



Review

Critical review of heavy metal pollution of traffic area runoff: Occurrence, influencing factors, and partitioning



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HIGHLIGHTS

- Heavy metal runoff concentrations in parking lots, bridges, and roads
- Large dataset of dissolved and total metal concentrations in traffic area runoff
- Description of site-specific and monitoring method-specific factors
- Summary of traffic-related and anthropogenic heavy metals in road runoff
- Reduction in Pb concentrations over time is one of the robust chemical results.

GRAPHICAL ABSTRACT



Site-specific



& Method-specific



Influencing Factors

Cd^{2+} Cu^{2+} Ni^{2+}
 Zn^{2+} Pb^{2+} **Dissolved** Zn^{2+}
 Cr^{6+} Zn^{2+} Ni^{2+} Zn^{2+} Cu^{2+}
Heavy Metals in Traffic Area Runoff
Categories: Parking Lots, Roads,
Bridges, and Highways

$ZnCO_3$ Pb $Zn(OH)_2$
Particulate $CuCO_3$
 Pb $Zn(OH)_2$ CrO_4^{2-}
 $ZnCO_3$

Occurrence & Partitioning

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ABSTRACT

A dataset of 294 monitored sites from six continents (Africa, Asia, Australia, Europe, North and South America) was compiled and evaluated to characterize the occurrence and fate of heavy metals in eight traffic area categories (parking lots, bridges, and three types each of both roads and highways). In addition, site-specific (fixed and climatic) and method-specific (related to sample collection, preparation, and analysis) factors that influence the results of the studies are summarized. These factors should be considered in site descriptions, conducting monitoring programs, and implementing a database for further research. Historical trends for Pb show a sharp decrease during recent decades, and the median total Pb concentrations of the 21st century for North America and Europe are approximately 15 µg/L. No historical trend is detected for Zn. Zn concentrations are very variable in traffic area runoff compared with other heavy metals because of its presence in galvanized structures and crumbs of car tire rubber. Heavy metal runoff concentrations of parking lots differ widely according to their use (e.g., employee, supermarket, rest areas for trucks). Bridge deck runoff can contain high Zn concentrations from safety fences and galvanizing elements. Roads with more than 5000 vehicles per day are often more polluted than highways because of other site-specific factors such as traffic signals. Four relevant heavy metals (Zn, Cu,

Abbreviations: AADT, average annual daily traffic; BR, bridge; d, dissolve; HL, highway with low AADT (>30,000) and non-urban land use; HU, highway with high AADT (>30,000) and urban land use; HWY, highway; p, particulate; Pb_21, dataset including only Pb concentrations measured in the 21st century; PL, parking lot; RL, road with low AADT (<5000); RM, road with medium AADT (5000 < AADT < 15,000); RU, road with high AADT (>15,000); t, total.

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Ni, and Cd) can occur in the dissolved phase. Knowledge of metal partitioning is important to optimize stormwater treatment strategies and prevent toxic effects to organisms in receiving waters.

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1. Introduction

Traffic area runoff summarize precipitation- and snowmelt-related discharges of mostly impervious surfaces (sidewalks, parking lots (PL), feeder streets, major roads, and highways (HWY)). The main contamination sources of traffic area runoff are related to traffic, surrounding land use, atmospheric contamination, and other meteorological and environmental conditions (Muschack, 1990; Ball, 2002; Crabtree et al., 2009; Valtanen et al., 2014). It is difficult to determine the dominant sources of pollutants because most substances have more than one origin and the water quality data of runoff from different sites are extremely heterogeneous because of differing background levels, types of uses (Göbel et al., 2007), and method-specific factors.

The substance spectrum analyzed in traffic area runoff waters includes organic parameters such as polycyclic aromatic hydrocarbons, total petroleum hydrocarbons, chemical oxygen demand, biological oxygen demand, and total organic carbon; heavy metals such as Pb, Zn, and Cu; and materials from de-icing salts such as chloride (Smullen et al., 1999; Moy et al., 2003; Eriksson et al., 2007; Kayhanian et al., 2012).

As HWY account for a small percentage of urban land use, they contribute only a small portion of pollutant loads compared with other road surfaces (Shelley et al., 1987). In urban catchments, all road surfaces represent approximately 10%–15% of the total area (Bannerman et al., 1993; Ball, 2002), and in commercial and industrial areas, PL can constitute up to 46% of the total area (Bannerman et al., 1993). Therefore, it is essential to consider all types of traffic area runoff.

In most cases, runoff waters from PL and road surfaces contain higher levels of the heavy metals than other types of runoff in drainage systems such as conventional roof runoff (Schueler, 2000; Ball, 2002). Metals in roof and road runoff contribute up to 80% of the total mass flow in combined sewer systems (Ellis et al., 1987; Boller, 1997). The present review focuses on all heavy metals in traffic area runoff that have either traffic or anthropogenic sources. These metals are transported by stormwater runoff either attached to solids or in dissolved form depending on the prevailing redox

and pH conditions (Ball, 2002). However, in most cases, only total metal concentrations are analyzed from runoff waters during measurement campaigns.

Some of these heavy metals can have acute or chronic impacts as a result of their accumulation in receiving waters in terms of aquatic habitats, drinking water resources, and recreational uses (Ellis and Revitt, 1982; Yousef et al., 1984). For potential toxic effects, the partitioning between the total and dissolved heavy metals is essential because the dissolved fractions are directly biologically available (Paulson and Amy, 1993; Crabtree et al., 2008). The toxicity of traffic area runoff has been investigated by various researchers (Gjessing et al., 1984; Pitt et al., 1995; McQueen et al., 2010). A particular link to the heavy metals was made by Tiefenthaler et al. (2001), who identified trace metals (particularly Zn) as important contributors to toxicity in PL runoff, and by Kayhanian et al. (2008), who identified dissolved Cu and Zn as the primary causes of toxicity in HWY runoff. In general, the toxicity of heavy metals is a function of several factors such as metal speciation and physical characteristics of receiving waters (Revitt and Morrison, 1987). If conditions change, particulate metals transported into receiving waters have the potential to repartition into the dissolved phase (Sansalone, 2002; Westerlund and Viklander, 2006). Metal partitioning is also important for designing appropriate stormwater treatment strategies (Hilliges et al., 2013; Maniquiz-Redillas and Kim, 2014).

The present review focuses on the objectives as follows:

- To describe site-specific factors (both fixed and climatic conditions) that influence the occurrence and partitioning of the heavy metals in traffic area runoff from different sites and that should be documented in investigation programs.
- To compile and evaluate the conditions of the monitoring methods, including sample collection, sample preparation, sample analysis, and calculation methods.
- To summarize the concentrations and fractionation of the heavy metals to produce a comprehensive dataset, to characterize different types of traffic areas, and to identify relevant heavy metals.

- To identify tendencies such as seasonal effects, phase-out measures, and land use influences that should be considered for planning stormwater treatment plants.

2. Material and methods

A dataset based on nearly 300 studies ($n = 294$) was analyzed. In addition to peer-reviewed journal papers, reports, books, and non-reviewed journal articles were considered after validation that presented heavy metal concentrations in traffic area runoff, including additional data concerning the monitoring program.

Several researchers evaluated street sweeping, the performance of grass swales, wetlands, detention ponds, biofiltration systems, porous asphalt surfaces, and artificial treatment plants. Only some characterized the quantity and quality of traffic area runoff containing heavy metal partitioning and influencing factors as their main objective. Therefore, the details of each monitoring program presented in publications differed and information regarding influencing factors such as those described in Section 3.1 are often missing.

To analyze historical trends and obtain a wide range of data, it is important to consider publications of the last decades. In North America and Europe, a large number of runoff data have been collected since the 1970s. Since the 1990s, data from Australia and New Zealand have been available, and since the 2000s, the number of publications from Asia, Africa, and South America has increased.

The date of the monitoring program was used instead of the publication date. If the date of the monitoring program was not available, the date of paper submission was used. All data are subdivided into eight categories: PL; bridges (BR); roads with an average annual daily traffic (AADT) of <5000 (RL), $5000 < \text{AADT} < 15,000$ (RM), and $>15,000$ (RU); urban HWY with an AADT of $>30,000$ (HU); non-urban HWY with an AADT of $>30,000$ (HN); and HWY with an AADT of $<30,000$ (HL). If the AADT was not mentioned, the classification was performed from the description of the monitoring site, e.g., a feeder street was classified as category RL. Each site was linked to one category. The identification (ID) of each site consists of two capital letters according to the category and two consecutive numbers, e.g., PL01. These IDs are used in Tables 1, 2, and 3 and in Sections 4.3 and 4.4.

One important step was to ensure that each dataset from one site was only considered once, although it might have been published twice or more in different forms. All data of one site was recalculated to obtain one dataset per site, except where the investigation period was not continuous (e.g., a new monitoring program after several years) or site-specific factors varied because of construction work (e.g., new road surface). In these cases, one site provides two or three datasets (highlighted in Tables 1, 2, and 3 by adding a letter a, b, or c at the end of an ID).

At data collection, total (t), dissolved (d), and particulate (p) concentrations were distinguished, as were mean and median concentrations. If authors presented both mean and median values, both values were used in this review. For sites including all single-event concentrations, both mean and median concentrations were calculated. Some extreme measured concentrations were deleted for plotting the figures but all these concentrations are mentioned in Section 4.

For statistical analysis and plotting histograms and box plots, the software package SPSS 22 (IBM) was used. For box and whisker plots, the bottom and top of each box are the first and third quartiles and the band inside the box is the median. The whiskers represent 1.5 times the interquartile range (IQR). Outliers (>1.5 times IQR) are marked as small circles and extreme values (>3.0 times IQR) as stars.

3. Occurrence and influencing factors

Literature datasets have a wide variation due to site-specific and monitoring-method-specific factors (Fig. 1). Site-specific factors can be divided into fixed site-specific factors (spatial variance) and climatic

site-specific factors (temporal variance); both affect the real pollution of surface runoff. Method-specific influences on the results, which should represent the real situation, are caused by the boundary conditions of a measurement program and occur during sample collection, sample preparation, sample analysis, and calculation.

3.1. Site-specific factors

3.1.1. Fixed site

Fixed site-specific factors can be divided into three categories: surrounding land use characteristics, traffic area site data, and operational characteristics (Shelley et al., 1987).

For the first factor, i.e., *surrounding land use characteristics*, several categories are used: non-urban, urban, ultra-urban, transportation, residential, open, agriculture, commercial, mixed, and industrial categories (Kayhanian et al., 2003; Flint and Davis, 2007). Driscoll et al. (1990) specified urban (undefined, commercial/residential, residential, and suburban) and non-urban (undefined rural, forest, undeveloped, agricultural, and desert) land use categories. Because the term residential land use could include a mixture of streets, driveways, rooftops, and lawns (Schueler, 2000), its use for analysis is restrictive and only comparisons of land use categories analyzed in one research program can provide an indication of their influence. E.g., the results of Dannecker et al. (1990) showed similar runoff pollution levels of an industrial road and a main street of an inner urban city.

The second factor, i.e., the *traffic area site data*, contains information about vegetation, topography, and road design. Only some data are available for surrounding *vegetation*, street surface covered with trees, and *topography*, which describes the slope and the configuration of the traffic area (cut, at grade, or elevated (Driscoll et al., 1990)). Jenewein and Schinner (1982) did not find a correlation of the slope with runoff concentrations. Most fixed site-specific data are available about *road design*, including information about cross-sections, pavement surfaces, and drainage areas. The cross-section describes the number of lanes per direction and the existence of curbs, median strips, hard shoulders, drainage systems, crash/noise barriers, and safety fences. The absence of curbs has a positive effect on reducing runoff concentrations (Driscoll et al., 1990), and further influences are described by Irish et al. (1998) and Li et al. (2008). Hard shoulders have an influence on water quality because they act as a sink for suspended solids and particle-associated metals (Berbee et al., 1999). Detailed information about the types of surfaces drained and sampled in monitoring programs is available in literature (62.9% of the datasets). Most surfaces are conventional asphalt surfaces ($n = 122$), followed by concrete ($n = 33$), asphalt and concrete ($n = 15$; mostly HWY with new lanes), porous asphalt ($n = 9$), asphalt and paver ($n = 3$; PL), paver ($n = 2$; PL), and crushed stone ($n = 1$; feeder street). Drapper et al. (2000) concluded that a concrete surface does not have a significant impact on pollutant runoff concentrations compared with asphalt surfaces. The surface condition has an effect on concentrations by either increasing runoff loads from decomposition products or reducing them during filtration into cracks and pot-holes (Driscoll et al., 1990). The size of the drainage area in the dataset of this study varies widely ($3\text{--}429,000 \text{ m}^2$) and has an effect on the first flush (Lau et al., 2009) and metal concentration, when the ratio of the drainage area to traffic volume is considered (CH2MHILL, 1998). The same applies to the imperviousness of the surface. Noise barriers lead to an increase in runoff concentrations (Dierkes, 1999; Kocher et al., 2010a,b), and concrete barriers can retain more solids on road surfaces (Barrett et al., 1998). Speed limits and traffic signals have an influence on runoff concentrations because braking and acceleration activities lead to increased abrasion of tires, higher use of brake linings, and increased automotive exhaust gas emissions (Muschak, 1990; Langbein et al., 2006). Drapper et al. (2000) measured higher concentrations of Cu and Zn at locations with exit lanes due to sharp braking by exiting vehicles and road signs release metals as a result of rain and splashing water (Van Bohemen and Van de Laak, 2003).

Table 1
Register of studies investigating parking lot runoff.

| ID | Literature | Location | Sampling period | Special conditions ^a | Discharge area[m ²] | Type and characteristics of parking lots | Number of events sampled | Type of sample collection | Sampling strategy | Pore diameter for filtration | Partition ^b | Metals detected ^c |
|-------|--|----------|-----------------------|---------------------------------|---------------------------------|--|--------------------------|---------------------------|----------------------------|------------------------------|------------------------|-----------------------------------|
| PL01 | Grotehusmann and Kasting (2002) | DEU | 2001-01–2001-12 | D, M, S | 10,000 | Tank farm and rest area for 41 trucks and four busses | 40 | Active-automatic | Flow-prop. | 0.45 | d, t | Cd, Cr, Cu, Hg, Pb, Pd, Pt, Zn |
| PL02 | Grotehusmann and Kasting (2002) | DEU | 2001-01–2001-12 | D, M, S | 5000 | Tank farm and rest area for 122 cars | 43 | Active-automatic | Flow-prop. | 0.45 | d, t | Cd, Cr, Cu, Hg, Pb, Pd, Pt, Zn |
| PL03 | Grotehusmann and Kasting (2002) | DEU | 2001-01–2001-12 | D, M, S | 17,700 | Tank farm and rest area for 18 trucks, 16 busses and 78 cars | 42 | Active-automatic | Flow-prop. | 0.45 | d, t | Cd, Cr, Cu, Hg, Pb, Pd, Pt, Zn |
| PL04 | Pick et al. (2002) | DEU | 1999-03–2000-11 | – | 300 | Campus parking lot | – | Active-automatic | Mixed | – | t | Cd (<1.0), Cr, Cu, Ni, Pb, Zn |
| PL05 | Engelhard et al. (2012) | AUT | 2010-04-02–2011-02-21 | D | – | Supermarket | – | Passive-automatic | Mixed | – | t | Cd, Cr, Cu, Ni, Pb, Zn |
| PL06 | Gromaire-Mertz et al. (1999) | FRA | 1996-07–1997-05 | – | – | Courtyard, old residential district with small businesses | 7 | Active-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Pb, Zn |
| PL07 | Gnecco et al. (2005) | ITA | 2002-01-23–2003-09-28 | – | 1000 | Campus parking lot, typical urban residential area | 9 | Active-automatic | Time-prop. (equally timed) | – | d | Cu, Pb, Zn |
| PL08 | Gnecco et al. (2008) | ITA | 2005-11-15–2006-07-12 | – | 14,000 | Airport parking area with 100 vehicles per hour | 4 | Active-automatic | Time-prop. (equally timed) | 0.45 | d, p | Al, Cu, Fe, Mn, Pb, Zn |
| PL09 | Nowakowska-Blaszczyk and Zakrzewski (1996) | POL | – | S | – | – | – | – | – | – | t | Pb |
| PL10 | Hogland et al. (1987) | SWE | 1985 | – | 470 | Research center | 2 | Active-automatic | Time-prop. (equally timed) | – | t | Al, Cd, Cr, Cu, Pb, Zn |
| PL11 | Wei and Morrison (1994) | SWE | 1992-05-26 | – | – | Park and ride | 1 | – | – | 0.45 | d, t | Pt |
| PL12 | Pitt et al. (1995) | USA | – | – | – | Park and ride | 1 | Manual | Random | 0.45 | d, t | Al, Cd, Cr, Cu, Ni, Pb, Zn |
| PL13 | Caltrans (2003) | USA | 2002-11-07–2003-04-24 | – | – | Park and ride, commercial site within the foothills | 8 | Active-automatic | Flow-prop. | – | d, t | As, Cd (<0.2), Cr, Cu, Ni, Pb, Zn |
| PL14 | Caltrans (2003) | USA | 2002-12-09–2003-02-12 | – | – | Park and ride | 8 | Active-automatic | Flow-prop. | – | d, t | As, Cd, Cr, Cu, Hg, Ni, Pb, Zn |
| PL15 | Caltrans (2003) | USA | 2002-11-07–2003-02-15 | – | – | Park and ride | 8 | Active-automatic | Flow-prop. | – | d, t | As, Cd, Cr, Cu, Hg, Ni, Pb, Zn |
| PL16a | Caltrans (2003) | USA | 2002-11-29–2003-05-02 | – | – | Park and ride, campus parking lot | 8 | Active-automatic | Flow-prop. | – | d, t | As, Cd, Cr, Cu, Ni, Pb, Zn |
| PL16b | Tiefenthaler et al. (2001) | USA | 2000-07-15–2000-10-07 | S | 608 | Park and ride, campus parking lot | 5 | Active-automatic | Time-prop. (equally timed) | 0.45 | d, t | Al, Cd, Cr, Cu, Fe, Ni, Pb, Zn |
| PL17 | Caltrans (2003) | USA | 2002-11-07–2003-04-14 | – | – | Park and ride | 8 | Active-automatic | Flow-prop. | – | d, t | As, Cd, Cr, Cu, Ni, Pb, Zn |
| PL18 | Caltrans (2003) | USA | 2002-12-20–2003-04-14 | – | – | Park and ride, rural area near foothills | 4 | Active-automatic | Flow-prop. | – | d, t | Cd, Cr, Cu, Ni, Pb, Zn |
| PL19 | Caltrans (2003) | USA | 2002-11-07–2003-04-12 | – | – | Park and ride, flat business/retail area | 8 | Active-automatic | Flow-prop. | – | d, t | As, Cr, Cu, Ni, Pb, Zn |

