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MONITORING OF STORMWATER BETWEEN 2002 AND 2010 – WHAT IS THE EVOLUTION OF STORMWATER QUALITY?

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ABSTRACT

The city of Toulouse with its separate storm sewer system is ideal for studying stormwater. That is why since 2002, three stormwater sampling campaigns were conducted. Samples were taken from the outlets of two storm drains located in heavily and moderately urbanized areas. Sampling was undertaken during wet weather and dry weather during the year 2002 for the first campaign, during the year 2007 for the second one and during the year 2010 for the last one. The overall pollution parameters were analyzed (chemical oxygen demand, biological oxygen demand, total nitrogen, ammonium, nitrate, total phosphorus, suspended solid matter, volatile suspended matter, pH, conductivity, turbidity) and the presence of some micropollutants was studied (6 PAHs, MTBE and total hydrocarbons). Results showed an evolution of stormwater quality between the three campaigns and an improvement of nutrient and suspended solid concentrations. They indicated also that dry weather had an impact on annual pollution load from separate storm sewer and that level of urbanization was also a factor whatever the year of sampling.

KEYWORDS:
overall pollution parameters, principal component analysis, separate sewer system, storm sewer discharge

INTRODUCTION

Municipalities have for long considered stormwater as a purely hydraulic phenomenon. However, it is now recognized as a pollution source for the receiving environment [1]. At present, stormwater may be collected in the same system as wastewater, i.e. combined sewer system, or else in an independent network, i.e. separate sewer system. It is important to study stormwater in the latter because it is discharged directly into the natural environment. Pollution of stormwater can be from different sources: rainwater quality, urban runoff from roofs/roads, illicit connections, illegal dumping, discharges from authorized companies and leaks from the wastewater sewer [2].

Worldwide, numerous studies have already been made on stormwater characterization especially concerning overall pollution parameters ([3] – [7]), but during these studies sampling was carried out for a limited time. For example, Zgheib et al. [7] collected samples during two months and Lee and Bang [3] sampled stormwater during two years. No evolution of stormwater quality during several years has already been published.

The present paper presents the results obtained during three campaigns occurred in 2002, 2007 and 2010 in the separate sewer system of city of Toulouse in France and the evolution of stormwater quality during these years. The overall pollution parameters were characterized to determine the pollution of water discharges from the two main storm drain outlets in Toulouse and some micropollutants were studied. The impacts of dry weather discharges were evaluated because some companies, such as car wash stations, are authorized to discharge their wastewater into the storm sewer after a pre-treatment, resulting in a constant flow here even in dry weather without rain. Respect of the French 1998 Act [8] limit values was checked and trace organic compound pollution levels were compared to requirements under the Water Framework Directive [9] because city's stormwater is discharged directly into the River Garonne.

MATERIALS AND METHODS

Studied sites. Two major outlets in the Toulouse storm sewer have been selected. The first, the Boulevards outlet, drains runoff water from the heart of downtown. Its watershed extends over an area of 439 hectares and represents a heavily urbanized zone.

The second, the Mirail outlet, drains runoff water from a residential zone composed of houses and blocks of flats with gardens. Its watershed extends over an area of 1428 hectares.

Sampling methods. For 2002 campaign, samplings were carried out from February 2002 to March 2003. Twenty and nineteen wet weather samples were collected for Boulevards and Mirail outlets respectively and four dry weather samples were collected per outlet. Dry weather samples were collected over 24 hours and wet weather samples were collected over a variable time from 2 to 12 hours. For 2007 campaign, samplings were carried out from February 2007 to March 2007. Two wet weather samples were collected per season and per outlet, eight samples in total per outlet. Two dry weather samples were collected per outlet. For 2010 campaign, samplings were carried out from January 2010 to February 2011. Two wet weather samples and three dry weather samples were collected per season and per outlet, eight wet weather samples and twelve dry weather samples in total. For these two last sampling campaigns, the water was collected over 24 hours for each sample.

A time based autosampler, ISCO 3700, was used to collect these representative average samples. The samples were then transferred into amber glass bottles and stored at -25°C for subsequent analysis. Flow was measured throughout the sampling.

Analyses. Initially, classical parameters were measured for the raw samples: pH, turbidity, conductivity. Commercially available tests (Spectroquant®, Merck) were used to determine the chemical oxygen demand (COD), total nitrogen (TN) and total phosphorus (TP) in the raw samples, as well as for ammonium ion (NH_4^+) and nitrate (NO_3^-) ion analysis in filtered samples. Suspended solid matter (SSM) and volatile suspended matter (VSM) were assessed by filtration according to NF-T90-105-1 and NF-T90-029 standards respectively.

