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Characterization of tire and road wear microplastic particle contamination in a road tunnel: From surface to release

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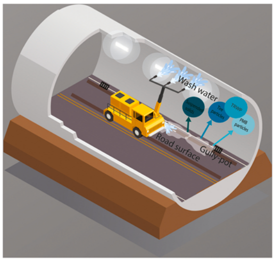
HIGHLIGHTS

- A new method for calculating and predicting mineral content of TRWP were proposed.
- Large variations of TRWP through the tunnel and across the driving lane.
- Concentrations of TRWP in gully-pots comparable to sedimentation basins.
- Sedimentation treatment of tunnel wash water only retained 63% of TRWP.
- Uncertainties related to calculation of tire and road wear particles were assessed.

GRAPHICAL ABSTRACT



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ABSTRACT

Road pollution is one of the major sources of microplastic particles to the environment. The distribution of tire, polymer-modified bitumen (PMB) and tire and road wear particles (TRWP) in different tunnel compartments were explored: road surface, gully-pots and tunnel wash water. A new method for calculating TRWP using Monte Carlo simulation is presented. The highest concentrations on the surface were in the side bank (tire:13.4 ± 5.67; PMB:9.39 ± 3.96; TRWP:22.9 ± 8.19 mg/m^2), comparable to previous studies, and at the tunnel outlet (tire:7.72 ± 11.2; PMB:5.40 ± 7.84; TRWP:11.2 ± 16.2 mg/m^2). The concentrations in gully-pots were highest at the inlet (tire:24.7 ± 26.9; PMB:17.3 ± 48.8; TRWP:35.8 ± 38.9 mg/g) and comparable to values previously reported for sedimentation basins. Untreated wash water was comparable to road runoff (tire:38.3 ± 10.5; PMB:26.8 ± 7.33; TRWP:55.3 ± 15.2 mg/L). Sedimentation treatment retained 63% of tire and road wear particles, indicating a need to increase the removal efficiency to prevent these from entering the environment. A strong linear relationship (R2-adj=0.88, $p < 0.0001$) between total suspended solids (TSS) and tire and road wear rubber was established, suggesting a potential for using TSS as a proxy for estimating rubber loads for monitoring purposes. Future research should focus on a common approach to analysis and calculation of tire, PMB and TRWP and address the uncertainties related to these calculations.

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1. Introduction

Road tunnels are considered pollution "hot spots", accumulating pollutants from both vehicles and the road surface over time. Several studies have therefore characterized and assessed the levels of traffic-associated pollutants in tunnel wash water, with examples of pollutants such as zinc (Zn), lead (Pb), copper (Cu), polycyclic aromatic hydrocarbons (PAH), and abrasion particles from brakes, tires, and the road surface (Allan et al., 2016; Hallberg et al., 2014; Meland et al., 2010a). Recently, attention has been given to microplastic particles associated with roads and traffic, as tire wear particles and road wear particles contain synthetic rubbers, and contribute a substantial amount of rubbers to the overall microplastic particle release into the environment (Boucher et al., 2020; Knight et al., 2020; Sundt et al., 2021). Previous studies have defined particles released from tire wear and subsequently mixed with road wear mineral particles as the hetero-aggregated tire and road wear particle (TRWP) (Kreider et al., 2010). These are estimated to contain 50% tire tread and 50% road wear (Kreider et al., 2010), in which the rubber concentration (SBR+BR) in the tire is estimated at 50% (Unice et al., 2012, 2013). However, the assumption of road wear content in TRWP is based on a small number of studies and the use of a fixed percentage estimation of 50% road wear in TRWP has been questioned by a recent study (Klöckner et al., 2021). Also, the assumption that all tires contain 50% synthetic rubber have recently been discussed, as new research show a large variation in Styrene Butadiene rubber (SBR) and Butadiene rubber (BR) between different tires (Rauert et al., 2021; Rødland et al., 2022b). It has also been reported that polymer-modified bitumen (PMB) typically added to the road asphalt where traffic density is high, also contain a synthetic rubber similar to the rubber used in tires (Rødland et al., 2022b). However, PMB concentrations have so far only been reported for road-side snow (Rødland et al., 2022a).

The present study aimed to provide a characterization of total suspended solids (TSS) and road-associated microplastic particles, including tire particles, PMB particles and TRWP, through a road tunnel system, from the road surface of various parts of the tunnel to the release of tunnel wash water, with an assessment of levels retained in gully-pots and sedimentation treatment. Previous studies of tunnels have reported that most of the road dust accumulates in the side bank area close to the tunnel walls and between wheel tracks and low particle concentrations are found in the wheel tracks, as well as reporting higher concentrations in the tunnel inlets compared to the outlets (NPRA (2017, 2021b)). Most tunnels have drainage systems that convey the tunnel wash water out of the tunnel. Gully-pots are an important part of the drainage system and are used to trap sediment, debris, and larger particles to avoid clogging of the pipes. Previous literature has however suggested that these gully-pots have a limited effect in removing TRWP from tunnel wash water due to the density of TRWP and the design of most gully-pots (Andersson et al., 2018; Vogelsang et al., 2018). In some tunnels, treatment facilities have been built, to remove pollutants before the water is released into the environment. The correlation between pollutants and TSS makes sedimentation an efficient treatment of tunnel wash water (Allan et al., 2016; Hallberg et al., 2014; Paruch and Roseth, 2008; Roseth and Amundsen, 2003; Roseth and Meland, 2006). Based on these previous studies, it has been assumed that sedimentation treatment potentially also retains a substantial portion of tire and road wear particles. The most common treatment methods for tunnel wash water are sedimentation ponds and basins (Meland et al., 2010a), as 40–90% of pollutants are bound to particles (Meland et al., 2010b; Roseth and Amundsen, 2003). TSS removal of $>$ 80% is demanded of a sedimentation treatment built for road and tunnel runoff in Norway (NPRA, 2021a), which has been confirmed possible with laboratory tests (TSS removal of 74 – 87%); (Garshol et al., 2015; Nyström et al., 2019)).

There is a need to generate concentration data of tire particles, PMB particles and TRWP for road runoff and tunnel wash water, especially for untreated tunnel wash water that is released directly into the environment. Currently there are no published studies on the tire, PMB and TRWP mass concentrations in tunnel wash water. One recent study, using Zn as a marker for tire wear, reports mass concentrations of TRWP between 110 and 120 mg/g (dry weight; dw) in tunnel road dust (Klöckner et al., 2021). This is approximately ten times higher mass concentration of TRWP than previously reported for road dust outside of tunnels using Zn (Klöckner et al., 2020; 76.7–9.4 mg/g). This suggests that TRWP do accumulate in tunnels and that tunnel wash water potentially contains high mass concentrations of TRWP compared to the levels currently reported for road runoff (3 – 180 mg/L) (Baumann and Ismeier, 1998; Kumata et al., 2000, 1997, 2002; Parker-Jurd et al., 2021; Reddy and Quinn, 1997; Wik and Dave, 2009).

