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ABSTRACT: The first flush phenomenon can be defined as the initial period during which the concentration of pollutants was higher than those in latter stage of rain events (Gupta and Saul 1996). Researches have shown that diverting the first flush from the harvested rainwater may improve the quality of stored water. In this study, a special first flush volume harvesting system has been setup in two houses located in two different areas in the Ile de France (France) and 13 storm events during 8 months have been collected. The results show the correlation between some parameters. Amount them, TSS and turbidity show a clear decline, which have the potential to describe the first-flush effect. Otherwise, there was no evidence that can prove the existence of such first flush phenomenon in terms of the microbial indicators. The first flush effect can be also linked to the rain events factors such as rain intensity and antecedent dry time.
KEY WORDS: roof-harvested rainwater, first-flush, water quality

1 INTRODUCTION

In recent years, water scarcity is one important issue faced by the population. In order to meet the increasing demand of water in terms of quality and quantities, effective rainwater harvesting is an essential water management strategies (Lee and *al.* 2012). Amount many methods used to capture rainwater, roof rainwater harvesting, which means collecting the rainwater from a rooftop by a convey system and storage in a tank, has been used in many countries for thousands of years (Simmons and *al*. 2001). It offers considerable potential as an alternative water supply and has been involved with the concept of sustainable development to reuse natural resources.

Although there is significant economic and environmental benefits of roof harvested rainwater (Hatibu and *al*. 2006; Sturm and *al*. 2009), it is necessary to investigate and to improve the water quality in order to reduce the potential health risks by removing chemical and microbial contaminants. Some studies show that the first-flush diversion is recognized as one of the useful methods to improve rainwater quality. It can treat roof runoff prior to storage and reduce both suspended and

dissolved contaminated loads in rainwater systems (Martinson and Thomas 2009; Amin and *al*. 2013; Lee and *al*. 2012; Zobrist and *al*. 2000). Such first flush system is relying on the concept of first-flush phenomena, which can be defined as the first portion of the total volume that contains the main portion of the pollutant during a storm event(Gupta and Saul 1996; Doyle 2008).
Nevertheless, several questions now have been raised: first of all, the variations of the runoff water quality need taking into account both the level of physic-chemical contamination and the level of microbial contamination. Secondly, actors impacting the phenomena such as the characteristics of roof, the precipitation event and other meteorological factors need further study. Finally, it is vital to analyse the rainwater quality and define the first-flush phenomenon by using common parameters.
The main objective of this study is to analyze the variation of the runoff water quality, the existence of the first-flush and the impact factors by monitoring both physical-chemical and microbiological parameters.

MATERIAL AND METHODS

1.1 Sampling site

The experimental equipments have been implemented on the roof of two houses located in two different areas in Ile de France (France), shown in Figure 1. It has a marine west coast climate that is mild with no dry season, warm summers. Heavy precipitation occurs during mild winters and the average annual precipitation is 619 mm. Seasonality is moderate.
The first experimental site is Vincennes, which is a commune in the eastern suburb of Paris. It is located 6.7 km from the center of Paris, 3.3 km from the largest public park in the city (bois de Vincennes) and 2.5 km from the highway. The roof is next to a small garden, covered with a 25 years tile and with a $10m^2$ projected area. The second site is located in Romainville, which is also a commune in the eastern suburb of Paris. It is located 7,2 km from center and 1 km from the highway A3. No industrial zone is near the site. The projected area of this tiles roof is $20m^2$.

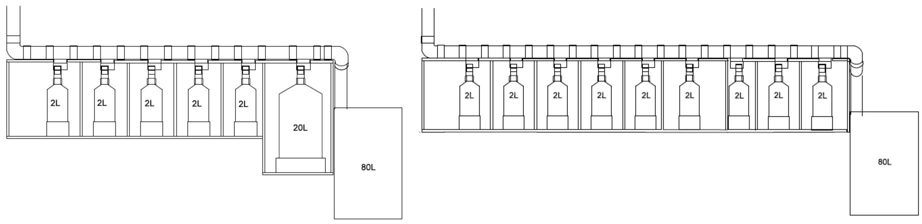
Figure 1. Location of the experimental sites

In both sites, a tipping bucket rain gauge (tipping every 0.2mm of the rain) is installed in the garden next to the experimental roof. The amount of rainfall, rainfall intensity and antecedent dry weather period were recorded.