Characterization was completed by analysis of six polycyclic aromatic hydrocarbons: fluoranthene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd) pyrene and benzo(g,h,i)perylene, of methyl *tert*-butyl-ether and of total hydrocarbons. The analytical methods used to quantify these compounds evolved over the years, therefore they are not detailed here but they followed European standards and were validated.

Multivariate analyses. Principal component analysis (PCA) was carried out in order to visualize the correlations between the variables studied (= overall pollution parameters) and to identify homogenous groups of observations or conversely, atypical ones. PCA was performed using the commercial software XLstat (Addinsoft, Paris, France).

RESULTS AND DISCUSSION

Annual concentrations. (1) Overall pollution parameters. Box plots for each campaign and each outlet were drawn to illustrate dispersion of data, and are shown in Figure 1 representing also average-median, minimum-maximum and extreme values. An LOQ / 2 value was applied for calculations when values were below the LOQ [10].

- **Nutrients and COD.** Comparing the three campaigns, COD, TP and TN concentrations were lower for the 2010 campaign. An improvement of stormwater nutrient concentrations appeared for this campaign. Furthermore, 2010 campaign had no sample with concentration above threshold value of French 1998 Water Act for COD, TP and TN unlike other campaigns. Indeed, for COD, 2002 campaign had six samples for Boulevards outlet with concentration above 125 mg O_2/L and 2007 campaign had two samples for Boulevards outlet and one for Mirail outlet. The maximum values in 2002 were 360 mg O_2/L for Boulevards outlet and 123 mg O_2/L for Mirail outlet. In 2007, they were 156 mg O_2/L and 146 mg O_2/L for Boulevards and Mirail outlets respectively. While, in 2010, the maximum values were 40 mg O_2/L and 41 mg O_2/L for Boulevards and Mirail outlets respectively. In 2002 and in 2007, Boulevards outlets had COD concentrations greater than Mirail outlet, it can be explained by its more urbanized watershed. Indeed, waters of downtown of Toulouse are discharged in Boulevards watershed.

Median values obtained for the three campaign are lower than COD concentrations found by Zgheib et al. [7] but 2002 and 2007 campaigns presented greater COD concentrations than Bressy et al. [11].

For TP, only 2002 campaign presented values greater than the threshold value of 2 mg/L, it was the case for four samples for Boulevards outlet (maximum value: 3.6 mg/L) and one sample for Mirail outlet (maximum value: 2.5 mg/L). Like COD concentrations, TP concentrations are higher for Boulevards outlet than for Mirail outlet.

For total nitrogen, two samples in 2002 presented values greater than 30 mg/L, the threshold value, one for Boulevards outlet (32 mg/L) and one for Mirail outlet (40 mg/L). In 2007, four values were greater than 30 mg/L, two samples for each outlet, for Boulevards outlet the maximum value was 54 mg/L and for Mirail outlet, it was 38 mg/L. Comparing the two outlets, TN concentrations were quite similar for each campaign.

