THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Occurrence of Traffic-Derived Microplastics in Different Matrices in the Road Environment

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Cover: Illustration of the major transport routes of traffic-derived particles (Ida Järlskog, 2022).

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ABSTRACT

The prevalence of microplastic contamination has raised concerns about the potential risk and impact on the global environment. Traffic-derived microplastics, i.e., tire wear particles (TWP), polymer-modified bitumen, and road markings contribute to the emissions, and TWP are assumed to be one of the largest sources of microplastic emissions. Due to analytical difficulties, there is still a knowledge gap regarding transport routes, environmental concentrations, and toxicity. This thesis aims to investigate the occurrence of traffic-derived microplastics in several traffic environments and thereby increase the understanding of the particles.

Samples were collected on the road surface (**Paper I–IV**), in the stormwater (**Paper I, II**, and **IV**), in the air (**Paper IV**), and in material collected by a street sweeper (**Paper I–II**). In addition to environmental samples, a road-simulator was used to generate TWP in a controlled environment enabling comparison between tire types and brands resulting in a deeper understanding of the characteristics and physicochemical properties of TWP (**Paper V**). Different sample preparation steps such as optimised density separation, solvent cleaning, and size fractionation have been assessed, and several analytical methods light microscopy, SEM/EDX, FTIR, and pyr-GC/MS have been evaluated and used for the analysis of traffic-derived particles. Further, a novel method, automated SEM/EDX single particle analysis coupled to a machine learning classifier, has been developed for the analysis of TWP and other traffic-derived particles (**Paper III**, implemented in **Paper IV**). The automated SEM/EDX determined the size, shape, surface texture, and elemental composition of the different particles, and was able to categorize the particles into several subclasses: TWP, bitumen, road markings, metals, organics, and minerals. The estimated absolute masses showed that the fine fraction (2–20 µm) corresponds to more than 50w% of the TWP and bitumen wear particles independently of the sample matrix indicating that TWP can both affect the PM10 concentrations and be transported long distances through water and air (**Paper IV**). Further, it was concluded that the stormwater system is an important transport route for traffic-derived particles, especially since the road runoff in Sweden is not commonly treated prior to release to recipients. Street sweeping as a potential measure to prevent the spreading of TWP was evaluated in **Paper I–II**. Even though the street sweeper collects considerable amounts of material containing high concentrations of TWP, metals, and organic pollutants, no clear reduction was detected neither on the road surface nor in the stormwater. Besides traffic-derived microplastics, **Paper II** analysed metals and organic pollutants. The results showed concentrations of metals, PAH, phthalates, and aliphatic hydrocarbons exceeding the national guidelines.

The result from this thesis contributes to an increased knowledge about the properties and composition of TWP as well as the occurrence of traffic-derived microplastics in different environments. The results can be used as validation against theoretical emissions and transport models. The results have also highlighted the importance of including fine particles (<20 µm) in forthcoming works.

KEYWORDS: traffic-derived microplastics; tire wear particles; road dust; street sweeping; automated SEM/EDX; optimised sample preparation; non-exhaust particles; organic pollutants; environmental samples; road simulator

SAMMANFATTNING

Den potentiella risk och påverkan på miljön som kontaminering av mikroplast kan tänkas ha på den globala miljön har väckt stor oro. Trafikrelaterade mikroplaster, det vill säga partiklar från däck, polymermodifierad bitumen och vägfärg bidrar till utsläppen och däckslitage antas vara den enskilt största källan till mikroplastutsläpp i naturen. Analyssvårigheter har resulterat i att det fortfarande saknas kunskap kring toxicitet, partiklarnas transportvägar samt koncentration i olika miljöer. Den här avhandlingen syftar till att undersöka förekomsten av trafikrelaterad mikroplast i ett flertal olika trafikmiljöer och därmed öka förståelsen för partiklarnas spridning.

Prover samlades in på vägytan (**Artikel I-IV**), i dagvatten (**Artikel I, II och IV**), i luften (**Artikel IV**) och i det material som en städmaskin samlat upp efter gatusopning (**Artikel I och II**). Som ett komplement till miljöprover användes en vägsimulator för att generera däckpartiklar i en kontrollerad miljö vilket möjliggjorde en jämförelse mellan olika däcktyper och däckmärken. Detta gav en fördjupad kunskap kring däckpartklar och dess kemiska sammansättning (**Artikel V**). Olika provberedningssteg så som optimerad densitetsseparation, tvätt med lösningsmedel, och storleksfraktionering har utvärderats vid analys av däckpartiklar i olika matriser. Därtill har ljusmikroskop, SEM/EDX, FTIR och pyr-GC/MS utvärderats för analys av trafikrelaterade partiklar (**Artikel I, II, V**). Utöver det har en ny metod utvecklats, en automatiserad SEM/EDX för analys av specifika partiklar som tillsammans med maskininlärning möjliggjorde klassificering av trafikrelaterade partiklar (**Artikel III**, implementerad i **Artikel IV**). Den automatiserade SEM/EDX-metoden analyserade storlek, form, ytstruktur, och grundämnessammansättning för varje enskild partikel. Den automatiserade klassificeraren delade in partiklarna i följande grupper: däckpartiklar, bitumen, vägmarkering, metallpartiklar, organiska partiklar och mineral. Den beräknade partikelmassan visade att de fina partiklarna (2–20 µm) stod för mer än 50w% av den totala vikten av däck- och bitumenpartiklar, oberoende av provmedia vilket indikerar att däckpartiklarna dels kan försämra luftkvaliteten i from av PM10, men att de även kan transporteras långa sträckor via både luft och vatten (**Artikel IV**). Detta tydliggör att de fina partiklarna spelar en mycket stor roll för de totala utsläppen. Det kunde även konstateras att dagvattensystemet är en viktig transportväg för trafikrelaterad mikroplast, särskilt eftersom en stor del av avrinningen i Sverige inte behandlas innan det släpps ut i naturen. Gatusopning utvärderades som en potentiell åtgärd för att förhindra spridningen av däckpartiklar (**Artikel I och II**). Sopmaskinen samlade in stora mängder material som vid analys visade sig innehålla höga koncentrationer av både däckpartiklar, metall och organiska miljögifter. Trots det gick det inte att detektera en tydlig minskning av dessa partiklar och ämnen vare sig på vägbanan eller i dagvattnet. Som ett tillägg till trafikrelaterad mikroplast analyserades både metaller och organiska miljögifter i **Artikel II**. Resultaten visade koncentrationer som överskred gränsvärdena för metaller, PAH, ftalater och alifatiska kolväten.

Resultaten från den här avhandlingen har bidragit till en ökad förståelse kring egenskaper och kemisk sammansättning hos trafikrelaterad mikroplast. Vidare har avhandlingen ökat kunskapen om förekomsten av trafikrelaterad mikroplast i olika trafikmiljöer. Resultaten kan bland annat användas som en validering av teoretiskt framtagna emissioner och transportmodeller. Resultaten visar även på vikten av att inkludera fina partiklar (<20um) i framtida arbeten.

NYCKELORD: trafikrelaterad mikroplast; däckpartiklar; vägdamm; gatusopning; automatiserad SEM/EDX; optimerad provberedning; icke avgasrelaterade partiklar; organiska miljögifter; miljöprover; provvägssimulator

LIST OF PUBLICATIONS

This thesis is based on the work contained in the following papers:

I. Järlskog, I., Strömvall, A-M., Magnusson, K., Gustafsson, M., Polukarova, M., Galfi, H., Aronsson, M and Andersson-Sköld, Y. (2020). Occurrence of tire and bitumen wear microplastics on urban streets and in sweepsand and washwater. *Science of the Total Environment;* **729**: 138950. 10.1016/j.scitotenv.2020.138950.

II. Järlskog, I., Strömvall, A-M., Magnusson, K., Galfi, H., Björklund, K., Polukarova, M., Garção, R., Markiewicz, A., Aronsson, M., Gustafsson, M., Norin, M., Blom, L and Andersson-Sköld, Y. (2021). Traffic-related microplastic particles, metals, and organic pollutants in an urban area under reconstruction. *Science of the Total Environment*; **774**; 145503. 10.1016/j.scitotenv.2021.145503.

III. Järlskog, I., Jaramillo-Vogel, D., Rausch, J., Perseguers, S., Gustafsson, M., Strömvall, A-M and Andersson-Sköld, Y. (2022). Differentiating and quantifying carbonaceous (tire, bitumen, and road marking wear) and non-carbonaceous (metals, minerals, and glass beads) non-exhaust particles in road dust samples from a traffic environment. *Water, Air, & Soil Pollution*; **233**: 375. 10.1007/s11270-022-05847-8.

IV. Järlskog, I., Jaramillo-Vogel, D., Rausch, J., Gustafsson, M., Strömvall, A-M and Andersson-Sköld, Y. Concentrations of tire wear and other traffic-derived non-exhaust particles in the road environment. (*Submitted Manuscript*).

V. Wilkinson, T., Järlskog, I., Aristéia de-Lima, J., Mattsson, K., Gustafsson, M., Andersson-Sköld, Y and Hassellöv, M. Tire and road wear particles (TRWP) – variable physicochemical properties and wear characteristics influenced by tire formulation and road surface. (*Manuscript*).

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Ida Järlskog´s contribution to the scientific papers is the following:

Paper I: Participation in field measurements, lab analyses, data collection, analysis and assessment of the data. Wrote most parts of the paper, editing and revising after consultation with co-authors and external review.

Paper II: Field measurements, analysis and assessment of the data, wrote most parts of the paper, editing and revising after consultation with co-authors and external review.

Paper III: Participation in planning of the study and collecting all field samples, determined a method for sample preparation, prepared the field samples, and performed some of the lab analyses. Analysed, compiled, and made the graphical illustrations of the data. Wrote some parts of the paper. Editing and revising after consultation with co-authors and external review.

Paper IV: Participation in planning of the study, responsible for all field measurements and the sample preparation. Analysis and assessment of the data, made the illustrations and wrote most parts of the paper. Editing and revising after consultation with co-authors and external review.

Paper V: Responsible for running the road simulator and for sample collection. Made some of the sample preparation after consultation with co-authors, wrote some parts of the paper and made some of the figures after data treatment.

Ida Järlskog has contributed to the following work and publications, which are not appended to the thesis:

Gomiero, A., Vianello, A., Vollertsen, J., Øysaed, K., **Järlskog, I** and Andersson-Sköld, Y. (2019). *Assessment of microplastics occurrence and composition in Swedish road wastewater through a thermal degradation method.* Conference: SETAC Europe 29th Annual Meeting (poster).

Gustafsson, M., Blomqvist, G., **Järlskog, I**., Lundberg, J., Janhäll, S., Elmgren, M., Johansson, C., Norman, M and Silvergren, S. (2019). Road dust load dynamics and influencing factors for six winter seasons in Stockholm, Sweden. *Atmospheric Environment.* Volume 2. 10.1016/j.aeaoa.2019.100014

Lundberg, J., Blomqvist, G., Gustafsson, M., Janhäll, S and **Järlskog, I**. (2019). Wet Dust Sampler- a Sampling Method for Road Dust Quantification and Analyzes. *Water, Air & Soil Pollution.* Volume 230. 10.1007/s11270-019-4226-6

Andersson-Sköld, Y., Johannesson, M., Gustafsson, M., **Järlskog, I**., Lithner, D., Polukarova, M and Strömvall., A-M. (2020). *Mikroplast från däck- och vägslitage. En kunskapssammanställning.* VTI Report 1028.

Andersson-Sköld, Y., Johannesson, M., Gustafsson, M., **Järlskog, I**., Lithner, D., Polukarova, M and Strömvall., A-M. (2020). *Microplastics from tyre and road wear: a literature review.* VTI Report 1028A.

Polukarova, M., Markiewicz, A., Björklund, K., Strömvall, A-M., Galfi, H., Andersson-Sköld, Y., Gustafsson, M., **Järlskog, I** and Aronsson, M. (2020). Organic pollutants, nano- and microparticles in street sweeping road dust and washwater. *Environment International.* Volume 135. 10.1016/j.envint.2019.105337

Järlskog, I., Rausch, J., Jaramillo, D., Gustafsson, M and Andersson-Sköld, Y. (2020). *Analyzes of tire and road wear particles (TRWP) in environmental road dust samples with an automated SEM/EDX and single particle classification system.* Conference: SETAC Europe 30th Annual Meeting (poster).

Strömvall, A-M., **Järlskog, I**., Gustafsson, M., Polukarova, M., Galfi, H., Andersson-Sköld, Y., Magnusson, K., Markiewicz, A., Aronsson, M., Garção, R., Norin, M and Blom, L. (2020). *Processes for Transport of Tyre and Road Wear Microplastic Particles, Metals and Polycyclic Aromatic Hydrocarbons in Road Runoff – Characterizations and a Sorption and Desorption Laboratory Study.* Conference: SETAC Europe 30th Annual Meeting (poster).

Johannesson, M., Andersson-Sköld, Y., Gustafsson, M and **Järlskog, I**. (2021). *Reporting of the Government commission on microplastics from road traffic.* VTI Report 1089.

Järlskog, I and Gustafsson, M. (2022). *Trafiken, ett miljöproblem som rullar på.* Chapter to a thematic report about wear particles, the Swedish Environmental Protection Agency.

Mattsson, K., Aristéia de Lima, J., Wilkinson, T., **Järlskog, I**., Ekstrand, E., Andersson-Sköld, Y., Gustafsson, M and Hassellöv, M. (submitted). *Tire formulations are the key to environmental emissions of tire and road wear particles.*

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It is simply this: do not tire, never lose interest, never grow indifferent—lose your invaluable curiosity and you let yourself die. It's as simple as that. Tove Jansson, Fair play

Linköping, August 2022
Ida Järlskog

DEFINITIONS AND ABBREVIATIONS

The following definitions and abbreviations are used in the text of the thesis:

AADT	Annual average daily traffic
ATR-FTIR	Attenuated Total Reflectance- Fourier-Transform Infrared Spectroscopy. A sampling technique used in conjunction with FTIR.
$CaCl_2$	Calcium chloride (≤ 1.4 g/cm^3), salt solution used for density separation
Elastomer	A viscoelastic polymer. Rubber is a typical elastomer
EMC	Event mean concentration
FTIR	Fourier-Transform Infrared Spectroscopy. Analytical method used to obtain an infrared spectrum of absorption or emission of a sample
GC/MS	Gas Chromatography-Mass Spectrometry. Analytical method used for quantification of polymers within a sample
Microplastics	Synthetic solid particles or polymetric matrices <5 mm of either primary or secondary manufacturing origin, which are insoluble in water.
ML	Machine learning
NaCl	Sodium chloride (≤ 1.2 g/cm^3), salt solution used for density separation
NaI	Sodium iodide (≤ 1.9 g/cm^3), salt solution used for density separation
NR	Natural rubber, used in tires. Polyisoprene is often used as a marker
OP	Organic pollutants
PAH	Polycyclic Aromatic Hydrocarbons
PM_{10}	Particles with an aerodynamic diameter <10 µm
PMB	Polymer modified bitumen
Polybutadiene	Used as a marker for TWP, e.g., in pyr-GC/MS analyses
Polyisoprene	Used as a marker for TWP, e.g., in pyr-GC/MS analyses
Polymer	A synthetic or natural chemical compound consisting of large molecules, composed of long chains with repeating subunits
POP	Persistent Organic Pollutants

Pyr-GC/MS Pyrolysis- Gas Chromatography-Mass Spectrometry. Analytical method, the sample is decomposed in the pyrolizer under an inert atmosphere. The smaller molecules are separated by the GC and detected in the MS. Used for e.g., characterization of polymers and elastomers

Road dust Collective name of all particles found on the road surface or adjacent to the road (e.g., minerals, brake wear, tire wear, road wear, road markings, organic particles, and vehicle wear)

Road simulator A laboratory equipment at VTI for simulating wear between tires and a pavement surface, consisting of four wheels running over a pavement ring.

Rubber Elastomer produced either from the rubber tree (known as natural rubber) or produced synthetically from different chemical substances.

Runoff The overflow of water that drains off the road surface

SBR Styrene-butadiene-rubber, synthetic rubber used in tires

SEM/EDX Scanning Electron Microscope with Energy-Dispersive X-ray spectroscopy. Analytical method. Particles are identified based on elemental composition, shape, size, and surface.

Sigma-2 Passive air sampler

SMA11 Stone mastic asphalt with a maximum gravel size of 11 mm

Stormwater Water originating from precipitation, that may infiltrate into the soil, evaporate to the air, or drain from impermeable surfaces and thereafter further transported to stormwater wells, ponds, or nearby streams without any treatment.

Sweepsand Solid material collected by the street sweeper

TBiWP Tire and bitumen wear particles, collective name for tire and bitumen particles <5 µm

Traffic-Derived Microplastics
TWP, polymer modified bitumen, and road marking wear. Microplastic particles generated by the traffic

TRWP Tire and Road Wear Particles

TWP Tire Wear Particles

Washwater Water collected by the street sweeper

WDS Wet Dust Sampler, a sampling equipment to collect road dust samples from the road surface

$ZnCl_2$ Zinc chloride (≤ 1.7 g/cm^3), salt solution used for density separation

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1 INTRODUCTION

Exhaust emissions (combustion from diesel and petrol engines) contribute to bad air quality and have severe impacts on health. The exhaust emissions are decreasing, mainly because of strict regulations connected to the EURO-classes, more effective engines, and the transformation into a more electric vehicle fleet. However, the non-exhaust emissions (NEE) are still increasing. NEE are an important source of environmental pollution and stems from the wear of tires, brakes, road surface, and road markings (Baensch-Baltruschat et al., 2020). Mineral particles derived from road wear, metal particles from brake wear, traffic-derived organic pollutants, and microplastics (MP) i.e., rubber from tire wear, plastics from road markings, polymer-modified bitumen, and particles from brake pads are sources of non-exhaust emissions (Amato et al., 2016; Evangeliou et al., 2020; Thorpe and Harrison, 2008).

The release of MP into the environment has become an area of significant global concern. During the last decades, attention has increased regarding microplastics as an environmental pollutant, and the occurrence of microplastic particles is widespread (Carney Almroth et al., 2018; Hann et al., 2018; Hartmann et al., 2019; Kole et al., 2017; Peeken et al., 2018; Peng et al., 2017). The definition of plastics differs between sources since the plastic material group is extensive and comprises a large number of materials with versatile properties and uses. Polymer materials are often divided into thermoplastics, thermosets, elastomers, and thermoplastic elastomers (Andersson-Sköld et al., 2020). Microplastics are defined as insoluble, solid, and polymer-based particles with an upper size limit of 5 mm (Boucher and Friot, 2017; Frias and Nash, 2019; Verschoor et al., 2016). Recently, a broader definition of the term microplastics have been implemented, which includes all manufactured polymer materials consisting of thermoplastic polymers or thermoset polymers with chemical additives, which therefore also includes rubber (tires), road markings, and polymer modified bitumen. Microplastics have been detected in all environments in urban, rural, and more remote locations. Still, little is known about the fate, transport routes, and distribution of MP in the environment (Akdogan and Guven, 2019). Moreover, there are growing concerns about the negative impact that microplastics may have on ecosystems. Especially with regard to the fact that MP can be carriers of toxic substances that can affect living organisms (Cole et al., 2011; Frias et al., 2016; Lee et al., 2014; Verla et al., 2019).

Traffic has been defined as one of the major sources of the global contamination of microplastics because of the generation of tire wear particles (TWP) (Sommer et al., 2018). TWP are formed during the interaction between tire and road surface, and research indicates that tire wear particles themselves contribute to 5–10 w% of the microplastic emissions entering the global marine environment (Baensch-Baltruschat et al., 2020; Hann et al., 2018; Kole et al., 2017; Lassen et al., 2015; Magnusson et al., 2016; Sommer et al., 2018). During the lifetime of a tire, approximately 10–20 % of the total mass is worn off resulting in an annual global release of 6 000 000 tons (Grigoratos et al., 2018; Kole et al., 2017).

Tire tread consists of 40–60% rubber (elastomers), both natural rubber (polyisoprene) and synthetic rubber (mainly polybutadiene and styrene-butadiene-rubber) together with various fillers (e.g., silicon and/or carbon black for an increased lifespan, rolling resistance and hardness), plasticizers, softening agents, chemicals for vulcanization, anti-aging agents and additional additives (Sommer et al., 2018; Sundt et al., 2014; Wagner et al., 2018). The wear rate of TWP is influenced by many factors such as driving behavior, speed, braking, the characteristics of the tire, the size and weight of the vehicle, the pavement properties and conditions, the vehicle/wheel conditions (e.g., wheel alignment and wheel pressure) and tire

type (studded, studless or summer tires) (Pohrt, 2019). Further, the ingredients in a tire differ between manufacturers, tire types, and models. U.S.EPA (2019) found 355 chemical substances and additives that could be related to recycled tire crumb rubber. When the TWP have been generated on the road surface, the particles are emitted to the air or instantly intermixed with road wear particles, brake wear particles, and minerals resulting in encrustations called tire and road wear particles (TRWP) (Thorpe and Harrison, 2008). Because of this aggregation, pristine TWP are rarely found in the environment (Baensch-Baltruschat et al., 2020; Dall'Osto et al., 2014).

The knowledge about the occurrence, fate, risk, transport routes and toxicity of TWP in the environment is limited. An important reason is that TWP in environmental samples are difficult to analyse (Klöckner et al., 2019; Mattonai et al., 2022; Wagner et al., 2018). The annual emissions are mainly based on models and estimations (Mennekes and Nowack., 2022), and many ecotoxicological studies are based on pure TWP or leachates from tires, most often at concentrations several times higher than environmental concentrations (Halle et al., 2021; Halsband et al., 2020; Knight et al., 2020; Marwood et al., 2011). With that said, more monitoring studies and evaluations of environmental concentrations in different matrices using comparable and trustworthy analytical techniques are needed. Until then, TWP emissions should be handled according to the precautionary principle, meaning that TWP should be seen and handled as an environmental risk until it has been properly evaluated.

1.1 Aim and objectives

Traffic, with tire wear particles (TWP) as the main contributor, has been identified as one of the major sources of microplastic contamination in the environment. This project aims to increase the understanding of traffic-derived particles and investigate the occurrence of TWP in different sample matrices in the environment (e.g., stormwater, air, road dust, and sweeping material). Within the project, street sweeping as a potential measure to reduce the emissions of traffic-derived microplastics was also investigated. The results from the project aim to increase the knowledge about environmental concentrations and thereby be used as a knowledge base for experts and authorities to develop and decide on measures to implement to decrease potential negative impacts of TWP and other non-exhaust emissions from road transport.

To reach the overall aims, the specific objectives of this research were to:

- Identify, characterize, and quantify traffic-derived microplastics and non-exhaust particles from different traffic environments (Paper I, II, and IV)

- Investigate the occurrence of traffic-derived microplastics, metals, and organic pollutants in different matrices from the road environment (Paper II)

- Evaluate the efficiency of street sweeping for the removal of traffic-derived microplastics, metals, and organic pollutants in urban areas (Paper I and II)

- Develop and apply an advanced method based on sieving/filtration, scanning electron microscopy, and machine learning for analysis of traffic-derived particles in environmental samples (Paper III and IV)

- Develop an optimised sample preparation method for the analysis of TWP and TRWP. Increase the knowledge about the characterization and physicochemical properties of TRWP generated in a road simulator (i.e., not affected by traffic and environment) by using a multi-method analytical strategy and thereby investigate what methods that should be suitable for environmental collected TWP (Paper V)

1.2 Thesis outline

The thesis is based on five papers, appended, and numbered as Paper I-V. A summarising illustration of the papers can be seen in Figure 1.

In **Paper I**, the occurrence of traffic-derived microplastics in an urban area was investigated. Samples were analysed with light microscopy (20–300 µm). Street sweeping was evaluated as a potential measure to prevent particles from reaching the stormwater. Finally, calculations were performed to roughly compare the potential theoretical emissions with the actual findings in environmental samples. The theoretical emissions were based on emission factors, traffic intensity, and fixed sizes and densities of the particles.

