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Occurrence of tire and road wear particles in urban and peri-urban snowbanks, and their potential environmental implications

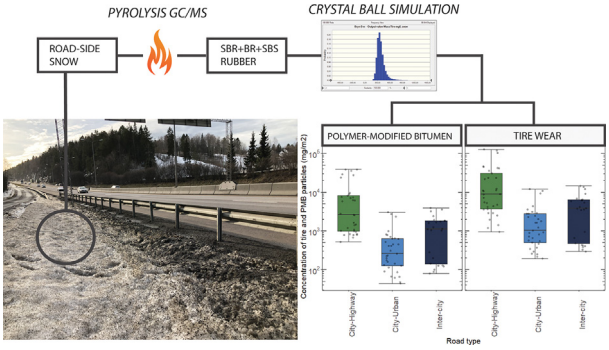
Elisabeth S. Rødland [a,b,*], Ole Christian Lind [b], Malcolm J. Reid [a], Lene S. Heier [c], Elvis D. Okoffo [d], Cassandra Rauert [d], Kevin V. Thomas [d], Sondre Meland [a,b]

[a] Norwegian Institute for Water Research, Økernveien 94, NO-0579 Oslo, Norway
[b] Norwegian University of Life Sciences, Faculty of Environmental Sciences and Natural Resource Management, P.O. Box 5003, NO-1432 Ås, Norway
[c] Norwegian Public Roads Administration, Construction, Postboks 1010, N-2605 Lillehammer, Norway
[d] Queensland Alliance for Environmental Health Sciences (QAEHS), The University of Queensland, 20 Cornwall Street, Woolloongabba, 4102, QLD, Australia

HIGHLIGHTS

- TRWP is estimated to be one of the largest sources of MP to the environment.
- Mass data of TRWP are limited and not before presented for roadside snow.
- Roadside snow from various road types were analysed with Pyr-GC/MS.
- Concentrations of TRWP showed large variations between and within road types.
- Speed and AADT explained the main variation observed.

GRAPHICAL ABSTRACT



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ABSTRACT

According to estimates put forward in multiple studies, tire and road wear particles are one of the largest sources to microplastic contamination in the environment. There are large uncertainties associated with local emissions and transport of tire and road wear particles into environmental compartments, highlighting an urgent need to provide more data on inventories and fluxes of these particles. To our knowledge, the present paper is the first published data on mass concentrations and snow mass load of tire and polymer-modified road wear particles in snow. Roadside snow and meltwater from three different types of roads (peri-urban, urban highway and urban) were analysed by Pyrolysis Gas Chromatography Mass Spectrometry. Tire particle mass concentrations in snow (76.0–14,500 mg/L meltwater), and snow mass loads (222–109,000 mg/m^2) varied widely. The concentration ranges of polymer-modified particles were 14.8–9550 mg/L and 50.0–28,800 mg/m^2 in snow and meltwater, respectively. Comparing the levels of tire and PMB particles to the total mass of particles, showed that tire and PMB-particles combined only contribute to 5.7% (meltwater) and 5.2% (mass load) of the total mass concentration of particles. The large variation between sites in the study was investigated using redundancy analysis of the possible explanatory variables. Contradictory to previous road studies, speed limit was found to be one of the most important variables explaining the variation in mass concentrations, and not Annual Average Daily Traffic. All identified variables explained 69% and 66%, for meltwater and mass load concentrations, respectively. The results show that roadside snow contain total suspended solids in concentrations far exceeding release limits of tunnel and road runoff, as well as tire particles in concentrations comparable to levels previously reported to cause toxicity effects in organisms. These findings strongly indicate that roadside snow should be treated before release into the environment.

* Corresponding author.
E-mail address: elisabeth.rodland@niva.no (E.S. Rødland).

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1. Introduction

Estimates suggest that tire and road wear particles (TRWP) emissions constitute one of the largest contributors to microplastics pollution (Boucher et al., 2020; Knight et al., 2020). The estimated release of synthetic rubber (Styrene Butadiene Rubber, SBR, and Butadiene Rubber, BR) from tire wear particles (TWP) varies in different countries. In Norway, the estimated release of microplastics from terrestrial sources is 19,000 t/year, where tire wear and road dust is estimated to contribute with 40% of the total estimated release (Sundt et al., 2021). In some countries, such as Australia, China, Denmark, Norway, Russia, Sweden and the United Kingdom (EAPA, 2018), it is common to add polymers to the bitumen of road asphalt in order to increase resistance to cracking and deformation (rutting) of the road surface (R.G. et al., 2012). This type of bitumen is referred to as polymer-modified bitumen (PMB). Various polymers are used for this purpose, such as styrene butadiene styrene (SBS), styrene ethylene butadiene styrene (SEBS), low-density polyethylene (LDPE), ethylene vinyl acetate (EVA), polypropylene (PP) and styrene isoprene styrene (SIS) (Chen et al., 2002; Giavarini et al., 1996; M. et al., 2003; Panda and Mazumdar, 1999; Polacco et al., 2005; Polacco et al., 2006; Sengoz et al., 2009). In Norway, only SBS rubber is used for the PMB asphalt (NVF, 2013; Rødland et al., 2022). The abrasion of the road surface and the release of road particles are estimated to be heavily impacted by the ratio of personal vehicles (PV) versus heavy vehicles (HV), as well as the use of studded winter tires (Rødland et al., 2022). The release of road abrasion particles from studded PV is estimated to be between 5 and 10 g per vehicle kilometer driven (g/vkm) for stone mastic asphalt (SMA) and between 15 and 20 g/vkm for asphalt concrete (AC) (SI-4 Table S3) (Bakløkk et al., 1997; Horvli, 1996; Snilsberg et al., 2016). The estimated road abrasion for studded HV tires is roughly 5 times the value for PV, and the abrasion from non-studded winter tires and summer tires are expected to be 40 times lower compared to studded tires (Snilsberg, 2008). The use of studded tires in Norway is extensive, covering over 80% of all vehicles in the northern part of Norway during winter. However, in the eastern part of Norway, where winters are considered milder and traffic density is high, the overall percentage in 2017 was approximately 20% (Reitan et al., 2017). The percentage of studded tires used in Oslo in 2019 was 8.6% for PV and 1.6% for HV (Rosland, 2020). Emitted TWP and PMB particles can be mixed with other road particles (such as mineral and organic matter). These are referred to as TRWP and are released to different environmental compartments through various pathways, such as road runoff, tunnel wash water, dry and wet atmospheric deposition and snow accumulation. Despite great efforts to improve analytical methods in recent years, especially developing new markers and methods for Pyrolysis GC/MS, analytical challenges still contribute to uncertainties associated with TRWP data on inventory and fluxes in the environment (Miller et al., 2021; Rauert et al., 2021; Rødland et al., 2022; Wagner et al., 2021). There are currently several different analytical methods for quantifying TWP in the environment (Chae et al., 2021; Goßmann et al., 2021; ISO, 2017a; ISO, 2017b; Klöckner et al., 2021; Parker-Jurd et al., 2021), however, only one method includes the presence of PMB-particles together with tire wear particles (Rødland et al., 2022).

