Stormwater retention basin efficiency regarding micropollutant loads and ecotoxicity

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ABSTRACT

Retention basin efficiency in micropollutant removal has not been very well studied, in particular for pollutants highlighted by the European Water Framework Directive of 2000 such as pesticides, polybrominated diphenyl ethers (PBDEs) and alkylphenols. This study is based on *in situ* experiments carried out on a stormwater retention basin with the aim of estimating the basin efficiency in trapping and removing micropollutants from stormwater run-off from an industrial catchment drained by a separate sewer system. Along with stormwater, the basin receives some dry weather effluent flows, which are supposedly non-polluted. Ninety-four substances from five families (metals, polycyclic aromatic hydrocarbons (PAHs), PBDEs, alkylphenols and pesticides) were analyzed during 10 event campaigns in urban wet weather discharges at the inlet and outlet of the basin. The ecotoxicity of the samples was also tested. The results show high inter-event variability in both chemical and ecotoxic characteristics. They indicate good event efficiency concerning heavy metals and most PAHs. The studied pesticides, mainly found in the dissolved fraction, were not trapped. Particulate fraction study highlighted that settling is not the main process explaining micropollutant removal in a retention basin, as was noted for alkylphenols and PBDEs.

Key words | dry retention basin, ecotoxicity, micropollutant, run-off, stormwater

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INTRODUCTION

In France, large retention basins have been implemented for several decades to mitigate stormwater flood and pollution impact in urban areas.

In 2000, the European Water Framework Directive (WFD) outlined high ambitions, with an objective of reducing of pollutant emission and discharge in receiving water by 2015. In addition to traditional macropollutants (total suspended solids (TSS), organic matter), the notion of micropollutants (MP) and EQS (Environmental Quality Standard) compliance values was raised (EC 2013).

The WFD prescriptions have led to studies on micropollutant behavior at a catchment scale (Bressy *et al.* 2011; Zgheib *et al.* 2012; Birch 2012), sometimes including identification of the contributions of atmospheric and wash-off sources (e.g. Eriksson *et al.* 2005). While MP loads have been studied at the outlet of urban catchments and in the receiving water courses, very little research has focused on the removal efficiency of large, dry retention basins across a wide range of MP (in particular organic compounds) at the outlet of a separate sewer system. Existing studies generally deal with a limited set of pollutants (heavy metals and polycyclic aromatic hydrocarbons (PAHs)) and/or specific systems like ponds (Hossain *et al.* 2005; Hares & Ward 1999).

The paper addresses this question and is completed by an ecotoxicity assessment. This additional insight is important, because ecotoxicity is also seldom reported in the stormwater treatment literature (Becouze-Lareure *et al.* 2012).

METHODS

Monitoring system

Description of the site

All experiments were conducted in a large, dry retention/settling basin (Django Reinhardt) situated at Chassieu near Lyon, France. This basin is located at the outlet of

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an industrial catchment (185 ha with 75% imperviousness) drained by a separate stormwater network. The retention basin (1.1 ha) is 32,000 m^3 in capacity with an outflow control limited to 350 $L.s^{-1}$. These values indicate the specific context of this study dealing with large basins (230 $m^3.ha\ imp^{-1}$), with a low outflow rate (2.5 $L.s^{-1}.ha\ imp^{-1}$) which is very common in France. The basin also receives dry weather flows, supposedly non-polluted, from the cooling of industrial processes. These dry weather effluents represent 26% of the total inlet volume and 20% of the total mass of suspended solids (values obtained from 2004 to 2010 by Gonzalez-Merchan (2012)). In 2006, the sediments at the bottom of the basin were totally removed.

Monitoring system

Event-mean pollutant concentrations and ecotoxicity were evaluated at a rain-event scale from mean samples taken with a refrigerated automatic sampler. Mean sample (corresponding to a sampling event) was composited on a flow proportional basis (i.e. by mixing *m* primary sub-samples in proportion to the flow; the number *m* and volume of primary samples depending on weather forecast).

