PROPER PROJECT
WP2 - ASSESSING THE VULNERABILITY OF EUROPEAN WATER BODIES FOR ROAD RUNOFFS DURING BUILDING AND OPERATING OF ROADS

Task 2.4. An evaluation of 'real world' vulnerability assessments of surface and ground water bodies to road-related pollution: case study database and critique to identify knowledge gaps

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Title

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Task 2.4. An evaluation of ‘real world’ vulnerability assessments of surface and ground water bodies to road-related pollution: case study database and critique to identify knowledge gaps

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Abstract

This report provides an overview of available case studies evaluating the effects of road runoff on the water quality in the watercourses and settlement tanks into which the runoff is discharged and impact on groundwater quality near roads. The first part of the report briefly describes the contaminants contained in the road runoff, their sources and factors that can influence the quality of the runoff. The following sections describe case studies that assess the impact of runoff on the quality of surface and groundwater and on aquatic organisms. The report includes results of 30 case studies from around the world which were identified as the most important ones based on their complex approach in description of impacts on living organisms and water quality. 23 of these studies deal with the influence of runoff on surface water. These studies that were conducted in the USA, Canada, Norway, UK and the Czech Republic, mostly included both the results of chemical analysis of surface water and runoff, as well as the results of toxicological analyses or biological monitoring. 10 studies were focused on groundwater issues namely contamination with chlorides and heavy metals. These studies were carried out in Canada, the USA, UK and Portugal. A weakness within the available literature is the narrow focus mostly only on a limited set of basic parameters.

Keywords: Road runoff, pollution, surface water, groundwater, ecotoxicity, biodiversity

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Acronyms and glossary

AADT	Annual Average Daily Traffic
ADP	Antecedent Dry Period
ADT	Average Daily Traffic
ASPT	Average Scores Per Taxon
BMWP	Biological monitoring working party scores
BOD	Biochemical Oxygen Demand
CCA	Canonical Correspondence Analysis
CCI	Chloride Contamination Index
COD	Chemical Oxygen Demand
DOC	Dissolved Organic Carbon
EC	Electrical Conductivity
EMC	Even Mean (Median) Concentration
EQS	Environmental Quality Standards
GTA	Greater Toronto Area
HEM	Hexane Extractable Materials
HMM	High Molecular Mass species of PAHs
LMM	Low Molecular Mass species of PAHs
MFO	Mixed Function Oxidase test
PAHs	Polycyclic Aromatic Hydrocarbons
PCA	Principal Component Analysis (multivariate statistical techniques)
PH	Purgeable Hydrocarbons
PWS wells	Public Water Supply wells
SPDM	Semipermeable Membrane Devices
SS	Suspended solids
TC	Total Hydrocarbons
TDS	Total Dissolved Solids
THP	Total Petroleum Hydrocarbons
TKN	Total Kjeldhal Nitrogen

TOC	Total Organic Carbon
TP	Total Phosphorus
TS	Total Solids
TSS	Total Suspended solids
USEPA	U.S, Environmental Protection Agency
VDS	Vehicles traveling During a Storm
VOCs	Volatile Organic Compounds

1 Introduction

The rapid growth of transport capacity and the quantity of the passenger and freight vehicles poses an increased environmental burden. Hence compounds emitted from all transportation modes occur in all environmental compartments. These compounds are widely reported to have a range of adverse effects on ecosystems, animals, plants and human health hence it is important to observe their occurrence in the environment (Bencko et al., 1996, Weijer et al., 2001, Tucková et al., 2001). In connection with an increasing environmental burden by transportation, increasing levels of air pollution is the most widely understood impact (Barek et al., 1998). However, the pollution also impacts other environmental compartments, such as surface and ground water, soil and biota. The source of contamination includes the emission of polluting substances from winter road maintenance and operation of vehicles or materials used for road construction both splashed by runoff waters, as well routine driving activities. The acute or chronic impact on the biota in the recipient depends on character of the use of a road, its surrounding land use, the type of a road, the rainfall characteristics and the type of receiving water body (Crabtree et al., 2009). Runoff from roads has been identified as a significant contributor to contamination of receiving waters (Gavens et al., 1982, Yousef et al., 1984).

Pollution of surface water by road runoff waters originates mostly from roads with high traffic intensity, especially highways and high-speed roads (see D1.1 and D2.1). Levels of pollution is influenced by the amount of precipitation falling on a road surface, where the concentrations of pollutants are the highest in the "first flush" (intense precipitation after a long dry period) with decreasing concentrations reported over time. Highest levels of pollution have been demonstrated directly behind the drain where the run-off water is not yet highly diluted (Kayhanian et al., 2012). A number of pollutants, such as metals, hydrocarbons and suspended particulate matter are generated by traffic, particularly by road surfaces and abrasion of tires. An important source of pollutants is also spills and leakage of fuel, when the range of pollutants, such as PAH, oil and grease, total petroleum hydrocarbons (TPH) and metals are released to the environment (Shinya et al.,2000).

Due to maintenance of roads, rest areas/stations and parking lots, contamination associated with the application of de-icing agents and anti-freeze mixtures occurs in winter. These activities are the main sources of chlorides, which can absorb in soil and rock environment and accumulate here under suitable conditions and then are gradually released into groundwater (Runge et al., 1989).

In addition to routine operations, areas surrounding roads, including ground water and surface water, may be also contaminated as a result of road construction activities. For example, water migrating into and out of the road body materials, can mobilise and transport pollutants released from these materials. However, this process depends on many factors, such as the nature of the subsoil and the surrounding terrain, the groundwater regime, including the capillary flow and the amount of precipitation that can infiltrate into the road body, particularly due to damage to its surface layer (cracks and tear) (Ličbinský et al., 2012).

The report presents and summarizes the results of a series of case studies that present the results of "real world" vulnerability assessments of surface and ground water bodies to road-related pollution. The report is conceived as an overview of the current state of knowledge within this issue written on the basis of available scientific literature and provides a comprehensive overview of field work undertaken within highway receiving water vulnerability assessments to-date.

2 Contaminants in runoff and their sources

2.1 Contaminants

The event mean concentration (EMC) is often used to characterize runoff water quality and serves to compare data from different locations and events in a comparative format and to evaluate the total pollutant load of an event entering a receiving water. EMCs represent a flow average concentration computed as the total pollutant load (mass) divided by the total runoff volume (Barrett et al., 1998).

2.1.1 Metals

Runoff water from roads maybe polluted by many chemicals. Metals are the most widely monitored and investigated group of contaminants. In contrast to organic pollutants (many of which can break down to simpler compounds), metals are persistent in the environment with only their forms of occurrence changing. A significant proportion of metals entering the environment from transport pass into or through surface water ecosystems. Here they may be taken up the aquatic environment by aquatic plants and/or animals. They tend to accumulate in sediments, which are deposited either in the treatment systems or are transported directly into the watercourses (depending on the type of road drainage). Many metals preferably bind to settled and/or suspended particulate matter (e.g. lead). However, some also remain in the dissolved form (e.g. cadmium) (Kayhanian et al., 2012, Shinya et al., 2000, Sansalone and Buchberger, 1997, Legret and Pagotto, 1999). When changing the physicochemical properties of the environment (e.g. pH, redox potential, oxygen ratios), metals previously bound to particulate mass may be released into the dissolved phase. This makes them potentially accessible to organisms living in water (Kayhanian et al., 2003; Sansalone and Buchberger, 1997). The most important metals occurring in watercourses and negatively affecting the quality of aquatic ecosystems are Cd, Cr, Cu, Ni, Pb and Zn. Cd, Cu and Zn are often present in a dissolved form while Fe, Al and Pb are mainly particulate bound (Sansalone and Buchberger, 1997). The behavior of metals within the aquatic environment is diverse, especially when there are frequent, rapid and sudden changes in conditions (antecedent dry period, storm events, rainfall events). Significant changes in flow and physicochemical properties of the aquatic environment occur especially in small watercourses both in urbanized and non-urbanized areas.

2.1.2 Organic compounds

Organic substances are often studied in runoff waters include polycyclic aromatic hydrocarbons (PAHs). They enter aquatic environment mostly due to oil spills, atmospheric deposition and accidents. The most PAHs found in highway runoff are pyrolytic (combustion related), while petrogenic (non-combustion-related) PAHs are found in lower concentrations (Lau et al., 2005). In runoff waters, they are mainly particle-bound and their concentrations strongly correlate with those of certain size fractions of suspended solids, while those in the aquatic phase are generally near detection limits (Opher and Friedler, 2010). Shinya et al. (2000) reported that the higher molecular weight PAHs were more associated with suspended solids in the runoff. Phenanthrene, fluoranthene and pyrene comprised about 50 % of fifteen quantified PAHs constituents in each sample (Shinya et al., 2000). Chen et al.

(2004) reported that PAHs are strongly adsorbed to organic sediment and soil fractions. Consequently, it can be concluded that sediments and soils are usually considered as sites of storage in the environment, and PAHs with 4 or more aromatic rings are more easily susceptible to sediment sorption and are persistent in the environment. Concentrations of PAHs and other pollutants in runoff water from pavements next to a motorway were studied in France. The highway daily traffic intensity was approximately 12,000 vehicles, of which about 7% were freight vehicles. A 275 meter long section of motorway was observed for one year, within which 50 precipitation events were recorded. Two different types of pollution have been revealed. One type can be defined as chronic and includes suspended solids, chemical oxygen demand, total hydrocarbons, Zn and Pb. The second type can be considered to be seasonal and incorporates chlorides, sulphates, suspended solids and heavy metals due to the use of de-icing salt in winter (Legret and Pagotto, 1999).

2.1.3 Suspended solids

The presence of solids in runoff can damage the river ecosystem by increasing turbidity and clogging of basal substrates. Solids also transport adsorbed pollutants e.g. metals which can contribute to instream acute or chronic toxicity. Their particle size determines how easily they are transported in the aquatic environment. Particles <63 μm are transported within flows the most easily and are associated with higher concentrations of pollutants than larger solids (Pontier et al., 2001).

It was found that the metals and suspended solids in the highway runoff had a very strong correlation in case of snow melting, but a low correlation in rainfall episodes. The same results were observed for the correlation between heavy metals and the size of suspended solids. From a physical point of view, this is expected because heavy metals and suspended solids are accumulated in snow piles in close proximity to the road for a longer period of time. It is believed that these metals will preferably be bound to finer and more surface-active solids (Sansalone and Buchberger, 1995).

2.1.4 De-icing materials

The surrounding area of roads is continuously loaded by the input of salts coming from winter road maintenance. The main contaminant from the road salts is the chlorine ion Cl^- (chloride), which passes through the environment without significant changes (in terms of chemical reactions, physical bonds and biological utilization). Chloride ions are transported from the road to surrounding soil, surface and ground water. Increased chloride ion content has a negative effect on ecosystems in the vicinity of roads. It changes the species composition of plant and animal communities of terrestrial and aquatic ecosystems (Hosek and Kaufman, 1992; Vyhnalek and Duskova, 2002). Extremely high concentrations of chloride ions in water have a toxic effect to organisms (Marsalek, 2003). Increased concentrations of chlorides in the outflow runoff from roads can cause the mobilization of metals (e.g. Cd, Cu, Pb and Zn) from soil into the water near the road (Backström et al., 2004). Huber et al. (2014) published results concerning the remobilization of Cu and Zn by NaCl and $CaCl_2$ for two soils and five filter materials used for stormwater treatment. In some cases, both heavy metals were released, and in some cases, only Zn showed higher mobility. These effects depended on the filter material and the de-icing salt type, e.g., some heavy metals were not released by NaCl alone but by a mixture of NaCl and $CaCl_2$. In addition, the pH of the effluent was often reduced by the use of de-icing salts.

Alternative de-icing agents for winter road maintenance are used including calcium chloride, magnesium chloride, urea, alcohols and glycols, calcium magnesium acetate and potassium acetate.

As an alternative to de-icing agents, salt – gravel mixture is used as well. Because salt concentrations in this mixture are lower, salinity and cyanide toxic shock should not be a problem (Novotný et al., 1998).

2.2 Sources of contaminants

Major sources of pollutants on highways are vehicles, dust, aerial deposition and precipitation. Many factors affect the type and amounts of these pollutants, including traffic volume and traffic flow composition, local land use, and weather patterns. Other possible sources of pollutants include spills of the operating liquids, accidents, particularly those carrying agricultural or chemical products. These losses are related to traffic volume and may go unnoticed but could result in a large pollutant load locally (Asplund et al., 1980). Roadway maintenance practices such as sanding and de-icing, may also act as sources of pollutants (Barret et al., 1993).

2.2.1 Vehicles

Sources of pollutants from vehicles include leakage of fuel, lubricants, hydraulic fluids; particles from abrasion of tires,clutch and brake linings; particulate exhaust emissions; dirt, rust, and decomposing coatings that drop off of fender linings and undercarriages. Among the elements contained in fuels (and thus in exhaust fumes) in measurable concentrations are: Cd, Cr, Cu, Ni, Pb, and Zn, which are commonly studied, as well as Al, Br, Ca, Co, Fe, K, Li, Mg, Mn, Na, Pt, Sb, Sr, Ti, and V. For many decades lead was added to vehicle fuel in average concentration of about 400 mg/L, resulting in an the emission of an estimated 7 million tons of Pb burnt as fuel additives between 1926 and 1985 in the United States alone and in an exposure to hazardous levels of environmental lead by large parts of the population (Nriagu, 1990). Contents of selected heavy metals in vehicles and road materials are presented in Table 1. Tire wear contributes oxidizing rubber components and zinc oxides. The quantity of matter resulting from wear of a light vehicle tire is assumed to be 68 mg/(km · vehicle) and twice that for heavy goods vehicles. The wear of brake linings is considered to be approximately 20 mg/(km · vehicle) for private vehicles, 29 mg/(km · vehicle) for light goods vehicles and 47 mg/(km · vehicle) for heavy lorries (Legret and Pagotto, 1999). Tire and brake wear were significant contributors to the load of copper on an urban road surface (47 %), while for the other metals these contribute only with 1–10 % (Davis et al. 2001). On motorways, brake linings were found to contribute nearly 100 % of Cu. Only 2 % of the expected annual load of Cu was detected in the storm water. Other studies have tried to quantify the amount of pollutants in highway runoff attributed directly to vehicles, but have been only partially successful (Opher and Friedler, 2010).

Table 1 Heavy metal contents in vehicle and road materials (mg/kg) (after Opher and Friedler, 2010)

Source	Pb	Cu	Cd	Zn
Vehicles				
Leaded gasoline	200	-	-	-
Unleaded gasoline	17	-	-	-
Brake linings	3900	142	2,7	21,8
Tire rubber	6,3	1,8	2,6	10,25
Road				
De-icing agent	3,3	0,5	0,2	0,5

2.2.2 Atmospheric deposition

Any pollutant present in the air over the road surface may settle upon it, a process often referred to as dry deposition. During rainfall further deposition (wet deposition) from the atmosphere takes place, as the rain washes down substances present in the atmosphere (Opher and Friedler, 2010).

It has been shown that atmospheric deposition is a very important source of cadmium loading and to a less extent for Cu and Pb (Davis et al., 2001). In rural highways, in the absence of buildings in the vicinity of the road, atmospheric deposition is expected to be the main source of cadmium. However, this has not yet been proven. Atmospheric deposition on impervious surfaces is a key source of mercury transported in urban runoff. Dry atmospheric deposition is also an important source of soluble, oxygenated VOCs (Opher and Friedler, 2010). Zhang et al. (2013) used multivariate statistical techniques (e.g. principal component analysis - PCA) to identify potential sources of pollution and found that dry and wet atmospheric deposition (TN, $NO_3^- - N$, $NH_4^+ - N$, dissolved Cu) was the most important source, followed by road sediment and materials (TSS, COD, and TP), and vehicle emissions (dissolved Pb and dissolved Zn) . The contribution of atmospheric deposition to emissions of trace metals in storm water runoff was investigated within a small, highly impervious urban catchment in Los Angeles (Sabin et al., 2005). This research demonstrated, that atmospheric deposition potentially accounted for 57–100 % of the trace metal loads in annual storm water discharges in this catchment and dry deposition appears to be the dominant mechanism. Atmospheric deposition was monitored by Aljazar and Kocher (2016) on three German motorways with an average traffic intensity of 70,000 vehicles per day. Bulk deposition was collected at two heights (1.5 m and 0.3 m above the ground) and in 1 m, 2.5 m, 5 m and 10 m distances from the road edge. The pollutant load in deposition measured near the ground surface was higher than those measured at 1.5 m above the land surface. At all three sites, a clear negative correlation between pollutant load and the distance from the roadside could be found. Individual elements showed a wide range of deposition values showing that the impact of traffic activities could reach large distances from the road edge. It was found that heavy traffic share clearly impacted deposition rate of traffic-related emissions. Wind and car-induced turbulence leads to a decrease in atmospheric deposition (Huber et al., 2016).