| | | | | | | | | | | | | |
|-------|---------------------------|-----|--------------------------|-------|--------|---|----|-------------------|-----------------------------------|-----------|------|--|
| PL20 | Caltrans (2003) | USA | 2002-11-08–2003-04-14 | – | – | Park and ride | 8 | Active-automatic | Flow-prop. | – | d, t | As, Cd, Cr, Cu, Ni, Pb, Zn |
| PL21 | Caltrans (2003) | USA | 2002-12-12–2003-03-14 | – | – | Rest area with planter boxes and restrooms | 8 | Active-automatic | Flow-prop. | – | d, t | As, Cd, Cr, Cu, Ni, Pb, Zn |
| PL22 | Caltrans (2003) | USA | 2003-02-12–2003-04-12 | – | – | Rest area, rolling grassy hills | 3 | Active-automatic | Flow-prop. | – | d, t | As, Cd, Cr, Cu, Ni, Pb, Zn |
| PL23 | Caltrans (2003) | USA | 2002-12-16–2003-04-17 | – | – | Rest area, flat agricultural land | 3 | Active-automatic | Flow-prop. | – | d, t | As, Cd, Cr, Cu, Ni, Pb, Zn |
| PL24 | Rushton (2001) | USA | 1998-11–1999-11 | – | 1050 | Florida Aquarium | 30 | Active-automatic | Flow-prop. | – | t | Cu, Fe, Mn, Pb, Zn |
| PL25 | Rushton (2001) | USA | 1998-11–1999-11 | – | 1050 | Florida Aquarium | 30 | Active-automatic | Flow-prop. | – | t | Cu, Fe, Mn, Pb, Zn |
| PL26 | Steuer et al. (1997) | USA | 1993-10, 1994-05–1994-08 | – | 54,000 | Commercial | 12 | Passive-automatic | Flow-prop. | 0.45 | d, t | Cu, Cd, Pb, Zn |
| PL27 | McQueen et al., 2010 | USA | 2006-10-16–2007-07-17 | nD, M | 1254 | Campus parking lot for 23 of 231 cars, residential/commuter | 19 | Active-automatic | First flush (2.54 mm) | – | t | Al, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn |
| PL28 | McQueen et al., 2010 | USA | 2007-01-05–2007-07-30 | nD, M | 6398 | Campus parking lot for 189 of 1089 cars, residential/commuter | 12 | Active-automatic | First flush (2.54 mm) | – | t | Al, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn |
| PL29 | Line et al. (1997) | USA | 1993-11-05 | – | 16,000 | Vehicle maintenance | 1 | Active-automatic | First flush (5 min) | – | t | <i>As (<10), Cd (<2), Cr (<5), Cu, Hg (<0.2), Ni (<10), Pb (<5), Sb (<50), Zn</i> |
| PL30 | Line et al. (1997) | USA | 1993-11-27 | – | 24,000 | Vehicle maintenance | 1 | Active-automatic | First flush (5 min) | – | t | <i>As (<10), Cd (<2), Cr, Cu, Hg (<0.2), Ni (<10), Pb, Sb (<50), Zn</i> |
| PL31a | Booth and Leavitt (1999) | USA | 1996-11–1996-12-25 | – | 18 | Employee parking lot with once-in, once-out daily usage | 3 | Passive-automatic | Flow-prop. | – | d, t | Ba, Cu, Fe, Mn, Pb, Zn |
| PL31b | Brattebo and Booth (2003) | USA | 2001-11–2002-03 | – | 18 | Employee parking lot with once-in, once-out daily usage | 9 | Passive-automatic | Flow-prop. | – | d, t | Cu, <i>Pb (<1)</i> , Zn |
| PL32 | Bannerman et al. (1993) | USA | 1991-05-05–1991-07-07 | – | – | Commercial | 5 | Passive-automatic | Flow-prop. | 0.7; 0.45 | d, t | Cd, Cr, Cu, Pb, Zn |
| PL33 | Bannerman et al. (1993) | USA | 1991-05-05–1991-07-07 | – | – | Industrial | 9 | Passive-automatic | Flow-prop. | 0.7; 0.45 | d, t | Cd, Cr, Cu, Pb, Zn |
| PL34 | Hatt et al. (2009) | AUS | 2007-01–2007-11 | – | 4500 | Campus parking lot, top level of a multi-level carpark | 17 | Active-automatic | Flow-prop. | – | t | Cu, Mn, Pb, Zn |
| PL35 | Maniquiz et al. (2010) | KOR | 2006-06–2008-10 | – | 10,700 | Vehicle registration office, commercial and light industrial | 45 | Manual | Time-prop. (first flush-enhanced) | – | t | Pb, Zn |

^a D = De-icing salt application, nD = no de-icing salt application, M = Maintenance, S = Sweeping.

^b d = dissolved, p = particulate, t = total.

^c Metals measured below study-specific detection limits are highlighted in this table with italics and the detection limits are presented in brackets behind these elements.

Table 2
Register of studies investigating road runoff.

| ID | Literature | Location | Sampling period | Special conditions ^a | Discharge area[m ²] | AADT [Vehicles per day] | Number of events sampled | Type of sample collection | Sampling strategy | Pore diameter for filtration | Partition ^b | Metals detected ^c |
|-------|----------------------------------|----------|------------------------------------|---------------------------------|---------------------------------|-------------------------|--------------------------|---------------------------|-----------------------------------|------------------------------|------------------------|--|
| RL01 | Xanthopoulos and Hahn (1995) | DEU | – | – | – | 3200 | 87 | – | – | – | t | Cd, Cu, Ni, Pb, Zn |
| RL02 | Dannecker et al. (1990) | DEU | 1987 | – | 210 | 500 | 17 | Active-automatic | Mixed | 250, 0.45 | t | Al, As, Ba, Be (<0.2), Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn |
| RL03a | Dannecker et al. (1990) | DEU | 1986 | – | 290 | 2300 | 16 | Active-automatic | Mixed | 250, 0.45 | p, t | Al, As, Be (<0.2), Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn |
| RL03b | Dannecker et al. (1990) | DEU | 1987 | – | 290 | 2300 | 16 | Active-automatic | Mixed | 250, 0.45 | p, t | Al, As, Ba, Be (<0.2), Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn |
| RL04 | Daub and Striebel (1995) | DEU | 1994 | – | – | <1000 | 3 | Active-automatic | – | 0.45, 0.1 | d, p | Cu, Pb |
| RL05 | Legret et al. (1996) | FRA | 1991–1994 | M | 2800 | 2000 | 39 | Passive-automatic | First flush | 0.45 | d, t | Cd, Cu, Pb, Zn |
| RL06 | Van Dam et al. (1986) | NDL | 1982-07–1985-12 | M, S | 176 | 3200 | 11 | – | – | – | t | Cd, Cr, Cu, Pb, Zn |
| RL07 | MacKay et al. (2011) | USA | 2006-08-29 | M | 549 | 3000 | 1 | Active-automatic | Time-prop. (equally timed) | 0.45 | d, t | Cu, Pb, Zn |
| RL08 | MacKay et al. (2011) | USA | 2006-08-29 | M | 1161 | 3000 | 1 | Active-automatic | Time-prop. (equally timed) | 0.45 | d, t | Cu, Pb, Zn |
| RL09 | MacKay et al. (2011) | USA | 2008-08-15 | M | 1302 | 4000 | 1 | Active-automatic | Time-prop. (equally timed) | 0.45 | d, t | Cu, Pb (<5), Zn |
| RL10 | MacKay et al. (2011) | USA | 2008-09-06 | M | 279 | 2000 | 1 | Active-automatic | Time-prop. (equally timed) | 0.45 | d, t | Cu, Pb, Zn |
| RL11 | Gilbert and Clausen (2006) | USA | 2002-06–2003-05 | – | 199 | – | 12 | Passive-automatic | Mixed | – | t | Cu, Pb, Zn |
| RL12 | Gilbert and Clausen (2006) | USA | 2002-06–2003-05 | – | 405 | – | 12 | Passive-automatic | Mixed | – | t | Cu, Pb, Zn |
| RL13 | Gilbert and Clausen (2006) | USA | 2002-06–2003-05 | – | 225 | – | 12 | Passive-automatic | Mixed | – | t | Cu, Pb, Zn |
| RL14 | Steuer et al. (1997) | USA | 1993-10, 1994-05–1994-08 | S | 104,000 | <3100 | 12 | Passive-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Pb, Zn |
| RL15 | Driscoll et al. (1990) | USA | 1970s and early 1980s | – | 5059 | 2000 | – | – | – | – | t | Cu, Pb, Zn |
| RL16 | Driscoll et al. (1990) | USA | 1970s and early 1980s | – | 1012 | 2500 | – | – | – | – | t | Cu, Pb, Zn |
| RL17 | Bannerman et al. (1993) | USA | 1991-05-05–1991-07-07 | S | – | 100–400 | 10 | Passive-automatic | Flow-prop. | 0.7, 0.45 | d, t | Cd, Cr, Cu, Pb, Zn |
| RL18 | Bannerman et al. (1993) | USA | 1991-05-05–1991-07-07 | S | – | 500–2150 | 9 | Passive-automatic | Flow-prop. | 0.7, 0.45 | d, t | Cd, Cr, Cu, Pb, Zn |
| RL19 | Waschbusch (1996) | USA | 1994–1995 | – | – | 378 | – | – | – | – | d, t | Cd, Cu, Pb, Zn |
| RL20 | Davis and Birch (2010) | AUS | 2007-08-19–2008-04-06 | – | 860 | 2000 | 4 | Active-automatic | Time-prop. (first flush-enhanced) | 250, 0.45 | d, p | Cu, Pb, Zn |
| RL21 | Maniquiz-Redillas and Kim (2014) | KOR | 2010-05–2012-11 | – | 520 | – | 24 | Manual | Time-prop. (first flush-enhanced) | 0.45 | d, t | Cd, Cr, Cu, Fe, Ni, Pb, Zn |
| RM01 | Robien et al. (1997) | DEU | 1990–1991 | D, S | 600 | 6000 | 4 | Active-automatic | – | 0.45, 0.1 | d, p | Cd, Cu, Fe, Ni, Pb, Zn |
| RM02 | Nadler and Meißner (2007) | DEU | 1996-10-01–2005-09-08 | D | 17.3 | 6800 | 36 | Passive-automatic | Mixed | – | t | Cd, Cr, Cu, Fe, Mo, Ni, Pb, Pt, Sb, V, Zn |
| RM02x | Nadler and Meißner (1999) | DEU | 1997-10-01–1997-12-31 | D | 17.3 | 6100 | 1 | Passive-automatic | Mixed | 0.45 | d, t | Cd, Cu, Pb, Zn |
| RM03 | Muschack (1990) | DEU | 1984-09–1985-08 | – | – | 14,200 | 31 | – | – | – | t | Cd, Cr, Cu, Fe, Hg, Ni, Pb, Zn |
| RM04 | Holthuis et al. (2012) | DEU | 2008-09–2010-07 | D | 41,200 | 15,000 | 8 | – | – | – | t | Cu, Fe, Zn |
| RM05 | SEH (2010) | DEU | 2008-08-20–2010-02-08 | D | 250 | 5500 | 18 | Passive-automatic | Flow-prop. | – | t | Cd (<50), Cu (<100), Pb (<100), Zn |
| RM06 | SEH (2010) | DEU | 2008-08-20–2010-02-08 | D | 250 | 5500 | 17 | Passive-automatic | Flow-prop. | – | t | Cd (<50), Cu (<100), Pb (<100), Zn |
| RM07 | SEH (2010) | DEU | 2008-08-20–2010-02-08 | D | 400 | 12,800 | 17 | Passive-automatic | Flow-prop. | – | t | Cd (<50), Cu (<100), Pb (<100), Zn |
| RM08 | Krauth and Stotz (1993) | DEU | 1991-12-17–1993-04-30 | D | 15,300 | 9870 | 111 | – | – | – | t | Cd, Cr, Cu, Fe, Ni, Pb, Zn |
| RM09 | Jenewein and Schinner (1982) | AUT | – | – | – | – | – | – | First flush | – | t | Cu, Pb, Zn |
| RM10 | Rutz (2009) | CHE | 2008-01-16, 2008-04-14, 2008-07-03 | – | 23,000 | 12,000 | 3 | Manual | Random | – | d, t | Cd, Cr, Cu, Pb, Zn |