Inflow and outflow rates were monitored continuously at a 2-minute time step, together with water pH, specific conductance, turbidity and temperature, by sensors. Details of the different sensors and measurement procedures can be found in Bertrand-Krajewski *et al.* (2008).

For the MP of the study, two types of samplers were used: one with 24×0.9 L glass bottles for most of the organic compounds (Hach Lange Bühler 4010, 4011) and one with 24×0.9 L polyethylene bottles for metals (Hach Lange Sigma 900) and specific pesticides such as glyphosate (Gly), ammonium glyphosate (GlA) and aminoethylphosphonic acid (AMPA) and ecotoxicity. Both the dissolved and particulate fractions of the samples were analyzed by research laboratories with methods specifically developed and calibrated for stormwater. Traditional blanks were also done. TSS were analyzed according to the AFNOR T.90–105 standard.

As huge volumes would have been necessary to analyze all of the selected MP, a sampling planning procedure was adopted according to the type of event. Therefore not all of them were systematically analyzed for all the events (see the Appendix (available online at http://www.iwaponline.com/wst/069/807.pdf) and Sébastian *et al.* (2011) for more details).

To characterize rain events, a rain gauge recorder was used on the site, collecting data at a 1-minute time step.

Micropollutants

Substances analyzed

The European WFD requirements recently integrated 45 substances defined as priority or priority hazardous substances (EC 2013). Most of these substances were studied in previous research programs, in particular at the outlet of the Chassieu catchment (Becouze-Lareure 2010).

In our study, not only most of the WFD micropollutants but also other emerging substances with potential sanitary hazard were analyzed. In total 94 substances from five groups (metals, PAHs, pesticides, alkylphenols and polybrominated diphenyl ethers (PBDEs)) were studied. Names and acronyms are presented in detail in the Appendix (available online at http://www.iwaponline.com/wst/069/807.pdf).

Experimental data processing

Micropollutant concentrations are event mean concentrations (*EMC*) and are defined both in dissolved and particulate fractions:

$$EMC = EMC_d + EMC_p \tag{1}$$

EMC_d and EMC_p event mean concentrations are analyzed in dissolved fraction and particulate fraction respectively.

MP Mass is then calculated according to Equation (2):

$$M_x = EMC_x.V_x \tag{2}$$

EMC_x, M_x and V_x are respectively the inlet or outlet MP event mean concentration, mass and volume during each event, calculated by using inflow and outflow values monitored at 2-minute time step.

Finally, the retention basin Event Mass Efficiency (E_M) in removing MP (%) is defined by:

$$E_M = \frac{M_i - M_o}{M_i}.100 \tag{3}$$

M_i and M_o are respectively the inlet and outlet MP masses.

Ecotoxicity

The ecotoxicological characterization of water samples from inlet and outlet was carried out using a set of additional bioassays. The set consisted of two chronic toxicity tests on *Heterocypris incongruens* (ostracods) and *Brachionus calyciflorus* (rotifers).

Ostracod mortality and growth inhibition were studied with the Ostracodtoxkit® standard procedure (ISO 14371 2012). This test was initially used to assess the toxicity of the sediments. In this work, it was decided to conduct this test on the total sample, using standard freshwater as the control test according to previous studies (Becouze-Lareure *et al.* 2012).

Rotifer reproduction was also studied on the total sample, with the Rotoxkit® standard procedure (PR NF ISO 20666 2007).

RESULTS AND DISCUSSION

Campaign characteristics and MP concentrations

The results presented in the paper were obtained during 10 sampling campaigns conducted both at the inlet and outlet of the retention basin. Characteristics of the rainfall events are given in Table 1. Minimum, maximum and median values of the different parameters between 2010 and 2012 are presented in the last row and give a good representation of the associated event. It can be noticed that 2011 was a very dry year in the mid-east of France.

