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OVERVIEW

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Merging patterns and processes of diffuse pollution in urban watersheds: A connectivity assessment

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Abstract

Urban diffuse pollution affects water resources as much as its rural counterpart does; however, it is considerably less studied. The full complexity of the urban landscape needs to be addressed to apprehend the diversity of surface layouts and covers, multiple pollution sources, and the diverse changes caused by different types of drainage systems. In this article, crucial *patterns* of pollution source areas are categorized, and current knowledge on their temporal and spatial variations are collated. Urban alterations of transport *processes* that enhance, delay, or inhibit diffuse pollution transport from source areas through the urban watershed are detailed. Current knowledge regarding diffuse pollution patterns and processes is conceptually merged by the simultaneous assessment of urban structural and functional connectivity relevant for pollutant transfer. Applying a more holistic approach is considered a prerequisite for identifying critical source areas of diffuse pollution within complex urban catchments, to minimize the transfer of particular harmful pollutants and to enhance future management of urban waters.

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KEYWORDS

connectivity, urban pollution, sustainable urban drainage design, critical source area

1 | INTRODUCTION

Diffuse pollution of urban water resources remains a serious global environmental problem, despite considerable efforts undertaken in the past. Urban diffuse pollution comprises fluxes of dissolved or particulate pollutants that enter urban water resources through precipitation, infiltration, or runoff processes from streets, yards, roofs, commercial areas, and heavily modified urban soils. Such pollutants have detrimental impacts on the quality of both surface water and groundwater. *Diffuse* pollution must be distinguished from urban *point-source* pollution, where contaminants enter the environment from easily identified sites, such as the outlet of an industrial or sewage treatment plant (Fletcher, Andrieu, & Hamel, 2013). In contrast, diffuse pollution, sometimes also called nonpoint pollution, originates from widespread activities with no definitive discrete source.

In the past, a ubiquitous drainage and sewage system in industrialized cities was believed to be the most practical solution to the pollution problem stemming from urban stormwater and sewage (Chocat et al., 2007). However, the

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deteriorating quality of urban waters has continuously raised concerns (Makepeace, Smith, & Stanley, 1995). At the same time, regulations such as the EU water framework directive demand good ecological status for urban waters. Common pollutants in urban waters include trace organics, heavy metals, nutrients, contaminated sediments, petroleum by-products, pesticides, and pathogens (see, e.g., reviews by Miller and Hutchins (2017) for urban rivers and Jurado, Vazquez-Sune, & Carrer, 2012 or Howard & Gerber, 2018 for groundwater). Some of the toxicants cause lethal and sub-lethal effects on aquatic organisms; O_2 deficit and eutrophication occur frequently due to elevated organic matter (Wenger et al., 2009). Hence, a sustainable management of urban waters has not yet been achieved.

A current research frontier is the analysis of how pollutants are retained within the complex landscapes of cities and released laterally toward rivers and vertically toward groundwater during high-intensity storm events. The identification of source areas and the resulting mobilization of pollutants during storm events from urban surfaces continue to be significant challenges in the research on urban water pollution (Fletcher et al., 2013; Pitt, Bannermann, Clark, & Williamson, 2004a, 2004b; Wang et al., 2017). Addressing this problem is not simple because the urban contribution to diffuse pollution varies widely as a complex function of surface cover and sealing, connection degree and type of drainage systems, soil types and their urban transformations, climate, topography, and management approaches, such as the frequency of street and snow cleaning routines (Duncan, 1995; Göbel, Dierkes, & Coldewey, 2007). The poorly defined and poorly identified spatial layout of pollutant sources in urban landscapes makes the identification and control of diffuse pollution particularly difficult.

Urban diffuse pollution is directly linked to the excessive alterations of the hydrological regime due to urbanization, that is, high levels of impermeable surface areas, altered river systems, and up to 100% of the city area connected to sewage and drainage networks (however, a significant smaller percentage is achieved in most cities of the global south as quantified by Corcoran et al., 2010). Impacts of the implementation of drainage systems are higher runoff and pollutant peaks, less recharge to the groundwater (i.e., much more lateral than vertical water distributions), and decreased low-flow conditions (Cristiano, ten Veldhuis, van de Giesen, 2017; Fletcher et al., 2013). Currently, there is a trend of moving away from traditional, hard engineering, and centralized urban drainage solutions toward a more natural drainage approach to reduce the peakedness of drainage and sewer overflow. This step-change involves an increasing extent of decentralized and natural measures for rainwater management (Golden & Hoghooghi, 2018) known under several names, such as sustainable urban drainage (SUD) measures, nature-based solutions, or green infrastructure (see Fletcher et al., 2015 for a full review of terminologies). This trend results in a paradigm shift from wanting to remove urban water rapidly toward wanting to keep it in the city for as long as possible.

For both centralized and decentralized drainage approaches, we have incomplete knowledge regarding the precise source of the pollutants, if and when they accumulate, and their pathways toward urban rivers or groundwater (Lundy & Wade, 2013). The transition from a traditional to a decentralized urban drainage is ultimately a change in the connectivity of vertical and horizontal water fluxes. This transition relates to a change from artificially increased lateral and heavily reduced vertical water fluxes in the centralized drainage approach to considerably more vertical fluxes down (groundwater) and up (evapotranspiration) in decentralized approaches (Golden & Hoghooghi, 2018). Figure 1 depicts the differences in the main flow directions between the two systems: (a) the centralized systems with mostly

