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Review

The pollution conveyed by urban runoff: A review of sources

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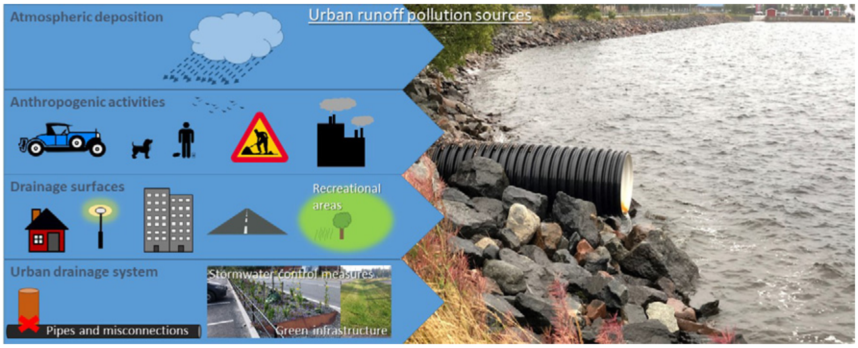
HIGHLIGHTS

- Current knowledge of stormwater pollution sources varies among source categories.
- Atmospheric deposition, transportation and metallic materials are major sources.
- Some data from older stormwater quality studies may be obsolete and no longer valid.
- New materials and pollutants necessitate future re-examination of pollution sources.
- More attention needs to be payed to pollutants of emerging concern.

GRAPHICAL ABSTRACT



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ABSTRACT

Urban stormwater and snowmelt pollution contributes significantly to the deterioration of surface waters quality in many locations. Consequently, the sources of such pollution have been studied for the past 50 years, with the vehicular transportation sector and the atmospheric deposition identified early as the major pollution sources. In search for mitigation of this pollution, source controls, besides other measures, were recognised as effective pollution mitigation tools, whose successful implementation requires a good knowledge of pollution sources. Even though great research efforts have been exerted to document specific sources of urban runoff pollution, or specific groups of pollutants present in urban runoff, a comprehensive overview of all known contributing sources is still missing. This review contributes to closing this gap by compiling findings of previous research and critically synthesizing the current knowledge of various stormwater pollution sources. As the emphasis is placed on the sources, the related issues of implications for urban surface water quality and possible source controls for individual sources are touched upon just briefly, where required. The review showed that the atmospheric deposition, vehicular transportation-related activities and metallic building envelopes continue to be among the major pollution sources, which have been studied in a far greater detail than other sources. Furthermore, it was noted that because of the rapid advances in clean manufacturing and pollution control technologies, a large part of the body of data on stormwater quality available in the literature should be considered as historical data, which may no longer describe well the current conditions. Progressing historical data obsolescence, combined with continuing releases of new materials and chemicals, and, in some cases of new substances of potential concern,

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into the environment, suggests that the identification of important stormwater runoff/snowmelt pollution sources, and the associated pollutants, has been and will remain to be a work in progress.

Contents

1. Introduction

Urban runoff, comprising stormwater and snowmelt in regions with seasonal snow, is widely recognised as a major transport vector of pollutants released in the urban environment, and, therefore, a significant contributor to the deterioration of urban receiving waters quality (Sartor and Boyd, 1972; Lee et al., 2007; Björklund et al., 2018). Urban stormwater as a concern in managing surface water quality has been studied since the mid-20th century and Åkerlindh (1950) and Weibel et al. (1964) were among the first to point out this concern. Some years later, the sources of pollutants occurring in urban stormwater and snowmelt attracted increased research interests, and the vehicular transportation-related sources were among those identified early as important pollution sources (Sylvester and DeWalle, 1972; Laxen and Harrison, 1977; Horkeby and Malmquist, 1977; Malmqvist, 1983). The early heuristic studies typically addressed such conventional pollutants as total suspended solids (TSS), chemical or biochemical oxygen demand (COD/BOD), trace metals (mainly Cd, Cr, Cu, Ni, Pb and Zn), and various species of N and P.

Starting in the 1980s, a number of ambitious, large-scale research programmes mapping stormwater quality over various land-use areas were established, with the U.S. Nationwide Urban Runoff Program (NURP) being the most extensive one (U.S. Environmental Protection Agency (EPA), 1983). In the U.S., a follow up on the NURP resulted in a recent National Stormwater Quality Database (NSQD), comprising data from >9100 urban runoff events, observed on six main urban land uses: residential, commercial, industrial, freeways, institutional, and open space. NSQD includes >100 various constituents, many of which are rather uncommon and documented only by sparse data (Pitt et al., 2018). Because of large variations of stormwater quality in time and place (Butler et al., 2018), the above extensive databases are helpful in e.g., estimating expected pollutant loads from whole catchments, planning treatment solutions where needed, or comparing pollution control options. However, these databases comprise data with a relatively coarse spatial resolution, equal to the catchment scale, and hence cannot be used for identifying the specific sources of diffuse pollution within the catchment, or their mitigation by source controls.

The EU Water Framework Directive (Directive 2000/60/EC) addressed the need to mitigate diffuse pollution and its amending priority pollutant directive (Directive 2013/39/EU) contributed to broadening the list of substances included in stormwater quality studies. The contemporary research often confirms the earlier findings concerning e.g., TSS, metals and polycyclic aromatic hydrocarbons (PAHs) (Zgheib et al., 2012; Gasperi et al., 2014), and produces new data on emerging pollutants in stormwater, such as phthalates (Björklund et al., 2011; Zgheib et al., 2012; Markiewicz et al., 2017; Müller et al., 2019), alkylphenols (APs) (Bressy et al., 2011; Björklund et al., 2011; Zgheib et al., 2012; Gasperi et al., 2014; Markiewicz et al., 2017; Müller et al., 2019), bisphenol-A (BPA) (Gasperi et al., 2014), and recently introduced pesticides (Zgheib et al., 2012; Gasperi et al., 2014). Moreover, because of rapid advancements in clean manufacturing and pollution control technology, much of the data from stormwater quality studies published in the literature may be dated and no longer valid in today's conditions. This resulted from the implementation of new technologies reducing emissions, as well as regulatory actions leading to virtual elimination of specific environmentally harmful substances. For instance, the earlier reported impacts of PAHs from steel plant emissions (Gu et al., 2003), or Pb from the use of leaded gasoline (Kayhanian, 2012) may no longer exist, or have been strongly abated.

Source controls, i.e. pollution prevention, represent fundamental steps towards minimising the presence of pollutants in urban stormwater and the concomitant potentially negative effects in receiving water bodies. Source control policies are recognised as the most cost-effective management tool in dealing with low-level diffuse pollution and there is a strong need to further advance this pollution control tool (Marsalek and Viklander, 2011). Often the opportunities for actually controlling the sources of pollution are limited or hard to achieve, and then it may be more feasible to control the release activities rather than the primary sources. Previous success stories concerning legislative actions leading to effective source control include the phasing out of lead from gasoline (Kayhanian, 2012; Huber et al., 2016). Recently implemented environmental policies such as the reduction of Cu in vehicle brake pads in the U.S. (U.S. EPA, 2015) and elsewhere are promising examples for future runoff quality improvements. For planning implementation of source control policies, adequate knowledge of the sources of urban stormwater pollution is essential (Loganathan et al., 2013).

Even though the urban stormwater pollution sources have been studied for more than half a century, a comprehensive overview of specific sources and the associated pollutant groups is still missing, and this review contributes to closing this gap by evaluating the current state of knowledge of various sources contributing to the pollution of urban stormwater. The scope of the review is fairly broad and focuses on well-documented sources, as well as the emerging sources, which have not been yet fully assessed in the scientific literature, and includes various types of pollutant groups, and the thermal pollution of stormwater. The approach used in this review, with focus on the sources rather than the nature or classification of pollutants, is a rather unique approach infrequently seen in other papers. In the context of this paper, urban stormwater was defined as urban runoff, generated by rainfall, snowmelt or both, normally conveyed by the public storm sewers in separate sewer systems, or other conveyance elements of contemporary drainage systems, including green infrastructure and stormwater control measures (SCMs). Hence, the terms 'urban runoff' and 'stormwater' are used synonymously, recognising that 'runoff' reflects the hydrological context and 'stormwater' focuses on the transported medium. Runoff conveyed in combined sewer systems, or in private drainage systems and/or treatment facilities, e.g. airport runoff, was considered outside the scope of this paper. Several conditions and factors may affect the transport and release of urban stormwater pollutants, but details about these factors were also considered outside the scope. When searching for relevant literature, contributions assessing urban runoff as a confirmed, or potential, transport vector were included. However, detailed discussions of the effects of urban runoff pollution on the receiving waters and control measures for mitigation of such effects were considered outside the scope of this review.

2. The classification of sources into categories

There are many ways of classifying and grouping sources of stormwater pollution, and the partition between the groups can be quite complex, with the boundaries between source categories rarely well defined. Depending on the classification approach taken, some sources may contribute the pollution to several source categories in different times or ambient conditions. For instance, one pollution source may be an operating vehicle releasing pollutants into the atmosphere (one source category), which may later be deposited on roads and other surfaces, and may thereby also contribute pollutants directly to road surfaces (another source category) and, hence, to stormwater runoff. The partitioning of sources is further complicated by source interactions (Thorpe and Harrison, 2008), caused, e.g., by resuspension of deposited pollutants, which in studies of stormwater quality and the application of study data may lead to double counting of pollutant contributions from a specific source. Thus, pollutant transformations and temporary storage during conveyance make it difficult to identify the contributions of pollution from specific sources on the basis of discharge concentrations (Lundy et al., 2012).

Various approaches to classification of stormwater pollution sources were noted in the literature. Loganathan et al. (2013) divided the sources contributing to road sediment pollution into the intrinsic sources (within the road environment, i.e., atmosphere, pesticide use, pedestrian littering, fences and railings, and all vehicular transportation related sources), and the extrinsic sources (buildings, industry, soils and vegetation). Lundy et al. (2012) classified sources based on land use types and activities, including, e.g., construction sites, highway surfaces, roof surfaces, open spaces, misconnections, and more. Similarly, Petrucci et al. (2014) grouped diffuse pollution sources into four main groups: activity-related (e.g., road transport emissions), land cover-related (buildings and infrastructure), behaviour related (e.g., pesticide and fertiliser use), plus atmospheric deposition (AD).