Seven campaigns were carried out on five heavy metals (nickel, lead, copper, zinc, cadmium), four on a larger list of metals, six on PAHs, three on alkylphenols, from one to four on pesticides depending on the family studied and just one on PBDEs. TSS concentration was systematically analyzed and ecotoxicity was tested in five campaigns.

Concerning MP occurrence, all the metals and alkylphenols were quantified at least once at the inlet and/or outlet. Dibenzo(a,h)anthracene was the only PAH not detected. Five PBDEs were quantified and only 11 of 45 pesticides were detected and 10 of 45 quantified (see Table B in the Appendix for campaign details and Table C and Table D for detection limits; available online at http://www.iwaponline.com/wst/069/807.pdf).

Concentrations of substances quantified at the inlet were in the range of common concentrations found in the literature (Bressy *et al.* 2011; Zgheib *et al.* 2012; Becouze-Lareure 2010). Table 2 presents inlet and outlet concentrations for some substances.

Retention basin impact on MP

Event mass efficiency

Figure 1 shows event mass efficiency (E_M) of the different substances for the *n* different campaigns when it was evaluated. The values are presented according to the total fraction. Comparisons with literature data were done even if treatment devices were different in terms of size, design and outflow control. Moreover, analytical uncertainties estimated by repeatability tests have not been calculated yet in detail so the results have to be carefully interpreted.

Table 1 | Rainfall characteristics

	Date	Rainfall duration h	Total rainfall depth mm	Antecedent dry weather period d	Mean intensity mm/h	Max. intensity (5-minute time step) mm/h
A	2011-07-08	4.4	15.4	1.2	3.5	68.5
B	2011-10-19	6.1	9.6	9.1	1.6	7.6
C	2011-12-07	30.3	5.3	0.5	0.2	2.2
D	2012-01-05	16.4	8.0	0.9	0.5	2.2
E	2012-03-18	11.8	11.5	0.7	1.0	4.7
F	2012-04-03	17.6	16.5	0.9	0.9	6.4
G	2012-04-11	4.7	7.6	0.2	1.6	6.2
H	2012-05-20	25.0	25.7	0.9	1.0	26.2
I	2012-07-03	31.6	50.0	1.8	1.6	22.7
J	2012-09-12	19.0	18.5	9.8	1.0	19.0
	2010-01-01 to 2012-12-31[a]	0.1–51.9 [3.8]	0.1–100.0 [2.0]	0.2–6.7 [0.7]	0.1–227.2 [0.6]	0.7–272.2 [3.5]

[a] Min–max [median] on 367 events.

Table 2 | Event mean concentrations of MP

	TSS		**Ni**		**Pb**		**Cu**		**Zn**		**Cd**		**PAHs $\sum_{15}$**	
	mg/l		**µg/l**		**µg/l**		**µg/l**		**µg/l**		**µg/l**		**ng/l**	
	In	**Out**	**In**	**Out**	**In**	**Out**	**In**	**Out**	**In**	**Out**	**In**	**Out**	**In**	**Out**
A	154.0	64.0	17.4	11.8	49.1	15.6	79.8	41.0	748.0	345.7	1.3	1.1	–	–
B	67.7	31.6	8.7	4.6	10.6	6.2	55.5	33.1	365.3	286.5	1.1	1.0	–	–
C	88.3	40.0	–	–	–	–	–	–	–	–	–	–	1391.3	597.0
D	67.5	42.4	–	–	–	–	–	–	–	–	–	–	–	–
E	177.6	69.2	12.8	2.3	70.2	7.1	187.0	21.9	1200.0	181.0	1.1	0.2	–	–
F	86.7	26.9	–	–	–	–	–	–	–	–	–	–	448.8	228.3
G	92.8	43.6	10.2	2.6	10.0	5.4	26.6	16.1	223.0	128.0	0.2	0.1	642.7	389.3
H	124.2	41.0	14.2	14.5	15.5	6.9	30.5	28.4	397.1	1054.1	1.1	1.3	506.8	227.7
I	100.7	13.6	5.0	1.7	11.3	2.9	31.8	8.1	201.0	57.7	0.3	0.1	485.8	165.1
J	114.6	48.9	6.7	3.5	16.0	7.1	41.5	22.1	243.0	114.0	0.3	0.2	782.2	448.3

