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Efficiency of source control systems for reducing runoff pollutant loads: Feedback on experimental catchments within Paris conurbation

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Highlights:

SUDS designed for peak flow control were monitored (flow, organic pollutants, metals)

Volume and contaminant loads were reduced by all SUDS even for ordinary events

Efficiency of SUDS depends strongly on the type of storage and its general conception

Strong reliance between pollutant mitigation and water volume reduction

Runoff reduction-oriented design of SUDS: an efficient solution for pollutant mitigation

Abstract:

Three catchments, equipped with sustainable urban drainage systems (SUDS: vegetated roof, underground pipeline or tank, swale, grassed detention pond) for peak flow mitigation, have been compared to a reference catchment drained by a conventional separate sewer system in terms of hydraulic behaviour and discharged contaminant fluxes (organic matter, organic micropollutants, metals). A runoff and contaminant emission model has been developed in order to overcome land use differences. It has been demonstrated that the presence of peak flow control systems induces flow attenuation even for frequent rain events and reduces water discharges at a rate of about 50 % depending on the site characteristics. This research has also

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demonstrated that this type of SUDS contributes to a significant reduction of runoff pollutant discharges, by 20 % to 80 %. This level of reduction varies depending on the considered contaminant and on the design of the drainage system but is mostly correlated with the decrease in runoff volume. It could be improved if the design of these SUDS focused not only on the control of exceptional events but also targeted more explicitly the interception of frequent rain events.

Keywords: flooding source control system; frequent rain event; micropollutants; stormwater; SUDS

1. Introduction

Stormwater management has become a critical issue in the field of sustainable urban development to protect civil society against flood and because runoff on urban surfaces has been recognised as a major cause of the degradation of receiving waters (Burton and Pitt, 2001). In the past, stormwater was collected by drainage networks, but with fast urbanization these networks have become inadequate, leading local authorities to develop strategies to prevent flooding.

The first strategy adopted was the large-scale management of urban drainage systems by building large reservoirs. It was not sufficient to remove the flooding risks and now a local stormwater management approach is preferred (Brombach *et al.*, 2005; Ellis and Revitt, 2010; Jefferies *et al.*, 2009; Roy *et al.*, 2008). In recently urbanised areas, facilities are developed simultaneously to the urban growth promoting retention or infiltration at a small scale. These facilities are often called “Sustainable Urban Drainage Systems” (or SUDS). Two major types of SUDS design are used worldwide: flow rate regulation and volume regulation. Both in France and in the USA, the most widespread regulation is based on a limited flow rate value (Petrucci *et al.*, 2013; Roy *et al.*, 2008). For example in the French department Seine Saint-

Denis, in the suburb of Paris, the local authorities have imposed a flow rate regulation at 10 l/s/ha since 1993 (DEA, 1992). Thus SUDS are typically intended to facilitate hydraulic management and have been designed for exceptional precipitation events; only on rare occasion are contamination mitigation objectives actually addressed (Martin *et al.*, 2007).

Studies have revealed that such SUDS are capable of: reducing the discharged volumes, delaying catchment response, slowing flow velocities and increasing water residence time within the various facilities (Jefferies *et al.*, 2004; Scholes *et al.*, 2008). Thus they can have a substantial impact on the pollutant fluxes being conveyed by stormwater and discharged into receiving waters. Purifying effects have indeed been observed at the system scale for several types of SUDS (Jefferies *et al.*, 2004; Pagotto *et al.*, 2000; VanWoert *et al.*, 2005). However, there are few studies highlighting the overall effect of SUDS on pollutant fluxes control, at a suburban catchment scale. The effect of SUDS that were designed for flow control and not pollutant control remains poorly documented. Moreover literature data is usually limited to metals and nutrients and few data is available on organic micropollutants (Diblasi *et al.*, 2009; Matamoros *et al.*, 2012).

Therefore, the objective of this research is to assess the effect of peak flow control policies, on the water and contaminant flows discharged during frequent rain events at a small catchment scale. A special attention has been given to a selection of priority substances listed in the Water Framework Directive (2000/60/EC), whose presence is significant in runoff (Bressy *et al.* 2012), but whose fate in SUDS is not much documented to date. Three catchments containing SUDS were compared to a reference catchment featuring a conventional separate sewer network, in terms of hydraulic behaviour and discharged contaminant fluxes (i.e., suspended solids (SS), organic carbon (OC), trace metals (copper, lead, zinc) and organic micropollutants: polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and alkylphenols). Moreover, the deposits formed in storage zones were characterised so as to better understand the fate of micropollutants during their transfer and in order to devise the best strategy for recovering and treating these wastes.

2. Materials and methods

2.1 Site characterisation

A residential site, characterised by low-density traffic and no industrial activity within a 5-km radius, was studied in a suburban area near Paris (France). The site was drained by a separate sewer system. Land use on this site was quite homogenous, while the stormwater management system featured a wide diversity.

On this site, four small catchments ranging from 0.8 ha to 1.9 ha were studied. The "Reference" catchment was drained by a conventional separate sewer system, while the other three catchments ("North", "Park" and "South") temporarily stored stormwater in various SUDS to comply with the 10 l/s/ha flow limitation imposed by local authorities. Stormwater on the North catchment was stored in a vegetated roof and in an underground pipeline for common rain events (up to 1 year return period) with an overflow onto a swale or on parking for exceptional events. In the Park catchment, stormwater was stored in a grassed detention pond that is part of a public garden. Stormwater management on the South catchment had been incorporated into the land use plan and the practices associated various types of storage facilities: underground tank for private parcels, swales and a public square covered by grass. The outlets of the catchments with SUDS are fitted with flow-rate regulators as usual in France (Table 1). According to Martin *et al.* (2007), these SUDS were representative of the kinds of solutions adopted in France.

The characteristics of the catchments are listed in Table 1. The four catchments displayed a homogeneous pattern of urbanisation and were located adjacent to one another (less than 400 m between two catchments), ensuring a relative homogeneity as regards atmospheric contributions (i.e. rainfall and deposits). Differences in land use breakdown appeared across these four catchments. The breakdown of the North catchment was similar to the Reference, though automobile traffic was heavier on North due to the presence of retail shops. The Park catchment was mainly composed of buildings and gardens and contains no streets. The South catchment was relatively devoid of streets and contained a higher density of pedestrian paths than the Reference. Consequently, the discharges for these four catchments could not be directly compared and required introducing a set of land use-based modelling tools.

With the objective of establishing a model, the potential contaminant entry paths (atmospheric fallout, pavement runoff and runoff from built parcels) were also examined. Atmospheric fallout was measured on the flat roof of the highest building within the study area. Both types of runoff (street and built parcel, i.e., roof and a private garden above slab underground parking) were evaluated on the Reference catchment. The characteristics of these two sub-catchments are provided in Table 1 and described with greater details in Bressy *et al.* (2011).

2.2 Rainfall and flow measurements

Rainfall and flows at the four catchment outlets were continuously measured (every 0.2 mm for the rain and every minute for the flow) for one year between July 2008 and August 2009.

2.2.1 Instrumentation

Rainfall depth was recorded using a rain gauge (3029, Alcyr) placed on a flat roof in the study area. Runoff flows were measured at the Reference catchment outlet with a Sigma 950 flow-meter (water depth with a bubble pipe and velocity by Doppler Effect). At the source-controlled catchment outlets, runoff flows were measured just beyond the flow regulation device with Sigma 950 flow-meters, by recording the water depth upstream of a V-notch weir.

2.2.2 Definition of a rain event

A rain event was defined as any precipitation leading to a flow signal. The beginning of an event was the time of the first precipitation data point during the 20 minutes preceding initiation of the flow signal for the Reference site and during the preceding 3 hours for the other three sites. The end of the event was defined as the time of the last precipitation data point during the flow signal period.

The following parameters were determined for each rain event:

- Peak flow: Q_{max} (in l/s/ha) was the maximum flow value during the rain event;
- Lag time: T_{lag} (h) was the time delay between maximum precipitation intensity and peak flow signal;
- Emptying time: T_{empty} (h) was the time delay between the end of precipitation and the end of the flow signal;
- Runoff water depth: H_{runoff} (mm) was the effective water depth discharged by the catchment during the rain event.

2.3 Sampling protocol and analytical procedure

2.3.1 Water sampling protocol

Both dry and wet bulk atmospheric depositions were sampled using 20-L bottles hermetically connected to a 1-m² stainless pyramidal funnel. The bottles were placed underneath the funnel just before the rain event and removed just afterward; they collected the wet deposition and the washoff of the contaminants deposited on the funnel during the previous dry weather period. Stormwater was collected from the storm sewer at the catchment outlet using automatic samplers (Bühler 1029) controlled via the flow meter. The sampling protocol was flow-proportional so as to obtain average concentrations throughout the event.

