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Combination of Lagrangian Discrete Phase Model and sediment physico-chemical characteristics for the prediction of the distribution of trace metal contamination in a stormwater detention basin

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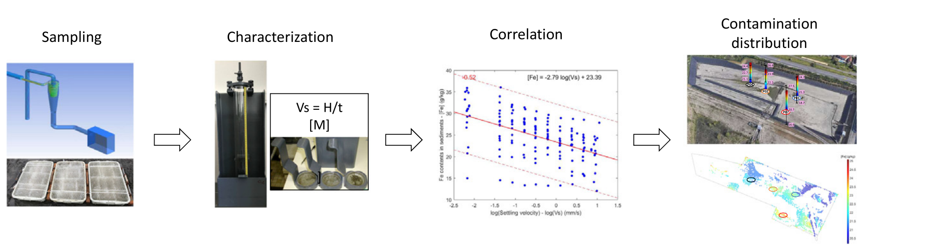
HIGHLIGHTS

- Significant correlation between metal contents and settling velocity is found.
- Fe is a good indicator of trace metal contamination in detention basins.
- The prediction of contamination distribution contributes to sediment management.

GRAPHICAL ABSTRACT



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ABSTRACT

Elevated trace metal concentrations in sediments pose a major problem for the management of stormwater detention basins. These basins provide a nature-based solution to remove particulate pollutants through settling, but the resuspension of these contaminated deposits may impact the quality of both surface and groundwater. A better understanding of trace metal distribution will help to improve basin design and sediment management. This study aims to predict the distribution of trace metal contamination in a stormwater detention basin through (i) investigation of the correlation between metal content in sediments and their settling velocity, and (ii) the coupling of such correlation with a Lagrangian Discrete Phase Model (LDPM). The correlation between Fe, Cr, Cu, Ni, Pb contents and the settling velocity is firstly investigated, based on the sediments collected from 6 sites (inlet and 5 traps at the bottom of a detention basin situated in Chassieu, France) during 5 campaigns in 2017. Results show that Fe is strongly correlated to settling velocity and can be considered as a good indicator of trace metal contents. The derived correlation is then combined with a LDPM for the prediction of trace metal distribution, producing results consistent with *in situ* measurements. The proposed methodology can be applied for other stormwater basins (dry or wet). As described in this article, the interactions between hydrodynamics and sediment physico-chemical characteristics is crucial for the design and management of stormwater detention basins, allowing managers to target the highest contaminated sediments.

1. Introduction

Stormwater detention basins, among the many stormwater control measures (SCMs), are used to intercept and trap sediments by sedimentation (e.g. Marsalek and Marsalek, 1997; Maniquiz-Redillas et al.,

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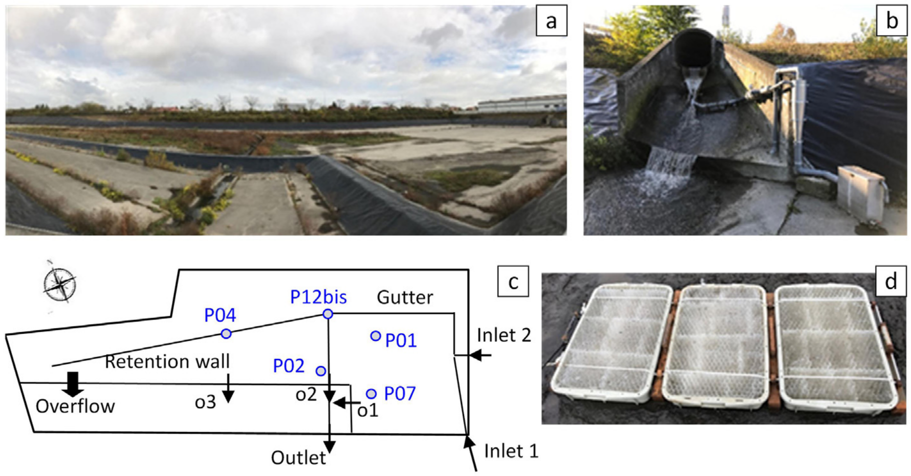


Fig. 1. (a) Panorama of DRB, (b) hydrocyclone trapping system at the inlet 1 of DRB, (c) sketch of DRB and sampling points, (d) honeycomb-like sampling traps.

2014). They are widely used in most developed countries, such as France, Canada, US and other European countries (Urbonas, 1994). Since most of the pollutants, such as trace metals, polycyclic aromatic hydrocarbons (PAHs) and some pathogenic bacteria, are transported in a predominantly particulate form in urban stormwater (e.g. Barbosa et al., 2012; Gasperi et al., 2014; Zgheib et al., 2011), stormwater detention basins play an important role in the decontamination of stormwater by means of settling processes. A better understanding of key mechanisms in these basins is important to improve hydraulic and pollutant removal performance. Sediment movement in detention basins or reservoirs depends on their characteristics, e.g. size, shape, cohesive aspect, density, and settling velocity (Loch, 2001). It also depends on the residence time, which in return is controlled by the basin's geometry and flow patterns (Verstraeten and Poesen, 2000; Zhang, 2009; Akan, 2010). While the interactions between hydrodynamics and physico-chemical characteristics are of importance, they have not commonly been considered during the basin design phase, which to date has been primarily focused on hydraulic performance (e.g. Persson, 2000). In addition, the accumulated contaminated deposits can be resuspended by incoming stormwater, subsequently being transported to the downstream aquatic environment. Sediment aerosolization could also pose a microbiological risk for humans through inhalation of aerosols during maintenance (Bernardin-Souibgui et al., 2018). Hence, appropriate management of accumulated contaminated sediments is crucial. Sustainable management of contaminated sediments is challenging, due to (i) expensive transportation, treatment and recycling fees, and (ii) limited reliable and suitable treatment and recycling options (e.g. Petavy et al., 2009). It is therefore essential to determine the distribution of contamination in stormwater detention basins and target the priority zones for treatment and intervention.