In **Paper II**, the concentrations of traffic-derived microplastics in an urban area under reconstruction were determined. In addition, metals and organic pollutants were analysed. Samples were collected from stormwater, at the road surface, and in material collected by a street sweeper. The MP samples were analysed with light microscopy (20–300 µm). The research evaluated several parameters at the same time, i.e., MP, metals, and organic pollutants which increased the understanding of the potential toxicity of traffic-derived particles found in an area under reconstruction.

In **Paper III**, a new and advanced analytical method was developed for the analysis of traffic-derived particles. The method is rapid and requires little sample preparation and enables detailed information about particles 2–125 µm. The method is based on a machine-learning algorithm coupled to an automated Scanning Electron Microscopy/Energy Dispersive X-ray spectroscopy (SEM/EDX). The particles were divided into several subclasses: TWP, bitumen wear particles (BiWP), road markings, reflecting glass beads, metallics, minerals, and biogenic/organics. This method development allowed analysis of finer particles from different matrices in the road environment.

In **Paper IV**, the methodology developed in paper III was used to investigate the occurrence of TWP and other traffic-derived particles in different parts of a rural traffic environment. Samples were collected on and in the surrounding area of a highway located in southern Sweden. Samples from the dust load on the road surface (in-between wheeltracks and adjacent to the kerb), runoff (stormwater well and roadside gully pot, both sediment and water phases), and atmospheric deposition were analysed. The results were presented both as relative number concentration (%) and as estimations of absolute concentrations. Paper IV increased the knowledge about sample composition and environmental concentrations in different matrices in the road environment.

Paper V aimed to optimise a method for sample preparation of TRWP, and to compare and evaluate analytical methods (light microscopy, SEM/EDX, pyr-GC/MS, and FTIR). Further, the aim was to increase the understanding for TRWP regarding characteristics (size, shape, density, surface) and physicochemical properties. The polymer composition was compared between road simulator generated TRWP and pristine tire treads. Different tire types from several manufacturers were tested, and the samples were collected in a closed environment (road simulator) to reduce the risk of contamination sources. Two different types of TWP were identified: *sub-elastic* and *firm-elastic* TWP. Further, the characteristics and properties of TRWP collected in a road simulator contain valuable information and can be seen as reference material for TRWP and TWP collected in environmental samples.

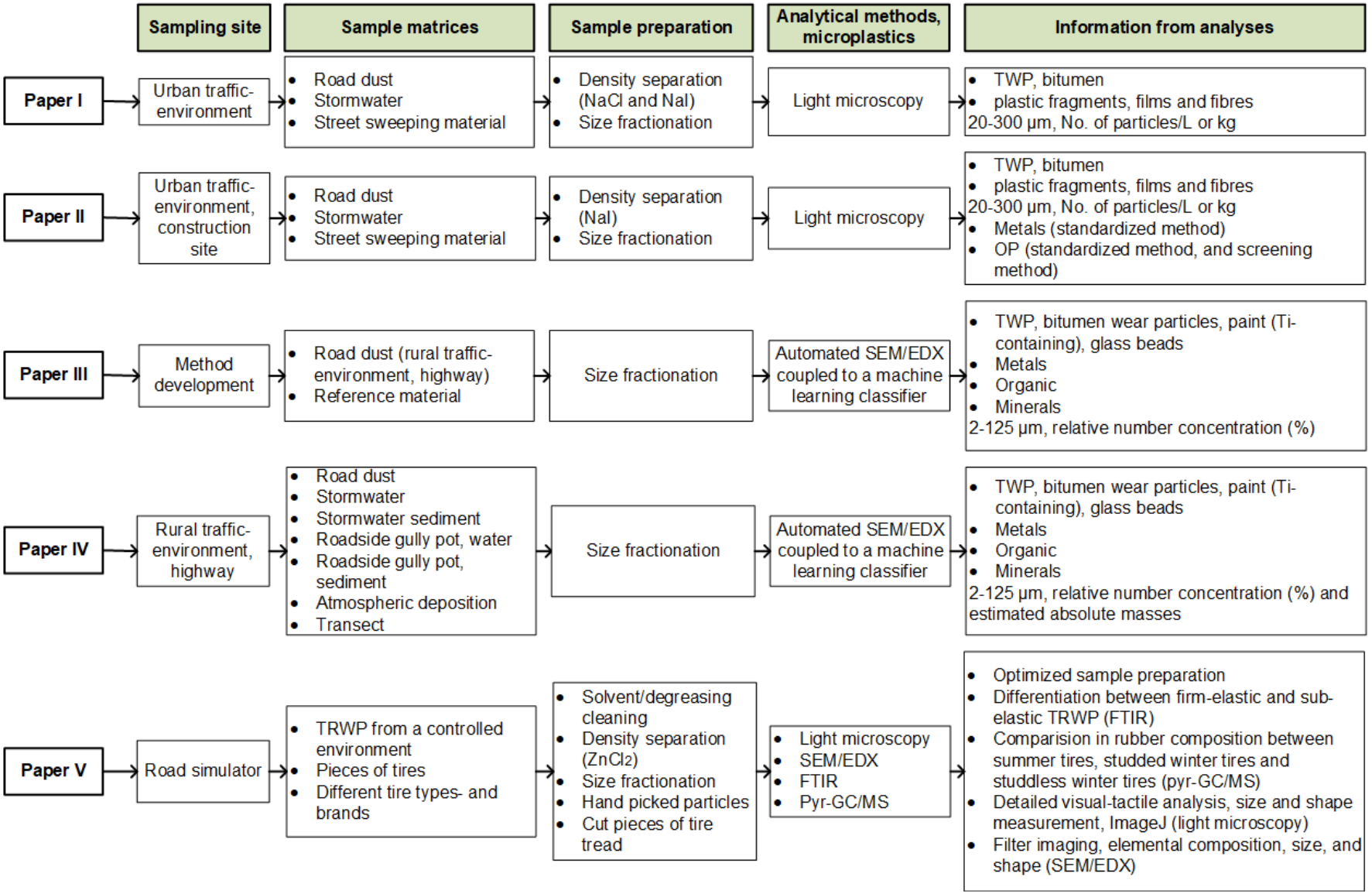

Figure 1. Overview of sampling sites, sample matrices, preparation, and analytical methods used in each paper.

2 THEORETICAL BACKGROUND

2.1 Traffic-derived microplastics and road wear particles

Besides microplastics, other types of exhaust and non-exhaust particles are generated during driving. Exhaust particles (combustion from diesel and petrol engines) are nowadays heavily regulated since exhaust particles are toxic, affect health negatively, and result in bad air quality. Even though the traffic amounts increase (Shepelev et al., 2022), the emissions of exhaust particles are reduced. More effective engines and stricter regulations connected to the EURO classes have resulted in lower exhaust emissions.

Approximately 100 000 tons of pavement wear are generated in Sweden every year (Gustafsson (2003), whereof 8 000 tons are bitumen wear, based on the assumption that asphalt consists of 8% bitumen (Asphalt Institute, 2014). The annual release of TWP in Sweden has been estimated to be about 11 000 tons (Polukarova et al., in prep). The estimations were based on emission factors (EF) and average annual driving distances, and the emissions were divided into heavy trucks, light trucks, buses, cars, and motorcycles. The annual emissions of brake pads were estimated to 600 tons (own calculation based on emission factors from Ntziachristos and Boulter (2016) and driving statistics from Trafa). The annual emissions of road marking paint have been estimated to 500 tons by Magnusson et al (2016), Figure 2. Tire and road wear particles are important from a health perspective since a relatively large fraction is within the inhalable size range (PM10) which can have severe effects on health (Adamiec et al., 2022; Gustafsson et al., 2009; Hesse et al., 2022).

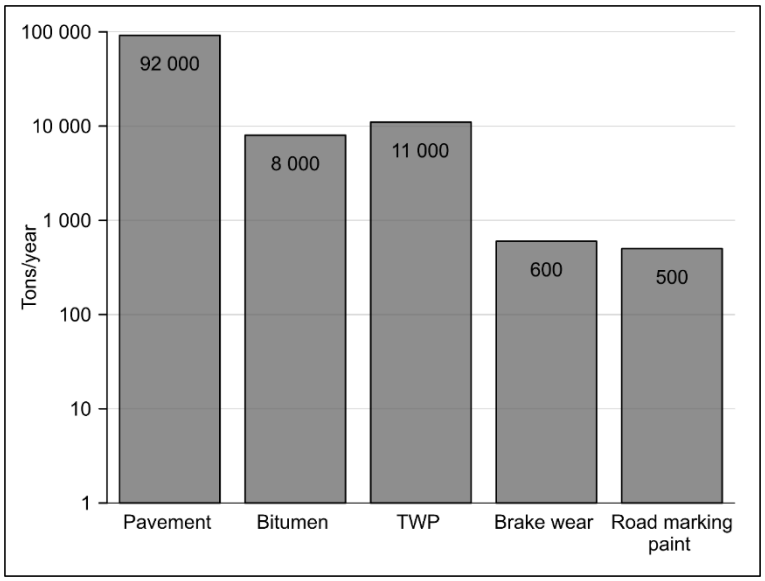

Figure 2. Approximate annual emissions in Sweden. Pavement (Gustafsson (2003)), bitumen (Asphalt Institute (2014), based on the assumption that asphalt consists of 8% bitumen), tires (Polukarova et al., in prep), brake wear particles (emission factors from Ntziachristos and Boulter (2016) and own calculations based on statistics from Trafa, https://www.trafa.se/vagtrafik/fordon/), road marking paint (Magnusson et al., 2016).

Traffic has been defined as one of the major sources of microplastic emissions. A general definition of plastics includes all materials consisting of thermoplastic polymers or thermoset polymers. TWP mainly consists of rubber, which is a polymer defined as an elastomer. It has been debated if TWP should be included in the microplastic definition (Hartmann et al., 2019). However, the majority of the research conducted has considered that TWP should be included in the microplastic definition because TWP contains polymeric rubber materials e.g., (Andrady, 2017; GESAMP, 2016; Verschoor et al., 2016).

Besides TWP, traffic-derived particles also consist of road wear, bitumen wear particles, BiWP, and wear particles from road marking paint. Bitumen is a viscoelastic binding agent used in the top layer of the pavement (4–8 w%) (Asphalt Institute, 2014)). Regular bitumen consists of heterogeneous mixtures of hydrocarbons and is not included in the microplastic definition. However, some bitumen is modified with polymers, polymer modified bitumen (PMB), where approximately 3–10 w% of the regular bitumen has been replaced with PMB. This is especially common in combination with porous pavements (Porto et al., 2019). PMB is used to increase the durability of the pavement Mushtaq et al. (2022), and styrene-butadiene-styrene (SBS) is one of the most used polymers. SBS is a thermoplastic elastomer that has the same elasticity as rubber materials at normal temperature but could melt, and thereby become liquefied at high temperature like plastics.

Tear and wear of road marking paint is also a source of microplastic emissions. Road markings are necessary to secure high safety on the roads and to guide the traffic. Road markings normally consist of glass beads, polymers, pigment, fillers, and additives (Fors and Johansen 2021). Both traffic intensity, tire type, and road maintenance (ploughing) affect the wear (Andersson-Sköld et al., 2020). The wear is higher in countries where studded tires are in use, and it has been shown that ploughing and snow maintenance severely affects the road markings. There are several different types of road marking paint on the market, both the color and content vary between countries and manufacturers. Often, the paint consists of thermoplastics, but solvent-based paints and thermosetting plastics (two-component systems, epoxy or acrylic based) are also used. (Chu, 2019). In Sweden, the annual demand for road markings on national roads is approximately 15,000 tons (roads maintained by municipalities are excluded due to individual contracts and procurements). The annual wear of road markings in Sweden is estimated to be 500 tons (Magnusson et al., 2016) but the numbers are uncertain.

2.1.1 Tire wear particles

Tire wear particles are generated from abrasion of the tire tread when the tire interacts with the pavement during friction. Abrasion is especially high during speed changes (acceleration, braking) and steering e.g., in roundabouts, at traffic lights, or at crossings. TWP differs from other microplastics in shape, color, density, and size. The differentiation depends on both the tire composition and the formation process. Heat is produced because of the friction in the interaction process softening the tread. The soft and sometimes sticky TWP mix and aggregates with road dust consisting of other wear particles e.g., minerals, metals, or sand. Therefore, TWP is rarely found as “pure tire particles”, but more likely as aggregates.

TWP found in environmental samples consists of a higher content of traffic-derived minerals (silica) and metals (aluminum, iron, titanium, or magnesium) than TWP from wear produced in controlled laboratory environments (such as the VTI road simulator) (Mattsson et al., submitted). TWP in field samples are commonly known as elongated black particles, and recent publications have shown that most particles are 20–200 µm (Figure 3) e.g., Kreider et al. (2010) and Sommer et al. (2018). However, other shapes and sizes have been reported, e.g., fragments with a more angular shape (Hassellöv et al., 2018; Kovochich et al., 2021; Rausch et al., 2022). According to reported density values, natural rubber (0.9 g/cm^3) and synthetic rubber (0.9–0.95 g/cm^3) are floating in water (Scinetific Polymer Products Inc., 2019). Pristine TWP has a density slightly higher than water (1.13–1.2g/cm^3) (Degaffe and Turner, 2011; Rhodes et al., 2012; Vogelsang et al., 2019; Wagner et al., 2018) and environmental collected TWP, with incrustations of road wear, has a higher density, (1.7–2.1g/cm^3) (Klöckner et al., 2021; Vogelsang et al., 2019). Further, TWP are often hydrophobic and tends to sorb organic

pollutants, road wear, or other particles which can result in an increased density and different properties (Wagner et al., 2022).

The size and shape of the particles are of importance as they can affect whether the particles sink or remain floating in the water. Also, the environmental context is important for the subsequent fate of the particles. For example, for particles ending up in stormwater, the water flow (speed and remixing conditions) affects how the particles can be expected to spread and where to be found. Small particles tend to settle more slowly than larger particles, and the finest particles (<20 µm) might stay in suspension even if they theoretically should sink (Waldschläger and Schüttrumpf, 2019). The final fate of TWP is dependent on the environment where it ends up, i.e., the opportunities for abilities to degrade as well as its chemical and physical properties. Figure 3 shows TWP particles found within this PhD project (Paper I, III, IV, and V). The particles are both elongated, angular, and irregular. The size varies and particles were found in the size range between 2 µm– several hundred µm.

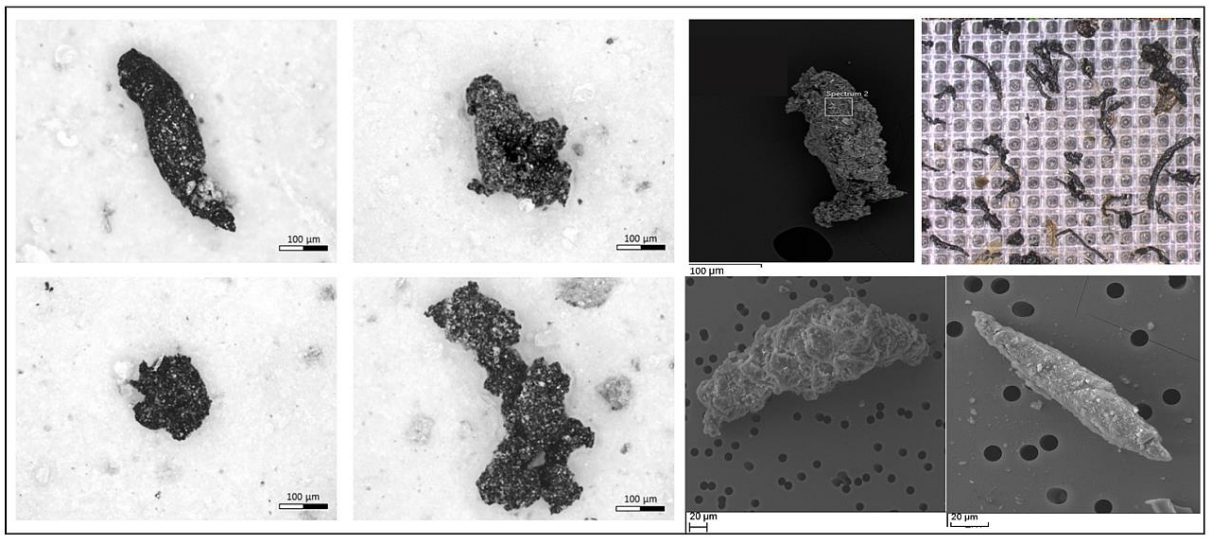

Figure 3. Example photos of tire wear particles. The four pictures to the left are taken with light microscopy and show TWP collected in the road simulator (Photos: Tim Wilkinson, University of Gothenburg, Paper V). Upper picture to the right shows environmental collected TWP 100–300 µm (Photo, Kerstin Magnusson, IVL, Paper I). The bottom pictures to the right are two TWP from the road simulator identified with SEM (Pictures: Tim Wilkinson, University of Gothenburg, Paper V), and the upper picture to the left is an environmental TWP identified with the automated SEM/EDX (Picture: David Jaramillo, Particle Vision, Paper III and IV).

2.1.2 Bitumen and road wear particles

Bitumen is used as a viscoelastic binding agent in the wearing course of the road surface. Conventional bitumen is not classified as an MP since it consists of heterogeneous mixtures of hydrocarbons, including insoluble asphaltenes (Hartmann et al., 2019). However, some bitumen is modified with polymers. PMB is e.g., used in porous pavements, and is common in southern Europe and Norway. PMB is rarely used in wearing courses in Sweden but is used at Testsite E18 (Paper III and IV). Bitumen wear particles are often a part of the encrustations between TWP and minerals in tire and road wear particles (Figure 4) and in the chemical analyses, it has been difficult to distinguish between TWP and bitumen (Jung and Choi, 2022). One solution has been to clean the samples with solvents (e.g., xylene) prior the analysis or to perform a melting test on all particles (i.e., bitumen melts, TWP remains unaltered). Pavements with the inclusion of recycled tires (ca 18–20 w%) in the binder have been tested in the United States and seem to reduce aging and resist fatigue and cracking from both traffic and variations in temperature better than conventional asphalt-concrete e.g., (Hung et al., 2017). However, the release of tire wear particles is not taken into consideration, and rubberized asphalt has not been

used in Sweden yet. Therefore, this will not be further discussed within the thesis. Minerals are the major particle class in road wear. Minerals are thereby an important source of non-exhaust emissions, especially in countries where studded tires are in use (Kupiainen et al., 2016). Mineral particles tend to be grainy, flaky, or shard-like (Gustafsson et al., 2008).

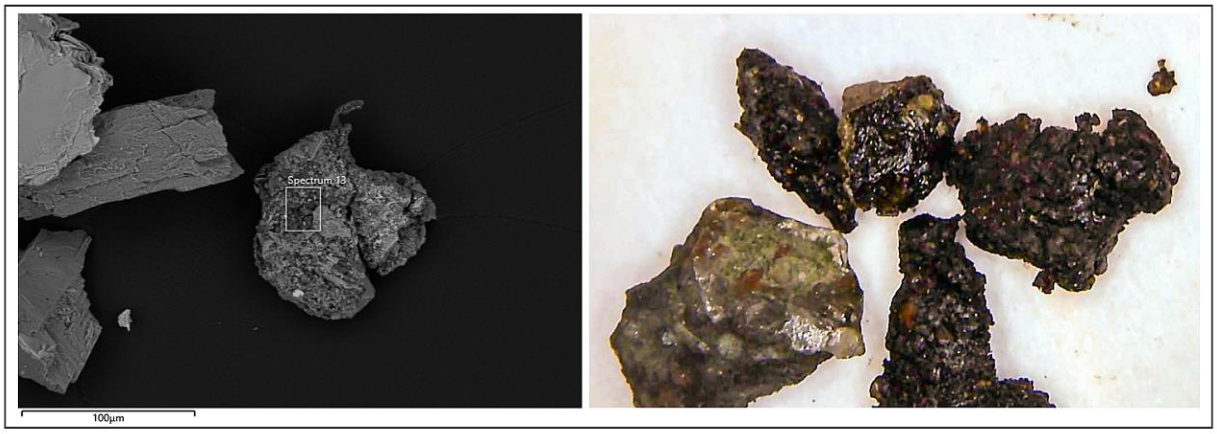

Figure 4. Left: bitumen particles analysed with SEM/EDX (David Jaramillo, Particle Vision, Paper III). Right: encrustations with minerals and bitumen. Photo was taken with a portable microscope.

2.1.3 Road markings

Road markings consist of reflective glass beads, fillers, pigments, and polymers. The binding agents in thermoplastic road markings usually consist of ethylene-vinyl acetate (EVA). EVA makes up 1–5% of the total road marking and is added to improve wear resistance and prevent crack formation. Studded tires, snow maintenance, and variations in the climate can affect the wear rate of road markings (Andersson-Sköld et al., 2020). In white road markings, as is the most common type in Sweden, titanium dioxide is used as a pigment. In yellow road markings, crocoite ($PbCrO_4$) is still used globally (O'Shea et al., 2021), but lead chromate pigments are prohibited in the Nordic countries and replaced with other pigments (Fors and Johansen 2021). Glass beads are included in both the paint and on the upper layer, after painting (Figure 5).

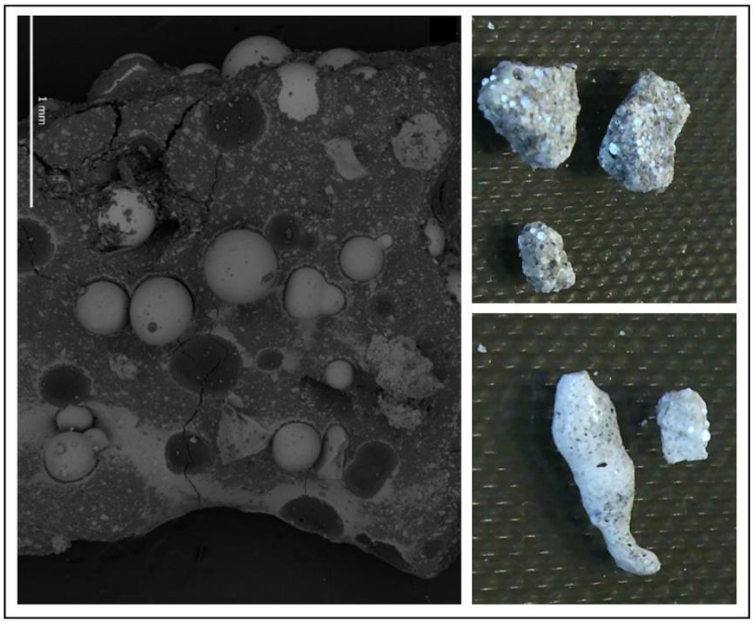

Figure 5. Photos of road marking particles collected in the environment. Left: SEM/EDX picture taken by David Jaramillo, Particle Vision, Paper III. Right: Photos taken with a portable microscope.

2.2 Occurrence, fate, and transport of traffic-derived microplastics

The main transport routes and dispersal pathways of traffic-derived microplastics are presented in Figure 6 (figure simplified from Paper IV). The particles are generated at the road surface through the interaction between the tire and the pavement. The particles are either transported into the environment through direct emissions to the air or to the road surface, and thereafter further transported through, runoff, or splash and spray. The particles accumulated on the road surface are thereafter available for suspension (e.g., resuspension and wind). The transport mechanisms are dependent on e.g., the traffic intensity, speed, characteristics of the road, and the surrounding environment. Previous research has assumed that the major part of TWP >10 µm (45–76% of the total emissions) deposit close to the road edge (Baensch-Baltruschat et al., 2021; Hann et al., 2018; Wagner et al., 2018), that 15–50% are further transported through runoff (Sieber et al., 2020; Unice et al., 2013; Unice et al., 2019; Verschoor et al., 2016), that 0.1–13% of the TWP are directly transported into the environment via atmospheric deposition (Panko et al., 2018) and that only a few percent reach the aquatic environment (Kole et al., 2017; Unice et al., 2019; Wagner et al., 2022). Moreover, some studies assume that the runoff is treated, e.g., in wastewater treatment plants or sedimentation ponds prior to release into the aquatic environment (Baensch-Baltruschat et al., 2021). In both the rural and urban traffic environments in Sweden, only a few percent of the stormwater is treated and the treatment systems available are not designed to specifically remove TWP. There are still large knowledge gaps regarding mass flows, dispersal routes, and total TWP emissions (Hüffer et al., 2018; Sieber et al., 2020), and the existing estimations are uncertain.