The campaigns conducted in order to analyse both organic contaminants (requiring the use of glass bottles) and metals (plastic bottles) were based on different sets of events. SS and organic matter were measured for all the events in glass or plastic bottles, but in this paper only SS and TOC data for rain events sampled simultaneously on atmospheric fallout, built parcel and street catchments were used. Table 2 provides the characteristics of the rain events considered for each parameter.

2.3.2 Soil and sediment sampling protocol

Sediment deposits were observed and sampled both in the North catchment storage pipe and upstream of the South catchment regulator. Several samples were collected in order to constitute a representative average sample of the sediment deposit. Average samples were reduced by quartering steps after homogenization. The soil of the public garden used as storage on Park catchment was also sampled. The retention basin surface was divided into three areas according to flooding frequency: one flooded at each rain event, another occasionally flooded and the last area was very seldom flooded. To constitute the average soil sample, 4 to 6 samples were collected in each area using a (20-cm long) corer, in following a 7-m^2 mesh grid pattern, and then combined. One sample was analysed in triplicate for a variability assessment: the signal deviation was below 20%.

2.3.3 Micropollutant analysis

The analytical procedures applied for organic compounds were previously described by Bressy *et al.* (2012). Briefly, it was based on separating the dissolved and particulate fractions (threshold: 0.45 µm). Dissolved fraction was extracted on a SPE C18 cartridge, while a microwave-assisted extraction procedure was applied to the particulate fraction. The three pollutant families (PCBs, PAHs and alkylphenols) were then separated during a purification step on silica columns. Contaminants were quantified by internal calibration using gas chromatography coupled with mass spectrometry (GC/MS, Focus DSQ, ThermoFisher Scientific). Results are displayed as the sum of 13 PAHs[3] (deriving from the US EPA list, excluding naphthalene, acenaphthene and acenaphthylene, which are too volatile to be correctly quantified), along with the sum of the 7 PCB indicators[4]. Among alkylphenols, nonylphenols (NPs) and octylphenols (OPs) were studied.

Trace metals were analysed in both the total and dissolved fractions. Raw samples were microwave acid-digested at 95°C with nitric and hydrochloric acids. Filtered samples on 0.45 µm cellulose acetate membranes were acidified to pH 1 with nitric acid. Metal concentrations were determined using Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES, Varian Vista MPX) through external calibration with a multi-element standard solution (PlasmaNorm Multi-Elements).

The analytical uncertainties and the detection limits are given in Table S.1 in supplementary files.

2.4 Methodology used for site comparisons

The various land use breakdowns did not exhibit the same runoff coefficient and moreover did not produce the same pollutant quantities. Consequently a direct comparison of water and contaminant fluxes between the Reference site and sites equipped with SUDS proved to be an impossible task.

A water and contaminant emissions model was developed for each type of land use relative to the Reference catchment. Then this model was applied to the land use breakdowns of each catchment equipped with SUDS in order to simulate what the catchment behaviour would have been in the absence of flow regulation. This methodology is shown Fig.1 and explained

[3] Fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3]pyrene, di-benzo[a,h]anthracene, and benzo[g,h,i]perylene.
[4] PCB 28, PCB 52, PCB 101 PCB 118, PCB 138, PCB 153, and PCB 180.

below. It is important to note that our methodology did not aim an accurate simulation of the pollutant masses but the estimation of SUDS effectiveness. For this purpose, the pollutant masses were simulated using low assumptions for sources, which allows to be sure that SUDS effect on pollutants is not overrated.

2.4.1 Tool for runoff volume simulation

The aim of this model was to simulate the runoff volume that would have been produced with conventional drainage systems based on the rainfall depth and land use dataset. The model was adapted from Berthier *et al.* (2001) and distinguished 3 types of surfaces: roofs, streets and gardens.

The initial losses on roofs and streets were modelled by a surface storage depth IL (mm). The filling level of this surface storage at the beginning of a rain event depended on the amount of rainfall during the previous 3 hours (H_{3h} in mm) for roofs and previous 6 hours (H_{6h} in mm) for streets. Infiltration through the street's pavement was modelled using a constant infiltration rate $K_{inf,s}$ (in mm/h). If H is the rainfall depth (mm), T_{rain} the rain event duration (h) and A_r and A_s the proportions of roof and street surface areas on the catchment, the runoff water depth (H_{runoff} in mm) was given by:

For roof runoff: $H_{runoff,r} = [H - \max\{IL_r - H_{3h};0\}].A_r$ **Equation 1**

For street runoff:

If $IL_s > H_{6h}$, then: $H_{runoff,s} = \max\{H - (IL_s - H_{6h}) - K_{\inf,s}.T_{rain};0\}.A_s$ **Equation 2**

otherwise: $H_{runoff,s} = \max\{H - K_{\inf,s}.T_{rain};0\}.A_s$

Private gardens above underground parking (50 to 100 cm soil) with drainage systems were modelled as a storage depth IL_g (mm), with their filling level at the beginning of the event being dependent on rainfall quantity over the previous 6 days (H_{6d} in h). The storage filling rate during the rain event was modelled by a constant infiltration rate $K_{inf,g}$;

evapotranspiration was not included in this modelling set-up. If A_g is the proportion of gardens covering the catchment, the runoff water depth from private gardens ($H_{runoff,g}$ in mm) was given by:

If $IL_g > H_{6d}$, then: $H_{runoff,g} = \max\{H - \min\{IL_g - H_{6d}; K_{\inf,g}.T_{rain}\}; 0\}.A_g$ **Equation 3**

otherwise: $H_{runoff,g} = H.A_g$

The runoff water depth from the public garden was assumed to equal zero because, in a conventional system, it would not have been connected to the sewer system.

The five model parameters (IL_r, IL_s, IL_g, $K_{inf,s}$ and $K_{inf,g}$) were calibrated using rainfall and flow data over a 12-month period from the Reference catchment, by means of minimising the sum of absolute error values. The calibrated parameter values listed in Table 3 lie within the same interval as those found in the literature (Berthier *et al.*, 2001).

2.4.2 Tool for micropollutant simulation

The objective of this tool was to simulate contaminant emissions from catchments equipped with SUDS as if these catchments were being drained with a conventional sewerage system.

The principle consisted, for the sum of sampled events i (Cf. Table 2), of comparing the mass measured at the outlet (M) with the mass simulated by summing the masses input via the various entry paths ($\overline{M}$) according to the equations 4 and 5. The validation of the tool was done with the Reference data.

$$M = \sum_i C_{Ref,i} \cdot V_i \qquad \textbf{Equation 4}$$

$$\begin{aligned}\overline{M} &= \overline{M}_{path} + \overline{M}_{street} + \overline{M}_{building} \\ &= \sum_i H_i \cdot \left[C_{atm,i} \cdot \overline{CR}_{s,i} \cdot S_p + C_{s,i} \cdot \overline{CR}_{s,i} \cdot S_s + C_{build,i} \cdot (\overline{CR}_{r,i} \cdot S_r + \overline{CR}_{g,i} \cdot S_g)\right]\end{aligned}$$

Where, for the event i, H_i is the rainfall depth; $C_{Ref,i}$, $C_{atm,i}$, $C_{s,i}$, $C_{build,i}$ the concentrations from Reference catchment outlet, atmospheric deposit, street runoff and building runoff according to Bressy *et al.* (2012);

$\overline{CR}_{s,i}$, $\overline{CR}_{r,i}$, $\overline{CR}_{g,i}$ the runoff coefficients calculated from the volumes simulated section 2.4.1 for street, roof and garden above underground parking; S_p, S_s, S_g the surfaces of path, street and garden; and V_i the water volume measured at Reference catchment outlet.

For zinc introduced via roofing materials, the corrosion models described in Gromaire *et al.* (2011) were used, distinguishing the zinc roofs of other:

$$\begin{aligned}\overline{M} &= \overline{M}_{path} + \overline{M}_{street} + \overline{M}_{building\neq zinc} + \overline{M}_{building=zinc} \\ &= \sum_i \begin{bmatrix} H_{tot,i} \cdot C_{atm,i} \cdot \left(\overline{CR}_{s,i} \cdot S_p + \overline{CR}_{g,i} \cdot S_g + \overline{CR}_{r,i} \cdot S_{r\neq zinc}\right) \\ + H_{tot,i} \cdot C_{s,i} \cdot \overline{CR}_{s,i} \cdot S_s \\ + \overline{M}_{corrosion,i} \end{bmatrix}\end{aligned} \qquad \textbf{Equation 5}$$

Where, for the event i, $S_{r=zinc}$ and $S_{r\neq zinc}$ are the roof surfaces with zinc and without and $\overline{M}_{corrosion,i}$ the mass from corrosion (Gromaire *et al.*, 2011).

