Mapping Microplastic Flows and Pathways

Microplastics sampling results in the FanpLESStic-SEA project

FanpLESStic-sea project 2021

Fanplesstic-sea
Interreg Baltic Sea Region
EUROPEAN UNION
EUROPEAN REGIONAL DEVELOPMENT FUND

Introduction

This document is a description of the work conducted within the *Activity 2.2 Mapping flows and pathways* of the "FanpLESStic-sea project – Initiatives to remove microplastics before they enter the sea", an Interreg co-funded project where HELCOM is partner. The activity aims to produce additional data through sampling and sampling analysis of several different sources by reflecting the actual sources, pathways, and recipients of microplastics in the Baltic Sea region (BSR). This new dataset is subsequently compared to previous studies done which used the same or comparable verifiable measurement methods.

Mapping microplastic flows and pathways

The problem

The current knowledge on microplastics (MPs, polymer particles less than 5 mm in size) is still limited, especially on the presence, pathways and exposure effects on human health and the environment. The research on the occurrence of MPs in the environment has started only recently and standardized analysis methods are still lacking which makes the comparison of results very difficult (Simon et al. 2018, Borg Olesen et al. 2019). Despite this, several facts are to be pointed out, which are convincing enough to develop measures to reduce MP release in the environment: the particles are likely to be persistent in the environment; and particles themselves, the hazardous substances in the particles or sorbed on their surfaces, may cause negative impacts on organisms (Andersson-Sköld et al. 2020).

Globally, methods of defining microplastics, sampling and measurement vary considerably among different studies, source sectors and geographical regions making it difficult to synthesize data across studies (GESAMP 2016, BASEMAN 2021).

At Baltic Sea level, research provides strong evidence of the occurrence of microplastics in water, sediments, beach, and biota in the area, but due to the varying monitoring methodologies applied, the comparison between studies, basins and regions is not possible (HELCOM 2019).

Therefore, the development of harmonized monitoring methods for microplastics in general and for the Baltic Sea, in particular, is of high importance to better understand the problem (HELCOM 2019).

Background

This document is a compilation of the work conducted within the FanpLESStic-sea project aiming at producing additional data through sampling and sample analysis of several different sources by reflecting the actual sources, pathways, and recipients of MPs in the Baltic Sea region (BSR). This new data set is subsequently compared to previous studies done which used the same or comparable verifiable measurement methods.

Sampling activities were performed in different places using a pumping system equipped with 10 µm filters. Samples were prepared according to the harmonized protocol described in the dedicated section and analyzed using µFTIR imaging to detect and identify microplastics down to 11 µm in size.

Sampling points were selected to complement previous studies and to holistically cover the main sources of microplastic pollutants in an efficient and comprehensive way. Sampling sources range from the potentially lowest concentration of microplastics (including only atmospheric deposition) to some of the heaviest polluted waters:

1) pristine water in the Arctic in Norway;
2) river inlet to a drinking water plant in Poland;
3) river from agricultural area in Latvia;
4) river from industrial area in Latvia;
5) household discharge to WWTP in Poland;
6) ditch or surface water pipes along a highway in Sweden;
7) roofs (and facades) run-off in Sweden;
8) artificial turfs soccer fields runoff in Norway;
9) small community storm water aggregated at pump station in Sweden;
10) river flowing through urbanized areas to estuary in Poland; and
11) WWTP outlet close to a river inflowing to the sea in Lithuania.

Methodology used

Sampling

A Universal Filtering Object (UFO - filtering device) was used for conducting the sampling. The sampling device samples water from the source by pumping it through different filters (300 µm and 10 µm). The 300µm filter was used to avoid fast clogging of the two 10µm filters that are filtering in parallel to save time. All operators conducting the sampling were trained on how to implement the sampling including the following:

- preparation before going to the field;
- testing and priming the sampling kit pre-sampling (in-situ priming, optional preparation of the sampling equipment in the laboratory);
- field sampling;
- cleaning and re-packing; and
- troubleshooting.

