KTH Architecture and the Built Environment

TREATMENT CONDITIONS FOR THE REMOVAL OF CONTAMINANTS FROM ROAD RUNOFF

Magnus Hallberg

June 2007

TRITA-LWR PHD 1032
ISSN 1650-8602
ISRN KTH/LWR/PHD 1032-SE
ISBN 978-91-7178-645-6
ISBN 978-91-7178-661-6

ACKNOWLEDGEMENTS

Skanska and the Skanska PhD program have provided financial support for this work to help increase knowledge and competence in the construction field. I would like to express especial appreciation to Associate Prof. Kyösti Tuutti and Environmental Manager Johan Gerklev for their important support. I would also like to extend the same appreciation to Professor Björn Täljsten at the Technical University of Denmark. Financial support for the fieldwork has been received from the Office of Regional Planning and Urban Transportation in Stockholm (RTK), the Road Bridge and Tunnel Consortium (VBT) in cooperation with the Royal Institute of Technology (KTH), the Swedish Road Administration in the Stockholm region, and the Swedish construction industry's organization for research and development-SBUF. The participants in the RTK stormwater and reference group have provided a valuable network for communicating results and important feedback. VBT has provided notable input during the seminars and the road group meetings. The support from Mr. Torbjörn Lundbom and the personnel at the Swedish Road Administration has been essential for the work by providing "first hand" access to the study sites and to traffic data. Two people have had to endure my many ideas, Mr. Bo Pettersson and Mr. Olli Kärki. Mr. Pettersson and I spent many evenings pondering construction and building the experimental equipment in Eugenia. Mr. Kärki has not only been exposed to diving in storm water basins in search of sediment traps but also provided skilled craftsmanship and good solutions during the difficult fieldwork. Also warm thoughts to my fellow colleagues at Land and Water Resource Engineering who have been exposed to my novel accent and furthermore the smell of cinnamon in the morning coffee. Moreover, thanks to Mr. Bertil Nilsson for his assistance with the monitoring equipments. I would also like to extend my thanks to Mrs. Gudrun Aldheimer and Mrs. Åsa Snith at Stockholm Vatten AB for their support. For review and comments on the work I would extend a thank you to Tech. Lic. Bernt Ericsson, Mr. Rolf Bergström, PhD Thomas Larm. The amount of data during the work has been extensive. Mr. Kjetil Sørhus has provided programming and good ideas for structuring the data and as my best friend he has had to put up with thoughts and changes during the strangest hours of the day. Thanks are also due to Prof. Jan Grandell for help with statistical problems and formula writing. Almost last, but not least I would like to extend a warm thank you and most sincere appreciation to my supervisor Associate Prof. Gunno Renman. In our work together Gunno's professionalism and positive commitment to research work has been invaluable as a constant source of encouragement for my thesis work. Last but absolutely furthest from the least I would like to extend a warm thank you to my girlfriend Monika Forsman. Monika has been valuable when elaborating on results and experiment since we also share the common ground of chemical engineering. But most importantly Monika, you have been a pillar in supporting me during the ups and downs of the work and endured my Skånska temper.

Magnus Hallberg

Stockholm April, 2007

SUMMARY

The pollutant load in road runoff is related to traffic densities and road maintenance activities. In urbanised areas treatment of road runoff is common and often considered necessary. The pollutants are partitioned between the particulate and dissolved matter. However, the contaminants tend to have an affinity to the particulate material. Sedimentation, the predominant treatment method for road runoff uses various types of ponds. Design tools used for stormwater treatment systems are based on extensive data from existing treatment systems. The variations in the empirical data make it difficult when attempting to evaluate precise conditions for pollutant removal and thereby minimising the land use for a treatment facility. This is a concern in highly urbanised areas where land use often is restricted.

In this work, field studies were conducted in three separate watersheds along the same motorway with an annual average daily traffic exceeding 120,000 vehicles. The aim was to assess treatment conditions for the removal of contaminants from road runoff.

The study of mass transport of total suspended solids used the EU Directive (1991/271/EEC) discharge requirement for urban wastewater treatment: 60 mg/l during winter and summer. The results showed that a capture of the total runoff volume was necessary during both seasons. Ten metals (Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn), as dissolved and particulate bound, were studied in the road runoff during a winter season and the following summer period. The dissolved part of Al, Cd, Co, Cr, Mn, and Ni was significantly higher in winter. The mass concentration (mg/kg) for all metals was significantly higher over the summer except for Al and Co, which showed a higher mass concentration during the winter. The total metal concentration showed a good correlation to total suspended solids (TSS) during winter with exception for Cd. Good correlation to TSS was also found for the summer period for Al, Cu, Fe, Mn, Ni, and Zn. A simple model could describe sedimentation by the initial concentration of TSS, albeit road salt (NaCl) had a significant impact on the sedimentation process during winter. Removal of dissolved metals was studied by column experiments using water granulated blast furnace slag. The result showed good removal for Cd, Cu, Ni, and Zn independent of NaCl concentrations. Sediment accumulation (mg sediment/mm precipitation) was relatively consistent for the studied summer seasons as opposed to winter. The sediment differed in metal mass concentrations (mg/kg) between the seasons. Concentrations of Cu and Zn were high in regard to the guidelines for sensitivity of sediment dwelling organisms and Swedish guidelines for contaminated soils.

The findings suggest that the entire runoff volume must be captured for treatment. The reduction of TSS concentration could be estimated for a specific surface load (m/h). This would also apply for majority of the studied metals that correlated well to the particulate material. Reactive filter technology using water granulated blast furnace slag could be applied for treatment of runoff for the reduction of dissolved metals. However, long-term studies are necessary for its practical implementation. Furthermore; the work shows that on-line turbidity measurements could be used for expedient process control for treatment facilities in similar watersheds dominated by roads. The work could be used together with existing design methods and models to evaluate and optimise road runoff treatment.

SAMMANFATTNING

Förutsättningar för behandling av dagvatten vid avskiljning av föroreningar

Föroreningsbelastningen i vägdagvatten är beroende av trafikbelastningen och vägunderhållet. I urbaniserade områden är behandling av dagvatten vanlig och ofta bedömd nödvändig. Föroreningarna är lösta och partikulära, men har vanligen en affinitet till det partikulära materialet. Den förhärskande behandlingsmetoden för dagvatten är sedimentering, vanligen i dammar. Designmodellerna bygger på data från olika befintliga dagvattenanläggningar. Det varierande ursprunget till det empiriska underlaget medför svårighet att precist värdera designförutsättningarna och således minimera behandlingsanläggningens storlek. I förtätad stadsmiljö, där tillgång på mark är begränsad, kan detta vara ett problem.

I detta arbete har fältförsök genomförts i tre avrinningsområden vilka domineras av en motorled med en årlig dygnstrafik större än 120,000 fordon för att utvärdera behandlingsförutsättningar för vägdagvatten.

Masstransporten av suspenderat material (SS) utvärderades utifrån EU Direktivet (1991/271/EEC) och gränsvärdet för avloppsvatten på 60 mg/l under vinter och sommar. Studien visade att hela avrinningsvolymen bör behandlas oberoende av säsong. Fördelningen mellan partikulärt och löst material studerades för tio metaller (Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn) under vinter och sommar. Den lösta delen av Al, Cd, Co, Cr, Mn och Ni var signifikant högre under vintern. Den partikulära koncentrationen (mg/kg) för samtliga metaller var högre under sommaren med undantag för Al och Co vilka förekom i högre halter under vintern. Totalhalten (µg/l) av metallerna korrelerade väl med SS under vintern med undantag för Cd. Likaledes uppvisade resultaten en god korrelation mellan SS och Al, Cu, Fe, Mn, Ni och Zn under sommaren. Sedimenteringsegenskaperna kunde beskrivas med en enkel modell utifrån koncentration av SS, men förhöjda halter av vägsalt (NaCl) befanns påverka sedimenteringen under vintern. Reduktion av lösta metaller studerades i pilotförsök med vattenkyld granulerad masugnsslagg. God avskiljning erhölls för Cd, Cu, Ni och Zn oberoende av förhöjda halter av vägsalt. Ackumulering av sediment (mg sediment/mm nederbörd) befanns vara konstant under sommaren i motsats till studerade vinterperioder. Sedimentkoncentrationerna av Cu och Zn var förhöjda med avseende på riktlinjer för känslighet hos sedimentlevande organismer samt för återanvändning av slam.

Resultaten visar att hela avrinningsvolymen måste behandlas. Reduktion av SS samt huvuddelen av metallerna, vilka visade god korrelation till det partikulära materialet, kan skattas utifrån en specifik ytbelastning (m/h). Reduktion av lösta metaller kan ske med granulerad masugnsslagg, dock bör långtidsstudier genomföras. Vidare visar studien på möjligheten att nyttja kontinuerlig turbiditetsmätning för en effektiv och praktisk processkontroll i reningsanläggningar för liknande avrinningsområden med hög trafikbelastning. Resultaten av arbetet kan användas för bedömning och optimering av vägdagvattenbehandling tillsammans med existerande designmetoder och modeller.

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LIST OF PAPERS

This thesis is based on the following papers that are referred to by their corresponding Roman numerals and can be found in appendix 1-6.

- I Hallberg M, Renman G. (2006) Assessment of suspended solids concentration in highway runoff and its treatment implication, Environmental Technology, 27: 945-950
- II Hallberg M, Renman G. (2007) Suspended solids concentration in highway runoff during summer conditions (Submitted to Polish Journal of Environmental Studies)
- III Hallberg M., Renman G., Lundbom T. (2007) Seasonal variations of ten metals in highway runoff and their partition between dissolved and particulate matter, Water, Air and Soil Pollution, in press (Published on-line Dec. 2006)
- IV Hallberg M., Renman G. (2007) Treatment of road runoff with sedimentation – Estimation of total suspended solids removal and the effect of seasonal conditions (Manuscript)
- V Hallberg M., Renman G. (2007) Removal of heavy metals from road runoff by filtration in granular slag columns (Manuscript)
- VI Hallberg M., Renman G. (2007) Seasonal generation and characteristics of sediment in a stormwater pond (Manuscript)

ABSTRACT

In highly urbanised areas, the existing land use may restrict the building of extensive ponds or wetlands for removal of particulate and dissolved pollutants from road runoff. To optimise treatment facilities for road runoff, treatment conditions must first be assessed. Field studies were conducted in three separate watersheds along the same highway. Based on the EU Directive (1991/271/EEC) maximum of 60 mg/l total suspended solids (TSS) in wastewater, it was found that treatment of the total runoff volume was necessary. The concentrations of dissolved Al, Cd, Co, Cr, Mn and Ni were significantly higher in winter compared with summer, but no significant difference was found for Cu, Pb and Zn. Total concentration of metals showed a good correlation to TSS (r^2>0.75). It was possible to estimate the reduction in TSS using an empirical model from the case study at one of the field sites. It was also possible to remove dissolved heavy metals at surface loads 5 to 10 times higher than in previous laboratory studies using a fixed filter bed of blast furnace slag. The mass concentration of metals (mg/kg) in particulate material varied between seasons. Sediment generation (mg sediment/mm precipitation) was found to be constant during summer. The field studies showed that turbidity measurement could be used for process monitoring and controlling treatment of road runoff. The findings of this study could be used to evaluate watersheds where traffic is the dominant source of pollutants, together with existing design methods to optimise treatment systems.