Many major ionic constituents originate from atmospheric pollution. Harrison and Wilson (1985) found that rainfall can contribute up to 78 % of the major ionic contaminants (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, and SO_4^{2-}) leaving the road surface in runoff and up to 48 % of the suspended solids.

2.3 Factors affecting highway runoff quality

There are many mechanisms for the removal of pollutants from highways. During dry weather, materials are continually blown on and off the highway, as well as on and off of vehicles by natural and vehicle-induced winds (Opher and Friedler, 2010). During wet weather storm water washes constituents off of the highway surface and simultaneously also off the traveling vehicles, onto the highway surface. Occasional road sweeping or other road maintenance activities add further complexity (Opher and Friedler, 2010).

The main factors influencing the runoff quality include traffic volume, precipitation characteristics, highway surface type and drainage mechanisms, traffic area categories and climatic factors (Barret et al., 1993, Opher and Friedler, 2010).

2.3.1 Traffic volume

Vehicles are one of the major sources of pollutants in highway runoff. The amount of traffic on a given highway section will influence the accumulation of pollutants on the highway surface. However, turbulence caused by vehicles can remove solids and other pollutants from highway lanes and shoulders, obscuring the relationship between traffic volume, pollutant loads, and concentrations in runoff. The volume of traffic is expressed on average daily traffic (ADT) (sometimes expressed as annual average daily traffic - AADT), which refers mainly to the dry period, and number of vehicles traveling during a storm (VDS). VDS is not often considered an optional variable for pollutant load prediction, because it is so closely related to the length of the storm, or simply because it is usually not readily available or difficult to measure (Opher and Friedler, 2010).

Speed limits and traffic signals indirectly affect runoff quality, because of higher breaks, break linings and tire abrasion due to braking and because of increased exhaust emissions from cars due to acceleration (Huber et al., 2016).

Review of available literature reveals a wide spectrum of conclusions as to the relationship between traffic volume and highway runoff pollution. Some findings indicate a strong correlation between the two, while others imply that such a correlation does not exist (Swadener et al., 2014). Most reports present findings of a weak relationship, in which traffic volume accounts for up to 40 % of the variability in pollutant concentrations in the runoff (Opher and Friedler, 2010, Drapper et al., 2000). Kayhanian et al. (2003) found that the average pollutant concentrations in runoff from urban highways (AADT > 30,000 vehicle per day) were two to ten times higher than those found in nonurban (AADT < 30,000 vehicle per day) highways. However, average concentrations of some pollutants - COD, TSS, TDS, turbidity, NH_3, and diazinon were found to be higher in runoff from nonurban highways than the runoff from urban highways, suggesting sources other than the transportation related activities. Kayhanian et al. (2012) reported that the concentration of metals on urban highways with AADT > 60,000 was higher than metal concentrations in rural highways with AADT < 30,000 in California. A higher level of motorway-derived heavy metal contamination was identified in storm water runoff from a road section with a higher average daily traffic density (Hares and Ward, 1999). Klimaszewska et al. (2007) found out that the concentration of heavy metals, pH and PAHs in runoff waters increased during the day, together with increasing traffic intensity. Huber et al. (2016) states in his work, that roads with more than 5000 vehicles per day are often more polluted than highway because of other site-specific factors such as braking and acceleration at traffic signals. Barret et al. (1995) compared the quality of the highway runoff in three places, which varied in traffic intensity, surrounding land uses, and highway drainage system types. He found that the mean traffic concentrations observed at the site with a medium traffic density are lower than those observed for the other two sites, including the low traffic site. It can be explained by the fact that the runoff from the highway at high traffic and low traffic sites is directly from the pavement to a catch basin where the samples are collected. However, the highway runoff at the medium traffic density site passes over approximately 60 m of grassy area (swale) before entering the storm drain pipe from which samples are collected. Highways which carry more traffic are usually those that are located in urban areas, whereas the lighter traffic-loaded highways are those that run through less polluted rural areas. Hence, it is often impossible to distinguish between the effect of the AADT and that of land-use on the concentrations of pollutants in the runoff (Kayhanian et al., 2003).

2.3.2 Precipitation characteristic

Three characteristics of a storm event which may be relevant to the ensuing water quality of runoff from a highway surface are: the number of dry days preceding the event, the intensity of the storm, and the duration of the storm (Barret et al., 1993).

Antecedent dry period (ADP) is a dry period preceding a runoff event. In this time pollutants accumulate on the road surface. The build-up occurs relatively quickly after a rain event but slows down after several days as redistribution occurs. The surface of the road has a maximum carrying capacity for dry matter, which is influenced by the type of asphalt and by ambient conditions contributing to the removal of sediments, such as wind, temperature, solar radiation and relative humidity (Opher and Friedler, 2010). In some studies, the relationship between the concentration of pollutants and ADT was found (Kayhanian et al., 2003, Opher and Friedler, 2009, Huber et al., 2016). Hewitt and Rashed (1992) found a highly significant correlation between the length of the antecedent dry period and the amount of lead and dissolved copper removed during a runoff event but this was not found for the other pollutants. According to other studies ADP has no effect on the concentration of pollutants (Crabtree et al., 2006, Harrison and Wilson, 1985, Swadener et al., 2014).

Attributes such as storm volume, rainfall intensity and storm duration may have various implications on the quality of runoff waters from road surfaces. Obviously all three variables are interconnected, as the storm volume is a function of its intensity and duration. A large-volume storm may be one of high intensity and short duration, or low intensity and long duration.

Rainfall volume was identified as one of the important parameters influencing the quality of runoff (Irish et al., 1995, Gan et al., 2008) On the other hand, Crabtree et al. (2006) found no relationship between pollutant concentrations and total event rainfall. While volumes may be poor indicators of pollutant concentrations in the runoff, they have been shown to be important factors in determining the total load of pollutants (Opher and Friedler, 2010).

The intensity of storm may have a marked impact on the type and quantity of pollutants in runoff. This is due in large part to the fact that many pollutants are associated with particles, which are more easily mobilized in high-intensity storms (Barret et al, 1993). It was found the positive correlation (Irish et al., 1995, Kayhanian et al., 2003, Crabtree et al., 2006) between certain pollutants loads or concentrations and storm intensity. The negative correlation (Kayhanian et al., 2003, Crabtree et al., 2008) was observed too. In the case of positive correlation, the constituents were associated with particles, indicating that higher rainfall intensities have the effect of mobilizing these particulate-associated parameters. Negative correlation was observed for dissolved constituents, indicating that the dilution effect is more common for parameters not associated with particulates (Kayhanian et al., 2003). It is generally accepted that constituent load (i.e. mass of constituent removed from highway per unit time and/or area) is positively correlated with rainfall intensity (Irish et al., 1995).

The storm duration is another important factor influencing the quality of runoff. Pollutant loads are generally higher in longer storms, as the transport of at least some constituents continues throughout the duration of the event, regardless of other factors such as intensity and flow (Barrett et al., 1995). Strong correlations exist between storm duration and other influencing factors, such as total rainfall or runoff volume, runoff flow or VDS. When a few variables are closely correlated it may be impossible to distinguish between their separate effects on the target variable (Opher and Friedler, 2010).

Pollutants that collect during a dry period on highways may tend to be carried off with the early portion of storm runoff, causing a “first flush” of pollutants. The first flush is a phenomenon in which a larger pollutant concentration or mass is discharged during the first portion of the storm water runoff compared with the rest of the runoff (Kayhanian et al., 2012). The first flush of highway runoff has been reported by numerous researchers (Sansalone and Buchberger, 1997; Mangani et al., 2005; Furumai et al., 2002; Barbosa and Hvittved-Jacobsen, 1999, Zhang et al., 2013, Yufen et al., 2008, Shinya et al., 2000, Di Modugno et al., 2015). The first flush effect had a significant influence on the removal of dissolved metals from road surface in the road runoff waters. This effect was clearly seen for the dissolved metals whilst the behaviour of the particle-associated material closely follows that of the total suspended solids (Hewitt and Rashed, 1992). Mass first flush effect for TKN (total Kjeldahl nitrogen), COD (chemical oxygen demand) and DOC (dissolved organic carbon) as well as Cu, Ni and Zn occurred more frequently than the other conventional and metal pollutants (Kayhanian et al., 2012).

2.3.3 Highway surface type and drainage mechanisms

The type of highway paving materials may also affect the amount of pollutants in highway runoff. However, Stotz (1987) investigated three highways with different paving materials and found that method of drainage influenced runoff quality more than pavement material. The impact of two types of pavement (conventional and porous) on the pollutant content in runoff was investigated by Pagotto et al. (2000). Results showed that porous pavement improved runoff water quality because reduced the amount of heavy metals and hydrocarbons mobilised by surface rainfall. The retention of particulate pollution by the porous pavement, which acts as a filter, was clearly demonstrated. Huber et al. (2016) reports that usage of porous asphalt for reconstruction of bridges causes significantly lower heavy metal concentration in runoff. One of the other important benefits of porous asphalt is its ability to reduce noise. The disadvantages are, for example, easy surface damage at high traffic volumes, mud and sand can fill the voids and reduce its drainage capacity. These surfaces require special winter road maintenance approaches and generally remain icy longer and require special patching and reconstruction techniques (Ahmad et al., 2017).

Concrete pavement compared to an asphalt surface contains alkaline substances to increase the pH of runoff and should have a stronger influence on the metal partitioning to the particulate phase than asphalt surfaces (Huber et al., 2016).

2.3.4 Traffic area categories

Huber et al. (2016) compiled a dataset of 294 monitored sites from six continents (Africa, Asia, Australia, Europe, North and South America) and based on evaluation and characterization of occurrence and fate of heavy metals, he divided traffic area in categories - parking lots, bridges, roads and highways.

Heavy metal runoff concentrations at parking lots differ widely according to their use (e.g., private companies, supermarket, rest areas for trucks). Although car parks have low traffic density, they show higher concentrations of pollution compared to non-congested roads due to drip losses, tire and brake wear and higher exhaust gas emissions. In particular, parking lots in urban and industrial areas are among the dominant contributors Cu, Zn and Cd in the runoff. Bridge deck runoff can contain high Zn concentrations from safety fences and galvanizing elements. Roads with more than 5000 vehicles per day are often more polluted than highways because of other site-specific factors such as traffic signals.

In another work by Huber and Helmreich (2016), there were summarized and discussed pollution levels at three types of traffic areas (highways, parking lots and roads) with selected seven metals (Sb, Cd, Cr, Co, Pb, Ni, Zn). Runoff from parking lots contained lower concentrations of these metals runoff from motorways and roads. Frequently used highway parking lots with a high percentage of trucks have increased runoff load and these values are comparable to the annual metal runoff load on roads. Knowledge of runoff in different traffic areas is important for rainwater management.

Rozkošný et al. (2014) presented results of water quality monitoring of road and parking lot run-off performed in the city of Brno in 2008-2009 and 2013-2014. The monitored parking areas were gradually loaded with traffic due to the completion of office buildings in their vicinity. Runoff concentrations of selected pollutants (chloride, PAHs, C10 – C40, Cd, Cr, Cu, Hg, Ni, Pb, Zn) from the two higher mentioned periods showed that the range of concentration varied in lower values than reported in the literature. The reason was the low occupancy of car parks by vehicles during their gradual commissioning.

2.3.5 Climatic factors

Various factors that relate to local conditions affect the build-up and removal processes of pollutants on and from the highway surface. These include both direct ambient influences, such as temperature and seasonal precipitation patterns, and indirect impacts caused by seasonal human actions such as the application of de-icing materials (Opher and Friedler, 2010). Hence seasonality may affect the quality of highway runoff. In regions where de-icing agents are used, higher concentrations of most components, especially metals, occur in the winter (Crabtree et al., 2006, Backström et al., 2003, Klimaszewska et al., 2007). The increased concentrations were due to more intense wearing of the pavement during the winter because of the use of studded tires in combination with the chemical effects caused by the use of de-icing salts (Backström et al., 2003). Helmreich et al. (2010) concluded that the de-icing salts had only a weak influence in terms of generating higher total pollutant load concentrations, and that the reported increase in metal concentrations were a function of increased wear and tear due to the application of gravel in cold weather conditions. A seasonal effect was also observed for PAH concentrations in runoff, which was manifested by lower concentrations of PAHs in summer (Crabtree et al., 2008). A considerable seasonal increase in pollutant concentrations was reported out for Cu, TOC, SS, pH value and especially for Zn during the cold season. The mean values during winter were several times higher than those measured during warmer seasons. In contrast, the fractionation of heavy metals was not affected by seasonal variations (Helmreich et al., 2010).

Climatic conditions in cold regions can affect runoff quality. Westerlund et al. (2003, 2006) compared the quality of road runoff during a melt period and a rain period. Runoff samples were analysed for suspended solids and heavy metals (Pb, Cu, Cd, Ni and Zn). The results showed that the concentrations of suspended solids, lead, copper and cadmium were higher for the melt period, compared to rain generated runoff on the catchment without snow, and the highest concentrations were found during the rain-on-snow events. It was also shown that more heavy metals were particulate-bound during the melt period as compared to the rain period (with a higher percentage of the dissolved fraction in this latter type of event).

3 Runoff effect on surface water

3.1 Introduction

The design of impermeable areas such as roads and airports in a catchment greatly alters the local hydrological cycle with larger volumes of water being discharged into nearby recipients in a shorter time frame (Ellis et al., 1997). The consequence is the increase in peak runoff during rainfall, which can cause basal erosion and increases the risk of flooding (Pontier et al., 2001). Runoff that enters - either directly or indirectly through a treatment system - a water recipient can alter their chemistry and quality (e.g. pH, dissolved oxygen, alkalinity, hardness, suspended solids, elevated chloride concentrations). The elevated temperature of the runoff can change dissolved oxygen levels and influence physiological processes. The first flush effect can generate comparatively high pollutant loads during the first phase of the storm event. These factors can affect the species composition of the aquatic ecosystem and may also have a toxic effect on aquatic organisms. In recent years this is particularly the problem of older and lower-class roads that do not have a drainage system. Newly constructed roads, especially motorways, are equipped with drainage system with retention of runoff in the retention tanks in accordance with new technical standards. These tanks serve as a pretreatment before draining the water into the recipient.

3.2 Case studies

Corsi et al. (2010) investigated the influence of road-salt runoff on surface water and aquatic organisms. Water-quality sampling was conducted on a local, regional and national scale. In the Milwaukee metropolitan area (USA, local scale), twelve streams were sampled in February and March 2007 which were located in an area of substantial urban land-use. A reference stream (Parnell Creek) with a catchment which was 80 % natural areas and no urban land use was also sampled. An additional stream, Wilson Park Creek, was monitored selectively from 1997 through 2007 during de-icing periods. Sample from these 14 streams were analysed for a range of parameters including chlorides, specific conductance, and were subjected to bioassays involving the use of *Pimephales promelas* and *Ceriodaphnia dubia*. In southern and eastern Wisconsin (regional scale), 11 streams were monitored continuously for specific conductance (as a surrogate for chloride), to assess potential impact of de-icing activities on aquatic organisms. These streams were selected to represent a gradient in land use typologies, with urban influence ranging from 6.0 to 100 %. On a national scale, analysis of historical data was conducted for 17 metropolitan areas. Data were retrieved from the U.S. Geological Survey (USGS) National Water Information System (NWIS) for chloride concentrations from streams sampled between 1969 and 2008. Data were compared to U.S. Environmental Protection Agency (USEPA) water-quality criteria and analysed for seasonal differences.

Dramatic impacts were observed on local, regional, and national scales. Locally, 7 samples of 13 from Milwaukee, Wisconsin area streams, exhibited toxicity towards *Ceriodaphnia dubia* and *Pimephales promelas* bioassays during road-salt runoff. Chloride concentrations above the EPA acute water-quality criteria concentration of 860 mg/L were reported for eight of these samples and concentrations above the EPA chronic water-quality criteria concentration of 230 mg/L in 11 of these samples, indicating potential for both acute and chronic aquatic toxicity effects. Another Milwaukee stream which was sampled from 1996 to 2008 exhibited toxicity towards *Ceriodaphnia dubia* in 72 % of 37 samples in chronic bioassays and in 43 % of samples subjected to acute bioassays. The maximum chloride concentration identified was 7730 mg/L in Milwaukee streams. Regionally, in southeast Wisconsin, continuous monitoring of specific conductance indicated increasing conductivity with increasing percentages of urban land use (Figure 1). Elevated specific conductance was observed between November and April at all sites, with continuing effects between May and October at sites with the highest specific conductance. Specific conductance was measured as high as 30 800 μS/cm (Cl^- = 11 200 mg/L). Chloride concentrations exceeded U.S. Environmental Protection Agency (USEPA) acute and chronic water-quality criteria at 55 and 100 % of monitored sites, respectively. Nationally, U.S. Geological Survey historical data were examined for 13 northern and 4 southern metropolitan areas. Chloride concentrations exceeded USEPA water-quality criteria at 55 % (chronic) and 25 % (acute) of the 168 monitoring locations in northern metropolitan areas from November to April. Only 16 % (chronic) and 1% (acute) of sites exceeded criteria from May to October. At the southern sites, very few samples exceeded chronic water quality criteria, and no samples exceeding the acute criterion. Bioassay results from runoff events confirmed that the observed high concentrations of road salt were associated with acute and chronic toxic in tested aquatic organisms (*Ceriodaphnia dubia* and *Pimephales promelas*). This review of road salt effects concluded that high concentrations may have immediate or long-term ecosystem population effects, and that lower levels of increased chloride concentrations may affect community structure, diversity, and productivity.