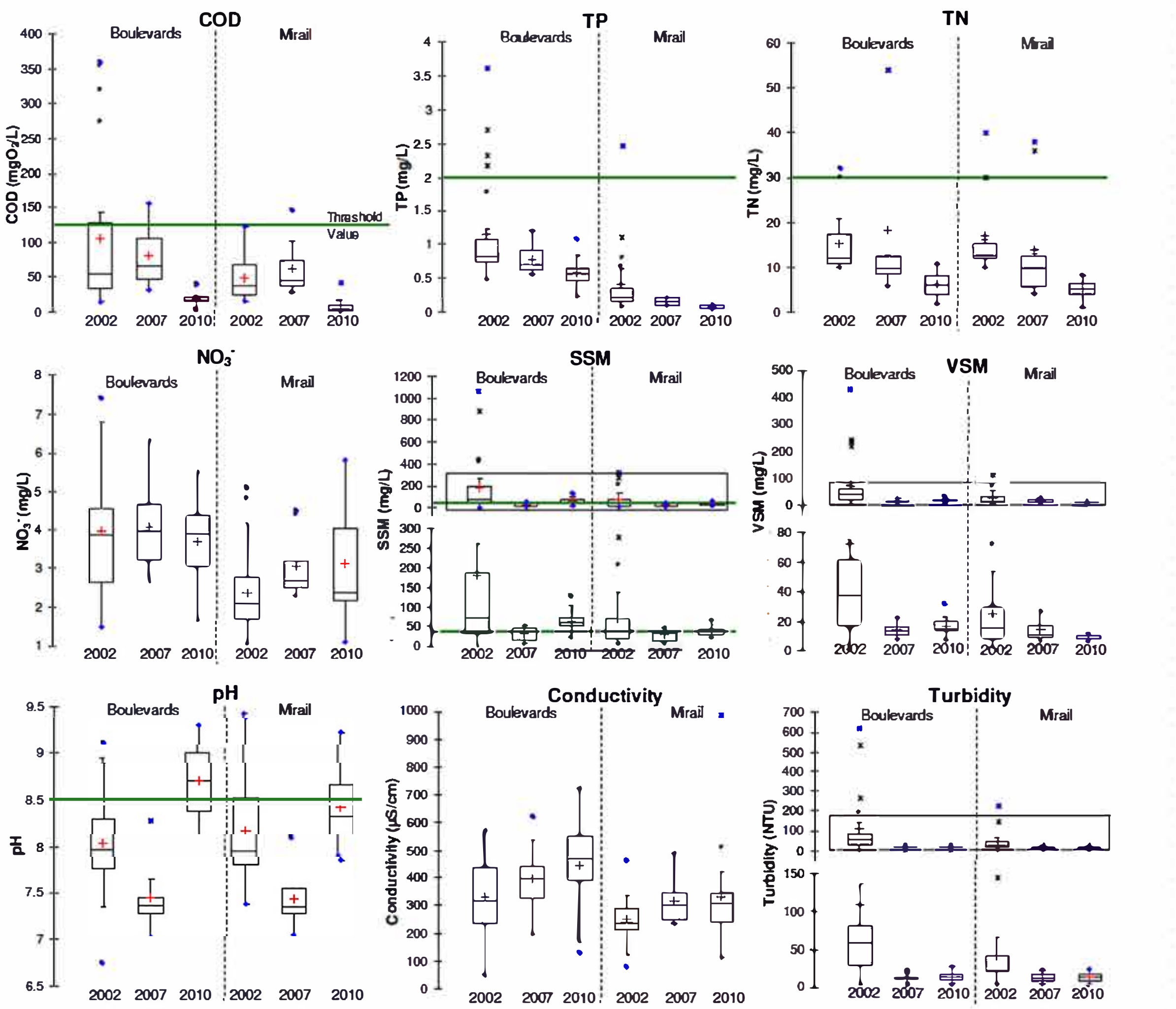

FIGURE 1

Box and whisker plots representing data distribution for overall pollution parameters in each outlet for the three campaigns –Threshold values of French 1998 Water Act [8] are shown by a line – A zoom on SSM, VSM and turbidity results is represented

Total phosphorus concentrations are very low comparing to results obtained by Zgheib et al. [7] and Terzakis et al. [5], but TN concentrations obtained in 2002 and 2007 are greater than the values of the same studies.

The decrease of COD, TN and TP between 2002 and 2010 can be explained by an enhanced performance of storm sewer policy and therefore a decrease of illicit connection and a reduction of leaks from the wastewater sewer and also by the end of use of detergent with phosphorus to clean pavements and by a better behavior of persons to clean animal excrement.

- **Suspended solids and turbidity.** Many samples for the three campaigns had SSM concentration higher than threshold value. Indeed, for 2002 campaign, sixteen and twelve samples had concentrations greater than 35 mg/L for Boulevards and Mirail outlets respectively. In 2007, five samples for each outlet presented also concentrations above the limit. And in 2010, 90% of samples for Boulevards outlet and 50% for Mirail outlet presented values greater than the threshold value. This raises questions as to the necessity for treating storm sewer discharge to reduce the quantity of SSM discharge into the natural environment. Some treatment systems can easily reduce the quantity of SSM and permit to have concentrations below the limit. Indeed, Borne et al. [12] obtained a mass removal efficiency for suspended solids of almost 60% with a stormwater treatment pond containing a floating treatment wetland and Fournel et al. [13] with constructed reed beds obtained a SSM removal greater than 83% on simulated combined sewer effluent. This reduction of SSM in stormwater will also permit to reduce the concentration of COD because almost 50% of COD are due to the particulate phase in stormwater as determined by Sablayrolles et al. in an internal study made in 2009 [14]. Bressy et al. [11] found also a median COD proportion in particulate phase equal to 47%.