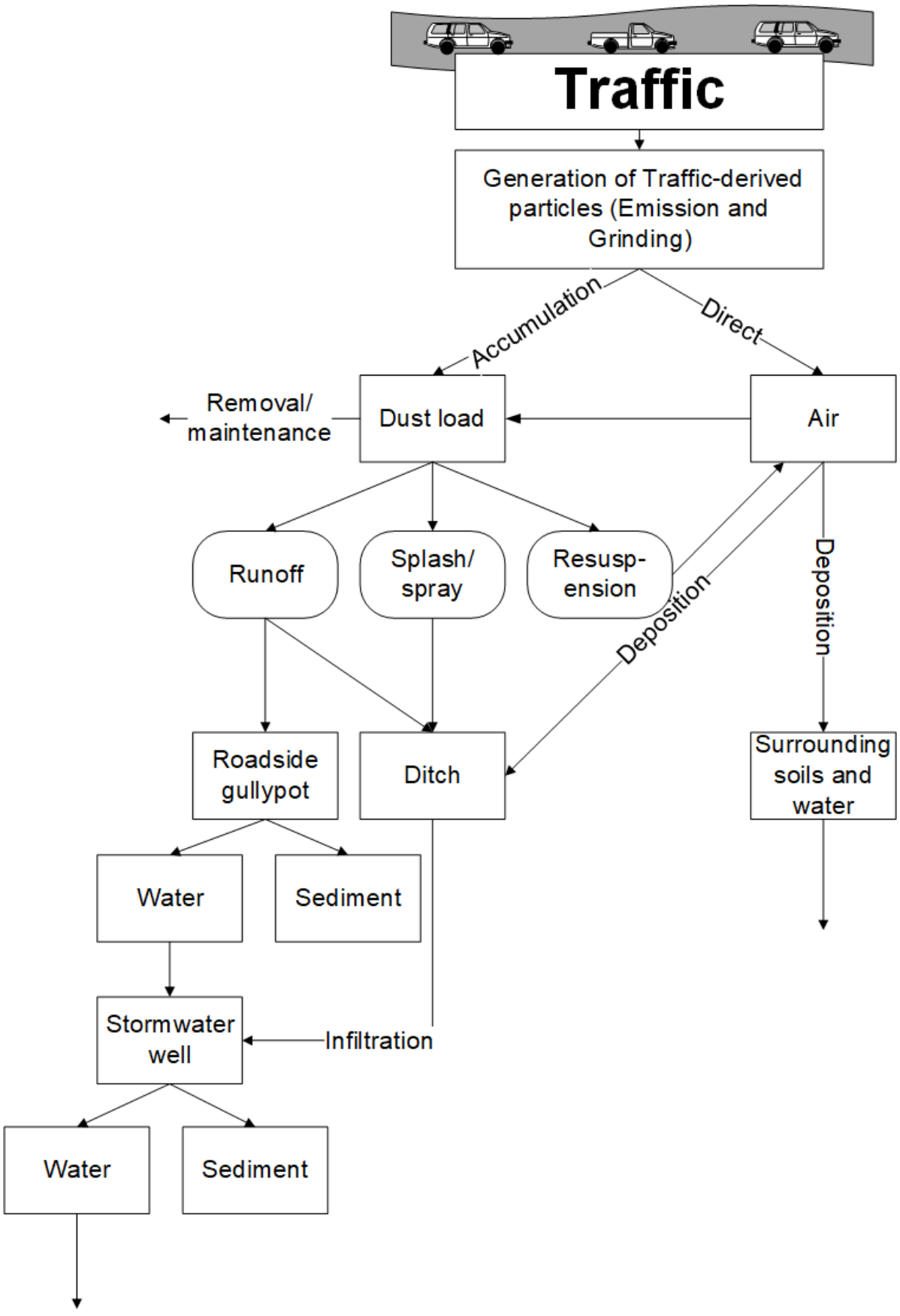

Figure 6. Simplified illustration of transport pathways of traffic-derived particles (Modified from Paper IV).

2.3 Organic pollutants

Several organic pollutants (OP) are regulated in environmental quality guidelines for the protection of the environment, both on global scales as quality standards e.g., the EU Water Framework Directive, the convention on long-range air pollution, and American Clean Water Act (EC, 2000; EC, 2008; UN-ECE, 1998; US.EPA, 2002), but also on local levels as national guidelines e.g., (Miljöförvaltningen, 2013; Naturvårdsverket, 2009; Naturvårdsverket, 2017). Organic pollutants are often persistent and degrade slowly in the environment. Further, many OP are non-polar with high lipophilicity and low solubility in water, meaning that organisms that are exposed to the OP easily accumulate the OP in the fatty tissues (Jones and de Voogt, 1999). Moreover, if an organism is exposed to high concentrations of OP, it is a risk that the concentrations of OP will be higher within the organism than in the surrounding environment due to bioaccumulation, and may therefore biomagnify, i.e., increase in concentrations in fatty tissues in organisms higher up in the food chain. Many OP comes with toxic properties and can for example cause cancer, and disrupt the hormone systems (Alharbi et al., 2018).

Emissions of organic pollutants into the environment occur from industrial production, wood burning, traffic, combustions, and other human-related activities, but also through leaching from landfills and contaminated sites and sediments. The Swedish Environmental Protection Agency states that benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, and indeno(1,2,3-cd)pyrene are the four substances that are among the most hazardous to the health of the most common analysed OP. Tires contain a large number of additives, and the composition of a tire is complex. Some of these additives are confirmed to be toxic, such as highly aromatic oils (HA-oils) that were used as a softener to improve the processability. Since 2010, the concentrations of HA-oils in tires are restricted by European legislation, which may result in reduced emissions. However, tires containing HA-oils can still be present on the roads, both due to the use of old tires, rubber recycling, international transports, and due to a delay in replacement (ETRMA, 2010; ETRMA, 2011). High concentrations of polycyclic aromatic hydrocarbons (PAH), aliphatics, and phthalates are commonly found in road dust and road runoff, and traffic is a major source of emissions of these pollutants (Björklund et al., 2009; Markiewicz et al., 2017; Polukarova et al., 2020), both as exhausts (combustion), and non-exhaust (Alves et al., 2020; Hwang et al., 2019). Calculations of emission factors from traffic and roads showed the following order of amounts of PAH emitted: phenanthrene > fluoranthene > naphthalene > pyrene, and the main sources were exhaust gases $\gg$ tires > motor oil leakage > road surface wear $\approx$ brake linings (Markiewicz et al., 2017).

2.4 Metals

Metallic particles are commonly found in runoff (McKenzie et al., 2009). The occurrence of copper (Cu) and zinc (Zn) have been frequently investigated and toxic concentrations far above both environmental quality guidelines and national guidelines have often been reported in urban stormwater, see Müller et al. (2020) and references therein. Road dust has often been used as an indicator of the pollution of heavy metals in the environment (Duong and Lee, 2011). TWP and brake wear particles are two of the most important non-exhaust sources of metal pollution (Adachi and Tainosho, 2004; Davis et al., 2001). A typical tire contains approximately 1.5 w% of Zn, and Zn is often used as a tracer for TWP in environmental samples. However, Zn is ubiquitous in other parts of the traffic environment e.g., in crash barriers and road furniture made of galvanized steel. Another considerable source of metal contamination is brake wear emissions. Brake pads consist of approximately 50% metals whereof iron (Fe), Cu, Zn, and tin (Sn) are the most abundant (Hulskotte et al., 2014). Previously, both lead (Pb) and asbestos were common ingredients in brake pads, but that has been replaced in modern brake pads (Grigoratos and Martini, 2014). Combustion of biomass is also an important source of Zn

emissions into the environment. Copper can be both traffic-derived (tires and brake pads) and occur as leachates from copper roofs. In the Nordic countries, where studded tires are in use, it is not unusual to find traces of tungsten carbide (WC) and cobalt (Co) in road dust and runoff. These metals can be related to the studs (Furberg et al., 2018). Metals are transported through the environment via wind or through soil and water as dissolved ions or attached to other particles.

2.5 Analytical challenges

Analysis of environmental collected TWP and TRWP is known to be difficult. The main reasons are the black color and the complex compositions of the particles (Kole et al., 2017). Wagner et al. (2022) divide the analysis of TWP into two main groups: analysis of bulk samples (e.g., destructive analytical methods such as pyr-GC/MS) and single particle analysis (light microscopy, SEM/EDX, FTIR). There is a lack of standardized methods and generalized protocols for sample preparation, Thomas et al. (2022) and one of the major concerns regarding the analysis of TWP and TRWP is that there is no universal method available, and the existing methods provide different information (Rauert et al., 2021). Another challenge for analytical method development and validation has been the difficulty of obtaining quantities of laboratory-generated tire wear particles with properties similar to real-life TWP (Mennekes and Nowack, 2022). As a result of the many analytical difficulties, several previously published microplastics studies have explicitly stated that TRWP was excluded from their analyses e.g., Liu et al. (2019), while differences in sampling techniques, sample preparations, and analytical methods, have complicated the comparability of studies (Knight et al., 2020).

Several methods have been used for identification of TWP and TRWP, light microscopy presents the number of particles within a certain size range (often) >20 µm. Scanning Electron Microscopy/Energy Dispersive X-ray spectroscopy (SEM/EDX) also presents the number of particles, and in addition, information about size, shape, surface, and elemental composition. An advantage of SEM/EDX is that even finer particles can be analysed (>2 µm). Fourier transform infrared spectroscopy (FTIR) determines the chemical fingerprint within a particle and detects what organic and inorganic compounds a particle contains. However, FTIR utilizes infrared light to scan a sample. The absorption, reflection, and emission of light are then registered. The amount of absorbed light at different wavelengths is specific for each compound (Löder and Gerdts, 2015). FTIR has not been suitable for analysis of TWP since dark particles tend to absorb the light resulting in insufficient quality of the signals (Eisentraut et al., 2018; Kovochich et al., 2021; Käppler et al., 2016; Mattonai et al., 2022). Thermal methods, (Pyrolysis- Gas Chromatography-Mass Spectrometry, pyr-GC/MS and Thermal Extraction Desorption- Gas Chromatography-Mass Spectrometry, TED-GC/MS) are commonly used for the identification of TWP and TRWP in environmental samples (Löder and Gerdts, 2015; Unice et al., 2013). During a pyr-GC/MS analysis, the sample is decomposed by heating under an inert atmosphere. The molecules within the sample are separated and detected in the MS. Pyr-GC/MS and TED-GC/MS only provide information about the concentration within a sample, the sample is also destructed during analysis.

2.5.1 Chemical markers

Zinc has been a common marker for TWP in environmental samples, e.g., (Halsband et al., 2020; Klöckner et al., 2019), but marker-based methods also have limitations. Zinc is also naturally occurring in the environment and can be emitted from several other traffic and non-traffic related sources (e.g., de-icing salts, road barriers, galvanized metal in cars, brake wear, and road markings) (Unice et al., 2013; Wagner et al., 2018). In studies by Halle et al. (2021) and Halsband et al. (2020), it was found that pristine TWP tends to have higher concentrations

of Zn compared to environmental collected TWP, indicating that Zn might leach from the tires into the environment (Councell et al., 2004). Several organic compounds, e.g., benzothiazoles (e.g., 2-(4-morpholinyl) benzothiazole (24MoBT) and N-cyclohexyl-2-benzothiazolamine (NCBA)), hexa(methoxymethyl) melamine (HMMM) (Johannessen et al., 2022; Kumata et al., 2000; Ni et al., 2008; Seiwert et al., 2020) and N-(1,3-Dimethylbutyl)-N'-phenyl-p-phenylenediamine-quinone (6PPD quinone) (Tian et al., 2021) have been used as markers for TWP. NCBA and 24MoBT are commonly used in the tire tread and can be biologically transformed under aerobic circumstances (Kumata et al., 2002). All tire manufacturers have their own formulas and the composition and concentrations of fillers, vulcanizers and additives vary between tire types. Further, all markers have drawbacks. Some compounds are volatile or degrade fast in the environment while others have additional origins making the source hard to determine. TWP can also attract/sorb organic pollutants resulting in higher concentrations of the specific markers, or specific compounds can be washed out during the weathering process. In summary, both TWP and TRWP consist of complex mixtures of chemical compounds, and it is difficult to distinguish whether a marker compound (e.g., Zn) originates from the TWP in environmentally collected samples.

2.5.2 Recent method developments

The development of advanced and rapid methods for the analysis of TWP is in progress, and several articles have published novel methods for the analysis of TWP e.g., SEM/EDX mapping in combination with time-of-flight secondary ion mass spectrometry (ToF-SIMS) mapping (Kovochich et al., 2021), simultaneous thermal analysis (STA) in combination with FTIR and parallel factor analysis (PARAFAC) (Mengistu et al., 2021), automated single-particle SEM/EDX in combination with machine learning (Rausch et al., 2022), pyr-GC/MS with multiple marker compounds (Rødland et al., 2022b), pyr-GC/MS enabling differentiation between car tires and truck tires (Goßmann et al., 2021), and TED-GC/MS (Müller et al., 2022). Further, it has been shown that FTIR can be used to get a decent spectrum for individual TWP particles and Aboelkheir et al. (2019) used FTIR as a complementary method to get information about the chemical groups in tire crumbs. Attenuated total reflectance (ATR)-FTIR has also shown promising results for the identification of larger TWP (>500 µm) (Manohar et al., 2017). However, for ATR-FTIR analysis, every single TWP must be isolated onto a glass slide, which complicates the analysis of finer particles, or samples with high concentrations of TWP.

3 METHODS AND MATERIALS

Most of the methodological aspects have been described in detail in Paper I–V. Therefore, this chapter presents and discusses the general aspects.

3.1 Manually and laboratory generated TWP

TWP was generated in three different ways:

- Manually created through scraping and rasping with a file
- Pieces were cut out from the tire tread with a steel knife
- The VTI road simulator was used to generate TRWP under controlled circumstances. The aim was to generate particles with similar properties as environmental collected TWP but without environmental contamination

3.1.1 Road simulator

The VTI road simulator consists of four wheels that run along a circular track with a diameter of 5.3 m diameter, Figure 7. The speed can be varied up to 70 km/h, and each wheel is driven by a separate motor. An eccentric movement of the vertical axis is used to slowly shift the tires from side to side over the full width of the track. Any type of pavement can be applied to the simulator track and all types of passenger car tires can be mounted on the axles. For paper V, the asphalt in the road simulator was a mixed stone mastic asphalt with granite and quartzite as the main aggregates, typical Nordic pavements. For comparison, and to avoid the influence of bitumen, a concrete pavement was used for additional tests. Moreover, the road simulator enables the possibility to collect samples from individual tires and specific manufacturers. For Paper V, three types of tires (summer tires, studded winter tires, and studless winter tires (soft and siped Nordic type) from four different manufacturers were tested. Each set of tires was driven on the pavement at 50-60 km/h for five hours. To be able to collect TWP in real-time, during a test, a sampling hood made of aluminum was mounted over one of the wheels (see Paper V). The generated particles were collected in a deposition box behind the tire.

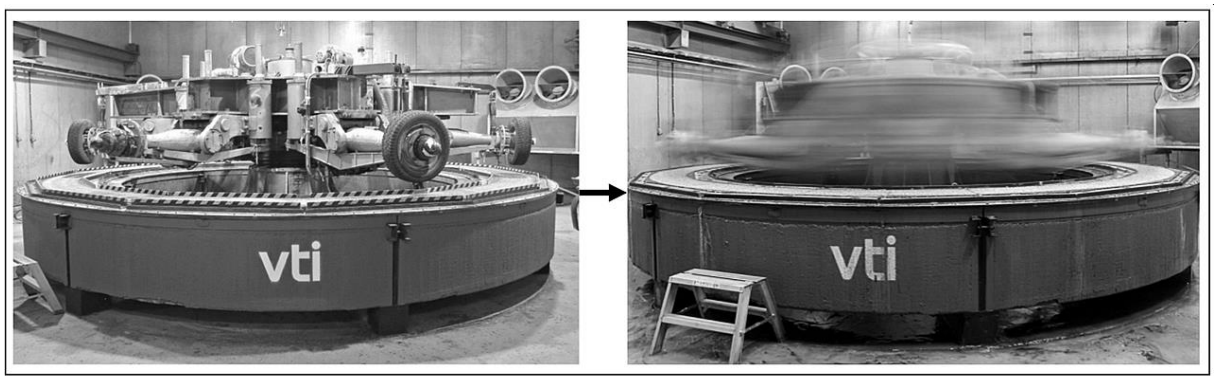

Figure 7. The VTI road simulator. Photos: Mats Gustafsson, VTI.

3.2 Sampling sites

Samples have been collected in several sampling matrices in different types of traffic environments during the period between 2017–2020.

3.2.1 Vitsippsbäcken, urban area

The sampling site was located in the outer central parts of Gothenburg, the second-largest city in Sweden (1 000 000 inhabitants). Gothenburg is located at the mouth of the river Göta älv, which feeds into the Kattegat, an arm of the North Sea (Figure 8). An area close to Sahlgrenska

hospital was the case study area for investigating the potential effects of frequent street sweeping and the concentrations of microplastics on the road surface, in the stormwater, and in the material collected by the street sweeper. The catchment area covers approximately 55 ha whereof 60% is pervious areas, 27% is paved surfaces e.g., parking lots, roads, and other paved areas) and 13% are buildings. On the streets within the catchment area, the annual average daily traffic (AADT) varies between 5 500–13 000 vehicles. The stormwater sewer system in the studied area is separated from the wastewater pipes, meaning that all runoff from impervious surfaces in the area drains, without treatment, into a small creek, Vitsippsbäcken. The creek is classified as a *sensitive water recipient* with a high pollution load of Cu, Zn, Cr, and Co, due to traffic, and corroded metal roofs in the catchment. It discharges through a natural reserve and the Botanical Gardens of Gothenburg, into the river Göta älv. Meteorological information was collected and downloaded from the SMHI open-source database (weather monitoring station 57.6996 N, 11.9673E). The data were compared with the data from the weather station placed adjacent to the stormwater measurement station.

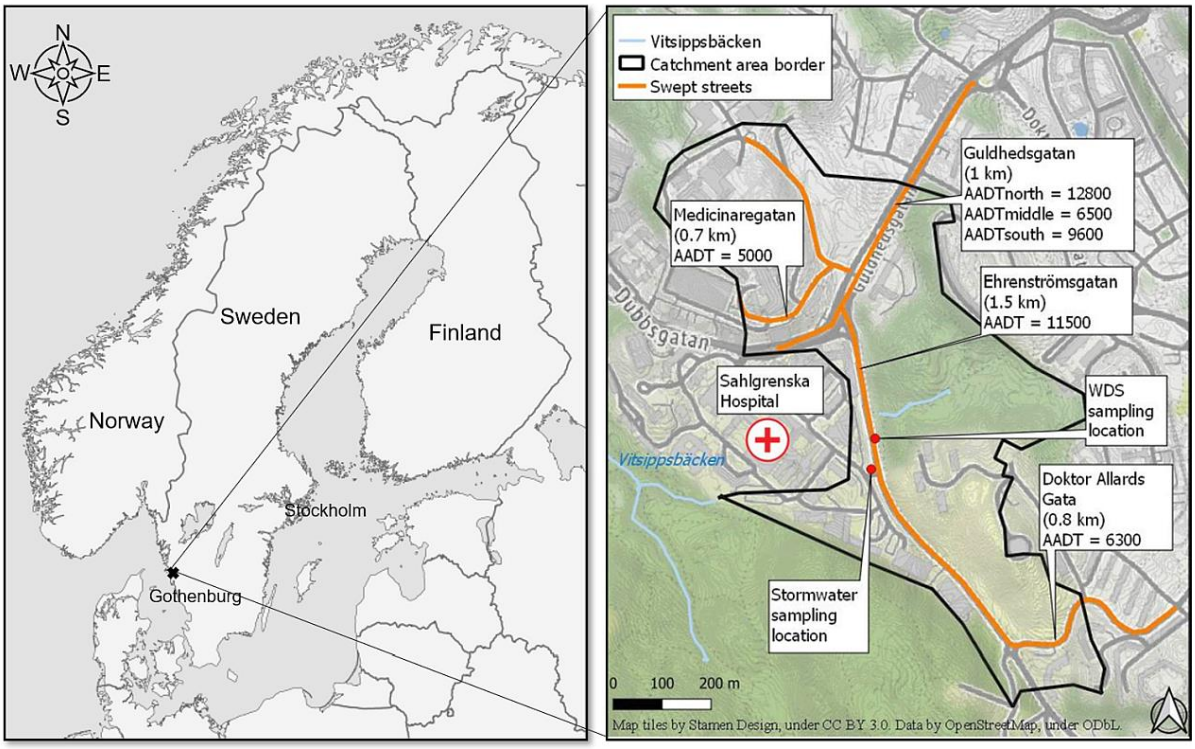

Figure 8. Sampling site, Vitsippsbäcken. Maps to the left were collected from Lantmäteriet. Map to the right made in Stamen Design under CC BY 3.0 Data by OpenStreetMap.

3.2.2 Gullbergsvass, urban area under reconstruction

The sampling site chosen for Paper II was a reconstruction area located in the central parts of Gothenburg, Sweden (close to the central station). The catchment area of Gullbergsvass is 9 ha and consists of 52% paved surfaces (roads and parking lots), 44% roofs of commercial buildings, and 4% permeable areas. It is located next to the river Göta älv, which is classified as a *less sensitive water recipient*, and the runoff within the catchment area is drained via separate storm sewers and discharged untreated into Göta älv which nearby feeds into Kattegat in the North Sea (Figure 9). The AADT in the catchment area is approximately 5 500 vehicles. However, during the measurement period, several large reconstructions took place within the area. The constructions included both renovations and new infrastructure constructions such as the construction of a new bridge. Each construction site has access routes to the surrounding road network, at which an appreciable number of trucks enter and leave daily. The truck traffic

through the study area varied considerably from week to week, but approximately 500 trucks/week passed through the construction site. During three weeks of the measurement period (only nighttime), one driving lane from E45, the major throughway, was redirected through Gullbergs strandgata resulting in an increased AADT during those weeks. Meteorological information was collected from the SMHI monitoring station (57.7156N, 11.9924E) and downloaded from the open-source database. The data from the monitoring station was compared with the data from the weather stations placed in connection to the stormwater sampling location.

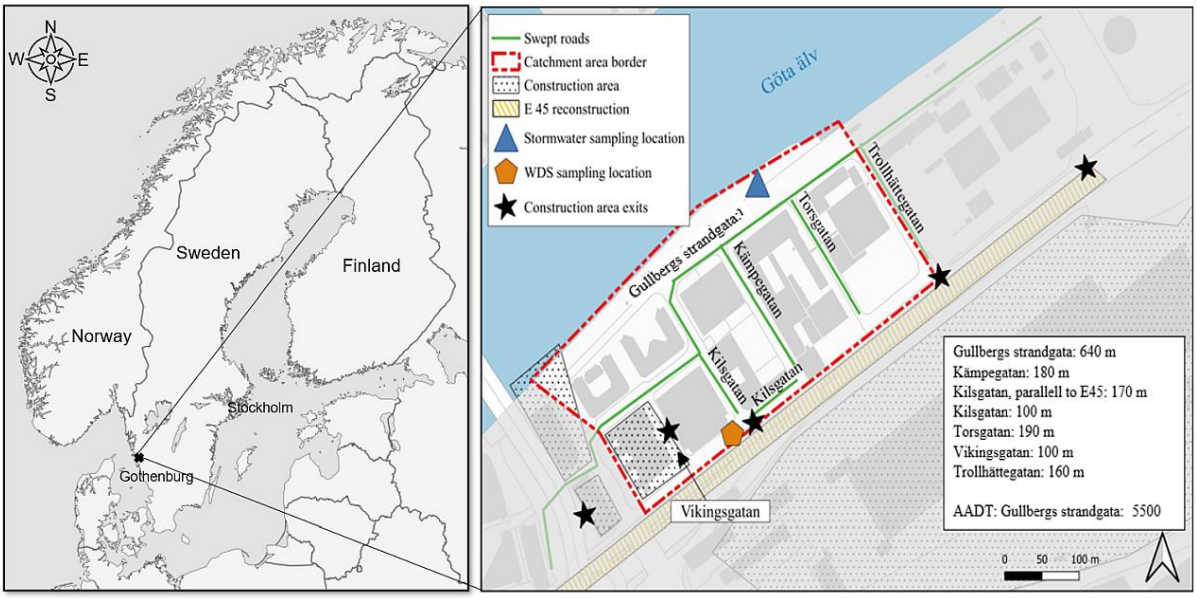

Figure 9. Sampling site, Gullbergsvass. Maps to the left were collected from Lantmäteriet. Map to the right made in QGIS.