Building on the classifications of sources by the earlier researchers, in our review, the sources are grouped into four main categories: atmospheric deposition, drainage surfaces, anthropogenic activities, and urban drainage systems. Anthropogenic activities were defined as the activities practiced in the urban environment by humans, or resulting from such actions of humans, with varying frequencies and intensities. Examples of activity-related dynamic sources are, e.g., vehicular transportation, winter road maintenance, or construction work. The strength of these sources varies in time while the activity progresses, as in the case of construction works, which are temporary sources of pollutants, or may vary with season, as in the case of winter road maintenance or fertilisation of lawns. Other sources in the urban environment are static and generally relate to land covers, or surfaces; they may contribute pollution in wet weather regardless of any anthropogenic activities. The latter sources include, e.g., building envelopes, parks and infrastructure elements. However, the magnitude of the pollutant releases from surfaces may vary with a number of factors, such as the material age (Robert-Sainte et al., 2009), rainfall intensity (Charters et al., 2016), and others. The quantitative contribution of pollution from specific sources have been accounted for where such information was available and considered applicable. For some specific source categories, especially the activity related sources, the quantitative contribution of pollutants cannot be allocated to specific sources because of several sources producing simultaneous pollutant contributions to the same mass of runoff water, or because of high unpredictability, e.g., in the case of spills. Fig. 1 further describes the classification of pollution sources used in this review and also serves as a table of contents for the topics discussed herein. The sources are listed in the boxes and arrows indicate the main transport pathways of the pollution, and interactions among the source categories. This way of grouping sources also facilitates the discussion of source controls. Building upon the discussion in Marsalek and Viklander (2011), the strength of sources related to anthropogenic activities may often be more feasible to control by restricting the activity leading to pollutant release, instead of eliminating the actual source. On the other hand, the sources constituting drainage surfaces may be controlled by substitution of environmentally friendly materials for those releasing pollutants.

3. Important sources of urban stormwater pollution

3.1. Atmospheric deposition

Atmospheric deposition (AD) facilitates transfer of polluting substances and materials in the atmosphere to the urban catchment surface, with either precipitation or deposition in dry weather. Thus, AD comprises wet and dry components, with the former one contributing directly to pollution of surface runoff and the latter one contributing potentially after washoff of substances and materials from catchment surfaces (Marsalek et al., 2008). With respect to the runoff pollution, AD is particularly important in urban areas, which are characterised by numerous sources of air pollution and high likelihood of pollutant washoff into stormwater runoff, facilitated by the presence of impervious areas and runoff conveyance networks (Hobbie et al., 2017). Consequently,

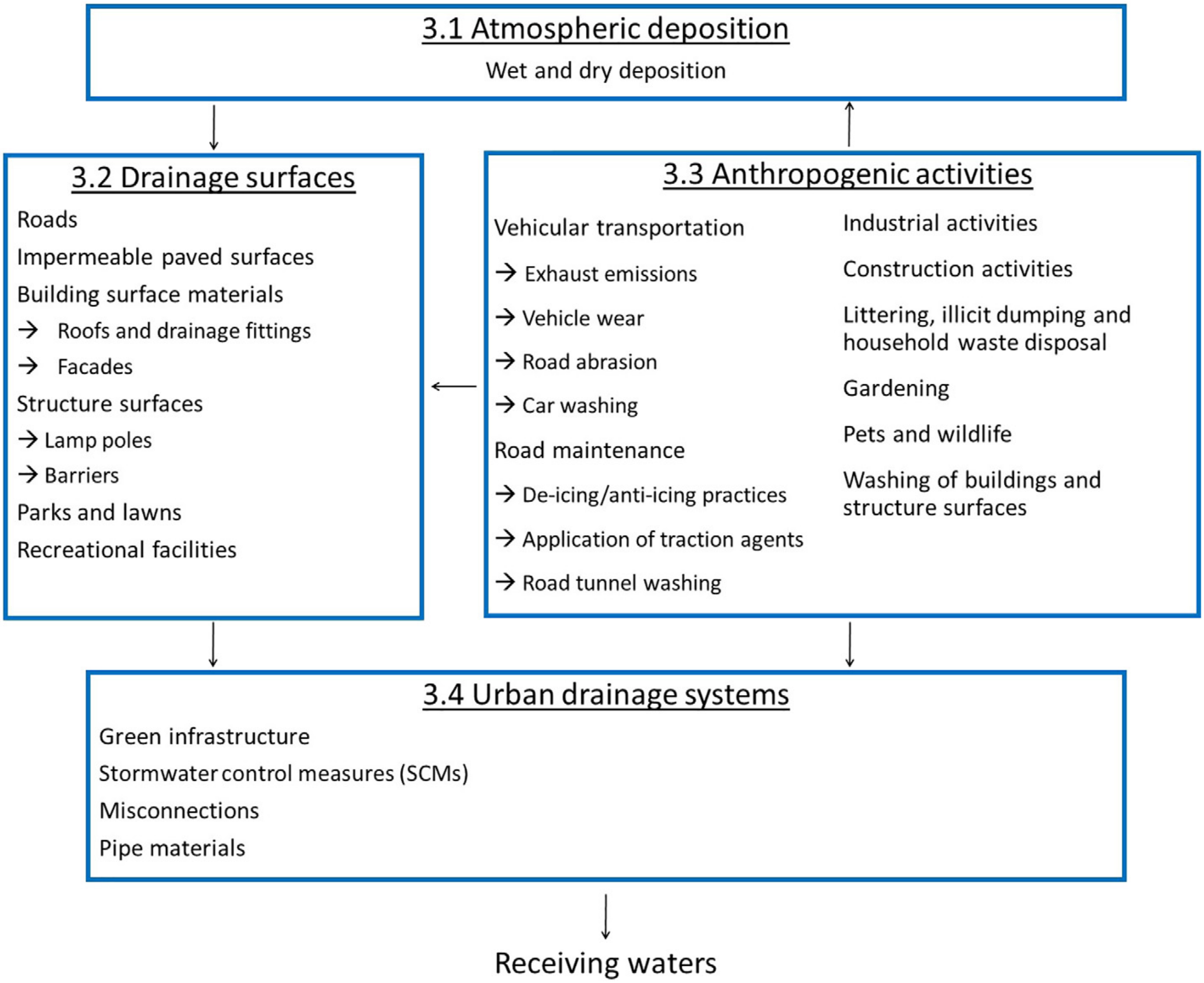

Fig. 1. Grouping of urban stormwater pollution sources, section numbers for the main source categories, and pollution transport pathways indicated by arrows.

AD has been generally identified as an important source of pollutants found in urban stormwater (e.g., Barkdoll et al., 1977; Malmqvist, 1983; Brinkmann, 1985; Davis et al., 2001; Pitt et al., 2004; Sabin et al., 2005; Davis and Birch, 2011; Gunawardena et al., 2012; Gunawardena et al., 2013; Petrucci et al., 2014; Omrani et al., 2017).

Concerning the stormwater pollution, the atmosphere serves more as a pollutant transport pathway, rather than an actual source of observed pollution (Petrucci et al., 2014). Atmospheric pollutants, from both natural and anthropogenic sources, are brought into the urban environment by emissions and atmospheric transport from local, regional and remote sources. The multitude of sources and atmospheric transport scales contribute to high variability of AD chemical characterisation and rates of deposition in both time and space (Malmqvist, 1983; Brinkmann, 1985; Boom and Marsalek, 1988; Rocher et al., 2004; Sabin et al., 2005). Among the sources listed in Fig. 1, AD is unique by including pollution sources located outside of urban catchments and contributing via long-range atmospheric transport, as documented, for example, by Li et al. (2007).

A quantitative assessment of AD contributions to pollutant exports with stormwater runoff from urban catchments is rather challenging because of such reasons as complexities of various pollution sources and air transport processes (Brinkmann, 1985; Morselli et al., 2003), changes in wet deposition chemistry on contact with surfaces (Polkowska et al., 2011), and uncertain pollutant entry from dry depositions into stormwater (Morselli et al., 2003; Murphy et al., 2015; Al Ali et al., 2017). Furthermore, Colman et al. (2001) pointed out that while the wet deposition is relatively simple to monitor, monitoring dry deposition and discerning between the vehicle-related deposition and the ambient urban atmospheric deposition is rather difficult and raises questions about comparison of data from various references. The lack of discernment between these two sources may then lead to double counting AD contributions. Finally, the historical AD data (e.g. Boom and Marsalek, 1988) have to be understood in the context of environmental conditions and AD sources existing when the data were collected. With time, some historical AD sources may be weakened by advances in implementation of clean air technologies and environmental policies reducing air pollution, but others (e.g. traffic) may be strengthened with the increasing intensity of the activity.

Among the chemicals transported through the atmosphere, there is a distinct class referred to as volatile organic compounds (VOCs). VOCs are widely used in many products (fuels, fumigants, paints, pesticides, precursors in manufacturing chemicals, refrigerants, and solvents) (Lopes and Bender, 1998; Moran et al., 2006), or are formed as by-products of processes occurring in urban areas (e.g., trihalomethanes formed as water disinfection by-products). Consequently, VOCs are ubiquitous in the urban environment, and may enter both surface waters, including stormwater, and groundwater from many sources (Lopes and Bender, 1998). Because of their toxicity, a number of VOCs were included in the U.S. EPA list of 129 priority pollutants and 12 of those were investigated in the NURP studies of urban runoff quality (U.S. EPA, 1983). In the list, this group includes three BTEX compounds, benzene, ethylbenzene and toluene, and their derivatives (including chlorinated benzenes). VOCs were rarely detected in the limited NURP sampling (U.S. EPA, 1983): four compounds in two to six of the 28 sampled cities, and in six samples per site or less. The primary sources of VOCs in stormwater can be identified as vehicle fuels (released as vehicle exhaust gases and particles, or at gasoline filling stations and in fuel spills), atmospheric transport and deposition, and industrial releases. Using the taxonomy of stormwater pollution sources introduced earlier (Fig. 1), and recognising broad uses and distribution of VOCs in urban areas, they are not discussed as a separate group, but in the appropriate review sections (Section 3.1 Atmospheric deposition, Section 3.3.1 Vehicular transportation, and Section 3.3.3 Industrial activities). One characteristic seems to be common to the stormwater studies of VOCs and the respective publications: although concerns about the stormwater pollution by VOCs were expressed, e.g., where stormwater serves for reuse (Liu et al., 2018b) or the recharging of groundwater (Yu et al., 2017), or that VOCs should be considered in planning community pollution reduction strategies (Li et al., 2018), the reported VOC concentrations were not assessed with respect to their environmental relevance and regulatory context.

Specific assessments of AD potential contributions to stormwater pollutant export loads were reported in about 20 references for total suspended solids (TSS), nutrients, metals, and trace organics. Such contributions were rated in this review with respect to their magnitude as "significant" (i.e., when representing 10–100% of stormwater export loads), or as "trace-to-low" contributions (up to several percent). A summary of such an assessment is presented in Table 1.

The references listed in Table 1 suggest that AD is a "significant" source of such stormwater quality parameters as TSS, nutrients (N and P), and metals associated with traffic and local industrial releases. The metals reported in AD included Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, Ti, V, W, Zn, and Zr, but most studies focused just on the most environmentally relevant metals in urban stormwater, Cu, Pb, and Zn. For TSS and the above chemicals in urban catchments, the annual loadings in AD typically equalled 20–100% (or more) of the respective loads exported with urban stormwater. A number of studies emphasised the importance of local stationary sources of AD, in comparison to mobile emission sources (traffic and road dust). Other studies reported the presence of trace organics (mostly priority pollutants) in urban stormwater. Those occurred in low or trace amounts, and mostly in wet atmospheric deposition.

Table 1
Two-level assessment of atmospheric deposition contributions to urban stormwater quality.