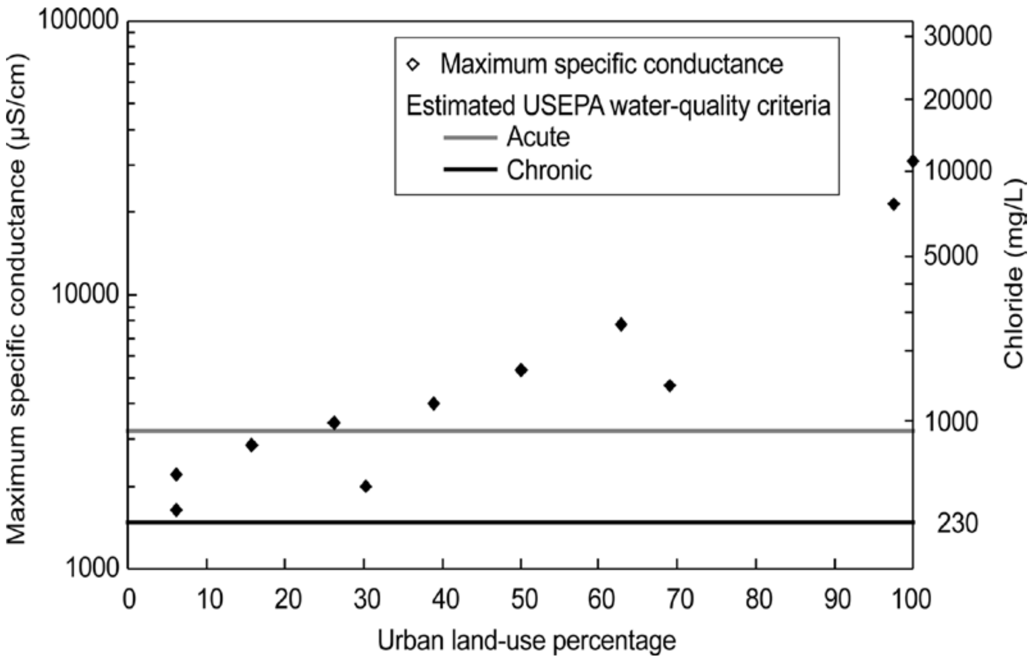

Figure 1 Maximum specific conductance compared to urban land-use percentage in 11 Wisconsin streams with reference to U.S. Environmental Protection Agency water quality criteria for chloride (after Corsi et al., 2010)

In Canada, Williams et al. (1999) examined 23 springs of water in the Greater Toronto Area (GTA) of southern Ontario to explore the relationship between spring macroinvertebrate communities and salt contamination, and to develop a biotic index. In order to assess the impact of road salting on surface and groundwater quality, surface water Cl levels in the study area (from the Ontario Ministry of Environment and Energy dataset) were compared with the spatial-temporal distribution of Cl levels recorded at the spring sites. Overall, as for surface waters, the Cl levels found in spring waters, were closely correlated with the degree of urbanisation. A major difference, however, was the far greater seasonal stability of the Cl level in the springs. The spatial pattern of Cl contamination indicated by spring water was more reliable and accurate than that measured in river water. It could be used for monitoring groundwater contamination, because it is much easier to sample springs than to collect groundwater directly (which typically involves drilling). The inter-year comparison of Cl levels at four of the most contaminated springs showed increases ranging from 21 to 34 % between 1996 and 1997. Canonical Correspondence Analysis (CCA) and subsequent cluster analysis divided spring-dwelling invertebrates into being either "tolerant" or "non-tolerant" of elevated Cl concentrations. A new biotic index of contamination - the Chloride Contamination Index (CCI), which summarised the Cl tolerance of a spring community - was significantly correlated with the Cl concentration measured at the springs. The amphipod *Gammarus pseudolimnaeus* was associated with source aquifers identified as only mildly contaminated with Cl. Absence of this species from a spring, particularly if nymphs of the stonefly *Nemoura trispinosa* were present indicated moderate to high levels of Cl contamination. Laboratory tests were conducted to determine the sensitivity of six species of spring macroinvertebrates to Cl contamination and for comparison with the field survey data. The laboratory assays identified the amphipod *Gammarus pseudolimnaeus* (and also, to a lesser extent, the amphipod *Crangonyx* sp.) as being intolerant of both acute and chronic exposure to elevated Cl concentrations, and in having its reproductive behaviour disrupted by the latter. For most of the spring invertebrate species tested Cl levels of 4500-6000 mg/L were lethal within 96 h. Springs were recommended as easy access points for study of both chemical and biological aspects of aquifer contamination.

Blasius and Merritt (2002) studied effects of road salt (NaCl) on stream macroinvertebrates by field and laboratory experiments. Field studies investigated possible differences in functional feeding group composition and litter processing rates between upstream (reference) and downstream (treatment) sites. The leaf pack studies were conducted from November 1995 through April 1996 in two northern lower Michigan streams (USA) which flow through culverts under major highways. Laboratory studies determined the effects of increasing NaCl concentrations on aquatic invertebrate drift, behaviour, and survival. Macroinvertebrates selected for drift and toxicity studies were chosen to represent a number of different trophic levels, habitat requirements, respiratory systems, and phenology. Field studies revealed that leaves were processed faster at upstream reference sites than at locations downstream from road salt point source inputs. However, it was sediment loading that resulted in partial or complete burial of leaf packs, that affected invertebrate activity and confounded normal leaf pack colonization. There were no significant differences that could be attributed to road salt between upstream and downstream locations in the diversity and composition of invertebrate functional feeding groups. Laboratory drift and acute exposure studies demonstrated that drift of *Gammarus* (Amphipoda) may be affected by NaCl at concentrations greater than 5000 mg/L for a 24-h period. This amphipod and two species of limnephilid caddis flies exhibited a dose response to salt treatments with 96-h LC50 values of 7700 and 3526 mg/L NaCl, respectively. Most other invertebrate species and individuals were unaffected by NaCl concentrations up to 10,000 mg/L for 24 and 96 h, respectively. The study considered short term/pulsed exposures of road salt with little or no residence time, but salt may accumulate in closed or restricted-flow systems, such as small urban ponds, storm water retention basins, wetlands, springs, and lakes. It can cause a chronic effect on the fauna.

Yousef et al. (1984) studied the influence of the bridge runoff on the quality of water in freshwater Lake Ivanhoe (Florida) and its effect on resident biota in this lake. A section of the central portion of the lake was filled in 1965 during the construction of Interstate 4 and the central island created was connected to northern and southern shores by means of two bridges. From the northern bridge, the water was drained to the adjacent land on either side, but from the south bridge drainage facilitated by scupper drains. Samples were taken from beneath the northern bridges, underneath two sets of scuppers on the southern bridges and in the main body of the western portion of the lake. Also, direct runoff water samples were collected directly from four different scupper drains during various storm events. The total concentrations of Zn, Pb, Ni and Fe in runoff water averaged 4.7, 20.8, 3.5 and 12.6 times higher than the average concentrations in Lake Ivanhoe (Table 2).

Table 2 Comparison of heavy metal concentrations in bridge runoff and Lake Ivanhoe water samples (after Yousef et al., 1984)

Metal	Form	Average concentration (µg/l)	
		Lake Ivanhoe	Bridge runoff
Zn	Total	103	498
	Dissolved	57	336
Pb	Total	76	1558
	Dissolved	53	187
Cr	Total	15	11
	Dissolved	5	2
Ni	Total	15	53
	Dissolved	9	49
Cu	Total	74	52
	Dissolved	24	27
Fe	Total	194	2429
	Dissolved	57	287
Cd	Total	4	5
	Dissolved	3	1

Heavy metals, particularly lead and iron in the scupper drain runoff, were mainly found in the particulate form, with an average of only 12% of the total concentration determined in the dissolved form. These particulate-associated fractions are most likely to settle out from the water column in the immediate vicinity of the point of release and become immobilized by the sediments. Sediment samples were collected from several locations in Lake Ivanhoe. The concentrations of Zn, Pb, Ni, and Fe were found to be significantly greater in the sediments underneath the south bridges with scuppers than in the sediments beneath the north bridge without scuppers. The plant samples collected from Lake Ivanhoe included submerged aquatic plants such as *Hydrilla*, the emergent species *Typha*, and the green algae *Spirogyra*. The benthic organisms included *Arthopodea* (*Crustacea*), *Molluska* (*Pelecypoda* and *Gastropoda*) and *Annelida* (*Tubifex*). Concentrations of Zn, Pb, Fe and Cd were reported to vary by species and location; for example, concentrations of all metals were significantly higher in *Spirogyra* collected from stations beneath bridges with two sets of scupper drains than those collected from stations beneath bridges without scuppers. Concentrations of most of the monitored heavy metals in *Spirogyra* were higher than in *Hydrilla.* This is probably due to the larger surface area to weight ratio for *Spirogyra* than other plants*.* The results support the use of *Spirogyra* as an indicator algae for contamination by highway runoff. In benthic organisms, it appeared that *Annelida* (*Tubifex*) concentrated more zinc, lead and chromium than other tested organisms indicating its potential role as an indicator benthic organism for highway runoff contaminants. Yousef et al. (1984) recommended that runoff from the bridge surface should be directed off the bridge surface towards either side to encourage percolation through the soil and removal of heavy metals by soil system.

Another study (Swadener et al., 2014) evaluated the quality of bridge deck runoff and determined the effects of bridge deck runoff on surface water bodies in Nebraska (USA) by evaluating water and sediment chemistry as well as their effects on aquatic life. Four sampling locations were selected to represent bridges with a range of AADT and receiving stream flow. Two sites were monitored for instream dry weather water quality at locations approximately 6 m upstream and downstream of the bridge. Runoff samples were collected from all four bridge locations by the gutter system. Instream water samples and bridge runoff samples were tested for a wide range of water quality parameters including TS, TSS, TDS, nitrate, nitrite, TKN, TP, organic compounds (n-hexane, MTBE, benzene, toluene, ethylbenzene, naphthalene, total xylenes, total purgeable Hydrocarbons), total metals (As, Cd, Cr, Cu, Fe, Ni, Zn), chloride, conductivity, *E. coli*, and Hexane Extractable Materials (HEM; Oil and Grease). Three surface sediment samples were collected upstream and downstream of one selected bridge with the lowest receiving stream flow at distances of zero, 3, and 6 meters from the edge of the bridge deck. All sediment samples were evaluated for metals. The upstream and downstream samples at 0 m upstream and downstream of the bridge deck were also analyzed for polyaromatic hydrocarbons (PAHs). Runoff toxicity was evaluated using a 48-acute exposure toxicity test with fathead minnow (*Pimephales promelas*). Metals, including Cr, Cu, Fe, and Ni were detected more routinely in runoff from all four bridge locations in comparison to organic compounds. The maximum observed concentrations of metals in the bridge deck runoff were 0.02 mg/L for total chromium, 0.05 mg/L for total copper, 14 mg/L for total iron, and 0.02 mg/L for total nickel. Metals that were detected the most frequently in bridge deck runoff were copper and iron. Copper was detected in 60 % of the runoff samples, while iron was detected in 100 % of runoff samples. Total arsenic and total cadmium were not detected in sediment samples above the detection limits of 10 mg/kg and 0.50 mg/kg respectively. Total iron was measured at the highest concentrations in sediment with a maximum observed concentration of 25 100 mg/kg (dry weight basis). Organic compounds, including benzene, total purgeable hydrocarbons, oil and grease, and MTBE were detected less frequently in this study compared to nutrients and metals. No clear relationship between the average concentration of contaminants in runoff at each site and AADT or ADP was observed. I The results of the toxicity testing showed that the bridge deck runoff was not toxic to the target organism. No statistically significant differences were observed in metal concentrations upstream and downstream of the bridges, providing additional evidence that bridges do not have long-term impact on receiving water quality. The sampling period in this study was limited to the summer months and does not include seasonal effects on the quality of bridge deck runoff.

The quality of tunnel wash water (the Nordby tunnel, Norway) and its influence on the small stream and impact on fish was studied by Meland et al. (2010a, 2010b). The first study (Meland et al., 2010a) was conducted in 2006 and the objective was to characterize and quantify *in situ* traffic related contaminants in runoff from a routine tunnel wash event which led to discharge from a sedimentation pond into the stream Årungselva and identify impact from these discharges for the downstream fish population, such as long-term changes in growth and density of juvenile sea trout (*Salmo trutta* L.). The tunnel cleaning intensity was 4 to 6 times per year. *In situ* size and charge fractionation techniques were applied to quantify traffic related metal species, while PAHs were quantified in total samples. Temperature, turbidity, conductivity, dissolved oxygen and redox potential increased rapidly in the pond outlet in response to the incoming tunnel wash water whereas pH remained circum-neutral. The concentrations of the various metals in the pond outlet were low prior to the tunnel wash and comparable with the concentrations measured upstream in the recipient stream, Årungselva. In addition, the concentrations of PAHs were below quantification levels both in the stream and in the pond outlet prior to the tunnel wash event. All metals and several PAHs appeared at elevated concentrations in the discharged wash water compared with concentrations measured in Årungselva upstream the pond outlet, and to concentrations measured in the pond outlet before the tunnel wash event. In addition, several contaminants (e.g. Cu, Pb, Zn, fluoranthene, pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(g,h,i) perylene and indeno(1,2,3-cd)pyrene) measured in the pond outlet, exceeded pertinent EQS for fresh waters obtained from EU (EC, 2006), USA (USEPA, 2009), Canada (CCME, 2007), Sweden (SWEPA, 2000) and Norway (Andersen et al., 1997; Lydersen et al., 2002). PAH and metals (including Al, Cd, Cr, Cu, Fe and Pb) were reported as primarily associated with particles and colloids, while As, Ca, K, Mg, Mo, Ni, Sb and Zn were more associated with low molecular mass species (LMM, <10 kDa). LMM species are believed to be more mobile and bioavailable than metals associated with particles. Calculated enrichment factors (based on total concentrations normalized by Si concentrations) in the tunnel discharged runoff revealed that many of the metals were derived from anthropogenic sources, such as wear of tires (Zn), brakes (Cu and Sb), combustion (Co) and from road salt (Na and Cl). The enrichment factors for Al, Ba, Ca, Cr, Fe, K, Mg and Ni were low, indicating that wear from the tunnel surfaces was their dominant source. Based on calculated PAH ratios, PAH are understood to originate from a mixture of sources such as wear from tires, asphalt and combustion. An evident growth reduction in sea trout (age 0+, fish less than one year old) in the lower parts of the stream Årungselva compared with sea trout in the upper parts may be attributed to contaminants discharged into the stream from the pond during the last 10 years. The authors recommended an additional treatment step after the pond outlet to effectively reduce the concentrations of contaminants entering the stream.

The second study (Meland et al., 2010b) in the Nordby tunnel was conducted in early December 2008 during washing of the tunnel. This study was designed to quantify chemical contaminants (trace metals, hydrocarbons, PAH and detergents) in wash water and assess the short-term impact on brown trout (*Salmo trutta* L.) based on *in situ* experiments. A chemical analysis of water samples, collected before and during the tunnel wash was carried out. The tunnel wash water contained elevated concentrations of trace metals. The highest concentrations were measured in the last sampling round in the end of the tunnel wash period. The greatest concentrations were observed for Na and Cl, ranged from 227-926 mg/L for Na and 514-1280 mg/L for Cl. The metals were strongly associated with particles and are hence normally considered not to bioavailable. In addition, wash waters showed high concentrations of Ca and DOC which most likely minimized the toxicity. PAH and hydrocarbons were below the detection limit in the tap water but showed elevated concentrations during the tunnel wash. Pyrene, chrysene and fluoranthene were the single PAH components that appeared with the highest concentrations (52 % of the total 16 PAHs concentration) and all are classified as being high molecular mass PAHs (HMM >4 rings). The experiment with fish was carried out in such a way that Brown trout (0+) were transported from the local hatchery and transferred into 2 black circular exposure tanks receiving municipal tap water. The fish were exposed to polluted washing water, which was pumped into exposure tanks during cleaning. Selected endpoints were accumulation of trace metals in gills, haematological variables and hepatic mRNA transcription of five biomarkers reflecting defence against free radicals, trace metals, planar aromatic hydrocarbons and endocrine disruptions which were measured prior (−3 h), during (1 and 3 h) and after the tunnel wash (14, 38 and 86 h). No mortality occurred during the experiment, but the fish exposed to tunnel wash water clearly changed their behaviour compared to the control fish and the rapid accumulation of trace metals in gills was observed immediately after the wash water entered the exposure tank. This was followed by a modest, but rapid, change in blood plasma ions and glucose. Both the accumulation of trace metals and the blood parameters recovered to control levels within 38–86 h. In contrast, the mRNA transcription of the CYP1A and the oxidative stress related biomarkers TRX and GCS did not increase until 14 h after the exposure start and did not return to background levels when the experiment was terminated 86 h after exposure start (Figure 2). This study has shown that discharging polluted water from the tunnel could be a potential threat to the living organisms in the receiving waters.