3.2.3 Testsite E18, rural highway

For Paper III and IV, samples were collected at Testsite E18 (59.63N, 16.86E), a national research station located on the highway between Västerås and Enköping, Sweden (Figure 10). The annual average daily traffic (AADT) is approximately 11 000 vehicles, and the speed limit is 120 km/h. During the sampling period (2018–2020), the pavement at Testsite E18 consisted of a stone mastic asphalt with a maximum gravel size of 11 mm (SMA11) and the binder was a polymer-modified bitumen. The road is surrounded by fields, and the area is flat. Testsite E18 is a large-scale testsite with the purpose to have easy access to monitor different aspects of the highway (e.g., contamination of the runoff, meteorology, salt amounts, particle loads, pH, and turbidity of the stormwater). Testsite E18 is a collaboration between the Swedish Road Administration, the Swedish National Road and Transport Research Institute (VTI), the Royal Institute of Technology (KTH), and the Swedish Meteorological and Hydrological Institute (SMHI). Meteorological data (precipitation, temperature, wind speed, and wind direction were collected from the SMHI's open-source database, the monitoring station is located in Enköping (59.65N, 17.12E). All road runoff at Testsite E18 (both splash and spray after infiltration of the ditches and water from the roadside gully pots) are transported into the same stormwater well. Thereafter, the water is released into a ditch and further transported to a small creek, Sagån, without any treatment.

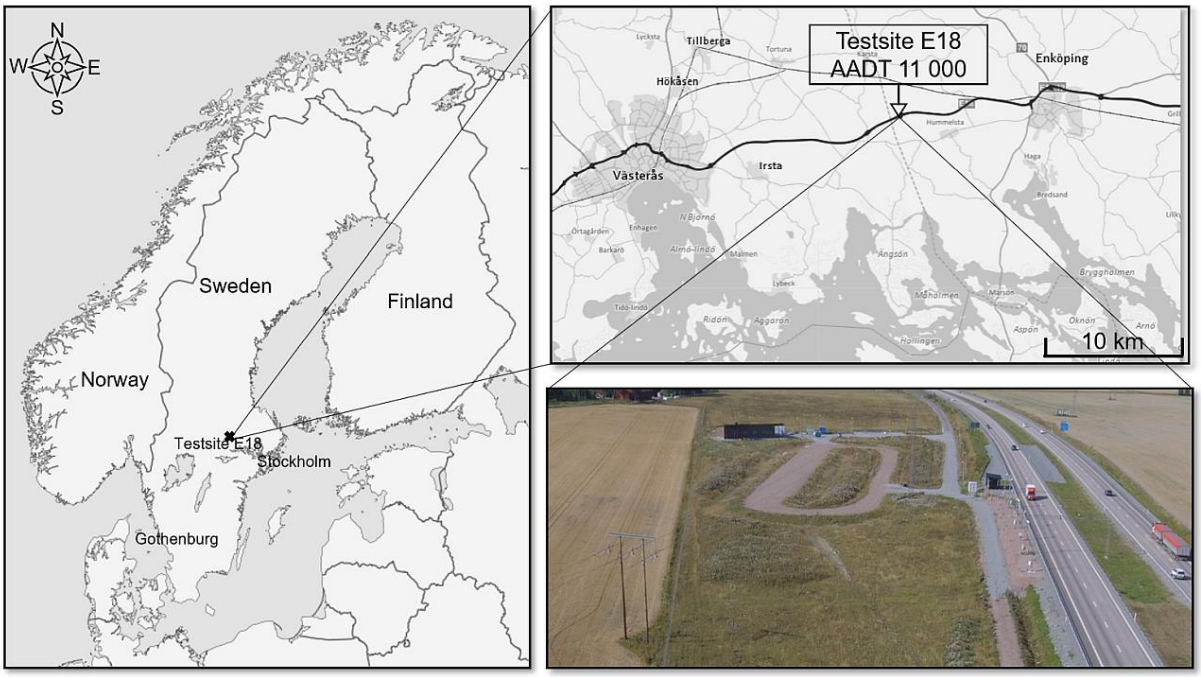

Figure 10. Sampling site, Testsite E18. Maps collected from Lantmäteriet. Photo Tobias Ulegård, Swedish Transport Administration.

3.3 Environmental samples

3.3.1 Road dust samples

Previous research has shown that road dust tends to accumulate adjacent to the kerb. In the driving lanes, more road dust is found in-between the wheeltracks than in the wheeltracks (Gustafsson et al., 2019). For Paper I, samples were collected in the left wheeltrack, in-between wheeltracks, and adjacent to the kerb. However, the dust load was very low in the wheeltrack, and it was decided to not collect samples in the wheeltracks for the remaining studies (Paper II–IV) due to the risk of too low concentration (i.e., too little material to detect traffic-derived microplastics). Therefore, all road dust samples for Paper I–IV were collected adjacent to the kerb and in-between wheeltracks (Figure 11). The kerb samples represent a "worst-case scenario", and the in-between wheeltracks samples show a more general concentration representative for the entire driving lane. Samples were collected from the road surface with a Wet Dust Sampler (WDS II) (Gustafsson et al., 2019; Jonsson et al., 2008; Lundberg et al., 2019). WDS II is a piece of sampling equipment developed at VTI, it is a wet sampling technique enabling collection of even the finest particles, both in dry and wet conditions. An advantage with the WDS II compared to vacuuming, sweeping, or shoveling, is that WDS II can collect samples even if the road surface is wet. Further, since the WDS II washes the road surface with high pressurized water, even particles that have sedimented in the pores of the pavement can be collected.

In an automated sampling procedure, WDS II cleans a known area of the road surface (20.4 cm^2) with a known amount of high-pressurized de-ionized water (340 mL). Compressed air is used to transfer the road dust-water-mixture from the sampling chamber into a glass bottle. Each sample bottle contains a composite sample where three WDS II samples have been pooled together (approximately 1 L), and the WDS II was moved slightly forward into the driving direction between each sample to secure that the sample was taken on a previous unsampled portion of the road surface (Figure 11). At least two sample bottles were collected from each position.

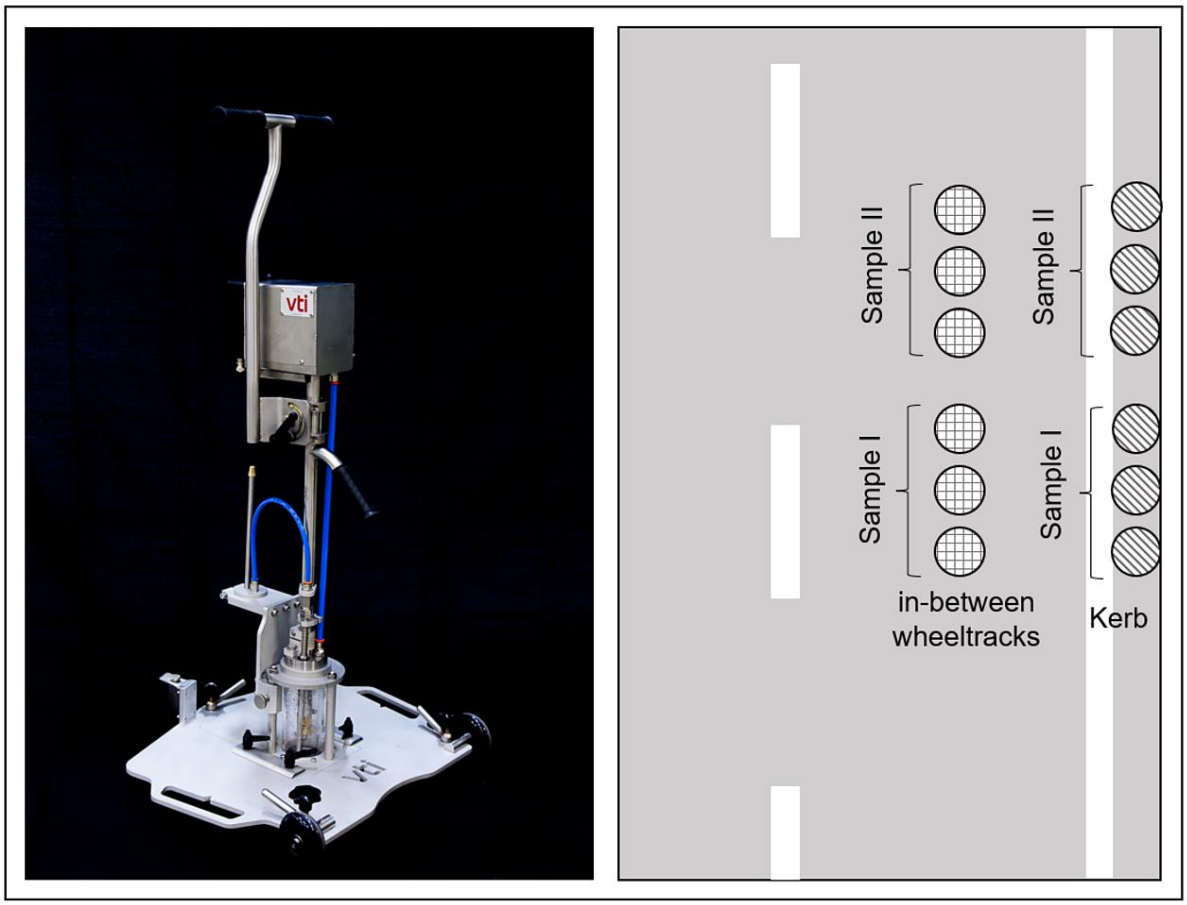

Figure 11. Wet dust sampler. Photo: Mats Gustafsson, VTI (left). Sampling set-up (right).

3.3.2 Stormwater samples

For paper I and II, stormwater samples were collected in a stormwater well with a flow-weighted automated sampler (ISCO 6712 with an area velocity flow meter, Triton+). The samplers were programmed to start to collect samples when a certain water level or water flow was achieved. The water flow depends on the precipitation and the trigger points that were chosen for Paper I and II were a minimum water depth of 75 mm and at least 5 mm of precipitation. One representative composite sample was collected during each rain event (>5 mm). To monitor the precipitation, a tipping bucket rain gauge (type MJK) was installed on a rooftop close to the sampling point. The samples can be seen as event mean concentrations (EMC).

The stormwater samples collected for Paper IV were grab sampled into 1L glass bottles with a telescopic surface water sampler (similar to the NASCO swing sampler but built at the VTI workshop). Samples were collected both in the roadside gully pot and in a stormwater well at Testsite E18, that enabled analysis of only road runoff compared to runoff in combination with splash and spray after infiltration in ditches. Further, it was possible to investigate eventual differences in particle size distribution and MP concentrations between the two wells. In addition, sediment was sampled both in the roadside gully pot and in the stormwater well with a tube sampler (Hydro-Bios, Sediment Corer) and stored in 1L glass jars.

3.3.3 Air samples

Boron substrates placed in passive air samplers, Sigma-2s (following the norm VDI2119:2013), were used to collect and measure the atmospheric deposition in the $PM_{1\text{-}80}$ µm fraction (Figure 12, Paper IV). Samplers were placed at Testsite E18, 3.1m south of the road edge at a height of 2 m above the ground level. The samplers collected particles for 14–30 days. The meteorology was monitored 30 days prior to and during the measurements. To investigate the atmospheric transport of particles, Sigma-2s were placed at additional 3 distances from the road edge (4.8, 27.1, and 100 m).

Figure 12. Sigma 2 sampler at Testsite E18. Photo: Mats Gustafsson, VTI and David Jaramillo, Particle Vision.

3.3.4 Samples from a street sweeper

In Paper I and II, a vacuum sweeper with metal brushes from Johnston Beam A/S (model S9000, VTJB61) was used for weekly sweeping. Sweeping was performed during the daytime, when the streets were not closed for traffic, resulting in slight variations between the sweeping occasions. The sweeping time and distance were recorded with Strava, a GPS-based smartphone app. The average driving speed varied between 4.7–10 km/h. Nozzles under the sweeper spray water on the road surface to prevent fine dust to emit into the air, and the amount of water varied depending on the weather conditions. Due to the use of water, no sweeping was performed when the temperatures were below zero, or during the winter period. Rotating brushes collect the road dust, and the material is transferred into the sweeper with vacuum suction nozzles.

The sweeping material was dumped on a temporal deposition site after each sweeping event. The liquid phase, herein called washwater, was systematically collected in 10L glass bottles with a hose that was connected directly to the sweeper. The solid material, called sweepsand, was manually collected once the masses had been emptied onto the ground. Sweepsand samples were taken randomly from the pile using a metal shovel. The samples were collected in 10L stainless steel buckets. To estimate the collected material, and to relate that to driving distance and speed, the sand pile, and the amount of washwater was measured after each sweeping occasion. Both the washwater and sweepsand samples should be seen as representative composite samples from the whole catchment area.

3.4 Experimental work

This chapter shortly describes the methods used for sample preparation and the analytical methods of traffic-derived microplastics tested throughout the project (Figure 13). For a more extensive description of the methods applied, see the paper referred to in the text.

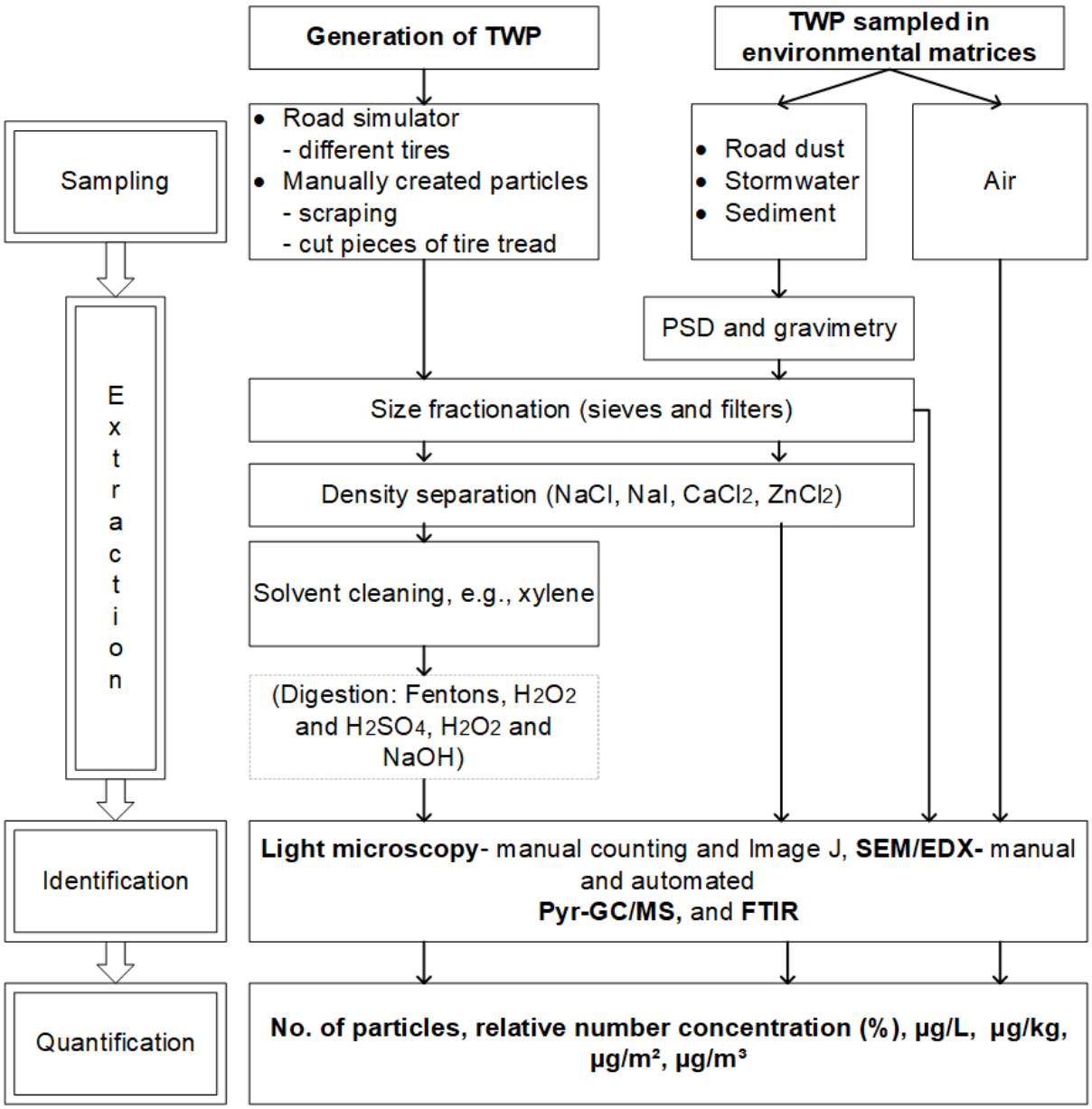

Figure 13. Simplified flow chart of the analytical steps for sample preparation, identification, and quantification of traffic-derived microplastics.

3.4.1 Analysed size ranges

Microplastics are defined as plastic particles smaller than 5 mm. Within this project, different size ranges of the traffic-derived particles have been analysed in each of the papers. A summary of the size ranges and analytical methods can be seen in Table 1. The size range for the particle size distribution was 0.125–2000 µm Paper I–IV) and the analytical limitation was set by the laser granulometer. Particles >2000 µm was inspected ocular, but not further analysed in any of the papers. For the gravimetric analysis, samples were sieved over a 180 µm mesh prior to the gravimetric analysis. The 180 µm mesh was used as a cut-off since previous research from the VTI lab where the WDS was used showed that the majority of the particles were finer than 180 µm. The 180 µm cut-off was chosen to enable comparisons with previously published studies e.g., (Gustafsson et al., 2019; Lundberg et al., 2020).

Paper I and II used light microscopy as the analytical method. The method was both time-consuming and dependent on the operator. The uncertainty increases with decreasing particle sizes, and it was not possible to differentiate between different plastic types for particles <20 µm. Even in the 20–100 µm fraction, it was complicated to distinguish between plastics, especially between TWP and BiWP in samples containing a large number of particles (i.e., sweepsand and washwater).

In Paper III and IV, the analysed size ranges were the same for PSD and gravimetry. The analytical method was SEM/EDX, and two size fractions were analysed, 2–20 µm, and 20–125 µm. The choice of size ranges had several explanations. The PSD indicated that only a few percent of the particles were coarser than 125 µm, independently of the sample matrix. It was desirable to analyse finer particles than in Paper I and II, but it was also of interest to separate the fine fraction from the coarse fraction to be able to calculate ratios between fractions (both relative and absolute). Moreover, it is harder to obtain a statistically significant result with the automated SEM/EDX for particles >125 µm due to a low number of particles.

In Paper V, several methods, various sample preparation steps, and different particle types were analysed (both manually generated TWP, and road dust collected in the road simulator). Particles >26 µm were analysed with light microscopy, SEM, pyr-GC/MS, and FTIR. The coarsest particle analysed with light microscopy was 4300 µm. The average particle (Feret diameter measured with ImageJ) was approximately 40–140 µm. Particles from studded tires tend to be finer than both studless tires and summer tires. Further, the finer particles (close to 26 µm) created noise in the data when the mosaics were analysed, therefore, a lower cut-off for light microscopy turned out to be 30 µm.

Table 1. Summary of the different size ranges analysed in each paper.

	Paper I	**Paper II**	**Paper III**	**Paper IV**	**Paper V**
	µm				
PSD	0.125–2000	0.125–2000	0.125–2000	0.125–2000	>26
Gravimetry	1–180	1–180	1–180	1–180	
Light microscopy	20–100 and 100–300	20–100 and 100–300			>26 (30)
SEM/EDX			2–20 and 20–125	2–20 and 20–125	>26
FTIR					-
pyr-GC/MS					26–63, 63–125 and >125

3.4.2 Particle size distribution

All samples (except for the air samples in Paper IV) from Paper I–IV were analysed for particle size distribution and gravimetry. The particle size distribution was determined by laser diffraction (Mastersizer 3000 from Malvern Panalytical) in the size range (0.125–2000 µm). A laser beam passes through the (dispersed) sample with particles (60 s at 1700 rpm), causing light scattering. The scattering pattern was analysed by a commercial software calculating the particle size distribution using the Mie scattering model. The refraction index used was based on earlier measurement experiences from Gustafsson et al. (2019). Measurements are performed until the measurement stabilizes, the measurement with the lowest weighted residual <1%, and the least spread is accepted. Due to the upper size limit (2 mm) of the laser granulometer, the samples were pre-sieved over an ISO 3310 VWR® 12" (mesh size 2 mm) and the measurements were performed in triplicates. Each laser granulometry measurement is performed in triplicates, and the average is calculated (Lundberg et al., 2020). The results from the measurements are presented both as *frequency distribution* (volume of particles i.e., the equivalent scattering of spheres) and *cumulative distribution* (particle size distribution for each size of particles in which percent of the sample has a size lower or equal than the values in the x-axis).

3.4.3 Gravimetric analysis

For the gravimetric analysis, a known amount of the samples (300–500mL for WDS samples and runoff, Paper I–IV, and 50g for sediment from the stormwater wells, Paper IV) were pre-sieved over an ISO 3310 VWR® 12" (mesh size 180 µm). Particles <180 µm were filtered through pre-weighted Munktell 00H filters. The filters were conditioned in an exicator and weighted whereafter the total amount of particles in the different sample matrices was calculated. In Paper I and II, the organic content was measured as loss of ignition. The weight loss was measured after incineration at 550ºC for eight hours in accordance with the procedure described in (Gustafsson et al., 2019)

3.4.4 Sample preparation

3.4.4.1 Density separation

A saturated sodium chloride solution, NaCl (1.2 g/cm^3) was used for density separation of the initial environmental samples in Paper I. Thereafter, a saturated sodium iodide solution, NaI (1.8 g/cm^3) was used to be able to collect TWP with mineral encrustations (Paper I–II). Density separation was performed in a stainless-steel separation tower (approximately 1 m high) (Imhof et al., 2012), and the samples were density separated for 6 h before the supernatant was transferred for further analysis.

In Paper III and IV, the aim was to analyse all particles within a sample, therefore no density separation was performed.

Paper V aimed to optimise the sample preparation, and a more detailed description can be seen in *Results and Discussion*, chapter 4.1.

3.4.4.2 Removal of organic matter

Environmental samples contain various amounts of organic matter which can affect and complicate the analysis. Therefore, several oxidation methods were tested on environmental samples. The methods that were tested have previously been described in the literature (Lusher et al., 2017; Tagg et al., 2017), and proved to be efficient for the removal of organic matter. Three methods were evaluated, 95% H_2SO_4 with 30% H_2O_2, 30% H_2O_2 with 1M NaOH, and

Fenton's reagent (H_2O_2 and $FeSO_4$). One of the concerns about the oxidation methods was that the TWP might be affected (e.g., degradation processes, changes in shape, change in chemical compositions) (Lusher et al., 2017). For the sediment samples collected in the stormwater well (Paper IV), a test was performed with Fenton's reagent. The oxidation was performed with satisfying results, but it was still decided to not perform an oxidation step on the samples since it was desirable to have as unaffected samples as possible.

