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Accumulation of Traffic-Related Trace Metals in Urban Winter-Long Roadside Snowbanks

S. Moghadas · K. H. Paus · T. M. Muthanna · I. Herrmann · J. Marsalek · M. Viklander

Abstract Accumulations of mass loads of selected chemicals in roadside snowbanks were studied at five sites with various traffic densities in the city of Trondheim (Norway) by collecting snow samples throughout the winter period and analyzing them for 13 water quality constituents: pH, electrical conductivity (EC), alkalinity, Cl, Na, total suspended solids (TSS), Cd, Cr, Cu. Ni, Pb, W, and Zn. The resulting dataset was then supplemented by similar data collected earlier in the city of Luleå (Sweden). Regression analyses for individual sites indicated linear trends in unit-area constituent accumulations with time ($0.65 < R^2 < 0.95$) and supported the assumption of linearity in further analyses. Principal component analysis (PCA) of the combined Luleå/Trondheim data revealed cause-effect relationships between the chemical mass loadings (TSS and trace metals) and three predictors: snow age (snow residence time (SRT)), traffic density (annual average density of traffic (AADT), and cumulative traffic volume (CTV=SRT×AADT). Cl and Na loads, originating from road salt applications in Trondheim only, did not display this trend. Two types of parsimonious models for predicting trace metal accumulations in winter-long roadside snowbanks were developed: (a) a linear regression model using CTV as a single predictor and predicting metal accumulations with a moderate certainty ($0.37 < R^2 < 0.66$) and (b) multiple regression models using SRT, AADT, and snow water equivalent (SWE) as predictors. The latter models indicated good correlations between the mass loads and the predictors ($0.64 < R^2 < 0.77$) and produced slightly better prediction accuracies ($0.44 < R^2 < 0.67$) than the simpler model.

S. Moghadas (✉) · I. Herrmann · M. Viklander
Department of Civil, Environmental and Natural Resources Engineering, Luleå University of Technology, 97187 Luleå, Sweden
e-mail: shahab.moghadas@ltu.se

I. Herrmann
e-mail: inga.herrmann@ltu.se

M. Viklander
e-mail: maria.viklander@ltu.se

K. H. Paus
Asplan Viak AS, Kjørboveien 20, 1337 Sandvika, Norway
e-mail: kimpaus@gmail.com

T. M. Muthanna
Department of Hydraulic and Environmental Engineering, Norwegian University of Science and Technology, S.P. Andersensvei 5, N-7491 Trondheim, Norway
e-mail: tone.muthanna@ntnu.no

J. Marsalek
National Water Research Institute, Environment Canada, 867 Lakeshore Rd., Burlington, ON L7R 4A6, Canada
e-mail: jiri.marsalek@ec.gc.ca

1 Introduction

Pollution accumulation in, and release from, snow deposits has been studied during the past several decades with the objective of examining pollutant transport

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pathways and potential impacts on the receiving environments during snowmelt. In this context, snowpacks in natural areas were used as "passive samplers" of atmospheric deposition of persistent pollutants (Carrera et al. 2001), including polycyclic aromatic hydrocarbons (PAHs), pesticides, and trace metals (Li et al. 2015). In urban areas, snow pollution processes are greatly modified (Marsalek et al. 2003), because snow deposits are exposed to the pollution released from anthropogenic activities (traffic, heating, road maintenance) and the concentrations of released substances in urban snow may exceed those associated with atmospheric deposition by two orders of magnitude (Viklander 1998; Bućko et al. 2011). Consequently, urban studies focused on snow deposits exposed to specific pollutant sources, most often traffic and winter road maintenance operations (e.g., Glenn and Sansalone 2002); spatial variations in pollutant distribution ranging from experimental plots (Westerlund and Viklander 2011) to whole catchments (Sillanpää and Koivusalo 2013); and pollution loads stored in bulk snow (Bækken 1994; Viklander 1997; Björklund et al. 2011; Telloli 2014), or released with snowmelt runoff in situ (Meyer et al. 2006; Meyer et al. 2009a), or at snow deposit and storage sites (Viklander 1996; Exall et al. 2011).

Among anthropogenic activities, the dominant effect on urban snow pollution is exerted by operation of motor vehicles, certainly with respect to trace metals (Viklander 1999) and PAHs (Kuoppamäki et al. 2014). The understanding of traffic-generated pollution in urban snow is partly based on the large knowledge base developed with respect to the pollution of road (highway) runoff, which was recently reviewed by (Kayhanian et al. 2012). This review and the literature cited below indicate that, besides the conventional water quality parameters, the most broadly reported constituents in highway runoff were trace metals, but data on nutrients, PAHs, toxicity, and herbicides/pesticides were relatively limited. Major specific sources of trace metals in highway runoff were identified by Bourcier et al. (1980), Frederick (1995), Davis et al. (2001), Ozaki et al. (2004), Peltola and Wikström (2006), Lee et al. (2012), and Zhong et al. (2012) as follows: vehicle exhaust/diesel soot—Ni, Pb, Zn; tire wear—Cd, Zn; tire studs—W; brake wear—Cd, Cu, Pb, Zn; catalytic converters—Pt, Pd, Rh; asphalt road surface wear—Ni; road marking paint—Cd, Pb; and galvanized metal structures (signs, drains, and guardrails)—Zn. The level of understanding of metal sources varies, with most

references addressing the three most ubiquitous metals, Cu, Pb, and Zn (Gunawardena et al. 2015), and relatively few studies examining the platinum group metals used in catalytic converters (e.g., Pt, Pd, Rh) (Lee et al. 2012; Zhong et al. 2012). The main sources of PAHs are vehicle exhaust/diesel soot (Takada et al. 1991). Both trace metals and PAHs are of particular interest in environmental studies, because besides chloride, they represent the major sources of toxicants in highway runoff (Mayer et al. 2011).

In urban areas with seasonal snowpacks, some part of the pollution load generated by traffic and winter road maintenance (applications of salt and grit) enters and is stored in roadside snow and eventually, it is released during snowmelt (Moghadas et al. 2015) into the receiving environments (Glenn and Sansalone 2002; Bækken 1994; Kuoppamäki et al. 2014; Lygren et al. 1984; Vasić et al. 2012; Mihailović et al. 2014). Pollutant accumulation in roadside snow increases with, among other factors, the annual average daily traffic (AADT) and the snow residence time (SRT) (Viklander 1998; Viklander 1999). Thus, particularly high concentrations of trace metals in roadside snow were reported for heavily trafficked highways in cold climate regions, where winter-long snowbanks may be exposed to traffic pollution inputs over the period of several months. For example, Lygren et al. (1984) reported Pb concentrations in a 4-month-old snow deposit along a highway in Oslo, Norway, ranging from 410 to 6880 µg/L (these data represent the period before phasing Pb out of gasoline), and for similar conditions, Bækken (1994) reported Zn concentrations ranging from 58 to 6114 µg/L. For less intense traffic in a smaller northern city (Luleå, Sweden), Viklander (1999) reported Cd concentrations in the range from 0.15 to 4.64 µg/L, and Westerlund and Viklander (2011) reported mean concentrations of Cu, Ni, Pb, and Zn as 239, 73, 63, and 851 µg/L, respectively. Additional pollutants enter roadside snow as a result of winter road maintenance, when in jurisdictions with "bare pavement" policies, large quantities of road salts are applied to pavement surfaces and strongly contribute to toxicity of produced snowmelt and changes in snowmelt (Marsalek 2003).