The main hypotheses of this study were:

I. Concentrations of tire and road wear particles on the road surface accumulates in the bank area close to the tunnel walls and in the outlet of the tunnel
II. Tire and road wear particles are not retained in gully-pots in tunnels
III. Untreated tunnel wash water contains higher concentrations of tire and road wear particles compared to road runoff
IV. Treatment of tunnel wash water by sedimentation is efficient in removal of tire and road wear particles ($<$80%)

2. Experimental

2.1. Sample collection and preparation

All samples were collected in the Smestad tunnel (westbound tube), which is 495 m long and consists of two tubes with two driving lanes in each direction (Oslo, Norway, 22 000 vehicles per day per tube (Annual Average Daily Traffic, AADT), 70 km/h speed limit, 59°56'10.4"N 10°40'47.7"E). Three different types of samples were collected: road surface particles suspended in water, gully-pot sediment, and tunnel wash water (Fig. 1).

2.1.1. Sampling from the road surface

The road surface samples were collected before a tunnel wash on November 6th, 2019. The road surface was collected with a Wet Dust Sampler (WDS II) (Gustafsson et al., 2019; Lundberg et al., 2019). Sampling was focused on the right driving lane as it is used for normal traffic, while the left lane is for passing traffic. In the inlet and outlet of the tunnel (100 m in), samples were collected in the right lane at the roadside bank (B), in the right wheel track (IW) and between wheel tracks (BW) (Fig. 2). In the middle of the tunnel, samples were taken across the right and left lane, from B, right IW, BW, left IW, middle between the lanes (M), right IW, BW, left IW and B (Fig. 2). The WDS II collects particles ($<$5 mm) in a small area (0.0028 m^2) of the surface by applying 330 mL high pressurized water in one "shot". Each area was collected by three "shots" and the sample from two areas along the road surface were pooled together (a+b, c+d). All WDS samples were collected in 2 L plastic bottles (HDPE plastic bottles, VWR Avantor) and stored at room temperature until analysis.

2.1.2. Sampling from gully-pots

Sediments from gully-pots were sampled at 100 m (GP-1), 250 m (GP-2) and 400 m (GP-3) from the tunnel entrance before the tunnel wash on the November 6th, 2019. The sediment was sampled using a small van Veen grab sampler. The gully-pots in this tunnel had not been emptied since February 2019. Multiple grab samples were collected in pre-cleaned (rinsed with RO-water) aluminium foil trays. Triplicate samples from each gully pots were pooled into glass jars (pre-treated in muffle furnace at 480̊C, Nabertherm, Germany).

2.1.3. Sampling of tunnel wash water

Untreated tunnel wash water was collected in a pre-basin in the

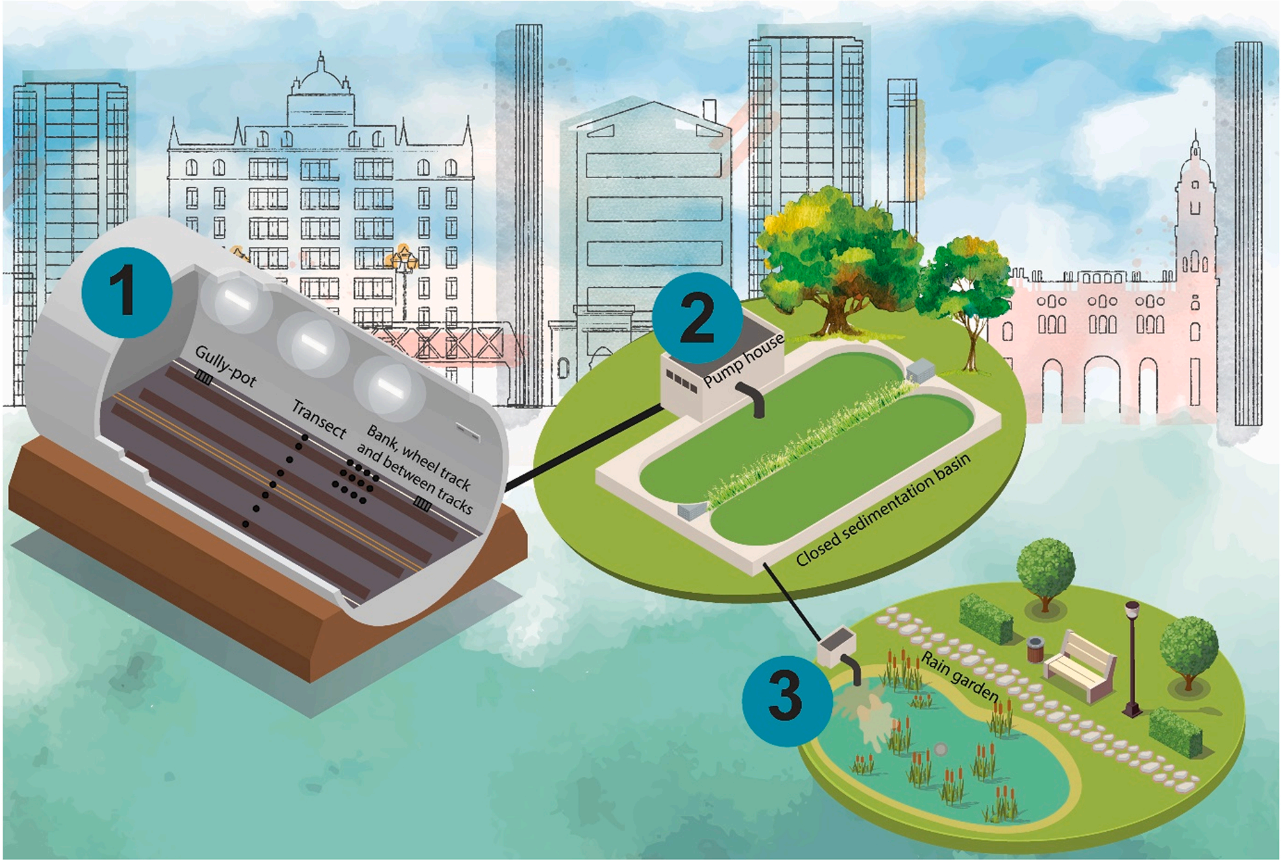

Fig. 1. Conceptual drawing of sampling locations: 1) Inside the Smestad tunnel (Road surface: transect of driving lane, road-side bank, in wheel tracks and between wheel tracks, and gully-pots), 2) the pump house (untreated tunnel wash water) and 3) outlet to the raingarden (treated tunnel wash water). Treatment was performed in the closed sedimentation basin before release to the rain garden.

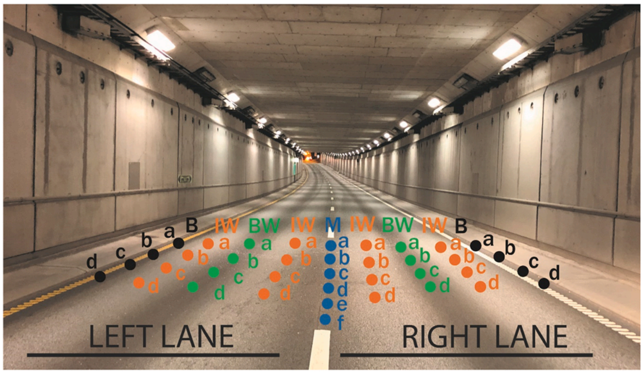

Fig. 2. Illustration of sampling with the Wet Dust Samples (WDS II) in Smestad tunnel. The circles indicate the "shot" where pressurized water has been applied to collect particles from the road surface. At the inlet, outlet and middle of the tunnel, samples are collected in the right lane in the bank (B), between wheel tracks (BW) and in right wheel tracks (IW). In the middle of the tunnel, samples were collected in B, BW, left and right IW for both left and right lane and in the middle between the two lanes (M). Photo: Kjersti W. Kronvall, NPRA.

pump house during the tunnel wash on April 21st, 2020, using a small drain pump (Brand Biltema) submerged in the water column. Samples were collected every third minute from the start to the end of the washing event, 14 samples in total. From the pump house, the tunnel wash water was pumped into sedimentation basins, one for each tunnel tube, where the water is let to settle for 21 days. After the 21 day sedimentation period, the water is released back into the pre-basin and then pumped into a rain garden. As the water is released into the rain garden, water samples were collected in a time series of 5x5min, 6x10mins and 3x15mins (May 12th, 2020). All wash water samples were collected in 1 L plastic bottles (high density polyethylene (HDPE) plastic bottles, VWR Avantor) and stored cool (4 °C) until analysis.