A thorough geochemical characterization of settled sediments can be used to determine the contamination distribution in stormwater detention basins (Jang et al., 2010; Tedoldi et al., 2017). However, a large number of samples are needed, and their collection and characterization are expensive and time-consuming. Numerical models, on the other hand, are an effective and low-cost approach to predict the distribution of contamination. A chemical model was proposed by Vezzaro et al. (2010) to simulate the removal of micropollutants in such basins based on relevant removal processes, such as settling, volatilization, sorption, biodegradation and abiotic degradation. This model, however, fails under complex hydrodynamic conditions in basins (Sebastian et al., 2014) as the interactions between hydrodynamics and physico-chemical characteristics are ignored. The combination of Lagrangian Discrete Phase Model (LDPM) and the correlation between physical and chemical characteristics can be an appropriate alternative. Indeed, the simulation of particle transport provides a quick, convenient and low-cost way to characterise particle movement and predict preferential deposition zones of the particles and their associated pollutants. Such an approach permits alternative basin geometries to be compared, facilitating the design process. For example, a LDPM implemented in computational fluid dynamics (CFD) package allows particle transport mechanisms in stormwater detention basins to be simulated, with particle physical characteristics used as model inputs (Adamsson et al., 2003; Yan et al., 2014). Finally, the transport mechanisms of pollutants can be simulated by implementing the correlation between particle physical and chemical characteristics using outputs of LDPM simulations or other solid transport model results.

Trace metals are considered as priority stormwater chemical pollutants related to urban activities. The decision No. 2455/2001/EC issued by the European Parliament and Council listed 33 priority substances,

Table 1
Characteristics of DRB.

Type of characteristics	Characteristics
Location	Chassieu, France
Type of drainage system	Separated stormwater drainage system
Watershed type	Industrial watershed of 185 ha
Impervious rate of the watershed	75%
Bottom surface area	11,000 m^2
Storage capacity	32,000 m^3
Max outlet flow rate limit	0.35 m^3/s (Bardin and Barraud, 2004)
Materials	Bottom in bitumen and banks covered with plastic lining
Compositions	2 inlets (mainly inlet 1 operates, illustrated in Fig. 1b), an outlet, a gutter guiding the flow to 3 orifices during dry periods and an overflow when the water height exceeds the retention wall (presented in Fig. 1c).
Construction and maintenance	Built in 1975, rehabilitated in 2002, cleaned in 2006 and total sediment removal in 2013.

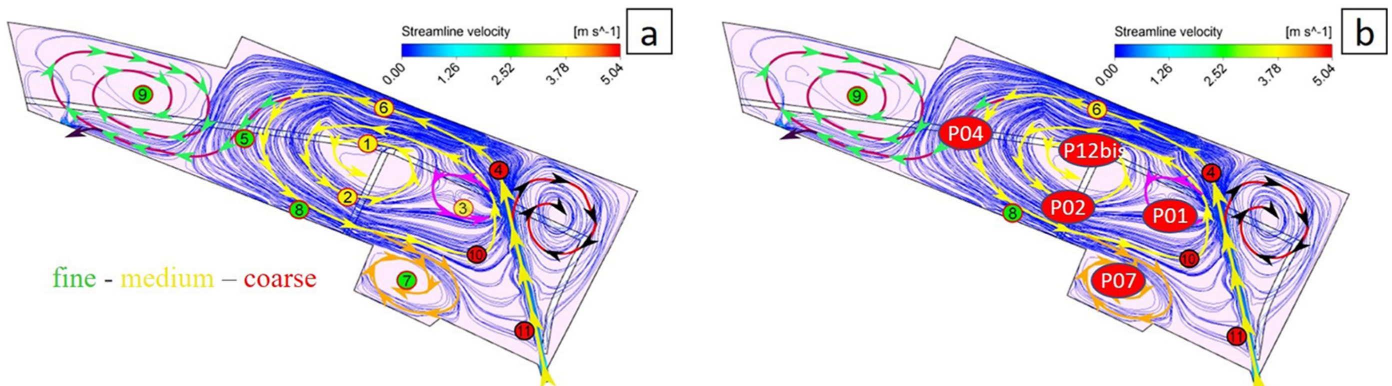


Fig. 2. Streamline of the liquid phase flow obtained by computational fluid dynamics simulations ($Q = 0.35\ m^3/s$, mean flow rate) and characteristics of sediments: (a) locations of settled particles of different sizes shown in different colors: red circles for coarse particles (D50 > 500 μm), yellow for medium ones (D50 around 40 μm), and green for fine ones (D50 around 25 μm). (b) locations of different sampling sites (P01, P02, P04, P07, P12bis) at the base of DRB. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

including Ni, Pb and their compounds. Zgheib et al. (2008) extended the list to 88 individual substances, including Cr and Cu. Iron, as a major earth element, plays a key role in the geochemistry of soils and sediments and is of great importance to many metals cycling processes (Taylor and Konhauser, 2011). Iron oxides are also important for the sorption of trace metals (Bradl, 2004; Whitaker and Duckworth, 2018). As far as physical characteristics are concerned, settling velocity is an integrated property of particulate pollutants, as it is related to multiple characteristics such as diameter, density, shape and surface roughness (Loch, 2001). Several investigations are reported in the literature regarding both trace metal contents (Becouze-Lareure et al., 2019; Sébastian et al., 2014a) and particles' settling velocities in stormwater detention basins (Torres et al., 2007; Yan et al., 2014). However, to the best of our knowledge, little is known concerning the relationship between metal contents, particularly Fe, and settling velocity in urban stormwater detention components. Bentzen and Larsen (2009) investigated such relationship in a wet detention pond of road runoff, where high Cd, Cr, Zn and Ni contents were found associated with low settling velocities while the tendency for Cu and Pb was unclear. However, the conclusions were empirically drawn without quantitative analyses. Torres et al. (2007) found a significant correlation among median settling velocity (V50) and contents of Cd, Cu, Pb and Zn in a stormwater detention basin, but metal contents with respect to different fractions of settling velocities were not considered. Overall, a thorough analysis of the correlation between metal contents and settling velocity and the importance of Fe have not been considered in previous work.

The objective of this article is to predict the spatial distribution of trace metal contamination through (i) the investigation of the correlation between metal contents (major element: Fe and some trace metals: Cr, Cu, Ni and Pb) in trapped sediments and their settling velocity in a stormwater detention basin, and (ii) the implementation of such correlations in a LDPM, taking into considerations the interactions between hydrodynamics and particle physico-chemical characteristics.

2. Methods and materials

2.1. Experimental site and sampling locations

Our work was conducted in the Django Reinhart stormwater detention basin (DRB), which remains dry during dry weather. The DRB is a part of the broader facilities investigated within the framework of the Field Observatory for Urban Water Management (or OTHU in French, http://www.graie.org/othu/ - see also Zhu and Lipeme Kouyi, 2019). Fig. 1a provides a panoramic photograph of DRB, while its characteristics are presented in Table 1.