3.4.4.3 Solvent cleaning

The differentiation between TWP and bitumen is problematic, and for Paper V it was decided to test six degreasing agents and ten solvents to find an optimised cleaning step. Xylene proved to most effective to remove bitumen without affecting TWP, and all road simulator samples were pre-cleaned with xylene prior further analyses (chapter 4.1.3).

3.4.4.4 Size fractionation

In Paper I and II the samples were sieved into two size fractions, 20–100 µm and 100–300 µm, while in Paper III and IV the samples were sieved into 2–20 µm and 20–125 µm. In Paper V, the road simulator samples were fractionated into three size fractions, 26–63 µm, 63–125 µm, and >125 µm. The different size fractions were dependent on the analytical methods. In Paper I and II, light microscopy was used for manual analysis of the particles, and the analysability was limited to 20 µm. Paper III and IV used automated SEM/EDX, where it was possible to distinguish between different particle groups down to 2 µm. In Paper V, the analysed particles were coarser than 27 µm due to limitations from the commercial lab performing the pyr-GC/MS analyses. To be able to compare results from different methods, the 26 µm mesh was used for all samples in Paper V.

3.5 Analytical methods

3.5.1 Metals and organic pollutants

In addition to the microplastic analyses, Paper II investigated the occurrence of metals and organic pollutants. Metals and OP (PAH-16, phthalates, aromatics, and aliphatics) were analysed with standardized, commercial methods (SS-EN ISO 17294-2; SS-EN ISO 17294-1,2 (mod) and EPA method 200.8 (mod); SS EN ISO 11885 (mod) and EPA method 200.7 (mod); SS-EN ISO 9377-2 (mod); US EPA8270, CSN EN ISO 6468; DIN EN ISO 18856; SS-EN ISO 18287:2008; E DIN 19742 (2012-04)). In addition, a non-target screening method was used to determine the occurrence of other organic pollutants in some of the samples. An extensive method description is presented in Paper II.

3.5.2 Methods for analysis of traffic-derived microplastics

Table 2 summarises the analytical methods used in this thesis work and the information that they provide. A more detailed description of each of the methods can be seen in Paper I–V.

The samples in Paper I and II were analysed with a stereo microscope (Leica M205 C 80–160x) by an external laboratory (IVL, research station at Fiskebäckskil, Sweden). Prior to the microscopic analysis, samples were density separated in a stainless-steel separation tower modified from Imhof et al. (2012). The analysed microplastic particles were identified as plastic fibers, plastic fragments, plastic flakes, plastic film, and road particles. The road particles were further separated into TWP and BiWP by a combination of tactile and visual identification as well as with melt tests. Light microscopy could provide information about size, shape, and surface, but the aim of Paper I and II was to identify the number of plastic particles in different sample matrices.

In Paper III and IV, an automated SEM/EDX (Zeiss Gemini 300 Field Emission Gun (FEG)-SEM equipped with an Oxford X-MAX EDS detector with an 80 mm^2 window, a high efficiency 4 quadrant backscatter electron (BSE) detector, and the particle analysis software AZtecFeature, ©Oxford Instruments) method was developed and used for all samples. The analyses were performed by an external laboratory (Particle Vision GmbH in Fribourg, Switzerland). Prior to the SEM/EDX analyses, all samples had been size fractionated at the laboratory at VTI, Linköping into two size fractions (2–20 µm and 20–125 µm). A subsample from each size fraction was dispersed on boron substrates using compressed air and a Morphology G3ID device from Malvern. Boron substrates have the advantage that they do not contain carbon which allows the identification of carbon-rich particles such as TWP, BiWP, road markings, and biogenic/organic particles. The samples were classified into different subclasses and thereafter quantified (relative number concentration and estimated absolute mass). The method provides information about size, shape, surface, number of particles, and elemental composition of each particle. The SEM/EDX method and the different subclasses are further described in the Result section 4.2.

Paper V used a multi-method analytical strategy to analyse TRWP from the road simulator and thereby increase the knowledge and understanding of properties and physicochemical composition. The light microscopy, SEM, and FTIR analyses were performed by an external laboratory (the University of Gothenburg at Fiskebäckskil, Sweden). Some of the sample preparation steps (size fractionation and solvent cleaning) were performed at VTI, Linköping. All pyr-GC/MS analyses were performed by an accredited and commercial laboratory.

A Leica Wild M8 dissection microscope was used to perform a visual-tactile differentiation between TWP and road wear. All black particles that were identified as possible TWP were further analysed and probed manually under a Leica M205 C dissection microscope with steel tweezers. Image analysis for particle measurement was performed by ImageJ. ImageJ presents 2D measurements, the height was manually measured on a selection of the TWP whereafter an estimated height was calculated for all individual particles. An ellipse-based formula was used to estimate the volume of the particles. A Zeiss Sigma VP high-resolution SEM was used to obtain additional information about the TWP particles. The morphology of the particle surfaces was obtained using a secondary electron detector.

Road simulator generated TRWP were analysed with SEM/EDS before and after the particles were cut in ultrathin sections using a diamond knife at -140ºC in a Leica EM FC6 cryo-ultramicrotome. After light microscopy and SEM/EDS analysis, the same particles were analysed with FTIR (Thermo Scientific Nicolet iN10 infrared microscope), the particles were placed in a diamond compression cell, and the transmission mode was used during the measurement.

To investigate the variability in tire composition between different tire types, manufacturers, and between road simulator generated TRWP and pure tire tread, the relation between polyisoprene (PIP) and polybutadiene (PBD) was quantified by pyr-GC/MS.

The methods that were evaluated in Paper V complement each other and when the results were combined, they provided novel information about TWP and TRWP, e.g., two different types of TRWP were detected in the samples called *sub-elastic* and *firm-elastic*. In addition to size, shape, particle surface, elemental composition, and aspect ratio (light microscopy and SEM), the pyr-GC/MS provides information about the polymer content, and the FTIR enables to distinguish between sub-elastic and firm-elastic TWP.

Table 2. Analytical methods used for identification and quantification of TWP and TRWP in Paper I-V.

Analytical methods						
	Light microscopy		**SEM/EDX**		**FTIR**	**Pyr-GC/MS**
	Manual counting	Image J	Manual	Automated		
Surface	x	x	x	x		
Size	x	x	x	x		
Aspect ratio	x	x	x	x		
Shape	x	x	x	x		
Elemental Composition			x	x		
Polymer type					x	x
No. of particles	x	x	x	x		
Relative number concentration (%)				x		
absolute mass (µg/L or µg/kg)				(x)		x

3.6 Calculations to assess the total number and absolute mass of TWP

In Paper IV, subsamples of the sieved fractions were analysed with SEM/EDX meaning that the total mass was not possible to determine directly. Therefore, the cumulative volume from the particle size distribution and the information from the gravimetric analyses were used in combination with an assumed homogenous density, 1.8 g/cm^3, (corresponding to the average density of the TWP in Klöckner et al. (2019)) and by calculating the volume of a sphere based on the equivalent circular diameter (ECD) obtained from the single particle analysis software Aztec Feature (Oxford). The absolute mass and number of particles were then extrapolated using the data from the relative mass concentrations.

Paper V used the results from pyr-GC/MS (Rubber content >26 µm (mg/g) polyisoprene and polybutadiene) and Image J to estimate the emissions of TWP from the tires used in the road simulator. A 3D ellipse-based formula was applied with the possibility to add the irregularity. The Feret diameter and an average TWP height were also implemented in the calculations. These results were then compared against literature values and the difference between methods was discussed.

4 RESULTS AND DISCUSSION

This chapter starts with the development for an optimised sample preparation, including density separation and solvent cleaning (Paper V) followed by method development (automated SEM/EDX, Paper III) and evaluation and comparison of methods (Paper V). Thereafter, the two different types of TWP, *sub-elastic* and *firm-elastic* are described (Paper V) whereafter the results from the environmental field samples are presented starting with the results from the analysis of metals and organic pollutants (Paper II) followed by traffic-derived microplastics (Paper I, II, IV).

4.1 Optimisation of sample preparation

One of the aims of the project was to find an optimised method for sample preparation. Environmental samples contain a mix of different particles, pollutants, and compounds which can make it difficult to distinguish if an effect depends on the TWP itself or on the compounds/particles sorbed on the TWP. To reduce the uncertainties with environmental samples, Paper V analysed samples collected in a controlled environment, i.e., a road simulator. Different preparation steps were also tested and evaluated on the environmental collected TWP.

4.1.1 Density separation

Several salt solutions have been evaluated for density separation of TWP and TRWP within this project. Prior to the work with environmental samples, initial tests with manually generated particles were performed with the aim to find a suitable salt solution for density separation. For that, calcium chloride, $CaCl_2$ (1.4 g/cm^3) was tested. $CaCl_2$ was chosen based on economic and environmental factors. Density separation was performed in specially made columns with a magnetic stirrer in the bottom and a valve in the middle enabling separation of the supernatant from the bottom phase. The samples were stirred for 6 hours and then left for 24 hours before the supernatant was collected. The density separation was performed in sequences and repeated twice prior to filtration and analysis. The separation worked well, but the $CaCl_2$ tends to clog the filters and heavily affect the (SEM) analysis since TWP and BiWP contain Ca. Therefore, it was decided to not use $CaCl_2$ for the upcoming analyses.

For the initial microplastic analysis in Paper I, a saturated sodium chloride solution, NaCl (1.2 g/cm^3) was used. However, the density of NaCl was too low to separate all TBiWP from minerals, especially for TBiWP encrusted with mineral particles. To evaluate potential underestimations, a test was performed on the same sample (sweepsand) with both NaCl and a saturated sodium Iodide solution, NaI (1.8 g/cm^3). The results indicated that up to ten times higher concentrations of TBiWP were detected when NaI was used. Therefore, NaI was used for the remaining samples in Paper I and II.

In Paper V, TRWP from the road simulator was used for a density separation test where six solutions with zinc chloride, $ZnCl_2$, of different densities were evaluated (1.3–1.6 g/cm^3). Both the supernatant and the bottom fraction were analysed. Repeated density extractions were also performed on aliquots of TWP from the road simulator, extracted over three consecutive 12-hour density separations ($ZnCl_2$ 1.55 g/cm$^{3)}$. The supernatants from each dense solution were filtered and the number of TWP was counted under a light microscope. $ZnCl_2$ solutions with densities <1.45 g/cm^3 seem to be equally effective for separation as a saturated salt solution with NaCl, and up to 25% of the total number of TWP were detected in the bottom fraction. The optimal $ZnCl_2$ density turned out to be 1.55 g/cm^3 where 6–12% of TWP was left in the bottom fraction. With a density above 1.55 g/cm^3, some road wear particles and bitumen particles were also detected in the supernatant which disturbed and delayed the TWP identification.

4.1.2 Solvent cleaning

In Paper V, two types of TRWP were identified, *firm-elastic* and *sub-elastic* TRWP, chapter 4.3 and Table 3. The sub-elastic TWP overlapped bitumen wear particles in form, properties, and tactile characteristics. Therefore, it was desirable to analyse TWP separately and thereby exclude BiWP from the samples. Six degreasing agents and twelve solvents were evaluated for the removal of bitumen from road simulator samples. Relative effectiveness was assessed by scoring post-treatment abundances of interfering black particles against an ACFOR scale (a method to describe the abundance of TWP within a given area, e.g., a quadrat with a known size). Xylene proved to be the most effective solvent whereof all samples in Paper V were cleaned with xylene and rinsed with methanol and de-ionized water prior to further analysis.

4.1.3 Summarizing discussion-sample preparation

During the project several methods and steps for an optimised sample preparation were evaluated. The choice of sample preparation is very dependent on the analytical method, and of course, the aim of the study. It was concluded that NaCl was not appropriate for density separation, but $ZnCl_2$ seem to be efficient- at least on TRWP collected in a road simulator. Density separation is efficient if minerals and heavy particles should be removed prior to analysis, but it comes with a risk of underestimations. If possible, it can be a good idea to avoid density separation, or at least use a salt with a much higher density than expected.
Xylene proved to be effective to remove bitumen without disturbing the TWP. So, xylene is a preferable choice if bitumen is not desirable in the analyses. Oxidation methods to remove organic matter were tested, but not further developed or evaluated. Previous research has shown that the TWP might be affected by oxidation e.g., Wagner et al. (2022), why chemical oxidation methods should be avoided if possible. Size fractionation was necessary for most analyses, especially for light microscopy and SEM/EDX where entire filters were analysed. Without size fractionation, it is more or less impossible to separate particles from each other. One of the main issues within this project is that all TRWP has been of interest, and it has not been limited to only TWP.

In Paper III and IV, it was desirable to include minerals in the analysis to be able to increase the information about ratios and the proportion between particle subclasses. While Paper I– IV focused on microplastics, metals, and organic pollutants. Paper V aimed to increase the understanding of particle properties, and TWP from different tires, therefore solvent treatment was used to remove bitumen, and density separation was performed in some of the analyses to remove minerals from road wear. All preparation steps come with uncertainties, and it is always a risk to lose particles in each step. Therefore, it is preferable to perform as little preparation as possible if the analytical method allows that. Within this project, it was not possible to determine a universal and optimised method for all sample types, but the main conclusions drawn concerning the sample preparation in this project are:

- the aim should be clarified and defined before the field campaigns are planned, and the following questions need to be answered: what kind of samples are of interest, how many samples (duplicates, triplicates, bulk samples?), and in what size interval,
- what analytical method/s should be used?
- sample collection and preparation: similar studies/samples? Possible to compare the results?
- is it possible to include additional analyses that can increase the understanding of the complex particle composition (e.g., metals, OP, size distributions, toxicity tests)
- is it possible to combine analytical methods to increase the amount of information? If so, is it possible to use the same sample preparation?

4.2 Method development

Paper III further developed a machine learning algorithm coupled to an automated Scanning Electron Microscopy/Energy Dispersive X-ray spectroscopy (SEM/EDX) analytical approach (Rausch et al., 2022) to classify and quantify the relative number concentration (%) of the following subclasses contained in environmental road dust: tire wear particles (TWP), bitumen wear particles (BiWP), road markings, reflecting glass beads, metallics, minerals, and biogenic/organics. Reference material from different road surfaces, road markings, bitumen, and reflecting glass beads was analysed both manually and automatically. To not disturb composition, texture, and size distribution, no sample preparation was performed on the reference materials prior to analysis. In addition, the reference materials were milled and dispersed on boron substrates to be more like real-life particles and thereafter analysed again. The results from the reference materials were implemented into the particle classifier enabling the recognition of these particle types in environmental samples (Paper III).

Environmental road dust samples from Testsite E18 were analysed with the automated SEM/EDX, and for each particle, an EDX analysis (with AZtecFeature software) and a picture with the backscatter electron (BSE) was performed. The data from the AztecFeature software was then treated by a machine learning algorithm. The ML-algorithm categorized the particles into subclasses based on 67 chemical and morpho-textural parameters. An image and elemental spectra of each particle are available if it is desirable to manually check some of the particles. It is also possible to check the "confidence score" and move particles between subclasses if they have been classified incorrectly by the ML-algorithm.

Even though environmental particles tend to have a certain heterogeneity, each particle subclass was defined by a clear fingerprint when the morpho-textural information was combined with elemental information. One challenge with the automated SEM/EDX classification occurs when the random forest classifier should distinguish between fine particles (<5 µm) of tire wear (TWP) and bitumen (BiWP). Both the shape and elemental composition of these particles tend to be very similar. For the coarser particles, a differentiation is possible based on other parameters (e.g., chemical, textural, and morphological), but for the fine particle fractions, where the particles are encrusted with each other, it was not possible to separate TWP from BiWP. Therefore, TWP and BiWP smaller than 20 µm was counted together in the subclass tire and bitumen wear particles, TBiWP. This has been illustrated in Figure 14, where the ML-algorithm has classified the TWP and the BiWP. Particles with similar characteristics are clustered together, and as can be seen, the coarse fraction (20–125 µm) creates well-defined clusters while the fine fraction (2–20 µm) clusters overlap to a larger extent.

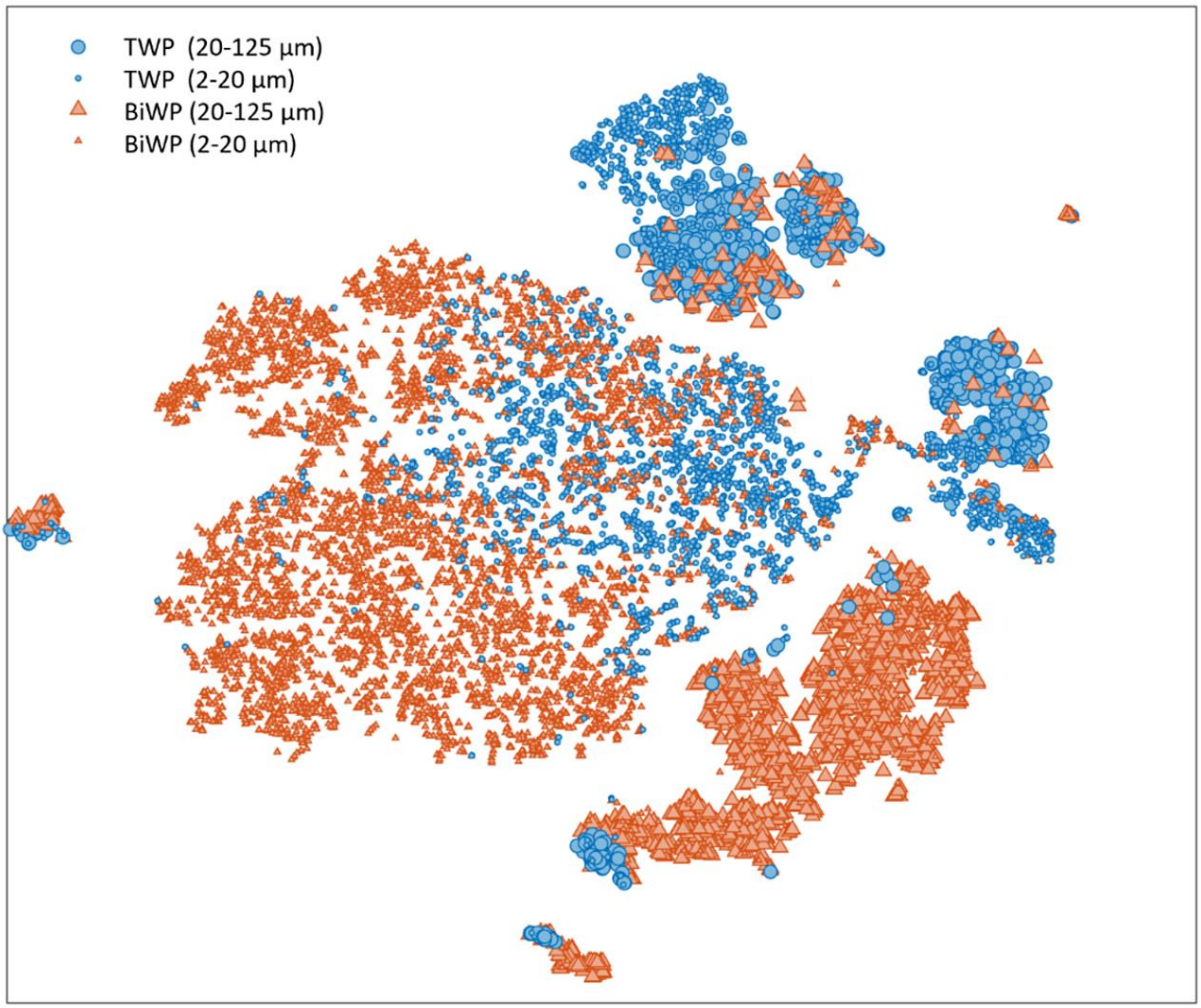

Figure 14. The TWP and BiWP subclasses was extracted from the main dataset of all particles classified with the road dust classification model. Particles with similar characteristics are located close to each other in the image. This graph shows that coarse particles (20–125 µm) tend to build better-defined clusters than fine particles (2–20 µm) before the classification model is applied. This illustration visualizes the uncertainties to distinguish between TWP and BiWP in the fine fraction. Figure from Paper III.

Overall, the SEM/EDX/ML method divided the particles into correct subclasses with high repeatability. The method proved to be rapid, require little sample preparation, and the samples were not destroyed after analysis enabling further analyses if desired. Lastly, it was possible to analyse particles down to 2 µm with detailed information about size, surface structure, elemental composition, and shape which is advantageous when the transport routes of TWP should be investigated and modeled. However, one disadvantage with the automated SEM/EDX is that it is not suitable for coarser particles (>125 µm), it is for example difficult to obtain a statistically relevant result if only a low number of particles can be analysed in each run considering the magnification settings used for the SEM. Further, coarser particles might conceal finer particles and thereby affect the result. This limitation results in a potential underestimation of traffic-derived microplastics. On the other hand, the particle size distributions have shown that the majority of the particles are finer than 125 µm, fine particles are transported longer distances and to a greater extent than coarser particles. Another limitation with the existing SEM/EDX method is that polymers cannot be defined. As previously mentioned, analysis of TRWP is challenging and the focus of Paper III has been to characterize and identify TWP, bitumen, and road markings. Other microplastics will probably be classified into “organic matter” due to the high content of hydrocarbons.

4.3 Characterization of manually and laboratory generated TRWP

Paper V investigated the differences between tire types and brands and showed that the rubber content within a tire group (e.g., summer tires, studless winter tires, and studded winter tires) were similar, but varied between groups. The ratio between natural and synthetic rubber were similar within the tire groups independent of tire brand, but a large variation was detected between the tire groups. The variation between tire groups was expected since winter tires contain softer rubber than summer tires (i.e., different proportions of polyisoprene, polybutadiene, and additives).

One of the major findings with Paper V was that two types of TRWP with slightly different properties were detected, Table 3. The different types were called *firm-elastic* and *sub-elastic* TRWP where the latter, more fragile particle class comprises 99 % of all the identified TWP, see example particles in Figure 15. When the TRWP was analysed with light microscopy and SEM; it was not clear if *sub-elastic* TRWP should be counted as a TWP, therefore other possible sources were investigated (i.e., lubricants from the road simulator, oil, bitumen). The *sub-elastic* particles tend to smear out when they were probed with tweezers, they were often elastic and elongated in shape and they were either glossy or matt-black with mineral encrustations on the surface. The *firm-elastic* TWP was firm and retained its original shape directly after poking, and felt more like suspected TWP. Moreover, the *firm-elastic* TWP was smooth and mat on the surface and they were often compact and angular to its shape, especially in the <300 µm fraction. The *firm-elastic* and *sub-elastic* TWP was distinguished from each other based on several tactile characteristics: firmness, brittleness (*sub-elastic* TWP were more fragile), the ability to regain original shape, and smearing.

Table 3. Description of the two types of TRWP detected in the road simulator samples. Table from Paper V.

	Sub-elastic TRWP	Firm-elastic TRWP
Color	Matt or glossy black. Normally with less noticeable surface mineral encrustation than firm-elastic TRWP.	Matt black, although normally with a coating of pale embedded mineral particles.
Form	Commonly present as elongated cigar-shaped "rolls", tapered at one or both ends, but also with variable, irregular shapes and, in summer tire dusts, including fluid forms.	The outline of smaller particles (with a largest diameter < approx. 500 µm) is often distinctive; normally gnarled, and/or knobbly/bobbly. Larger particles also occasionally had a smoother matt surface, and include angular chunks or rolled shapes.
Feel	Normally with little or no elasticity. A few particles had some degree of elasticity, in that they started to pull back towards their original shape after being depressed with tweezers, but the response was slower than that for pristine tire rubber. Particles could either be easily smeared into a broken or unbroken film or, in the case of larger brittle summer tire particles, be broken up into brittle angular pieces. Resistance to smearing was always greater than that for grease particles.	When poked with a tweezer, these particles feel firm and springy, and respond to poking as expected for a piece of tire. >100 µm particles normally regain their original shape to a great extent, but smaller examples tend to be destroyed (crumbled or spread out) by light tweezer pressure. Similar descriptions of tire rubber have been reported in the McCrone Atlas of microparticles.
Abundance	The dominant black particle type in the asphalt associated road simulator dusts.	Rare

The results from light microscopy, SEM/EDX, and ImageJ showed that a considerable proportion of the black non-minerogenic particles consisted of black particles covered in mineral crystals that were most common in the road simulator samples from studded winter tires, probably a result of the increased road wear generated from the studs. The encrustations made it difficult to quantify and characterize individual TRWP, and the particles were not captured by density separation resulting in possible underestimations due to heavier densities than expected. A variety of different *sub-elastic* and *firm-elastic* particles can be seen in Figure 15.