	4-OP		**4-NP**		**B209**		**Di**		**Isop**		**Gly**		**AMPA**	
	ng/l		**ng/l**		**ng/l**		**ng/l**		**ng/l**		**ng/l**		**ng/l**	
	In	**Out**	**In**	**Out**	**In**	**Out**	**In**	**Out**	**In**	**Out**	**In**	**Out**	**In**	**Out**
A	–	–	–	–	–	–	–	–	–	–	–	–	–	–
B	–	–	–	–	–	–	–	–	–	–	–	–	–	–
C	55.1	40.9	189.5	121.9	–	–	58.1	65.6	7.2	8.4	–	–	–	–
D	–	–	–	–	–	–	6.3	33.7	60.5	64.9	–	–	–	–
E	45.4	36.2	422.5	468.6	216.9	93.5	–	–	–	–	<	<	<	<
F	38.4	33.7	1016.7	879.1	–	–	45.9	59.4	6.8	7.4	–	–	–	–
G	–	–	–	–	–	–	–	–	–	–	–	–	–	–
H	39.4	39.8	1286.1	1331.7	–	–	20.4	20.8	0.8	0.8	3.7	1.7	2.8	9.2
I	–	–	–	–	–	–	12.2	1401.3	2.8	17.5	–	–	–	–
J	–	–	–	–	–	–	2.8	3.0	2.8	2.7	–	–	–	–

PAHs $\sum_{15}$: Acy, Ace, Flu, Phe, A, Flh, Pyr, BaA, Chr, BbF, BkF, BaP, IP, D(a,h)A, Bper.
– Not analysed.
< Below limit of detection.

Event mass efficiency of the basin for nickel, lead, copper and zinc (Figure 1(a)) showed median values ranging from 60% to 74% ($n = 7$) which is consistent with the results found in the literature (e.g. Hares & Ward 1999; US-EPA 2008).

The values of cadmium efficiency (median: 55%, mean: 53%) are a little bit lower but still higher than those in the literature (e.g. a US-EPA study (US-EPA 2008) reported mean values around 34%, observed on a dataset of 25 retention basins). However, an inter-event variability can be noticed whatever the metal studied.

Regarding the 17 other metals, median values (not presented in Figure 1) are generally higher than 50% except for vanadium, strontium, calcium, potassium and sodium whose median values are 46, 41, 41, 31 and 23% respectively.

For PAHs (Figure 1(b)), E_M values seem to increase with the number of aromatic hydrocarbon rings. Benzo(k)fluoranthene (five rings) is better trapped than acenaphthene (three rings) with median E_M values of 67% and 24% respectively. This is also consistent with the literature (e.g. Pitt *et al.* 1999; Hwang & Foster 2006). But once again, inter-event variability can be high for certain substances such as benzo (a)pyrene, fluoranthene and anthracene (substance not presented here). Naphthalene E_M (two rings) varies from 4% to 31%, and this result can be compared to literature data indicating this compound is not trapped (Moy *et al.* 2003).