The potential pollution of rivers due to surface-water discharges from newly-built sections of the A74(M) motorway in south-west Scotland was examined by McNeill and Olley (1998). A method for assessing the theoretical risks of pollution of road drainage was compared with the results of discharge monitoring. This method is based upon the assessment of dissolved copper and total zinc concentrations because EQSs are established for these two metals and they are omnipresent in highway runoff. The assessment assumes that if the runoff does not cause the EQS of copper and zinc to be exceeded in the receiving waters, no other dissolved contaminant will cause pollution. The assessment is carried out in two stages. Firstly, a simple procedure is used to identify outfalls which will need a more detailed evaluation to establish if mitigation or treatment of the runoff is required. The simple assessment is essentially based upon three criteria: (a) the water-quality classification of the receiving watercourse, (b) the dilution afforded by the receiving waters at the 95%ile level (minimum flow), and (c) the number of vehicles which will use the road each day. The hardness of the receiving waters (as determined by the calcium carbonate concentration) also needs to be considered at this stage, because the EQS for copper and zinc varies with hardness. Any discharges to receiving waters with $CaC0_3$ concentrations below 50 mg/L require a more detailed assessment because the toxicity of the two metals increases considerably in soft water. The detailed assessment for dissolved copper and total zinc uses the same criteria as described for the simple test but, in addition, the concentration build-up of these metals over a 5-day period is required. Also, background concentrations of the metals in the receiving waters above the points of discharge are needed. If it is accepted that filter drains remove up to 80 % of pollutants, the detailed assessment is re-calculated on this basis. The impact of motorway runoff on the invertebrate fauna of watercourses was also investigated. 63 discharge samples were collected over a twelve-month period, from eleven of the twenty-two outfalls within the study area. Samples were analysed for dissolved copper, total zinc and suspended solids. Dissolved copper and total zinc concentrations in discharges were generally very low and well below relevant EQS. The average results for copper and zinc of all of the samples collected correlated fairly closely with the detailed assessment for these metals (which include the 80 % purification factor). Therefore the assessment method appeared to be fairly accurate, although it errs towards the upper end of the scale in its predictions. Suspended-solids concentrations were generally satisfactory, although there were occasions when relatively high concentrations were recorded. The concentrations of copper closely mirrored those of suspended solids because this metal is associated with suspended solids. Total zinc concentrations showed similar trends to copper and suspended solids during the summer and autumn, but were at a variance with these parameters in the winter and early spring. The highest results for zinc were recorded during the winter and spring whilst, in contrast, the lowest average concentrations for copper and suspended solids were obtained at this time. This probably reflects the application of road salts for de-icing, because zinc is known to be present in many types of salts which are used for this purpose. Samples of invertebrates were collected from a selection of watercourses upstream and downstream from the outfalls. The results of biological monitoring are presented as biological monitoring working party (BMWP) scores and average scores per taxon (ASPT). Invertebrate fauna in streams below the A74(M) outfalls, where the highest levels of solids were recorded in discharges, were unaffected with no obvious drop in diversity taking place. *Gammarus* were found at every monitored site, and there was no evidence that this shrimp had been affected by solids or any associated toxins.

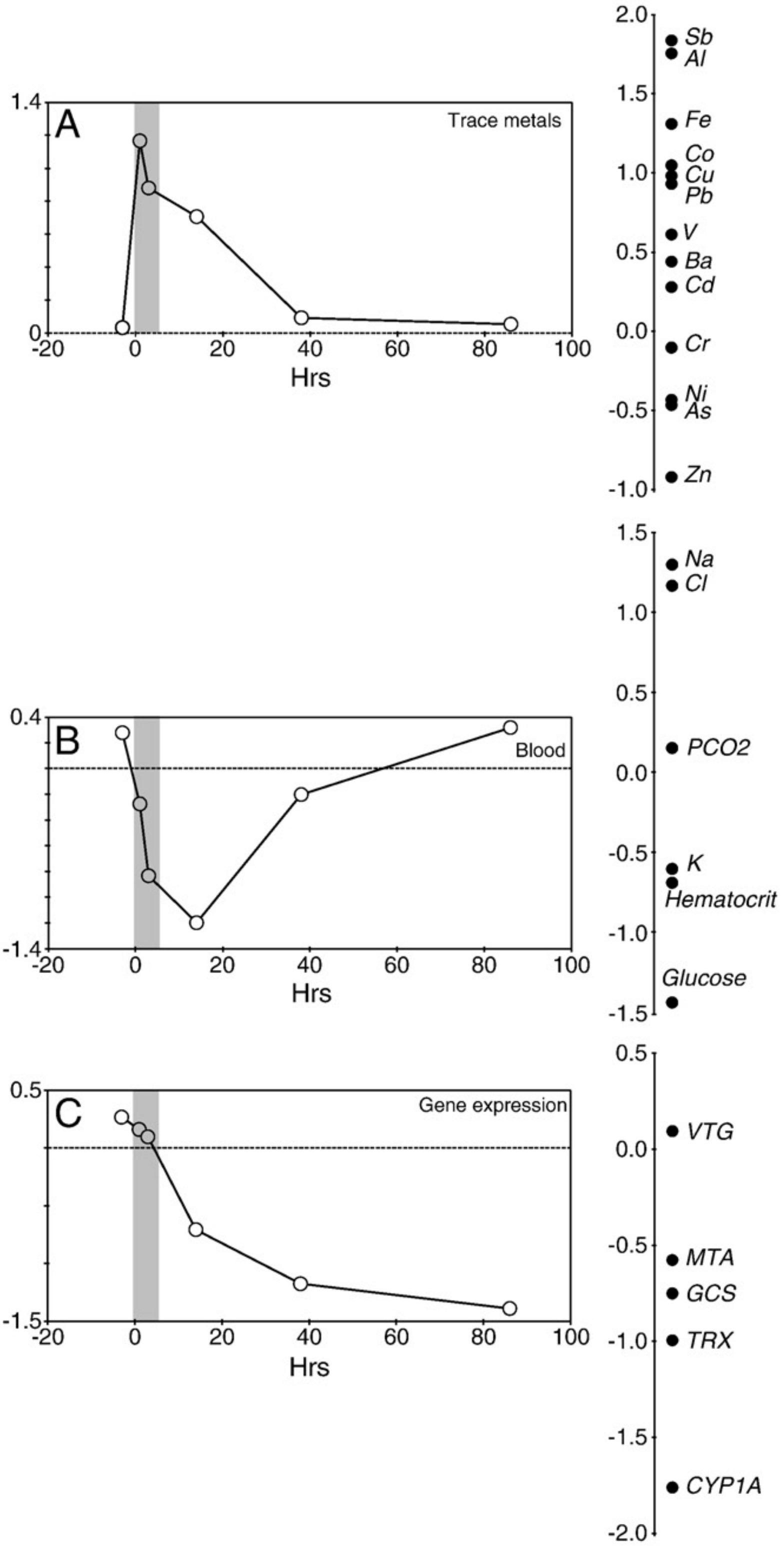

Figure 2 Diagrams showing the first component of the principal response curves (PRC) of trace metals in gills (A), blood physiology (B) and gene expression over selected biomarkers (C). Each diagram has its corresponding set of endpoint weights displayed on the right-hand side. The control group in each diagram is displayed with a dotted line through zero. The tunnel wash is indicated by a grey vertical line (after Meland et al., 2010b)

A battery of bioassays was used to assess the degree of runoff toxicity in a study carried out in Southern Ontario, Canada (Mayer et al., 2011). Runoff samples were collected from three sites, representing different classes of highways high (92,000 vehicles/day), intermediate (31,100 vehicles/day) and low (15,460 vehicles/day) traffic intensity). The acute toxicity tests included *Daphnia magna* (water flea) static non-renewal toxicity tests, the commercially available standard toxicity bioassay - Microtox™ test and Rainbow trout (*Oncorhynchus mykiss*) acute toxicity test. Sub-mitochondrial particle (SMP) assays were also used to detect bioavailable toxicants in some samples. Estimates of sub-lethal toxicity were performed using *Ceriodaphnia dubia* (water flea) chronic tests and rainbow trout mixed function oxidase (MFO) tests. Toxicity of the runoff solids was determined using the nematode (*Panagrellus redivivus*) test and Microtox™ solid phase test. *Daphnia magna* and *C. dubia* whole organism tests were shown to be sensitive to elevated concentrations of salts and metals in runoff. Generally, higher toxicities were observed with increasing levels of conductivity, which likeiwse corresponded to increasing concentrations of chloride. Moderate to strong acute and chronic toxicity responses were also generated by runoff samples identified as containing elevated levels of Zn. The changes in acute and chronic toxicity over the course of the runoff event were shown in *C. dubia* survival and reproduction tests (Figure 3). Acute mortality in *C. dubia* was reduced and the number of neonates produced increased in response to samples collected over the duration of the storm, indicating a decline in toxicity of runoff during a storm event. The 'first flush' effect was confirmed, with the decline in runoff toxicity attributed to corresponding changes in Zn concentrations, as the majority of toxic metals, except for Zn, did not decrease appreciably over the profile of the monitored event. A similar toxic response was also observed on the exposures of rainbow trout (*O. mykiss*) to highway runoff.

Many contaminants (e.g. metals, PAHs) commonly present in highway runoff are known to have a strong affinity for particulates. Therefore, solid phase toxicity testing should be an important component of highway runoff toxicity assessment. Benthic invertebrates from receiving water bodies can take up bioavailable toxicants from sediments by ingestion of contaminated particles and concentrate them in their tissues. The results of the Microtox™ sediment tests performed on samples collected in the study by Mayer et al. (2011) were indicative of increasing toxicity during the course of the runoff event, a trend that was opposite to that displayed by the toxicity of the aqueous portion of runoff. The results showed that surface runoff from the major multilane divided highway, with the highest traffic intensity, had the highest levels of contaminants and that this runoff exerted the highest acute and chronic toxicity on aquatic organisms in laboratory bioassays. Co-analysis of toxicity tests with analytical data identified the role of de-icing salts in contributing to toxic responses. For example, the runoff samples with the highest Cl concentrations were also the most toxic to test organisms. The runoff with the highest MFO induction was collected from the precipitation event with the highest rainfall. As shown by the chemical analysis of runoff samples, this runoff sample also had the highest concentration of 16 priority PAHs associated with runoff solids. The toxicity of runoff increased with the increasing concentrations of high-molecular-weight PAHs in runoff solids, particularly with increasing concentrations of benzo[a]pyrene and indeno[1,2,3-cd]pyrene (both identified as mutagens and carcinogens). The information presented in this study represents the 'worst case scenario', in which untreated highway runoff would directly enter a small receiving water body, with little dilution. This investigation contributed useful information necessary for planning and implementation of remediation strategies for the mitigation of impacts of highway runoff pollution.

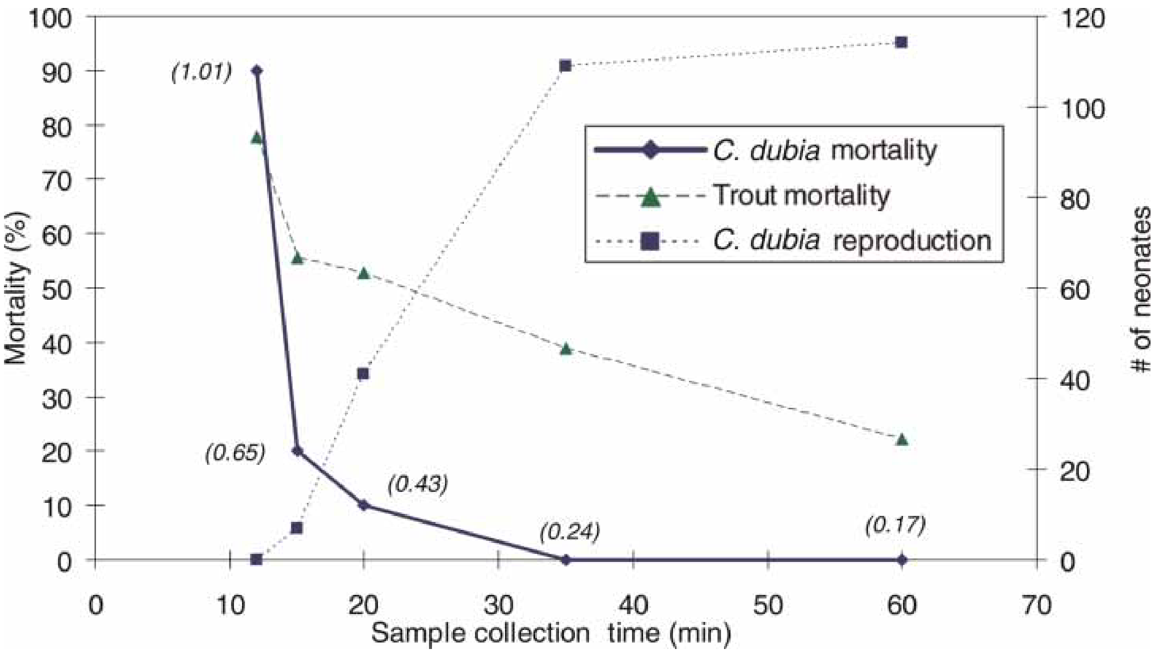

Figure 3 Changes in runoff toxicity during the course of the runoff event as indicated by time series data, from Skyway Bridge (SW), of *Ceriodaphnia dubia* survival and reproduction and rainbow trout acute toxicity tests (17 September), (after Mayer et al., 2011)

The toxicity of stormwater runoff during various time-based stages was measured in both grab and composite samples collected from three highly urbanized highway sites in Los Angeles, California between 2002 and 2005 (Kayhanian et al., 2008). Stormwater runoff samples were tested for toxicity using three freshwater species (the water flea *Ceriodaphnia dubia*, the fathead minnow *Pimephales promelas*, and the green algae *Pseudokirchneriella subcapitatum*) and two marine species (the purple sea urchin *Strongylocentrotus purpuratus*, and the luminescent bacteria *Photobacterium phosphoreum* using Microtox™). Samples were also analyzed for major trace metals (Cu, Pb, Ni, and Zn). Toxicity to both freshwater and marine species was frequently observed, but varied between storm events, test species, and monitoring locations. The sea urchin fertilization test was the most sensitive of the five methods evaluated. Samples from all storms exhibited toxicity to all test species, but there was little correlation between the marine and freshwater tests. Fathead minnows were more sensitive than *Ceriodaphnia* for most of the stormwater samples tested. The toxicity responses for green algae and Microtox™ were less consistent, and occasionally showed stimulation or atypical dose response relationships due to the presence of nutrients in stormwater. A first flush effect was almost always observed with both freshwater and marine species. In most cases, more than 40 % of the toxicity was associated with the first 20 % of discharged runoff volume. Furthermore, on average, 90 % of the toxicity was observed during the first 30 % of storm duration. However, toxicity was occasionally not related to the first flush samples and occurred in grab samples collected later during a storm. While all species responded to the first flush, toxicity in samples collected after the first hour of each storm often differed between species.