Sediments were collected from six sampling sites, five of which are situated at the base of DRB (P01, P02, P04, P07, P12bis, illustrated in Fig. 1c). The sampling sites were selected according to sediment accumulation zones and previous studies (Sébastian et al., 2014b; Torres et al., 2007; Yan et al., 2014). Stormwater enters from the inlet situated at the lower-right side, and creates a counterclockwise swirl at the centre of the basin (shown in yellow in Fig. 2). There are then several flow paths within the basin (in orange, green, red and magenta in Fig. 2). Sampling points P01 and P02 are prone to trap medium-sized particles (median size D50 of 40–200 μm), given their locations near the basin inlet and at the middle of a flow path, respectively. Points P04 and P07 are situated at the end of the flow path where fine particulate particles (median size D50 of 25–70 μm) are expected to be trapped. Indeed, Jacopin et al. (1999) revealed the same trend i.e. the finest particles are found in the furthest part from the inlet of detention/retention basin. Point P12bis is a specific sampling site near a small tank designed to trap hydrocarbons, where medium and coarse particles are usually found (median size D50 of 200 μm). For each point, 3 honeycomb-like traps (Fig. 1d) were placed at the bottom. A hydrocyclone system was designed as a sampling device and installed at the inlet (illustrated in Fig. 1b) in order to intercept particulate pollutants from the inlet of basin (Zhu et al., 2016).

2.2. Presentation of campaigns

Five campaigns were conducted during rain events in 2017 (from spring to winter). Nineteen sediment samples from 6 sites were collected and characterized (summarized in Fig. 3). The main characteristics of the sampled rain events and their corresponding hydraulic parameters in the detention basin are presented in Table 2.

2.3. Methodology to predict distribution of trace metal contamination

2.3.1. Physical and chemical characterizations and determination of their correlation

In this study, particle-size distribution, settling velocity distribution and metal contents in sediments were analysed, according to the standards and protocols listed in Table 3.

The VICAS protocol (a French acronym for Effluent Settling Velocity, Chebbo and Gromaire, 2009) is applied to measure settling velocity distribution. It is based on the principle of homogeneous suspension, where the solids are uniformly distributed over the entire sedimentation height. The measurement is realised with a plexiglas sedimentation column in the laboratory. The settled solids at a set of 10 predefined time points ($t = 1, 2, 4, 8, 16, 32$ min, 1, 2, 4, >24 h) are manually collected at the bottom of the sedimentation column in aluminium receptacles. The evolution of accumulated mass of settled particles over time is then determined by measuring the mass in each receptacle, which yields the settling velocity distribution curve. Meanwhile, the average settling velocity in each receptacle defined in Eq. (1) is used to investigate the correlation between metal contents and settling velocity:

$$Vs = \frac{H}{t} \quad (1)$$

where Vs denotes particle settling velocity (mm/s), H is water height in the sedimentation column (mm) and t is the time point (s) when the receptacle with collected particles is removed.

Metal contents in particles are then analysed with ICP-OES (inductively coupled plasma optical emission spectrometry) method. The total suspended solids (TSS) are filtered through glass fibre filters (0.45 μm) and dried at 105 °C (French standard NF EN 872, 2005). Mineralization is then applied in order to extract soluble elements from particles with aqua regia (NF EN 16174, 2012). The obtained solution is finally diluted to 50 ml, filtered at 0.45 μm and analysed with ICP-OES equipment. For each element, the limit of detection (LOD) is calculated from the calibration. The limit of quantification (LOQ) is considered to be 3.3 times of LOD. In our study, as a precaution, only the values larger than 3 times of LOQ were considered as quantifiable.

Fig. 4 shows the methodology of physical and chemical characterizations. Two groups of data (physical and chemical characteristics) were gathered from the collected samples, in order to, respectively, (i) obtain overall physical and chemical characterization of samples and associated global relationships and (ii) reveal in particular the correlation between metal contents and settling velocity, as well as its significance. The first group (Data of group I) consists of particle-size distribution, settling velocity distribution and metal contents (Fe, Cr,

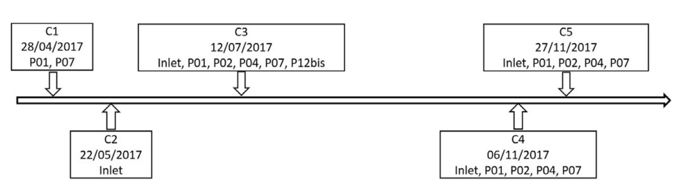


Fig. 3. Campaign timeline and sampling points for each campaign

Table 2
Characteristics of the sampled rain events and corresponding hydraulic parameters in DRB.

Campaign date	C1 28/04	C2 22/05	C3 12/07	C4 06/11	C5 27/11
Rainfall characteristics					
Rain Duration (hour:min)	19:22	08:14	03:02	18:26	16:28
Total Depth (mm)	26.5	17.5	10.5	30.9	11.2
Mean Intensity ($mm \cdot h^{-1}$)	1.4	2.1	3.4	1.7	0.7
Max intensity ($mm \cdot h^{-1}$)	6	7.5	58.2	11.7	6.9
ADWP (days)	23	4	8	12	11
DRB hydraulic parameters					
Mean inflow rate (m^3/s)	0.21	0.31	0.24	0.31	0.11
Max inflow rate (m^3/s)	0.76	0.81	1.27	1.08	0.77
Max water level (m)	0.51	0.54	0.39	0.69	0.50

ADWP: antecedent dry weather period.

Cu, Ni and Pb) of 19 sub-samples (deriving from the 19 original samples). The second group (Data of group II) consists of settling velocities and metal contents (Fe, Cr, Cu, Ni and Pb) of 190 sub-samples under the VICAS protocol. For each of the 19 samples, 10 sub-samples, i.e. receptacles of particles removed from the bottom of experimental column at all the 10 predefined time points *t*, have different settling velocities (described by Eq. (1)). For each receptacle, metal contents in collected sediments are measured.

Several statistics have been computed, including: (i) principal components of physical and chemical characteristics of group I via PCA (Principal Component Analysis), (ii) correlation between metal contents and settling velocity based on the data of group II and test of significance, and (iii) correlation matrix for all metals accounting for data of group II. In this study, Spearman's rank correlation coefficient is applied as the settling velocities are not normally distributed according to the Shapiro-Wilk test (Shapiro and Wilk, 1965) and a monotonic relationship is observed between metal contents and settling velocity. Significance tests of correlation coefficient are also carried out by comparing *p*-value to a significance threshold (denoted as α, equal to 0.05 by default). Bonferroni correction (Dunnett, 1955) is applied to reduce false positive (Type I) errors when multi-comparisons are applied. In order to set FWER (Family-wise error rate) lower than α, it rejects the null hypothesis with $p - value$ as follows:

$$p{-}value \leq \frac{\alpha}{m} \tag{2}$$

where *m* is the total number of null hypotheses.

