

The Effects of Urbanization on Runoff Pollutant Concentrations, Loadings and Their Seasonal Patterns Under Cold Climate

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Abstract The effects of urbanization on runoff pollutant concentrations and pollutant loading were studied in three urban catchments of varying imperviousness and land use type in the city of Lahti, Finland. Imperviousness of the catchments were 19 % (“Low”), 62 % (“Intermediate”) and 89 % (“High”). During the 2-year study period, runoff quantity was measured continuously and samples were taken for water quality analysis. Besides imperviousness, land use type strongly affected pollutant concentrations: differences in total phosphorus (tot-P), Al, Cr, Zn and Pb concentrations were observed especially between the city centre catchments (High and Intermediate) and the residential catchment (Low), while total suspended solids (TSS), total nitrogen (tot-N), Mn, Co, Ni and Cu concentrations increased with increasing imperviousness. As for pollutant loads, imperviousness was strongly related to TSS, tot-P, Al, Mn, Zn, Cr, Co, Ni and Cu export. The effects of urbanization on runoff quality were season dependent: urbanization increased runoff volumes and, hence, pollutant loads, especially during warm seasons. Still, highest pollutant export in the catchments occurred

during spring. Nevertheless, the warm period produced comparable loads to spring at the city centre catchments. Pollutant concentrations, especially in the city centre catchments, exceeded thresholds set for surface waters, indicating a need for runoff treatment in water quality protection.

Keywords Heavy metals · Imperviousness · Land use type · Nutrients · Total organic carbon · Total suspended solids

1 Introduction

Urban runoff is considered one of the greatest sources of diffuse pollution of surface waters worldwide (Arnold and Gibbons 1996; Burton and Pitt 2002; Kim et al. 2003). Among the most common and abundant pollutants in urban runoff are nutrients in various forms, suspended solids and heavy metals (Pitt et al. 1999; Mitchell 2001; Moy et al. 2003; Göbel et al. 2006; Ellis and Mitchell 2006). The level of urbanization, such as the degree of imperviousness, is known to directly relate to runoff pollutant concentrations and their loadings (Schueler 1994; Hatt et al. 2004; Dougherty et al. 2006). The type of pollutants and their discharge rates also depend on the type of land use and land use activities, such as traffic levels (Mitchell 2001; Moy et al. 2003; Ellis and Mitchell 2006). In Finland and various other countries, untreated urban runoff is conveyed to nearby surface waters via separate sewer systems, thereby causing acute and chronic effects, such as eutrophication, water quality degradation, health risks and

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habitat loss (Harremoës 1988; Davis et al. 2001; Burton and Pitt 2002).

Most of the urban runoff studies that focus on runoff quantity and quality have been conducted in warm and temperate climates where the generation of pollutant loads is determined by rainfall-induced runoff. In cold climates, with frozen soils and snowfall during winter, hydrological cycles are strongly affected by seasonal variation in temperature and precipitation (Buttle 1990; Brezonik and Stadelmann 2002; Matheussen 2004; Helmreich et al. 2010; Valtanen et al. 2014). In relatively undisturbed rural sites in cold climates, runoff volumes and pollutant loads have two peaks per year—spring and autumn (Gottschalk et al. 1979; Laurén et al. 2005). However, in urbanized areas, it was recently shown that highly constructed catchment generates proportionally much more runoff during warm seasons than during cold ones (Valtanen et al. 2014). Although studies carried out in road areas and in urbanized catchments in cold climates have shown seasonal patterns of runoff generation and pollutant export, they have also shown considerable site-specific differences in both urban runoff quantity and quality (Melanen 1981; Brezonik and Stadelmann 2002; Bäckström et al. 2003; Semádeni-Davies and Titus 2003; Dougherty 2004; Westerlund and Viklander 2006; Hallberg 2007; Helmreich et al. 2010; Sillanpää 2013). Consequently, evidence on the catchment-scale generation of various types of urban-derived pollutants and their transport throughout the year are still meagre (Semádeni-Davies and Titus 2003). Knowledge on pollutant load generation is particularly scarce from highly impervious city centres, which presumably generate the highest loads in urban settings. In previous studies, monitoring have often lasted for less than a year (Bäckström et al. 2003; Westerlund and Viklander 2006; Hallberg 2007; Helmreich et al. 2010) and as such, data are needed from longer measurement periods to cover large annual variation in pollutant export (Hatt et al. 2004; Groffman et al. 2004; Sillanpää 2013).

Our main objective was to improve knowledge on the impacts of urbanization (defined as percentage of imperviousness and land use type) and season on urban runoff quality in cold climate. We hypothesized that urbanization increases runoff pollutant concentrations and loadings. We also hypothesized that, due to strong seasonality experienced in cold climate, the effects of urbanization on stormwater quality are season dependent. Based on our previous research on runoff

generation (Valtanen et al. 2014), we hypothesized that the greatest differences between different levels of urbanization take place during warm months. Finally, we hypothesized that urbanization alters the seasonality of pollutant loadings by shifting the largest seasonal loads from cold to warm periods of the year, mirroring patterns observed for the generation of runoff. This study is part of a long-term study programme that investigates runoff in urban areas (see Valtanen et al. 2014).