To avoid overestimating the simulated masses when data from Reference were missing due to technical problems, i.e., to avoid overvaluing the SUDS effect during comparisons with measurements, the simulated values were deliberately minimised by adopting hypotheses based on entry path concentrations and runoff volumes. Only atmospheric input was considered for PCB assuming the recent buildings or cars do not release them. For PAHs and alkylphenols, when concentrations from road runoff were missing, the lowest measured value was used. Uncertainties on model results induced by input data uncertainty (especially measurement uncertainty on experimental data) were estimated with the law of propagation of uncertainties (explanation in supplementary files Annexe A.1).

3. Results and discussion

3.1 Performance of the simulation tools

The errors and relative errors between simulated and measured H_{runoff} values are shown in Figure 2. Over a one-year period, model behaviour proved to be satisfactory (0.1% error between simulated and measured annual volumes) since the simulations have yielded good results for the events producing the majority of yearly discharged water volume (60% of events were simulated with a margin of error less than ±30%, representing 80% of total annual rainfall). To minimise errors, this model was always applied to the sum of studied events, i.e., over the year for hydraulic simulation and over the sampled events for micropollutant simulation.

Table 4 offers a comparison, for the Reference catchment, of measured mass vs. simulated mass when aggregated over all sampled events (using Table 2 data). The simulated values lie within the uncertainty of the corresponding measurement values (uncertainty on both water volume and concentration measurements). This uncertainty does not cover scenario uncertainty according to the classification described by Warmink *et al.* (2010) since the scenario with the lowest value was initially chosen for sources. Our goal was not to develop a model to simulate actual masses of pollutant but to assess the SUDS effect. This choice allowed us to minimize the measured masses and therefore to avoid overestimation of the SUDS efficiency.

3.2 Effects of source control systems on discharged water

3.2.1 Flow dynamics at the event scale

Flow dynamics were studied for all rain events between July 2008 and August 2009. Over this period, 140 events could be distinguished on the Reference catchment (100 on North, 77 on Park and 114 on South). The difference in number of events across catchments was due to the slower dynamics of source-controlled catchments for which one event might correspond to several for Reference. These results have been analysed with a focus on frequent rain events, which represent most part of annual runoff volumes.

Peak flow reduction (see Fig. 3):

For Reference, Q_{max} spanned a wide range of values (0.2 to 209 l/s/ha as the 1^{st} and 9^{th} deciles), yet it remained below 10 l/s/ha for 69% of rain events suggesting that for the majority of rain events the 10 l/s/ha regulatory flow threshold did not necessarily imply water retention. The situation would be quite different with lower thresholds: for instance, the 2 l/s/ha level would be exceeded for 66% of events.

Figure 3 provides a peak flow reduction for all source-controlled catchments. This effect was observed for almost all rain events, even ordinary rainfall episodes whose flow did not reach the nominal regulator flow (10 l/s/ha). The flow rate actually exceeded 2 l/s/ha for 19% and 13% of events at the North and Park outlets. At the South outlet, flow exceeded 2 l/s/ha for 63% of events; consequently, source control systems in place at the South site caused less impact on frequent rain events.

In the case of North catchment, peak flows were mainly controlled by the flow regulator. However, for Park and South catchments, the level required for the initiation of the regulator was not reached for most rain event and peak flow attenuation was due to the natural retention time in the garden and in the swales.

Staggering and lag time of discharged water (see Fig.4):

For the Reference catchment, flow dynamics were close to the rainfall dynamics: T_{lag} varied between 3 and 14 minutes (1^{st} - 9^{th} deciles), and T_{empty} ranged from 0.8 to 5.8 hours. These high T_{empty} values were due to drainage in the private gardens above underground parking.

For source-controlled catchments, the peak flows shifted in time relative to the rain peak: T_{lag} varied from 16 minutes to 2.3 hours for North, 23 minutes to 2.6 hours for Park, and 8 minutes to 1.4 hours for South (1^{st} - 9^{th} deciles).

The North and Park catchments also showed much longer emptying times for the aggregate of all rain events than either Reference or South. Storage emptying lasted between 1.7 and 17 hours for North and 2.3 to 10 hours for Park. The South catchment took between 0.9 and 3 hours to empty, which was of the same order of magnitude as Reference. The reactivity of the South site could be explained by the type of regulation system installed (vortex regulator, pump) with which flows quickly reached the nominal regulator flow. It may induce fewer effects when a small quantity of water is being stored, i.e., for ordinary rain.

These results indicate that the design of retention devices and, more importantly, the choice of regulation system have proven to be determinant as regards flow dynamics.

3.2.2 Effect on discharged volumes

For the three source-controlled catchments, the annual discharged volumes that would be generated with a conventional storm sewerage system were simulated with the model described in the section 2.4.1 and then compared in Table 5 to actual measured volumes as regards annual runoff coefficient (calculated as the ratio of rainfall amount to runoff water depth).

The annual runoff coefficient simulated for a conventional sewerage system was very close to the proportion of impervious surfaces for all 4 catchments. A large reduction (43% to 55%) in annual runoff volumes, compared to the hypothetical volumes with a conventional storm sewer, was observed for all 3 source-controlled catchments. This significant reduction was not surprising for the Park catchment since all its stormwater flowed through a garden and potentially infiltrated into the soil. For South, the reduction was correlated with longer and more extensive contact with vegetated surfaces (grass strips, swales, grassed basin). The reduction was much less expected on the North site, where storage facilities are mostly composed of impervious materials. One part of this reduction could be explained by vegetated roof and the other part by greater initial losses within the source control systems (dead volumes).

These reductions in annual runoff volumes represent very promising developments for stormwater quality management. Discharged masses are indeed partly determined by discharged volumes, as already observed by other authors (Davis *et al.*, 2009; Trowsdale *et al.*, 2011), and lower annual runoff volumes can also induce a reduction in the discharged pollutant load.

3.3 Effects of source control systems on pollutant loads in stormwater

Figures 5a, 5c and 5e display, for all contaminants studied, both the masses measured (grey histogram) at the outfall of each catchment and the simulated masses (white histogram) as if the storm drainage system was conventional, for the sum of all monitored events. Water volume data have been added for comparison. Figures 5b, 5d and 5f present the same types of data for average concentrations. Uncertainty bars associated with measured data correspond to measurement uncertainties and those associated with simulated data are model uncertainties (with a 80 % confidence interval) (explanation in supplementary files Annexe A.1).

3.3.1 North catchment

For the North catchment, the mass values simulated for a situation with no SUDS ever installed were higher than the measured masses on the whole, a finding that reveals a drop in onsite contamination. The effect on contaminant concentrations depended on the type of substance. Three categories of behaviour could be distinguished:

- For SS, total PAHs and zinc, the decrease in contaminant mass (50%, 60% and 72%, respectively) was greater than the drop in water volume (43%). The measured concentrations were thus lower than the simulation results. Since SS and PAHs are both particulate, we assumed the level of settling would be substantial in the underground storage zones, given that these decreases amounted to the same order of magnitude as the mass reductions evaluated for large storage basins (Aires *et al.*, 2003; Calabro and Viviani, 2006; Clark and Pitt, 2012). The introduction of zinc, on the North site, occurred mainly in dissolved form via the corrosion of metallic roof materials (80% in dissolved form according to Bressy *et al.* (2012)). At the outfalls of large catchments, zinc has been proved to be 50% bind to particulate matter (Zgheib *et al.*, 2011), which proves that zinc tends to bond with particles. It is suggested that part of the dissolved zinc became attached to particles and settled with them or else bonded with either the drainage system or deposits in the storage zone.

- For PCBs and NPs, in their total and dissolved form, the decrease in contaminant mass (between 24% and 36%) was assumed equal to the drop in water volume since the simulated concentrations lied within the uncertainty interval of measured concentrations. These mass amounts were thus lowered by the presence of storage, yet at a constant concentration. Let's recall that these substances were at around 70% in dissolved form in our samples (Bressy *et al.*, 2012). This speciation was not expected to differ like it did for zinc since the distribution here is the one measured in stormwater (Zgheib *et al.* 2011) or for the natural environment (Cailleaud *et al.*, 2007). It is therefore likely that a portion of the contamination has been trapped during water losses due to sedimentation / filtration for the particulate fraction and to adsorption / infiltration for the dissolved fraction.

