system</td>
<td>50% ±20%</td>
<td>tnormal</td>
</tr>
<tr>
<td>$k_3 = 1 - k_4$: stormwater from railways joining the combined sewage system</td>
<td>35% ±20%</td>
<td>tnormal</td>
</tr>
<tr>
<td>$k_5 = 1$: wastewater going to CSO$_1$</td>
<td>100% ±20%</td>
<td>tnormal</td>
</tr>
<tr>
<td>$k_7 = 1 - k_6$: wastewater going from CSO$_1$ to CSO$_2$</td>
<td>96.6% ±20%</td>
<td>tnormal</td>
</tr>
<tr>
<td>$k_9 = 1 - k_8$: wastewater going from CSO$_2$ to CSO$_3$</td>
<td>94.9% ±20%</td>
<td>tnormal</td>
</tr>
<tr>
<td>$k_{11} = 1 - k_{10}$: wastewater going from CSO$_3$ to WWTP</td>
<td>80.9% ±20%</td>
<td>tnormal</td>
</tr>
<tr>
<td>$k_{12} = 1 - k_{13}$: transfer to sludge</td>
<td>75% ±20%</td>
<td>tnormal</td>
</tr>
<tr>
<td>$k_{14}$: transfer to sediment</td>
<td>60% ±20%</td>
<td>tnormal</td>
</tr>
</tbody>
</table>

The standard deviations of the transfer coefficients were estimated to be 20% based on expert judgment and previous measurements in the urban water system (Rossi 1998; Service de l’Assainissement, Lausanne, personal communication). The probability density of the distribution is assumed to be truncated normal between 0 and 1 (Schaffner et al. 2009).

Current state simulation and sensitivity analysis

Modelling was performed by using the software SIMBOX® developed at the Swiss federal institute for aquatic sciences (Eawag), Dübendorf, Switzerland.

Water quality criteria

A value of 8.2 µg/L was proposed as water quality criterion for dissolved copper under the EU Water Framework Directive (Crane & Babut 2007). In the US, the water quality criteria for copper is expressed as a function of pH, hardness and dissolved organic carbon (DOC) in water. The values vary between around 2 and 200 µg/L (Reiley 2007). Swiss legislation (Swiss Government 1998) gives the criteria of 5 and 2 µg/L for the total and dissolved copper, respectively.

For sediment, MacDonald et al. (2000) propose a threshold effect concentration (TEC) of 31.6 mg/kg dry weight and a probable effect concentration (PEC) of 149 mg/kg dry weight. The first value represents a concentration below which harmful effects are unlikely to be observed, and the second a concentration above which harmful effects are likely to be observed.

RESULTS AND DISCUSSION

SFA of copper in Lausanne

Figure 2 illustrates the results of SFA of copper applied to Lausanne city. The yearly input is estimated to be around 2,000 kg/year. The roofs as well as catenaries from trolley-buses are clearly identified as the major sources of this heavy metal in the urban water system. The mass of copper issued from these two inputs reaches 1,500 kg/year. The third and the fourth main sources are car brakes and catenaries from trains, which contribute about 300 kg/year. These four sources were also highlighted as major sources of emission in Stockholm (Bergbäck et al. 2001). The other inputs are more than ten times lower than the major ones. Inputs through car tires, motor oils as well as train wheels seem to be negligible in comparison. In particular, the input through car tires is twenty folds lower than the input through car brakes. This ratio (input through car tires/input through car brakes) is lower than the ratio estimated from Hjortenkrans et al. (2007) in Stockholm, Sweden. One explanation could be the overestimation of the load through car tires, as we considered that the average of 6,000 vehicles per day in Lausanne are driving on the total of the road network.

The main source of entry of copper in urban water is therefore through stormwater. A large amount of this substance, around 880 kg/year, directly reaches surface water without treatment and thus contributes to an enrichment of surface water and sediment in copper. This result is in accordance with a SFA analysis conducted in Australia, where the main source of water contamination by copper was road stormwater (Seelsaen et al. 2007). Wastewater and part of the stormwater reach the wastewater treatment plant (WWTP), where copper is mainly retained in sludge. The discharge of copper through the WWTP is five times lower than that through stormwater, reaching 180 kg/year. Note that the same amount of copper is released through CSO directly to surface water. This point highlights the...
importance of monitoring CSO discharges as they may contribute significantly to surface water pollution.

Based on these results, we can estimate an amount of around 1,500 kg/year of copper entering the Vidy Bay, of which around 910 kg/year ends in sediment. By extrapolating this result to the entire watershed of the Vidy Bay, i.e., Lausanne and surroundings (factor 1.8; Rossi 1998) and by considering an accumulation rate of 0.5 g dry weight cm$^{-2}$ year$^{-1}$ (Poté et al. 2008), we can estimate an average concentration in the sediment of 250 mg/kg dry weight. This value is above the probable effect concentration of 149 mg of copper/kg dry weight proposed by MacDonald et al. (2003). The input of copper through the urban catchment could therefore induce toxicity for the organisms living in the Bay.

Of the 1,500 kg of copper which enters the Bay yearly, around 600 kg/year is dissolved in water, and 95% of these inputs occur during rain events. Assuming 100 rain days in Lausanne per year, we can roughly estimate an average of 10 kg copper/event reaching surface water (always with the factor of 1.8 to extrapolate to the whole catchment). By dividing this load by the volume of water in the Bay (1.32 × 10$^7$ m$^3$), we obtain an average concentration of 0.8 µg/L of copper in the surface water at the end of the rain events. This concentration is lower than the water quality criteria (for example 2 µg/L for dissolved copper in Switzerland). However, as most of the pollution occurs in the first few hours of the event, we can expect water quality limits to be exceeded at the beginning of the rain event, which also may induce toxic effects for the aquatic organisms.