Concerning sampling, there is always the issue of a sufficiently large sample volume. This is the case especially when expecting low (or completely unknown) MP concentrations. In this sampling, at least several hundred litres were sampled reaching 1 m^3 of water so that results could be considered representative.

For raw wastewater, only 2 litres were analysed but considering the high MP concentrations as well as the large variety of polymer types detected, it can be concluded that the sampling was also representative.

However, for road runoff samples (highway and parking lot), the volume sampled was very low, usually less than 1 litre, and thus some higher degree of extrapolation has been made. This is, however, not unusual (it has been done in other studies as well e.g. Bergmann et al. 2019) and since the MP concentrations are high in these samples, the results can be trusted.

Sample preparation

Sample preparation was conducted to extract microplastics from the samples by reducing the sample matrix and removing bioorganic and inorganic matter. Six different laboratories participated in the sample preparation and all operators involved received specific training on how to prepare the samples following a widely used multistep protocol. The procedure consisted of a chemical treatment to remove any bioorganic matter utilizing a surfactant, various enzymes and an oxidation as well as a physical treatment to remove inorganic matter through density separation. Furthermore, all the laboratories provided procedural laboratory blanks.

Figure 1 Microplastic sampling device (UFO- universal filtering object).

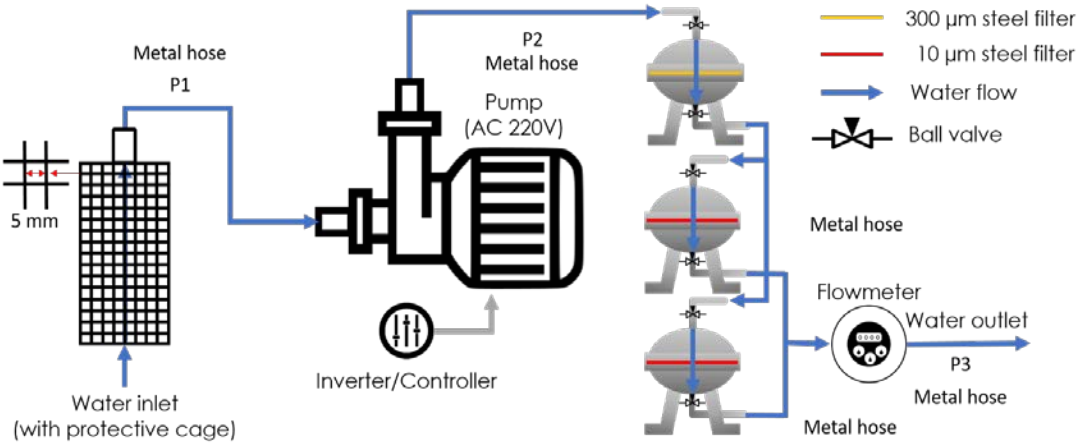

Figure 2 Sampling setup schematics, graphics by Lucian Iordachescu.

Some challenges have been faced due to the differences between the sampled matrices as with rust (in the snow samples), refractive material (e.g. wood, humic substances) and coal like particles. Therefore, it is worth pointing out that there is not one sample preparation technique that fits all sample matrices but since the protocol used in this report is quite modular, certain steps can be repeated if necessary to achieve better results.

Sample analysis

All prepared samples were analysed in the same laboratory using µFTIR imaging to detect and identify microplastics down to 11 µm in size. In µFTIR imaging, high spatial resolution FTIR sample's mapping is performed where the chemical composition of particles is determined down to a few micrometers in size, hereby distinguishing natural material from plastics. µFTIR is supported by automatic microplastic detection (siMPle). Additional information on the software can be found in the publication by Primpke et al 2020. As car-tyre rubber cannot be analysed by FTIR, a complementary technique, Pyr-GC/MS (Pyrolysis-Gas-Chromatography-Mass Spectrometry), was used to determine tyre wear particle (TWP) concentration. In Pyr-GC/MS, the sample is rapidly heated up to 600-1000

Celsius in an inert atmosphere, subsequently the sample's components are separated (GC), identified, and quantified (MS), providing the concentration of different plastics and rubbers in the sample.