The actual release of pollutants from melting snow is related to the release of water and, in general terms, was described as preferential elution of dissolved chemicals and delayed elution of hydrophobic chemicals (Meyer et al. 2009a; Schöndorf and Herrmann 1987; Meyer and Wania 2008; Meyer et al. 2009b). The elution

chemodynamics may magnify the concentrations of pollutants in bulk snow during various phases of snowmelt. Comparisons of trace metal concentrations in bulk snow to ambient freshwater quality guidelines indicate high risks of biological effects (Swedish EPA 2000; CEPA 2007) resulting from urban snow disposal, or snowmelt discharge, into receiving waters. Thus, the levels of trace metal concentrations in roadside snow and their variations in time are of great interest when considering used snow disposal; fresh, little contaminated snow could be disposed in local receiving waters, to save on transportation costs and reduce pollution caused by snow transportation, but old snow may need to be disposed of at more distant snow storage and disposal sites equipped with treatment of meltwater (Exall et al. 2011).

To provide guidance for quick estimation of trace metal pollution in urban winter-long road snowbanks, a study of such pollution was undertaken in the northern city of Trondheim (Norway) and the resulting dataset was enhanced by inclusion of similar data from an earlier study conducted in the city of Luleå (Sweden). Following the exploratory statistical analysis of the combined dataset, parsimonious models of trace metal accumulation in roadside snowbanks were formulated and proposed for assessing the quality of urban snow in roadside snowbanks.

2 Materials and Methods

2.1 Study Sites

The study sites were located in two cities, Trondheim (Norway) and Luleå (Sweden), and their basic characteristics are listed in Table 1. The city of Trondheim is the third largest city in Norway, with about 170,000 inhabitants (in 2010), located at 63.42° N, 10.39° E. The average annual precipitation for Trondheim study sites was taken as 920 mm, as recorded at the Risvollan Urban Hydrological Research Station (RUHRS). Snowfall represented about 30 to 40 % of the total precipitation. The city of Luleå is located in northern Sweden at 65.58° N, 22.15° E, and its population was about 70,000 inhabitants in 1995, when the snow data were collected. Average annual precipitation in Luleå is 500 mm, of which 50 % is snow staying on the ground between November and April (Hernebring 1996). In each city, four study sites were selected, in the built-up parts of the

city, according to their representative ranges of traffic intensities (AADTs), and used for snow sampling during the winter of 1995 in Luleå and the winter of 2010 in Trondheim. These four sites represented snowbanks exposed to pollution input from traffic, characterized by a specific AADT estimated by manual counts and standard calculation procedures (Norwegian Public Road Administration 2009). Additional two sites in Luleå and one site in Trondheim were chosen as reference sites with no direct exposure to traffic. In Trondheim, the reference site was located 40 m from the access road, and in Luleå, the reference sites were in a city park and a residential area. Trace metal burdens in snow at the reference sites were assumed to originate from atmospheric deposition and represented background levels (Viklander 1999).

In Trondheim, snow clearance by ploughing is carried out when precipitation as snow exceeds 7 mm for roads with AADT >3000 vehicles/day and 10 mm for roads with 1500<AADT≤3000 vehicles/day. In Luleå, snow clearance starts when snowfall reaches 40–50 mm in main streets and 100–120 mm in residential streets (using the practical rule of thumb, 1 mm of precipitation=10 mm of snowfall). Snow ploughed off the roads, and streets is typically stored along the road, in the form of snowbanks. Approximately 2900 t of road salt (NaCl) was applied on the city roads in Trondheim during the winter of 2009/2010, while no salt was used in Luleå. According to the logs of winter road maintenance operations in Trondheim (i.e., snow ploughing and removal, road salt and sand applications), all the studied sampling sites with traffic were subject to salt applications during the study period. Furthermore, no abrasives were applied at the sites studied, and no snow was removed during the sampling period.

2.2 Snow Sampling and Laboratory Analyses

Snow samples were collected as vertical cores from the top of the snowbanks to the bottom. Nine samples were taken between 17 January and 26 April 1995 in Luleå and seven samples between 29 January and 15 March 2010 in Trondheim. The samples were stored in plastic bags and transported to the laboratory in a frozen state within 1 h of sampling. Snow density was calculated from the measured volume and weight of snow samples.

In the laboratory, the snow samples were slowly melted at a room temperature (about +20 °C) and the

Table 1 Characteristics of study sampling sites in Luleå and Trondheim

City	Site ID	AADT vehicles/day	Comment
Luleå	L_{ref1}	0	Residential area—away from streets
	L_{ref2}	0	Park in downtown
	L1	1500	Residential street—low traffic
	L2	4500	Connecting road—intermediate traffic
	L3	5000	Connecting road—intermediate traffic
	L4	20,000	Arterial road—higher traffic
Trondheim	T_{ref}	0	Research site—no public access
	T1	2256	Residential street
	T2	7300	Connecting road—intermediate traffic
	T3	12,364	Arterial road—higher traffic
	T4	14,955	Arterial road—higher traffic

meltwater was filtered through a nylon screen with the size of openings of 1 mm to remove coarse particles, debris, and litter. The resulting filtrate was then analyzed for alkalinity, pH, electrical conductivity (EC), total suspended solids (TSS) and ten elements, including seven trace metals. Analytical methods applied to Trondheim and Luleå samples somewhat differed, as explained below.

Trondheim samples were analyzed by the Department of Chemistry of the NTNU (Norwegian University of Science and Technology). TSS was measured according to the Norwegian Standard NS-EN 872, using Whatman GF glass microfiber filters with the pore size of 1.2 μm and vacuum-assisted filtration. For trace metal analysis, sample aliquots were collected in 50 mL Sarstedt tubes and stored in a refrigerator. Subsequently, 6.5 mL of the aliquots was digested in a microwave oven (UltraClave) with 20 % HNO_3 and then diluted six times. Subsamples were analyzed using high-resolution inductively coupled plasma-mass spectrometry (HR ICP-MS) for total concentrations of ten elements. The reported detection limits, in μg/L, were as follows: 0.01 (Cd), 500 (Cl), 0.025 (Cr), 0.1 (Cu), 50 (Na), 0.065 (Ni), 0.01 (Pb), 0.025 (Pt), 0.005 (W), and 0.125 (Zn). Standard QA/QC procedures were implemented, with blanks analyzed and the results verified against certified reference materials. Alkalinity and pH were measured using a Metrohm 726 Titroprocessor, Norwegian Standards NS 4754 and NS 4720, and EC was measured using a microprocessor conductivity meter (LF 537), Norwegian Standard NS 4721.

