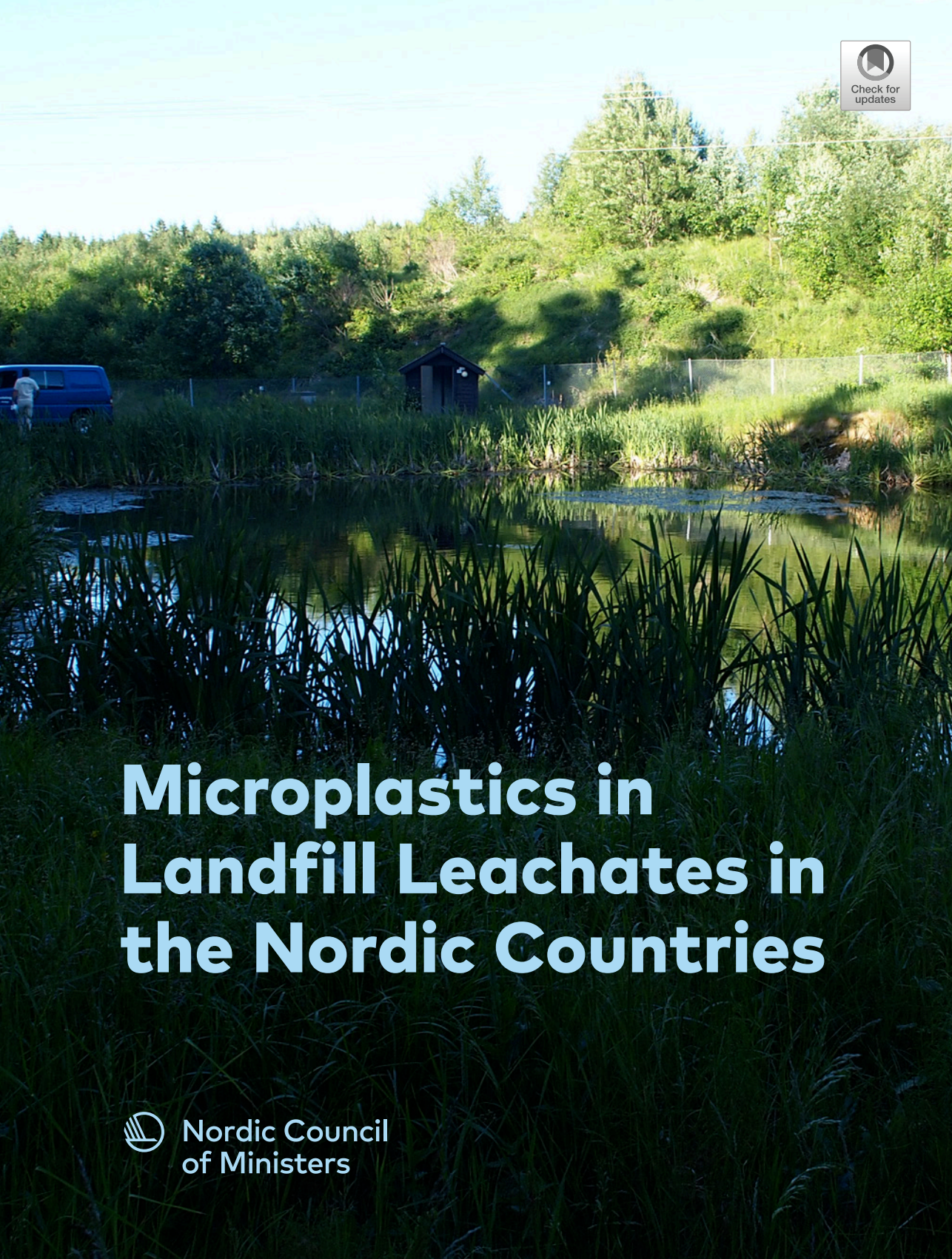

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Nordic Council of Ministers

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Nordic Council of Ministers/Publication Unit
Ved Stranden 18
DK-1061 Copenhagen K
Denmark
Phone +45 3396 0200
pub@norden.org