2.2. *Sample treatment*

WDS and tunnel wash water samples were shaken to ensure representative subsampling. For samples with high particle content (by visual inspection), 30 mL water was first transferred to 50 mL Falcon tubes and centrifuged (Thermo Scientific Multifuge 3 S/S-R Heraeus, 3000 rpm/min), in order to make the filtration step more efficient. Separation of the particles from the water by centrifugation will help to get a larger column of water through the filters, before adding the particle fraction. The supernatant was filtered (13 mm glass fibre filter, GF/A, Whatman, pore size 1.6 µm pre-treated in muffle furnace at 480 °C) using glass filtration equipment under vacuum. The particle fall-out as resuspended with a small volume (2 mL) of filtered RO-water and filtered onto the same filter and dried. Filters were weighed before and after filtration to obtain the mass of total suspended solids (TSS, >1.6 µm) in mg/L filtered water. The TSS measurements for tunnel wash water were performed on 1 L replicate samples using 47 mm RTU filters (1.5 µm) (analysed by Eurofins Norway). The whole 13 mm filter was folded up and put directly into pyrolysis cups before analysis.

For size distribution in tunnel wash water, the distribution (0.4–2000 µm) was measured by laser diffraction (Beckman Coulter LS 13 320; Pye & Blott, 2004). Samples were prepared by mixing the samples (250 mL) with a dispersant (~15% 0.05 M tetrasodium pyrophosphate) and ultrasonicating for 5 min. All samples were analysed for

60 s four times and reported as an average of the four. The obscuration limit was set between 8% and 12%. Fraunhofer's optical model was applied for the analysis (refraction index 1.333 and absorbation index 0.1). The size distribution was calculated on a volume percentage, and classification was based on a previous size distribution of TRWP (Kreider et al., 2010).

For the sediment samples from the gully-pots, the glass jars were frozen (−20 C, >24 H) and freeze dried (3–4 days, Leybold Heraeus Lyovac GT2). Next, the sediment samples were dry sieved (1 mm sieve, VWR) and weighed directly into pyrolysis cups (3.4–13.3 mg/cup) before analysis.

2.3. 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), following the method of Rødland et al. (2022b). Samples were pyrolyzed in 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 selected markers for Styrene Butadiene rubber (SBR), Butadiene Rubber (BR) and Styrene Butadiene Styrene (SBS) 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, and the method uses the combined peak heights of the four markers normalized against an internal standard (deuterated Polybutadiene, d6- PB). To demonstrate the presence of the four markers, the total ion chromatograms (TIC, pyrogram) of one tunnel wash water sample (TW-1–1) and one 30 µg SBR (quality control) sample are presented in the SI (SI-6). The calibration curve was created with three different ratios of SBR and SBS (20:80, 40:60 and 80:20). A total mass of SBR + SBS of 1 µg, 5 µg, 25 µg, 50 µg and 100 µg was inserted into pyrolysis cups (n = 3 for each ratio of SBR:SBS) 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) (Figure SI-1). The signal to noise ratio (S/N) is determined by the Agilent Masshunter software for each of the selected markers. The limit of detection (LOD) is calculated as 3 x S/N and the limit of quantification (LOQ) is calculate as 10 x S/N. The lowest limit for any marker compound (if they are different) will determine the LOD and LOQ for the analysis.

2.4. Concentration calculations

2.4.1. Tire and PMB concentrations

Tire and PMB concentrations are calculated based on the SBR+BR+SBS concentrations following the method described in detail in Rødland et al. (2022a). A detailed calculation example is given in SI-4 and SI-5. The value for SBR+BR+SBS for each sample is reported in the Supplementary for each tunnel compartment (SI-2 and SI-3).

2.4.2. TRWP calculations

TRWP are defined as the hetero-aggregates of tire and road wear particles, where the tire tread is mixed with mineral particles from the road surface when abraded. According to previous morphology studies, the mineral encrustment of tire particles collected from road surfaces ranges from 6% to 53% (Kreider et al., 2010; Sommer et al., 2018). The encrustment level was found to be highest where the speed limit is lower and with higher frequency of "stop and go" driving, as more road wear particles are left on the road surface and available for mixing with the tire wear particles (Sommer et al., 2018). Based on density analysis, a recent study of tunnel road dust reported a 25% mineral content of TRWP (Klöckner et al., 2021). To calculate the TRWP concentrations, a Monte Carlo simulation (Crystal Ball) was performed with the predicted mean concentration of tire particles and the expected level of encrustment based on previous studies. A triangular distribution was chosen to incorporate the minimum (6%), mean (30%) and maximum (53%) encrustment levels.

$$M_{TRWP} = \frac{M_T}{1 - R_{ENCR}} * 1$$

where.

M_{TRWP} is the mass of tire particles with road wear encrustment in a sample (mg);.

M_T is the mass of tire in a sample (mg);.

R_{ENCR} is the ratio of encrustment covering the tire particle.

2.5. Statistical analysis

The tire and PMB concentrations were calculated and predicted by Monte Carlo Simulation (Crystal Ball Add-In, Microsoft Excel), as described in Rødland et al. (2022b). Normal distribution was applied for both datasets of personal vehicle (PV) and heavy vehicle (HV) tires and triangular distribution for the SBS dataset. The TRWP concentration was predicted by Monte Carlo Simulation with a triangular distribution for the mineral content data for TRWP. For all three models, 100,000 simulations were applied, and the prediction statistics obtained for tire, PMB and TRWP were mean, median, standard deviation, minimum, maximum, and the 10th, 25th, 75th and 90th percentiles for each sample.

The statistical analysis of the data was conducted in RStudio 1.3.109 (Team, 2020), R version 4.0.4 (2021–02–15), 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).

All ANOVAs for WDS were performed on log-transformed data and all ANOVA for GP and TWW were performed on original data. The assumption of normal distribution of residuals was tested using an Andersen-Darling normality test. 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 TSS and total concentration of SBR+BR+SBS in tunnel wash water. The residuals of the regression model were checked for normality using an Andersen Darling Normality test.

The variation in size distribution of tunnel wash water was tested using Aitchison-weighted-logratio-PCA/Aitchison-weighted-logratio-RDA for compositional data (Canoco 5.12,Braak and Šmilauer, 2018).

