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LEON-T

Low particle Emissions and lOw Noise Tyres

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Revision History

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Contents

Abbreviations and Units

A2 / A27 / A28	Highways in The Netherlands
A61	Highway in Germany
AADT	Annual average daily traffic
ACTRIS	Aerosol, Clouds and Trace Gases Research Infrastructure
APS	Aerodynamic Particle Sizer
BAM	Bundesanstalt für Materialforschung und -prüfung
BSE	Backscattered Electrons
BR	Polybutadiene
CEDR	Conference of European Directors of Roads
CL	Cathodoluminescence
CPC	Condensation Particle Counters
DCMR	DCMR Milieudienst Rijnmond
DP	Dipentene
E18	Highway in Sweden
EC	External calibration
EDX(MA)	Energy-dispersive X-ray spectroscopy (microanalysis)
EMEP	Co-operative Programme for Monitoring and Evaluation of the Long-range Transport of Air Pollutants in Europe
GC	Gas chromatograph
GCMS	Gas Chromatography-Mass Spectrometry
GC-MSMS	Gas Chromatography with tandem mass spectrometry
ISO	International Organization for Standardization
KNMI	Dutch Royal Meteorological Institute
Leon-T	Low particle Emissions and lOw Noise Tyres
LML	Landelijk Meetnet Luchtkwaliteit
MPs	Microplastics
MPS	Multi-purpose Sampler
N210	Provincial road in The Netherlands
NR	Natural rubber
PET	Polyethylene terephthalate
PM	Particulate matter
PM10	Particulate matter with an upper limit of 10 µm
PM2.5	Particulate matter with an upper limit of 2.5 µm
PM1	Particulate matter with an upper limit of 1 µm
PMB	polymer-modified bitumen
PS	Polystyrene
PSD	Particle size distribution
RWS	Ministerie van Infrastructuur en Waterstaat
SA	Standard addition
SB	Cyclohexenylbenzene
SBB	Phenyl-[4.4.0]bicyclodecene
SBR	Styrene-butadiene rubber

SEM	Scanning electron microscope
SEM-EDX-CL	Scanning electron microscopy with energy-dispersice x-ray spectroscopy and cathodoluminescence
Si	Silicon
SMA	Stone Mastic Asphalt
SMPS	Scanning Mobility Particle Sizer
TDU	Thermal Desorption Unit
TED-GCMS	Thermal Extraction and Desorption combined with Gas Chromatography-Mass Spectrometry
TGA	Thermogravimetric analysis
TOHA	9-tetradecyl-1,2,3,4,5,6,7,8-octahydro anthracene
TSP	Total Suspended Particles
TSS	Total Suspended Solids
TRWPs	Tyre and road wear particles
TWPs	Tyre wear particles
4VCH	4-vinylcyclohexene
VSCC	very sharp cut cyclone
VTI	The Swedish National Road and Transport Research Institute
ZOAB	Zeer Open Asfalt Beton
µ-FTIR	Micro-Fourier Transform Infrared Spectroscopy

1 Introduction

1.1 Background

In the past decade, microplastics have drawn much attention. They have been found in every environment of the world, including Antarctica, the Alps and all the world's oceans (Avio et al., 2017; Bergmann et al., 2019; Bessa et al., 2019; Waller et al., 2017). Tyre wear particles (TWPs) are estimated to be the largest source of microplastics (MPs) emission (Schwarz et al., 2023; A. J. Verschoor et al., 2017; A. J. Verschoor & de Valk, 2017), and compared to other sources of microplastics they could potentially account for more than 90% of all MP emissions (Koelmans et al., 2022; Rutgers et al., 2022). TWPs are also referred to as tyre and road wear particles (TRWPs) when encountered in the environment due to ubiquitous road wear encrustations they contain. In the future TWP emissions are also expected to grow with the increasing use of electric cars, which are heavier and have high instant torque (Jan Kole et al., 2017). Although little is reported on the health effects of TWPs specifically, it has been shown that particulate matter (PM) increases hospital admissions for cardiovascular and respiratory diseases (Makar et al., 2017).

In the last years, numerous research activities have started on quantifying and tracing TWP and microplastics in the environment (Koelmans et al., 2017; J. Panko et al., 2019; Unice et al., 2019a; A. Verschoor et al., 2016). This is done by monitoring studies using different measurement techniques, with varying specificity to TWP. For instance, based on historically reported values, the contribution of TWPs to ambient PM10 and PM2.5 is estimated to vary between 0.8-8.5% and 1-10% by mass, respectively (J. Panko et al., 2018). The broad range is mainly caused by the differences in chemical markers used (i.e. zinc, benzothiazoles, pyrolysis). Limited monitoring data in water bodies and in the ground is available and the majority of data lacks quality, implying a significant need to improve quality of sampling, sample treatment and final detection (Koelmans et al., 2019). Besides monitoring studies, alternatively, studies on release of TWP to the environment (Sieber et al., 2020) and further environmental fate modelling (Unice et al., 2019b) of TWP as well as microplastics in general (Koelmans et al., 2017; Quik et al., 2023) have been conducted. However, none of these models have been truly validated with real-world monitoring data. Although, recently high quality quantitative data was reported on TWP in runoff, soil, waterbodies (Dröge & Tromp, 2019; Min. I&W, 2023; Tamis et al., 2021) and in air (J. Panko et al., 2019), more high quality field data (more locations & environmental matrices, longer periods, TWP specific mass/number concentration, sizes and densities) is needed (Dröge & Tromp, 2019; Jekel & TU Berlin, 2019).

For environmental fate and behaviour, photo- and biodegradation processes play a central role but reliable quantitative data on degradation rates is almost non-existent (Baensch-Baltruschat et al., 2020). In environmental compartments biodegradation processes are expected to be slower (Jekel & TU Berlin, 2019) compared to changes in chemical composition, leaching and fragmentation of TWP into smaller particles due to aging and weathering. The physical and chemical changes of TWP will also influence processes like soil-fixation, resuspension and wash-off and therefore determine the fate of TWP in road-side soils and further long range transport via waterbodies. The transport of TWP via runoff and drainage systems are the primary source for aquatic emissions (Unice et al., 2019a). The knowledge on particle sizes, particle morphology and the effective density in runoff and drainage systems is not known yet, but are major determining factors for the further fate of TWP in aquatic systems (Jekel & TU Berlin, 2019). The material flow between pathways is highly variable and depends on location, rainfall events and the rural and urban drainage design. Also in waterbodies (rivers, lakes, estuaries and oceans) the properties of TWP, such as size and density, are important parameters for calculating settling rates to aquatic sediments and consequently determining the fate of TWP (Unice et al., 2019b). Estimates for TWP emissions into estuaries and oceans are limited and not yet validated due to complex analytical issues.

1.2 Objective

In the LEON-T project, we aim to address the lack of real-world measurements and provide much-needed quality data on which emission and fate models can be based. In previous work (P. Tromp et al., 2022) within the LEON-T project (D3.1) the different methods of physicochemical characterisation were assessed, and a workflow was developed to be suitable for the analysis of TWPs in environmental samples. The current report focusses on the real-world measurements of TWPs in various environmental compartments on sampling locations with different vehicle behaviour (i.e. highway, urban street) to gain insight into the emission, dispersion and fate of TWP in the environment. For this objective, several measurements and collection methods are employed in air, deposited dust, water, sediment, soil and runoff at varying distances from the road and are spread out over The Netherlands, along with a sampling location in both Germany and Sweden. Also, several sampling points on the river Rhine and Meuse in the Netherlands are selected for additional measurements in waterbodies and sediments.

2 Measurement strategy

The main goal of the measurements within the LEON-T project is to provide high quality data on concentrations and particle sizes of TWPs in various environmental compartments, such as air, water, soil and sediments and to assess the dispersion of TWPs across these compartments. For this objective several measurement campaigns at roadside locations and in waterbodies were organized. For the collection and analysis of environmental samples the methodology discussed in the LEON-T Deliverable 3.1 (P. Tromp et al., 2022) has been used. More details on the sampling and measurement sites are described below and in Section 2.2 and 2.3. The obtained results serve as input and validation for the emission and fate model which will be developed and employed within LEON-T by RIVM (Deliverables 3.3 & 3.4). To provide quality data, unambiguous identification and quantification of TWP in the environmental samples is employed with TGA-TED-GCMSMS using two different quantification techniques: the TNO-method, based on the composition of real (used) tyres and the BAM/ISO-method, based on the determination of rubber. Additional selective SEM-EDX single particle analysis were performed for information on morphology and particle density of TWP. Particle size distributions were obtained via wet cascade sieving (25 – 1000µm) and subsequent cascade filtration (1 – 25µm). Details on pre-treatment and analytical techniques are described in section 2.4. An overview of the sampling periods for all locations is given in Appendix A.

2.1 Roadside locations

To assess the primary emission and dispersion of TWP into the environment, in the Netherlands three roadside locations with different traffic behaviour: traffic load, traffic speed and accelerating/decelerating behaviour, were selected (Figure 1). The first measurement location was at a rural background location (Cabauw), at the roadside and approximately 350 meters away from the provincial road N201, the second location was at a busy intersection within a city (Rotterdam), and the third location was near the A2 highway between Amsterdam and Utrecht (exit Breukelen). Additional selective measurements were performed near the A27/A28 traffic junction (Utrecht) for method verification and for insight into the annual variation of the emission of TWP at the roadside. The three main measurement sites were located in the vicinity of measuring stations of the national air quality monitoring network (LML: Landelijk Meetnet Luchtkwaliteit), to get additional information on general air quality parameters, such as PM10, PM2.5, elemental carbon (EC), nitrogen dioxide (NO2) and nitrogen oxide (NO). As the LEON-T project aims to assess the Europe wide emissions, runoff samples were collected near the E18 highway in Sweden, and additional soil samples collected within the Microproof project (Dröge & Tromp, 2019) near the E18 in Sweden and the A61 highway in Germany were used.

The measurement campaigns at the three main roadside locations have been coordinated in order to allow an objective comparison without the influence of weather conditions. In this way insight can be given on the influence of traffic behaviour on the release of TWP. Measurements campaigns for the location Cabauw and Rotterdam were carried out simultaneously from May to September 2022 (20 weeks). The campaign at the A2 highway started later, from August to October 2022 (11 weeks), but during two months the three campaigns were overlapping. At each road side locations, besides on-line particle and black carbon measurements, sampling of particulate matter (PM), atmospheric deposition, runoff and soil was carried out. Figure 1 displays the different measurement locations within the LEON-T project along with the associated measurements for each of the locations.

2.2 Waterbodies

The composition of river water at a sampling location is not constant over time. There are seasonal influences (i.e. rainfall), hydrological influences (i.e. flow profiles at high and low tide) and daily variation (due to inputs from different sources). To be able to compare TWP concentrations in river water and to assess the dispersion and fate of TWP in these rivers, all of these factors need to be evaluated. Above all, there is a variation caused by the sampling method as well. Therefore, a collaboration was setup with the Ministry of Infrastructure and Water Management (Rijkswaterstaat, RWS-CIV) to be able to use their samples. These samples consist of surface water (suspended matter), sediment and riverbank samples from the Dutch rivers the Rhine and Meuse and the river Delta and were collected during 2020 – 2021 as part of a pilot monitoring study of microplastics in river water (Min. I&W, 2023). Also a validation study was conducted within this project, on sampling techniques and methods. Representative suspended matter samples for the LEON-T project were collected with continuous flow centrifuges for 24 hours (appr. 30-40 m^3

water) and sedimentation boxes for several weeks (appr. 80 – 160 m^3 water). Together with these samples information was shared on suspended matter concentration (mg/L) and flowrates of the riverwater (m^3/s). In addition near the A2 and A27 surface water and sediment of the ditches along the highways were sampled.

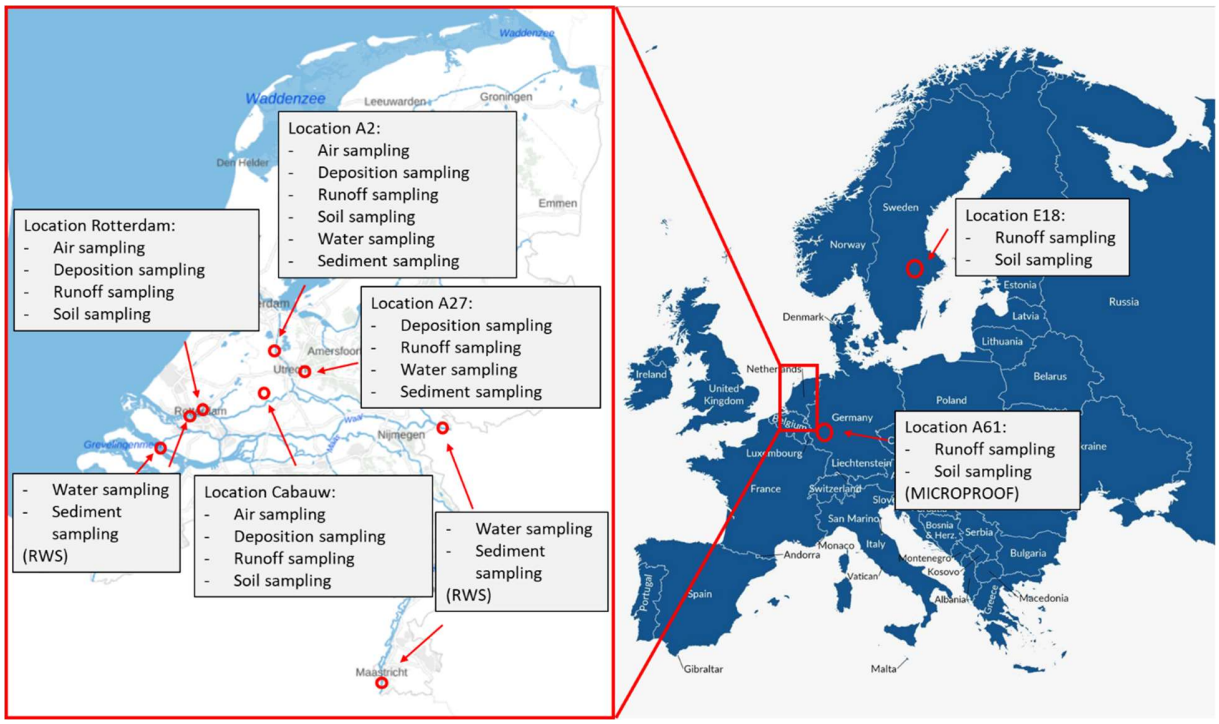

Figure 1 Sampling locations measurement campaign LEON-T. Water sampling was partially done by Rijkswaterstaat (RWS).

3 Sampling sites

3.1 Cabauw (rural background and provincial road)

The measurement location at Cabauw serves as a rural background location of the national air quality monitoring network (LML). The site is located near a measuring tower of the Dutch Royal Meteorological institute (KNMI), and the location has been used in several other studies such as the Co-operative Programme for Monitoring and Evaluation of the Long-range Transport of Air Pollutants in Europe (EMEP) (www.emep.int) and the Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS) (www.actris.eu). Air, soil and atmospheric deposition measurements took place near the KNMI tower, at approximately 350 m from the road. In addition to the background measurements near the KNMI tower, some measurements were also performed near the main provincial road (N210, M.A. Reinaldaweg) in the area at 1.5m from the road, as illustrated in Figure 2.

The N210 is a provincial road with 2 lanes and speed limit of 80 km/h. The traffic intensity was approximately 5.100 movements per weekday in 2022 (1 direction), of which heavy vehicles made up approximately 10% (4% heavy freight traffic and 6% medium freight traffic). The road type is closed asphalt, which has a limited effect on the dispersion of particles. The surrounding area consist of trees and agricultural grounds, with a ditch running along the road at approximately 7 m distance. The dominant wind direction is south – southwest: crosswise to the road. The runoff is infiltrated in the road shoulder and the embankments, and partially in a storm drain, from where it is either deposited in nearby water bodies or transported to the wastewater treatment plant through the sewage system.

Runoff sampling took place in one of the storm drains close to the bus stop. The storm drain was present at the bus stop, which was located at approximately 2 m from the road. Deposition sampling took place at approximately 1.5 meter from the road. Soil sampling took place at 1 m and 5 m from the road. At each distance from the road 5 individual soil samples were taken with a mutual distance of 20 m.

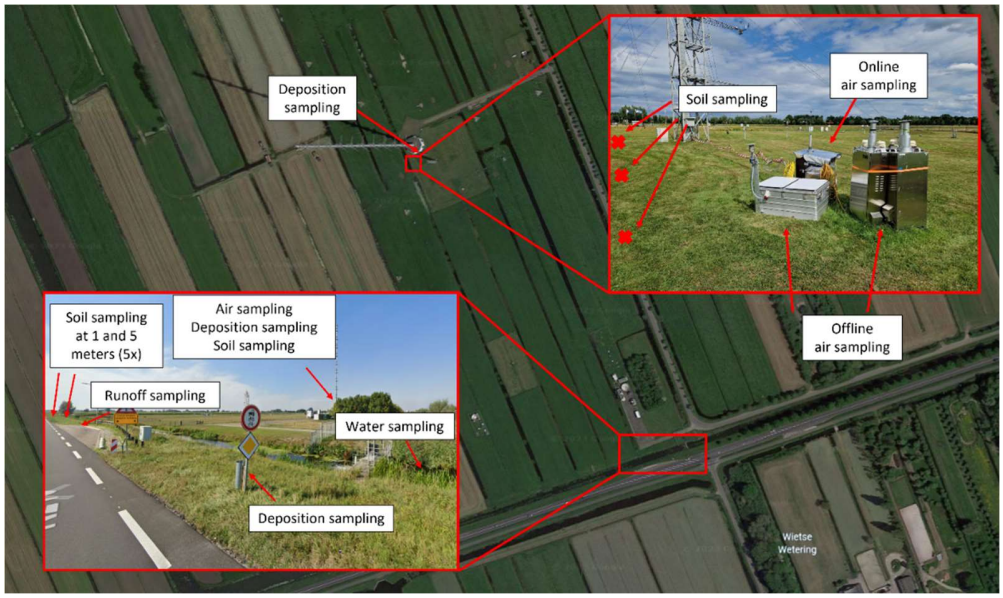

Figure 2 Sampling locations along the N210 M.A. Reinaldaweg. Coordinates: 51.970073N, 4.926620E (based on satellite imagery from Google maps)

3.2 A2 (highway)

The measurements near the highway A2 took place near the measurement station 'A2-Breukelen' used in the national air quality monitoring network (LML). The location is situated along the A2 highway, running North-South between Amsterdam and Utrecht, near the exit Breukelen. The A2 highway is a busy highway with five lanes in each direction (ten total), along with an emergency lane. The maximum speed is 100 km/h during the day (06:00-19:00) and 130

km/h during the night, and the traffic intensity in 2022 was approximately 85600 vehicles per weekday in 2022 (1 direction), of which heavy vehicles made up approximately 13% (6% heavy freight traffic and 7% medium freight traffic). The asphalt on the A2 consists of ZOAB (Zeer Open Asfalt Beton), which is a porous type of asphalt. ZOAB potentially reduces the amount of TWP that could reach the roadside. Figure 3 shows the A2 location, along with the sampling setup at the location. Along the highway runs a guardrail, and the highway is slightly higher than the surrounding area, which is mostly flat, with few obstacles. The dominant wind direction is west – southwest: crosswise to the road. The runoff is infiltrated in the road shoulder and the embankments.

PM, deposited dust and soil samples were collected at several distances from the road. As the A2 has an emergency lane, the distance from regularly used traffic lanes was approximately 7 m. PM samples were collected at 7 and 18 m, deposited dust at 7, 20 and 50 m and soil at 7, 15 and 30 m from the first (nearest) driving lane. At each distance from the road 5 individual soil samples were taken with a mutual distance of 20 m.

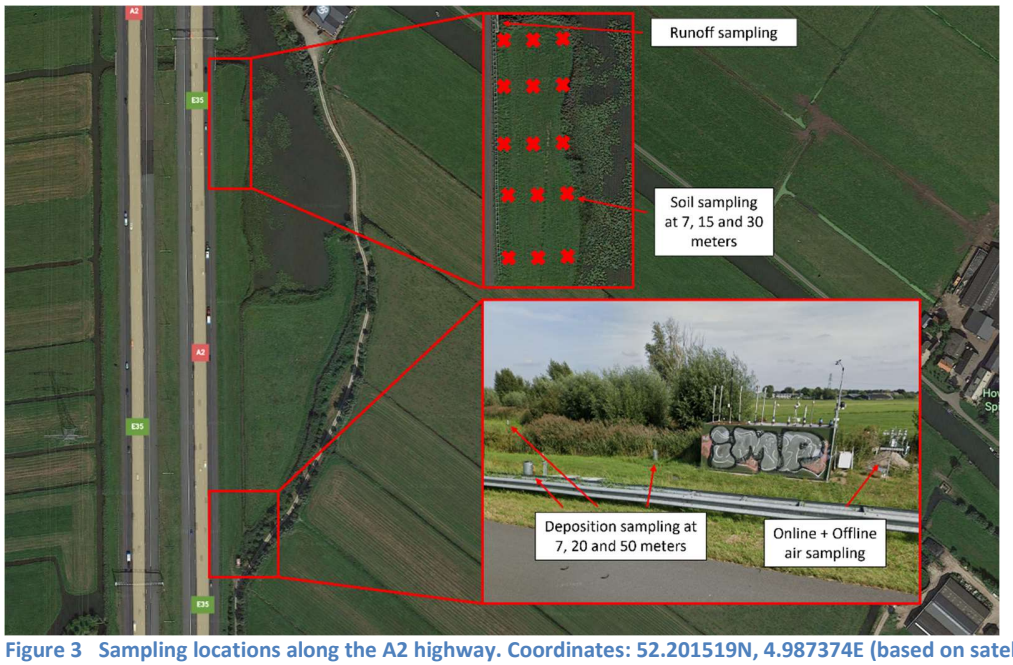

Figure 3 Sampling locations along the A2 highway. Coordinates: 52.201519N, 4.987374E (based on satellite imagery from Google maps)

3.3 Rotterdam (city intersection)

The measurement location in Rotterdam was close to a busy intersection along the Statenweg, near Bentinckplein. This location is also used in the national air quality monitoring network (LML). The Statenweg is a busy road in the city of Rotterdam, with two lanes in both directions (four total) with approximately 16700 vehicles per weekday in 2022 (1 direction), of which approximately 6% is made up of heavy vehicles (1% heavy freight traffic and 5% medium freight traffic). The maximum speed is 50 km/h. There are several trees and bushes in proximity to the road. The dominant wind direction is south-southwest, meaning that the measurement location is downwind from the intersection. The runoff runs towards a storm drain, from where it is either deposited in nearby waterbodies, or transported via the sewage system to a wastewater treatment plant.

The runoff was collected in two storm drains close to the site where the LML location is situated. The deposition sampling took place at approximately 1 m from the road. The soil samples were collected at 1 and 5 m from the road; at each distance from the road 3 individual soil samples were taken with a mutual distance of 20 m. The soil was on nearly the same level as the road, but separated a curb, so runoff would not reach the soil directly, only splashwater.

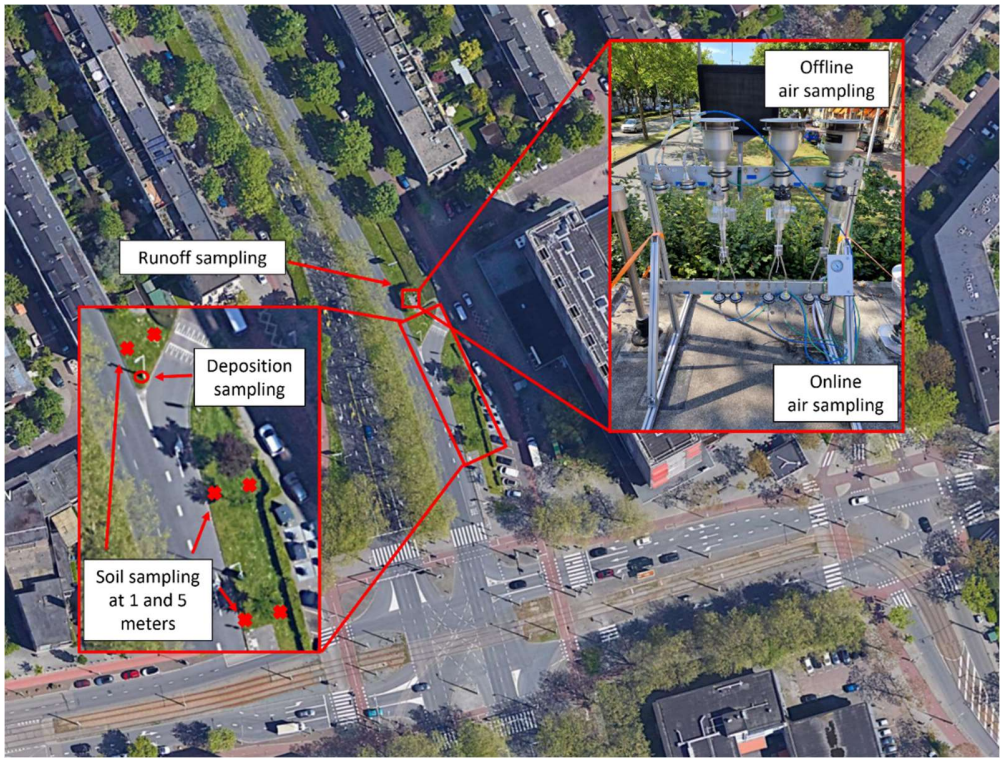

Figure 4 Sampling locations in Rotterdam. Coordinates: 51.927082N, 4.461398E (satellite imagery from Google maps)

3.4 A61 (highway, Germany)

In Germany, samples have been gathered of runoff and soil from the highway A61 between Kreuz Meckenheim and Dreieck Bad Neuenahr-Ahrweiler (at the parking place 'Goldene Meile'). These samples were collected on the 18th of March 2019 within the Microproof project (Dröge & Tromp, 2019) and were used for additional TWP particle size analysis. The A61 highway is a busy highway with 5 lanes in total, along with an emergency lane and with approximately 38500 vehicles per day in 2019, of which approximately 22% is made up of heavy vehicles. The maximum speed is 130 km/h. The road consists of regular asphalt, and the surrounding area is flat, with some bushes in the area. The dominant wind direction is crosswise to the road and there is no collection or treatment system present for the runoff, which is therefore infiltrated in the soil on the road shoulder and the embankments.