Results

The tables below summarize the microplastic sampling results obtained. An overview of the results can be seen in Table 1, where each of the eleven sampling matrices include microplastic concentration range in both particle numbers (MP m^{-3})/ (MP L^{-1}) and mass (µg m^{-3})/ (µg L^{-1}). The per litre values are due to low volumes in some samples. In addition, the polymer composition of each sample was analysed. A more detailed description of each sample matrix can be seen in Annex 1.

Table 2 provides information on the sampling dataset and conclusions extracted from the results obtained.

Table 1 List of the sampling matrix and summary of results.

Code	Sampling matrix	No. of samples	MP concentration range in				Ref. Concentrations [MP m^{-3}]/[MP L^{-1}]	Ref.	Comments
			Number	Unit	Mass	Unit			
1	Pristine water in the Arctic in Norway	6	0–45	MP m^{-3}	0–112	µg m^{-3}	0–14	Bergmann et al. 2019	3 lakes with 2 samples each
2	Inlet drinking water plant in Poland	3	126–232	MP m^{-3}	10–25	µg m^{-3}			Inlet to drinking water plant sampled on three consecutive days
3	Agricultural river	4	306–2.1x10^{3}	MP m^{-3}	47–239	µg m^{-3}	67–11532	Mintenig et al. 2020	Agriculture impacted river
4	Industrial river	4	770–3.3x10^{3}	MP m^{-3}	70–350	µg m^{-3}	67–11532	Mintenig et al. 2020	Industrial river, samples hard to process and analyse
5	Raw waste water	3	1.3x10^{3}-5.4x10^{3}	MP L^{-1}	94–1540	µg L^{-1}	533	Rasmussen et al. 2021	
6	Parking lot/highway runoff	6	95–1.7x10^{3}/ 267–1.1x10^{4}	MP L^{-1}	3–114	µg L^{-1}	490–23000	Liu et al. 2019	Highway and parking lot run off with 3 samples each, low sample volume -> extrapolation to MP per litre (MP L^{-1})
7	Rooftop runoff	3	467–1.1x10^{3}	MP m^{-3}	19–132	µg m^{-3}	490–23000	Liu et al. 2019	Roof-top run off
8	Artificial turfs football field	4	150–239	MP m^{-3}	13–876	µg m^{-3}	67–11532	Mintenig et al. 2020	2 rivers with 2 samples each
9	Storm water aggregated at pump station	4	1.2x10^{3}-1.1x10^{4}	MP m^{-3}	40–1734	µg m^{-3}	490–23000	Liu et al. 2019	Aggregated stormwater
10	River in Poland	3	2.2x10^{3}-5.1x10^{3}	MP m^{-3}	80–1292	µg m^{-3}	67–11532	Mintenig et al. 2020	Urban stream
11	WWTP outlet close to a river	3	202–4.3x10^{4}	MP m^{-3}	52–31641	µg m^{-3}	67–11532	Mintenig et al. 2020	River and outlet WWTP

Table 2 Microplastic sampling dataset with conclusions.