The pyr-GC/MS results confirmed that the *sub-elastic* particles contain high concentrations of tire polymers i.e., polyisoprene and polybutadiene and that both particle categories should be counted as TWP. To ensure the origin of *sub-elastic* and *firm-elastic* TWP, particles from each category were handpicked and analysed with pyr-GC/MS. The results indicated that concentrations of polyisoprene and polybutadiene were lower in the *sub-elastic* TWP, which can be explained by the fact that the *sub-elastic* TWP was softer and had a higher density which might have led to more encrustations with minerals. Moreover, when the same tires were run on a concrete pavement instead of an asphalt pavement, the amount of *sub-elastic* TWP was reduced to 18% (compared to 99% on an asphalt surface) indicating that the road surface influence the generation and properties of TWP.

The wheels in the road simulator run on a 5-meter circular track which probably results in higher TWP emissions due to a continuous turn-slip abrasion compared to driving at the same speed on a straight road. However, one major advantage with the road simulator is the possibility to compare and evaluate different tires and road surfaces (asphalt pavement and concrete). A difference in tire hardness, tread depth, and polymer composition was detected between tire types and tire brands. This information is of importance and should be further investigated since variations in hardness (differences in additive composition) and polymer ratios can affect the wear rate.

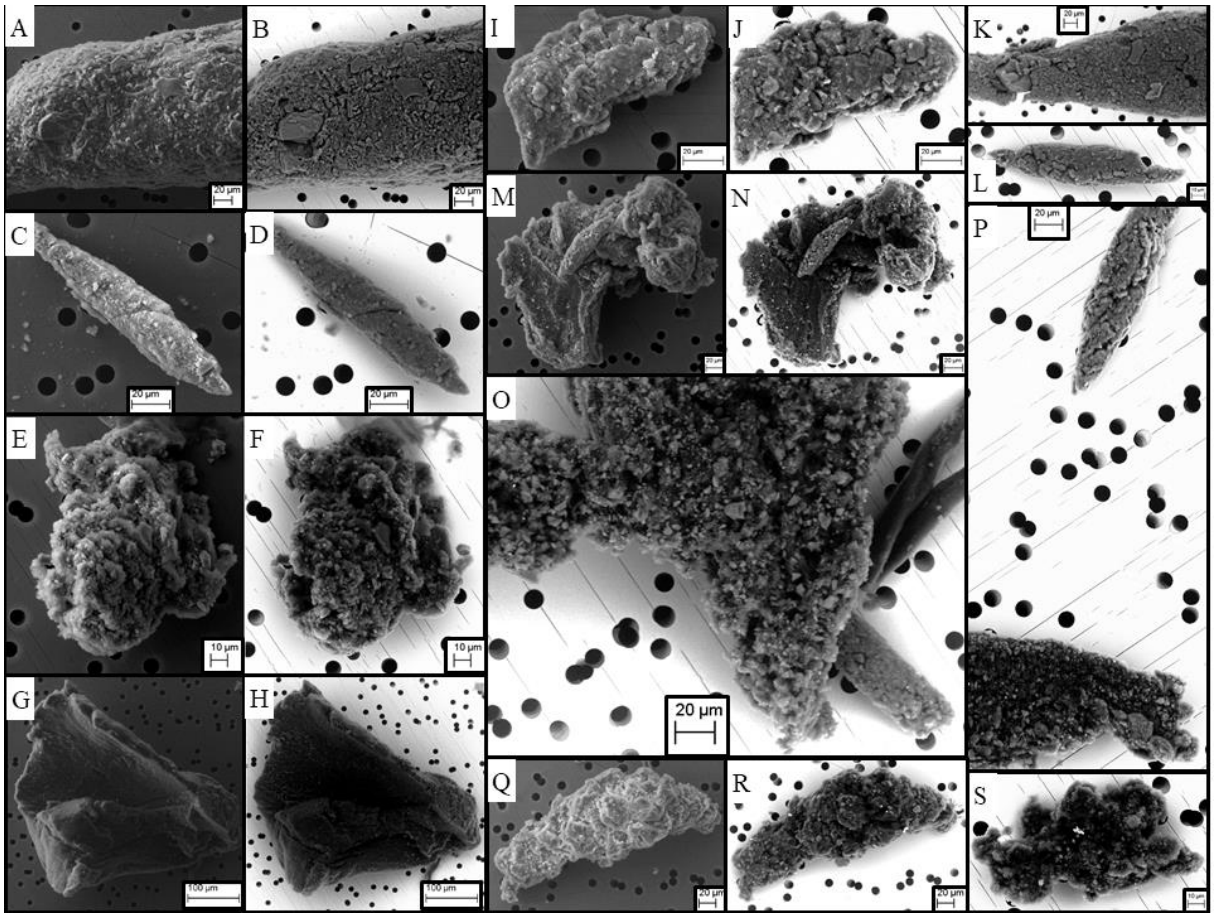

Figure 15. ***A)*** *Secondary electrons (SE) from a large brittle* ***sub-elastic*** *TRWP, summer tire.* ***B)*** *Backscattered electrons (BSE) on the same particle as in A.* ***C)*** *SE from a* ***sub-elastic*** *TRWP from a studded tire.* ***D)*** *BSE on the same particle as in C.* ***E)*** *SE from a* ***firm-elastic*** *TRWP, studded tire.* ***F)*** *BSE on the same particle as in E.* ***G)*** *SE from a large* ***firm-elastic*** *TRWP from a studded tire.* ***H)*** *BSE on the same particle as in G.* ***I)*** *SE from a* ***sub-elastic*** *TRWP, studded tire.* ***J)*** *BSE on the same particle as in I.* ***K)*** *BSE from a large brittle* ***sub-elastic*** *TRWP, summer tire.* ***L)*** *BSE from a* ***sub-elastic TRWP,*** *studded tire.* ***M)*** *SE from a large* ***firm-elastic*** *TRWP, studded tire, cradling a smaller, elongate,* ***sub-elastic*** *TRWP from a studded tire.* ***N)*** *BSE from the same large* ***firm-elastic*** *TRWP cradling a smaller, elongated,* ***sub-elastic*** *TRWP.* ***O)*** *BSE from a large* ***firm-elastic*** *TRWP from a studded tire, partly covering a smaller, elongated,* ***sub-elastic*** *TRWP from a studded tire (a smooth dark contaminating fibre extends to the right margin of the image).* ***P)*** *BSE from a large* ***firm-elastic*** *TRWP, studded tire, at bottom of image and a smaller, elongated,* ***sub-elastic*** *TRWP from a studded tire extending from the top of the image.* ***Q)*** *SE from a* ***firm-elastic*** *summer TRWP* ***R)*** *BSE on the same particle as in Q.* ***S)*** *BSE from a* ***firm-elastic*** *TRWP from a studded tire. All particles are supported on a platinum-coated polycarbonate substrate, with a 10µm pore size. Figure from Paper V.*

4.4 Metals and organic pollutants

Metals and organic pollutants were analysed in all samples collected in stormwater, road dust, sweepsand, and washwater (Paper II and Table 4). The local guidelines for stormwater were exceeded for PAH, petroleum hydrocarbons, and all metals except chromium, indicating that the stormwater should be treated prior to release. For the stormwater, the event mean concentration (EMC) EMC was in the same magnitude throughout the study period, and it was not possible to identify a trend or a specific source for the high concentrations. The PAH concentrations in the stormwater were in the same size range as Hou and Li (2018) and Zgheib et al. (2012) but lower than in Masoner et al. (2019). For sweepsand and washwater, no specific guideline values are available, but the metal concentrations were several times higher than both the local guideline values for release of polluted water to recipients, Miljöförvaltningen (2013), and the Canadian interim sediment quality guidelines, CCME (2010), who present guideline values for both metals and OP in sediment.

The construction work, excavators, and heavy traffic are possible sources, and the results are in line with previous studies (Ma et al., 2021; Nawrot et al., 2020). In the sweepsand samples, only a few of the analysed PAH were detected in concentrations above the quantification limit, however, the average concentrations were still 490 µg/kg which is above the Canadian Sediment quality guidelines. When a more sensitive method was used, up to 732 µg/kg were detected (Paper II). Similar results were presented in studies from Iran (Abbasnejad et al., 2019) and Vietnam (Anh et al., 2019a). Even petroleum hydrocarbons (aliphatic C_{16}–C_{35}) were found in high concentrations in both sweepsand and washwater, with traffic and exhausts from diesel engines as the most likely sources (Anh et al., 2019b).

Table 4. Concentrations of organic pollutants and metals found in different sample matrices in an area under reconstruction (Paper II).

	Stormwater	**Washwater**	**Sweepsand**	**WDS**
	[µg/L]		[µg/kg dw]	[µg/m²]
	n=9	n=6	n=7	n=2
As	1,6	27,0	644,5	390,1
Cd	1,1	3,4	235,8	54,8
Co	3,3	116,7	4 480,0	2 090,3
Cr	6,5	306,2	12 681,4	4 938,0
Cu	210,2	1 351,7	41 728,6	14 960,8
Mo	BQL	30,7	3 228,9	280,0
Ni	5,2	212,8	8 454,3	4 766,6
Pb	24,8	535,0	12 091,4	7 917,1
Zn	341,1	5 851,7	134 142,9	76 803,8
V	BQL	514,8	14 142,9	9 500,5
naphthalene, NAP	0,3	0,1	BQL	2,1
acenaphthylene, ACY	0,0	0,1	BQL	BQL
acenaphthene, ACE	0,0	0,1	BQL	BQL
fluorene, FL	0,0	0,2	BQL	2,0
phenanthrene, PHE	0,2	2,2	BQL	10,4
anthracene, ANT	0,0	0,4	BQL	4,1
fluoranthene, FLR	0,4	4,6	143,7	29,5
pyrene, PYR	0,3	4,1	135,3	27,8
benzo(a)anthracene, BaA	0,2	2,1	BQL	13,1
chrysene, CHY	0,2	2,5	BQL	32,9
benzo(b)fluoranthene, BbF	0,2	3,4	108,0	21,8
benzo(k)fluoranthene, BkF	0,1	0,9	BQL	5,6
benzo(a)pyrene, BaP	0,1	2,5	BQL	9,4
dibenzo(ah)anthracene, DBahA	0,0	0,5	BQL	3,3
benzo(ghi)perylene, BPY	0,1	2,2	BQL	10,9
indeno(123cd)pyrene, INP	0,1	2,1	BQL	9,5
∑PAH-L	0,3	0,3	BQL	1,1
∑PAH-M	1,0	11,5	313,0	70,7
∑PAH-H	1,0	16,2	200,5	104,8
∑PAH16	**2,3**	**28,0**	**490,0**	**176,5**
aliphates >C10-C12	8,0	20,2	BQL	BQL
aliphates >C12-C16	24,5	80,2	BQL	BQL
aliphates >C16-C35	755,6	3 148,0	167 571,4	24 077,7
aromates >C8-C10	N.A	0,5	BQL	BQL
aromates >C10-C16	N.A	2,2	BQL	BQL
methyl pyrenes + methyl fluoranthenes = ∑MP +MF	N.A	4,2	BQL	BQL
benzo(a)anthracenes = ∑MC +MB(a)A	N.A	5,2	BQL	BQL
aromates >C16-C35 = ∑MP +MF + MC + MB(a)A	N.A	9,4	BQL	BQL
di ethyl phthalate	N.A	3,2	BQL	BQL
di-n-butyl phthalate	N.A	1,3	70,0	BQL
di-iso-butyl phthalate	N.A	1,2	BQL	BQL
di-(2-ethyl hexyl)phthalate (DEHP)	N.A	17,1		857,0
di-iso-nonyl phthalate (DINP)	N.A	98,0	BQL	BQL

High concentrations of metals and organic pollutants were found in stormwater, road dust samples, washwater, and sweepsand (commercial methods). Zn and Cu were the dominating metals in all sample matrices, followed by Pb and V. When the results from each sample media presented with the same unit, µg/L (i.e., WDS, washwater, and stormwater) were correlated to each other it was clear that the metal concentrations in the WDS samples had a strong correlation to washwater (R^2=0.99), but a weaker correlation to stormwater (R^2=0.83). Consequently, washwater also had a weak correlation with stormwater (R^2=0.36). For PAH-16, the pattern was similar, but with slightly weaker correlations: stormwater against WDS, R^2=0.55, stormwater against washwater, R^2=0.60, and washwater against WDS, R^2=0.79. This may be explained by the longer transport route for the stormwater over impermeable surfaces and long transport route in pipes before reaching the stormwater sampler. During this transport, PAH could sediment with coarser particles and the most volatile PAH could also evaporate causing a change in the relative PAH composition. In Polukarova et al (2020), it was clearly seen that the stormwater samples contained relatively more of the light weight and more water-soluble PAH than the WDS and washwater samples. In addition, a non-target screening (non-commercial) for organic pollutants were tested resulting in findings of approximately 100 organic compounds of anthropogenic origin (e.g., alkanes, phthalates, and solvents) (Paper II). The average concentrations were 310 µg/L for stormwater, 78 000 µg/L for washwater, and 36 000 µg/kg for sweepsand. These results highlight the importance of rapid analytical methods that cover more organic compounds (e.g., phthalate isomers) since the existing methods tend to underestimate the concentrations of organic pollutants. The screening also showed the occurrence of N-(1,3-Dimethylbutyl)-N'-phenyl-p-phenylenediamine (6PPD) in the street sweeping washwater which is together with its degradation product a highly toxic pollutant (Tian et al., 2021). In summary, high concentrations of metals, and organic pollutants were detected in all sample matrices indicating that the traffic is a plausible source of large emissions of metals and organic pollutants. In the environment, a contaminant is seldom found on its own, the particles consist of a complex mixture of minerals, metals, OP, MP, TWP, and organic material. If a study is limited to a specific group of contaminants, the overall knowledge will continue to be limited. If possible, it is always of interest to perform additional analyses e.g., complement MP analyses with PSD, OP, and metals. Even though the content of metals and OP in road dust has been well investigated over the years, the combination with MP and TWP is rare.

4.5 Dust load and particle size distribution - comparison between environments

When TRWP has been generated at the road surface, a large amount of the particles deposit in the road dust. During precipitation, some road dust is transported to the runoff, while other parts are sedimented in the structure of the road surface or at the kerbside. When the road dust has settled, it acts as a temporary sink for traffic-related microplastics since the MP can stay at the road surface for a long period (i.e., until resuspension, road maintenance or heavy rain/wind occurs). Road dust can act both as a sink and a source of the TRWP emissions in the runoff. Therefore, it is of interest to investigate the road dust and to analyse the particle size distribution in different traffic environments, especially with complementing information about pavement, meteorology, road dust composition, and road maintenance.

Road dust was sampled and analysed in Paper I–IV. The amount of road dust in the size range 1–180 µm (g/m^2) for each sampling occasion (Paper I, II and IV) can be seen in Figure 16. The left figure shows samples collected adjacent to the kerb, and the figure to the right are samples collected in-between wheeltracks. Based on Figure 16, the total amount of road dust seems to be larger in-between wheeltracks in the urban area in Paper I even though the streets were more frequently swept than the rural highway. This is likely an effect of much lower speeds (less

removal by resuspension), more braking and acceleration, and graveling of nearby cycle paths during the winter. Precipitation and wind can also affect the amount of road dust, the urban streets in Gothenburg are surrounded by buildings while the rural highway is surrounded by open fields. It was also clear that the largest amount of road dust was found adjacent to the kerb, as previously described in Gustafsson et al. (2019). In-between the wheeltracks, the dust loads varied between 2–40 g/m^2, while in the samples collected adjacent to the kerb, the lowest amount of road dust was 180 g/m^2 and the highest over 1200 g/m^2.

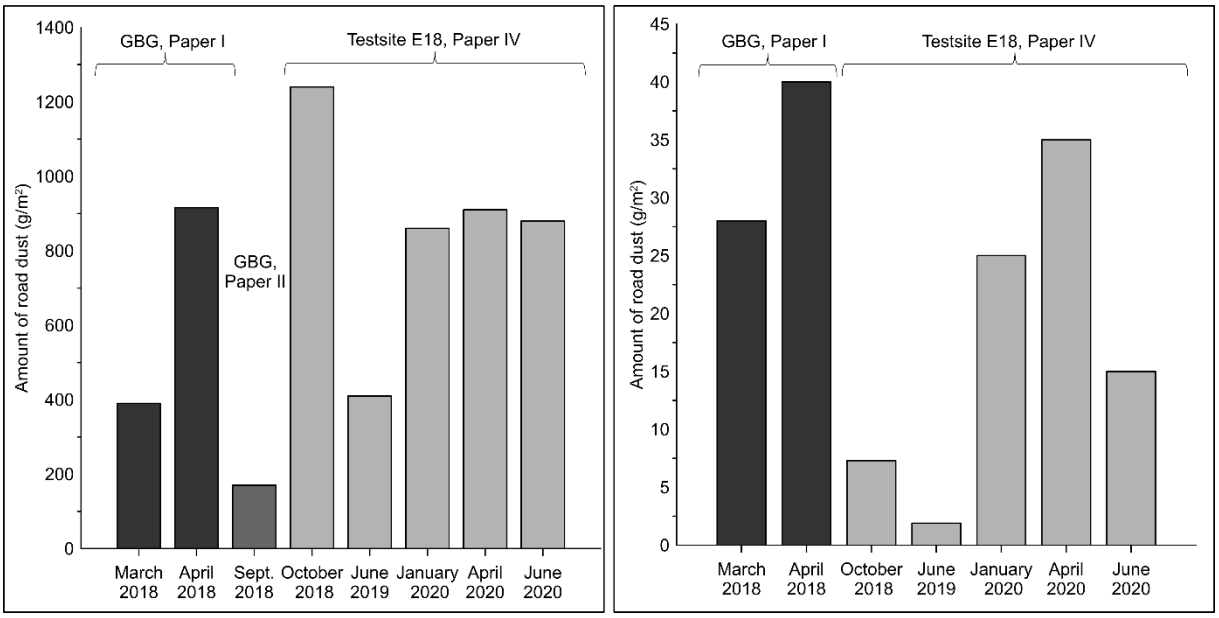

Figure 16. Amount of road dust (g/m^2) for samples collected adjacent to the kerb (left) and in-between wheeltracks (right). Paper I and II collected samples in an urban area (streets in the inner city of Gothenburg (GBG)), and Paper IV in a rural area (highway).

In Figure 17, the particle size distribution (frequency distribution) is shown as mean values. Particles in the size range 0.025 µm–2000 µm were analysed, and the results clearly show the importance to include also fine particles in microplastic analyses to avoid underestimations. The size distribution of road dust samples collected at Testsite E18 (highway, rural area) were slightly finer than road dust samples collected in Gothenburg (urban area). Some explanations can be that the speed and AADT are higher at Testsite E18, that the speed varies more in Gothenburg (start-stops), and that more gravel (i.e., from the bike- and pedestrian lanes) is transported onto the driving lanes in the urban areas. Another explanation is that the roads in Gothenburg were frequently swept, compared to the highway that is seldom swept resulting in more grinding of the road dust. Further, the pavements contain different stone aggregates at the different sites which might affect the size distributions. Another explanation, as could also be seen in Paper V, is that the use of studded tires generates finer particles than summer tires, especially when the speed is higher i.e., at Testsite E18 (120km/h) compared to Gothenburg (50km/h). The average particle size was 42 µm (in-between wheeltracks, Gothenburg), 37 µm (kerb, Gothenburg), 25 µm (in-between wheeltracks, E18), and 20 µm (kerb, E18). Gothenburg is located by the coast, more south than Testsite E18, the climate in Gothenburg is milder, and a smaller proportion of the cars have studded tires.

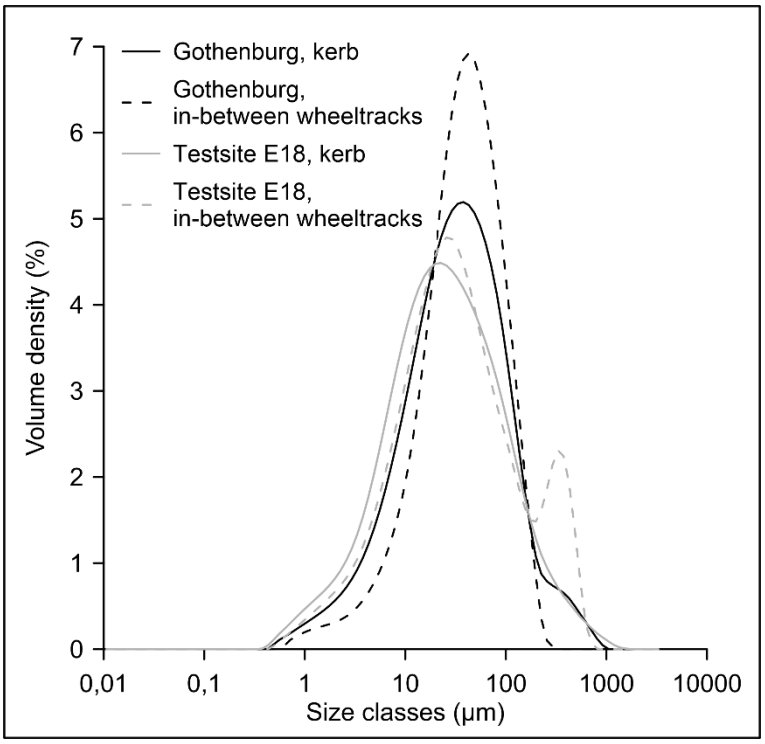

Figure 17. Average Particle Size Distribution of road dust samples collected at two positions in Gothenburg (Paper I and II) and at two positions at Testsite E18 (Paper IV).

The cumulative distribution (volume) from the Testsite E18 samples (mean value from the kerb samples, published in Paper III) shows that 85% of the particles (by volume) are within the interval 2–125 µm (5% <2 µm and 10% >125 µm), Figure 18. This information was important to decide what sizes to analyse further with the SEM/EDX. In Paper I and II, particles <20 µm were not analysed due to analytical challenges. Kreider (2010) reported that less than 1% (volume) were <10 µm and it has previously been discussed that the fine fraction probably accounts for a negligible part of the total particle mass or volume even though the total number of particles are higher for fine particles. However, Figure 18 shows that the fine fraction (2–20 µm) stands for a considerable part of the sample (≈45% by volume) and should definitely not be excluded from the analyses. Also, the cumulative distribution of particles collected in the road simulator (Paper V), showed that more than 50% of the particles were <60 µm. Particles finer than 30 µm were not analysed in Paper V, but it is plausible that a large amount of the particles were finer than 30 µm even for the road simulator samples. The coarsest particles were detected when the road simulator runs with summer tires or studless winter tires (85% of the particles 30–140 µm) and the finest particles were generated when studded tires were in use (85% of the particles 30–80 µm). The difference in particle size is probably dependent on the studs since the hard metal studs generate more- and finer particles, the studs is also responsible for an increased road wear resulting in a higher ratio of road wear particles. One of the tires diverged from the pattern, Summer tire-C, where only 50% of the particles were <300 µm (Paper V). One explanation to the variations in particle size distributions can probably be related to the ratio between natural and synthetic rubber as well as the inclusion of additives and carbon black (i.e., different tire recipes) as discussed in Paper V.