| | | | | | | | | | | | | |
|-------|-------------------------------------|-----|------------------------------------|-------|--------|---------------|----|-------------------|-----------------------------------|-----------|------|--|
| RM11 | von Ballmoos (2007) | CHE | 2006-04-11, 2006-08-09 | - | - | - | 2 | Manual | Random | - | t | Cd (<1), Cr (<5), Cu, Pb (<5), Zn |
| RM12 | Gromaire-Mertz et al. (1999) | FRA | 1996-07-1997-05 | M, S | 602 | - | 7 | Active-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Pb, Zn |
| RM13 | Barbosa and Hvitved-Jacobsen (1999) | PRT | - | - | 5970 | 6000 | 10 | Active-automatic | Time-prop. (first flush-enhanced) | 0.45 | t | Cd (<1), Cr (<1), Cu, Pb, Zn |
| RM14a | Westerlund et al. (2003) | SWE | 2000-03-25-2000-04-25 | - | 660 | 7400 | 4 | Active-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Ni, Pb, Zn |
| RM14b | Westerlund et al. (2003) | SWE | 2000-05-25-2000-06-26 | - | 660 | 7400 | 3 | Active-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Ni, Pb, Zn |
| RM15 | Pitt et al. (1995) | USA | - | - | - | - | 1 | Manual | Random | 0.45 | d, t | Al, Cd, Cr, Cu, Ni, Pb, Zn |
| RM16 | MacKay et al. (2011) | USA | 2007-08-09, 2008-04-28, 2008-05-02 | M, S | 1079 | 8000 | 3 | Active-automatic | Time-prop. (equally timed) | 0.45 | d, t | Cu, Pb, Zn |
| RM17 | MacKay et al. (2011) | USA | 2006-09-03 | M, S | 1898 | - | 1 | Active-automatic | Time-prop. (equally timed) | 0.45 | d, t | Cu, Pb, Zn |
| RM18 | MacKay et al. (2011) | USA | 2006-09-03 | M, S | 866 | - | 1 | Active-automatic | Time-prop. (equally timed) | 0.45 | d, t | Cu, Pb (<5), Zn |
| RM19 | Fulkerson et al. (2007) | USA | 2005-03-03-2005-07-30 | nS | 8100 | 5000 | 14 | Manual | Time-prop. (equally timed) | 0.7 | d, p | MeHg, THg |
| RM20 | Steuer et al. (1997) | USA | 1993-10, 1994-05-1994-08 | S | 16,000 | 10,600 | 12 | Passive-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Pb, Zn |
| RM21 | Steuer et al. (1997) | USA | 1993-10, 1994-05-1994-08 | S | 21,000 | 3100-5100 | 12 | Passive-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Pb, Zn |
| RM22 | Steuer et al. (1997) | USA | 1993-10, 1994-05-1994-08 | - | 49,000 | - | 12 | Passive-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Pb, Zn |
| RM23 | Driscoll et al. (1990) | USA | 1970s and early 1980s | - | 1133 | 7300 | - | - | - | - | t | Cu, Pb, Zn |
| RM24 | Bannerman et al. (1993) | USA | 1991-05-05-1991-07-07 | S | - | 2850-7300 | 10 | Passive-automatic | Flow-prop. | 0.7; 0.45 | d, t | Cd, Cr, Cu, Pb, Zn |
| RM25 | Bannerman et al. (1993) | USA | 1991-05-05-1991-07-07 | - | - | - | 10 | Passive-automatic | Flow-prop. | 0.7; 0.45 | d, t | Cd, Cr, Cu, Pb, Zn |
| RM26 | Novotny et al. (1998) | USA | - | D | - | - | - | - | - | - | d, t | Cu, Pb, Zn |
| RM27 | Novotny et al. (1998) | USA | - | D | - | - | - | - | - | - | d, t | Cu, Pb, Zn |
| RM28 | Waschbusch, 1996 | USA | 1994-1995 | D | - | 6157 | - | - | - | - | d, t | Cd, Cu, Pb, Zn |
| RM29 | Novotny et al. (1998) | CAN | - | D | - | - | - | - | - | - | d, t | Cu, Pb, Zn |
| RM30 | Novotny et al. (1998) | CAN | - | D | - | - | - | - | - | - | d, t | Cu, Pb, Zn |
| RM31 | Brockbank et al. (1999) | AUS | - | - | - | - | - | - | - | - | d, p | Cu, Fe, Mn, Pb, Zn |
| RM32 | Prestes et al. (2006) | BRA | 2002-09-2004-03 | S | 1300 | 9000 | 21 | Manual | Flow-prop. | 0.45 | d, t | Cd, Cu, Pb |
| RM33 | Zhang et al. (2013) | CHN | 2010-06-06-2011-08-04 | nD | 540 | 10,000 | 9 | - | Time-prop. (first flush-enhanced) | - | d | Cu, Pb, Zn |
| RU01 | Wüst et al. (1994) | DEU | 1990-03-28, 1990-05-08, 1990-06-02 | D, S | 133 | 25,000 | 3 | Manual | - | 0.45, 0.1 | d, p | Cd, Cu, Pb, Zn |
| RU02 | Herrmann et al. (1992) | DEU | 1991-07-14, 1991-08-15, 1991-08-17 | nD, S | 30 | 16,000 | 3 | Active-automatic | - | 0.45, 0.1 | d, p | Cd, Cu, Fe, Ni, Pb, Zn |
| RU02x | Laschka et al. (1996) | DEU | 1991 | D, S | 30 | 16,000 | 6 | Active-automatic | - | - | t | Pt |
| RU03a | Helmreich et al. (2010) | DEU | 2003-11-2005-11 | D, S | 400 | 57,000 | 63 | Passive-automatic | First flush (1 mm rain) | 0.45 | d, t | Cd (<0.5), Cu, Ni, Pb, Zn |
| RU03b | Hilliges et al. (2013) | DEU | 2006-07-01-2007-06-30 | D, S | 100 | 57,000 | 24 | Active-automatic | Flow-prop. | - | t | Cu, Pb, Zn |
| RU04 | Dannecker et al. (1990) | DEU | 1987 | - | 250 | 16,200 | 12 | Active-automatic | Mixed | 250, 0.45 | t | Al, As, Ba, Be (<0.2), Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn |
| RU05 | Dierkes (2010) | DEU | 2009-01-12-2009-11-27 | D | 2300 | 18,000 | 11 | Manual | Random | - | t | Cd, Cu, Pb, Zn |
| RU06 | Gäth et al. (1990) | DEU | 1988-12-01-1989-01-13 | - | 162.2 | 18,129 | 8 | Active-automatic | Time-prop. (equally timed) | - | t | Cd, Cu, Pb, Zn |
| RU07a | Kasting et al. (2003) | DEU | 2000-10-2001-09 | D | 15,000 | 45,000 | - | Manual | Random | - | d, t | Cu, Pb, Zn |
| RU07b | Kasting and Grotehusmann (2007) | DEU | - | D | 15,000 | 45,000 | - | Manual | Random | - | d, t | Cu, Pb, Pt, Zn |
| RU07c | Grotehusmann and Kasting (2009) | DEU | 2007-02-09-2008-01-07 | D | 15,000 | 45,000 | 10 | Manual | Random | - | d, t | As (<5), Cd, Co, Cr, Cu, Fe, Hg (<0.2), Mo (<5), Ni, Pb, Sb, V, Zn |
| RU08 | Dierkes (1999) | DEU | 1997-06-1999-02 | D | - | 26,000 | 28 | Passive-automatic | Mixed | 0.45 | d, t | Cd, Cu, Pb, Zn |
| RU09 | Daub and Striebel (1995) | DEU | 1994 | - | 100 | 22,000 | 3 | Active-automatic | - | 0.45, 0.1 | d, p | Cu, Pb |
| RU10 | Schütte (1997) | DEU | 1996-09-1996-12 | - | - | 15,000-20,000 | 10 | - | - | - | t | Cd, Cu, Fe, Hg (<0.5), Ni, Pb, Zn |
| RU11 | Steiner et al. (2010) | AUT | - | - | - | - | - | - | - | - | t | Cr, Cu, Ni, Pb, Zn |
| RU12 | Windhofer et al. (2012) | AUT | 2011-10-21-2011-12-10 | D | 3300 | 26,000 | 4 | - | Flow-prop. | - | t | Cu, Rh, Pt, Zn |
| RU13 | Langbein et al. (2006) | CHE | 2002-10-23-2004-07-08 | - | 1500 | 17,000 | 7 | Active-automatic | Time-prop. (first flush-enhanced) | 0.45 | d, t | Cd, Cr, Cu, Fe, Ni, Pb, Zn |

(continued on next page)

Table 2 (continued)

| ID | Literature | Location | Sampling period | Special conditions ^a | Discharge area[m ²] | AADT [Vehicles per day] | Number of events sampled | Type of sample collection | Sampling strategy | Pore diameter for filtration | Partition ^b | Metals detected ^c |
|-------|--|----------|------------------------|---------------------------------|---------------------------------|-------------------------|--------------------------|---------------------------|-----------------------------------|------------------------------|------------------------|------------------------------|
| RU14 | Steiner et al. (2006) | CHE | 2002–2004 | – | 14.1 | 17,000 | – | Passive-automatic | Mixed | – | t | Cd, Cr, Cu, Fe, Pb, Sn, Zn |
| RU15 | Klimaszewska et al. (2007) | POL | 2003-01–2003-05 | S | – | 59,000 | 13 | Passive-automatic | Flow-prop. | – | t | Cd, Pb, Zn |
| RU16 | Klimaszewska et al. (2007) | POL | 2003-01–2003-05 | S | – | 39,400 | 8 | Passive-automatic | Flow-prop. | – | t | Cd, Pb, Zn |
| RU17 | Nowakowska-Błaszczyk and Zakrzewski (1996) | POL | – | S | – | – | – | – | – | – | t | Pb |
| RU18 | Wei and Morrison (1994) | SWE | 1992-05-26 | – | – | – | 1 | – | – | 0.45 | d, t | Pt |
| RU19a | Bannerman et al. (1993) | USA | 1991-05-05–1991-07-07 | S | – | 20,000 | 10 | Passive-automatic | Flow-prop. | 0.7, 0.45 | d, t | Cd, Cr, Cu, Pb, Zn |
| RU19b | Waschbusch (1996) | USA | 1994–1995 | – | – | 18,600 | – | – | – | – | d, t | Cd, Cu, Pb, Zn |
| RU20 | Bannerman et al. (1993) | USA | 1991-05-05–1991-07-07 | S | – | 19,800 | 9 | Passive-automatic | Flow-prop. | 0.7, 0.45 | d, t | Cd, Cr, Cu, Pb, Zn |
| RU21 | Davis and Birch (2010) | AUS | 2007-08-19–2008-04-06 | – | 1095 | 84,500 | 6 | Active-automatic | Time-prop. (first flush-enhanced) | 250, 0.45 | d, p | Cu, Pb, Zn |
| RU22 | Lloyd and Wong (1999) | AUS | 1999-01-22, 1999-01-29 | nS | 1500 | 32,000 | 2 | Manual | Time-prop. (equally timed) | – | t | Cd, Cu, Pb, Zn |
| RU23 | Timperley et al. (2005) | NZL | 2002-08-24–2002-09-18 | – | – | 17,000 | 4 | Active-automatic | Flow-prop. | – | d, p | Cu, Pb, Zn |
| RU24 | Chang et al. (2009) | CHN | 2005-06-28, 2005-09-21 | – | – | – | 2 | – | Time-prop. (equally timed) | 0.45 | d, t | Cd, Cr, Cu, Ni, Pb, Zn |
| RU25 | Huang et al. (2007) | CHN | 2005-06–2006-09 | – | 3875 | 30,000 | 5 | Active-automatic | Time-prop. (first flush-enhanced) | – | t | Cu, Pb, Zn |
| RU26 | Maniquiz et al. (2010) | KOR | 2006-06–2008-10 | – | 5000 | – | 45 | Manual | Time-prop. (first flush-enhanced) | – | t | Pb, Zn |

^a D = De-icing salt application, nD = no de-icing salt application, M = Maintenance, S = Sweeping, nS = no sweeping.

^b d = dissolved, p = particulate, t = total.

^c Metals measured below study-specific detection limits are highlighted in this table with italics and the detection limits are presented in brackets behind these elements.

Table 3
Register of studies investigating bridge runoff.

| ID | Literature | Location | Sampling period | Special conditions ^a | Discharge area [m ²] | AADT [vehicles per day] | Number of events sampled | Type of sample collection | Sampling strategy | Pore diameter for filtration | Partition ^b | Metals detected ^c |
|-------|--|----------|------------------------------------|---------------------------------|----------------------------------|-------------------------|--------------------------|---------------------------|-----------------------------------|------------------------------|------------------------|---|
| BR01a | Legret and Pagotto (1999) | FRA | 1995-03-28–1996-02-26 | D, S | 3200 | 12,000 | 49 | Active-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Pb, Zn |
| BR01b | Pagotto et al. (2000) | FRA | 1997-06-09–1997-11-09 | D | 3200 | 12,000 | 25 | Active-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Pb, Zn |
| BR02a | Hallberg et al. (2007) | SWE | 2005-05-03–2005-08-26 | D | 13,700 | 108,300 | 4 | Active-automatic | Flow-prop. | 0.45 | d, t | Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn |
| BR02b | Hallberg et al. (2007) | SWE | 2005-12-14–2006-02-14 | D | 13,700 | 108,300 | 5 | Active-automatic | Flow-prop. | 0.45 | d, t | Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn |
| BR03 | Driscoll et al. (1990) | USA | 1970s and early 1980s | – | 6070 | 42,000 | – | – | – | – | t | Cu, Fe, Pb, Zn |
| BR04 | Yousef et al. (1984) | USA | – | – | 6825 | 55,000 | 11 | – | – | – | d, t | Cd, Cr, Cu, Fe, Ni, Pb, Zn |
| BR05 | Yousef et al. (1984) | USA | 1982-08–1983-05 | – | 19,800 | 11,500 | 15 | – | – | – | d, t | Cd, Cr, Cu, Fe, Ni, Pb, Zn |
| BR06 | Driscoll et al. (1990) | USA | 1970s and early 1980s | – | 5787 | 70,000 | – | – | – | – | t | Cd, Cr, Cu, Pb, Zn |
| BR07 | Dean et al. (2005) | USA | 2002-04-11–2002-06-16 | – | 544 | – | 4 | Manual | Time-prop. (equally timed) | 0.45 | d, p | Cd, Cu, Pb, Zn |
| BR08 | Wu et al. (1998) | USA | 1995-09-22–1996-07-01 | D, M | 1497 | 25,000 | 10 | Active-automatic | Time-prop. (pre-set time) | – | t | Cd (<0.5), Cr, Cu, Ni, Pb |
| BR09 | Barrett et al. (1998) | USA | 1994-04-29–1995-05-18 | – | 526 | 8780 | 26 | Active-automatic | Flow-prop. | – | t | Cu, Fe, Pb, Zn |
| BR10 | Barrett et al. (1998) | USA | 1994-10-17–1995-05-08 | – | 1060 | 47,000 | 8 | Active-automatic | Flow-prop. | – | t | Cu, Fe, Pb, Zn |
| BR11 | Bourcier and Hindin (1979); Bourcier et al. (1980) | USA | 1976-10-14–1977-03-23 | M, S | 2323 | – | 3 | – | – | <i>d</i> | d, t | Cr, Fe, Pb, Ti, W, Zn |
| BR12 | Driscoll et al. (1990) | USA | 1970s and early 1980s | – | 401 | 42,000 | – | – | – | – | t | Cu, Pb, Zn |
| BR13 | Driscoll et al. (1990) | USA | 1970s and early 1980s | – | 890 | 17,000 | – | – | – | – | t | Cu, Pb, Zn |
| BR14 | Wilson (2006) | USA | 2003-04-08, 2003-10-06, 2004-01-14 | – | 250 | 100,000–180,000 | 3 | Active-automatic | Flow-prop. | – | d, t | As, Ba, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Sb, V, Zn |
| BR15 | Driscoll et al. (1990) | USA | late 1970s and early 1980 | – | 8498 | 53,000 | 35 | – | – | – | t | Cd, Cr, Cu, Fe, Hg, Pb, Zn |
| BR16a | Frazer, 1990 | CAN | – | – | – | – | 29 | – | – | – | t | Cd, Cu, Ni, Pb, Zn |
| BR16b | Marsalek et al. (1997) | CAN | 16 months | – | – | 83,700–102,100 | 53 | Passive-automatic | Flow-prop. | 0.45 | d, t | Cd, Cu, Ni, Pb, Zn |
| BR17 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 5200 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR18 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 3548 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR19 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 15,524 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR20 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 23,672 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR21 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 15,766 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR22 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 27,000 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR23 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 12,156 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR24 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 13,302 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR25 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 14,312 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR26 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 33,000 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR27 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 33,000 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR28 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 35,248 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR29 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 33,191 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR30 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 26,950 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR31 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 26,826 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR32 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 50,304 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR33 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 46,235 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR34 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 32,000 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR35 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 32,000 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR36 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 45,000 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR37 | Drapper et al. (2000) | AUS | 1997-07–1999-01 | – | – | 5900 | 12 | Passive-automatic | First flush (20 l) | – | t | Cu, Pb, Zn |
| BR38 | Gan et al. (2008) | CHN | 2006-04-06–2006-06-12 | M | 110 | 31,000 | 7 | Active-automatic | Time-prop. (first flush-enhanced) | – | t | Cd, Cr, Cu, Ni, Pb, Zn |
| BR39 | Yu and Zhao (2012) | CHN | 2007-04-22–2007-09-01 | – | – | – | 3 | – | Time-prop. (equally timed) | – | t | Cd, Cr, Ni, Pb, Zn |

^a D = De-icing salt application, M = Maintenance, S = Sweeping.

^b d = dissolved, p = particulate, t = total.

^c Metals measured below study-specific detection limits are highlighted in this table with italics and the detection limits are presented in brackets behind these elements.

^d Centrifuging 50 ml of sample for 5 min at 681 g.

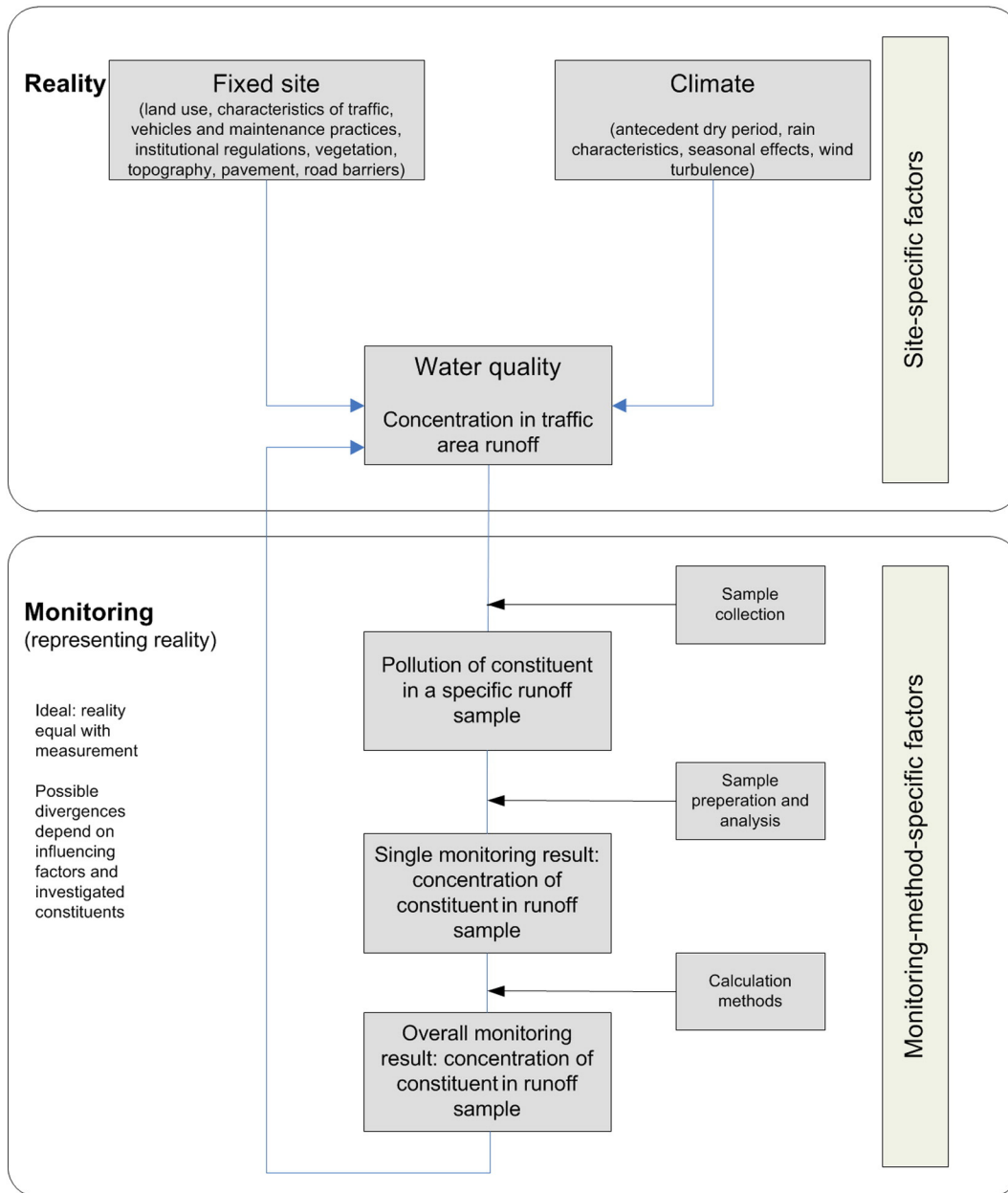


Fig. 1. Factors influencing monitoring programs and general flow schema for analysis of runoff characteristics and concentrations.

