Particle Pathways during Urban Snowmelt and Mass Balance of Selected Pollutants

C. Westerlund[1*], M. Viklander[1], K. Nordqvist[1], H. Galfi[1] and J. Marsalek[2]

[1] *Department of Civil, Environmental and Natural Resources Engineering, Lulea University of Technology, S-971 87 Lulea, Sweden*
[2] *National Water Research Institute, Environment Canada, 867 Lakeshore Rd, Burlington, ON L7R 4A6, Canada*
**Corresponding author, e-mail cam@ltu.se*

ABSTRACT
The pathways and mass balance of selected pollutants, released during the snowmelt process, were investigated for urban bulk snow placed in small, intermediate, and large-scale lysimeters. The results showed that low percentages of TSS (total suspended solids) and heavy metal (Cu, Zn, Pb) loads contained in snow were transported with snowmelt, the rest remained in situ with the particulate residue. The TSS loads transported with snowmelt were 3, 3.4, and 4.8% of the initial TSS mass in the small, intermediate and large lysimeters, respectively. Particulate heavy metal loads transported with snowmelt, during the whole melting process, were measured in the intermediate lysimeter for copper and zinc, and for lead in the large lysimeter. The measured mass loads in snowmelt leaving the intermediate lysimeter were 7.5 and 7.2% for copper and zinc, respectively, and 1.7% for lead leaving the large lysimeter. The remainder of the loads stayed in situ with the particulate residue. The loads transported with snowmelt were independent of the initial TSS and metal concentrations in bulk snow. These findings have implications for siting and operating snow disposal facilities; most of the initial TSS and particulate heavy metal loads can be retained on site, rather than released with snowmelt into the receiving environments.

KEYWORDS
Urban drainage; total suspended solids (TSS); heavy metals; mass balance; snowmelt; residual particles

INTRODUCTION
Snow and snowmelt-induced runoff along roads and highways often contains major loads of pollutants, such as sediment and total suspended solids (TSS), heavy metals, bacteria, salts, and trace organic compounds originating from anthropogenic activities, such as traffic (vehicle component wear, fluid leakage, tire and pavement wear), corrosion and atmospheric deposition (Hvitved-Jacobsen and Yousef, 1991; Viklander, 1997). The resulting environmental impacts on receiving waters vary according to the loads and concentrations of pollutants in snowmelt runoff as well as according to the sensitivity and types of receiving environments. Commonly reported impacts include increased turbidity, reduced oxygen levels, eutrophication, and toxic levels of chloride, heavy metals and trace organic substances exerting adverse effects on biota (Marsalek *et al*., 2003). A large fraction of road runoff pollutants, such as heavy metals and PAHs (polycyclic aromatic hydrocarbons), are particle bound and transported with TSS (Lau and Stenstrom, 2005). Furthermore, the pathways of

pollutants in a cold climate urban environment are rather complex reflecting such sources of pollutants as local land use activities, as well as wet and dry depositions resulting from local and long-rage air transport. It was also reported that with respect to atmospheric scavenging, snow is more polluted than rain when reaching the ground, because of its larger specific surface and lower fall velocities (Gjessing and Gjessing, 1975).

Additional loads of particles and pollutants arise during cold climate due to, for example, cold starts of vehicle engines, pavement wear by studded tires, and heating (Malmqvist, 1983). Winter road maintenance contributes to the urban pollution through vehicle exhausts during snow-handling operations, and the application grit and salts, which increase solids loads in snowmelt runoff as well as the corrosion of road guard rails and other metal structures along roads and highways (Malmqvist, 1983).

Viklander (1997) and Reinosdotter (2007) noted that the choice of methods for removing, transporting, and dumping used snow will greatly affect the pathways of pollutants and the loads transported. In particular, it is of interest to determine what fraction of the pollutants in the snow will be transported with the snowmelt runoff and what fraction will stay with the solid residue in-situ on the surface, and this partitioning between the dissolved and particulate phases depends on the pollutant chemistry and the concentrations of solids in the snowmelt (Viklander, 1999; Glenn and Sansalone, 2002; Reinosdotter; 2003; Westerlund *et al.*, 2003). For example, Viklander (1996) reported that in a laboratory study, the percentages of Cu, Pb, and Zn leaving the snow deposit with snowmelt were 28, 42, and 30%, respectively. However, much different results were found in a pilot scale study (10, 0.5, and 30%) and in a full-scale study (6.5, 1, and 10%), for the same metals (Viklander and Malmqvist, 1993; 1994).