As illustrated in Figure 1, the campaign of 2002 showed SSM and VSM concentrations and turbidity values more important than the other campaigns. For SSM, the median values were 72.3 mg/L and 35.5 mg/L for Boulevards and Mirail outlets respectively in 2002, 35.8 and 32 mg/L in 2007 and 58.9 and 35.1 mg/L in 2010.

For VSM, the median values were 37.2 mg/L and 14.8 mg/L for Boulevards and Mirail outlets respectively in 2002, 13.2 and 10.5 mg/L in 2007 and 14 and 8.1 mg/L in 2010.

In 2002 and in 2010, SSM and VSM concentrations were higher in Boulevards outlet than in Mirail outlet, as COD, this difference can be explained by the more urbanized watershed of Boulevards outlet. Indeed, a more urbanized watershed significates also more car circulation and therefore a more important particles release into the air or on the road than in a more residential watershed as Mirail outlet. In 2007, the difference between the two outlets was negligible.

For turbidity, the median values were 58.3 NTU and 22.6 NTU for Boulevards and Mirail outlets respectively in 2002, 11.8 and 11.3 NTU in 2007 and 13.5 and 13 NTU in 2010. In 2007 and in 2010, the difference for turbidity values between the two outlets was negligible, whereas in 2010, the difference for SSM concentration was significative, therefore, we can suppose than the proportion of colloidal phase is different between the two outlet and it can be greater in Mirail outlet.

- **Ions, conductivity and pH.** Nitrates concentrations were quite similar between the three campaigns for the two outlets. Median values for 2002 campaign were 3.9 mg/L and 2.1 mg/L for Boulevards and Mirail outlets respectively, 4 and 2.7 mg/L in 2007 and 3.9 and 2.4 mg/L in 2010.

Box plots for ammonium ions are not presented in Figure 1 because many values were below limit of quantification. Except for Boulevards outlet in 2010, all others campaigns and outlets had an ammonium median values less than limit of quantification (LOQ=0.8 mg N/L in 2002 and 0.1 mg N/L in 2007). Ammonium median value obtained in 2010 for Boulevards outlet is 1.55 mg N/L.

Conductivity values were greater for 2010 campaign for Boulevards outlet. For Mirail outlet, 2007 and 2010 campaigns presented almost the same values. Median values for 2002 campaign were 317 µS/cm and 237 µS/cm for Boulevards and Mirail outlets respectively, 398 and 303 µS/cm in 2007 and 470 and 309 µS/cm in 2010.

pH values were greater in 2010 than during the years 2002 and 2007. 2007 campaign presented the best results because no sample had value above the upper limit: 8.5. In 2002, three and six samples had pH values above 8.5 for Boulevards and Mirail outlets respectively, and in 2010, fourteen (70% of total samples) and seven samples were above this limit. The maximum pH value obtained in 2002 was 9.43 for Mirail outlet and 9.34 in 2010 for Boulevards outlet. These values are greater than maximum values found by Terzakis et al. in 2008 (8.31) [5], and Zgheib et al. in 2011 (7.35) [7]. These basic pH values can be explained by several roadworks led in the center of Toulouse in 2010 and maybe, fresh concrete use increased pH values. These values were not only caused by rainwater where pH values were between 4.1 and 8.7 in 2009 in Toulouse [14], median value equal to 5.8.

White and Cousins [15] developped a floating treatment wetland where a reduction of 30% of pH values was observed. This treatment system could be sufficient to reduce pH values below the legislative limit.

(2) Principal component analysis. Principal component analysis (PCA) was conducted for datas of each outlet from the three campaigns for COD, TP, NO_3^-, SSM, VSM, pH, conductivity and turbidity. The others parameters were not taken into account because of many results less than LOQ and for this reason, the software did not permit us to make PCA for all the parameters.

Figure 2 presents two correlation circles with the first two principal components for each outlet and each line in circles corresponds to the vector of each illustrated parameter. This representation permits to underscore easily correlations between parameters because co-linear variables are significantly correlated.

The first two principal components aggregate 71% of the variance for Boulevards outlet and 67% for Mirail outlet. The F1 component seems to represent the organic matter (COD, SSM, VSM, turbidity) and the F2 axe can be taken as the ionic component (conductivity and nitrate ions) for the two outlets.