The final part of site-specific factors describes *operational characteristics* concerning traffic, types of vehicles, maintenance, and institutional regulations. Traffic volume is mostly characterized by AADT and varies in this review between 100 and 328,000 vehicles per day. Although some authors have concluded that AADT has the greatest influence on runoff concentrations of metals (Mangani et al., 2005; Crabtree et al., 2008), it might not be the best indicator for traffic area runoff concentrations and should not be used as a sole predictor because it only explains approximately 30% of the variation in these concentrations (Driscoll et al., 1990; Drapper et al., 2000; Kayhanian et al., 2003). Special cases of AADT are the traffic flow volume during sampling and between two sampling periods (Shinya et al., 2003; Hallberg et al., 2007; Li and Barret, 2008). CH2MHILL (1998) concluded that splashing and washing of pollutants from vehicles is more important than the wash-off of pollutants accumulated on road surface during dry periods and splashing corresponds with average vehicle speed during storms (Hallberg et al., 2007). It has not been specified whether the stated

AADT is only valid for the monitored lanes or for all lanes in both directions; this makes it difficult to compare different studies, e.g., the site described by Aryal and Lee (2009), and to evaluate the impact of AADT on runoff concentrations. Another traffic-related aspect is that the *percentage of trucks* can be very high in industrial zones (approximately 37%) (Dannecker et al., 1990) and can be as much as 60% of the AADT (Gan et al., 2008). Further factors influencing runoff quality are construction works, car accidents, firefighting foams, and leakages (Hogland et al., 1987; Barrett et al., 1998; Holthuis et al., 2012). *Maintenance* factors, which differ widely among countries (Gan et al., 2008), are mowing of the roadside shoulders (Barrett, 2008), street sweeping (Gromaire-Mertz et al., 1999; Waschbusch, 2003; Horwath and Bannerman, 2009; Helmreich et al., 2010), and winter services (Sansalone and Glenn, 2002; Bäckström et al., 2003; Crabtree et al., 2006; Hallberg et al., 2007; Klimaszewska et al., 2007; Tromp et al., 2012). The most important *institutional regulation* was the phase-out of Pb (cf. Section 4.1).

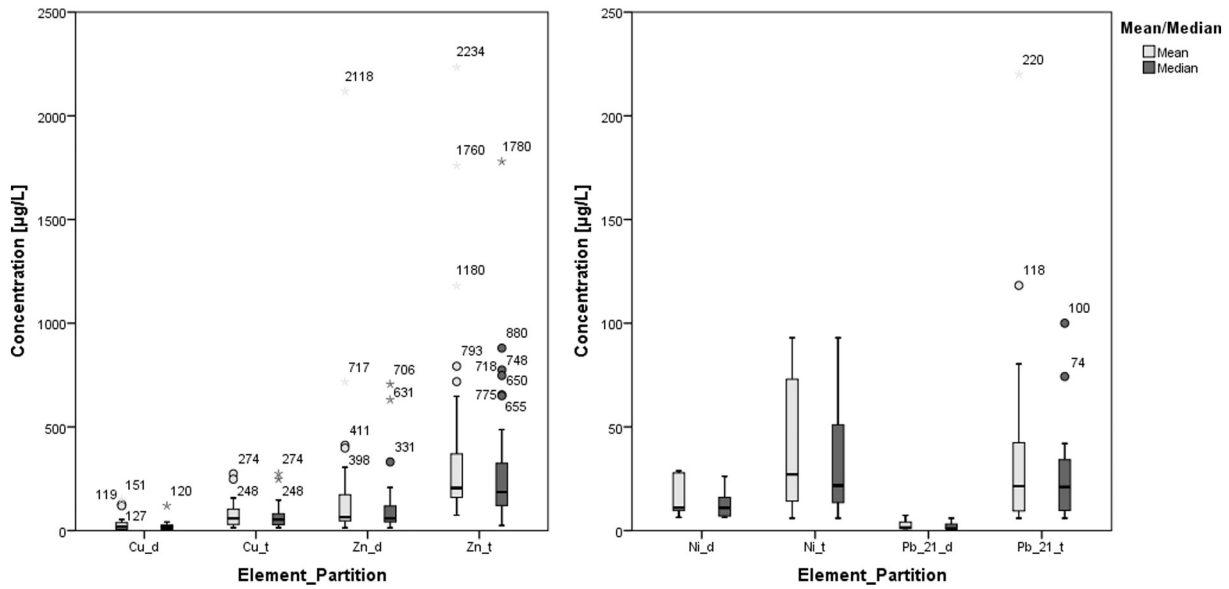


Fig. 2. Differences between mean and median values (d and t) for Cu, Zn (both left), Ni and Pb (both right) for highway runoff data (n = 41). Data were obtained from monitoring programs with more than two observed rain events, and both mean and median data were calculated for each site.

3.1.2. Climate

Climatic site-specific factors are dry deposition rates, antecedent dry periods (ADP), rain characteristics (volume, intensity, and duration), seasonal effects, and wind turbulence. Significant non-traffic-related sources are *dustfalls and dry deposition* during periods without rain, which entrains contaminants and removes them from the atmosphere (Hamilton et al., 1987; Ball, 2002). ADP were found to be a statistically significant factor for runoff concentrations due to the accumulation of substances on the traffic area surface (Harrison and Wilson, 1985; Hewitt and Rashed, 1992; Pitt et al., 1995; Irish et al., 1998; Drapper et al., 2000; Shinya et al., 2003; Westerlund et al., 2003; Mangani et al., 2005; Han et al., 2006a; Prestes et al., 2006; Wilson, 2006; Alo et al., 2007; Gan et al., 2008; Li and Barrett, 2008; Moores et al., 2010; Lee et al., 2011). However, Moy and Crabtree (2003), Shinya et al. (2003), Langbein et al. (2006), and Nason et al. (2012) found no relation between ADP and metal concentrations. This is most probably because of street sweeping, wind turbulence, and air turbulence caused by traffic

(Bourcier et al., 1980; Helmreich et al., 2010). However, not all storm events completely clean the road surface and this effect prolongs the actual ADP (Cristina and Sansalone, 2003). A special case in (semi)arid regions is the first storm of each season characterized by higher runoff concentrations (Stenstrom and Kayhanian, 2005; Terzakis et al., 2008).

For *rain characteristics*, mostly poor correlations were obtained (Moy et al., 2003; Han et al., 2006a; Desta et al., 2007; MacKay et al., 2011; Nason et al., 2012; Zhang et al., 2013) and only rain history appears to have an effect on the washed-off substances (Steiner and Goosse, 2007). In contrast, some researchers found a correlation between runoff concentrations and storm intensities (Pitt et al., 1995; Tiefenthaler et al., 2001; Crabtree et al., 2006).

Moy et al. (2003) have observed an influence of *seasonal effects* on runoff concentrations; however, the conclusions are heterogeneous. Several studies have indicated that metal concentrations are higher in snowmelt runoff than in rainfall runoff because of the snow accumulation period, and the following melting period appears to be a critical

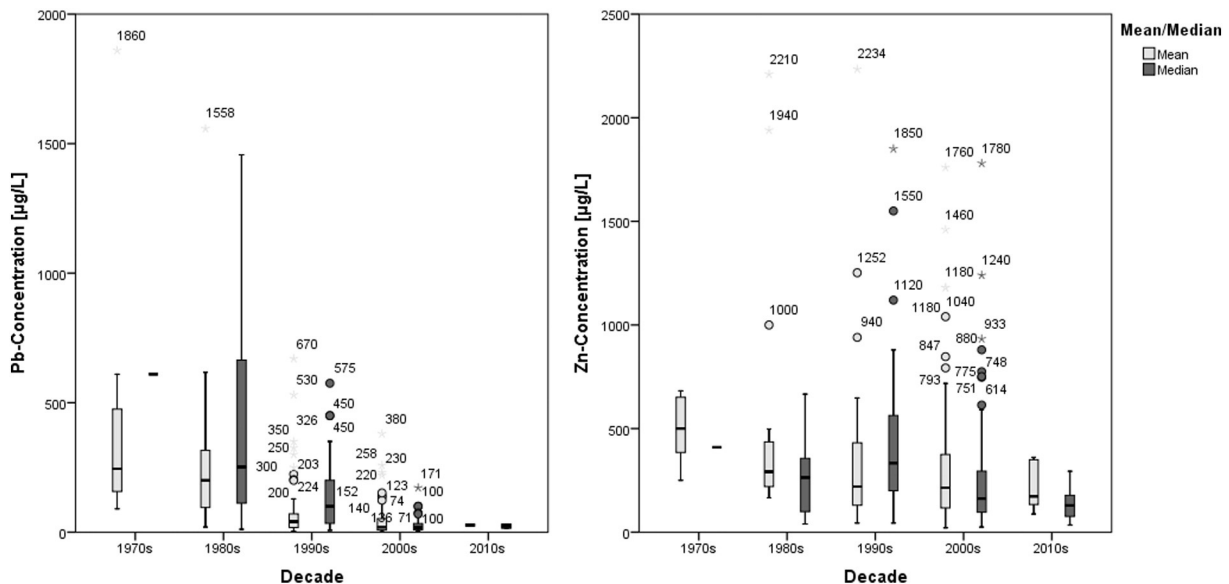


Fig. 3. Historical trends of Pb (n = 194 for mean; n = 146 for median) and Zn (n = 206 for mean; n = 148 for median) in traffic area runoff from the 1970s to the 2010s.

Table 4
 Statistics of Fig. 3 – historical trends of lead (n = 194 for mean; n = 146 for median) and zinc (n = 206 for mean; n = 148 for median) in traffic area runoff from the 1970s to date.

| Element | Decade | Calculation for all mean values from the literature | | | | | | | | | | Calculation for all median values from the literature | | | | | | | | | |
|---------|--------|---|--------|----------------|------|------|------|------|--------|------|------------------|---|--------|----------------|------|------|------|------|------|------|------------------|
| | | Min | Median | Geometric mean | Mean | 75th | 90th | 95th | Max | SD | 95% ^a | Min | Median | Geometric mean | Mean | 75th | 90th | 95th | Max | SD | 95% ^a |
| Pb | 1970s | 90.0 | 293 | 346 | 558 | 923 | 617 | 617 | 1860 | 662 | 694 | 610 | 610 | 610 | 610 | 610 | 610 | 610 | 1457 | 377 | 146 |
| Pb | 1980s | 20.0 | 200 | 162 | 277 | 328 | 319 | 339 | 1558 | 346 | 167 | 252 | 230 | 393 | 685 | 995 | 1281 | 1457 | 377 | 146 | |
| Pb | 1990s | 4.0 | 41.0 | 40.7 | 79.6 | 71.5 | 227 | 339 | 670 | 119 | 28.8 | 100 | 73.3 | 133 | 200 | 350 | 450 | 575 | 133 | 38.2 | |
| Pb | 2000s | 1.4 | 19.2 | 20.7 | 42.2 | 56.0 | 118 | 152 | 380 | 59.9 | 12.0 | 18.0 | 17.2 | 26.1 | 34.2 | 59.0 | 92.3 | 171 | 28.2 | 7.0 | |
| Pb | 2010s | 24.0 | 27.5 | 27.3 | 27.5 | 27.3 | b | b | 31.0 | 4.9 | 44.5 | 15.0 | 37.3 | 52.7 | b | b | b | 112 | 52.0 | 129 | |
| Zn | 1970s | 250 | 455 | 776 | 3540 | 5240 | b | b | 19,100 | 7624 | 8000 | 410 | 410 | 410 | 410 | 410 | 410 | 410 | 2892 | 565 | 238 |
| Zn | 1980s | 166 | 292 | 366 | 535 | 467 | 1994 | b | 2210 | 614 | 316 | 40.0 | 217 | 366 | 364 | 655 | 2336 | 2892 | 565 | 238 | |
| Zn | 1990s | 43.8 | 220 | 230 | 329 | 450 | 620 | 940 | 2234 | 342 | 89.1 | 44.0 | 332 | 445 | 564 | 846 | 1486 | 1850 | 372 | 116 | |
| Zn | 2000s | 21.0 | 215 | 201 | 295 | 375 | 652 | 857 | 1760 | 287 | 52.4 | 25.0 | 170 | 264 | 304 | 667 | 891 | 1780 | 296 | 68.0 | |
| Zn | 2010s | 88.0 | 173 | 188 | 213 | 353 | b | b | 361 | 115 | 121 | 35.0 | 189 | 545 | 861 | b | b | 2560 | 991 | 1040 | |

^a 95% confidence errors on the means.

^b No values because of small universe.

one for metal runoff concentrations (Lygren et al., 1984; Sansalone and Buchberger, 1996; Westerlund et al., 2003). A general increase of metals in traffic area runoff in winter was detected due to higher wear and tear as well as to an increased corrosion enhanced by de-icing salts and lower rain intensities (Legret and Pagotto, 1999; Hallberg et al., 2007; Helmreich et al., 2010). In contrast, Mungur et al. (1995) and Brezonik and Stadelmann (2002) detected the highest runoff loads in spring and summer, and Langbein et al. (2006) did not observe any seasonal dependency.

The final climatic factor, i.e., *wind and car-induced turbulence*, leads to a decrease in atmospheric deposition and reduces metal runoff concentrations (Stotz, 1987; Ball et al., 1998; Barbosa and Hvitved-Jacobsen, 1999; Preciado and Li, 2005). Wind turbulence increases with increasing vehicle speeds and decreases with the existence of hard shoulders and road barriers (Scheiwiller, 2008). In special cases such as in Auckland, New Zealand, frequent rain does not allow contaminants to be blown off the road surface (Timperley et al., 2005).

Further description of several site-specific factors for HWY can be found in Opher and Friedler (2010).

3.2. Monitoring-method-specific factors

In most studies, mean or median metal concentrations from runoff of a particular surface type are analyzed; other studies used pollutographs of single events. Further differences occur in sample preparation and analysis. The largest possible deviations arise during sampling and sample preparation, while the actual sample analysis causes less variation, if appropriate analytical methods are used.