3.5 E18 (highway, Sweden)

The samples from Sweden were collected at the VTI test site, located along the highway E18 in the central part of Sweden, shown in Figure 5. The E18 is a state road and one of the country's most trafficked, which constitutes an important route for passenger and freight transport between Oslo-Stockholm. The E18 has two lanes running in both directions (four total), and the road type consists of stone mastic asphalt with a maximum stone size of 11 mm (SMA11) and with polymer-modified bitumen (PMB) as the binder. The climate is mild with an annual mean temperature of 6 ◦C and a yearly precipitation of approximately 700 mm. The annual average daily traffic (AADT) is around 11100 in 2019 and 11900 in 2022, of which respectively 13% (2019) and 8% (2022) was made up of heavy vehicles. The speed limit is 120 km/h. The surrounding area is flat with no trees or bushes. The lanes heading from east to west and west to east are separated by a 10 m patch of green. The main wind direction is crosswise to the road. The runoff can infiltrate at the shoulder of the road but is also collected in a storm drain. The water and sediment samples were collected on the 6th of October 2022 from a well where runoff is collected, illustrated in Figure 5. This well is a storm water well which contains road runoff water. The well contains several measuring devices, as well as an overflow mechanism where large particles will settle, while clean water can flow out. Soil samples were collected on 14 June 2019 within the Microproof project (Dröge & Tromp, 2019) and were used for additional TWP particle size analysis.

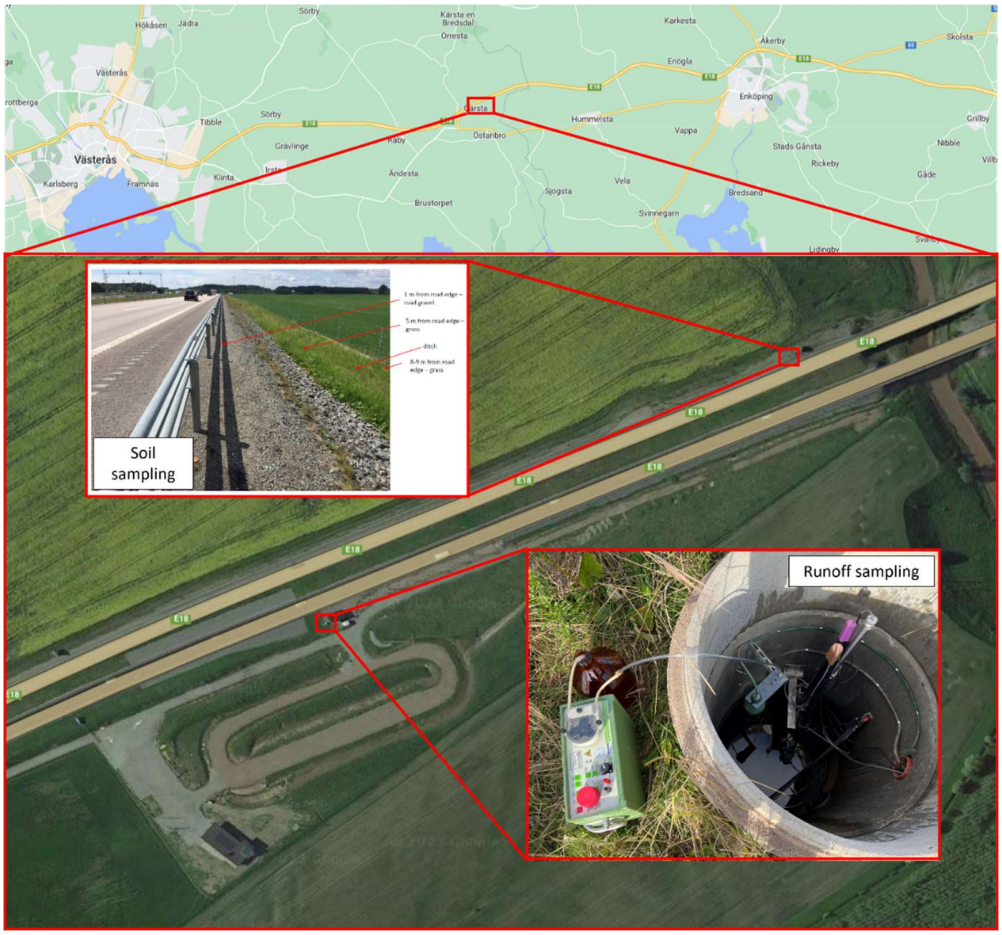

Figure 5 Sampling location at the E18 test site in Sweden. Coordinates: 59.633677N, 16.855227E. (Source: Google).

3.6 River Rhine, river Meuse and river Delta

Total suspended solid (TSS) and sediment samples were taken by Rijkswaterstaat at different locations on the river Rhine, Meuse and the river Delta between November 2020 and December 2021 (Min. I&W, 2023). In the river Rhine, near Lobith (the border between the Netherlands and Germany) and the river Meuse, near Eijsden (the border between the Netherlands and Belgium) TSS samples were collected using a continuous flow centrifuge system. The river Rhine is a large European river with an average flow of 2200 m^3 per second (near Lobith). The river Meuse is a European river with an average flow of 200 m^3 per second (near Eijsden). The Rhine-Meuse-Scheldt delta is a river delta in the Netherlands and Belgium formed by the confluence of the Rhine, Meuse and Scheldt. The result is a multitude of former islands and estuaries that converge in Zeeland, which forms the heart of the Delta. Most of the TSS samples were taken at the RDM dock in the New Meuse in Rotterdam with sedimentation boxes. Sediment samples were taken at different locations in de New Meuse, Old Meuse, Haringvliet and Hollands Diep. Yearly average characteristics of the Meuse and Rhine are presented in Table 1. For the new Meuse no yearly average characteristics are known; suspended dust concentrations are determined based on the sampling campaign at the RDM dock in 2021.

Table 1 Yearly average flow rate and suspended dust of the Meuse and Rhine

	Rhine	Meuse	New Meuse	
Flow rate	2350	230	-	m^3/s
	$7,4*10^{10}$	$7,3*10^{9}$	-	m^3/year
Suspended dust	20	15	2 – 12 *	mg/L
	$1,5*10^{9}$	$1,1*10^{8}$	-	kg/year

* min – max values from the sampling campaign at RDM dock in Rotterdam between March – September 2021

4 Sampling methods

Sampling techniques and methods for air (particulate matter), atmospheric deposition, soil, water, sediment and runoff, which were used during the sampling campaigns are described below. An overview of actual sampling times for each location and measurement is given in Appendix A.

4.1 Air

Air sampling took place from the 4th of May 2022 until the 21st of September for the location Cabauw and Rotterdam (20 weeks), while sampling at the A2 highway took place from the 27th of July until the 13th of October (11 weeks). For collection of PM in different size fractions size selective filter based techniques and multi-stage cascade impactors were used. The collected fractions were TSP (Total Suspended Particles), PM10, PM2.5 and PM1. The TSP fraction was collected using an open sampling head, with a protruding cylindrical head, which served to protect the filters from incoming rain. The PM10 fraction was collected using a PM10 sampling head (Anderson type 246B), while PM2.5 and PM1 were sampled with the same sampling head with a VSCC (very sharp cut cyclone) for further particle fractionation. With the use of a splitter the PM size fractions were simultaneously collected on 47 mm quartz (Whatman® QM-A quartz filters) and Teflon (Pall PTFE Membrane Disc Filters) filters, each at a flowrate of 8.35 L/min. The flowrate was kept constant by using a critical flow capillary. For analysis with SEM/EDX, PM was collected with 25mm open-face samplers on gold-coated polycarbonate filters (0.4 µm, 25 mm, Nuclepore), with a flowrate of 1 and 3 L/min. All sampling heads were mounted on an aluminium frame. A schematic overview of the offline filter collection is given in Figure 6.

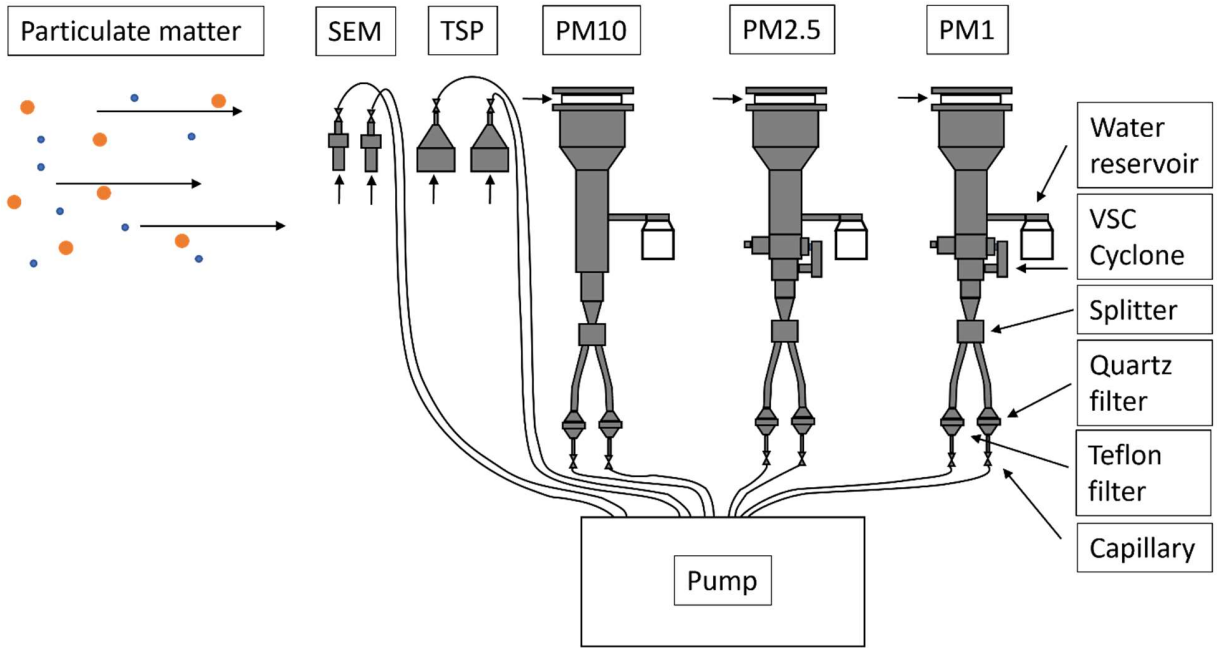

Figure 6 Schematic overview of the main offline sample collection on filters

For the rural background location (Cabauw), Leckels (SEQ47/50-RV) were used to achieve a higher flowrate. The general principle of the Leckels is similar to the other filter based measurements, but the Leckels allows for automatic filter exchanging, whilst only being able to collect PM on one filter at a time. The flowrate of the Leckels was 38 L/min and alternated between quartz and teflon filters each week. The sampling time of all filter based sampling devices was 1 week during the whole sampling campaign (20 week for Cabauw and Rotterdam, 11 weeks for A2 highway).

Additional size fractions smaller than PM1 were collected using two different multi stage cascade impactors, which were alternated between locations. Impactors use the principle of inertia to capture particles on collection plates. Particles enter through the sampling nozzle by an air flow guiding them towards a collection plate. After passing the nozzle, the particles enter a chamber where the flow of air is parallel to the collection plate. Larger particles will not

be able to make the turn, and impact on the collection plate. Smaller particles will be able to follow the flow of air past the first collection plate(s). By progressively decreasing the nozzle size, smaller particles will deposit in a later stage of the cascade impactor. This results in a separation of particles based on their aerodynamic diameter.

The DGI (Dekati Gravimetric Impactor) was employed at the rural background location (Cabauw) and the A2 highway location; the DLPI (Dekati Low Pressure Impactor) was used at the A2 highway location and the urban street location in Rotterdam. An overview of the two impactors and their 50% cut-off value can be found in Figure 7. The DGI is a high flow impactor with 5 stages, which was operated at 70 L/min, and allowed for the collection of size fractions <0.2-2.5 µm on 47mm quartz and teflon filters, with a 70 mm glass fibre (Pallflex Emfab) backup filter for the size fraction <0.2 µm. The DLPI impactor is a 13-stage impactor, which operates at 30 L/min, and allows for the collection of PM from <0.03-10 µm on 25 mm quartz and Teflon filters. The sampling time for the cascade impactors varied from 1 to 2 weeks, with a total sampling period of 4 weeks per location.

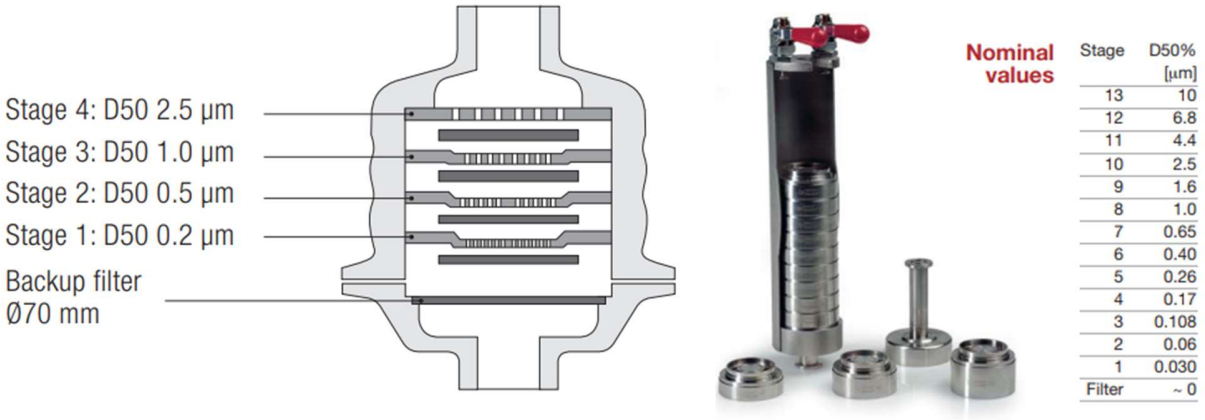

Figure 7 Cascade impactors. DGI (left) and DLPI (right). The D50 values indicate the 50% cut-off diameter (source: dekati.com)

In order to gain more insight into the hourly and daily fluctuations of PM in the air along with the corresponding size distribution, online measurements were performed with an Aerodynamic Particle Sizer (APS – Model 3321; TSI), a Scanning Mobility Particle Sizer (SMPS – Model 3082; TSI) and a black carbon monitor (Aethalometer – Model A33). The online measurement setup was deployed at all locations for approximately 4 weeks.

The APS enables real-time aerodynamic size measurements of particles between 0.5 and 20 µm with 32 size bins and operates at a sampling flow rate of 1 L/min with a time resolution of 1 second. The SMPS consists of a differential mobility analyser (DMA) covering a size range of 2-1000 nm and a Condensation Particle Counter (CPC 3775, TSI Inc., USA) with a 50% cut-off at 4 nm and a concentration range up to 10^4 particles per cm^3. The sampling flow rate was 2.5 L/min and the measurement scan was set at 10 – 430nm with 96 size bins, which takes about 3 min. With a DMA particles are classified according to their electrical mobility diameter (Brouwer et al., 2014). In addition to the particle counters, an aethalometer was used to measure the black carbon concentration at 7 wavelengths (350 – 950 nm). The measuring frequency was set at 10 seconds with a flow rate of 2 L/min.

4.2 Atmospheric deposition

The collection of deposited dust was performed with total (wet + dry) atmospheric deposition samplers. The deposition samplers consist of a glass funnel with a diameter of 25 cm connected to a 10L amber-coloured glass flask, which collect both dry and wet deposited dust. The funnel and flask were protected by an aluminium housing, which also protects the sample from direct sunlight. By collecting deposited dust, the total deposition rate of TWP in mg/m2/year is determined at several locations near and at further distances from the road Figure 8 shows the deposition sampler at Cabauw.

Figure 8 Total deposition sampler at Cabauw

The deposition samplers were located near two highways in The Netherlands (A2 at three distances from the road and A27), at a rural background location and at the roadside of a provincial road and a busy city road. The sampling period for each individual sample was approximately 1 month. Deposited dust samples were collected during the whole air sampling measurement campaign.

4.3 Road runoff

The collection of road runoff gives insight in the total amount and particle size distribution of TWP deposited onto the road surface. This material is transported to storm water wells, drainage wells and/or roadside soil during rainy periods. The runoff samples were collected in drainage wells (provincial road N201 in Cabauw, city road in Rotterdam) and on the edge of the road with funnels to guide the water into a 5L collection flask (highways A2 and A27). These runoff samplers were installed on the 16th of August 2022 at all four locations, during an extended period of dry weather (approximately 2 weeks), and were collected after a day of rain. This ensured a relatively high concentration of material in the runoff samples. Additional road runoff sampling was performed on the 6th of October 2022 in a storm water well at the test location at the E18 in Sweden. A grab sample was taken from sedimented material and the water layer above was sampled via a hose using a peristaltic pump.

4.4 Soil

Samples of the upper layer of the soil were taken by excavating carefully measured out sections of 10x10 cm of soil using a spade. The sampling depth was approximately 3-5 cm. Soil samples were taken at several distances from the road side. At each distance from the road 3 - 5 individual soil samples were taken with a mutual distance of 20 m and combined in a glass container. The collection of soil samples allows for the determination of TWPs in the top layer of the soil, which originate from road runoff, splash water and atmospheric deposition, and provides insight into the accumulation of TWP over the years, in mg/m^2.

4.5 Water and sediment

Sampling of total suspended solids (TSS) and sediment in rivers in the Netherlands was conducted by Rijkswaterstaat. In the river Rhine, near Lobith and the river Meuse, near Eijsden TSS samples were collected using a continuous flow centrifuge system with flow rate of 1000 – 2000 L/hour for 24 hours. In the New Meuse, TSS samples were taken at the RDM dock in Rotterdam with sedimentation boxes with a flow rate of 1 – 8 L/min for 2 – 4 weeks. The upper sediment layer was sampled at different locations in de New Meuse, Old Meuse, Haringvliet and Hollands Diep with sediment grabbers. Sampling of surface water and sediment in ditches along the A2 highway and the A27 highway was performed by TNO. Sampling of surface water was performed using a peristaltic pump with a hose, which sampled approximately 100 L of water 10 cm below the water surface. At the same locations sediment sampling was performed with a gouge. The top 5 cm of sediment was collected at several places in each ditch.

5 Analytical techniques

For the analysis of environmental samples from the measurement campaigns the methodology discussed in the LEON-T Deliverable 3.1 has been used (P. Tromp et al., 2022), without the separation techniques: digestion and density separation. The resulting workflow of this methodology is presented in Figure 9.

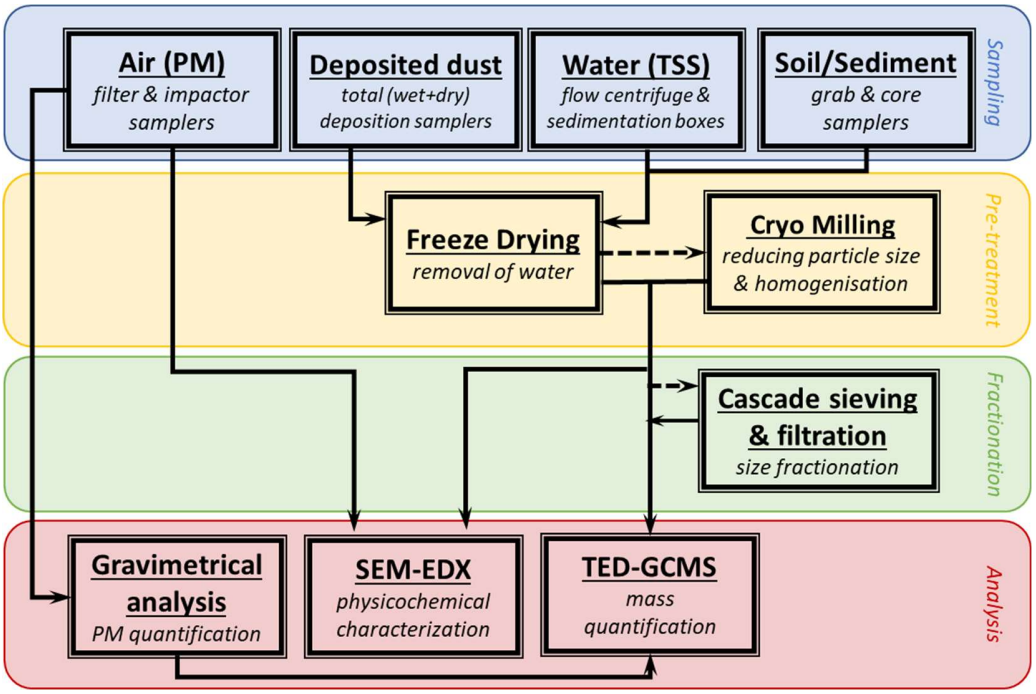

Figure 9: Flow diagram of the workflow for the sampling, pre-treatment, fractionation and analysis of environmental TWP samples.

5.1 Pre-treatment

Deposited dust, water (including total suspended solids), sediment and soil samples required some form of pre-treatment before analysis. The collected PM on filters and impactor stages were analysed directly without pre-treatment. The collected water samples from the ditches were fractionated without pre-treatment.

To remove the water, deposition, runoff, total suspended solid (TSS) and sediment samples were freeze-dried. First the 'wet' samples were divided over several 850 ml jars and were frozen at -40 °C for 24 h. The frozen samples were then transferred to a Lablyo Mini freeze dryer where they were kept under vacuum at - 50 °C until complete dryness. The remaining solids were transferred to glass vials and were weighed using a microbalance. Until fractionation or direct analyses the samples were stored in the freezer. The 'field moist' soil samples were dried at room temperature by spreading the material out onto large, stainless steel plates in a laminar flow cabinet and leaving them to dry for approximately 5 days. A part of the soil, sediment and TSS samples were cryogenically ground with a rotor mill to particle sizes below 0.25mm. The selected samples for fractionation were not cryogenically milled.

5.2 Fractionation

Fractionation took place in three steps: dry sieving with a sieve of 1 mm to remove coarse particles (1), a cascade wet sieving setup for particles of 25-1000 µm (2) and a cascade filtration setup for fine particles <25 µm (3). The wet sieving setup consists of a cascade of five sieves of 400, 200, 100, 50 and 25 µm. The cascade filtration setup consists of four 25 mm Swin-Lock Filter holders or Sartorius Stedim stainless steel filter holders stacked together. Pre-weighed track-etched membrane polycarbonate filters (Nuclepore and ipPORE) with pore sizes 10, 5, 2 and 0.8 µm were used

in the cascade filtration. The filter holders were placed in an ultrasonic bath (VWR ultrasonic cleaner USC-TH) to avoid clogging.

After dry sieving over 1mm and screening this coarse material for tyre tread particles under a microscope (5 – 60x), remaining material was brought into suspension with MiliQ and was sieved using the cascade sieving setup. The fractions 25-1000 µm were dried at room temperature in a laminar flow cabinet for 24 hours. A part of the remaining suspension with particles < 25 µm was used in the cascade filtration setup. Prior to cascade filtration, 1 drop of Tween20 (Sigma Aldrich) was added for every 25 mg of material. The solution was forced through the filter holders using a peristaltic pump, at an RPM of 60. Afterwards the setup was rinsed with an equal amount of MilliQ as the starting volume. After drying the filters at room temperature in a laminar flow cabinet for 24 hours the recovered material was determined gravimetrically using a Mettler Toledo AX205.

5.3 Gravimetrical analysis

The concentration of collected PM on Teflon filters is determined using gravimetric analysis in accordance with NEN-EN 12341 (NEN, 2014). The Teflon filters were weighed before and after sampling using a microbalance (Mettler Toledo AX205; ± 1 µg) to constant weight in a conditioned room (50% relative humidity, 20 °C).

5.4 TGA-TED-GCMS

To determine the mass concentration of TWP, TED-GCMS analysis was performed following the procedure of (Duemichen et al., 2019). The analysis is a two-step method. A sample is first decomposed in a thermogravimetric analyzer (TGA) and the gaseous decomposition products are then trapped on a solid-phase adsorber. Subsequently, the solid-phase adsorber is analyzed with thermal desorption gas chromatography mass spectrometry (TD-GC-MS). A representation of the used TGA-TDU-GCMSMS set-up is shown in Figure 10.

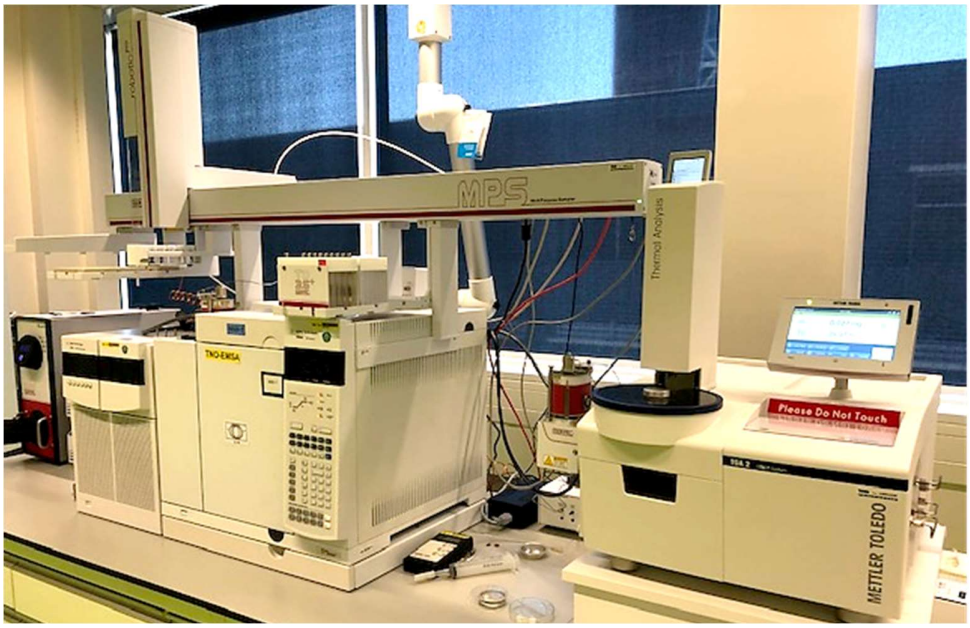

Figure 10: the TGA-TDU-GCMSMS system used for the analysis of TWP in environmental samples.