1.2 Experimental design and sample collection

Self-made movable experimental equipment had been designed and constructed. It consists of three parts: 1) a series of PVC pipes for connection; 2) several brown glass bottles SCHOTT DURAN which can be sterilized easily by autoclave. Ideally, the first rain falls on the roof and fills the first bottle. Once a bottle is full, the runoff can flow directly over to the following bottle one after another; 3) an 80L PVC bucket that harvest the remaining runoff in order to calculate the runoff coefficient. Two equipments have successively been used. The first equipment had 6 bottles (Figure 2-a), which was consisted by five bottles of 2L and one bottle of 20L. Then it was upgrade to 9 bottles of 2L (Figure 2-b) in order to improve the accuracy of measurement.

So far, six available rainfall events were captured by the first equipment and then two events captured by the second in Vincennes. These two systems can capture respectively 3 mm and 1.8 mm of runoff. 5 events were captured by second equipment in Romainville and 0.9mm of runoff could be collected. 89 rainwater samples were collected and analyzed over a period of 10 months (from May 2015 to February 2016).



(a) Equipment 6 samples (b) Equipment 9 samples

Figure 2. Schematic diagram of the equipment design

All the sterile bottles were placed less than one day before each rain event storm thanks to the weather forecast. The samples were collected within 24 hours after the first drop of the rainfall and were immediately transferred to the laboratory to carry out the water quality analyses. The measurements of all the parameters were carried out in 24 hours after the collection.

1.3 Sampling analyses

The parameters, the methods and the equipment are summarized in Table 1. The choices of these physical-chemical and microbial parameters were based on the previous relevant studies (Yaziz and *al*. 1989; Lee and *el*. 2012; Vialle and *al*. 2011) and samples were analyzed on the basis of norms and some water quality standards (Table 1).

1.4 Data analysis

Three methods have been used to achieve the qualitative characterization of the rains events. 1) Basic statistics: Ranges, mean, medians and standard deviation were used to describe rainfall quality; 2) Correlation analysis to determine relationships between the parameters. The analysis should use the nonparametric Spearman coefficient because that the sample size of this study is too small; 3) concentration versus time plots to describe the variation of the pollutant during runoff events, and the dimensionless M (V) curve that compare the shape of the pollution/volume distributions from the different rainfall events.

The dimensionless M (V) curve is an assessments frequently used for the evaluation of first flush, which establish by the dimensionless cumulative mass fraction M (t) against the cumulative volumes fraction V (t). Equation (1) and (2) can be used for it.

$$M\ (t) = (\sum C\ (t)Q(t)\ \Delta t)/M \quad (1) \qquad V\ (t) = \ (\Sigma Q\ (t)\ \Delta t)/V \quad (2)$$

Where C (t) is the pollutant concentration at time t, Q (t) the flow rate at time t (L/sec), Δt the time stem (sec), M the total pollutant mass and V the total runoff volume (V).

Table 1. Review of analytical parameters and methods.

Parameters	**Methods and equipment**	**Reference standard**
	Physical-chemical parameters	
pH	Probe (Multi 340iWT®)	NF T 90-008
Conductivity ($\mu S.cm^{-1}$)	Probe (Multi 340iWT®)	NF EN 27888
Turbidity (NTU)	Turbidimeter (2100P Turbidimeter HACH®)	NF EN ISO 7027
TSS ($mg.\ L^{-1}$)	Filtration GF/F (Verrerie Sartorius® and filter)	NF EN 872
DOC ($mgC.\ L^{-1}$)	DOC meter (TOC-Vcsn SHIMADZU®)	NF EN 1484
	Microbiological parameters	
Enterococcus (ufc per 100ml)	Microplate MUG/EC BIORAD®	NF EN ISO
E. coli (ufc per 100ml)	Microplate MUG/SF BIOerRAD®	NF EN ISO
HPC at 22 °C (log ufc per ml)	Culture methods (PCA)	NF EN ISO-6222
HPC at 36°C (log ufc per ml)	Culture methods (PCA)	NF EN ISO-6222

2 RESULTS AND DISCUSSION

2.1 Quality assessment.

Table 2 summarizes the values maximum, minimum, mean, median and standard deviation in both Vincennes and Romainville.