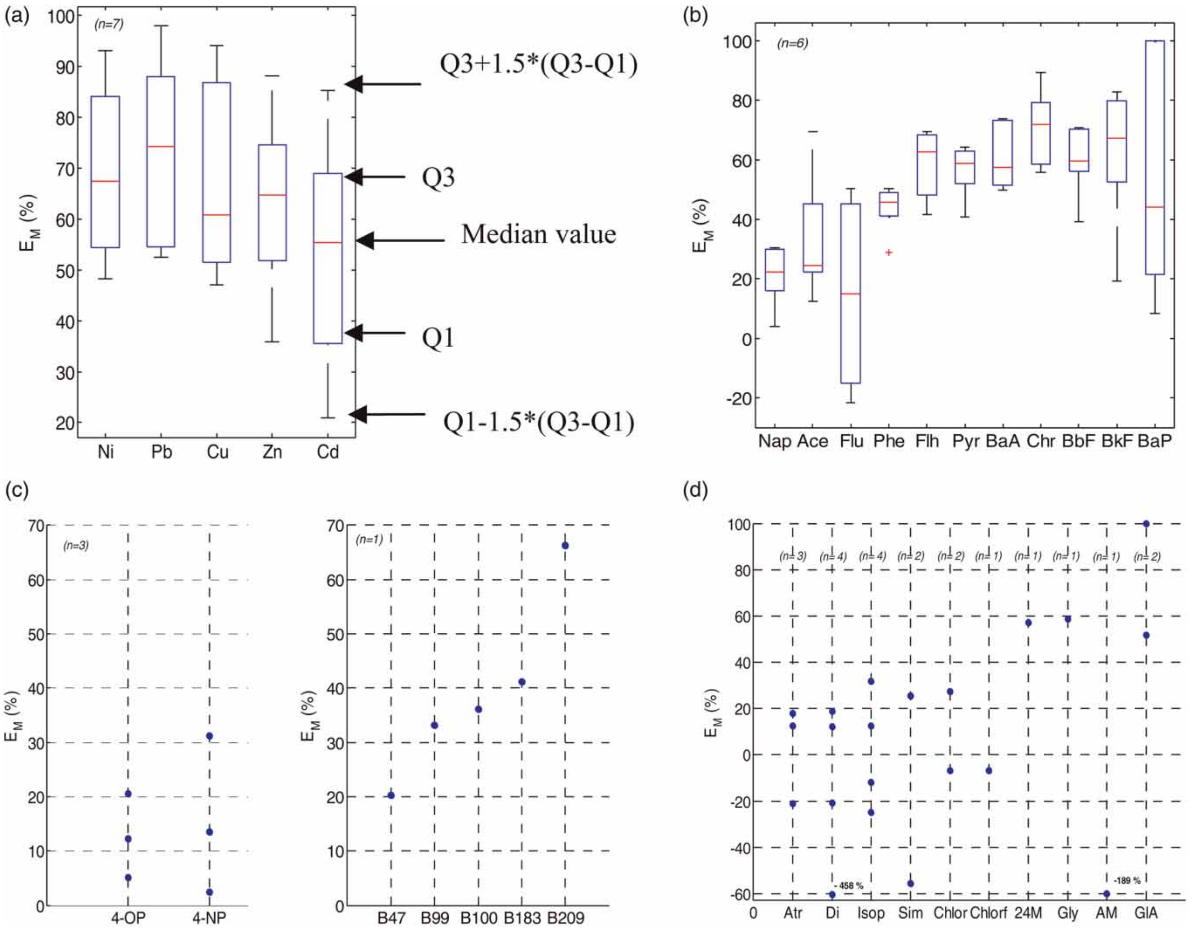

Figure 1 | Event mass efficiency (E_M) depending on the number of campaigns *n*. Boxplot (Q3: 75% of values and Q1: 25% of values): (a) heavy metals, (b) PAHs. Scatterplot: (c) alkylphenols and PBDEs, (d) pesticides.

The efficiency in terms of alkylphenols varies between 2% and 31% with a median value of 14% for 4-nonylphenol (4-NP) and from 5% to 21% with a median value of 12% for 4-tert-octylphenol (4-OP) (Figure 1(c)). Whatever the campaign, the efficiency remains low.

PBDE removal efficiency was only evaluated on one campaign. Figure 1(c) shows median values ranging from 20% to 60% depending on the compound. These not well-known MP are flame-retardants whose use is strictly regulated (EC 2003). BDE209, the most wide spread in the environment, presents the highest bromine atom number and seems to have the best efficiency (around 66%) compared to the others (from 20% for BDE47 to 41% for BDE183).