Keywords: Dissolved matter; filter bed; particulate matter; sedimentation; suspended solids

INTRODUCTION

Roads are an integral part of our infrastructure, providing crucial transport and communication links in the urban and rural environment. However, they are also significant pollution sources, in the case of stormwater for suspended solids, metals and organic contaminants.

The adverse effect of stormwater pollution on the receiving water environment and the need for treatment was recognized in the 1960s. The implementation of the Water Frame Work directive in the European Union in 2005 underlined the focus on pollutant control measures for stormwater.

Stormwater displays attenuated variations in quality and hydraulic behaviour during runoff events. The pollutant content has an affinity to the particulate matter but elevated concentrations of e.g. dissolved metals can be found. The contaminants' affinity to the particulate matter has rendered the treatment in different types of sedimentation ponds effective.

The performance of the treatment plant depends on the process control and handling of removed pollutants such as in wastewater treatment the sludge, or in the case of runoff, accumulated sediments.

The key design criteria for any treatment system depend primarily on the pollutant concentration and the partition between the particulate matter and the dissolved matter. This and the knowledge of the hydraulic transport patterns of pollutants provide the basis for a successful design of a stormwater treatment facility.

One generally used design method is an empirical method based on desired reduction efficiency of particulate material as a function of the relation between a permanent volume in the pond and the average runoff volume (Schueler, 1987).

The basis for empirical design methods, as well as for design models is the need for relevant input data. This can be obtained by laboratory experiments, field trials, and follow up of existing treatment system.

The extensive use of ponds for stormwater handling and treatment has over the years generated extensive field data from grab samples to extensive monitoring by flow proportional sampling (e.g., Pettersson, 1999). Applying the data, as part of a design tool, the variations in sampling conditions, stormwater quality, and treatment systems will ensure corresponding uncertainty when seeking to optimise the runoff treatment facility. In rural areas and urban areas where land use allows for construction of extensive ponds, this is less of a problem.

In highly urbanised areas, where the need for treatment is emphasised and it is practically and economically exigent to erect treatment facilities, specific design data are necessary to optimise and minimise the land use for the treatment plant.

A management strategy for reducing the size of the runoff treatment facilities is the use of "first flush" as design criteria to contain and treat only the most polluted volume during a runoff event. However, no unified definition exists and several studies have shown that the mass transport behaviour varies significantly even between similar catchment areas during comparable runoff events (e.g., Deletic, 1998; Lee and Bang *et al.*, 2002). The function of the sedimentation unit is of course essential for the treatment of runoff. However, sedimentation will not remove dissolved pollutants. In road runoff elevated concentrations of dissolved metals can be found. A reduction of dissolved metals could be carried out with fixed reactive filter beds. This would be pre-conditioned by a good removal of particulate material before the filter unit. Furthermore, the impact of road salt (NaCl) on the filter removal process is of interest. Elevated concentrations of the monovalent positive sodium ion could interfere with the sorption process of the divalent and trivalent positive metal ions. Very few field studies have been performed to assess the use of reactive filter media for treatment of road runoff when road salt is used during winter.

In Sweden, the dominating treatment system for stormwater is wet ponds. However, monitoring and follow up of the treatment facilities are rare (Lundberg *et al.* 1999). Usually, no monitoring equipment is installed and no provisions are made for future fitting of water sampling equipment when the stormwater treatment plant is built. This, in combination with the cost for executing a monitoring program, decreases the motivation for evaluating the treatment process.

Over 400 stormwater treatment plants have been built for the Swedish public roads and the majority of them are ponds (Starzec *et al.* 2005). In addition, a study of the sediments in 26 selected runoff treatment ponds suggested that the pollutant removal capacity was not optimal (Starzec *et al.* 2005).

Several studies of road runoff quality have been carried out (Asplund *et al.*, 1982; Lygren 1984; Hvitved-Jacobsson and Yousef, 1991; Sansalone and Buchberger, 1997a; Sansalone and Buchberger 1997b; Westerlund *et al.*, 2003; Polkowska *et al.*, 2005; Westerlund and Viklander, 2006).

Important data for removal efficiencies in ponds are continuously elaborated on. However, field studies of the sedimentation behaviour of road runoff, especially in regard to elevated salt concentrations, are rare and are needed as a complement to prevailing design methods and models. In addition, removal techniques for dissolved metals, foremost in winter, needs to be addressed by *in situ* studies with reactive filter media.

Knowledge of sediment accumulation rates and characteristics of sediments constitutes vital understanding for the successful operation and maintenance of stormwater treatment systems.

Continous measuring equipment for the monitoring and operation of stormwater treatment facilities are of interest. A parameter of interest would be TSS in regard to the affinity of the pollutants to the particulate material

OBJECTIVES AND SCOPE

The objective was to study the treatment conditions for contaminant removal in road runoff. This was carried out by field studies. The field work objectives were to study (i) the transport of total suspended solids (TSS) during runoff events (ii) the mass concentration (mg/kg) of particulate bound metals and the dissolved metal concentration (μg/l) in road runoff, (iii) the sedimentation behaviour of road runoff, (iv) *in situ* removal of dissolved metal from road runoff and (v) road runoff sediment generation and characterisation.

Fieldwork was executed in three separate watersheds, all dominated by a major motorway with an annual average daily traffic (AADT) exceeding 120,000 vehicles. The field studies included the summer and winter periods between October 2004 and December 2006. In winter the effect of studded tyres used on vehicles and road salting (NaCl) were of particular interest.

URBAN RUNOFF

Urban runoff is known to have an adverse impact on surface waters (USEPA, 2005). In 1996, an extensive investigation was initiated to elaborate on a best management practice for runoff in the heavily urbanised area of Stockholm, Sweden (Stockholm Vatten 2000; Stockholm Vatten, 2001a; Stockholm Vatten, 2002). It was proposed to classify runoff into three concentration ranges: low-, intermediate- and high concentration level (Table 1).

Based on the classification (Table 1) and the recipient the need for treatment of the stormwater can be assessed. For stormwater with low pollutant concentrations the guidelines (Stockholm Vatten, 2001a) treatment was not deemed necessary. Runoff from roads with an AADT exceeding 30,000 vehicles as rule needed treatment according to the guideline.

ROAD RUNOFF –PROBLEM FORMULATION

The ambient air quality and the traffic environment will govern the pollutant composition of road runoff.

The atmospheric deposition in urbanised areas is significantly influenced by anthropogenic sources such as industrial activities as well as traffic (Lopez *et al.* 2005). Studded tyres and/or the use of traction sand have a major impact on the pollutant loads during winter. The use of studded tyres in winter will increase the wear of the pavement dramatically (Jacobson, 1994; Jacobson and Hornvall, 1999; SLB 2004; Gustafsson *et al.*, 2005). Experimental studies indicate that traction sand has greater impact on the generated airborne particles than studded tyres alone (Kupiainen *et al.,* 2003).

The contaminant load in road runoff is affected by the traffic density (Asplund *et al.*, 1982; Hoffman *et al.*, 1985; Barrett *et al.*, 1998a; Hares and Ward, 1999). Traffic related road pollutants are generated from abrasion of the road surface, tyres and brake linings as well as leakage of hydrocarbons and residues from combustion (e.g. Lygren *et al.*, 1984; Kobriger and Geinopolos, 1984; Muschak, 1990; Hewitt and Rashed, 1990; Takada *et al.*, 1991; Lindgren, 1998; Legret and Pagotto, 1999; Bäckström *et al.*, 2003; Grant *et al.*, 2003; Folkeson, 2005).

The contaminants are deposited on roadway surfaces, median areas and right-of-ways from moving vehicles, stationary constructions and atmospheric fallout. The magnitude and pattern of accumulation appear to be a function of the roadway pavement and grade, traffic volume, maintenance activities, seasonal characteristics and adjacent land use (Hvitved-Jacobsen and Yousef, 1991).

Pollutant characteristics

The pollutants in road runoff are particulate and dissolved but tend to show an affinity to the particulate material (Xanthopolus and Hahn, 1990; Urbonas and Stahre, 1993;

Table 1 *Classification of runoff according to Stockholm Vatten 2001a LowC = Low Concentration, IntC = Intermediate Concentration, HigC = High Concentration*

	LowC	*IntC*	*HigC*
TSS (mg/l)	*<50*	*50-175*	*>175*
TotN (mg/l)	*<1.25*	*1.25-5.0*	*> 5.0*
TotP (mg/l)	*< 0.1*	*0.1-0.2*	*>0.2*
Pb (µg/l)	*< 3.0*	*3.0-15.0*	*>15.0*
Cd (µg/l)	*< 0.3*	*0.3-1.5*	*>1.5*
Hg (µg/l)	*<0.04*	*0.04-0.20*	*>0.20*
Cu (µg/l)	*< 9.0*	*9.0-45.0*	*>45.0*
Zn (µg/l)	*<60.0*	*60.0-300*	*>300*
Ni (µg/l)	*<45.0*	*45.0-225*	*>225*
Cr (µg/l)	*<15.0*	*15.0-75.0*	*>75.0*
Oil (mg/l)	*< 0.5*	*0.5-1.0*	*>1.0*
PAH (µg/l)	*< 1.0*	*1.0-2.0*	*>2.0*

Shinya *et al.*, 2000; Hallberg *et al.*, 2007). The particle size distribution in road runoff is usually below 100 μm (Roger *et al.*, 1998; Durand *et al.,* 2004; Stockholm Vatten 2001b; Hallberg, 2006). The particles with a diameter below 50 μm have been found to exhibit cationic exchange capability (Roger *et al.*, 1998). Correlation between some metals and organic matter has been indicated in particle fractions between 0.45 μm and 20 μm in urban runoff (Characklis and Wiesner, 1997). The specific surface area of the particles in road runoff increases with decreasing particle size (Sansalone *et al.*, 1998) and the particulate concentration (mg/kg) of metals increases with decreasing particle size (Sansalone and Tribouillard, 1999).