- For the total (TOC) and dissolved organic (DOC) carbon, copper, dissolved PAHs and both total and dissolved OPs, the decrease in contaminant mass when assuming no SUDS had been installed was less than the drop in water volume or lied within the uncertainty interval of the measures. The mass of released contaminants was in fact lower by use of on-site storage, although the concentration released was slightly superior to that simulated for conventional sewer. These substances are in the both fractions (Bressy *et al.*, 2012) and should therefore undergo at least the same decrease as the other substances, i.e. by sedimentation for the particulate fraction and adsorption for the dissolved fraction. One hypothesis for these findings might be that our simulation has underestimated the masses of these substances, as automotive traffic is a major source of copper, PAHs and OPs (Bjorklund *et al.*, 2009; Bressy *et al.*, 2012; Motelay-Massei *et al.*, 2006). Automotive traffic is more intense on the North catchment than the Reference site, which was used to calibrate the street-based contaminant production function. But as explained in paragraph 3.1, our model intentionally underestimates the simulated masses in order to avoid overestimation of the SUDS effect. As a consequence, for these substances, our methodology does not allow us to conclude about the effectiveness of SUDS.

3.3.2 Park catchment

Downstream of the Park catchment, the majority of simulated mass values exceeded the measured values (except for SS and PAHs in the total fraction), thus indicating a mass drop due to the use of open space storage. As mentioned for the North catchment, this decreasing effect depended on the type of substance under consideration:

- For zinc, dissolved PAHs, total and dissolved NPs, and total and dissolved OPs, the decrease in contaminant mass (80%, 71%, 70% and 60%, respectively) was greater than or equal to the loss of water volume (60%). For these substances, which are mainly in dissolved form (80% for zinc, 79% for NP and 74% for OP according to Bressy *et al.* (2012)), an adsorption effect in the public garden was to be assumed, as demonstrated by Ray *et al.* (2006) on tree bark samples or by Scholes *et al.* (2008).
- For TOC, DOC, PCBs, dissolved PCBs and copper, the contaminant mass decrease was less than the actual volume loss. The simulated mass exceeded measurement results by respectively 29%, 29%, 42%, 50% and 22%, with measured concentrations topping the

simulated concentration values. It was possible that the adsorption effect for these substances, which were at 57% in particulate fraction for TOC, 36% for PCBs and 72% for copper (Bressy *et al.*, 2012), was less pronounced than for the group of substances described above.

- The mass of SS and total PAHs did not appear to be reduced by the upstream management systems introduced. The measured masses of these substances exceeded the simulated values, as if the fact of regulating discharges were raising the level of water contamination for these parameters. It was likely that our simulation has underestimated the actual SS mass produced, given that this value did not take into account particle production from the garden and playground (sandpit). As above, for these substances, our methodology does not allow us to conclude about the SUDS efficiency.

On this site, the decrease in water volume was high (over 50%), and the water residence time in the basins was quite long according to Section 3.2.1 (median drainage time exceeds 5 hours). The contact time between water and potential substrates (plants and soil particles) was also increased, thereby promoting both the adsorption of dissolved contaminants and their infiltration into the soil. Moreover, since the storage facilities were not enclosed, it was considered likely that the phenomenon of volatilisation, photolysis and biodegradation eliminated a portion of soil contamination during dry weather periods (Scholes *et al.*, 2008; Weiss *et al.*, 2007).

3.3.3 South catchment

On the South catchment, all simulated mass values exceeded measurements, which definitely points to contaminant interception within the various SUDS systems. This decrease in mass release however remains small in magnitude given that all simulated concentrations was less than or equal to the measured concentrations, with the exception of zinc.

- As regards zinc, the mass decrease (60%) exceeded the drop in water volume (46%), and the simulated zinc concentration was 24% higher than the measured value. On this catchment, the majority of zinc (90%) entered in dissolved form through the corrosion of roofing materials (Gromaire *et al.*, 2011) and the roof runoff was recovered in both an underground tank and planted swales. In these storage zones, sorption may indeed occur.
- As regards DOC, dissolved PCBs, dissolved PAHs, total and dissolved NPs, total and dissolved OPs and copper, the mass decline was similar to the reduction in water volume. Since all these parameters were essentially dissolved (Bressy *et al.*, 2012), sorption or infiltration effects were clearly apparent.
- The decreases in SS mass (23%), TOC (33%), total PCBs (15%) and total PAHs (19%) were all less than the loss of water volume (46%): these parameters tend to be more particulate in nature.

The decrease in South catchment contaminant mass thus appeared to be less pronounced than that of the other sites. Flow control systems on this catchment were less efficient on current flow rates than the other studied sites (Section 3.2.1), which results in lower residence times and hence worse efficiency.

3.3.4 Fate of micropollutants in these systems

<u>Level of deposit contamination:</u>

Figure 6 presents the contaminant contents in deposits at both the North (underground pipe) and South (storage zone upstream of the regulator) catchments.

The average PCB contents varied between 0.034 µg/g.dw for South and 0.058 µg/g.dw for North: this range was 3 times weaker than contents measured in the Reference stormwater SS (0.10 µg/g.dw, according to Bressy *et al.* (2012)). The particles held in storage prove to be the coarsest as well as the least contaminated; in addition, they are comparable to those detected by Jartun *et al.* (2008) in sediments from a separate urban sewer system in Norway (0.029 µg/g.dw) and below the limit established by the French decree relative to the spreading of sewage sludge (Decree No. 97-1133, 1998), i.e. 0.8 µg/g.dw.

Average PAH contents was equal to 6.9 µg/g.dw in South sediment and 7.0 µg/g.dw in North sediment, which places them at roughly 5 times less than the contents measured in the Reference SS (33 µg/g.dw). These results were comparable to the values measured by Gasperi *et al.* (2005) in particles from water used for street cleaning and above the contents recorded by Jartun *et al.* (2008): 3.4 µg/g.dw. The contents were between 3.5 and 4 times weaker than the limits established for the spreading of sewage sludge.

NPs contents ranged from 0.26 µg/g.dw at South to 6.3 µg/g.dw at North (0.04 and 0.70 µg/g.dw for OPs, respectively). The differences identified between these two sites may be explained by a smaller proportion of road on South and a building age effect subsequent to the European NP use restriction Directive (Directive 2003/53/EC). More specifically, the South catchment contained more recent construction, meaning that the materials employed could contain less NP. The contents found in Reference SS were measured at 6.8 µg/g.dw (0.27 µg/g.dw for OPs). Bjorklund *et al.* (2009) found lower contents in sediments from a separate sewer network in Sweden: below 1.5 µg/g.dw for NPs, and below detection limits for OPs. No regulation has been adopted for alkylphenols as regards their spreading.

For trace metals, the contents recorded at South revealed: 0.059 mg/g.dw copper, 0.041 mg/g.dw lead, and 2.8 mg/g.dw zinc. North catchment results yielded: 0.16 mg/g.dw copper, 0.12 mg/g.dw lead, and 0.79 mg/g.dw zinc. The higher copper and lead contents found in North stemmed from the greater volume of road traffic. The lower zinc values in North were due to a much larger proportion of zinc roofs in South (Table 1). These entire values were lower than those for Reference particles: 0.28 mg/g.dw copper, 0.26 mg/g.dw lead, and 5.5 mg/g.dw zinc. The order of magnitude remained the same as for measurements conducted by Jartun *et al.* (2008), i.e., 6 times less than the spreading limits for copper, 7 times less than those for lead and 4 times for zinc.

These deposits were thus only slightly contaminated, especially when compared to the concentrations measured in stormwater SS: the coarsest, and hence least contaminated, particles are those retained in the SUDS. This category of particles does not require any special treatment, as opposed to the sludge generated by combined sewer networks.

Impacts of urban runoff retention on soil contamination:

To understand the fate of contaminants and to assess whether stormwater storage actually degrades garden soil quality in Park catchment or not, contaminant contents were measured in the soil as a function of the flooding gradient. Figure 7 displays the contents recorded across the various zones. OPs are not displayed because contents were around the LOQ.