The measurements were carried out with a thermogravimetric furnace TGA 2 with an auto sampler (Mettler Toledo GmbH, Germany) using 150 or 900µL crucibles and a purge gas flow (N2) of 30 ml min-1. The following temperature program was used: 35°C (2min), 35-290°C (20°C/min), 290°C (2min), 290-550°C (20°C/min), 550°C (2min), 550-1000°C (50°C/min), 1000°C (5min). Between 35-290°C the released gaseous products were discarded and only between 290-550°C the decomposition products were sampled. Between 550-1000°C the purge gas was synthetic air (50ml/min) to burn residual carbon (mainly elemental carbon) in the crucibles and to clean the TGA system.

The TGA is coupled to a thermal desorption unit (TDU 3.5, Gerstel, Germany) via a heated transfer line (250°C), a heated coupling (130°C) and a TAU interface that is cooled to 25°C. Tenax TA 100mg (Industry standard; length 3.5inch 1/4inch diameter) was used as solid-phase adsorbent agent. A 7000 GC/MS Triple Quad (Agilent Technologies, United States) was used to analyse the decomposition products. The GC was equipped with a cooled injection system (CIS 4, Gerstel) and a TDU 2 unit (Gerstel). The sample robot for the transfer of the thermal desorption tubes from the TGA to the TDU was designed with a MultiPurpose Sampler (MPS, Gerstel). The thermal desorption was carried out in solvent vent mode with a total He flow of 25 ml min-1. The sample was thermally desorbed with a temperature program of 50 to 280 °C at a heating rate of 60 °C min-1 and hold time at 280 °C for 10 minutes. The compounds were trapped on a Tenax TA trap in the CIS at -25°C, using a CCD2 system (Gerstel). For the chromatographic separation, a HP-5MS column (30 m, 0.25 mm i.d., df = 0.25 µm) with a flow rate of 1 ml min-1 He was used. The temperature program for the GC was: 40°C (2 min), 40-300 °C (10°C/min), 300°C (2 min). A temperature of 300 °C for the MS source was selected. The MS was used in MRM mode. Dipentene and 4-phenylcyclohexene (4PCH, also referred as cyclohexenylbenzene) were used as markers for qualification and quantification of natural rubber (NR) and styrene-butadiene rubber (SBR). Identification and quantification of these markers was achieved via a comparison of retention times and MRM transitions with those of an external standard mixture. The following MRM transitions were used: quantifier (m/z) 93-77 (CE 15eV) and qualifier (m/z) 67-41 (CE 15eV) for dipentene; quantifier (m/z) 104-78 (CE 15eV) and qualifier (m/z) 104 – 51 (CE 50eV) for 4PCH.

Different quantification methods are used to determine the mass of TWPs in a sample based on the pyrolysis products measured. An in-depth discussion of these methods is presented in LEON-T Deliverable 3.1 (P. Tromp et al., 2022). Briefly, in the TNO method the average concentration of the marker 4PCH in tyres has been determined by analysing approximately 40 tyres available on the market and used as a conversion factor to calculate the mass of TWP in a sample based on the average concentration of 4PCH in tyres (0.17 mg/gr tyre). This conversion factor takes into account the most used car tyres in Europe and the percentage of freight traffic (6%). An external standard calibration was used with directly spiked 4PCH in methanol to Tenax TA tubes. To ensure comparability with other studies, a second quantification method was used based on the determination of the rubbers SBR and NR, adapted from the Bundesanstalt für Materialforschung und -prüfung (BAM). In this method, two pyrolysis markers are used – 4PCH and dipentene – as markers for SBR and NR, respectively. The SBR and NR concentrations are calculated using an external standard calibration of reference rubbers SBR1500 and isoprene rubber (IR) in accordance with ISO/TS 21396 and ISO/TS 20593; for SBR a correction factor of 0.9 is applied due to the difference in styrene content between SBR 1500 (23,5%) and SBR in tyre tread (15%). The conversion to TWP is based on an estimation of 45% rubber in tyres.

5.5 SEM-EDX

The gold-coated polycarbonate filters (0.4 µm, 25 mm, Nuclepore) with PM sampled at the locations A2-highway, Cabauw and Rotterdam were analysed with single particle analysis using electron microscopy in combination with X-ray microanalysis (SEM-EDX). The analysis was performed with a Tescan MAIA3 XMH FESEM in combination with a Bruker QUANTAX 800 EDX microanalysis system and a 30 mm2 XFlash 6 | 30 Silicon Drift Detector. Each filter was randomly examined at magnifications between 200 - 10.000X. The images were recorded at an accelerating voltage of 10 keV in the so-called backscattered electron mode (BSE: contrast) based on atomic number. From the BSE image the differences in rubber (dark), mineral encrustations (light gray) and metal encrustations (white) are made clearer. An in-depth discussion of the SEM-EDX method and the application of physico-chemical characterization techniques is presented in LEON-T Deliverable 3.1 (P. Tromp et al., 2022). In this study SEM-EDX is used for determining the TWP morphology and percentage of mineral encrustations in TWP in five size classes: 25-50, 10-25, 5-10, 2.5-5 and 1-2.5 µm. In total more than 50 TWP particles were analysed in detail. The particle size and element composition of the total TWP particle and the encrustations within this particle were determined, using FEATURES within the ESPRIT 2 software system. Based on the 2D particle diameter a 3D equivalent-volume diameter was estimated. Particle densities of the encrustations were estimated based on the elemental composition. The density of pure TWP was determined at 1.5 g/cm^3, based on following average composition: SBR 22%, BR 15%, NR 8%, carbon black 24%, silica 15%, ZnO 2% and oil/organic additives 14% (see also LEON-T Deliverable 3.1 for details on tyre composition (P. Tromp et al., 2022)).

6 Results

6.1 General meteorological data

As the meteorological circumstances can play a large role in the emission and dispersion of TWP across different environmental compartments, close attention was paid to temperature, precipitation, wind velocity and wind direction during the measurement campaigns. Figure 11 shows the hourly average wind direction and velocity for all three locations in the Netherlands, and Figure 12 shows the yearly average values based on the past 10 years. In general, the wind direction and velocity were similar to the 10-yearly average, indicating that no large anomalies in terms of weather conditions were registered during the measurement campaign. Figure 13 shows the daily average temperature and precipitation at the main three measurement locations during the sampling period.

Together with the PM10, PM2.5, black carbon and NOx data from the national air quality monitoring network (LML), the meteorological data was used to determine the most optimal sampling weeks with high expected TWP concentrations. In general, samples were selected with the measurement station down winds from the road, with minor precipitation and with relative high PM, black carbon and NOx concentrations (due to traffic). Figure 14 shows the weekly average concentrations of PM10 and PM2.5 and the selected sampling weeks for TWP analysis.

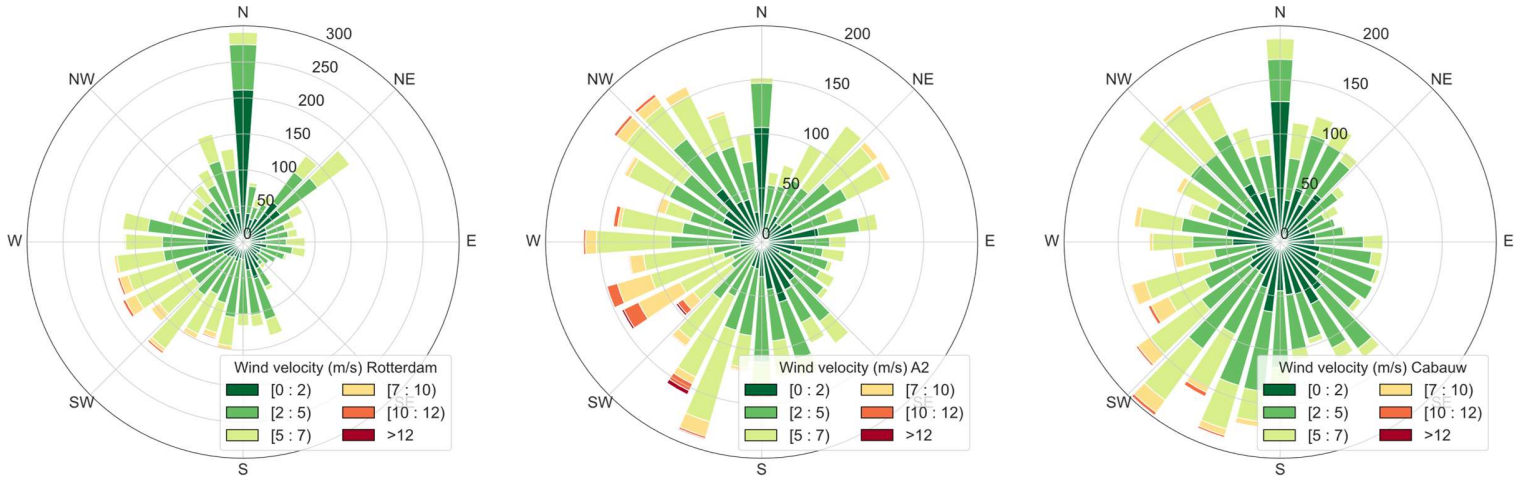

Figure 11 Hourly average wind direction and wind velocity data for the measurement period in 2022 for all three locations according to the nearest KNMI weather stations. The radial axis indicates the amount of times wind with a certain direction and velocity occurred, and the colour indicates the wind velocity.

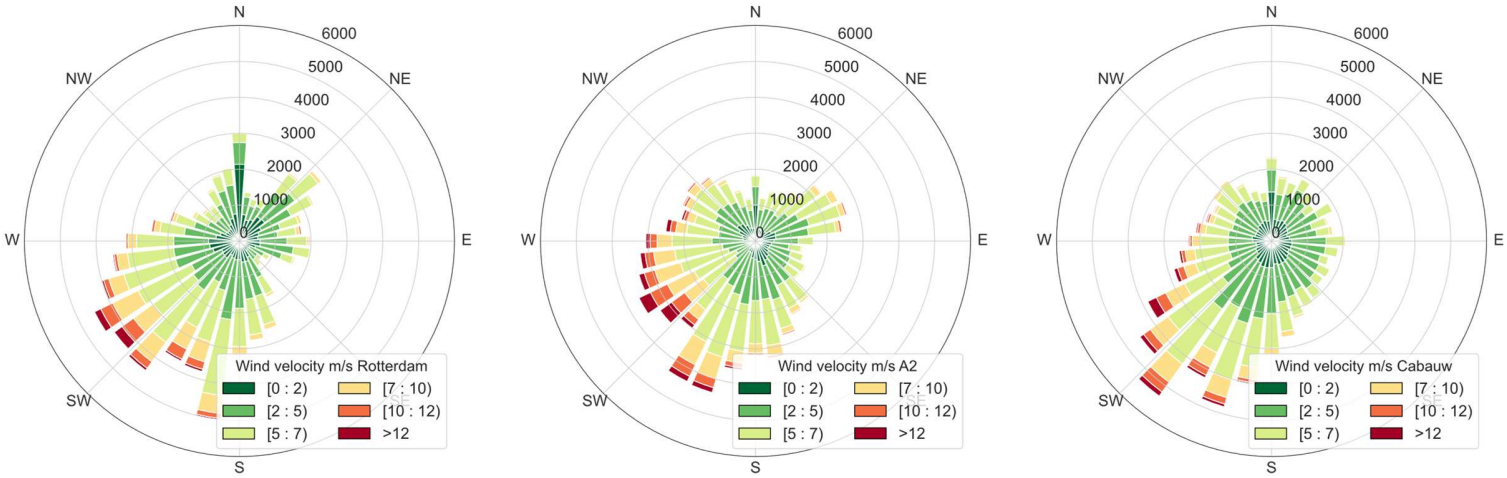

Figure 12 Hourly average wind direction and wind velocity data for the past 10 years at the measurement locations according to the nearest KNMI weather stations. The radial axis indicates the amount of times wind with a certain direction and velocity occurred, and the colour indicates the wind velocity.

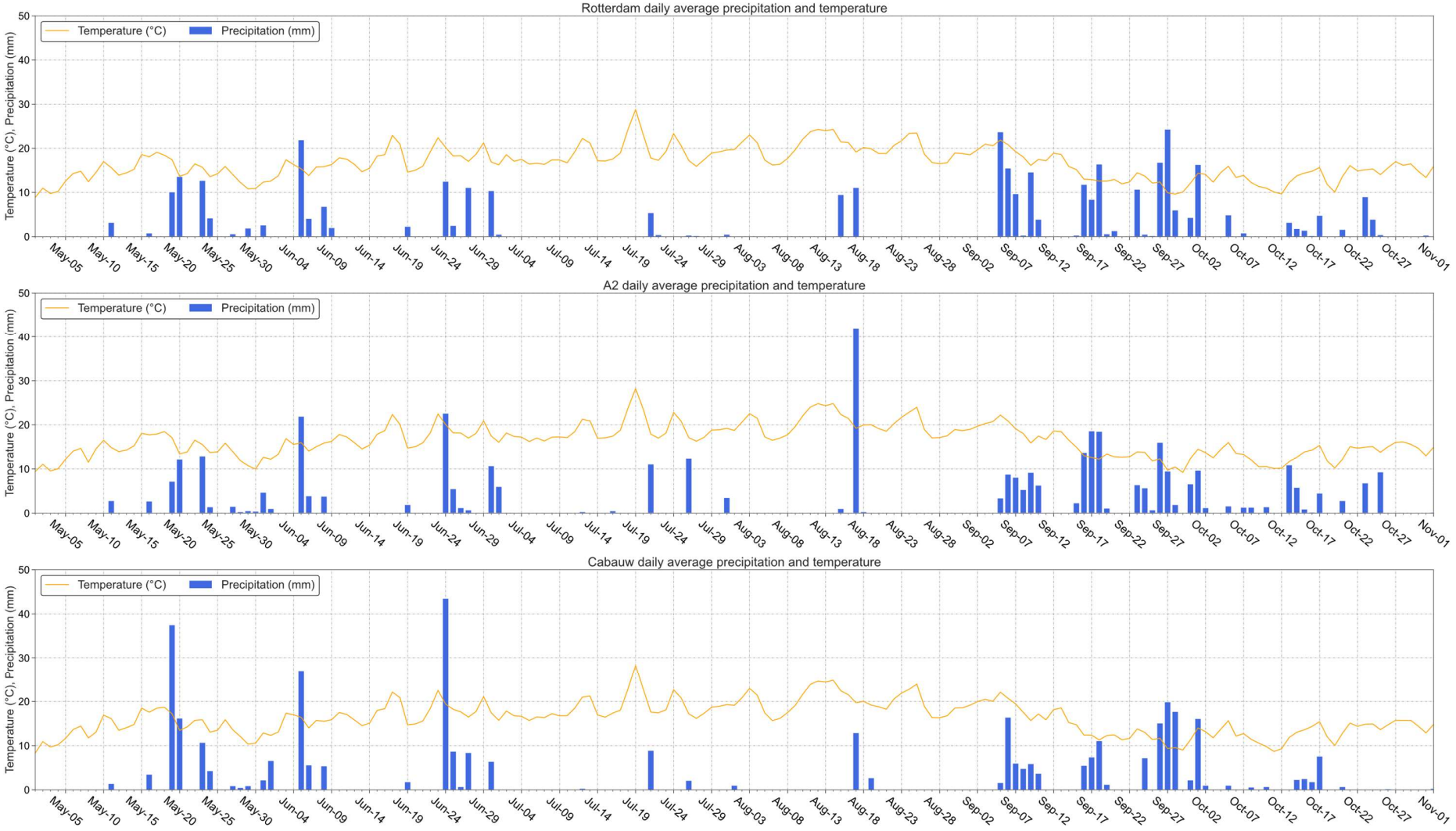

Figure 13 Daily average temperature and precipitation at all three measurement locations during the measurement period according to the nearest KNMI weather station.

6.2 Ambient air

The collected samples within the measurement campaign allow for a detailed analysis of tyre wear particles in the air. As mentioned before, a distinction is made between three different types of locations in such a way that vehicle behaviour can also be considered. This project's measurement locations were situated near measurement locations within the national air quality monitoring network (LML). Within the monitoring network LML a.o. PM10, PM2.5, black carbon and NOx are measured (www.luchtmeetnet.nl). This allows for an extrapolation of the measurement data within LEON-T to the whole year. Figure 14 shows the weekly average concentrations of PM10 and PM2.5 measured within the LEON-T and LML in 2022, with periods where additional real-time particle number and black carbon measurement took place marked in blue, and where PM filter and impactor sampling took place marked in grey. The figure indicates that the concentrations found within the LEON-T measurement campaign align with the concentrations from the LML stations.

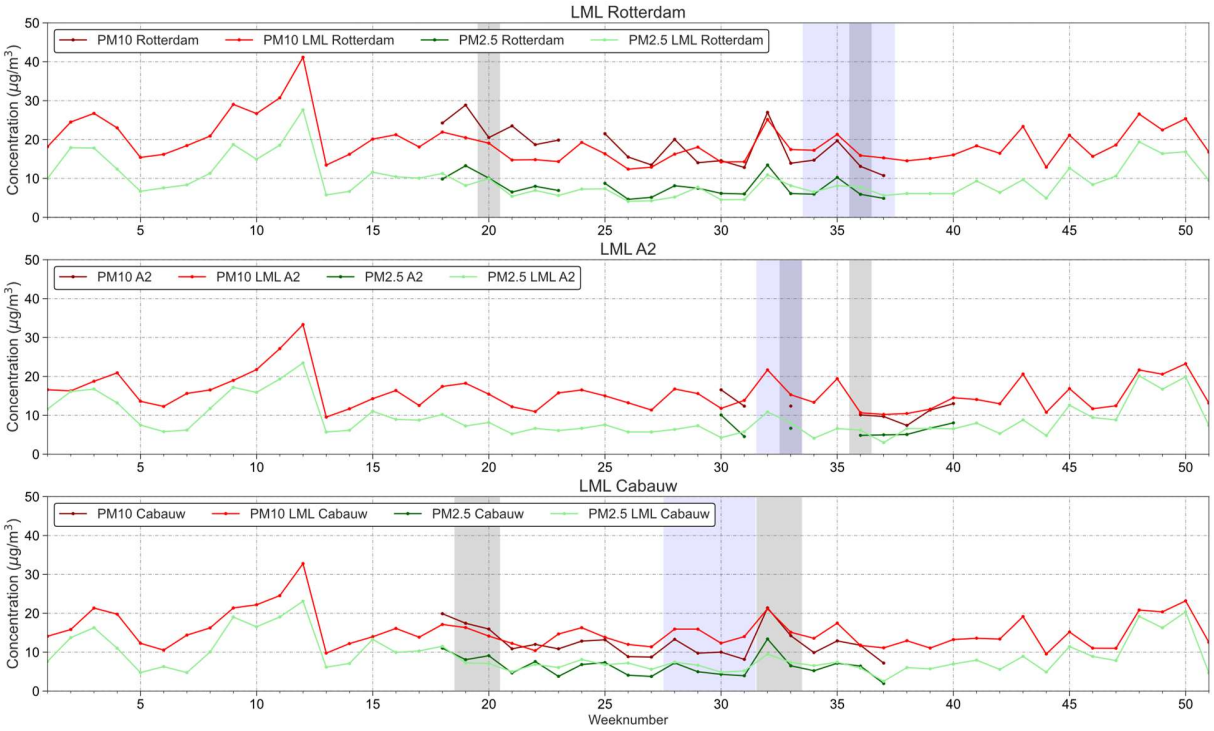

Figure 14: Weekly average concentrations of PM10 and PM2.5 throughout 2022 within the national air quality monitoring network (LML, red and light green) and LEON-T (dark red and dark green), for the locations Rotterdam (top), A2 (middle) and Cabauw (bottom). The area marked in blue represents the period where the real-time monitors were present, and the area in grey represents the periods where PM filter and impactor sampling took place.

Figure 15 shows the NOx concentrations next to the PM concentrations found at the LML stations, giving an indication of the amount of traffic at all three locations. The concentrations of NOx clearly show that the traffic contribution is highest in Rotterdam, followed by the A2 highway.

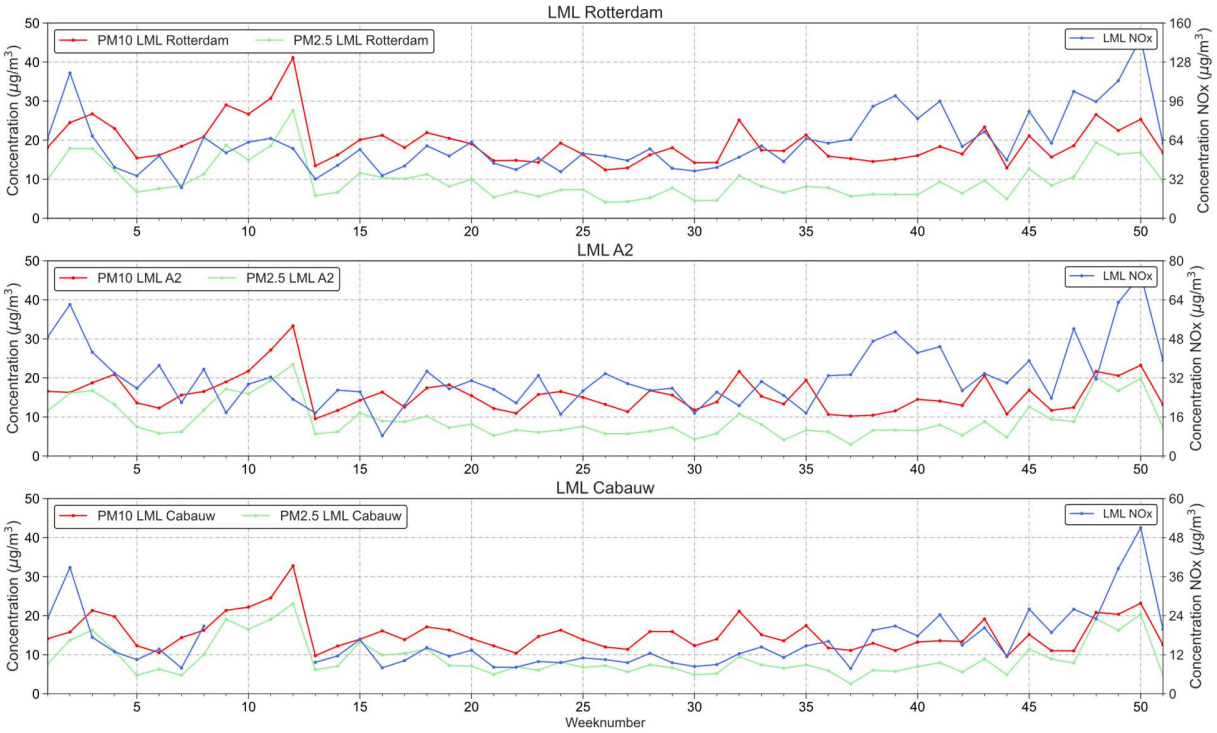

Figure 15 LML PM and NOx concentrations for the locations Rotterdam (top), A2 (middle) and Cabauw (bottom). The NOx concentration is displayed on the right y-axis.

6.2.1 Particulate matter concentrations

In addition to the measurements of PM10 and PM2.5, TSP (total PM) and PM1 fractions were also measured, along with fractions below PM1 using the cascade impactors. Figure 16 shows the weekly concentrations of TSP, PM10, PM2.5 and PM1.0 for locations Cabauw (rural background), A2 highway and Rotterdam (urban city) in the Netherlands. The measured PM concentrations of the urban city location in Rotterdam are markedly higher than those at the rural background location in Cabauw. This holds true for all PM fractions but is most noticeable for the TSP fraction. The concentrations at the A2 highway are similar to the concentrations in Rotterdam, with only the TSP concentrations at Rotterdam being higher. The higher TSP concentrations at Rotterdam are likely due to the shorter sampling distance from the road. The sampling at Rotterdam was performed 3.5 meters from the road, while the A2 highway sampling was carried out at approximately 15 meters from the road, but due to the emergency lane this is 18 meters from the nearest driving lane. The difference between the A2 highway and Rotterdam can be explained by the coarser particles in the TSP fraction, that are more susceptible to gravitational settling what results in a low dispersion rate at longer distances. This can also be seen from the difference between the TSP, PM10 and PM2.5 concentrations at each location; the further away from the road the greater the difference in concentration. For instance at Cabauw the difference between TSP and PM 10 is appr. 5 µg/m^3 and at the urban city location in Rotterdam this is 10 – 15 µg/m^3.