2 Methodology

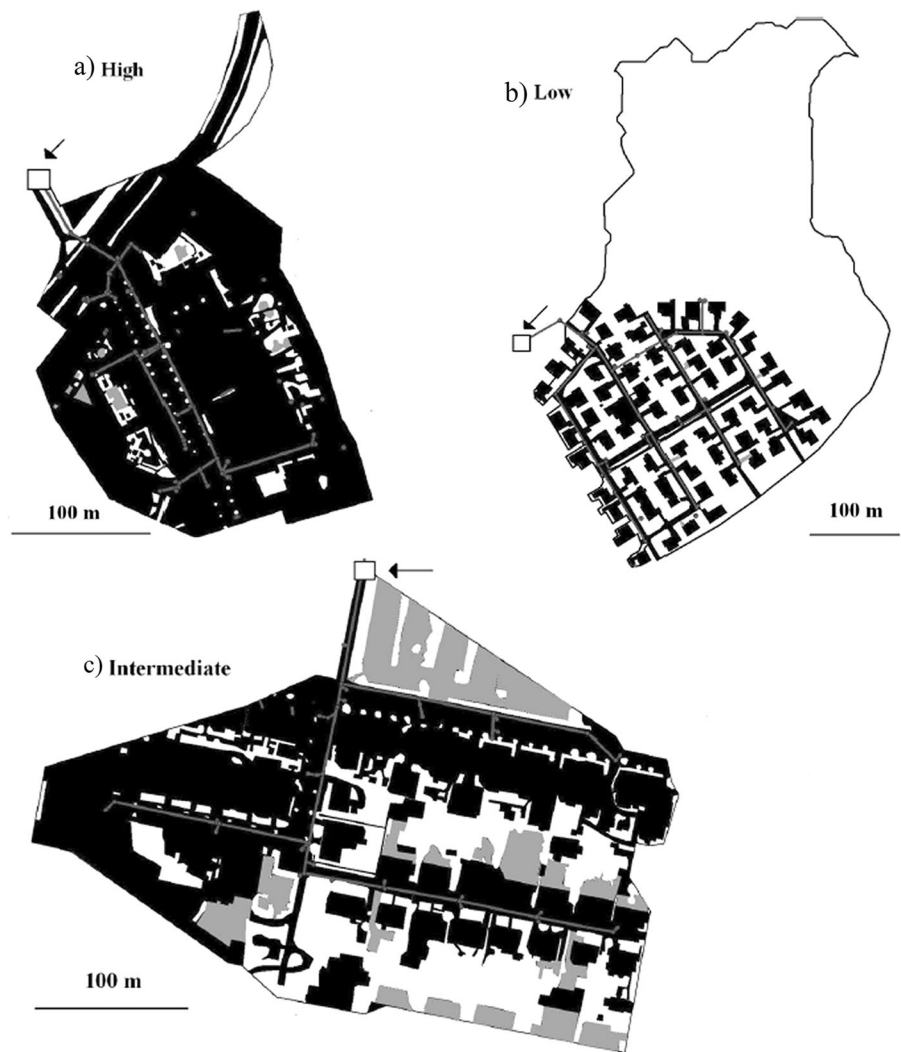
2.1 Study Catchments

The study was conducted in the city of Lahti (60°59'00" N, 25°39'20", population 102,000 in 2011), southern Finland, where the winter period lasts for 135–145 days and the summer period for 110–120 days with an annual mean precipitation of 633 mm (Kersalo and Pirinen 2009). Three catchments were selected based on imperviousness and land use type: Taapelipolku (total impervious area (TIA) 89 %, hereafter referred to as “High”), Ainonpolku (TIA 62 %, “Intermediate”) and Kilpiäinen (TIA 19 %, “Low”) (Fig. 1). The High and Intermediate catchments were located in the city centre characterized by commercial and residential apartment buildings, having a population density of ca. 11,000 and 7,000 inhabitants/km², respectively (The city of Lahti 2013). The Low catchment was situated in a residential area of single-family houses with ca. 1,400 inhabitants/km². At the High and Intermediate catchments, respectively, 80 and 30 % of the roads have traffic densities typical of city centres (public transport and commutation) and the rest of the streets are walkways and mixed-use streets of low traffic density. At the Low catchment, traffic was low density, with mixed-use streets and walkways. The High and Intermediate catchments represent one land use type, while the Low catchment another. A more detailed description of the study catchments, weather conditions and runoff volumes during the study years can be found in Valtanen et al. (2014).

2.2 Measurements and Sampling

Measurements took place from 15 October 2008 until end of August 2010. Each catchment included a measurement station that was equipped with an automatic water sampler (ISCO 3700) and an ultrasonic flow

Fig. 1 Maps of the three study catchments: **a** High, **b** Low and **c** Intermediate (modified from Valtanen et al. 2014). *Black areas* indicate impermeable surfaces (roof tops present 40 % at the High and Intermediate catchment and 50 % at the Low catchment). *Light grey areas* indicate gravel surfaces. *White areas* indicate urban green areas. *Dark grey lines* indicate stormwater pipe lines. *Black arrows* point to the locations of the measuring stations (*white squares*)



meter (Nivus PCM4) placed inside a stormwater pipe. At the Intermediate and the Low catchments, precipitation was measured with a tipping bucket rain gauge (Rainew 111) at 0.2-mm resolution. Precipitation data for the High catchment were obtained from the Intermediate catchment, located 1 km away. Both flow (l/s) and precipitation (mm) were measured continuously at 1-min intervals.

Flow-proportional samples were taken at irregular time intervals based on the accumulated runoff volume: sampling densities varied between 10 and 200 m³ depending on the season and on upcoming predicted precipitation. Three to 51 samples/event were taken during rainfall-runoff events (runoff volumes varying 0.1–16 mm) and during snowmelt and rain-on-snow events (runoff volumes varying 0.1–145 mm). From the

samples, total nitrogen (tot-N), total phosphorus (tot-P), heavy metals (Cr, Mn, Co, Ni, Zn, Cu, Pb, Al), total organic carbon (TOC) and total suspended solids (TSS) were determined.

2.3 Runoff Sample Analysis

Tot-N, tot-P and TOC samples were stored at -18°C in plastic bottles before analysis. The concentration of tot-P was analysed using the Lachat instruments QuickChem 8000 (method 10-155-01-1-Q with oxidation SFS 3026; samples taken in 2009) or the Shimadzu UV-2401PC spectrophotometer (SFS 3026:1986; samples taken in 2010). The concentration of tot-N was analysed using the Lachat QuickChem 8000 (method 10-107-04-1-I with oxidation SFS 3026) or the modified

method of Hioki and McLaren (2008) with oxidation (SFS 3031: 1990). In the latter, the column Waters IC-Pak Anion HC (150 mm, 4.6-mm diameter), the eluent 0.04 M NaCl (2.34 g/l) and UV absorbance at 225 nm were used in the analysis. The two methods gave similar concentrations for tot-N. The concentration of TOC was measured using the Apollo 9000hs TOC Analyser applying the SFS-EN 1484 method.

Samples for total metals were fixed with 65 % nitric acid (1:100) and stored at 5 °C in plastic bottles before analysis. Dissolved metal samples were filtrated using glass microfiber GF/C filters before fixation. Both total and dissolved metals were analysed using the Perkin Elmer Elan 6000 ICP-MS with the modified EPA method 2008.8, revision 5.4. Before analysis, total metal samples were diluted depending on the concentration of metals in a sample and oxidized with 65 % nitric acid (1:5) in an autoclave (121 °C, 1 bar, 30 min). Indium was added to the samples as an internal standard.

TSS were analysed immediately after the samples were taken using the ESS Method 340.2, except during the winter of 2009 when samples were first stored at -18 °C in plastic bottles.

2.4 Data Analysis

Pollutant loads generated at each catchment were calculated using total runoff volumes previously presented in Valtanen et al. (2014). For each of the sampled runoff events, event mass loads (EML [kg]) was calculated as the sum of pollutant mass loads determined by multiplying each discrete concentration within an event with its corresponding runoff volume, a method described in Huber (1993) and Charbeneau and Barrett (1998). After this, event mean concentration (EMC [mg/l, µg/l]) was defined as the ratio of EML to event runoff volume.

Seasonal pollutant loads were calculated for 3-month periods representing winter (December to February), spring (March to May), summer (June to August) and autumn (September to November). The “warm period” consists of summer and autumn months, and the “cold period” refers to winter and spring months. Because the measurements terminated in August 2010, the two study years represent data collected from December 2008 to November 2009 and from September 2009 to August 2010. Seasonal loads were calculated by summing the measured EMLs and the loads between the sampled events. To estimate mass load between the sampled events, runoff volumes that occurred before

and after the sampled event were divided by two and then multiplied by the EMC of the sampled event. The relationship between total runoff (per season) and seasonal pollutant loading and mean seasonal EMC was tested using Spearman correlation (PASW Statistics programme 18.0).