PAH concentrations varied between 0.33 and 1.1 µg/g.dw, while PCB contents ranged from 0.026 to 0.060 µg/g.dw and NP values from 0.22 to 0.45 µg/g.dw. These PAH and PCB contents are comparable to measurements recorded in Seine River basin soils by Motelay-Massei *et al.* (2004) and are below the values from the Canadian Soil Quality Guidelines (CCME, 2007) and far from the intervention values for remediation given by the regulation of Netherlands (VROM, 2000). In contrast, the garden soil seems to be contaminated by NPs, given the findings of Vikelsoe *et al.* (2002), who reported contents equal to 0.034 µg/g.dw in runoff storage areas, but this value stay below the Canadian Soil Quality Guidelines (CCME, 2007).

Copper contents varied between 0.014 and 0.040 mg/g.dw, lead between 0.020 and 0.083 mg/g.dw, and zinc between 0.056 and 0.10 mg/g.dw. These values are of the same order of magnitude as Paris region soils, according to Thévenot *et al.* (2007) and they respect guidelines (CCME, 2007; VROM, 2000).

A comparison across the 3 zones of increasing flood frequency did not reveal any difference in content for PCBs, and NPs. For PAHs and 3 trace metals, however, the most commonly flooded zones showed contents of between 1.3 and 3.2 times less. These differences may be attributed to sol heterogeneity or to an infiltration or degradation process, depending on contaminants. Moreover, these results demonstrate that the Park catchment soil is not significantly contaminated following its use for storage.

3.4 Discussion on micropollutant reduction measures for stormwater

Section 3.2 highlighted: i) a significant reduction in annual runoff volumes at the outlets of the source-controlled catchments due to losses on permeable surfaces and higher initial losses on impervious surfaces and ii) an increase in water retention time, which strongly depend on facilities design. When the design of SUDS considers only exceptional rainfalls, retention efficiency of common rainfall is highly variable depending on the type of storage (permeable or impervious surfaces) and the method of flow regulation. For the facilities that naturally increase the retention time, like the large grassed detention garden in Park catchment, the effects due to the flow regulator itself become insignificant for common rainfall.

Section 3.3 proved that the presence of SUDS considerably lowered the contaminant masses released by the catchments. This conclusion was based on a comparison of fluxes measured with SUDS and simulated without SUDS. This methodology takes into account the measurement uncertainty. Scenario uncertainty was not evaluated, however the simulated scenario is an underestimating scenario. It is intentionally based on minimal emissions from the different pollutant sources, so as to avoid overestimation of the SUDS effectiveness.

This decrease varied from 20% to 80% of the total released mass, depending on both the study site and the type of pollutant. For underground storage facilities, the drop is substantial for particulate pollutants (SS, particle-bound contaminants) which are sensitive to sedimentation, and less pronounced for dissolved contaminants which might adsorb to the structures or deposits. For storage scenarios in permeable planted zones, the most significant contaminant decreases involve dissolved pollutants. Other effect of the SUDS can be observed on the substances primarily introduced into stormwater in dissolved form (i.e. metals from the corrosion of roofing materials) and that exhibits a strong affinity for organic matter. In this case, the binding of these substances to deposits and soil is assumed, inducing a stronger reduction of their release. Our findings should apply to other sites equipped with SUDS if they are designed with the same criteria. These results are interesting at local scale because it provides original data on organic micropollutants in SUDS and particularly useful at global scale because they allow to make effective recommendations for design criteria in terms of reducing pollution.

The effects on discharged contaminants may be correlated with hydraulic effects, namely the followings.

- Discharged masses are strongly correlated to discharged volumes and a reduction in annual runoff volumes may explain the highlighted mass decreases. Consequently, the volume drop for ordinary rain events needs to be assigned a top priority when developing stormwater management processes and, moreover, can be maximised by adding storages in grassy areas.
- During runoff, the reduction in flow velocity may lower contaminant erosion at the surface.
- In water storage facilities, reduced flow velocity allows for particle settlement (Calabro and Viviani, 2006). A longer residence time increases contact time between water and substrate (i.e., soil, sediments, building materials) and is therefore favourable to both

pollutant filtration trough permeable materials and sorption of dissolved fractions (Mason *et al.*, 1999; Ray *et al.*, 2006).

- On a long-term basis and during dry weather periods, some of the pollutants retained in the source control devices may undergo evaporation or degradation (biodegradation, photodegradation, etc.) (Scholes *et al.*, 2008; Warren *et al.*, 2003).

In order to improve water quality, the design of SUDS should focus on systems that retain all rain events, even those with the lowest intensity. To intercept ordinary events without increasing the size of storage facilities, flow rate regulation may be adjusted according to the importance of the event. It may be feasible to store the first few millimetres of rain in porous materials without discharge and by emptying via infiltration and/or evapotranspiration. As an example, "rain garden" and "bioretention" type systems are currently being promoted in other countries (Dietz and Clausen, 2005; Jefferies *et al.*, 2004). These recommendations are consistent with Petrucci *et al.* (2013). They showed that flow-rate based regulations can produce negative impacts on water discharge at the catchment scale and that volume-based regulations should be encouraged for example local infiltration facilities.

4. Conclusion

The research presented herein has demonstrated that the use of SUDS systems, initially aimed at peak flow attenuation, can also serve to slow and delay water flow for frequent rain events, and result in a significant reduction of the annual discharged volumes. The masses of discharged contaminants are also decreased. This phenomenon is correlated with hydraulic effects: greater initial and continuous losses limit contaminant transfer downstream while an extended residence time enhances the phenomena of substance sedimentation and adsorption. This reduction in discharged contaminants is primarily explained by a drop in runoff water volumes. These results do not systematically reveal any kind of "purifying effect" in the classical meaning (i.e. lower concentrations), but instead an overall effect of reducing mass discharges.

The effects are however highly variable from one site to another, and from one contaminant to another. They depend on the hydraulic interception of small rainfalls. For impervious storage structures the retention time of small events depends mainly on the characteristics of the flow regulator device, whereas for pervious and vegetated storage structures natural losses (infiltration, evapotranspiration) greatly contribute to the interception of frequent rain events.

Bressy *et al.* (2012) have exposed the benefit of managing stormwater upstream, for the purpose of locally discharging the slightly contaminated water; refraining from mixing with heavily polluted water, and avoiding the network cross contamination process.

Nowadays, in France, the design of SUDS is mainly intended to protect against flooding and to limit discharged flows mainly by intercepting the exceptional rain events. To ensure efficiency in terms of pollutant mitigation, this upstream stormwater management approach must limit water transfer downstream and take into account frequent rain events when designed. A regulation system needs to be introduced for retaining ordinary events without excessively increasing the storage volume for ten-year return period rainfalls. As an example, the first few millimetres of rainfall could be systematically stored in a porous material or a vegetated zone without any discharge to the network but with drainage provided by infiltration and/or evapotranspiration.

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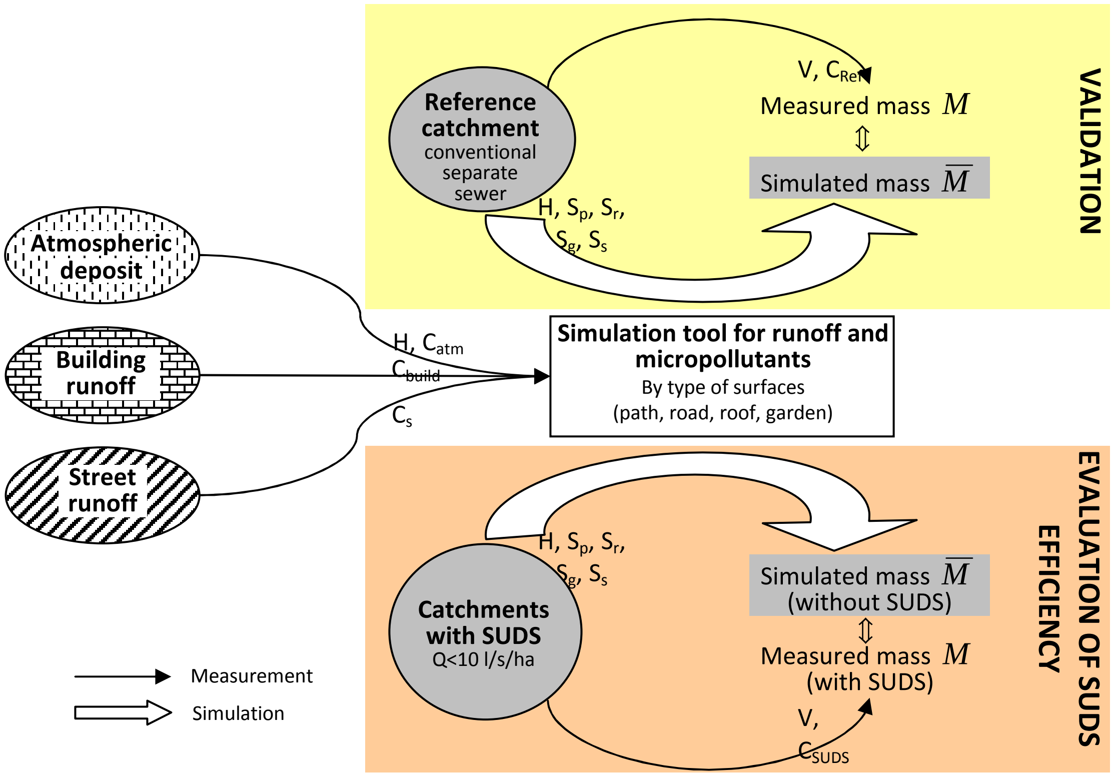