2.1.1 Physical-chemical parameters.

Table 2. Quality of the roof runoff for the sample rain events

		pH	Conductivity (µS /cm)	Turbidity (NTU)	TSS (mg/l)	DOC (mg C/l)	Enterococcus (ufc/100ml)	E. *coli* (ufc/100ml)	HPC 22°C (log ufc/ml)	HPC 37°C (log ufc/ml)
Vincennes	**Maximum**	9.53	211	27.1	133.50	44.37	2900	260	5.2	5.0
	Minimum	6.76	31	2.6	2.00	1.78	32	0	2.7	2.3
	Median	7.67	105	6.7	14.24	6.20	370	0	4.0	3.9
	Mean	7.90	94	8.7	27.95	9.15				
	Standard deviation	0.77	39.16	5.95	34.01	8.06	669.33	62.88	0.63	0.68
Romainville	**Maximum**	9.67	99	15.9	122.74	9.72	1900	110	4.4	4.2
	Minimum	6.68	12	1.2	0.68	0.85	0	0	1.0	0.0
	Median	7.38	39	3.5	6.23	1.92	10	0	2.6	1.5
	Mean	7.60	41	4.3	12.41	2.61				
	Standard deviation	0.70	20.17	3.14	21.14	1.92	365.17	19.35	0.78	1.12

Table 3. Spearman's correlation between different variables. Values in italic are significant (Significant at the 5% level)

	pH	Conductivity	Turbidity	TSS	DOC	Enterococcus	E. *coli*	HPC 22°C
Conductivity	*0.412*							
Turbidity	*-0.240*	*0.503*						
TSS	*-0.311*	*0.352*	*0.919*					
DOC	0.207	*0.861*	*0.632*	*0.458*				
Enterococcus	0.113	*0.551*	*0.438*	*0.250*	*0.654*			
E. *coli*	-0.091	*-0.020*	*0.267*	*0.249*	0.055	*0.084*		
HPC 22°C	0.081	*0.407*	*0.552*	*0.479*	*0.597*	0.519	*0.235*	

pH. The pH of the harvested rainwater from all of the samples collected in Vincennes varies between 6.76 and 9.53, while it varies between 6.68 and 9.67 in Romainville. The average values are respectively 7.60 and 7.90. By comparison, the literatures have reported the following pH ranges for runoff water: 5.8-8.4 (Schriewer, Horn, and Helmreich 2008), 6.4-7.7(Doyle 2008), 5.6-10.4(Vialle et al. 2011).
Conductivity. The maximum values of conductivity are 211µS /cm in Vincennes and 99 µS /cm in Romainville, while the minimum values are respectively 31 µS /cm and 12 µS /cm. The mean values are 94 µS /cm and 41 µS /cm
Turbidity and Total Suspended Solids (TSS). The range of the turbidity values varies from 2.6 NTU to 27.1 NTU with an average value of 6.7 NTU in Vincennes and it varies from 1.2 NTU to 15.9 NTU with an average value of 4.3 NTU in Romainville. The ranges of TSS are 2 mg/l to 133.50 mg/l in Vincennes and 0.68 mg/L to 122.74mg/l. The means of TSS are respectively 12.41 mg/l and 27.95 mg/l.
Dissolved organic carbon (DOC). DOC values are between 1.78 mg C/l to 44.37 mg C/l in Vincennes and ranges from 9.72 mg C/l to 0.85 mg C/l in Romainville. The concentrations in Vincennes are much higher than Romainville.