Using these circle interpretations and linear coefficients (R^2), four significant correlations were found (at an α=0.05 level) for Boulevards outlet: TP-SSM (R^2=0.553); COD-SSM (R^2=0.525) and two strong correlations between SSM and Turbidity (R^2=0.921) and between TP and COD (R^2=0.750). The correlation between COD and SSM can be explained by an important part of COD due to particulate phase and especially to suspended matter. No correlation between COD and turbidity was found, therefore colloidal matter had no significant impact on COD. The important correlation between turbidity and SSM is normal because turbidity takes into account SSM and colloidal matter. The correlation between total phosphorus and COD can be explained by any leakage from the wastewater sewer or by any illicit connection in 2002 for highest values and also by a better use of rubbish by persons

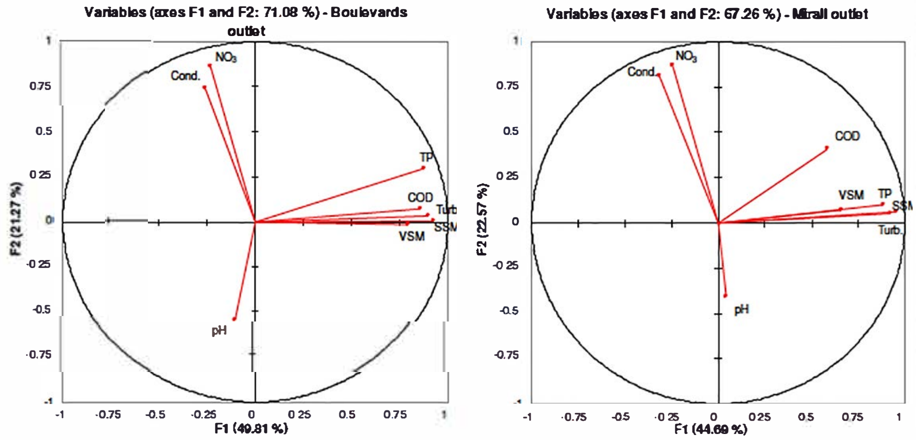

FIGURE 2
Correlation circles for the F1-F2 and F1-F3 axes constructed from PCA of all the datas from the three campaigns

for their waste and for their animal excrement in 2010 with lowest values.

For Mirail outlet, correlations were different. But the correlation between SSM and turbidity is still present (R^2=0.860). For this outlet, total phosphorus is also correlated with turbidity (R^2=0.828) and SSM (R^2=0.693), this can be explained by a possible presence of phosphorus on particulate matter. A last correlation between conductivity and NO_3^- was observed (R^2=0.516). Indeed, conductivity takes into account ions present in water and it is possible the concentration of nitrates represents an important part of ions present in the stormwater and it significantly influences conductivity values.

Urbanization of watershed can be a reason of different correlations between outlets.

These correlations were different than correlations found by Deffontis et al. [16] for the 2010 campaign, so the correlations between the overall pollution parameters are different considering a campaign during a year and different campaigns conducted during three different years. The quantity of datas can also explain this difference.

Another PCA illustration permits to see each sample on the new F1, F2 and F3 axes (Figure 3). Each coloured point represents one collected sample during each campaign. This PCA confirms a significant difference between the 2007 and 2010 campaigns because they are grouped in two distinct clusters as illustrated in the figure whereas 2002 campaign presents a wide dispersion of datas.

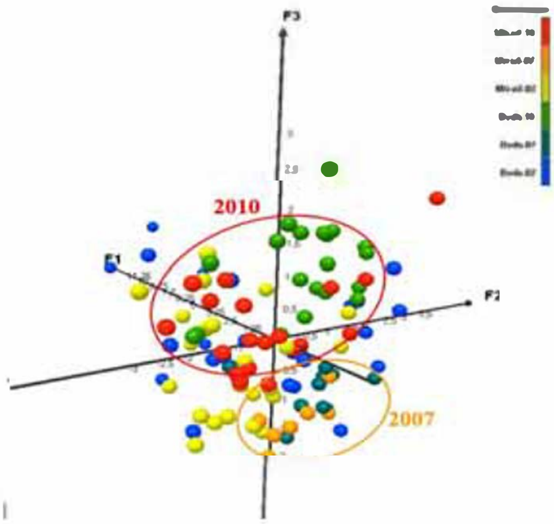

FIGURE 3
Correlation circles for the F1-F2 and F1-F3 axes constructed from PCA of all the datas from the three campaigns