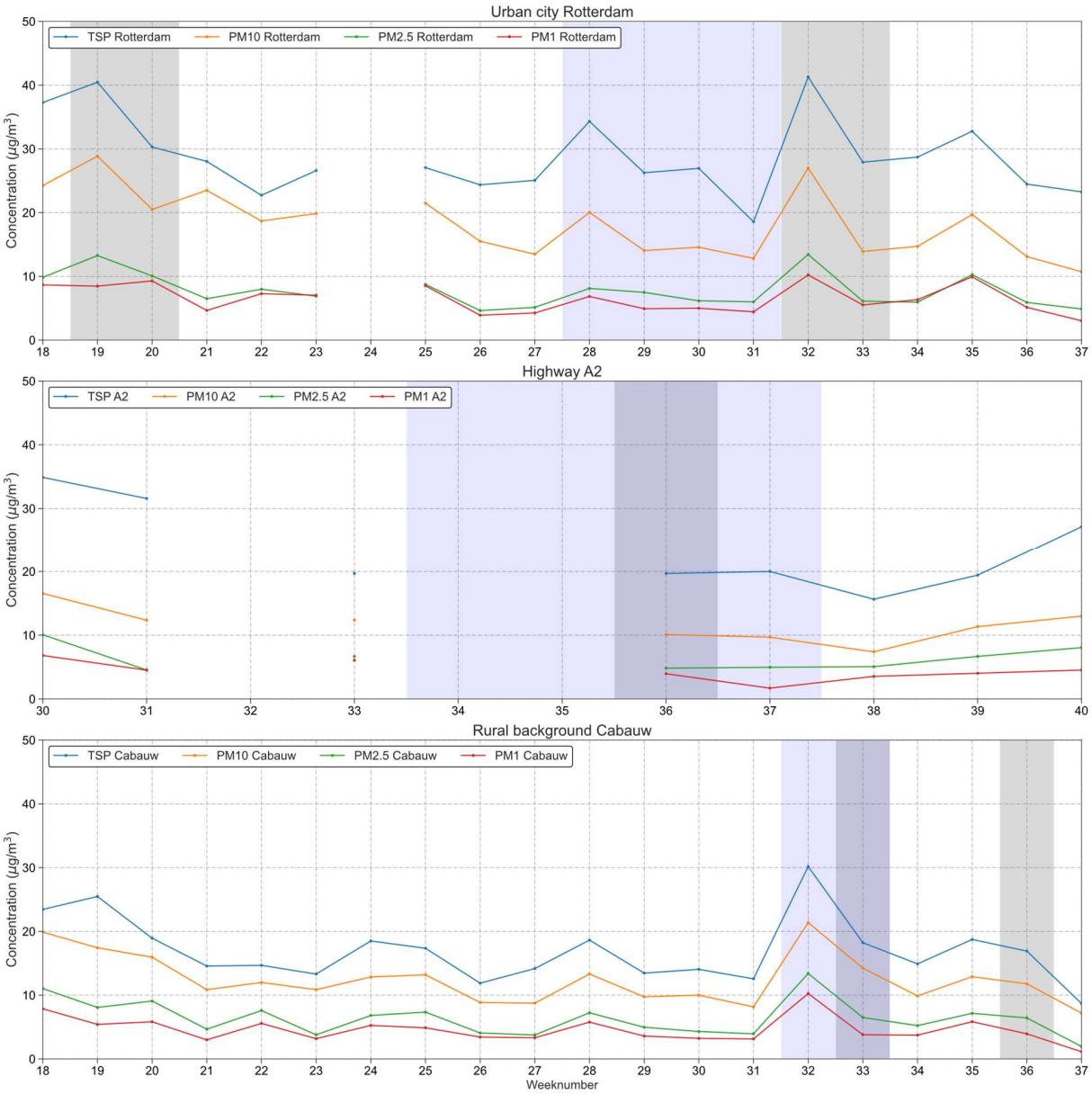

Figure 16 Weekly measured concentrations for TSP, PM10, PM2.5 and PM1 within the LEON-T campaign at the locations Rotterdam (top), A2 (middle) and Cabauw (bottom). Take note of the changed x-axis for the A2 concentrations. The area marked in blue represents the period where the real-time particle and black carbon monitors were present, and the area in grey represents the periods where PM filter and impactor sampling took place.

6.2.2 TWP concentrations

For all measurement locations the amount of TWP in different PM fractions has been determined in two separate periods, illustrated by the greyed-out area in Figure 14 and Figure 16. As mentioned in section 5.4, the quantification of TWP is based on two techniques: (1) the TNO method with a direct calculation of TWP based on an average conversion factor from approximately 40 used car and truck tyres and (2) the BAM/ISO method, based on the quantification of SBR and NR rubber and an estimation of the amount of rubber in car and truck tyres. Tables 2-5 shows the TWP concentrations calculated with both methods (TWP-TNO and TWP-BAM/ISO) and the average TWP concentration from those two methods (TWP average). In addition the difference between the two methods is presented (% BAM/ISO vs TNO), where 100% means that both methods yield the same concentration and a lower and higher percentage means that the BAM/ISO method yields a lower and higher concentration than the TNO method respectively.

In Table 2 and Table 3 TWP concentrations are presented for the location highway A2 at two different distances from the roadside (respectively 18 m and 7 m from the first driving lane). In general, the TWP concentrations in PM2.5 (<2.5µm) and PM1 (<1µm) is higher in the measurements closer to the roadside. It should be noted, however, that the measurement periods are not the same.

Table 2 Ambient air TWP concentrations in different size fractions (in µg/m3) on LML station Breukelen (NL), 18 m from the roadside (first driving lane) of highway A2.

Location	Highway A2 – LML station A2-Breukelen										
Sampling period	Week 33				Week 36				Week 32		
PM fraction	TSP	PM10	PM2.5	PM1	TSP	PM10	PM2.5	PM1	<1µm	<0.5µm	<0.2µm
PM (µg/m^3)	19.7	12.4	6.7	6.1	19.4	9.8	4.7	4.1	7.8	4.4	0.36
SBR (µg/m^3)	0.081	0.020	0.009	0.007	0.061	0.014	0.004	0.004	0.013	0.012	0.001
NR (µg/m^3)	0.011	0.003	0.001	0.001	0.008	0.002	0.001	0.001	0.001	0.001	0.000
TWP-BAM/ISO (µg/m^3)	0.18	0.046	0.021	0.016	0.14	0.031	0.010	0.010	0.028	0.025	0.002
TWP-TNO (µg/m^3)	0.20	0.036	0.013	0.006	0.16	0.039	0.009	0.006	0.016	0.014	0.002
% BAM/ISO vs TNO	93%	129%	162%	249%	87%	81%	105%	152%	141%	147%	107%
TWP average (µg/m^3)	0.19	0.041	0.017	0.011	0.15	0.035	0.009	0.008	0.022	0.019	0.002
% NR in rubber	12%	12%	14%	10%	12%	11%	15%	13%	13%	8.1%	13%
% TWP/PM	1.0%	0.3%	0.3%	0.2%	0.8%	0.4%	0.2%	0.2%	0.3%	0.5%	0.6%

Table 3 Ambient air TWP concentrations in different size fractions (in µg/m3) near LML station Breukelen (NL), 7 m from the roadside (first driving lane) of highway A2.

Location	Highway A2 – roadside			
Sampling period	Week 35			
PM fraction	<2.5µm	<1µm	<0.5µm	<0.2µm
PM (µg/m^3)	7.3	5.9	2.7	1.5
SBR (µg/m^3)	0.010	0.009	0.006	0.003
NR (µg/m^3)	0.001	0.001	0.001	0.000
TWP-BAM/ISO (µg/m^3)	0.022	0.019	0.014	0.007
TWP-TNO (µg/m^3)	0.015	0.013	0.009	0.005
% BAM/ISO vs TNO	148%	146%	150%	151%
TWP average (µg/m^3)	0.018	0.016	0.011	0.006
% NR in rubber	8.1%	8.9%	11%	7.2%
% TWP/PM	0.3%	0.3%	0.4%	0.4%

Table 4 Ambient air TWP concentrations in different size fractions (in µg/m3) on LML station Bentinckplein in Rotterdam (NL), 1 m from the roadside.

Location	LML station Bentinckplein – Rotterdam (urban city)										
Sampling period	Week 20				Week 36				Week 34		
PM fraction	TSP	PM10	PM2.5	PM1	TSP	PM10	PM2.5	PM1	<1µm	<0.5µm	<0.2µm
PM (µg/m^3)	30.3	14.6	10.1	9.3	23.6	9.4	5.9	5.1	8.6	6.6	1.4
SBR (µg/m^3)	0.65	0.12	0.050	0.009	0.59	0.073	0.007	0.009	0.008	0.006	0.004
NR (µg/m^3)	0.034	0.009	0.008	0.002	0.037	0.006	0.002	0.002	0.001	0.001	0.001
TWP-BAM/ISO (µg/m^3)	1.4	0.25	0.12	0.023	1.3	0.16	0.017	0.023	0.019	0.015	0.010
TWP-TNO (µg/m^3)	1.4	0.31	0.074	0.018	1.3	0.20	0.015	0.014	0.016	0.012	0.007
% BAM/ISO vs TNO	99%	81%	156%	126%	96%	81%	116%	161%	119%	118%	144%
TWP average (µg/m^3)	1.4	0.28	0.095	0.023	1.3	0.18	0.016	0.018	0.018	0.013	0.008
% NR in rubber	5.0%	7.5%	13%	22%	5.9%	7.2%	24%	20%	11%	11%	19%
% TWP/PM	4.5%	1.9%	0.9%	0.2%	5.5%	1.9%	0.3%	0.4%	0.3%	0.4%	0.6%

Table 5 Ambient air TWP concentrations in different size fractions (in µg/m3) near the regional background LML station in Cabauw (NL).

Location	LML station Cabauw (regional background)											
Sampling period	Week 20				Week 32				Week 31			
PM fraction	TSP	PM10	PM2.5	PM1	TSP	PM10	PM2.5	PM1	<2.5µm	<1µm	<0.5µm	<0.2µm
PM (µg/m^3)	18.9	15.9	9.1	5.8	30.2	21.4	13.4	10.3	6.0	4.9	3.3	1.2
SBR (µg/m^3)	0.020	0.007	0.009	0.001	0.030	0.027	0.007	0.013	0.011	0.010	0.008	0.004
NR (µg/m^3)	0.002	0.001	0.000	0.000	0.002	0.001	0.000	0.000	0.001	0.001	0.001	0.000
TWP-BAM/ISO (µg/m^3)	0.044	0.016	0.019	0.003	0.065	0.054	0.016	0.026	0.024	0.022	0.019	0.008
TWP-TNO (µg/m^3)	0.048	0.016	0.010	0.003	0.078	0.064	0.017	0.015	0.016	0.014	0.012	0.005
% BAM/ISO vs TNO	93%	98%	197%	101%	83%	84%	91%	175%	153%	160%	153%	162%
TWP average (µg/m^3)	0.046	0.016	0.014	0.003	0.071	0.059	0.017	0.021	0.020	0.018	0.016	0.006
% NR in rubber	8.3%	7.4%	4.7%	11%	7.2%	5.7%	5.4%	3.8%	7.0%	7.2%	9.1%	3.7%
% TWP/PM	0.2%	0.1%	0.2%	0.05%	0.2%	0.3%	0.1%	0.2%	0.3%	0.4%	0.5%	0.5%

In general, the TWP concentrations and the contributions of TWP to the PM concentrations (%TWP/PM) in Rotterdam are much higher than along the A2 highway and in Cabauw (2-9 and 5-20 times higher respectively), with increasing differences with increasing PM size. The difference between Rotterdam and Cabauw is expected due to the difference in traffic intensity and the distance to the road. The difference between Rotterdam and A2 highway was not entirely expected due to the much higher traffic intensity (85600 vs 16700 movements per day) along the highway A2. Also, on both locations the wind direction was crosswise to the road. Partially the difference can be explained by the greater distance to the roadside (18 m vs 3.5 m) of the measurements, but comparing the <2.5µm and <1µm concentrations from the measurements nearer to the roadside (7 m, Table 3), concentrations are similar to Rotterdam even with 5 times higher traffic intensity. A major part of the difference can be explained by the accelerating and braking of vehicles in Rotterdam, because the measurements are performed close to a busy intersection. Braking and acceleration causes more friction on the tyres which results in higher emissions.

Another explanation could be the application of open graded asphalt (ZOAB) on the A2. Open graded asphalt is known to partially trap particles that are deposited onto the road surface and therefor reduces the resuspension of these particles and not only prevents that particles are washed to the soil or surface water (Geilenkirchen, 2022) but also reduces the TWP concentrations in ambient air near the road side. However, air monitoring studies along highways with open graded asphalt are contradictory. Tests in Rotterdam showed that the PM concentration in the air along the porous asphalt part was approximately 30% lower than along the dense asphalt concrete (Heijden & Hooghwerff, 2011). Contradictory, Blokland & Hooghwerff (2009) showed that emission of PM_{10} is about 30-35% higher on open

graded asphalt than on dense asphalt concrete. By far the largest part (95%) of the material loss is from rubber wear, and only a small portion (5%) from asphalt wear. Moreover, a study performed by Svensson et al. (2023), with atmospheric measurements of NO_x and PM_{10} for 7 months along a road stretch containing porous and dense asphalt, showed significantly lower PM_{10} concentrations at the porous pavement during all meteorological conditions. An average reduction factor of approximately 2 was estimated, based on the ratio PM_{10}/NO_x. Also particle emissions measured behind the wheel of a moving car were lower at the porous pavement. The reasons for the improved air quality are suggested to be the removal of road dust from the surface into the pores of the pavement, the reduction of the air-pumping suspension forces of the tyres, and the prolonged drying of the surface.

In Figure 17 a schematic presentation of the percentage of NR in the total rubber (SBR+NR) content is given (% NR in rubber) from which the contribution of truck tyres (freight traffic) can be derived. Natural rubber is more abundant in truck tyres as opposed to passenger car tyres, and therefore is expected to be higher at locations with more freight traffic. As such, the fraction of NR is expected to be highest at the A2 highway, as 8% of traffic at this location is reported to consist out of trucks. In Rotterdam and Cabauw this percentage is lower, with 1% and 3% respectively.

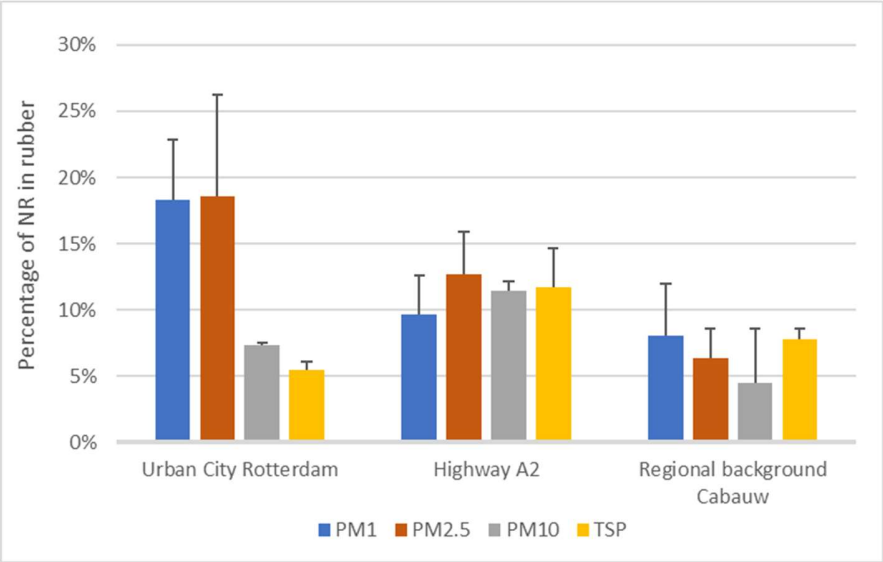

Figure 17 Average percentage of natural rubber (NR) in rubber (SBR+NR) of TWP in ambient air from two measurement periods in 2022 at three locations in the Netherlands.

The fraction of NR in total rubber in the PM samples was found to be approximately 12% at the A2 highway, which is indeed higher than the values found in Rotterdam (5- 7%) and Cabauw (7-8%). Assuming a linear relation between the amount of freight traffic and the percentage of NR, the PM emission from car tyres (at 0% freight traffic) consist of approximately 2-5% of NR rubber and each percent of freight traffic contributes to an equal increase in NR in rubber. For the location Rotterdam the contribution of NR increases with smaller PM fractions; in PM1 and PM2.5 the percentage of NR is in the range of 13 – 24%, 3-4 times higher than in PM10 and TSP. The reason is not clear, but could be an artefact due to matrix interferences from the organic matter in PM. Another explanation could be that braking and accelerating of heavy vehicles (with rubber tyres) results in a higher emission of small TWP.

The concentrations found within LEON-T for the locations Rotterdam and Cabauw are comparable to the concentrations found in an earlier measurement campaign in 2012-2013, indicating that the TWP concentrations over the years have remained relatively stable. Table 6 shows the results of the 2012-2013 measurement campaign within the NanoFASE project. As expected, TWP concentrations in the Maastunnel are higher. The Maastunnel has two tunnel tubes with two traffic lanes each and a maximum speed of 50 km/h. In 2012 the traffic intensity in the tunnel was 30.000 – 35.000 vehicles per 24 h. The TWP concentrations are slightly lower than previously reported (P. Tromp et al., 2021) due to new insights in applied markers from earlier work in LEON-T (P. Tromp et al., 2022). In the new results shown in Table 6, the TWP concentrations are calculated with only 4-vinylcyclohexene as marker, discarding styrene and dipentene, which were used previously.

Table 6 Results of measurement campaign in 2012-2013 for the EU project NanoFASE: ambient air TWP concentrations in TSP, PM10, PM2.5 and PM1 (in µg/m³) on different locations in Rotterdam and near the regional background LML station in Cabauw (NL), using 4VCH as tracer. The percentage values indicate the %TWP/PM.

Location	n	TWP in µg/m3 (%TWP in PM)			
		TSP	PM10	PM2.5	PM1
Bentinckplein - Rotterdam	3	0.78 (5%)	0.16 (0.9%)	0.07 (0.4%)	<0.03 (<0.2%)
Maastunnel – Rotterdam	2	1.5 (3%)	0.68 (1.5%)	0.12 (0.3%)	0.06 (0.1%)
Cabauw (regional background)	1	-	0.04 (0.20%)	-	-

The concentrations found within LEON-T are comparable to ambient air concentrations of TWP found for other cities in Europe. For instance, 81 PM10 measurements in 2012 within 11 cities in France, USA and Japan yielded concentrations ranging from < 0.004-0.67 µg/m^3, with an average of 0.08 µg/m^3, representing about 0.42 (0.07-1.4) % of total PM (J. M. Panko et al., 2013). Sampling locations were selected to represent a variety of settings including both rural and urban core. The PM10 concentrations found within the city Rotterdam are similar with an average concentration of 0.23 µg/m^3, but representing a higher contribution to PM (1.9%). The TWP PM10 concentrations found along the highway A2 and at the rural location Cabauw are lower (0.02 – 0.06 µg/m^3) but still in the same range. Other historically published values (1973-2005) for high traffic areas show slightly higher TWP concentrations in ambient air ranging from 2 to 5 µg/m^3 for TSP or PM10 and from 0.2 to 2.2 µg/m^3 for PM2.5 (J. M. Panko et al., 2013). The air concentrations of TWP were associated with traffic load and population density, but trends were not statistically significant. Generally, air concentrations of TWP were higher for samples collected near the road (<10 m) than further away (10-300 m), however, the trend was not significant.

In 2018 a follow-up study was performed in three major cities (Los Angeles, London and Tokyo) with 82 PM2.5 and 23 PM10 measurements (J. Panko et al., 2019). TWP concentrations in PM10 in these cities were higher than found in (J. M. Panko et al., 2013), with an average of 0.46 µg/m^3, ranging from 0.047-2.2 µg/m^3 and contributions to PM10 of 0.97 (0.23-1.2)%. TWP concentrations in PM2.5 were considerably lower than in PM10, ranging from 0.002-0.15 µg/m^3 with an average concentration of 0.015 ug/m^3 and contributions to PM2.5 of 0.12 (0.05-0.25)%. These PM2.5 concentrations are similar to those found at the LEON-T sites, where TWP concentrations range from 0.009-0.095 µg/m^3 with a contribution to PM2.5 ranging from 0.2-0.9%. The TWP levels in PM2.5 were significantly different between the three cities; no significant correlation between TWP in PM2.5 and traffic count was observed. The highest PM2.5 and PM10 concentrations were recorded at the Blackwall Tunnel Approach in London (mean 0.052 µg/m^3 for PM2.5 and 0.67 µg/m^3 for PM10). These levels are comparable to the concentrations found in the Maastunnel in the Netherlands.

A recent monitoring study conducted in 2020 at the E18 test site in Sweden with Sigma-2 passive samplers and subsequent SEM-EDX analysis showed TWP concentrations near the road side in the same orders of magnitude (Järlskog et al., 2022). At 1.5 m from the roadside TWP concentrations varied between 1.4 – 3.2 µg/m^3 for the TSP fraction and 0.3 – 0.8 µg/m^3 for the fraction < 5µm. For the fine fraction a clear distinction between TWP and bitumen wear particles (BiWP) from the stone mastic asphalt road could not be made, so the values might overestimate actual concentrations. Seasonal differences in TWP concentrations were minor; lowest values were measured in January, but significance is not clear. TWP concentrations at further distances from the road were decreasing, with maximum TWP concentrations in TSP of 3.2, 2.9, 1.4 and 0.45 µg/m^3 at 1.5m, 4.8m, 27m and 100m.

Figure 18 gives a schematic presentation of particulate matter found at all three locations, with the corresponding TWP concentrations for all PM fractions. No major seasonal differences in TWP concentrations were observed between spring and summer; also the distribution over the PM fractions was not affected, however temperatures were not all that different (10-15 vs 20-25°C), and it remains unclear if cold temperatures during wintertime will have an effect on the TWP concentrations. During the whole year deposited dust was sampled near the highway A27 (see section 6.3); no significant seasonal differences in TWP concentrations were observed.

From the results it becomes clear that most TWP are found in the coarse fraction >10µm, ranging from approximately 80% for Rotterdam and the A2 highway to around 50% for the rural background location Cabauw. The mass concentration of TWP in air decreases rapidly with smaller particle sizes. This could also be observed in Figure 19, where the particle size distribution of TWP is shown, based on current filter and impactor measurements (bar chart)

compared with measurements from 2012 for the EU project NANOFase (line chart). From this data it can be seen that the emission of TWP in the nano sizerange is negligibly low. For the rural background location Cabauw the proportion of nanosized TWP seems larger, however on this location the high volume impactor has been used with the smallest impactor stage of 0.2µm instead of 0.1µm. Also TWP concentrations in the TSP fraction were considerably lower than the other locations, so relatively the TWP proportion in the smaller size range is larger.

The fact that the difference between TWP in TSP and PM10 is quite large will have an effect on comparing data between measurement studies. Different types of samplers exist to collect the PM10 fraction (i.e. Anderson cyclones, Harvard impactors); but the cut-off diameters of these samplers can deviate which for TWP will have a major effect on the measured concentrations.

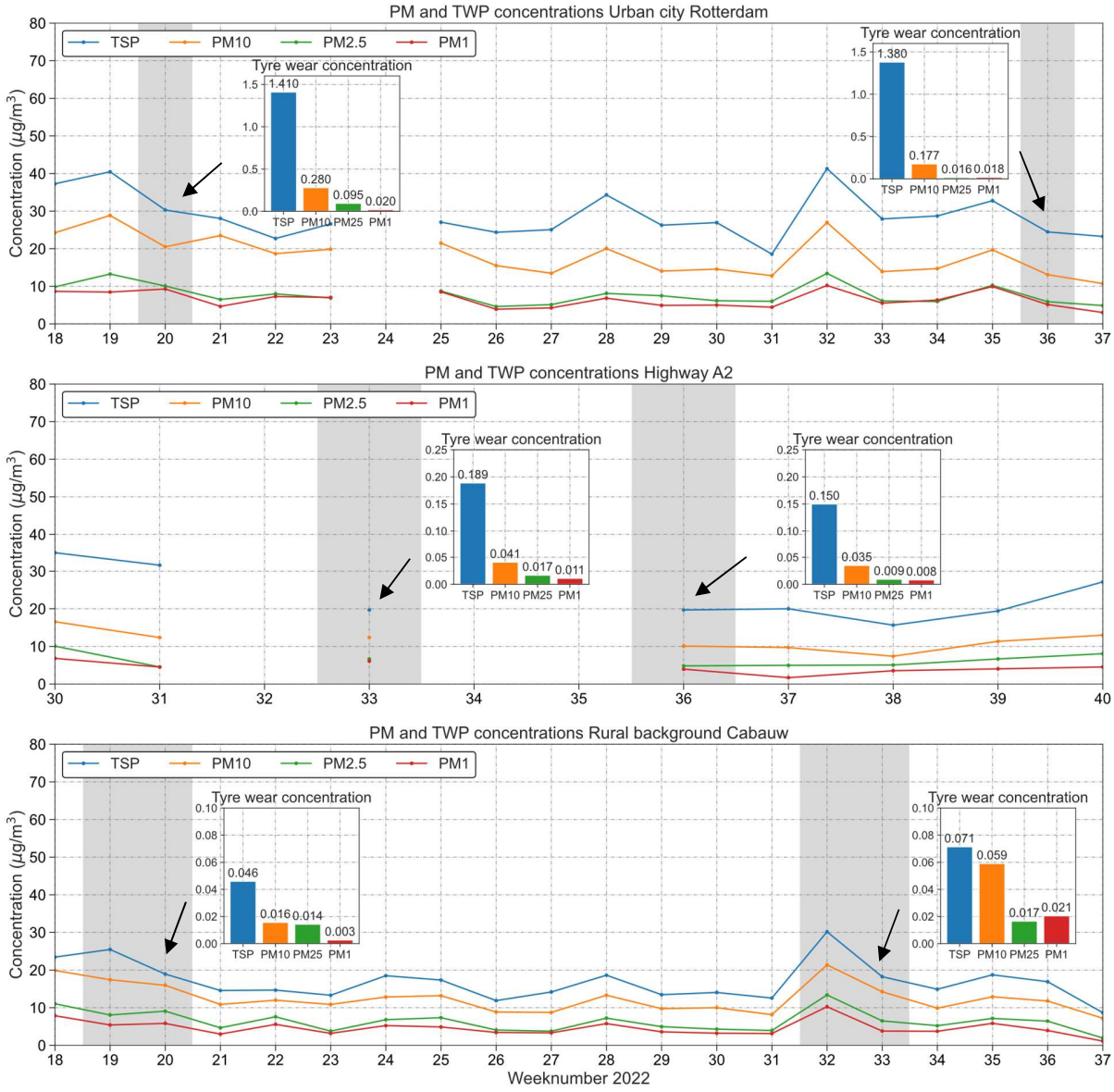

Figure 18 Measured particulate matter concentrations along with tyre wear concentrations as determined by TGA-GCMS for the air fractions TSP, PM10, PM2.5 and PM1 within the LEON-T measurement campaign at all three locations. The area marked in grey represents the periods where chemical characterisation of the air samples took place.