3. Results

3.1. Limit of detection and limit of quantification

Different types of blank samples were analysed. For the sampling with WDS, 6 field blank samples and 3 lab blank samples were analysed. No SBR+BR+SBS were detected in these. For the tunnel wash water, 6 lab blanks were analysed. No SBR+BR+SBS were detected in these. During the pyrolysis runs, three blanks were analysed per 48 samples (autosampler), 12 blanks in total. These were blanks run without pyrolysis cups, to evaluate carry-over between samples. No SBR+BR+SBS were detected in these. Blank samples of the solvent used for the calibration samples and internal standard (Chloroform) were also analysed (n = 2). No SBR+BR+SBS were detected in these. The limit of detection (LOD, 3xS/N) for the four pyrolysis markers was < 1 µg of SBR+SBS. The limit of quantification (LOQ, 10xS/N) was < 1 µg. (Table 1).

Table 1
Summary of the Limit of detection (LOD) and the Limit of quantification (LOQ) based on the average signal to noise (S/N) of 1 µg of Styrene Butadiene rubber (SBR) and Styrene Butadiene Styrene (SBS) analysed in ratios of 20:80, 60:40 and 80:20.

1 µg SBR+SBS	Average S/N	LOD (3 x S/N)	LOQ (10 x S/N)	Concentration of SBR+SBS
m/z 78	58.5	175.4	584.8	< 1
m/z 117	7.3	21.9	73.1	< 1
m/z 118	0.3	0.9	2.9	< 1
m/z 91	0.5	1.6	5.4	< 1

3.2. Road surface

The total concentration of particles (TSS) collected per square meter road surface (m^2) varied greatly between the sample locations within each area (bank, in wheel track and between wheel tracks) and between inlet, middle and outlet of the tunnel (Fig. 3, SI Table SI-10). The average TSS concentration across all locations was 47.8 g/m^2, with a large standard deviation of 56.9 g/m^2 ($n = 27$). Comparing the inlet, mid area and outlet of the tunnel, the highest concentrations of TSS were found in the inlet (103 ± 74.7 g/m^2), the second highest in the mid area (34.0 ± 45.1 g/m^2 and the lowest in outlet (23.9 ± 10.9 g/m^2). The concentration of tire particles, PMB and TRWP were highest in the bank area of the outlet and lowest in the wheel track of the middle area right lane (Fig. 3, Table SI-10, Table SI-11). Tire particles were reported in the range of 25.3–4820 mg/m^2 (893 ± 1210 mg/m^2), the PMB in the range of 20.2–3840 mg/m^2 (712 ± 960 mg/m^2) and the TRWP in the range of 36.6–6970 mg/m^2 (1290 ± 1740 mg/m^2). The difference between inlet, middle and outlet, as well as between the right and left lane in the middle, was not statistically significant (ANOVA, $p > 0.05$). The difference between the sampling locations (B, IW, BW, M) was statistically significant (ANOVA, $p < 0.0001$). The percentage of tire, PMB and TRWP were highest in the outlet (tire: 6.4%, PMB: 5.1%, TRWP: 9.2%), compared to the middle (tire: 2.1%, PMB: 1.7%, TRWP: 3.1%) and the inlet (tire: 0.94%, PMB: 0.75%, TRWP: 1.4%). The relative standard deviation of the predicted mean tire concentrations using Monte Carlo simulation was 9.4% (Table SI-1). The relative standard deviation of the predicted mean PMB concentration was 11% (Table SI-2). Overall, the relative standard deviation of the predicted mean values of TRWP using Monte Carlo simulation was 14.2% across all samples (Table SI-3). For comparison with previous literature on tunnel and road dust, the concentration of TRWP on the road surface is also reported in mg/g, where the concentrations ranged between 0.835 and 373 mg/g (57.2 ± 99.1 mg/g).

3.3. Gully-pots

The tire and PMB concentrations found at the inlet (GP-1: tire: 53.1 ± 1.33 mg/g; PMB: 42.3 ± 1.06 mg/g) were an order of magnitude higher compared to the middle of the tunnel (GP-2: tire: 4.75 ± 1.53; PMB: 3.78 ± 1.22 mg/g) and the outlet (GP-3: tire: 7.32 ± 2.86; PMB: 5.83 ± 2.28 mg/g) (Fig. 4, SI Table SI-12). The difference between the three gully-pots was statistically significant (ANOVA, $p < 0.0001$; Tukey post hoc, $p < 0.0001$). By visual inspection, the sediments at the middle and outlet of the tunnel were significantly drier compared to the inlet sediment. The predicted standard deviation of the tire and PMB concentrations in the gully-pot samples was 9.4% and 11%, respectively (Table SI-4 and SI-5). The results for TRWP in the gully pots varied between 4.37 and 78.4 mg/g (31.4 ± 34.2 mg/g), with the highest concentrations found for the inlet. The predicted % standard deviation of the TRWP values were 14.1% (Table SI-6).

3.4. Tunnel wash water

The average total suspended solids (TSS) concentration for tunnel wash water before treatment ranged from 930 to 3500 mg/L (1620 ± 930 mg/L; Fig. 5, Table SI-13, Table SI-14). The predicted concentration of tire particles before treatment ranged between 14.5 and 47.8 mg/L (33.6 ± 9.20) and had standard deviation in the Monte Carlo

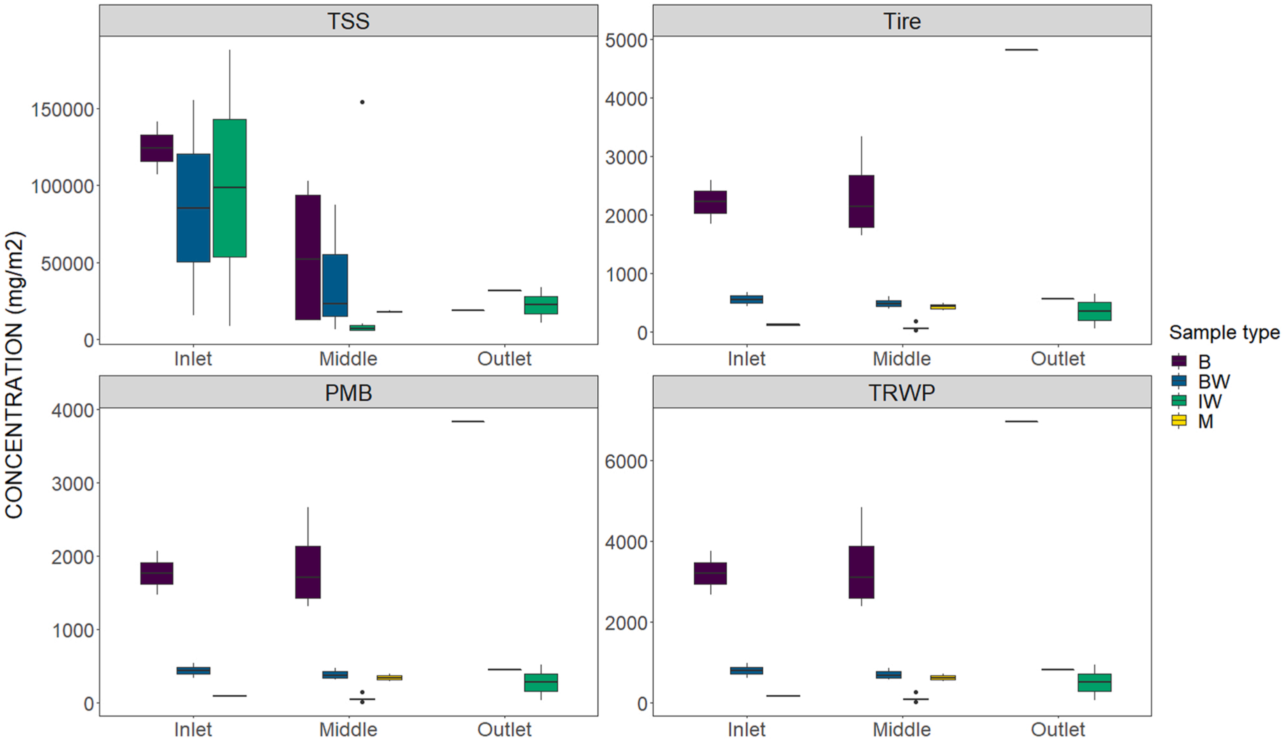

Fig. 3. Concentration of TSS, tire, PMB and TRWP through the tunnel from inlet, middle and outlet of the tunnel, as well as across the driving lane from the bank area (B), between wheel tracks (BW) in the wheel tracks (IW) and in the middle between lanes (M).