3 Results

3.1 Runoff Pollutant Concentrations

3.1.1 Total Nitrogen and Total Phosphorus

Concentrations of tot-N in runoff increased with increasing imperviousness (Fig. 2, Table 1). In the High catchment, median EMC of tot-N (2,200 µg/l) was 40 and 70 % higher than in the Intermediate and Low catchment, respectively. For tot-P, the city centre catchments had equal median concentrations (100 µg/l), with EMCs twice as high as in the Low catchment.

For nutrients, the biggest differences in median EMCs between the study catchments were observed during cold periods (Table 1): tot-N and tot-P concentrations were, respectively, approximately 40 and 80 % higher at the city centres than in the Low catchment. Median tot-P EMCs were twofold higher during cold periods in the city centre catchments (High and Intermediate), while median EMCs in the Low catchment were twice as high during warm periods than during cold periods. For tot-N, twofold higher median EMCs were observed during cold periods in all catchments.

3.1.2 Total Suspended Solids

The concentration of TSS was affected by imperviousness (Fig. 2, Table 1): median EMC in the High catchment (190 µg/l) was 50 and 90 % higher than in the Intermediate and Low catchments, respectively (Fig. 2).

Fig. 2 Event mean concentrations (EMC) of total nitrogen and total phosphorus, total suspended solids and selected heavy metals calculated for the entire study period and of total organic carbon for the first study year at the three study catchments. High, $n=65$; Intermediate, $n=52$; Low, $n=30$. For TOC High, $n=40$; Intermediate, $n=22$; Low $n=14$. Box plots represent medians (horizontal lines), quartiles (white and grey boxes), minimums and maximums (horizontal lines) and means (black dots). Half of the concentrations for total Cr, Co and Cu and most of the concentrations for Pb and Ni were below the reliable quantitation value. Note the logarithmic scale of y-axis

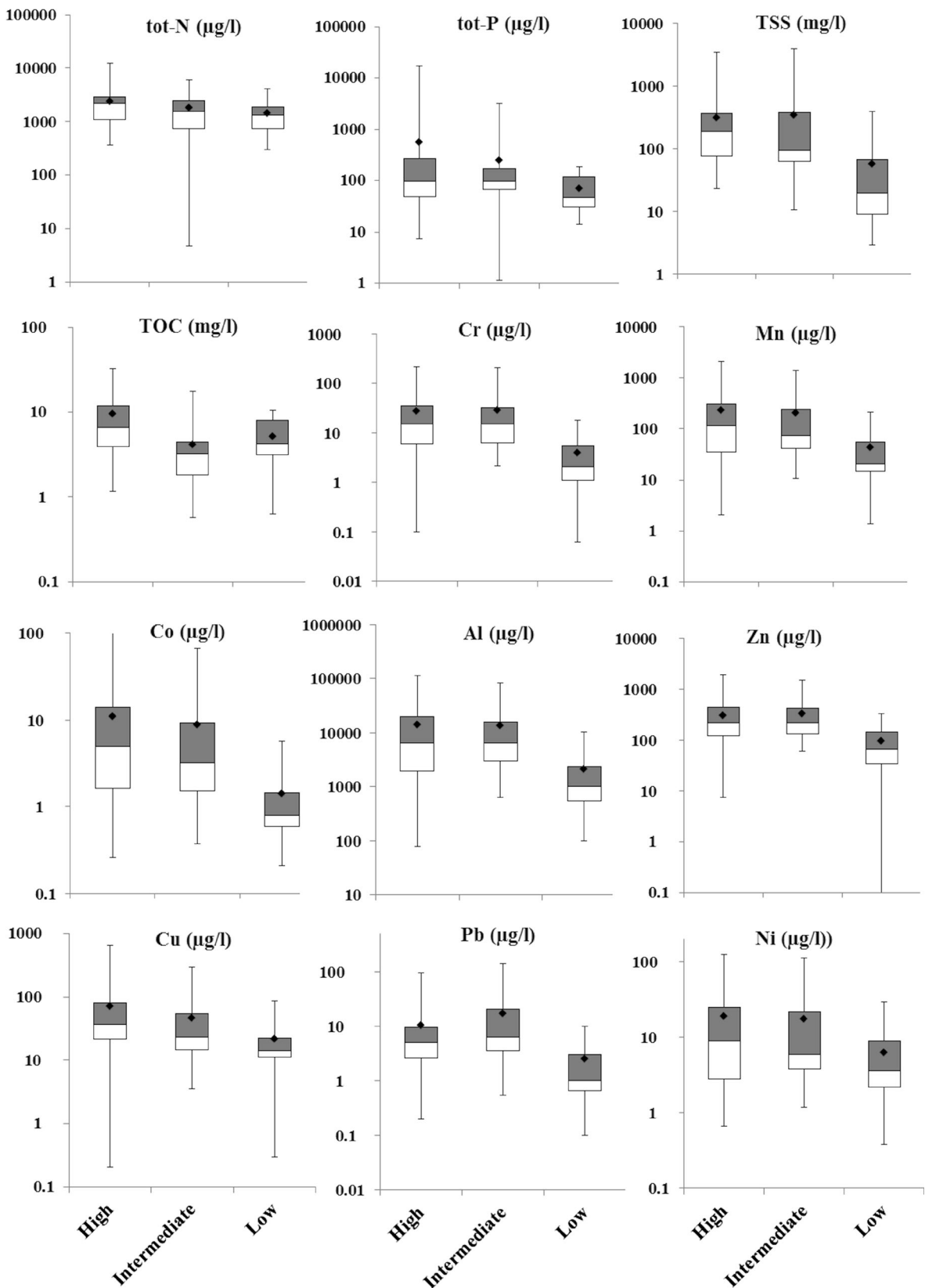


Table 1 Event mean concentrations (EMC) of total nitrogen and total phosphorus, total suspended solids, total organic carbon and selected heavy metals in cold and warm periods at the three study catchments. High, $n=65$; Intermediate, $n=30$. For: TOC High, $n=40$; Intermediate, $n=22$; Low $n=14$. For total Cr, Co and Cu, half of the concentrations and for total Pb and Ni and dissolved Pb most of the concentrations were below the reliable quantitation value