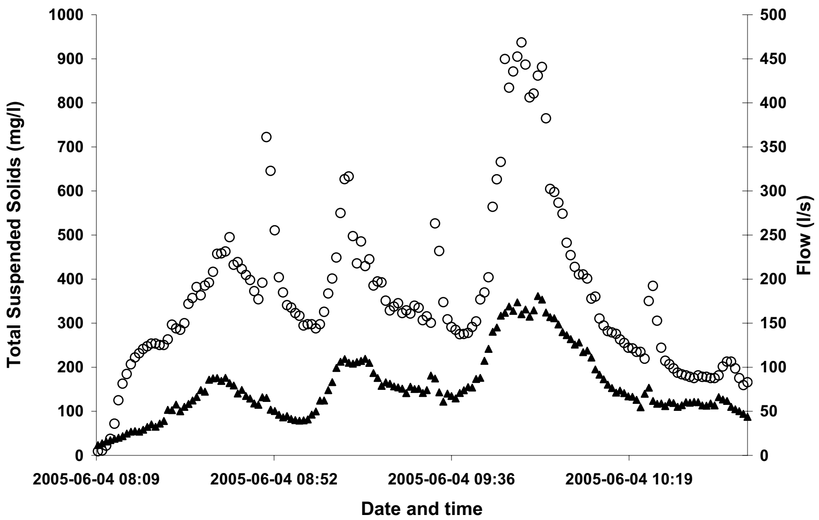

Figure 1. *Example of variations in TSS concentrations and flow during a runoff event in the Eugenia watershed (Paper I).* ○ = *TSS,* ▲ = *Flow*

During winter the pollutant loading will increase from roads (Lygren *et al.*, 1984; Westerlund *et al.*, 2003; Bäckström *et al.*, 2003; Westerlund and Viklander, 2006; Hallberg and Renman, 2006; Hallberg *et al.,* 2007). The length of the winter period influences the increase in pollutant load by accumulation of pollutants in snow (Reinosdotter and Viklander, 2005). Snow accumulates heavy metals and their affinity to the particulate material increases in the runoff during winter compared to summer (e.g., Glenn and Sansalone, 2002; Sansalone and Glenn, 2002, Westerlund and Viklander, 2006). The use of road salt (NaCl) as a de-icing agent influences the pollutant load during winter. Salt also adversely effects the environment (Marsalek, 2003). In a study by the UK Highway Agency and UK Environmental Agency (UK Environmental Agency, 2003), it was concluded that metals were found in higher concentration following winter salting. A three-year study of car corrosion was executed between 1986 and 1988 in the island of Gotland and the town Västervik on the Swedish mainland (Hedlund, 1995). The two areas are in the same geographical vicinity on the east coast of Sweden and the Baltic Sea. On Gotland no road salt was used. Västervik used road salt. The cars that were studied belonged to the Swedish postal service. It was found that the cars driven on salted roads in the Västervik area displayed 2-3 times the corrosion damage compared to the vehicles driven in Gotland. Road salt is transported away during or shortly after being spread (Buttle and Labadia, 1999), which affects the wet exposure time and accordingly also the corrosion (Bertling, 2005) in the road environment.

The use of traction sand, de-icing agents, and studded tyres and winter road maintenance generates elevated pollutant loads from road surface and vehicles compared to summer conditions. The seasonal variations are emphasised and affects the composition of particulate material and dissolved matter. Thus it is important to study seasonal variation in regard to mass transport and treatment of road runoff.

Mass transport during a runoff event

The variations in flow and contaminant concentrations are emphasised during a runoff event (Fig. 1).

One descriptor of mass transport in stormwater is the first flush. First flush is signified by higher concentrations of pollutants in the initial part of a runoff event. Over time, the concept of first flush has been applied as a design criterion using arbitrary precipitation to estimate the appropriate capture volume (Barrett *et al.*, 1998a). The relationship between the cumulative and total mass load (kg/kg) and the cumulative and total runoff volume (m^3/m^3) during a runoff event has been used to describe and study first flush for pollutants (Urbonas and Stahre, 1993; Bertrand-Krajewski *et al.,* 1998; Deletic, 1998; Barbosa and Hvitved-Jacobsen, 1999; Lee and Bang, 2000; Lee *et al.,* 2002; Li *et al.*, 2005).

The variations in pollutant mass transport are notable even in comparable watersheds and runoff events. Traffic-related pollutant load increases with traffic density.

The highly intermittent operating pattern of a runoff treatment facility calls for good knowledge of runoff properties to enable an optimisation of the treatment strategy. This is particularly true for urban road runoff. Hence there is an interest in assessing the possibility to optimise treatment facilities by a partial treatment of the runoff volume. This could be carried out by studying the partial event mean concentration of a particular pollutant during individual runoff occasions in regard to a discharge demand, as an alternative to first flush criteria.

Pollutant removal

The particle size distribution and the density of the material determine the gravimetric removal of particulate matter. Street sweeping removes coarser particulate matter (Grottker, 1990; Stone and Marsalek, 1996: German, 2003). Coarser particles in road runoff are trapped in the gully pots of the drainage system, at least until intense runoff can erode and transport them away. Finer particles and colloidal and dissolved material in the runoff are transported through the pipe network of the drainage system.

Treatment practices most commonly used for stormwater are sedimentation ponds, infiltration ponds and also different types of vegetated filter strips.

These types of treatment methods remove the particulate pollutants and to some extent colloidal and dissolved matter. An alternative to infiltration for removal of colloidal and dissolved material is the use of filters. These are not common, but have been studied in laboratory experiments and pilot-scale field tests (e.g. Färm, 2003a).

Gravimetric separation of particles

The particle-fluid separation processes are difficult to describe by a theoretical analysis, mainly because the particles involved are not regular in shape, density and size (AWWA, 1999). However, for most theoretical and practical computations of settling velocities the shape of particles is assumed to be spherical (AWWA, 1999).

Settling of discrete particles

When the concentration of particles is small, each particle will settle discretely, i.e. unhindered and independently of other particles in the solution. Starting from rest, the velocity of a single particle settling in a liquid will increase where the density of the particle is greater than the density of the liquid. Acceleration continues until the resistance to flow through the liquid, or drag, equals the effective weight of the particle. Thereafter, the settling velocity remains essentially constant.

This velocity is called the terminal settling velocity, v_t (AWWA, 1999). For laminar flow and Reynolds number between 10^{-4} and 0.2 the terminal settling velocity for a spherical particle can be calculated according to Stokes equation for laminar flow (1):

$$v_t = \frac{g(\rho_p - \rho)d^2}{18\mu} \qquad (1)$$

where g is the gravitational constant of acceleration, ϱ_p is the density of the particle, ϱ the density of the liquid, d is the diameter of the particle and μ is the dynamic viscosity of the liquid.

In an ideal sedimentation tank where the flow is evenly distributed over tank surface a particles settling velocity (m/h) is identical with the surface load (m/h) i.e. the flow (m^3/h) divided by the surface area (m^2) of the tank (m/h) (AWWA, 1999). The sedimentation is accordingly not dependent on the depth of the sedimentation basin. This can be used for assessing the surface load by carrying out column experiments.

In a sedimentation column experiment the settling velocity of a particle, v_s ,is measured in a water column with a certain height, h_{wc}, over time, t_{sed},(2)

$$v_s = \frac{h_{wc}}{t_{sed}} \qquad (2)$$

The settling velocity, v_s, is accordingly an estimation of the surface load needed for removing particles with a settling velocity greater than v_s .

In stormwater the density of the particles and the particle size distribution can vary (Bondurant *et al.*, 1975; Urbonas and Stahre, 1993; Cristina *et al.*, 2002). Particles densities are also influenced by formation of flocs (Krishnappan *et al.*, 1999). The runoff water temperatures also influence the sedimentation properties during treatment. This suggests that assessment of sedimentation behaviour of stormwater is site specific and less geographically transferable. Studies of the sedimentation removal process have been carried out by laboratory column experiment but although predominantly by monitoring of inlet and outlet concentration of pollutants from many runoff treatment facilities.

Sedimentation column experiment

Whipple and Hunter (1981) studied runoff from five urban watersheds. The result of the study was based on one composite sample from the each of the five watersheds. The results emphasised differences in settleability between the individual studied pollutants and the watersheds. Randall *et al.*, (1982) studied stormwater from three watersheds with impervious surfaces and found that the initial concentration of total suspended solids (TSS) correlated well with relative removal (%) of TSS. These types of sedimentation studies are not common and are often carried out after transportation and storage of the collected stormwater. These handling procedures could affect the water quality and accordingly the settleability of particulate matter in a study.

In situ sedimentation studies over the winter and summer seasons could complement existing design tools for runoff treatment facilities.

Design of runoff sedimentation units

The most used design criteria for wet ponds and detention basins are either planning-level design for calculating the required facility area or more detailed design of the permanent or detention volumes in these facilities.

The permanent water area can in a planning-level be designed as a certain share of the reduced watershed area (=area*runoff coefficient) (Larm, 2000). There have also been compiled empirical data of estimated reduction efficiency as a function of basin relative area, i.e. pond area divided by the reduced watershed area as compiled in Walker (1987) and Pettersson (1999) and the database of the stormwater model StormTac (Larm, 2007).

According to Guo and Urbonas (1996), the percentage of stormwater runoff volume, or the number of runoff events, captured is a key factor for design.

One generally used (Schueler, 1987) method for designing wet ponds is an empirical method based on desired reduction efficiency as a function of the relation between permanent pool volume and average runoff volume and are used in design models (e.g. Larm, 2007)

Design methods appeal to extensive data from different types of treatment facilities and watersheds. Important parameters that affect the reduction efficiency are the inlet concentration of pollutant, the shape of the facility including the location of inlet and outlet, the outlet construction, and the detention volume and sedimentation time for particles (Pettersson, 1998; Vikström *et al*, 2004). In addition, seasonal variations in pond performance also have to be considered (Semadeni-Davies, 2006). The variations will influence the accuracy when used as empirical data for design models.

In Sweden, Stockholm treatment facilities for road runoff use a batch-wise operation procedure with a typical sedimentation time of 36 hours (Vägverket, 2007). However, very few studies have been carried out on the performance of the treatment plants.