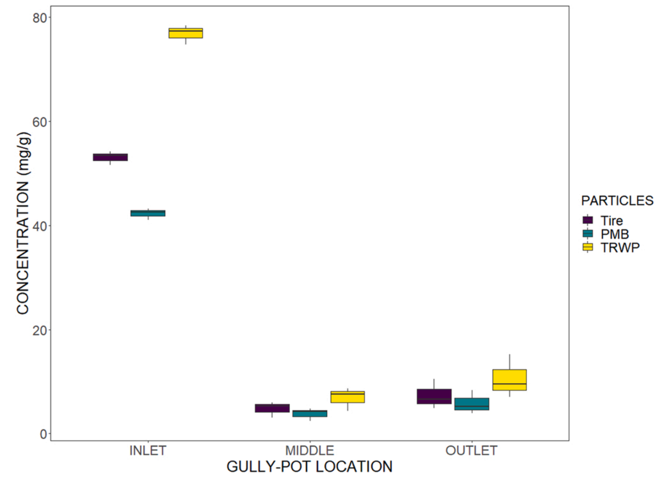

Fig. 4. Concentration of tire, PMB and TRWP particles in gully-pots from the inlet, middle and outlet of the tunnel.

simulation of 9.4% for all samples (Table SI-7). For the PMB, the concentration ranged from 11.5 to 38.1 mg/L (26.8 ± 7.33 mg/L) and the predicted standard deviation was 11% for all samples (Table SI-8). The percentage of tire, PMB and TRWP compared to TSS increased slightly between the untreated samples (tire: 2.2%, PMB: 1.8%, TRWP: 3.2%) and the treated samples (tire: 3.1%, PMB: 2.5%, TRWP: 4.5%), due to sample TWW-15, which had 5 times higher percentage of tire, PMB and TRWP compared to the average across all samples. The predicted concentration of TRWP before treatment ranged from 20.9 to 69.2 mg/L before treatment (48.6 ± 13.3 mg/L), with a predicted standard deviation of 14.2% (Fig. 5, Table SI-7, Table SI-13, Table SI-14).

After treatment, the average concentration of TSS was reduced by 69% (average 500 ± 300 mg/L), with a range of 82–1300 mg/L. The predicted concentration of tire ranged from 6.78 to 29.4 mg/L (12.5 ± 6.00 mg/L) and the standard deviation of the prediction was 9.4% for all samples. The predicted PMB concentration ranged from 5.40 to 23.4 mg/L (10.0 ± 4.78 mg/L), with a 11% standard deviation of the Monte Carlo prediction. The concentrations of TRWP varied between 9.81 and 42.5 mg/L after treatment (18.1 ± 8.68 mg/L) with an 11.2% standard deviation from the Monte Carlo simulation (Fig. 5, Table SI-7, Table SI-13, Table SI-14).

The first sample collected after treatment (TWW-15), had low TSS (82 mg/L) compared to the average of 500 mg/L as well as a high percentage of tire and PMB (12%) compared to the overall percentage excluding TWW-15 (2.57%). For the relationship between TSS and tire, PMB and TRWP, the linear regression was performed on the SBR+BR+SBS rubber values and not the predicted values, to reduce the uncertainty related to the prediction of these values. A strong relationship between TSS and SBR+BR+SBS was confirmed (adjusted $R^2 = 0.88$, Fig. 6). This relationship indicates that TSS is a possible proxy for SBR+BR+SBS rubber and subsequently tire and road wear particles in tunnel wash water.

The difference in size distribution of particles in the tunnel wash water before and after treatment was small, but significant (RDA, $p = 0.008$, Fig. 7, Table SI-15) when sample TW-15 is excluded as an outlier. The most visible difference between the before and after samples was the presence of particles > 350 µm after treatment (TWW-16, TWW-17and TWW-18), which were not present in the samples before treatment. The largest mass of particles was found in the 10–30 µm size class, with an average contribution of 42% in the untreated samples and 47% in the treated samples. In fact, over 83% of the particles in both untreated and treated samples were < 30 µm in size. The untreated samples had a higher percentage of the smallest particles (>2.5 µm and 2.5–10 µm) For the fraction 50–350 µm, where the main mass of tire particles is expected (Kreider et al., 2010), the mass of particles was

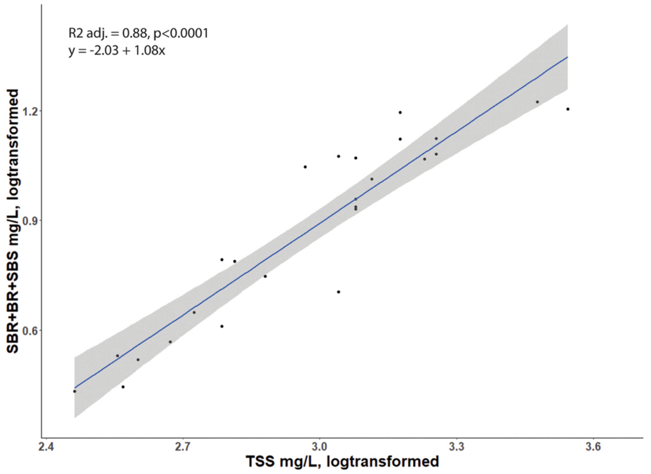

Fig. 6. Linear regression between TSS (log transformed) and SBR+BR+SBS (log transformed) for all samples before and after treatment, except TWW-15 (low TSS).

