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SUSPENDED SOLIDS AND METALS IN HIGHWAY RUNOFF – IMPLICATIONS FOR TREATMENT SYSTEMS

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SUMMARY

It is known that traffic is a source of pollutants and that pollutant loads increase with elevated traffic densities. Studies executed in Stockholm, Sweden advocate that highway runoff from roads with an annual average daily traffic (AADT) exceeding 30,000 vehicles need treatment before discharge to the receiving water. It is common knowledge that sedimentation is the most expedient method for stormwater treatment. However, sedimentation units are area demanding and in highly urbanised watersheds the land use is often restricted. Studies have implied the occurrence of first flush, i.e. an initially higher pollutant load in the beginning of the runoff event, in highway watersheds. With an emphasized first flush it would be possible to treat only a part of the total runoff volume reducing the area needed for a sedimentation basin. In general two methods are used to design stormwater treatment ponds. One method is based on the reduced catchment area and pond surface and the other is based on an average runoff volume and a permanent pond volume. The methods are relaying on data from routine monitoring of various treatment systems and suggest removal efficiencies for pollutants. Applying general removal efficiencies for design it can be intricate to estimate an outlet concentration when the specific removal efficiency may be dependent on the initial concentration of the pollutant. Consequently, knowledge of the removal efficiencies dependence on initial concentration would be helpful to optimise stormwater treatment systems. This research has studied runoff from highly trafficated watersheds. The aim has been to evaluate the mass transport, stormwater quality and sedimentation behaviour and their implications for stormwater treatment. The study sites, Eugenia and Fredhäll, are located along the six-lane highway E4 through Stockholm that has an AADT load of 120,000 vehicles and a speed limit of 70 km/h. In lack of a unified definition of first flush the mass transport was studied using the EU directive 1991/271/EEC discharge demand for TSS of 60 mg/l. It was found that for the majority of the runoff events during winter the event mean concentration exceeded 60 mg/l suggesting that the complete runoff volume should be captured during winter. The dissolved concentration of metals showed significant variations between winter and summer, as did the concentration in the particulate matter (mg/kg). It was possible to correlate total metal concentration to total suspended solids with good correlation (r^2 >0.90) for the majority of studied metals in winter and summer. The findings would imply that a successful treatment of the studied metal pollutants could be carried out by sedimentation. However, depending on discharge criteria, the elevated levels of dissolved matter, especially during winter, have to be considered with regards to the selection of the appropriate water treatment process. The sedimentation process could be described by a logarithmical function and initial turbidity. Good correlation (r^2 >0.90) was indicated between turbidity and TSS. The sedimentation process of the studied highway runoff varied significantly ($p<0.05$) when elevated levels of NaCl could be found in the runoff. A significant difference ($p<0.05$) was shown for turbidity and TSS between summer and winter, which was assumed to be related to the use of studded tires. This study implies that the entire runoff volume must be treated and that the use of first flush as a design criterion is less applicable for the winter period. The study implies good correlation between total metal concentration and TSS. In addition the indicated correlation between turbidity and TSS would point to the possibility to use turbidity as a surrogate measurement for TSS and the studied metals. Moreover, the possibility to describe the sedimentation process by the initial concentration of turbidity would infer the utilisation of turbidity as a tool for process control for stormwater treatment systems. In addition, the novel results for the dependence on the sedimentation process could be incorporated in existing models for design of stormwater treatment systems in similar watersheds.

SAMMANFATTNING

Det är allmänt känt att trafiken är en källa till föroreningar i dagvatten och att föroreningarna ökar med trafikintensiteten. Vid framtagandet av en dagvattenstrategi för Stockholm förordades en behandling av dagvatten från vägar med en årlig dygnstrafik (ÅDT) överstigande 30 000 fordon. Den helt dominerande behandlingsmetoden för dagvatten är sedimentering i varierande typer av ytkrävande dammar. I urbana och tätbebyggda områden med ytterst begränsad tillgång till markområden är möjligheterna att anlägga större dammar få och ofta obefintliga. Undersökningar har visat på ett masstransportfenomen benämnt ”first flush” eller initial flödes- alternativt smutspuls (förf. övers.) i avrinningsområden med hårdgjorda ytor. En initial smutspuls beskrivs av en högre föroreningsbelastning i början av avrinningstillfället. En utpräglad initial flödespuls skulle innebära en möjlighet att endast behandla en del av den totala avrinningsvolymen och således minska behövlig yta för sedimentering. Nuvarande designmetoder för dagvattenbehandling baseras främst på reducerad avrinningsyta eller en avrinningsvolym. Underlaget till metoderna utgörs av mätningar i befintliga dagvattensystem av in- och utgående halter och anvisar en generell relativ reduktion (%) av en förorening. Användandet av en generell relativ reduktion kan vara vanskligt i det fall att reduktionen är avhängig den inkommande föroreningskoncentrationen. Således är det intressant att studera reduktionens beroende av inkommande koncentration. Detta skulle möjliggöra en komplettering av nuvarande designmetoder och underlätta optimeringen av dagvattenbehandlingar. I föreliggande licentiatarbete har dagvatten studerats från högtrafikerade avrinningsområden. Fokus har varit på masstransport, vattenkvalitet samt sedimenteringsegenskaper utifrån ett reningsperspektiv. Arbetet har genomförts på två platser, Eugenia samt Fredhäll, vilka båda är lokaliserade längs Essingeleden (E4) med en ÅDT av 120 000 fordon samt en skyltad hastighet av 70 km/timme. I avsaknad av en gemensam definition för initial smutspuls nyttjades EU direktivet 1991/271/EEC med utsläppskrav på 60 mg/l för undersökning av masstransport. Under vintern studerades totalt 30 på varandra följande avrinningstillfällen och det konstaterades att för merparten översteg medelkoncentrationen av suspenderat material 60 mg/l. Följaktligen bör hela avrinningsvolymen behandlas vid vinterförhållanden i jämförbara dagvatten. Undersökningen av lösta metaller visade att koncentrationen (µg/l) varierade mellan vinter och sommar vilket även var fallet för metallinnehållet i det partikulära materialet (mg/kg). En god korrelation mellan suspenderat material och totala halten metaller konstaterades under vinter och sommar. Det borde härför vara möjligt att framgångsrikt avlägsna de studerade metallföroreningarna med sedimentering. Noteras bör dock de förhöjda halterna av löst material under vintern och val av behandlingsmetod visavi utsläppskrav. Sedimenteringsegenskaperna kunde beskrivas logaritmiskt och med initial turbiditet. En acceptabel korrelation ($r^2 > 0.90$) mellan suspenderat material och turbiditet erhölls även. I försöken befanns vägsalt ha en betydande påverkan ($p<0.05$) på sedimenteringsprocessen. Vidare konstaterades en väsentlig skillnad ($p<0.05$) för sambandet mellan turbiditet och suspenderat material vilken bedömdes vara avhängig dubbdäcksanvändningen. Utifrån genomförda undersökningar kan konstateras att hela avrinningsvolymen under vintern måste kvarhållas för behandling varför detta indikerar att nyttjandet av initial flödes- eller smutspuls (first flush) är mindre tillämpbart vid projektering av dagvattenanläggningar i liknande avrinningsområden. Vidare pekar undersökningarna på att turbiditet kan användas för att indirekt kvantifiera suspenderat material och studerade metaller. Av detta följer även möjligheten använda turbiditet för processkontroll vid dagvattenrening. Slutligen, bör den nyvunna kunskapen avseende sedimenteringsegenskaperna komplettera och nyttjas i existerande designmodeller vid projektering i jämförbara trafikbelastade avrinningsområden.

