Stormwater retention basin efficiency regarding micropollutants loads and ecotoxicity

Efficacité évènementielle d'un bassin de retenue-décantation des eaux pluviales urbaines sur les flux de micropolluants et l'écotoxicité des rejets

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RÉSUMÉ

L'efficacité des bassins de retenue-décantation des eaux pluviales a été peu étudiée notamment vis-à-vis de certains polluants pointés par la directive Cadre sur l'Eau en 2000 comme les pesticides, les PBDEs ou encore les alkylphénols. Le travail présenté ci-après s'appuie sur l'étude *in-situ* d'un bassin de retenue-décantation des eaux pluviales afin d'évaluer sa capacité à piéger les micropolluants à l'exutoire d'un bassin versant industriel drainé par un réseau séparatif pluvial recevant quelques eaux de temps sec supposées propres. Le suivi de 94 substances réparties dans 5 familles de polluants (Métaux, HAPs, PBDEs, Alkylphénols et pesticides) a été réalisé, au cours de 10 campagnes sur des rejets urbains par temps de pluie en entrée et en sortie du bassin. En parallèle, l'effet du bassin sur l'écotoxicité des rejets a été évalué. Les premiers résultats montrent une variabilité inter-évènementielle importante d'un point de vue chimique et écotoxicologique. Ils indiquent un abattement évènementiel de la pollution métallique et de certains hydrocarbures aromatiques polycycliques. Les pesticides présents principalement sous forme dissoute ne sont pas retenus. L'étude de la fraction particulaire des micropolluants a mis en évidence que le phénomène de décantation n'est pas le seul processus responsable de l'élimination des polluants notamment pour les Alkylphénols et PBDEs.

ABSTRACT

Retention basins efficiency in micropollutants removal has not been very well studied in particular for pollutants pointed out by the European Water Framework Directive in 2000 such as pesticides, PBDEs or Alkylphenols. This study is based on *in-situ* experiments carried out on a stormwater retention basin. The aim is to estimate the basin efficiency in trapping and removing micropollutants from stormwater run-off at the outlet of an industrial catchment drained by a separate stormwater sewer system receiving some dry weather effluent supposed to be clean. 94 substances from 5 families (Metals, PAHs, PBDEs, Alkylphénols and pesticides) were analyzed during 10 event campaigns in urban wet weather discharges at the inlet and outlet of the basin. The effect on ecotoxicity was also assessed. First results show high inter-event variability as regards both chemical and ecotoxic characterization. They indicate a good event efficiency concerning heavy metals and most of PAHs. Studied pesticides, mainly found in dissolved fraction were not trapped. Particulate fractions study highlighted settling is not the main process occurring in the retention basin and responsible for micropollutant removal in particular for Alkylphenols and PBDEs.

KEYWORDS

Dry retention basin, Ecotoxicity, Micropollutant, Run-off, Stormwater

1 INTRODUCTION

In France, since decades, large retention basins were implemented to mitigate stormwater floods impacts and pollution in urban areas.

In 2000, the European Water Framework Directive (WFD) outlined high ambitions, with an objective of reduction of pollutants emissions in receiving water by 2015. In addition to traditional macropollutants (Total Suspended Solids (TSS), organic matter), the notion of micropollutants (MP) and compliance values EQS were raised (EC, 2000).The European Directive prescriptions have entailed numerous studies on micropollutants behaviour at a catchment scale (Zgheib, 2009; Bressy, 2010; Lamprea, 2010; Dembele; 2010; Birch, 2012) where contributions from the catchment and wash-off were highlighted and assessed (e.g Eriksson *et al*., 2005; Becouze-Lareure; 2010).

If micropollutant loads have been studied at the outlet of urban catchments and in the receiving water courses, very few research work have focused on the effect of a large dry retention basin on a wide list of micropollutants (in particular organic ones) at the outlet of a separate sewer system. Existing researches generally deal with stormwater ponds (not dry) and/or with few micropollutants. The paper addresses this question and is completed by an ecotoxicity assessment which is also poorly reported in the literature.