2.1.2 Microbiological parameters

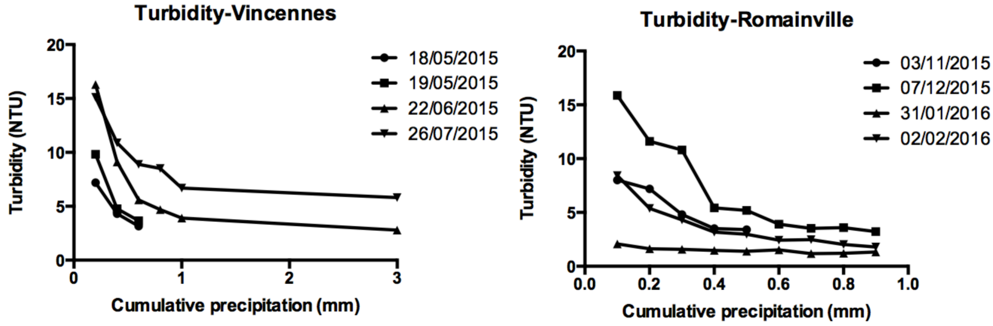
Enterococcus and Escherichia. coli. (*E.* coli). The contamination with enterococcus has showed higher level in Vincennes with the maximum value of 2900 ufc/100ml and median value of 370 ufc/100ml, while it is not detectable or only present rarely in Romainville with the median value of 10 ufc/100ml. This difference could be explained by environmental factors (more birds in Vincennes which is nearby to the Bois de Vincennes) or the seasonality (the data in Vincennes were collected in summer). Regarding another indicator of faecal contamination, only 35% of samples tested are positive for *E.coli* while 65% of samples are negative. The maximum values are 260 ufc/100ml in Vincennes and 110 ufc/100ml in Romainville. Since there are so few colonies found, the graphical representation of data is not suitable. The fact that the concentrations of enterococcus are much higher than *E.coli* can prove that enterococcus has better survival ability in rainwater than *E.coli*.
Heterotrophic Plate Count (HPC) 22°C and 37°C. The HPC results are reported as log ufc per 1ml of sample. The values of HPC at 22°C vary from 2.7 log ufc/ml to 5.2 log ufc/ml with a median value of 4 log ufc/ml in Vincennes and from 2.6 log ufc/ml to 4.4 log ufc/ml with a median value of 1 log ufc/ml in Romainville. For the HPC at 36°C, le values ranges between 2.3 log ufc/ml and 5.0 log ufc/ml in Vincennes and between 1.5 to 4.2 log ufc/ml. The median values are 2.3 log ufc/ml in Vincennes and 0 log ufc/ml in Romainville. For most of the results, the numbers of HPC are greater at 22° than at 37°C. So the lower temperature could be more suitable for the growth of HPC, which has the same conclusion with Martin et *al*. (2010).

2.2 Correlation analysis

Table 3 shows the correlation analyses that we carried out to detect the possible relation between the physic-chemical parameters and the microbiological parameters. Except that pH is correlated negatively with turbidity, TSS and *E*.coli. Other parameters are positively correlated with each other.
The pH seems related to conductivity (ρ=0.41) and not significantly correlate with all the microbiological parameters, with ρ = 0.11, -0.09, 0.08 and 0.22 for enterococcus, E.coli, HPC at 22°C and 36°C. The conductivity exhibits strongly correlation with DOC with a Spearman coefficient equal to 0.86. The presence of Turbidity and TSS are significantly correlated with each other. The Spearman coefficient is equal to 0.92 with a corresponding regression coefficient of 0.75. Refers to DOC, the correlations with the microbiological parameters are relatively significant. The Pearson's coefficient are 0.65 for enterococcus, 0.60 for HPC at 22°C and 0.63 for HPC at 36°C.
The correlation between the two indicators of faecal contamination, *E*.coli and enterococcus is not significant. The Spearman's correlation coefficient is equal to 0.06. This result is much different in the other literatures who have higher correlations between them (Vialle et al. 2011, Whitman et al. 2003). This outcome can be interpreted by the low detection of *E*.coli both in Vincennes and Romainville.