The TRWP contain a large variety of chemicals, and a recent study identified 214 different organic chemicals in tires, in which 145 were classified as leachable (Müller et al., 2022). About 60% of the leachables were classified as mobile compounds, indicating a large potential for transport in the environment (Müller et al., 2022). Examples of tire-derived chemicals that have been found to be harmful to organisms are benzothiazoles, N-1,3-dimethylbutyl-N 0-phenyl-*p*-phenylenediamine (6-PPD), 1,3-diphenylguanidine (DPG) and different polycyclic aromatic hydrocarbons (PAHs) (Halsband et al., 2020; Seiwert et al., 2020; Tian et al., 2021; Unice et al., 2015).

The number of studies of road contamination in snow are limited, but those that are published have reported high levels of heavy metals, polycyclic aromatic hydrocarbons (PAHs), salt and overall particulate material in roadside snow (Moghadas et al., 2015; Vijayan et al., 2021; Viklander, 1999). Using snow as the target matrix is useful both for showing the concentration levels accumulating over a short-time window, and as a potential extreme pulsed contaminant release event when the snow melts during spring. Previous studies have looked at the number of road-related rubber particles in different snow samples and reported between 190 and 193,000 particles/L in melted snow (Bergmann et al., 2019; Vijayan et al., 2019), however no studies have so far, to the best of our knowledge, measured the mass concentrations of TRWP in snow samples. The overall objective of the present study was to provide new knowledge on the concentration levels of TRWP rubbers along roads, and the potential impact on the environment if measures are not taken. The objective was further divided into three goals. The first was to assess the concentrations of tire and PMB particles in roadside snow and compare the levels to other road-related releases such as road runoff. The second goal was to utilize the accumulation of TRWP rubber in snowbanks to explore the impact of traffic variables such as Average Annual Daily Traffic (AADT) and speed limit on the accumulation. The third goal was to assess the potential environmental impact of tire and PMB particles from roadside snow.

2. Materials and method

2.1. Sample collection and processing

Snow samples were collected from several sites in three areas around the City of Oslo, Norway, on 26th and 27th of February 2019. There are no weather stations present for all locations, so the Hovin weather station covering the city of Oslo is used for weather data (Yr, 2019). The weather in the end of January 2019 was warm, up to 14 °C on January 30th and the whole month had low precipitation (2.8 mm). In February, the weather fluctuated more, with a cold period (−9.9 °C up to 4.8 °C) with higher precipitation (6.9 mm) from 1st to 12th of February. Then there was a warmer period (−1.4 °C to 10.2 °C) from the 13th of February until the first sampling day on the 26th of February with 4.1 mm of precipitation. Three sampling sites were along a peri-urban highway (Inter-city Highway) outside of Oslo; Holstad (HO), Vinterbro (VI) and Skullerud (SK). An additional four sites were along the City Highway (Bryn (BR), Storo (ST), Ullevål stadion (US) and Lysaker (LY)), and the final four sites were in the area in the inner city (City Urban; Tøyen (TØ), Carl Berner (CB), Ila (IL) and Frogner (FR)) (Fig. 1, SI-1 Table S1). The three road types are associated with different driving styles. The Inter-city Highway has a higher speed limit (80 km/h) and is associated with long-distance driving. The City Highway has a speed limit of 70 km/h and includes areas with more braking and accelerating (crossings, roundabouts) compared to the Inter-city Highway. The urban city roads are associated with an urban driving style, including lower speed limits (40–50 km/h) and frequent braking and accelerating (traffic lights, crossings, roundabouts, zebra crossings). All sites within each area were chosen to provide a spatial dispersion. They also represent different AADT level within the area, based on data collected from (Vegkart, 2019) (SI-1 Table S1). A city-reference snow sample was taken from a lawn area in the centre of the Tøyen Botanical garden, approximately 60 m from the closest urban road and on a hilltop. The winter maintenance for all sites are similar. In general, snow from all three site groups are pushed to the sides by snow ploughs where there is available space. In the City Urban sites where space for snow storage is limited, the snow is transported to a melting facility in the harbour when needed (BYM, 2021; NPRA, 2022). Road-deicing salts (mainly NaCl and $MgCl_2$) are also used on all the roads in this study when the temperatures fluctuate around 0 °C.

The samples were collected using a metal snow corer (inner diameter 4.2 cm) in snowbanks at 0 m (0–1 m), 1 m (1–2 m) and 3 (3–4 m) m from the road. At each site, 5 cores were taken within a 1 m square at random. Some sites had low snow depth, at which 10 cores were sampled to obtain a representative snow volume. All snow cores were measured for length. The 5–10 cores from each site were then mixed together in one bag per site (ziplock, polyethylene (PE) bag). The mixed samples were stored in a freezer (−20 °C) until processing and chemical analysis. The