2.3.2. Strategy to combine derived correlations with LDPM

LDPM was employed to simulate sediment transport in DRB (Yan et al., 2014). The flow was firstly simulated in steady state condition and then used as a base for further sediment transport modelling with a Lagrangian approach. The model was evaluated and had a good fit with both in situ free surface velocity field measurement results (Zhu and Lipeme Kouyi, 2019) and measured accumulated sediment distribution at the bottom of DRB (Yan et al., 2014). LDPM approach allows the computation of particles trajectories. The interaction between sediments and the bottom wall was modelled by an original boundary

Table 3
Standard and protocols for physical and chemical analyses.

Analysis	Method	Standard
Particle-size distribution	Mastersizer 2000 laser diffraction granulometer	NF ISO 13320-1 (2000)
Settling velocity distribution	VICAS protocol (Chebbo and Gromaire, 2009)	
Metal content	ICP-OES	NF EN ISO 11885 (1998)

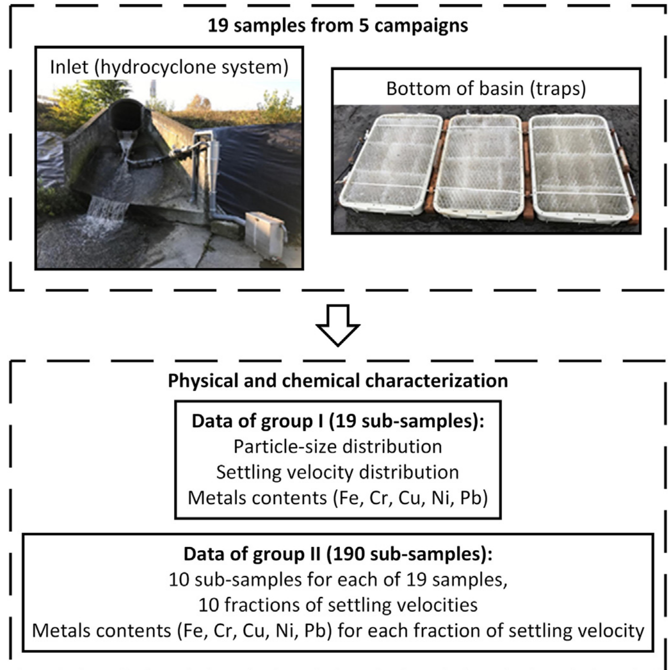


Fig. 4. Methodology of physical and chemical characterizations.

Table 4
Physical and chemical characteristics of trapped sediments.

Sample		D50 (μm)	V50 (m/h)	Fe contents (g/kg)	Trace metal contents (mg/kg)			
					Cr	Cu	Ni	Pb
C1	P01	69.2	7.0	nm	74.3	360.1	56.7	98.8
	P07	54.5	6.6	nm	78.3	325.7	54.1	114.3
C2	inlet	55.6	6.7	13.5	96.5	222.0	237.7	99.8
C3	inlet	313.5	11.6	18.2	71.5	242.3	85.2	114.1
	P01	52.9	9.6	20.4	73.5	233.5	74.6	105.7
	P02	37.7	3.4	nm	nm	nm	nm	nm
	P04	45.3	6.9	nm	nm	nm	nm	nm
	P07	45.5	4.1	27.5	93.1	278.8	93.9	137.1
	P12bis	78.3	7.5	15.4	62.7	226.8	70.5	95.8
	mean	95.5	7.2	20.4	75.2	245.4	81.1	113.2
	stdv	107.7	3.2	5.2	12.8	23.1	10.6	17.6
C4	inlet	nm	1.9	21.0	154.3	584.4	93.8	200.6
	P01	144.6	5.9	18.7	76.3	308.7	58.1	96.7
	P02	199.3	2.9	25.6	96.4	457.4	68.7	136.6
	P04	129.4	2.6	24.4	80.5	428.2	61.9	140.7
	P07	79.8	6.7	17.9	89.6	355.3	63.1	111.5
	mean	138.3	4.0	21.5	99.4	426.8	69.1	137.2
	stdv	49.2	2.2	3.4	31.7	105.9	14.3	39.8
C5	inlet	870.5	4.3	14.9	110.4	429.0	80.4	69.7
	P01	78.5	6.0	24.7	94.9	349.2	65.7	115.7
	P02	100.2	2.1	22.2	89.1	378.3	72.7	167.2
	P04	433.3	5.4	21.1	65.8	258.1	59.2	112.3
	P07	109.8	5.7	16.6	66.3	275.0	42.7	88.9
	mean	318.5	4.7	19.9	85.3	337.9	64.1	110.8
	stdv	341.6	1.6	4.0	19.2	71.4	14.4	36.6
inlet	mean	413.2	6.1	16.9	108.2	369.4	124.3	121.1
	stdv	416.5	4.2	3.4	34.7	170.9	75.8	56.2
P01	mean	86.3	7.1	21.3	79.7	312.9	63.8	104.2
	stdv	40.3	1.7	3.1	10.1	57.3	8.2	8.6
P02	mean	112.4	2.8	23.9	92.8	417.8	70.7	151.9
	stdv	81.5	0.6	2.4	5.1	55.9	2.9	21.6
P04	mean	202.7	5.0	22.7	73.2	343.2	60.5	126.5
	stdv	204.1	2.2	2.4	10.4	120.3	1.9	20.1
P07	mean	72.4	5.8	20.7	81.8	308.7	63.5	113.0
	stdv	28.9	1.2	6.0	12.1	38.7	22.0	19.7

D50: median particle size, V50: median settling velocity, nm: not measured, stdv: standard deviation.

condition based on turbulent kinetic energy threshold (Yan et al., 2014). During LDPM simulations, the location in the DRB as well as the physical characteristics (settling velocity, particle size and density) of sediments was recorded. The focus was placed on the settled particles as they represent accumulated sediments or deposits in the DRB. The metal contents distributions were then computed by combining the correlation between metal contents and settling velocity with recorded simulated data deriving from LDPM approach.

3. Results

3.1. Physical and chemical characteristics from data of group I

Physical and chemical characteristics are illustrated in Table 4. In general, settling velocity varies with campaign and site, consistent with previous work (e.g. Torres et al., 2007). The samples collected from the inlet show the most significant temporal variability, with median settling velocity ranging from 1.9 to 11.6 m/h. Trapped particles from P01 have higher settling velocities than those from the other sampling sites at the base of DRB. Indeed, the location P01 is near the inlet where heavy particles settle rapidly, as demonstrated e.g. by Jacopin et al. (1999).