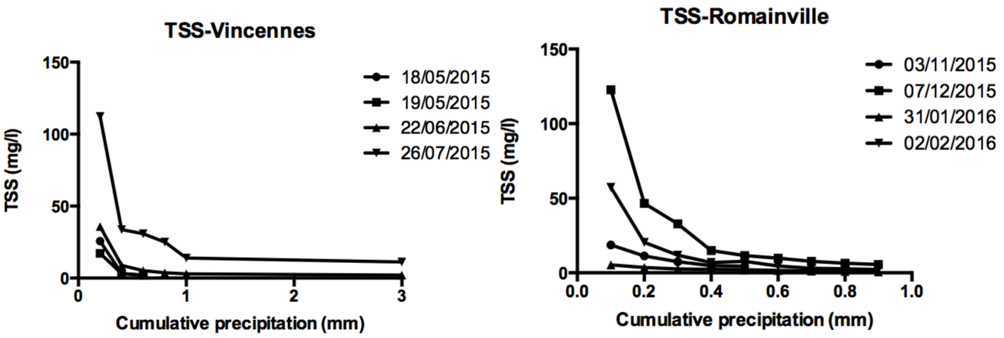
2.3 Changes et distribution

By analyzing the evolution with time of the parameters for each rainfall events, the values of pH, conductivity not always vary regularly, thus the phenomenon of first-flush was not observed for these two parameters. Likewise, for the microbiological parameters, none of the results exhibits regular decay trends. For some of the storm events, the values in the last bottles were higher than others. No observation could prove the existence of first flush effect.
On the other hand, turbidity and TSS show a clear decline for all of the storm events as expected. In Vincennes, the turbidity values can decrease 55 % on average by removing the first 1mm rainfall and decrease 62% by removing 3 mm (Figure 3-a). Regarding the TSS, the first 1mm rainfall can take away 83% of suspended solids and 88% for 3 mm (Figure 3-a). In Romainville, turbidity decreases 64% for the first 0.8mm rainfall (Figure 3-b), while TSS decreases 83% (Figure 4-b).



(a) Collection from Vincennes (b) Collection from Romainville

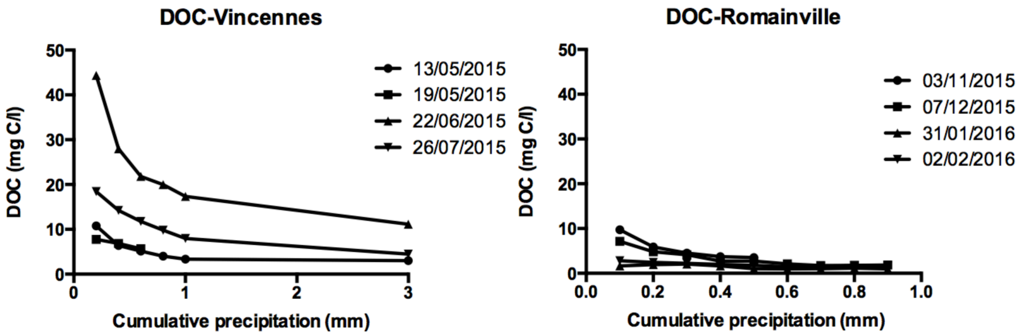
Figure 3. Turbidity profiles in Vincennes and Romainville



(a) Collection from Vincennes (b) Collection from Romainville

Figure 4. TSS profiles in Vincennes and Romainville

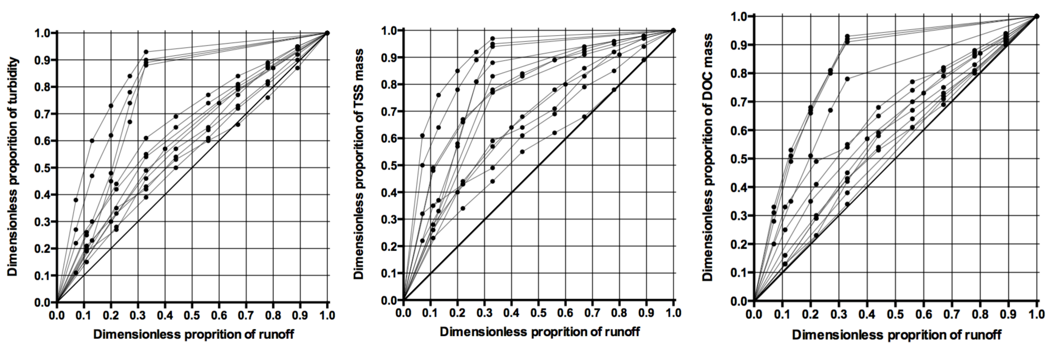
The variations of DOC also show a clear decline both in Vincennes and Romainville (Figure 5). Therefore removing the first portion can reduce the quantity of DOC.



(a) Collection from Vincennes (b) Collection from Romainville

Figure 5. DOC profiles in Vincennes in Romainville

In order to qualitatively characterize the pollutant reduction for all the storm events, the curve of cumulative proportion of mass vs. proportion runoff volume were bring out for turbidity, TSS and DOC. These three parameters have the potential to definer the first flush in the roof runoff system. Equivalency is indicated by the first bisector, which means the concentration of the pollutant remained constant throughout the runoff.



(a) Curve for Turbidity (b) curve for TSS (c) curve for DOC

Figure 6. Curves of cumulative proportion of mass vs. cumulative proportion of runoff volume for 13 storm events collected from May 2015 to February 2016

The Figure 6 presents that almost every curves are above the first bisector, which means that the first portion of runoff contain the more portion of pollutant. For example, for some of the curve of TSS (Figure 6) 30% volume of samples contains 80% of TSS. This result can prove the existence of the first flush phenomena in the roof runoff water harvesting system.