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The Nordic Council of Ministers
Nordens Hus
Ved Stranden 18
DK-1061 Copenhagen K, Denmark
Tel.: +45 3396 0200 www.norden.org

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Contents

Summary

The Nordic Waste Group (NAG) and the Marine Group (HAV) under the Nordic Council of Ministers commissioned ÅF Infrastructure to investigate the occurrence of microplastics in landfill leachates in Finland, Iceland and Norway. For the purpose of this work, microplastics are defined as particles with an upper size limit of 5 mm and a lower limit of 50 µm (micrometres), made of man-made polymers, deriving from petroleum or petroleum by-products, and non-synthetic polymers such as natural rubber and polymer modified bitumen. A total of 11 landfills were included in the study. In three cases both untreated and treated landfill leachate was sampled and analysed, together with a reference sample of tap water. Samples were collected as collective samples by means of pumping leachate with the help of a stainless steel submergible pump through a stainless steel unit equipped with stainless steel filters with different pore sizes. Sampling was carried out by ÅF staff in Norway and Finland, and by Resource International in Iceland.

The filters were sent to the laboratory at the Austrian EPA (Umweltbundesamt) for a two-step analyses. In the first step, samples were pre-treated in order to remove unwanted organic and inorganic material, split into particles size classes above 500 µm and above 50 µm, respectively. Samples were screened for occurrence of microplastics including tire rubber and polymer modified bitumen. Subsequently, the samples with positive detects were analysed by FT-IR spectroscopy in order to quantify 10 different polymers microplastic particles, tire rubber and polymer modified bitumen excluded. Results show the following:

- most samples tested positive for multiple microplastics. Three leachates however, including a blank test, tested positive only for tire rubber and/or polymer modified bitumen;
- few if any microplastic particles were larger than 500 µm, and most likely in the range of 100–50 µm within the particle size range included in this study;

- a paired comparison of treated leachates versus untreated generally exhibited decreased microplastic counts by orders of magnitude.

Compared to other quantified sources of microplastics such as raw or treated sewage, the results for landfill leachates indicate that landfills are likely to be a small source of microplastic particles larger than 50 µm. The considerably lower count in treated leachate samples suggests that local treatment of leachate has an impact on microplastic concentration in leachate outflow. This appears to be true even for robust, non-complex treatment options such as settlement basins or sand filtration.

The variation in particle count and concentration in landfill leachates extends over several orders of magnitude. Continued data acquisition and improved understanding of variability is, therefore, necessary. As particles smaller than 50 µm were not included in the study, these should be included in future investigations. Additionally, when viable analytical protocols have been developed for tire rubber and polymer modified bitumen, microplastics of these materials should be quantified, as well. Microplastics as potential carriers of priority pollutants such as plasticizers, flame retardants etc. from landfills to water bodies should be addressed. Future studies should target landfills in Denmark and Sweden in addition to the Nordic countries covered in this study.

Abbreviations

ATR	Attenuated total reflection
FT-IR	Fourier Transform Infrared (Spectroscopy)
GDP	Gross Domestic Product
Haz	Hazardous waste
IW	Industrial Waste
MSW	Municipal Solid Waste
Non-haz	Non-hazardous waste
n.d.	Non-detect
PE	Polyethylene
PP	Polypropylene
PVC	Polyvinyl Chloride
PS	Polystyrene
PET	Polyethylene Terephthalate
PA	Polyamide (Nylon)
PU	Polyurethene
PC	Polycarbonate
PMMA	Polymethylmethacrylate
PMB	Polymer Modified Bitumen
POM	Polyoxymethylene (Acetal)
SBR	Sequencing Batch Biological Reactor
STD	Standard Deviation
TR	Tire Rubber
WWTP	Waste Water Treatment Plants