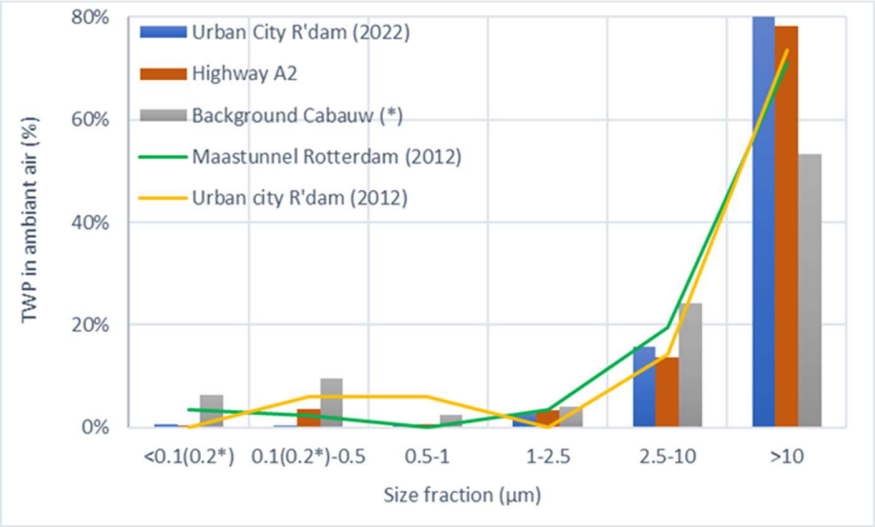

Figure 19 Average particle size distribution of TWP in ambient air (average from the two measurement periods in 2022) at three locations in the Netherlands (for Cabauw the smallest size fraction is 0.2µm). For comparison also the particle size distributions are given from the measurement campaign in 2012 for the EU project NANOFase.

6.3 Deposited dust

Total deposition samples were collected at different locations, and at several distances from the road. Total deposited TWP is the amount of TWP that is deposited during dry periods as dry deposition and during rainy periods as wet deposition and gives insight in the contamination and accumulation of TWP on soil and water surfaces. Table 7 shows the TWP concentrations (in mg/m^2/day) found in deposited dust for all locations.

Table 7 TWP concentrations in deposited dust on four locations in the Netherlands, at various distances from the roadside.

Location	Rotterdam		A2						A27				Cabauw / N201			
Sampling period (month/year)	3/22	9/22	8/22			9/22			3/22	6/22	8/22	11/22	3/22		9/22	
Sampling days	44	33	27	27	27	35	35	35	32	32	31	37	44	44	26	26
Distance from road	1 m	1m	7m	20m	50m	7m	20m	50m	17m	17m	17m	17m	1m	350m	1m	350m
Analyses (n)	n=1	n=1	n=1	n=1	n=1	n=1	n=1	n=1	n=3	n=1	n=1	n=1	n=1	n=1	n=1	n=1
SBR (mg/g)	19	34	7.0	4.3	3.2	3	3.6	2.0	13 ± 1	3.6	2.9	6.4	6.2	3.6	23	0.89
NR (mg/g)	3.9	4.2	1.6	1.2	0.69	1	1.4	0.74	3.3±0.3	2.0	1.5	4.5	2.4	0.48	3.5	0.50
% NR in rubber	17%	11%	18%	22%	18%	29%	28%	27%	21±2%	36%	35%	41%	27%	12%	25%	36%
TWP-BAM/ISO (mg/g)	46	76	17	11	7.7	10	9.9	5.5	32 ± 3	11	8.9	22	17	8.3	28	2.8
TWP-TNO (mg/g)	54	71	21	13	5.4	7.7	11	7.0	36 ± 1	10	9.3	18	21	13	23	2.7
% BAM/ISO vs TNO	86%	107%	80%	87%	145%	126%	89%	79%	96±1%	109%	96%	117%	81%	65%	121%	102%
PM (mg/m2/d)	343	329	206	89	92	279	94	93	64	148	164	53	132	76	127	82
TWP (mg/m2/d)	17	23	4.0	1.1	0.61	1.5	0.38	0.35	2.0±0.2	1.6	1.5	1.1	2.5	0.80	3.7	0.23
% TWP/PM	5.0%	7.0%	1.9%	1.2%	0.7%	0.5%	0.4%	0.4%	3.1%	1.1%	0.9%	2.1%	1.9%	1.1%	2.9%	0.3%

The concentrations of TWP in deposited dust vary depending on location, and distance from the road. The highest concentrations of TWP are found nearest to the road and decrease with increasing distance to the road (see Figure 20). From the three measurements at highway A2 an exponential decrease can be derived. Just like the TWP concentrations in air, in Rotterdam near the busy intersection the highest deposited TWP concentrations were measured, most likely due to accelerating and braking of vehicles. As explained before (section 6.2.2), the application of open graded asphalt (ZOAB) on the A2 could also have an effect. Open graded asphalt is known to partially trap particles that are deposited onto the road surface and therefor reduces the resuspension of these particles (Geilenkirchen, 2022) which could result in lower concentrations of TWP in deposited dust.

Near the roadside deposition is caused by splash water and gravitational settling while further away from the road only gravitational settling plays a role. The results from the A27 highway give an indication of seasonal differences in the deposition of TWP throughout the year. Unlike total deposited PM, where concentrations were higher in spring and summer compared to autumn and winter, for TWP no significant seasonal differences were observed. No clear relationship between deposited PM and TWP could be observed; near the roadside the percentage TWP in PM is the highest and decreases with increasing distance.

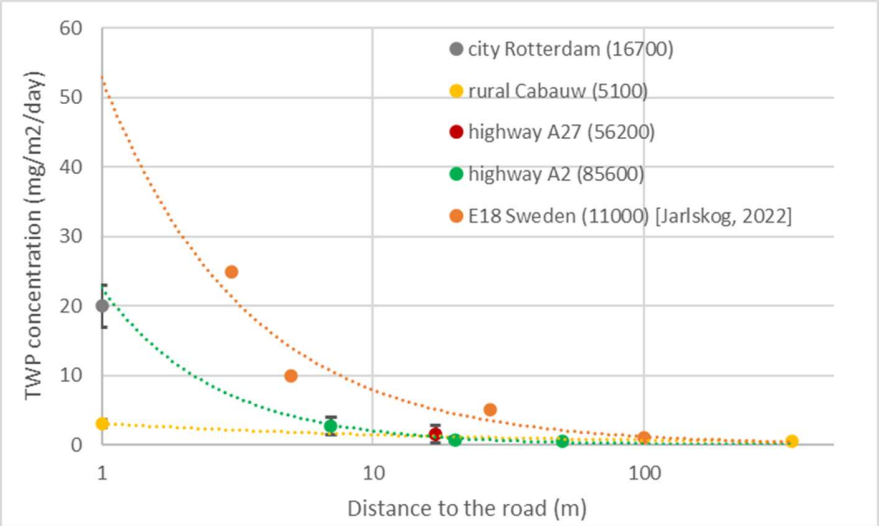

Figure 20 Deposited TWP concentrations (mg/m2/day) at different distances from the road side for the city of Rotterdam, rural location Cabauw and the highways A2 and A27. For comparison the data from the monitoring campaign of VTI at the test site E18 is included (Järlskog et al., 2022). In the labels the daily traffic intensity in vehicles per day is presented.

In Figure 20 also data of VTI (Järlskog et al., 2022) of the test site at the E18 is included for comparison. In this study at several distances from the road (3.1, 4.8, 27 and 100m) bulk deposition samplers were applied with a measurement period of 30 days, similar to the LEON-T campaign. Total collected PM concentrations were very similar to the values of the LEON-T campaign and ranged from 67 – 430 mg/m^2/day, with decreasing values further away from the road side. As can be observed in Figure 20, TWP concentrations were a bit higher than the values in present study. The TWP concentrations, in mg/m^2/day were calculated from the 'raw' data as published in the paper, where, besides total PM mass, only relative percentages of TWP in the fractions 2-20µm and 20-125µm were given. Results are based on the assumption that the same ratio between fine and coarse PM exists as measured in present study. The resulting values are most likely to be an overestimation of actual concentrations due to the number to mass conversion methodology used by (Rausch et al., 2022). A density of 1.8 g/cm^3 for TWP is assumed based on 50% attached road wear particles; for fine particles (<20 µm) this percentage is lower (see section 6.8.2) what will result in a lower density of about 1.6 g/cm^3. However, the largest overestimate comes from the assumption of an equal 3D volume diameter and 2D projected particle diameter (observed from the 2D SEM-image). For irregular shaped particles (deviating from the spherical shape) a correction is needed to convert 2D diameters into 3D diameters (P. C. Tromp et al., 2017). This correction factor (the volume shape factor) has a big effect on the derived particle mass, which can overestimate the concentration up to a factor 2-3 for irregular shaped particles, such as TWP. Another complicating factor is that adsorption of semi-volatile organic substances on TWP may influence the particle size. As can be seen in Deliverable 2.3 the evaporation of tyre additives and subsequent condensation is an important emission source which can cause TWP to become larger.

The concentrations of TWP in deposited dust are much lower than recently published deposited TWP values in the UK (Parker-Jurd et al., 2021). At 20 m and 50 m from two urban roads, two rural roads and two motorways in the cities Bristol and Plymouth 24h deposited dust samples were collected. Average TWP concentrations were 30 ± 6, 11 ± 2 and 43 ± 7 mg/m^2/day for respectively the urban road, rural road and motorway. In general, 20 m from the roadside the concentrations were higher than 50 m from the road; 28 ± 5 vs 19 ± 2 mg/m^2/day, however the difference as not significant. The difference could be explained by the traffic load, but no traffic count data was published. Also it remains unclear if concentrations relate to TWP or TRWP; this makes a difference of a factor 2. Furthermore, the applied analytical method was different than the TED-GCMS technique in this study. Pyrolysis GC-MS was used, however benzothiazole was used as a marker. A study performed by Rodland et.al. (2022) showed widely variating TWP concentrations deposited in roadside snow, between 220–109,000 mg/m^2. These large concentration differences relate to the road surface and traffic variables (speed limit and AADT). In agreement with previous estimates the study of Rodland et al. (2022) showed that concrete asphalt surface layers contribute three times more road abrasion compared to those with stone mastic asphalt. Concentrations could not be converted in mg/m^2/day because deposition times were not clear, however the contribution of TWP to the total particle load of 5 – 6% is in agreement with our findings in Table 7, with 1 – 7% TWP in total deposited PM.

For the deposition sampler located nearest to the road side mass size distributions of deposited TWP has been determined, using cascade fractionation and filtration. Figure 21 shows a similar bimodal size distributions of TWP for all three locations, with peaks between 5-25 µm and 50-200 µm. This is in agreement with a number of other studies that report bimodal mass size distributions with one peak in the coarse fraction and another one in the fine fraction (S. Wagner et al., 2018). A detailed explanation is given in section 6.8.

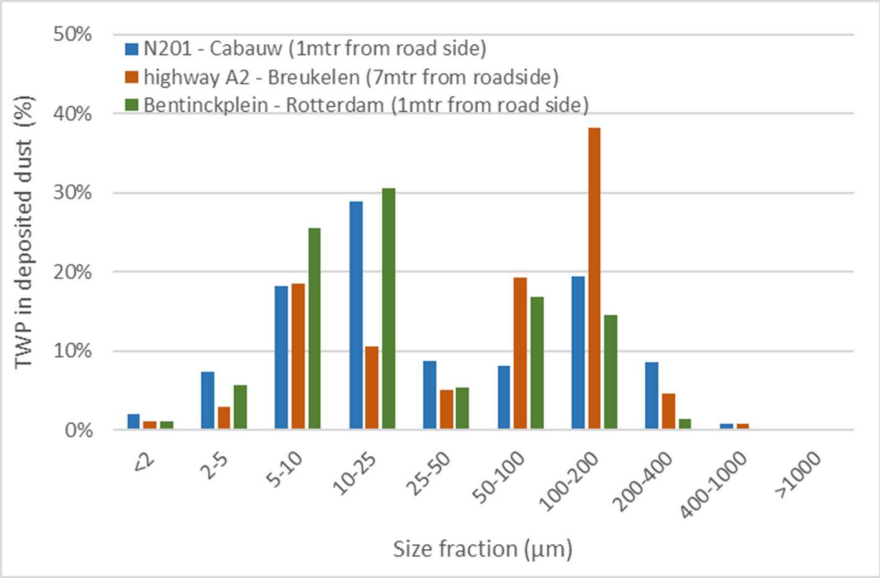

Figure 21 Particle size distribution of TWP in deposited dust at three road side locations in the Netherlands (TWP in fraction < 2 µm is based on the LOD)

6.4 Runoff

Runoff sampling in the Netherlands took place after approximately 2 weeks of dry weather. The dry period is given in Table 8 TWP concentrations in runoff (in mg/g and mg/L) on three road side locations

Location	Bentinckplein Rotterdam	A2 - Breukelen	E18 – Sweden (storm well)
Sampling period	16-8-2022	16-8-2022	6-10-2022
Dry days	24*	16*	-
Analyses (n)	n=1	n=1	n=7
SBR (mg/g)	44	7.1	2.9 ± 0.9

NR (mg/g)	4.0	3.6	2.1 ± 0.7
% NR in rubber	8%	34%	42 ± 12%
TWP-BAM/ISO (mg/g)	95	21	9.9 ± 1.4
TWP-TNO (mg/g)	106	16	7.8 ± 1.5
% BAM/ISO vs TNO	90%	130%	128 ± 18%
TWP (mg/g)	101	19	8.8 ± 1.3
Suspended solids (mg/L)	520	1220	215
TWP (mg/L)	52	23	1.9 ± 0.3

*only a rough indication, local precipitation was different from precipitation at KNMI weather station

Measured TWP concentrations in runoff water were in the same range as recent published data. In storm water wells TWP concentrations of 2.5 mg/L (Parker-Jurd et al., 2021) and 0.6-179 mg/L (Kumata et al., 2002) were reported. In sediments from roadside gully pots TWP concentrations were in the range of 3 - 54 mg/g (Rødland, Samanipour, et al., 2022) and 1 – 150 mg/g (Mengistu et al., 2021) and in street runoff TWP values of 35 – 82 mg/g (Eisentraut et al., 2018) and 16 – 150 mg/g (Klöckner et al., 2019) were measured. Based on a literature review (Rødland, Lind, et al., 2022) states that the current available data show TWP are found in the range of 3–180 mg/L in road runoff.

In a recent study from (Järlskog et al., 2022) the runoff water and sediment layer in the same storm water well at the E18 once again was measured. Total TWP concentration in runoff water and sediment were 130 – 280 mg/L and 86 mg/L. These concentrations were much higher than measured concentrations with the Leon-T and Microproof project. Partly this could be explained by the very high total suspended solid content of 1100 mg/L, that was 5 – 40 higher than in the previous studies. Another contributing fact is most likely the applied SEM-EDX analysis and quantification method, as explained in section 6.3. From this study the division between fine (2-20um) and coarse (20-125um) TWP could be calculated; most of the TWP was present in the fine fraction, with a percentage of 54 – 83%. This is in agreement with the mass size distribution of TWP in the Leon-T study. Figure 22 shows the size distribution of TWP found in the runoff of the E18 in Sweden together with the runoff of the urban street in Rotterdam and the A2 Highway. From the bimodal size distributions it becomes visible that the share of TWP in the fine fraction is much bigger in the runoff from the E18 and the urban street than in the runoff from the A2, were the coarse fraction dominates. A detailed explanation is given in section 6.8. The bimodal size distribution is in agreement with other studies where it's stated that the runoff waters are typically characterized by a bimodal particle size distribution with maxima between 6-10 um and 70-100 um (Brombach et al., 2005; Charters et al., 2015). The mass percentage of particle sizes <63 mm, so-called fine sediments, varies between 17 wt% and 100 wt% (Charters et al., 2015). In a recent study of (Goedecke et al., 2022; Klöckner et al., 2020) a runoff water sample from a street in Berlin was fractionated, with 25% of TWP in the fine fraction (10-50um). Also here a bimodal distribution was visible with peaks between 10 – 50um and 100 – 500um.

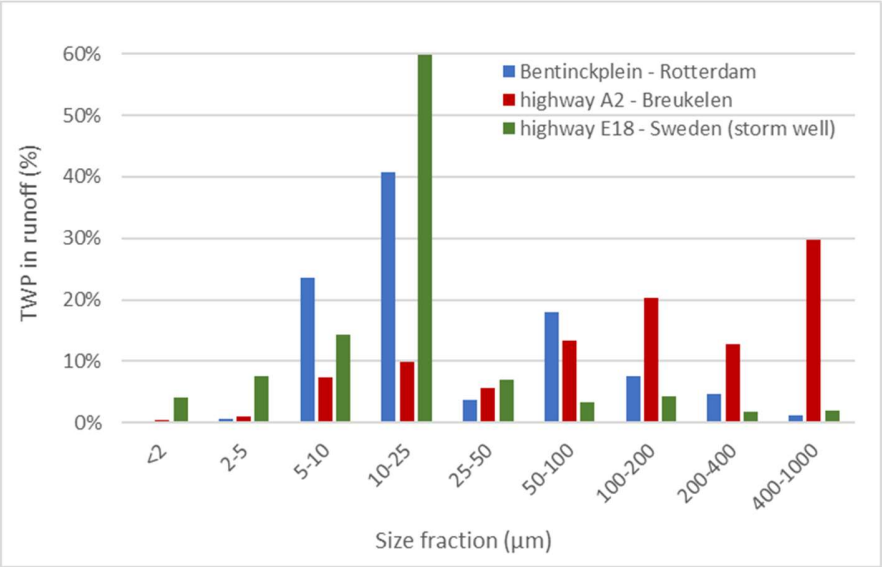

Figure 22 Particle size distribution of TWP in runoff of highway A2 near Breukelen, highway E18 in Sweden (sampled in a storm well) and Bentinckplein in Rotterdam (TWP in fraction < 2 µm is based on the LOD)

according to the precipitation data from the nearest KNMI weather station. Runoff from the E18 in Sweden was sampled in a storm water well adjacent to the road. All runoff samples were fractionated, to determine the mass size distribution of TWP. Despite the 3 times higher traffic intensity at the highway A2 compared to the urban street in Rotterdam, the TWP concentration in road runoff in Rotterdam was approximately two times higher than at the highway A2. This can be explained by difference in traffic behaviour (braking and accelerating) and the difference in the road surface. The highway A2 consist of open graded asphalt while the urban street in Rotterdam consist of dense graded asphalt. Open graded asphalt (ZOAB) is known to partially trap coarse particles that are deposited onto the road surface and therefor reduces the resuspension of coarse particles and prevents that particles are washed with runoff water to the soil or surface water (Geilenkirchen, 2022). Just like the air samples and deposited dust samples, also in runoff at the highway A2 and E18 the percentage of natural rubber is higher than the urban street in Rotterdam, which is in agreement with the amount of freight traffic.

Compared to the road runoff locations in the Netherlands, the runoff water in the storm water well along the E18 showed relatively low TWP concentrations. In runoff water from the same storm water well, sampled in 2018 for the CEDR project Microproof, a higher concentration was measured in the total suspended solid fraction (39 mg/g) but is in the same order of magnitude when values are converted to concentrations per litre runoff water (1.0 mg/L), due to the low total suspended solid fraction of 25 mg/L (Dröge & Tromp, 2019). The TWP concentration in the sediment layer in the storm water well was 13 mg/g. In the same project at the German highway A61 a TWP concentration of 140 ± 10 mg/g and 55 ± 7 mg/L was measured, which is similar to the values found at the urban street in Rotterdam and higher than the concentrations along the highway A2. Both highways have a similar traffic intensity, with an AATD of 38500 for the A61 and 85600 for the A2, but differ in road pavement (dense vs open graded asphalt).

Table 8 TWP concentrations in runoff (in mg/g and mg/L) on three road side locations

Location	Bentinckplein Rotterdam	A2 - Breukelen	E18 – Sweden (storm well)
Sampling period	16-8-2022	16-8-2022	6-10-2022
Dry days	24*	16*	-
Analyses (n)	n=1	n=1	n=7
SBR (mg/g)	44	7.1	2.9 ± 0.9
NR (mg/g)	4.0	3.6	2.1 ± 0.7
% NR in rubber	8%	34%	42 ± 12%
TWP-BAM/ISO (mg/g)	95	21	9.9 ± 1.4

TWP-TNO (mg/g)	106	16	7.8 ± 1.5
% BAM/ISO vs TNO	90%	130%	128 ± 18%
TWP (mg/g)	101	19	8.8 ± 1.3
Suspended solids (mg/L)	520	1220	215
TWP (mg/L)	52	23	1.9 ± 0.3

*only a rough indication, local precipitation was different from precipitation at KNMI weather station

Measured TWP concentrations in runoff water were in the same range as recent published data. In storm water wells TWP concentrations of 2.5 mg/L (Parker-Jurd et al., 2021) and 0.6-179 mg/L (Kumata et al., 2002) were reported. In sediments from roadside gully pots TWP concentrations were in the range of 3 - 54 mg/g (Rødland, Samanipour, et al., 2022) and 1 – 150 mg/g (Mengistu et al., 2021) and in street runoff TWP values of 35 – 82 mg/g (Eisentraut et al., 2018) and 16 – 150 mg/g (Klöckner et al., 2019) were measured. Based on a literature review (Rødland, Lind, et al., 2022) states that the current available data show TWP are found in the range of 3–180 mg/L in road runoff.

In a recent study from (Järlskog et al., 2022) the runoff water and sediment layer in the same storm water well at the E18 once again was measured. Total TWP concentration in runoff water and sediment were 130 – 280 mg/L and 86 mg/L. These concentrations were much higher than measured concentrations with the Leon-T and Microproof project. Partly this could be explained by the very high total suspended solid content of 1100 mg/L, that was 5 – 40 higher than in the previous studies. Another contributing fact is most likely the applied SEM-EDX analysis and quantification method, as explained in section 6.3. From this study the division between fine (2-20um) and coarse (20-125um) TWP could be calculated; most of the TWP was present in the fine fraction, with a percentage of 54 – 83%. This is in agreement with the mass size distribution of TWP in the Leon-T study. Figure 22 shows the size distribution of TWP found in the runoff of the E18 in Sweden together with the runoff of the urban street in Rotterdam and the A2 Highway. From the bimodal size distributions it becomes visible that the share of TWP in the fine fraction is much bigger in the runoff from the E18 and the urban street than in the runoff from the A2, were the coarse fraction dominates. A detailed explanation is given in section 6.8. The bimodal size distribution is in agreement with other studies where it's stated that the runoff waters are typically characterized by a bimodal particle size distribution with maxima between 6-10 um and 70-100 um (Brombach et al., 2005; Charters et al., 2015). The mass percentage of particle sizes <63 mm, so-called fine sediments, varies between 17 wt% and 100 wt% (Charters et al., 2015). In a recent study of (Goedecke et al., 2022; Klöckner et al., 2020) a runoff water sample from a street in Berlin was fractionated, with 25% of TWP in the fine fraction (10-50um). Also here a bimodal distribution was visible with peaks between 10 – 50um and 100 – 500um.

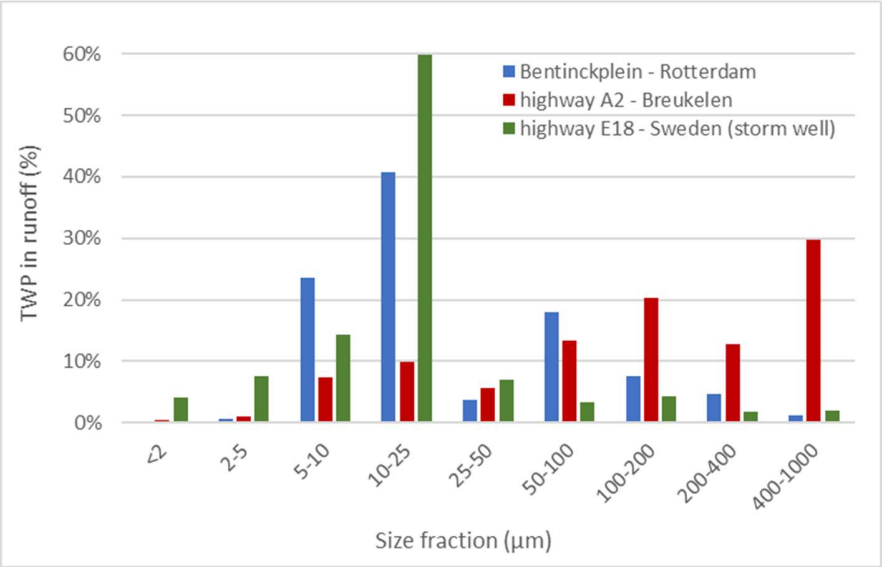

Figure 22 Particle size distribution of TWP in runoff of highway A2 near Breukelen, highway E18 in Sweden (sampled in a storm well) and Bentinckplein in Rotterdam (TWP in fraction < 2 µm is based on the LOD)

Based on emission factors (EFs) per vehicle, it is possible to calculate the expected concentrations in the runoff. EFs were calculated based on the latest TNO & Deltares factsheet on tyre abrasion emission factors (Deltares & TNO, 2022) taking into account the distribution between passenger traffic and freight traffic and differences in city traffic, highways and rural roads. For passenger cars calculated average EFs for a route with approx. 60% urban and suburban traffic and 40% motorway were similar to the values of ADAC (ADAC, 2021), based on real tyre abrasion measurements on the same route. For Rotterdam, A2 highway and E18 highway calculated average EFs were about 140 mg/km/vehicle. Calculating the expected runoff concentrations becomes possible when combining the EF with the annual average daily traffic (AADT), and the precipitation and road dimensions: concentration (mg/L) = AADT x 365 x EF / (road width x road length x precipitation). For Rotterdam, A2 highway and E18 highway in Sweden the expected concentrations are 200, 320 and 190 mg/L. All three calculated runoff concentrations are higher than the actual measured concentrations. The measured TWP concentrations within runoff for Rotterdam (52 mg/L) and the A2 highway (23 mg/L) are respectively 4 and 14 times lower than calculated. For Sweden the measured concentration is even 100 times lower. This difference stems from the fact that the calculated emission is the total emission of tyre wear, whereas the current concentrations are only the concentrations present in the runoff. For urban areas coarse TWP will end up in the sewer via runoff (appr 60%) and road side soil (appr. 40%) and the majority of fine particles (appr. 5% PM10) will disperse in the air (Geilenkirchen, 2022). Taking into account these factors, the measured value of TWP in runoff in Rotterdam approaches the calculated value for runoff (110 mg/L).