Sedimentation is the most expedient method for treatment of runoff; however in urbanised areas land use may hamper the use of sedimentation facilities (Aldheimer and Bennerstedt, 2003). Restricted land use has led to construction of treatment facilities partly or completely underground (Stockholm Vatten, 2001b; Vägverket 2007) resulting in high investment costs. Additional knowledge in regard to sedimentation properties, in particular for highly polluted road runoff to optimise the treatment plant and minimise its land use are accordingly of interest.

Mechanical removal of pollutants

Mechanical removal of particles can be performed by vegetation in filter strips and grassed swales, as well as in ponds where vegetation has been established over time (e.g. Barrett *et al.*, 1998b; Hares and Ward, 1999; Bäckström and Viklander, 2000; Bäckström, 2001; Bäckström, 2003). Filter strips and grassed swales remove particles and also reduce the stormwater volume by infiltration. Constructed wetlands combine gravimetrical removal and mechanical filtration of particulate pollutants. Shutes *et al.* (1999) describe the design and function of constructed wetlands with respect to highway runoff. Hares and Ward (1999) and Sriyaraj and Shutes (2001) have elaborated on this. The required land use (m^2) to treated watershed area (ha) is considerably less for ponds compared with filter strips, grassed swales and wetlands (Larm, 2000). The use of filters with sand or other filter media for particulate removal is not common practice, but some laboratory studies and preliminary field studies have been performed (e.g. Tenney *et al,* 1995). The utilisation of porous road surfacing for retention of particulate matter has been studied by Pagotto *et al.* (2000). The use of filters for particulate removal from runoff would be questionable, in particular for road runoff as the elevated TSS levels could clog the filter media. Backwashing of the filter would be necessary, a procedure that renders it less practical to use.

Reduction of colloidal and dissolved pollutants

Metal contaminants in road runoff have an affinity to the particulate material (e.g. Urbonas and Stahre, 1993; Shinya *et al.,* 2000; Glenn and Sansalone 2002; Reinosdotter and Viklander, 2005; Hallberg *et al.,* 2007). However, elevated dissolved concentrations of metals are not uncommon, (e.g. Charackis and Wiesner, 1997; Sansalone and Buchberger, 1997b; Gromaire-Mertz *et al.*, 1999; Sansalone and Glenn 2000; Hallberg *et al.*, 2007).

The use of biofilters for metal removal from highway runoff has been studied in laboratory trials (Lau *et al.*, 2000). Several low-cost sorbent materials for removal of heavy metals have been investigated (Bailey *et al.,* 1999) and these could be included in systems based on filter-bed techniques (Kängsepp *et al.,* 2003).

Some pilot scale tests with reactive filter media for removal of heavy metals from runoff have been conducted (Färm, 2003a; Hallberg and Renman, 2007; Renman *et al.*, 2007). Hallberg and Renman (2007) studied two reactive filter materials and showed good reduction for selected heavy metals. The surface load to the filter was low (0.06

m/h) and no increase in pressure drop was found during filtration. However, a negative impact of road salt was suggested. Dimitrova (2002) showed that the sorption capacity of Pb to slag filters was reduced at elevated concentrations of Na. Provided a reactive filter media with a good sorption capacity of metals, it is important to optimise the surface load to minimise the area needed for the filter in the treatment facility. A treatment step with reactive filter would be preconditioned by an effective removal of particulate material.

Further studies are of interest for applying reactive filter media for treatment of road runoff. Particular interest is the impact of road salt and feasible surface loads to optimise the size of the filter unit.

Sediment characteristics

The removal of pollutants in ponds results in a sediment accumulation. The build up of sediments is critical for the function of the pond since the sediments decrease the available volume for sedimentation and thereby increase the risk of resuspension. Several studies have elaborated on the accumulation rate of sediments and times in between emptying of sediments (Yousef *et al.*, 1990; Yousef *et al.* 1994a; Yousef *et al.*, 1994b; Marsalek, 1995: Yousef *et al.*, 1996; Marsalek and Marsalek, 1997; Färm, 2001; Färm, 2003b; Durand *et al.*, 2004). The sediment accumulation depends on the watershed and shape and size of the pond and intervals between emptying has been suggested between 10 years and 25 years.

If anaerobic conditions occur in the sediment, this will result in a drop in pH. At pH below 5, there is a risk for mobilisation of metals (Yousef *et al.*, 1990). Stone and Marsalek (1996) studied the bioavailability of metals in street sediment. It was found that Cu, Cd, Pb and Zn were predominantly in a bio available form. Durand *et al.* (2004) found that the mobility of metals in sediments from highway runoff in an increasing order was Cr ≤ Ni ≤ Pb ≤ Cu ≤ Zn ≤ Cd. Stead-Dexter and Ward (2004) showed an increased potential for mobility of metals (Cd, Cu, Ni, Pb, Zn) in road runoff sediments compared to freshwater sediments not affected by road runoff. The bioavailability increases with decreasing particle size (Preciado and Li, 2006).

The seasonal variations in pollutant characteristics and load in road runoff makes additional studies of sediment accumulation rates, particulate contaminant concentrations (mg/kg) and sediment particle size distribution, a concern. Such studies would be of importance for planning and design of road runoff treatment facilities.

STUDY SITES

The three study sites were located along the six-lane E4 motorway through Stockholm and were, from North to South, Eugenia, Fredhäll, and Lilla Essingen. The E4 motorway has an annual average daily traffic (AADT) load exceeding 120,000 vehicles and a speed limit of 70 km/h.

Eugenia

The E4 motorway including a 235 m long tunnel section dominates the watershed. The total asphalt surface is 54,000 m^2 of the watersheds total area of 67,000 m^2 (Table 2).

The watershed is divided into four separate areas in regard to piping network i.e. South West 1 (SW1), South West 2 (SW2), South East (SE) and North West (NW) (Table 2, Figs. 2-9). SW1 receives some runoff water

Table 2 *Description of the four parts of the catchment area.*

Catchment area	*Total area (m^2)*	*Asphalt surface (m^2)*	*Green areas (m^2)*	*Inclination (‰)*	*Main pipe diameter (mm)*	*Gully pot pipe diameter (mm)*
South West 1	*6,900*	*5,900*	*1,000*	*40*	*300*	*225*
South West 2	*26,000*	*21,000*	*5,000*	*20*	*400*	*225*
South East	*1,500*	*1,500*		*30*	*225*	*225*
North West	*32,600*	*25,600*	*7,000*	*20*	*400/500*	*225*
Sum	*67,000*	*54,000*	*13,000*			

from a pedestrian walk. SW2 includes the Solna Bridge that passes over the motorway and a parking lot from which the runoff is discharged via a sand trap to the piping network. SE includes runoff water from a park area and pedestrian walk. NW exclusively receives water from the motorway.

In 1991, a treatment plant was constructed and commissioned to reduce pollutant load from the watershed. After treatment, the stormwater runs into Brunnsviken, a small freshwater lake. The treatment plant, Eugenia, is located below ground and the runoff is transported by gravity to the intake chamber. The runoff then overflows to a step screen and passes two separate Parshall flumes before it discharges to the retention basin.

Fredhäll

The total drainage area is 13,700 m^2 and the road surface is covered with asphalt. The watershed includes a tunnel with a road area of 7,800 m^2 (Figs. 10-11). The recipient is part of the Lake Mälaren. To reduce the pollutant load from the runoff, a treatment plant was built and commissioned in 2003 (Fig. 12). The treatment plant, Fredhällsmagasinet, is located below the south tunnel entrance. The runoff is transported under gravity to the treatment plant from the bridge, tunnel and motorway at the north entrance to the tunnel. The runoff water enters the treatment plant's grit chamber and overflows via a Thompson weir to a sedimentation basin. Level sensors in the sedimentation basin are used for process control.