Luleå samples were analyzed by a commercial laboratory accredited by the Swedish Board for Technical Accreditation (SWEDAC) as fulfilling the requirements of international standards EN45001 and ISO/IEC Guide 25 (Viklander 1998). TSS was measured according to the Swedish Standard Method SS 028112 for Water Investigations, using Whatman GF glass microfiber filters with the pore size of 1.6 μm and vacuum-assisted filtration. Trace metals were analyzed by inductively coupled mass spectrometry (ICP-MS), with pneumatic nebulization. An Ingold meter for ion weak water at room temperature of 20 °C was used to measure pH, and a radiometer, CDM 210, was used to measure the conductivity in Luleå samples.

2.3 Meteorological Data and Snowbank Development

The primary objectives of field data collection in both cities were to estimate the time of the onset of snowbank development (i.e., when SRT=0), identify the cold period when accumulation of trace metals in snowbanks occurs, and illustrate the potential eutectic melt period of snowbanks. Meteorological data (i.e., daily precipitation and air temperature) in Trondheim were collected at the RUHRS and in Luleå, at the Luleå Airport, located approximately 4.5 km from the city center. It should be noted that meteorological stations in both cities were less impacted by anthropogenic influences (e.g., heat fluxes, solar radiation reflection from buildings) than the study sites with traffic, and this may explain some uncertainties in the occurrence and timing of minor snowmelt episodes.

Roadside snowbank evolution during the study period, with respect to snow water equivalent (SWE[mm]),

was derived from the observed meteorological data and presented in Fig. 1 for both cities, together with air temperatures, precipitation, and the sampling dates (i.e., denoted LD1–9 for Luleå and TD1–7 for Trondheim) over the course of the study. In Luleå, the monthly average air temperatures in January, February, March, and April were −6.7, −5.4, −2.1, and 0.4 °C, respectively. A snowfall of 12.3 mm (i.e., SWE) on December 29/30, 1994, was assumed to be the onset of evolution of seasonal roadside snowbanks in the city (i.e., SRT=0). Air temperatures exceeded 0 °C on five occasions during the sampling events LD1–7; however, the SWE values indicated only minimal snow melting in the roadside snowbanks. Therefore, samples LD1–7 were assumed to be representative of the snow accumulation period, but samples LD8 and LD9 clearly represented the snow melting period with a rapidly declining SWE.

In Trondheim, the monthly average air temperatures in January, February, and March of 2010 were −7.0, −5.7, and −0.1 °C, respectively. A snowfall of 32 mm (i.e., SWE) on January 4 initiated the development of a winter-long snow cover lasting throughout the study period (i.e., SRT=0). Furthermore, this snowfall was followed by a snow ploughing operation on all roads in the city, which resulted in the development of roadside snowbanks along all traffic routes. Further increases in snowbank SWE resulted from snowfall events and two snow ploughing operations (i.e., on January 25 and February 8); hence, for the first three sampling operations (TD1–TD3), a positive growth in SWE was observed. In March 2010, the mean SWE started to decline. Road salt was applied on the roads shortly after the snow ploughing operations on five occasions during the study period (i.e., on January 27, February 9 and 18, and March 13 and 21). Although the daily average air temperatures exceeded 0 °C on three occasions, no snowmelt events were recorded at the RUHRS station before March 9. From March 9 on, the air temperatures were fluctuating around 0 °C and caused three snowmelt events within the study period and a decrease in SWE at all the study sites with traffic. It was noted that even during the snow accumulation period, determined from observations at both meteorological stations, the sampled SWEs were not continually increasing and occasional SWE declines were observed at LD3, LD5, TD3, and TD4 before the final snowmelt period started (Fig. 1).
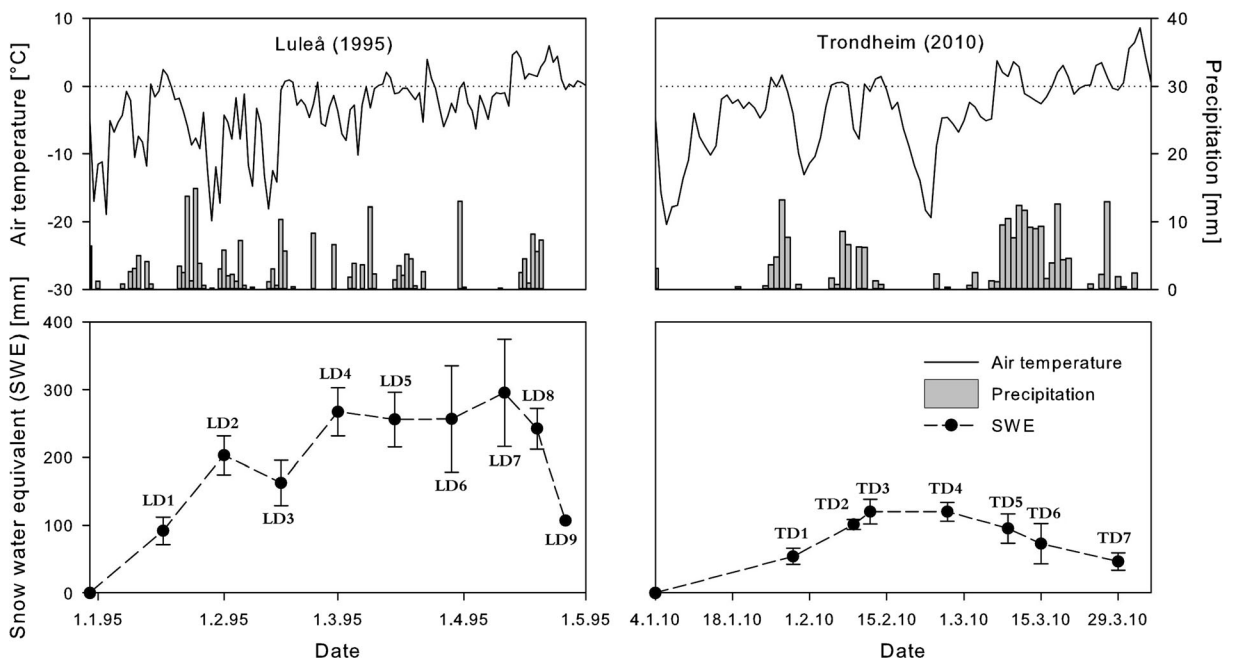
Fig. 1 *Top panels*: air temperature and precipitation (measured at the Luleå Airport and RUHRS). Bottom panels: mean SWE with standard deviations in the studied snowbanks (with traffic), sampled in Luleå on nine dates (LD1–LD9) and in Trondheim on seven dates (TD1–TD7)

2.4 Statistical Analysis

When dealing with spatially varying pollutant loads, it is useful to estimate the loads stored in snow as unit-area mass loads [mg/m^2], which can be readily compared among different sites (Boom and Marsalek 1988; Viklander 1997; Reinosdotter and Viklander 2005). Hence, trace metal concentrations in bulk snow were converted to unit-area mass loads using Eq. 1:

$$ML = K \cdot C_S \cdot SWE \qquad (1)$$

where ML is the constituent mass load per square meter of a snow deposit [mg/m^2], K is a unit conversion coefficient (=1000), C is the constituent concentration in the melted snow sample [mg/L], and SWE is the snow water equivalent of the sample [m], calculated from the measured weight and volume of each snow sample.