Code	Sample matrix	MP concentration range in numbers (MP m^{-3}/MP L^{-1}) and in mass (µg m^{-3}/ µg L^{-1})	MP concentration level (low/medium/high)	Conclusions
1	Pristine water in arctic regions	0–45 MP m^{-3} 0–112 µg m^{-3}	Low	• Pristine lakes relatively unaffected but not at all free of MP. • Concentrations are **low** (0-45 MPs/m^3), one order of magnitude lower than in Arctic marine waters (Rist et al. 2020). • Only few (n=4) polymer types, dominated by polypropylene (PP) and followed by polyester. • A likely source of MPs is atmospheric deposition. • Bergmann et al. (2019) found MP concentration up to 10^4 times higher than this in Arctic snow.
2	Inlet drinking water plant in Poland	126-232 MP m^{-3} 10–25 µg m^{-3}	Relatively low	• Inlet to drinking water plant. • Concentrations are **relatively low** (126-232 MPs/m^3), and in the same order of magnitude as from Swedish drinking water (Kirstein et al. 2021). • 11 different polymer types identified, dominated by polyester followed by PP and polyamide (nylon) (PA). • Concentration as well as polymer composition does not differ immensely between samples.
3	Agricultural river	306–2.1x10^3 MP m^{-3} 47-239 µg m^{-3}	Medium range	• Concentrations are in a **medium range** (306-2050 MPs/m^3), and in a similar order of magnitude compared to a study from the Netherlands (Mintenig et al. 2020). • 15 different polymer types identified, dominated by polyester followed by polyethylene (PE), PA, polyurethane (PU), polyvinyl chloride (PVC) and cellulose acetate (CA). • The concentration was highest in one sample, but the polymer composition does not differ immensely between the samples.
4	Industrial river	770–3.3x10^3 MP m^{-3} 70–350 µg m^{-3}	Medium range	• Concentrations are in a **medium range** (770-3350 MPs/m^3), and in a similar order of magnitude compared to a study from the Netherlands (Mintenig et al. 2020). • 15 different polymer types identified, dominated by polyester followed by PE, PA, PP, PU and CA. • The concentration was highest at one sample in particular but the polymer composition does not differ immensely between the samples. • MP concentration and mass were 2 times higher than for the agricultural river but the polymer composition was similar.

5	Raw wastewater	$1.3x10^3$-$5.4x10^3$ MP L^{-1} 94–1540 µg L^{-1}	High	• Concentrations are **high** (1284-5371 MPs/L^1), and one order of magnitude higher compared to a study from Sweden (Rasmussen et al. 2021). • 14 different polymer types identified, dominated by polyester followed by PU. • The concentration was 4 times higher for one of the samples, but the polymer composition did not differ immensely between samples. • MPs were dominated by particles, but the portion of fibers was with 33% higher than for other sample matrices (13-25%).
6	Parking lot runoff/ highway runoff	Parking lot: 95–$1.7x10^3$ MP L^{-1} 6–114 µg L^{-1} Highway runoff: 267–$1.1x10^4$ MP L^{-1} 3–101 µg L^{-1}	High both in parking lot runoff/ highway runoff	Parking lot runoff: • Concentrations are **high** (95-1694 MPs/L^1), and up to 2 orders of magnitude higher compared to a study from stormwater in Denmark (Liu et al. 2019). • 11 different polymer types identified, clearly dominated by PP followed by PE and polyester. • The concentration was 2 orders of magnitude higher for one of the samples with almost exclusively PP. • Sampled volume quite small (1.5-4.2 L). Highway runoff: • Concentrations are **high** (267-11422 MPs/L^1), and up to 3 orders of magnitude higher compared to a study from stormwater in Denmark (Liu et al. 2019). • 5 different polymer types identified, clearly dominated by PP. • The concentration was 2 orders of magnitude higher for one of the samples with almost exclusively PP compared to the other two samples. • Sampled volume quite small (0.6-2 L).
7	Rooftop runoff	467–$1.1x10^3$ MP m^{-3} 19–132 µg m^{-3}	Medium to high	• Concentrations are **medium to high** (469-1123 MPs/m^3), and much lower than what was found in the Arctic or in urban snow (Bergmann et al. 2019). • 6 different polymer types identified, mostly dominated by PP followed by PE, PU and polyester. • MP concentration for one of the samples was less than half of that of the others but more diverse in polymer composition with more polyester and PU instead of PP.
8	Artificial turfs football field	150–239 MP m^{-3} 13–876 µg m^{-3}	Relatively low	• Concentrations are **relatively low** (150-239 MPs/m^3), and in a similar or rather lower order of magnitude compared to a study from the Netherlands (Mintenig et al. 2020). • 12 different polymer types identified, dominated by PP and polyester followed by PE, PS and PA. • MP concentrations and polymer compositions do not differ greatly between samples. • MP concentrations downstream were slightly lower than upstream. • Two samples from one of the rivers sampled in northern Norway contained cellulose acetate while the other did not, two samples from this river contained a higher percentage of acrylates compared to the other river.