1. Introduction

1.1 Background

Although discovered more than 40 years ago, awareness of microplastics in the seas has been raised considerably only in recent years. Global plastic production is increasing, and microplastics in the seas are of great concern, as they can be consumed by organisms, subsequently move up the food chain with negative consequences for both the ecosystem and human health. Some studies have shown that microplastics can have negative effects on aquatic and terrestrial organisms [1]. Therefore, the identification and quantification of major sources of microplastics ought to be prioritized, and actions should be taken to reduce the flow of microplastics from these sources. However, at the time of commission of this project, no reliable data on the nature and size of the majority of microplastic sources were available. Still, waste management, e.g. landfilling of waste, has been pinpointed as one of the potential primary sources [2], in particular by the pathway of landfill leachates (the liquid emissions from waste in landfills) emitted to surface waters. Leaching of various contaminants from landfills, including plastic additives, is well known and documented. Neither information on microplastic content in landfill leachates, nor on the potential impact of landfill leachate treatment on potential microplastic content has been previously available [3]. If landfills are indeed a significant source of microplastics, which have detrimental effects in the environment, this should be addressed, for example by eliminating the source or by reducing the outflow. Landfill operators might be requested to install or improve on-site leachate treatment.

In order to address the questions of landfill leachates as potential sources of microplastics and the efficacy of existing landfill treatment processes, the Nordic Waste Group and the Marine Group have tendered and commissioned the assignment, which is presented in this report.

1.2 Aim and scope of the study

The aim of this study is to acquire quantitative data on the amounts of microplastics in landfill leachates from Nordic Countries. Furthermore, the aim is to acquire information on the impact of different methods of leachate treatment on the amounts of microplastics and to relate potential emissions of microplastics in leachate to other potential sources.

1.3 Definition of microplastics

Microplastic particles, as defined in the call for tender this study, are particles with an upper size limit of 5 mm and a lower limit of 50 µm, made of man-made polymers, deriving from petroleum or petroleum by-products, and non-synthetic polymers such as natural rubber and polymer modified bitumen.[1]

Microplastic particles are a heterogenous, complex group of different polymers, sizes, colours, shapes, age, tear and additives.

1.4 Background and project management

This project was initiated and steered by Guðmundur B. Ingvarsson, Yvonne Augustsson, Hanna Salmenperä and Jon Fonnlid Larsen amongst others (all members of the Nordic Waste Group) and in close co-operation with the Marine Group. The project was managed by Martijn van Praagh, ÅF Infrastructure, and carried out together with Cornelia Hartman, Emma Brandmyr and Ola Wik, also ÅF. Sampling equipment was provided by Resource International, Iceland, and sampling in Iceland conducted by Jamie McQuilkin and Lea Böhme under supervision of Nicolas Marino Proietti, Resource International.

[1] Please observe that "microplastics", or "plastics", is not discriminately defined; e.g. "natural rubber" might be included only if it has been modified, and even bioplastics, polymers produced from non-fossil sources, could be included. Additionally, suggestions have been made to define the size range for microplastics as between 5 mm and 1 µm.

Analytical work was supervised by Bettina Liebmann and carried out and administrated by Sebastian Köppel, Gerrit Hermann, Felizitas Zeitz, Martina Göß and Helmut Weber, Umweltbundesamt, Austria.

This report is authored by Martijn van Praagh with the help of Cornelia Hartman and Emma Brandmyr and has been reviewed by members of the Nordic Waste Group and the Marine Group.

2. Methodology

2.1 Selection of landfills

The type of landfills, their location and information on the respective leachate treatment can be derived from table 1. Landfills have been selected in order to reflect different levels of leachate treatment and operational status; estimated age of the landfilled wastes; and types of landfilled class (hazardous or non-hazardous). In total, samples were taken from 11 different landfills in three different countries.

Table 1: Overview of landfills comprised in this study

Country	Nr	Landfill	Location	Landfill class	Type of waste	Leachate treatment type	Technical level of treatment
Finland	1	Topinoja	Turku/Åbo	Non-haz	MSW	None	-
Finland	2	Korvenmäki	Salo	Non-haz	MSW, IW	None	-
Finland	3	Anonymous 1*	South-west	Haz	IW	Filtration and active carbon	Medium-high
Finland	4	Hollola	Lahti	Non-haz	MSW, IW	Artificial soil filtration	Moderate
Finland	5	Kujala	Lahti	Non-haz	MSW	None	-
Norway	6	Böler	Skedsmokorset	Non-haz	MSW, IW	SBR	High
Norway	7	Gjerdrum	Ask	Non-haz	Mixed	None	-
Norway	8	Anonymous 2*	South-east	Non-haz	MSW, IW	None	-
Iceland	9	Fiflholt new cell	Fiflholt	Non-haz	MSW, IW	Sand bed filtration	Moderate
Iceland	10	Fiflholt old cell	Fiflholt	Non-haz	MSW, IW	None	-
Iceland	11	Álfsnes	Álfsnes	Non-haz	MSW, IW	None	-

Note: * two landfill operators asked to remain anonymous, their identity is known to the steering group.