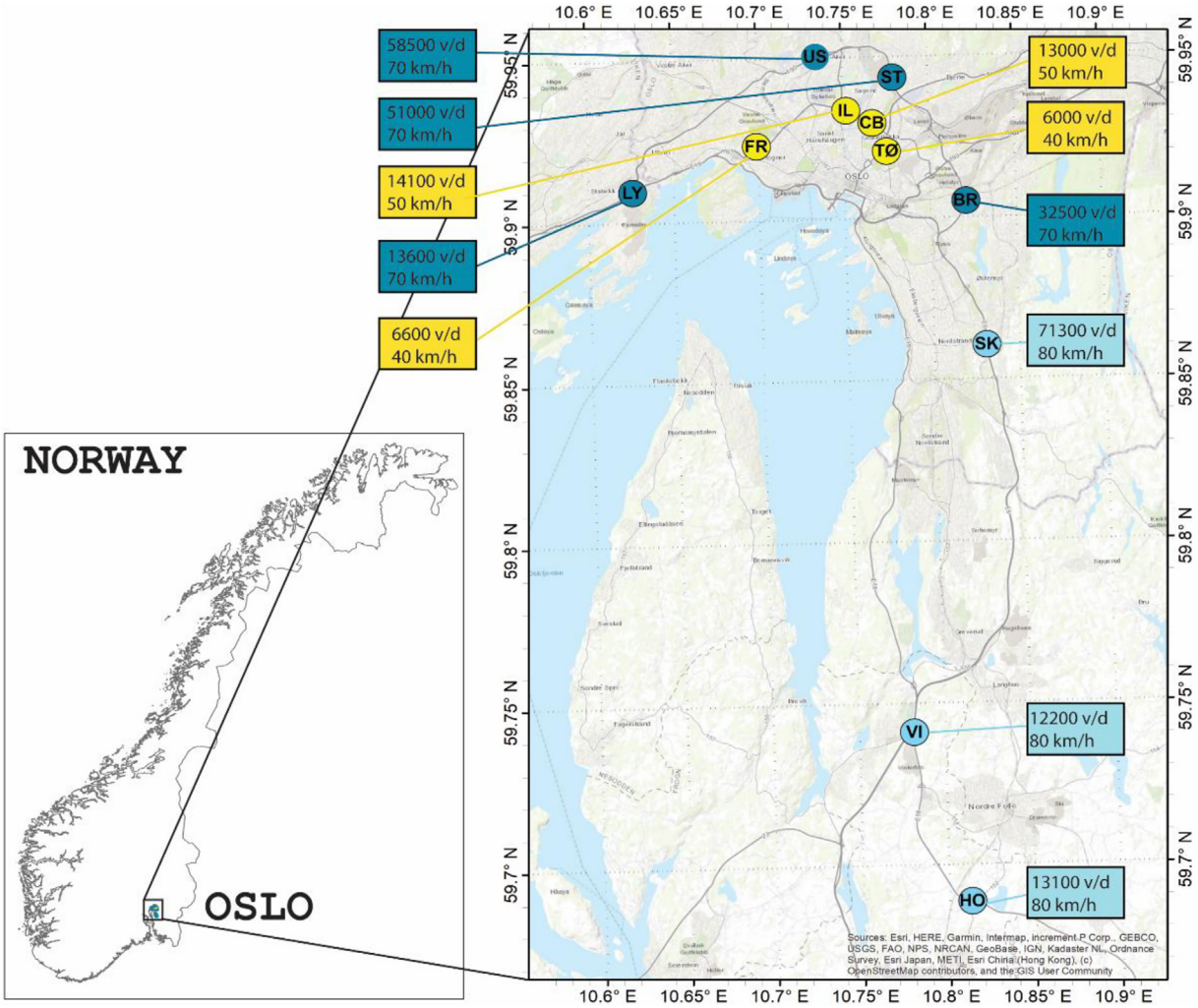

Fig. 1. Map of the sampling locations: Inter-city Highway: Holstad (HO), Vinterbro (VI), Skullerud (SK). City Highway: Bryn (BR), Storo (ST), Ullevål stadion (US), Lysaker (LY). City Urban: Frogner (FR), Ila (IL), Carl Berner (CB), Tøyen (TØ).

frozen snow samples were weighed and then melted in the zip-lock bag at room temperature. The volume of melt water was recorded and then transferred to pre-cleaned glass beakers using 1 mm sieves to remove large items. The samples were stirred by handshaking for 20 s before 16 mL sub-samples were transferred to glass jars. These jars had been pre-treated in the muffle furnace (Nabertherm, Germany) at 550 °C in order to remove any contamination from other polymer particles that could interfere with the analysis results. Contamination could include polyvinyl chloride (PVC), polyethylene terephthalate (PET), acrylonitrile-butadiene-styrene copolymer (ABS) and Polystyrene (PS) (Rødland et al., 2022). The sub-samples were frozen (−20C, 24H) and freeze dried (3–4 days, Leybold Heraeus Lyovac GT2). Dried snow material were weighed directly into the pyrolysis cups for analysis.

2.2. Pyrolysis GC–MS

Samples were analysed with a Multi-Shot Pyrolyzer (EGA/PY-3030D) equipped with an Auto-Shot Sampler (AS-1020E) (Frontier lab Ltd., Fukushima, Japan) coupled to gas chromatography mass spectrometer (GC/MS) (5977B MSD with 8860 GC, Agilent Technologies Inc., CA, USA). Samples were pyrolyzed with single-shot mode at 700 °C for 0.2 min (12 s). Injections were made using a 50:1 split and with a pyrolyzer interface temperature at 300 °C. The pyrolysis method followed Rødland et al. (2022), and uses the combined peak heights of four selected markers normalized against an internal standard (deuterated Polybutadiene, d6-PB). The selected markers consisted of *m/z* 78 Da for *benzene*, *m/z* 118 Da for *α-methylstyrene*, *m/z* 117 Da for *ethylstyrene* and *m/z* 91 Da for *butadiene trimer* (first trimer in the TIC) (SI-2 Table 2).

The calibration curve was created with three different ratios of SBR and SBS (20:80, 40:60 and 80:20). Total mass of SBR + SBS of 1 μg, 2 μg, 5 μg, 25 μg, 100 μg and 150 μg, were inserted into pyrolysis cups (n = 3) and spiked with 25 μg d6-PB as internal standard. The normalized sum peak of all marker compounds is plotted against the mass of SBR + SBS at each calibration level to form the calibration curve ($R = 0.99$, $p = 2.2 \times 10^{-16}$, Supplementary Information (SI) Fig. S1).

2.3. Meltwater concentrations vs mass load calculations

The rubber concentrations (SBR + BR + SBS) in samples were analysed as meltwater concentrations in mg/L. Tire and PMB results for acute release through meltwater were reported as mg/L concentrations. In addition to meltwater concentrations, values were converted to mass load (ML) concentrations, in mg/m^2. The ML conversion is a useful tool when comparing measured contaminant concentrations in snow from various sites, where the snow has been frozen and thawed at different times. This has been previously applied in studies of road-related contaminants in snow (Boom and Marsalek, 1988; Moghadas et al., 2015; Reinosdotter and Viklander, 2005; Viklander, 1997).

The conversion from mg/L to ML can be done using the following equation according to Moghadas et al. (2015):

$$ML = K * Cs * SWE \quad (1)$$

where *ML* is the mass load of the given pollutant per square meter of a snow deposit (mg/m^2);

K is a unit conversion coefficient (0.1 to convert from cm snow cores to m);
Cs is the pollutant concentration C in the melted snow sample s (mg/L);
SWE is the snow water equivalent of the sample (cm), calculated from the measured snow core height (cm) multiplied with the snow density (g/cm^3) for each snow sample.

A calculation example for ML is given is SI-5 and all snow core sample data can be found in SI-10.

2.4. Calculation of tire and PMB particles

The concentration of SBR + BR + SBS per cup (μg/cup) was calculated with the added weight of dried snow material (mg) to give the concentration of SBR + BR + SBS μg/mg dried snow material. The amount of dried snow from each sample was related to the volume of melted snow (mL) for each sample, and then upscaled to give the concentration of SBR + BR + SBS mg/L per sample. The concentration of SBR + BR + SBS in meltwater (mg/L) was used to calculate the concentration of tire particles and PMB particles in the sample. The calculation is described in detail in Rødland et al. (2022). This method utilizes emission factors (EFs) for tire wear (Klein et al., 2017) and for road wear (Bakløkk et al., 1997; Horvli, 1996; Snilsberg, 2008; Snilsberg et al., 2016) to find the expected ratio of tire to PMB in each sample (SI-4 Tables S3 and S4). The EFs for road wear is adjusted to include the ratio of studded tires used for both personal and heavy vehicles at each site. Then the SBR + BR + SBS values are used to calculate the concentration of tires (M_T) and PMB (M_{PMB}) separately by applying the following Eqs. (2)–(5). A calculation example can be found in the Supplementary (SI-5)

$$M_T = \frac{M_S - (M_S * R_{SBS}) * Sc}{(S_{PV} * R_{PV}) + (S_{HV} * R_{HV})} \quad (2)$$

$$M_{PMB} = \frac{(M_S * R_{SBS})}{C_{PMB}} \quad (3)$$

where

M_T is the mass of tire in a sample (mg);
M_{PMB} is the mass of PMB in a sample (μg);
M_S is the mass of SBR + BR + SBS in a sample (μg);
R_{SBS} is the estimated ratio of SBS from the total SBR + BR + SBS concentration for each location;
Sc is the conversion factor for styrene content in standards vs tires;
S_{PV} is the mass of SBR + BR in personal vehicle tires (μg/mg);
R_{PV} is the ratio of personal vehicles at the sampling location;
$S_{HV}$is the mass of SBR + BR in heavy vehicle tires (μg/mg);
R_{HV} is the ratio of heavy vehicles at the sampling location;
C_{PMB} is the conversion factor for SBS to PMB, based on the percentage SBS in PMB (0.05).