Conventional pollutants	AD contributions equal to 10–100% of stormwater export loads
TSS	Murphy et al. (2015)
Nutrients (N, P)	Hobbie et al. (2017); Hou et al. (2012)
Metals associated with traffic (Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, Ti, V, W, Zn, Zr)	Colman et al. (2001); Davis et al. (2001); Davis and Birch (2011); Gunawardena et al. (2012); Gunawardena et al. (2013); Huston et al. (2009); Liu et al. (2018a); Morselli et al. (2003); Murphy et al. (2015); Omrani et al. (2017); Ruban et al. (2010); Sabin et al. (2005); Al Ali et al., 2017 (a small, high traffic catchment, with AD contributions < 10%)
Trace organics (mostly priority pollutants)	AD contributions reported in low (only PAHs) or trace amounts (other substances than PAHs)
PAHs	Boom and Marsalek (1988); Ruban et al. (2010); Schifman (2014); Al Ali et al. (2017)
Pesticides	Ruban et al. (2010)
PFCs (perfluorinated compounds) or poly- fuorinated chemicals	Kim and Kannan (2007); Kwok et al. (2010); Murakami et al. (2009); Scott et al. (2006); Xiao et al. (2012); Zhao et al. (2013); Zushi and Masunaga (2009)
Xeno-estrogenic compounds	Peters et al. (2008)

In summary, the current level of knowledge of AD indicates that depending on local circumstances, AD may be an environmentally important source of solids and conventional and emerging pollutants conveyed by urban stormwater, with some of such pollutants originating outside of the catchment. While wet deposition can be readily measured, dry deposition measurements and the entrainment of deposited chemicals by runoff are highly uncertain. In relation to other pollutant sources, AD may expand the list of pollutants found in urban stormwater and can be effectively controlled only at the source.

3.2. Drainage surfaces

3.2.1. Roads and impermeable paved surfaces

Paved surfaces contribute to pollution of stormwater through two different processes: Mechanical wear of pavement surface by vehicle tires (discussed later in Section 3.3.1), and by elution of chemicals into water running off the pavement surface, discussed in this section. Coal tar-based sealants, used to enhance the structural integrity and visual appearance of asphalt driveways, sidewalks, walking trails, parking lots, roads and some playgrounds, were found to be a major source of PAHs in urban stormwater pond sediments in Minnesota, USA, accounting for 67% of the detected PAHs (Crane, 2014). Bitumen is a constituent of asphalt that contains PAHs and, therefore, a potential source of PAHs in runoff. Because of this environmental risk, the release of PAHs and selected metals from bitumen and asphalt has been studied in static and dynamic leaching tests (Brandt and De Groot, 2001; Legret et al., 2005). Both studies demonstrated that pollutant leaching was weak and most concentrations were below the existing regulatory limits for potable water, and concluded that leaching from asphalt is not an important source of metals or PAHs.

Similar to the effect of concrete pipes and other concrete structures on stormwater quality, the use of concrete pavement has the potential to provide the alkalinity needed to raise the runoff pH and alter the metal element partitioning (Sansalone and Buchberger, 1997). Concrete pavement materials were also reported to leach Cr; the source of Cr was Portland cement used in the concrete production (Kayhanian et al., 2009). The leaching of Cr was mainly influenced by the material age, permeability, temperature, and contact time. In a study comparing pollutant contributions from three pavement types, asphalt and crushed stone driveways produced the highest pollutant concentrations, while paver driveways had significantly lower pollutant concentrations (Gilbert and Clausen, 2006). Even though not explicitly stated by the authors, it is likely that the pollutants in their study were originally derived from e.g. AD, or traffic related activities, deposited on the driveways, rather than from the driveway material. Zhang et al. (2017) reported that natural stone-paved pedestrian paths produced the highest PAH surface load because of a high total solids content, with solids acting as pollutant carriers. However, the PAHs deposited in their study on the pedestrian path originated from other sources (i.e., vehicle emissions, coal and wood combustions, engine oil spills and vehicle tire debris). Furthermore, as urban impermeable surfaces are heated up by solar radiation, especially during summer months, they become an important source of thermal pollution of runoff, threatening cold water fisheries (Van Buren et al., 2000; Thompson et al., 2008; Janke et al., 2009).

In summary, notwithstanding the mechanical pavement wear by vehicle tires, discussed in Section 3.3.1, urban paved surfaces elute measurable contributions of PAHs and alkalinity. This source is closely related to, though less important than, the pavement wear by vehicle tires (Section 3.3.1). In regions with cold water fisheries, the thermal pollution of urban runoff may also be of concern to be fully considered in stormwater management planning.

3.2.2. Building materials and structure surfaces

All surfaces in contact with rainwater or stormwater surface runoff can potentially affect the quality of such waters. Various structures exposed to rainwater or stormwater exist in the urban environment: buildings, sculptures, lamp poles and crash barriers, to name a few. Their surfaces comprise a wide range of different materials, many of which have been studied extensively from the stormwater quality perspective. Similar to other impervious surfaces in the built environment, these surfaces can also act as sources of thermal pollution (Van Buren et al., 2000). Clay and concrete roofing tiles have been reported to increase runoff pH (Sulaiman et al., 2009; Lee et al., 2012), and corrosion of metal structures, commonly found on e.g. roofs and facades, is known to be a major source of Cu and Zn in runoff (Malmqvist, 1983). Gromaire et al. (2001) demonstrated that roof runoff was the main source (>80%) of Cd, Pb and Zn in wet weather flows in a combined sewer system serving a densely populated residential area in central Paris, because of the corrosion of roof cover materials. Slate roofs, window frames, ventilation holes, etc. on historical buildings were the main source of Pb, while zinc sheets used in roofs and gutters were the main source of Zn and Cd, which is a minor constituent of zinc products. The architecture in the catchment studied was considered representative of many large cities in Europe (Gromaire et al., 2001). Galvanised steel, used on various surfaces in the urban built environment such as building envelopes, crash barriers and lamp poles, is another important source of Zn. For example, Robert-Sainte et al. (2009) reported an annual average Zn concentration of 3081 μg/L in runoff from galvanised steel panels. Moreover, Boller and Steiner (2002) estimated that copper used on roofs accounted for up to 50% of the total copper load in urban drainage systems. Cu concentrations in runoff from copper panels were characterised by a median concentration of 1905 μg/L, over 20 rain events (Winters et al., 2015), and Müller et al. (2019) reported an average Cu concentration of 3090 μg/L from tests of triplicate panels for six rain events. In recent years, Corten weathering steel has been increasingly used on sculptures, architectural features and other surfaces in the urban environment, and was shown to contribute Ni to runoff (Raffo et al., 2016; Müller et al., 2019). However, metal sheets protected by surface coatings often exhibit lower releases of metals compared to raw metal sheets (Robert-Sainte et al., 2009; Müller et al., 2019). Furthermore, even though often considered as minor sources, non-metallic materials may also contribute metals to runoff. For example, pressure-treated and waterproof wood was reported to contribute Cu to runoff (Clark et al., 2008; Winters et al., 2015) and the EPDM (Ethylene propylene diene monomer) roof cover released Zn (Winters et al., 2015).

The release of various pesticides from building materials was reported in a number of studies. For instance, Burkhardt et al. (2011) showed that large amounts of pesticides (diuron, terbutryn, carbendazim, irgarol 1051) used in building envelopes entered stormwater. Similarly, Gromaire et al. (2015) reported high concentrations of benzalkonium chloride, used as a de-mossing agent, in roof runoff. Bucheli et al. (1998) concluded that bituminous roofing membranes were a major source of the pesticide Mecoprop, added as root penetration protector, and this finding was later confirmed by e.g. Vialle et al. (2013), who reported occurrence of Mecoprop in all samples of runoff collected in a suburban catchment, with measured concentrations of up to 4.8 μg/L. Paints applied to different kinds of surfaces can act as a source of Pb (Davis and Burns, 1999), pesticides (Jungnickel et al., 2008) and polychlorinated biphenyls (PCBs) (Jartun et al., 2009). Old façade plaster (mainly mid-20th century) was also found to be a potential source of PCBs (Andersson et al., 2004). Compared to the state of knowledge of metal and pesticide releases from building materials and other structure surfaces in the built environment, much less is known about releases of other organic micropollutants. Buildings were found to be a dominant source of APs in field measurements of atmospheric deposition and stormwater runoff in a suburban catchment near Paris (Bressy et al., 2011). They reported a median concentration of 0.56 μg/L nonylphenols (NPs) in building runoff and hypothesised that these NPs originated from additives in paints, pesticides, plastics and civil engineering materials. However, the specific materials (and, thus, potential sources) present in the catchment were not accounted for in their publication. Björklund (2010) performed a substance flow analysis and identified roofing and façade materials as important sources of phthalates, and concrete as an important source of NPs and nonylphenol ethoxylates (NPEOs). Pilot-scale outdoor experiments of runoff from building surface materials confirmed the release of phthalates (with average concentrations up to 455 μg/L Diisononyl phthalate, DINP), NPs (up to an average concentration of 26 μg/L) and lower levels of NPEOs from PVC roofing membranes, as well as the release of NPs (average concentration 0.99 μg/L) and lower levels of NPEOs from bitumen felt roofing (Müller et al., 2019).

In summary, a plethora of materials, both organic and inorganic, have been used in building envelopes to protect buildings and structures against adverse climatic effects and those of atmospheric deposition and stormwater. Most of these coatings, representing biocides and paints, contain potentially harmful substances, which eventually find their way into stormwater. Continuing development of new materials, surface coatings, paint formulations, etc. and their applications in the urban environment represent new potential sources of known or emerging pollutants in urban runoff. Material substitutions, policy controls and locally implemented stormwater treatment processes represent feasible control options. This source is closely related to the washing of building surfaces (Section 3.3.8), which represents a maintenance activity conducted with relatively low frequency (compared to the frequency of occurrence of wet weather).

3.2.3. Green areas (parks, lawns, urban forests and sport facilities)

Contemporary urban planning recognises the value and benefits of sustaining or expanding green areas in towns and cities (Swanwick et al., 2003). Such areas, of various forms and sizes, range from boulevards with grass and trees, to neighbourhood parkettes, district or central parks, and various sport fields, including golf courses. Among the benefits of urban green areas, one can name contributions to mitigation of urbanisation impacts with respect to the water cycle, local microclimate, air pollution, noise, urban ecology, and carbon footprint, and thereby the green areas contribute to the health and general well-being of urban population. Particularly significant are the benefits of urban forests, which were defined as the sum of all the trees within an urban area (City of Toronto, 2013), and in Toronto (Canada), such a forest encompasses 10.2 million of trees. The presence of green areas leads to inevitable interaction with traditional “grey” drainage infrastructure, including releases of dissolved organic matter (DOM) and its mixing and transport with stormwater from other sources in street gutters and storm sewers. DOM not only plays a major role in forming aquatic life systems, but also affects the fate and transport of pollutants (McElmurry et al., 2014; Zhao et al., 2015; Zhao et al., 2019; Yuan et al., 2019).