Statistical analysis of snow quality was based on the hypothesis that trace metal accumulation rates in roadside snow are quasi-linear during the freezing periods (i.e., no metal mass losses during snowmelt, which is particularly well justified for metal fractions associated with solids; Schöndorf and Herrmann (1987)), and therefore, AADT and SRT could be used as independent variables for predicting the quality of roadside snow with respect to trace metals. The data analysis was performed in two steps: firstly, conducting an exploratory analysis examining relationships among the experimental variables, and secondly, using the first step findings, a predictive general linear model of constituent loads in snow exposed to traffic was developed.

The exploratory analysis tested the metal accumulation hypothesis in two stages: (a) for individual sites, considering temporal and spatial variations, and (b) analyzing the combined data sets as a general representation of metal accumulation. Individual site data were subject to regression analyses examining the linear accumulation behavior of mass loads and normalized mass loads (i.e., normalized by traffic loads, AATD), with the overall objective of examining the feasibility of combining the site data into a single (citywide) dataset for further analysis. In such analysis, the metal accumulation hypothesis was examined for the combined datasets by applying principal component analysis (PCA) to investigate correlations among the influential factors and the studied water quality constituents.

Using the findings of the PCA, predictive parsimonious models for metal mass accumulations in roadside snowbanks were developed using two approaches of different complexity: in a simpler approach, a single predictive variable, the cumulative traffic volume (CTV=SRT×AADT) was used in a simple linear regression model, and in the more complex approach, multiple regression analysis was applied to develop a general linear model including several independent variables (i.e., AADT, SRT, and SWE) and their interactions. The statistical analyses were carried out using the Minitab software (Minitab 2013).

3 Results and Discussion

3.1 Trace Metals in Roadside Snow

Examples of snow physical properties and snowmelt quality characteristics, described by five physicochemical parameters and nine selected elements, are shown in Table 2, for three sites in each city: a reference site, a low-traffic site (AADT=1500 and 2256 in Luleå and Trondheim, respectively) and a high-traffic site (AADT=20,000 and 14,955, in Luleå and Trondheim, respectively).

When comparing the complete datasets from the two cities studied (for reasons of brevity, Table 2 contains just partial datasets), the statistical significance test (t test) confirmed that in terms of magnitudes, concentrations of TSS, Cr, Cu, Ni, Zn, and pH values were significantly higher in Trondheim than in Luleå (Table 2). This can be explained by two facts: Greater snow depths in Luleå (hence, higher dilution) and generally higher AADTs in Trondheim (the average value of four site AADTs in Trondheim, 9375, was 1.2 times higher than that in Luleå, 7750; Table 1). It is generally acknowledged in the literature that concentrations of traffic-related constituents increase with AADT both in road runoff and sediments (Gunawardena et al. 2015) and in roadside snow (Viklander 1999; Gunawardena et al. 2015; Li et al. 2015). The above authors also confirmed that traffic was the main source of such pollution, greatly exceeding atmospheric deposition and other anthropogenic sources. Li et al. (2015) also showed a clear decline in trace metal concentration in urban roadside snow in the direction from the urban

Table 2 Summary of selected hydrologic and water quality parameters measured in roadside snow Luleå and Trondheim

City [–]	SRT [days]	SWE [mm]	pH	EC [µS/cm]	Alkalinity [mmol/L]	TSS [mg/L]	Cl	Na	Cd [µg/L]	Cr	Cu	Ni	Pb	Z	W
L$_{ref}$	70	88.3	6.4	13	0.07	5	7.2	1.8	0.01	1.3	3.0	0.6	0.6	12.6	1.2
L1 AADT=1500	25	68.0	8.6	1073	0.78	3755	492.5	251.8	0.6	335.4	304.5	172.9	36.1	607.7	83.6
	36	112.1	8.9	863	0.79	1820	306.6	157.9	0.3	171.1	146.9	89.3	18.2	278.3	41.9
	39	104.8	8.8	2310	0.8	2415	803.6	428.0	0.1	83.2	62.6	40.5	7.8	127.4	27.4
	53	118.8	8.6	927	0.76	1740	487.0	270.7	0.4	226.7	218.7	110.1	22.5	416.7	63.1
	64	90.7	8.8	249	0.87	3385	111.1	78.2	0.7	367.8	297.5	177.4	32.4	624.2	84.3
	70	45.9	7.4	643	0.47	1870	320.0	181.3	0.1	112.6	63.8	50.8	8.3	127.0	10.5
	84	39.7	7.1	50	1.46	730	16.4	18.8	0.2	176.0	158.8	92.3	16.0	288.5	29.8
L4 AADT= 20,000	25	54.8	8.6	945	1.79	8425	576.4	272.2	0.2	68.7	105.6	34.7	12.1	216.8	105.8
	36	95.2	8.9	622		5405	207.3	138.3	0.6	419.5	366.9	202.6	43.0	672.8	173.6
	39	105.8	8.9	880		5950	337.2	190.8	0.5	336.1	320.3	173.1	33.2	559.4	155.0
	53	103.8	9.0	680		8290	317.2	185.3	1.1	737.7	621.1	330.2	72.3	1229.9	207.5
	64	79.4	9.1	959		11145	66.8	49.0	0.8	487.7	462.8	241.1	49.2	1080.6	172.4
	70	51.1	7.8	1525		2920	859.3	475.5	0.8	526.3	413.2	246.4	44.2	856.8	132.4
	84	61.3	7.6	87	1.87	12695	6.3	29.5	2.6	1351.8	1248.2	678.3	163.7	2403.5	143.0
T$_{ref}$	75	86.9	4.9	6.92	0.04	14		0.2	0.1	0.5	1.4	0.9	3.3	18.5	
T1 AADT=2256	18	103.3	6.4	13.7	0.1	790		5.4	0.5	45.3	85.7	28.0	60.6	164.0	
	33	187.6	6.3	15.2	0.09	630							56.0		
	47	190.2	6.3	11.6	0.09	400		1.9	0.8	23.0	63.3	14.8	42.5	155.0	
	61	226.4	6.3	10.48	0.09	550							32.6		
	75	246.0	6.3	10.24	0.08	410		5.8	0.8	59.6	88.2	31.4	59.4	226.0	
	89	207.8	6.4	12.2	0.09	480		4.7	0.4	28.4	49.0	18.7	36.6	119.0	
	102	323.0	6.0	5.2	0.07	440							47.4		
	110	196.3	5.9	10.57	0.06	310		1.8	0.2	21.8	40.5	11.7	39.9	71.0	
	117	114.3	5.8	18.1	0.07	1300							75.8		
T4 AADT= 14,599	18	101.7	7.6	3.16	0.3	1600		16.6	2.3	58.8	211.0	69.5	274.0	590.0	
	33	230.5	7.2	21.1	0.16	480							82.2		
	47	166.7	7.1	22.4	0.16	350		5.5	1.0	27.5	119.0	20.2	100.0	228.0	
	61	297.4	7.4	29.2	0.24	1100							194.0		
	75	239.9	7.9	30.6	0.36	3000		29.8	4.6	181.0	574.0	125.0	567.0	1390.0	
	89	267.3	7.6	25.7	0.23	820		11.0	1.4	45.7	168.0	40.6	169.0	448.0	
	102	366.7	7.3	19.5	0.21	1300							188.0		
	110	295.7	7.1	14.3	0.18	2000		11.8	1.6	62.3	202.0	54.9	202.0	574.0	
	117	130.3	7.1	12.48	0.32	7100							270.0		