	High (TIA 89 %)						Intermediate (TIA 62 %)						Low (TIA 19 %)					
	Cold		Warm		Cold		Warm		Cold		Warm		Cold		Warm			
	Median (average)	Min-max	Median (average)	Min-max	Median (average)	Min-max	Median (average)	Min-max	Median (average)	Min-max	Median (average)	Min-max	Median (average)	Min-max	Median (average)	Min-max		
Tot-N $\mu\text{g/l}$	2,400 (2,920)	683–12,354	1,021 (1,314)	683–3,236	2,368 (2,509)	4.6–6,038	927 (1,174)	105–5,544	1,567 (1,920)	843–4,095	764 (1,035)	298–2,304						
Tot-P $\mu\text{g/l}$	160 (793)	7.4–17,459	92 (144)	7.4–906	135 (360)	1.1–3,263	74 (165)	19–1,098	30 (39)	14–120	64 (92)	31–186						
TSS mg/l	256 (402)	23–3,359	88 (152)	22.8–676	129 (408)	21–3,810	72 (278)	11–1,756	19 (29)	4.2–93	28 (76)	2.9–384						
TOC mg/l	7.0 (10)	2.3–33	5.9 (7.8)	2.3–31	3.8 (5.3)	0.8–18	3.4 (3.6)	0.6–7.3	5.7 (6.2)	0.63–10	3.2 (3.6)	1.3–8.6						
Total heavy metals																		
Al $\mu\text{g/l}$	17,639 (26,258)	78–113,236	2,155 (4,718)	78–23,083	10,528 (17,136)	2,843–85,217	2,715 (8,511)	635–48,752	1,266 (2,134)	101–10,369	840 (2,096)	293–10,458						
Mn $\mu\text{g/l}$	262 (424)	2.1–2,088	42 (80)	2.1–318	140 (254)	41–1,414	39 (135)	11–825	21 (33)	1.4–132	24 (54)	4.4–214						
Zn $\mu\text{g/l}$	269 (509)	7.3–1,937	140 (233)	7.3–703	277 (340)	73–1,473	208 (316)	60–1,082	34 (61)	0.017–156	91 (138)	33–400						
Cr $\mu\text{g/l}$	30 (48)	0.1–222	7.8 (12)	0.1–50	20 (35)	4.5–207	7 (20)	2–119	2.3 (4.2)	0.06–18	1.9 (4.1)	0.72–17						
Co $\mu\text{g/l}$	12 (22)	0.26–100	1.8 (3.6)	0.26–20	7 (12)	1.6–67	1.5 (5.3)	0.4–35	0.83 (1.6)	0.21–5.8	0.71 (1.3)	0.25–5.6						
Ni $\mu\text{g/l}$	18 (33)	0.66–122	6.6 (13)	0.66–125	9 (20)	1.2–113	5.0 (15)	1.2–79	5.0 (8.0)	0.378–30	3.6 (5.2)	0.93–13						
Cu $\mu\text{g/l}$	72 (126)	1.8–656	22 (33)	1.8–129	29 (58)	11–293	15 (34)	3–179	12 (12)	0.29–28	21 (303)	11–87						
Pb $\mu\text{g/l}$	6.8 (17)	0.2–98	3.2 (9.7)	0.2–68	7 (18)	0.54–110	5 (16)	1.0–142	1.4 (2.7)	0.10–9.8	1.4 (2.6)	0.15–10						
Dissolved heavy metals																		
Al $\mu\text{g/l}$	58 (70)	4.7–234	26 (40)	4.7–101	87 (106)	26–254	75 (69)	31–103	186 (284)	87–911	241 (202)	76–288						
Cr $\mu\text{g/l}$	1.6 (3.2)	0.11–26	2.4 (2.1)	0.11 3.1	2.1 (4.9)	0.41–49	0.5 (1.7)	0.3–4.4	1.3 (1.5)	0.87–2.6	0.50 (0.44)	0.34–0.50						
Mn $\mu\text{g/l}$	16 (35)	0.2–161	2.9 (3.3)	0.2–5.3	10 (45)	1.1–257	8 (104)	4–302	6.3 (6.3)	1.8–11	3.0 (2.6)	0.59–4.4						
Ni $\mu\text{g/l}$	2.0 (3.8)	0.23–25	0.76 (0.88)	0.23–1.9	1.6 (3.7)	0.06–30	0.7 (2.0)	0.1–5.2	1.7 (2.7)	1.1–12	1.1 (0.99)	0.77–1.1						
Zn $\mu\text{g/l}$	56 (61)	3.7–120	93 (93)	3.7–120	66 (77)	12–187	144 (145)	107–183	55 (52)	22–80	53 (58)	46–73						
Cu $\mu\text{g/l}$	8.2 (9.0)	0.47–23	9.5 (12)	0.47–24	6.4 (7.4)	4.9–14	7.2 (7.8)	4.1–12	9.3 (9.7)	6.1–21	11 (11)	8.3–13						

Large differences in concentrations occurred during cold periods between the Low catchment and city centre catchments: median EMCs in the Low catchment were only 2 and 4 % of those in the High and Intermediate catchments, respectively (Table 1). Median EMCs of TSS peaked during the cold months in the High and Intermediate catchments and were 70 and 40 % higher than during warm periods, respectively. In contrast, the warm season yielded 30 % higher median EMCs of TSS compared to the cold season in the Low catchment.

3.1.3 Total Organic Carbon

Concentrations of TOC correlated neither with imperviousness nor with land use type as the lowest median EMC (3.2 µg/l) occurred in the Intermediate catchment (Fig. 2). Irrespective of season, median TOC EMCs were approximately twice as high in the High catchment compared with the other less impervious catchments (Fig. 2, Table 1). In all study catchments, the highest median TOC EMCs occurred during the cold period but in the city centres, the difference in the concentrations between the warm and cold period was only around 10 %, while in the Low catchment it was 50 %.

3.1.4 Heavy Metals (Al, Mn, Zn, Cr, Co, Ni, Cu, Pb)

When expressed as total metals, median EMCs of Mn, Co, Ni and Cu (120, 5, 9 and 40 µg/l at the High catchment, respectively) increased with increasing imperviousness (Fig. 2). The concentrations of Cr, Zn and Al were more strongly related to land use, since median EMCs of these metals were similar in the two city centre catchments (15, 220 and 6,500 µg/l, respectively) and were ca. 85, 70 and 85 % higher than in the Low catchment, respectively. However, some seasonal differences were observed for Al and Zn during warm periods, with highest median EMCs in the Intermediate catchment (Table 1).

The proportion of dissolved metals of the total heavy metals in the runoff varied greatly depending on the given metal and the study catchment (Table 2). In the Low catchment, a larger proportion (12–98 %) of metals occurred in dissolved form than in the two city centre catchments, particularly during the cold period. Dissolved concentrations of Al, Ni and Cu at the Low catchment even exceeded concentrations detected in the two city centre catchments (Table 1). Furthermore, the fractions of dissolved metals differed between the cold

Table 2 Proportions of dissolved heavy metals of total heavy metals in the warm and cold periods during the first study year at the three study catchments

		Dissolved metals (% of total metals)					
		Al	Cr	Mn	Ni	Zn	Cu
Cold period	High	0.9	21	14	21	33	20
	Intermediate	1.1	12	9.6	38	31	17
	Low	12	46	23	61	98	61
Warm period	High	2.3	38	9.0	44	61	56
	Intermediate	1.9	8.3	25	8.9	49	37
	Low	15	36	15	44	63	88

and warm periods in all study catchments (Table 2). In the High catchment, the proportion of dissolved metals in the runoff was about twice as high during the warm season than during the cold season, except for Mn. The same occurred in the Intermediate catchment except that the dissolved fraction of Ni was fourfold higher during the cold season than during the warm one. In the Low catchment, differences in the fractions of dissolved metals between the warm and cold periods were smaller than in the city centre. In the Low catchment, a larger proportion of Al and Cu in the runoff occurred in dissolved form during the warm season than during the cold season, while for Cr, Ni and Mn, it was the opposite.