TABLE 1
Median concentrations and median annual loads for PAH, MTBE and total hydrocarbons for Boulevards and Mirail outlet for 2002, 2007 and 2010 campaign and for dry and wet weather
The occurrence of results above LOQ is indicated into parenthesis

			Wet weather			Dry weather		
		Units	2002-03	2007	2010	2002-03	2007	2010
Boulevards outlet								
∑ PAH (6)	Concentration	µg/L	0.072 (75%)	<0.01 (0%)	<0.02 (0%)	<0.04 (0%)	<0.01 (0%)	<0.02 (0%)
	Load	kg/an	0.06	ND	ND	0.04	ND	ND
MTBE	Concentration	µg/L	0.25 (7%)	<10 (0%)	<0.5 (0%)	0.68 (50%)	<10 (0%)	<0.5 (0%)
	Load	kg/an	0.32	ND	ND	1.18	ND	ND
TH	Concentration	mg/L	0.20 (75%)	0.05 (13%)	0.03 (13%)	0.06 (50%)	<0.1 (0%)	0.03 (8%)
	Load	kg/an	140.7	45.9	30.8	112.4	ND	62.6
Mirail outlet								
∑ PAH (6)	Concentration	µg/L	0.039 (64%)	0.005 (13%)	<0.02 (0%)	0.02 (25%)	<0.01 (0%)	<0.02 (0%)
	Load	kg/an	0.08	0.001	ND	0.09	ND	ND
MTBE	Concentration	µg/L	<0.5 (0%)	<10 (0%)	<0.5 (0%)	<0.5 (0%)	17.5 (50%)	<0.5 (0%)
	Load	kg/an	ND	ND	ND	ND	5.6	ND
TH	Concentration	mg/L	0.10 (53%)	<0.1 (0%)	0.03 (13%)	<0.05 (0%)	<0.1 (0%)	<0.05 (0%)
	Load	kg/an	117.3	ND	45.8	ND	ND	ND

(3) Trace organics compounds. The median values of all samples were calculated for each type of sampling weather, for each outlet studied and for each campaign. For values below the LOQ, LOQ / 2 was used in calculations [10]. Results are shown in Table 1.

Concentrations of trace organic pollutants were very low for each campaign as indicated by the low occurrence of results quantified in samples. Indeed, in literature, Terzakis et al. [5] obtained an average value for six PAHs equal to 0.5 µg/L in runoff water and Rianawati and Balasubramanian [17] obtained an average value for sixteen PAHs of 1.1 µg/L. The concentration of studied PAHs are below NQE of Water Framework Directive [18], therefore, the quality of stormwater in Toulouse can be considered as good considering these micropollutants.

For total hydrocarbons, a threshold value was defined in French 1998 Water Act [8] as 10 mg/L. No sample presented a concentration above this limit. The maximum value for total hydrocarbons was obtained in 2002 for Boulevards outlet and was equal to 4.6 mg/L.

Dry weather loads and impact. (1) Overall pollution parameters. The pollution generated by each outlet is due to the pollutant concentrations during samplings but also to the flow, because the higher the latter the greater the quantity of pollutants that will be carried along. Thus, to make a significant comparison between the campaign, the outlets and the sampling weather for this pollution, it is better to look at pollutant loads. These calculations were made as explained by Deffontis et al. [16]. For values below the LOQ, LOQ / 2 was used in calculations.

To calculate the dry weather pollutant load, the annual rainfall and the dry weather duration needed to be known. In 2002, the annual precipitation was 706 mm for the Toulouse region. During the year 2007, 589 mm of rain fell and during the year 2010, the precipitation was 599 mm. In 2002, dry weather lasted 252 days. During the year 2007, there were 276 days dry weather and in 2010, there were 260.

The results for each campaign and each outlet are illustrated in Figure 4 where each bar represents the total annual load and is separated in two parts: one corresponding to dry weather load and the other to wet weather load. The figure also shows the percentage impact of dry weather on the annual pollutant load.

Student or Mann-Whitney statistical tests were made to verify the significance of the difference between the two outlets and between sampling weather. When a significant difference (at the α=0.10 level) between the pollution resulting from the Boulevards outlet and from the Mirail one was determined, the year of campaign was framed in Figure 4 and when a significant difference between the sampling weather was observed, the percentage impact was underlined (Figure 4).