9	Small community storm water aggregated at pump station	$1.2x10^3$-$1.1x10^4$ MP m^{-3} 40–1734 µg m^{-3}	Relatively high	– Concentrations are **relatively high** (1220-11257 MPs/m^3), and in a similar range compared to a study from storm water in Denmark (Liu et al. 2019). – 15 different polymer types identified, clearly dominated by PP and followed by PE. – Cellulose acetate could be detected in all samples. – MPs were dominated by particles, but the portion of fibers was 28% higher than for most of the other sample matrices (13-25%).
10	River in Poland	$2.2x10^3$-$5.1x10^3$ MP m^{-3} 80–1292 µg m^{-3}	Medium to high	• Concentrations are **medium to high** (2228-5053 MPs/m^3), and in a similar order of magnitude compared to a study from the Netherlands (Mintenig et al. 2020). • 13 different polymer types identified, clearly dominated by PP followed by polyester, CA and PE. • Cellulose acetate could be detected in all samples. • MP concentration in one of the samples was 2 times as high as the other ones and contained relatively more CA.
11	WWTP outlet close to a river	202–$4.3x10^4$ MP m^{-3} 52–31641 µg m^{-3}	Medium to high	• Concentrations are **medium to high** (202-42929 MPs/m^3), and in a similar or slightly elevated range compared to a study from the Netherlands (Mintenig et al. 2020). • 15 different polymer types identified, clearly dominated by polyester followed by PU. • Polymer composition differed greatly between the samples. • Cellulose acetate could be detected in all samples. • MP concentration of one of the samples was 2 orders of magnitude higher than the other two. • MPs were dominated by particles, but the portion of fibers was 37% higher than for most of the other sample matrices (13-25%).

Table 3 provides a summary of the results of tyre wear particle analysis that was also included in the sampling activities. A technique called Pyr-GC/MS (Pyrolysis-Gas-Chromatography-Mass Spectrometry) was used to determine tyre wear particles (TWPs) concentration as this cannot be analysed by FTIR. For TWPs only mass values are possible to obtain (due to the technique). The results show very high concentrations for parking lot run-off ($6.66x10^4$–$2.20x10^6$ µg/L) and highway run-off ($2.85x10^5$–$1.30x10^6$ µg/L).

Table 3 Summary of results of tyre wear particles (TWPs) concentration in mass per litre (µg/L).

Code	Sample matrix	No. of samples	TWPs concentration in [µg/L]
6	Parking lot run-off	3	$6.66x10^4$–$2.20x10^6$
6	Highway run-off	3	$2.85x10^5$–$1.30x10^6$
8	Artificial turfs football field	4	5.71-63.55
9	Stormwater aggregated at pump station	4	73.22-$4.81x10^3$

Key observations

From the sampling results the following key observations can be extracted:

- **Pristine lakes** are relatively unaffected by MP presenting MP concentrations one order of magnitude lower than in Arctic marine waters (Rist et al. 2020), a likely source for the MPs is atmospheric deposition.
- **Agricultural river**: concentrations are in a **medium range** (306-2050 MPs/m^3), and in a similar order of magnitude compared to a study from the Netherlands (Mintenig et al. 2020).
- **Industrial river**: concentrations are in a **medium range** (770-3350 MPs/m^3), and in a similar order of magnitude to the fundings by a study from the Netherlands (Mintenig et al. 2020).
- **Raw wastewater**: concentrations are **high** (1284-5371 MPs/L^1), and one order of magnitude higher compared to a study from Sweden (Rasmussen et al. 2021), 14 different polymer types identified, dominated by polyester followed by PU; MPs were dominated by particles, but the portion of fibers was 33% higher than for other samples matrices (13-25%).
- **Parking Lot runoff/ Highway runoff**: **high** concentrations of MPs in both parking lot (95-1694 MPs/L^1), and up to 2 orders of magnitude higher compared to a study from stormwater in Denmark (Liu et al. 2019)) and highway runoff (267-11422 MPs/L^1), and up to 3 orders of magnitude higher compared to a study from stormwater in Denmark (Liu et al. 2019), 5 different polymer types identified, **clearly dominated by polypropylene (PP).**
- **Rooftop runoff**: concentrations are **medium high** (469-1123 MPs/m^3), and much lower than what has been found in Arctic or urban snow (Bergmann et al. 2019).
- **Artificial turfs football field:** concentrations are **relatively low** (150-239 MPs/m^3), and in a similar or rather lower order of magnitude compared to a study from the Netherlands (Mintenig et al. 2020).

- **Small community storm water aggregated at pump station**: concentrations are **relatively high** (1220-11257 MPs/m^3), and in a similar range compared to a study on storm water in Denmark (Liu et al. 2019). **Cellulose acetate was detected in all samples,** MPs were dominated by particles, but the portion of fibers was with 28% higher than for most of the other sample matrices (13-25%).
- **River in Poland:** concentrations are **medium to high** (2228-5053 MPs/m^3), and in a similar order of magnitude compared to a study from the Netherlands (Mintenig et al. 2020)
- **WWTP outlet close to a river:** concentrations are **medium to high** (202-42929 MPs/m^3), and in a similar or slightly elevated range compared to a study from the Netherlands (Mintenig et al. 2020). **Cellulose acetate was detected in all samples**. MPs were dominated by particles, but the portion of fibers was with 37% higher than for most of the other sample matrices (13-25%).
- **Parking lot and highway**: concentrations are the **highest** detected, 6.66×10^4–2.20×10^6 µg/L for the run-off from a parking lot and 2.85×10^5–1.30×10^6 µg/L for a highway.

Discussion

This section summarises some discussion points which could be used for further managing microplastics sources in the Baltic Sea:

- the results obtained show the presence of microplastics in pristine lakes, but in very low concentrations (10^1);
- hundreds of MPs (which is in the same order of magnitude as for sea surface water) were detected in two Norwegian rivers. MPs concentrations in sampled stormwater were one to three orders of magnitude higher (10^3-10^5) than in the analyzed rivers;
- generally, most MP concentrations fall well into the range of known literature values;
- polypropylene (PP) and polyester were the most dominant polymer types in most of the samples;
- samples related to stormwater were clearly dominated by Polypropylene (PP);
- samples related to rivers usually contained cellulose acetate (CA) (except for one river);
- samples related to WWTPs showed a higher share of fibers (>25%) than samples from other matrixes;
- high MP concentrations were found in raw wastewater and parking lot/highway runoffs and relatively high at small community storm water aggregated at pump station;
- medium to high MP concentrations: rooftop runoff, river in Gdansk and WWTP outlet close to a river;
- it can very cautiously be speculated that a large part of polyester comes from laundry (the elevated amounts in wastewater and a higher proportion of fibres could imply this) and that the source of cellulose acetate might be cigarette butts; and
- in urban areas which are related to traffic, tyre wear particles are the largest source of microplastics (in mass).

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Glossary of terms

CA	Cellulose Acetate
PA	Polyamide (nylon)
PE	Polyethylene
PP	Polypropylene
PS	Polystyrene
PU	Polyurethane
PVC	Polyvinyl chloride
TWR	Tyre wear particle