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ABSTRACT

Elevated levels of pollutant can be found in runoff from catchment areas with dense traffic loads. It is understood that the major pollution from stormwater is related to the content of particulate matter. A treatment practice can be based on the mass transport phenomena known as first flush, by detention of the initial part of the runoff, which is considered to contain the highest concentrations of pollutants. However, no unified definition is available for a first flush criterion. The knowledge of the partitioning between dissolved and non-dissolved matter of pollutants during the runoff event is of concern, as is its seasonal variations for design of a water treatment process. Moreover, the existing design tools apply general removal efficiencies. Removal efficiencies witch may be dependent on the initial concentration of the contaminants. The effluent standard for wastewater of 60 mg TSS per litre applied in EU was used to assess the mass transport. The concentration of total suspended solids was studied in 30 consecutive runoff events during the winter. In only two of the events the event mean concentration was below 60 mg/l. Consequently, during winter these findings imply that a capture of the total runoff volume is necessary for treatment. The partition between dissolved and particulate matter of ten metals (Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) was studied during winter and the subsequent summer. The dissolved part of Al, Cd, Co, Cr, Mn and Ni was significantly higher for winter compared to the summer ($p<0.01$). For Fe, however, the dissolved part was lower in the winter. No significant difference was found for Cu, Pb, and Zn between the two seasons. 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 over the winter. The concentration of selected metals vs. total suspended solids showed a linear relationship ($r^2>0.95$) during winter runoff events except for Cd. A good correlation ($r^2>0.90$) was also found over the summer period for Al, Cu, Fe, Mn, Ni and Zn. The impact of initial concentration on the sedimentation process was assessed by turbidity. The sedimentation process could be described by a logarithmical expression. It was found that the sedimentation process was dependent on the initial turbidity. The use of road deicing salt had significant impact on sedimentation behaviour ($p<0.05$). Turbidity was well correlated with total suspended solids ($r^2 >0.90$) for all runoff events. These findings suggest that TSS can be used as a substitute parameter to assess the metal pollutant load. Furthermore, the sedimentation process for a specific surface load can be approximated from initial concentration of total suspended solids for the studied type of highway runoff.

Key words: Dissolved matter; particulate matter; sedimentation process; suspended solids; turbidity

INTRODUCTION

The adverse effect of stormwater pollution on the receiving water environment and the need for treatment was recognized in the 1960's (e.g. Muschak, 1990). Stormwater may inflict pernicious toxic and/or erosion effects in the recipient. The contaminant transport during a single storm event is often characterised by higher load in the beginning of the runoff event. This mass transport phenomenon is well known as a first flush (Bertrand-Krajewski *et al.*, 1998; Urbonas and Stahre, 1993). However, studies of catchment areas between 211 m^2 (Deletic, 1998) up to 560 ha (Lee and Bang *et al.*, 2002) have shown that the mass transport behaviour vary significantly even between similar catchment areas during comparable runoff events. The composition of pollutants and pollutant load will be influenced by seasonal variations and the diversity of activities in the catchment environment. Important are the variations in flow depending on precipitation depth and duration. In winter the variations in temperatures in the course of snowmelt exert an influence on the flow and concentration of the contaminants.

A watershed with roads as the dominating land use describes the complexity of the variations in runoff. Most evident are the variations in regards to summer and winter conditions in cold climate. In the course of winter the pollutant load increases dramatically when de-icing agents are utilized (Legret and Pagotto, 1999). Additionally, the use of studded tires contributes to an augment of the wear of the asphalt pavement (Jacobsson and Hornwall, 1999) resulting in increased transport of particles. The particle size influences the erosion and transport properties of the particulate matter, generated from vehicles, road surface and airborne deposition and accordingly impact the pollutant load during rain or snowmelt. These variations, as a whole, make it very difficult to predict flow or pollutant concentration for an individual runoff event.

For abatement of the harmful effects of runoff, various types of detention basins or retention systems have been utilised. A detention basin is designed to intercept and temporarily store the accumulated stormwater for a period of less than 24 hours (USEPA, 1999). A retention system may consist of a single unit or a combined system of underground piping and caverns and wet ponds (USEPA, 1999). The purpose of the retention system is to provide for a storing capacity between runoff events. Since the majority of the pollutants in runoff are associated with the particulate matter (e.g. Hvitved-Jacobson and Yousef, 1990; Sansalone and Buchberger, 1997a), detention basins and retention systems reduces pollutants by means of sedimentation. Furthermore, the contaminants in runoff are predominantly associated with the particulate material and increases with decreasing particle size (Hvitved-Jacobsen and Yousef, 1991; Xanthopolus and Hahn, 1990). The retention system could accordingly furnish an enhanced reduction of particulate pollutants by extended sedimentation times in comparison with a detention basin.

Over time the importance of stormwater facilities for treatment of the runoff has become increasingly important. Nix *et al.* (1988) concluded that design guidelines for stormwater focused on the need for detention and not primarily on sedimentation. Ten years later Barret *et al.* (1998a) noted that many stormwater treatment systems were designed to capture the initial runoff from storms and thus remove and treat the runoff that contains the highest concentrations of pollutants. Numerous studies of existing stormwater handling systems, e.g. detention basins and retention systems, by monitoring of inlet and outlet concentration of pollutants and analysis of accumulated sediments (e.g. Färm, 2003; German, 2003; Bäckström, 2001; Pettersson, 1999) have been executed. The treatment capacity is commonly given by the removal efficiency i.e. the ratio between outlet and inlet concentration of a pollutant. The hydraulic conditions vary depending on construction of the studied treatment systems in regards to non-uniform distribution over the surface, counter currents, hydraulic shortcuts from inlet to outlet, erosion and bioturbation. Another factor that will influence particulate removal is the extent of vegetation in a pond or in the drainage network (e.g. Bäckström, 2001; Barret *et al.*, 1998a). In a study of ponds for reduction of highway runoff it was found that 450 m^2 of wet pond surface per ha of watershed area was needed to achieve a good reduction of TSS (Pettersson, 1999). The StormTac model (Larm 2000; Larm, 2005) indicates a minimum reduced surface area per watershed ha for wet ponds of 150 m^2 (70 m^2 – 400 m^2). However, the removal efficiency does not consider the dependence of outlet concentrations on the inlet concentration of the pollutant. Furthermore, the existing design guidelines do not provide information as to seasonal effects on the sedimentation process. However, when ample space is available for construction of ponds and filter strips the prevailing design procedures are adequate. In highly urbanised areas however, the land use often restricts the construction of extensive basins. Within a watershed traffic has been recognized as a source of pollutants and that a relationship exists between an increasing number of vehicles and a rise in the pollutant load (Hvitved-Jacobson and Yousef, 1991). Runoff from urban roadways often contributes to significant loads of metal elements, particulate and dissolved solids, organic compounds and inorganic constituents (Sansalone and Buchberger, 1997b). Barret *et al.* (1998a) concluded that high pollutant loads were related to elevated traffic flows. Hares and Ward (1999) recognized that a higher level of motorway-derived heavy metal contamination existed in runoff from a road section with an elevated average daily traffic density. Studies executed in Stockholm, Sweden advocate that highway runoff from roads with an annual average daily traffic (AADT) exceeding 30,000 vehicles need treatment before discharge to the receiving water (Aldheimer and Bennerstedt, 2003). In the EU directive 1991/271/EEC guidance as to discharge value for suspended solids is given. The directive defines stormwater as sewage water. For domestic wastewater the directive gives a discharge concentration of 60 mg/l for TSS. Given the known affinity of stormwater pollutants to the particulate material the discharge value could be applied as an alternative for assessment of mass transport during a runoff event.

OBJECTIVES

The fieldwork was conducted from 25 October 2004 to 28 August 2005 including the winter period in Stockholm, Sweden, when vehicles are equipped with studded tyres and salt is regularly used on the roads. Two study sites were chosen along the same major highway with an AADT load of 120,000 vehicles. The aim was to (i) examine the TSS concentration during a runoff event to study the mass transport for assessing first flush behaviour and treatment possibilities (ii) investigate seasonal variations between winter and summer with regards to the dissolved metal concentration (µg/l) and the mass concentration (mg/kg) of particulate bound metals (iii) study the sedimentation process of runoff water from a highly trafficated area during winter and summer conditions.

HIGHWAY RUNOFF-A BACKGROUND

The transport of contaminants has been described from different catchments areas by different models (e.g. Larm, 2000; Larm 2005). It is however recognized, that pollutant concentrations and loads will vary even between very similar catchments areas. In Stockholm an elaborate investigation was executed to assess the impact of runoff water on the receiving water bodies, treatment needs and costs (Stockholm Vatten 2000; Stockholm Vatten, 2001a; Stockholm Vatten, 2002). The investigation resulted in guidelines for best management practice (Stockholm Vatten, 2001a). The runoff water was classified in three group's i.e. low concentrations, intermediate concentrations and high concentrations as described in Table 1.

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

	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*

For runoff water with high concentrations it was concluded that stormwater treatment was necessary. Roads and motorways was identified as important sources of pollutants. In addition, runoff from highways with an AADT exceeding 30,000 vehicles as a rule needed treatment to reduce the contaminant concentrations in regards to expected pollutant concentrations.