Defining the pathways and loads of solids and the associated pollutants during snowmelt is of great interest when developing pollution-mitigation measures serving to prevent polluted runoff from strongly impacting on receiving waters. To develop some guidance for such mitigation measures, a study of pollutant pathways and loads, during the snowmelt process, was performed in small, intermediate and large lysimeter installations, with the objective of determining the pollutant pathways and loads in lysimeters mimicking the melting of urban snow deposits.

METHODS

Mass balance calculations

The notation used in mass balance calculations is shown in Fig. 1. A snowpack in the lysimeter contains a mass of water, M_{SW}, and a mass of suspended solids, M_{TSS}. A mass balance equation indicates that the initial mass of water should equal the mass of snowmelt water leaving the lysimeter plus any losses, such as ablation. Similarly, the initial mass of TSS should equal the mass of suspended solids leaving the snowpack with snowmelt plus the mass of TSS residue left on the bottom of the lysimeter at the end of the snowmelt process. Finally, the same consideration would apply to heavy metals in snow.

$$M_{SW} = M_{SM} + M_{RES} + M_L \qquad (1)$$

Where M_{SM} is the mass of snowmelt, M_{RES} is the mass of liquid residual left on the lysimeter plastic sheet, and M_L is the mass loss (e.g., by ablation).

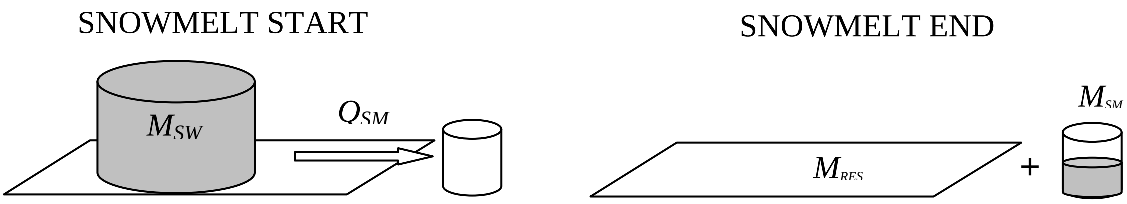

Figure 1. Schematics of snowmelt experiments

Data collection and sampling procedures

The small, intermediate and large lysimeter snowmelt experiments were performed at different times in 1991, 2004 and 2006, respectively. All three experimental setups were well controlled with impermeable lysimeter bottoms. Polluted snow from roadsides in Luleå, in the north of Sweden, with traffic intensities ranging from 7,400 to 21,900 vehicles/day, was collected and placed into lysimeters. For the three different size lysimeters, the initial snow volumes, weights, densities, TSS and heavy metal concentrations were measured and solids loads were calculated. For the small scale lysimeter, snow was divided into nine cylindrically shaped sub-samples of 30 litres each. The snowmelt runoff volume was measured and TSS were sampled during the entire snowmelt process with temperature settings between 5 and 20 °C. The intermediate scale lysimeter was a single unit which was placed inside an unheated garage, well exposed to and affected by the outside temperature. The snow pile melted during a three to four weeks period. The large scale lysimeter was setup on an asphalt pavement plot covered with a polyethylene film. At the lowest point of the plot was a polyethylene pipe that drained runoff into a well, where measuring equipment had been placed. Snowmelt flows were continuously measured using a tipping-bucket type flow meter during low flows and a standard water flow meter during high flows. Samples for TSS and metal analyses were collected twice a day, two days a week throughout the entire snowmelt period. For all the three different sized lysimeters, all snowmelt was collected and analysed for concentrations of TSS and heavy metals. After melting, the residual particles remaining on the plastic sheets were collected and weighed.

Laboratory analyses

After collection, the samples were immediately transported to and processed (within a few hours after collection) in the laboratory at Luleå University of Technology for pH, conductivity, and total suspended solids (TSS). The TSS concentration was measured according to the standard method SS-EN872, which has the status of the European Standard EN 872: 1996. This standard specifies a method for determination of TSS by filtration through a glass fibre filter. The lower limit of detection is 2 mg/l; no upper limit has been established. The concentrations of TSS were determined by immediately filtering the samples through a Whatman GF/A filter (Whatman International Ltd., Maidstone,UK). The filter has an average nominal pore size of 1.6 µm, a thickness of 0.25 mm, and a rate of filtration of 13 ml/s. The filters were weighed before the filtration and subsequently dried and weighed again after the filtration to determine the concentration of TSS (EN 872, 1996 and SIS, 1996). Copper and zinc concentrations in the intermediate lysimeter were analysed with a plasma mass spectrometer (ICP-MS) with detection limits for Cu and Zn of 1 and 4 µg/l, respectively. The lead concentrations in the large lysimeter were analysed after addition of ultra pure water (10 ml) and 3 M sodium acetate pH (200 ml) by differential pulse anodic stripping voltammetry at a mercury drop electrode.