2 METHODS

2.1 *Monitoring system*

2.1.1 Description of the site

All experiments were conducted in a large dry retention basin (Django Reinhardt) situated at Chassieu near Lyon, France (see Figure 1). This basin is located at the outlet of an industrial catchment (185 ha and 75% of 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. These values indicate the specific context of this study dealing with a large basin (230 $m^3.ha\ imp^{-1}$) and a low outflow rate (2.5 $l.s^{-1}.ha\ imp^{-1}$) which is very common in France. The basin was completely scrapped and the sediment removed in early 2006. The pipe network and then the basin collect dry weather flows, in fact “clean” water (or supposed to be clean) coming from cooling of industrial processes. Dry weather effluents represent 26% of the total inlet volume and 20% of the total mass of suspended solids between 2004 and 2010 (Gonzalez-Merchan, 2012).

Figure 1. Django Reinhardt retention basin (source : Google map - 2012)

2.1.2 Monitoring system

Pollutant concentrations and ecotoxicity were evaluated from samples taken with a refrigerated automatic samplers and composed on a flow proportional sampling basis. For the micropollutants selected in the study, two types of samplers were used: one with 24 x 0.9 L glass bottles for most of the organic compounds (*Hach Lange Bühler* equipment) and one with 24 x 0.9 L polyethylene bottles for metals, specific pesticides such as Glyphosate, Glyphosate ammonium and AMPA (Aminoethylphosphonic acid) and ecotoxicity (*Hach Lange Sigma* equipment). Samples were sent to specific research laboratories for the analysis of both fractions dissolved and particulate, which is

rather innovative in this field, the analytical procedures being specifically calibrated for stormwater and sediments found in the basin. Traditional blanks were also done. As huge volumes would have been necessary to analyze all of the selected micropollutants, a sampling planning procedure was adopted according to the type of event. Therefore all of them were not systematically analyzed for all the events (see part 3.1 and (Sébastian *et al.,* 2011) for more details).
Total suspended solids (TSS) and Volatile Suspended Solids (VSS) were analyzed according to AFNOR NF T.90-105 and NF T90-029 standards.

As the samples were composed with a flow proportional strategy, a continuous measurement system (2 minute time step) was installed at the inlet and outlet of the basin for inflow, outflow rate, and for qualitative parameters evaluation such as pH, specific conductance, turbidity and temperature.
To characterize rain events, a rain gauge recorder measuring rainfall intensity at 1 minute time step was used on the site.

2.2 Micropollutants

2.2.1 Substances to analyze

Initially, the European Water Framework Directive requirements integrated 33+8 substances defined as priority or priority hazardous substances (EC, 2000; EC, 2008).These substances were studied in previous research programs, in particular at the outlet of Chassieu catchment (Becouze-Lareure, 2010). In our study, most of these micropollutants have been analyzed completed by emerging substances not well known in terms of potential sanitary hazard but supposed to have some. Table 1 presents the list of the 94 substances studied in this paper. Names and acronyms are presented in details in the Annex and for better understanding, several groups were created.

Table 1. Number of Micropollutants studied (total, priority and hazardous substances according to (EC, 2000; EC, 2008) per family of substances

Family	Number of substances studied	Number of Priority substances	Number of Priority hazardous substances
Metals	22	2	1
PAHs	16	2	6
Pesticides	45	13	3
Alkylphenols	2	1	1
PBDEs	9	0	9
Total	**94**	**18**	**20**

2.2.2 Experimental Data processing

Micropollutants concentrations are Event Mean Concentrations (EMCs) and are defined both in dissolved and particulate fractions, according to previous works (Zgheib *et al*., 2011):

$$EMC = EMC_d + EMC_p \quad (1)$$

With EMC_d Event Mean Concentration measured in dissolved fraction (µg.L^{-1} for metals or ng.L^{-1} for organic compounds) and EMC_p Event Mean Concentration in the particulate fraction (µg.L^{-1} for metals or ng.L^{-1} for organic compounds) obtained by the MP mass concentration (µg.g^{-1} or ng.g^{-1}) multiplied by the TSS concentration (g.L^{-1}).

Mass of micropollutants is then calculated according to Equation (2):

$$M_i = EMC_i * V_i \quad (2)$$

With M_i the inlet or outlet MP mass (µg for metals or ng for organic compounds) and V_i inlet or outlet volume during the run-off event (L), 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:

$$E_M = \frac{M_I - M_o}{M_I} * 100 \quad (3)$$

With M_I and M_O, respectively the inlet and outlet MP masses (µg for metals or ng for organic compounds).