For highways it's assumed that 70% of the TWP enters the road side soil via runoff and 30% via drift (Deltares & TNO, 2022), so the measured TWP concentration at the A2 is still a factor 9 lower than the calculated value for runoff (210 mg/km/vehicle). A large part of the difference can be explained by the type of asphalt; the highway A2 consists of open graded asphalt (ZOAB). In principle, the coarser texture of the open graded asphalt road surface causes more abrasion and an increase in tyre wear. However, this porous type of asphalt is known to partially trap coarse particles that are deposited onto the road surface and therefore reduces the resuspension of coarse particles and prevents particles from washing out to the soil or surface water (Geilenkirchen, 2022). Furthermore, open graded asphalt reduces not only the spread of TWP by reducing water runoff to the verge but also by reducing splash water (Dröge et al., 2019). Based on a memorandum from Centre for Water Management (Van den Roovaart, 2000) it was estimated that the emission of (coarse) particles to the soil and surface water for open graded asphalt mixes is between 11 and 40 times lower than for closed graded asphalt mixes, provided that the asphalt is cleaned periodically. For particle emission calculations from road surfaces in the Netherlands an average reduction factor of 20 is used for open graded asphalt compared to dense asphalt. (Deltares & TNO, 2022). Taking into account these factors, the measured value of TWP in runoff along the A2 highway is in the same range of the calculated value for runoff (8 – 29 mg/L).

The highway E18 has road side gully pots to drain the runoff. As explained before approximately 60% of released TWP will be drained by the runoff system and 40% ends up in the road side soil. Furthermore, for stone mastic asphalt, Rodland et al. (2022) showed that road abrasion was 3 times less compared to dense asphalt. Taking into account these factors the calculated value for runoff (38 mg/L) is still 20 times higher than the measured value. However there are several other factors that play a role as well. Because of the presence of a runoff system, the sampling was performed in a storm water well a few meters away from the road. Part of the TWP in the storm water well will settle depending on the duration between a precipitation episode and the time of sampling. This could also be observed in the 2.5 – 6 times lower total suspended solid (TSS) concentration compared to the runoff in the highway A2 and Rotterdam (Table 8). Assuming a TSS concentration for the E18 the calculated value for run off would be 3 – 8 mg/L and approaching the measured value. The importance of the time of sampling could also be seen, by comparing the TSS and TWP concentrations found by Järlskog et al. (2022) in the same storm water well (1100 mg/L and 130 – 280 mg/L). As already explained before (paragraph 6.3), due to the analytical/quantification technique used it is likely that the TWP concentration has been overestimated.

6.5 Soil

Soil samples were collected at three roadside locations in the Netherlands and at several distances from the road, to determine the amount of TWP deposited on road side soil. Only the top layer of 5 cm was analysed, as it is expected that no migration of TWP to deeper soil layers will occur. Table 9 shows the results of the analyses for all three locations, in combination with the results of samples from Sweden and Germany (re-analysed), collected in the Microproof project, and

Table 10 shows the original results from the Microproof project (Dröge & Tromp, 2019). The TWP concentrations are calculated as mg/g dry soil and g/m^2.

Table 9 TWP concentrations in soil (in mg/g and g/m^2) on five locations in Europe, at various distances from the roadside (the soil on locations E18 and A61 were sampled in 2019 for the CEDR project Microproof and re-analysed)

Location	Bentinckplein Rotterdam		Highway A2 Breukelen			Cabauw / N201			E18 Sweden	A61 Germany
Sampling week	2	2	1	1	1	1	1	1	24	12
Year	2023	2023	2023	2023	2023	2023	2023	2023	2019	2019
Distance from road	1 m	5 m	7 m	15 m	30 m	1 m	5 m	350 m	1 m	1 m
Analyses (n)	n=4	n=1	n=1	n=1	n=2	n=1	n=2	n=1	n=3	n=7
SBR (mg/g)	0,99±0,11	0,35	0,37	0,14	0,19±0,01	0,93	0,44±0,02	0,09	0,96±0,22	5,2±1,0
NR (mg/g)	0,11±0,02	0,05	0,05	0,022	0,02±0,001	0,06	0,04±0,02	0,04	0,10±0,02	0,75±0,15
% NR in rubber	10 ± 1%	11%	13%	13%	12 ± 0%	6%	9 ± 5%	29%	9 ± 1%	13 ± 2%
TWP-BAM/ISO (mg/g)	2,2 ± 0,3	0,79	0,85	0,33	0,42±0,01	2,0	0,97±0,00	0,25	2,1±0,5	12 ± 2
TWP-TNO (mg/g)	2,7 ± 0,3	0,71	0,76	0,29	0,34±0,01	1,8	0,86±0,14	0,14	1,8±0,5	13 ± 3
% BAM/ISO vs TNO	82 ± 8%	111%	99%	114%	122 ± 2%	112%	115 ± 20%	179%	116 ± 14%	91 ± 21%
TWP average (mg/g)	2,4 ± 0,2	0,75	0,85	0,38	0,38 ± 0,01	1,9	0,94 ± 0,04	0,40	2,0 ± 0,5	13 ± 2
TWP (g/m^2)	126 ± 12	44	43	18	15 ± 1	97	32 ± 1	8,2	30 ± 5	260 ± 40

Table 10 Results from the CEDR project Microproof: TWP concentrations in soil (in mg/g and g/m²) at various distances from the roadside of the E18 (Sweden) and A61 (Germany)

Location	E18 - Sweden			A61 – Germany		
Sampling week Year	24 2019	24 2019	24 2019	12 2019	12 2019	12 2019
Distance from road	1 m	5 m	9 m	1 m	5 m	10 m
Analyses (n)	n=1	n=1	n=1	n=1	n=1	n=1
TWP (mg/g)	3.0	1.1	0.5	13	1.5	1.5
TWP (g/m2)	30	10	6.2	261	35	29

TWP can be transported from the road to the surrounding soil via several pathways: (1) direct airborne dispersion and deposition, (2) resuspension from road surfaces and deposition, (3) splash and spray from wet road surfaces and (4) road runoff during precipitation (infiltration into road shoulders). However, most pathways are relatively short and only atmospheric transport (of smaller particles) can be responsible for large distances. TWP deposited at a distance further than 30 meter from the road are usually at the background deposition level (Dröge et al., 2019). This is in agreement with the results in
Table 9, where the TWP concentrations in soil decreases at greater distances from the road and the TWP concentration at 30 mtrs from the A2 (15 g/m²) is in the same range of the background deposition level in Cabauw (8.2 g/m²).

Comparing the different locations, the roadside soil next to the A61 highway has the highest concentration of TWP at 260 ± 40 g/m², followed by Rotterdam (126 ± 12 g/m²) and Cabauw (97 g/m²). The lower TWP concentration in roadside soil at the A2 highway (43 g/m²) can be explained by the broad emergency lane of approximately 6 meters and the open graded (porous) asphalt. The effect of asphalt type and the presence of an emergency lane is shown in a study by de Best et al. (2002). This study showed a reduced amount of pollutants that is transported to the surrounding environment via runoff and via drift for a porous asphalt compared to regular asphalt. This is caused by the open structure of the road, where more water and pollutants are caught in the pores. Also, on secondary roads transported amounts of pollutants were higher than on highways, probably due to the presence of an emergency lane.

The TWP concentrations at roadside soils, in mg/gr, are in good agreement with results of Unice et al. (2013), where a total of 69 roadside surface soil samples were collected near three watersheds: Seine (France), Chesapeak (USA) and Yodo (Japan), with an average TWP concentration of 2,0 (0,1-10) mg/g. The average road side soil concentrations near the individual watersheds were 4,6 (0,1-10) mg/g (Seine, n=22), 0,7 (0,23-2,1) mg/g (Chesapeak, n=20) and 0,7 (0,14-2,9) mg/g (Yodo: n=27). In this study road side soil concentrations were grouped based on the population density and the distance to the road. For a population density >3000 person per sq. km the average TWP concentration was ca. 2,5 mg/g, with 4 mg/g at a distance less than 3 m from the road and ca. 1,2 mg/g at a distance between 3 – 15 m from the road. For a population density less than 3000 person per sq. km the average TWP concentration was ca. 1,1 mg/g, with 1,7 mg/g at a distance less than 3 m from the road and ca. 0,6 mg/g at a distance between 3 – 15 m from the road.

Also the TWP concentrations found within the Microproof project for the E18 and A61 samples (Table 10) are similar to the results found when re-analysing the samples (Table 9). **Figure 23** shows the TWP concentration in soil (mg/m²) with decreasing distance from the road side for the measurements performed within the Leon-T project and the Microproof project. In addition, the recalculated TWP concentrations, published by Muller et. al (2022) are shown. In this study, the SBR concentration in soil was determined with TED-GCMS at 0.3, 2, 5 and 300 meter from the roadside of the federal road B27. Concentrations in g/m² were derived from the mg/g values from the known sampling layer (in cm) and the assumption of a soil density of 1.8 kg/dm³. The TWP concentrations calculated by Muller et. al (2022) were based on 11.3% SBR, using the estimation of Eisentraut et. al (2018). As in the LEON-T Deliverable 3.1 (P. Tromp et al., 2022) it has already been discussed that this results in an overestimation of the TWP concentration, we recalculated the TWP concentration using the quantification method described in 5.4. Based on a similar percentage of heavy vehicles as the highway A2 (appr. 6%), the percentage of NR was assumed to be appr. 13% (Table 9).

Taking into account the average annual daily traffic intensity (AADT), the TWP load in roadside soils of the different locations are quite similar, with the exception of the highway A2 and E18. In Figure 23 the TWP concentration in road side soil at several locations with a different AADT and a different type of asphalt is shown. For the A2 the soil concentration at 1m from the roadside is calculated based on an exponential decrease further away from the road using the data for the other roads (see Figure 24). For the regular (dense graded – concrete) asphalt types there is a good correlation between the TWP concentration in soil and the AADT. For the A2 with open graded asphalt and for the E18 with stone mastic asphalt the concentrations in soil are approximately 3 times lower. For stone mastic asphalt this is in agreement with the study of Rodland et al. (2022) which also showed three times more road abrasion on concrete asphalt surface layers compared to stone mastic asphalt. However, besides the surface layer, for highway E18 also the presence of road side gully pots to drain the runoff will have an effect on the road side soil concentration. At other highways (i.e. A61) the runoff will infiltrate into the road shoulders, which is an additional contribution to the airborne deposition of TWP.

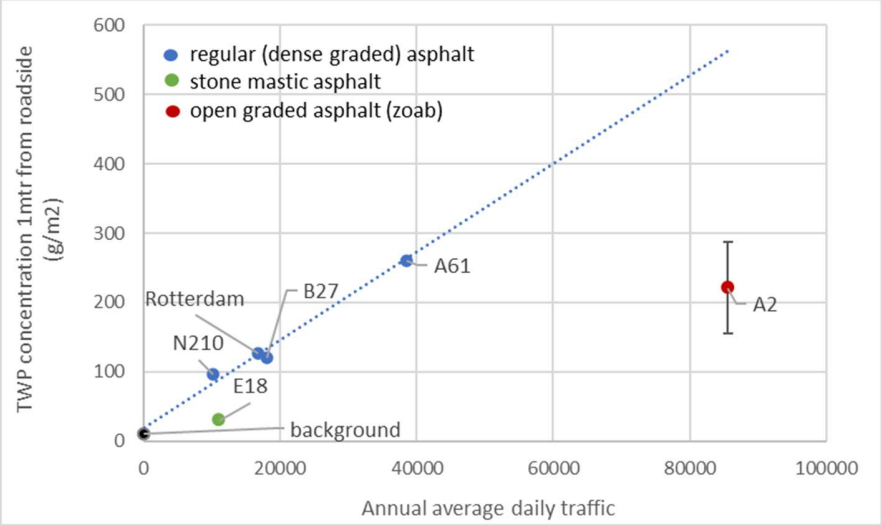

Figure 23 TWP concentrations in road side soil (mg/m2) at several locations with a different Annual average daily traffic (AADT) intensity and a different type of asphalt. For comparison the data from the monitoring campaign of the CEDR project Microproof at the test site E18 in Sweden and the highway A61 in Germany is included (Dröge & Tromp, 2019) and the measurements performed by Muller et. al (2022) at the roadside of the federal road B27 in Germany.

When comparing the results of the TWP concentration in road side soil between the highway A61 and E18 the effectiveness of the drainage system can be calculated. At a distance of 5 and 10 meter from the road the difference in soil concentration (factor 3.5 and 4.5) can be almost fully explained by the difference in traffic intensity (factor 3.5). At these distances airborne deposition of TWP is the major pathway and road runoff will not contribute. At a distance of 1 meter from the road the difference in soil concentration is a factor 9 and taking into account the difference in traffic intensity there still is 2.5 times more TWP in the soil surface next to the A61 than next to the E18. The same comparison can be made between the urban road in Rotterdam with gully pots for runoff drainage and the provincial road N210 in Cabauw, without runoff drainage system. At distances of 1 and 5 meter from the road the difference in soil concentration (corrected for the difference in traffic intensity) is still a factor 2.5. Thus in both situations, highways and (sub)urban roads, approximately 60% of TWP in road runoff is drained via road side gully pots. This is in agreement with the findings of Geilenkirchen (2022).

Figure 23 shows that the background concentration at Cabauw (350mtr from the roadside; 8.2 g/m^2) is similar to the background concentrations determined by Muller et. al (300mtr from the roadside: 12 g/m^2). Probably the concentration at Cabauw and other locations is a bit higher because Muller et. al (2022) demonstrated that the soil layer deeper than 5 cm also contains minor amounts of TWP (appr. 20-25%).

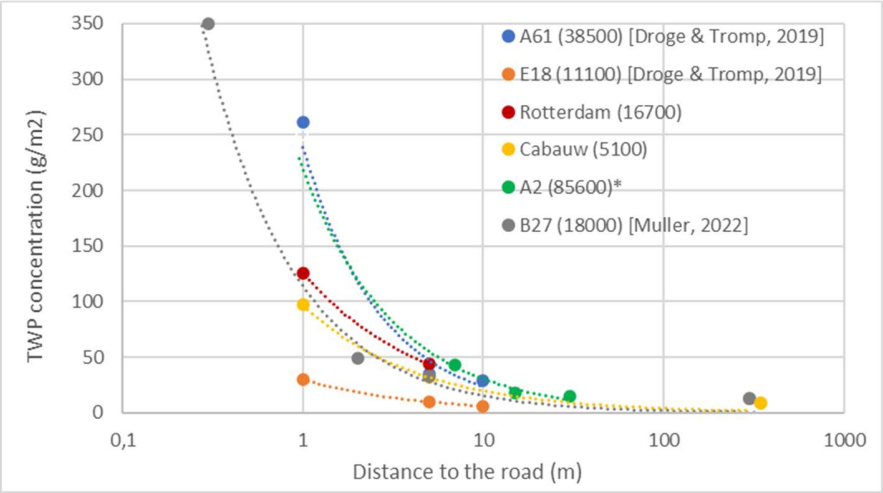

Figure 23 TWP concentrations in soil (mg/m2) at different distances from the road side for the city of Rotterdam, rural location Cabauw and the highways A2. For comparison the data from the monitoring campaign of the CEDR project Microproof at the test site E18 in Sweden and the highway A61 in Germany is included (Dröge & Tromp, 2019) and the measurements performed by Muller et. al (2022) at the roadside of the federal road B27 in Germany. In the labels the annual average daily traffic intensity in vehicles per day is presented.

Figure 24 shows the particle size distribution of TWP in soil at the A61 and the E18. For the highway E18, the particle size distribution in road side soil is similar to the distribution in runoff sampled from the storm water well, with a large share of TWP particles in the fine fraction. The particle size distribution of the highway A61 is very similar to the runoff and deposited dust size distribution of the A2, were the coarse fraction dominates. Compared to the TWP particle size distribution of Muller et. al (2022), determined in roadside soil from the federal road B27 in Germany, the fraction of 'fine' particles (< 50µm) is much higher in roadside soil sampled at the A61 (Germany) and E18 (Sweden) (see Table 11). Most likely this is due to the sieving method; with dry sieving agglomerated 'fine' particles will end up in coarse sieve fractions, while with wet sieving these agglomerated particles break up into single 'fine' particles.

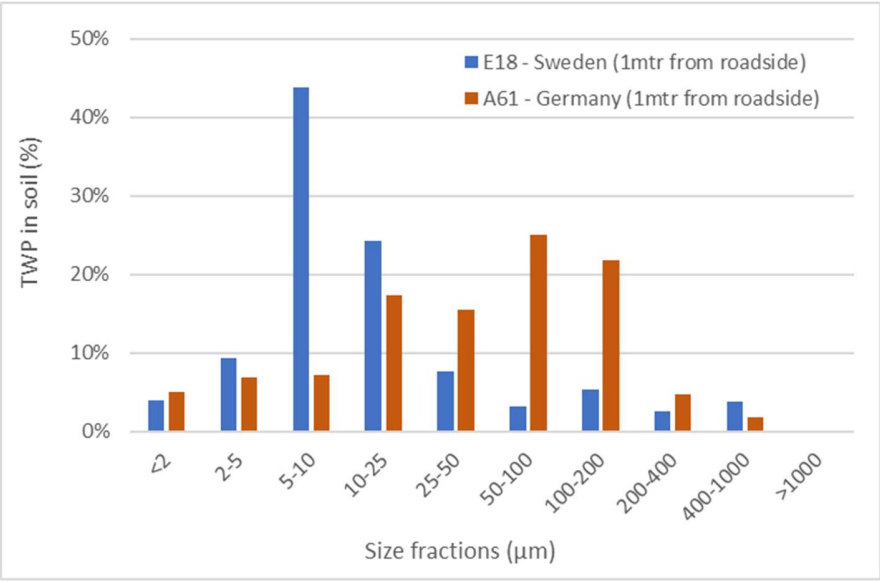

Figure 24 Particle size distribution of TWP in soil 1 mtr from the road side along highway E18 in Sweden and highway A61 in Germany (TWP in fraction < 2 µm is based on the LOD)

Table 11 Comparison of particle size distributions of TWP in roadside soil at the E18, A61 and B27 (Muller et.al, 2022).

Fraction (µm)	Wet - sieving E18	A61	Dry-sieving B27
400-1000	27%	9%	35%
100-400	24%	39%	36%
50-100	18%	12%	22%
<50	31%	39%	8%

From the soil concentration in g/m^2 at different distances from the road the total amount of TWP in the verge in a 1 km stretch of 30 meters wide is calculated for each location by extrapolation (
Table 11). As stated before, the TWP levels further than 30 meter from the road are usually at the background deposition level (Dröge et al., 2019). For comparison, the total potential emission from the roads, based on average EFs, derived from the latest TNO & Deltares factsheet (Deltares & TNO, 2022) and the traffic intensity (AADT) are given. The total emission is corrected for PM10 that will disperse at longer distances (5% reduction), the presence of a runoff drainage system (60% reduction), open graded asphalt (90% reduction) and stone mastic asphalt (67% reduction). As can be seen in
Table 11, the accumulation: measured TWP concentration divided by the calculated yearly emission that end up in road side soil, is 2 – 3 years, with the exception of the highway A61, which is less than a year. The main reasons for that are most likely the higher EF due to the higher share of heavy traffic (22%) on the A61 compared to the other roads and the higher proportion of natural rubber (due to heavy traffic) that degrades faster than SBR (see Deliverable 3.3).

Table 11 Theoretical number of years of accumulation of TWP on road side soil surface, from comparison of potential road side soil concentrations based on calculated EFs and measured (extrapolated) road side soil concentrations (30 meters wide) for different locations.

Location	Average EF	AADT	Total TWP emission	Road side soil TWP	Measured road Side soil TWP	Accumulation
	mg/km/vehicle	vehicles/day	kg/km/yr	kg/km/yr	kg/km	Years
A61	167	38500	2300	2200	1500	1
E18	140	11100	570	72	240	3
A2	142	85600	4400	470	1500	3
Rotterdam	142	16700	860	490	970	2
Cabauw	101	5100	370	360	770	2

The accumulation of TWP derived from EFs are most likely an underestimation as these EFs are purely based on measurements of mass loss of the tyres. Not all mass loss is related to the emission of TWP, also the emission of (semi-) volatile compounds from the additives in tyres play an important role. As can be seen in Deliverable 2.3 actually the emission of (semi-)volatiles from tyres is approximately 5 – 10 times higher than the emission of TWP. In addition, a part of the emitted TWP will stick to the asphalt and will end up in the road-side soil at all. Thus, the potential road side soil concentration is lower than calculated from the EFs, which contributes to an extra underestimation of the accumulation.

Also, the comparison of deposition values (in mg/m^2/day) and soil concentrations (mg/m^2) on the same location can give some insight into the accumulation and degradation processes of deposited TWP (see Table 12). Again, this can only be considered as a very rough estimate because deposition sampling was only performed in two periods in 2022 and the prevailing meteorological conditions during these sampling periods may differ from average long-term conditions. As deposited dust concentrations are highly dependent on these meteorological condition (i.e. wind speed/direction, precipitation) the outcomes of these calculations have a high degree of uncertainty. In addition, the TWP concentrations in road side soil closest to the road are not only the result of atmospheric deposition but are also influenced by runoff. Therefore, for the provincial road N210 in Cabauw and the highway A2, TWP concentrations are corrected to exclude the runoff (60%) and only take into account atmospheric deposition. The urban road in Rotterdam has a runoff drainage system, so in this location TWP in road side soil is not affected by runoff. Unlike the N210, for the highway A2, with the open graded asphalt and a broad emergency lane, it's not

clear what the percentage of runoff is in road side soil, so in Table 12, corrected values are between parenthesis. Another factor that can have an effect on the result is the difference in height of the deposition samplers (appr. 1m) compared to the height of the soil surface. Especially for deposition samplers near the road (1m distance: Rotterdam and N210) this could give an underestimation of the deposited dust values and overestimation of the accumulation. Based on the difference in soil concentration between 1 and 5 m distance from the road, the overestimation is a maximum of a factor of 3.

Table 12 Theoretical number of years of accumulation of TWP on road side soil surface, from comparison TWP levels in soil and deposited dust on the same location

Location	Street	Distance from the road side (mtr)	TWP in soil (g/m^2)	Deposition rate TWP (mg/m2/day)	Accumulation (years)
Rotterdam	urban street	1 mtr	126 ± 12	17-23	13 – 22
Breukelen	highway A2	7 mtr	43 (17) [1]	1.5-4.0	29 – 76 (12 – 31)
		20 mtr	15	0.38-1.1	39 – 107
		50 mtr	8,2	0.35-0.61	36 – 63
Cabauw	provincial road	1 mtr	97 (39) [1]	2.5-3.7	72 – 105 (29 – 42)
	N210	350 mtr	8.2	0.11-0.8	28 – 98

1) For road side soil at 1 m distance from the N210 and A2 (between parenthesis) the TWP concentrations are corrected to exclude the runoff (60-70%) and only take into account the airborne deposition.

The accumulation derived from the road side soil and deposition samples (Table 13) is higher than the accumulation derived from the EFs (Table 12). The average accumulation derived from EFs is 2 (± 1) years and from soil/deposition samples it's around 40 (± 30) years. Although both methods give different results, it has to be stated that they are rough estimates and the accumulation derived from EFs have to be considered as underestimates. Apart from this, a first insight is given on the possible degradation of TWP in road side soil. Moreover, the accumulation is in the same range of the calculated accumulation from the UV aging experiments in Deliverable 3.3.

6.6 Surface water

Water samples were collected in rivers by RWS with sedimentation boxes and flow-through centrifuges. In ditches along the road side of highway A27 and highway A2 TNO performed the sampling with cascade sieving for the coarse fraction (>25µm); remaining water was collected in glass bottles for analysis of the fine fraction (<25µm). Resulting TWP concentrations are summarized in

When applying the SBR-BR conversion factor of current study to the measured SBR-BR levels of Rauert et al. (2022) and taking into consideration that sampling was performed in the dry season, TWP concentrations are in the same range (26 (3 – 130) µg/L). In another recent study by Kittner et al. (2022) microplastic levels on 18 locations in the Danube river catchment were investigated. Applying the conversion factor of current study, TWP concentrations varied between 0.2 – 2.6 mg/g with an average concentration of 0.57 mg/g suspended solids, which is in line with current study with TWP levels ranging from 0.37 – 1.0 mg/g.