Concentrations of total Ni and Pb in the runoff were generally low and below the level set for a reliable quantitation value (Ni 63 µg/l, Pb 38 µg/l). However, high concentrations sometimes occurred in all study catchments independent of season. Roughly, every second runoff sample of total Cr, Co and Cu was below the reliable quantitation value (Cr 33 µg/l, Co 7.5 µg/l, Cu 48 µg/l). Higher values were detected during spring melt (especially at the beginning of the melt season) and in summer after long dry periods. Concentrations of dissolved Pb in the runoff samples were mostly below the reliable quantitation value (2.2 µg/l). It was very rare that heavy metal concentrations were below their analytical detection limit; in such cases (e.g. as for Cd), the element was excluded from the data.

3.2 Loading of Runoff Pollutants

3.2.1 Total Nitrogen and Total Phosphorus

Tot-N export resulting from the 2-year study at the High catchment was 650 kg/km², while the Intermediate

catchment generated about 70 % and the Low catchment 80 % of that load (Fig. 3). Tot-P load was related to increasing imperviousness: the Low catchment

generated ca. 10 % and the Intermediate catchment ca. 70 % of the 137 kg/km² tot-P load produced at the High catchment during the study.

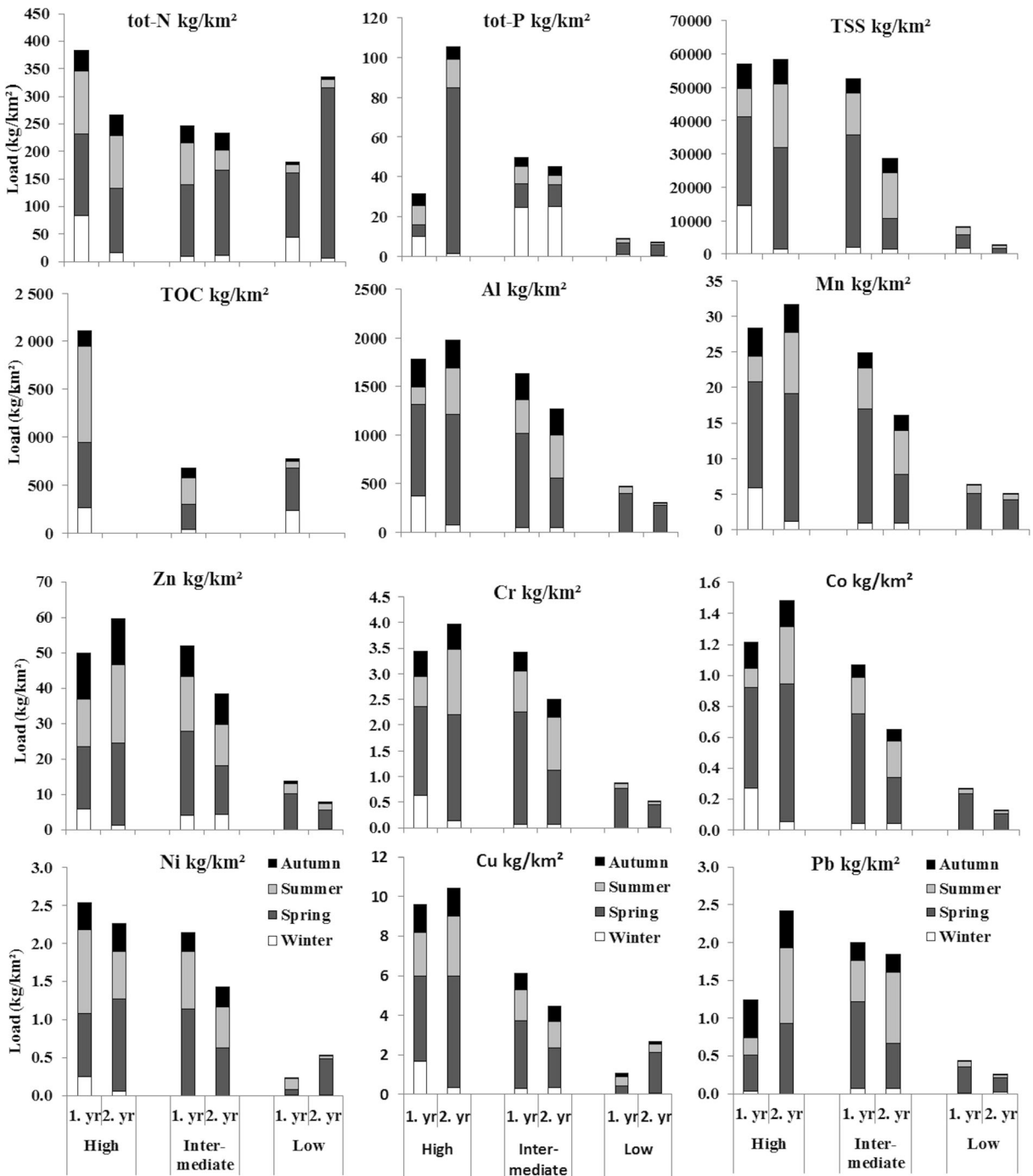


Fig. 3 Annual and seasonal loads for total nitrogen (*tot-N*) and phosphorus (*tot-P*), total suspended solids (*TSS*) and selected heavy metals for the entire study period and for total organic carbon (*TOC*) for the first study year at the three study

catchments. About half of the measured concentrations of total Cr, Co and Cu and most of the concentrations of total Pb and Ni used for load calculations were below the value set for reliable quantitation

For tot-N load, large differences between the catchments were observed during both the warm and cold periods depending on the year. For tot-P, differences between the catchments were often similar during the two periods (Fig. 3). As for tot-N, the cold period generated 60–90 % of the annual load at each study catchment. In the case of tot-P, 70–80 % of its annual load was generated during the cold period in all catchments. The only exceptions occurred in the High catchment: during the first study year, the contribution of the warm and the cold period to tot-P load was similar and during the 2nd year similarities occurred for tot-N load.

3.2.2 Total Suspended Solids

The export of TSS in runoff increased with increasing imperviousness during both years (Fig. 3). The 115,000 kg/km² of TSS load generated in the High catchment during the study was about 30 % higher than in the Intermediate catchment, while load detected in the Low catchment was only 10 % of that generated by the two city centre catchments.

During the warm months, TSS loads in the Low catchment were only 1 % of the loads generated in the two more urbanized catchment (Fig. 3). However, large differences in TSS loads (15–75 %) were detected during the cold months between the High and Intermediate catchment. During the first study year, around 70 % of the annual TSS load occurred during the cold period in each study catchment. During the 2nd year, differences between the two periods were minimal in the High catchment, while in the Intermediate catchment, notably, more TSS was exported during summer than during spring.