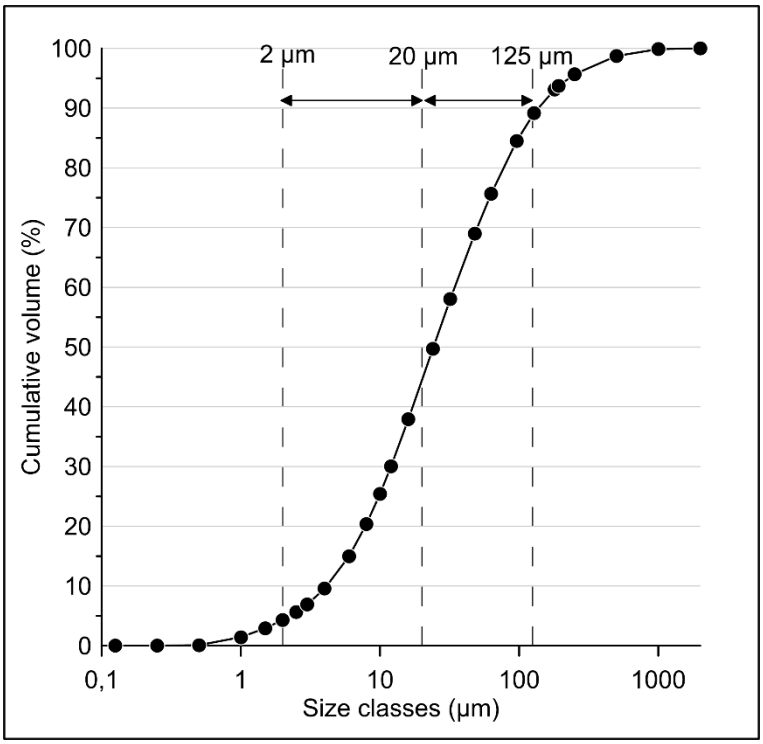

Figure 18. Cumulative volume (%) of the particle size distribution (PSD) for road dust samples. The PSD is presented as a mean value from the five sampling occasions, figure from Paper III.

4.6 Particle size distribution in runoff

In Paper IV it was desirable to compare the particle size distributions in the different sample matrices to detect potential differences, Figure 19. The results show some interesting patterns, with coarse particles in the roadside gully pot (placed at the roadside), slightly finer particles at the road surface, and the finest particles was found in the stormwater well. Higher intensity and duration of precipitation enable the transport of coarser fractions of road dust. When coarse particles reach the roadside gully pot, they settle into the sediment, while finer particles are more easily further transported to the stormwater well. In the stormwater well (sediment and water), the PSD:s are finer, with a maximum below 20 µm. A possible explanation for the large number of coarse particles in the roadside gully pot water is that coarse road dust from the kerbside is continuously resuspended by traffic turbulence and deposited into the gully pot. A high dust load (880 g/m^2) was also detected in the area adjacent to the kerb, i.e., where the roadside gully pot is located, Figure 16. Moreover, road dust has been shown to have hydrophobic properties which might explain why some coarse particles remain floating on the water surface.

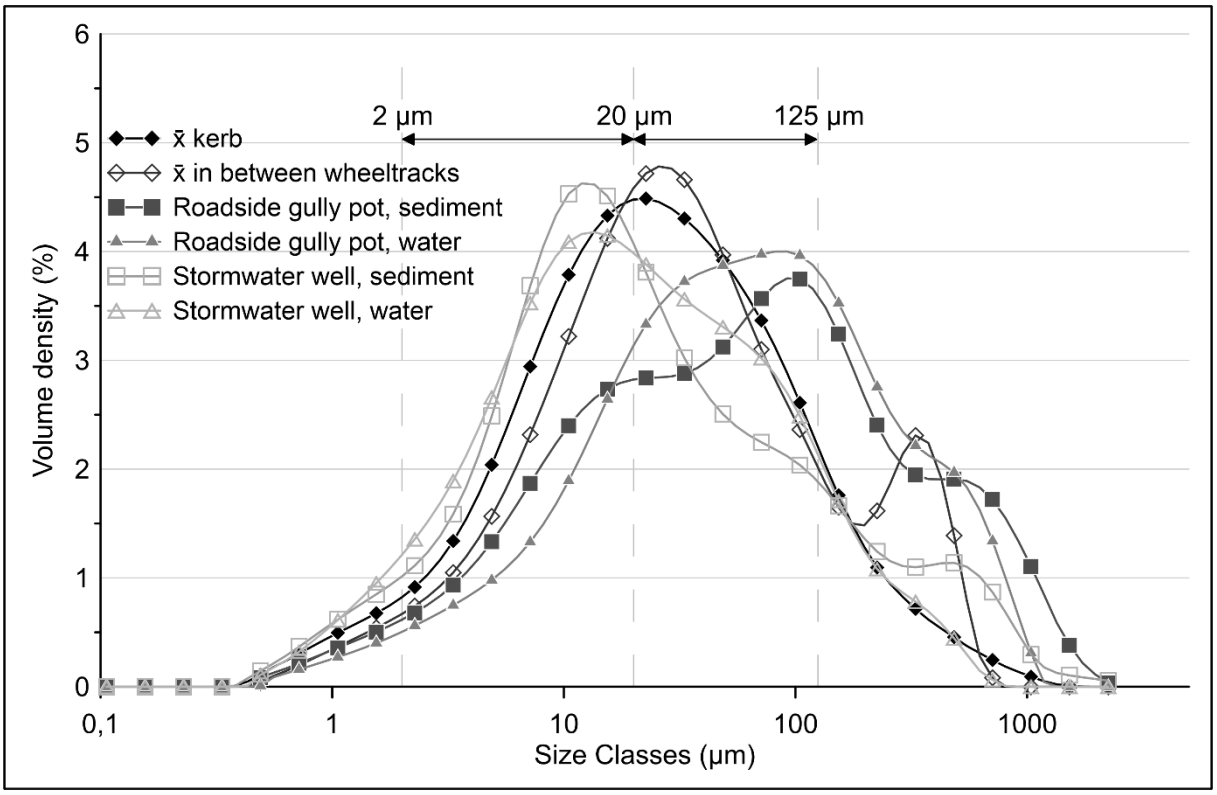

Figure 19. Average Particle Size Distribution of road dust (two positions) and runoff samples (both sediment and water from the stormwater well and the roadside gully pot) at Testsite E18 (figure based on data from Paper IV).

4.7 Traffic-derived microplastics in different environments

4.7.1 Urban areas

The same methodology, light microscopy, was used for Paper I and II enabling comparison between the two sites. The AADT was higher in the study area of Paper I (13 000 vehicles compared to 5 000 vehicles), but the amounts of heavy traffic were much higher in the study area of Paper II due to an extensive reconstruction within the area. For the first batch of samples (Paper I), sodium chloride (NaCl 1.2g/cm^3) was used for density separation. However, even if NaCl theoretically has a higher density than rubber, it was concluded that the results were heavily underestimated since TWP contains mineral incrustations which increase the density. Therefore, sodium iodide (NaI, 1.8 g/cm^3) was used for the upcoming samples.

To evaluate potential underestimation, Paper I divided homogenous sweepsand samples from two sampling occasions into two and density separated them in parallel with NaCl and NaI. In the samples separated with NaI, 2–9 times higher concentrations of TWP were found. Only the supernatant was analysed, and no analysis was performed on the sedimented material. For upcoming studies, it had been of interest to use a solution with a higher density than NaI to determine if the samples separated with NaI were underestimated as well.

4.7.1.1 Total number of tire wear particles

In Figure 20, the number of TWP/L (100–300 µm) in the different sample matrices from Paper I and II have been compared. Samples analysed with NaCl have been excluded. The results indicate that the mean number of TWP was higher in all sample matrices (stormwater, washwater, road dust, and sweepsand) in the area under reconstruction (Paper II) in comparison to the urban area (Paper I). However, the variation within a sample matrix was larger in samples from Paper II, especially for the washwater, where the maximum value was 23 120 TWP/L and the minimum value was 1 TWP/L. The large variations within a sample matrix can probably be related to *when* the sweeping was performed. If the sweeping was performed directly after a heavy rain event, the first flush has probably transported most of the TWP into the stormwater resulting in low concentrations at the road surface while a long dry period accumulates the TWP resulting in high road surface concentrations. The amount of heavy traffic can also affect the variations.

The variations between study areas can be explained by several reasons such as meteorology, precipitation, AADT, maintenance, and sampling dates. As an example, all traffic from highway E6 was redirected through the study area of Paper II on several nights during the measurement period. The precipitation 10 days prior to the street sweeping varied between 6–10 mm (Paper I) and 0–91.3 mm (Paper II). However, there was no correlation between recent precipitation and high/low TWP in the collected sweeping material. Neither had the high concentrations in the stormwater any correlation with concentrations in the sweeping material. Further, the sweeping material was collected during the seasons when no studded tires were in use (summer and autumn), and stormwater was collected during all seasons. That might also affect the results, i.e., higher TWP emissions during periods without studded tires.

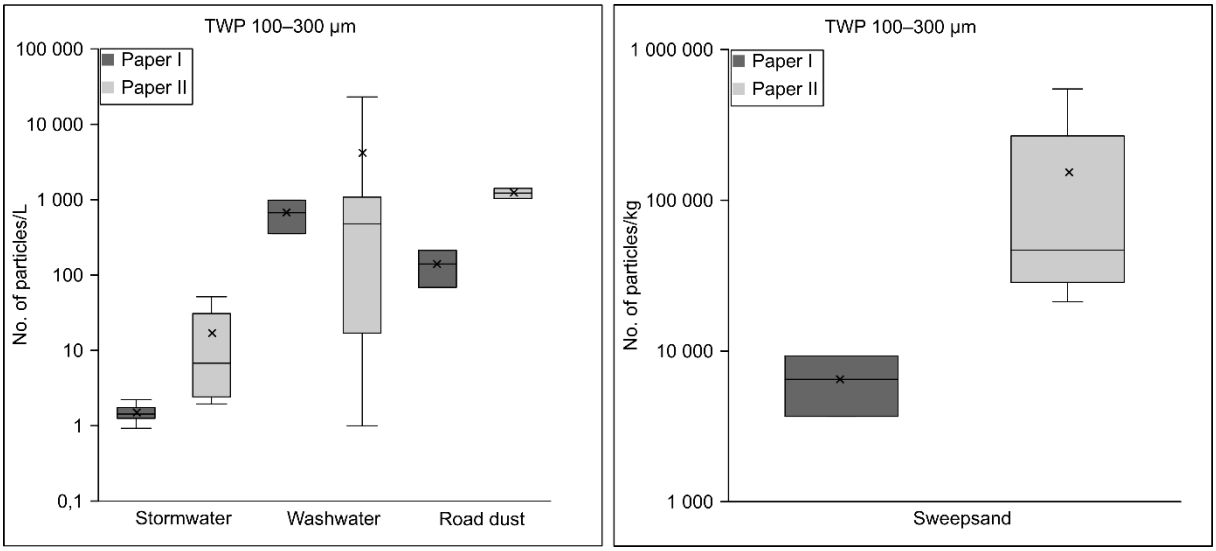

Figure 20. Comparison of the number of TWP in stormwater, washwater, road dust, and sweepsand. The whiskers show the 90 and 10 percentiles, the line within the box is the median and the cross represents the mean value.

Fine particles <50 µm are difficult and uncertain to analyse with light microscopy. It is time-consuming and requires an experienced operator (Primpke et al., 2020). However, for some samples in Paper I and II, particles down to 20 µm were analysed (20–100 µm). In Paper I, no differentiation between TWP and bitumen was made. Therefore, in Figure 21, the results are presented as tire and bitumen wear particles, TBiWP. The number of TBiWP/L was several times higher in Paper I compared to Paper II, which is the opposite pattern compared to the coarser fraction (100–300 µm) where Paper II had the highest concentrations. One possibility

is that the heavy traffic generates coarser particles, or that the finer particles stay longer on the road surface, due to cavities in the pavement. Moreover, a stormwater sample from Paper II was in parallel analysed with the automated SEM/EDX method used in Paper III and IV. The same sample preparation and size range was used. With SEM/EDX, the number of TBiWP was more than 100 times higher, highlighting the uncertainties to analyse fine particles with light microscopy only, Figure 21. For the coarse fraction (100–300 µm), the results were in the same size range, indicating that light microscopy is accurate for the coarser particles, but uncertain for fine particles.

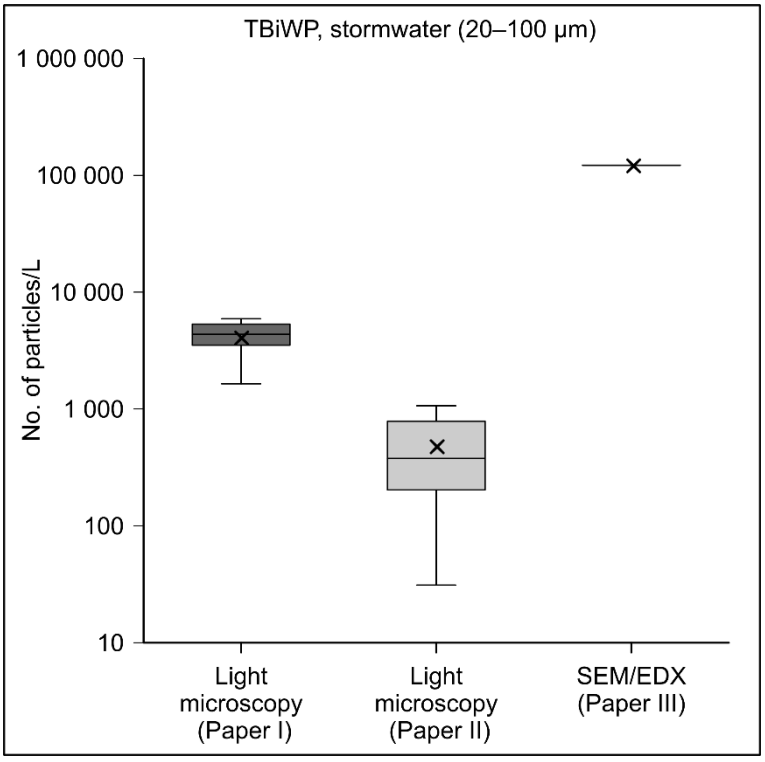

Figure 21. Comparison of the number of TBiWP/L (20–100 µm) in stormwater. The whiskers show the 90 and 10 percentiles, the line within the box is the median and the cross represents the mean value.

4.7.2 Rural area

In addition to urban samples analysed with light microscopy (Paper I and II), rural highway samples were analysed with the developed automated SEM/EDX method (Paper III and IV). Based on the total number of analysed particles, fixed densities, and the assumption that all particles are spherical, the relative mass- and number concentration of all particle subclasses were calculated. The relative number concentration (%) presents the composition of the sample and enables comparison between the different sample matrices (independently of unit or sample media). The SEM/EDX analyses were performed on subsamples, and a rough estimation of the absolute masses of each particle subclass was performed by combining the information from the relative mass concentration with information from PSD, gravimetric analyses, and the estimated mass per particle. The estimated absolute masses for the two size fractions were then extrapolated into g/m^2 (road dust), g/kg (runoff sediment), and g/L (runoff water). The advantage to have the estimated absolute masses is that they can be compared with previously published studies presenting their findings as concentrations (e.g., pyr-GC/MS).

4.7.2.1 Estimated absolute masses in road dust (g/m²)

For the fine fraction, 2–20 µm, tire and bitumen were counted together, hereafter called TBiWP (see chapter 4.2). The amount of TBiWP on the road surface varied between 21–94 g/m^2 (kerb) and 0.1–1.5 g/m^2 (in-between wheeltracks), Paper IV. For the mineral concentrations, the lowest values were found in the summer samples which confirms that studded tires generate more road wear than summer tires. Winter maintenance of the road surface such as sanding, and salting might also affect the mineral concentrations (i.e., sanding results in more minerals on the road surface and salting keep the road surface moist and thereby prevent fine particles to suspend into the air). For the coarse fraction (20–125 µm), the TWP content varies between 3–43 g/m^2 (kerb) and 0.02–0.2 g/m^2 (in-between wheeltracks). As can be seen in Figure 22, less particle mass was found in the coarse fraction (20–125 µm) than in the fine fraction (2–20 µm) of the road dust, especially in-between the wheeltracks (Figure 22). This is an important finding since the cut-off is a critical point and the finer fractions have often been excluded from previous works due to analytical difficulties. Particles accumulated at the road surface are available for transport into the stormwater system through runoff. However, the kerbside may act as a temporary sink for particles, and thereby prevent the particles to reach the stormwater. This needs to be further evaluated, and samples should be collected after heavy rain events to increase the knowledge about the transport mechanisms. Further, when traffic touches the kerb or drives close to the kerbside, it is possible that settled particles are released from the road dust sediment and thereby are available for further transport. One solution could be that increase the street sweeping, but focus on the area adjacent to the kerb, this needs further investigation and various sweeping strategies, brush techniques, and sweeping machines should be tested. Moreover, this result indicates that results from previous studies (including Paper I and II) might be underestimated. It is also clear that the majority of the particles accumulate adjacent to the kerb, and that the amount of particles found in the driving lanes is very low as also found in the urban studies (Paper I and II). This is also of importance to have in mind for upcoming field studies, models, estimations, and when it is discussed what measures to implement to reduce the amount of TWP and to prevent further transport of TWP. It is also important to have in mind that the total number of particles varies drastically from total mass and that the fine particles are easily transported further into the air and to the stormwater system (and recipients) while the coarse particles stay closer to the source to a greater extent. The fine particles can have a negative impact on health and the environment (e.g., bad air quality). All particle sizes can harm both health and the environment, but it is an urgent need that the size distribution and transport routes to be properly evaluated to increase the chance to implement the most efficient measures.

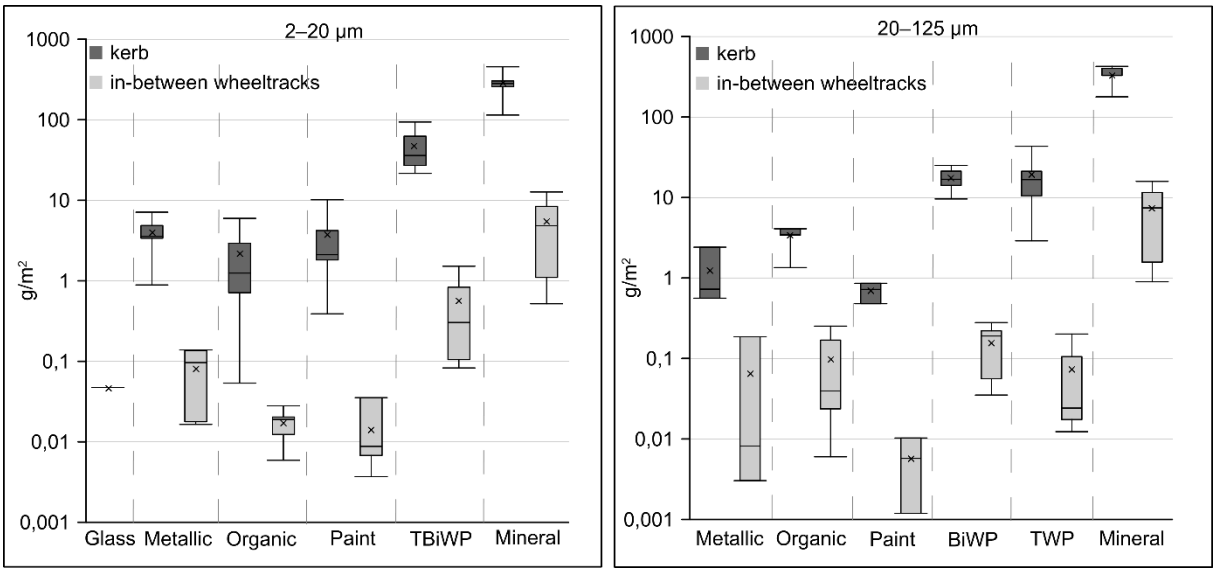

Figure 22. The results are divided into the different subclasses and presented as g/m^2 (estimated mass). The road dust samples were collected adjacent to the kerb and in-between the wheeltracks. The whiskers show the 90 and 10 percentiles, the line within the box is the median and the cross represents the mean value. Figure from Paper IV.

4.7.2.2 Relative number concentration (%)

Figure 23 shows boxplots from the samples collected at Testsite E18 (Paper IV) presented as the relative particle number concentration of the subclasses (%) analysed in the sediment (kg), road dust (g/m^2), and runoff (g/L). All runoff and sediment samples from the roadside gully pot and the stormwater well are presented together, called *stormwater* and *sediment*. The highest relative TWP number concentration was found in the stormwater, followed by the sediment, kerb, and in-between wheeltracks. The same pattern could be seen for both size fractions. Up to 47% of the particles analysed in the runoff samples (fine fraction) consist of TBiWP, while in the coarse fraction the relative number concentration of TWP varied between 24–38%.

Particles within the analysed size ranges seem to be transported from the road surface to the roadside gully pot, and further into the stormwater well, and the TBiWP seems to remain floating in the stormwater. This can be thought of as surprising since the density of TBiWP is higher than water, but during the sample preparation, it was evident that the particles behaved very hydrophobic. The hydrophobicity might explain why the particles remain floating longer than expected. Turbulence in the water, and particles making clusters which can increase the buoyancy might be another explanation. This is in contrast to the more expected particle size distribution (PSD) found in the wells (Figure 19 and Paper IV), where the particles found in the roadside gully pot in general were coarser than the particles in the stormwater well.

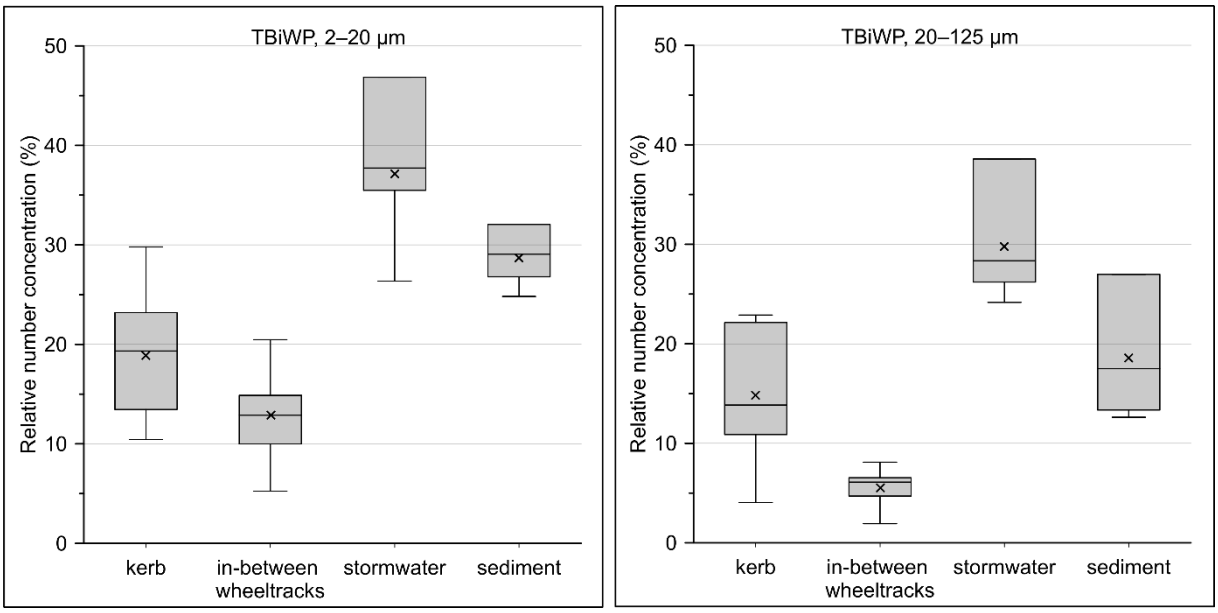

Figure 23. The relative number concentration (%) of samples collected from the different sample matrices are presented as boxplots. The whiskers show the 90 and 10 percentiles, the line within the box is the median and the cross represents the mean value. The kerb represents by 10 samples, in-between wheeltracks by 9 samples, for stormwater and sediment, 4 samples were analysed in each matrix.