Fig. 1. Methodology for comparison between sites (with H the rainfall depth, V the runoff volume, S the surface, C the concentration, Q the flow rate and M the mass)

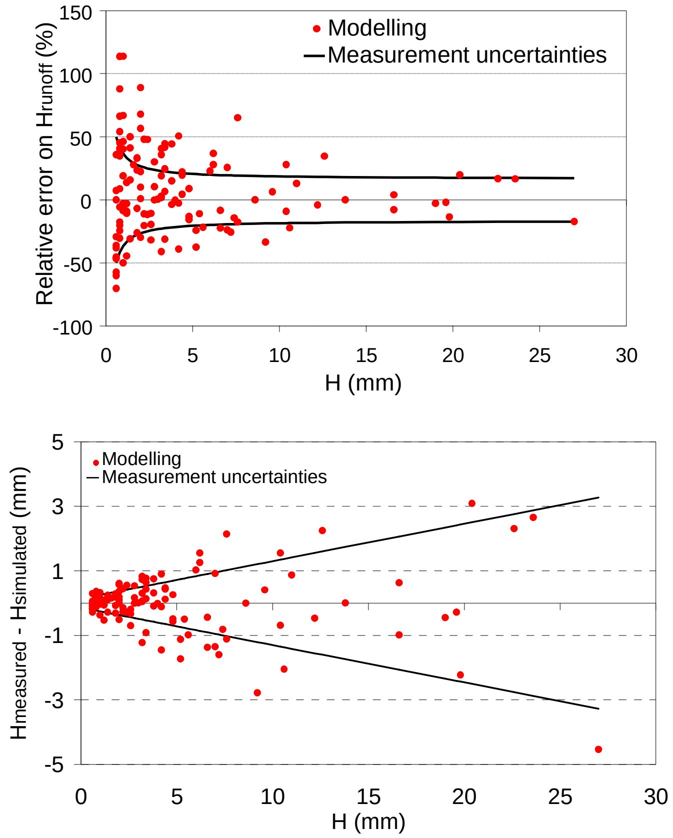


Fig. 2. Errors and relative errors of the model vs. rainfall depth

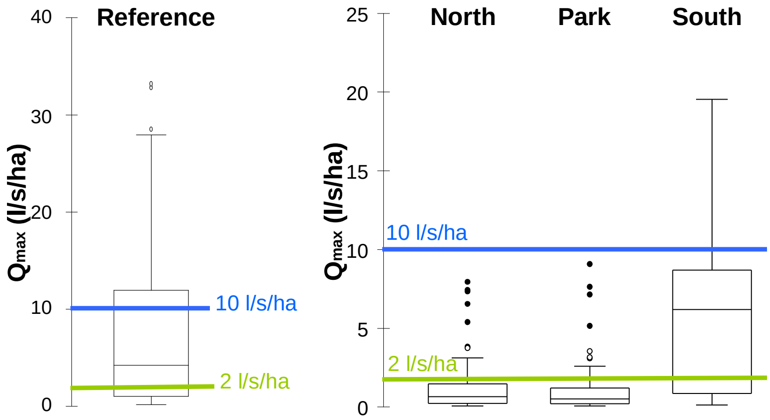


Fig. 3. Box plots[5] of peak flows observed during events from a one-year monitoring period

[5] The box represents the 25^{th} and 75^{th} percentiles and the band inside represents the median. The ends of the whiskers depict the lowest data remaining within the 1.5 interquartile range (IQR) of the lower quartile and the highest data remaining within 1.5 IQR of the upper quartile.

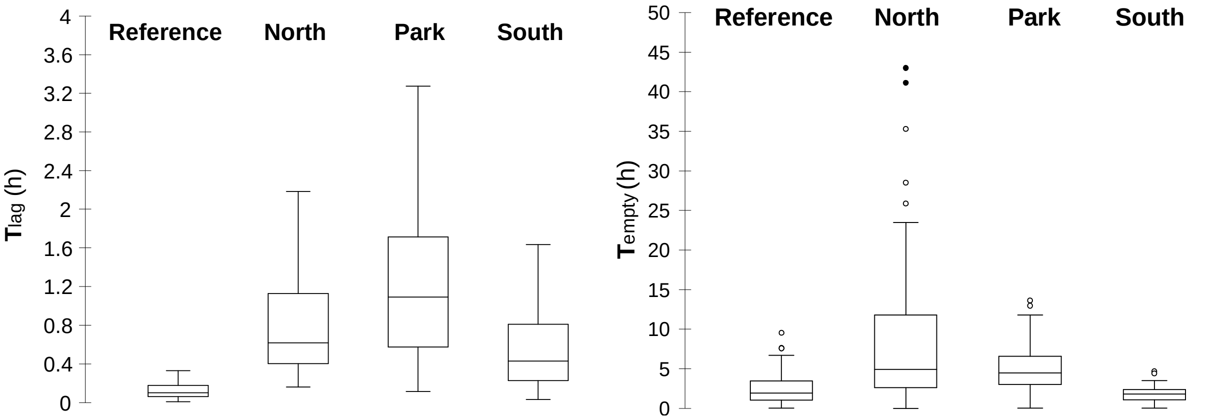
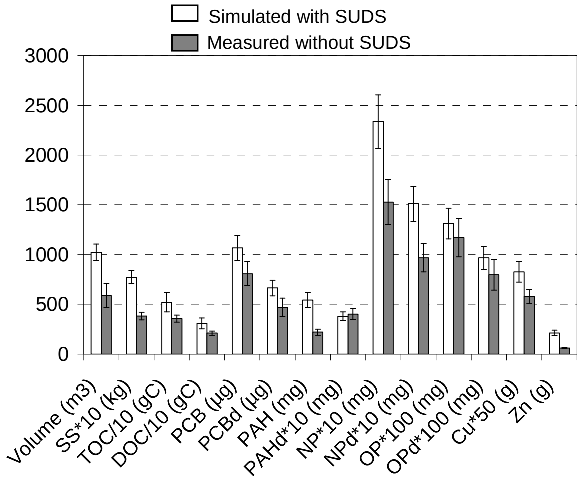
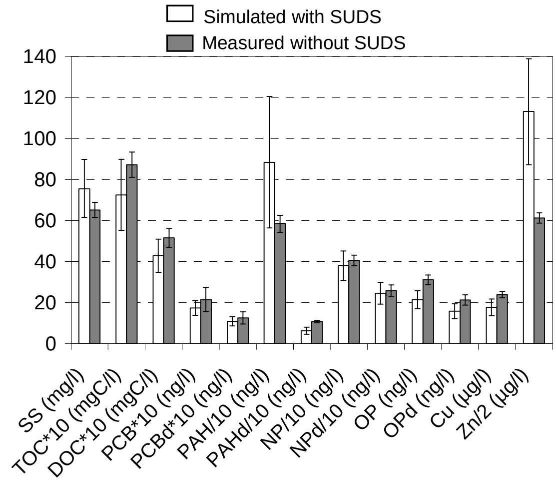


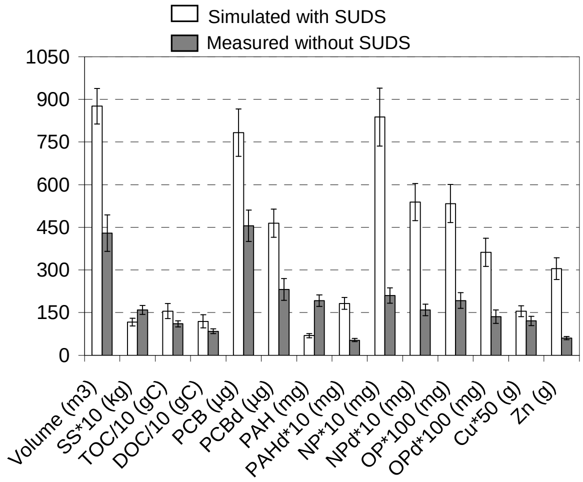
Fig. 4. Box plots of T_{lag} and T_{empty} for each catchment and all rain events between July 2008 and August 2009