Figure 2 *Picture of the SW1 Catchment Area*

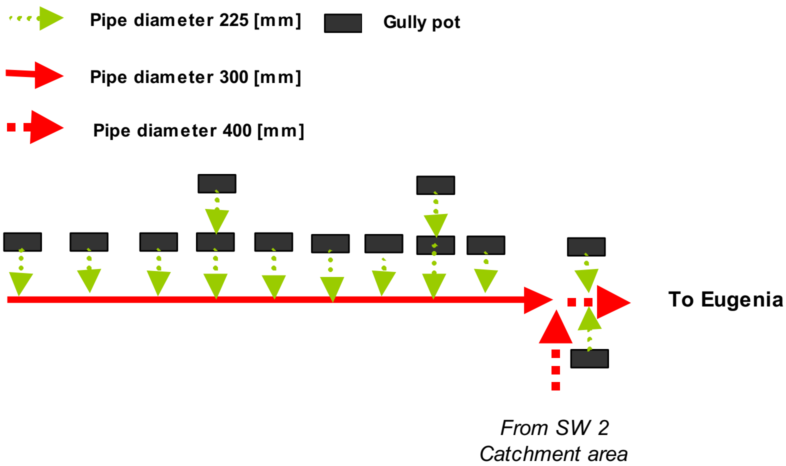

Figure 3 *Schematic layout of drainage system SW1 Catchment area*

Figure 4 *Eugenia catchment area SW2 (Picture taken from Solna Bridge)*

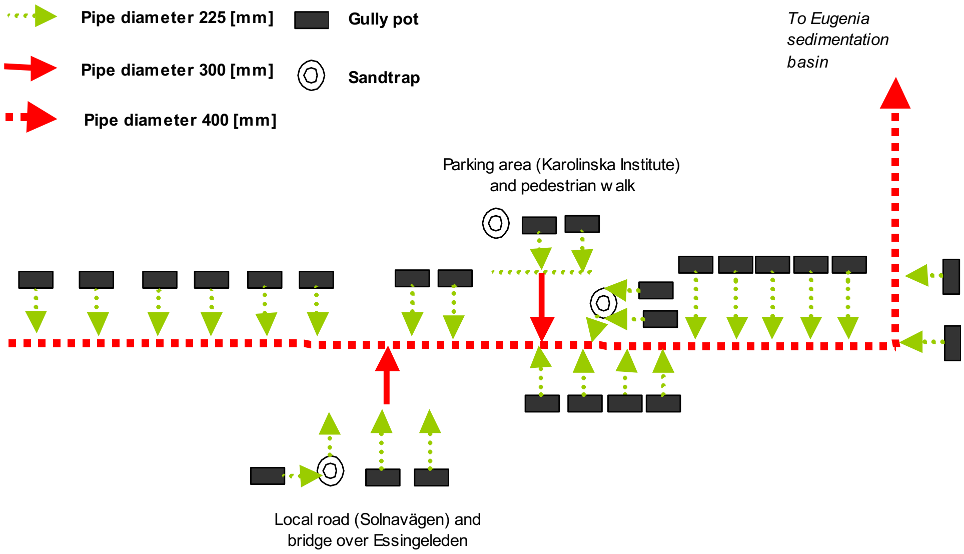

Figure 5 *Schematic layout of drainage system SW2 catchment area*

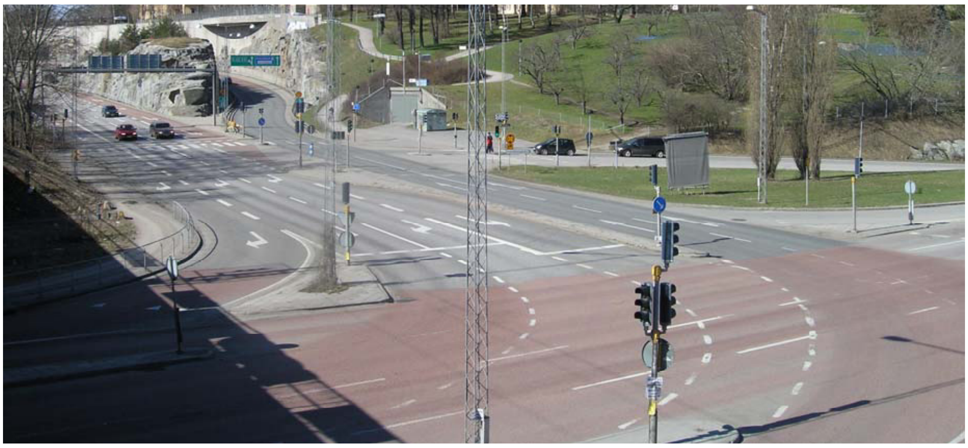

Figure 6 *Eugenia catchment area SE*

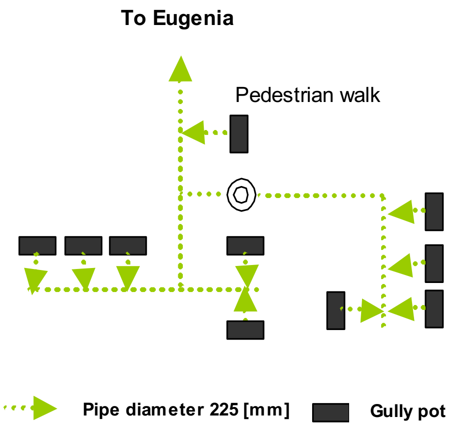

Figure 7 *Schematic layout of drainage system SE catchment area*

Figure 8 *Eugenia catchment area NW*

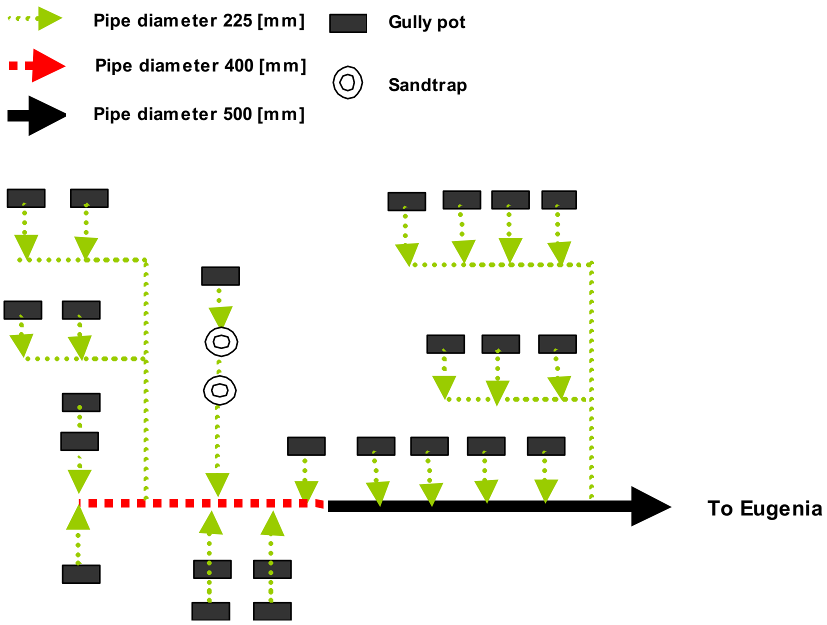

Figure 9 *Schematic layout of drainage system NW catchment area*

Figure 10 *Fredhäll catchment area (north bound traffic in left lane)*

Figure 11 *Fredhäll catchment area (north bound traffic in right lane)*

Figure 12 *Fredhäll treatment plant for stormwater*

Lilla Essingen

The watershed is located in the central area of Stockholm and is dominated by local roads and the E4 motorway (Table 3, Figs. 13-15).

Table 3 *Lilla Essingen watershed*

Description	*Total area (m^2)*	*Surface*
Highway	*3,800*	*Asphalt*
Road	*4,980*	*Asphalt*
Road	*600*	*Concrete*
Parking lot	*1,500*	*Asphalt*
Roof	*800*	*Tiles*
Green areas	*140*	*Grass*
Sum	*11,820*	

To reduce the pollutant load from runoff a pond was built in 2003 as part of a stormwater treatment system. The recipient for the treated water is Lake Mälaren. The pond is located under a motorway bridge that covers 80 % of the pond surface. The runoff is transported by gravity to the pond. The pond has a permanent volume of 150 m^3 and a maximum total volume of 200 m^3 with a minimum depth of 0.90 m and a maximum depth of 1.40 m. The pond has a maximum surface area of 3.8 % of the catchment area. In a study by Stockholm Water (2007) the reduction of TSS was found to be 93 %.

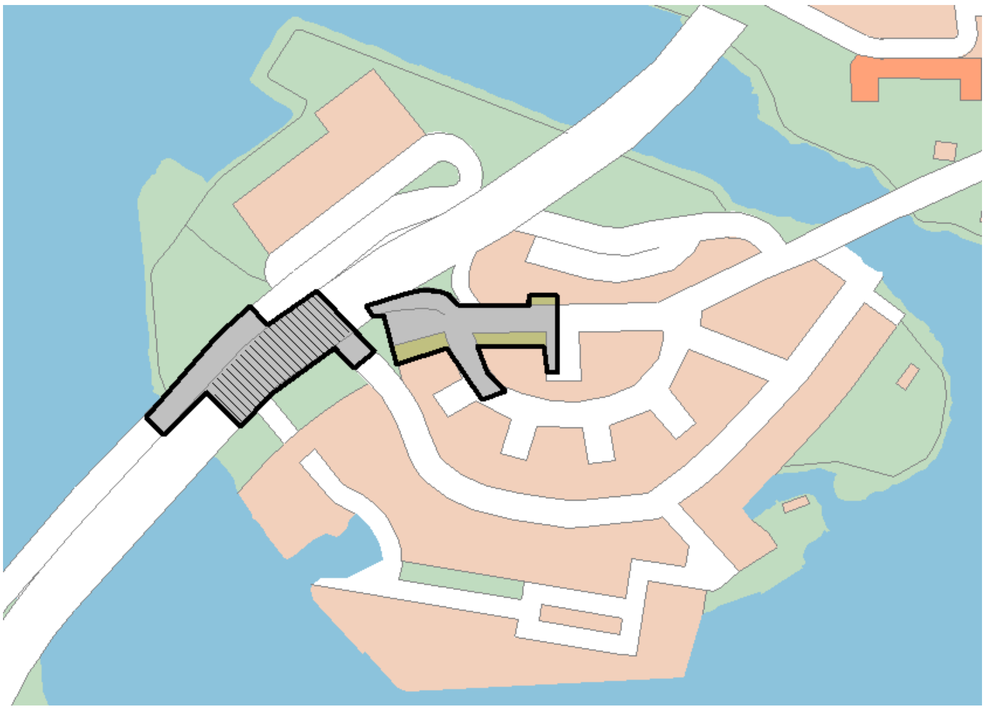

Figure 13 *The watershed at Lilla Essingen. The areas shaded grey shows the catchment area. The grey area marked with black lines identifies the heavily trafficated E4 motorway part of Essingeleden.*

Figure 14 *Lilla Essingen catchment area*

Figure 15 *Lilla Essingen catchment area*

EXPERIMENTAL SET-UP

Eugenia

Measurement of total suspended solids

Continuous measurement of total suspended solids was carried out using a Cerlic ITX suspended solids meter. The measuring wavelength for the instrument was 880 nm. Cleaning of the measuring probe was executed automatically with compressed air. Calibration of the instrument was achieved by correlating the analysed TSS concentration to the registered value from the Cerlic ITX instrument.

Flow measurement

Flow measurement from 1 l/s to 600 l/s was performed with two Parshall flumes. Flows between 1 l/s to 20 l/s were registered with a Chanflo Open Channel (Danfoss) flowmeter (0 m to 0.3 m) with a Sonolev sensor (100 KHz). Flows between 20 l/s to 600 l/s were registered with Chanflo Open Channel (Danfoss) flowmeter (0 m to 1 m) with a Sonolev sensor (100 KHz).

Precipitation and ambient air temperature measurement

A rain gauge was located 6 m above the ground level in the central part of the watershed. The rain gauge registered every 0.5 mm rain. The rain gauge was without heating capabilities thus making registration of precipitation at temperatures below and around 0°C uncertain.

Conductivity and water temperature measurements

To measure conductivity a Campbell Scientific 247 Conductivity and Temperature Probe was used. The cell constant (K_c) of the conductivity sensor was 1.399 and the measuring range was 0.005 mS/cm to 7.5 mS/cm. The temperature sensor used a Betatherm 100K6A1 thermistor and the measuring range was from 0°C to +50°C.

Data collection

All sampled data from the on-line measurements were collected with a Campbell Scientific CR10X data logger.

Sampling of runoff water

For the case study a system for collecting the runoff samples for the sedimentation study was constructed. The experimental set-up consisted of two separate parts i.e. the runoff collection system (RCS) and the sampling tank (ST) (Fig. 16). The RCS consisted of six individual vessels each with a diameter of 550 mm. The ST was made up of a single tank. The settling studies were carried out in the RCS. The ST was used for sampling for analysis of TSS, particles sizes, Na and Cl. The RCS allowed for consecutive samples of runoff. The purpose of the RCS was to collect samples with different concentrations of suspended solids to assess settling. By using the ST for sampling of TSS, sodium and chloride less than 3 % of the total volume of a RCS vessel was used for assessment of sedimentation.