Pollutant generation and characteristics

Highway 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). Traffic related pollution originates from abrasion of tire and brake linings, leakage of hydrocarbons and residues from combustion (Muschak, 1990). In a study by Roger *et al.* (1998) 90 % of the particulate matter had a diameter of less than 100 μm and 78 % of the material had a diameter below 50 μm. The content of these clay fractions displayed cationic exchange capability indicated by a high correlation between zinc, the clays and the organic matter. This correlation between zinc and organic matter was also found in the particle fraction 0.45 μm to 20 μm by Characklis and Wiesner (1997). Furthermore, the specific surface area of particles in highway runoff increases with decreasing measured particle diameter (Sansalone *et al.*, 1998). The sediment from highway runoff also displays very fine particle fractions typically less than 100 μm (Durand *et al.,* 2004; Stockholm Vatten, 2001b)

During winter the pollutant loading will increase from roads (Westerlund *et al.*, 2003). A study in northern Sweden concluded that a longer winter period generated higher concentrations of pollutants (Reinosdotter and Viklander, 2005). Snow in the road environment will accumulate heavy metals and other anthropogenic constituents (Glenn and Sansalone, 2002) and as a consequence the pollutant concentration will be elevated during snowmelt. Studded tires 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 (Jacobsson and Hornvall, 1999; Jacobsson, 1994). Traction sand is also a pollutant source during winter and experimental studies indicate that traction sand have greater impact on the generated airborne particles than studded tires alone (Kupiainen *et al.,* 2003). Furthermore, 90 % of the generated airborne particles originates from the traction sand and road pavement and for the remaining the assessed source was tire and bitumen abrasion (Kupiainen and Tervahattu, 2005). In a study with studded and non-studded winter tires (friction tires), studded tires generated a multiple of 40-50 times of particles measured as PM10. The generated PM10 particles had a maximum around 3 μm to 4 μm but all particles generated were greater than 1 μm (VTI, 2005). A study conducted over three years in Stockholm concluded that 87 % of PM10 particles originated from wear of the pavement, 8 % could be attributed to exhaust, 3 % to brake wear and, 2 % from tire wear. The use of studded tires accounted for 40 % to 70 % of the road wear (SLB, 2004).

Another factor that contributes to the pollutant loading during winter is the use of salt (NaCl) as a de-icing agent. In a study by Buttle and Labadia (1999) it was found that the used road salt was transported away from the road surface during or shortly after its application. In an extensive study by the UK Highway Agency and UK Environmental Agency (UK Environmental Agency, 2003) it was deducted that metal was 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 (Korrosionsinstitutet, 1995). The two areas are in the same geographical vicinity on the east coast of Sweden and the Baltic Sea. In Gotland no road salt was used during the study period as opposed to Västervik. The cars that were studied were employed by 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. Another issue is the wet exposure times, which affect the corrosion. Bertling (2005) notes that the metal runoff rate was higher for lower rain intensities for a given total rain volume compared to higher intensities, because of the longer contact time at lower rain intensities.

The use of traction sand, de-icing agents, studded tyres and, winter road maintenance will generate elevated pollutant loads from road surface and vehicles compared to summer conditions. The particulate matter generated during colder periods is fine and typically below 50 µm. The impact of salt may cause an accretion in the dissolved part of metal pollutant. The seasonal variations are emphasised and affects the composition of particulate material and dissolved matter. Thus it is important to study seasonal variation in regards to mass transport and treatment of stormwater.

Mass transport during a runoff event

One of the key transport phenomena discussed during a runoff event is first flush. First flush is described 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 depths to estimate the appropriate capture volume (Barett *et al.*, 1998a). Different descriptions of first flush exist, however, to quantify a first flush the dimensionless M(V) curve can be used (1).

$$\frac{\sum_{i=1}^{j} CiQi\Delta t}{\sum_{i=1}^{N} CiQi\Delta t} = f\left(\frac{\sum_{i=1}^{j} Qi\Delta ti}{\sum_{i=1}^{N} Qi\Delta ti}\right) = f\left(\frac{\sum_{i=1}^{j} Vi}{\sum_{i=1}^{N} Vi}\right) \quad (1)$$

The M(V) curve describes the relative mass load of the contaminant in relation to the relative volume during the time of the storm event. The most stringent definition of first flush is when 80 % of the pollutant mass is transported within 20 % of the initial runoff volume as described by Urbonas and Stahre (1993) and Bertrand-Krajewski *et al.* (1998). Lee *et al.* (2002) studied 13 separate urban watersheds and implied a first flush if relative pollutant mass load was greater than the relative volume for the whole runoff event. Barbosa and Hvitved-Jacobsen (1999) suggested a first flush when 50 % of the total volume carries 61 % to 69 % of the total solids.

In the study by Barrett *et al.* (1998a) it was concluded that a pronounced initial higher concentration of TSS was evident in the catchment with the highest daily traffic load, approximately 60,000 vehicles. The elevated initially higher pollutant load was found during rain events with short duration and constant rainfall. In addition, the traffic was constantly generating pollutants so that a complete wash off never occurred. Characklis *et al.* (1997) studied a large urban watershed (240 km^2) and found no indication of first flush effects. Lee and Bang (2000) found that watersheds of less than 100 ha with an impervious surface covering 80 % and for watersheds larger than 100 ha and 50 % covered by impervious area the peak concentration of the pollutants preceded the peak flow. Deletic (1998) studied two urban asphalt catchments with an area of 211 m^2 and 270 m^2 and could not identify a pronounced first flush. Thus, the complexity of factors influencing a runoff event render it difficult to predict mass transport behaviour for the particulate matter for a single runoff event as pointed out by e.g. Charbenau and Barett (1998). There is no unified definition of a first flush criterion. Furthermore, variations in mass transport are emphasised even in comparable watersheds and runoff events. However, a “fixed” discharge concentration could provide an additional possibility to assess mass transport during a storm event. Moreover, it would be helpful to determine if only a part of the runoff needs to be detained for treatment.

Pollutant removal

A focus in highway runoff has been on suspended solids and heavy metals. Common treatment practices for runoff water are sedimentation basins or ponds, infiltration ponds and different types of vegetated filter strips. Aldheimer and Bennerstedt (2003) found that sedimentation was the most expedient treatment for runoff. They noted that in a city, such as Stockholm, the size of sedimentation basin might restrict its use. Pettersson (1998) studied pollutant removal in a pond receiving runoff from a predominantly highway watershed. Pettersson concluded that a detention pond could be designed to capture the complete storm volume due to the risk of short-circuiting. A recent study in Southern Sweden showed a decrease in pond performance during winter (Semadeni-Davies, 2006). The lowered performance was attributed to impaired retention time due to ice and salt induced stratification. Barett *et al.* (1998b) studied vegetated highway median and found a reduction of suspended solids of 85 %. Hares and Ward (1999) studied the removal efficiency for heavy metals in combined systems of wet biofiltration and dry detention ponds. In the extended system removal efficiencies around 90 % was reached. Hence, there are a number of options for treatment of stormwater. However, sedimentation as treatment method prevails. As exemplified by Pettersson (1998) constructed dams or ponds imply uncertainties in the hydraulic behaviour. In addition, as demonstrated by Semadeni-Davies (2006) there is also the issue of effects of seasonal variations, and yet the majority of the data for design of stormwater treatment system rests on routine monitoring of existing ponds or dams. The studies provide a good "rule of thumb" for design when no restriction as to land use exists. Thus, there is a need for complementing existing knowledge by detailed studies of the sedimentation process.

STUDY SITES

The study sites, Eugenia and Fredhäll, were selected along the six-lane highway E4 through Stockholm that has an annual average daily traffic (AADT) load of 120,000 and a speed limit of 70 km/h.

Eugenia

The highway is passing through the 235 m long Eugenia road tunnel. Data on the drainage area and land use are presented in Table 2. The watershed was divided into four separate areas in regards to piping network i.e. South West 1 (SW1), South West 2 (SW2), South East (SE) and North West (NW) as shown in Table 2 and Fig. 1-8. SW1 receives some runoff water from a pedestrian walk. SW2 includes the Solna Bridge that passes over the highway 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 highway. A treatment plant was constructed and commissioned in 1991 in order to reduce pollutant load from the watershed. The recipient of the stormwater after treatment is the small freshwater lake Brunnsviken. The treatment plant, named 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.