Lastly the retention basin does not seem to trap pesticides (Figure 1(d)). Some negative E_M values are found and indicate that mass at the outlet can be higher than at the inlet. This could indicate a partial release and/or transformation of pollutants in the sediments accumulated in the basin. For example, glyphosate and ammonium glyphosate present median E_M values higher than 50% but AMPA, which is a glyphosate degradation product, is released ($E_M = -189\%$). Results concerning glyphosate removal must be confirmed because there is variability. For example, a previous study indicated highway retention basin efficiency from 0% to 60% concerning this herbicide (Scholes *et al.* 2005).

Particulate distribution

The TSS E_M value was evaluated during the 10 campaigns. The median value is about 65% (from 35% to 87%), which is in the range of values found in the literature on such systems (e.g. Li & Pyatt 2004; Hossain *et al.* 2005; US-EPA 2008).

For other pollutants several tendencies can be identified.

Inlet and outlet particulate distributions of heavy metals, PAHs and most of the pesticides can qualitatively explain the different E_M ranges. For instance, copper that enters the retention basin mainly in particulate fraction (median value of 86%) and is also released with a particulate distribution (about 59%) presents a rather good median efficiency (61%). On the contrary, acenaphthene inlet/outlet particulate fractions which are respectively about 53% and 23% (so present rather in dissolved phase) show a low efficiency (median value of 24%). Atrazine, diuron, isoproturon, simazine and chlorfenvinphos are mainly in dissolved phase both at inlet and outlet ($EMC_p < LOD_p$ for diuron) and are not trapped (median $E_M = -4\%$ for diuron).

Therefore we could have thought that the more particulate the pollutants are at the inlet and outlet, the more efficient the basin would be. However, some exceptions were found. Alkylphenols, although poorly trapped (median E_M values about 12% for 4-OP and 14% for 4-NP), present particulate distributions at the inlet and outlet which are not especially low (54% and 43% for 4-OP and 57% and 48% for 4-NP).

Another tendency can be observed for PBDEs. According to the first results (campaign E), all of the nine PBDEs were mainly particulate, both at inlet and outlet (about 85%). However, the E_M values depend on the PBDE studied and range from 20% for BDE47 to 66% for BDE209. So, as for alkylphenols, it also seems that particulate distribution and pollutant removal are not so well linked.

The last observation is related to glyphosate and its product of degradation, AMPA. These two compounds are mainly particulate (like ammonium glyphosate) but glyphosate is trapped ($E_M = 59\%$) whereas AMPA is released ($E_M = -189\%$). So, glyphosate could be trapped in the basin and transformed into AMPA which could be released further. This conclusion will have to be confirmed by other campaigns and analyses.

In conclusion of this part, settling phenomena and particulate distribution of pollutants are not the only processes explaining pollutant removal in a large, dry retention basin. Other processes, developed in different studies (e.g. Scholes *et al.* 2008), can be responsible for the behavior of the chemical contaminants and have to be taken into account, in particular in models.

Retention basin impact on ecotoxicity

Ecotoxicity tests were conducted during five campaigns on event mean samples both at inlet and outlet.

The ecotoxic effects on ostracods and rotifers are presented in Figure 2.

According to the standard ISO 17616 (2008), biological effects on ostracods and rotifers could indicate (i) a significant inhibition of growth (ostracods) or reproduction (rotifers) when more than 30% of the population is impacted, or (ii) no significant inhibition when less than 30% of the population is impacted, or (iii) a stimulation when less than −30% of the population is impacted.

An inter-event variability can be observed for the two chronic tests.

Ostracod tests showed a low effect on stormwater for all campaigns (i.e. the inhibition results are below 30% or above −30%) except for one campaign (F), which highlighted a beneficial effect of the retention basin. The outlet sample showed a high, significant stimulation of organism population growth. This stimulation could have been due to the presence of nutritive elements in this sample. However the results obtained with the rotifer test were totally the opposite. These results highlighted the mixture effects of pollutants regarding the sensitivity of organisms (Perrodin *et al.* 2010). For this campaign the rainfall characteristics did not present a significant difference from the other ones.