2.3 Ecotoxicity

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

Ostracods mortality and growth inhibition were studied with Ostracodtoxkit® standard procedure (ISO 14371, 2012). This test was initially used to assess the toxicity of the sediments. In this work, it was chosen to conduct this test on the total sample, using standard freshwater as control test according to previous studies (Angerville, 2009; Becouze-Lareure *et al*., 2012).

Rotifers reproduction was also studied on total samples with Rotoxkit® standard procedure (PR NF ISO 20666 2007).

3 RESULTS AND DISCUSSION

3.1 Campaigns characteristics and MP occurrence

The results presented in the paper were obtained during 10 sampling campaigns conducted in the retention basin, both at inlet and outlet. 7 campaigns were carried out on 5 heavy metals (Ni, Pb, Cu, Zn, Cd), 4 on a larger list of metals, 6 on PAHs, 5 on ecotoxicity, 3 on Alkylphenols, from 1 to 4 for the pesticides depending on the family studied and just one for PBDEs. TSS concentration was systematically analyzed (Table 2).

Table 2. Campaigns and occurrence of MP

N°	date	TSS	VSS	MeI	MeII	PAH	Ap	PBDE	PeI	PeII	PeIII	PeIV	Ecot.
A	2011-07-08	X		X									X
B	2011-10-19	X		X									X
C	2011-12-07	X				X	X		X		X		X
D	2012-01-05	X	X							X	X		
E	2012-03-18	X	X	X	X			X				X	
F	2012-04-03	X				X	X		X		X		X
G	2012-04-11	X	X	X	X	X						X	
H	2012-05-20	X		X		X	X		X		X		X
I	2012-07-03	X		X	X	X							
J	2012-09-12	X		X	X	X							
n		*10*	*3*	*7*	*4*	*6*	*3*	*1*	*3*	*1*	*4*	*2*	*5*
N. of substances		-	-	5	17	16	2	9	15	20	7	3	-
N. of substances detected **		-	-	5	17	15	2	6	3	2	3	3	-
N. of substances quantified **		-	-	5	17	15	2	5	3	2	2	3	-

**at least once at inlet and/or outlet

The ten campaigns correspond to 10 rain events whose total rainfall depth and duration are plotted among all the events observed between 01/01/2010 and 18/09/2012 (367 events) (see Figure 2). The graph shows a good representativeness of rainfalls on the period. Other characteristics of the rainfalls are given in Table 3. It can be noticed that 2011 was a very dry year in the middle-Est of France.

Concerning micropollutants, concentrations of all substances quantified at the inlet of the basin were in the range of common concentrations found in literature (Zgheib, 2009; Becouze-Lareure, 2010; Bressy, 2010). We can also notice that micropollutants masses were not linked with antecedent dry weather periods but with run-off volumes. For example, inlet mass of Benzo(a)pyrene varied from 583 mg to 230 mg between campaigns I and J (antecedent dry weather period being resp. equal to 1.8 d and 9.8 d).

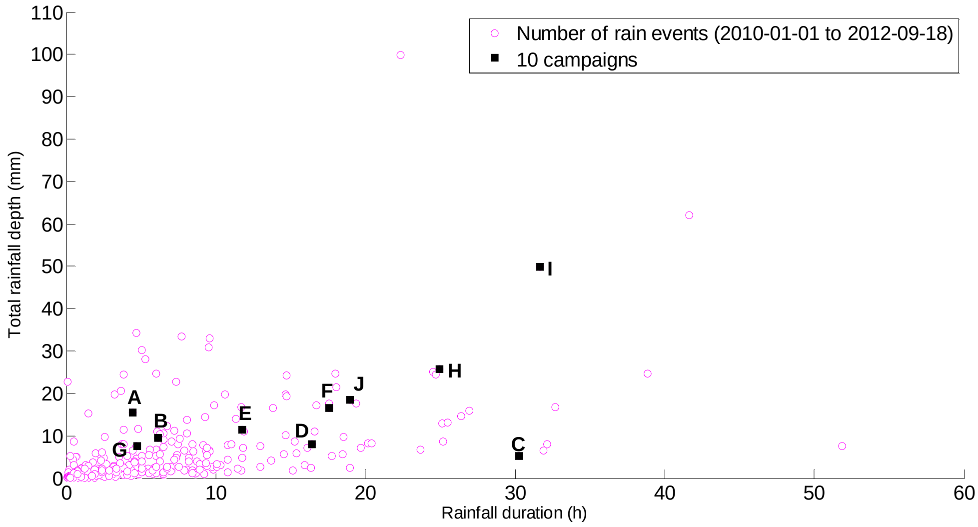

Figure 2. Total rainfall depth Vs rainfall duration of rainfalls observed between 01/01/2010 to 18/09/2012 (dark dots correspond to our 10 campaigns)