center toward suburbs of Urumqi, China. Concerning the potential dilution by greater storage of water in Luleå snowbanks, the larger amount of snow in Luleå (i.e., the mean SWE value in Luleå was 2.7 times that in Trondheim) would have diluted the constituent concentrations and reduced pH. However, this is not the case for Cd and Pb, which both occurred at higher concentrations in Luleå. Lower concentrations of Cd and Pb in Trondheim were attributed to the implementation of more stringent environmental legislation introduced during the 1990s (e.g., the Cadmium Directive 91/338/ECC; phasing Pb out of gasoline, OECD and UNEP (1999)), thus after the completion of the Luleå study in 1995. The effects of Pb phasing out on reduced concentrations of Pb in highway

runoff were demonstrated, e.g., by Viklander and Marsalek (2011) and Gunawardena et al. (2015). Road salt applications in Trondheim contributed to much higher concentrations of Cl and Na and higher values of EC, compared to those reported in Luleå, where no salt was applied. As discussed later, the concentrations of Cl and Na are clearly not correlated with traffic (AADTs).

When comparing the chemistry of the roadside snow with samples collected from the reference sites unaffected by traffic, heavy impacts of traffic on snow quality were obvious (Table 2). Roadside snow samples exhibited consistently higher pH values, compared to those from the reference sites (Table 2). The higher pH and alkalinity values in the roadside snow can be attributed to the concentrations of TSS, which were higher in the roadside snow samples. The same tendency was also noted for trace metals (i.e., Cd, Cr, Cu, Ni, Pb, W, and Zn) in the roadside snow, whose concentrations exceeded those in reference site snow by one to two orders of magnitude (Table 2). Thus, it appears that the mass of metals scavenged by falling snow from the air is insignificant compared to the metal accumulations contributed by traffic. The same finding was also reported by Kuoppamäki et al. (2014), who noted that traffic was the dominant source of snow pollution in cities, with pH, alkalinity, and conductivity all displaying trends closely related to traffic density, particularly within a 5-m band adjacent to the road; outside this band, the snow pollution concentrations drastically decreased. In highly industrialized areas, Li et al. (2015) showed that in spite of the importance of traffic pollution sources, anthropogenic effects including factory pollution emissions, construction fugitive dust, and oil refinery emissions may also have considerable impacts on the pollution accumulating in urban snow.

Mass loads of the trace metals studied were ranked similarly in both cities studied, and could be presented, in a descending order, as Zn > Cu > Cr > Ni > Cd. An exception to this order was noted for Pb in Luleå, reflecting the fact that those data predated the lead phasing out from gasoline (OECD and UNEP 1999). W was studied only in Trondheim, where W concentrations were ranked just below those of Ni.

3.2 Statistical Data Analysis

3.2.1 Exploratory Analysis

In the first stage of exploratory analysis, data at individual sites were examined separately using linear regression (Fig. 2) to investigate if the accumulation of constituents in roadside snow increased linearly with time, as assumed in further statistical data analyses of the combined city datasets serving for developing general predictive relationships between the individual constituents and independent variables (i.e., SRT, AADT, and SWE).

Linear regression analyses were performed on data for consecutive sampling days showing continuous snow accumulation (with no disturbance), which were three sampling days in Trondheim (SRTs varying from 25 to 53 days) and four in Luleå (SRTs varying from 18 to 75 days). The same trend was observed for all the constituents studied, except Na and Cl. The results of regression analysis showed linearity ($0.65 < R^2 < 0.95$) in both cities and all the sites, except site T1, where the SWE was fluctuating during the analyzed period. As an example of these analyses, plots of Cu accumulations in sampled snow are shown in Fig. 2.

Furthermore, spatial variations affecting the dataset characteristics were quantified by analyzing metal loads normalized by traffic densities (AADTs), yielding metal accumulations in roadside snow per vehicle. Plots of such data are shown for Cu in Fig. 3; all the other constituents studied showed similar tendencies, except for Cl and Na. In general, mass accumulations were increasing with SRT in a consistent way, except for the two sites with the lowest traffic loads (L1 and T1).