2.2 Sampling method

Sampling and analysis of microplastics in landfill leachates constitutes specific challenges due to the following:

- the small sizes of the particles;
- unknown concentrations and fluxes;
- complicated chemical matrixes of landfill leachate;
- the abundance of plastics in the working environment (protective gear, hoses, beakers etc.);
- a lack of standardization.

The sampling method was chosen in order to address these challenges.

Sampling was carried out by means of pumping leachate through 3 custom made stainless steel filter plates with falling mesh sizes incorporated in a stainless steel unit (5,000, 411, and 47 µm wide, respectively; see picture 1).

Figure 1: Sampling equipment

Note: (l, courtesy of Resource International), close-up of loaded filter plate (r).

A submersible, stainless steel pump (*Proril X-SMART*, 400A, 2", 0.4kW/230V/50 Hz) was connected via a hose to the stainless steel filter housing. The filter system was developed and constructed by Resource International, who acted as a subcontractor to this assignment.

The filtering unit is equipped with a flow meter and a pressure release valve. The flow meter reading was annotated prior and after each sampling occasion. At sampling occasions in Finland and Iceland, the measured flows were cross-refenced by volume estimations with the help of buckets with known volumes.

As can be derived from picture 1, a PVC-hose was used between the submersible pump and the filter unit, rather than a stainless steel one. This was primarily due to practical and occupational safety reasons: In order to collect samples as close to the source as possible, in certain cases the pump had to be submerged several meters into a pumping station or manhole. This would not have either impossible or unsafe with the additional weight of several meters of steel hosing.

In some cases, mainly due to either very low water levels after an extremely dry spring or the technical prerequisites at the sampling location, the leachate was caught in a (plastic) bucket and subsequently pumped through the filter unit. In order to evaluate potential cross-contamination of leachate samples due to a) occupational safety gear (mainly PVC-gloves and PET-clothing); b) the PVC hose and power cable connected to the pump; and c) the use of plastic buckets, a blank sample with drinking water was collected and analysed. The presence of plastic materials used at or in the vicinity of the sampling point were documented (photographed and identified).

Sampling was carried out by a subcontractor in Iceland (Resource International) and ÅF's staff in Norway and Finland.

Sampling took place during the following time periods:

- 8 May 2018 in Iceland;
- 16–17 May 2018 in Finland;
- 23–24 May 2018 in Norway.

The sampling locations and volume of leachate/water collected until pressure built-up at each landfill are described in the table below.

Table 2: Sampling locations

Landfill	Untreated leachate	Sample volume [m³]	Treated leachate	Sample volume [m³]	Comment
Álfsnes	Leachate pond	0.044			
Fiflholt old cell	Outflow pipe	0.307			
Fiflholt new cell	Bore hole	0.295	Outflow pipe	0.255	
Kujala	Settlement pond	0.01			Particle-rich leachate
Topinoja	Pump station	0.12			Mixed leachate*
Hollola	Leachate pond	0.07	Outflow pipe + PE-bucket	0.21	Particle-rich untreated leachate
Korvenmäki LSJH	Pump station	0.08			Clear but foaming leachate
Anonymous 1	Pump station + PE-bucket	0.07	Pump station + PE-bucket	0.19	Clear leachate
Böler	Pump station	0.02	Pump station	0.005	Foam (untreated)
Gjerdrum	Well (with flow)	0.01			Particle-rich leachate
Anonymous 2	Pump station	0.04			
Blank	Tap, PE-bucket	0.105			At "Anonymous 1"

Note: * Due to extremely low water levels (<8 m), the leachate in the appropriate manhole could not be reached with the equipment; instead, a sample was taken from another pumping station which receives both landfill leachate and drainage from waste sorting activities.