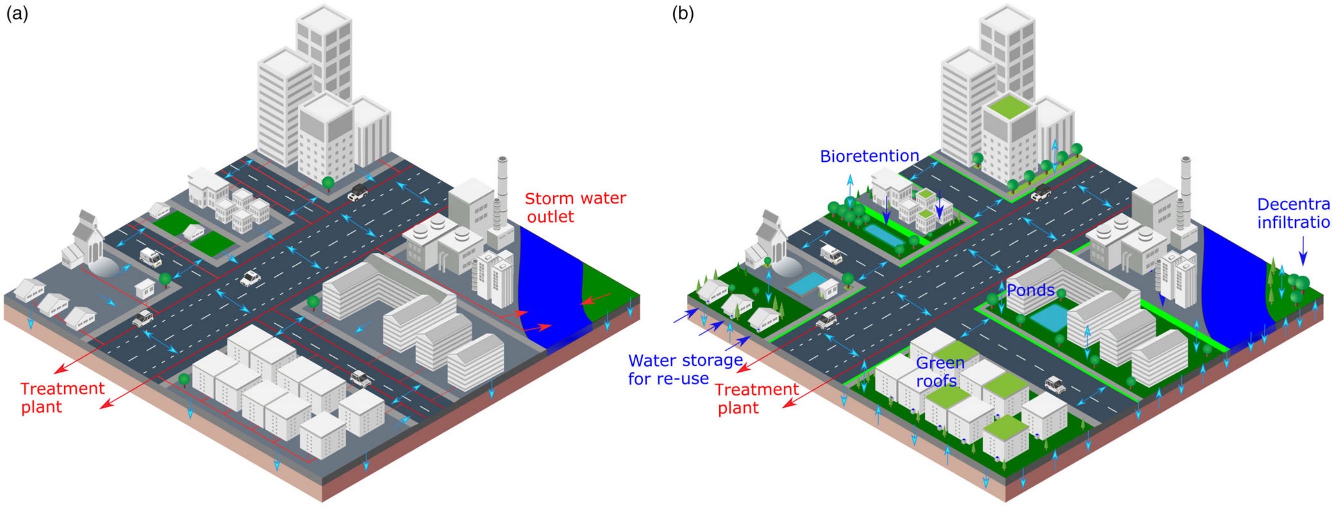


FIGURE 1 (a) Centralized versus (b) decentralized approaches for urban drainage (red line: drainage network, red arrows: storm-water outlet points into rivers, blue arrows: street runoff, infiltration, and evapotranspiration) horizontal flow patterns (blue lines indicate the street runoff, red lines represent the drainage and sewage network, and the red arrows locate flows toward the treatment plant or toward storm-water outlet points (also called combined sewer overflows from which excess waste and drainage water is discharged directly into the rivers during heavy rainfall); (b) the decentralized systems with a significant increase in vertical flow patterns toward groundwater or the atmosphere (blue arrows), retention and storage of water in different storage units, and no storm-water outlet points into the river.

We argue that the handling of diffuse pollution should address the prevalent and changing nature of the connectivity of water and pollutant flows in either drainage system. The concept of hydrological connectivity has proven to be very useful for understanding diffuse pollution in rural catchments (such as studies on nutrient export causing eutrophication, e.g., Heathwaite, Quinn, & Hewett, 2005, Dupas et al., 2015, Gonzalez-Sanchis et al., 2015, Stachelek & Soranno, 2019), but has not yet been applied to urban settings. We use the connectivity definition by Turnbull et al. (2018), who define hydrological and pollutant connectivity as the degree to which a system facilitates the transfer of water and pollutants through itself, through coupling relationships between its components. In this review, we take a holistic perspective on urban diffuse water pollution dynamics to address the full complexity of associated urban connectivity, patterns and processes including urban surface heterogeneity, meteorological, hydrological, and soil variability, drainage systems, and decentralized drainage measures. Process-descriptions of (dis)connectivity of water and pollutant movement throughout urban catchments will facilitate the identification of critical source areas of the cityscape, where significant amounts of pollution that end up in urban waters are generated (Brierley, Fryirs, & Jain, 2006). Consequently, pollution control of the critical source areas is likely to be more cost-effective than attempts to control pollution across the cityscape (Steuer, Selbig, Hornewer, & Prey, 1997).

We presume that only a better understanding of the interlinked dynamics of source areas and pathways will enable better management of urban water resources. In this study, we assemble the current knowledge to achieve this goal. (a) We classify crucial *patterns* of pollution source areas and variables that dominate or influence urban diffuse pollution. (b) We then discuss the urban alterations of transport *processes* that enhance, delay, or inhibit diffuse pollution transport from source areas through the urban watershed. (c) Finally, we examine how we can theoretically merge patterns and processes of urban diffuse pollution within a hydrological and pollutant *connectivity* framework and practically enhance future management of urban waters.

2 | SPATIAL AND TEMPORAL PATTERNS OF DIFFUSE POLLUTION ON URBAN SURFACES

2.1 | Spatial patterns of diffuse pollution

The quantification of diffuse pollution patterns involves two methodologies: an adapted end-of-pipe field approach and the source area sampling.

Adapting an end-of-pipe field approach, diffuse pollution originating from sealed surfaces is frequently quantified in stormwater at sampling points inside the drainage systems, at sewer overflow points, or directly in urban rivers. Recent studies by Eriksson et al. (2007), Pal, He, Jekel, Reinhard, and Gin (2014), and Corada-Fernández et al. (2017), for example, quantified emerging organic contaminants such as surfactants, algal toxins, or priority substances in urban rivers or aquifers. However, these studies do not relate contamination directly to potential source areas. Thus, in these examples, diffuse pollution is sampled applying a method that is more applicable for point source pollution using storm-water outlets as collectors of pollution originating from various sources. Figure 2 illustrates this "simplified" end-of-pipe perspective by showing the efforts of quantifying pollution loads from storm-water outlets or inside water-bodies (highlighted circle) but ignoring the hidden complexity "upstream" of these outlet points (depicted as a gray shaded cityscape).

The source area sampling examines the degree of pollution directly on urban surfaces by quantifying either the potentially available pollution load or pollutant concentration in sheet flow running on these surfaces (Figure 3). The reviews by Duncan (1995), Pitt et al. (2004a, 2004b), and Göbel, Dierkes, and Coldewey (2007) compiled literature on a wide range of different surface types and parameters that showed the highest contribution to diffuse pollution. Sampled source area pollution included heavy metals and organic salts on roads, sidewalks, car parks and different roof surfaces, organic litter from urban green (parks, yards, and street trees) and contaminants due to atmospheric deposition. These studies demonstrate the vast extent of current knowledge on contaminant concentrations and their potential sources.

However, a coherent categorization of all different source types, their intrinsic spatial patterns, and temporal dynamics have not yet been performed consistently. We propose to group urban patterns that dominate or influence

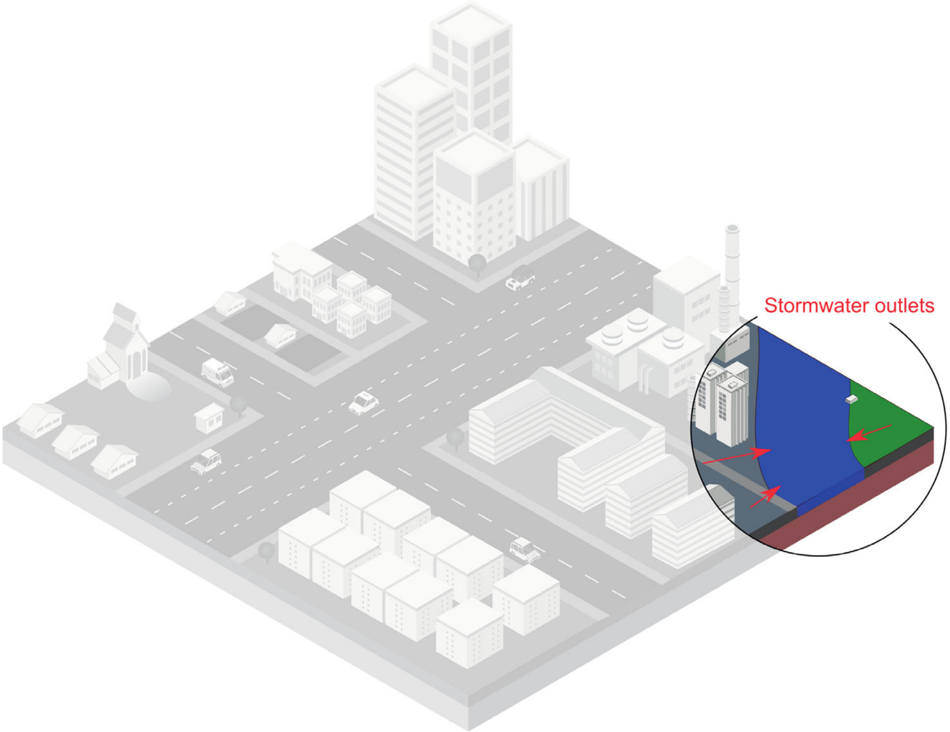
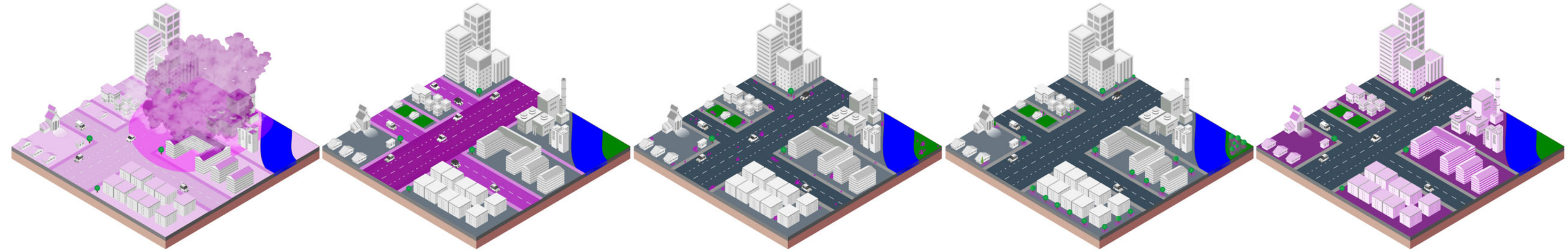