3 CONCLUSION

During 10 month (from May 2015 to February 2016), 13 series of rainwater, which consist 89 samples, were collected and analyzed. This work presents results of roof runoff quality by measuring both physicochemical parameters and microbiological parameters. The results obtained, based on a first limited data set, can be summarized as follows:

1. The values of the pH and conductivity do not always vary regularly which means that the first flush phenomena cannot be described by these two parameters. Removing the first amount of the runoff can not ensure the quality improvement;
2. The turbidity, the TSS and the DOC show a clear decline for nearly all of the storm events. The changes of the results can clearly show that the quality tends to improve after the first flush. We could use these two parameters to define quantitatively the first flush phenomena and establish reliable models for the roof runoff properties. The affects of the rainfall intensity and antecedent period of dry days need a further detailed study.
3. None of the microbiological parameters shows the first flush phenomena. And some values in the last samples were higher than the others. Some of the values were so high to meet the water standard (even the treated water standard). This conclusion was matched with some other studies that the high levels bacterial contaminations make the poor quality in the rainwater collect system. The level of values is different from both sites, meaning that the environment of the roof or the season of experiments could be determining factors of influence.

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HPC 22°C	0.081	*0.407*	*0.552*	*0.479*	*0.597*	0.519	*0.235*	
HPC 36°C	0.218	*0.474*	*0.526*	*0.448*	*0.634*	*0.636*	*0.268*	*0.850*

pH. The pH of the harvested rainwater from all of the samples collected in Vincennes varies between 6.76 and 9.53, while it varies between 6.68 and 9.67 in Romainville. The average values are respectively 7.60 and 7.90. By comparison, the literatures have reported the following pH ranges for runoff water: 5.8-8.4 (Schriewer, Horn, and Helmreich 2008), 6.4-7.7(Doyle 2008), 5.6-10.4(Vialle et al. 2011).

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2.1.2 Microbiological parameters

Enterococcus and Escherichia. coli. (*E.* coli). The contamination with enterococcus has showed higher level in Vincennes with the maximum value of 2900 ufc/100ml and median value of 370 ufc/100ml, while it is not detectable or only present rarely in Romainville with the median value of 10 ufc/100ml. This difference could be explained by environmental factors (more birds in Vincennes which is nearby to the Bois de Vincennes) or the seasonality (the data in Vincennes were collected in summer). Regarding another indicator of faecal contamination, only 35% of samples tested are positive for *E.coli* while 65% of samples are negative. The maximum values are 260 ufc/100ml in Vincennes and 110 ufc/100ml in Romainville. Since there are so few colonies found, the graphical representation of data is not suitable. The fact that the concentrations of enterococcus are much higher than *E.coli* can prove that enterococcus has better survival ability in rainwater than *E.coli*.

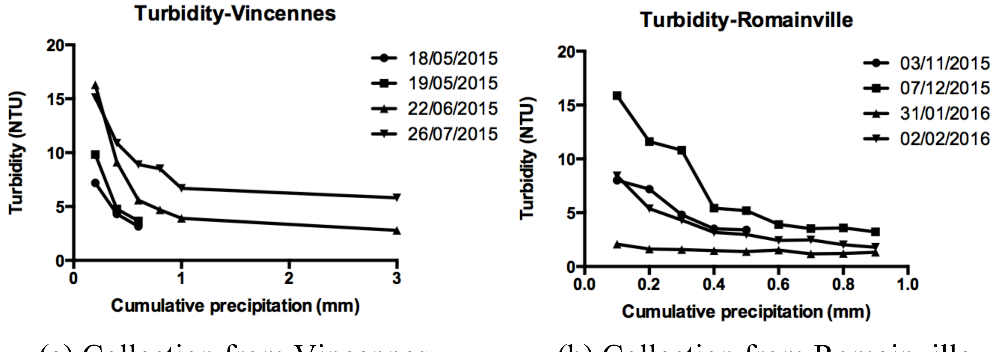
Heterotrophic Plate Count (HPC) 22°C and 37°C. The HPC results are reported as log ufc per 1ml of sample. The values of HPC at 22°C vary from 2.7 log ufc/ml to 5.2 log ufc/ml with a median value of 4 log ufc/ml in Vincennes and from 2.6 log ufc/ml to 4.4 log ufc/ml with a median value of 1 log ufc/ml in Romainville. For the HPC at 36°C, le values ranges between 2.3 log ufc/ml and 5.0 log ufc/ml in Vincennes and between 1.5 to 4.2 log ufc/ml. The median values are 2.3 log ufc/ml in Vincennes and 0 log ufc/ml in Romainville. For most of the results, the numbers of HPC are greater at 22° than at 37°C. So the lower temperature could be more suitable for the growth of HPC, which has the same conclusion with Martin et *al*. (2010).