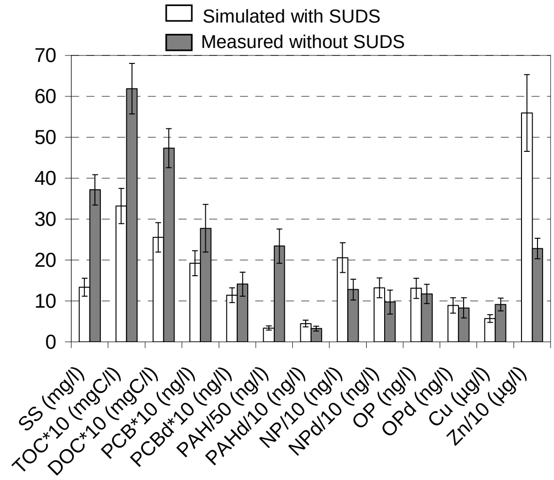
5a. Contaminant masses on North



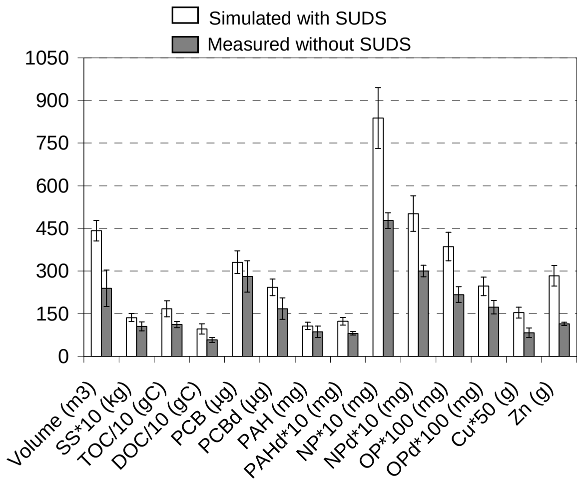
5b. Contaminant concentrations on North



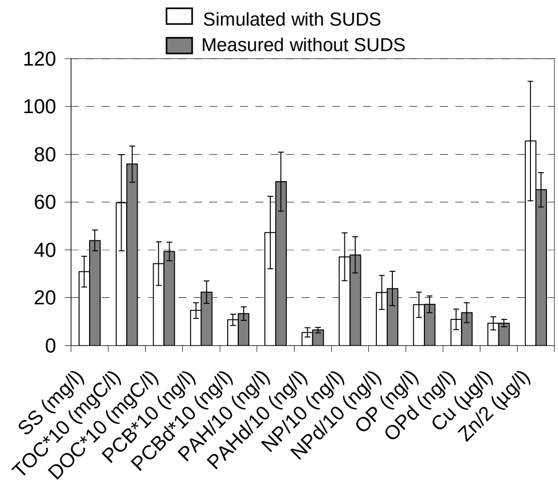
5c. Contaminant masses on Park



5d. Contaminant concentrations on Park



5e. Contaminant masses on South



5f. Contaminant concentrations on South

Fig. 5. Masses and concentrations simulated without SUDS and measured at the catchment outlet for the sum of monitored events

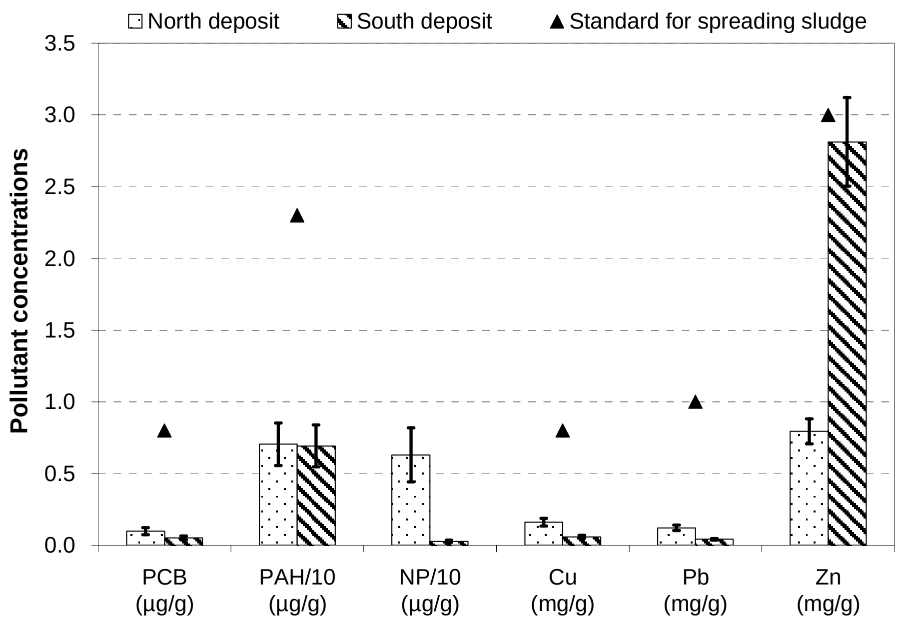


Fig. 6. Pollutant concentrations (in dry weight) in the North and South catchment deposits

 DOI : 10.1016/j.watres.2014.03.040

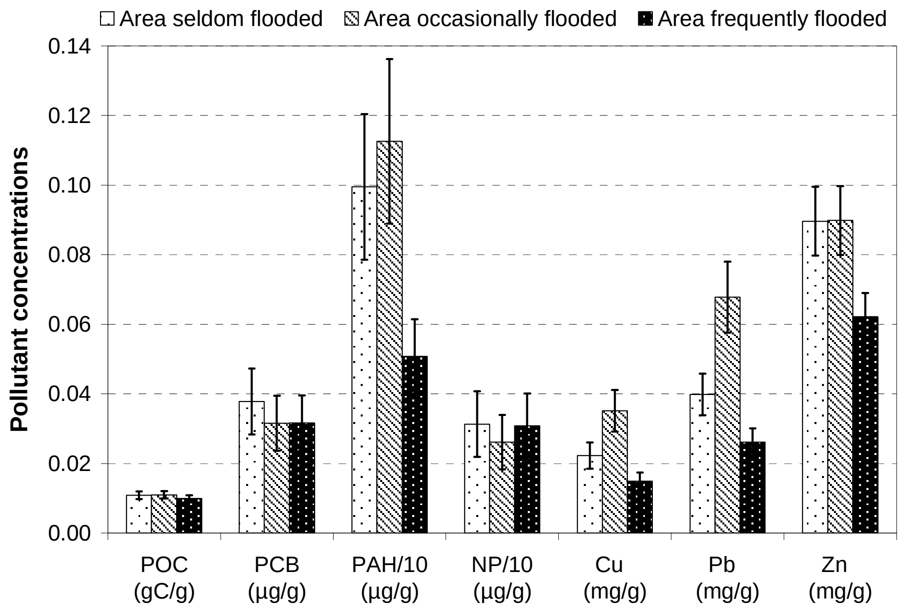


Fig. 7. Pollutant concentrations in the soil of Park catchment retention basins

 DOI : 10.1016/j.watres.2014.03.040

Table 1. Description of the studied catchments

Name	Size (ha)	Land use (%)						Retention system	Flow regulation
		R≠Zn*	R=Zn*	S*	P*	Gs*	G*		
Reference	0.82	36	7	28	3	25	0	Conventional stormwater system	-
North	1.5	47	2	24	4	18	6	Vegetated roof + retention in an oversized pipe (return period of up to 2 years), possible overflow into a swale and a parking lot	16 l/s vortex flow regulator
Park	2.0	12	4	0	19	26	39	2 grassed retention basins in a public park	23 l/s float valve flow regulator
South	0.92	28	10	8	19	17	17	Swales + grassed retention basin in a square + underground tank	2 × 1 l/s nozzles + 5.6 l/s vortex flow regulator + 3 l/s pump
Built parcel	0.13	22	43	0	4	31	0	Conventional stormwater system	-
Street	0.031			100				Conventional stormwater system	-

*R≠Zn: Roof without zinc; R=Zn: roof made with zinc; S: street; P: walking paths; Gs: garden above underground parking; G: garden

Table 2. Characteristics of sampled rain events
(*n* is the number of rain events and *h* the cumulated water depth in mm)