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 dimension (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*			

***Figure 1** Picture of the SW1 Catchment Area*

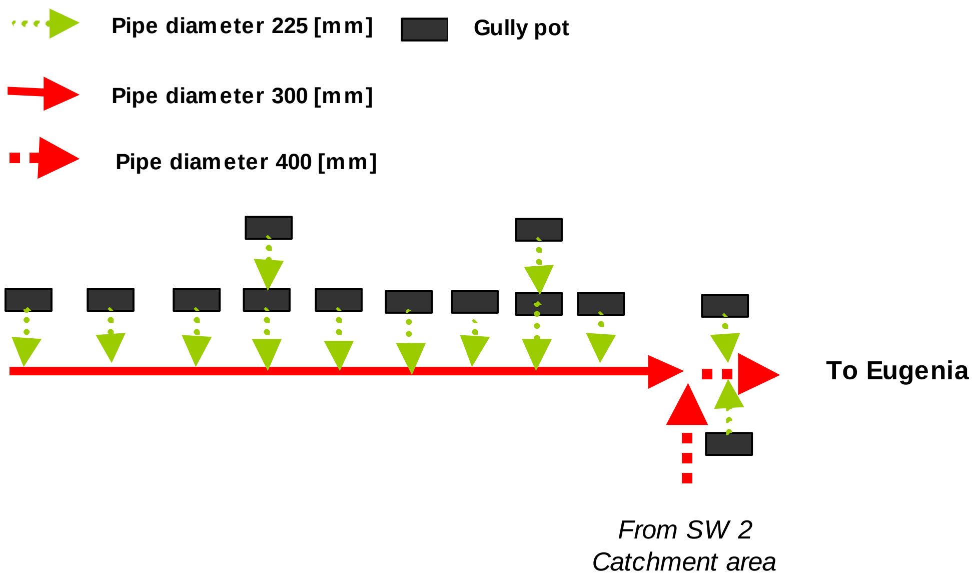

***Figure 2** Schematic layout of drainage system SW1 Catchment area*

Figure 3 *Eugenia catchment area SW2 (Picture taken from Solna Bridge)*

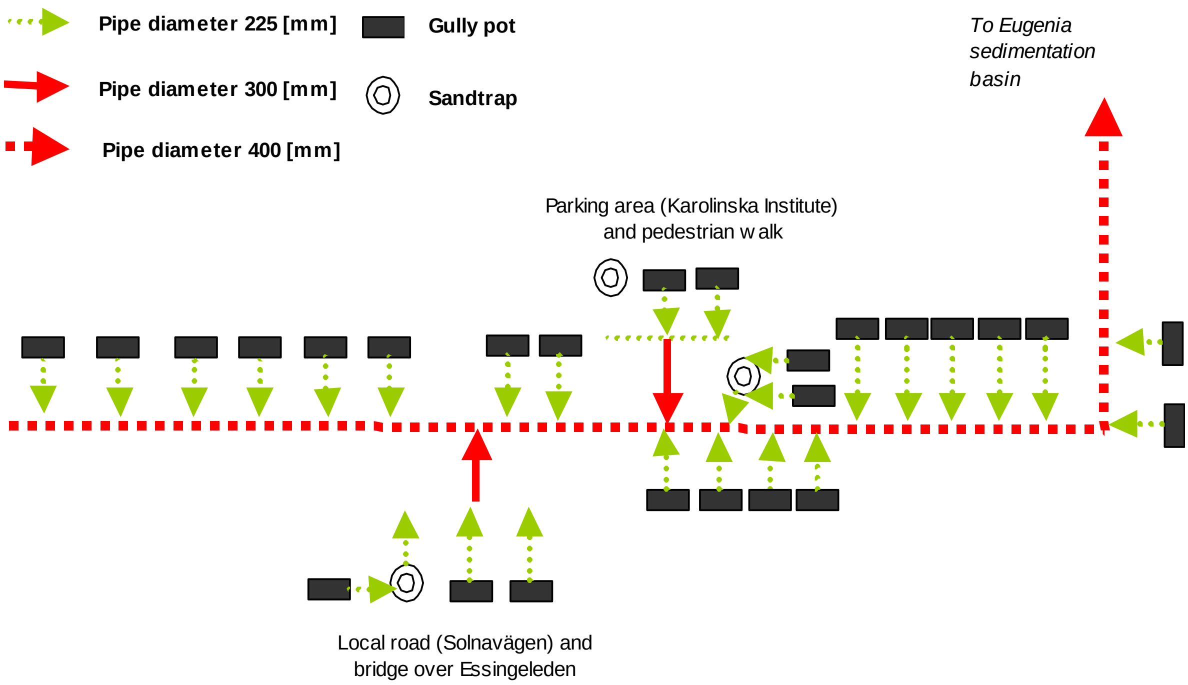

Figure 4 *Schematic layout of drainage system SW2 catchment area*

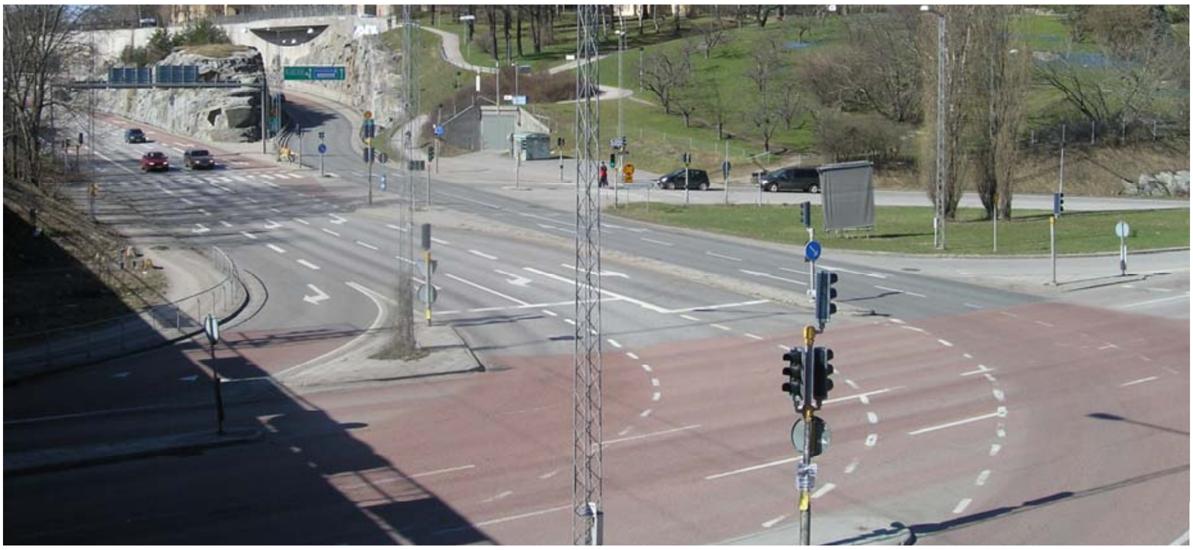

Figure 5 *Eugenia catchment area SE*

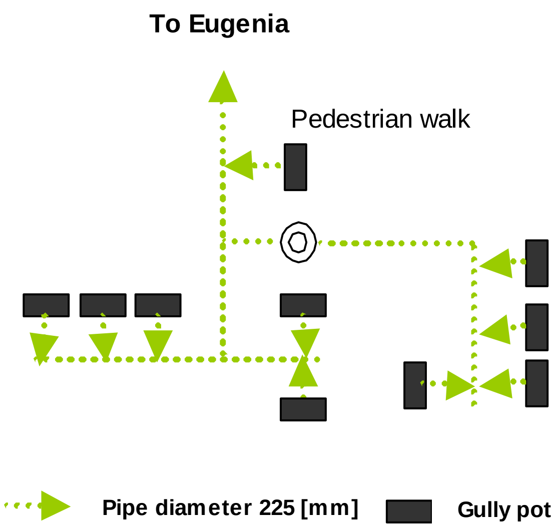

Figure 6 *Schematic layout of drainage system SE catchment area*

Figure 7 *Eugenia catchment area NW*

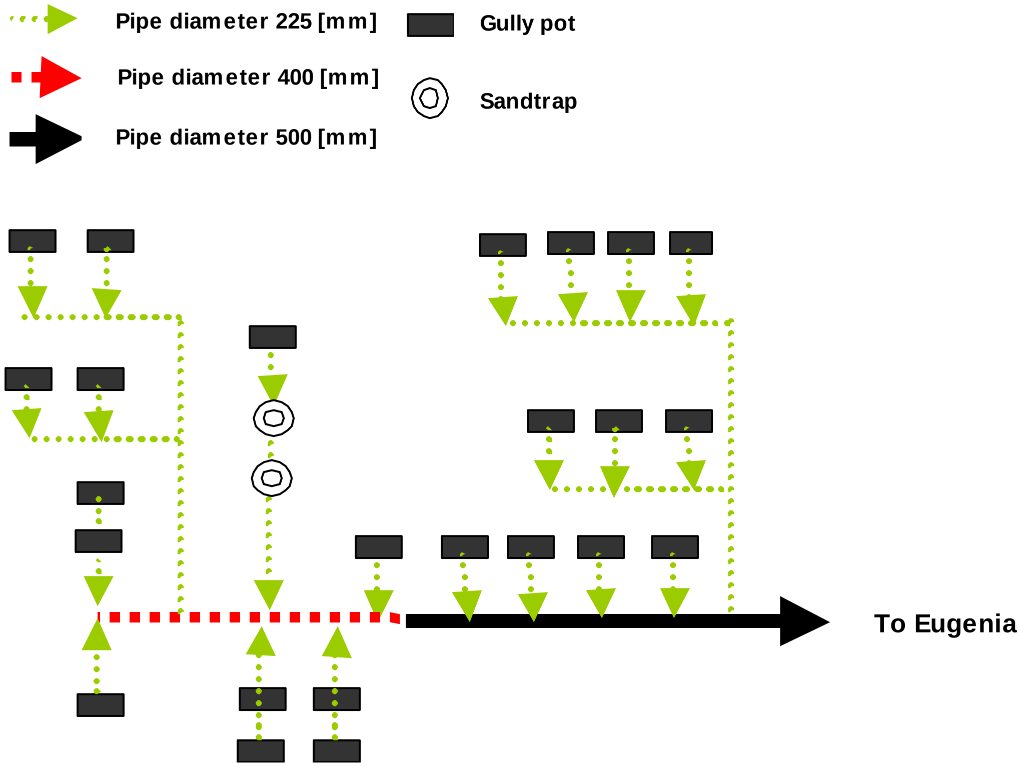

Figure 8 *Schematic layout of drainage system NW catchment area*

Fredhäll

Figure 9 *Fredhäll treatment plant for stormwater*

The total drainage area was 13,700 m^2 and the road surface was covered with asphalt. The watershed includes a tunnel with a road area of 7,800 m^2 (Fig. 9). The recipient, seen in Figure 10, is part of the Lake Mälaren. In order to reduce the pollutant load from the runoff, a treatment plant was built and commissioned in 2003 (Fig. 9). The treatment plant, named Fredhällsmagasinet, is located below the South tunnel entrance. The runoff is transported under gravity to the treatment plant from the bridge, tunnel and highway 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.

EXPERIMENTAL SET-UP

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. *In situ* calibration of the instrument was achieved by correlating the analysed TSS concentration to the registered value from the Cerlic ITX instrument.

Data collection – On line measurements

In Eugenia all sampled data from the on-line measurements was collected with Campbell Scientific CR10X data logger. In Fredhäll all sampled data from the on-line measurements was collected in the operating panel (ABB type 245B).

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

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

Eugenia

Flow measurement

Flow measurement from 1 l/s to 600 l/s was executed 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).

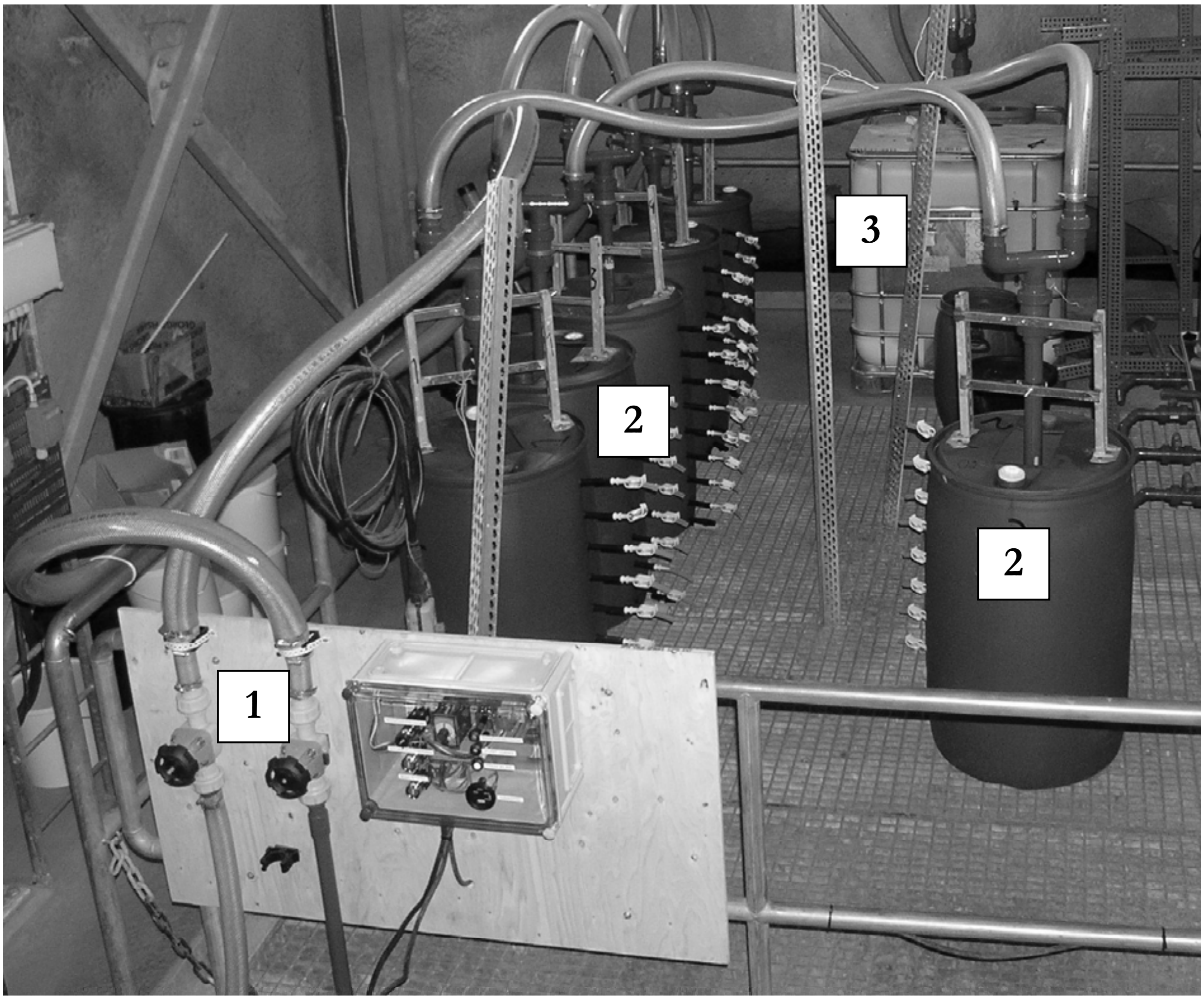

Figure 12 *Collection system for runoff during field trials. The two valves used to set the flow [1], the six sedimentation vessels in the HACS [2], and Sampling Tank (ST) [3]*

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.

Sampling of runoff water