In general, the 2002 campaign had a higher pollution load than the other ones except for COD load in Boulevards outlet where the higher load was obtained in 2007 and for SSM where 2010 campaign had higher load for the two outlets. For TP, NH_4^+, NO_3^-, SSM and VSM, 2007 campaign has generated a less important pollution load than the other campaign. For example, for NO_3^-, the pollution load in 2007 for Mirail outlet was almost 13 times smallest than the pollution load in 2002.

An improvement of pollution generated by stormwater was therefore observed for these

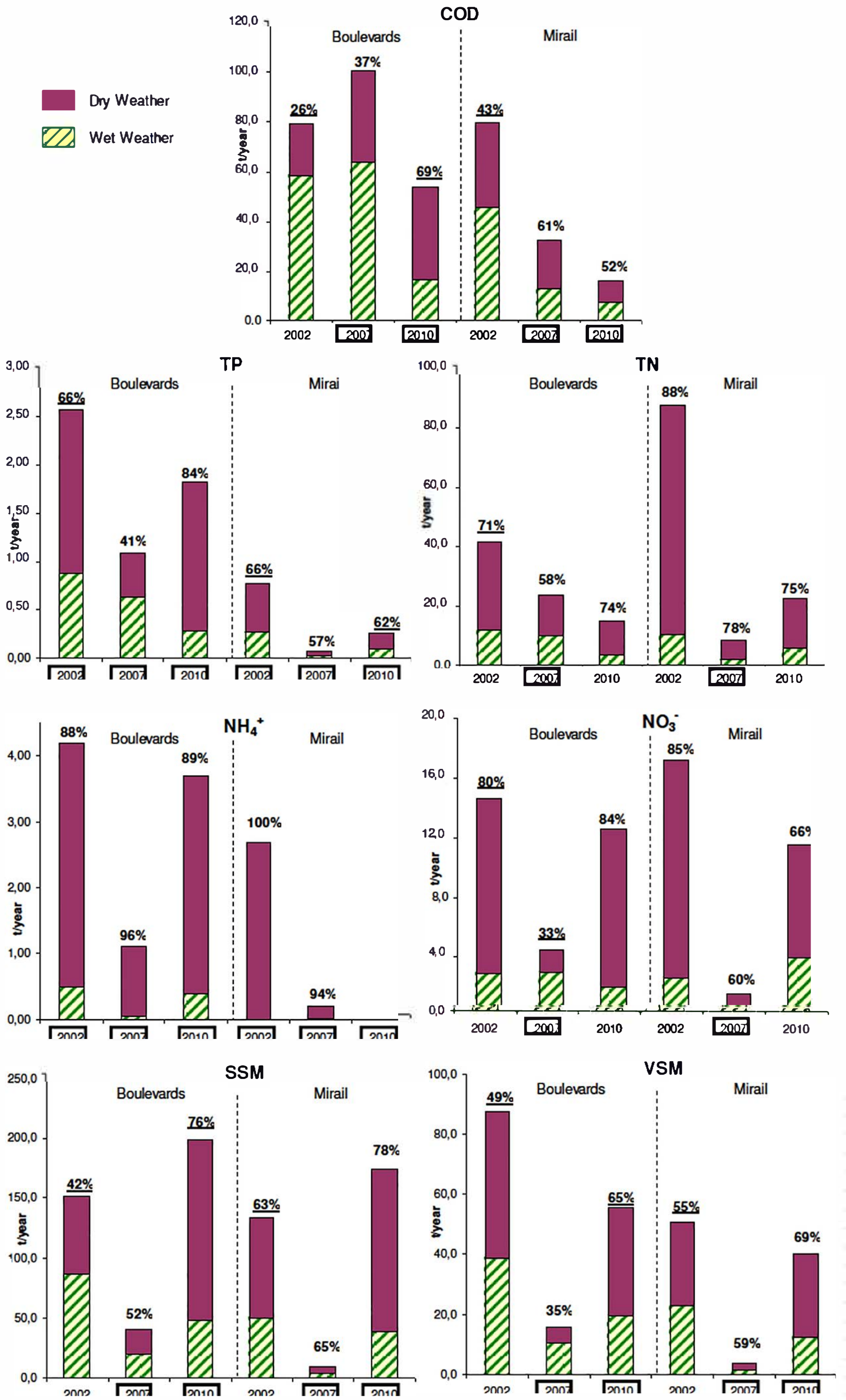

FIGURE 4

Annual dry and wet weather loads for the overall pollution parameters at the Mirail and Boulevards outlets for each campaign – The impact of dry weather discharges on the annual pollutant load are expressed as percentages