Table 13 in mg/g suspended matter and µg/L water. Results show that the concentration of TWP varies per type of waterway, and was found to range from 0.55-30 µg/L for the rivers Rhine, Meuse and New Meuse, and from 3.8-6.5 µg/L for the ditches along the A2 and A27 highways. Unice et al., (2019a) used a mass balance model to predict the concentration of TRWP in subcatchments of the river Seine. The calculated concentrations of TRWP was between 3.7 and 120 µg/L, which translates to a TWP concentration between 1.85 and 60 µg/L, which is in the same range as results found in the current study. TRWP concentrations found by Rauert et al. (2022) in surface water at 21 different locations in Queensland are considerable higher, ranging from approximately 9 – 1990 µg/L with an average of 230 µg/L (TWP: 110 (4 – 1000) µg/L). However they used a SBR+BR rubber content in tyres of 4.5 – 13% to calculate TWP

concentrations, while in this study we use a rubber content of 45% with ca. 35% of SBR+BR, based on own measurements and reported values in literature (see Deliverable 3.1).

When applying the SBR-BR conversion factor of current study to the measured SBR-BR levels of Rauert et al. (2022) and taking into consideration that sampling was performed in the dry season, TWP concentrations are in the same range (26 (3 – 130) µg/L). In another recent study by Kittner et al. (2022) microplastic levels on 18 locations in the Danube river catchment were investigated. Applying the conversion factor of current study, TWP concentrations varied between 0.2 – 2.6 mg/g with an average concentration of 0.57 mg/g suspended solids, which is in line with current study with TWP levels ranging from 0.37 – 1.0 mg/g.

Table 13 TWP concentrations in three rivers in the Netherlands (Rhine, New Meuse and Dordtsche Kil) and two road side ditches along the highways A2 and A27 (in µg/g and µg/L)

Location	Lobith				Eijsden		Rotterdam	A2-Breukelen	A27-Utrecht
Waterway	Rhine				Meuse		New Meuse	Ditch	Ditch
Sampling period	15-7-2009	31-1-2018	13-1-2021	6-4-2022	14-6-2005	26-6-2018	18-5-2021	3-4-2023	26-1-2023
Analyses (n)	n=1	n=1	n=2	n=2	n=1	n=1	n=1	n=1	n=1
SBR (mg/g)	0.48	0.28	0.37 ± 0.00	0.21 ± 0.05	0.11	0.49	0.22	0.34	0.18
NR (mg/g)	0.02	0.02	0.05 ± 0.01	0.03 ± 0.00	0.01	0.02	0.02	0.03	0.03
% NR in rubber	4%	8%	13 ± 1%	14 ± 5%	10%	4%	8%	8%	16%
TWP-BAM/ISO (mg/g)	1.0	0.60	0.84 ± 0.02	0.48 ± 0.09	0.23	1.0	0.49	0.73	0.42
TWP-TNO (mg/g)	0.83	1.1	0.82 ± 0.00	0.57 ± 0.14	0.51	1.0	0.39	1.0	0.40
% BAM/ISO vs TNO	121%	57%	103 ± 3%	86 ± 6%	46%	100%	125%	72%	107%
TWP (mg/g)	0.91	0.83	0.83 ± 0.01	0.53 ± 0.12	0.37	1.0	0.44	0.87	0.55
solids (mg/L)	6.0	36	16	16	4.0	5.5	1.25	4.4	12
TWP (µg/L)	5.5	30	13 ± 0.2	8.5 ± 1.9	1.5	5.6	0.55	3.8	6.5

The concentration of TWP in rivers and ditches is highly dependent on the amount of suspended solids within the waterways. Rijkswaterstaat (Min. I&W, 2023) analysed the suspended solids within multiple rivers in The Netherlands, and reported SBR and NR concentrations. From these rubber concentrations the total TWP concentration was calculated according to the methodology in section 5.4. which results in concentrations ranging from 0.20-0.82 mg/g and 0.60-5.1 µg/L. The concentrations found within the current study are similar, ranging from 0.37-1.0 mg/g and 0.55 – 30 µg/L. Figure shows the TWP concentration in mg/g suspended solids for the samples in current study, along with calculated concentrations from SBR and NR values reported by Rijkswaterstaat (Min. I&W, 2023). In addition to Figure 25, Table 14 shows the average TWP concentrations found within several waterways in mg/g and µg/L, along with the share of TWP in the total microplastic load in suspended solid. In the rivers Meuse, Rhine and New Meuse, the share of TWP in the total microplastic load (PE, PP, PET, PS, PA, PMMA, SBR, NR) is similar and ranges between 18 – 26%. The most dominant microplastic, with the highest concentration in surface water, is polyethylene (PE); 76% of the total amount of microplastic consists of PE.

Table 14 Average TWP concentrations in various rivers and ditches in the Netherlands (in µg/g suspended solid and µg/L), including the share of TWP of the total microplastic concentration. Results are largely based on the measurements performed by Rijkswaterstaat (*) (Min. I&W, 2023).

Location - waterway	No. of samples	SBR (mg/g)	NR (mg/g)	TWP (mg/g)	TWP (µg/L)	%NR	%TWP in total microplastics

Eijsden - Meuse	2 + 2*	0,29 ± 0,14	0,04 ± 0,03	0,70 ± 0,23	3,6 ± 2,9	15 ± 11%	26 ± 8%
Lobith - Rhine	4 + 5*	0,22 ± 0,13	0,03 ± 0,02	0,53 ± 0,23	14 ± 11	15 ± 8%	18 ± 2%
Rotterdam - New Meuse	1 + 23*	0,16 ± 0,06	0,02 ± 0,01	0,37 ± 0,13	1,3 ± 0,9	13 ± 6%	22 ± 6%
A2 (Breukelen) - Ditch	2	0,34	0,03	0,74 ± 0,19	4,2 ± 0,5	8%	
A27 (Utrecht) - Ditch	1	0,18	0,03	0,55	6,5	16%	

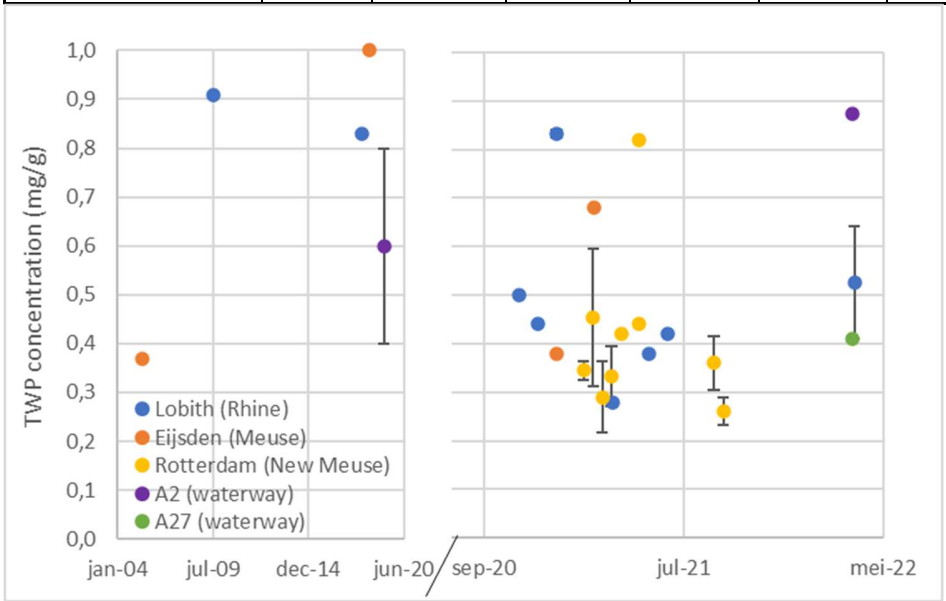

Figure 26 TWP concentrations in various rivers and ditches (in mg/g), measured between 2005 – 2022. Besides measurements for LEONT (between 2021 – 2022), results are taken from the CEDR project Microproof (between 2005 – 2020) (Dröge & Tromp, 2019) and are derived based on extensive measurements performed by Rijkswaterstaat (between 2020 - 2021) (Min. I&W, 2023).

The variability (RSD) of the TWP concentrations, expressed as mg/g suspended matter is 44%; when converted to µg/L surface water the variability is a factor of 4 – 5 higher (180%). The small variability of TWP and microplastics in suspended matter can be explained by adsorption of these polymers to the organic fraction of suspended matter, which makes the concentrations less dependent on the flow rate and depth of the surface water. The particle size distribution of TWP and suspended matter for the collected ditch samples is shown in Figure 25. Within the ditch samples, 60-70% of TWPs exists in the fraction <25 µm. The TWP size distribution is similar to the size distribution of suspended matter, which is in agreement with the findings reported by Rijkswaterstaat (Min. I&W, 2023).

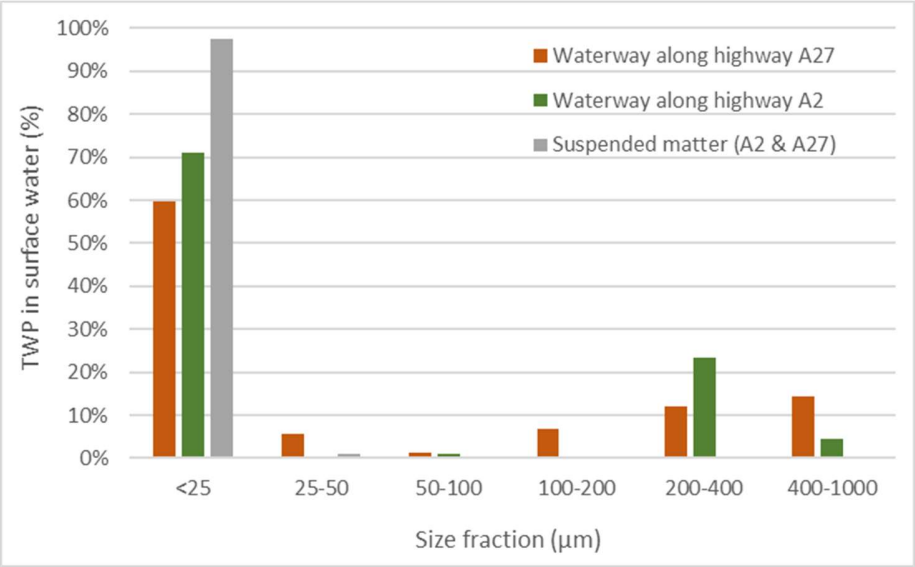

Figure 25 Particle size distribution of TWP and suspended matter in two waterways along highway A27 and highway A2 (TWP in fraction < 2 µm is based on the LOD)

6.7 Sediment

Sediment samples were collected in rivers by RWS and in ditches along the road side of highway A27 and highway A2 by TNO. Resulting TWP concentrations are summarized in
Table 15 in mg/g dry weight.

Table 15 TWP concentrations (in mg/g) in four sediment samples from two rivers New Meuse and Hollands Diep and two road side ditches along the highways A2 and A27

Location	Rotterdam	Rotterdam	A2-Breukelen	A27-Utrecht
Waterway	New Meuse	Hollands Diep	Ditch	Ditch
Sampling date	6-12-2021	12-4-2021	8-10-2022	9-10-2022
Analyses (n)	n=1	n=4	n=1	n=1
SBR (mg/g)	0.30	0.31 ± 0.12	0.24	0.67
NR (mg/g)	0.02	0.01 ± 0.01	0.05	0.05
% NR in rubber	6%	4 ± 2%	16%	7%
TWP-BAM/ISO (mg/g)	0.64	0.65 ± 0.25	0.58	1.4
TWP-TNO (mg/g)	0.45	0.66 ± 0.15	0.41	1.2
% BAM/ISO vs TNO	144%	97 ± 19%	142%	116%
TWP (mg/g)	0.54	0.65 ± 0.20	0.49	1.3

The TWP concentrations found within the sediment of the rivers New Meuse and Hollands Diep are 0.54 and 0.65 ± 0.20 mg/g respectively. The concentrations within sediments in the ditches along the A2 and A27 highways were found to be 0.49 and 1.3 mg/g respectively. The concentration of TWP within sediments was also analysed for the ditch along the A2 highway in Microproof (Dröge & Tromp, 2019), yielding a similar concentration, at 0.3 mg/g. Concentrations reported by Min. I&W (2023) for sediment in rivers were found to vary depending on the location, but are on the same scale at 0.007 - 0.84 mg/g (calculated from reported SBR concentrations). Table 16 shows the average concentration of TWPs within sediments, along with the contribution of TWPs to the total microplastic concentration.

Table 16 Average TWP concentrations in sediments from various rivers in the river delta and ditches in the Netherlands (in mg/g), including the share of TWP of the total microplastic concentration. Results are largely based on the measurements performed by RWS (*).

Location	No. of samples	SBR (mg/g)	NR (mg/g)	TWP (mg/g)	%NR	%TWP in total microplastics
River Delta Netherlands	2 + 17*	0.12 ± 0.11	0.01 ± 0.01	0.24 ± 0.22	7 ± 6%	20 ± 10%
A2 (Breukelen) - Ditch	2	0.24	0.05	0.40 ± 0.14	16%	
A27 (Utrecht) - Ditch	1	0.67	0.05	1.3	7%	
River banks Netherlands	9*	0.09 ± 0.08	0.01 ± 0.01	0.20 ± 0.17	14 ± 15%	16 ± 6%

The reported contribution of SBR rubber to the total microplastics concentration by Min. I&W (2023) is 7%. When accounting for the SBR content in TWP, the values in LEON-T are similar to the reported values. The average TWP concentrations in sediments are equal to those in suspended matter. Due to the sedimentation of suspended solids from surface water, the amount (kg) of microplastics in sediment is increasing. But there is no measurable increase in microplastics concentrations (g/kg) in sediments or banks compared to microplastics concentrations in suspended solids. Also the size distribution of TWP in sediment is similar to the size distribution in suspended matter. Figure 26 shows the particle size distribution for the A2 and A27 waterways, along with a sample from Hollands Diep.

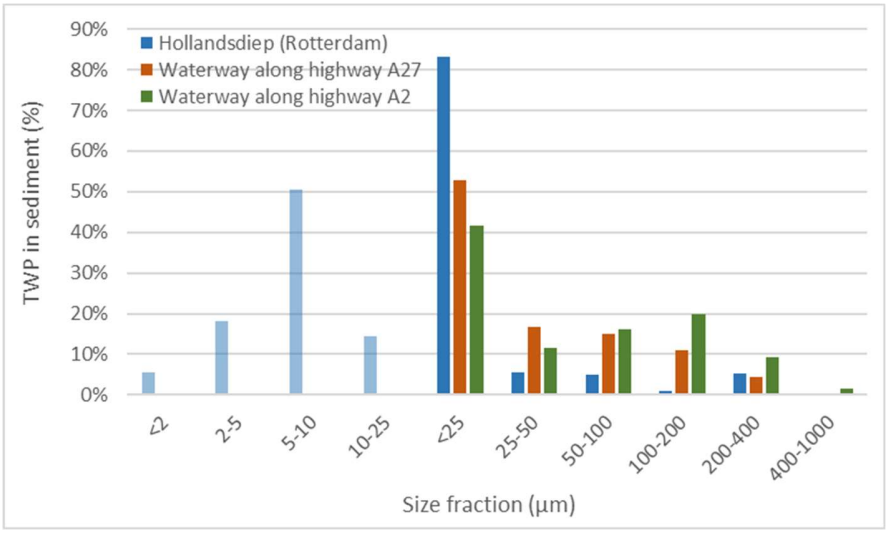

Figure 26 Particle size distribution of TWP in sediments of the river Hollands Diep and two waterways along highway A27 and highway A2.

A monitoring study performed by Unice et al. (2013) in the surficial sediment of watersheds in the Seine (France), Chesapeake (USA), and Yodo-Lake Biwa (Japan) watersheds, with ca. 150 sampling points, show higher TWP concentrations in sediments from the Seine (2.2 (0.03 – 5.8) mg/g), but comparable results in the sediments from Chesapeake (0.42 (0.05 – 2.2) mg/g) and Yodo-Lake Biwa (0.38 (0.03 – 2.3) mg/g) as in current study. In general , maximum concentrations were observed downstream of high population density urbanized areas. Most likely, the higher concentrations in sediments from the Seine could be explained by a higher traffic density and more urbanized areas. This is supported by the study of Kumata et al. (2002) which showed higher TWP concentrations in river sediments in proximity to heavily trafficked areas, in rivers receiving storm water run-off. Prior studies suggesting even higher TWP concentrations, ranging from 0.3 – 55 mg/g, however these studies are based on different tyre markers (e.g. benzothiazoles, organic zinc) with concerns about marker specificity & stability under environmental conditions (Unice et al., 2013).

Wagner et al. (2018) suggested that TWP concentrations in sediment are higher than those in surface water, because particles settle down and accumulate in the sediments. This seems valid for rivers and surface waters receiving direct storm water run-off with a certain proportion of coarse particles. Also in current study near the highway A27 in a ditch receiving direct storm water run-off the TWP concentration in sediment (1.3 mg/g) was

higher than in surface water (0.44 mg/g). However, as mentioned before in Dutch rivers receiving no direct road run-off TWP concentrations in surficial sediments are similar to concentrations in suspended solids.

6.8 Physicochemical characterization

6.8.1 Morphology & density

Table 17 shows the volume percentages of encrustations of road wear particles in TRWP, determined with SEM-EDX. In Figure 27 the SE-images and BSE-images of some example particles are shown; the BSE-images are used for image analyses to calculated the percentage of road wear encrustations in TRWP. In total 53 particles in deposition samples and ambient air samples were analysed, with particle sizes ranging from 1 to 50 µm. The encrustations are a mixture of minerals (road and soil dust) and metals (mainly iron(oxides)). The percentages of these encrustations range from 5 – 56 % with an average of 29 %. This is similar to Sommer et al. (2018) with a percentage between 10 – 50% based on SEM-EDX analyses of 171 'super-coarse' airborne TRWP in the size range of 10 – 80 µm collected near a highway (A555) and provincial road (B31). Tyre abrasion particles attract fine road dust once they are deposited on the road surface, due to their flexibility, shape (rounded cross section) and morphology (rough surface). Sommer et al. (2018) reported a lower percentage of encrustations in TRWP collected near a highway (10%) than near a provincial road (50%), which was explained by the difference in driving speed. On roads with high velocities (e.g., motorways), a more efficient removal of material from the road surface due to vehicle-induced turbulence is observed and therefore, less road dust can be accumulated on the tire-abrasion particles. In current study this effect could not be observed; actually the percentage of encrustations in TRWP collected near the urban street in Rotterdam (28 %) was slightly lower than the particles collected near the highway A2 (33 %). There is no clear explanation for this difference, however it could be related to the difference in asphalt (the highway A2 has open graded asphalt instead of dense graded asphalt) and/or the analysed particle sizes (current study: 1 – 50 µm vs Sommer et al. (2018): 10 – 80 µm). Moreover, the results in Table 18 show an increase in percentage of incrustations with increasing particle size. This is also the reason why the percentage of road wear encrustations on the background location (24 %) is a bit lower than the other two locations. Particle sizes of TRWP were smaller, because the larger particles will deposite near the road and will not reach the background location further away from the road.

Table 17 Percentage of encrustations (road wear) in TWP in ambient air from single particle analysis with SEM-EDX

PM fraction / location	Particles analysed	Encrustations (%)			Density (g/cm3)
		mean ± SD	geomean	median	
25-50 µm	11	33 ± 10	32	32	1.8
10-25 µm	30	31 ± 11	29	30	1.8
5-10 µm	5	29 ± 10	27	29	1.8
2-5 µm	6	13 ± 4	13	13	1.7
1-2.5 µm	1	5	5	5	1.5
total TWP 1-50 µm	53	29 ± 12	26	30	1.8
urban street Rotterdam	30	28 ± 12	24	29	1.7
highway A2	16	33 ± 11	32	30	1.8
background Cabauw	7	24 ± 12	21	20	1.7

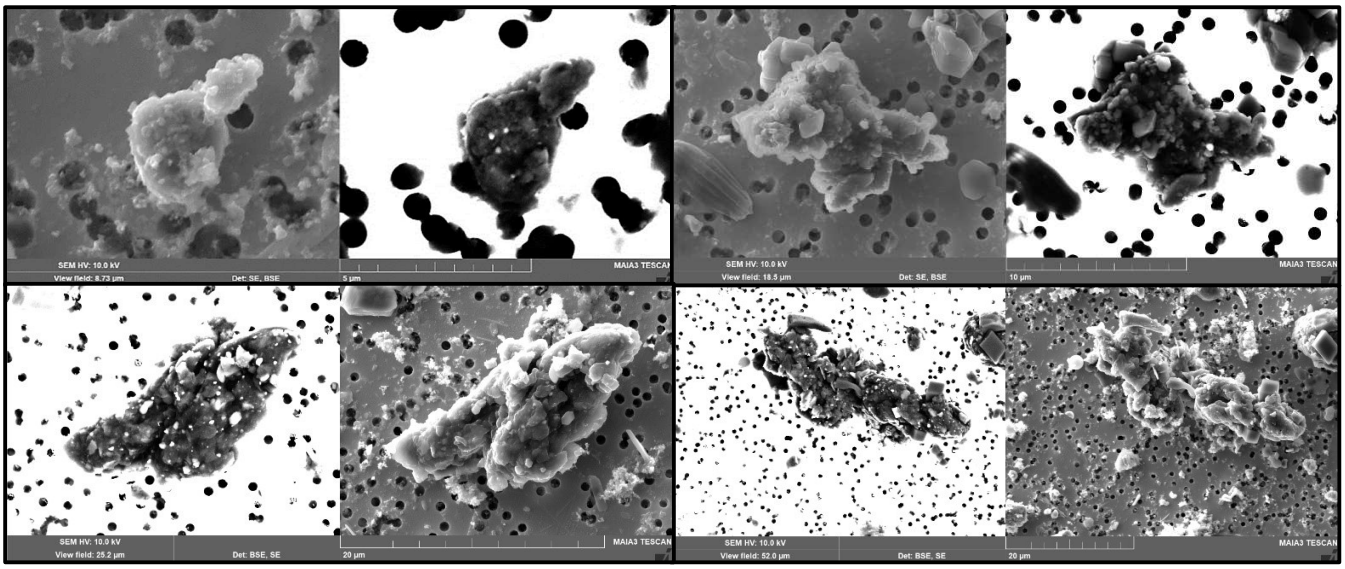

Figure 27 SEM images from TWP of different size classes: 2-5 µm (top left), 5-10 µm (top right), 10-25 µm (bottom left) and 25-50 µm (bottom right). On the BSE images (with white background) the encrustations are visible as light grey and white spots.

The amount of encrustations has also an effect on the density of the particles. EDX-analysis show that incrustations contain 10% iron(oxides) and 90% quartz & clay minerals; based on these findings we calculated a density of around 2.4 g/cm^3 for the incrustations. Based on the composition of tyres (45% rubber, 24% carbon black, 15% silica, 2% ZnO and 14% oils & organic additives) we calculated a density of around 1.5 g/cm^3 for pure TWP. This translates to a density range of 1.5 – 2.0 g/cm^3 for all analysed particles with an average of 1.8 g/cm^3. This is in agreement with other reported density values of TRWP between 1.3 and 2.2 g/m3 (Kayhanian et al., 2012; Sommer et al., 2018).

From current study and the study of Sommers et al. (2018) the average volume percentage of road wear encrustations in all analysed TRWP is approximately 30% corresponding to a mass percentage of approximately 40%. Unice et al. (2013) and Panko et al. (2013) assume a slightly higher mass percentage of 50% to calculate the concentration of TRWP from TWP. This conversion factor is also used in the ISO Standards for sediment, soil and ambient air (ISO/TS 20593 and ISO/TS 21396). Moreover, the average volume percentage of TRWP in PM_{10} is 20% corresponding to a mass percentage of approximately 30% what varies more with the conversion factor applied in the ISO Standards.

6.8.2 Composition

Tyres consist of a rubber mixture of different kind of polymers: SBR, BR and NR. The average rubber percentage in tyres is approximately 45%, based on the average of published data in literature (Baensch-Baltruschat et al., 2020; Grigoratos et al., 2018; Rødland, Samanipour, et al., 2022; Unice et al., 2019a; Wagner et al., 2018) and own data from the physicochemical characterization of 35 car & truck tyres (P. Tromp et al., 2022). There is a difference in composition between car tyres and truck tyres: truck tyres mainly consist of NR and car tyres mainly consist of SBR and BR. The current freight traffic contribution in NL is approximately 7% (CBS, 2023); this is comparable with data in literature, with an estimated contribution between 6 – 9% (Goßmann et al., 2021). Table 18 shows the amount of NR found in samples from different locations and in the different environmental matrices. From the percentage of natural rubber in analysed environmental samples collected at the road sides, insight can be obtained about the share of freight traffic at certain locations.

Table 18 Percentage of natural rubber (NR) in rubber (SBR+BR+NR) of TWP collected at different locations and from different environmental compartments

Location / environmental compartment	No. of analysis	Freight traffic (%)	Natural rubber (%)		
			mean ± SD	geomean	median
Background Cabauw	28	8.0	14 ± 11%	10%	11%

Urban street Rotterdam	52	6.0	14 ± 9%	11%	11%
Highway A2 - Breukelen	58	13.2	19 ± 13%	16%	15%
Highway A27 - Utrecht	17	11.4	20 ± 14%	15%	19%
Highway E18 - Sweden	23	13.2	19 ± 14%	15%	16%
Highway A61 - Germany	17	22.2	14 ± 6%	13%	13%
Ambient air	43		10 ± 6%	9%	8%
Runoff	45		21 ± 15%	16%	17%
Deposited dust	41		24 ± 12%	21%	22%
Soil	43		13 ± 6%	12%	11%
Sediment	13		13 ± 8%	11%	11%
Surface water	28		13 ± 12%	9%	9%

In general the percentage of NR at locations near highways is higher than urban street and background locations. The highway A61 is the exception with a lower percentage of NR compared to other highways. This could be an indication that there is less freight traffic, however the percentage of freight traffic is even higher than on the other highways. The main reason for this is the fact that the NR percentage is only based on soil samples, without runoff and deposited dust. As can be seen in Table 18, in runoff and deposited dust samples the share of NR is higher than in other environmental matrices. This clear difference can be explained by UV- and biodegradation of TWP. As shown in Deliverable 3.3, natural rubber degrades faster than SBR, which reduces the proportion of NR in the rubber matrix in soil, sediment and surface water samples. In contrast, in runoff and deposited dust samples, which contain mainly freshly generated TWP, no degradation has occurred yet. However, the percentage of NR in TWP collected in ambient air samples is lower than in runoff and deposited dust samples, despite the fact that also here the proportion of freshly generated TWP is high. This can be explained by the lower share of NR when particle sizes decrease, as shown in Figure . From the environmental samples treated with cascade sieving and filtering the percentage of NR in each size fraction is be determined. Because in ambient air the majority of TWP is smaller than 10 µm, the percentage of NR is low. As the percentage of NR in TWP decreases with particle size and freight traffic is the main emission source of NR, one could reason that trucks emit more coarse TWP than cars.