3.2.1. Sample collection

The *type of sampling* (automatic or manual) depends on the research objective. For short-term sampling, *manual (grab) samples* are regularly used because they are a sampling requirement for stormwater permit applications (Line et al., 1997) and are easily manageable for an evaluation of the effectiveness of decentralized stormwater treatment plants (van Ballmoos, 2007; Dierkes, 2010; Rutz, 2012). *Automatic composite samplers* are used for a convenient collection of representative samples with up to several hundred subsamples of entire storm events (Ma et al., 2009; Steiner and Goosse, 2011b). Within the literature study, 13% of the datasets containing information about the sampling type were based on manual sampling, whereas 38% conducted passive-automatic sampling and 49% conducted active-automatic sampling. Active-automatic sampling can lead to the loss of particulate metals (Pagotto et al., 2000). This depends on the runoff composition, the mixture of the flowing water, the sampler intake orientation, and the tube and suction pump used. To minimize the fractionation effect during sampling, highest flow rates and shortest sampling tubes possible should be used; the loss of metals by adsorption can be avoided using polytetrafluoroethylene sampling tubes (Hewitt and Rashed, 1992). For the determination of pH and temperature, automatic samplers should not be used and manual sampling is recommended (Minervini, 2008). This recommendation is applicable to all rapidly degradable compounds and to substances that can adhere on container surfaces. To avoid cross-contamination of runoff samples, bottles must be changed for each rain event to prevent the mixing of samples from two events. Therefore, criteria for detecting a runoff are essential, as well as the event definition, which is directly linked to the sampling strategy and considers both the beginning and end of each event, to prevent a sampling of overlapping events. An appropriate cleaning of tubes, bottles, and the parts of the sampling device in contact with the runoff is necessary. For manual sampling, only bottle cleaning must be considered. Detailed examples of passive-automatic sampling devices have been given by Bannerman et al. (1993) and Li et al. (2008); an active-automatic sampling device has been described by Striebel et al. (1994). A comparison of grab and composite sampling techniques by Ma et al. (2009) concluded that a large number of grab samples (>30)

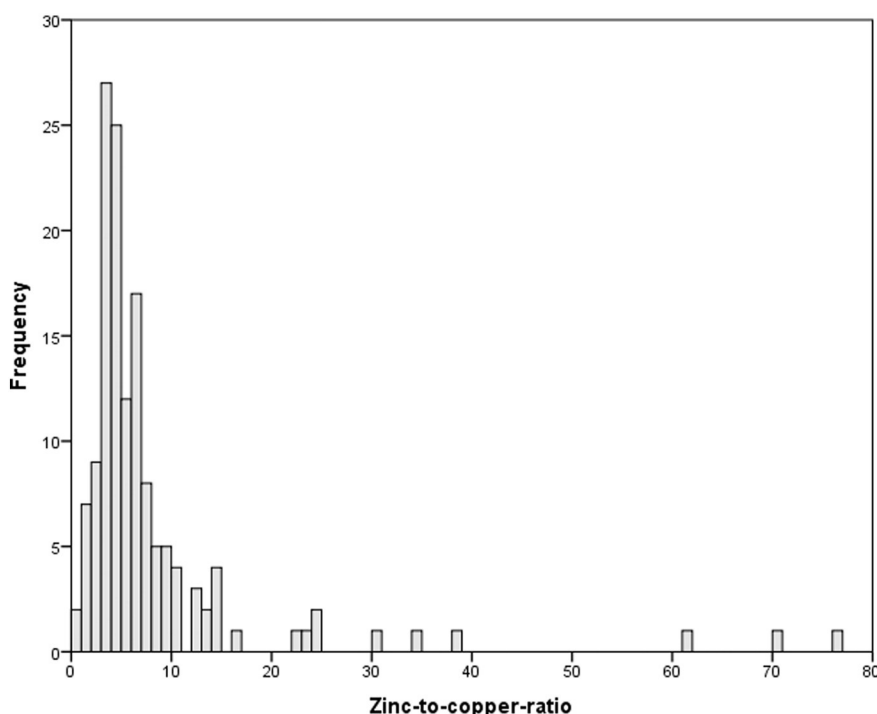


Fig. 4. Frequencies of Zn-to-Cu-ratios for all datasets containing median values ($n = 141$; mean = 8.1, SD = 10.9).

is required to approach the precision and accuracy of flow-proportional automatic sampling and for detecting a first flush effect, even more grab samples must be obtained. For manual sampling, the best strategy is to collect grab samples flow-proportionally (Stenstrom and Kayhanian, 2005). A storing of samples at 3 °C–6 °C is preferred (Hallberg et al., 2007; Hilliges et al., 2013; Scheiwiller, 2014).

The *sampling strategy* differs between flow-proportional (38% of datasets with information about the sampling strategy), time-proportional (24%), first flush (21%), mixed (12%), and random (5%) sampling.

In *flow-proportional sampling*, combined samples are mostly collected but discrete sampling is used for producing pollutographs (Legret and Pagotto, 1999) and a second device is required to trigger the sampling device. The second device can be, e.g., a magnetic inductive flow meter (Hilliges et al., 2013), a V-shaped overflow with an immersed ultrasonic flow meter (Legret and Pagotto, 1999; Hatt et al., 2009), a weir with a pressure probe (Hallberg et al., 2007), or a bubbler flow meter (Barrett et al., 1998; Nason et al., 2012). Criteria for sampling are very site-specific and can be 20 mL of sample for every 100 L of a runoff (Tromp et al., 2012). This strategy is limited by the detection limit of the second device sensor (Lau et al., 2009).

Time-proportional sampling can be subdivided into first flush-enhanced and equally timed sample collection. Han et al. (2006b) performed first flush-enhanced sampling, in which five grab samples were collected every 15 min in the first hour after the start of a detectable runoff. Subsequently, one sample was obtained every hour for the following 7 h and one or two additional grab samples were collected for events lasting longer than 8 h. Equally timed samples were obtained at specific time intervals by considering the sampling type and site-specific factors such as rain intensity, the slope of the catchment area and the season (Stotz, 1987). In most cases, the time interval is 5 min but it varies between a few minutes (2 min (Tiefenthaler et al., 2001) or 3 min (Chang et al., 2009)) up to 45 min (Golwer and Schneider, 1983) and 60 min (Gäth et al., 1990). Discrete time-proportional samples can be handled similar to mixed/combined flow-proportional samples by considering the data for simultaneous and continuous flow measurements.

For *first flush sampling*, which is a special case of time-proportional sampling, three types of triggering signals are used: time, volume, or rain depth. Line et al. (1997) sampled water of the first 5 min per rain event, Mangani et al. (2005) collected the first 10 L, Drapper et al. (2000) collected the first 20 L, Helmreich et al. (2010) collected the first 400 L (equaling a rain depth of 1 mm for this monitored site), and McQueen (2008) collected the first 2.5 mm runoff of a storm event. For an evaluation of the first flush effect, which might occur in the first 30 to 60 min, a sampling of the entire rain event is necessary (Han et al., 2006a). Definitions of the first flush effect are given by Wanielista and Yousef (1993), Saget et al. (1996), and Bertrand-Krajewski et al. (1998).

Mixed sampling by collecting the complete runoff volume of one event is an ideal method to obtain representative concentrations of a storm event. However, this strategy is only practical for small catchment areas (<20 m²) and a settling of particles in a storage tank must be avoided (Kasting et al., 2003). Some mixed samples are taken over a time period containing several rain events (Nadler and Meißner, 2007; Engelhard et al., 2012).

All *randomly collected samples* were taken manually during storm events to evaluate the effectiveness of best management practices.

The type and strategy of sampling have an influence on metal concentration, as does the *location*, from which runoff samples are taken. E.g., sediments can be retained by the overflow of a measurement channel and are not sampled by the suction pump of an automatic sampler (Legret and Pagotto, 1999). Further aspects are related to the characteristics of the channel. A concrete channel has an influence on pH and long distances between the runoff edge, and the sampling device might allow the settling of solids in the channel during rain events with low intensities. Coatings and tube materials (e.g., stainless steel) can increase Ni concentrations in the samples (Nadler and Meißner, 1999).

Overall, as a result of rain events without runoff, equipment failure, and operational reasons such as holidays, not all rain events can be sampled. Grotehusmann and Kasting (2009) sampled and analyzed about half of all events during a one-year investigation period, Lee et al. (2011) monitored 50 out of more than 100 storm events during a

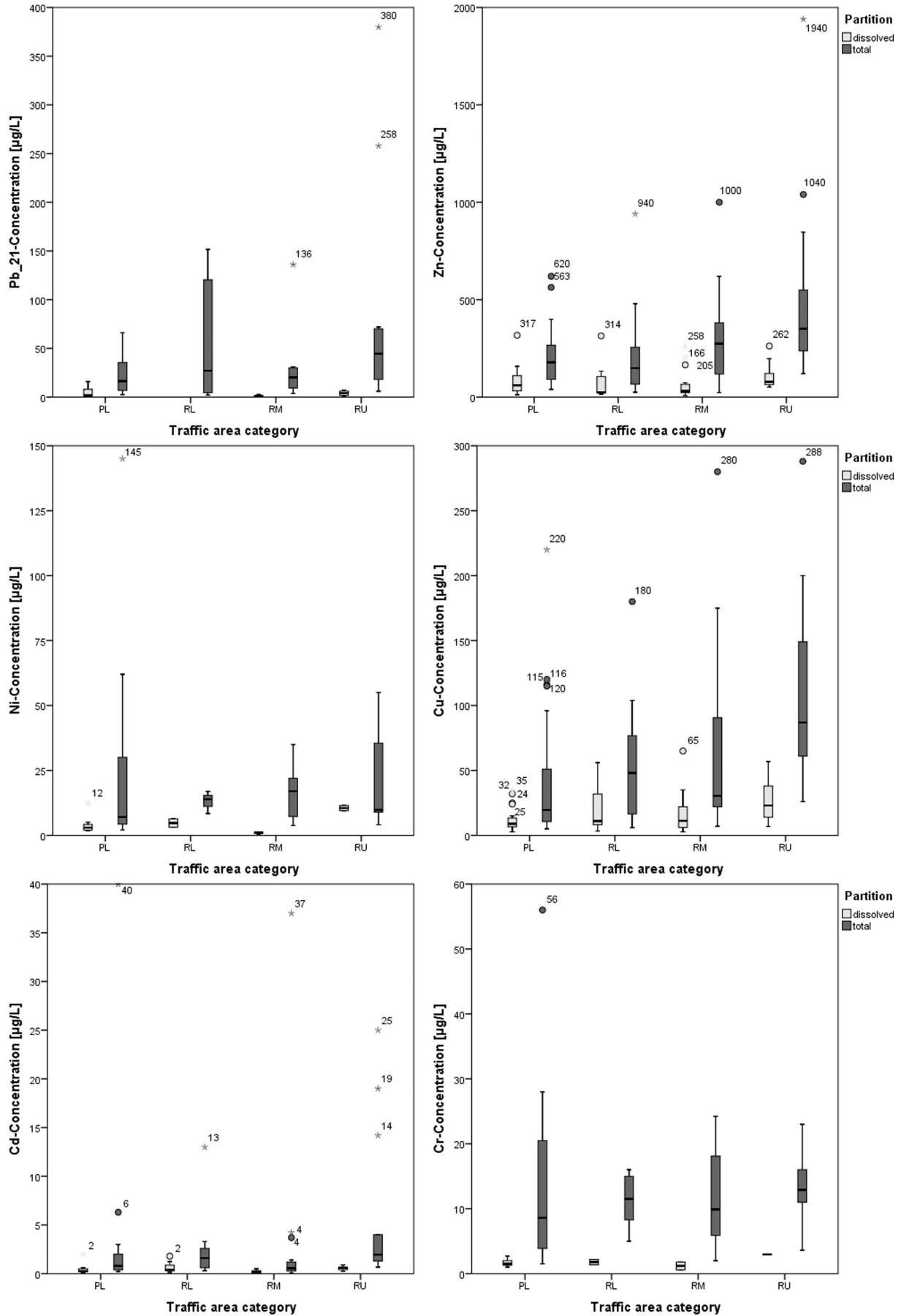


Fig. 5. Mean metal concentrations (d and t) in parking lot (PL) and road (RL, RM, and RU) runoff.

three-year period, and Terzakis et al. (2008) sampled 41 out of 58 rain events during a one-year period.

In conclusion, sampling programs had different objectives and constraints, and the sampling types and strategies used varied widely. Further influencing factors arise from equipment and operators.

3.2.2. Sample preparation and analysis

Prior to analysis, samples must be prepared according to the objectives. Only 36% of the datasets contain information about *preparation methods* (e.g., homogenization, subsampling, and filtration) and only 55 sets describe the holding time. For all metal analyses except those for total concentrations, the *holding time* is essential by having a critical effect on pH and on the fractionation of metals. Runoff samples were held for less than 1 h (Dean et al., 2005; Fulkerson et al., 2007), for 2–3 h (Barbosa and Hvitved-Jacobsen, 1999), for 7 h (Lau et al., 2009) or for a maximum of 24 h (Stotz, 1987; Stotz and Krauth, 1994; Booth and Leavitt, 1999; Brattebo and Booth, 2003; Flint and Davis, 2007; Moores et al., 2010; Lee et al., 2011; Nason et al., 2012; Borne et al., 2013) before delivery to a laboratory and beginning of preparation. Other studies stated the holding time as “immediately” or did not specify it properly. As Minervini (2008) discussed, the description of the sample collection and analysis often does not state how the pH analysis was performed, especially considering the maximum recommended holding time of 15 min (APHA, 2005). Only pH measurements in the field can comply with this requirement (Sansalone et al., 2005). As on-site filtration is risky owing to possible contamination of aqueous samples, samples should be fractionized as soon as practically possible within hours after collection in a laboratory to preserve the partitioning at the time of interest (Gnecco et al., 2008). For the *fractionation process*, a 0.45- μm -pore-diameter membrane filter is used (APHA, 2005) and the metals passing through the filter are the *dissolved part*. Filters with smaller pores (0.1 μm (Herrmann et al., 1992), 0.2 μm (Kluge et al., 2014), and 0.22 μm (Mangani et al., 2005)) and larger pores of up to 0.8 μm (Golwer and Schneider, 1983; Fulkerson et al., 2007) have also been applied. In addition, centrifuging (Bourcier and Hindin, 1979) and decanting before filtration (Bäckström et al., 2003) have been conducted. After filtration, the samples are mostly conserved with nitric acid to a pH of less than 2. The *soluble part* is determined as the dissolved part but from an acid preserved solution. Further factors associated with the filtration process are the diameter of the filter, the manufacturer, the volume of sample processed, and the amount of suspended solids in the sample, from which an inclusion/exclusion of colloidal-associated trace metals can occur (Horowitz et al., 1996). Measurements by Langbein et al. (2006) detected that the filtrate might contain fine colloids and particulate material in the nanometer range. An example of contamination during preparation and analysis of aqueous samples that was detected using blanks is described by Bannerman et al. (1993), in which the samples were contaminated with Zn during filtering procedure.

Preciado and Li (2005) described a procedure of using a chelex resin in a glass column to obtain the *resin-exchangeable part* of heavy metals, which is highly bioavailable.

To determine the potentially bioavailable part of a sample that can be *mobile* in different environmental conditions (i.e., for most metals at a lower pH), one part of a sample can be decanted and acidified followed by filtration after 24 h (Bäckström et al., 2003).

The *acid-exchangeable part* of metals is also called the total recoverable or pseudo-total concentration and is determined by adding a strong acid to the sample. Depending on the oxidizing potential of the acid and other equipment used (i.e., autoclave, microwave), a different part of all metals (i.e., all dissolved/soluble and a varying part of the particulate bound metals) is analyzed.

For the determination of *total concentration* of metals, aqua regia digestion (Nadler and Meißner, 1999; Hilliges et al., 2013), aqua regia digestion with a microwave (Shinya et al., 2000; Shinya et al., 2003), or lithium metaborate digestion must be used to destruct any particulate matter.

The difference between the results of total and acid-exchangeable concentrations depends on the element, fixed site-specific factors, and climatic factors.

The particulate content is measured by separating and weighing the particulates gravimetrically. The metal concentration in *particulates*, i.e., the particulate bound metals, is usually taken as the difference of total or acid-exchangeable and dissolved or soluble concentrations but can also be determined in the filter residue by acid digestion, autoclaves, microwave digestion, or a combination of these digestion methods.

After sample preparation, the concentrations were determined in most cases using *atomic absorption spectroscopy* (AAS, 62 times), *inductively coupled plasma optical emission spectrometry/inductively coupled plasma mass spectrometry* (ICP-OES/ICP-MS, 108 times), or both techniques consecutively to reduce the detection limits of several elements (25 times), and to improve the differentiation of solid and liquid phases (Sansalone and Buchberger, 1997b).

To obtain precise and accurate data, it is essential to conduct several *quality assurance/quality control* (QA/QC) procedures. First, field replicate samples should be collected and analyzed. Second, field and method blanks, certified reference materials, matrix spikes, and duplicates or triplicates must be considered and analyzed to prevent failures during sample preparation/analysis (Line et al., 1997; Wilson, 2006). Line et al. (1997) used standard and spiked samples for a minimum of 10% of their field samples. A further QA/QC procedure is the use of materials from two manufacturers for every elemental standard used in the analysis (Westerlund and Viklander, 2006).

3.2.3. Calculation methods

Thomson et al. (1997) and Drapper et al. (2000) concluded that a minimum of 17 runoff events provides a statistically representative sample database. In the dataset of this review, the number of events sampled varied between 1 and 211 (Moxness, 1986), with a mean of 18.

The most influential factor for the calculation of mean or median values concerns the analyzed concentrations of samples below the *detection limits*. There are several methods of dealing with this type of result: they could be discarded and only observed concentrations could be used for the calculation (Pitt et al., 1995), or they could be set as the detection values (Barrett et al., 1998; Dierkes, 1999; Lange et al., 2003; Borne et al., 2013), or as numerical values of one-half the detection level (Herrmann et al., 1992; Bannerman et al., 1993; Laschka et al., 1996; CH2MHILL, 1998; Brattebo and Booth, 2003; Flint and Davis, 2007; Grotehusmann and Kasting, 2009; Lau et al., 2009). None of the studies included in the dataset specified, when a mean or median value is stated as “not detected” or when it is calculated using either the full or one-half the detection values and it is often not specified, how many of the analyses were below the detection limit. This has a crucial effect on the concentrations of elements near the detection limit such as Cd or dissolved Cr concentrations and might lead to an overestimation of the dissolved fraction. A comparison of the mean and median concentrations of a HWY runoff shows that the median Cd concentration was as high as the detection limit (1.0 $\mu\text{g/L}$) and only the mean concentration was slightly higher (1.6 $\mu\text{g/L}$) (Lange et al., 2003).

Some researchers selected the storm events used for analysis. The *main criterion for the selection* is associated with ADP to distinguish between different events or to discard several rain events. In the first case, different lengths of time between 30 min (Steiner et al., 2008) and 10 h (Barrett et al., 1998) of dry weather are used to separate two events and the concentration of each event is used for the calculation of mean/median values. In the second case, an ADP between 24 h (Moy et al., 2003; Wilson, 2006) and up to five days was maintained before stormwater sampling was deployed (Hares and Ward, 1999). This approach, together with the influence of the ADP on runoff concentrations (longer ADP leads to higher concentrations, cf. Section 3.1), might lead to higher metal concentrations in a runoff in comparison with measurements obtained without a selection. A minimum rain depth, which varied between 1.5 mm (Steiner et al., 2008), 3.8 mm

(Nason et al., 2012), 5.1 mm (Horwath and Bannerman, 2009), 6.4 mm (Wilson, 2006), and 13 mm (Brattebo and Booth, 2003), is considered to ensure that sufficient and representative sample volumes could be collected. Nason et al. (2012) considered a minimum rain duration of 1 h.