FIGURE 2 End-of-pipe sampling of diffuse pollution at storm-water outlets in urban catchment

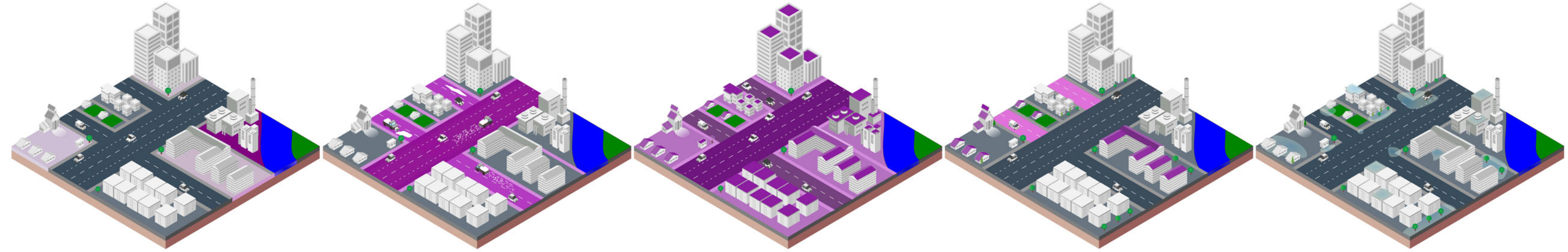
diffuse pollution into four categories: (a) pollution patterns that accumulate or build up on the surface, (b) static pollution patterns (considered relatively stationary from years to decades), (c) management patterns that influence pollution structure, and (d) hydrological response factors that influence the generation of overland flow or infiltration toward groundwater (Figure 3). Current knowledge on pollution types, their spatial compositions, and temporal dynamics of the four categories are discussed in the following paragraphs by identifying existing detailed review studies or gaps in the literature.

Pollution patterns that accumulate

a. *Dry and wet atmospheric deposition* result in the vertical transfer of a wide range of pollutants in dissolved and particulate form from the atmosphere to all urban surfaces (streets, buildings, and plant surfaces). These pollutants include nitrogen, sulfur, and phosphorus deposition (see, e.g., Vet et al., **2014** for a recent global assessment), heavy metals such as Pb, Zn, Cu, Cd, and Cr (Göbel et al., **2007**), Hg (recent studies, e.g., Lynam et al., **2016**), and polycyclic aromatic hydrocarbons (PAHs, e.g., Kim & Young, **2009**). Sources are typically related to major anthropogenic air pollution due to power stations, industries, traffic fumes, and heating.
   Deposition patterns show strong spatial variations within cities with generally more pollution in central and industrial areas and thus depend considerably on city size, structure, climate, and traffic volume (Göbel et al., 2007).
   Temporal variations include seasonal variations due to different annual rainfall distributions and different intensities of air pollution from power stations and heating systems in the winter season (Pitt et al., 2004b). Long-term patterns are detected for sulfur, whose emissions declined significantly in line with reduction policies over the last two decades (Vet et al., 2014). Other long-term variations have resulted from a significant increase in pollution loads in rapidly growing cities over the last several decades, specifically for Hg, as assessed by Wu et al. (2018) for Beijing.
b. *Street pollution due to cars* originate from automobile emissions and inadequate automotive maintenance (Campbell, D'Arcy, Frost, Novotny, & Sansom, 2004; Wada, Takei, Sato, & Tsuno, 2015). Pollutants include airborne heavy metal particulates, such as Pb attributed to emissions from motor vehicle exhausts and heavy metals, PAHs, and microplastics originating from mechanical operation wear, such as road surface abrasion, tire abrasion, and brake pad abrasion (Barjenbruch, 2018; Crabtree, Dempsey, Johnson, & Whitehead, 2008; Göbel et al., 2007; Pitt et al. 2004b; Poudyal, Chochrane, & Bell-Mendoza, 2016). Drip losses lead to local contamination with mineral oil hydrocarbons (Göbel et al., 2007). Tire wear has been identified as a significant source of Zn (Pitt et al., 2004a).



(a) Atmospheric depositions (b) Street pollution due to cars (c) Spot pollution (d) Organic litter (e) Abrasive sources



(f) Background pollution of urban soils (g) Street cleaning, salt application (h) Degree and type of sealing (i) Slope (j) Antecedent moisture patterns

FIGURE 3 Spatial source area patterns that dominate or influence urban diffuse pollution: (a–d) pollution patterns that accumulate, (e–f) static pollution patterns, (g) management routines affecting pollution patterns, and (h–j) hydrologically relevant patterns—(h) degree and type of sealing, (i) antecedent moisture pattern, and (j) slope. Pink shading shows the extent and magnitude of the different pollution patterns (except it depicts the degree of sealing in h) and the slope magnitude in (i). Gray shading in (j) shows the magnitude of soil moisture

The degree of contamination depends primarily on the amount and fraction of the total traffic (Pitt et al., 2004b, Wada et al., 2015), rainfall patterns including dry spell length defining the length of accumulation time (Schiff, Bay, & Greenstein, 2016), and cleaning routines.

Studies on the temporal variations of street pollution have not yet been conducted.

c. *Spot pollution* comprises single-point pollution on a relatively small spatial extent. It may be as diverse as rubbish (e.g., paper cups and packaging, chewing gum), cigarettes (Green, Putschew, & Nehls, 2014), animal droppings on streets and pavements, accidental spills from cars (Duncan, 1995; Poudyal et al., 2016; Revitt, Lundy, Coulon, & Fairley, 2014), petrol stations or building sides (paints, tar, concrete, dust, etc.; Björklund, 2010).