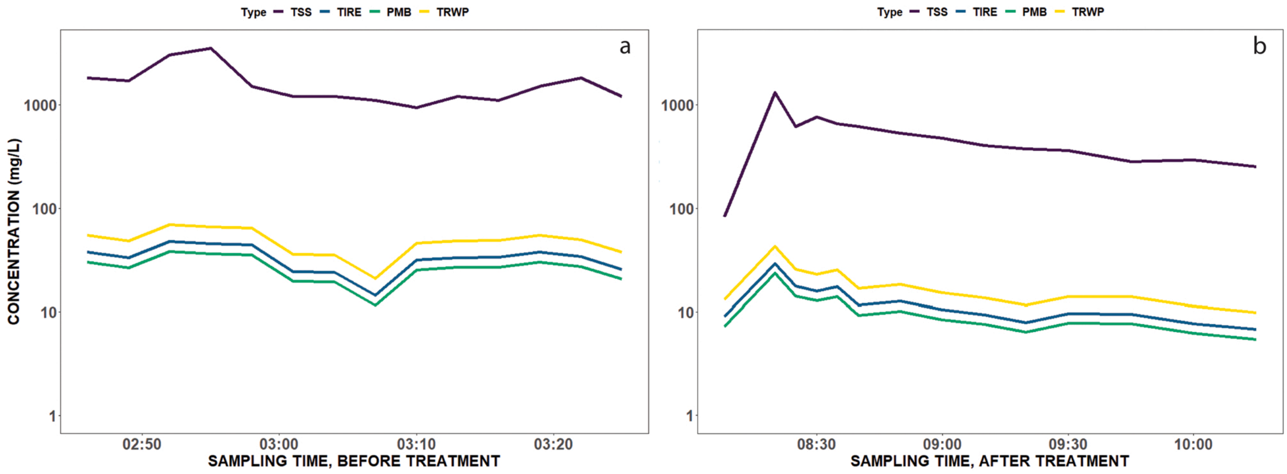

Fig. 5. Concentrations of total suspended solids (TSS), tire particles, PMB particles and tire and road wear particles (TRWP) in the tunnel wash water before (a) and after (b) treatment. The samples are displayed as a time-series for the sampling, from 02:46–03:25 (April 21st, 2020) before treatment and from 08:08–10:15 (May 12th, 2020).

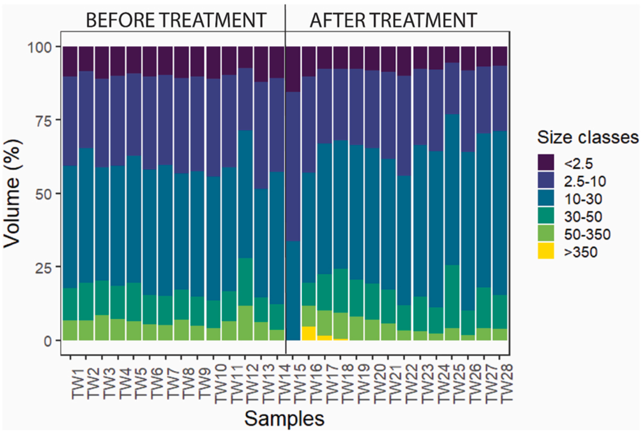

Fig. 7. Size distribution of particles (total suspended solids $<2000\ \mu m$) in tunnel wash water before and after treatment. Sample TW15 is the first sample released of the treated tunnel water and differs significantly from the others (outlier). The difference between samples before and after treatment was significant (RDA, $p = 0.008$).

6.4% before treatment and reduced to 5.3% after treatment. The difference in concentration of tire and PMB before and after treatment was statistically significant (ANOVA, $p < 0.0001$).

4. Discussion

4.1. Road surface

The concentrations of tire, PMB and TRWP were highest in the outlet and bank area, which supports hypothesis I. For TSS, the high accumulation in the bank area agrees with previous studies of tunnel road dust using WDS (NPRA, 2017, 2021b), however, in these studies the concentrations were higher in the inlet compared to the outlet and the overall concentrations reported in the present study were significantly lower compared to previous studies using WDS (200–400 g/m^2 (Gustafsson et al., 2019; NPRA, 2021b).

Previous studies have reported up to 10% organic components in tunnel road dust (NPRA, 2017), which agrees with the percentage of tire, PMB and TRWP found at the tunnel outlet in the present study. However, the percentage of tire, PMB and TRWP was significantly lower in the inlet and in the middle of the tunnel. The reason for this difference could be that the inlet area is the highest point of the tunnel and receives a lot of runoff from outside of the tunnel. This could potentially include a higher concentration of other particles, which therefore dilutes the concentration of tire and PMB particles in this area. Another reason might be that tire and PMB particles are transported through the tunnel by the suspension made by traffic and wind (piston effect; Moreno et al., 2014), as well as runoff when it precipitates, and accumulating in the lower areas of the tunnel. Another possibility is that a larger portion of the tire and PMB particles in the inlet area ends up in the gully-pots of that area.

The macrostructure of the pavement can have a substantial impact when comparing the particle load retained in the road surface (Lundberg et al., 2017). In the Smestad tunnel, the road surface is asphalt concrete (maximum aggregate size of 11 mm), which has a lower texture and less area for particles to be retained (especially in the wheel tracks), compared to coarser stone mastic asphalts with maximum aggregate size of 16 mm (Gustafsson et al., 2019; NPRA, 2017). The number of comparable studies for the mass of tire and road-wear rubber, tire particles, PMB and TRWP is limited. However, one study of street runoff from Germany (Eisentraut et al., 2018) used a mass-based analysis (Thermal Desorption GC/MS) and found SBR concentrations between 3.9 and 8.9 mg/g in the street runoff, which is over 40 times lower than the highest values of rubber reported for tunnel road surface in this present study, although the rubber concentration in the present study also includes SBS rubber from the PMB surface. One likely reason for this major difference may be the sampling procedure. A previous study using a Wet Dust Sampler has demonstrated that 90% of particles ($<180\ \mu m$) are collected using three shots of each area (NPRA, 2021b), whereas only 60% of particles 180–5000 μm were collected by three shots. Compared to a previous study of tunnel road dust (Klöckner et al., 2021), where the middle bank area (110 mg/g) and the outlet bank area (120 mg/g) were analysed, the TRWP concentration in Smestad reported in the present study is more than three times higher for both areas. Another study analysed road dust mixtures collected by road sweeper trucks (Klöckner et al., 2020) and the concentrations were more than four times lower (8.1–14 mg/g) than the average concentrations in road dust in the present study. Different sampling procedures, such as using multiple sample shots with a WDS compared to applying a commercial vacuum cleaner, might be the main reasons for these differences. Other explanations might be local, such as the different length, slope and AADT for the different roads analysed.

4.2. Gully-pots

The results for the gully-pots confirmed that tire, PMB and TRWP can be retained in high concentrations and thus supported a rejection of hypothesis II. Previous studies where the possibility of retention in gully-pots are discussed, have suggested low treatment efficiency for tire and road wear particles gully-pots (Blecken, 2016; Vogelsang et al., 2018) due the density and size of the particles. Studies that have tested the efficiency of gully-pots have found that the efficiency depends on the particle size, the particle geometry and the flow within the gully-pot, where the efficiency decreases as the sediment builds up in the gully pot (Rietveld et al., 2020). The concentration of TRWP at the tunnel inlet in the present study was comparable to the concentrations found in gully-pots from municipality roads (0.8–150 mg/g; (Mengistu et al., 2021) and sediment from a road runoff treatment (130 ± 15 mg/g; Klöckner et al., 2019), while it should be kept in mind that these studies were based on different analytical approaches. The major difference between the inlet gully-pot and the outlet in the current study is the opposite of the results for the road surface, where the concentration in the outlet was significantly higher compared to the other areas. One possible reason for the differences between inlet and outlet, as was also observed for the road surface, is that the inlet area is the highest point of the tunnel and receives a lot more runoff from outside of the tunnel. This runoff is likely to flow into the gully-pots, causing a higher percentage of tire and PMB particles to accumulate in the gully-pot sediment, as well as bringing in a higher percentage of other particles from outside of the tunnel to the inlet area. The particle concentration in mid area and the outlet of the tunnel are affected by the traffic inside the tunnel with little water flowing through during normal conditions. This is also supported by the observation of drier sediment present in the middle and outlet gully-pots.