RESULTS AND DISCUSSION

Water mass balance

Earlier investigations showed that in melting snowpacks, the pollutant mass balance is greatly affected by the water mass balance since water serves as the pollutant vector (Westerlund et al., 2007). Subsequently, to improve the understanding of pollutant pathways a water mass balance has to be established first as described earlier by Eq. (1).

To determine the water mass equivalent for the initial snowpack in the small lysimeters (M_{SW}), the total snow volume and mass of the initial snowpack were measured and the density was calculated (Table 1). Due to the size of the intermediate and large scale lysimeters, measurements of mass and volume were made only for representative sub-samples taken from each lysimeter snowpack, and the density was estimated as an average of the sub-sample readings. The amount of water left on the plastic sheets after snowmelt, M_{RES}, was considered negligible for all the three lysimeters.

Table 1. Snow and snowmelt laboratory experiments: Measured and estimated data

	Small[1]	**Medium**	**Large**
INITIAL SNOWPACK			
Volume (m^3)	0.032**	0.83**	202**
Weight (kg)	20**	310*	113,000*
Density (kg/m^3)	625*	373*	560*
M_{SW} (m^3)	0.020*	0.310*	113*
SNOWMELT			
M_{SM} (m^3)	0.0202**	0.287**	100**
RESIDUAL			
M_{RES}	Negligible	Negligible	Negligible

[1] Average of 9 lysimeters
*Calculated
**Measured

To satisfy the water mass balance, M_{SW} (i.e., the initial snow mass) and M_{SM} (total snowmelt mass) are assumed to be about equal, since M_{RES} (residual water mass) and M_L , potential losses by snow ablation (applicable to the large lysimeter, exposed to open air), were considered negligible. According to Table 1, M_{SW} was 100, 93 and 88 % of M_{SW} , for the small, intermediate and large lysimeters, respectively. There were two sources of errors in the measures water mass terms: Water mass measurements by sampling and possible losses due to ablation (or other losses), particularly in the case of the large lysimeter. In general, all the snowpack parameters in the small lysimeter were measured and any errors should be negligibly small. The volume of the intermediate and large lysimeter snowpacks was measured, but their density was only sampled and this could be a significant source of error, in spite of taking good care in density measurements in snow samples. The total mass of snowmelt, M_{SM}, was measured fairly accurately using both volumetric and flowrate measurements. For all the lysimeters, there was a small risk of leakage, contributing to errors in mass balance. However, in the final assessment, the calculated water mass balance was considered acceptable for all the three lysimeters and suitable for addressing the pollutant mass balances.

Pollutant mass balance

Pollutant mass balance can be expressed as the initial mass of the pollutant PL in bulk snow, M_{SWPL}, obtained by multiplying the initial snowpack volume by the mean PL concentration in snowpack samples, which should equal the PL mass leaving with snowmelt, M_{SMPL} , plus any PL residue left on the lysimeter bottom at the end of the experiment, M_{RESPL}. From the measurements of pollutant (TSS and heavy metal) concentrations in the initial snowpack as well as continuous measurements and sampling of the snowmelt, mass loads for the specified pollutants were calculated (Eq.2). For both TSS and heavy metals, the different start concentrations of the three lysimeter setups depended on the different origins and the different time of collecting the snow.

$$M_{RESPL} = M_{SWPL} - M_{SMPL} \qquad (2)$$

TSS

The difference in TSS mass in the initial snow and the snowmelt was assumed to be the residual left on the bottom of the lysimeters after all snow has melted. The mass of TSS transported by snowmelt represented 3, 3.4 and 4.8% of that in the lysimeter snow, for the small, intermediate and large lysimeters, respectively. Consequently, the different initial TSS concentrations in snow did not seem to influence the TSS mass transported by snowmelt. Similar results were reported by Johannessen and Henriksen (1978), who concluded that the release of pollutants from snow did not depend on the initial pollutant concentration in snow. To calculate the TSS mass in the residual, Eq.2 was used and the results are presented in Table 2.