Further influencing factors involve data processing and plausibility tests for all relevant data such as flow measurements, clock time adjustments, and communication among sampling devices.

The most important criterion for representative datasets must be a sampling of a full range of seasonal conditions for a complete range of rainfall intensities, depths, and durations (Thomson et al., 1997; Moy et al., 2003; Stagge et al., 2012; Tromp et al., 2012) and all sources of critical pollutants should be considered (Sansalone and Buchberger, 1997b). However, most studies have been performed within a single year or a part of a year and their conclusions often do not reflect the seasonality and environmental variability of the site (Uhl and Kasting, 2002; Steiner and Goosse, 2011a).

Overall mean and median values presented in previous studies, which were calculated using different methods and should describe the real water quality (Fig. 1), are not assumed to be comparable. To prove the correctness of this hypothesis, mean and median concentrations that are both available for each HWY site and contain a minimum of three events are presented in Fig. 2. For the mean datasets, more extreme values and outliers are computed (for all four elements and for the dissolved and total fractions) in comparison with the median values. Therefore, for this review, mean and median datasets are not combined and are used separately to characterize the central tendency and variability of runoff concentrations among sites or site categories.

4. Occurrence and partitioning

4.1. Historical trends

Historical trends of Pb and Zn in traffic area runoff published in literature were analyzed from the 1970s to date. As described by Kayhanian (2012), for HWY runoff, Pb concentrations have been decreasing continuously in the last few decades. The decrease of Pb use as an anti-knocking agent started in the USA in the mid-1980s and was completed in 1996 (FHWA, 1999). In Europe, the situation was not as homogenous as in the USA. In Germany, the beginning of the phase-out of leaded gasoline began in 1976, when the permitted concentration of Pb in gasoline decreased sharply to 150 mg/L (Golwer and Schneider, 1983). The phase-out in other countries of Western Europe started at the latest in 1986 (Berbee et al., 1999) and ended with the elimination of leaded gasoline in Italy in January 2002 (Mangani et al., 2005). At the same time, leaded gasoline was banned in China (Gan et al., 2008). Further reductions in Pb runoff concentrations are related to the substitution of Pb in other sources such as tires and brake linings (Sansalone et al., 1996; Legret and Pagotto, 1999; Davis et al., 2001), lubricating oil and grease (Ball et al., 1998), weights added to vehicles for tire balance (Root, 2000; Bleiwas, 2006), and Pb-based paints on roads and HWY (Mosley and Peake, 2001; Preciado and Li, 2005; Masanao et al., 2006). However, tires still contain Pb (Shinya et al., 2006; McKenzie et al., 2009).

A decline in Pb concentrations can be observed in the dataset of this review. Because of different numbers of measurements per year, the collected data were aggregated into decades to ensure that there were sufficient data for statistical analysis. Less data is available for the 1970s and 2010s compared to the other three decades (Fig. 3). The decline of Pb starting in the 1980s correlates with the statements mentioned above and is comparable to the one presented by Kayhanian (2012) for HWY runoff. Overall median concentrations of Pb in the 1980s are 252 µg/L (for all median values) and 200 µg/L (for all mean values). In the 2000s, overall median concentrations are one order of magnitude lower at 18.0 µg/L (for all median values) and 19.2 µg/L (for all mean values). All data is summarized in Table 4. The highest concentrations in the 21st century illustrated in Fig. 3 were measured in

Poland by Klimaszewska et al. (2007), South Korea by Maniquiz-Redillas and Kim (2014), and China by Zhao et al. (2001). To increase the benefit of all other data analysis, only Pb data for the 21st century were used. These Pb concentration datasets shown in Figs. 5 and 6 are highlighted by the use of the index “21”. However, in this selected dataset, artifacts caused by the wash-off of Pb deposited on the upper part of road soils cannot be excluded completely.

For Zn, the trend is different. Neither a significant decline nor an increase in measured concentrations can be identified for the last few decades (Fig. 3). For the medians, overall concentrations of Zn in the 1980s are 264 µg/L (for all median values) and 292 µg/L (for all mean values). In the 2000s, overall median concentrations are 162 µg/L (for all median values) and 215 µg/L (for all mean values). All data is summarized in Table 4. Kayhanian et al. (2007) compared different Zn datasets obtained in California and concluded that there was a decrease in measured concentrations. In contrast, an increase in runoff concentrations is proposed as a result of an increasing use of Zn for safety fences (Dierkes and Geiger, 1998) and of galvanized car parts containing a Zn plating that should prevent corrosion (Sansalone and Buchberger, 1997a). In addition to these increasingly common applications, Zn has been used for decades in several structural elements, batteries (Davis et al., 2001), tires (Van Bohemen and Van de Laak, 2003; McKenzie et al., 2009), brake linings (Legret and Pagotto, 1999), motor oils and grease (Ball et al., 1998). Consequently, Legret and Pagotto (1999) indicated that Zn concentrations remain highly variable depending on the use of safety barriers made from galvanized steel and site-specific factors that have an influence on tire and brake wear. This is confirmed by the occurrence of several outliers and extreme values shown in Fig. 3. The highest values in the 21st century are affected by the corrosion of galvanized elements (Wilson, 2006; Gan et al., 2008) or by highly trafficked and congested roads (Helmreich et al., 2010; Moores et al., 2010) and HWY (Lau et al., 2009; Kluge et al., 2014). Preciado and Li (2005) identified Zn as a metal of future potential concern because it was second in concentration to Fe in most dust, air, and water samples.

4.2. Variability of Zn

Zn concentrations are very variable in traffic area runoff compared with other heavy metals as shown in Section 4.1. In recent years, most monitoring programs measured Zn and Cu, and frequencies of Zn-to-Cu-ratios are calculated for all datasets containing median values ($n = 141$, Fig. 4) to show the variability of Zn compared to Cu.

Calculation of Zn-to-Cu-ratios was first performed by Moores et al. (2010) for total median concentrations of HWY runoff in New Zealand. The calculated ratios in untreated runoff waters were within the range of 4–5 at all four sites (6–8 rain events per site; active-automatic and flow-proportional sampling) except at one site with porous asphalt that had a lower median concentration ratio of 2.4.

The most frequent ratios of Fig. 4 are between 3 and 5 with a larger deviation (mean = 8.1, SD = 10.9). For the highest ratios, either sample contamination or special site conditions are responsible. The highest value of 76.5 was calculated for a PL runoff that received water from a slate roof with Zn gutters (Gnecco et al., 2005). The ratio of 70.5 could be explained by the presence of a nearby Zn smelter (Driscoll et al., 1990), the ratio of 30.8 by vehicle maintenance (Line et al., 1997), and the ratio of 24.5 by the surrounding old residential district (Gromaire-Mertz et al., 1999). The ratio of 23.4 is correlated with a failure of galvanizing material of a BR (Wilson, 2006). Mangani et al. (2005) described the presence of safety fences at a construction site with a nearby agricultural land use, where metal-containing pesticides are used, that might have led to the high Zn-to-Cu-ratio of 22.7. Further ratios might be influenced by the presence of guardrails (ratios 14.0 and 16.5) (Gäth et al., 1990; Nadler and Meißner, 1999), and the erosion of galvanized utilities and noise barrier walls (ratio 14.8) (Gan et al., 2008). The Zn-to-Cu-ratios of 24.9, 34.2, 38.0, and 61.0 are caused by a temporal contamination of samples by a Zn-containing anti-corrosive paint have (Dierkes, 1999).

Table 5

Statistics of Figs. 5 and 6 – mean metal concentrations (dissolved and total) in parking lot (PL), road (RL, RM, and RU), bridge (BR), and highway (HL, HN, and HU) runoff.

| Element | Traffic area category | Dissolved concentrations | | | | | | | | | | Total concentrations | | | | | | | | | |
|---------|-----------------------|--------------------------|--------|----------------|------|--------------|--------------|--------------|------|--------------|------------------|----------------------|--------|----------------|------|--------------|--------------|--------------|--------|------|------------------|
| | | Min | Median | Geometric Mean | Mean | 75th | 90th | 95th | Max | SD | 95% ^a | Min | Median | Geometric Mean | Mean | 75th | 90th | 95th | Max | SD | 95% ^a |
| Pb_21 | PL | 1,0 | 1,7 | 2,5 | 4,3 | 8,0 | 12,0 | ^b | 15,9 | 4,6 | 2,6 | 2,5 | 16,3 | 15,5 | 23,1 | 36,3 | 59,8 | 65,8 | 66,0 | 19,5 | 9,1 |
| Pb_21 | RL | – | – | – | – | – | – | – | – | – | – | 2,0 | 27,0 | 21,5 | 61,5 | 123 | ^b | ^b | 152 | 66,2 | 61,3 |
| Pb_21 | RM | 0,13 | 0,4 | 0,4 | 0,9 | 2,3 | ^b | ^b | 2,8 | 1,3 | 2,0 | 3,7 | 20,3 | 18,3 | 32,3 | 30,5 | ^b | ^b | 136 | 43,1 | 36,0 |
| Pb_21 | RU | 0,5 | 4,0 | 2,7 | 3,9 | 6,5 | ^b | ^b | 7,0 | 2,8 | 4,4 | 6,0 | 44,5 | 38,8 | 79,3 | 71,0 | 331 | ^b | 380 | 112 | 67,4 |
| Pb_21 | BR | 0,13 | 0,14 | 0,4 | 1,4 | ^b | ^b | ^b | 3,8 | 2,1 | 5,3 | 24,0 | 41,7 | 42,5 | 49,9 | 83,7 | ^b | ^b | 92,3 | 32,3 | 51,4 |
| Pb_21 | HL | 0,01 | 1,0 | 0,3 | 1,3 | 2,6 | ^b | ^b | 3,1 | 1,3 | 1,3 | 2,5 | 25,0 | 32,1 | 64,4 | 118 | 212 | ^b | 230 | 73,1 | 49,1 |
| Pb_21 | HN | 12,8 | 12,8 | 12,8 | 12,8 | ^b | ^b | ^b | 12,8 | ^b | ^b | 4,4 | 16,0 | 18,7 | 31,7 | 64,5 | ^b | ^b | 90,0 | 34,1 | 35,8 |
| Pb_21 | HU | 0,8 | 2,1 | 2,3 | 3,0 | 4,9 | ^b | ^b | 7,4 | 2,4 | 2,6 | 1,4 | 12,6 | 16,8 | 33,1 | 39,7 | 81,7 | 155 | 220 | 44,9 | 16,5 |
| Zn | PL | 12,0 | 60,4 | 56,1 | 77,2 | 113 | 157 | 293 | 317 | 69,1 | 30,6 | 39,0 | 178 | 155 | 201 | 295 | 397 | 586 | 620 | 147 | 53,7 |
| Zn | RL | 15,0 | 24,0 | 42,6 | 76,2 | 119 | ^b | ^b | 314 | 98,8 | 76,0 | 25,0 | 149 | 141 | 212 | 272 | 525 | ^b | 940 | 219 | 109 |
| Zn | RM | 7,9 | 31,0 | 41,4 | 68,0 | 73,0 | 226 | ^b | 258 | 77,2 | 42,8 | 23,0 | 274 | 203 | 285 | 384 | 533 | 886 | 1000 | 217 | 89,6 |
| Zn | RU | 51,0 | 78,3 | 97,7 | 113 | 159 | ^b | ^b | 262 | 71,6 | 55,1 | 120 | 351 | 379 | 474 | 556 | 982 | 1805 | 1940 | 397 | 176 |
| Zn | BR | 50,0 | 148 | 191 | 354 | 646 | ^b | ^b | 1226 | 432 | 332 | 77,0 | 498 | 611 | 2231 | 700 | 15,572 | ^b | 19,100 | 5607 | 3766 |
| Zn | HL | 5,0 | 66,5 | 54,3 | 76,9 | 108 | 177 | ^b | 191 | 53,0 | 30,6 | 32,3 | 217 | 207 | 306 | 371 | 642 | 1540 | 1760 | 359 | 155 |
| Zn | HN | 8,6 | 66,8 | 90 | 204 | 453 | ^b | ^b | 577 | 231 | 177 | 52,5 | 216 | 248 | 385 | 388 | 1202 | 1875 | 2210 | 477 | 193 |
| Zn | HU | 11,0 | 50,0 | 77,1 | 217 | 166 | 595 | 1838 | 2118 | 449 | 194 | 21,0 | 213 | 237 | 338 | 414 | 682 | 986 | 2234 | 358 | 103 |
| Ni | PL | 1,8 | 3,0 | 3,2 | 3,8 | 4,4 | 10,2 | ^b | 12,4 | 3,0 | 1,9 | 2,1 | 7,1 | 10,5 | 23,7 | 35,0 | 86,9 | ^b | 145 | 37,0 | 19,7 |
| Ni | RL | 3,1 | 4,7 | 4,5 | 4,7 | ^b | ^b | ^b | 6,3 | 2,2 | 20,2 | 8,4 | 13,9 | 12,5 | 13,1 | ^b | ^b | ^b | 16,9 | 4,3 | 10,7 |
| Ni | RM | 0,5 | 1,0 | 0,9 | 0,9 | ^b | ^b | ^b | 1,3 | 0,4 | 1,0 | 3,8 | 17,0 | 12,7 | 16,3 | 23,0 | ^b | ^b | 35,0 | 11,2 | 11,3 |
| Ni | RU | 9,5 | 10,5 | 10,5 | 10,5 | ^b | ^b | ^b | 11,5 | 1,4 | 12,7 | 4,1 | 9,8 | 13,8 | 20,5 | 40,4 | ^b | ^b | 55,0 | 20,2 | 21,2 |
| Ni | BR | 3,0 | 4,8 | 8,3 | 15,9 | 35,5 | ^b | ^b | 49,0 | 19,5 | 20,5 | 6,0 | 26,7 | 28,0 | 82,0 | 65,0 | ^b | ^b | 458 | 154 | 129 |
| Ni | HL | – | – | – | – | – | – | – | – | – | – | 6,0 | 12,0 | 15,6 | 22,6 | 35,2 | ^b | ^b | 73,0 | 25,3 | 26,5 |
| Ni | HN | 4,0 | 11,0 | 10,6 | 14,0 | ^b | ^b | ^b | 27,0 | 11,8 | 29,3 | 4,0 | 20,0 | 16,1 | 29,3 | 57,0 | ^b | ^b | 83,0 | 30,8 | 23,7 |
| Ni | HU | 1,9 | 9,6 | 9,9 | 14,9 | 28,3 | ^b | ^b | 28,8 | 12,5 | 15,6 | 2,0 | 8,7 | 9,5 | 19,2 | 31,5 | 68,8 | ^b | 93,0 | 25,1 | 12,1 |
| Cu | PL | 2,7 | 9,0 | 9,4 | 11,8 | 13,5 | 28,5 | 34,3 | 35,0 | 8,7 | 3,7 | 5,0 | 19,6 | 23,4 | 40,7 | 56,0 | 116 | 165 | 220 | 49,2 | 15,4 |
| Cu | RL | 3,3 | 11,0 | 14,5 | 20,3 | 37,8 | 53,2 | ^b | 56,0 | 17,9 | 11,4 | 6,0 | 48,0 | 37,5 | 53,7 | 77,4 | 104 | ^b | 180 | 43,9 | 21,2 |
| Cu | RM | 2,7 | 11,2 | 11,2 | 16,0 | 23,0 | 35,0 | ^b | 65,0 | 14,9 | 7,2 | 7,0 | 30,5 | 38,6 | 64,6 | 100 | 175 | 264 | 280 | 71,3 | 31,6 |
| Cu | RU | 6,8 | 23,0 | 22,2 | 26,4 | 39,0 | 55,0 | ^b | 56,8 | 15,9 | 9,6 | 26,0 | 86,9 | 86,9 | 105 | 149 | 197 | 275 | 288 | 66,6 | 29,5 |
| Cu | BR | 16,0 | 23,5 | 24,7 | 26,1 | 31,1 | ^b | ^b | 47,0 | 9,9 | 8,3 | 20,0 | 54,0 | 54,9 | 63,9 | 93,2 | 133 | ^b | 136 | 36,1 | 25,8 |
| Cu | HL | 5,7 | 18,0 | 18,9 | 23,3 | 31,1 | 56,9 | ^b | 64,0 | 16,4 | 9,9 | 13,3 | 48,0 | 49,4 | 60,7 | 91,7 | 124 | 138 | 140 | 38,8 | 17,2 |
| Cu | HN | 4,0 | 28,7 | 25,8 | 34,6 | 51,5 | 88,0 | ^b | 100 | 26,9 | 17,1 | 23,0 | 53,0 | 61,8 | 84,4 | 97,0 | 205 | 394 | 430 | 90,4 | 39,1 |
| Cu | HU | 4,1 | 15,4 | 18,8 | 35,5 | 49,1 | 122 | 144 | 151 | 42,0 | 17,3 | 13,0 | 56,4 | 51,4 | 63,5 | 74,7 | 118 | 145 | 274 | 45,5 | 13,2 |
| Cd | PL | 0,05 | 0,3 | 0,3 | 0,4 | 0,6 | 1,6 | ^b | 2,0 | 0,5 | 0,3 | 0,2 | 0,8 | 0,9 | 3,0 | 2,0 | 5,6 | 36,6 | 40,0 | 8,6 | 3,9 |
| Cd | RL | 0,14 | 0,4 | 0,5 | 0,7 | 1,2 | ^b | ^b | 1,8 | 0,6 | 0,6 | 0,3 | 1,6 | 1,5 | 2,7 | 2,8 | 12,0 | ^b | 13,0 | 3,8 | 2,7 |
| Cd | RM | 0,01 | 0,2 | 0,1 | 0,2 | 0,3 | 0,4 | ^b | 0,5 | 0,1 | 0,1 | 0,06 | 0,6 | 0,7 | 3,2 | 1,3 | 14,1 | ^b | 37,0 | 9,1 | 4,8 |
| Cd | RU | 0,25 | 0,6 | 0,5 | 0,6 | 0,7 | ^b | ^b | 0,9 | 0,2 | 0,2 | 0,67 | 2,0 | 2,7 | 5,6 | 6,5 | 22,0 | ^b | 25,0 | 7,8 | 4,5 |
| Cd | BR | 0,04 | 0,5 | 0,3 | 0,5 | 1,0 | 1,0 | 1,0 | 1,0 | 0,4 | 0,4 | 0,28 | 3,5 | 3,9 | 10,3 | 15,0 | 43,8 | ^b | 47,0 | 14,4 | 10,3 |
| Cd | HL | 0,02 | 0,1 | 0,1 | 0,5 | 0,8 | ^b | ^b | 2,6 | 1,0 | 1,1 | 0,05 | 0,9 | 0,7 | 1,8 | 1,9 | 7,6 | ^b | 9,0 | 2,6 | 1,6 |
| Cd | HN | 0,50 | 1,1 | 1,2 | 1,4 | 1,8 | ^b | ^b | 3,0 | 0,8 | 0,8 | 0,14 | 1,3 | 1,2 | 2,6 | 4,6 | 6,2 | 11,6 | 11,9 | 3,0 | 1,4 |
| Cd | HU | 0,10 | 0,9 | 0,8 | 1,3 | 1,7 | ^b | ^b | 3,8 | 1,2 | 1,1 | 0,06 | 1,7 | 1,6 | 4,1 | 2,4 | 12,5 | 31,3 | 35,0 | 7,9 | 2,9 |
| Cr | PL | 1,0 | 1,5 | 1,6 | 1,7 | 2,2 | 2,7 | ^b | 2,7 | 0,6 | 0,4 | 1,5 | 8,6 | 8,6 | 13,1 | 21,0 | 26,4 | 50,4 | 56,0 | 12,5 | 5,4 |
| Cr | RL | 1,4 | 1,8 | 1,7 | 1,8 | ^b | ^b | ^b | 2,2 | 0,6 | 5,3 | 5,0 | 11,5 | 10,4 | 11,2 | 15,3 | ^b | ^b | 16,0 | 4,3 | 4,5 |
| Cr | RM | 0,6 | 1,2 | 1,0 | 1,2 | ^b | ^b | ^b | 1,8 | 0,8 | 7,6 | 2,0 | 9,9 | 9,0 | 12,0 | 24,2 | ^b | ^b | 24,2 | 8,9 | 8,3 |
| Cr | RU | 3,0 | 3,0 | 3,0 | 3,0 | ^b | ^b | ^b | 3,0 | ^b | ^b | 3,6 | 12,9 | 11,6 | 13,2 | 17,8 | ^b | ^b | 23,0 | 6,4 | 6,7 |
| Cr | BR | 1,3 | 3,2 | 3,3 | 4,0 | 7,1 | ^b | ^b | 7,5 | 2,6 | 2,7 | 7,5 | 11,0 | 26,7 | 83,5 | 140 | ^b | ^b | 381 | 140 | 129 |
| Cr | HL | – | – | – | – | – | – | – | – | – | – | 4,9 | 12,5 | 14,4 | 18,6 | 38,0 | ^b | ^b | 40,4 | 14,4 | 13,3 |
| Cr | HN | 0,5 | 11,3 | 3,3 | 11,3 | ^b | ^b | ^b | 22,1 | 15,3 | 137 | 2,7 | 9,3 | 12,7 | 25,0 | 36,8 | 88,7 | ^b | 89,0 | 33,4 | 23,9 |
| Cr | HU | 1,5 | 3,2 | 3,3 | 4,5 | 8,0 | ^b | ^b | 11,9 | 4,3 | 5,3 | 1,0 | 7,3 | 8,6 | 17,9 | 13,8 | 69,3 | 103 | 105 | 26,8 | 12,5 |