For the study of the sedimentation process a pilot scale sampling system was constructed. The sampling system accommodated for collecting consecutive samples during a runoff event. It was important to provide for sampling during the initial part of the runoff event when rapid increase in flow and pollutant concentration can be expected. Furthermore, the individual sample should have an ample volume to monitor the sedimentation process with a minimum impact on the total volume. To accommodate for this a custom made sampling system was build comprising of two separate lines. One line was the *Hallberg Collecting System* (HACS) and the other was the sampling tank (ST) as described in Fig. 12. The HACS consisted of six individual vessels with a volume of 175 l. In all sedimentation trial less than 3 % of the total volume of a vessel was used for turbidity measurements. The ST was made up of a single tank with a volume of 1000 l. The flow was adjusted so that the filling time for the HACS and ST was the same. The runoff water filled the vessels of the HACS consecutively by use of a floating switch in combination with a check valve mounted in the individual vessels. The same floating switch was used in the ST.

Two pumps of type Flygt SXM 3 and Flygt SXM 2 were placed in the intake chamber. The pumps were located approximately 0.5 m above the intake chamber floor. The Flygt SXM 3 lifted the water to the HACS. The Flygt SXM 2 pumped to the ST. Sampling 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 valves controlled the flow to the HACS and ST. All material was PVC, PP or PE with exception for the two pumps of stainless steel. Sampling during sedimentation was executed in the HACS from seven taps on the individual vessels (Fig.12). The distance between the taps (centre to centre) was 100 mm. After each trial the HACS and ST was dismantled and thoroughly washed and hosed down with potable water.

Turbidity measurements

A HACK 2100P ISO turbidity meter was utilised 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 1000 FNU with a resolution of 0.01. The sample volume was minimum 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 by assessing the deviation from the standard solutions 0.1 FNU, 20 FNU, 100 FNU and, 800 FNU.

Fredhäll

Part of the experimental equipment can be seen in Fig. 13.

On-line measurements

Conductivity

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

Continuous flow measurement

Flows between 1 m^3/h-60 m^3/h were measured using a Thompson weir in combination with a pressure gauge, of type Cerlic FLX, with a measuring range of 0 m -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

In order 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.