2.2 Correlation analysis

Table 3 shows the correlation analyses that we carried out to detect the possible relation between the physic-chemical parameters and the microbiological parameters. Except that pH is correlated negatively with turbidity, TSS and *E*.coli. Other parameters are positively correlated with each other.
The pH seems related to conductivity (ρ=0.41) and not significantly correlate with all the microbiological parameters, with ρ = 0.11, -0.09, 0.08 and 0.22 for enterococcus, E.coli, HPC at 22°C and 36°C. The conductivity exhibits strongly correlation with DOC with a Spearman coefficient equal to 0.86. The presence of Turbidity and TSS are significantly correlated with each other. The Spearman coefficient is equal to 0.92 with a corresponding regression coefficient of 0.75. Refers to DOC, the correlations with the microbiological parameters are relatively significant. The Pearson's coefficient are 0.65 for enterococcus, 0.60 for HPC at 22°C and 0.63 for HPC at 36°C.
The correlation between the two indicators of faecal contamination, *E*.coli and enterococcus is not significant. The Spearman's correlation coefficient is equal to 0.06. This result is much different in the other literatures who have higher correlations between them (Vialle et al. 2011, Whitman et al. 2003). This outcome can be interpreted by the low detection of *E*.coli both in Vincennes and Romainville.

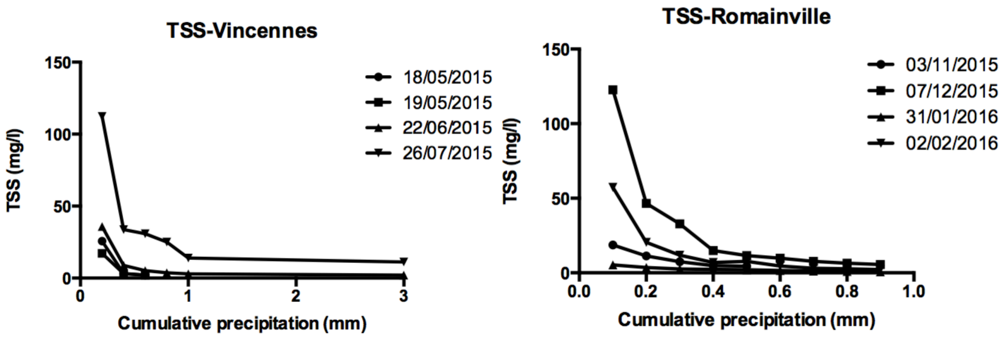
2.3 Changes et distribution

By analyzing the evolution with time of the parameters for each rainfall events, the values of pH, conductivity not always vary regularly, thus the phenomenon of first-flush was not observed for these two parameters. Likewise, for the microbiological parameters, none of the results exhibits regular decay trends. For some of the storm events, the values in the last bottles were higher than others. No observation could prove the existence of first flush effect.
On the other hand, turbidity and TSS show a clear decline for all of the storm events as expected. In Vincennes, the turbidity values can decrease 55 % on average by removing the first 1mm rainfall and decrease 62% by removing 3 mm (Figure 3-a). Regarding the TSS, the first 1mm rainfall can take away 83% of suspended solids and 88% for 3 mm (Figure 3-a). In Romainville, turbidity decreases 64% for the first 0.8mm rainfall (Figure 3-b), while TSS decreases 83% (Figure 4-b).