The mass of SBR + BR in personal vehicles and heavy vehicles were obtained by analysis of reference tires representing the Norwegian tire use as reported in Rødland et al. (2022). The equations were performed using the Excel Add-in package Crystal Ball, where 100,000 Monte Carlo simulations were applied. This gives the predicted statistics of the tire and PMB concentrations from each sample. For this study, the mean, median, standard deviation, minimum, maximum, and the 10th, 25th, 75th and 90th percentiles for both tire and PMB concentrations were reported. The results using meltwater were converted to ML (mg/m^2) after simulation and both results are presented to facilitate comparison with previous work.

2.5. Statistical analysis

The statistical analysis of the data was conducted in RStudio 1.3.109 (Team, 2020), R version 4.0.4 (2021-02-15), specifically using the ggplot2-package (Lai et al., 2016) (gplot2_3.3.3), the car-package (Fox and Weisberg, 2019) and the dplyr-package (Wickham et al., 2018) for creating boxplot graphs, linear regression and for performing Analysis of Variance (ANOVA). The uncertainty analysis of tire and PMB calculation was performed by using Excel Monte-Carlo Add-In Crystal Ball, as described in Section 2.2.

2.5.1. Univariate statistics

All ANOVAs were performed on log-transformed data. The assumption of normal distribution of residuals was tested using Andersen-Darling normality test. If the assumption of normality was not met, ANOVA was still applied when number of samples (n) in each group were >15. The assumption of equal variance was tested using Levene's Test of Homogeneity of Variance. Whenever this assumption was not met, Welch's one-way ANOVA was used. The statistically significant level was set to $p = 0.05$.

Linear regression was used to assess the relationship between total suspended solids (TSS) and total concentration of rubbers for both meltwater and mass loads concentrations. The residuals of the regression model were checked for normality using Andersen Darling Normality test. If assumption of normality was not met, the linearity was tested using assumption free Redundancy analysis (RDA), with rubber concentration as response variable and TSS concentration as the explanatory variable.

2.5.2. Multivariate statistics

To assess the relationship between the response variables (SBR + BR + SBS concentrations in meltwater and mass load) and explanatory variables (traffic variables, road type and distance from the road), multivariate statistical analyses were conducted by using Canoco 5.12 (Ter Braak and Smilauer, 2018). Redundancy analysis (RDA) was used to explore the observed variation in the concentration of SBR + BR + SBS using the explanatory variables. Different variants of RDA were performed. Both the meltwater (mg/L) and massload (mg/m^2) data was log-transformed by the default transformation setting in Canoco. First, a constrained RDA with all variables were performed to explore the total variation explained by all identified variables. Second, RDA with forward selection were tested, where the explanatory variables contributing the most to the variation can be selected until there are no more variation to explain. In the forward selection mode, both the simple effects (the effect of each independent variable) and the constrained effects (the effect of the variable considering the other variables) were tested. The significance level in the RDA is derived by Monte Carlo permutation tests (9999 permutations performed). For all tests, $p < 0.05$ is set as the level of significance.

3. Results

3.1. Concentrations of SBR + BR + SBS rubber in snow

The reference sample location was chosen as a site close to one of the sample locations in Urban city to be relevant and to show possible background concentration in snow further away from a road. The SBR + BR + SBS concentration found in the reference sample was 8.9 mg/L in meltwater concentration and 3.2 mg/m^2 in mass load concentration. As the location is approximately 60 m distance from the nearest busy road, it is not possible to calculate the tire and PMB contribution to the SBR + BR + SBS concentrations. The concentrations of total suspended solids (TSS) in the reference snow were 556 mg/L and 198 mg/m^2.

All samples were analysed in triplicates and the standard deviation (in percentage) ranged from 3 to 34%, with an average of 15% s.d. The concentrations varied largely between different sites, road types and distance from

the road (Fig. 2, SI-7 Table S8). The lowest concentration of SBR + BR + SBS was detected at Frogner 0 m, for both meltwater and mass load concentrations (32 ± 5.9 mg/L, 119 ± 22 mg/m^2). The highest concentration for meltwater was found at Lysaker 0 m (4438 ± 191 mg/L) and the highest for mass load at Storo 0 m (29,686 ± 2949 mg/m^2). The City Highway sites had the largest mean concentrations of SBR + BR + SBS rubber for both meltwater (1290 ± 1510 mg/L) and mass load (6224 ± 8565 mg/m2), the Inter-city Highway sites had the second-highest (284 ± 222 mg/L, 1173 ± 1112 mg/m^2) and the City Urban sites had the lowest mean concentrations (124 ± 153 mg/L, 574 ± 760 mg/m^2) (Fig. 2, SI Table S3).

The difference in concentrations between the three road types were significant (ANOVA) for both meltwater ($p < 0.0001$) and mass load ($p < 0.0001$). For the concentrations in meltwater, there was a significant difference between all pairs of road types (City Urban - City Highway $p < 0.0001$; Inter-city highway – City Highway $p < 0.0001$; Intercity Highway – City urban $p < 0.0001$). For the concentrations in mass loads, only the difference between City Urban and City Highway ($p < 0.0001$) and Inter-city Highway and City Highway ($p < 0.0001$) were significant. Within the City-Highway and the City-Urban sites, the mean concentrations both in meltwater and mass loads showed that the presence of SBR + BR + SBS rubbers are highest at 0 m distance from the road and decreases towards 3 m distance (Fig. 3, SI-7 Table S8,). However, this decreasing pattern was not found to be significant (ANOVA, $p > 0.05$) when using the distance as categorical variable. In the Inter-City samples, the sites at 1 m distance had a higher mean and median concentration of rubbers compared to the samples at 0 m and 3 m (SI Table S8, Fig. 3), and the difference between 3 m and 1 m was found to be significant for both mass load ($p = 0.0131$) and meltwater ($p < 0.0001$) concentrations. For concentrations in meltwater, also the difference between 3 m and 0 m was found to be significant ($p < 0.0001$). Overall, when combining all snow samples analysed, the difference between the samples collected at 0 m, 1 m and 3 m was statistically significant both for values in meltwater ($p = 0.00056$), and for mass load ($p = 0.00157$). However, the difference was only significant between the 3 m and 0 m (meltwater $p = 0.0012$; mass load $p = 0.0024$) and between 3 m and 1 m (meltwater $p = 0.0040$; mass load $p = 0.012$) (Fig. 2).