Table 3. Rainfall characteristics

	Rainfall duration	Total rainfall depth	Antecedent dry weather period	Max. intensity	Max. intensity (5 minute time step)
	h	*mm*	*d*	*mm/h*	*mm/h*
A	4.4	15.4	1.2	3.5	68.5
B	6.1	9.6	9.1	1.6	7.6
C	30.3	5.3	0.5	0.2	2.2
D	16.4	8.0	0.9	0.5	2.2
E	11.8	11.5	0.7	1.0	4.7
F	17.6	16.5	0.9	0.9	6.4
G	4.7	7.6	0.2	1.6	6.2
H	25.0	25.7	0.9	1.0	26.2
I	31.6	50.0	1.8	1.6	22.7
J	19.0	18.5	9.8	1.0	19.0

3.2 Retention basin impact on MP

3.2.1 Event Mass Efficiency

Figure 3 shows Event Mass Efficiency (E_M) of the different substances and for the 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, uncertainties were not calculated yet in details so results have to be carefully interpreted. A first approach indicates analytical uncertainty around 25 %.

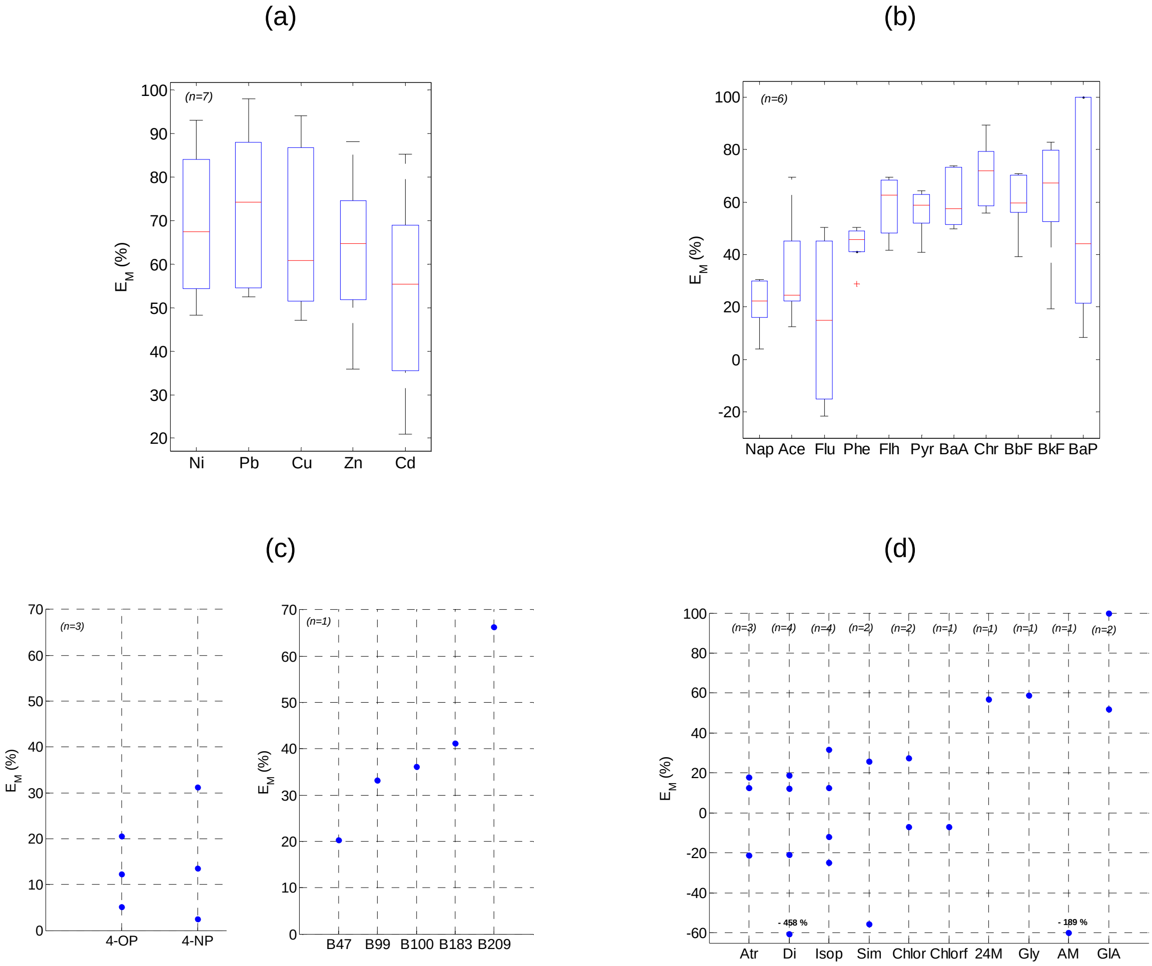

Figure 3. Event mass efficiency (E_M) depending on the number of campaigns *n*

(a) Heavy metals, (b) PAHs, (c) Alkylphenols and PBDEs, (d) Pesticides.

Event Mass Efficiency of the basin for Nickel, Lead, Copper and Zinc (Figure 3(a)) showed median values ranging from 60 to 74% (n=7) which is coherent with the results found in the literature (e.g. Chebbo and Bachoc, 1992; Hares *et al.,* 1999). The values of Cadmium efficiency (median: 55 %, mean: 53%) is a little bit lower but higher than those of the literature (e.g. (US-EPA, 2008) which indicates 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 (Metals II (Annex)) obtained on 4 campaigns, median values (not presented in Figure 3) are generally higher than 50% for most of them except Vanadium, Strontium, Calcium; Potassium and Sodium whose median values are respectively 46%, 41%, 41%, 31% and 23%.

For PAHs (Figure 3(b)), analyzed on 6 campaigns, E_M values seem to increase with the number of aromatic hydrocarbon rings. Benzo(k)fluoranthene (5 rings) is better trapped than Acenaphtene (3 rings) with a median E_M value of 67% and 24% respectively. This is also coherent with literature (e.g. Pitt *et al*, 1999; Hwang *et al.*, 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). Naphtalene (2 rings) E_M varies from 4% to 31%, this result can be compared to literature data indicating this compound is not trapped (Moy *et al.,* 2003).