4.7.2.3 Distribution of TBiWP in different environmental compartments

Figure 24 presents an updated version of Figure 6, where the main pathways for traffic-derived particles have been complemented by results from the field measurements at Testsite E18. The figure presents pie charts where both size fractions (2–20 µm and 20–125 µm) and TWP and BiWP have been counted together as TBiWP. The pie charts present the relative number concentration (%) (road dust and runoff) and relative mass concentration (air and deposition). Below each pie chart, the estimated absolute mass (average values) is shown, note that the absolute masses are presented with different units. Based on the relative number concentrations, most TBiWP were found in the water, roadside gully pot (38%), and stormwater well (34%), followed by sediment from the stormwater well (33%), sediment from the roadside gully pot (26%), road dust from the kerb (20%), road dust from in-between wheeltracks (12%), and air (4%). The same pattern was detected if the size fractions was separated (Paper IV). One possible explanation of the results is that the mineral content is higher closer to the source resulting in a lower concentration of TBiWP. The heavy mineral particles deposit closer to the source, resulting in an increased relative TBiWP content in other sample matrices. The estimated absolute masses showed that the stormwater sediment contained more TBiWP than sediment from the roadside gully pot, 112g/kg compared to 34g/kg. The opposite pattern was detected for the water, where the roadside gully pot water contained 0.3g/L compared to 0.03g/L that was found in the stormwater. Road dust collected at the kerb contained 85g/m^2 compared to 0.8g/m^2 in the in-between wheeltracks samples, Figure 24. One of the major findings in Paper IV was that the estimated absolute masses were higher in the fine fraction than in the coarse fraction if the two size fractions were separated both for the sediment and in the road dust. This is of importance especially since particle size has been a limiting factor in previous studies, and some articles exclude fine particles (<20 µm) from their analyses with the explanation that they do not contribute significantly to the total mass. However, the result from Paper IV shows the opposite, and forthcoming studies are encouraged to include even the fine particles and to perform particle size distributions to better understand the occurrence of TWP and TBiWP in each size interval.

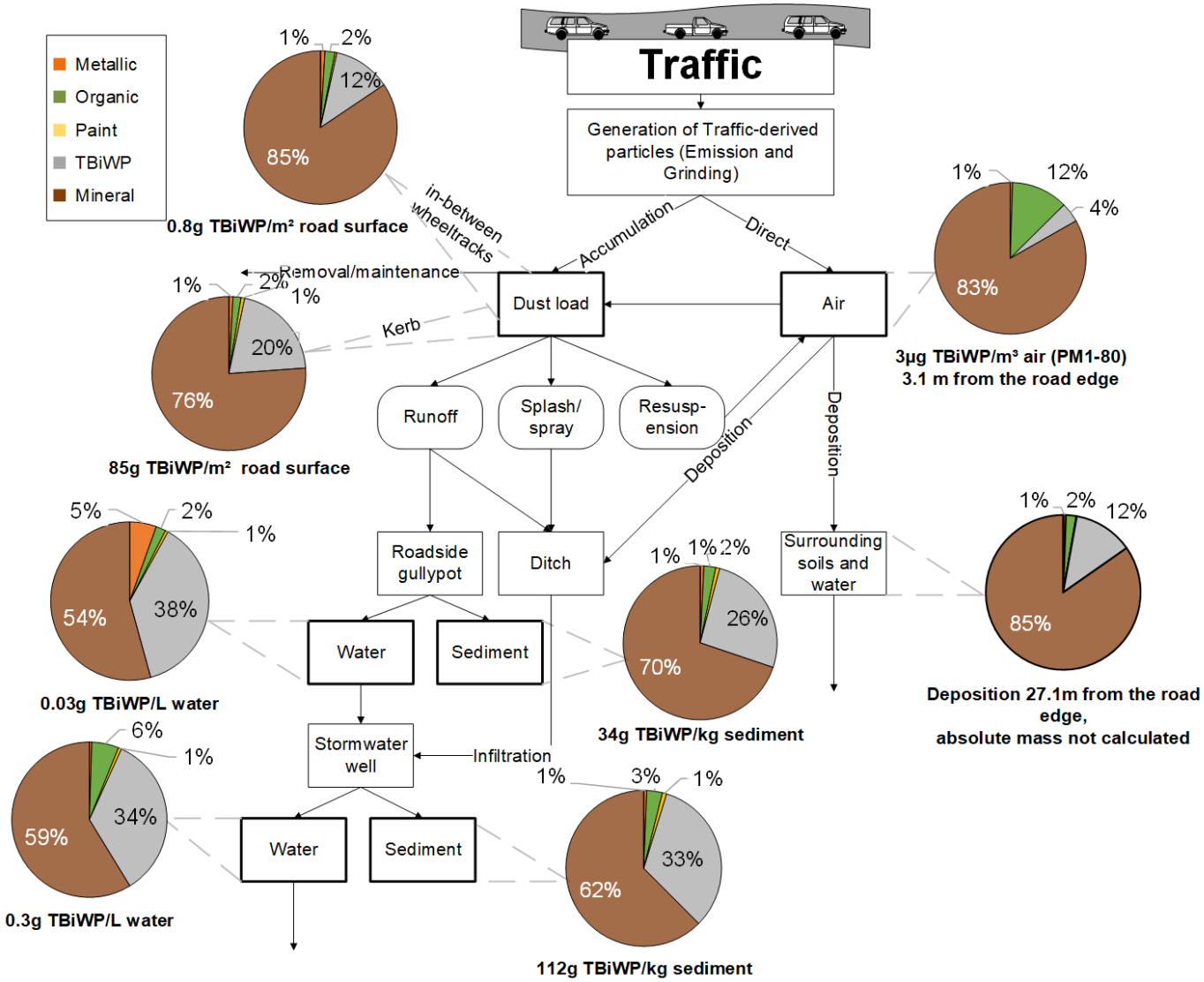

Figure 24. Simplified illustration of the transport routes of TBiWP, the bold boxes show the analysed sample matrices. The pie charts show the relative number concentration (%), and the masses below the pie charts are the result of the estimated absolute masses. NB! Different units for the sample matrices. Figure from Paper IV.

4.7.2.4 Results from paper IV in comparison to other studies

The estimated mass concentrations of TWP (only the coarse fraction, 20–125 µm) from Paper IV were compared with 25 max and min values from 14 field studies presenting TWP findings as concentrations (Amato et al., 2014; Eisentraut et al., 2018; Fauser et al., 1999; Fomba et al., 2018; Knight et al., 2020; Kumata et al., 2002; Mengistu et al., 2021; Ni et al., 2008; Panko et al., 2013; Panko et al., 2019; Parker-Jurd et al., 2021; Rausch et al., 2022; Rødland et al., 2022a; Schauer et al., 2002) (Figure 25). The result indicates that the estimations based on the findings from Paper IV are within the same order of magnitude as previously published studies. The results might not be directly comparable since the studies differ in sampling techniques, preparation steps, and analytical methods. Further, it was also difficult to identify the analytical size ranges, and information about *how* and *where* the samples were collected (e.g., sampling position at the road surface, information about meteorology, or AADT). However, even if the comparisons, as well as the estimations from the SEM/EDX results, come with uncertainties, the results can still be informative and indicate that the concentrations of TWP are similar independently of location, AADT, or road type.

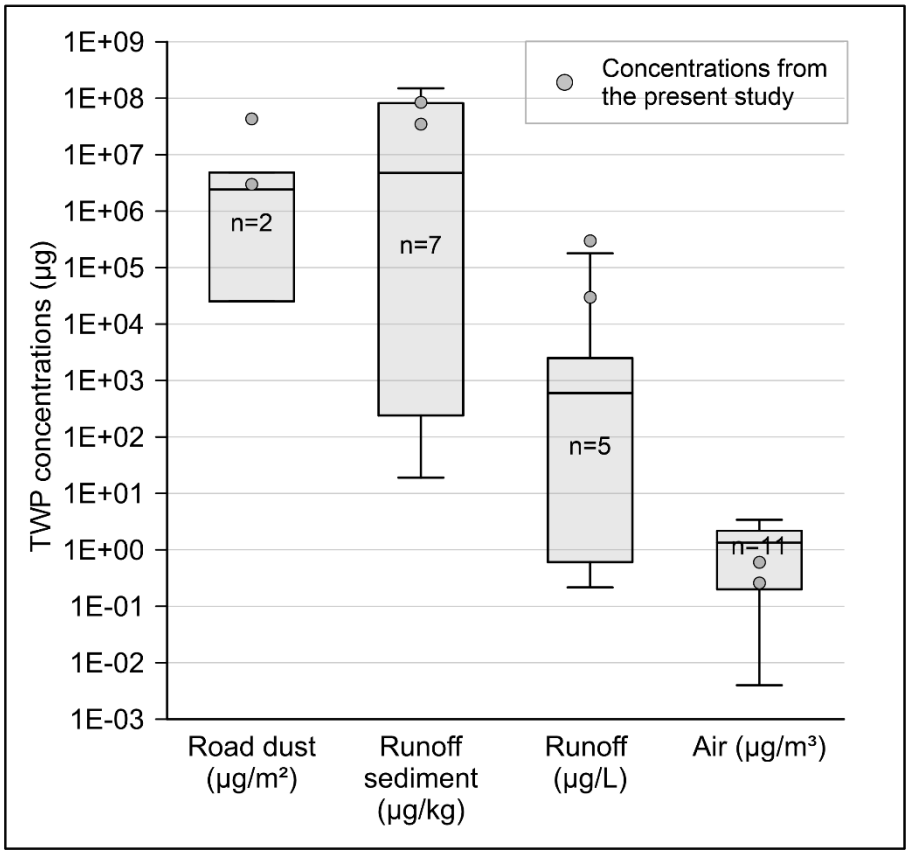

Figure 25. Comparison of TWP concentrations in the different environmental compartments obtained in this study and 14 previously published studies. The TWP concentrations from the present study (dots) have been compared with findings from the literature (boxplots). The number of values included in each box is named n=. Figure from Paper IV.

4.8 The results in relation to potential measures to prevent and reduce traffic-related microplastic emissions

Several measures could affect the emissions of traffic-related particles. For example, smooth driving, reduced speed, and less braking reduce the wear emissions. Small, light vehicles with smaller tires and low torque emit less TWP than heavier vehicles with a high torque. Further, correct wheel pressure and wheel alignments will also help to reduce the emissions (Johannesson and Lithner, 2022). However, these measures have not been evaluated within this project and cannot be related directly to the results from the present project. Another important change is the global transformation in the vehicle fleet with an increased number of electric vehicles, why the exhaust emissions decrease. Electric vehicles are heavier than conventional vehicles, due to heavy batteries and larger vehicles in general, which cause larger emissions of non-exhaust particles i.e., heavier vehicles generate more tire and road wear particles. Further, electric vehicles can accelerate faster and have an increased torque which might result in higher emissions of TWP and TRWP. The emissions of TRWP are not regulated and the emissions will probably continue to increase as long as the traffic volume increase (Beddows and Harrison, 2021).

4.8.1 Street sweeping

Street sweeping as a measure to reduce the transportation of fine, inhalable particles, PAH, and metals to surrounding recipient waters has been the focus of previously investigations and field studies (Amato et al., 2010; Bogacki et al., 2018; Norman and Johansson, 2006; Polukarova et al., 2020). Polukarova et al. (2020) conclude that street sweepers collect large amounts of particles, including nanoparticles and organic pollutants, indicating that street sweeping can be a good measure to reduce the amount of road dust and thereby likely also preventing the TWP to get further transported into the stormwater system if it is treated properly afterward. Recent compilations indicate that street sweeping may be a relevant measure to reduce the spreading of microplastics from urban streets (Vogelsang et al., 2019). The results from Paper I and II confirmed previous studies that street sweeping collects considerable amounts of road dust, it was also shown that the collected sweeping material (sweepsand and washwater) contained high concentrations of TWP, PAH, metals, and several organic compounds. However, the road surface and the stormwater did not exhibit a reduction in TWP concentration after sweeping, probably because the sweepers tend to collect the coarse particles and leave the fine particles in the cavities of the road surface. All particle sizes can harm both health and the environment, but it is an urgent need for the size distribution and transport routes to be properly evaluated to increase the chance to implement the most efficient measures. Therefore, street sweeping as a measure needs to be properly investigated. A suggestion is to only sweep adjacent to the kerb, and perform the sweeping more frequently, especially prior to heavy rain events. Another suggestion is to evaluate different types of street sweepers (e.g., sweepers with rotating brushes compared to vacuum sweepers) as well as different driving speeds to find an optimised sweeping routine. Another thing to have in mind is the fact that many street sweepers have brushes made of plastic. These brushes might be a source of microplastic emissions, and it could be a good idea to investigate that properly before frequent street sweeping is implemented as a measure to reduce the emissions of traffic-derived microplastics. However, without street sweeping, the concentrations of organic pollutants and MP might be even higher in the environment.

4.8.2 Different tire types

Paper V detected a variation between different tire types, e.g., summer, studless, and studded winter tires. The physicochemical properties a between brands were similar within a tire type, but a large differentiation between tire types was detected. The proportion between natural and synthetic rubber varied, as well as the concentration of carbon black and additives. The polymer content varied between TWP from the road simulator compared to tread samples from the tested tires indicating that simulator based TRWP or environmental TRWP should be used for upcoming ecotoxicology studies. The wear rate varied between tires from different brands, and between pavements. ADAC (2022) performed real-life tests with almost 100 summer, winter, and all-season tire models from 15 manufacturers and found a large variation between different brands. One positive result was that the best tires performed well both regarding safety and tire abrasion. The tire wear varied between 59–171 g/vkm (average 95–136 g/vkm), the lowest tire abrasion was generated from smaller summer tires (i.e., small vehicles) and the highest values were from large winter tires (heavy vehicles) (ADAC, 2022). Paper V evaluated tires from three well-known manufacturers. For upcoming studies, it had been of interest to include more tires from all price classes as well as include both pristine and used tires.

4.8.3 Tire labeling

There is an ongoing discussion in Europe about regulations for the wear rate of tires (treadwear grade), and several initiatives (both from the academia and the tire industry) tries to find a suitable method to test, measure, and evaluate the wear rate. The European Commission, the European Tire and Rim Technical Organization (ETRTO), and the TRWP platform discuss the possibility to include a "wear label" on tires, in addition to the existing labels (wet grip, rolling resistance, and noise). Rolling resistance and wear rate stands often in conflict against each other and traffic safety (i.e., wet grip and friction) is generally ranked higher than noise and wear rate by the society. However, a high friction might generate higher TRWP emissions that can have negative effects on environment and health (Trudsø et al., 2022). This can be worth having in mind, and before a label can be implemented, the risks with TWP must be properly evaluated. However, a label might result in greater awareness about TWP, and a tire with a slow wear rate will probably have a longer lifespan which is an advantage both from an economic and resource perspective. Moreover, a new regulation titled *Measures aiming to reduce the presence in the environment of unintentionally released microplastics from tires, textiles, and plastic pellets* is under development in the European Commission. One of the intentions with the regulation is to develop labelling standardization, certification, and regulatory measures to prevent and reduce the release of microplastics in the environment (EC, 2022). Further, another solution, as a contrary to restrictive regulations could be that the tire industry become more transparent regarding their tire recipes. If the additives and tire specific ingredients continues to be a secret, it is difficult to perform realistic risk assessments.

4.9 How to handle TWP as a potential risk?

Based on the existing data found in literature, and the data conducted within this project, it was not possible to perform a proper risk analysis. More studies are needed, both field studies to investigate the concentrations of TWP in different sample matrices and traffic environments on a global scale, but also more ecotoxicological studies, models/estimations about mass flows, and lifecycle analyses. Previous studies have tried to assess the risk with TWP and TRWP, but they have also come to the conclusion that more data is needed before a proper risk analysis can be performed (Tamis et al., 2021; Wik and Dave., 2009).

It is also important to aim to answer research questions and use methods adapted for the local traffic environment to make sure to measure the "right thing" meaning that, it is not appropriate to investigate the occurrence of polymer-modified bitumen on a road with conventional bitumen or examine the wear-rate of studded tires in southern Sweden where the majority have studless tires. Otherwise, it is a risk that the study become deceptive, and the results will not be comparable.

Until that the risk with traffic-derived microplastics is properly evaluated, the emissions should, due to their potential ecotoxicity, be assumed to have a negative impact on the environment. Therefore, it is recommended to follow the precautionary principle and thereby work preventative and implement measures to reduce the emissions and prevent existing particles to reach the environment.

5 CONCLUSIONS

This thesis contributes to increasing the knowledge about the occurrence of traffic-derived microplastics in a variety of sample matrices within different traffic environments and in laboratory-generated TRWP. The work performed within this project resulted in novel information about the properties and characteristics of TWP from different tire types and brands. It could also be concluded that shape, polymer content, morphology, size, and properties vary drastically, between tire types but also between environmental TWP and TWP generated using a laboratory method (road simulator). Lastly, it has been shown that the fine fractions (2–20 µm) of TWP and bitumen correspond to more than 50w% of the estimated absolute mass (in the size range 2–125 µm) independently of the sample matrix. The results from this thesis are of importance from several aspects including environmental risk assessment, health effect assessments, and microplastic transport modeling. Hopefully, the work can inspire forthcoming studies to focus on transport routes, innovative measures to reduce and prevent TWP emissions and transport to recipients, and as reference material for other studies conducted in different parts of the world.

The main conclusions from the thesis are listed below:

- A universal method for an optimised sample preparation was not able to find. The sample preparation is dependent on the analytical method, but the less preparation the better. Especially since almost every step tends to result in a loss of particles. In general, xylene proved to be effective to remove bitumen from TRWP without affecting the TWP, and $ZnCl_2$ with a density of $1.55 g/cm^3$ proved to separate TWP from TRWP to a great extent (<90%), at least for TRWP generated in a road simulator

- It is of great importance to specify the aim, research questions, and hypotheses before choosing the analytical method for the analysis of TWP and TRWP. No single optimal method is available, and all methods have limitations. With a thermal method, valuable information about size, shape, and surface will be lost, SEM/EDX cannot identify specific polymers, and light microscopy is unable to distinguish between particles <20 µm. A combination of methods is preferable. The present work got the opportunity to apply a multi-method analytical strategy and thereby combine several analytical methods that provide complementary insight about the abundance and properties of TWP and TRWP.

- The number of particles could be a unit of interest, especially for coarser particles. For fine particles, billions of TWP per volume or mass units used are found in all sample matrices, why it is not appropriate to present the results in absolute numbers. It is therefore preferable to use concentrations and/or relative proportions, at least for comparisons between matrices and/or samples. It is also important to conclude what unit is relevant for the study and clarify if the aim is to specify the total number of particles in a certain size, or if it is more relevant to get the total TWP concentration.

- It is important to have information about the particle size distribution within a sample. Without that, it is a risk that the results are underestimated, and that fine (or coarse) particles are excluded from the analyses.

- The TWP, both from laboratory and field samples, appeared in several different shapes and sizes. Also, properties such as density, surface structure, and brittleness varied. The conventional picture of a TWP as elastic, elongated, and with mineral encrustations has

been joined by TWP with fragile, soft, or brittle properties and with more angular shapes. It has been discussed if the different types of TWP can be deduced to aging or degradation.

- Analysis of metals and organic pollutants showed that all sample matrices (stormwater, road dust, sweepsand, and washwater) from an urban area under reconstruction, contained high concentrations of both metals and OP. Further, a strong correlation was found between the sweeping material and the road dust, while a weak correlation was detected between stormwater and the road dust indicating that most of the metals and OP stay in the nearby road environment, and that the runoff has been transported over impermeable surfaces resulting in a potential loss of OP and metals, OP and metals can also sediment with coarser particles and the most volatile OP can evaporate resulting in a change in OP composition.

- Street sweeping collects large quantities of road dust and could be an effective measure to reduce microplastics, metals, and organic pollutants. However, sweeping strategies, brush techniques, and different sweepers need to be properly evaluated before sweeping as a measure can be optimised.

- The material collected by the street sweeper (sweepsand and washwater) contains high concentrations of microplastics, metals, and organic pollutants. Therefore, the material should either be handled properly and treated prior to further use or placed in a landfill.

- One of the major advantages of the analytical methods used in Paper I–IV (light microscopy and automated SEM/EDX) compared to thermal methods is all the valuable information about the particle properties that was gained. A combination of particle size distribution, gravimetry, tactile and visual observation, and information about the elemental composition, shape, and particle surface can for example increase the knowledge about transport routes and degradation.

- The automated SEM/EDX coupled to a machine learning classifier enables differentiation between TWP, BiWP, and road markings (all being carbon-rich traffic-derived wear particles). The method showed high repeatability and an accurate classification into particle subclasses. Minerals were the largest particle subclass independent of the sample matrix and size fraction (>70%), followed by TWP and bitumen (ca. 20%). The major limitation with the automated SEM/EDX is that no other microplastics are detected and quantified, and that the method, with the present settings, is not suitable for analysis of coarser particles >125 µm.

- The estimated absolute masses indicated that the fine fraction (2–20 µm) corresponds to more than 50 % by weight of the TBiWP in the samples from all sample matrices. This result is of great importance and clearly shows that the fine particles (<20 µm) cannot be neglected, and it should rather be prioritized in forthcoming studies. Further, fine particles are easily transported with air and runoff, and they can also be difficult to capture in wastewater treatment plants. The particle mass decreased with an increased distance from the source (i.e., the road surface), however, with time, the accumulated loads of traffic-derived microplastics might be high also at distant recipients.

- When the estimated absolute masses were compared with previously published studies conducted in highly trafficked areas, the concentrations were in the same size range

indicating that the calculation based on the results from the gravimetry and PSD, in combination with fixed shapes, and densities seem to result in realistic absolute masses.

- Large amounts of road dust are transported through runoff into the stormwater system and confirm previous findings that the stormwater is an important transport path for traffic-derived microplastics.

- Two different types of TWP were detected in the road simulator samples, *firm-elastic* and *sub-elastic*. The properties and the physical characteristics differed (density, shape, size, firmness, fragility), but the analysed ratios of polyisoprene and polybutadiene were similar indicating that both particle types were related to TWP. A concrete pavement generated less *sub-elastic* TWP than an asphalt pavement indicating that the road surface can affect the generation and emissions of TWP.

- The road simulator samples evidenced that the road surface (asphalt compared to concrete) strongly affects the generation of TWP, which can be of interest for forthcoming infrastructure planning.

- Studded tires generate more TRWP than summer tires and studless winter tires. Particles from studded tires are finer than summer and studless tires which can have a negative impact on the health and the environment. In contradiction, larger amounts of TWP were detected during the periods without studded tires (e.g., due to rough road surfaces). In general, studded tires cause immensely more road wear than studless tires. Road wear is a strong source to PM_{10} and adds to particle air pollution. The polymer ratio varied between tire types (summer tires, studless winter tires, and studded winter tires), which further complicates the emission models.

6 FUTURE WORK

Recommendations and suggestions for future work based on the results from this PhD project are as follows:

- The vehicle weight increases which might result in increased TWP emissions during the upcoming years. Electric vehicles reduce exhaust emissions but tend to generate more TWP due to increased weight, heavy torque, and heavy batteries. However, there is a chance to reduce the TWP emissions with smoother driving and less braking (Eco-Driving). Upcoming studies should evaluate the actual wear rate of electric vehicles compared to conventional cars.

- When the particles reach the stormwater, size, density, and water flow are important factors if the particles sink or remain floating. A minority of the stormwater in Sweden is treated before release to water courses. Sedimentation ponds, filters in the roadside gully pots, bioretention filters, and rain gardens can be efficient traps for TWP in urban areas. This needs to be further investigated.

- Paper II analysed metals and organic pollutants in addition to microplastics. In all sample matrices, the metal and PAH concentrations were above the national and global guidelines. This is also an important finding since microplastics, metals, and organic pollutants have been well investigated on their own, but seldom in parallel.

- Street sweeping is a potential measure to prevent the TWP to get further transported into the environment. The effectiveness should be properly evaluated, and different sweeping techniques need to be tested, and the use of plastic brushes should be taken into consideration.

- Two different types of TWP were detected in the road simulator dust (Paper V), *sub-elastic* and *firm-elastic*. The finding raised several questions about the eventual occurrence of *sub-elastic* TWP in the environment, and if both TWP are present, do they behave differently in the environment regarding weathering resistance, and environmental fate?

- The road surfaces that were sampled and investigated during this project are all relatively new. It had been of interest to investigate the occurrence of traffic-derived microplastics on old roads, both concerning for construction and the age of the pavement.

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