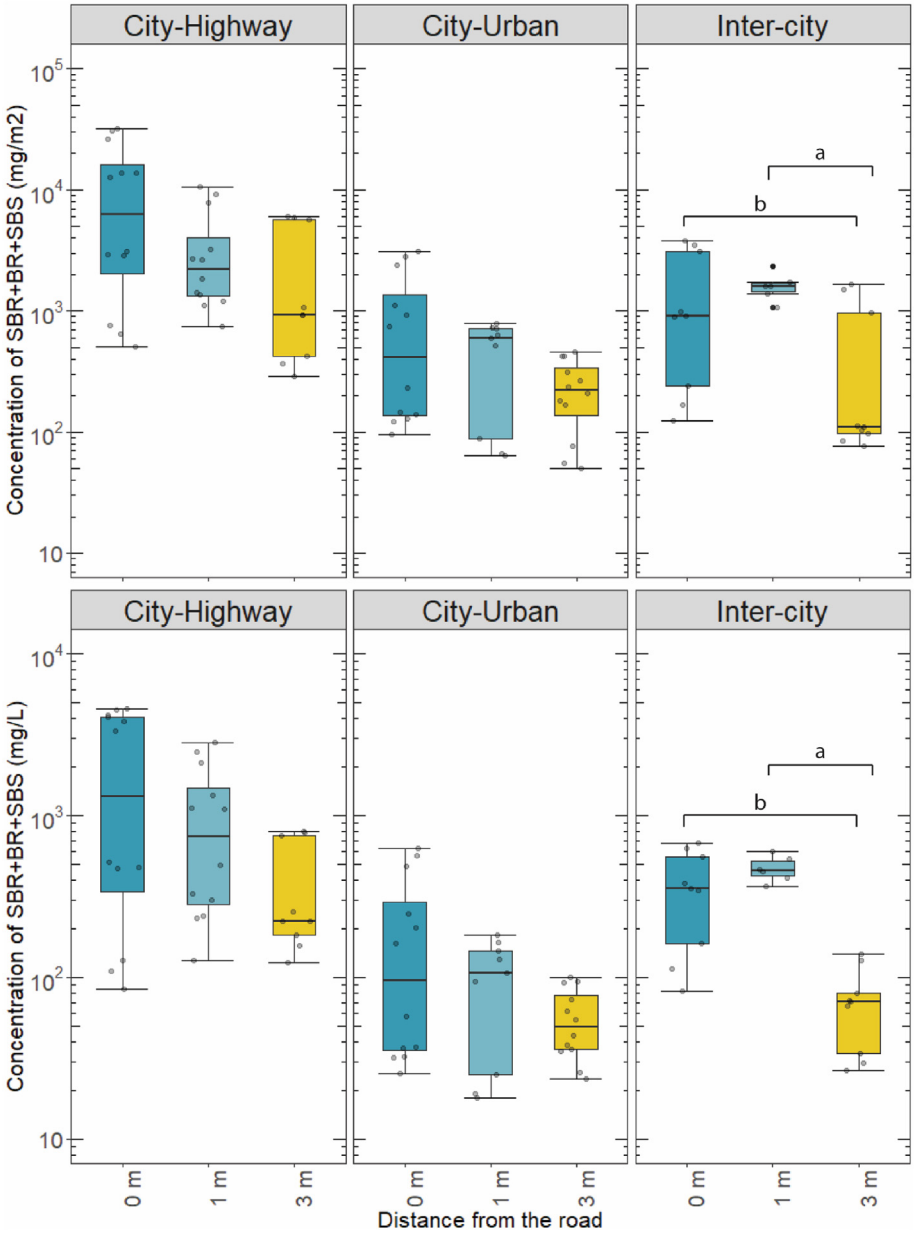

Fig. 2. Concentration of SBR + BR + SBS in meltwater (mg/L, below) and mass load (mg/m^2, above) at distances 0 m, 1 m and 3 m distance from the road The difference is significant ($p < 0.05$) between Inter City 1 m and 3 m (a) and between Inter City 0 m and 3 m (b).

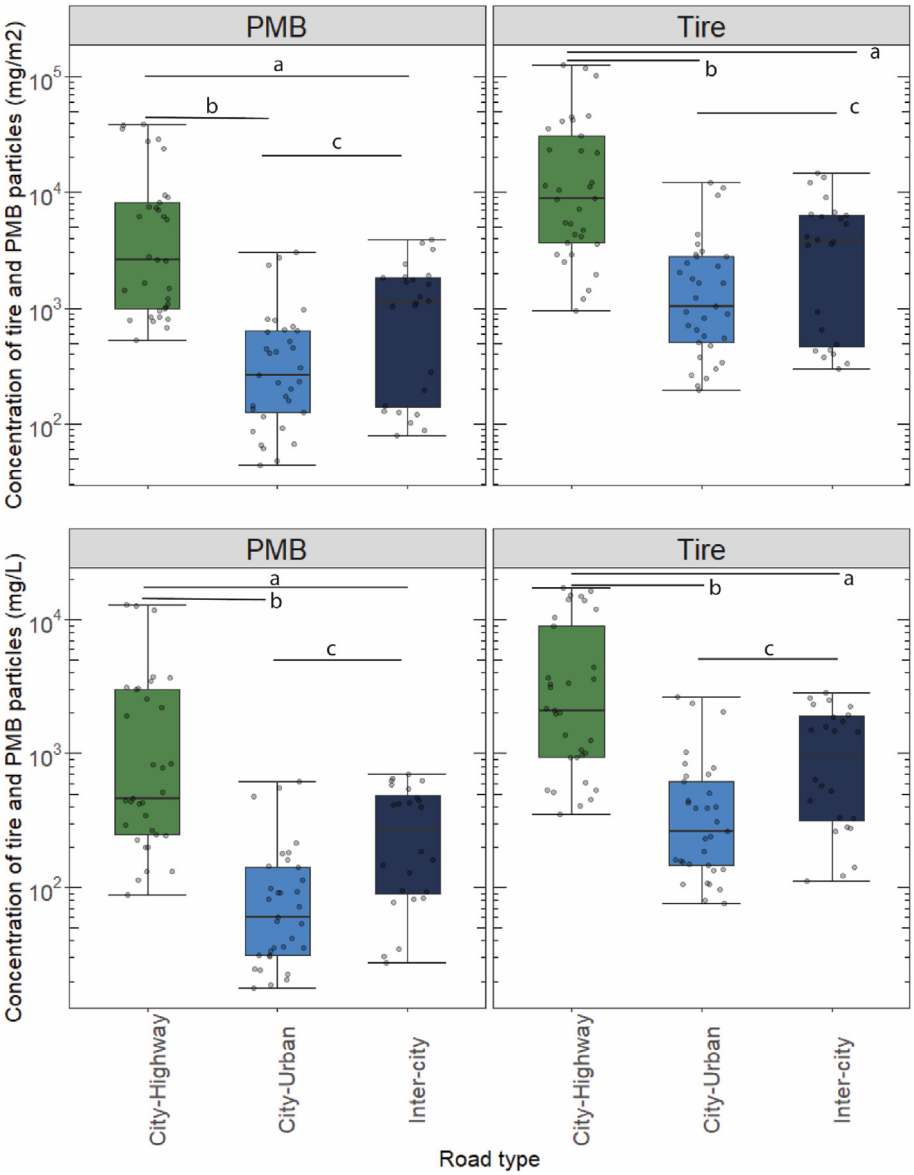

Fig. 3. Concentration of tire and PMB particles for all distances (0, 1 and 3 m) pooled, in mass load (mg/m^2, above) and meltwater (mg/L, below). The difference is significant ($p < 0.05$) between Inter-city and City highway (a), between City Highway and City Urban (b) and between City Urban and Inter City (c).