As illustrated in Table 4, Fe contents are at least 36 times higher than Cu contents and up to 200 times higher than the other trace metal contents (Cr, Ni and Pb). As for trace metals, Cu, Ni and Pb contents exceed the target values of Dutch standards (DTIV, 2000). Cu contents, as well as Pb contents in some cases, surpass even the intervention threshold. It is noticed that Fe contents at the inlet are lower (average 16.9 g/kg DM) than those observed in the basin (average 20.7–23.9 g/kg DM), in opposition to trace metals. Besides, samples collected from the inlet show a significant temporal variability, with a coefficient of variation of approximately 50%.

Fig. 5 shows PCA results of all physical and chemical variables (particle size, settling velocity and metal contents). Principal component 1 (PC1) explains 48.9% of variance, while PC2 explains 22.9%. Overall, settling velocities are highly related among themselves (V20, V50 and V80). The same conclusion can be drawn for particle sizes (D10, D50 and D90). The relationship among different metals, as well as that between metal contents and settling velocities or sizes, is unclear. Temporal variability is observed as expected. Event C3 is a storm of high rain intensity and maximum inlet flow rate but low total depth and rain duration. Event C4 is a rain event of the highest total depth and maximum water level in DRB. This may lead to strong variation of spatial distribution of sediments due to shear stress (Adamsson et al., 2003) or turbulence in DRB (Sechet and Le Guennec, 1999). Significant temporal variability is particularly observed for particles intercepted by the hydrocyclone device at the inlet of the basin. Among particles trapped at the base of DRB, those at P02 seem to be different from particles observed at other sites, characterized by their low settling velocities and high metal contents, regardless of the campaign. In fact, particles with high settling velocities tend to settle near the inlet (P01), while P02 is in the main recirculation zone with long residence time which enables particles to settle progressively (Zhu and Lipeme Kouyi, 2019).

3.2. Correlation between metal contents and settling velocity, and correlation matrix for different metals based on data of group II

All the results from different campaigns (C1–C5) and sampling sites (inlet, P01, P02, P04, P07 and P12bis) are presented in Fig. 6(a–e) to analyse the correlation between metal contents and settling velocity. Fig. 6 (f–j) are boxplots of different metal contents with respect to settling velocity. In general, results show that contents of the metals in settled sediments are relatively stable, with values varying within ranges of 15–35 g/kg, 20–120, 200–650, 20–150 and 50–250 mg/kg DM for Fe, Cr, Cu, Ni and Pb, respectively, except for a few outliers. [Fe] and [Pb] have significant correlations with log(Vs) after Bonferroni correction, especially in the case of [Fe], with a correlation coefficient equal to −0.52. Higher [Fe] and [Pb] are related to lower settling velocity. The best fit of their correlations are illustrated in Fig. 6a and e and can be defined by the equation shown at the upper-right side. [Cu] tends to decrease slightly with the increasing of Vs, in opposite to [Cr]. Indeed, previous works suggest that Fe contents are relatively stable, while Cu

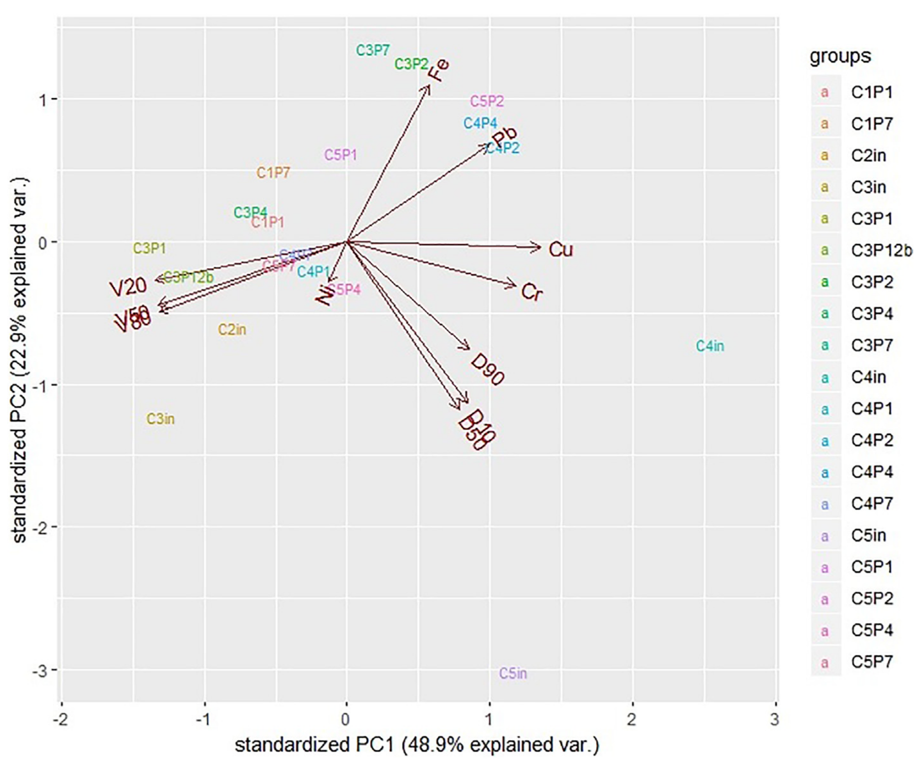
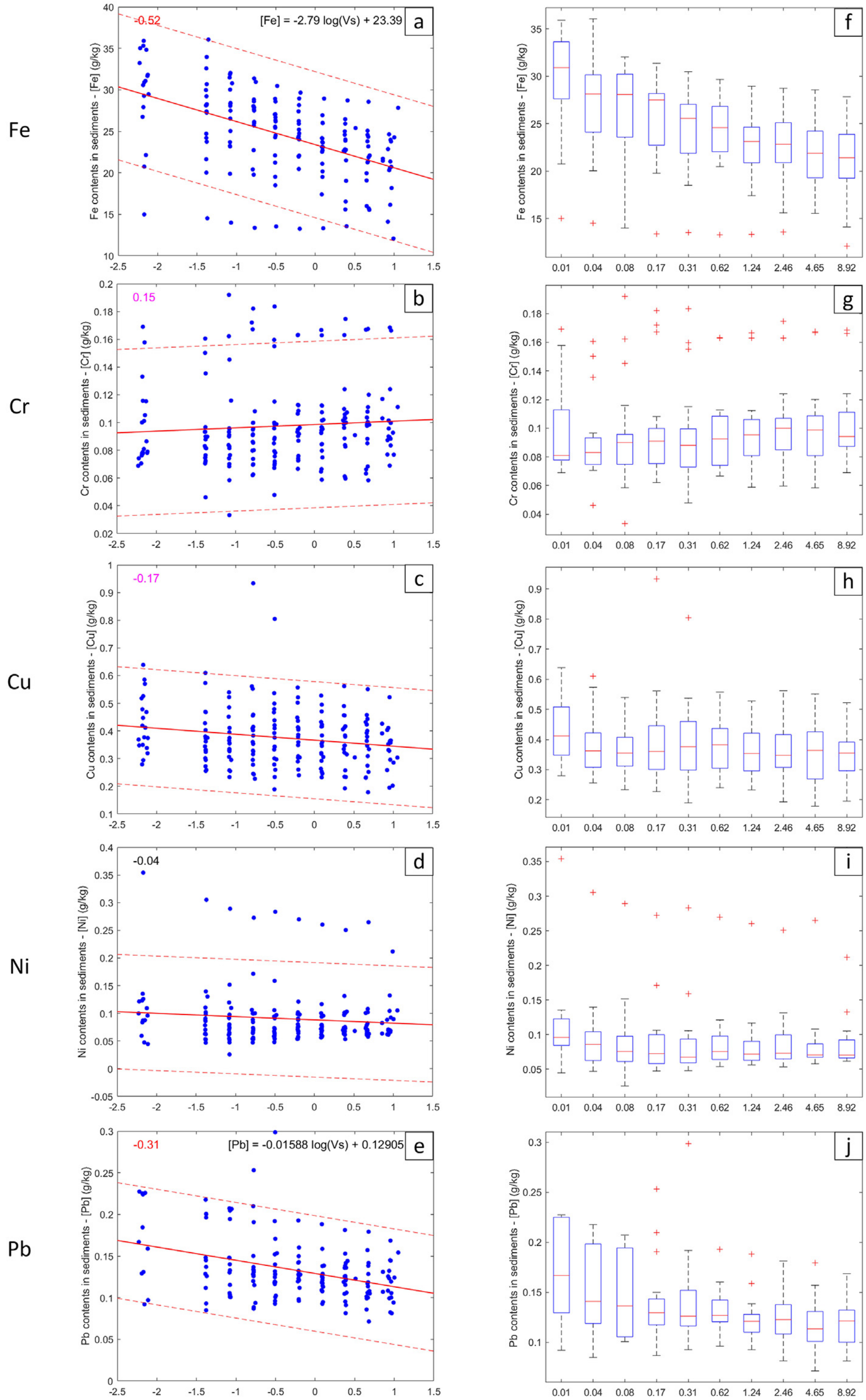