Alkylphenols have been studied on 3 campaigns until now. The efficiency varies between 2% and 31% with a median value of 14 % for 4-Nonylphenol and from 5% to 21% with a median value of 12 % for 4-Tert-Octylphenol (Figure 3 (c)). Whatever the campaign, the efficiency remains low even if further campaigns are necessary to confirm it.

PBDEs removal efficiency was only evaluated on 1 campaign (Figure 3(c)). The median values range from 20 to 60 % depending on the compound. These micropollutants, not well-known, are flame-retardants whose use is strictly regulated (Ayrault *et al*., 2009). BDE209, the most spread in the environment (La Guardia, 2006), presents the highest Brome atoms number and seems to have the best efficiency (around 66%) compared to the others (from 20 % for B47 to 41% for B183).But other measurements are also necessary.

Finally, according to the results obtained on pesticides (Figure 3(d)), it seems that the retention basin does not trap these organic compounds. Some negative E_M values are found and indicate that mass at the outlet can be higher than at inlet. This can be explained by partial release and/or transformation of pollutants in the sediments accumulated during 6 years. For example, Glyphosate and Glyphosate ammonium that present median E_M values higher than 50% whereas AMPA, which is a Glyphosate degradation product, is released (E_M=*-189%)*. These results must be confirmed but previous study showed efficiency variability concerning Glyphosate removal in two highway retention basins (from 0 to 60%) (Scholes *et al*., 2005).

3.2.2 Particulate distribution

Particulate distribution of the different pollutants was analyzed, thanks to particulate content, in order to detect a potential relationship with the event mass efficiency E_M .The first results highlighted several tendencies.