^a 95% confidence errors on the means.

^b No values because of small universe.

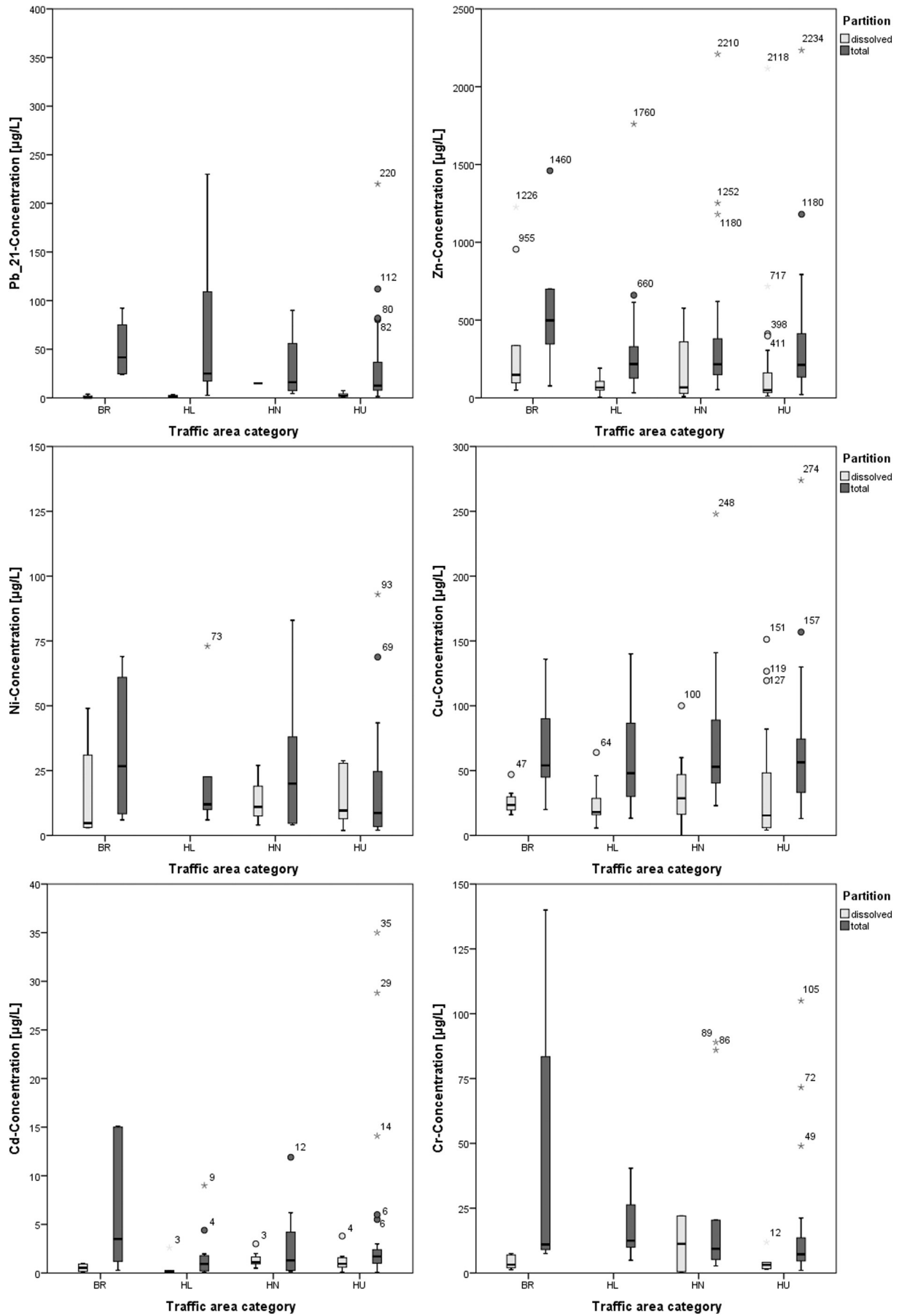


Fig. 6. Mean metal concentrations (d and t) in bridge (BR) and highway (HL, HN, and HU) runoff.

Table 6
Elements that are detected in sampling programs and that are related to traffic or might be used as a tracer for anthropogenic sources.

| Element | Literature (partition) |
|---------|---|
| Co | Dannecker et al. (1990) (p, t); Hares and Ward (1999) (t); Bäckström et al. (2003) (d, t); Wilson (2006) (d, t); Hallberg et al. (2007) (d); Herrera (2007) (*, d, t); Steiner and Goosse (2007) (t); von Ballmoos (2007) (t); Grotehusmann and Kasting (2009) (t) |
| Mn | Ellis and Revitt (1982) (*, d, t); Golwer and Schneider (1983) (t); Harrison and Wilson (1985) (d, t); Dannecker et al. (1990) (p, t); Sansalone and Buchberger (1997a) (d, t); Booth and Leavitt (1999) (d); Brockbank et al. (1999) (d, t); Hares and Ward (1999) (t); Shinya et al. (2000) (t); Rushton (2001) (t); Bäckström et al. (2003) (t); Shinya et al. (2003) (p); Preciado and Li (2005) (d, t); Hallberg et al. (2007) (d); Gnecco et al. (2008) (t); Hatt et al. (2009) (t); McQueen (2010) (t) |
| PGE | Wei and Morrison (1994) (Pt; d, t); Laschka et al. (1996) (Pt; t); Grotehusmann and Kasting (2002) (Pd; d, t; Pt; d, t); Moy et al. (2003) (Pd; t); Hilliges (2007) (Rh; t); Kasting and Grotehusmann (2007) (Pt; d, t); Nadler and Meißner (2007) (Pt; t); Windhofer et al. (2012) (Pt; Rh; t) |
| Sb | Dannecker et al. (1990) (p, t); CH2MHILL (1998) (t); Hares and Ward (1999) (t); Wilson (2006) (d, t); Herrera (2007) (*, t, d); Nadler and Meißner (2007) (t); Stachel et al. (2007) (*, t); Scheiwiller (2008) (t); Grotehusmann and Kasting (2009) (t); Steiner and Goosse (2009) (t) |
| Ti | Bourcier et al. (1980) (t); Golwer and Schneider (1983) (d, t); Fürhacker et al. (2013) (t) |
| W | Bourcier et al. (1980) (t); Bäckström et al. (2003) (d, t) |

^a Literature study.

The explanation for low Zn-to-Cu-ratios related to porous asphalt could only be confirmed by one Zn-to-Cu-ratio with a value of 1.2 (Berbee et al., 1999). For comparison, mean ratios of porous asphalt vary between 1.0 and 3.9 with one exception of a ratio of 8.5 (Stotz and Krauth, 1994), which strengthens the conclusion of Moores et al. (2010). Examples of low median ratios resulting from higher Cu concentrations could not be identified.

In conclusion, the calculation of median Zn-to-Cu-ratios can be used to evaluate datasets to detect possible sample contamination and to emphasize the importance of detailed site descriptions.

4.3. Characteristics of traffic area categories

The data for all traffic areas are subdivided into eight categories, as described in Section 2. The data for five categories, including PL, different roads without HWY, and BR, which are presented for the first time, are shown in detail in Tables 1, 2, and 3.

In Table 1, 35 PL are included, of which two were investigated in two monitoring programs (PL16a + b and PL31a + b). Most PL samples were collected in the USA; some samples were obtained in Europe (Germany, Austria, France, Italy, Poland, and Sweden). Only one PL runoff was collected in each of Australia and South Korea.

Generally, PL have low traffic densities. Thus, even in high-use parking areas such as at supermarkets, vehicle changes only occur up to four times per hour and might not exceed 50 vehicles per day per parking slot (Göbel et al., 2007). However, drip losses, tire and brake wear due to braking and steering activities, and exhaust gas emissions because of acceleration are higher compared with non-congested roads. Therefore, most of the Cu loads in runoff waters from industrial land uses originate from PL (Bannerman et al., 1993; Steuer et al., 1997) and PL runoff are a dominant contributor of total Zn and Cd loads in urban runoff (Steuer et al., 1997). Another factor is the contamination of parked vehicles (Pick et al., 2002). PL maintenance includes regular sweeping and cleaning of drains about three times a year (Grotehusmann and Kasting, 2002).

All types of road categories with an AADT of <5000 (RL, 21 sites), 5000 < AADT < 15,000 (RM, 33 sites), and >15,000 (RU, 26 sites) are presented in Table 2.

For RL, most published data are for US sites, with four sites in Germany and one site each in France, the Netherlands, Australia, and South Korea. The land uses are mostly urban and residential but also rural (RL07–RL10), industrial (RL03 + RL18), non-urban desert (RL15), and non-urban agricultural (RL16). RL mostly includes residential, feeder, and smaller collector streets, often with sidewalks and sometimes with side parking zones and curbs. Urban sites have single-family houses and two- to six-story buildings nearby. For RM, 14 datasets have been published for US sites, eight for German sites, and a total of twelve for sites in eight other countries. The land uses are mostly urban and residential but also include rural (RM11, RM13, RM16–18) and non-urban agricultural (RM23) uses. RM include collector roads, residential driveways, and other medium-traffic major roads with mostly two lanes, curbs, and limited on-street parking. RL and RM can be maintained by daily sweeping of street gutters and pressurized washing of sidewalks and gutters several times a week (Gromaire-Mertz et al., 1999). For porous asphalts pressurized water spraying and suction to prevent clogging is used (Legret et al., 1996; Dierkes et al., 1999). For RU, ten sites are located in Germany, three in Poland, three in the USA, and a total of ten sites in six other countries. All sites are highly trafficked arterial roads with almost no hard shoulders. The number of lanes varies between two and five for urban sites, which sometimes have medians, and only RU08 is a two-lane rural road with hard shoulders.

All BR are summarized in Table 3. 21 of the 39 sites are from a huge Australian study by Drapper et al. (2000), and 13 sites are located in the USA. One site in France was monitored with original asphalt surface and after renovation with porous asphalt (BR01a + b), one site in Sweden

was selected to evaluate seasonal variations (summer/winter; BR02a + b), one site in Canada was monitored in two different decades (BR16a + b), and two sites are located in China. BR are monitored because contaminant sources are assumed to be exclusively traffic-related (Wilson, 2006), minimal infrastructure modifications are required for sampling (Drapper et al., 2000), and sites are better protected against vandalism (Marsalek et al., 1997). BR decks are intensively maintained (Wu et al., 1998).

All HWY sites are classified depending on the AADT according to Strecker et al. (1990). For HWY with an AADT of >30,000, all sites are distinguished between urban (HU, 64 sites) and non-urban (HN, 33 sites) land uses, which have been proposed to be the only factor having a statistically significant influence on pollutant concentrations for US datasets (Strecker et al., 1990; Kayhanian et al., 2007; Nason et al., 2012). The dataset also includes 35 HL sites. Data are available for five of six continents (no data for South America); 47 of the 132 sites are located in the USA, 24 in Germany, and eleven in each of Switzerland and the United Kingdom. The main difference between RU and HWY in European countries is that the latter mostly have hard shoulders and roads may have traffic signals in contrast to HWY.

4.4. Runoff concentrations of traffic area categories

All published mean metal concentrations (d and t) in PL, RL, RM, and RU runoff for Pb (n = 74), Zn (n = 154), Ni (n = 55), Cu (n = 164), Cd (n = 103), and Cr (n = 64) are plotted in Fig. 5 (Table 5). As datasets are not plotted because of fewer available data (n = 24 with 18 for PL). In most cases, the overall median concentrations for PL are lower than those for roads. Differences in pollution are related to the type of PL use: Employee PL (mostly once-in and once-out per day usage, PL31) are less polluted than highly frequented car PL (supermarket, PL05) and PL for trucks are higher loaded for almost all parameters in comparison to car PL (Grotehusmann and Kasting, 2002). RU is the road category with the highest runoff concentrations for all metals except Ni. Zn has the highest total runoff concentration, followed by Cu, Pb, Ni, Cr, and Cd. The highest dissolved concentrations have Zn and Cu, followed by Ni, Pb, Cr, and Cd. Thus, Pb and Cr are mostly in the particulate phase, whereas Ni and Cd are almost dissolved. For PL, median values for As (2.1 µg/L t; 1.1 µg/L d) are higher than those for Cd (0.8 µg/L t; 0.3 µg/L d). The total concentrations of PL08 (only one event analyzed in contrast to the dissolved values) and the very high values of RL21 are not plotted in Fig. 5.