Sampling of runoff could be started automatically or manually. A timer was used to register the sampling period. Automatic start of sampling was initiated by a signal from the water treatment plants programmable logic controller (PLC) when the incoming flow exceeded 3 l/s, indicating the start of a runoff event. Two Flygt pumps of type SXM 3 and Flygt SXM 2 were placed in the intake chamber and used for pumping the water to the sampling system. The pumps were started at the same time and the flow from the pumps was adjusted so that the filling time for the RCS and ST was the same. The runoff water filled the vessels of the RCS consecutively by use of a floating switch in combination with a check valve mounted in the individual vessels. The same type of floating switch was used in the ST.

Two valves controlled the flow to the RCS and ST. All material was plastic with exception for the two pumps of stainless steel. Sampling of turbidity during sedimentation was executed in the RCS from seven taps on the individual vessels (Fig. 16). The distance between the taps (centre to centre) was 100 mm. Sampling for analysis of turbidity was executed from the sampling taps of the ST (Fig.16).

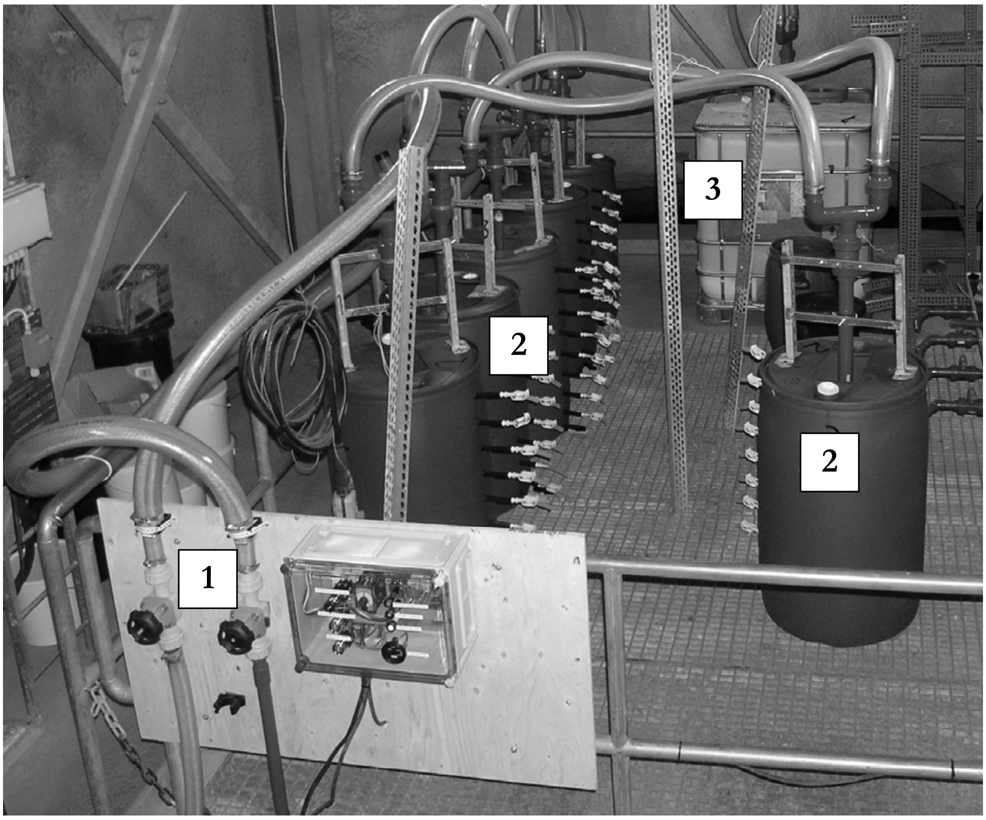

Figure 16 *Collection system for runoff during the experiment, showing the two valves to set the flow (1), the six sedimentation vessels in the RCS (2) and sampling tank (3)*

Turbidity measurements

A HACK 2100P ISO turbidity meter was used for turbidity measurements. The instrument complies with EN ISO 7027. The operating wavelength of the instrument was 860 nm. The measuring range was from 0 FNU to 1,000 FNU with a resolution of 0.01. The sample volume was a minimum of 15 ml. Before collecting the water sample, the tap was open for a minimum of 5 s and kept open until the sample vial had been washed with four volumes of sample water. The turbidity meter function was regularly checked using standard solutions of 0.1 FNU, 20 FNU, 100 FNU and 800 FNU.

Fredhäll

On-line measurements

Measurement of total suspended solids

Continuous measurement of total suspended solids was carried out using a Cerlic ITX suspended solids meter. The measuring wavelength for the instrument was 880 nm. Cleaning of the measuring probe was executed automatically with compressed air. Calibration of the instrument was achieved by correlating the analysed TSS concentration to the registered value from the Cerlic ITX instrument.

Conductivity

The on-line measurements of conductivity were made with a Jumo dTransLf01 type 202540. The measuring range of conductivity was between 0 mS/m and 2,000 mS/m and the cell constant (K_c) was 1.00.

Continuous flow measurement

Flows between 1 m^3/h and 60 m^3/h were measured using a Thompson weir in combination with a pressure gauge (Cerlic FLX) with a measuring range from 0 m to 1 m.

Precipitation

A rain gauge was located 10 m above the ground level in the central part of the watershed. The rain gauge registered every 0.2 mm of rain and was equipped with sensors so that the temperature in the collecting part of the gauge did not fall below 2°C when the outside temperature was below 0°C. The gauge had a capacity of 6 mm/min.

Level measurement in sedimentation basin

To measure the level in the sedimentation basin, a Swedmeter submersible DS/mA pressure probe was used with an operating range of 0 m to 5 m. The DS/mA probe had automatic temperature compensation.

Data collection

All sampled data from the on-line measurements was collected in the operating panel (ABB type 245B).

Water sampling

The conditions for sampling were increased flow, increased conductivity and increased TSS. Flow proportional sampling was executed when the registered flow exceeded 1 m^3/h and sampling was carried out every 4 m^3 except for the sampling event on 15 December 2004 when the sampling interval was 1 m^3. If the conductivity increased by 30 % or the TSS exceeded 200 mg/l, sampling was performed with an interval of 1 h.

Water sampling equipment and procedure

A CO/TECH 750 water-sampling pump was placed at the end of the grit chamber, before the Thompson weir. Water was pumped through a sampling loop and back to the grit chamber and discharged before the Thompson weir. The material of the piping and parts of the sampling loop were made from PVC. The retention time in the sampling loop was less than 5 s. Water was extracted from the loop using an ISCO 3700RF sampler. To rinse the sampling tubing, flushing was executed with three sampling tube volumes. The time for the rinsing cycle was 30 s and the sample volume was 800 ml. The sampling pump was started every six hours for 60 s to flush the sampling loop during the study period.

Pilot trials with reactive filter media

The pilot plant system consisted of two identical lines. The layout of the pilot plant was governed by available space in the Fredhäll treatment plant. A line consisted of three vessels. Each vessel had a total volume of about 220 l. The runoff water was siphoned from the sedimentation basin of the Fredhällsmagasinet to the two vessels before the filter columns. Road salt (NaCl) was added to one line. Sampling was executed in runoff immediately after the vessels were filled. Sampling after salt addition and mixing was carried out after approximately two hours to dissolve the salt. The sedimentation time for the runoff used in the pilot trials was never less than 36 hours.

Vessel 1 and 2 was separated with a valve. The runoff was always pumped from vessel 1 to the filter columns. The pilot trial was carried out in two steps. In the first step, the valve was closed and runoff was pumped only from vessel 1. Road salt was added in vessel 1. When the level was low in vessel 1, the pump was stopped and sampling of the filtrate was carried out. The filtrate vessels were emptied and thoroughly cleaned. In the second step, road salt was added to vessel 2. After about two hours the valve was opened and pumping was performed from the now communicating vessels 1 and vessel 2. After the pump was stopped, sampling of the filtrates and measurement and sampling of the remaining volume in vessel 1 and vessel 2 of the lines were carried out.

The runoff water was discharged to the top of the filters with a laboratory peristaltic pump. Because the outlet from the filters was 100 mm above the top of filter material the filtration was carried out under saturated conditions. This also allowed for an even hydraulic distribution to the filters. By measuring the distance from a fix point on the filter column to the water level, the pressure drop over the filter material was monitored.

Lilla Essingen

Sediment traps

Six sediment traps were constructed and placed in the pond (Fig. 17). The traps (STr) were made solely from PVC to avoid metal contamination. The STr consisted of two parts, the holder and the collection vessel for the sediments (Fig. 17). The inside diameter of the collection vessel was 104 mm with a height of 500 mm. The total height of the STr was 850 mm. The bottom plate of the holder was used to place weights to fix the position of the STr. The collection vessel was removed from the holder when the collected sediment was retrieved. The holder was never moved from its position during the study.

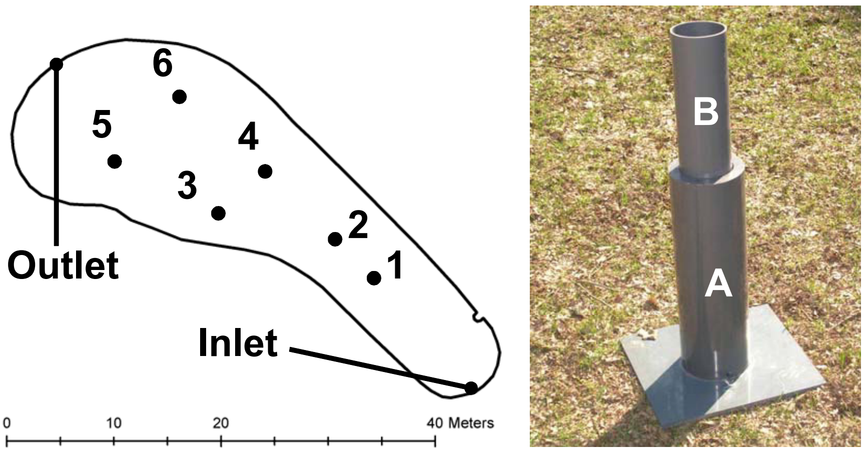

***Figure 17** Placement of sediment traps (STr 1 to STr 6) and the sediment trap with the holder (A) and removable collection vessel (B)*

PAPER OVERVIEW

An overview of appended papers is given below.