Fig. 5. Principal component analysis (PCA) of physical and chemical characteristics based on the data of group I.



Parameters in (a-e)	Explications
Correlation coefficient	Left-upper side in red (p -value < 0.05/190 according to Eq. 2 with Bonferroni correction) when the correlation is significant; in magenta when p-value < 0.05.
Correlation equation	Right-upper side when the correlation coefficient is significant.
Correlation best fit	Least-squares error minimization, illustrated by red solid lines
95% confidence interval	Illustrated by red dashed lines

Fig. 6. (a–e) Correlation between metal contents [M] in g/kg DM and settling velocity (*Vs*) in mm/s based on data of group II; (f–j) Boxplot of different [M] with respect to each *Vs*.

can be variable in stormwater environment given its affinity with organic matter (Camponelli et al., 2010). No obvious correlation is observed between [Ni] and *Vs*. The dispersion around the regression line (marked as red dashed lines for 95% confidence interval) is related to spatio-temporal variabilities of metal contents and settling velocity distributions, physico-chemical processes and characteristics, as well as uncertainties in sampling and analytical measurements (Torres and Bertrand-Krajewski, 2008; Sébastian et al., 2015). The physico-chemical analyses conducted by Becouze-Lareure et al. (2018) show a high organic matter content for the trapped sediments in DRB. This may lead to changes in the physico-chemical conditions in the sediments, which could affect the solubility and availability of certain trace metals. These different processes and assumptions have already been observed in various investigations focusing on urban sediments, such as those from rivers (Chapman et al., 1998) and from reservoirs (Frémion et al., 2016; Frémion et al., 2017). Torres et al. (2007) also showed the spatial and temporal variability of settling velocity of DRB sediments.

The correlation matrix among different metal contents is also calculated (shown in Fig. 7). Results show that almost all the studied metals have significant correlations with each other except for Cr versus Fe and Ni versus Pb. Fe and Pb are the most related metals, with a correlation coefficient of 0.61.

3.3. Prediction of trace metals contamination distribution

Given the significant correlations of Fe and Pb contents with settling velocity, the derived correlations (equation in Fig. 6) were applied on LDPM outputs to simulate the spatial distribution of Fe and Pb contents in DRB. Fig. 8 illustrates the comparison of the measured and simulated distribution of Fe and Pb contents at the base of DRB. The simulated [Fe] at P01, P02, P04 and P07 are consistent with the measurements obtained from group I, where P07 and P02 have the highest [Fe] in sediments. P07 shows a significant variation in [Fe]. Indeed, P07 is situated at the end of pathway, where sediments tend to be resuspend, as previous work has suggested (Zhu et al., 2017). In addition, P07 is near an orifice, which may lead to maximum flow shear and velocities around this site and through the orifice, increasing variability due to dispersion of sediments (e.g. interaction between burst and bedload transport, see e.g. Sechet and Le Guennec, 1999). Simulated [Pb] in DRB is around 129 mg/kg DM, which is not consistent with the measurements due to temporal variability. However, the results are consistent when comparing the simulated Fe contents distribution with the measured Pb contents at different sampling points. Indeed, significant positive correlation between Fe and Pb is revealed in Fig. 7. Hence, it is interesting to use Fe as an indicator to predict other metal contents distribution. Uncertainties with 95% confidence interval of ±8.8 g/kg and ±69.4 mg/kg for Fe and Pb, respectively, are observed. In DRB, P02 and P07 are highly contaminated by Pb according to the analyses obtained, not only throughout this study, but also in many investigations carried over the last 10 years. For example, Becouze-Lareure et al. (2016) show that the areas farther from the inlet are more contaminated and that P02 presents highly contaminated sediments. The sediments at P02 and P07 settle slowly, with a mean settling velocity of around 2 mm/s, explained by the two locations being remote from the inlet.