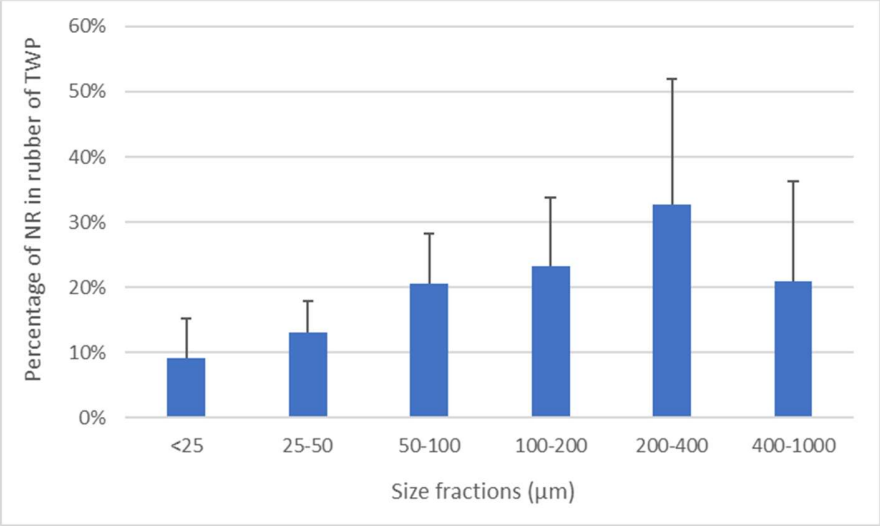

Figure 30 Percentage of natural rubber (NR) in rubber (SBR+BR+NR) of TWP with different size classes (µm).

As can be seen in Figure 31 there is a good correlation between the percentage of freight traffic and the amount of NR in TWP collected in ambient air samples and deposited dust samples near the road side. In these samples mainly freshly generated TWP are collected, which makes the amount of freight traffic a good predictor of natural rubber in TWP. As for TWP collected in soil degradation processes affect the amount of NR in TWP the correlation between freight traffic and natural rubber is poor.

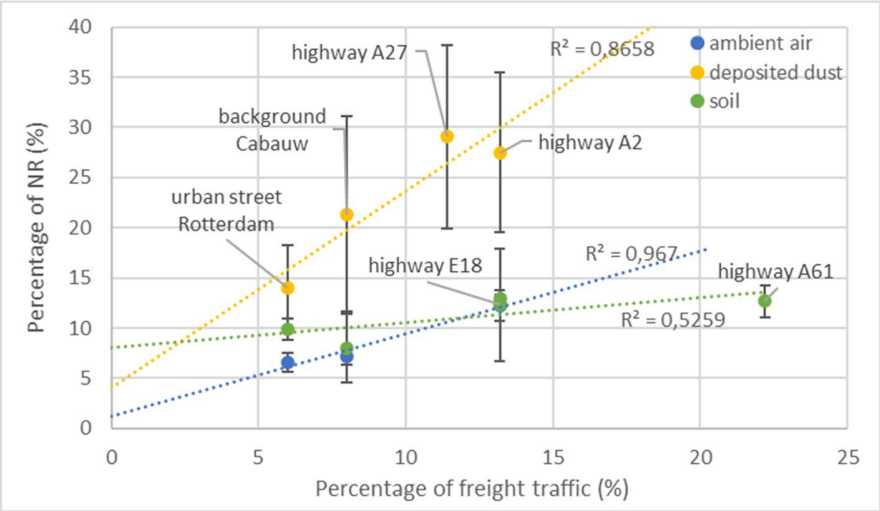

Figure 31 Percentage of natural rubber (NR) in rubber (SBR+BR+NR) of TWP found in environmental samples collected at road side locations with a different percentages of freight traffic.

6.8.3 Particle size distribution

Within LEON-T, several environmental samples (runoff 'R' , deposited dust 'D' and soil 'S') have been fractionated to determine the mass size distribution, shown in Figure 28. For each location an average size distribution has been determined based on the available environmental samples that have been fractionated. In general, the particle size ranges are in agreement with other published data (i.e. Kovochich et al., 2021; Kreider et al., 2010, Wagner et al., 2018) that report average sizes (mode) between 50 – 100 µm, with lower and upper limits of around 5 and 350 µm. It can be seen that the particle size distribution actually is bimodal with a peak between 5 – 25 µm and 50 – 200 µm. Also this has been reported before in other studies with a bimodal mass size distributions with one peak in the coarse fraction and another one in the fine fraction (Fukahori & Yamazaki, 1994; Gustafsson et al., 2008; Wang et al., 2017). A bimodal size distribution of TWP can be explained from the different abrasion mechanisms: small particles (<PM_{10}) originate from micro vibration and larger particles in the range of a few hundred micrometers are attributed to stick-slip motion (Fukahori & Yamazaki, 1994).

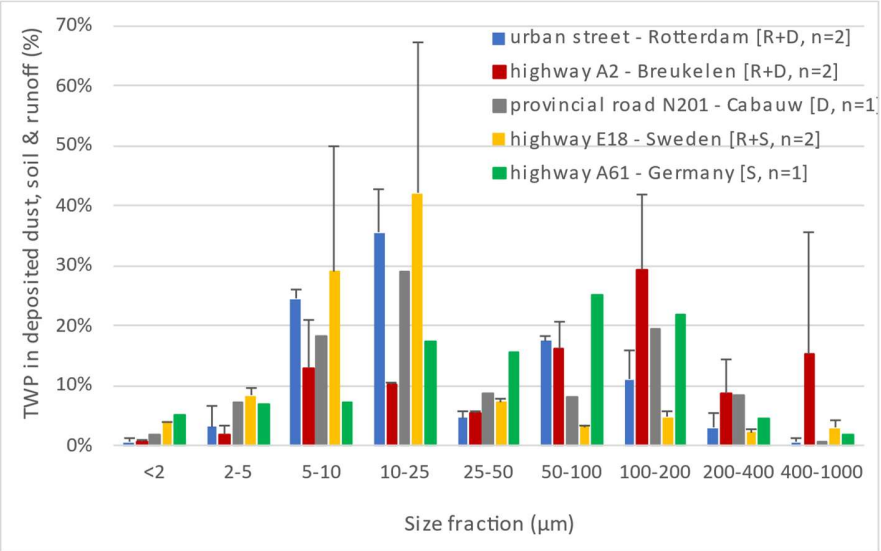

Figure 28 Average particle size distributions of TWP in deposited dust [D], soil [S] and runoff [R] on various road side locations with different traffic and road surface characteristics (TWP in fraction < 2 µm is based on the LOD)

For highway E18, urban road Rotterdam and provincial road Cabauw the amount of fines (<25µm) are respectively 5, 2 and 1.5 times higher than coarse TWP particles. For highway A61 and highway A2 it's the other way around, with 1.5 and 2.5 times higher amounts of coarse particles than fine TWP's. The difference between the two extremes (E18 and A2) could be explained by the difference in road surfaces. The road surface of highway E18 is very smooth and made of stone mastic asphalt, while the road surface of the highway A2 is made of 'open graded (porous) asphalt' (ZOAB) with a very rough structure. The other roads (A61, N201, urban street) all consist of dense graded asphalt with probably little differences in (micro)structure. The road surfaces of the urban street and the N201 appear to be rather smooth, just like the E18, which could explain the higher amount of fines. The road surface of the A61 was not visually inspected. As explained before in section 6.2 – 6.5, open graded asphalt is known to partially trap particles that are deposited onto the road surface and therefor reduces the resuspension of these particles and prevents that particles are washed to the soil or surface water via runoff (Geilenkirchen, 2022; Svensson et al., 2023). The shift to a coarser particle size distribution for run-off and deposited dust could be very well explained by the entrapment of finer particles by the pores of the asphalt layer on the highway A2.

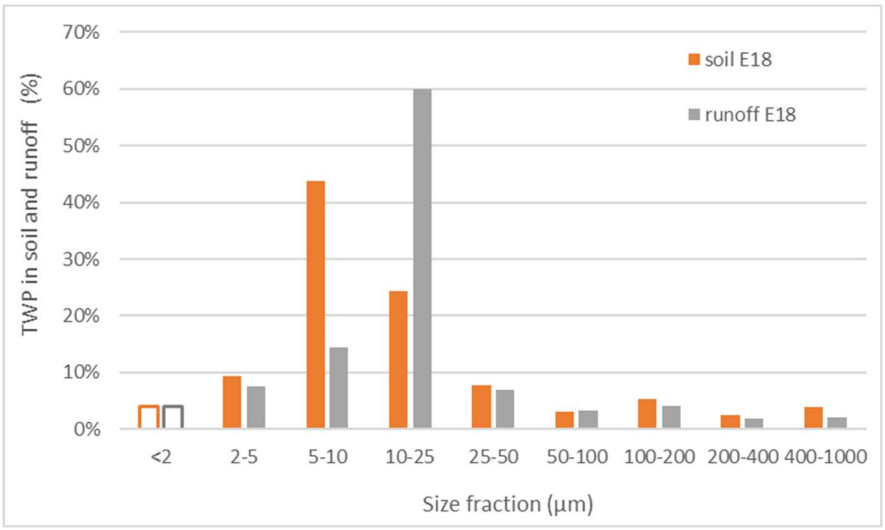

Figure 29 Particle size distributions of TWP in soil and runoff sampled next to the highway E18 in Sweden (TWP in fraction < 2 µm is based on the LOD).

While the particle size distributions of TWP in the runoff and deposited dust samples from the urban street and the highway A2 location are quite similar (relatively small error bars in Figure 31), there is a big difference in particle size distribution of TWP in the runoff and soil sample from the highway E18 location (see Figure 33). This clear difference can be explained by UV- and biodegradation of TWP on the soil surface, as already explained in section 6.8.2., concerning the decrease in NR. As shown in Deliverable 3.3, UV degradation processes enhance fragmentation of TWP which results in smaller particles.

7 Quality control and analytical uncertainties

TNO Earth, Life and Social Sciences operates in compliance with the Quality System standard ISO-9001 (certificate no. 00680-97-AQ-ROT-RvA). This study was performed in compliance with that Quality System standard. Two quantification methods were used: TNO method is based on the actual composition of used reference car and truck tyres with a direct calculation of TWP with conversion factors based on pyrolysis tyre markers 4-VCH or 4-PCH (TNO-method). In this method, the variability in rubber composition of the reference tyres causes an uncertainty in the TWP quantification, however our work in Deliverable 3.1 showed that the concentration of pyrolysis tyre markers in reference tyres remained comparable over the past 10 years and were similar to the concentrations determined by Rauert et al. (2021). The ISO method and the method used by BAM are based on the analysis of the polymers SBR and NR and the calculation of TWP is based on assumptions about the proportion of rubber in tyres (SBR+NR-method). This method has the advantage that the SBR and NR rubber content can be quantified separately and allows the use of internal standards to correct for deviating responses of the tracers due to matrix effects. However, the quantification of NR, based on the tracer dipentene can be challenging due to the presence of dipentene (limonene) in organic material.

To determine performance characteristics of both methods on several occasions duplicate samples were analysed with TED-GCMS. Duplicate samples were representative for the total collection of analysed samples with different environmental matrices and concentrations and from varies locations. The repeatability of the TNO method and SBR+NR method, determined with the same 25 duplicate samples, is 14% and 12% respectively. For the individual rubbers SBR and NR the repeatability is 12% and 19% respectively. The quasi intra-laboratory reproducibility (same instrument, same operator, but different analysis days) of the TNO method and SBR+NR method, determined with the same 6 duplicate samples, is 14% and 18% respectively. For the individual rubbers SBR and NR the reproducibility is 18% and 16% respectively. With a repeatability and intra-laboratory reproducibility below 20%, in principle both methods perform well.

As can be seen in Figure 34, without internal standard correction the SBR+NR method and the TNO method give the same TWP concentrations (slope 1.02 = 2% deviation) and correlate well (R^2 = 0.98). With correction for the internal standard (ISTD) 4-fluorostyrene, the correlation between both methods is still satisfactory (R^2 = 0.93), but the SBR+NR method gives approximately 24% higher TWP values (slope 1.024) than the TNO method according to correlation statistics. Looking at the mean ratio of individual TWP concentrations determined by both methods (Table 19) then the SBR+NR method gives appr. 10% lower values; with correction for the internal standard the SBR+NR method gives 18% higher values. These values are slightly different than the correlation statistics, because the slope of the trendline is biased towards higher concentrations and the mean ratio is not dependent on concentration levels.

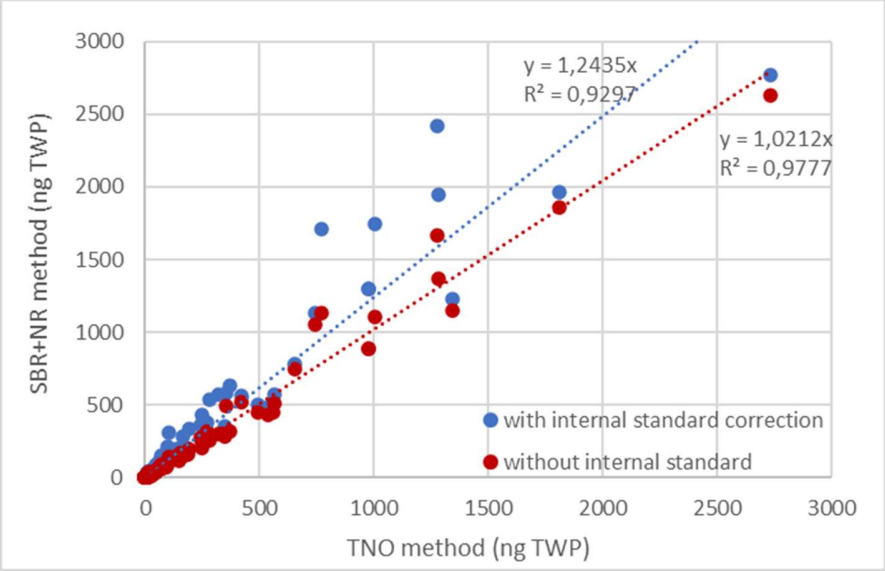

Figure 30 Correlation between both applied quantification methods (SBR+NR and TNO) with and without internal standard correction for the SBR+NR method.

For all individual environmental matrices linear correlation statistics (slope, R^2) and mean ratios (including standard deviation, SD) of TWP concentrations determined by both methods have been summarized in Table 20. For ambient air for four samples SBR and NR concentrations were outside the linear external standard range of the SBR+NR method, so the calculated slope is underestimated. For all individual environmental matrices the deviation between both applied methods is less then ca. 10%, except for surface water. For this matrix the SBR-NR method (without correction for internal standard) gives 20 % lower values then the TNO-method. With correction for internal standard, for all environmental matrices the SBR+NR method gives higher values (5 – 25%). Also, comparing values of the SBR+NR method with and without internal standard correction, values are 16 – 33 % higher with internal standard correction, depending on the matrix. Due to matrix effects the response of the same quantity of the internal standard 4-fluorostyrene in the environmental samples is lower than in the external calibration standards. It is debatable if the matrix has the same effect on the response of the tyre markers dipentene and 4-phenylcyclohexene. Either way correction for the internal standard has a big effect on calculated TWP concentrations. In this study TWP values of both the TNO method and the SBR+NR method with internal standard correction, are reported. The final TWP concentrations are average values of both methods.

Table 20 Correlation (slope / R^2) and mean ratio individual samples, between both applied quantification methods (SBR+NR and TNO) with and without internal standard correction for the SBR+NR method for all environment matrices.

Environmental sample	Samples analysed	SBR+NR method without / with ISTD		SBR+NR method / TNO method			SBR+NR method + ISTD / TNO method		
		Slope	R2	Slope	R2	Ratio ± SD	slope	R2	Ratio ± SD
Ambient air	45	0.72	1.00	(0.72)	0.98	1.03 ± 0.29	(0.99)	0.98	1.26 ± 0.35
Deposited dust	38	0.69	0.97	0.99	0.98	1.03 ± 0.27	1.39	0.97	1.26 ± 0.42
Runoff	39	0.79	0.97	1.02	0.98	1.02 ± 0.19	1.24	0.93	1.24 ± 0.35
Soil	36	0.84	1.00	1.01	0.98	0.88 ± 0.17	1.21	0.97	1.12 ± 0.39
Sediment	28	0.74	0.98	0.86	0.97	0.91 ± 0.17	1.14	0.96	1.18 ± 0.27
Surface water	19	0.67	0.99	0.80	0.97	0.79 ± 0.23	1.16	0.94	1.05 ± 0.35
Total samples	205	0.79	0.97	1.02	0.98	0.95 ± 0.24	1.24	0.93	1.20 ± 0.37

For the environmental matrices with more freshly generated TWP (e.g. ambient air, runoff, deposited dust) the difference in TWP values between both methods is negligible. For the other matrices the SBR+NR method (wo ISTD) result in slightly lower TWP concentrations. This is in agreement with degradation mechanisms (see Deliverable 3.3). As NR degrades faster than SBR rubber, and only the SBR+NR method is dependent on the NR concentration for calculated TWP values, the difference between both methods should be bigger in environmental matrices susceptible to degradation. As can be seen in Figure 35, for all environmental matrices the ratio of the SBR+NR method (wo ISTD) vs TNO method increases with an increasing percentage of natural rubber in TWP. Due to more freshly generated TWP in deposited dust and runoff the proportion of samples with a high NR percentage is higher. For ambient air NR percentages are lower due to smaller particle sizes with less NR in the rubber matrix.

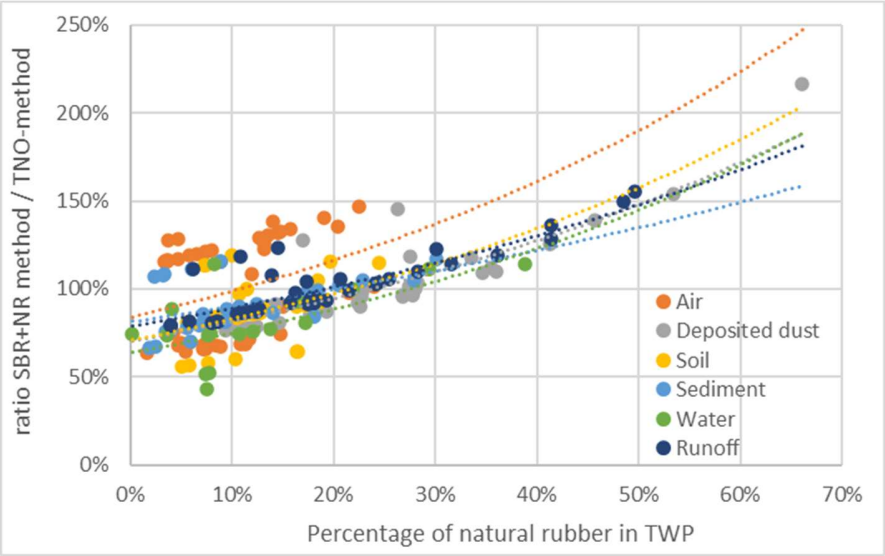

Figure 31 Correlation between the percentage of natural rubber in TWP and the ratio of TWP values determined with the SBR+NR method (wo ISTD) and TNO method

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10 Appendices

10.1 Appendix A: Measurement periods

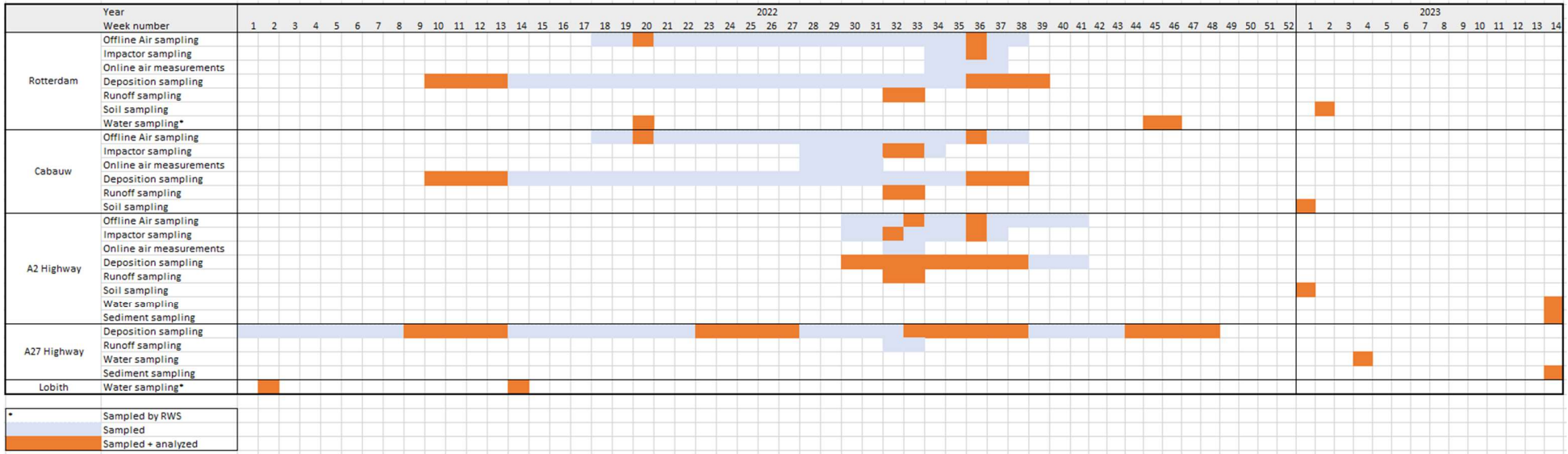

Figure 32 Sampling dates LEON-T

10.2 Appendix B: Meteorological conditions A27

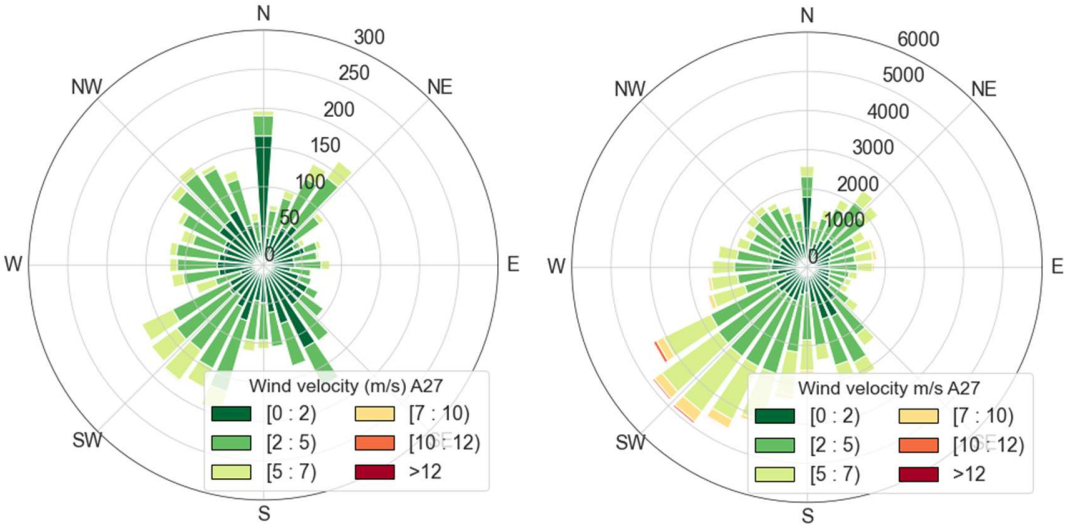

Figure 33 Wind direction and velocity A27 during the measurement period of LEON-T (May 2022-September 2022) (left) and the 10 year average (right).

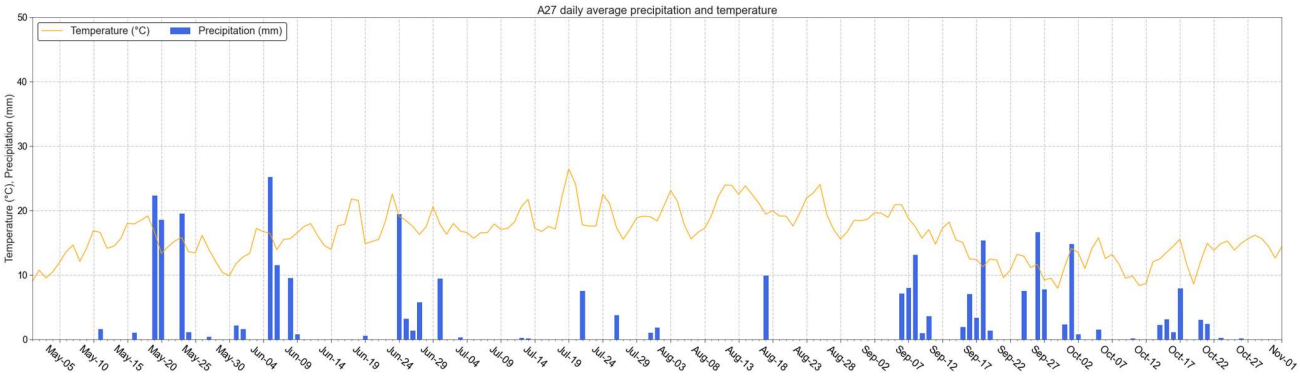

Figure 34 Daily average temperature and precipitation at the A27 during the measurement period of LEON-T (May 2022-September 2022) according to the nearest KNMI weather station.