4.3. Tunnel wash water

Compared to previous studies, the concentrations of TRWP in untreated tunnel wash water agrees with values reported for road runoff (3–180 mg/L: (Baumann and Ismeier, 1998; Kumata et al., 2000, 1997, 2002); Parker-Jurd et al. (2021); (Reddy and Quinn (1997); Wik and Dave (2009)). It should be noted that these studies all represent different analytical methods and calculations, so comparisons should be made with caution. A recent study has also reported TRWP in the range of 6.4–18 mg/L in an Australian urban creek receiving stormwater runoff (Rauert et al., 2022), although these values represent the diluted runoff mixed with the river water. The hypothesis that untreated tunnel wash would far exceed the road runoff concentrations due to the accumulation in tunnels was not supported based on these comparisons. On the contrary, the concentrations of tire and PMB particles found in the untreated tunnel wash water were significantly lower compared to a recent study of tire and PMB particles in road-side snow in Oslo, Norway (Rødland et al., 2022a). One explanation might be that the tire, PMB and TRWP particles in the tunnel system are divided between different tunnel compartments, such as the road surface and the gully-pots, whereas the road-side snow traps more of the total tire, PMB and TRWP production and therefore features higher concentrations. Different types of road surfaces could be one contributing factor, as discussed in the study of road-side snow that the sites with concrete asphalt had a higher concentration of TSS and a higher calculated contribution of SBS based on road abrasion factors for concrete asphalt (Rødland et al., 2022a). As the road surface in the Smestad tunnel is also made of concrete asphalt, the observed high TSS compared to tire, PMB and TRWP might indicate that there is a higher road wear contribution here compared to sites with stone mastic asphalt.

The strong correlations found between TSS and SBR+BR+SBS rubber could potentially provide a valuable tool for environmental monitoring of tunnel wash water. Applying online sensors in tunnels, the TSS and turbidity data could then be collected in real-time for a large number of tunnels, and TSS and turbidity could be used as proxy tire, PMB and TRWP concentrations. More data is needed to establish the basis for such a tool, however, the impact on environmental monitoring could be high, as monitoring with sensors would potentially reduce the costs for sampling and analysis.

The retention efficiency reported in this study (63%) is lower than expected according to the previous assumption of > 80% retention in sedimentation treatment. This supports the rejection of hypothesis IV, stating that the current treatment of tunnel wash water in Smestad is efficient in retaining tire, PMB and TRWP. The possible issues with the sedimentation treatment in Smestad are also highlighted by sample TW-15. This is the first treated sample released into the rain garden, and it was characterized by low TSS (5 times lower than the average) and a high percentage of tire and PMB compared to the average and particle sizes > 30 μm. This indicates that the first samples may represent the water that has been treated inside the pump house basin and not the treated water from the sedimentation basin. The following samples may have higher TSS and particle size distribution due to the turbulence and resuspension of particles upon the release. This is also supported by the size distribution analysis, where the difference in size distribution before and after treatment was low. A small decrease in the volume of particles > 50 μm was observed for the after-treatment samples compared to before treatment, however, three of the after-treatment samples also contained particles > 350 μm, which had not been observed in the previous samples. This further indicates that turbulence and resuspension of particles occurs when the treated water is released into the rain garden and may be the reason why the retention efficiency for TSS, tire and PMB was lower than expected for this tunnel.

The size distribution data may provide us with more understanding of the treatment process. Although the total particle retention (TSS) had a 69% retention in the treatment basin, this did not have a significant impact on the size distribution. Only a small decrease was observed in the second largest size class (50–350 μm), where the main mass of tire particles is expected to be found (Kreider et al., 2010). The Swedish road authorities have reported that treatment by sedimentation is not suitable for particle sizes < 10 μm, where infiltration treatment is needed (Andersson et al., 2018). As 41% of the particles before treatment and 37% after treatment were < 10 μm, this indicates that the sedimentation basin is less efficient in the removal of particle-bound pollutants. Although low efficiency was found for this tunnel, different tunnels do have different types of treatment, and these should be investigated to evaluate to overall efficiency of wash water treatment for tire, PMB and TRWP.

Other factors that could impact the treatment efficiency is the use of soap. As previously mentioned, soap is applied in the Smestad tunnel, and the percentage of soap used in this specific tunnel is approximately 0.2% (NPRA, 2022). However, the use of soap has been demonstrated to lower the treatment efficiency for several metals (Aasum, 2014): the use of 0.3% soap in the wash water reduced the retention of Zn from 98% (no soap) to only 33%, and for Cu, the use of 3% soap reduced the retention from 99% without soap to as low as 25% retention. The treated tunnel wash water in Smestad still had soap in it when released into the rain garden, indicating that the soap could be a crucial factor in the low treatment efficiency. Furthermore, temperature could impact the treatment of tunnel wash water, where lower temperatures (4 °C) cause slower sedimentation compared to higher temperatures (20 °C) (Garshol et al., 2015). The average outdoor temperature for the treatment period was 8.4 °C (Yr, 2020), which could have an impact on the treatment efficiency.

4.4. *Uncertainty evaluation*

For studies applying mass-based methods such as pyrolysis GC/MS, the use of reliable pyrolysis markers with low variability is crucial. This has been discussed as a major issue when it comes to analysing tire particles, as different pyrolysis products have displayed large variations in different reference tires tested (Rauert et al., 2021; Rødland et al., 2022b). The major impacting factor causing variability is the different microstructures in the composition of SBR and BR rubber, which can cause variability in the pyrolysis products (Choi, 2001; Choi and Kwon, 2020; Miller et al., 2021). The use of different types of SBR in tires, such as emulsion-SBR and solution-SBR has also been brought to attention in previous literature (Miller et al., 2021; Rødland et al., 2022b). Another important aspect, which has also been brought to attention by Wagner et al. (2022), is the need to address how aging of tire particles in the environment impacts the SBR+BR content and the pyrolysis products used as marker compounds. Aging is also important for the SBS rubber content in PMB. The presence of SBS rubber in samples has so far only been addressed by one environmental study from Norway (Rødland et al., 2022b), however, several countries such as Australia, United Kingdom, Russia, Denmark and Sweden apply PMB asphalt on roads with high traffic volume (EAPA, 2018). As various polymers and rubbers, not just SBS, can be applied, it is important to investigate the presence of SBS in the road surface before analysing samples for SBR+BR. SBR and SBS have identical pyrolysis products, as well as BR sharing overlapping products with SBS, so without separation between SBR+BR and SBS, TRWP concentrations will be overestimated in a sample that contains both. To reduce the uncertainty caused by variations in the pyrolysis products, the present study applies a method where the combination of four different pyrolysis products (*benzene, α-methylstyrene, ethylstyrene and butadiene trimer*) are used for quantification. This method has displayed lower variability (40% S.D) in reference tires compared to the single markers previously proposed by other studies (62–85% S.D.) (Rødland et al., 2022b), suggesting that the method reduces the uncertainty related to variation in single markers compared to previous methods. A second challenge in analysing tire particles concerns the variable rubber content in different commercial tires. Previous studies have reported a rubber content of 50% in personal vehicle tires (44% SBR+BR) and 50% in truck tires (45% NR) (Unice et al., 2012), however, in our recent study we found large variations of SBR+BR content in commercial tires, not in line with the 50% assumption (PV: 19–47% SBR+BR, HV: 11–68% SBR+BR) (Rødland et al., 2022b). Variations in commercial tires have also been reported by other studies (Goßmann et al., 2021; Rauert et al., 2021). To reduce the uncertainty in calculations from rubber concentrations to tire concentrations, the SBR+BR concentrations in relevant seasonal tires are used in the present study, and the calculations are applied in Monte Carlo simulations to predict the tire concentrations present in a sample. The use of these simulations allows us to predict the possible mean concentrations of tire present in the sample, as well as a variety of statistics such as predicted standard deviations, minimum and maximum values and percentiles. For the tire concentrations, % predicted standard deviation from the predicted mean was 9.4% (SI Table SI-1, SI-4 and SI-7), which demonstrates that there are some uncertainties related to the calculation of tire particles. This uncertainty is influenced by the large variation of SBR+BR content in the PV tires (96.8%, Crystal ball sensitivity analysis), which underlines the need for relevant and reliable reference tires. The estimated SBS rate contributes 2.4% to the variation, whereas the SBR+BR variation in HV tires only contributes 0.8% of the variation. For the PMB particles, the variation of SBS reported in PMB asphalt is low compared to the variation in tires because the input data has a lower variance and the model is only influenced by the SBS ratio for Smestad (100%, Crystal Ball sensitivity analysis), and therefore the % predicted standard deviation of PMB concentrations is lower (11%).