Characteristics of DOM at the catchment drainage outlet reflect both the catchment cover, determined by land use (Zhao et al., 2015), and local environmental factors. In urban areas, DOM measured by DOC (dissolved organic carbon) was in runoff at lower concentrations than in natural landscapes, and among the environmental factors, significance was attributed to solar radiation, and water temperature and conductivity (McElmurry et al., 2014), which are all modified in urban areas. Concerning the pollutant transport, Zhao et al. (2019) demonstrated that DOM in stormwater runoff from various land use areas exhibited strong binding affinities for heavy metals (Cd, Cu, Ni and Pb) and hence influenced their migration and transformation. Such processes are of great interest in stormwater treatment by vegetated surfaces, as demonstrated by Yuan et al. (2019) for grassy swales. While the environmental significance of DOM in urban stormwater has been demonstrated for a limited range of samples and environmental conditions, there is a need for continuing research expanding the published findings and defining their consequences for urban drainage planning and design.

A summary of sources of chemicals and faecal indicator bacteria in urban green areas is presented in the following paragraphs of the current section, under the catchment surfaces (covers), and in Section 3.3.6 on gardening, under activities.

Residential lawns and turf areas (e.g., sports fields, golf courses and parks) in the urban environment were shown to be 'hotspots' of nutrient input into stormwater (Center for Watershed Protection, 2003; Hobbie et al., 2017). Lawns and their soils are among the most important sources of total and dissolved phosphorus in urban runoff (Waschbusch et al., 1999; Hobbie et al., 2017), and Waschbusch et al. (1999) reported an average concentration of 0.79 mg/L P in runoff from lawns. Moreover, fallen leaves are a significant source of nutrients, especially phosphorus, in urban stormwater and accounted for >50% of the annual P output, winter season excluded (Selbig, 2016). Shaver et al. (2007) reviewed the literature data and reported that nutrient concentrations in runoff from grassy areas can be up to four times greater than those from other urban areas. Golf courses were found to be a source of pesticides in runoff (Metcalfe et al., 2016). Pervious surfaces, such as lawns, may contribute suspended solids to runoff during high intensity rain events causing soil erosion (Gromaire et al., 2001). The issues concerning soil erosion in urban areas are addressed in more detail in Section 3.3.4 on construction activities.

In recent years, the use of artificial turfs or recycled rubber mulch (e.g., in playgrounds) has become relatively common. Such a rubber mulch is an obvious source of microplastics (Magnusson et al., 2016) and a source of Zn, PAHs and phthalates released to runoff (Kanematsu et al., 2009; Bocca et al., 2009; Llompart et al., 2013). In addition, Celeiro et al. (2018) reported the presence of Cd, Pb and Cr in some rubber mulch samples and confirmed the leaching of several hazardous substances (including PAHs, phthalates, Cd, Pb and Cr) from synthetic play surfaces to runoff water.

In summary, recreational areas, parks and lawns are generally areas with high pedestrian traffic, and thus the places where littering (Sundt et al., 2014) and deposition of pet (or wildlife) faeces would commonly occur. Thus, these areas may be sources of faecal bacteria, nutrients and solids to urban runoff, assuming that they produce runoff, in spite of relatively high degree of perviousness. More information on pollution sources related to these areas can be found in Sections 3.3.5 and 3.3.7, respectively. Moreover, these areas are often closely connected to gardening activities and the associated pollution sources, such as fertiliser and pesticide applications, described in Section 3.3.6.

3.3. Anthropogenic activities

3.3.1. Vehicular transportation

Traffic related pollution has been studied extensively and the main pollution sources attributed to vehicular transportation have been identified as vehicle operation (Brinkmann, 1985), including exhausts, automotive fluid leakages (Markiewicz et al., 2017) and wear (Muschack, 1990); vehicle washing (Sörme et al., 2001; Björklund, 2010); and, road abrasion (Hvitved-Jacobson and Yousef, 1991), generating a complex mixture of pollutants. In cool temperate climate with seasonal snow, there is one more important source of pollutants: winter road maintenance involving applications of road salts and traction agents (grit materials) as discussed later in Section 3.3.2. Considering the urban stormwater pollution, the level of detail of specific transport related sources and the list of substances they contribute is much greater compared to other source categories, likely because road runoff was early identified as a major pollution source, depending on traffic intensity. In this connection, Marsalek et al. (1999) reported that among the selected sources of urban stormwater, runoff samples from two freeways (Average Daily Traffic (ADT) > 100,000 vehicles/24 h) produced the highest frequencies of moderate-to-severe toxicity detection by a battery of bioassays. In response to the reported sources of pollution in road runoff, the parameters traditionally studied include suspended solids (Westerlund and Viklander, 2006), petroleum hydrocarbons, polycyclic aromatic hydrocarbons (PAHs), BOD and COD as well as trace metals in both solid and liquid fractions (Huber et al., 2016). Even though the specific sources contributing to pollutants released by vehicular traffic have been studied extensively, their quantitative contributions to actual concentrations in urban runoff cannot be readily discerned, because of simultaneous releases from several sources (e.g., brakes, tires and exhausts). A summary of pollutants released by vehicular traffic in urban areas is presented in Table 2; a detailed commentary follows.

Vehicle exhaust gases from internal combustion engines contain such pollutants as particulate matter (particles of soot and metals), hydrocarbons (including PAHs; Markiewicz et al., 2017), benzene series (BTEX) pollutants (Liu et al., 2018b), nitrogen oxides (NO_x), carbon monoxide (CO), sulphur dioxide (SO_2), and hazardous air pollutants (e.g., Pb; Brinkmann, 1985). Vehicle exhaust pollution is partly controlled by catalytic converters, which in turn were identified as sources of Rh, Pd and Pt (Rauch et al., 2005). The phasing out of Pb from gasoline resulted in a steady decrease in Pb concentrations in road runoff in recent decades (Kayhanian, 2012), but new additives were introduced and found their way into the environment, including oxygenates, such as Methyl tert-butyl ether (MTBE), ethanol, and alkylates (Nadim et al., 2001), and methylcyclopentadienyl manganese tricarbonyl (MMT) (Geivanidis et al., 2003). However, stormwater has yet not been confirmed as an important transport vector for these substances with the exception of MMT, which may cause increased Mn concentrations in road runoff (Huber et al., 2016). Exhausts from diesel engines were found to be a source of Ni (Duong and Lee, 2011). Other related

Table 2
Sources of pollutants released by vehicular traffic in urban areas.

Specific source	Pollutants released	References
Vehicle operation		
Exhaust gases and particles	Hydrocarbons, PAHs, NOx, Ni, BTEX	Markiewicz et al. (2017); Brinkmann (1985); Huber et al. (2016); Kayhanian (2012); Duong and Lee (2011); Liu et al. (2018b)
Catalytic converters	Rh, Pd, Pt	Rauch et al. (2005)
Vehicle wear		
Tires	TSS, Cd, Cu, Zn, PAHs, microplastics	Muschack (1990); Councell et al. (2004); McKenzie et al. (2009); Legret and Pagotto (1999); Kose et al. (2008); Horton et al. (2017a)
Tire studs	W	Huber et al. (2016)
Brakes	TSS, Cd, Cu, Ni, Pb, Sb, Zn, PAHs	McKenzie et al. (2009); Hjortenkrans et al. (2007); Markiewicz et al. (2017)
Engine and vehicle body wear	Cr, Ni	Gupta et al. (1981); Ward (1990)
Body paint	Pb	Kayhanian (2012)
Wheel balance weights	Pb, Fe (steel), Zn	Root (2000); Bleiwas (2006)
Vehicle washing		
Commercial car washing facilities	Pb, Cd, Cr, Zn	Sörme et al. (2001)
	Phthalates, NPs, NPEOs	Björklund (2010)
Road abrasion		
Abrasion by tires (non-studded and studded)	TSS	Hvitved-Jacobson and Yousef (1991); Van Duin et al. (2008) Lindgren (1996)
	PAHs	Markiewicz et al. (2017)
	Microplastics	Magnusson et al. (2016); Horton et al. (2017b); Vijayan et al. (2019a)

sources of pollutants are leakages of automotive fluids, among which the engine oil is the most important source of PAHs in the traffic environment (Markiewicz et al., 2017). Gasoline stations were reported as 'hotspots' of BTEX pollutants (Liu et al., 2018b) and, thus, represented a potential source of BTEX pollutants in stormwater.

Among the non-exhaust pollution sources, tire and brake wear were identified as the most important sources (Muschack, 1990), with respect to both the environmental relevance and the number of reported studies. The main pollutants associated with these sources were identified (Table 2) as follows:

- Tires: Zn (Councell et al., 2004), Cu (McKenzie et al., 2009), Cd (Legret and Pagotto, 1999), PAHs (Kose et al., 2008) and microplastics (Horton et al., 2017a)
- Brakes: Cu (McKenzie et al., 2009), Zn, Ni, Sb, Pb (Hjortenkrans et al., 2007), Cd (McKenzie et al., 2009) and PAHs (Markiewicz et al., 2017).

Other types of vehicle wear contributing to pollution included engine wear and welded metal plating (the vehicle body), both contributing Ni and Cr (Gupta et al., 1981; Ward, 1990). Paints (Kayhanian, 2012) and wheel balancing weights (Root, 2000; Bleiwas, 2006) were suggested as important sources of Pb in the traffic environment. Concerning the latter source, the use of Pb has been banned in some jurisdictions (e.g., EU) and other materials, Fe (steel) and Zn, were substituted. Tire studs used in some northern regions are made of W and are recognised as a source of this heavy metal in road runoff (Huber et al., 2016). However, W is rarely reported in stormwater studies and does not generate specific environmental concerns, considering its absence from lists of priority substances, e.g. the EU Directive (2013/39/EU) on priority pollutants (2013) and the U.S. EPA Priority Pollutant List (US EPA, 1983). Finally, it is worth mentioning that a range of material types and compositions are used in the vehicle industry (Thorpe and Harrison, 2008), e.g., for different vehicle types or in different regions, which creates large variations in the potentially released pollution. For instance, Hjortenkrans et al. (2007) stated that their results from 2005 showed that tires could be excluded as a significant source of Cr, Cu, Ni and Pb emissions in Stockholm.

Vehicle washing releases various chemicals and materials attached to the vehicles (Sörme et al., 2001), or contained in car-care products. Rain falling on stationary or moving cars could potentially have similar washing effects. Sörme et al. (2001) studied sources of heavy metals in urban wastewater in Stockholm (in both combined and separate sewer systems) by substance flow analysis and found that commercial car washes were an important contributor of Pb, Cd, Cr and Zn. Paint attrition during car washing may produce Pb (Kayhanian, 2012) and vehicle under-spray may activate other earlier mentioned sources of pollutants (e.g., leaked engine oil deposits). Using a substance flow analysis, Björklund (2010) reported that vehicle washing and wear were important sources of phthalates, NPs and NPEOs in urban stormwater. NPEOs are commonly used in commercial car wash detergents and degradation to NPs is a plausible contribution to NP concentrations found in stormwater (Rule et al., 2006). Where commercial car washes drain to storm sewers, they are obviously also a similar source, with the difference that the commercial wash water usually passes through an oil separator.