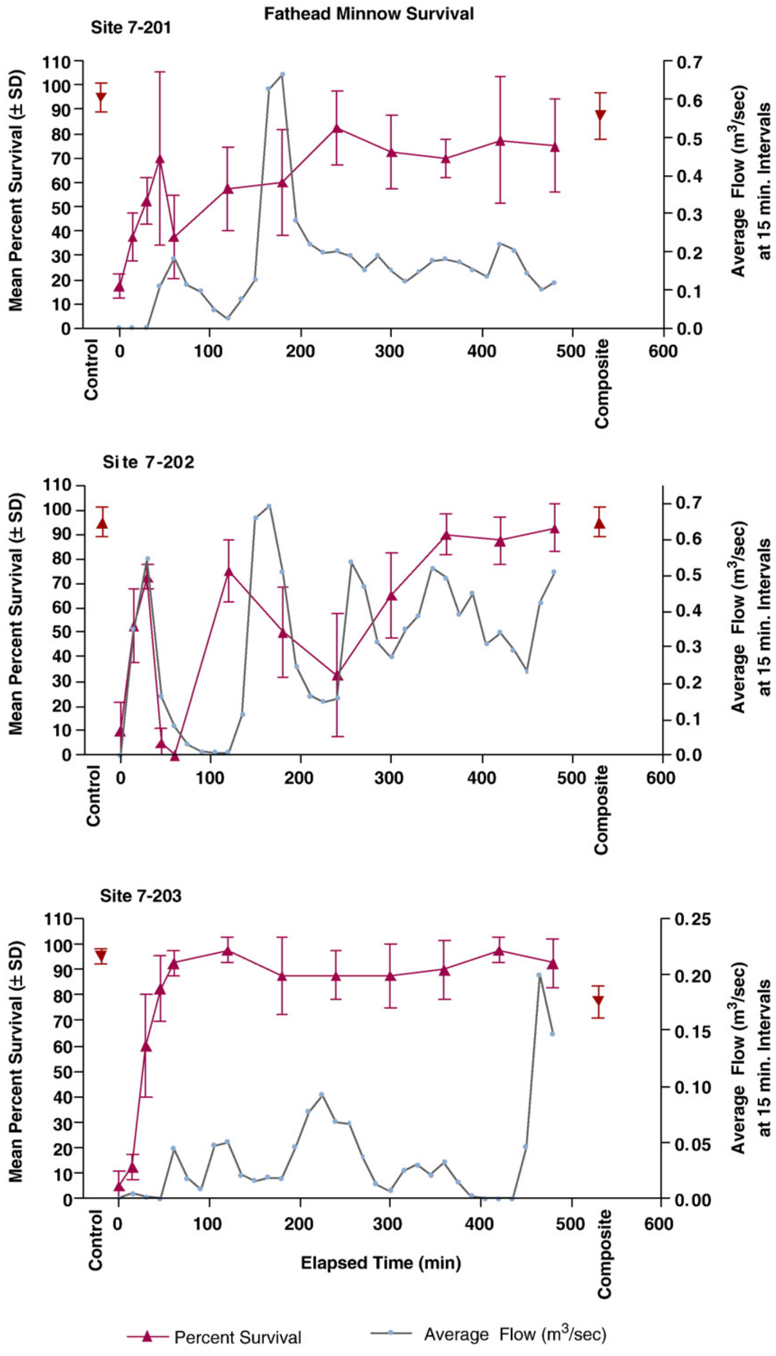

Figure 4 Fathead minnow survival test toxicity response to runoff samples (October 26–27 2004 storm event) (after Kayhanian et al., 2008)

The proportion of toxicity attributable to the first flush suggested that efforts focused on treating the first portion of stormwater runoff volume may be more beneficial in protecting the environment while also being more cost effective. A majority of the composite samples were non-toxic to freshwater test species, even when a strong first flush effect was observed in individual grab samples (Figure 4). A toxic response in composite samples was more prevalent with the marine species tested. Toxicity identification evaluation results identified copper and zinc to be the primary cause of toxicity to *Ceriodaphnia* and fathead minnows in about 90 % of the evaluated. Surfactants were also found to be the cause of toxicity in less than 10 % of the samples.

Invertebrate samples from nine East Anglian (UK) rivers were collected above and below crossings of the A12 and A14 trunk roads in spring and summer 1996 to assess any impact due to road run-off discharges (Perdikaki and Mason, 1999). The BMWP score, ASPT and Shannon diversity index values were compared between upstream and downstream sites. Only the BMWP score was significantly lower in summer at downstream sites in those rivers crossed by the A12. It suggested that road run-off might have an impact during low flow conditions on these rivers. Zinc, lead and cadmium concentrations in sediments and invertebrates (*Asellus aquaticus*, *Gammarus pulex*, *Sialis lutaria*) did not differ significantly between upstream and downstream sites. This implies that either the metals discharged from road run-off are not in sufficiently high concentrations to cause discernible build-up of metals at downstream sites or that discharges of metals upstream (from agricultural and urban sources, including treated sewage effluents) are sufficiently elevated to mask the contribution from roads. There was a significant relationship between metal concentrations in sediments and invertebrates only for lead. The significant correlations between the three metals implied that the metals were released to the streams simultaneously. Road run-off from these road trunks appeared to have no major impact on the receiving streams.

Moy and Crabtree (2002a, b, c, d, e, f) monitored the quantity and quality of highway surface water drainage and the receiving waters at five sites incorporating eight different drainage treatment facilities (UK). Monitoring was focused on collecting water samples and *in situ* measurements at upstream and downstream watercourse monitoring points, taken at monthly intervals, under dry weather conditions. Sediment samples were collected at the beginning and conclusion of the monitoring period from the highway runoff ditch and the watercourse at the upstream and downstream monitoring points. Biological surveys of the receiving watercourse were undertaken on three occasions upstream and downstream of the point of discharge and Biological Monitoring Working Party (BMWP) scores and ASPT (Average Score Per Taxon) scores were calculated. Hydrologic data (rainfall, river flows) were collected and water samples analysed for BOD, COD, NH_3, TSS, hardness, temperature, pH and dissolved oxygen on a basis. Storm event sampling involved the analysis of samples for BOD, COD, NH_3, TSS, hardness, temperature, pH, DO, turbidity, conductivity, metals, PAHs, herbicides and de-icing salts. Concentrations of metals, polyaromatic hydrocarbons and weathered hydrocarbons, were identified in the sediments samples together with the analysis of particle size distribution and organic matter content. Additional data, such as traffic density, herbicide application and road salt application, were also reported.

The first monitoring site selected was on the M4, where surface drainage for a section of motorway to the west of junction 16, discharges to the Brinkworth Brook, a tributary of the River Avon (Moy and Crabtree, 2002a). This site was selected as a control site, where surface water drainage received no treatment prior to discharge to the receiving water. Traffic density was in the range of 62 230 to 79 433 vehicles per day (two ways). Untrapped gullies discharge directly into open, unlined ditches on either side of the motorway embankment which in turn discharge directly to Brinkworth Brook. There was a significant difference ($P < 0.02$) in BMWP scores between the upstream site and the three sites downstream, equating to the absence of 2 or 3 sensitive taxa downstream compared to upstream. This suggested that the untreated runoff had a relatively small – but statistically significant - adverse effect on the macroinvertebrate community downstream. However, the results should be interpreted with caution, since the gravels of the upstream reach of the brook were of higher quality for macroinvertebrates than the downstream reaches, providing a greater proportion of coarse sediments on a firmer underlying substrate. This would tend to give the upstream site higher BMWP scores than downstream sites even in the absence of water quality impacts. Given these habitats and the relatively small differences observed in BMWP score, the researchers concluded that it was not possible to say with any certainty that run-off from the motorway had an impact on the macroinvertebrate community. A clear tendency for depressed BMWP scores and ASPT values at all three sampling sites were noted during the "late Summer" sampling round. Overall the study concluded that whilst there were statistically significant differences in biological quality between sites upstream and downstream of the point of run-off entry, it was not possible to discount contributing physical habitat and life cycle influences as the cause.

The second monitoring site was on the A417, discharging to the River Frome (Moy and Crabtree, 2002b). Traffic density was in the range of 20 890 to 26 323 vehicles per day (two ways). Surface runoff from the carriageway passes through a bypass oil separator to a dry balancing pond. The throttled pond discharge is piped for a distance of 600m and outfalls to a small spring fed rill, which in turn discharges to the River Frome at Brimpsfield Park. The results suggested that the treated runoff had no deleterious effect upon the River Frome. The ditch containing the runoff had the highest BMWP and ASPT in the late autumn sampling. Even in late spring and late summer, where the ditch sample was of comparatively poorer quality, the downstream samples scored more highly than upstream. Assuming the runoff was toxic, it appeared that the dry balancing pond and bypass oil separator used in this system provided sufficient treatment to the runoff, such that it caused no detectable impact on the receiving water at these dilutions.

The third monitoring site was also located on the M4 (Moy and Crabtree, 2002c). Traffic density was in the range of 29 872 to 41 201 vehicles per day (one way). Surface runoff from the carriageway between junctions 15 and 16 passes through an oil trap to a sedimentation tank. The tank discharges over a weir to the River Ray, a tributary of the River Thames. There were some marked differences in diversity throughout the sampling period. The first two sets of samples were taken within five months of each other and yielded similar biotic scores. The third set of samples were taken a year later and, while the upstream sample scored almost identically to previously, the downstream sample scored higher for taxa number and BMWP scores. This may indicate an improvement in water and or habitat quality at this site. The aim of the sampling regime was to monitor the two sites in late spring, late summer and late autumn. The most contaminated runoff might be expected to occur in late summer following a long dry period during which contaminants are deposited on the road surface and not washed off until the first heavy rainfall of autumn. The improvement in BMWP that was observed during this sampling period may be due simply to each successive sample being taken during a relatively less contaminated runoff period. Were a further sample to be taken in late summer 2000, the biotic scores could well resemble those of late summer 1998, i.e. the changes identified may be due to seasonal variations.

The fourth monitoring site was on the M40, between junctions 10 and 11, where surface runoff discharges to the Souldern Brook in the Cherwell valley (Moy and Crabtree, 2002d). Traffic density is in the range of 71 870 to 87 348 vehicles per day (two ways). Surface runoff from the carriageway passes through an oil trap to a large balancing pond. The pond discharges via a throttled outlet to the Souldern Brook. The discharge from the balancing pond appeared to have little effect on the chemical composition of the brook. However, some changes were found in several sensitive species being recorded both up- and down-stream of the outfall. It is possible that these observed changes in habitat in biodiversity may be due, at least in part, to the flow of sediment from the pond into the brook in times of flood. Such potentially contaminated sediments may be carried some distance downstream before being deposited and creating the deeper substrate at the sites where the invertebrate community was found to be less diverse (downstream 2 and 3) during summer sampling.

The fifth monitoring site was on the A34 approximately 1 mile south of its junction with the M40 at junction 9 at the Family Farm Services area near Weston on the Green (Moy and Crabtree, 2002e). Traffic density is in the range of 58 460 – 69 461vehicles per day (two ways). This section of highway is largely drained by filter drainage on either side of the carriageway with some sections, notably adjacent to service areas, junctions etc., drained via gully pots and piped carrier drains. There was no consistent pattern between upstreams and downstreams sites that might suggest an impact from the highway drainage. The factors likely to contribute to spatial differences in macro-invertebrate communities included the effects of highway drainage and differences in habitat (substrate type and degree and type of vegetation cover). The upstream and tributary sites may differ as a result of the flow regimes of Gallos Brook and the nature of the catchment above these sites. The tributary site was noted to receive sediment wash-off from agricultural fields. There was a possibility that highway drainage was modifying the composition of the macro-invertebrate fauna of the downstream sites in Gallos Brook. However, it was not possible to draw firm conclusions because the effect was not consistently noted throughout the sampling period.

In a study undertaken in Norway, Grung et al. (2016) investigated if fish from sedimentation ponds were affected by road pollution and sought to identify the sources of PAHs in associated sediment, plants and biota. The study involved the collection of common minnow (*Phoxinus phoxinus*) from both the pond and from the receiving river nearby. Minnow collected from the sedimentation pond had higher levels of CYP1A enzyme and DNA damage than minnow from the nearby river, but high concentrations of PAH-metabolites in bile of all fish revealed that both populations were highly exposed. Minnow from a lake un-affected by traffic reported much lower levels of PAH-metabolites (60-80 ng/g) than the exposed fish (2000-3000 ng/g), and also an improved condition. The latter results indicate that fish health was affected by road runoff. A further investigation of PAH levels within the two sedimentation ponds and nearby environments were conducted. The concentration of the 16 EPA PAHs in sediments of the sedimentation ponds ranged from 1900–4200 ng/g, and even higher levels were observed in plants. The PAH concentrations in the different plants varied considerably between sites and species. The levels in animals were lower than those observed in plants, indicating either lower bioaccumulation in these species, or possibly biotransformation of PAHs. The plants preferentially accumulated the high molecular PAHs, both from sedimentation ponds (where a petrogenic PAH isomer ratio was identified in sediments); and from a lake (where the PAH isomer ratio in sediments indicated a pyrogenic origin).

A small lake ecosystem (Lake Padderudvann) in Norway close to a highway was investigated to reveal the possible effects of highway pollutants (Baekken, 1994). Comparing data on dissolved oxygen and specific conductivity levels in this lake before construction of the highway suggested no effects on the oxygen condition but considerable effects on the conductivity after its construction. Lake Semsvann was selected as a reference lake for accumulation studies. This lake is 4 km north-east of Lake Padderudvann, and there is very low traffic density in its vicinity. The concentrations of cadmium and zinc in bivalves were about 2 to 3 times higher in Lake Padderudvann than in the reference Lake Semsvann, suggesting an impact from the highway. The values were 0.76 µg/g w.w. and 0.38 µg/g w.w. for cadmium and 46.2 µg/g w.w. and 16.0 µg/g w.w. for zinc. The concentrations of copper, mercury, nickel, lead and polycyclic aromatic hydrocarbons (PAH) in bivalves were low, presumably close to the background levels and there were no differences in the accumulation in bivalves from Lake Padderudvann and Lake Semsvann. The concentrations of lead in liver tissue from fish in Lake Padderudvann were above background level presumably as a result of the local input of lead from the highway pollutants. The concentration of PAH in the flesh of perch specimens from Lake Padderudvann was more than five times higher than that of Lake Semsvann: about 112 µg/g, compared with 23 µg/g. Most of the difference was due to naphthalene in the PAH complex, presumably an effect of the local highway pollutants. The diversity and abundance of the benthic communities were reduced on the highway side of the lake, suggesting negative effects of the highway pollutants. This lake is relatively uncommon with its high calcium content. The effects of highway pollutants may be very different on small acid lakes that are low in calcium.

In two retention ponds on the highway D5 (Rozvadov and Heřmanova Huť, Czech Republic), water quality was monitored between the year 2002 – 2004 (Adamec et al., 2004). Water sampling was undertaken in two ways: spot sampling and passive sampling using semipermeable membrane devices (SPMD). Concentrations of mineral oils and 16 PAHs according to US EPA were monitored. The results of PAHs indicated slightly polluted waters. Concentration values of 16 PAHs in spot samples were in the range of 12 - 190 ng/L and were higher than the concentration in SPMDs samples (1 – 25 ng/L). Concentrations of PAHs in the spot samples showed a fluctuating trend, while PAHs concentrations in SPMDs showed clearly increasing trend. Due to the type of sampling, the results of SPMDs can be considered more accurate. SPMD simulates a living organism. In the case of SPMD, only PAHs contained in the water are captured in the triolein within the membrane, while in the spot sampling the analyses determine not only the PAHs contained in the water but also the PAHs adherents on the solid particles suspended in the water samples. Mineral oils concentrations were in the range of 0.03 – 1.5

mg/L for spot samples. Spot water samples taken in 2004 were also subjected to ecotoxicology testing at three trophic levels (algae *Scenedesmus quadricauda*, crustaceans *Thamnocephalus platyurus*, and bacteria *Vibrio fisheri*). Water samples were non-toxic, except for two cases sampled on the locality Heřmanova Huť on July (*Scenedesmus quadricauda*) and on December (*Thamnocephalus platyurus)*.

Five settlement tanks near the motorway D1 (Prague-Brno-Vyskov, Czech Republic) were chosen for the sampling in a study by Marešová et al., 2002. These settlement tanks collected surface water from the motorway. Water washed away from the road surface enter the settlement tank in which suspended particles settle and runoff waters which exceed system capacity are freely discharged from the settlers to a water stream or to soil. Water and sediment samples from these tanks were analysed for polycyclic aromatic hydrocarbons (PAHs), nitro-polycyclic aromatic hydrocarbons (nitro-PAHs) and mineral oils. Ecotoxicological tests on bacteria (Microtox), crustaceans (Thamnotox) and algae (*Scenedesmus quadricauda*) were performed. Toxicity test showed that the most sensitive test for these types of waters was the test on crustacean *Thamnocephalus platyurus*. Porous water of sediment samples was generally more toxic than water samples. In several water samples algal test showed slight toxicity and the Microtox test showed toxicity only in porous water from one locality. Concentrations of mineral oils and PAHs were higher in sediments then in water samples. Both the non-toxic samples and the samples which showed toxicity in some of the tests were used for selected heavy metals concentrations determination. Most heavy metals entering the aquatic system are associated with particles and accumulate in sediments. Metals Cr, Cu, As, Cd, Pb, Ni and Zn showed higher concentrations which we can consider to slightly pollution or pollution water in comparison to Government Regulation defining indicators and values of permissible degree of water pollution No. 82/1999 Coll. Identification of the relation between chemical pollutants and ecotoxicological biotests is generally difficult. Possible connection could be suggested between the crustacean test and the amount of arsenic in the one case of water samples from one locality. Concerning PAHs and mineral oils, the results did not suggest any connection with the toxicity. Concerning the other samples, analogical relations were not suggested.