At each sampling location, the supposed "cleaner" sample was taken first, i.e. treated leachate prior to untreated. The filter equipment was cleaned between samples, either with tap water and/or paper towels. Every stainless steel filter plate was used only once.

Pumping and filtration took place until pressure built-up stalled further sampling and flow was minimal or came to a halt. The pressure release valve never opened during sampling, which means that pressure was always below 1 bar.

To produce reference spectra for tire rubber, samples of regular summer and winter tires were collected. Sampling took place the 26 June 2018 at a tire company in

Malmö. Sampling was carried out by the staff by cutting off small pieces from old tires. 3 samples were taken in total, one sample of a winter tire and two samples of different summer tires. The samples were then put in an envelope and sent to the lab as soon as possible.

2.3 Sample transport

To avoid contact with the filter mesh, loaded filter plates were carefully removed from the filtering unit with a stainless steel knife, placed in stainless steel boxes, sealed, packed and delivered to the laboratory in Vienna, Austria, as soon as possible, i.e. at the end of every sample campaign, via courier.

2.4 Analytical method

All samples were send to the laboratory at the Environment Agency Austria (Umweltbundesamt GmbH). Samples were stored, prepared and analysed in two steps: screening and quantification.

As of today, there are no standardized methods published for the analysis of microplastics. However, one of the more common and robust analytical method employs FT-IR (Fourier Transform Infrared Spectroscopy) in order to identify the material of the microplastic particles. This method was used for all leachates samples, as well as the blank sample.

The analysis was conducted in three steps as follows:

1. pre-treatment of samples in two size fractions;
2. occurrence of microplastics and determination of total sample mass in fractions;
3. counting of particles.

These steps are described more in detail below.

2.4.1 *Sample pre-treatment*

Filter plates were carefully back-flushed with deionized water and filtered onto 500 and 50 µm aluminium screens. This means that, although smaller particles might have been caught on the 47 µm stainless steel screens during sampling, particles between 50 and 47 µm are excluded from the analysis.

Landfill leachate might be rich in particles or suspended matter. In order to be able to identify and count microplastic material, as much as possible of the non-plastic material has to be removed chemically. The aim of chemical sample pre-treatment for microplastic analysis is to remove the interfering matrix of samples, and to make the plastic particles easier accessible for subsequent material identification by FT-IR micro-spectroscopy. Based on experience with other water samples, chemicals are selected that dissolve the biological matrix while ensuring the integrity of potential microplastics, in this case mainly Hydrogen peroxide. The amount of solid matter remaining after chemical treatment was separated into two size fractions: (1) particles larger than 500 µm and (2) particles of size 50–500 µm. The total mass of residue after pre-treatment was weighed.

2.4.2 *Microplastic determination*

The following, most common, plastics were searched for in the samples: PE, PP, PVC, PS, PET, PA, PU, PC, PMMA, POM, as well as tire rubber (TR) and polymer modified bitumen (PMB).

Microplastic determination for particles larger than 500 µm

The material of the particles larger than 500 µm was determined by means of ATR-FT-IR micro-spectroscopy. It is possible to roughly characterize the particles by the naked eye (ocular inspection) into categories such as "biological matter", "plastic fragments", "plastic fibres", "plastic foils", etc. This characterization method was, however, not part of the assignment. Instead, it was verified or falsified which plastic materials were present by analysing random subsamples from the filter residue.

Microplastic determination for particles of size 50 µm to 500 µm

As there is little or no prior knowledge of which materials to be expected in the leachates, some loaded filters from pre-treatment were pre-screened and checked for whether and which of the ten most common polymers are present in the particles (result: present yes/no). This screening procedure can be done relatively quickly and facilitates for a swifter analytical process later on; the information gathered can be used for the analysis of the remaining samples.

Sample preparation for the smaller size fraction includes representative subsampling, and loading of particles onto a filter material (aluminium oxide) that is then analysed by micro-FT-IR-spectroscopy and imaging. For each sample, a total area of 12.5 cm^2 was scanned via imaging for acquisition of infrared spectra.