In the second stage of exploratory analysis, the relationships among the experimental variables were examined by applying the PCA. The loading plot of constituent concentrations and their predictors (i.e., AADT, SRT, and CTV) produced by PCA provides an overview of correlations among the variables, with the first and second orthogonal components accounting for 58 and 15.8 % of the data variability, respectively (Fig. 4). Acute angles between any two vectors plotted in Fig. 4 show correlation between those variables. Thus, Fig. 4 indicates possible correlations between the group of trace metals and such predictors as AADT and SRT, or their product CTV. Among such correlations, the strongest ones were noted for the well-known traffic-related metals (Cd, Cr, Cu, Ni, Pb, Zn) and the weakest ones were noted for less well-understood W. Practically, no correlation was noted for Na and Cl. The CTV variable had a longer vector than AADT and SRT, thus showing that the interaction between these two predictors is
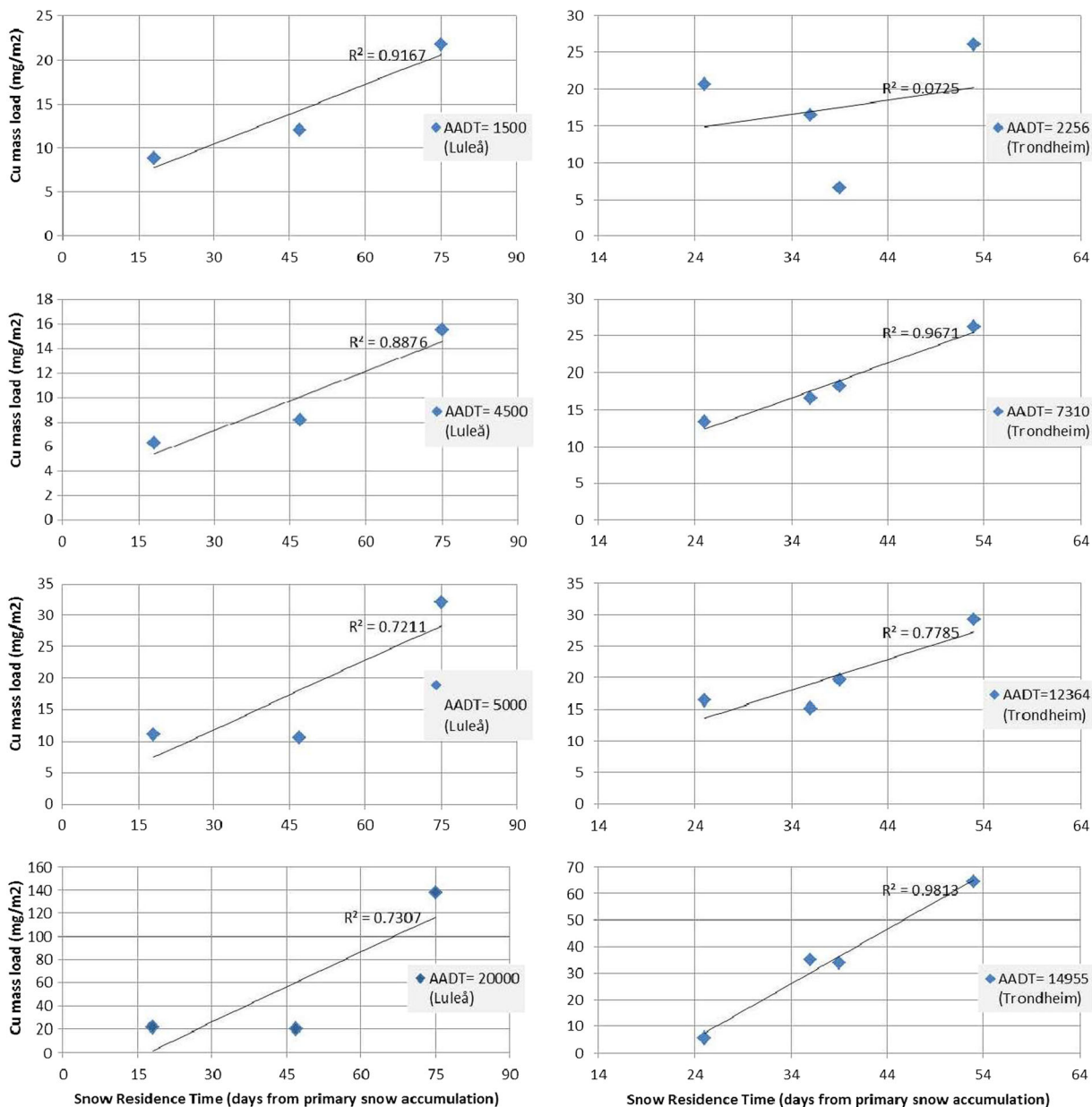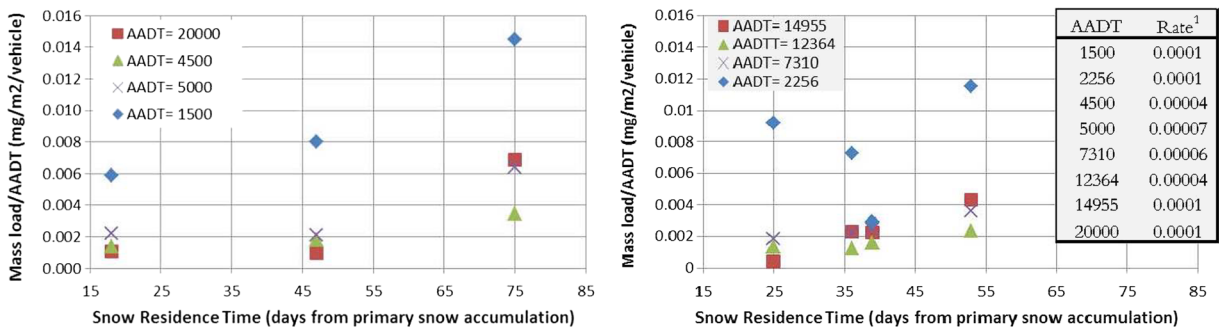
Fig. 2 Cu unit-area mass loads versus SRT, with linear regression lines and coefficients of determination (R^2) displayed for individual sites in Luleå and Trondheim

important and could be captured by the combined variable (CTV). Salt chemicals Cl and Na were not correlated to traffic density (AADT), because road salt is applied according to snowfalls, which occur independently of traffic descriptors.

The results summarized in the loading plot generally agree with those of other studies, showing significant cause-effect relationships between traffic characteristics and trace metal accumulations in roadside snow, before their potential release with snowmelt runoff. For example, Helmreich et al. (2010) studied the seasonal changes of urban road runoff quality and showed that pollutant concentrations of TSS and heavy metals in snowmelt exceeded those in rainfall runoff multiple times. It was also reported that the dissolved fraction of heavy metal did not change seasonally. Therefore,

Fig. 3 Accumulated Cu unit-area mass loads per vehicle versus SRT, for all sites in Luleå (*left*) and Trondheim (*right*). [1]Cu accumulation rates at study sites with various AADTs in Luleå and Trondheim [mg/m^2/vehicle/day]

under stable conditions, the period of pollutant accumulation (equivalent to the antecedent dry period in runoff studies) can conceivably be set equal to the SRT, whose significant correlation with trace metals can be observed in the PCA plot in Fig. 4. This assumption is also supported by Li and Barrett (2008) and Finney et al. (2010), who identified the AADT and the antecedent dry period (ADP) as two significant factors affecting the mass loads

of constituents accumulated on the road surfaces during dry weather.

3.2.2 Prediction of Trace Metal Accumulations in Roadside Snow

Multiple Regression Model Employing SRT, AADT, and SWE as Predictors A general linear model was applied to the observed data assuming that no snowmelt occurs
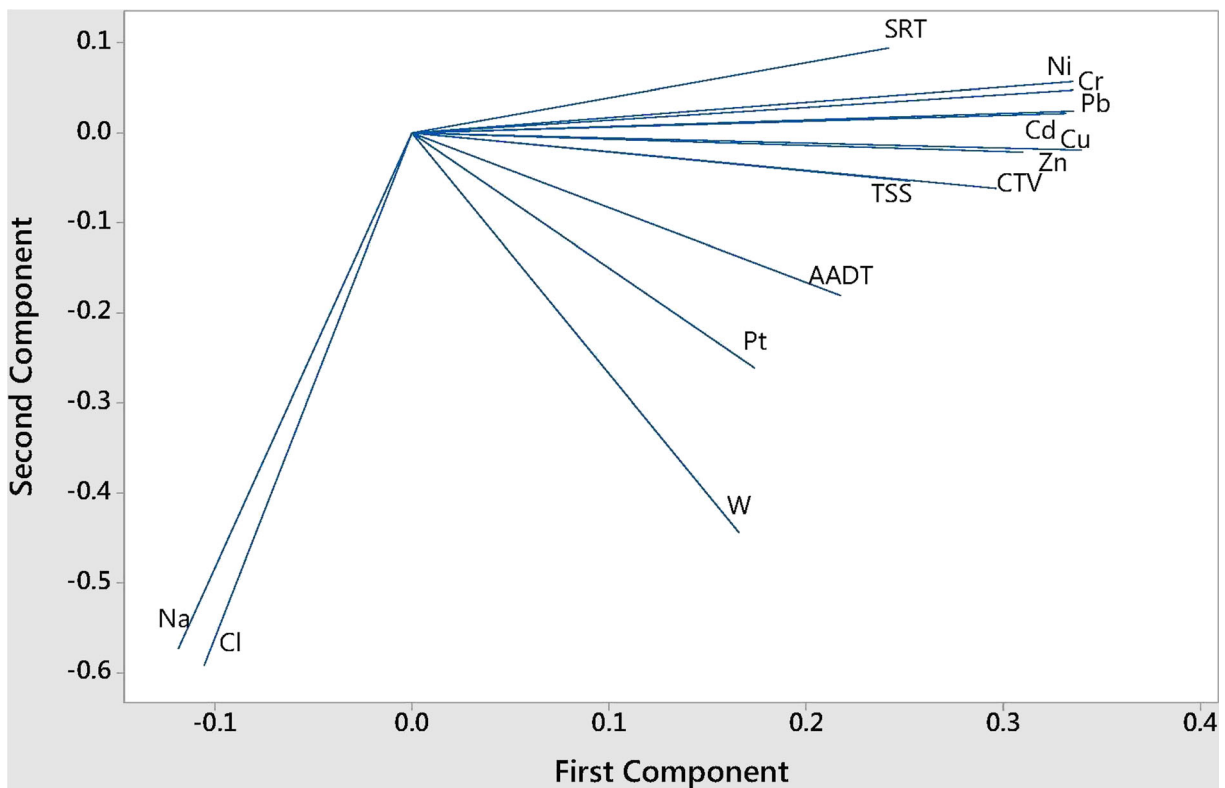
Fig. 4 PCA loading plot for combined Luleå/Trondheim data