Paper I

The possibility to treat only a partial runoff volume has been suggested with reference to first flush behaviour. A criterion for first flush is given by Bertrand-Krajewski *et al.* (1998). Other definitions exist but Bertrand-Krajewski *et al.* (1998) is stringent in regard to a minor portion of the total volume, less than 30 % carrying a major fraction, exceeding 80 % of the total pollutant mass. The first flush concept as described by Bertrand-Krajewski *et al.* (1998) is a relative measure for assessment of the mass transport during a runoff event. The methodology used to describe first flush can be employed describing the partial event mean concentration (PEMC) for the course of the runoff event. This could be used to assess a partial treatment of the total stormwater volume.

In Paper I, the EU Directive 1991/271/EEC requirements for discharge from a wastewater treatment plant for TSS (≤60 mg/l) was used to evaluate a partial capture of the runoff volume during winter conditions. The partial event mean concentration (PEMC) was calculated from the end of the runoff event to determine the PEMC of the latter runoff volume for the selected discharge demand and consequently the need for treatment. The average TSS EMC for the studied events was 670 mg/l and the average total runoff volume was 315 m^3.

It was found that for the majority of the studied runoff events the TSS PEMC was over 60 mg/l for the duration of the runoff events. None of the studied runoff events fulfilled the criterion for first flush according to Bertrand-Krajewski *et al.* (1998). Paper I notes that the capture of the total runoff volume is necessary in this type of watershed in winter when studded tyres and road salt are used. However, it is necessary to study the mass transport for summer conditions in regard to design of treatment facilities. This was carried out in Paper II.

Paper II

The study in Paper II used the same methodology as described in Paper I to study the possibility to treat a part of the runoff volume during summer.

The average TSS EMC was 280 mg/l, and the mean total volume for the studied runoff events was 530 m^3. Paper II shows that for most of the runoff events the TSS PEMC were greater than 60 mg/l. At only one runoff event a significant first flush was found using the criteria identified by Bertrand-Krajewski *et al.* (1998). The result shows that the design criterion for stormwater treatment facilities in similar watersheds must consider the entire runoff volume during summer.

Paper III

In winter, the asphalt surface is exposed to studded tyres and salt is regularly used as a de-icing agent. In addition, snow removal and the accumulation of pollutants in snow are factors that account for seasonal variations in the runoff quality during winter compared to summer. Seasonal variations of TSS and ten selected metals were studied in Paper III. The TSS EMC, dissolved metal concentration, metal mass concentration (mg/kg) of the particulate material, and the correlation between TSS and total metal concentration were studied. The TSS EMC was higher during the winter for the studied stormwater events. The dissolved concentration for Al, Cd, Co, Cr, Mn, and Ni was higher for winter compared to the summer ($p<0.01$). There was no significant difference between the sampling periods with regard to the dissolved part for Cu, Pb and Zn. A higher dissolved part during summer was found for Fe ($p<0.01$).

In the summer, Cd, Cr, Cu, Mn, Ni, Pb, and Zn displayed elevated content in the particulate matter ($p<0.01$) as well as Fe ($p<0.05$). Particulate Al and Co during the summer was lower than during the winter ($p<0.01$).

The total metal concentration showed good correlation to the TSS concentration. In the winter, the linear correlation for the total metal concentration and TSS showed a correlation factor (r^2) greater than 0.95 for all metals except for Cd ($r^2=0.92$). In summer, all metals showed a good correlation ($r^2>0.90$) with the exception of Cd, Co, Cr, and Pb having correlation factors of 0.82, 0.77, 0.84, and 0.89 respectively. The analysed TSS range for the winter was 13 mg/l to 4,800 mg/l and 14 mg/l to 520 mg/l for the summer.

Paper III shows that a successful treatment of the studied motorway runoff can be accomplished by sedimentation and that seasonal variation in removed sediment concentrations (mg/kg) can be expected. However, the elevated concentrations of dissolved metals during winter, e.g. Zn, must be considered with regard to discharge demands and appropriate treatment method.

Paper IV

Paper III suggests that sedimentation is an effective treatment method for the studied road runoff. Road salt and studded tyres are two parameters that have a notable impact on water quality and the particle size distribution in road runoff in winter. Paper IV studied the sedimentation behaviour of runoff during winter and the subsequent summer period.

Paper IV showed that the sedimentation behaviour can be estimated by the concentration of total suspended solids in the studied runoff for the winter and summer period. However, a significant difference in the sedimentation properties was shown for elevated levels of NaCl ([Cl^-] > 2,000 mg/l). The results give a simple model for estimating the outlet concentration of TSS for a specific inlet concentration of TSS. This model provides a design tool to optimise design of stormwater treatment facilities in similar watersheds.

Good correlation was noted for turbidity and TSS, which concurs with the findings in Paper I and Paper II.

The possibility to indirectly measure the TSS concentrations by turbidity provides a mean for a practical and undemanding process control for stormwater treatment facilities. In treatment systems such as wet ponds a continous monitoring of the particulate pollutant load would be possible. For batch-wise operated treatment system, discharge to the recipient could be governed by on-line TSS measurements.

Paper V

Removal of dissolved metals could be carried out in systems using reactive filter media prepared from natural minerals or from by-products of steel production, such as blast furnace slag (BFS).

Several studies have shown good removal of dissolved metals using BFS (Dimitrova, 1996; Dimitrova and Mehandgiev, 1998; Dimitrova and Mehandgiev, 2000; Berggren Kleja and Gustafsson, 2005; Kietlinska and Renman, 2005). These studies have been carried out with fabricated water matrices using metal concentration far exceeding common stormwater concentrations. In addition, other waters than road runoff e.g., landfill leachate that differs notably from road runoff in composition have been evaluated. The use of road salt (NaCl) during winter is an important factor that could affect the sorption process in reactive filters (Dimitrova, 2002). This was also suggested by Hallberg and Renman (2007).

The study in Paper V was performed *in situ* at the Fredhäll treatment stormwater treatment plant. The pilot trials were carried out October 2006 to December 2006 when studded tyres were used. The conditions for the study were favourable since the runoff was collected directly from the sedimentation basin of the treatment plant. Hence the runoff for the pilot trials was not transported or stored before the trials, and importantly the properties of the runoff were very similar to actual field conditions.

Five metals where chosen (Cd, Cu, Cr, Ni, and Zn) for the study. They were selected based on SEPA (1999).

The results showed good removal of dissolved Cd, Cu, Ni, and Zn independent of elevated NaCl concentrations common in road runoff during winter. The reduction of dissolved Cr was low or neglectable. It was possible to perform the trials with a surface load of 0.25 m/h without any tendencies of hydraulic clogging. The surface load was four to five times higher than typical for other studies with granulated blast furnace slag. This would suggest that after efficient removal of TSS there would be a potential to use the reactive filter media for effective reduction of dissolved metals in similar road runoff.

Paper VI

The knowledge of sediment accumulation and characteristic are pivotal tools for planning and operation of stormwater treatment facilities.

Previous studies have described the accumulation rate for stormwater ponds in height difference over time (cm increase of sediment annually) in the individual ponds. The seasonal variations of accumulation and characteristics of sediment from road runoff have, to the author's knowledge, not been elaborated on prior to this study. The accumulation rate or generation of sediments was studied in regard to the precipitation - i.e. mg generated sediment/mm precipitation. This approach was used for being able to make comparison between seasons.

The study showed that the sediment generation (mg sediment/mm precipitation) during the study period was 44 % higher during the winter periods. The sediment generation showed little variation during summer as opposed to winter.

In winter, the particle size changed to finer particle size distributions, which could be attributed to the use of studded tyres and traction sand in the watershed.

The sediment concentration (mg/kg) of Cd, Cu, Pb, and Zn and organic material differed significantly between the summer and the winter periods. The sediment concentration was elevated (mg/kg) for Cr, Cu, Ni, and Zn during both seasons in regard to guidelines for sensitivity of sediment living organisms. In addition, the levels of Zn and Cu were high with regard to guidelines for contaminated soils

The concentrations of Zn and Cu are of concern for sediment re-use and the risk of mobilisation.

RESULT AND DISCUSSION

Pollutant characteristics

The dissolved metal concentrations (μg/l) and the particulate metal concentrations (mg/kg) displayed significant seasonal variations between the studied winter and summer period. In general, a good correlation between the total metal concentrations and TSS was found. The dissolved part of Al, Cd, Co, Cr, Mn, and Ni was significantly higher in winter in comparison with summer. For Fe, however, the dissolved part was lower during winter. No significant difference was found for Cu, Pb and Zn between the two seasons. The mass concentrations (mg/kg) for all metals were significantly higher over the summer except for Al and Co, which showed higher mass concentrations during the winter. The total concentrations of selected metals and TSS showed a good linear relationship during winter runoff events except for Cd. A good correlation was also found for the summer period for Al, Cu, Fe, Mn, Ni, and Zn.

In the winter season in colder climates, such as in Sweden, the measures for ice prevention, snow removal and the use of studded tyres give notable repercussions on pollutant generation and characteristics.

An extensive study performed in Sweden (Hedlund, 1995) showed that cars driven on roads where de-icing salts were applied displayed two to three times more corrosion damage than those cars driven on un-salted roads. De-icing salt is thus an important contributing factor to a higher metal load, both particulate and dissolved, originating from traffic. Bertling (2005) pointed out that the metal runoff rate was higher for lower rain intensities for a given total rain volume, than for higher intensities. The increased metal runoff rate (i.e. corrosion) could be explained by the extended wet contact time in winter.

The elevated pollutant load of particles during winter runoff events found in Paper III agrees with the findings in Paper I and Paper II and also other studies (Legret and Pagotto, 1999; Westerlund and Viklander, 2006).

In summer atmospheric fallout (Hvitved-Jacobson and Yousef, 1991) could be an important source given the location of the watershed in a heavily industrialised and metropolitan area.

The studied dissolved metal concentrations in Paper III displayed seasonal variations but were constant for the periods.

Dissolved metals are a concern due to the increased risk for acute or long-term toxic effects in the recipient. In Table 1, a classification of total metal concentrations for stormwater is given. The dissolved concentrations of Cu and Zn are somewhat elevated (Table 1). The Swedish Environmental Agency (SEPA) classifies freshwater recipients in regard to metal concentrations in five classes (Class 1-5). Metal concentrations falling into Class 1 are deemed non-low risk and vice versa (SEPA, 1999). The dissolved concentrations of Cu and Zn found in Paper III would be classified as high (Class 4). The reduction of the dissolved fraction cannot be accomplished with sedimentation. One alternative to remove dissolved metal pollutants could be a reactive filter as elaborated on in Paper V.