Road abrasion is an important source of particles (TSS) (Hvitved-Jacobson and Yousef, 1991), PAHs (Markiewicz et al., 2017), and microplastics (Magnusson et al., 2016), and the abrasion process is exacerbated by the use of studded tires (Lindgren, 1996) and applications of grit in winter road maintenance. Grit particles are ground by vehicle tires against the pavement and produce fine particles, which may either be washed off, or be incorporated into pavement pores (Van Duin et al., 2008). Microplastics may originate from both the bitumen used in road pavement construction and from road marking paints (Magnusson et al., 2016; Horton et al., 2017b).

When assessing traffic related pollution at specific sites, it has to be recognised that emissions of pollutants from their sources in the traffic environment greatly depend on such factors as traffic intensity (ADT) and composition (i.e., passenger cars vs. trucks), driving patterns, such as speed and brake use, and the pavement type. For instance, the rate of brake wear is largely dependent on the composition of the brake lining and the mode of driving, in which the brakes are engaged (Thorpe and Harrison, 2008). As synthesised by Loganathan et al. (2013), several studies showed that sediments deposited on road sections with high braking, accelerating and decelerating activities generally contained higher metal concentrations compared to other locations along the road. A literature review by Huber et al. (2016) concluded that roads with high annual ADT belong to the road category with the highest runoff concentrations of metals, because of braking and acceleration activities at traffic signals. The same literature review reported that concentrations in parking lot runoff varied widely depending on the main use of the parking lot (i.e., the frequency of entries and exits, presence of heavy vehicles, etc.). Revitt et al. (2014) reported that a coarser surface in car park areas and the pavement abrasion from increased frequency of stopping and starting of vehicles would result in larger sized particles released from the pavement. Increase of speed and traffic density was shown to increase pollutant concentrations in road dust mainly because of higher exhaust emissions and increased road abrasion (Duong and Lee, 2011), and the vehicle speed was found to be more important than traffic density (De Silva et al., 2016). Increased PAH concentrations in runoff were also explained by higher vehicular traffic (Burant et al., 2018). Duong and Lee (2011) reported that metal concentrations in road dust were higher for concrete highway pavements than for asphalt-concrete mixtures, mainly because of higher abrasion in the former case.

In summary, the current state of knowledge clearly shows that the sources related to vehicular transportation are major sources of many of the pollutants usually found in urban stormwater. Depending on traffic intensity (ADT), heavy metals may occur at toxic levels. The major contributors of such a pollution are vehicle exhausts, wear, washing and road abrasion. Thus, vehicle operation is one of the traffic pollution sources depending on the activity volume; hence, this source can be partly controlled by limiting the use of conventional vehicles and distances driven. Substantial reductions in exhaust gases pollution will result from a greater uptake of alternative fuel vehicles and the use of substitute materials reducing or eliminating some traditional sources of pollution. Other activities shown in Fig. 1 may also contribute to increased vehicular transportation intensity – e.g., catchment development in the form of urban sprawl, and industrial (Section 3.3.3) and construction (Section 3.3.4) activities. Finally, road maintenance, particularly in winter months, also contributes to runoff pollution, as discussed in Section 3.3.2.

3.3.2. Road maintenance

In climates with seasonal snow, snow clearance and de-icing practices are used to maintain roads passable and safe. In jurisdictions where the "bare pavement" policy was adopted for winter road maintenance (i.e. keeping roads and highways essentially clear of snow and ice during winter season), large amounts of road salts are applied to meet these policy requirements. In the U.S., it was estimated that 18 million tonnes of road salt were used annually (Corsi et al., 2010); the usage in Canada was 5 million tonnes annually (Government of Canada, 2018), and in Sweden, on average 240,000 t/y were used on state roads between the years 2000–2013 (Swedish EPA, 2013).

Common road salts contain chloride (typically about 60% by weight, in NaCl), anti-clumping agents, ferrocyanide, chromate or phosphate (Ramakrishna and Viraraghavan, 2005), and impurities reaching up to 5% of the total salt weight. Significant environmental effects have been associated with high concentrations of chloride discharged into receiving waters during the periods of snowmelt, as reviewed by Marsalek (2003) and Vignisdottir et al. (2019). Chemical effects of road salts are of two-fold nature, firstly as direct sources of pollutants (e.g., chloride), and secondly as agents increasing the dissolved phase (and effects on biota) of metals (Reinosdotter and Viklander, 2007). Ferrocyanide may form toxic free cyanide, which is removed by volatilization and, therefore, does not represent a significant environmental concern (Ramakrishna and Viraraghavan, 2005; Exall et al., 2011). Salt impurities contain mostly phosphorus, sulphur, nitrogen, copper and zinc. Other salts used in road de-icing include calcium, potassium and magnesium chlorides; those are generally more expensive than NaCl, but can be used effectively at lower temperatures. Urea, which is used mainly in aircraft and airport de-icing operations, and exceptionally was used in highway de-icing (Ramakrishna and Viraraghavan, 2005), is a potent source of nitrogen to runoff.

Common snow management practices also include application of traction agents (sand and grit) on pavements, streets, roads, highways and parking lots, and such materials may accumulate during the winter periods (Westerlund and Viklander, 2006) and remain in road gutters or roadside areas or gully pots, or become resuspended during snowmelt periods. The application of sand and grit is the main source of the high concentrations of TSS or coarser sediments often found in snowmelt-induced runoff (Westerlund et al., 2003; Galfi et al., 2016). Such solids serve as a carrier of pollutants, e.g. metals and PAHs (Stone and Marsalek, 1996; Helmreich et al., 2010; Loganathan et al., 2013; Zhang et al., 2015; Borris et al., 2016; Vijayan et al., 2019b). While small particles often hold the highest concentrations of metals, the highest metal loads were generally found in the largest particle size fraction (Loganathan et al., 2013), and large particles may act as collectors of smaller particles that can be released during runoff events (Borris et al., 2016). Helmreich et al. (2010) attributed heavy metal concentration increases during the snowmelt season to the increased tear and wear of vehicles and pavements due to applications of grit. The mineral composition (affecting e.g., material hardness) as well as particle size distribution of the abrasive materials applied was reported to affect the formation of road dust, and materials with high resistivity to fragmentation were preferred (Räisänen et al., 2003). Similarly, the mineral composition (i.e. natural metal content) of sand, grit and gravel materials may affect their release of metals. For instance, in some cases, sand used for anti-skid purposes was shown to contain high concentrations of phosphorus and several metals (Oberts, 1986). Furthermore, snow clearance from roads, parking lots and paved surfaces can increase abrasion of such surfaces and, thus, the occurrence of asphalt wear particles in runoff (Vijayan et al., 2019a).

Washing of road structures, e.g., traffic tunnels (Meland et al., 2010), or washoff of safety barriers or traffic signs by rain (Mayer et al., 2011), may also contribute to road runoff pollution. Depending on whether the wash water is collected and treated, it may be a source of pollution of stormwater. The washing process leads to removal of exhaust particles attached to tunnel walls and their entrainment by water. Meland et al. (2010) studied wash water from a traffic tunnel; such water was presettled in a small settler prior to discharge to a small stream. The settler effluent contained high concentrations of metals and PAHs, mostly associated with solids, and the estimated loadings from one wash event were 13 g Cu, 2.4 g Pb, 245 g Zn and 0.8 g of 16 PAHs. Wash waters may also contain detergents used in the washing process (Meland et al., 2010), which can increase their pH (Hallberg et al., 2014). The pollutants emitted in road tunnels showed tendencies to accumulate outside the tunnel and, thus, increase pollutant concentrations in runoff from road sections receiving wash water from road tunnels (Barbosa et al., 2007).

In summary, road maintenance in cool temperate climate involves applications of large quantities of road salt and grit during winter months, to ensure road safety. Both materials can be harmful in the receiving waters, by interfering with water quality processes, and in the case of salts, by causing toxicity. Future use of road salts may decline for several reasons: increasing awareness of environmental impacts of chloride, increasing occurrence of mild winters attributed to climate change, and advances in road salt management, including “smart” road salting and the development and increased usage of substitute de-icers (Stone and Marsalek, 2011). Such advances are somewhat offset by the lack of control over salt use on private or privately operated properties (e.g., parking lots) and the population growth increasing demands on road snow management (Chapra et al., 2009). Another maintenance activity contributing to runoff pollution by trace metals and PAHs (typical for road runoff) is the washing of traffic tunnels.

3.3.3. Industrial activities

Industrial activities are recognised as sources of pollutants (Duke and Chung, 1995), which may enter stormwater in two ways (Ellis, 1986): Via pollutant emissions into the atmosphere and subsequent wet and dry atmospheric deposition described earlier in Section 3.1, or by a direct surface runoff from industrial land. The contributions of pollution from industries to stormwater evidently depend on the types of industrial activities, and will change as such activities change with market demands and introduction of new technologies for both industrial production and pollution control. Some generic features of industrial sites may be common for most sites and include: vehicle operation, washing and storage (discussed earlier in Section 3.3.1); storage and handling of products and materials used in the industrial production; waste management, including pre-treatment of wastewaters; operation of equipment outdoors (Duke and Chung, 1995); and, operation of the on-site stormwater management system with quality control measures. Vehicular transportation activities at industrial sites cover a broad range of vehicles from passenger cars to heavy industrial trucks and equipment, with associate consequences for pollution emissions. Examples of industrial sources of pollutants entering stormwater follow, starting with conventional pollutants and followed by micropollutants.

Pitt et al. (1995) investigated sources of stormwater pollutants and concluded that in industrial and commercial areas the likely conventional pollutant hotspots were vehicle service areas, and parking and storage facilities. The industrial sites included in the U.S. NSQD produced some of the highest concentrations of Zn (median 199 μg/L), among the various land use categories (Pitt et al., 2004). Brown and Peake (2006) measured runoff quality from a catchment with combined residential, commercial and light industrial land use in Australia, and attributed the elevated levels of Pb and Cu to the industrial activities in the area. This finding was confirmed by the results of Tiefenthaler et al. (2008) and Liu et al. (2018a), who found that industrial and commercial areas produced higher metal concentrations in stormwater runoff, compared to other land uses, e.g., residential areas, especially with respect to Zn, Cu and Pb.

Gasperi et al. (2014) measured micropollutant concentrations in three French urban catchments of different land use, with the industrial catchment producing particularly interesting results. The catchment produced runoff with the highest concentrations of Cr and Ni (mean concentrations of 6 μg/L Cr and 7 μg/L Ni, respectively), most likely because of related industrial activities, but somewhat unexpectedly, the catchment generated the lowest PAH concentrations in runoff, possibly because of a low traffic density within the catchment. Further studies were conducted in this catchment, because it was considered representative of industrial catchments found in developed countries (Wiest et al., 2018). Contrarily to the Gasperi et al. (2014) results, a subsequent stormwater quality study in this catchment found that it had the highest concentration of PAHs, e.g., anthracene, with the event mean concentration (EMC) as high as 993 ng/L (Becouze-Lareure et al., 2019). Sediment samples from a detention basin in the catchment revealed the presence of NPs and NPEOs, which was consistent with the ubiquitous use of such chemicals in several industrial operations within the catchment. BPA was also present in the sediment samples, and was likely derived from the construction materials and automotive coatings, or from a paper recycling industrial facility also present in the catchment (Wiest et al., 2018). Moreover, Rule et al. (2006) found higher concentrations of

NPEOs in industrial developments (average concentration of 170 μg/L from one rain event) compared to housing developments (generally <20 μg/L), possibly because of the presence of such chemicals in commercial cleaning products, such as car wash detergents. Similarly, Xiao et al. (2012) found industrial activities to be an important source of perfluorooctane sulfonate (PFOS) released from local industrial sources, and Liu et al. (2018b) reported that industrial land use produced higher BTEX pollutant loads in stormwater.