parameters but in 2010, an enhancement of this pollution was observed again although the pollution is less important than in 2002. Hypotheses were made to explain this: an increase of fertilizers use was made in 2010 because of an enhancement of "green" zone in Toulouse between 2007 and 2010. For SSM and VSM, this increase of pollution load in 2010 can be explained by the enhancement of car traffic and also by many roadworks made in 2010. The COD pollution decrease in 2010 can be explained by an enhanced performance of storm sewer policy and therefore a decrease of illicit connection and a reduction of leaks from the wastewater sewer but also by a better behavior of persons to clean animal excrement. This hypothesis can also explain the decrease of nitrogen and phosphorus loads between 2002 and 2010.

In general, for 2007 and 2010, pollution loads were greater for Boulevards outlet than for Mirail one. This can be explained by the more urbanized watershed of Boulevards outlet. In 2002, COD, TN and NO_3^- pollution loads were greater for Mirail outlet. During this campaign, the greatest pollution loads of Mirail outlet can be explained by a chemical industry explosion occurred in 2001. Indeed, the Mirail outlet watershed is closer to this industry than the Boulevards outlet one. But this difference between the two outlets was not confirmed by statistical tests. However, these tests confirm a significant difference between Mirail and Boulevards outlets in 2002 for TP and NH_4^+ pollution loads with a greater pollution for Boulevards outlet. The statistical tests confirm a significant difference (at the α=0.10 level) between the pollution resulting from the two outlets for all the parameters for 2007 campaign. In 2010, a significant difference was found for all the parameters except for TN and NO_3^-. In general, dry weather had an impact on annual pollution load because dry weather loads represents more than 50% of the annual pollution load for most of the parameters as illustrated in Figure 4 where the percentage impact of dry weather on the annual pollution load is indicated above the annual load bar. In 2002, except COD load for Boulevards outlet, all other parameter dry weather loads represent more than 50% of annual load and this difference between dry and wet weather discharge is significant for COD, TP, SSM and VSM for both outlet, for TN and NO_3^- for Boulevards outlet only. In 2007, the only significant difference was observed for NO_3^- in Boulevards outlet, this can be explained by the low number of dry weather sampling. Indeed, only two dry weather samples were sampled during this campaign for each outlet. In 2010, significant differences between the sampling weather were demonstrated for COD, SSM and VSM for Boulevards outlet and for TP for Mirail outlet, all calculated impact of dry weather are above 50%.

(2) Trace organic compounds. The annual trace organic compounds loads were calculated in the same way as for the pollution parameters. The results are shown in Table 1.

Given the low occurrence of results above the LOQ for 2007 and 2010 campaigns, no conclusion was made on annual pollution load.

In 2002, a significant difference was found for total hydrocarbons and PAHs between dry and wet weather for Boulevards outlet. For these molecules, wet weather loads represents a more important impact on annual pollution loads than dry weather loads, this impact of wet weather is equal to 60% and 56% for PAHs and TH respectively. This difference between the two weathers can be explained by road leaching during rain days.

CONCLUSION

The study of water quality at two major outlets of the Toulouse storm sewer during three campaigns of one year was completed with the aim to look at the evolution of stormwater quality. 107 events were collected: 47 samples between February 2002 and March 2003, 20 samples between February 2007 and March 2008 and 40 samples between January 2010 and February 2010 with dry and wet weather samples. Chemical oxygen demand, biological oxygen demand, total nitrogen, ammonium, nitrate, total phosphorus, suspended solid matter, volatile suspended matter, pH, conductivity, turbidity were characterized for all the 104 samples. Six PAHs, methyl *tert*-butyl-ether and total hydrocarbons were also researched.

A decrease of nutrient and suspended solid concentrations was observed over the years. The first campaign conducted in 2002 had in general the highest concentrations and the highest pollutant loads, this can be explained by illicit connection, leaks from the wastewater sewer and also by a chemical industry explosion occurred in September 2001 in Toulouse. Certain samples taken in 2002 had levels of suspended solid matter, total phosphorus, total nitrogen and pH that did not respect legislation limit values, but in 2010, only SSM and pH had values above legislation limit. Concerning trace organic compounds, concentrations found in the samples were very low. An impact of dry weather discharges and of urbanization was confirmed on annual pollutant loads whatever the year of sampling. These results, together with strengthened environmental quality standards raise serious questions as to the necessity for treating storm sewer discharge, prior to discharge into the natural environment.

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