3.2.3 Total Organic Carbon

The export of TOC was only quantified in 2009 when the High catchment generated 2,100 kg/km² TOC that was over 60 % more than the two less urbanized catchments (Fig. 3). The difference in TOC load between the Low and Intermediate catchment was greatest (85 %) during the cold period, while the difference between the Low and High catchments was pronounced (85 %) during both periods. The cold period yielded the highest TOC export at the Low catchment, while the warm period contributed most to the annual load in the city centre catchments.

3.2.4 Heavy Metals (Al, Mn, Zn, Cr, Co, Ni, Cu, Pb)

The export of total metals increased with increasing imperviousness for all metals except Pb (Fig. 3). Exports resulting from the study period varied depending on the metal; from 3–3,800 kg/km² in the High catchment to 0.7–800 kg/km² in the Low catchment (Fig. 3). The Low catchment generated 20 % of the heavy metal of the High catchment, except for Co (10 %). At the Intermediate catchment, the exports of Al, Cr and Zn was 80 %, Mn and Ni 70 %, Co 60 % and Cu 50 % of the exports occurring at the High catchment. Only Pb loads were similar between the city centre catchments.

During warm seasons, differences in metal loads between the Low catchment and the two more urbanized catchments were greater than during the cold months. Between the two city centre catchments, differences in loads were greater during the cold periods. In the first study year at each catchment, Al, Cr, Mn and Co loads were 60–90 % higher during the cold period than during the warm one (Fig. 3). In the second year, such seasonal difference diminished in the two city centre catchments. Ni, Zn and Cu loads were of the same magnitude between the cold and the warm periods in the city centre catchments, and Pb was the only heavy metal with highest loads during the summer. In the Low catchment, spring contributed substantially (70–90 %) to annual loads of all the metals, except for Cu.

3.3 Relationship Between Seasonal Pollutant Loads and Runoff

Runoff volumes used here are based on data in Valtanen et al. (2014). The largest volumes of runoff occurred during the warm periods in the High catchment, while in the Low catchment, spring time runoff was high (Table 3). In the High catchment, seasonal pollutant loads were independent of the seasonal total runoff volume (Table 4). In the Intermediate catchment, seasonal loads correlated positively with seasonal runoff for most of the pollutants, while in the Low catchment, positive correlations were observed for half of the pollutants. Of the studied pollutants, the loads of heavy metals were typically related to runoff quantity. The correlations between seasonal total runoff volumes and mean seasonal EMCs were insignificant (Spearman correlation, $p > 0.05$).

Table 3 Seasonal runoff volumes (mm) for the two study years in the three study catchments. Data from Valtanen et al. (2014)

		Low	Intermediate	High
1st study year	Winter	24	26	32
	Spring	78	63	67
	Summer	27	82	133
	Autumn	10	62	70
2nd study year	Winter	3	19	7
	Spring	156	104	70
	Summer	9	49	63
	Autumn	10	62	70

4 Discussion

4.1 Imperviousness- and Land Use-Induced Variation in Runoff Quality

We showed that runoff quality was differently influenced by urbanization: besides imperviousness, concentrations were often strongly affected by land use type but pollutant loads responded more to the degree of imperviousness. Previous studies have shown that runoff quality depends not only on imperviousness but also

Table 4 Spearman correlation (r) and its significance (p) between seasonal total runoff volumes (mm/season) and pollutant loadings (kg/km²/season)

Seasonal load (kg/km ² /season)	Seasonal total runoff (mm/season)		
	High r (p)	Intermediate r (p)	Low r (p)
tot-N	0.319 (0.441)	<i>0.904 (0.002)</i>	0.687 (0.060)
tot-P	0.123 (0.772)	-0.253 (0.545)	<i>0.827 (0.011)</i>
TSS	0.025 (0.954)	0.590 (0.123)	0.703 (0.052)
Al	-0.074 (0.862)	<i>0.759 (0.029)</i>	<i>0.759 (0.029)</i>
Cr	0.111 (0.793)	<i>0.752 (0.032)</i>	0.415 (0.307)
Mn	-0.074 (0.862)	<i>0.759 (0.029)</i>	0.681 (0.063)
Co	-0.148 (0.726)	<i>0.805 (0.016)</i>	<i>0.681 (0.041)</i>
Ni	0.663 (0.073)	<i>0.768 (0.026)</i>	<i>0.759 (0.029)</i>
Zn	0.417 (0.304)	<i>0.831 (0.011)</i>	<i>0.715 (0.046)</i>
Cu	0.196 (0.641)	<i>0.855 (0.007)</i>	0.581 (0.131)
Pb	0.314 (0.448)	0.630 (0.094)	0.542 (0.165)

The table combines seasonal data from the whole study period (December 2008 to August 2010) and catchment data (High, Intermediate, Low). Statistically significant correlations at $p < 0.05$ are indicated by italic text. Seasons, $n = 7$

strongly on land use, which affects water quality through different (anthropogenic) activities related to traffic, parking and population densities and snow handling (Bengtsson and Westerström 1992; Novotny 1992; Semádeni-Davies and Bengtsson 1999; Viklander 1999; Mitchell 2001, 2005; Göbel et al. 2006; Clark and Pitt 2012). The two land use types in our study differed particularly in traffic quantities, road management (such as de-icing and snow ploughing and transportation), population density and probably also in gardening. Logically, therefore, differences in the concentrations of tot-P, Al, Cr, Zn and Pb were evident between the residential catchment (TIA 19 %) and the two city centre catchments that support our hypothesis. The impact of imperviousness on runoff pollutant concentrations was clear when distinct differences were also observed between the city centre catchments (TIAs 62 and 89 %) that occurred in particular for TSS, tot-N, Mn, Co, Ni and Cu. This indicates that, even though land use related pollutant sources may be the same at the city centre catchments, pollutant emissions can increase with increasing TIA.

The loads of most pollutants (TSS, tot-P, Al, Mn, Zn, Cr, Co, Ni, Cu) increased with increasing imperviousness, which supports our hypothesis and evidence that the impact of imperviousness arises from the positive correlation between runoff volumes and pollutant loads (Novotny 1992; Hatt et al. 2004; Dougherty et al. 2006; Bedan and Clausen 2009). During the study, the High catchment generated around 140 mm more direct runoff than the Intermediate catchment (Valtanen et al. 2014), which resulted in considerably higher exports from the High catchment despite the similar concentrations of tot-P, Al, Cr and Zn at these two catchments. A smaller difference, 40 mm (Valtanen et al. 2014), in direct runoff volume occurred between the Low and Intermediate catchments. Thus, the large difference in TSS, tot-P and all heavy metal loads between these sites are explained also by distinct pollutant concentrations and, hence, land use-related pollutant sources. As no large industrial plants are situated in or close to our study catchments, a large part of TSS, tot-P and metals in the city centre catchments likely derive from traffic that is one of their main pollutant sources for runoff in urban areas (House et al. 1993; D'Arcy et al. 2000; Moy et al. 2003). In the Low catchment with very low traffic density, tot-P and TSS can originate from green areas, pets and housing, and the metals can originate from the soil or buildings (i.e. rooftops). It is known, however, that

numerous sources of metals exist in urban areas and that their transport is affected by several environmental factors and mechanisms, such as land use type and hydro-meteorological factors (House et al. 1993; Chiew and McMahon 1998; D'Arcy et al. 2000; Moy et al. 2003).