Industrial activities such as production of microplastic pellets used in personal care products, plastic media used in abrasive blasting, plastic pellet production and various manufacturing processes, e.g. rubber, chemicals, paints and varnishes manufacturing represent potential sources of microplastics in stormwater from industrial sites (Cole et al., 2011; Sundt et al., 2014; Magnusson et al., 2016; Horton et al., 2017a). However, stormwater was not confirmed as the main transport vector of the microplastics from industrial activities, even though the stormwater detention ponds serving industrial and commercial areas produced the highest concentrations of microplastics (Liu et al., 2019).

In summary, industrial activities contribute both conventional pollutants and micropollutants to stormwater runoff. While the former group is connected with sources common to most industrial activities, the latter group of greater importance reflects the substances used in specific industrial operations. Industrial activities also contribute to increased traffic, which was discussed in Section 3.3.1.

3.3.4. Construction activities

Construction sites and related activities are known to contribute great amounts of sediments and soil particles to surface runoff and ultimately to the receiving waters. During catchment development, soil erosion is intensified in urbanising areas for two reasons: the stripping of natural protective vegetation cover of soils during construction, and increased runoff flows contributing to sheet erosion and scouring and transport of sediment in unlined channels (Marsalek et al., 2008). The issue of stormwater entrainment and transport of suspended solids and coarser sediments was first time identified in the literature more than fifty years ago (Wolman and Schick, 1967; Leopold, 1968). Wolman and Schick (1967) observed in Maryland (USA) that annual sediment yields from natural catchments could be as low as 100 t/km^2/y, but increased >100 times during urbanisation and catchment development. Marsalek (1992) pointed out that these high rates of sediment export represented a transitional state, because after completion of the urban development and establishment of the soil surface cover, those high yields would drop down to the predevelopment level, or even lower. In spite of advances in soil erosion protection during construction activities, in recent literature, TSS or turbidity still are among the most commonly documented parameters in runoff from construction sites (Wang et al., 2013; Murphy et al., 2014; Sillanpää and Koivusalo, 2015; Shen et al., 2018; Sajjad et al., 2019). Shaheen (1975) reported that a roadway across the street from a construction site received 14 times the expected amounts of deposited sediment based on contribution of traffic alone. TSS concentrations of 200–1200 mg/L were reported in stormwater runoff from construction sites (Center for Watershed Protection, 2003). Moreover, Sajjad et al. (2019) measured higher mean concentrations of TSS during the active construction phase, including excavation works (annual average EMCs of 1175 and 748 mg/L TSS, respectively, for two years of active construction), compared to post-construction phase, when the annual average EMCs for the following two years were around 600 mg/L TSS. Besides the physical impacts of stormwater sediment loads (e.g., silt blanketing of spawning beds in streams (Shaver et al., 2007)), stormwater sediment also carries adsorbed pollutants, e.g., metals and PAHs (Stone and Marsalek, 1996; Loganathan et al., 2013; Zhang et al., 2015; Borris et al., 2016), and thereby contributes to the pollution in the receiving waters. Ellis and Mitchell (2006) identified the construction industry to be a significant source of sediments, a documented source of nitrogen, and a possible source of metals, based on a survey of data in the UK literature. However, it was not stated what specific sources or activities within the catchments with ongoing construction activities were producing the sediments, nitrogen and metals.

Construction activities were shown to have profound impacts on water quality in a Finnish developing catchment (Sillanpää and Koivusalo, 2015), and earth moving works, paving, house construction and temporary wastewater discharges were among the most important factors explaining the variations in water quality. Construction work is often associated with the use of heavy vehicles and equipment, and materials that can potentially contribute various pollutants to the surroundings. Therefore, increased levels of pollution associated with vehicular transportation (Section 3.3.1), littering and oil spills (Section 3.3.5), and building materials (Section 3.2.2) may apply to construction sites as well. US EPA (2005) identified the fertiliser use (an important source of N and P), spills or littering, solid and sanitary wastes and debris, application of pesticides and other construction chemicals, and washout from concrete trucks as other important sources of pollution in construction areas. In demolition of old buildings, façade plaster was suggested to be a potential source of PCBs (Andersson et al., 2004). Construction and demolition work may also disperse microplastics to the surroundings, through the use of plastic building materials such as Polyvinyl chloride (PVC) and Polyethylene (PE), or through construction or maintenance work (e.g. sawing and drilling) using plastic materials (Magnusson et al., 2016). Expanded polystyrene (EPS) foam, used for insulation of e.g., pipes, roofs and walls, and house basements, was reported to break easily into smaller fragments during manipulation, and those can be transported by wind because of their low density (Magnusson et al., 2016). Such material properties may also increase the likelihood of transportation by stormwater.

In summary, construction activities represent a major source of stormwater solids generated by soil erosion and impacting on the receiving waters; a frequently reported source of nutrients (P and N), trace metals and PAHs attached to solids; and, episodically documented source of pesticides, construction chemicals, PCBs, and microplastics. In production of stormwater solids, construction activities are clearly the strongest source listed in Fig. 1, but little is known about the magnitude of contribution of other pollutants, including nutrients and trace metals.

3.3.5. Littering, illicit dumping and household waste disposal

Intentional and unintentional littering results in litter deposition both on catchment surfaces, where litter can be dislodged and transported with runoff, or directly into stormwater inlets and gully pots. Shaheen (1975) reported variations in seasonal roadway depositions of litter, unrelated to traffic, varying from 14.3 kg/km/day in winter to 24.8 kg/km/day in summer; such litter was defined as particles or objects >3.4 mm. However, Shaheen stated that "*it has already been determined that such litter is of minimal importance as a water pollutant*". Contrarily, recent findings point to the importance of fragmentation and degradation of e.g., plastic litter (bottles, bags used for collecting pet droppings, and others) as a source of microplastics to the marine environment (Lambert and Wagner, 2016; Magnusson et al., 2016). Plastic litter may also contain and carry adsorbed trace metals and organic pollutants that may be released to the surrounding environment and runoff (Nakashima et al., 2012; Hahladakis et al., 2018). As reasoned by e.g. Horton et al. (2017a) and Hahladakis et al. (2018), common plastic additives that were found to be released from plastic litter include many trace organic pollutants, such as phthalates, BPA and polybrominated diphenyl ethers (PBDEs), and metals, that are often used as or in colourings. Moreover, littering by disposing cigarette butts in the urban environment was considered a relevant threat to urban water quality, considering their rapid release of nicotine (Roder Green et al., 2014), as well as nano-scale particles and several metals (Chevalier et al., 2018). Littering may also attract rodents and other animals, which may contribute nutrients and faecal microorganisms to stormwater runoff, as further described in Section 3.3.7.

Illicit or accidental disposals and spills of wastes or chemicals from e.g. households, industries and other sources, may deteriorate stormwater quality, and can evidently be a source of various substances. Accidental spills occur on catchment surfaces draining into storm sewers, intentional disposal usually targets the sewer inlets (e.g., disposal of used engine oil, old paint) and can occur both in wet and dry weather. Accidental spills are likely to occur at industrial, commercial or vehicular transportation sites and are a significant source of illicit discharges to the stormwater sewers (Brown et al., 2004). A common example of dumping of liquids into the storm sewers is the cleaning of deep fryers in the parking lots of fast food operations (Brown et al., 2004). Furthermore, household cleaners and motor fluids or lubricants are also occasionally spilled or discarded into storm sewers (Butler et al., 2018). Dumping or spills of motor vehicle fluids may contribute hydrocarbons, phthalates, BPA, oil and grease, metals as well as other pollutants to storm sewers (Brown et al., 2004; Markiewicz et al., 2017). Moreover, personal communication with staff of Swedish municipalities revealed that illegal dumping of drilling residues from installations of downhole heat exchangers into storm sewers has occurred and thus contributed to increased TSS concentrations in stormwater. Other activities reported to produce improper discharges to storm sewers include draining of swimming pools involving discharges of chlorinated water into the sewers, and car washing in driveways, contributing sediments, nutrients and other pollutants to the storm sewers (Brown et al., 2004). Car washing was described with more detail in another section (3.3.1). A literature search revealed the lack of data on, and reporting of, such incidents, with practically no data on the measured effects on stormwater quality. Butler et al. (2018) estimated that domestic sources of chemical pollutants are usually minor compared to industrial spills or illicit toxic waste disposal.

In summary, the probabilistic nature of littering and illicit dumping and household waste disposal contribute to the unpredictability of these sources and their contributions to the pollution of stormwater. Recent identification of microplastics as pollutants of the marine environment exacerbates the concerns about the sources in this group, which are known to contribute plastics to urban runoff.

3.3.6. Gardening

Relatively few studies have been published on the topic of gardening activities as sources of stormwater pollution, perhaps because very little to no runoff is expected to occur from garden plots. In heavy thunderstorms, or when the ground is already saturated, runoff from pervious surfaces may occur and, thus, contribute suspended solids (Gromaire et al., 2001), or nutrients originating from e.g., fertilisers applied to grassed turfs (Malmqvist, 1983) to stormwater. In a recent study of nutrient budgets of urban catchments of various land use in the U.S., lawn fertilisers were among the dominating inputs of N and P (Hobbie et al., 2017). Gardening activities may also generate garden wastes, such as grass clippings or leaves, which, if deposited on nearby impervious surfaces, are a source of biodegradable organics, N and P to runoff (Dorney, 1986; Selbig, 2016). Pesticides used in gardening to control insects, weeds, and fungi can also potentially influence stormwater runoff quality (Bollmann et al., 2014). Wittmer et al. (2010) found elevated concentrations of the pesticide Mecoprop during the period May – September (up to 32 μg/L) compared to the rest of the year (below 0.1 μg/L), and attributed applications of this pesticide on lawns and in gardens as a likely source. Botta et al. (2012) reported frequent detections of 18 pesticides applied in the urban study area during the period of most frequent applications (May–July). Gardening activities are often closely connected to those concerning parks and recreational areas, which were described earlier in Section 3.2.3.

In summary, relatively little is known about gardening as a source of pollutants to urban runoff. This is probably caused by the fact that garden properties are highly pervious and may contribute little runoff. One can assume that gardening contributes solids originating from soil erosion of cultivated soils, and nutrients from over-fertilised gardens or lawns. In general, this source is closely connected with parks, lawns and recreational areas discussed in Section 3.2.3.