Concerning rotifer reproduction, two events indicated an inhibition effect higher at inlet than outlet. One campaign (H) presented significant inhibition (56%) at the

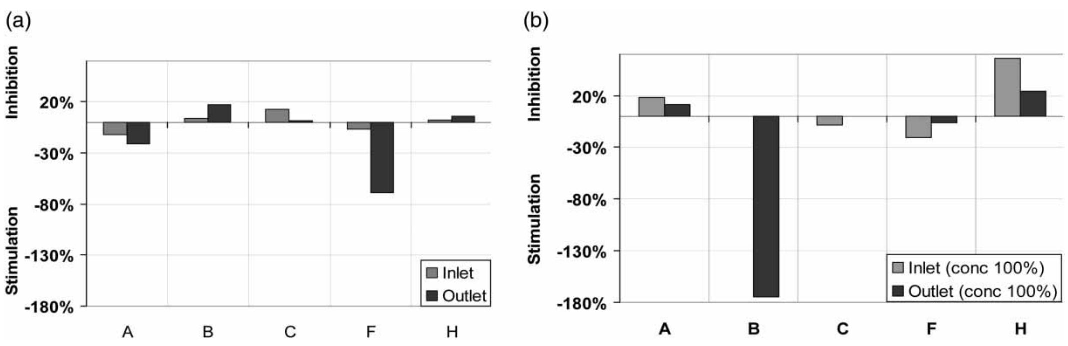

Figure 2 | Ecotoxic effects both at inlet and outlet, (a) on ostracods, (b) on rotifers.

inlet, and another (B) significant stimulation at the outlet (inhibition = −175%). With reference to the rainfall characteristics (Table 1), the dry weather duration before campaign (B) was the greatest of important of the five campaigns (nine days). In this specific situation, the high rotifer reproduction stimulation effect at the outlet could indicate a beneficial effect of the retention basin on ecotoxicity. During campaign (H), rainfall duration was about 25 hours coupled with the highest total rainfall depth (25 mm). In this situation, very different from the previous one, there was also a positive effect of the basin with a decrease of rotifer reproduction inhibition between inlet and outlet.

With these few campaigns, it is difficult to identify a real effect of a retention basin on ecotoxicity. The tests applied to a global water sample are maybe not the most relevant methods for evaluating basin ecotoxicity. Nevertheless, these preliminary results repeatedly showed a potential positive effect of the retention basin and a low (but not insignificant) toxicity of stormwater, as already observed in another study (Tang *et al.* 2013). Additional campaigns are now necessary to reach conclusions on the relevance of the methodology or on clear tendencies. Other types of catchments and other bioassays applied to the solid phase as previous works suggested (Gonzalez-Merchan *et al.* 2014) should be tested.

CONCLUSIONS

Analysis of the micropollutant removal efficiency and toxicity of a stormwater dry detention basin was undertaken over 10 campaigns, with samples taken both at the inlet and outlet.

The retention basin impact is consistent with the particulate distribution of some mineral and organic compounds (heavy metals, PAHs and pesticides), but several results indicate that settling is not the main process responsible for pollutant removal, in particular for alkylphenols and PBDEs. The accumulation of sediments and vegetation at the bottom of the basin could be responsible for other processes like biodegradation or volatilization. These aspects will be the focus of future research. The retention basin impact on ecotoxicity could also be confirmed.

The next step of this research work will concern the verification of existing models from the experimental data. For that purpose, the Stormwater Treatment Unit model for Micro-Pollutants (STUMP) (Vezzaro *et al.* 2011) will be tested, because of its integration of a wide range of processes. Further study will also be undertaken on the combined effect of mixed pollutants on ecotoxicity.

From a global point of view, impact assessment of such a ‘real’ system is not trivial. Accumulation of sediments and hydrodynamic and chemical transformation processes impact the behavior of micropollutants. More sampling events are obviously required and other sites need to be observed to explore a wider range of situations linked to catchment activities and land uses.

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