Beránková et al. (2008) reported the results of runoff monitoring that was performed in the period of 2005 – 2007 on the highways D1 Praha-Brno (Czech Republic) between 61.5 and 81.5 km. The intensity of transport on this stretch was approximately 40 thousand cars/24 hours. Water quality was monitored at the inflow to a series of runoff settlement tanks and in the adjacent recipient (streams into which the water is drained from settlement tanks). Also, samples of snow from the shoulder and sediment from the bottom of these settlement tanks were analysed. The second monitored site (with the same density of traffic was monitored with an automatic sampling device) was situated on the highway bridge on the 149.5 kilometre of D1. The third monitored area was a new stretch of road located at 233.0 kilometre of highway D1 with a very low traffic intensity and which has been operating only for a short period. Water and sediment leachate samples were analysed for a series of basic chemical parameters and priority substances as well as being subjected to ecotoxicity testing. Table 3 presents the results of the monitoring campaign together with the limit values defined in Regulation of. Gov. 229/2007 and Working qualitative goals for 2005 (Rosendorf et al., 2004). These limit values were used in the Czech Republic for period of the characterization of water bodies. The measurement of precipitation and outflow has also brought findings about the variability of runoff coefficient on these localities. Almost high-risk pollution levels (those exceeding defined standards) were found in the samples of settled sludge as a consequence of accumulation and fixation of organic compounds and metals to particulates. Results of ecotoxicity testing in run-off water indicated higher acute toxicity towards the algae *Scenedesmus quadricauda*, in comparison with the testing on invertebrate *Daphnia magna*, and are connected also with higher concentration of chloride in water during winter.

Table 3 Parameters of water quality of highways runoff (after Beránková et al., 2008)

Parameter	Unit	Average	Median	Q90	Reg. 229/ 2007	Qualitative limits (2005)
Pb	$\mu g.l^{-1}$	**3,82**	2,40	**6,10**	14,4	5
Cd	$\mu g.l^{-1}$	**0,406**	0,190	**0,770**	0,7	0,2
Ni	$\mu g.l^{-1}$	**45,3**	21,8	**132**	40	5
Hg	$\mu g.l^{-1}$	**0,199**	0,140	**0,270**	0,1	0,1
Cr	$\mu g.l^{-1}$	**4,83**	4,50	**6,80**	35	2
Cu	$\mu g.l^{-1}$	**19,0**	13,7	**52,8**	25	2
Zn	$\mu g.l^{-1}$	**142**	69,0	**400**	160	10
Cl	$mg.l^{-1}$	**1095**	726	**1 510**	250	-
Hydrocarbons C10-C40	$mg.l^{-1}$	**0,145**	0,145	**0,88**	0,1	-
Benzo(b)fluoranthene	$ng.l^{-1}$	**7,66**	3,75	**20,4**	60	30
Benzo(k)fluoranthene	$ng.l^{-1}$	**5,87**	3,65	**15,7**	60	30
Benzo(a)pyrene	$ng.l^{-1}$	**5,63**	2,10	**11,8**	100	50
Benzo(g,h,i)perylene	$ng.l^{-1}$	**6,29**	3,33	**13,1**	30	16
Indeno(1,2,3-cd)pyrene	$ng.l^{-1}$	**5,69**	3,25	**15,5**	30	16
Fluoranthene	$ng.l^{-1}$	**21,2**	9,80	**63,0**	200	90
Σ 6 PAH	$ng.l^{-1}$	**7,66**	3,75	**20,4**	200	-

Q90 – value of 90 % of exceeding

Beránková et al. (2010) monitored the quality of highway runoff in the period 2008-2009 on the D1 highway and the R46 expressway (Czech Republic). The concentrations of selected metals (Cd, Cr, Cu, Hg, Ni, Pb, Zn), PAHs (fluoranthene, benzo (b) fluoranthene, benzo (k) fluoranthene, benzo(a)pyrene, benzo (g,h,i) perylene, indeno (1,2,3-cd)pyrene), C10-C40 and chlorides were monitored. Toxicity tests have been carried out for a comprehensive assessment of the impact on aquatic organisms. The results of analyses of water samples taken on established sampling profiles in 2008-2009 confirmed a relatively low load by the monitored groups of pollutants (metals, polyaromatic hydrocarbons). High concentrations of chlorides have been confirmed in the samples (Table 4). Due to the differences in the nature of the sampling points (open tank, underground tank, waterline, and collector) and other influencing factors, it was not possible to draw conclusions about the extent of the load on the individual sections. Part of the pollutants occurred as particulate associated substances flowing off the highway surface and then settled in the resting zones. To measure the content of pollutants, single samples were taken in sludge deposited at the bottom of settlement tanks. Increased concentrations of Cr, Cu, Zn, slightly even Cd were detected, which already indicate anthropogenic load (Table 5). The concentration of Ni in the sludge exceeded the limits for contents in the waste (limit value 80 mg/kg) and for all-purpose land use. The PAHs sludge load was not high. The C10-C40 content reached below-limit values (about 80-90 mg/kg). In 2008, a sample of duckweed (*Lemna* sp.) was collected from the settlement tank at the end of the growing season. The sample was analysed for selected heavy metals. The results of the analysis showed high concentrations especially Zn (802 mg.kg^{-1}). The toxicity of the runoff was tested on three organisms - the green alga *Desmodesmus quadricauda* and the crustaceans *Daphnia magna* and *Thamnocephalus platyurus*. Results indicated acute toxic effects on all of the tested aquatic organisms. Acute toxicity to invertebrates and algae was detected in samples of highway runoff taken from D1 (61.5, 72 km) and R46 in February and March (Figure 5) The inhibitory effect on the growth of the algal culture *Desmodesmus quadricauda* has been demonstrated in more than 60 % of the samples of highway runoff. Regression relationship between concentrations of harmful substances and acute toxicity to algae was determined. The results showed that the increase in chloride, nickel and zinc concentrations contributed to increased acute toxicity to algae. The negative impacts of highways and parking lots runoff on the aquatic ecosystem was confirmed.

Table 4 Summary results of measurements on profiles D1 and R 46 for the period 2008-2009 (after Beránková et. al, 2010)

limit by Reg. 229/2007	**250**	**0,7**	**35**	**25**	**0,1**	**40**	**14,4**	**160**	**200**	**200**	**0,1**
Parametre	**Cl**	**Cd**	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**	**Fluoranthene**	**Σ 6 PAH**	**C10-40**
Profile	**mg/l**	**µg/l**	**µg/l**	**µg/l**	**µg/l**	**µg/l**	**µg/l**	**µg/l**	**ng/l**	**ng/l**	**mg/l**
D1 - 5,3 km	871,9	0,3	5,3	36,8	0,2	5,6	6,0	102,4	26,0	52,9	0,355
D1 - 35 km	560,4	0,2	13,8	6,9	0,1	4,8	2,3	23,7	10,1	25,1	0,05
D1 - 61,5 km	4263,2	2,0	3,3	28,0	0,098	135,3	6,3	338,2	9,5	19,4	0,163
D1 - 72 km	2359,0	0,783	5,2	27,1	0,079	35,7	5,4	177,5	11,6	19,5	0,182
D1 - 149 km	269,9	3,7	20,0	199,7	0,1	47,6	56,4	3108,3	19,1	27,3	1,9
D1 - 233 km	140,1	0,2	1,7	2,2	0,1	3,4	1,6	6,2	10,2	17,7	0,01
R46 - 13,5 km	1794,1	0,5	6,1	26,0	0,1	4,6	2,7	86,9	51,2	72,4	0,10
R46 – 17 km	3870	0,81	15,5	59,6	0,1	11,7	7,7	281,8	38,2	55,8	0,26

Legend: grey colour – values above limit defined in Reg. 229/2007.

Table 5 The results of chemical analyses of the sludge from settlement tanks on 61.5 and 72.0 km of D1 in 2006 and 2009 (Methodological Instruction of the Ministry of the Environment, 1996 – limit A - natural substance in nature, limit B - an artificially established value, this value exceedance may have a negative impact on human health and the environment), (after Beránková et. al, 2010)

Sample number	Pb [mg/kg]	Cd [mg/kg]	Ni [mg/kg]	Cr [mg/kg]	Cu [mg/kg]	Hg [mg/kg]	Zn [mg/kg]	As [mg/kg]
limit A/B	**80/250**	**0,5/10**	**60/180**	**130/450**	**70/500**	**0,4/2,5**	**150/1500**	**30/65**
limit by Reg. 294/2005	**100**	**1**	**80**	**200**		**0,8**	**-**	**10**
2006 61,5 km	42,5	0,61	213	126	148	1,6	-	7,3
2006 81,5 km,	128	0,28	121	516	76,3	0,02		4,9
2009 61,5 km	71,3	0,68	214	134	228	0,07	1140	6,3
2009 72 km	81,3	1,12	214	147	379	0,13	1490	7,4
Sample number	**anthracene [mg/kg]**	**fenanthrene [mg/kg]**	**fluoranthene [mg/kg]**	**pyrene [mg/kg]**	**benzo(a)pyrene [mg/kg]**	**indeno(c,d) pyrene [mg/kg]**	**benzo(g,h,i,) perylene [mg/kg]**	**benzo(b) fluoranthene [mg/kg]**
limit A/B	**0,1/40**	**0,15/30**	**0,3/40**	**0,2/40**	**0,1/1,5**	**0,1/4**	**0,05/20**	**0,1/4**
2006 61,5 km	0,049	0,545	0,582	0,509	0,055	0,027	0,026	0,098
2006 81,5 km	0,023	0,15	0,132	0,097	0,01	0,003	0,003	0,016
2009 61,5 km	0,002	0,025	0,061	0,032	0,023	0,006	0,042	0,038
2009 72 km	0,001	0,009	0,03	0,043	0,006	0,010	0,022	0,019

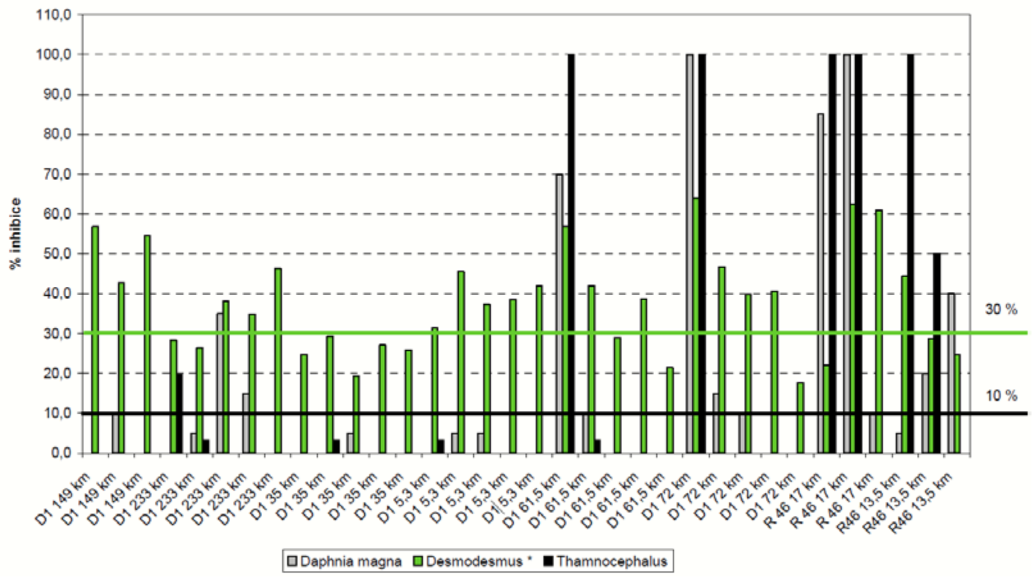

Figure 5 Results of toxicity measurements on D1 and R46 profiles in 2009 (after Beránková et al., 2010)

4 Runoff effect on groundwater

4.1 Introduction

Whilst both surface and subsurface waters are susceptible to contamination from road runoff, impacts on ground water are generally considered to be of a lower magnitude due to the presence of overlying soils which can act as a barrier by offering the opportunity for processes such as absorption to and/or immobilisation by the soil, bacterial degradation and storage effects to take place. Contaminants introduced into the soil-rock-groundwater system will spread within the system only if a transport mechanism is available, for example, a flowing liquid. As soon as the contaminant reaches the subsurface water in the unsaturated or saturated zone, its fate determine various processes such as physical processes (advection, dispersion, evaporation, filtration and degassing); geochemical processes (acid-base reactions, adsorption-desorption, ion exchange, oxidation-reduction, precipitation-dissolution, retardation and complexation) and biochemical processes (transpiration, bacterial respiration, decay and cell synthesis). Contaminants are carried by moving groundwater (advection) and travel at the same rate as the average linear velocity of groundwater. The process of dispersion acts to dilute the contaminant and reduces its concentration. For example, because of hydrodynamic dispersion, the concentration of a waste plume will decrease with distance from the source. Dispersion increases with increasing groundwater velocity and aquifer heterogeneity. Chemical reactions, such as adsorption-desorption and ion exchange, can retard the rate of contaminant movement. Various physical factors are used to assess groundwater vulnerability (e.i. recharge, unsaturated zone characteristics, the groundwater flow system). Groundwater vulnerability to contamination varies depending on the degree of protection provided by the physical environment. It is necessary to evaluate the various physical and hydrogeological conditions affecting the vulnerability. Results of this evaluation of the groundwater vulnerability will be the subdivision of an area into subareas that have different classes of vulnerability (Zaporozec et al., 2002).

The risk can, however, be higher in karst areas where runoff may drain directly in to the aquifer with little or no natural attenuation. Once underground water resources are polluted, the water may then pose a threat to public health, either through consumption of the water or body contact, the cost for treatment increases and the aquatic environment is impaired or destroyed (Bruen et al., 2006).

4.2 Case studies

Howard et al. (1993) determined annual retention rates of de-icing salts in an urban watershed in Canada. A chloride mass balance study was performed on the Highland Creek basin; one of 14 major sub-catchments in the Metropolitan Toronto and Region watershed. The catchment had an area of 104 km^2 and groundwater recharge was estimated to be 162 mm per year. Total aquifer thickness was estimated to be 30 m and the water table depth varied up to 20 m. Groundwater flow velocities were thought to be in the range of 10-100 m per year. The basin was crossed by Highway 401, and by a grid of two- and four-lane arterial roads about 1.5-2 km apart. These arteries and numerous secondary roads were regularly salted throughout the winter season. Chloride input to the catchment was determined from municipal records. These showed that the catchment received approximately 10,000 tonnes of chloride annually, predominantly in the form of NaCl de-icing chemicals. Chloride output was estimated from stream flow and electrical conductivity measurements recorded at 15-minute intervals over a two-year period. The results of the mass balance revealed that 45 % of the salt applied to the catchment was removed by surface run-off before the following winter, when a new salting season began. Most of the chloride removed was flushed from the catchment surfaces during the winter in which it was applied (Table 4). The remainder entered temporary storage in shallow sub-surface waters within the monitored aquifer. If present rates of salt application were maintained, it was predicted that average steady-state chloride concentrations in ground water discharging as springs within the monitored basin would reach an unacceptable 426 ± 50 mg/L potentially within a 20-year time frame. The value of 426 mg/L represents a three-fold increase over present average baseflow concentrations and is nearly twice the drinking water quality objective of 250 mg/L maximum acceptable concentration.

Table 6 Chloride balance summary for the period 1 November 1988 to 31 October 1991 (after Howard et al.,1993)

	Salting Season	Total Input (t)	Total Output (t)	Baseflow Load (t)	Corrected Output[1] (t)	Salt Output (as % of salt applied during salt season)	Total %
1988-89	Winter (1 Nov.–30 April)	10,486	2137[2]	486[2]	1651[2]	>15[2]	>34
	Summer (1 May–31 Oct.)	NIL	2889	867	2022	19	
1989-90	Winter (1 Nov.–30 April)	11,228	4562	1135	3427	31	45
	Summer (1 May–31 Oct.)	NIL	2699	1089	1609	14	
1990-91	Winter (1 Nov.–30 April)	9173	3651	1318	2333	26	>26
	Summer (1 May–31 Oct.)	NIL	–	–	–	–	

Notes [1] Output has been corrected for baseflow load.
[2] Data only available for March and April.
– Not determined.

A study by Perera et al. (2013) re-examined the findings of Howard et al. (1993) using a more comprehensive dataset. It analysed the temporal variability of chloride concentration in Highland Creek baseflow for the purpose of gaining greater insight to the mechanisms of watershed salt transport and release. Monitoring of dry-weather flow chloride concentrations in the Highland Creek watershed of the eastern Greater Toronto Area indicated the presence of a previously unrecognised, dual porosity aquifer system whereby preferential flow associated with “urban karst” exerts a significant influence on baseflow chloride concentrations early in the year. Annual chloride mass balances undertaken for the studied area indicated approximately 40 % of the chloride applied enters the shallow aquifer each year. Since this chloride exceeds the amount of chloride released via baseflow there is a net accumulation that leads to a gradual long-term increase in baseflow chloride concentrations. Historical data for chloride in late season baseflows are included in Figure 6. Chloride concentrations increased by approximately 200% between 1970 and 2010. The rate of increase in groundwater chloride concentration is consistent with the rate of development of the study area. Once salt application rates stabilise, the system will work towards steady state such that chloride entering the watershed system is matched by the chloride released. Assuming current salt application rates, it was determined that chloride concentrations in stream baseflow during late season will eventually reach an average, steady state concentration of 505 mg/L, significantly higher than current levels of about 300 mg/L in late summer/fall. To achieve a reduction in chloride concentration to 250 mg/L (the Canadian drinking water quality aesthetic guideline) would require reducing the mass of chloride entering the aquifer annually by 50 %.