3.2. Concentrations of tire and PMB in snowbanks

The calculation from SBR + BR + SBS concentrations to tire and PMB concentration are reported in SI-6 Tables S6 and S7, and all concentration data is summarized in Table S8. The concentration of tire particles in meltwater varied from 76.0 mg/L (Tøyen 1 m) to 14,500 mg/L (Lysaker 0 m), with an average of 2090 ± 3700 mg/L for all sites (Fig. 3, Table S8). The PMB concentration ranged from 14.8 (Tøyen 1 m) to 9550 (Lysaker 0 m) mg/L, with an average concentration of 731 ± 1810 mg/L (Fig. 3, Table S8). The concentration of tire particles in mass load varied from 222 mg/m^2 (Ila 3 m) to 109,000 mg/m^2 (Storo 0 m), with an average of 10,600 ± 2200 mg/m^2. For PMB, the concentration in mass load varied from 45.0 mg/m^2 (Ila 3 m) to 28,800 mg/m^2(Lysaker 0 m), with an average of 2960 ± 6410 mg/m^2. The three road groups show a large spread in concentrations of tire and PMB and the variation between the groups are statistically significant (ANOVA: Meltwater $p < 0.0001$, massload: $p < 0.0001$). The City Highway sites show slightly higher concentrations of both tire and PMB for meltwater and for mass loads. The difference in concentration of tire and PMB between 0 m, 1 m and 3 m distance is also significant for both meltwater ($p < 0.0001$) and for mass load ($p < 0.0001$).

3.3. Total concentrations of particles

The total mass of particles (TSS) in the samples were calculated based on the total weight of particles (<1 mm) per L melted snow. The concentration of TSS varied between 1810 and 355,000 mg/L, with an average of 49,700 ± 76,800 mg/L for meltwater and 6340 to 2,810,000 mg/m^2 for mass loads, with an average of 247,000 ± 516,000 mg/m^2 (SI-7 Table S8). Comparing the levels of tire and PMB particles to the total mass of particles (W_{Tire}/W_{TSS}, W_{PMB}/W_{TSS}) showed that tire and PMB-particles only contribute 5.7% of the particles for the meltwater concentrations and 5.2% of the particles for the mass load concentrations.

A linear relationship was found between the total particle concentrations and the synthetic rubber concentrations. The model of SBR + BR + SBS (log-transformed) ~ TSS (log-transformed) in mass load concentration (Fig. 4) did not have normally distributed residuals, and an assumption free Redundancy Analysis (RDA) was performed. With TSS as the explanatory

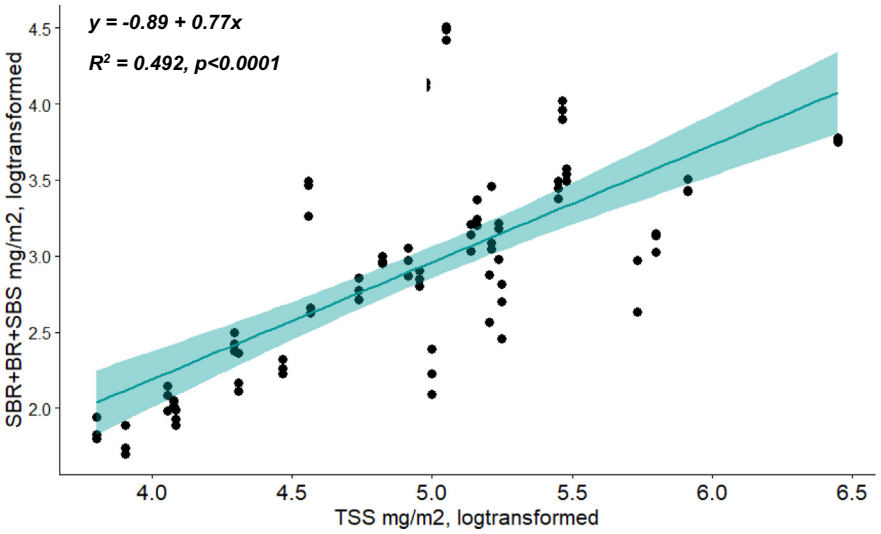

Fig. 4. Assumption-free linear regression of TSS (mg/m^2, log transformed) and rubber (SBR + BR + SBS, mg/m^2, log transformed) performed by RDA, $R^2 = 0.492$ ($p < 0.0001$) and $y = -89 + 0.77x$.

variable, 49% of the variation in SBR + BR + SBS concentration (mg/m^2) was explained ($p < 0.0001$). For concentrations in meltwater (mg/L), the residuals of the linear regression model were normally distributed; $R^2 = 0.48$ ($p < 0.0001$, SI-8 Fig. S2).

3.4. Relationship between SBR + BR + SBS, traffic features and road type

To assess any relationship between tire and PMB particles, road type and traffic features, the concentration of SBR + BR + SBS was applied instead of estimated tire and PMB particles. Tire and PMB are calculated using AADT, which consequently exclude AADT to be used as an independent explanatory variable. Thus, using the total concentration of these rubbers will allow the exploration of all traffic variables, including AADT.

The explanatory variables explored were site *Group* (Inter-city Highway, City Highway and City Urban), *AADT* (total), *PV%, HV%*, *STUD%* (studded tires %), *Speed limit*, *STOP-GO* (areas with acceleration/braking), *Distance* from the road (0, 1 and 3 m) and the interaction between *AADT x Speed limit*. Using an assumption free multiple linear regression with RDA on all explanatory variables, 69% of the variation found for concentrations in meltwater could be explained ($p = 0.0001$). For the concentrations in mass loads, 66% could be explained by the identified explanatory factors ($p = 0.0001$). (SI-9 Table S8). Exploring the variation using only the traffic variables and distance (removing road groups) lowered the percentage explained to 50% ($p = 0.0001$) for meltwater and 47% mass load concentrations ($p = 0.0001$). The Simple (i.e. the unique effect of each explanatory variable) and conditional effect (i.e. the unique effect of each explanatory variable after considering already chosen explanatory variable(s)) of each explanatory variable are presented in SI-9 Table S8. For meltwater concentrations, the statistically significant variables that explain the observed variation in SBR + BR + SBS concentrations are *Group-City-Highway* (34%), *Group-City-urban(30%)*, *Speed limit (22%)*, *distance (15%)*, *AADT*speed limit (12%)* and *AADT (12%)*. For mass loads, the variables responsible were slightly different. *Group-City-Highway* was still the main explanatory variable (34%), followed by *Group-City-Urban (23%)*, *Speed limit (16%)*, *AADT (14%)*, *AADT*Speed limit (14%)* and *Distance (13%)*. The significance of *Speed limit* was reduced for concentrations in mass loads compared to meltwater. The weather in Oslo before the snow was collected was fluctuating a lot, with both dry periods and precipitation, as well as both very cold and very warm temperatures (Yr, 2019). Even though the weather was not monitored for each exact sample location, the weather data available indicates that the sampled snow represents several melt and freeze episodes. This is also supported by the density data (SI-10), where the density varies between 31.5 and 72.2 g/m^3. In the calculation of mass load concentrations, density of the snow sample is one of the key parameters. Therefore, density is accounted for when applying the RDA analysis on mass load concentrations but not for the meltwater concentrations. The correlation (R^2 adjusted, RDA) between density and the SBR + BR + SBS concentration in meltwater was 36% and by including density as a variable together with the other identified variables mentioned above, the variation explained increased to 74% for meltwater.