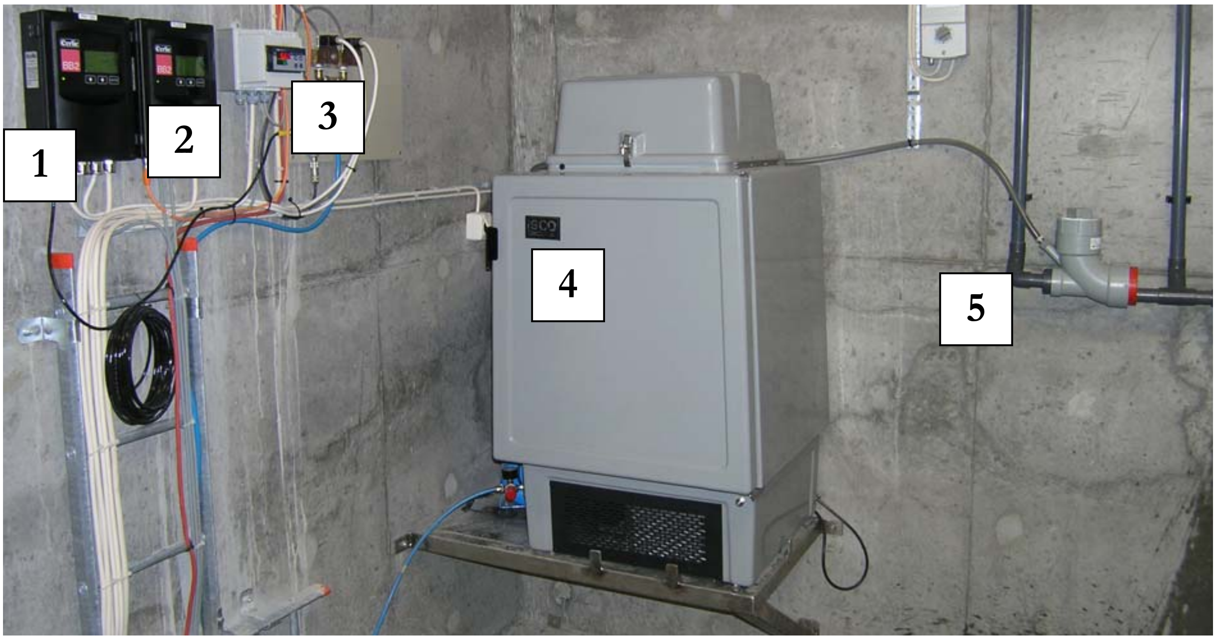

Figure 13 *Equipment for on-line measurements and sampling, pH and TSS [1], Flow [2], Conductivity [3], Sampler (ISCO3700FR) [4], Sampling loop [5]*

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 was 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 to the sampler, flushing was executed with three sampling tube volumes. The time for the rinsing cycle was 30 s and the sample volume was 800 ml. Between runoff events the water sampling pump was started every six hours for 60 s to flush the sampling loop.

PAPER OVERVIEW

An overview of appended papers is given here.

Paper I

In Paper I the possibilities for utilizing the first flush to optimise a treatment system are discussed. If the mass transport in the runoff displays emphasised first flush behaviour only a part of the total volume needs to be captured for treatment. Thus, a lesser sedimentation basin would be necessary. The process of sedimentation should be applied for primary removal of particulate pollutants as a pre-treatment step before consecutive units such as filters. Data for the dissolved fraction for Cr, Cd, Cu, Zn and Pb was obtained for annual average daily traffic loads (AADT) of 20,000, 71,000 and 120,000 based on data from three Swedish studies. For the highest AADT the dissolved fraction was between 24 % (Pb) and up to 50 % (Cd). It is not explicitly stressed in Paper I, but optimisation of the sedimentation unit is crucial, when land use is restricted, for removal of the particulate matter before treatment in a filter unit.

Paper II

In Paper II the EU Directive 1991/271/EEC requirements for discharge from a waste water treatment plant for TSS of 60 mg/l was used to assess mass transport in regards to treatment options 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 need for treatment. The measured TSS mean EMC for the studied events (Table 3) was 670 mg/l.

It was found that in the majority of the studied runoff events TSS exceeded a concentration of 60 mg/l. A definition of the first flush is given by Bertrand-Krajewski *et al.* (1998). Other definitions exists but Bertrand-Krajewski *et al.* (1998) is the most stringent, and evident, in regards to a minor portion of the total volume (X=20 %) carrying a major portion of the total pollutant mass (Y>80 %). Moreover, if the first flush is not emphasised caution should be advised for use of first flush as one of the design criterion. In Table 4 it can be seen that in

***Table 3** Studied runoff events (Paper II)*

Date	*Type of runoff **	*Antecedent dry period (h)*	*Duration (h)*	*TSS EMC (mg l^{-1})*	*Total runoff volume (m^3)*
25 October 2004	*1*	*17*	*5.0*	*230*	*160*
30 November 2004	*2*	*863*	*32.5*	*440*	*640*
2 December 2004	*1*	*1.8*	*12.6*	*830*	*460*
3 December 2004	*1*	*8.4*	*14.9*	*280*	*400*
15 December 2004	*1*	*274*	*6.3*	*1,640*	*220*
15 December 2004	*1*	*9.3*	*3.2*	*950*	*80*
17 December 2004	*1*	*32.0*	*3.6*	*1,670*	*16*
18 December 2004	*1*	*31.3*	*14.6*	*520*	*330*
22 December 2004	*3*	*76.3*	*15.4*	*870*	*740*
23 December 2004	*3*	*13.0*	*5.4*	*500*	*150*
30 December 2004	*3*	*152*	*22.0*	*410*	*540*
2 January 2005	*1*	*51.3*	*5.3*	*800*	*380*
4 January 2005	*1*	*54.7*	*10.1*	*580*	*420*
6 January 2005	*1*	*25.3*	*4.3*	*760*	*215*
8 January 2005	*1*	*46.6*	*5.7*	*920*	*270*
8 January 2005	*1*	*1.0*	*15.9*	*620*	*850*
10 February 2005	*3*	*538*	*8.8*	*1,800*	*610*
7 March 2005	*2*	*597*	*2.7*	*300*	*20*
11 March 2005	*2*	*90.0*	*7.4*	*960*	*180*
16 March 2005	*3*	*113*	*5.5*	*1,020*	*140*
17 March 2005	*3*	*10.4*	*16.3*	*700*	*450*
22 March 2005	*2*	*112*	*8.8*	*470*	*120*
23 March 2005	*2*	*15.1*	*9.1*	*400*	*140*
24 March 2005	*2*	*14.0*	*12.6*	*390*	*190*
25 March 2005	*2*	*11.0*	*13.1*	*210*	*170*
26 March 2005	*2*	*11.7*	*9.0*	*50*	*730*
7 April 2005	*1*	*272*	*4.9*	*592*	*180*
7 April 2005	*1*	*5.8*	*1.9*	*<10*	*200*
14 April 2005	*1*	*166*	*4.0*	*1,010*	*200*
14 April 2005	*1*	*1.3*	*9.7*	*280*	*220*

** 1 = Rain. 2 = Snowmelt. 3 = Rain combined with snowmelt* none of the studied runoff events a total pollutant load exceeding 80 % (Y) was carried in a total relative volume less than 20 % (X). The mass transport based on the findings in Paper II infers that a capture of the total runoff volume is necessary in this type of stormwater in winter. In addition, the elevated levels of TSS (EMC > 60 mg/l) were found for the majority of the runoff events. This demonstrates the necessity of a sedimentation unit before a filtration unit as discussed in Paper I.