Studies on the spatial and temporal variations of spot pollution have not yet been conducted.

d. *Organic litter* comprises leaves, blooms, pollen, fruits, honeydew, and branches from urban green, mostly from street trees and green facades, and can be found on streets, pavements, and car parks. Compared with artificial litter, the mass of organic litter is significantly higher (Duncan, 1995). Organic litter may be contaminated by dry deposition, retention, and accumulation of particulate air pollutants (heavy metal, sulfur, etc.) on leaves and pollen (see Section 2.1.1a).

Organic litter production is highly seasonal and peaks when trees bloom and shed their leaves (in moderate climates generally in April/May and autumn, respectively). A comprehensive analysis of these seasonal distributions or a spatial assessment is not yet available.

Static pollution patterns

(e) *Abrasive sources* from building surface materials (vertical and horizontal) such as concrete, asphalt/tar shingles, galvanized metals, bitumen-based roofing felt, roofing fabric, plastic/vinyl/fiberglass roofing panels, wood products, paints and (incorporated) additives have the potential to release significant amounts of pollutants into urban runoff (Clark et al., 2008; Clark, Pitt, & Field, 2002; Göbel et al., 2007). The release of pollutants from building surfaces depends strongly on the material, its age, slope, exposure of the surface, and climatic variables such as temperature, pH of precipitation, rainfall energy, and drop size (splash erosion; Burkhardt et al., 2011; Duncan, 1995; Göbel et al., 2007; Müller, Österlund, Nordqvist, Marsalek, & Viklander, 2019). While weathering is the dominant process for metal release of construction materials, additives are mostly leached. Most research on pollutants from building surface materials focuses on metals (e.g., Bürgel et al., 2016; Clark et al., 2008; Pitt et al., 2004b). This research showed that for both pilot-scale field tests and laboratory experiments, traditional galvanized metal roofing contributed to significant concentrations of Cu, Zn, and Pb due to corrosion. Several studies have investigated the leaching amounts of additives, such as fungicides (e.g., Carbendazim), herbicides (e.g., Mecoprop, Isoproturon), and pesticides (Diuron, Terbutryn), which are mostly used in roof and facade paints to prevent undesirable growth (moss, lichen, and algae), and root penetration (Burkhardt et al., 2011; Wicke, Cochrane, & O'Sullivan, 2012). Little is known about other organic micropollutants, such as industrial additives (e.g., nonylphenols and phthalates) that are commonly used in plastic products such as PVC materials (Müller et al., 2019) and are listed as priority substances in Annex 1 of the European Union directive on priority substances (Directive 2013/39/EU, 2013).

The dynamics of pollution from abrasive sources appears to be particularly challenging because no estimates exist on their spatial extent and variations for urban built environments and new building additives with different chemical compositions emerge on a yearly basis (Müller et al., 2019). The study of their seasonal variations is a new research field and some first laboratory experiments indicate higher pollution loads in the summer months, linked to higher radiation rates, which promotes increased leaching of additives (Bollmann et al., 2016).

(f) *Background pollution of urban soils* refers to the extensive presence of pollutants that originate from the historical urban usage and contamination of the ground, which may be washed or leaked out. The pollution in urban soils is manifold and originates from modifications during building construction and demolition, former sewage farms, old waste or debris landfills (including rubber from war damages), and former industrial and brownfield sites. The types of pollution are very diverse and include nutrient leaching (mostly by former sewage farms, Hass, 2012), PAHs, heavy metals, and sulfates (from debris landfills and industrial sites, Mekiffer, 2008, Mekiffer & Wessolek, 2011, Nehls, Rokia, Mekiffer, Schwartz, & Wessolek, 2013), mineral oil hydrocarbons, biocides, dioxins, furans, and PCBs (from former urban industrial and brownfield sites, Bürgel et al., 2016, Wessolek, Kluge, Trinks, & Facklam, 2016).

Spatial information on pollution patterns, such as heavy metal concentrations as well on physical properties und buffer capacities of urban soil is available for cities that foster urban soil data management systems such as the environmental atlas of Berlin (Umweltatlas Berlin, 2021) or the New York City Soil Survey (NYCSS, 2021). Temporal information is normally not readily available.

Management routines affecting pollution patterns

(g) *Street cleaning* clears pavements, streets, gullies, and parking spaces, from litter (organic and others), mostly by manual sweeping or by cleaner trucks equipped with vacuum and sprayers to loosen particles (Calvillo, Williams, & Brooks, 2015; Chang, Chou, Su, & Tseng, 2004). With modern machines, sediment, and pollutant particles down to the size of PM10 are cleaned from street surfaces (Chang et al., 2004). A special form of cleaning is the application of pesticides for weed control on mosaic pavements (Wessolek et al., 2011). Weed control of pavements is considered necessary in many cities as pavements covered with moss can easily get slippery which may affect pedestrian safety. In particular, the herbicide glyphosate was applied in many cities around the world; however, its adverse impacts on the environment put this practice into question and led to a reduction in its usage in some regions (e.g., in Denmark, Kristoffersen, Larsen, Møller, & Hels, 2004).

Cleaning intervals vary widely as a function of traffic magnitude (major streets more frequent than smaller streets), season (more frequent cleaning of leaf fall in autumn), region (daily cleaning in some southern European cities, daily to monthly cleaning, e.g., in Boston (City of Boston, 2018), London (London Borough of Bromley, 2019), and Berlin (Berliner Stadtreinigung, 2018).

(h) *Road salt application for de-icing* on road and pavement surfaces for winter traffic safety is a common practice in many areas in the temperate zone since the 1930s. With sodium chloride (NaCl) or calcium chloride ($CaCl_2$) applied during the winter months (Blomqvist, 1998; Göbel et al., 2007; Ramakrishna & Viraraghavan, 2005).

The spatial distribution and frequency of application vary widely as a function of climatic variables and the traffic safety category of the streets (major streets more frequent). Most cities have detailed management plans detailing under which streets need salt applications (e.g., Berliner Stadtreinigung, 2018). Due to detrimental environmental impacts on street trees and urban waters (e.g., review by Amundsen, Haland, French, Roseth, & Kitterod, 2010), de-icing has recently been prohibited on pavements and most minor streets or is only allowed to be used under extreme conditions in several German and Austrian cities. In comparison, de-icers remain a common method in countries with particularly long and cold winters, such as Russia, Canada, and Scandinavia.

Hydrologically relevant patterns affecting diffuse pollution mobilization

The following three spatial patterns are not pollution patterns per se, but hydrologically relevant surface properties that control if overland flow is generated for the mobilization and transfer of diffuse pollution.