In Fig. 6, all published mean metal concentrations (d and t) in BR, HL, HN, and HU runoff are plotted separately (Table 5). For BR, the number of datasets varies between seven for Pb and 20 for Zn. For HWY runoff, much more data are available for Pb (n = 61), Zn (n = 144), Ni (n = 42), Cu (n = 142), Cd (n = 83), and Cr (n = 44). For BR, the order of total and dissolved concentrations is equal to the ones presented in Fig. 5 but the Zn concentrations of BR runoff are higher compared with all other categories (cf. Sections 3.1, 4.2, and 4.3). Median values of total Cr concentrations (n = 13) are very high compared with all other datasets. This might be related to the generally very high total concentrations of BR11 (19,100 µg/L Zn, 1860 µg/L Pb, 140 µg/L Cr) and to specific conditions in China (BR38, BR39; 26.8–381 µg/L Cr). From a comparison of two monitoring programs of a French BR with asphalt and after renovation using porous asphalt (BR01a + b), it can be concluded that heavy metal runoff concentrations of porous asphalt are markedly lower and the type of BR deck surface does not affect heavy metal partitioning. A comparison of the effects of winter and summer on the runoff concentrations of a BR in Sweden (BR02a + b) showed higher mean concentrations during winter, and a comparison of two Canadian BR runoff (BR16a + b) indicated a sharp historical decrease in Zn and Pb concentrations and an increase in Cu and Ni concentrations during the 1990s.

For HWY, no significant difference among the three categories HL, HN, and HU could be detected. This contrasts with the conclusions

obtained using US datasets that HL and HN have lower runoff concentrations compared with HU (Strecker et al., 1990). For comparison with this dataset, note that in this evaluation, all HN of US sites are in category HL. Thus, in HN, only European and two New Zealand sites are included. According to Strecker et al. (1990), the concentrations of HL should be lower than those of HN and HU. However, by considering worldwide data, the correlation among HWY runoff concentrations, AADT, and land use could not be verified. Thus, other site-specific aspects such as hard shoulders, median strips, noise barriers, and grade of congestion might be better indicators to characterize HWY runoff concentrations. This is confirmed by a comparison of RU data with HWY datasets. For RU, nearly all overall heavy metal median concentrations are higher in comparison to each of the three HWY categories. For special site conditions such as for RM03, the runoff concentrations of an urban road can be higher than most measurements of HWY because of braking and acceleration activities at traffic signals. The results of an evaluation of runoff concentrations of a HWY construction site not yet in use with two other sites on the same HWY showed that Zn runoff concentrations of the construction site are sometimes higher compared with the other two sites and that Cu and Pb concentrations of the construction site are lower (Mangani et al., 2005). This demonstrates the importance of site-specific factors for HWY runoff.

An analysis was performed to examine the hypothesis that a lower pH of the runoff corresponds to a higher proportion of dissolved metals (Mikkelsen et al., 1994; Sansalone and Buchberger, 1997b; Genç-Fuhrman et al., 2007). The dissolved concentrations in runoff samples are influenced by the capacity for rainwater and pH-modified runoff (cf. Section 4.6) to dissolve metals from a surface, which is influenced by kinetic factors, and by the rate of dissolution, which increases as pH decreases. An acidic pH of one entire event confirms the ionic form for Cu as most relevant (Gnecco et al., 2008). However, an evaluation of datasets for the percentages of Cu and Zn dissolved at the prevailing runoff pH could not confirm the speciation proposed by Genç-Fuhrman et al. (2007). The dissolved fraction of Cu in runoff from different sites was analyzed as 52% at a pH of 6.2 (Caltrans, 2003), 81% at a pH of 6.7 (Lau et al., 2009), and 72% at a pH of 8.0 (Mangani et al., 2005). The dissolved Cu fraction varies between 1% at a pH of 7.6 (Pitt et al., 1995) and 93% at a pH of 7.2 (Golwer and Schneider, 1983). For Zn, the dissolved fraction varies between 2% at a pH of 8.1 (Mangani et al., 2005), 11% at a pH of 7.1 (Lange et al., 2003), and 91% at a pH of 6.7 (Lau et al., 2009). Thus, nearly no decrease of the dissolved fraction of Zn and Cu can be found in the pH range between 6.2 and 8.1, and the hypothesis could not be confirmed. Potential reasons include the likelihood that dissolution is kinetically limited, the relative independence of dissolved and particulate sources, or differences in the sample preparation and analysis.

The results from all datasets show that Cu and Zn mostly exhibited intermediate behavior (Harrison and Wilson, 1985; Dean et al., 2005; Herrera, 2007; Kayhanian et al., 2007) but sometimes, they were more dissolved (Timperley et al., 2005; Wilson, 2006; Hilliges et al., 2013). The concentrations of Pb and Cr are mostly in the particulate phase; this was confirmed in several individual monitoring programs (Harrison and Wilson, 1985; Shinya et al., 2000; Timperley et al., 2005; Herrera, 2007; Kayhanian et al., 2007; Kayhanian, 2012; Hilliges et al., 2013). Ni (Shinya et al., 2000) and Cd (Wilson, 2006) are mainly in the dissolved form, although they are sometimes equally partitioned between the two phases (Harrison and Wilson, 1985; Dean et al., 2005; Kayhanian et al., 2007). The particulate fraction of heavy metals might be higher for snowmelt events than rain events (Westerlund et al., 2003). Bäckström et al. (2003) confirmed this for Pb and Zn, although other metals remained at a constant fraction during winter, and Hallberg et al. (2007) determined higher dissolved fractions of Cd, Cr, and Ni in winter compared to summer, whereas no significant differences occurred for Cu, Pb, and Zn. Helmreich et al. (2010) concluded that the fractionation of the heavy metals was not affected by seasonal variations but remarkable fluctuations were observed between

different rain events with dissolved fractions greater than 90%. Similar results were confirmed by Dauber et al. (1979). However, Dean et al. (2005) stated that the partitioning of Pb was unaffected by flow rate.

4.5. Other elements

Other elements that are detected in sampling programs and that are related to traffic or might be used as a tracer for anthropogenic sources are listed in Table 6. As tracers, Co and W are potentially suitable for roads and traffic because of their low natural concentrations in environmental waters (Bäckström et al., 2003). Nadler and Meißner (1999) and Huang et al. (1994) suggest that Sb should be used as a marker, too.

Co is mainly used in steel and paints (Makepeace et al., 1995) and in diesel (Roeva et al., 1996). Steiner et al. (2006) concluded that traffic is not the main source for Co in soils, whereas Ward (1990) observed increased levels of Co in soils because of traffic activities. Bäckström et al. (2003) clearly demonstrated that roads act as a source for Co. Total mean concentration of Co varies between 1.5 µg/L (Grotehusmann and Kasting, 2009) and 13.3 µg/L (Bäckström et al., 2003) and dissolved mean concentrations are between 0.1 µg/L (Dannecker et al., 1990) and 15.1 µg/L (Hallberg et al., 2007); dissolved fractions vary between 4.1% (Dannecker et al., 1990) and 67% (Bäckström et al., 2003). The highest mass of Co (around 94%) in runoff was transported during winter at one site in Sweden (Bäckström et al., 2003).

Bourcier et al. (1980) measured W, which is used in tire studs, in concentrations between non-detectable (2.5 mg/L) and 25 mg/L in road runoff (completely in the particulate phase) and did not detect it in precipitation and dust samples. Bäckström et al. (2003) measured W at two sites in summer and winter and found that the particulate fraction is low and the concentrations increased by a factor of ten during winter (5.8–9.2 µg/L).

Sb is a very toxic element, and the main traffic-related sources are brake pads (Von Uexküll et al., 2005). Concentrations of Sb have been presented in ten publications (cf. Table 6). Total mean Sb concentrations are between 0.8 µg/L (Dannecker et al., 1990) and 10.7 µg/L (CH2MHILL, 1998). Dissolved concentrations were analyzed at three sites and vary between 1.2 µg/L (Dannecker et al., 1990) and 5.05 µg/L (Wilson, 2006).

Anthropogenic sources of Mn are moving engine parts (Ball et al., 1998; Drapper et al., 2000), gasoline additives such as methylcyclopentadienyl manganese tricarbonyl (MMT) (Young et al., 1996; Preciado and Li, 2005), brake wear (Ward, 1990; Sansalone et al., 1996), and tire wear (Hares and Ward, 1999). Values of Mn concentrations in runoff from different sites have been presented in 17 publications (cf. Table 6). Shinya et al. (2000) reported that Mn is mainly particle-bound, whereas Sansalone and Buchberger (1997a) and Harrison and Wilson (1985) reported that it is mainly dissolved. Total mean concentrations of Mn vary between 11.1 µg/L (Rushton, 2001) and 659 µg/L (Gnecco et al., 2008) (both PL), and dissolved mean concentrations are between 9 µg/L (Booth and Leavitt, 1999) and 255 µg/L (Sansalone and Buchberger, 1997a). Ward (1990) measured elevated levels of Mn in soils with increased traffic density.

Of the PGE only Rh, Pd, and Pt are related to traffic sources. Since the end of the 1980s, these three elements have been primarily used as catalytic converters and the number of automobiles equipped with these converters has increased sharply (Wei and Morrison, 1994). A three-way catalytic converter contains a total of 2–3 g of Rh, Pd, and Pt (Geiger-Kaiser and Jäger, 2005). Fritsche and Meisel (2004) determined the distribution of PGE in soils along major Austrian HWY and concluded that Rh, Pd, and Pt significantly exceeded natural background values. In road runoff, Rh was only detected by Hilliges (2007) at a concentration of 1.14 µg/L and by Windhofer et al. (2012) at a concentration of 0.08 µg/L. Pt concentrations vary between 0.0065 µg/L (Wei and Morrison, 1994) and 24 µg/L (Moy and Crabtree, 2003), and Pd concentrations are between 0.34 µg/L (Moy et al., 2003) and 8 µg/L (Grotehusmann and Kasting, 2002). The dissolved phases of Pd and Pt are high (65%–81%,

Grotehusmann and Kasting, 2002). According to Laschka et al. (1996), Pt runoff concentrations depend on ADP, rain intensity, and rain duration.

Ti was only measured in three studies, and its main source is the wear of white road marking paint containing titanium dioxide as pigment (Makepeace et al., 1995), which is often renewed once a year (Golwer and Schneider, 1983). Another source is tire studs (Bourcier et al., 1980). Ti was not measured in precipitation samples by Bourcier et al. (1980) but was measured in road runoff with a mean concentration of 12.7 mg/L. Golwer and Schneider (1983) detected that Ti was mostly particulate and only 26 µg/L was dissolved. Fürhacker et al. (2013) measured Ti in one HU runoff sample with a total concentration of 630 µg/L.

4.6. Correlation between pH and type of surface

The partitioning of metals in runoff is strongly influenced by the pH of the rainfall, the solids present, the solubility of all substances, the pavement residence time, and the type of surface (Prestes et al., 2006). For asphalt, no significant level of alkalinity is provided to neutralize the pH of rainfall (Sansalone and Buchberger, 1997a), whereas a concrete pavement provides alkalinity to raise the runoff pH more than that of an asphalt pavement (Dierkes, 1999). Therefore, concrete surfaces should have a stronger influence on metal partitioning to the particulate phase than asphalt surfaces.

The evaluation of pH for 63 asphalt, 13 asphalt and concrete, 21 concrete, and four porous asphalt surfaces cannot provide significant differences. The median pH of asphalt surfaces was the lowest (6.8) and varied between 5.4 (Driscoll et al., 1990) and 8.2 (Zhang et al., 2013). A combination of asphalt and concrete surfaces had a median pH of 6.9 and varied between 6.4 (Lau et al., 2009) and 7.5 (Grotehusmann and Kasting, 2009). For concrete surfaces, a mean value of 7.2 was calculated, which is 0.4 units higher than the value for asphalt. The values varied between 5.6 (Driscoll et al., 1990) and 7.9 (Geiger-Kaiser and Jäger, 2005). For porous asphalts, the median value is 7.5.

The non-significant evaluation results of all datasets can be explained by two factors. First, the pavement residence times vary between each monitored site and samples were sometimes collected from a concrete channel that would have had an influence on the pH (Dierkes, 1999). Second, the pH of the rain itself can be different. Two-thirds of the measurements with concrete surfaces were conducted in the 1980s, when rain was more acidic. Conversely, the pH of the runoff of a concrete BR deck increased slightly with increasing flow rates (Dean et al., 2005). Drapper et al. (2000) compared asphalt and concrete BR decks and did not observe any variance in the pH or runoff concentrations. Aspects that might have an effect on porous asphalts have been described by Muschack (1990), and a literature review was provided by Barrett et al. (2006).

4.7. Continental trends

The runoff concentrations of Pb, Zn, and Cu that occur in the highest concentrations compared with other heavy metals were evaluated separately for each continent for which more than five datasets were available (i.e., Asia, Australia, Europe, and North America). For Pb, the recent concentrations are very low and the older, higher concentrations correlate well with the phase-out of leaded gasoline for all continents (cf. Section 4.1). The overall median Pb values for Europe and North America are approximately 30 µg/L and 15 µg/L for the 21st century. For Asia, both Pb values are approximately 50 µg/L, which correlates with the later Pb-phase out in Asia compared with Europe and North America (cf. Section 4.1). The Australian overall median Pb concentration is approximately five times higher than those of Europe and North America for the complete dataset and approximately three times for the dataset of the 21st century. The same results were obtained by Kayhanian et al. (2012) for HWY runoff, whereas the Asian HWY values consist out of one extreme value (Maniquiz et al., 2010). For Cu,

the overall median concentrations are approximately 30 µg/L for North America and approximately twice as much for Asia, Australia, and Europe. For Zn, the overall median concentrations are approximately 170 µg/L for North America, 250 µg/L for Europe, 300 µg/L for Australia, and 620 µg/L for Asia. For Asia, it must be considered that fewer data are available compared with the other three continents and the data are mostly for heavily loaded traffic areas. In conclusion, the median values in North America are the lowest for Pb, Zn, and Cu, and the variance of Pb and Zn runoff concentrations among the continents is higher than the variance of Cu.

4.8. Planning stormwater treatment plants

Several factors concerning the occurrence and partitioning of heavy metals must be considered when planning stormwater treatment plants because the differentiation between the dissolved and particulate phases is of great concern for a successful treatment of stormwater to prevent negative effects on groundwater and surface water. As not all metals are present predominantly in the particulate phase (Cd, Ni, Zn, and Cu; cf. Section 4.4), an additional priority should be set for the reduction of dissolved metal concentrations in runoff treatment programs (Tiefenthaler et al., 2001). Because dissolved metals cannot be removed mechanically with solids (e.g., by sedimentation and filtration), a second treatment step (e.g., filter media) is necessary to reduce higher metal runoff concentrations (Hilliges et al., 2007; Steiner and Goosse, 2008; Minervini, 2010; Kayhanian et al., 2012). Maniquiz-Redillas and Kim (2014) investigated the partitioning of heavy metals in runoff and in the effluent of a stormwater treatment plant (manufactured eco-biofilter) and concluded that the fractionation of heavy metals plays an important role on the performance of the system and a reduction of dissolved metal concentrations is necessary. It is recommended that not only the initial runoff or the first 25% of the total runoff should be treated but the entire water volume (Flint and Davis, 2007; Maniquiz-Redillas and Kim, 2014). In addition, the individual site and climatic influencing factors (cf. Section 3.1) are relevant for the construction and operation of treatment plants to prevent negative toxic effects on ecosystems (Schueler, 2000; Nason et al., 2012; Dierkes et al., 2015). As discussed in Section 4.3, the treatment of runoff from large PL should be considered for an effective control of heavy metal loads. Furthermore, a proper control of BR runoff, runoff of congested streets, and arterial roads with traffic signals becomes a concern. A better knowledge of the historical trends (cf. Section 4.1) and seasonal variations of heavy metal runoff concentrations (cf. Section 4.4) can be used for optimization of treatment processes (Hallberg et al., 2007).

5. Conclusions

The conclusions of this review are summarized as follows:

- Trace element pollutant loads determined at a given site depend on the unique subset of site-specific (surrounding land use characteristics, traffic area site data, operational characteristics, and climatic factors) and method-specific factors (sample collection, preparation, analysis, and calculation). In literature, inconsistent conclusions have been obtained regarding how specific variables affect traffic area runoff. Thus, a combination of several interacting factors results in heavy metal runoff pollution, and these factors should be described in detail for each monitoring site. These factors should also be implemented in databases to allow further specifications in future.
- The calculation of median Zn-to-Cu-ratios can be used to detect possible sample contamination and relevant site-specific factors.
- Historical trends for Pb show a sharp decrease during the last decades, consistent with the phase-out of leaded gasoline; no historical trends were detected for Zn.
- The heavy metal runoff concentrations of PL differ widely according to their use (e.g., employee, supermarket, rest areas for trucks). BR deck runoff can contain high Zn concentrations from safety fences and

galvanizing elements. Roads with more than 5000 vehicles per day are often more polluted than HWY because of other site-specific factors such as braking and acceleration at traffic signals. Worldwide HWY runoff concentrations are not significantly influenced by AADT and urban/non-urban land uses.

- Pb and Cr are mostly particle-bound, while Zn, Cu, Ni, and Cd occur at a higher fraction in the dissolved phase. Therefore, the treatment of runoff waters containing dissolved heavy metal pollutants by stormwater treatment strategies is important.
- Further traffic-related elements are Mn, Rh, Pd, Pt, and Ti. As tracers, Co, W, and Sb, which have anthropogenic sources, are potentially suitable for traffic areas.

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