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 literature on such systems (e.g. Adams and Papa, 2000; Hossain *et al.,* 2005; Lie and Pyatt, 2004; US-EPA, 2008).

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 enters the retention basin mainly in particulate fraction (median value of 86%), is released with a particulate distribution too (about 59%) and presents a rather good median E_M *value of* 61%. On the contrary, particulate fraction of Acenaphtene is about 53% and 23%, respectively at the inlet and outlet (so rather present in dissolved phase). In this case, the efficiency is not so good (median value of 24%). Atrazine, Diuron, Isoproturon, Simazine and Chlorfenvinphos are mainly in dissolved phases both at inlet and outlet ($EMC_p < LOD_p$), except for Diuron for which EMC_p <LOQ_p. No surprisingly, they 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 a particulate distributions at the inlet and outlet which are not especially low (54% and 43 % for 4-Tert-Octylphenol and 57% and 48 % for 4-Nonylphenol).

Another tendency can be observed for PBDEs. According to the first results (campaign E), all of the 9 PBDES were mainly particulate, both at inlet and outlet (about 85%). However, the E_M values depend on the PBDE studied and ranges from 20% for B47 to 66% for B209. So, like for Alkylphenols, it also seems that the particulate distribution and pollutant removal are not so well-linked.

The last tendency is linked to Glyphosate and its product of degradation AMPA. These two compounds are mainly particulate (like Glyphosate ammonium) but Glyphosate seems to be 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 further released. This conclusion has to be confirmed by other campaigns and analyses.

In conclusion of this part, settling phenomenon and particulate distribution of pollutants are not the only parameters explaining pollutant removal in a large dry retention basin. Other process, 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.

3.3 Retention basin impact on ecotoxicity

Ecotoxicity tests were conducted during 5 campaigns on event mean samples both at inlet and outlet (seeTable 2).

The ecotoxic effects on ostracods and rotifers are presented in the next figure (see Figure 4).

According to the standard (ISO 17616, 2008), biologic effects on ostracods and rotifers indicate:

- a significant growth (ostracods) or reproduction (rotifers) inhibition when more than 30% of the population is impacted,
- no significant inhibition when less than 30% of the population is impacted,
- a stimulation when less than -30% of the population is impacted.

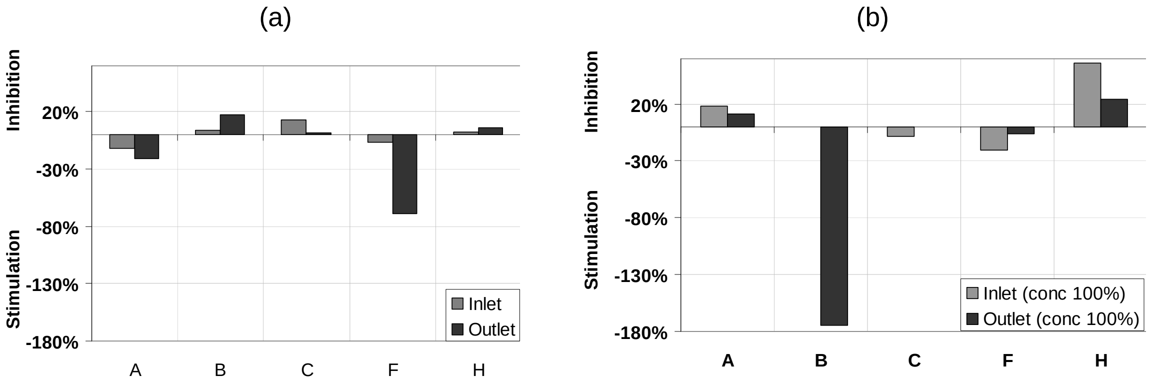

Figure 4. Ecotoxic effects both at inlet and outlet. (a) on ostracods (b) on rotifers

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

Concerning ostracods, 3 events indicate growth inhibition and 2 events indicate stimulation both at inlet and outlet. All of the inhibition results are below 30% or above -30%, except for one campaign (F), so there is rarely significant ecotoxic effect, according to the standard (ISO 17616, 2008).

Concerning rotifers reproduction, 2 events indicate inhibition higher at inlet than outlet. One campaign (H) presents significant inhibition (56%) at the inlet, and another (B) significant stimulation at the outlet (inhibition = -175%).