(i) *The degree and type of surface sealing* determine infiltration rates and is one of the most influential factors for runoff generation and pollution mobilization (Shuster, Bonta, Thurston, Warnemuende, & Smith, 2007, Miles & Band, 2015, Lim, 2016, more details on process implications in Section 3.1). The degree of sealing varies between 10 and 25% for less densely built-up areas to 55–85% for inner-city areas (DWA-M 609-1). Sealed surfaces can consist of a single continuous cover (e.g., asphalt, metal, or concrete) or assemblies of individual pavers with joints in between (e.g., cobblestone, sett stones, stone or concrete plates, or grass pavers; Timm, Kluge, & Wessolek, 2018 for a review) with larger infiltration and lower runoff rates for the latter group. The terms degree of sealing and fraction of impervious area cannot be equally set as the latter term suggests nearly no infiltration, whereas the former is characterized by a wide range of different infiltration rates. The type of sealing has a significant influence on runoff generation. New sealing materials such as super porous asphalt and other mixed paver types specifically designed as stormwater remediation methods exhibit considerably larger infiltration rates than conventional asphalt (Timm et al., 2018). Sound spatial information exists for the urban degree of sealing with large resolution data up to 1 m are widely available (see Weng, 2020 for a full review on current techniques), whereas spatial information on the *type* of sealing appears to be much less readily available.

(j) *Antecedent moisture patterns* have been neglected in urban hydrology until recently. The prevailing thought was that urban catchments consist of mostly impervious surfaces where antecedent moisture patterns do not influence rainfall-runoff responses (e.g., Smith, Smith, Baeck, Villarini, & Wright, 2013). However, the modeling study by Hettiarachchi, Wasko, and Sharma (2019) for an urban catchment in Minnesota (USA) showed that antecedent

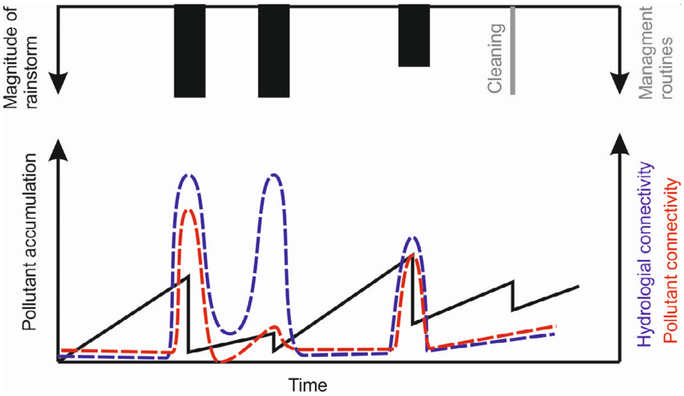


FIGURE 4 Concurrency dynamics of pollutant accumulation and hydrological and pollutant connectivity as a function of rainfall-runoff response and management routines (reprinted with permission from Bracken, Turnbull, Wainwright, and Bogaart (2016)

moisture patterns can have a significant impact on runoff generation, especially where parts of the urban catchment employ decentralized approaches of rainwater management such as enhanced local infiltration with swales. Moisture patterns are highly surface- and time-dependent and an important driver for runoff generation (see more in Section 3.1).

(k) *Slope and surface roughness* determines runoff generation and pollution transport, with steep streets and high-pitched roofs being the main contributors. High-resolution data on slopes are readily available through several remote-sensing products (e.g., QuickBird, 2016; Klemas, 2015) and are frequently used to locate steep streets and traffic areas prone to flooding. Roof-top analysis tools exist to evaluate the spatial distribution of flat and steep roof slopes (Grunwald, Heusinger, & Eber, 2017) thus making them a valuable information source for a detailed slope assessment of the urban landscape.

2.2 | Temporal dynamics of diffuse pollution

The previous sub-sections provided some first indications on the seasonal to decadal dynamics of various pollutants. However, it became apparent that for many pollution sources, the temporal variations are not known. The categorization of the pollutant sources reflects their different temporal dynamics during the year: static pollutants (abrasive sources and background pollution of soils) are thought to not vary significantly across the year. Pollutants originating from management routines (road salt or pesticides for weed control) vary according to a prescribed management schedule following city safety regulations. Pollutants that accumulate (e.g., litter, street pollution, wet and dry deposition) show more complex variations during the year. They are significantly influenced by the length of intermittent dry periods, the magnitude and timing of rainstorm events, and the timing of management routines such as the cleaning of street surfaces.

The importance of timing for accumulated pollution is illustrated in Figure 4. During dry periods, there is a linear increase in pollutant amount, resulting in a build-up over days to weeks followed by a rapid decrease after a large rainstorm event (first black bar) or street cleaning actions (gray bar). However, if a second rainstorm event of a similar magnitude occurs only a short time later (second black bar), pollutant accumulation is insignificant and unlikely to cause any diffuse pollution.

This concurrency dynamics of pollutant accumulation and transport mechanisms (in the form of hydrological and pollutant connectivity) are addressed in the next two sections.

3 | DISTRIBUTION PROCESSES OF DIFFUSE POLLUTION WITHIN URBAN CATCHMENTS

We established that diffuse pollution originates from various sources in the city stemming from a complex city layout. Similarly, complex is the series of pathways through which pollutants can be discharged via the drainage systems into

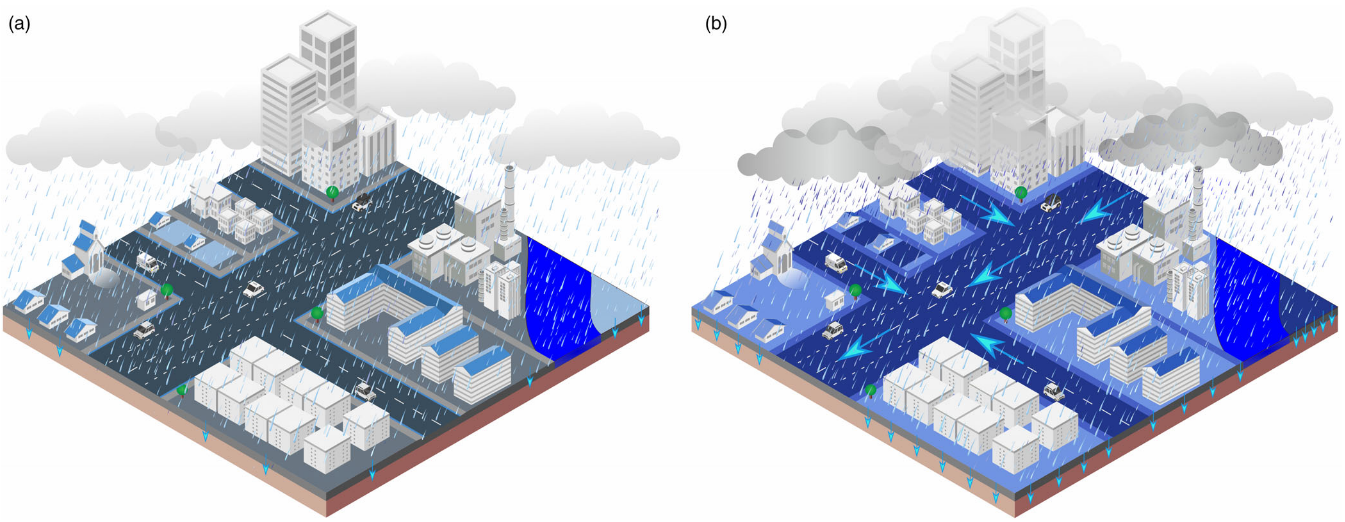


FIGURE 5 Two scenarios illustrating the different spatial extents of urban variable contributing areas for a (a) small and (b) large rainfall event (shades of blue) and associated potential to transport pollutants

the urban river or via runoff into the upper soil layer or groundwater resources in the direct vicinity of source areas. Transport processes are significantly altered in an urban catchment compared to their rural counterparts. The characteristic modifications in their slope distributions, infiltration rates, localized pollution patterns, and completely different setup of flow pathways (or their obstructions) require novel approaches for studying and understanding the interplay of runoff generation and transport mechanisms.