3.3.7. Pets and wildlife

Droppings from pets and birds are confirmed sources of nutrients and faecal microorganisms in urban stormwater. Concerning the former source, Hobbie et al. (2017) estimated household pet waste (particularly dog waste) as one of the top inputs of N and P to urban watersheds in St. Paul (Minnesota). In the case of N, pet waste was typically the third largest source (after residential fertilisers and atmospheric deposition), and with respect to P inputs, pet waste was the greatest source. Malmqvist (1983) estimated that a dog could contribute 700 g N and 90 g P per year to runoff, and a corresponding estimate for one bird in a catchment was 40–400 g N and 3–25 g P per year. These estimates were based on several assumptions, including the number of animals in the catchment and the probability of the droppings deposited on impermeable surfaces. Considering the large populations of pet dogs and cats worldwide (e.g., 90 and 94 million in the U.S. in 2017, respectively; https//en.wikipedia.org/wiki/Dogs_in_the_United_States, accessed Aug. 1, 2019), pet waste potentially represents a huge source of P, N, and faecal microorganisms. It is generally assumed that N comes from animal urine and P from faeces. This implies that the pet contribution of P could be controlled by pet owners picking up the droppings, while pet contribution of N is more difficult to control. Little has been published on dog owner practices of waste pickup; Hobbie et al. (2017) estimated that 40% of dog waste was left in the landscape.

Moreover, animal droppings are a source of faecal bacteria that can infect humans. Contamination of the urban environment by dog faeces is particularly of concern, because such faeces may harbour pathogenic bacteria (e.g., *Campylobacter, Salmonella* and *E. coli*) and parasites (e.g., *Giardia* and *Cryptosporidium*) threatening human health, and antibiotic resistant bacteria (e.g., *vancomycin-resistant enterococci* and *methicillin-resistant Staphylococcus aureus*) (Cinquepalmi et al., 2013). Faecal indicator bacteria (FIB), mostly *E. coli* and *enterococci*, are commonly used to assess the risk of microbiological contamination of urban stormwater and receiving waters (Galfi et al., 2016). Paule-Mercado et al. (2016) found that among agricultural, mixed land use and urban catchments, urban stormwater had the highest concentrations of faecal contaminants and domestic animal wastes were one of the sources. However, sanitary sewer contributions through e.g. misconnections (Section 3.4.2) or sewer overflows were probably the main source. O'Keefe et al. (2005) reported that droppings from dogs and birds were among the main sources of the FIBs found in their study catchment in Edinburgh. These FIBs sources were likely associated with a pigeon roost below a railway bridge and a guard dog kennel. Equestrian facilities in recreational areas were also shown to be a source of FIBs to stormwater (Tiefenthaler et al., 2011). Furthermore, microbial source tracking methods using molecular markers associated with specific sources make it possible to identify specific sources of bacteria. This was demonstrated by Steele et al. (2018), who detected animal faeces in stormwater, primarily associated with an avian marker (birds), but also associated with a canine marker. Microbial source tracking on urban beaches in Hamilton (Edge and Hill, 2007) and Toronto (Edge et al., 2007; Edge et al., 2010) indicated that in shallow water, the dominant FIB sources were bird droppings (stored in wet beach sand), rather than dog faeces or municipal wastewater in combined sewer overflows. Baral et al. (2018) found that stormwater runoff provided the main input of microorganisms to an urban creek during wet-weather, and that 75% of the microorganisms in stormwater was from materials washed off impermeable surfaces in the watershed. Pigeon faeces were predicted to account for a median of 14 to 21% of the microorganisms found in street sweepings, which were assumed to correspond to the materials washed off impermeable surfaces (Baral et al., 2018). Moreover, pharmaceuticals and personal care products (PPCPs) were detected in stormwater canals in New Orleans, USA, (Boyd et al., 2004) and even though the main source was wastewater contamination of stormwater, a potential source would also be veterinary pharmaceuticals found in pet waste.

In summary, pet waste represents an important source of N, P and FIBs in urban stormwater. Dog droppings are of particular concern, because of a potentially large mass of dog waste in urban areas, the presence of pathogenic bacteria and parasites threatening humans, occurrence of antibiotic resistant bacteria in such a waste, and an uncertain effectiveness of waste pick up by pet owners. Bird droppings are of concern in areas with large bird populations and opportunities for bacteria survival, such as interstitial water on urban beaches. This source is likely to be connected with those concerning urban green areas, which are likely to be frequented by pets and wildlife (Section 3.2.3).

3.3.8. Washing of buildings and structure surfaces

Building roofs and facades, and other structure surfaces, found in the urban environment collect deposited pollutants, may suffer from moss or algae fouling, and be subject to applications of protective or biocidal chemicals and materials. Occasionally, such surfaces need to be washed, by little trained individuals or specialised companies, to maintain or enhance their visual appearances. A similar activity of potential stormwater quality concern is the removal of graffiti from buildings. Sundt et al. (2014) reported washing of plastic-treated surfaces (coated or painted) as a source of microplastics and estimated the loss of particles due to surface maintenance as 5% of the surface layer. However, this estimate was based on scarce data, because relatively few studies were published on the building surface maintenance (Sundt et al., 2014). Furthermore, pressure washing can be applied to remove flaking paints from e.g. metal roofs. Depending on actions taken to collect and treat the wash water, the maintenance of building and structure envelopes by washing is a potential source of pollution of surface runoff and urban stormwater.

The wash waters contain constituents from atmospheric deposition, particles and dissolved constituents from the actual surface material, and any chemicals previously applied to the surface, e.g., biocides, or detergents used in cleaning. The contribution of pollution from building surface materials and structure surfaces is described in detail in Section 3.2.2. No reports on implications of these cleaning practices for stormwater quality were found in the scientific literature, but one report was found in Swedish media, the first washing in 27 years of the façade of the Stockholm Globe Arena (Lund, 2016). The 22,000 m^2 façade was pressure washed using hot water and concerns about the stormwater quality were addressed by treating the wash water, containing algae as well as metals and other pollutants originating from car and airplane emissions, by using filters inserted in road gullies. The washing of road tunnels as a source of stormwater pollution was described under the Road maintenance Section 3.3.2, because the associated pollutants are mainly derived from vehicular transportation and related activities.

In summary, there is little information available on generation of pollutants during the washing of buildings. The processes involved in this type of maintenance are similar to those described in Section 3.2.2 for Building surfaces and structures, with three distinctions: (i) the washing is likely to be more mechanically intensive (using e.g., pressure washers or other tools), (ii) wash detergents or chemicals may be used, but (iii) wash waters may be captured, contained and treated before disposal.

3.4. Urban drainage systems

The contemporary drainage systems represent complex systems of man-made and natural elements serving to convey stormwater runoff, improve its quality in green infrastructure (GI) and stormwater control measures (SCM), serve for beneficial uses of stormwater, and protect the receiving waters. In such systems, there are many opportunities for stormwater to elute or entrain, or reject various pollutants, as discussed in this section focusing on the underlying processes. The sources discussed are classified as Green infrastructure and stormwater control measures (SCMs), Sewer pipe materials, and Misconnections. GI and SCMs are considered as a part of the urban drainage systems, as they are specifically designed to collect and convey urban runoff, and often their effluents are connected to the storm sewers.

3.4.1. Green infrastructure and stormwater control measures (SCMs)

In general, GI and SCMs are designed to reduce runoff volumes and peak flows, and improve stormwater quality, and it is, therefore, somewhat counter-intuitive to discuss them in the context of sources of stormwater pollution. However, for such reasons as conflicting demands on these measures (e.g., stormwater treatment vs. aesthetical and recreational amenity features), outdated design, lack of maintenance, or lack of communications in the planning stage, the control measures may lead to deterioration of some aspects of stormwater quality within such systems. Examples of such cases, where GI and SCM facilities contributed to the pollution of stormwater, are relatively common, and some are listed below.

Permeable pavements were reported to increase significantly TSS concentrations in runoff due to the washout of sediments from the subgrade (Winston et al., 2016). Furthermore, such sediments may carry particle-bound metals, and possibly other substances as well. Concrete porous pavements and pervious concrete were shown to increase runoff pH and alkalinity (Kuang and Sansalone, 2011; Pilon et al., 2019), and sulphate concentrations (Pilon et al., 2019), though not necessarily to harmful levels. Green infrastructure, employing vegetated pervious surfaces, may leach out environmentally harmful chemicals into stormwater. Kondo et al. (2016) studied elemental concentrations in GI soils in Philadelphia (USA) and observed that even though the three elements of concern to human health, Cd, Hg, and Pb, were either no different or lower in GI soils, compared to non-GI soils, those concentrations were up to four times higher than soil clean-up objectives for residential use.

Green roofs may act as a source of nutrients, depending on enrichment of nutrients in the soils used and fertilising routines (Czemiel Berndtsson, 2010). If pesticides are applied to green infrastructure, to control insects, weeds, or fungi, they may also be present in the effluent water from such systems. At an experimental green roof site in Italy, Gnecco et al. (2013) found that green roofs could act as a source of solids, COD and some metals, and that the leaching of substances was greatly dependent on substrate characteristics. The characteristics of facility construction materials are of high importance also in other SCMs. For instance, Flanagan et al. (2019) showed that various pollutants, including phthalates, APs and BPA, were released from construction materials used in stormwater biofilters, such as a drain filter fabric, drain made of plastic, geomembrane, and asphalt. Similar conclusions were drawn by Gromaire et al. (2014) regarding the release of APs and BPA from materials used in green roof structures. In all SCM facilities with free water surface or conveyance materials exposed to solar radiation, the temperature of stormwater rises (Van Buren et al., 2000), and this temperature increase contributes to the thermal pollution of receiving waters, causing potential harm to cold water fisheries and other biota.

Primarily, SCMs or GI facilities may be acting as “secondary sources” of pollution, i.e. pollutants originally derived from other sources are released to effluent waters due to such processes as resuspension or leaching. Infiltration based systems bring up concerns about deterioration of soil quality (Kondo et al., 2016; Tedoldi et al., 2017) due to accumulation of road sediments and contamination from bypassing runoff, and such systems may therefore leach pollutants to the effluent water (Flanagan et al., 2019). The presence of road salts in the runoff water may mobilise metals and increase TSS levels in effluent waters (Winston et al., 2016; Søberg et al., 2017; Flanagan et al., 2019). In stormwater ponds, or other treatment facilities designed to trap stormwater sediments, there is a risk of sediment resuspension or releases of mobile elements (e.g. metals) in the case of high flows or changes in the ambient chemistry (Marsalek and Marsalek, 1997;

Marsalek et al., 2006; Wiest et al., 2018). Such changes can be induced by e.g., influx of chloride-laden snowmelt (Vijayan et al., 2019b). Chemical characteristics of sediments from six types of SCMs (ponds, constructed wetlands, an infiltration basin, biofilter, stormwater treatment clarifier, and oil grit separators) were assessed by Marsalek et al. (2006) against the Ontario Sediment Quality Guidelines and 80–100% of collected samples were found marginally to intermediately polluted by heavy metals. Severely polluted sediments were found for Cr, Cu, Mn, and Zn at several facilities, and sediment toxicity was confirmed at several sites (Marsalek et al., 2006).