**Comparison of predictions with field measurements**

The results of SFA were compared with real measurements. It turns out that the modelled and measured data were of a similar order of magnitude. Indeed, we estimated a concentration of 250 mg/kg dry weight in sediment (see above). This value is within the range of the measured concentrations reported by Poté et al. (2008), which vary between 33 to 727 mg/kg in sediment of the Vidy Bay. We also compared the estimated accumulation of copper in sediment of the Vidy Bay with measurements published elsewhere. An enrichment of 87.8 tons of copper was estimated for a period of 24 years (Poté et al. 2008). In this study, the estimated accumulation of copper during the same period is lower, corresponding to 39 tons (extrapolation factor to the entire watershed of the Vidy Bay: 1.8), which is lower than the estimation by Poté et al. However, Loizeau et al. (2004) showed that the concentrations of copper in cores of sediment from the Bay have decreased during the last
decade (the highest concentrations being measured in the 1970s). Our estimations, being based on current sources and concentrations of copper, may explain the lower load calculated in this study.

A concentration of 0.67 to 2.11 g of copper per imperious hectare and millimetre of rain (g/ha/mm/mm) were measured in stormwater runoff in Lausanne (separated sewer system catchment; (Rossi 1998). Assuming an average of 1,122 mm of rain per year, the catchment of 42 km² considered in this study and 50% of stormwater reaching the aquatic environment directly (k₂), we can estimate a copper load between 1,600 and 5,000 kg per year for stormwater runoff. The load estimated in this study, around 1,100 kg/year, is in the same order of magnitude. These results confirm the validity and the plausibility of the hypotheses made in our study.

Comparison of predictions with data from literature

To compare with literature data, we calculated the yearly copper load per person. We estimated a yearly load of 11.6 g Cu/Cap for input to the aquatic compartment, 7 g Cu/Cap reaching the sediment. A yearly amount of copper to surface water of 54 g/hab was reported for a catchment area near Sydney, Australia (Seelsaen et al. 2007). For the Stockholm area, a yearly load of 17.1 g Cu/Cap was estimated for input to surface water and soil, and of 2–6 g/Cap for input to sediment (Bergbäck et al. 2001). In the same area Lindström et al. (2001) estimated 10 g Cu/Cap for input to sediment. The load of copper entering surface water and sediment of this study are therefore in good agreement with similar estimations in the Stockholm area. The input to surface water in the Australian study is much higher. A reason might be the high content of copper in fuel additives reported in this specific study.

Sensitivity analysis

Sensitivity analysis allows identifying the sensitive parameters of the system variables and, in particular, of the key variables considered. The two key variables in this study are the net input to surface water and the accumulation rate to sediment. For these two variables, the most sensitive parameters are: I₂ (roofs), I₇ (trolleybus catenaries) and k₂ (transfer coefficient from stormwater to separate sewer system; results not shown here). Field measurements campaigns focusing on these specific inputs and this specific coefficient would lead to a better approximation of the annual balance of copper in Lausanne.

Uncertainty analysis

The uncertainty of the parameters have been estimated as described above. According to the data available on the parameters, either a lognormal, a normal or a truncated normal density distribution have been assumed (see Tables 1 and 2). Based on these probability density distributions, the probability distribution of the variables were calculated using the Monte Carlo Calculation with a sample size of 100,000. The results show that the net input to surface water varies between 350 and about 954 kg/year (5–95% confidence interval) and the accumulation rate in the sediment between around 560 and 1,385 kg/year (5–95% confidence interval) when all the estimated uncertainties are included. These ranges of values are also in agreement with the field measurements.

Discussion of the hypotheses

Some hypotheses support the SFA presented in this paper and they may be discussed. One critical hypothesis is linked to the assumption that no infiltration of stormwater occurs in the city, i.e., we assume that the surface is imperious. This is not true as the city of Lausanne has many parks and gardens. However, as the city is on a slope (between around 400 and 900 m above sea level), we can argue that stormwater rapidly joins the sewer system, especially because the soils are drained.

We also assume no resuspension of copper from sediment to water. Such a transfer may occur during stormwater events and is certainly important during storms. It is, however, punctual and such a transfer of copper hasn’t been considered for a one-year period.

CONCLUSIONS

Coupled with water and sediment quality criteria, SFA is an interesting management tool as it allows detecting the main problems in a system (for example the main sources of pollution). It also allows studying different scenarios when action is planned. However, its application is based on several hypotheses and estimations, which should be justified by, for example, point measurements in the system. In this particular study, the results show that copper might represent a risk for the aquatic environment of the Vidy Bay, especially for the sediment compartment. We have also shown that stormwater presents the major source of contamination. There is therefore a need to take action to
reduce this pollution in the city of Lausanne. Different technical solutions, from at-source solutions (infiltration of stormwater, with or without adsorption material) to end-of-pipe solutions (detentions ponds, constructed wetlands) have already been proposed in Switzerland. The next step would therefore be to implement different scenarios in the model to help choose the most adequate investments to limit copper contamination of the Vidy bay.

ACKNOWLEDGEMENTS

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