below 0 °C, recognizing that this assumption may be better applicable in Luleå, where no road salt is used, than in Trondheim, where road salt is used. The concerns about salt lowering the eutectic temperature are borne out by temporal variations of SWE, indicating that it was not rising consistently but decreasing on some occasions, even during the freezing periods. A number of studies have demonstrated that snow and ice freezing point can be lowered by using deicing chemicals. For example, Ketcham et al. (1996) reported that a 23.3 % NaCl concentration lowered the eutectic temperature to −21 °C, and Zhang et al. (2009) recommended a practical salt application range of temperatures between −10 and 1 °C. As illustrated in Fig. 1, about half a time, temperatures during the study period were above −10 °C, and depending on salt concentrations, snowmelt could be expected on a number of occasions, even though it would not be detected by flow meters. The evidence of such snowmelt episodes at individual study sites is provided by the SWE data (Fig. 1, bottom panel), which fluctuated even during the subzero temperature period. Sublimation may be another reason for water losses from the snowbanks, though several studies showed that it represents an insignificant fraction (e.g., 1 % for a rural catchment) of snow ablation (Szczypta et al. 2015). Hence, the assumption of no melt during the observation period will introduce some uncertainties into the results.

On the other hand, experiments with repeated snow thaw/freeze cycles indicate that dissolved constituents leave the snowpack with early snowmelt (i.e., the preferential elution), but those attached to particulates remain in the snowpack until the final melt (Schöndorf and Herrmann 1987). This mechanism would tend to retain most of the trace metal burden, typically attributed to the particulate fraction, in roadside snow until the final snowmelt and minor intermittent snowmelt episodes would not significantly affect metal accumulations. Dissolved metals would leave with early snowmelt, and deicing salts (applied in Trondheim) were found to increase the dissolved metal content of snow (Reinosdotter and Viklander 2007).

In setting up the multiple regression model, SWE was considered as one of the predictors.

Statistical tests showed that SWE and SRT were not strongly correlated, with Pearson correlation coefficient of 0.36 and a low coefficient of determination ($R^2 = 12.6$ %) for a linear regression between predictors SWE and SRT. Thus, SWE was not considered as a multicollinearity phenomenon and could be safely included in the multiple regression analysis (O'Brien 2007).

Results in Table 3 indicate that TSS and trace metals (Cd, Cr, Cu, Ni, Pb, and Zn) are well correlated (p values <0.05) with all the predictors, i.e., SRT, AADT, and SWE. Furthermore, multiple regression models could be used to explain the correlations in the form of a polynomial equation, which could serve as a prediction tool. R-squares (i.e., adjusted and predicted) calculated by the regression models are also presented in Table 3. An average adjusted R-square of 67.75 % reflects the explanatory weight of the regression model, and the predicted R-squares, with an average of 62.18 %, show the goodness of fit for the predicted response of a new observation. This was done by removing one of the actually observed values (one at a time) from the data set and comparing it with the associated modeled response. The predicted R-squares showed the numerical percentage of predicted responses, which were explained by the model, with a 95 % confidence. Comparison of adjusted and predicted R-squares shows differences equal to or less than 9 %, which indicates that the model explains well predictors and is not over-fitted (Minitab 2013). Concerning the individual elements, the best model fits can be arranged

Table 3 Multiple regression model for trace metal and TSS and predictors (i.e., SRT, AADT, and SWE) for combined data sets from Luleå (1995) and Trondheim (2010)

Model	Multiple regression		
Constituent	R^2	Adj-R^2	Pre-R^2
TSS	0.66	0.63	0.6
Cd	0.71	0.68	0.63
Cr	0.64	0.61	0.55
Cu	0.75	0.72	0.65
Ni	0.65	0.62	0.56
Pb	0.72	0.7	0.67
Zn	0.77	0.75	0.67
Pt	0.74	0.58	0.44
W	0.73	0.66	0.48

Adj adjusted, *Pre* prediction

Table 4 Predictor coefficients and their interactions estimated by multiple regression models

	C	a	b	c	d	e	f
TSS	1.78	1.78	−1.96E-02	8.26E-04	5.95E-02	1.20E-05	−2.39E-04
Cd	−2.46	−2.46	−2.25E-03	2.60E-05	1.03E-02	1.00E-06	−5.00E-05
Cr	0.07	0.07	1.00E-04	1.65E-04	2.83E-02	6.00E-06	−2.01E-04
Cu	−0.02	−0.02	−2.30E-03	1.27E-04	3.21E-02	6.00E-06	−2.00E-04
Ni	0.11	0.11	−2.20E-03	1.10E-04	2.05E-02	4.00E-06	−1.29E-04
Pb	−0.93	−0.93	2.05E-03	5.10E-05	1.02E-02	1.00E-06	−4.40E-05
Zn	−0.20	−0.2	4.00E-03	1.57E-04	4.78E-02	9.00E-06	−3.11E-04
Pt	0.01	0.01	−1.45E-04	0.00	−1.11E-04	0.00	1.00E-06
W	0.75	0.75	−7.40E-03	3.20E-05	7.30E-03	2.00E-06	6.20E-05

in the following descending order: Pb > TSS > Cd > Cr > Ni > Zn > Cu.

The results further indicate that TSS, Pb, Cd, Cr, Ni, Zn, and Cu accumulate over time in roadside snow, as a result of continuous input of traffic pollution. The rate of trace metal accumulations in snow increased with increasing AADT and SRT, for several metals, and was further influenced by SWE. Owing to the satisfactory fits of the multiple regression models, mass loads of Cd, Cr, Cu, Ni, Pb, Zn, and TSS may be expressed as functions of SRT, AADT, and SWE, with fairly high certainty, and the prediction model can be defined by a polynomial equation with specific coefficients for individual constituents:

$$
\begin{aligned}
ML = {}& C + (a \times SRT + b \times AADT) \\
& + (c \times SWE) + (d \times SRT \times AADT) \\
& + (e \times SRT \times SWE) \\
& + (f \times AADT \times SWE)
\end{aligned} \quad (2)
$$

where ML [mg/m^2] is the predicted unit-area mass load and C, a, b, c, d, e, and f are the coefficients generated by the regression model and listed in Table 4.