4. Discussion

4.1. Meltwater concentrations vs. mass loads

Snowpacks are continuously affected by freezing and thawing processes which vary in time and space. Consequently, the occurrence of contaminants in roadside snow may also vary greatly in both time and space which makes it difficult to compare concentration data between different sites. Hence, previous studies on contaminants in roadside snow (Moghadas et al., 2015; Vijayan et al., 2021; Viklander, 1998) have presented results as concentrations in mass load to overcome this issue. Considering the environmental impact, meltwater concentrations are potentially more relevant. Most tire and PMB particles in snow will be transported to the environment in meltwater during the snowmelt periods. Meltwater concentrations are also more comparable to other studies of tire and road particles in other environmental matrices. A snowpack is by no means a homogenous sample matrix, especially when considering particle pollution. However, using sampling strategies where multiple samples are collected and combined to represent a larger area (multi-incremental sampling), as applied in the present study, could be important to obtain representative samples. Such strategies have been suggested in previous studies of snow (Vijayan et al., 2021) as well as for roadside soil sampling (Johnsen and Aaneby, 2019).

4.2. Tire and PMB particle concentrations in snow

The SBR + BR + SBS concentrations in meltwater and mass load in the reference snow sample were very low compared to the roadside snow samples, only 1.5% and 0.11% of the average concentration of SBR + BR + SBS in roadside snow. Despite the low concentrations, quantifiable concentrations of rubbers were found in the reference site at such a distance from nearby roads. The rubbers may originate from atmospheric deposition from nearby roads. In the review of TRWP release in Germany, Wagner et al. (2018) estimated that 10% of the total tire wear mass on highways are deposited from the atmosphere. It is also a possibility that some of these

rubbers originate from small maintenance vehicles used in the park, although no information on the use of these vehicles in the park could be obtained.

The ratio of tire vs PMB particle based on the calculated ratio of SBS (RSBS) varied between the sites, however, for all sites except Lysaker, the mean calculated percentage of SBS was between 3.7 and 4.7%.For Lysaker, the only site with asphalt concrete surface layer, the mean calculated SBS percentage was 10.8%. The increased road wear for sites with concrete asphalt compared to the sites with stone mastic asphalt is supported by the fact that Lysaker had the highest concentration of SBR + BR + SBS in meltwater and the second highest for mass load concentrations, although the traffic density (AADT) at Lysaker is only 13,600 v/day compared to between 14,000 and 71,000 v/day at Ila, Bryn, Storo, Ullevål stadion and Skullerud. Our data support the notion that the type of road surface does have an important impact on the release of road abrasion particles and the total release of rubber particles from roads with polymer-modified bitumen.

A limited number of relevant studies of tire wear and road wear particles in the environment, and additionally that different analytical approaches have been used, makes it difficult to have a direct comparison of our results with earlier findings. Only one study in which tire wear material concentrations were quantified in roadside snow could be identified. Using Benzothiazole (BT) as the marker, Bauman and Ismeier measured tire and PMB particle concentrations of 563 mg/L (Baumann and Ismeier, 1998). However, BTs may not be a reliable marker for tire wear studies since the concentration of different BTs varies between different tires and as they have shown the ability to transform during different environmental conditions (Asheim, 2018; Bye and Johnson, 2019; Zhang et al., 2018).

In the lack of other snow-studies, tire wear particles in road runoff are the most relevant environmental matrices to compare with. The current available data show that tire particles are found in the range of 3–180 mg/L in road run-off (Baumann and Ismeier, 1998; Kumata et al., 2000; Kumata et al., 1997; Kumata et al., 2002; Parker-Jurd et al., 2021; Reddy and Quinn, 1997; Wik and Dave, 2009). The results found for meltwater snow in the present study are significantly higher (76.0–14,500 mg/L), suggesting that snowpacks accumulate tire wear over time and potentially poses a higher acute release risk to the environment compared to road runoff. Similar trends were found for metals and PAHs in snow during melting in previous studies (Viklander, 1996).

4.3. Total particle concentrations

Total particle concentrations have been reported for a range of environmental samples, such as roadside snow (5–12,700 mg/L: Moghadas et al. (2015), Viklander (1999), tunnel wash water (8–31,000 mg/L, Meland and Rødland (2018), Hallberg et al. (2014))) and sedimentation pond effluent (<15 mg/L, Hallberg et al. (2014)). Compared to previous studies, the total particle concentrations found for the roadside snow in this study are significantly higher, from 1800 to 355,000 mg/L in meltwater and 6340 to 2,810,000 mg/m^2. The variable AADT might be a reason for the large variations found for TSS in these studies, as the AADT for snow-sites in our study are between 6000 and 71,250 vehicles (v)/day, compared to 1500–20,000 v/day in the studies of Moghadas et al. (2015). The TSS values found in tunnel wash water in Norway, where the AADT varied between 1550 and 77,000 v/day, were in fact more comparable to the values found for roadside snow in this study. The relationship found between TSS and SBR + BR + SBS concentrations suggests that there is scope to predict tire and PMB concentrations based on the measured TSS. This also suggest that historical TSS data may be used to estimate the presence of tire and PMB particles in previous studies, where the tire and PMB concentrations have not been analysed specifically.

4.4. Exploring the variations

There are several possible explanatory variables that impact the variation in rubber content in the roadside snow. Exploring all the identified traffic variables as well as deposition as a function of distance from the road, explained 50% of the variation for meltwater and 46% for mass load concentrations. However, adding the road groups as a variable increased the overall explanation to 69% for meltwater and 66% for mass loads.

Previous studies of road-related pollution have also explored the possible explanatory variables for other contaminants. Especially *AADT* has been proposed as one of the main drivers behind the variation of road-related pollutants in roadside snow (Li et al., 2014; Moghadas et al., 2015; Viklander, 1999), roadside soil (Werkenthin et al., 2014) as well as in road dust (Gunawardena et al., 2015) and in tunnel wash water (Meland and Rødland, 2018), as increased number of vehicles causes increased abrasion on tires and road surface as well as increased release of other pollutants related to vehicles. The results of our study suggest that *AADT* is less important compared to the road location (*Group*), followed by speed limit and distance from the road. The high impact found for the road group locations could also be explained by the physical difference between these roads, where both City Highway and City Urban are urban roads with numerous traffic lights, crossings, roundabouts and other obstacles. Another possible explanation for the high impact of road groups could be different snow handling and road maintenance for the different road groups. As described in Viklander (1998) the snow handling procedures can have an substantial impact on the concentrations of road-related contaminants in mass load of snow. The impact of snow handling could also have an impact on the meltwater vs. mass load concentrations, as mass loads take into consideration the density of the snowpack as a measure of melting and packing. Our study, however, indicates the opposite, as the variables explained the variation slightly better for meltwater concentrations compared to mass load concentrations. Another aspect not explored in this present study is how atmospheric deposition from nearby roads have impacted the study sites. As shown in this study, SBR + BR + SBS rubbers were detected even at 60 m distance from the nearest road, suggesting that tire and/or PMB particles can be transported for relatively long distances.