Table 4 *Minimum and maximum values of relative mass of suspended solids (Y) for 30 studied runoff events for assessment of first flush criteria in regards to the relative volume (X)*

X	*Average (Y)*	*Median (Y)*	*Minimum (Y)*	*Maximum (Y)*
0.2	*0.17*	*0.16*	*0.04*	*0.40*

Paper III

In Paper III the seasonal variations between summer and winter conditions were studied for EMC, dissolved metal concentration, metal content (mg/kg) of the particulate material, and the correlation between TSS and total metal concentration. During winter, the asphalt surface is exposed to studded tyres and salt is regularly utilized as a de-icing agent on the highway. Both of these factors are expected to have a noticeable impact on the runoff water quality. The EMC was higher during the winter as compared to the summer for the studied stormwater events. This would suggest an elevated pollutant load during winter, which could be expected. The dissolved part 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 regards to the dissolved part for Cu, Pb and Zn. A higher dissolved part during summer was found for Fe ($p<0.01$). Throughout 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 ($p<0.01$). Compared to the findings of Stockholm Vatten (2001b) the mass concentrations found in this study was elevated both for summer and winter with the exception for Pb. The catchment area (Stockholm Vatten, 2001b) was located in the same geographical area and had the same AADT as the studied catchment area in Paper III. The findings in Paper III were based on runoff water samples and not from sediment samples, as was the case in the Stockholm Vatten (2001b) study. According to Stockholm Vatten (2001b) 65 % of TSS was reduced in the stormwater treatment plant. The removal efficiency could affect metal content in the sediments if mass concentration of metal pollutant increases with decreasing particle sizes. Another possibility can be changes in the pollutant sources since the execution of the study in 1994-1995. This could be the case with lead that was phased out during 1994. Paper III suggests variations in the metal content between summer and winter. Furthermore, according to Stockholm Vatten (2001b) traction sand was used during winter. Tractions sand was not used in the studied watershed in Paper III. Moreover, as suggested in Paper III difference in the atmospheric downfall could attribute to the variations for the studies. In addition the studied watershed includes a tunnel section, which makes up 36 % of the total road surface. A possibility could be the accumulation of airborne particles, typically less than 10 μm, in the tunnel section. During runoff events the traffic will spray the walls and roof of the tunnel and thereby "washing" off the accumulated pollutants. The contaminant concentration (mg/kg) in these finer particles could contribute to the elevated levels of metals in the particulate material compared to Stockholm Vatten (2001b). There is a tunnel section in the watershed (Stockholm Vatten, 2001b) but it represents less than 5 % of the total surface area.

Stormwater total metal concentrations and TSS concentrations was found to be correlated during summer and winter. In winter

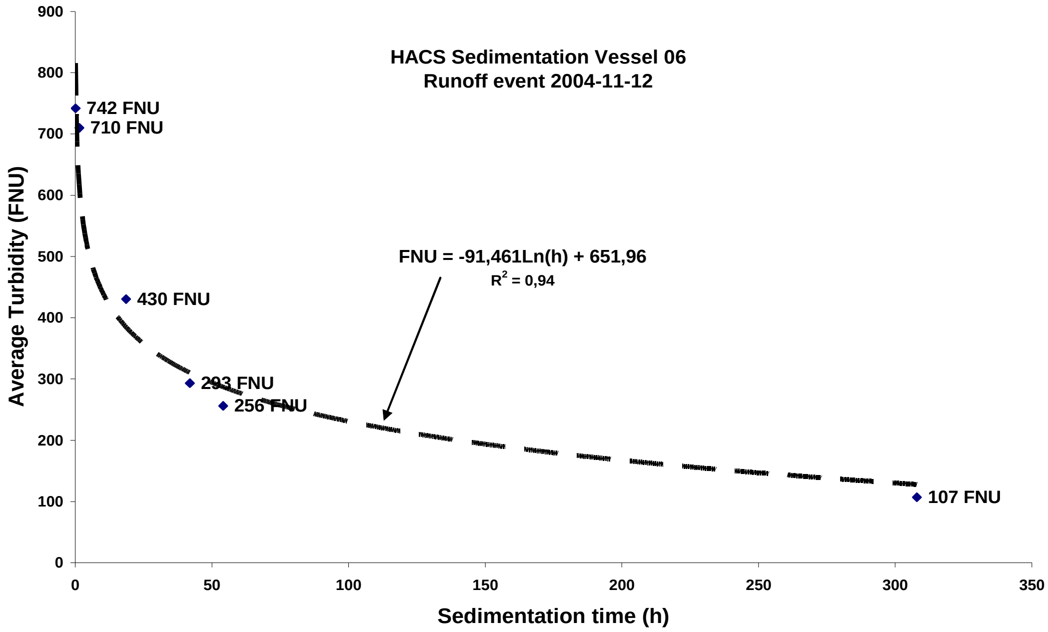

Figure 14 *Turbidity during sedimentation. Fitted line is described by [— —]*

the linear correlation for the total metal concentration and TSS showed a correlation factor (r^2) equal or greater than 0.98 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. Thus, it should be feasible to assess the pollutant transport for metals based on measurements of TSS. Furthermore, these findings can be used as a modelling input (e.g. Yuan *et al.*, 2001). The findings of Paper III suggest that a successful treatment of the studied metal pollutants could be carried out by means of sedimentation. However, depending on discharge criteria, the elevated levels of dissolved matter, especially during winter, have to be considered with regards to the selection of the appropriate water treatment process.

Paper IV

Sedimentation behaviour of runoff was studied, and the influence of winter and summer conditions were investigated and compared. Turbidity was used for assessment of the sedimentation process. The average turbidity in the sampling vessel was plotted against time. It was found that the sedimentation process could be described, for all the studied storm events, by a logarithmical expression as exemplified by the sedimentation trial in November 2004 (Fig. 14).

Based on the data from the sedimentation trials the surface load was calculated to describe the sedimentation process. It was found that the sedimentation behaviour could be described by initial turbidity. The use of road de-icing salt had a significant impact on the sedimentation process. Turbidity correlated well with total suspended solids (r^2 >0.90). However, a difference between winter and summer was found and suggested to be attributed to the finer particle size distribution during winter (Table 5). The findings in Paper IV suggest that the

***Table 5** Maximum particle size in 10 %, 50 % and 90 % of the total volume d(0.1), d(0.5) and d(0.9) respectively*

Field Trial	*d(0.1) (µm)*	*d(0.5) (µm)*	*d(0.9) (µm)*
2004-10-24	*	*	*
2004-11-12	*2.0*	*10.2*	*40.8*
2004-11-28	*2.0*	*7.5*	*24.0*
2005-01-10	*2.0*	*8.4*	*25.4*
2005-02-10	*1.0*	*3.4*	*11.4*
2005-03-17	*1.7*	*6.0*	*23.0*
2005-05-28	*3.1*	*16.1*	*73.7*
2005-06-22	*	*	*
2005-08-26	*4.1*	*23.0*	*118.0*

** missing data*

sedimentation properties for a specific surface load can be estimated from initial turbidity and concentration of total suspended solids for this type of highway runoff. The variation in flow and concentration of TSS during the runoff events does not affect the sedimentation properties suggesting that the water quality and the particle size distribution are similar over the duration of the runoff event. De-icing salt (NaCl) was shown to have a "positive" influence on the sedimentation properties in increasing the rate of sedimentation. The results from this study could be used for estimation of sedimentation properties in this type of runoff.

RESULT AND DISCUSSION

Mass transport

The findings in Paper I identify the need for detailed studies of first flush phenomena and in particular during the winter period when a higher pollutant load can be expected. In Paper II 30 consecutive runoff events in winter conditions was studied. Different hydrological definitions of the first flush effect exists, some of them stringent other more general. This presents a problem since no unified definition exists and the stringent definitions such as e.g. Bertrand-Krajewski *et al.* (1998) is less applicable for assessing design implications for treatment systems. The problem was addressed in Paper II by selecting a discharge demand for TSS of 60 mg/l (EU Directive 1991/271/EEC). In Paper II the PEMC for TSS was calculated from the end of the runoff event to study if the mass transport for the latter part of the runoff volume was below the target concentration of 60 mg/l. For the studied winter season it was concluded (Paper II) that the entire runoff volume needed to be captured for treatment. In regards to the definitions of Bertrand-Krajewski *et al.* (1998) no evident first flush could be found in the study (Paper II)(Table 4), on the contrary. These findings concur with several studies that imply only a diffuse and not a pronounced first flush as defined by Bertrand-Krajewski *et al.* (1998) for highway watersheds. Sansalone and Buchberger (1997b) studied a 300 m^2 road section with an average daily traffic count of 150,000 vehicles. The study was based on five separate runoff events. A first flush effect was found for dissolved Zn and Cu. For the particulate matter the first flush effect was related to the dissolved fraction and thus not well defined. Barett *et al.* (1998) studied three catchment areas with a runoff surface of 526 m^2, 5,341 m^2 and 104,600 m^2 with an average daily traffic of 8,780, 58,150 and 47,240 vehicles respectively. Barett *et al.* (1998) only noted a first flush phenomenon in storm events of short duration and constant rainfall in the watershed with the highest traffic flows. The main reduction of TSS occurred during the first 5 mm and then stabilized at elevated levels for the duration of the runoff event. Deletic (1998) studied two small catchments areas, a medium trafficated street with a surface area of 211 m^2 and a parking lot with an area of 270 m^2. In order to asses the first flush phenomena the pollutant mass carried in 20 % of the total volume was studied (Deletic, 1998). It was found that a slight first flush effect for suspended solids could be observed. Furthermore, in an extensive study by the UK Environmental agency (UK Environmental Agency, 2003) the overall effect of the first flush was small or negligible The study was based on watersheds with an average daily traffic ranging from about 20,000 vehicles to 80,000 vehicles.