Uncertainty is also important to consider for the calculated TRWP concentrations. Previous literature has suggested that urban roads with lower speed limits and traffic density have a high percentage of encrusted particles (>73%, Klöckner et al., 2020) compared to highways with higher speed limits and traffic density (<10%:Sommer et al., 2018; 25%: Klöckner et al., 2021). Increased speed limit and traffic density increases the distance a tire wear particle is transported from the point of release (Gustafsson et al., 2009; Rødland et al., 2022a), thus, decreasing the potential mixing with mineral particles from the road surface. In tunnels, the semi-enclosure of the tunnel walls inhibits the transportation, however, as demonstrated by the present study and others (NPRA, 2017, 2021b), a substantial proportion of tire and road wear particles accumulate in the side bank area, which is outside of the driving lane and not contributing to the increased mixing of tire and mineral particles on the surface. Thus, assuming a generalized 50% mineral encrustment (Kreider et al., 2010; Unice et al., 2013) may overestimate TRWP concentrations for highways, including in road tunnels, as well as underestimating the TRWP concentrations for urban roads. In the present study, Monte Carlo simulation was applied to calculate the predicted TRWP concentrations based on tire concentrations and the reported distribution of mineral encrustment. However, the available data on mineral encrustment on TRWP for different road types and sample matrices are currently limited to three studies (Klöckner et al., 2021; Kreider et al., 2010; Sommer et al., 2018), hence these calculations are associated with large uncertainties. Even so, the use of Monte Carlo simulations for predicting the expected TRWP concentrations in the sample is promising and the method can be improved by increasing the data available for mineral encrustment of TRWP. Future studies should investigate the impact on mineral content by different variables such as driving conditions (highway, urban, rural), traffic speed, the use of studded tires and different types of road surfaces. As the input variable in the TRWP model is the predicted tire values, the TRWP model is also subject to the variations in tire reference data. Hence, improving both the data available for mineral encrustment and SBR+BR content in relevant reference tires will improve the prediction of TRWP. Another aspect not mentioned in any previous literature is how road wear particles with polymer-modified bitumen interacts with and impacts the TRWP. In this study, tire and PMB particles are reported and discussed as separate particles. This is mainly because there is not enough research available on how these particles interact with each other in the environment. To fully understand the transport mechanisms and the possibilities with mass-based analysis, more research is needed on the impact of PMB particles on TRWP.

The demonstrated use of multiple pyrolysis products as markers as well as Monte Carlo simulations for tire, PMB and TRWP calculations demonstrates the possibility of applying local conditions relevant for each sample (reference tires, asphalt abrasion, mineral encrustment) to improve the results and reduce the uncertainties.

5. Conclusions

The lowest concentrations of tire and PMB particles were found on the road surface of the tunnel, with the second highest concentration in the gully pots and the highest concentration in the tunnel wash water. For the road surface, the concentrations were high compared to previous studies, and validated the first part of hypothesis I, that most of these particles accumulate in the side bank area. However, in contrary to previous studies, the highest concentrations were found in the outlet of the tunnel and not in the inlet, rejecting the second part of hypothesis I. Our findings confirm that it is important to clean the surface both before the tunnel wash and after the wash. This will reduce the particle load in the tunnel wash water, which will decrease the release of pollutants from tunnels without water treatment. Removing particles in the smaller size range (<50 μm), which had the highest number of particles, from the road surface may also increase the retention efficiency of sedimentation treatment. Cleaning the road surface again after the tunnel wash helps to remove particles from the surface before it settles in the road surface macrostructure. High concentration of tire, PMB and TRWP were also reported in the inlet gully-pot, with concentrations comparable to sediment in road treatment basins. This was in contrast to previous studies, thus rejecting hypothesis II. The concentration was lower in the middle and outlet gully-pots, also displaying a different pattern compared to the accumulation of tire, PMB and TRWP on the road surface. For the tunnel wash water, the concentration of tire, PMB and TRWP in untreated water was comparable to previous studies of road runoff, however, significantly lower compared to meltwater from road-side snow, thus rejecting hypothesis III. The retention of tire, PMB and TRWP (63%) and TSS (69%) were also lower than expected (>80%) based on previous literature for tunnel wash water, rejecting hypothesis IV. Factors such as soap and temperature could be influencing the treatment, as well as the large fraction of small sized particles that potentially hampers removal by sedimentation alone. The second treatment step at Smestad (rain garden) could not be analysed and assessed in this study, and future research on this tunnel should aim to include this second treatment step for comparison. The concentrations of tire, PMB and TRWP in the untreated tunnel wash water are relevant for the high number of tunnels in Norway that release untreated tunnel wash water into freshwater and marine recipients. There are still issues related to using different analytical approaches in different studies, making comparisons between different matrices such as road runoff, road-side snow or road dust difficult. Large uncertainties are also related to the analysis of tire and road wear rubber with Pyrolysis GC/MS, as well as the calculations of tire, PMB and TRWP based on the rubber concentrations.

Future research should focus on finding a common approach to both analysis and calculation of tire, PMB and TRWP, as well as addressing the uncertainties related to these calculations. The impact of aging on the pyrolysis markers applied should be addressed, and an increase of available data for SBR+BR content in tires, road abrasion including PMB and mineral content of TRWP is needed.

CRediT authorship contribution statement

Elisabeth Rødland: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft, Writing – review & editing, Visualization. **Ole Christian Lind**: Conceptualization, Writing – review & editing. **Malcolm J. Reid**: Conceptualization, Writing – review & editing. **Lene S. Heier**: Conceptualization, Writing – review & editing. **Emelie Skogsberg**: Investigation, Writing – review & editing. **Brynhild Snilsberg:** Investigation, Writing – review & editing. **Dagfin Gryteselv:** Investigation, Writing – review & editing. **Sondre Meland:** Conceptualization, Investigation, Writing – review & 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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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Appendix A. Supporting information

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