Contrary to our hypothesis and the pronounced impacts of urbanization discussed above, our study showed that a clear increase in imperviousness or a change in land use type did not have systematic effects on all studied pollutants. For instance, the concentration of TOC was lower in the Intermediate compared to the Low catchment. In addition, the smallest loads of both TOC and tot-N were observed in the Intermediate instead of the Low catchment. A similar pattern for nitrate was found in the yard soil of our catchments: the highest concentrations were detected at the High catchment and lowest at the Intermediate catchments (H. Setälä, unpublished results) that supports our finding. Nitrogen sources for runoff are various and may differ between land use types (Malmqvist 1983; Pitt et al. 1999; Mitchell 2001; Moy et al. 2003). The high proportion of green areas including gardening, fertilization and animal faeces in the Low catchment might be responsible for the disproportionately high leaching of nitrogen there. In the High catchment, the minor proportion of green surfaces does not enable much gardening. As such, load of nitrogen in the High catchment most likely originates from local traffic and low N-retention capacity of the catchment. The difference between the two city centre catchments likely arises from the relatively higher traffic emission at the High catchment in comparison to the Intermediate catchment as the High catchment receives air contaminants deriving from the high traffic main road of Lahti. This is supported by the air quality measurements conducted around the city of Lahti showing that NO₂ concentrations increase along growing traffic density (Setälä et al. 2014). The sources for TOC in runoff can be linked to green areas and their permeable soils (Ågren et al. 2008), which probably explain the relatively high concentrations and loads generated in the Low catchment. The relatively high TOC-loads generated at the High catchment are more difficult to trace but most likely relate to organic dust and organic contaminants found in city centres (Foster et al. 2000). It is also to be noted that differences in runoff quality between our study catchments are not dependent on long-range pollutant transport as the catchments are located only a few kilometres away and therefore sources are to be found within the catchments.

4.2 Annual and Seasonal Differences in Runoff Quality

The study confirmed our hypothesis that pollutant concentrations and loads in the urban runoff exhibit strong seasonal variation, which is enhanced by imperviousness and land use. The largest differences in pollutant loads between the study catchments were detected during the warm periods when loads in the city centre catchments were about twice as high as in the low-density residential catchment. This was expected since differences in runoff volumes between the catchments were greatest during the warm months (Valtanen et al. 2014). Similar seasonal differences in runoff generation have been detected for residential areas in Finland (Sillanpää 2013) and in an urbanizing catchment in Washington D.C. (Dougherty et al. 2006).

Surprisingly, the observed differences in TSS, tot-P, Mn, Co and Pb loads (both years) and in Al, Cr, Ni, Zn and Cu loads (one of the years) between the two city centre catchments were most pronounced during the cold periods. This can be explained by winter time pollutant concentrations that were about twice as high at the High catchment compared to the Intermediate one. In each of the catchments, concentrations of most of the studied pollutants peaked during cold months supporting some previous studies that have shown that concentrations of TSS, TOC and several heavy metals in stormwater are many times higher during snowmelt than during summer rainfall events (Brezonik and Stadelmann 2002; Bäckström et al. 2003; Westerlund et al. 2003; Westerlund and Viklander 2006; Helmreich et al. 2010). The air quality measurements conducted in the city of Lahti also showed that NO₂ and particle concentrations during the cold month (March) are higher than during the summer time (August) (Setälä et al. 2014), which can partially explain the higher element and nitrogen concentrations during the winter time than summer time. However, previous studies show that seasonal runoff pollutant concentration patterns may also depend on land use and the pollutant in question (Sillanpää 2013; Semádeni-Davies and Titus 2003). Furthermore, we showed that the concentrations of TSS, tot-P and some heavy metals (Al, Cr, Co, Ni) were highest during the warm season instead of the snowmelt period in the Low catchment. This is most probably explained by the low runoff volumes generated on polluted impervious areas during the warm period, which results in higher pollutant concentrations in the runoff. It has also been shown that during cold seasons,

pollutant concentrations dilute in low-density urban areas due to snowmelt from clean snow on pervious green surfaces (Sillanpää and Koivusalo 2013), a phenomenon also likely to occur in our study.

As to the pollutant loads in the two city centre catchments, summer periods generated some of the highest seasonal loads indicating that loads generated during the warm period are particularly prone to urbanization. During the cold months—when the evapotranspiration rate is at the minimum and infiltration capacity of soils were reduced by freezing temperatures or saturated soils—the difference in imperviousness, runoff quantities (Valtanen et al. 2014) and pollutant loadings between our study catchments was far less pronounced than during the warm months. The cold period has a clear effect on the characteristics of the catchments as the frost period in Lahti lasts for several months and frost depth in Lahti area is on average 1–1.2 m (Finnish Meteorological Institute 2014). Cold periods—and spring in particular—generated the highest pollutant loads at each catchment. Previous studies have shown that besides rural areas, snowmelt and rain-on-snow events can generate some of the highest loads in urban areas (Hautala et al. 1995; Bäckström et al. 2003; Laurén et al. 2005; Sillanpää 2013). This was expected to occur in the Low catchment where most of the annual runoff volumes are generated during spring snowmelt (Valtanen et al. 2014). However, contrary to our hypothesis at the High catchment loads of nutrients, TSS and most heavy metals (Al, Mn, Zn, Cr, Co, Ni, Cu) did not follow the same seasonal pattern with runoff volumes that were the largest during warm months (Valtanen et al. 2014). As mentioned before, annual loads were affected by imperviousness for which it is also surprising that in the High intensity catchment runoff volumes did not correlate with pollutant loadings at a seasonal scale (see Table 4). The emergence of high spring time loads is most likely due to the fact that pollutant accumulation encompasses the entire cold period. Also, as the high winter time concentrations at the city centres of our study already indicated, pollutant emissions are often higher during cold months due to slipperiness control, high emissions of gaseous impurities by traffic and heating, increased erosion of roads by snow ploughing and the use of friction tyres (Armhein et al. 1992; Hautala et al. 1995; Bäckström et al. 2003). Thus, the importance of the spring season in generating loads implies that urbanization affects the most generative season by increasing pollutant concentrations during winter in contrast to rural

areas where it is more the massive runoff volume that leads to high spring loadings.