(a) Collection from Vincennes (b) Collection from Romainville

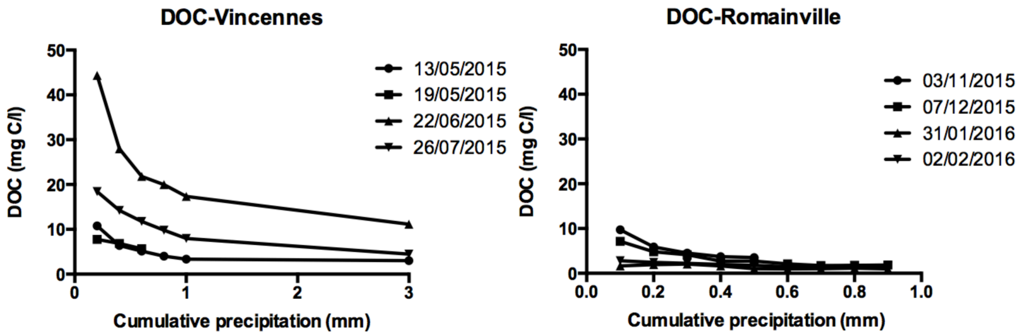
Figure 3. Turbidity profiles in Vincennes and Romainville



(a) Collection from Vincennes (b) Collection from Romainville

Figure 4. TSS profiles in Vincennes and Romainville

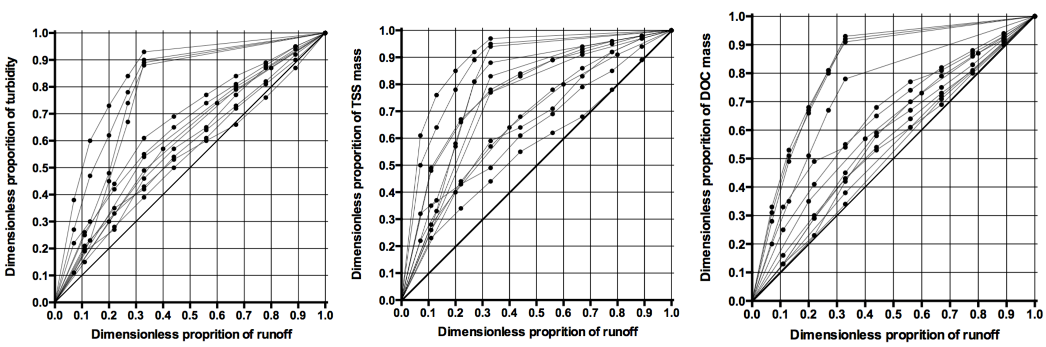
The variations of DOC also show a clear decline both in Vincennes and Romainville (Figure 5). Therefore removing the first portion can reduce the quantity of DOC.



(a) Collection from Vincennes (b) Collection from Romainville

Figure 5. DOC profiles in Vincennes in Romainville

In order to qualitatively characterize the pollutant reduction for all the storm events, the curve of cumulative proportion of mass vs. proportion runoff volume were bring out for turbidity, TSS and DOC. These three parameters have the potential to definer the first flush in the roof runoff system. Equivalency is indicated by the first bisector, which means the concentration of the pollutant remained constant throughout the runoff.



(a) Curve for Turbidity (b) curve for TSS (c) curve for DOC

Figure 6. Curves of cumulative proportion of mass vs. cumulative proportion of runoff volume for 13 storm events collected from May 2015 to February 2016

The Figure 6 presents that almost every curves are above the first bisector, which means that the first portion of runoff contain the more portion of pollutant. For example, for some of the curve of TSS (Figure 6) 30% volume of samples contains 80% of TSS. This result can prove the existence of the first flush phenomena in the roof runoff water harvesting system.

3 CONCLUSION

During 10 month (from May 2015 to February 2016), 13 series of rainwater, which consist 89 samples, were collected and analyzed. This work presents results of roof runoff quality by measuring both physicochemical parameters and microbiological parameters. The results obtained, based on a first limited data set, can be summarized as follows:

1. The values of the pH and conductivity do not always vary regularly which means that the first flush phenomena cannot be described by these two parameters. Removing the first amount of the runoff can not ensure the quality improvement;
2. The turbidity, the TSS and the DOC show a clear decline for nearly all of the storm events. The changes of the results can clearly show that the quality tends to improve after the first flush. We could use these two parameters to define quantitatively the first flush phenomena and establish reliable models for the roof runoff properties. The affects of the rainfall intensity and antecedent period of dry days need a further detailed study.
3. None of the microbiological parameters shows the first flush phenomena. And some values in the last samples were higher than the others. Some of the values were so high to meet the water standard (even the treated water standard). This conclusion was matched with some other studies that the high levels bacterial contaminations make the poor quality in the rainwater collect system. The level of values is different from both sites, meaning that the environment of the roof or the season of experiments could be determining factors of influence.

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