	SS and organic matter		Organic micropollutants		Trace metals	
Catchment	*n*	*h* (mm)	*n*	*h* (mm)	*n*	*h* (mm)
Reference	8	57	9	81	5	36
North	7	65	5	58	9	99
Park	8	73	6	66	8	94
South	6	49	4	42	7	63

 DOI : 10.1016/j.watres.2014.03.040

Table 3. Runoff modelling parameters

Surface	IL (mm)	K_{inf} (mm/h)
Roof	0.33	
Street	0.65	0.2
Private garden	27.6	19.7

 DOI : 10.1016/j.watres.2014.03.040

Table 4. Validation of micropollutant load simulations on the Reference site
(*t* for total fraction and *d* for dissolved fraction)

	Measured	Simulated		Measured	Simulated
SS (kg)	15.2 ± 1.5	15.4 ± 1.6	NP*t* (mg)	204 ± 18	202 ± 19
TOC (g)	2.08 ± 0.21	2.42 ± 0.28	NP*d* (mg)	142 ± 15	141 ± 13
DOC (g)	1.16 ± 0.12	1.38 ± 0.15	OP*t* (mg)	12.2 ± 1.3	11.7 ± 1.1
PCB*t* (µg)	994 ± 99	992 ± 79	OP*d* (mg)	8.51 ± 1.1	8.23 ± 0.83
PCB*d* (µg)	615 ± 92	695 ± 55	Cu (g)	4.50 ± 0.51	4.20 ± 0.41
PAH*t* (mg)	467 ± 42	434 ± 57	Zn (g)	146 ± 16	150 ± 15
PAH*d* (mg)	36.6 ± 2.9	34.9 ± 3.7			

value ± uncertainty (80% confidence level)

Table 5. Proportion of impervious surfaces and annual runoff coefficients (measured and simulated) (± measurement uncertainty)

	Reference	North	Park	South
Proportion of impervious surfaces	0.75±0.018	0.76±0.013	0.35±0.007	0.65±0.015
Measured annual runoff coefficient	0.72±0.086	0.40±0.048	0.15±0.018	0.32±0.038
Simulated annual runoff coefficient*	0.71±0.056	0.69±0.054	0.34±0.026	0.64±0.050
Annual runoff volume reduction attributed to SUDS		43%	54%	55%

* Simulated for a conventional stormwater pipe system as described in paragraph 2.4.1

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Supplementary files:

Table S.1: log K_{OW}, analytical detection limits in both dissolved and particulate fraction

	Log K_{OW}	LQ in dissolved fraction (ng/L) for 2.5 L extracted	LQ in particulate fraction (ng/g.dw) for 100 mg extracted
Fluorene	4.14	0.8	20
Phenanthrene	4.46	0.4	10
Anthracene	4.54	0.8	20
Fluoranthene	5.22	1.2	30
Pyrene	5.18	1.4	35
Benzo[a]anthracene	5.91	1.6	15
Chrysene	5.61	0.6	10
Benzo[b]fluoranthene	5.80	0.4	10
Benzo[k]fluoranthene	6.00	0.4	10
Benzo[a]pyrene	6.04	0.4	10
Indeno[123]pyrene	7.00	0.6	15
Dibenzo[ah]anthracene	6.75	0.4	10
Benzo[ghi]perylene	7.23	0.4	10
PCB28	5.68	0.16	4
PCB52	5.68	0.42	10
PCB101	5.73	0.16	4
PCB118	6.41	0.16	4
PCB153	6.57	0.2	5
PCB138	6.79	0.16	4
PCB180	6.72	0.16	4
Para-tert-octylphenol	4.12	7.6	190
4-nonylphenol	4.48	3.2	330
MES	-	0.5 mg/L	
COD	-	0.5 mgC/L	-
COP	-	-	1.7 mgC/g

Table S.2: Analytical uncertainties at 80% confidence level

	Analytical uncertainties
MES	10 %
COD	5 %
COP	10 %
ΣHAP	29 %
ΣPCB	26 %
NP	23 %
OP	30 %
Cu	17 %
Pb	15 %
Zn	11 %

Annexe A.1: uncertainties calculation

Relative uncertainty (u_r) calculations of simulated or measured data were made according to the law of propagation of uncertainties.
In the following part: S is the surface, V is the volume, H is the rainfall, C is the concentration, CR is the runoff coefficient. The subscript i depends on the i[th] event and the subscript *j* depends on the type of surface: p for pathways, s for street and b for building.

For measured rainfall H_i:
The uncertainty of measured rainfall is due to uncertainty of the pluviometer. V_{runnel} is the volume of the runnel which is equal to 0.2 mm and $u_r(V_{runnel})$ was estimated at 5.6 %. The relative uncertainty on the surface of the runnel, $u_r(S_{runnel})$, was estimated at 1 % by the constructor. N is the number of tipping of the runnel and $u_r(N_{runnel})$ was estimated at one over the whole rain.

$$u_r(H_i) = u_r\left(\frac{V_{runnel}.N_{runnel}}{S_{runnel}}\right) = u_r(V_{runnel}) + u_r(S_{runnel}) + u_r(N_{runnel}) = 0.056 + 0.01 + \frac{0.2}{H_i}$$

For measured surface S:
The uncertainty of measured surface is due to uncertainty of the delimitation of the surface. The uncertainty on the delimitation of the boundaries was estimated to 0.5 m.

$$u_r(S) = u_r(l.l) = 2.u_r(l) = 2\frac{0.5}{l} \approx 2\frac{0.5}{\sqrt{S}} \approx \frac{1}{\sqrt{S}}$$

For simulated annual runoff coefficient (Table 5):

$$u_r^2(CR) = u_r^2(S) + \sum_i \frac{V_i^2}{V^2}\left[u_r^2(H_i) + u_r^2(CR_i)\right]$$

$$u_r^2(CR) \approx \frac{1}{S} + \sum_i \frac{V_i^2}{V^2}\left(\left(0.066 + \frac{0.2}{H_i}\right)^2 + u_r^2(CR_i)\right)$$

$u_r^2(CR_i)$ was assessed from the comparison between measured data and simulated data on Reference catchment. It was evaluated at 23 % if $H_i > 4$ mm and 47 % if $H_i < 4$ mm.

For simulated volumes (Figure 5):

$$u_r(V_j) = \sqrt{\frac{\sum_i V_{i,j}\left[u_r^2(H_i) + u_r^2(CR_{i,j})\right]}{V_j^{\,2}}}$$

And $u_r(V) = \dfrac{\sqrt{u_r^2(V_p)\cdot V_p^2 + u_r^2(V_s)\cdot V_s^2 + u_r^2(V_b)\cdot V_b^2}}{V}$

For simulated concentrations (Figure 5):

$$u_r(C_j) = \sqrt{\frac{\sum_i \left[u_r^2(C_{i,j}) \cdot \left(\frac{C_{i,j} \cdot V_{i,j}}{V_j} \right)^2 + \left(u_r^2(CR_{i,j}) + u_r^2(H_i) + u_r^2(S) \right) \cdot \frac{\left(V_{i,j} \cdot C_{i,j} \cdot (V_j - V_{i,j}) \right)}{V_j^4} \right]}{C_j^{\,2}}}$$

$u_r^2(C_{i,j})$ is the relative uncertainty for the concentration of the event i and the type of surface j, evaluated according to Bertrand-Krajewski *et al.* (2001):

$u_r^2(C_{i,j}) = u_r^2(C_{i,j}1) + u_r^2(C_{i,j}2) + u_r^2(C_{i,j}3) + u_r^2(C_{i,j}4) + u_r^2(C_{i,j}5) + u_r^2(C_{i,j}6)$

with $u_r^2(C_{i,j}1)$ the uncertainty due to sampling ≈ 5 %

with $u_r^2(C_{i,j}2)$ the uncertainty due to analysis given in Table S1 for each pollutant

with $u_r^2(C_{i,j}3)$ the uncertainty due to sub sampling ≈ 2 %

with $u_r^2(C_{i,j}4)$ the uncertainty due to sampler installation ≈ 20 %

with $u_r^2(C_{i,j}5)$ the uncertainty due to flow measurement ≈ 10 %

with $u_r^2(C_{i,j}6)$ the uncertainty due to difference between the event duration and the sampling duration ≈ 20 %

And $u_r(C) = \frac{\sqrt{u_r^2(C_p) \cdot C_p^2 + u_r^2(C_s) \cdot C_s^2 + u_r^2(C_b) \cdot C_b^2}}{C}$

For simulated masses (Figure 5):

$u_r(M_j) = \sqrt{u_r^2(C_j) + u_r^2(V_j)}$