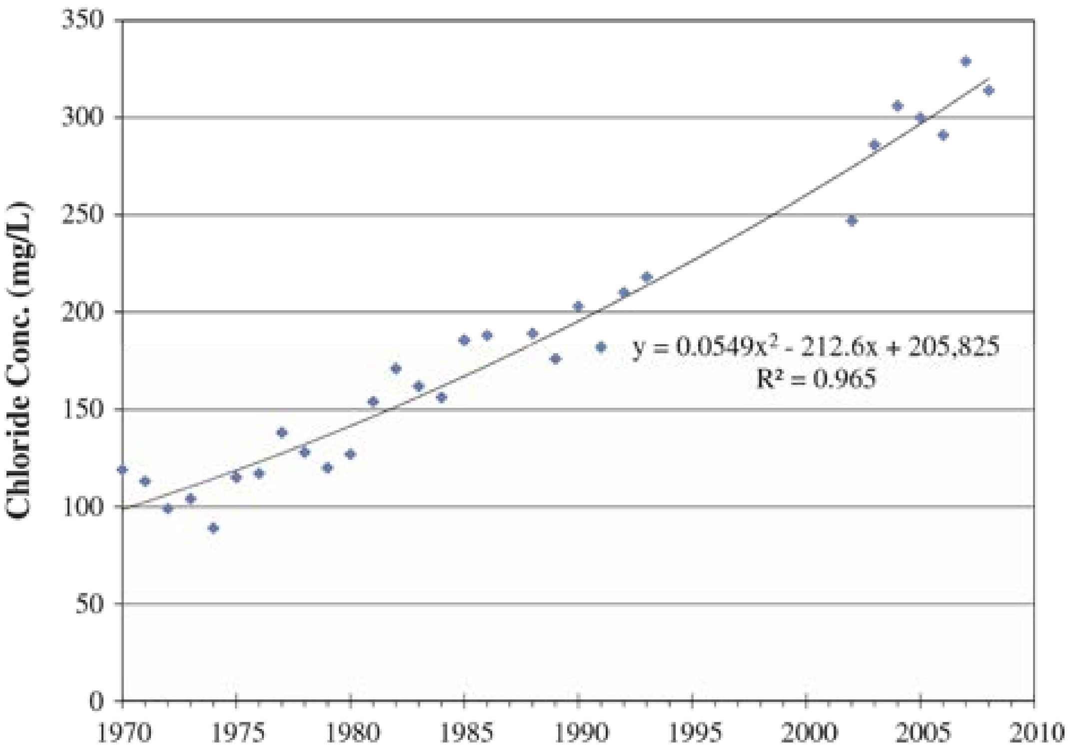

Figure 6 Chloride concentrations in Highland Creek stream samples under dry-weather flow conditions during the late season months of September and October (after Perera et al., 2013)

Granato et al. (1995) studied the mobilization of the major and trace constituents of highway runoff in groundwater potentially caused by de-icing-chemical migration. Analyses of groundwater samples collected at test sites adjacent to Route 25 in south eastern Massachusetts (USA) during February and August 1991 and March, August and November 1993 indicated that concentrations of targeted constituents in groundwater were substantially higher down-gradient of the highway in comparison to those determined up gradient from the highway (Figure 7). For some constituents, down-gradient concentrations were more than an order of magnitude greater than up gradient concentrations. Highway runoff containing road salts and calcium-magnesium acetate seemed to have the greatest effect on groundwater quality at a test site where highway runoff discharges locally to the land surface. This site had an open-drainage system typical of many highways. Seventy-five percent of annual recharge, which carries highway runoff through the unsaturated zone to groundwater, occurred during the winter road-salting months. Cation exchange was indicated by decreases in sodium concentrations and corresponding increases in calcium concentrations in infiltrating water. pH decreased significantly in down-gradient groundwater due to acidification from road salt. None of the measured concentrations of the major and trace constituents in groundwater exceeded national primary drinking-water standards (Report EPA/822/R-94-001). However, secondary standards were exceeded for chloride (250 mg/L) and manganese (0,05 mg/L), and recommendations for surface-water criteria (Report EPA/440/5-86-001) were exceeded for chloride (230 mg/L), cadmium (0,0039 mg/L), and copper (0,018 mg/L).

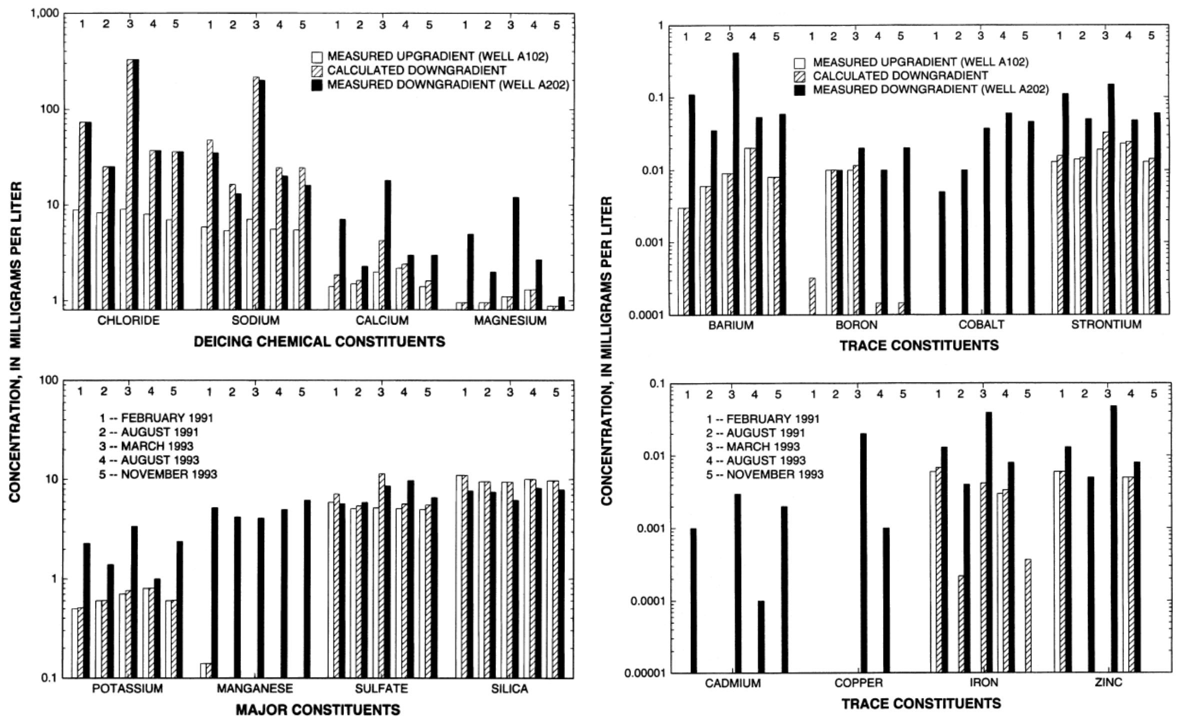

Figure 7 Constituent concentrations in groundwater measured up gradient and downgradient from the highway compared with constituent concentrations expected from road salt, based on the excess chloride measured in downgradient groundwater at site A along State Route 25 in southeastern Massachusetts (after Granato et al., 1995)

Rivett et al. (2016) evaluated the dynamic impact on the receiving water environment of de-icing salt application at a major highway (motorway) interchange in the UK for two recent severe winters. The possible influence on the public water supply (PWS) wells that were situated relatively close to both the motorway network and a local small watercourse (Battlefield Brook) was monitored. The Battlefield Brook is known to recharge groundwater within the study area, and thus has potential to serve as a line source to the underlying aquifer. Logged stream electrical-conductivity (EC) to estimate chloride concentrations, streamflow, climate and motorway salt application data were used to assess salt fate. Stream chloride loadings were responsive to salt applications and climate variability influencing salt release. Salt persistence on the highway under dry-cold conditions was inferred from stream observations of delayed salt removal. During warm periods of increased precipitation, salt application is occasional and estimated chloride stream is low. Steep chloride declines were ascribed to precipitation onset causing rapid salt runoff, increased streamflow and dilution. Peaks occurred typically immediately following snowfall where road salt applications had been increased, or where there was yet further snowfall, or else there were sustained cold conditions with a lack of precipitation as rain. The limited water entering the stream was hence mostly derived from salt-induced melting of highway snow as other precipitation water was largely locked up as snowfall or ice in the wider catchment. Small spikes of stream chloride were attributed to low-level salt applications intended to address night time freezing risks, but rapidly washed off by ensuing rainfall. Alternatively, salt may persist on the highway and possibly increase with repeated salt applications. Accumulated highway salt was being eventually washed off by later precipitation. This study has quantitatively demonstrated that winter discharge to groundwater from a stream receiving highway storm discharges containing de-icing salt may constitute a significant line-source of chloride to the underlying aquifer. Figure 8 provides a conceptual model of de-icing salt application pathways to an abstraction well alongside other example chloride sources.

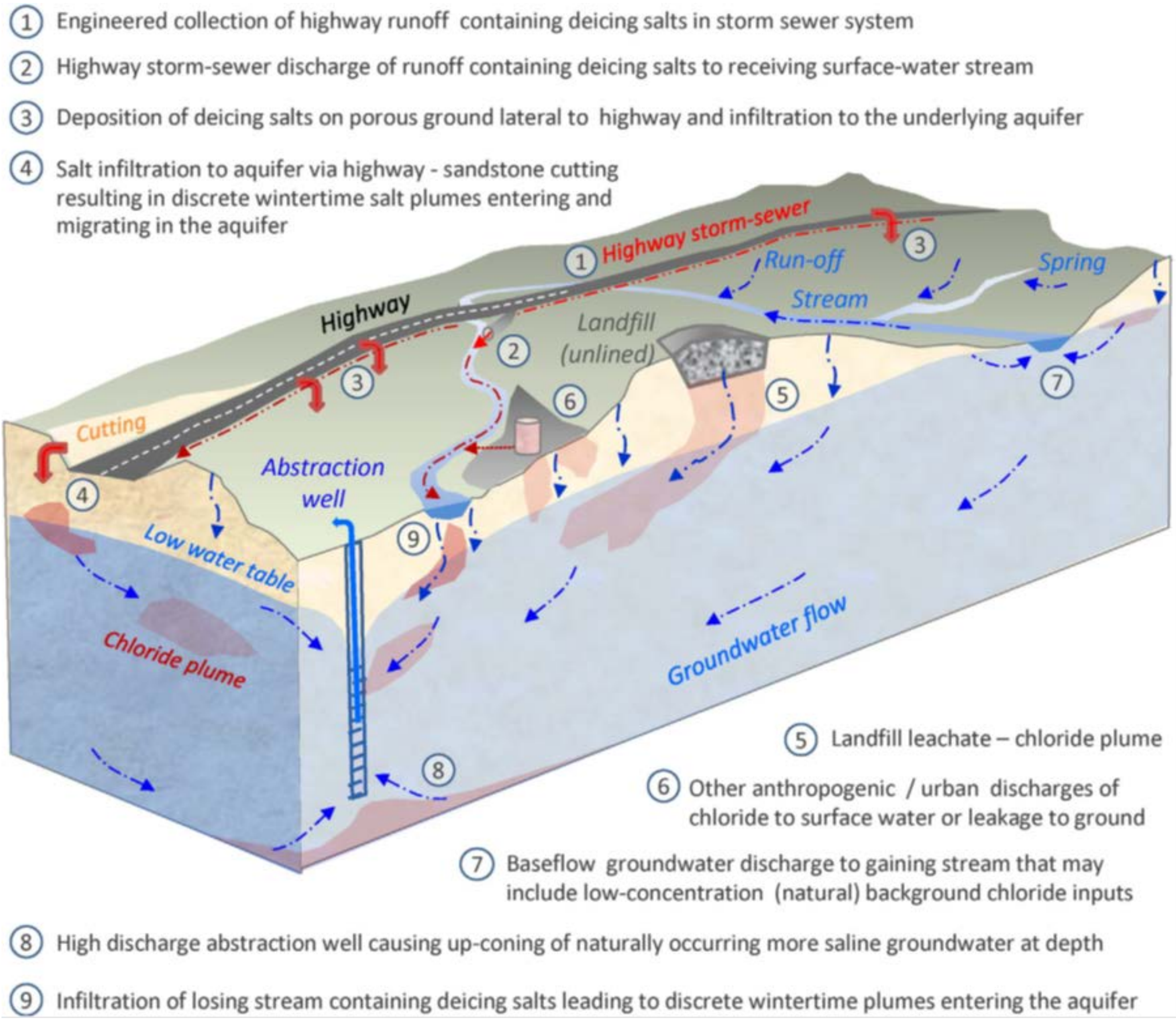

Figure 8 Conceptual model of de-icing salt application and other potential sources of chloride that pose risks to a supply well (after Rivett et al., 2016)

The model is anticipated to have generic relevance. The annual mass of de-icing salt imported to catchments remains very high (approx. 237-257 t) and is likely to significantly exceed other anthropogenic inputs. Stream chloride (via EC) was predicted to exceed the stream Environmental Quality Standard (250 mg/L) for 33 % and 18 % of the two winter time periods. Maximum stream concentrations (3500 mg/L; 15 % sea water salinity) were ascribed to salt-induced melting and drainage of highway snowfall without dilution from, still frozen, catchment water. Mild and severe winter stream infiltration may account for around 21 to 54 % respectively of the long-term PWS well chloride increase. Around 80 % of the storm de-icing salt discharges (and majority of highway salt applied) remained within the stream.

Watson et al. (2002) studied the effects of highway-de-icer application on ground-water quality at a site in North Western Indiana (USA) using a variety of geochemical indicators. Forty-three monitoring wells were installed in an unconfined sand aquifer (the Calumet aquifer) near Beverly Shores along two transects that approximately paralleled groundwater flow in the Calumet aquifer and crossed US–12. US–12 is a highway that receives Indiana's highest level of maintenance to maintain safe driving conditions. Ground-water quality and water-level data were collected from the monitoring wells, and precipitation and salt-application data were compiled from 1994 to 1997. The water-quality data indicated that chloride was the most easily traced indicator of highway de-icers in ground water. The principal source of chloride and sodium in ground water from the uppermost one-third to one-half of the Calumet aquifer and down-gradient from US–12 was from a halite highway-de-icer source. Chloride and sodium in the deep parts of the aquifer originated from natural sources. Chloride and sodium from highway de-icers were present in the aquifer throughout the year. The highest concentrations of chloride and sodium in ground water were determined in samples collected during the spring and summer from wells open to the water table within about 9 feet of the highway. Peak concentrations in water-table wells down-gradient from US–12 were as much as 1,000 times greater than concentrations in ground water from up gradient water-table wells. For Cl, the downgradient wells values are up to about 1000 mg/L, while the upgradient wells have values of about 1-10 mg/L. For Na downgradient wells have the highest values up to 600 mg/L, upgradient wells 1-80 mg/L. Peak concentrations decreased with increased distance from US–12. Chloride concentrations in ground water that were attributable to highway de-icers also were found in tested wells about 400 feet down-gradient from US–12 during the fall and winter and at greater depths than in wells closer to US–12. Chloride concentrations exceeded the U.S. Environmental Protection Agency's (USEPA) secondary maximum contaminant level of 250 milligrams per litre for drinking water at seven wells down-gradient from the highway during late winter, spring, and summer samplings. The chloride standard was exceeded only in water from wells with total depths that are less than 10 feet below land surface. Sodium concentrations in water periodically exceeded the USEPA drinking-water equivalency level of 20 milligrams per litre in both the uppermost (de-icer affected) and lower one-thirds of the aquifer. Sodium concentrations in ground water down-gradient from US–12 and in the upper 5 feet of the aquifer also occasionally exceeded drinking-water standards for sodium (160 mg/L) as set by the State of Florida and a standard for taste (200 mg/L) as set by the World Health Organization. Chemical analyses of the sand composing the aquifer indicated that cation exchange decreased the mass of de-icer-related sodium in ground water, although the sand has a limited capacity to sustain the process. Automated daily measurements of specific conductance, correlated to chloride concentrations, indicated that some de-icer was retained in the aquifer near the highway throughout the entire year and acted as a continuous chloride source for ground water. The relative effects of processes affecting the distribution of de-icers in the subsurface will vary from site to site depending on differences in hydrogeology, de-icer application methods and climatic history.