The two study years yielded some clear differences in pollutant loads between the three catchments. During the 1st year, the loads of tot-P, Zn and Pb were the same order of magnitude between the two city centre catchments but the Intermediate catchment yielded the largest loads. However, during the second study year, the Low catchment generated the highest tot-N load. Reasons for non-systematic differences between the city centre catchments are difficult to trace but may be because of differences in runoff volumes due to weather conditions and snow transportation. The two study years experienced equal precipitation but 65 % of annual precipitation occurred during the warm period in the first year, while precipitation was roughly the same between the warm and cold periods in the 2nd year (Valtanen et al. 2014). During the second study year, high winter time precipitation caused a massive spring flow at the Low catchment from which the high tot-N load most probably originated, despite the low nitrogen concentrations during the cold months. In the Low catchment, nitrogen is mainly exported from the rural areas as there most of the runoff in spring is generated in green spaces. During cold periods, 50–90 % of the area in the High catchment and 40–70 % of the area in the Intermediate catchment was ploughed, almost free of snow. Almost all of the ploughed snow in the two city centre catchments is transported out of the catchments (Jani Tuhkanen, the city of Lahti, personal communication). In the Low catchment, only 10–25 % of its area is ploughed, almost free of snow and virtually all of the ploughed snow retains in the catchments. Approximately 130,000 m³ (2009) and 220,000 m³ (2010) of snow was transported out of the city of Lahti during the study winters (The City of Lahti 2011). Thus, during the second study year with high winter time precipitation, the transport of snow must have had a stronger impact on pollutant export. Due to the lack of data on snow removal at the study catchment scale, accurate estimates of the amount of pollutants removed/left in the three study catchments cannot be made. Nevertheless, previous studies have shown that snow transportation reduces runoff volumes in city centres (Bengtsson and Westerström 1992; Semádeni-Davies and Bengtsson 1999; Valtanen et al. 2014). Differences between years indicate that uncertainties in short-term studies are substantial and that long-term measurements are needed to determine specific pollutant export patterns.

4.3 Variation in Dissolved Fraction of Heavy Metals

Our study showed that the dissolved metal fraction was impacted by land use, imperviousness and season. Even though total metal concentrations in the city centre catchments were orders of magnitude higher than in the low-density residential catchment, the proportion of dissolved metals of the total metals was higher in the Low catchment than in the city centres. Furthermore, dissolved Al, Cu and Ni concentrations were highest in the Low catchment. This indicates that the dissolved metal fraction is dependent on land use and, in fact, decreases along increasing imperviousness. Based on previous studies, most of the heavy metals in runoff are particulate-bound and correspond to the solubility of the metal (Foster and Charlesworth 1996; Sansalone and Buchberger 1997) that occurred also in our study. However, the dissolved metal fraction varied from 1–61 % in the High catchments to 12–98 % in the Low catchment, which is higher than in previous research (Clark and Pitt 2012).

In the current study, seasonal differences in the fractions of dissolved heavy metals were also detected but we were unable to identify systematic patterns for seasonal behaviour. Our results also contradict previous studies. Findings from the two most impervious catchments support work by Bäckström et al. (2003) and Helmreich et al. (2010) who showed that dissolved fractions of Zn and Pb diminish during cold months. Besides these metals, dissolved fractions of Al, Cu, and Zn also decreased during the cold seasons in the city centre catchments of our study, indicating that cold conditions increase the fraction of particulate-bound pollutants, particularly in dense urban settings. Surprisingly, the fraction of dissolved Ni in the Intermediate catchment was fourfold higher during the cold season compared to the warm season, but an explanation for this eludes us. Contrary to city centres, dissolved metal fractions of Zn, Cr, Mn and Ni in the Low catchment were highest during cold seasons. Higher dissolved fractions in road site runoff during cold months have also been shown for Al, Cd, Co, Cr, Mn and Ni (Hallberg et al. 2007), which partly supports our findings, especially from the Low catchment. Based on previous studies, the main reasons for dissolved form of metals in meltwater are most probably due to the use of salt as a de-icing agent (Oberts 1994; Marsalek 2003; Reinosdotter 2007), which is also applied in the study catchments of the present study (The City of Lahti

2011). It is also known that sources of metals differ between warm and cold periods (Hallberg et al. 2007; Bäckström et al. 2003), which may explain differences in the forms and concentrations of metals between the two distinct periods. In summary, our results showed that mechanisms controlling dissolved metals in runoff are complex and sometimes contradictory, which indicates that more research is needed for understanding the behaviour of metals in urban runoff in cold climates.

4.4 The Impacts of Urban Runoff Quality on Surface Waters

Our study indicated that urbanization has runoff-induced impacts on receiving water bodies. As such, urban runoff requires treatment. Concentrations detected in the current study were compared with thresholds set for stormwater quality by the local water company in Stockholm, Sweden (Stockholm Vatten 2001). According to these criteria, urban runoff should be treated if moderate or high threshold values are exceeded. In all catchments studied here, median concentrations of tot-N, Cu and Zn exceeded these thresholds, while median concentrations of TSS, Cr and Pb exceeded these thresholds in the city centre catchments. These thresholds were mainly exceeded during cold months, but maximum concentrations also exceeded these thresholds during warm periods in all studied catchments. Thresholds for the receiving water bodies also exist for Cu, Zn, Cr, Ni and Pb, set by the Swedish Environmental Protection Agency (2000). Exceeding these thresholds can pose a high risk to the aquatic biota and their functions in surface water courses. In our study, median concentrations exceeded these thresholds for Cr and Pb in the city centre catchments (especially during cold months) and for Zn and Cu in all catchments year-round.

5 Conclusions

The aim of our study was to shed light on the effects of urbanization on runoff-induced pollutant transport and to study the mechanisms that control pollutant loads and concentrations during vastly divergent seasons in cold climate conditions. The study showed that, irrespective of land use-induced pollutant concentrations, pollutant loads increased with increasing imperviousness during a 2-year period, confirming the importance of runoff

volumes and imperviousness in pollutant export. Nevertheless, during different seasons, urbanization affected pollutant loads and concentrations in distinct ways: (i) in city centres during warm seasons, loads were more controlled by runoff volumes, i.e. imperviousness, but during cold months, loads were also affected by high pollutant emissions as a result of land use type and (ii) in the low-density residential area, high cold period loads were affected by high runoff volumes even though some of the pollutant concentrations were diluted during cold months. On the whole, increasing levels of urbanization altered pollutant load generation especially during warm seasons, which implies that runoff pollutant generation, especially in highly urbanized areas, is an all year-round phenomenon.

The study showed that urbanization, defined either as imperviousness or land use type, increases runoff-induced pollutant export of several common pollutants (nutrients, solids, heavy metals, TOC) and, thus, has potential to affect the quality of receiving water bodies. Based on threshold values set for stormwater quality, the treatment of urban runoff is recommended for northern countries. In addition, seasonality of the pollutant discharge should not be neglected when designing runoff management strategies that require functionality throughout the year. The study also suggests that in cold climate, year-round measurements that span several years are important when estimating pollutant export patterns and stormwater treatment requirements.

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