Previous studies have reported that urban driving styles with increased braking and accelerating results in higher emission factors for tire wear than highway driving (Dannis, 1974; LeMaitre et al., 1998; Luhana et al., 2004; Stalnaker et al., 1996), as summarized in Vogelsang et al. (2018). Increasing speed has been found to cause increasing abrasion in tires in previous studies (Li et al., 2011) as higher speed will cause higher friction between tire and the road surface as well as generate higher temperatures in the tire tread, adding to the abrasion of the tires. As for tires, driving speed has also been found to impact the road pavement abrasion, as demonstrated for increasing PM_{10} concentrations with increasing driving speed from driving simulators (Gustafsson et al., 2009). Higher speed may also cause smaller tire and road particles to be deposited further away from the roadsides due to suspension and splash and spray as well as being caused by increased speed (Gustafsson et al., 2009). This could explain why the mean and median concentrations of SBR + BR + SBS in both meltwater and mass loads are higher at 1 m distance compared to 0 m distance in the Inter-City Highway sites, where also the highest speed limits are found. It should, however, be underlined that the speed limit used for the analysis in this study is the official speed limit on each site and not the actual average driving speed of the vehicles, as this information was not available. In future studies, atmospheric deposition, monitoring of real-time driving speed and driving behavior, the use of different driving lanes for specific vehicle types and impact of different snow handling procedures should be investigated further.

4.5. Environmental impact

The high concentrations of total particles, tire and PMB-particles present in roadside snow suggests that melting snow could pose a potential threat to the environment. The meltwater from large accumulations of snow should be considered for treatment before release into the aquatic environment. There are currently no limits set for the release of tire and PMB-particles from roads, or for other road-related matrices such as road and

tunnel runoff. The current legislation in Norway demands that a permit for release is applied for and granted if there is a release of pollutants that can have an environmental impact (Pollution Control Act, 1987), such as for the release of tunnel wash water. For TSS, there is limit value set for release of TSS in general, however, in most permits given to road runoff release in Norway, the limit for TSS to freshwater recipients is 100 mg/L and to marine recipients 400 mg/L (Rødland and Helgadottir, 2018). Thus, the TSS values in the range of 1800–355,000 mg/L observed for snowpacks in the present work are far exceeding these limit values set for road and tunnel runoff. The specific impact on the environment from TRWP is still being studied. The current published research on tire toxicity are performed on different types of artificially generated tire particles, tire leachates and/or environmental samples such as runoff sediments, which makes comparisons difficult (Baensch-Baltruschat et al., 2020; Rødland, 2019). The recently discovered acute toxicity of 6-PPD-quinone, a transformation product coming from the tire antiozonant chemical 6-PPD, on both juvenile and adult coho salmon (*Oncorhynchus kisutch*) (McIntyre et al., 2021; Tian et al., 2021) has gained significant attention by both the research community and regulators. Acute toxicity was found for adult coho salmon exposed to tire leachates of 320 mg/L tire particles (1.3–1.8 μg/L 6PPD-quinone). However, the observed acute toxicity found for coho salmon might be species-specific, as recent studies have found no acute toxicity for other organisms or even other salmon species tested (chum salmon, *Oncorhynchus keta*) (Hiki et al., 2021; McIntyre et al., 2021). As the concentrations of tire particles found for roadside snow (76–19,000 mg/L) is within the range where toxic effects on some organisms have been confirmed by previous studies (Gualtieri et al., 2005; Khan et al., 2019; McIntyre et al., 2021; Panko et al., 2013; Tian et al., 2021), it is possible that toxic effects could be observed if meltwater is released directly into a recipient, and especially smaller recipients with lower dilution capacity. Müller et al. (2022) showed that less than 20% of the organic chemical in tires had leached completely by 28 days in water, suggesting that tire particles left in an aquatic environment will continue to leach out chemicals over time unless they are removed. However, as the current knowledge on tire toxicity lacks standardization and there are no published studies on the toxicity of PMB-particles, more research is needed to address the possible environmental impact of TRWP into the environment.

5. Conclusions

The present study is the first study on mass concentrations of both tire and PMB particles in roadside snow. The study also contributes to the understanding of which traffic-related processes can be attributed to the production of these particles alongside different roads. The results show that the concentrations of tire particles along roads vary widely at over three orders of magnitude in meltwater (76–14,500 mg/L). The results also show that the total concentration of particles (1800–355,000 mg/L) in roadside snow far exceeds the limits for TSS set for other types of road-related runoff such as tunnel wash water. This shows that roadside snow in peri-urban and urban environments are highly polluted and thus, should be treated before meltwater runoff is released into the environment. The large concentration differences relate to the difference in road surface and traffic variables identified for the different sites. The high concentration of TSS and rubbers at Lysaker is in agreement with previous estimates that concrete asphalt surface layers contribute three times more road abrasion compared those with stone mastic asphalt. The main traffic variables driving the variation was found to be speed limit and AADT, where speed limit was found to be the most important variable explaining the variation in both meltwater and mass load concentrations. This is contradictory to previous road runoff studies, where AADT has been reported as the main explanatory variable. Increased speed causes higher friction between tire and the road surface, which generates higher temperatures in the tire tread and consequently increased tire abrasion. For the PMB, the road surface abrasion also increases with driving speed due to the increased friction with tires. Higher speed is also related to increased suspension and splash and spray-effect, which can cause smaller tire and PMB particles to be deposited further away from the roadsides. Increased AADT generates more abrasion on the road surface due to the increased number of vehicles, as well as the release of tire abrasion particles due to increased number of tires passing the area. Part of the variation was also found to be related to the road type of each site, e.g. urban city road (municipality road), peri-urban highway road (state road) and inter-city highway road (state road). Within each road type, the sites include variable AADT, speed limit and areas of braking and acceleration. There is, however, also a variation explained by the road groups that could not be assigned to any known traffic variable. This suggests that there may be other variables that impact the production of tire and PMB particles that are yet to be explored, such as the importance of different snow handling procedures and winter maintenance. This should be prioritized for future research.

CRediT authorship contribution statement

Elisabeth Rødland: Conceptualization, Methodology, Investigation, Formal analysis, Writing – Original Draft, Writing - Reviewing and Editing, Visualization. **Ole Christian Lind:** Conceptualization, Writing - Reviewing and Editing. **Malcolm J. Reid:** Conceptualization, Writing - Reviewing and Editing. **Lene S. Heier:** Conceptualization, Writing - Reviewing and Editing. **Elvis D. Okoffo:** Investigation, Writing - Reviewing and Editing. **Cassandra Rauert:** Investigation, Writing - Reviewing and Editing. **Kevin V. Thomas:** Writing - Reviewing and Editing. **Sondre Meland:** Conceptualization, Writing - Reviewing and Editing, Formal analysis, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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