The particulate concentrations (mg/kg) of the studied metals in Paper III and Paper VI showed that to predict sediment characteristics in a similar watershed the seasonal variations must be considered.

Thomson *et al.* (1997) studied the correlation between metals and TSS in road runoff. Thomson *et al.* (1997) used databases comprising of data from rain events, snowmelt events, and combined snowmelt and rain events ("mixed events") for studying correlations between TSS and metals. Thomson *et al.* (1997) also included TOC (total organic content) and TDS (total dissolved solids). In regard to data transferability or "portability", Thomson *et al.* (1997) discusses the near-site portability. Near-site portability refers to sites located in close geographical proximity to the site used where the relationships were developed. At these sites the environmental differences and road maintenance practices are considered to be minimal. Thomson *et al.* (1997) found good near-site portability for metals in their study.

The findings in Paper I, Paper II, and Paper IV show a good correlation between turbidity measurements and TSS in the studied road runoff. Hence it would be feasible to assess the metal pollutant load using indirect measurement of TSS in similar watersheds.

In the studied catchments areas in Paper I through Paper IV, no traction sand was used for winter maintenance. The impact of traction sand should be of importance as shown in laboratory studies of particle generation from the abrasion of roads (Gustafsson *et al.*, 2005; Kupiainen *et al.*, 2003; Kupiainen and Tervahattu, 2005). Consequently there is a further need to study the impact of traction sand on runoff quality.

Mass transport during a runoff event

Many stormwater treatment systems are designed to capture the initial runoff from storms due to the conception that the majority of the pollutants are contained in the first flush (Barret *et al.,* 1998a). In Paper I and Paper II the possibility to treat only a part of the total runoff volume was studied evaluating the TSS PEMC in regard to a selected demand of 60 mg/l (EU Directive 1991/271/EEC).

The results of Paper I and Paper II showed that the TSS PEMC for majority of the runoff events exceeded 60 mg/l. This suggests that the total runoff volume is necessary to capture and treat in similar runoff.

Several studies of first flush behaviour have been performed and different criterion for first flush has been discussed (Sansalone and Buchberger, 1997b; Bertrand-Krajewski *et al.,* 1998). In motorway watersheds the traffic will be a continous source of pollutants during the runoff event (Barrett *et al.,* 1998a). In a study of two small impervious trafficated watersheds (<300 m^2), no significant first flush was found (Deletic, 1998). In an extensive study of runoff from roads, with AADT between 20,000 to 80,000 vehicles, it was found that the first flush effect was low or negligible (UK Environmental Agency, 2003). To evaluate the treatment options in a watershed the methodology presented in Paper I and Paper II could be applied as an alternative to first flush. The utilisation of a discharge demand is transparent in regard to design of an optimal capture volume.

The evaluation methodology used in Paper I and Paper II in combination with the on-line measurement of TSS could be used for detailed assessment of watersheds in the planning stage. The condition for applying the methods would be the correlation between TSS concentration and turbidity measurement.

Removal of pollutants

Sedimentation

In Paper IV, the sedimentation process during winter and summer was studied. The surface loads for removal of TSS in regard to the initial concentration were estimated. Paper IV showed that the sedimentation properties in winter and summer could be assessed from the initial concentration of TSS. However, elevated concentrations of road salt had a significant impact on the sedimentation behaviour of the particulate matter. Elevated concentrations of NaCl were suggested to increase the settleability of the particulate matter.

The particle size distribution for winter period changed towards finer particle sizes. The particle size distributions found in Paper IV, agreed with the particle size distributions found in road runoff by Andral *et al.* (1999) and Roger *et al.* (1998). In the study by Roger *et al.* (1998) it was shown that 90 % of particles in the studied road runoff had a diameter of less than 100 μm and 78 % were smaller than 50 μm. Particles smaller than 50 μm were composed of clays (56 %) with an organic content of 9 %. Andral *et al.*, (1999) showed that 50 % (w/w) of particles smaller than 50 μm fell at velocities less than 2.98 m/h. Paper IV studied settling velocities below 4.4 m/h in regard to the study by Andral *et al.* (1999). The findings in Paper VI show an organic content for the sediments of 16.7 % in winter and 19.1 % in summer, which is significantly higher than the study by Roger *et al.,* (1998).

Randall *et al.*, 1982 collected runoff samples from three shopping centre parking lots. In the study it was concluded that 80 % of the particles had a diameter of less than 25 μm and 57 % had a particle diameter less than 15 μm. The study found that the initial concentration of TSS corresponded well with the removed part of the TSS. In this case study it was shown that the sedimentation behaviour was similar between the runoff events with exception for elevated concentrations of road salt. In watersheds where traffic is a dominant and continous pollutant source a consistency in sedimentation properties could be expected.

The findings in Paper IV shows a good correlation between turbidity and TSS was found. However, a significant difference between the winter period and summer period was found. The difference could be attributed to the finer particle size that occurs in runoff during winter. Turbidity measurement is an alternative to gravimetrical analysis of TSS and has been elaborated on other studies (e.g. Pavanelli and Bigi, 2005; Suk *et al.*, 1998; Grayson *et al.*, 1996). However, organic material and colour of the water could also influence the turbidity measurements. Turbidity measurements are commonly used to assess drinking water quality.

Filtration

Paper V showed that a reactive filter could be used successfully for removal of dissolved metals in road runoff. Elevated levels of dissolved metals in road runoff in particular, Cu and Zn, were demonstrated in Paper III. The runoff was siphoned to the pilot plant after a minimum of 36 hours sedimentation. It was possible to achieve a good reduction of dissolved Cd (>90 %), Cu (>75 %), Ni (>40 %), and Zn (>97 %) independent of elevated concentrations of NaCl. The filtration was carried out with a surface load of 0.25 m/h with a pressure drop less than 0.01 m water column. The precondition for the comparatively high surface load would be the effective reduction of suspended solids from the runoff.

Pollutant removal from road runoff using porous material or filters combined with infiltration in soil have been studied by e.g. Hogland *et al.* (1987), Tenney *et al.* (1995), Legret and Colandini (1999) Legret *et al.,* (1999), and Teng and Sansalone (2004). Barrett *et al.* (1998b) and Bäckström (2003) studied particle removal and infiltration using grassed swales.

The employment of various constructed filters for pollutant removal must consider the unpredictability of clogging and the long-term operation and maintenance (cf. Paper I and II). The deposition of metals in the soil infers an absence of pollutant control - e.g., the mobilization of metals at elevated chloride concentrations (Amrhein *et al.*, 1992) and in regard to soil quality and pH (Barbosa and Hvitved-Jacobsen, 1999).

Sediment characteristics

The sediment accumulation dependence on precipitation (mg sediment/mm precipitation), particle size distribution and metal and organic content were studied (Paper VI). The sediment generation rate during winter was 44 % higher. Sediment accumulation showed little variation during summer. In winter, the particle size shifted to finer particle size distributions, which was attributed to the use of studded tyres. The sediment concentration of Co, Cu, Pb, and Zn and organic material differed significantly between the summer and winter periods.

The elevated sediment generation rate during winter could be expected. The difference in sediment accumulation between the two winter periods could be explained by e.g. the usage of road salt and traction sand as well as variations in snow accumulation, snow melting, and snow handling (e.g. Viklander, 1997). In addition, the hydraulic conditions in the pond also influence the sediment accumulation in winter. The relatively consistent accumulation rates for the summer would infer that the pollutant load could be estimated from total precipitation for that season. Thus, the impact of duration of precipitation as well as dry weather condition is of less importance for the assessment of sediment generation during summer.

The sediment concentrations (mg/kg) of Cr, Cu, Ni and Zn were elevated in regard to sensitivity of sediment living organisms (GPMAS, 1993) for both summer and winter. In addition, the levels of Zn and Cu were high in regard to re-use of the sediments (SEPA, 1997).

The particle size distributions found during winter and summer were less than 200 μm. Stone and Marsalek (1996) suggested that the finer fractions (< 250 μm) were retained by drainage systems. The particle size distributions found in Paper IV are similar to the findings of Paper VI. The findings of Stone and Marsalek (1996) and Paper IV would suggest that the coarser particles were retained by the drainage system of the studied watershed in Paper VI.

The selected discharge criteria used in Paper I and II show that the entire runoff volume must be captured. Paper III shows the variations in the metal concentration in the particulate material (mg/kg) and Paper IV shows the settleability of the particulate matter in road runoff. In all, the function of the sedimentation unit for treatment of road runoff is essential.

Further long term studies should be performed on sediment in regard to seasonal variations of pH and dissolved oxygen concentration in the sediment. In addition studies are also of interest for re-use of the sediments as an alternative to landfilling. This would be important for the assessment of sediment removal intervals and a sustainable handling of the sediments.

CONCLUSION

The mass transport in regard to the TSS reference value of 60 mg/l showed that the entire runoff volume must be retained and treated. The majority of metal pollutants in the studied road runoff showed good correlation to TSS for the studied periods. This implies that sedimentation will also remove metals. However, elevated dissolved concentration of Cu and Zn were found during both seasons. For this reason, the filter bed technology using reactive materials, such as blast furnace slag (BFS), could be applied for further treatment.

A column study with BFS and a surface load of 0.25 m/h showed that the dissolved metals were successfully removed.

Good removal was achieved independent of elevated concentrations of road salt (NaCl), which is of importance in regard to winter conditions when road salting is common. The condition for using filters is the removal of TSS before filtration to avoid clogging.

The removal of TSS could be described, for winter and summer, with an empirical model using only the influent concentration of TSS to estimate the effluent concentration of TSS in regard to a chosen surface load. Elevated concentrations of salt (NaCl) did have an impact on the settleability of the particulate material. The affinity of the metals to the particulate material would suggest that the metal reduction also could be assessed with the model. However, the dissolved metal concentration must be considered in regard to discharge demands. The sediment accumulation (mg sediment/mm precipitation) was found to be relatively consistent for the studied summer seasons as opposed to winter. The particulate metal concentration (mg/kg) did show variations between the seasons. The findings of this work show the conditions for removal of TSS and particulate and dissolved metals from road runoff and sediment handling in regard to design of treatment facilities. The case study performed also indicates the possibility of using on-line measurement of TSS as a powerful tool for process control and monitoring of a treatment facility.

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