In summary, in modern drainage systems, with SCM or GI facilities, attention needs to be paid to pollutant pathways and their ultimate fate. In most stormwater control facilities, pollutant “removal” is achieved by immobilisation and storage, with associated risks of subsequent releases. Hence, the facilities need to be designed with sufficient storage (e.g., of polluted sediment), be well and timely maintained and the removed pollutants need to be properly disposed at special sites.

3.4.2. Misconnections

The term misconnections used here includes both intentional and unintentional cross-connections of wastewater and stormwater sewers, and includes entry of household as well as industrial or commercial wastewaters (i.e. wash water, process water or other inappropriate flow) into storm sewers. Another closely related term found in the U.S. literature is “illicit discharges” defined as any direct or indirect non-stormwater discharges to the stormwater conveyance systems. Cross-connections with municipal sewage can occur where combined sewer systems were converted to separate sewers, while old industrial or commercial areas are more likely to have cross-connections with industrial or commercial wastewaters (Brown et al., 2004) and thus contribute with different types of constituents to stormwater. Illicit household discharges to storm sewer systems were described by Revitt and Ellis (2016) as a *“ubiquitous problem for urban receiving water quality”*. They reported that the biodegradable organic and nutrient loads from misconnections were likely to require dilution of up to 100:1 to conform to ecological criteria and that, in the UK, toilets, kitchen sinks and washing machines pose the main problems (Revitt and Ellis, 2016).

The best indicators of stormwater contamination by wastewater include chemical (e.g. caffeine) or microbiological human waste markers (Panasiuk et al., 2015). However, as reported by Irvine et al. (2011) because of wide ranges of concentration of many substances found in stormwater, it is difficult to find effective indicators for source identification. The detection of PPCPs in stormwater was interpreted as an indication of stormwater contamination by wastewater (Boyd et al., 2004). Misconnections are among the main sources of faecal coliforms found in stormwater sewers (Makepeace et al., 1995). Human source markers and pathogens were found present in stormwater regardless of the storm severity or sampling location, and were therefore assumed to come from a chronic pollution, such as sanitary sewer misconnections (Paule-Mercado et al., 2016; Steele et al., 2018). Considering the high occurrence of misconnected washing machines, misconnections may also be an important source pathway for microplastic fibres from synthetic clothes (Magnusson et al., 2016; Horton et al., 2017b) and household detergents, containing e.g. PO_4-P (Revitt and Ellis, 2016). The occurrence rate of such misconnections reported in the literature and summarised by Ellis and Butler (2015) showed great variation, but a valid average misconnection rate was 3–4% for UK, European and U.S. data, although specific areas with up to 30% misconnection rates were reported.

In summary, the risk of misconnections and stormwater contamination by wastewater depends on the structural integrity of the sewer infrastructure and its proper operation and maintenance. Generally, the presence of wastewater is commonly determined by the associated markers, which may be chemicals (e.g., caffeine) or faecal indicators. Higher risks of misconnections were reported in cities with an older sewer infrastructure.

3.4.3. Pipe materials

The quality and composition of stormwater may change during conveyance by storm sewers to the receiving waters, with both pipe materials and stormwater composition playing important roles. Davies et al. (2010) investigated the impact of PVC and concrete pipes on urban water chemistry by laboratory experiments and found that concrete pipes can affect water quality significantly, mainly by increasing its pH. Wright et al. (2011) also concluded that concrete pipes are responsible for changes in the geochemistry in urban streams, resulting in higher pH compared to non-urban streams. Metal release from different pipe materials in laboratory submersion tests was studied by Ogburn et al. (2013) and they found that concrete, high-density polyethylene (HDPE) and vinyl materials released small or no concentrations of metals and can, therefore, safely be used as stormwater drainage materials. Borris et al. (2017) conducted laboratory tests with conveyance of three types of stormwater by sewer pipes made of concrete, galvanised corrugated steel, and PVC. Their results confirmed the earlier reported increases in pH in concrete pipes, and indicated reductions in stormwater turbidity in concrete and corrugated steel pipes. Flocculated settling of fine solids in relatively new concrete pipes, and solids settling and retention in bottom corrugations might be the causes of such reductions. Furthermore, significant releases of Zn (up to a concentration of 1406 μg/L Zn) from the galvanised corrugated steel pipe were observed. The results also depended on the type of stormwater used: two synthetic stormwaters mimicking the locally observed chemical composition of stormwater and one actual stormwater enhanced by gully pot sediments. Among these stormwater media, the last one was considered as the best with respect to mimicking the actual field conditions.

Among trenchless technologies for pipe repair, sewer relining with synthetic liners is becoming a more common practise and a number of studies addressed the effect of this technology on water quality. For instance, Grella et al. (2016) suggested coating of insides of concrete pipes with epoxy to avoid large pH increases. It has been demonstrated that inorganic and organic contaminants, including carcinogenic and endocrine disrupting compounds, may be released from certain storm sewer coatings into the conveyed waters (Whelton et al., 2013; Tabor et al., 2014; Ra et al., 2018; Li et al., 2019). Ren and Smith (2012) studied the potential release of BPA, di-(2-ethylhexyl) phthalate (DEHP), and benzyl butyl phthalate (BBP) from two common sewer pipe repair procedures in laboratory batch experiments, but did not detect any of the aforementioned three plasticisers in the water exposed to the pipe-repair materials. However, the experimental setup of their experiments do not mimic well the conditions in actual storm sewers, characterised by continual changes of properties of the conveyed stormwater, including changes of temperature, flow rate, and stormwater chemical composition, with respect to, e.g. organic matter content and sediment concentrations, which may potentially affect releases of substances. Furthermore, the analytical method used in their study had relatively high detection limits (0.029, 0.191, and 0.447 mg/L for BPA, DEHP, and BBP, respectively), compared to the environmentally relevant concentrations. For example, the maximum allowable concentration of DEHP in surface waters is 1.3 μg/L, according to the EU directive on priority substances (Directive 2013/39/EU).

In summary, the choice of sewer pipe material (i.e., concrete, plastics, or corrugated steel) may affect the quality of conveyed stormwater. Where such changes would bear adverse consequences for conditions in the receiving waters, pipe materials preventing these changes should be used. Alternately, special coatings (e.g., by epoxy) hold promise in avoiding interactions between the pipe material and stormwater quality.

4. Implications of stormwater pollution source analysis and future outlooks for urban stormwater quality

The knowledge of the specific sources contributing to diffuse pollution through transport vectors such as stormwater is necessary for identifying opportunities for implementation of source controls and other SCMs, and to ensure their effective design and operation. This review summarises the current knowledge of the sources contributing to the pollution of stormwater and, thus, the sources of urban diffuse pollution. Several of the sources described herein were identified as requiring more research. These included anthropogenic activities such as washing of buildings and structure surfaces, and construction activities (concerning other pollutants than TSS) as well as specific elements of urban areas or surfaces, such as non-metallic building surface materials, and gardens, parks and other green areas, among others. In the context of the ongoing climate change and global warming, which may lead to, e.g., longer dry periods and increased rain intensities in many parts of the world, the effects that these factors can exert on runoff quality may increase concentrations of harmful substances in runoff, and may also increase the problems associated with the thermal pollution of stormwater. The major stormwater pollution sources, identified in this review as: vehicular transportation, metallic building envelopes and AD will likely remain major sources due to their widespread presence in the urban environment. Moreover, AD should undoubtedly be considered in stormwater quality studies (Liu et al., 2018a), even in relatively unpolluted areas, because the global AD pollution load due to contributions from highly polluted areas may impact also unpolluted catchments (Omrani et al., 2017).

For stormwater quality concerns, a number of stakeholders need to contribute to achieving sustainable management of stormwater quality. Researchers should continue the work of identifying and quantifying the contributing pollution sources, as well as developing control strategies and treatment technologies, in collaboration with the industry. Future stormwater quality programs should to a larger extent reflect the societal developments and include the pollutants of emerging concern, defined e.g. in the lists of priority pollutants (such as the Directive 2013/39/EU, 2013) or released with the introduction of new products, materials, or chemicals to the urban environment. Synergistic environmental effects of the micropollutants present in urban stormwater are poorly understood (Masoner et al., 2019) and need to be studied further in parallel with collecting the data on their occurrences in stormwater. Furthermore, policy makers should: (i) develop control policies and regulations for reducing the actual entry of harmful pollutants into the environment, including the stormwater, (ii) provide guidance to practitioners implementing such policies, and (iii) establish programs for follow up and evaluation of these pollutant control policies (Marsalek and Viklander, 2011). Past successes of pollutant control policies supporting stormwater quality, including the earlier mentioned phasing lead out of gasoline (Kayhanian, 2012; Huber et al., 2016), or substitution of harmless materials for copper in brake pads, indicate that source controls are much more effective in actually removing diffuse pollutants from the environment than stormwater treatment facilities (Marsalek and Viklander, 2011). Regarding the building surfaces and structures exposed to rainwater or stormwater, material substitutions are effective pollution control measures, when comparing various materials. One exception could be historical buildings, where the original materials need to be retained to preserve the historical appearance. In that case, on-site treatment of polluted building runoff should be implemented. In any case, cooperation and communications among different stakeholder groups, including the public, is necessary to build acceptance for the implementation of innovative acceptable solutions (Barbosa et al., 2012).

5. Conclusions

The knowledge of sources of pollution of urban stormwater and snowmelt is required for selecting effective control options, including source controls, for preventing or mitigating such a pollution and its impacts on the receiving waters. The status of knowledge in this field varies among the source categories, with much of the past research focused on highly influential sources including the atmospheric deposition, vehicular transportation, and metallic building envelopes and structure surfaces. Such sources have been considered as major contributors of urban runoff pollution, and are likely to retain this assessment, because of their central role in urban land use activities involving releases of atmospheric pollutants (contributing to atmospheric deposition, e.g., industries), vehicular transportation, and the use of metallic materials exposed to rainfall.

Concerning the other urban stormwater pollution sources, a number of knowledge gaps were identified: the extent of potential contributions of pollution from washing of buildings and structure surfaces; the contribution of other pollutants than TSS from construction activities; the contribution of pollutants from non-metallic building surface materials (mainly with respect to organic micropollutants other than pesticides); the significance of the pollution contribution from gardens, parks and other green areas, especially in anticipation of future climate changes characterised by increased rainfall depths and intensities in many regions of the world; and, the significance of the faecal pollution caused by urban pets and wildlife.

More attention needs to be paid to the emerging pollutants by conducting systematic studies (reaching beyond opportunistic episodic sampling) reporting the concentrations needed for the environmental risk assessment of emerging pollutants. Such data would serve to broaden the database of stormwater pollutants and guide material substitutions, where needed.

Rapid advancements in clean manufacturing and pollution control technologies, as well as increased awareness concerning the environmental impacts of anthropogenic (behavioural) activities among the public, may further increase the differences in pollutant emissions between historical and recent data. Hence, caution is urged when using historical data in current environmental studies. The tendency of progressing data obsolescence is likely to continue in the future, possibly at even faster rates. Moreover, the continuing introductions of new materials and products, and potentially of new pollutants, into the urban environment suggests that the identification of important stormwater pollution sources, and of the associated pollutants, is a continuing process.

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Declaration of competing interest

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

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