Linear Regression Model with a Single Predictor, CTV The results of PCA showed that CTV (=AADT× SRT) might be a better descriptor of the metal accumulation in roadside snow, than SRT and AADT, considered separately. Furthermore, consideration of CTV facilitates prediction of the mass loads of TSS and trace metals using a small number of independent variables, which is generally a feature of parsimonious models. Also, this approach has another advantage of expressing metal mass loads as a linear function of CTV, whose estimated slope of the regression line yields the accumulation rate, which has some practical use. Thus, linear regression analyses were performed to further evaluate the CTV as an independent variable, and the results of such analyses are summarized in Table 5.

Using this method makes the correlation less evident for metals (i.e., R^2 ranging from 0.37 to 0.66), when compared with the previous analyses (i.e., multiple regression). However, considering the high variations in snow quality observed in this study, the results suggest that there is a potential of using CTV to estimate metal accumulations by conducting more

Table 5 Linear regression correlation coefficients (R^2) between TSS and trace metal unit-area mass loads and the snow accumulation period traffic volume (CTV) for Luleå, Trondheim, and the combined datasets

Constituent	Luleå CTV [vehicles]	Trondheim CTV [vehicles]	Combined CTV [vehicles]
TSS	0.48	0.60	0.35
Cd	0.61	0.42	0.49
Cr	0.44	0.39	0.24
Cu	0.55	0.55	0.57
Ni	0.46	0.37	0.35
Pb	0.66	0.50	0.57
Zn	0.60	0.60	0.60
W	–	0.78	–

field measurements. In any case, it is feasible to express unit-area mass loads, ML [mg/m^2], of Cd, Cr, Cu, Ni, Pb, and Zn as functions of CTV (Eq. 3):

$$ML = f_{acc} \cdot CTV + ML_{BG} \qquad (3)$$

where f_{acc} is the accumulation rate coefficient [mg/ (m^2 vehicle)] and ML_{BG} [mg/m^2] is the background mass load due to other sources than traffic.

Figure 5 shows the estimated accumulation rate coefficients for Luleå, Trondheim, and the combined datasets and the previously estimated accumulation rates from Oslo (Bækken 1994). Accumulation rate coefficients for Luleå and Trondheim were significant at a level of 0.05. For Cu and Zn, the accumulation rate coefficients were comparable for the two cities studied; for other parameters, the differences were larger. The variations in the accumulation rates can be caused by local differences in the type of asphalt pavement, tires, distribution of vehicles, and traffic density during the winter months, hydrologic characteristics, and snow management practices, including the use of de-icing and traction agents.

Even though the proposed parsimonious model (Eq. 3) does not account for the complexity of snow hydrology and quality, it is well suited for obtaining first-order approximations of the net accumulation rates of contaminants in roadside snow during cold periods. By applying unit-area accumulation rate coefficients (Fig. 3), it may be possible to predict the mass of constituents accumulated in urban roadside snow without extensive sampling and analyses. Potentially, this approach would assist municipalities in distinguishing between heavily and less polluted snow, on the basis of SRT, and thereby helps decide where to dispose snow of various age bearing different levels of contamination. Note that relatively clean fresh snow can be disposed locally, even in receiving waters, but heavily contaminated old snow may require disposal at special snow storage and disposal sites (Exall et al. 2011).

4 Conclusions

An experimental exploratory study of pollution accumulation in roadside snow exposed to traffic was conducted in the city of Trondheim (Norway) by means of snow sampling and statistical analysis of measured data. Toward this end, snow samples were collected at five sites on seven dates between January and March 2010 and analyzed for snow physical properties (i.e., SWE) and 12 water quality constituents (pH, alkalinity, TSS, Cl, Na, and seven trace metals, Cd, Cr, Cu, Ni, Pb, W, and Zn). The resulting dataset was compared to, and enhanced by including, an earlier similar dataset from Luleå (Sweden), where snow samples were collected at five sites on nine dates between January and April 1995, and analyzed for pH, TSS, Cd, Cr, Cu, Ni, Pb, and Zn. In general, the concentrations in roadside snow were higher in Trondheim than in Luleå, because of higher AADTs in Trondheim (by 20 %, on average) and lower dilution of deposited traffic contaminants by smaller snow depths, representing about one third of those observed in Luleå. There were two exceptions to this finding, higher Cd and Pb concentrations in Luleå, explained by the fact that the collection of these data (1995) predated the full implementation of stringent environmental legislation controlling environmental releases of Cd and Pb during the 1990s.

Regression analyses carried out for individual study sites indicated a linear trend in unit-area constituent accumulations over time ($0.65 < R^2 < 0.95$), which satisfied the assumption of linearity for further statistical data

Fig. 5 Accumulation rates of six trace metals and TSS derived from a regression model using CTV as the sole independent variable
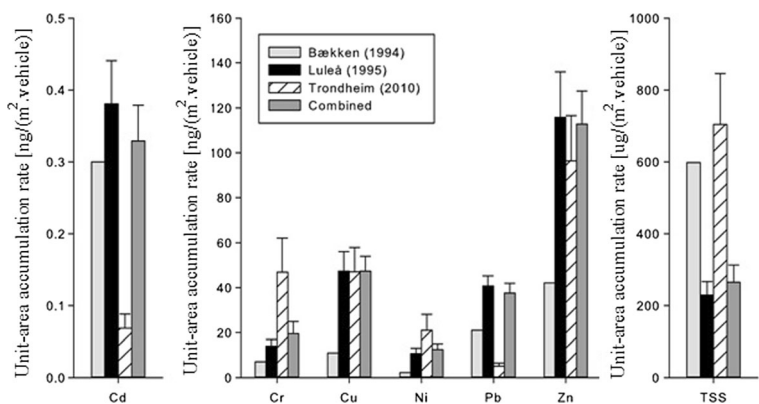
analyses. A PCA model including the constituent mass loads and the three predictors (AADT, SRT, and CTV) illustrated the cause-effect relationship between traffic and trace metal accumulations; Cl and Na originating from road salt applications did not follow this trend. CTV (the product of AADT and SRT) correlated more strongly with trace metal mass loads than AADT and SRT, which showed the importance of interactions among the predictors.

Two types of models for predicting the trace metal accumulations in roadside snow were developed: a linear regression model and multiple regression models. The linear regression model used CTV as a single predictor and showed moderate certainty for metal accumulation predictions ($0.37 < R^2 < 0.66$). However, this model could be further improved by collecting more field data. A multiple regression model was developed using SRT, AADT, and SWE as predictors, which correlated well with the metal unit-area mass loads ($0.64 < R^2 < 0.77$). This model can be described by a polynomial equation with specific coefficients for individual constituents and used to predict contaminant accumulation in roadside snow (prediction $0.44 < R^2 < 0.67$) with a greater accuracy than the linear regression model, using CTV as single predictor. Thus, the study enhanced the understanding of metal accumulations in roadside snow and led to the development of parsimonious models of trace metal accumulations in roadside snow.

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