4. Discussion

4.1. Use of Fe as indicator of pollution and for the prediction of contamination distribution

In this study, [Fe] in trapped sediments is strongly related to settling velocity and this relationship is relatively stable from one site to another and from one event to another. In addition, significant correlations

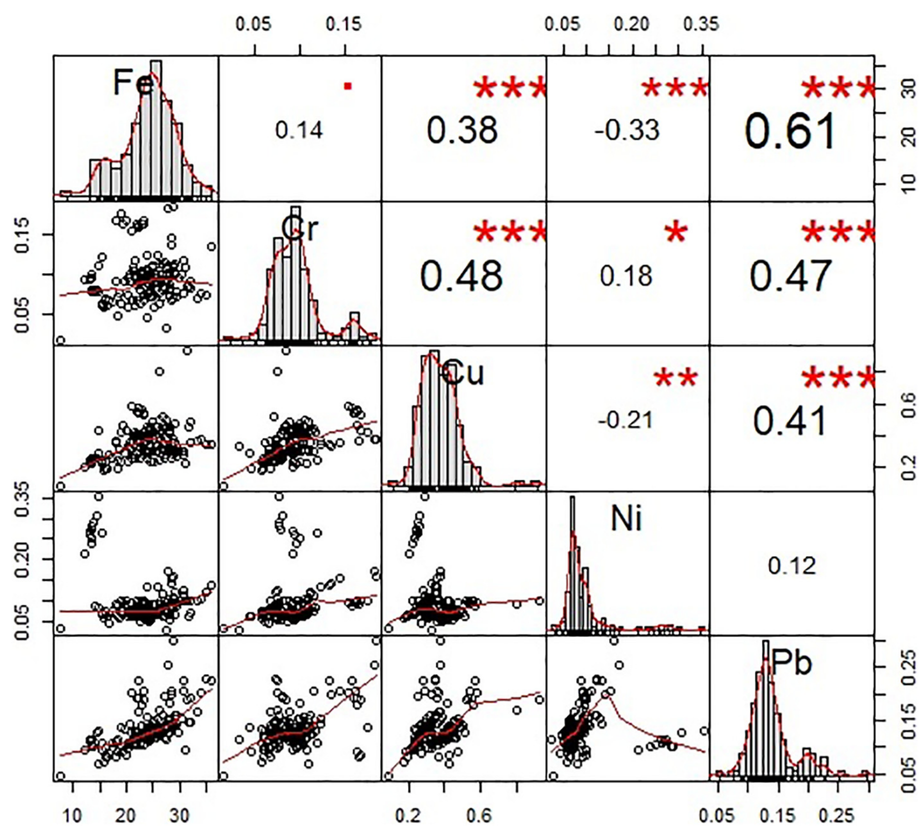


Fig. 7. Correlation matrix of different trace metal contents in trapped particles: the distribution of each variable is listed on the diagonal. The bivariate scatter plots with their linear fits are displayed below the diagonal. The upper triangle contains the correlation coefficients and their significance levels. Each significance level is associated to a symbol: *p*-values of 0.001, 0.01, 0.05, 0.1 correspond to symbols of ***, **, * and ■, respectively.

between Fe and trace metals are observed (Fig. 7). Other studies have also observed positive correlations between Fe and Cr, Cu, Pb, Ni and Zn in highway stormwater runoff (Kayhanian et al., 2007). Drapeau et al. (2017) revealed a close link among Al, Fe and Si, as well as linear regression between Fe and Cu, Zn, P, S, Si and organic matter in a stormwater infiltration basin. Given the significant correlation between Fe and certain pollutants (here Pb and Cu), the simulated Fe content distribution can then be utilised to understand or to explain the distribution of other particulate pollutants that have affinities with Fe. Besides, iron isotopes have been widely used as tracers to analyse biochemical processes (e.g. metal transport, microbial redox reactions and transformation of organic matter-ferrihydrite coprecipitates), and track sources and other components such as suspended Fe-organic carbon aggregates (Gould et al., 2008; Ingri et al., 2018; Owens et al., 2012; Tishchenko et al., 2015). In DRB, P07 and P02 present the highest maximum values of [Fe] and are also highly contaminated areas given the significant positive correlation between Fe and certain trace metals. Hence, P07 and P02 should be the preferential zones for cleaning.

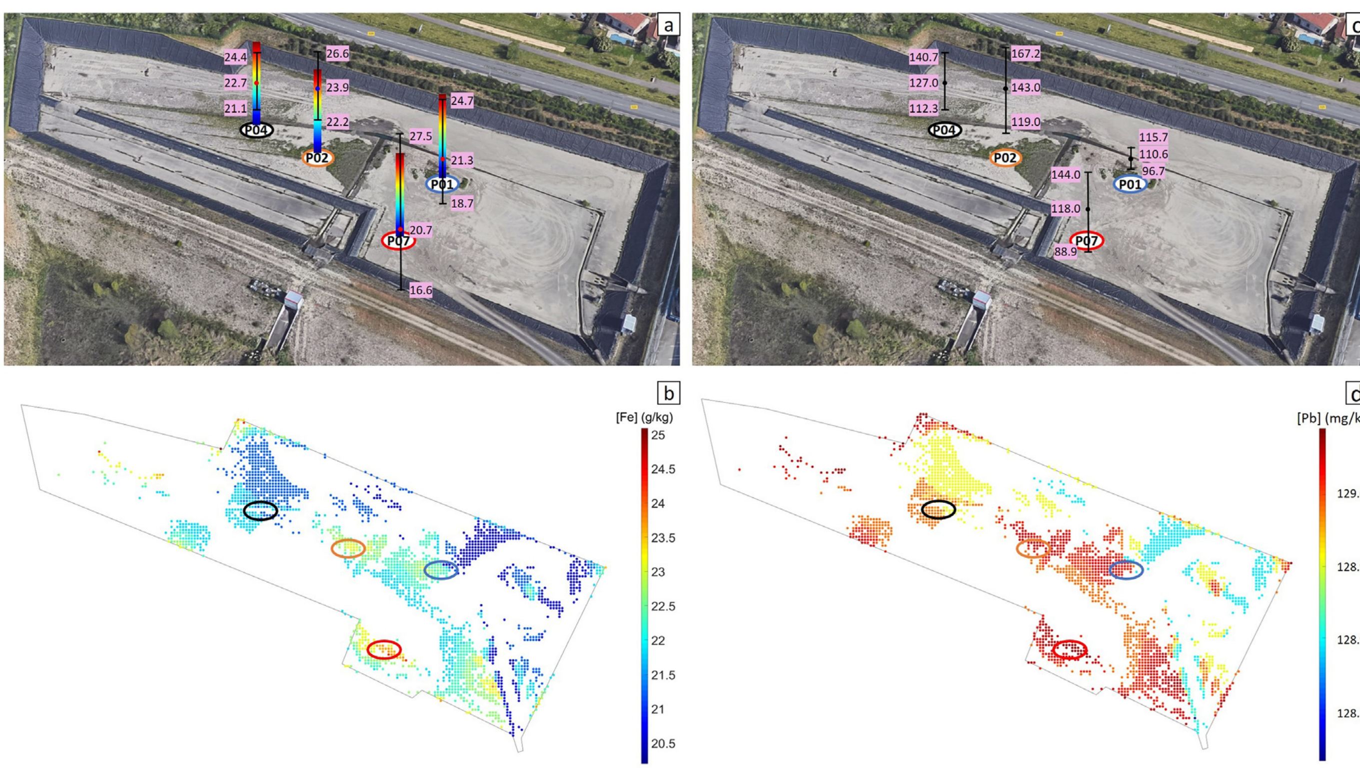
4.2. Use of settling velocity for stormwater detention basin sediments management