Table 2. TSS concentrations and loads in the bulk snow, snowmelt and residual for the small, intermediate and large lysimeters

	Small [1]	**Intermediate**	**Large**
INITIAL SNOW			
TSS concentration (mg/l)	6567	2000	630
TSS mass (kg)	0.131	0.620	71
SNOWMELT			
TSS concentration (mg/l)	200	73	34
TSS mass transported (kg)	0.004	0.021	3.4
RESIDUAL			
Calculated residual TSS mass (kg)	0.127*	0.599*	67.6*
Measured (TSS + sediment) residual mass (kg)	0.547**	3.064**	1870**

[1] Average of 9 lysimeters
*Calculated TSS load in the residual
**Measured total solids load in the residual (i.e., TSS plus sediment)

A comparison of the calculated TSS residual mass with the measured mass of total solids in the residual (i.e., considering both TSS and sediment), it can be noted that 23% of the total solids load was represented by TSS in the small-scale lysimeter and the rest of the particles were settleable solids, which had not been suspended during the snowmelt process. The TSS percentages of the total solids in the intermediate and large lysimeters were 18 and 4%, respectively. It was noted that when preparing samples for TSS analysis, large particles in the sample container did not become suspended when stirring the container volume and extracting sub-samples for TSS filtration, but remained on the bottom of the sample container. Consequently, such coarse particles are not included in the standard method of TSS analysis. Thus estimating total solids loads in the initial snow without considering settleable solids may lead to significant underestimation of solids loads in urban snow and snowmelt, with potential consequences for underestimating the costs of street cleaning/sweeping, and the risk of sewer pipe clogging and coarse solids deposits in receiving waters.

Finally, the data in Table 2 can be used to estimate total solids masses in the initial snow pack, by adding the TSS mass in snowmelt (i.e., TSS mass transported by snowmelt) to the mass measured in the final residual (i.e., measured [TSS + sediment] residual mass). This calculation yielded the total mass of solids in the initial snowpack as 0.551, 3.085 and 1873.4 kg, for small, intermediate and large lysimeters, respectively. When comparing these values to the measured residual mass, the mass of TSS transported by snowmelt is negligible and generally represents less than 1% of the total solids load in snow.

Heavy metals

The same mass balance approach was applied to heavy metal concentrations and loads in the intermediate and large scale lysimeters. Copper (Cu) and zinc (Zn) concentrations were measured in the intermediate lysimeter and lead (Pb) concentrations in the large lysimeter, in both, the initial snowpack and during the entire melting process. Furthermore particulate mass loads were calculated for all the three metals in the snowmelt runoff (Table 3). The particulate load transported by snowmelt from the intermediate scale lysimeter was 7.5% for Cu and 7.2% for Zn, compared to the initial snow burden. The particulate load percentage for the snowmelt runoff from the large lysimeter was 1.7% for Pb. This low percentage of released lead agrees with the fact that lead is strongly fractionated to solids. As noted earlier for TSS, different initial concentrations of metals in snowpack did not seem to influence the heavy metal loads transported by snowmelt.

Table 3. Heavy metal concentrations and calculated particle loads in the initial snow, snowmelt and residual for the medium and large-scale lysimeters.

	Cu	**Zn**	**Pb**
INITIAL SNOW			
Metal concentration (µg/l)	210	574	790
Mass in snow (g)	0.065	0.178	89.5
SNOWMELT			
Conc. (µg/l)	17.1	44,9	15
Transported mass (g)	0.0049	0.0129	1.5
RESIDUAL			
Residual mass (g)	0.0601*	0.1651*	88*

*Calculated particle load in residual

CONCLUSIONS

The TSS mass balance for the small, intermediate and large lysimeters used in this study showed that the TSS load transported by snowmelt outflow from the lysimeters was 3, 3.7, and 4.8% of the initial TSS load, respectively. Similar estimates for heavy metal releases from the lysimeters were 7.5 and 7.2% for Cu and Zn, respectively (observed in the intermediate lysimeter experiments) and 1.7% for Pb, in the case of the large lysimeter. Furthermore, the loads transported with snowmelt were independent of the initial pollutant concentrations in bulk snow for both TSS and heavy metals. It was also noted that the TSS analysis did not adequately described the total load of solids contained in bulk snow or in snowmelt runoff. Thus, estimates of the total solids loads in urban snow based on TSS analysis were significantly underestimated, because the TSS analysis generally neglects settleable solids. Further studies of analytical methods accounting for all particles sizes in urban snow and snowmelt are planned.

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