3.1 | Variable hydrologic partitioning in cities

Transport of pollutants occurs either in dissolved form (such as road salt), suspended or semi-suspended (larger components such as litter) or in the particulate form attached to suspended sediments as a function of the infiltration rate, runoff amount and velocity. The urban area does not uniformly generate runoff toward the drainage network or vertical fluxes toward groundwater following rainfall events. As in rural catchments, the size of the contributing area is event-dependent with a nonconstant ratio in rainfall-runoff transformation (Duncan, 1995; Lim, 2016).

Smaller rainfall events result in immediate runoff from heavily sloped and sealed surfaces such as roofs and steep streets; some infiltration into the upper soil layer of unpaved surfaces may occur (Figure 5a, and a smaller peak for hydrological connectivity in Figure 4). However, most urban surfaces do not become hydrologically active during smaller rainfall events (Shuster et al., 2007). In contrast, larger rainfall events with durations exceeding 15–30 min and rainfall heights above a certain threshold result in much larger hydrologically connected areas, including both sealed and unsealed surfaces, such as urban green spaces such as parks, yards, street tree pits, or vacant lots. Extreme rainfall events with magnitudes above a certain threshold (according to Westra et al., 2014 and Guerreiro, Glenis, Dason, & Kilsby, 2017, higher than ~20 mm/hr, but heavily depending on surface conditions and antecedent moisture conditions) are likely to result in urban flash floods. Such an occurrence relates to all urban surfaces contributing to runoff generation, groundwater recharge, or excessive ponding on sealed surfaces (Figure 5b and larger peak for hydrological connectivity in Figure 4). For the last two scenarios, limited amount of empirical data are available to correlate rainfall intensity and duration with the surface condition and runoff generation on the district scale; we identify this fact as a major research gap. A notable exception is the study by Kelleher, Golden, Burkholder, and Shuster (2020), who estimated the extent to which vacant lots in cities modulate hydrologic partitioning under varying rainfall intensities.

In drainage studies, the effective impervious area (EIA) or directly connected impervious area (DCIA) is frequently employed to describe the impervious area fraction of an urban watershed that is hydraulically (as a function of rainfall-response) or physically connected to the storm sewer system, respectively (Ebrahimian, Gulliver, & Wilson, 2016; Hwang, Rhee, & Seo, 2017). A problem with the impervious attribute is that for its quantification, the different types of surface sealing are often not considered (see Section 2.1i for a wide range of sealing types). Miles and Band (2015) and Lim (2016) adopted the variable source area (VSA) concept to urban catchments to include the influence of pervious surface areas as a contributing area of urban runoff generation. However, all indicators have several drawbacks, which limit their use when dealing with diffuse pollution transfer. They only evaluate lateral water fluxes but not the connectedness to the groundwater, and only describe the hydrological connectivity but do not provide information if the connected surfaces are actually polluted and hence contribute to diffuse pollution at all.

3.2 | Critical source area concept for urban diffuse pollution

Identification (and subsequent management) of areas that contribute most of the pollutants to urban water resources are of critical importance. These so-called "critical source areas" after Shore et al. (2014) are usually small in size, particularly heavily polluted (Wang et al., 2017) and located in such a way that pollutant supply and transport coincide in time and space (Heathwaite et al., 2005). Those fractions of the urban surfaces that contribute disproportionately large amounts to or transport pollutants within the drainage network or groundwater are referred to as "effective catchment areas." They are thought to be considerably smaller than quantified by the indicators EIA, DCIA, and VSA.

The concept of critical source areas appears to be more suitable for a system's evaluation of multiple-source diffuse pollution in urban catchments than the impervious area indicators. The concept of the critical source area was previously used for erosion and diffuse pollution assessment in rural catchments to isolate sediment and nutrient transfer from runoff generation (Fryirs, Brierley, Preston, & Kasai, 2007; Fryirs, Brierley, Preston, & Spencer, 2007; Heathwaite et al., 2005). In agricultural catchments, it has allowed a much better way of targeting and managing pollution hazards. Confining mitigation to critical source areas, which tend to be the source of disproportionately large amounts of pollution, was significantly more effective (and costs less) than employing universal controls (Strauss et al., 2007).

Despite its conceptual advantages, the critical source area concept has been applied only in a few studies on urban diffuse pollution. Wang et al. (2017) and Tuomela, Sillanpää, and Koivusalo (2019) applied the concept to identify nutrient, sediment, and pollutant sources in residential areas. Steuer et al. (1997) measured contaminant concentrations from different relatively homogenous urban source surfaces including rooftops, parking lots, and residential lawns of 1–72 ha in an attempt to relate the measured pollutant concentrations at the outlets to the specific surface types. In the study by Björklund (2010), a substance flow analysis was employed in a section of an urban motorway to identify source areas of contaminants (phthalates, with detrimental effects on hormone balance of animals), and scale up the potential amount of phthalate emission from annual stormwater discharge.

A transfer of the critical source area concept to the multiple pollutant patterns across urban surfaces is still pending. A comprehensive analysis of the overlap of pollution supply and transport potential will result in a multi-dimensional matrix of relevant source areas. First, the critical source areas and corresponding effective catchment areas are likely to be very different areas for urban rivers and groundwater. Second, one has to expect very different kinds of critical source areas for the diverse urban pollution patterns, as described in the previous section.

3.3 | Buffers, barriers, and boosters in urban watersheds

The transfer of diffuse pollution is accelerated, delayed, clogged, or inhibited by different elements in an urban watershed, which may function accordingly as pollutant buffers, barriers, and boosters. The terms are borrowed from recent

FIGURE 6 Buffers, barriers, and boosters of urban diffuse pollution: (a) swale directly after rain event, (b) blocked gully, and (c) rain gutter and storm-water outlet

geomorphological and erosion research, where they provide a valuable framework for understanding the various processes involved in matter movement from source areas through a watershed to its outlet (Blanco-Canqui & Lal, 2010; Fryirs, 2013).

Buffers are elements in the urban watershed that prevent or delay diffuse pollution from entering the drainage system. Most of the vegetated areas in cities function as buffers through enhanced infiltration and retention of fine matter (Tedoldi, Chebbo, Pierlot, Kovacs, & Grmaire, 2017). Special forms of buffers are most elements of decentralized urban drainage measures for stormwater control, including infiltration areas, swales, and urban wetlands (Miles & Band, 2015 for a recent review). Indeed, one of the main functions of decentralized drainage measures is to disconnect impervious surfaces such as streets and rooftops from the central drainage system and at the same time enhance infiltration of rainfall (Ebrahimian et al., 2016). In a study by Driscoll et al. (2015), the capacity to capture or leak stormwater and pollutants (nitrogen, phosphorus, and chloride) was quantified for individual bioretention systems. Ahiablame, Engel, and Chaubey (2012) study contains an extensive database of heavy metal and nutrient loads retained by decentralized drainage measures. Another form of retention is given by "accidental" types of buffers as compiled by Palta et al. (2017), which refer to urban wetlands that developed not deliberately on abandoned or low-lying urban areas and become retention areas of water and matter flow.