Comparing to the rainfall characteristics (Table 3), dry weather duration before campaign (B) is the most important of the 5 campaigns (9 days). In this specific situation, the high rotifers reproduction stimulation effect 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 is also a positive effect of the basin with a decrease of rotifers reproduction inhibition between inlet and outlet.

During campaign (F), there is a beneficial effect of the retention basin on ostracods growth (stimulation).

With these few essays, it is difficult to conclude on the real effect of a retention basin on ecotoxicity. Nevertheless, we have observed positive effects of the retention basin repeatedly. Additional campaigns are now necessary to confirm these first results.

4 CONCLUSIONS AND OUTLOOK

In conclusion, the 10 campaigns conducted both at inlet and outlet of a large basin provide several results on chemical and ecotoxic characterization of retention system effect assessment.

The retention basin impact is coherent with the particulate distribution of some mineral or organic compounds (heavy metals, PAHs and pesticides) but several results indicate the settling parameter is not the main criterion responsible for pollutants removal in particular for Alkylphenols and PBDEs. The accumulation of sediments and vegetation at the bottom of the basin could be responsible for other process like biodegradation or volatilization. These aspects have now to be investigated. Retention basin impact on ecotoxicity could also confirm if the other process are predominant.

The next step of this research work will concern (i) the verification of existing models thanks to the experimental data, in particular the Stormwater Treatment Unit model for Micro-Pollutants (STUMP) (Vezzaro *et al.,* 2011) because numerous processes can be integrated and (ii) further study on combined effect of mixed pollutants on ecotoxicity.

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ANNEX

Metals I	Nickel Ni	Lead Pb	Copper Cu	Zinc Zn	Cadmium Cd
Metals II	Arsenic As	Chrome Cr	Strontium Sr	Titane Ti	Vanadium V
	Aluminum Al	Iron Fe	Manganese Mn	Mollybdène Mo	Platine Pt
	Phosphore P	Sodium Na	Potassium K	Magnésium Mg	Calcium Ca
	Baryum Ba	Cobalt Co			
PAHs	Naphtalene Nap	Acenaphthylene Acy	Acenaphtene Ace	Fluorene Flu	Phenanthrene Phe
	Benzo(b)fluoranthene BbF	Benzo(k)fluoranthene BkF	Benzo(a)pyrene BaP	Indeno(1,2,3-cd)pyrene IP	Dibenzo(a,h)anthracene Dah
	Fluoranthene Flh	Pyrene Pyr	Benzo(a)anthracene BaA	Chrysene Chr	Anthracene A
	Benzo(g,h,i)perylene Bper				
PBDEs	BDE28 (tri) B28	BDE47 (tétra) B47	BDE99 (penta) B99	BDE100 (penta) B100	BDE154 (hexa) B154
	BDE183 (hepta) B183	BDE205 (octa) B205	BDE209 (nona) B209	BDE153 (hexa) B153	
Pesticides I	Alachlor Ala	Atrazine Atr	Simazine Sim	Chlorpyrifos Chlor	Delta hexa Dhex
	Op DDT Op DDT	Pp DDT Pp DDT	Endrine End	Alpha hexa Ahex	Endosulfan beta Enb
	Gama hexa Ghex	DDD pp DDD pp	DDE pp DDE pp	Beta hexa Bhex	Trifluralin Tri
Pesticides II	Metaldehyde Meh	Mecoprop Mec	2_4_D 24D	2_4, MCPA 24M	S-metolachlore Sme
	Carbendazim Car	Isothiazolinone Itz	Chlorothalonil Clo	Pendimethalin Pen	Acetochlore Ato
	Metazachlor Met	Tebuconazole Teb	Epoxiconazole Epo	Diflufenicanil Dif	Deltamethrine Del
	Fenpropidine Fen	Trichlopyr Trp	Folpel Fol	Irgarol 1051 Irg	Terbutryne Ter
Pesticides III	Diuron Di	Endosulfan Alpha Ena	Aldrin Ald	Isodrin Iso	
	Chlorfenviphos Chlorf	Isoproturon Isop	Dieldrin Die		
Pesticides IV	Glyphosate Gly	Glyphosate ammonium GlA	AMPA AM		
Alkylphenols	4-Tert-Octylphenol (4-OP)	4-Nonylphenol (4-NP)			