Most studies on the correlation between particle physical and chemical characteristics in stormwater detention basins focus on the relationship between particle size (e.g. Kayhanian et al., 2012; Tuccillo, 2006) or sediment density (El-Mufleh et al., 2014) and trace metal contents. However, settling velocity should be emphasized given its relationship with other physical characteristics (Loch, 2001). It has been widely applied in urban stormwater management and its importance has been demonstrated (Ciccarello et al., 2012; Yun et al., 2010). Settling velocity is also a key element, when combined with the residence time, for the sizing of stormwater detention basins.

The design and management of stormwater detention basins not only need knowledge of hydrodynamic behaviour, but also an understanding of the spatial distribution of highly contaminated sediments. Various modelling approaches (e.g. LDPM) can be used to address the understanding of the velocity field and the spatial distribution of sediment in stormwater detention basins (Adamsson et al., 2003). Given that the particulate fraction of certain metals and PAHs in stormwater from industrial catchments is above 60% and 80%, respectively (Becouze-Lareure et al., 2019) and most other pollutants are conveyed in particulate phase (Ashley et al., 2004), the investigation of the correlation between metal contents in sediments and settling velocity may support the coupling between chemical and physical aspects. The final simulation results help to predict pollutant deposition and determine the priority cleaning zones in stormwater detention basins. They may also be used as a support for the design of stormwater detention basins using trapping or removal efficiency as targeted criteria.

4.3. Relationship between sediments' physical and chemical characteristics

The significant correlation between metal contents in sediments and their settling velocity is a key finding of this study. Badin et al. (2008) suggested that organic matter in urban stormwater sediments is related



	[Fe] (g/kg DM)	[Pb] (mg/kg DM)
Measured [M] at P01, P02, P04 and P07: minimum, averaged and maximum values are plotted, respectively.	a (data of C3-C5 in group I)	c (data of C3-C5 in group I and previous campaign data by Becouze-Laureure et al. (2018))
Simulated median [M] spatial distribution at the bottom of DRB	b: uncertainty of ± 8.8 g/kg (95% confidence interval)	d: uncertainty of ± 69.4 mg/kg (95% confidence interval)

Fig. 8. Comparison of measured and simulated Fe and Pb contents distribution: P01, P02, P04, P07 locations are coloured in blue, orange, black and red, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

to grain size. Bernardin-Souibgui et al. (2018) found that *Nocardia* counts are positively linked to volatile organic matter. Wiest et al. (2018) revealed that BPA is associated with fine particles. Concerning the physical characteristics, we can analyse particle-size distribution, density and shape related to each fraction of settling velocity to understand their links with chemical characteristics. Given the strong correlation of Fe content with settling velocity, it is interesting to investigate more thoroughly the oxidation states, isotopes of Fe and the related physical, chemical and biological processes, such as sorption and precipitation (Taylor and Konhauser, 2011). Besides, other pollutants such as PAHs, organic matter and some pathogenic bacteria can be investigated to see if they are related to metals or settling velocity.

4.4. Sediment management based on targeted dredging strategy

The methodology proposed in this article can be applied in any other stormwater detention or retention basin (dry or wet) constructed on a catchment (industrial, urban or periurban) in order to compare and evaluate deriving results against the obtained correlation in this study. This methodology also informs the intervention and treatment of sediments with a focus on the highly contaminated area. In general, the intervention criteria could be determined by comparing the metal contents to the Dutch target and intervention values (DTIV, 2000) and taking into account site specificities. The outcomes of this paper suggest that targeted dredging may be a good alternative for sediment management. In the case of DRB, P02 and P07 are highly contaminated and should be treated promptly. In theory, these contaminated sediments could be stored in places where less recirculation and sediment resuspension phenomenon occur (e.g. the upper-left side of DRB). Indeed, Becouze-Lareure et al. (2018) showed that the ecotoxicity level decreases over time and sediments should thus be left in situ before being discharged to a dedicated resource recovery plant or a separation device. Sediments from these most problematic zones could then be separated by means of screening and attrition (Petavy et al., 2009). As the focus is only placed on the problematic areas, less sediments need to be extracted and can be directly treated in situ. Such an approach could substantially reduce cost related to sediments transportation, treatment, and reuse.

5. Conclusions

Better understanding of the spatial distribution of trace metal contamination in stormwater detention basins is vital to better manage accumulated sediments and better design such facilities, taking into account the interactions between hydrodynamics and sediment physico-chemical characteristics. Our work investigated the locations of highly contaminated sediments with trace metals in a settling and detention basin by coupling a LDPM with the relationships of sediment' physico-chemical characteristics (correlation between Fe, Cr, Cu, Ni, Pb contents and settling velocity). Based on a large dataset, Fe and Pb contents have a significant correlation with settling velocity, followed by Cu and Cr. The observed significant correlation between Fe content and settling velocity remained stable for all campaigns and sampling sites. An equation describing this correlation is coupled with simulated spatial distribution of sediments to predict Fe content distribution. Obtained results are consistent with in situ measurements. Accounting for hydrodynamic behaviour (streamlines exhibiting recirculations, flow shear and turbulence characteristics), particles carrying Fe could then be tracked and used as an indicator to comprehensively identify trace metals contamination areas (e.g. deposition and resuspension zones). This may help to determine the priority cleansing zone in detention basins and better design the stormwater detention basin by taking into consideration all these correlations as well as hydrodynamic parameters in sediments transport equations.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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