Barriers disrupt diffuse pollution flow moving along major flow routes in and outside the drainage network and include intentional barriers such as bioretention systems (Ahiablame et al., 2012), stormwater retention ponds, and artificial ponds or unintentional barriers such as blockages in gutters and clogging of gully holes (Figure 6). There are no systematic studies that quantify the timing and location of unintentional barriers for cities, and their effects on pollutant transfer are not clear but are likely to result in the dispersal of pollutants upstream of the barrier.

Boosters are surface elements that enhance the propagation of diffuse pollution and may have diverse boosting functions: (a) rain gutter and gully holes are boosters toward the drainage network, (b) stormwater outlets of combined sewer systems are boosters toward urban surface waters, whereas (c) decentralized drainage measures such as infiltration areas and swales may function as boosters for dissolved pollution flow toward the groundwater. Interestingly, decentralized drainage measures can function both as buffers and as boosters depending on flow directions. Although initially designed as a pollutant buffer toward the drainage network, they concurrently function as a booster that enhances the propagation of water and dissolved pollutants toward groundwater.

4 | MERGING PATTERNS AND PROCESSES OF URBAN DIFFUSE POLLUTION

Diverse, frequently changing pollution patterns and components (Figure 3), nonlinear rainfall-runoff, and transport relationships (Figure 5) determine the magnitude of urban diffuse pollution. We argue here that an end-of-pipe perspective of evaluating pollutant transfer at the outlets of urban catchments (see Section 2) cannot comply with the complexity of a typical urban layout. We see here an analogy to recent efforts of diffuse pollution management in rural catchments. The redistribution of nutrients and sediments could not be understood by simply quantifying nutrient and sediment fluxes at the outlets of agricultural catchments (Bailey et al., 2013; Heathwaite et al., 2005). Whether pollution occurs toward urban rivers or groundwater depends on how well the pollutants are connected to the outlet of the catchment or the groundwater. The previously mentioned critical source area concept, but also the notion of buffers, barriers, and boosters are part of recent advancements in connectivity studies. This science has become a transformative concept in understanding and describing what is considered to be complex systems (Turnbull et al., 2018).

4.1 | Structural and functional connectivity of diffuse pollution in cities

Although widely used in environmental disciplines, the concept of connectivity (as defined in the first chapter) has, to our knowledge, not yet been systematically applied to study transfer processes of water and diffuse pollution in urban systems. However, ecological connectivity research on movements of animals or green bands in urban areas is considerably more advanced, as reviewed by LaPoint, Balkenhok, Hale, Sadler, and van der Ree (2015), but is not further considered in this article.

Approaches to the study of hydrological and pollutant connectivity are frequently divided into the aspects of structural connectivity and functional connectivity (Turnbull, Wainwright, & Brazier, 2008, Wainwright et al., 2011 or

Bracken & Croke, 2007). Both aspects of connectivity were discussed in the previous sections, but not under these names.

Structural connectivity refers to the extent to which urban surfaces are physically linked or connected (e.g., Bracken et al., 2013; Wainwright et al., 2011) and, according to Turnbull et al. (2018), thus derives from the urban system's spatial configuration or “anatomy.” Examples of structural connectivity relate to the physical connectedness of sealed areas channeling water fluxes toward the urban drainage networks, decentralized drainage measures such as swale infrastructure next to streets, increasing infiltration rates into the upper soil layer, and contiguous pollution patterns such as on metal roofs or street surfaces. The effective catchment area, already described in Section 3.2, reflects the degree to which urban catchment pollution is structurally connected laterally and longitudinally (i.e., along major flow lines) toward the drainage network or vertically to the groundwater. Finally, buffer, barrier, and booster may modulate the structural connectivity by intentionally or unintentionally increasing, decreasing, or disrupting it.

Functional connectivity describes dynamic processes operating within structurally connected surfaces and induces the actual transport and fluxes of water and matter between source areas and outlets (Wainwright et al., 2011). According to Bracken and Croke (2007), functional connectivity can be understood to mean both the short-term variations in antecedent conditions and nonlinear rainfall-runoff catchment response (as discussed in Section 3.1 under the name of variable hydrologic partitioning) and the longer-term catchment development, such as the long-term changes of urban infrastructure and the drainage network (as presented in Figure 1 the current move from central to decentral drainage measures). Even the gradual increase in surface sealing through omnipresent infill housing development, which we can see in many cities at the moment, can be assigned to functional connectivity, as it affects hydrological partitioning on a larger time scale.

An urban complex system possesses structural and functional connectivity. According to the systematic review by Turnbull et al. (2018), the structure always affects the function and often (but not always) function affects the structure. Figure 4 underlines this point by visualizing how pollutant connectivity depends on the accumulation state of pollutants on urban surfaces. A large rainfall event might lead to large pollutant connectivity if a particular pollutant had accumulated over some time (peak 1 in Figure 4). However, the same rainfall event might result in much smaller pollutant connectivity otherwise (peak 2 in Figure 4). It is important to point out here that the degree of hydrological connectivity and pollutant connectivity can be very dissimilar (as illustrated in Figure 4), and high hydrological connectivity does not always coincide with high pollutant connectivity. Therefore, it is particularly important to simultaneously quantify and monitor the structural and functional components of an urban system. To understand pollution dynamics over time, it needs to be taken into account that surface runoff and infiltration processes change pollutant patterns, redistribute pollutants horizontally and vertically, and will set new conditions for the coming storm events.

Recent advances in connectivity science include the development of novel monitoring and modeling tools that explicitly consider structural and functional aspects of catchment system. Connectivity monitoring tools are currently being applied to evaluate for example structural changes in topography influencing runoff generation, and pollutant and sediment transfer. Topography changes are evaluated through morphological budgeting (Heckmann & Vericat, 2018), runoff and flow path network analysis for runoff connectivity assessment (Ferreira, Dominic Walsh, de Lourdes Costa, & Alves Coelho, 2016; Masselink et al., 2017), sediment fingerprinting to identify temporal and spatial variability of source areas and transport pathways (Masselink, Temme, Giménez, Casalí, & Keesstra, 2017; Sherriff, Rowan, Jordan, & Uallacháin, 2018), and analysis of hysteresis loops to link hydrological and pollutant connectivity (Keesstra et al., 2019; Lloyd, Freer, Johnes, & Collins, 2016). Monitoring of source areas and pathways was carried out so far mostly for rural settings; urban settings have not received much attention yet (Russell, Vietz, & Fletcher, 2019).