Further investigations comparable with the ones executed in Paper II are necessary to evaluate seasonal variations in mass transport from similar watersheds. This is emphasized by seasonal variations in TSS and metal load found in Paper III. Moreover, studies for assessment of the applicability of models for calculating the total mass of solids as suggested e.g. by Lord (1987) in this type of watershed are of interest for different times of the year.

Seasonal variations

The findings in Paper III for EMC of TSS confirm the elevated pollutant load during winter as shown in other studies. Furthermore, in Paper III the dissolved metal concentration (µg/l) and particulate metal content (mg/kg) was studied, as was the correlation of TSS to the total concentration of individual metals during winter and summer. The study showed that the metal content in the particulate material was similar between the runoff events in the respective season. However, significant differences between winter and summer were found. A number of studies have been executed showing the partition between particulate and dissolved matter in highway runoff. However, the study in Paper III has a novel approach by comparing the metal content during a runoff event and also between the storm events for assessing the transported particulate matter. Paper III imply a good correlation for total metal concentration and TSS concentration for the studied highway runoff. The found correlations in Paper III would imply that TSS could be used as a substitute parameter for assessing metal pollutant loads in comparable catchment areas. Thomson *et al.* (1997) used databases comprising of data from rain events, snowmelt events and combined snowmelt and rain events ("mixed events") for establishing correlations between TSS and metals. One evaluated catchment had an area of 6.60 ha with an average daily traffic of 65,000 vehicles. A total of 112 runoff events were elaborated on for the catchment area. As can be seen from Table 6 and Table 7 k values (Paper III) and β values (Thompson *et al.* 1997) for Cr, Cu, Fe, Ni and Zn are in the same range. However, for Al, Pb, and Cd there were notable differences. It is important to recognize that Thomson *et al.* (1997) also included TOC and TDS for correlation of surrogate parameters. In regards 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 minimal. For metal constituents relationships (e.g. Zn, Ni, Cu, Pb) Thomson *et al.* (1997) found good near-site portability. The Thompson findings would infer that the findings of Paper III would be transferable to highly trafficated roads in similar watersheds.

Table 6 *Correlations for metals (Thomson et al., 1997), TSS (mg/l), TOC (mg/l), TDS (mg/l)*

Metal (µg/l)	β_1	β_2	β_3	r^2
Al	1.995	2.819*TSS		0.846
Cd	0.0115	0.00371*TSS		0.783
Cr	0.0632*TSS	0.00988*TDS		0.949
Cu	10.91	0.248*TSS		0.896
Fe	40.05*TSS	1.448		0.968
Ni	2.12	0.0449*TSS	$6.08*10^{-4}$*TOC	0.879
Pb	3.845*TSS	0.274*TDS		0.959
Zn	0.955*TSS	0.0749*TDS	3.672*TOC	0.910

Table 7 *TSS and total metal concentration (Paper III) Me = k*TSS [mg/l]+M P=Period, S=Summer, W=Winter*

Metal µg/l)	*k*	*M*	*r²*	*P*
Al	*19*	*520*	*0.96*	*S*
Al	*25*	*2300*	*0.99*	*W*
Cd	*0.0005*	*0.08*	*0.82*	*S*
Cd	*0.0004*	*0.30*	*0.92*	*W*
Cr	*0.09*	*4.3*	*0.84*	*S*
Cr	*0.08*	*9.4*	*0.99*	*W*
Cu	*0.44*	*29*	*0.92*	*S*
Cu	*0.25*	*14*	*0.99*	*W*
Fe	*36.5*	*1100*	*0.93*	*S*
Fe	*46.4*	*1570*	*0.99*	*W*
Ni	*0.03*	*3.8*	*0.93*	*S*
Ni	*0.03*	*5.3*	*0.99*	*W*
Pb	*0.10*	*2.6*	*0.89*	*S*
Pb	*0.06*	*1.0*	*0.99*	*W*
Zn	*1.76*	*72*	*0.94*	*S*
Zn	*1.15*	*132*	*0.99*	*W*

In the studied catchment area 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 wear of roads (VTI, 2005; Kupiainen *et al.*, 2003; Kupiainen and Tervahattu, 2005). Consequently there is a further need to study the impact of traction sand on roads with elevated traffic loads. Although, Paper III indicate a correlation between the studied metals and TSS further studies to evaluate the e ffects of TOC and TDS are of interest in comparable watersheds. In addition, further studies as executed in Paper II are necessary but also for roads were traction sand is utilised for evaluating seasonal variations.

Sedimentation process

In Paper IV the sedimentation process during winter and summer was studied. In Paper III the findings suggest that the source of the pollutant differs between the seasons but that there is a consistency in material composition for a specific season. Paper IV implies that the particle size distributions are similar within a season but differs between winter and summer (Table 5). The particle size distributions found in Paper IV were in agreement with the particle size distributions found by Andral *et al.* (1999) and Roger *et al.* (1998). The results of Paper IV show that the sedimentation properties in winter and summer can be estimated from the initial concentration of TSS. Studies of sedimentation behaviour in runoff have been executed in laboratory studies (Whipple and Hunter, 1981; Randall *et al.*, 1982; Urbonas and Stahre, 1993). Whipple and Hunter (1981) collected a total of four samples during runoff events from two urban streams and two shopping malls for sedimentation trials. The finding of the study was inconclusive in regards to settling properties and particle concentrations. However, 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. The findings of Randall would thus be in accordance with the findings of Paper IV. The results in Paper II demonstrates a magnitude of 10 in the variations in mass transport between the minimum and maximum relative mass of pollutant transported in 20 % of the total runoff volume (Table 4). However, the results in Paper III and Paper IV would suggest that the particulate material is consistent during the runoff event and differs only in concentration for a specific season. Furthermore, this could infer the same dependence of sedimentation properties for highway runoff in similar catchment areas as described by Andral *et al.* (1999) and Roger *et al.* (1998). An explanation for this could be that the traffic is consistent and dominant as a pollutant generator, particularly during winter (e.g. Glenn and Sansalone, 2002). During summer the pollutant generation is less from the traffic and as a consequence the impact of atmospheric downfall increases in importance as noted in Paper III. Traffic as a constant source of pollutants was also suggested by e.g. Barett *et al.* (1998a). Furthermore, the findings in Paper IV suggested that salt had an influence on the sedimentation. Elevated concentration of NaCl increased the sedimentation velocity of the particulate matter.

Implications for design of treatment systems

Paper IV provides a complement to the existing design criteria as to the dependence of initial concentration for the sedimentation process. The possibility to use concentrations is important mainly for two reasons (i) detailed design of a sedimentation unit i.e. be able to estimate sedimentation performance for single runoff events and (ii) knowledge of the sedimentation properties allows for implementation of process control. Furthermore, Paper IV also shows that it could be possible to use turbidity as a measurement for TSS. Turbidity measurements provide the possibility for on-line measurements which are of interest for process control. The findings of Paper III suggest that, in particular, during winter time the metal pollutant load could be indirectly monitored by on-line turbidity measurements. The study in Paper IV is novel in its execution and further studies are of interest in similar watersheds. Modelling of existing dams or ponds should also be studied to evaluate efficiency factors for the found sedimentation properties as well as incorporating the findings in models for design of treatment systems.

CONCLUSION

The study off mass transport showed that the majority of runoff events displayed higher concentration than the reference value of 60 mg/l during winter. Seasonal variations were found for the dissolved part of the metals and also for the particulate matter. The studied metals were mainly associated with the particulate material. The study further suggests that sedimentation process can be described by initial turbidity and concentration of total suspended solids. Good correlation was implied for turbidity and TSS. The metal pollutant load could be assessed indirectly from the measurement of TSS during winter, without considering Cd and Co. Furthermore, the metal pollutant load of Al, Cu, Fe, Mn, Ni, Zn and possibly Pb, can also be determined by measuring TSS in summer. The generated particulate matter differs between the seasons but appears similar within the season for the studied runoff. A successful treatment of the studied metal pollutants could be carried out by means of sedimentation. However, depending on discharge criteria, the elevated levels of dissolved matter, especially during winter, have to be considered with regards to the selection of the appropriate water treatment process. In addition, the study implies that the entire runoff volume must be treated and that the use of first flush as a design criterion is less applicable for the winter period in this type of runoff.

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