Leitão (2007) summarized the results of 37 case-studies which reviewed soil and groundwater pollution by heavy metals in the vicinity of roads. Findings indicated that Cadmium (Cd) is a very mobile element with a toxic effect at very low levels. It was found that its presence in soil and groundwater close to the roads was usually low, in concentrations lower than the intervention values (for soil, the quality values used are defined in Dutch legislation (MHSPE, 1994) as target and intervention) and VMA (for groundwater, the quality values used are defined in Portuguese legislation (DL 236/98, 1998) as recommended (VMR - 5 µg/l) and admissible (VMA - 1 µg/l)) defined, respectively. It is possible to observe clear variations over time and with distance to the road, due to its high mobility, with a few samples exceeding the target value (1 µg/l) for groundwater. Cr is a metal that tends to accumulate in the soils due to its low mobility. Usually, Cr concentration is constant with depth and distance showing a low capacity for mobilization to the groundwater. This is also evidenced by the low concentration of Cr in groundwater (all values reported were lower than 5 µg/L). Cu is a low mobility metal, especially in areas with some organic matter content, not significantly influenced by changes in pH or salt content. The concentration of Cu in the vadose zone was higher than 20 µg/L, but all samples except one were below 50 µg/L. The higher values were closer to the road. In most case-studies a reduction of Pb with the distance to the road and in depth was observed. In 10 of the 14 case-studies analysed under the POLMIT project (POLMIT, 2000), the water in the vadose zone had concentrations below 10 µg/L. Groundwater had Pb concentrations below VMA (50 µg/L) too, and the pattern of concentrations was very dependent on soil conditions. Zn is a very mobile element especially in sandy and acid soils. The existence of Zn in groundwater varied from case to case, depending on the existing physico-chemical conditions. Most of studies found Zn concentrations below 500 µg/L.

Effects of highway runoff percolating through unsaturated sand on the quality of water in the Biscayne aquifer (Florida) were studied by Howie and Waller (1986). The scope of the study included chemical analyses of lithologic material at two test sites and one control site, and chemical analyses of storm water runoff collected from an adjacent swale, water from the unsaturated zone, and water from the saturated zone underneath the swale at two test sites. The chemical constituents selected as tracers were lead, zinc, iron, cadmium, chromium, nickel, mercury, nitrogen, and phosphorus. Results of this study suggested that storm water runoff from highway surfaces was a contributing factor in the accumulation of selected constituents in near-surface lithologic material. Analyses of lithologic material in highway swales that have been in contact with percolating storm water at two test sites indicated concentrations of lead ranging from 1,000 to 6,600 µg/kg, iron from 490 to 2,400 µg/kg, and zinc from 90 to 1,800 µg/kg between 0 and 0.5 foot below land surface. Concentrations of this magnitude were not detected in lithologic samples collected at an unaffected control site, with concentrations of lead bellow 10 µg/kg, iron from 9 to 40 µg/kg, and zinc from 6 to 24 µg/kg. Results of trace metal and nutrient analyses indicated that there were no obvious water quality effects on the surficial aquifer caused by highway runoff. The data collected for dissolved trace metals indicated slight concentrations in storm water samples with subsurface water samples usually indicating decreases in concentration below about 0.5 to 1 foot of unsaturated material. For most trace metals, the apparent decreases with depth were statistically significant based on nonparametric analysis of variance. Concentrations of dissolved nitrogen and phosphorus, however, were somewhat homogeneous and were not inferred to be significantly different with depth. Water hardness as calcium carbonate at both sites indicated a significant difference with depth.

Wang et al. (2018) quantified the impact of highway transport on groundwater in turfy swamps in the Changbai Mountain area from the two aspects of heavy metal and hydrochemical characteristics. This region is the main headstream of the Mudan River and an important water conservation area in Jilin Province. Groundwater samples were collected in July–August 2017 at distances of 5, 15, 50, and 200 m, from the highway, respectively at three sites. Nineteen parameters (Cu, Pb, Zn, Cd, Cr, Ni, Hg, As, pH, TDS, Ca^{2+}, Mg^{2+}, Na^+, K^+, $SO4^{2-}$, Cl^-, $HCO3^-$, $NO3^-$ and F^-) were determined. Results indicate that metals Cu, Pb, Zn, Cr, Cd, the ions Na^+, K^+ and Cl^- in groundwater were negatively affected by highway transportation, and the maximum affected distance of these pollutants varied from 15 to 100 m. The content of the most of these pollutants in roadside groundwater decreased exponentially with the distance from the highway, as did the heavy metal pollution index (HPI) and index indicates the relative contamination of different metals separately and manifests the combined effects of all metals (C_d). The values of HPI and C_d in these three sites ranged from 46.8 to 78.4 and −4.9 to −2.9, respectively. However, neither the concentrations of pollutants, nor the HPI and C_d indexes, exceeded the water quality standard. The low pollution levels of heavy metals are related to the strong adsorption capacity of turfy soil towards metals. Road transport activities increased the Cu, Pb, Zn, Cr, Cd, Na^+, K^+ and Cl^- content in roadside groundwater in turfy swamp. With the increase of highway operation time, it will inevitably have a great influence on the groundwater quality of these wetlands. For this reason, Wang et al (2018) suggested long-term monitoring to protect the sustainable development of turfy swamp.

Kaminski and Korzak (2007) examined the concentration of selected elements (zinc, lead, nickel, cadmium, chromium, cobalt, copper and iron) in soil and ground water and evaluated the level of migration of these elements in perpendicular system in regard to road axis. Four experimental sections of forest roads situated in marshy areas have been selected for chemical analysis. As a result of analyses carried out it was noted that total content of heavy metals in ground water taken from wells located in the vicinity of forest roads as well as concentration of heavy metals in soils collected near forest roads were low and did not exceed current Polish standards.

Ličbinský et al. (2012) presented results of a study focused on the leaching of compounds from materials used for road construction and on groundwater contamination by these materials. Boreholes were drilled at three localities with a different age of the pavement. Samples of individual structural layers were taken at all localities and at one locality a seeping waters sampler was installed. Concentrations of EPA's 16 priority pollutant polyaromatic hydrocarbons (PAHs) including coronene were determined in samples taken from individual layers of the road body. The obtained results indicate that concentrations of substances decreased with increasing depth. The highest concentrations of PAHs at localities with an older pavement surface from the nineteen fifties or nineteen sixties were found in the layer below the asphalt pavement surface. Samples from individual layers were also analysed for the concentration of selected metals. Iron was found to be the most frequent element that was present in concentrations by two orders higher than Ba (182 - 1733 mg.kg^{-1}) and Mn (380 - 880 mg.kg^{-1}) and by three orders higher than other analysed elements. The water extracts of soil samples were tested for toxicity at one trophic level. The tests were conducted using the freshwater crustacean *Thamnocephalus platyurus* as the indicator. Toxicity tests showed the negative effects on tests organisms at all localities in the first layer, i.e. the asphalt pavement surface, and in the third or fourth layer. On the other hand, samples with the highest concentrations of PAHs were classified as non-toxic. Two samples of percolation water were taken and were analyzed for the determination of organic and inorganic pollutants and for toxicity tests. In the samples the same PAHs as in soils were detected. The results of analyses indicate that the highest concentrations in groundwater were found for pyrene, benzo(a)pyrene and fluoranthene. With the exception of the last mentioned, these are the same compounds that were also present in the highest concentrations in road layers. Concentrations of metals were also determined in groundwater. Concentrations of alkali metals were relatively high, particularly of sodium and potassium. This could be caused particularly by the interaction of the percolation water with rubble and a binder, by which these compounds are released into the water. There is probably also a significant contribution of spreading used for road maintenance in winter. Both samples of groundwater were classified as slightly toxic.

5 Case study database

An integral part of this study is a case study database (Annex 1). The database matrix has 7 main fields of information, namely:

(i) Country;

(ii) Title of study;

(iii) Name of authors;

(iv) Name of institutions;

(v) Abstract

(vi) Methodology

(vii) Conclusions

The case study database contains 70 studies that were selected from more than 100 analysed reports, articles and studies. 36 of the case studies in the developed database are focused on the chemical characterization of road runoff and were undertaken within a range of European countries (e.g. Sweden, Denmark, France, UK, Switzerland, Germany, Poland, Czech Republic, Italy, Portugal and Slovenia) and in Australia, New Zealand, the USA, China and Japan. Another 23 studies deal with the influence of runoff on surface water. These studies that were conducted in the USA, Canada, Norway, UK and the Czech Republic, mostly include both the results of chemical analysis of surface water and runoff, as well as the results of toxicological analyses or biological monitoring. 14 studies are focused on groundwater issues namely contamination with chlorides and metals. These studies were carried out in Canada, the USA, UK, France and Portugal.

6 Conclusions

This report provides an overview of available case studies evaluating effects of road runoff on the water quality of waterbodies into which highway runoff is discharged either directly or indirectly (through drainage and treatment systems). Analysed studies evaluated impact on receiving water bodies both from chemical and ecotoxicological perspectives. From a chemical point of view, a wide range of parameters such as pH, temperature, COD, BOD, NH_3, TSS, hardness, DO, organic compounds, metals, chlorides have been evaluated in several studies (Moy and Crabtree, 2002a, b, c, d, e, f; Swandener et al., 2014, Meland et al., 2010a, b). Some studies included only metal concentrations measurements (Yousef et al, 1984; McNeill and Olley, 1998, Kayhanian et al., 2008, Perdikaki and Mason, 1999), the others were focused only on PAHs (Grund et al., 2016, Adamec et al., 2004). Water pollution by both PAHs and metals was quantified and evaluated by Marešová et al. (2002), Beránkova et al. (2008) and Beránkova et al. (2010). Elevated concentrations in receiving waters were most commonly observed for chlorides, which are released into water courses mainly in winter from de-icing agents due to winter road maintenance. They are often determined by conductivity analysis rather than chloride determination. Corsi et al. (2010); Williams et al. (1999) and Blasius and Merritt (2002) focused on contamination of surface waters by salts (chlorides) and consequent influence on aquatic invertebrates. Corsi et al. (2010) monitored the increase in conductivity with the increasing urban land use. During the sampling programme, increased conductivity was observed between November and April. Chloride concentrations exceeded USEPA acute and chronic water-quality criteria at 55 and 100 % of monitored sites. Williams et al. (1999) recommended the monitoring of groundwater contamination by chlorides through the monitoring of spring water because the sampling of springs is simpler than the direct collection of groundwater. During one year, the concentration of chlorides in the four of the most contaminated springs increased by 21-34 %.

Impacts on living organisms were evaluated *in situ* or in laboratories in several case studies. Ecotoxicological tests on aquatic organisms (invertebrates, fish and algae) have been carried out in laboratories. Mayer et al. (2011) applied a battery of bioassays involving daphnia, bacteria and fish to assess the toxicity of road runoff from the roads with different transport intensities. Results found that *Daphnia magna* and *Ceriodaphnia dubia* were sensitive to increasing concentrations of salts (chlorides) and metals in runoff. The toxicity of runoff increased with the increasing concentrations of high-molecular PAHs in runoff solids. Kayhanian et al. (2008) assessed the toxicity of runoff from 3 highways using three species of freshwater organisms (water flea, fathead minnow, green algae) and two species of marine organisms (purple sea urchin and luminescent bacteria). The most sensitive test species was the sea urchin; fathead minnows were more sensitive than water flea. The main source of toxicity was the presence of copper and zinc in the runoff. Corsi et al. (2010) found toxicity for *Ceriodaphnia dubia* and *Pimephales promelas* in 7 of 13 samples of streams due to high concentrations of chlorides. For the other monitored streams, chronic toxicity was demonstrated for 72 % of samples and acute toxicity for 43 % of samples. Blasius and Merritt (2002) demonstrated that *Gammarus* (Amphipoda) may be affected by NaCl concentrations greater than 5000 mg/L over a 24 h exposure period. Williams et al. (1999) identified the amphipod *Gammarus pseudolimnaeus* as being intolerant to acute and chronic exposure to high concentrations of chlorides that later had an effect on reproductive behaviour. In some cases, however, the toxicity of the runoff was not confirmed (Swadener et al., 2014, Adamec et al., 2004). Marešová et al. (2002) identified crustaceans as the most sensitive of the tested organisms (algae, bacteria, crustaceans) when testing the runoff toxicity from settling reservoirs. Beránková et al. (2010) found the acute toxicity of highway runoff for algae and invertebrates in February and March. Based on a regression analysis, it found that increased concentrations of chloride, nickel and zinc resulted in increased algae toxicity. Some studies confirm the first flush effect, i.e. the reduction of runoff toxicity during the storm (Mayer et al., 2011, Kayhanian et al., 2008). Greater toxicity of sediments than aqueous samples was found due to their higher contamination with metals and PAHs (Beránková et al., 2008, 2010, Mayer et al., 2011).

Several studies focused on biological monitoring and assessing the diversity of communities using BMWP, ASPT and Shannon Index of Diversity. Blasius and Merritt (2002) did not detect differences that could be attributed to road salt between upstream and downstream locations in the diversity and composition of invertebrate functional feeding groups. Similarly, studies by McNeil and Olley (1998) and Moy and Crabtree (2002a, b, c, d, e, f) mostly did not observe variation in the diversity in stream below the outfalls of highway runoff. The reduction of the diversity and abundance of the benthic communities was observed on the highway side of the lake (Baekken, 1994). Greater runoff influence on diversity was observed in summer, when flow rates were lower. (Perdikaki and Mason, 1999). Pollution by tunnel wash water has led to a change in behaviour and a reduction in fish growth in downstream areas (Meland et al., 2010, a, b). Grung et al. (2016) reports that elevated concentrations of PAHs in the sediment pond resulted in DNA damage and higher levels of CYP1A enzyme in fish.

Bioaccumulation studies evaluated the accumulation of metals (Perdikaki and Mason, 1999, Yousef et al., 1984) and PAHs (Grund et al., 2016, Baekken, 1994, Meland et al., 2010b) in aquatic plants and organisms. Baekken (1994) observed an increased concentration of Cd and Zn in bivalves and lead concentrations in perch liver and PAH in perch flesh above the background levels. Beránková et al. (2010) monitored high concentrations of Zn in duckweed (*Lemna* sp.). Meland et al. (2010b) reported the accumulation of trace elements in the gills of the fish.

Infiltration of runoff may also affect the quality of the groundwater in the vicinity of roads. The quality of groundwater is threatened by increased concentrations of chloride due to winter road maintenance, and also by contamination of metals that enter the groundwater from the runoff. Howard et al. (1993) reported that 10,000 tonnes of chlorides were received annually by the Toronto catchment area. Of this mass, 45 % of applied salts were removed from the catchment by surface water with the rest infiltrating to sub-surface waters. Perera et al. (2013), in a follow-up study, found that approximately 40 % of the chlorides applied each year enter local aquifers. The rate of increase of chlorides in groundwater was calculated to correspond to the rate of development in this area. To maintain the current chloride concentrations in drinking water, it would be necessary to reduce the mass of chlorides entering the aquifers by 50 %. Due to acidification from road salt, the pH of groundwater was reduced (Granato et al., 1995). For some monitored chemical constituents in groundwater, concentrations determined down-gradient of a highway crossing point were more than an order of magnitude higher than up-gradient concentrations. None of the measured concentrations of the major and trace constituents in groundwater exceeded national primary drinking-water standards. However, secondary standards were exceeded for chloride and manganese (Granato et al. 1995). Salt discharges in winter were a significant source of chloride for aquifers (Rivett et al., 2016). EQS (250 mg/L) in surface water was exceeded for 33 % and 18 % of two monitored winters. Exceedances of this standard were also observed by Watson et al. (2002) in 7 out of 43 monitored wells during winter, spring and summer. Some de-icing agents retained in the aquifer near the highway throughout the entire year and acted as a continuous chloride source for ground water. Concentrations of metals in groundwater are mainly related to their mobility and physical-chemical characteristics (Leitão, 2007). Zn is very mobile and its occurrence in groundwater was the highest among the monitored metals (concentrations less than 500 µg/L). The concentrations of other monitored elements (Cr, Cu, Pb, Cd) in groundwater were comparatively low. Wang et al. (2018) quantified the impact of highway transport on groundwater in turfy swamps and found out that road transport activities increased the Cu, Pb, Zn, Cr, Cd, Na^+, K^+ and Cl^- content in roadside groundwater. For this reason, suggested long-term monitoring to protect the sustainable development of turfy swamp. Equally Kaminski and Korzak (2007) examined the concentration of selected elements in forest roads situated in marshy areas. They reported that concentration of heavy metals near forest roads were low and did not exceed current Polish standards. The same organic and organic contaminants were found in the percolating water passing through the pavement layers as in the road layers (Ličbinský et al., 2012).

A deficiency identified within the analysed studies is that they focus mostly only on a short-list of parameters with effects separately considered e. g. as pollutants concentrations or ecotoxicity tests. Important parameters that should also be monitored in water are temperature, pH, amount of dissolved oxygen and water hardness. Some organisms may be sensitive to changes of these parameters in addition to their impacts on the physico-chemical behaviour of monitored substances (and hence their biological effect). In most studies, hydrological characteristics of monitored streams, such as flow rates that may affect contaminant concentrations, are not reported. Some studies only involve sample collection over a limited period of time and do not include the influence of seasonal variability in concentrations of pollutants which can lead to distortion of the results. Mainly, acute effects are evaluated, with little if any data available on chronic effects. Evaluation of the impact on organisms would be strengthened by the inclusion of both toxicological tests and by monitoring species diversity and abundance.

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