Modeling of pollutant redistribution within urban sub-catchments or in the upper soil zone is currently not implemented in drainage models. Current drainage models, such as the SWMM model (Rossman, 2010), include pollutant build-up, wash-off processes, and reduction in pollutant build-up due to street cleaning operations (see Tu & Smith, 2018, table 1 for a recent review of field and modeling studies on build-up processes of suspended sediment, total nitrogen, and phosphorus). However, neither an implicit nor an explicit representation of connectivity is included in urban catchment models yet. An improved urban model should for example include structural connectivity by incorporating interconnected pollution patterns typical for urban surfaces or specific model algorithms that can reproduce functional connectivity, as suggested by Nunes et al. (2017) for rural catchments.

4.2 | Implications for the management of diffuse pollution in cities

The linchpin of diffuse pollution management is source control, that is, the reduction or avoidance of pollutant accumulation on urban surfaces wherever possible. As a universal source control of complex urban surfaces is not possible, a connectivity assessment of pollutants may help in the identification of critical urban source areas, for which management practices should be prioritized (see Section 3.2 and Wang et al., 2017).

After providing a comprehensive list of the diverse forms of diffuse pollution in Section 2, we used the summary term diffuse pollution in the last two sections. What we do not assess in this study is which of the pollutants are particularly harmful or have toxic impacts on surface or groundwater resources concerning usage capacities, human health, and habitat impact. The development of a hierarchical ranking of priorities (as, e.g., suggested conceptually by Aschonitis et al., 2018) for interventions both spatially explicit (i.e., critical source areas) and requiring pollutants, were beyond the scope of this article. The water framework directive list of priority substances may be used as guidance (Directive 2008/105/EC, 2008, Annex I).

Beyond source control, we argue for a step-change toward the active management of structural and functional connectivity aspects in urban catchments. To enable this process, a process-based understanding of the connectivity of urban systems needs to be established, just as understanding the role of connectivity for diffuse pollution in a rural area, so that "conceptual rather than solely empirical understanding drives how water managers interpret" the urban and specifically the drainage system (Bracken et al., 2013, p. 18). For this purpose, monitoring guidelines for diffuse pollution need to be updated so that not only first-flush concentration measurements at the outlet of sub-catchments (Poudyal et al., 2016, see Section 2) is used for pollution quantification. Monitoring techniques for the structural and functional aspects of connectivity need to be developed for the urban context, such as for the spatial–temporal identification of effective catchment areas of diverse pollutants, runoff and flow path assessments, and redistribution patterns of matter as a function of rainstorm events. Up to this point, connectivity monitoring techniques are only employed in rural catchments (see Section 4.1).

The spatial and temporal interplay of pollutant buffers, barriers, and boosters (Fryirs, 2013) and their distributions in urban catchments (see Section 3.3 for details) require a coordinated assessment. Knowledge regarding their functioning would help urban drainage managers in identifying when and where connectivity patterns need to be maintained or altered, thus enabling or avoiding intentional or unintentional pollutant fluxes to urban water resources, respectively. A multi-scale analysis of the functioning and breaching capacity of buffers and barriers should identify when urban surfaces are strongly disconnected, diffusely connected, or completely connected (Fryirs, 2013). This analysis may guide future re-design of urban drainage strategies, as is already underway with the recent ubiquitous expansion of decentralized SUD measures (Zhao et al., 2018) in cities. Particular care must be taken here so that altering connectivity patterns at one location does not result in detrimental effects in the system (e.g., from buffer to booster: decoupling roof areas from the drainage network to diminish effects of combined sewage systems on surface water may result in increased groundwater pollution due to the installation of rain gardens).

Continuous urbanization across the globe (McGrane, 2016) and a predicted increase in pluvial flooding due to climate change in many regions (e.g., Kendon et al., 2014; Miller & Hutchins, 2017) have set the conditions for more urban activities, pollution accumulation, and runoff events. Therefore, very likely creating an increase in diffuse pollution in the nearer future. A system's approach may help tackle current and future pollution rates under different sets of environmental conditions.

4.3 | Future research directions

The previous sections identified several knowledge gaps: the limited knowledge on the temporal variations of pollutant source areas concerning their size and pollution intensity (Section 2), limited availability of empirical data, and limited process understanding of rainfall-runoff responses on mixed urban surfaces (Section 3), and ultimately limited prediction of the timing of pollutant connectivity for different boundary conditions. In order to overcome the apparent lack of data, future field studies should focus on data collection within individual street canyons, that is, entire sections of streets including road, pavement and tree pit surfaces, building facades, and roofs of the surrounding buildings. Street canyons of different compositions and layouts or sub-districts containing a mixture of built and vacant lots to study changing pollution and runoff patterns needs to be assessed for intrinsic pollution patterns. Sampling locations should include different city types (new/old/industrial) and sizes (small town-to-city scales), building types, central to suburban locations, high to low degrees of sealing, different traffic amounts, and high to low-income areas. Weekly sampling over several seasons, similar to the data collection in rural catchments, will then provide a reliable base for further process studies. The major drawback is the high demand for resources that would be required for such a study and the question to which extent the results would be scalable to other cities.

To scale pollution to larger areas and to predict pollution under changing boundary conditions, urban catchment models need to evolve. Their current inability to handle complex terrain and pathways calls for a paradigm shift away from classical drainage models toward urban process models comprising structural and functional connectivity parameterizations and process descriptions. For the validation of these new model tools, it is essential to have beforementioned field data.

Finally, diffuse pollution is seen to be more of a problem in industrialized cities in the developed world. It is less in focus in cities of the developing world, where maintenance of public hygiene and extreme pollution of urban waters mostly from point sources leads the agenda (Chocat et al., 2007; Corcoran et al., 2010). Findings on the effects and functioning of urban connectivity may help to guide future research and re-design of urban drainage strategies to achieve sustainable management and at the same time, avoiding repeating past urban drainage mistakes.

5 | CONCLUSION

This study illustrated that very diverse spatial pollution patterns exist, which can become potential source areas of diffuse pollution in cities. Some pollutant patterns, for example, heavy metal originating from street surfaces or metal roofs, are better studied than others, such as street litter, abrasive sources, or priority substances. A comprehensive survey showed that little is known about the temporal variations of most pollution patterns (e.g., seasonal or annual variations) and the concurrency dynamics of pollutant build-up and pollutant connectivity as a function of rainfall timing. For active management of diffuse pollution, distribution processes must be studied, by considering the typical spatial layout of urban critical source areas and the interplay of pollutant buffers and boosters (e.g., in the form of decentralized drainage measures) and barriers (intentional as an integral part of, and unintentional due to the malfunctioning of the drainage system).

We conclude that a holistic approach for the simultaneous investigation of urban structural and functional connectivity relevant to pollutant transfer is essential for the identification of particular harmful diffuse pollution within complex urban catchments. Understanding pollutant connectivity is, in turn, a prerequisite to improve buffer and increase barrier functions against particular harmful pollutants and to enable compliance tests for pollution control for emerging pollutants such as microplastics, novel additives or priority substances from urban surfaces.

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CONFLICT OF INTEREST

The authors have declared no conflicts of interest for this article.

AUTHOR CONTRIBUTIONS

Eva Paton: Conceptualization; funding acquisition; investigation; methodology; writing-original draft; writing-review & editing. **Nasrin Haacke:** Conceptualization; investigation; methodology; visualization; writing-original draft; writing-review & editing.

DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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