Via software assisted comparison with reference materials from a spectra database, the "chemical image" of the filter (2D map plus IR spectra) was evaluated for a variety of the most common plastic materials (see above), with an exemption for rubber and polymer modified bitumen.

Once the most relevant material types are known, the chemical image of the filter could be evaluated for the number of the particles consisting of the respective materials. The total mass of microplastic particles was estimated with the help of the counts of particles (see chapter 3).

Polymer modified bitumen and tire rubber

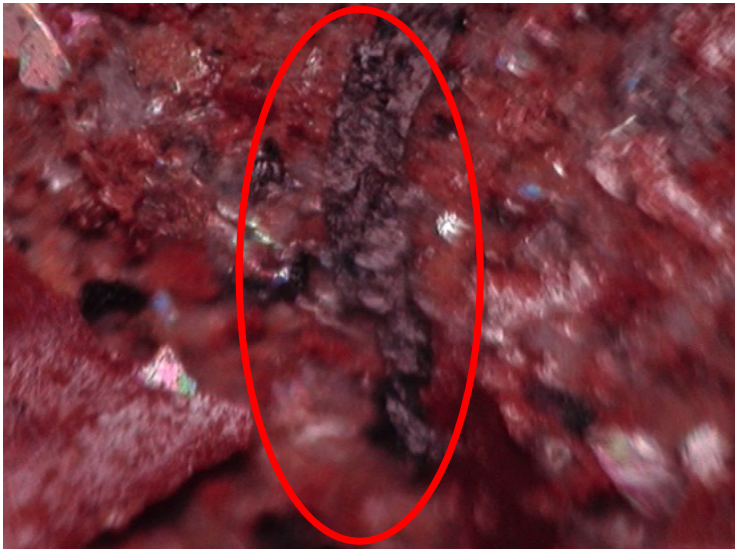
The employed analytical method is regarded state-of-the art for identifying and quantifying microplastics of various polymers. The underlying approach of analysing the reflection from the particle in the infrared spectrum (IR), however, reaches its limitations when it comes to tire residues and bitumen. The reason for these limitations is the lack or absence of reflection due to the black nature or black additives of these materials. Therefore, the following, differing approaches have been used to identify tire rubber (TR) and polymer modified bitumen (PMB) particles:

- Reference data:
  - samples of 3 different PMB from two different Swedish producers were scanned and added to the laboratory's reference database;

  - Samples of TR were collected by removing circa 3x3 cm large parts from disposed tires at a garage. Subsamples were then used in the lab in the same way as for bitumen to produce reference spectra.
- Screening:
  - for screening of PMB and TR particles, a different measurement technique of ATR-FT-IR micro-spectroscopy was applied; rather than scanning from a distance, contact was established between a germanium crystal and suspected particles. The crystal was positioned onto 5 different spots on the filter, each of which exhibited an area of 0.16 mm², where potential PMB/TR particles were preliminary identified visually. The total sample area that was screened for the presence of PMB/TR particles resulted in 0.8 mm².
- The ATR-measurements were combined with imaging.

See the picture below for an identified tire rubber particle.

Figure 2: Image of tire rubber particle



3. Analytical Results

3.1 Analytical results from screening of microplastic, tire rubber and polymer modified bitumen

The mass of the remaining particles after chemical treatment can be used as an indicator of microplastics in samples. The chemical treatment prior to the analysis described in chapter 2.4 does, however, not completely remove non-target matter. In the table below, the microplastic, tire rubber and polymer modified bitumen detects from the screening procedure are displayed together with the total amount of particulate matter remaining in the sample after chemical pre-treatment.

Table 3: Analytical results from microplastic screening for particles in landfill leachates (n.d. = none detect)

Sample name	Identified microplastic	47 µm filter Approximate maximum total mass in mg*	411 µm filter Approximate maximum total mass in mg*	Approximate maximum total mass in mg*
Analytical Method		**2.4.2.2**	**2.4.2.1**	**2.4.1**
Alfnes Untreated	PE, PP, PS, PU, PMB, TR	16	46	62
Filfhold Old Cell untreated	TR	11	1	12
Filfhold New Cell untreated	PE, PS, PET, PU	4	5	9
Filfhold New Cell treated	TR, PE	21	12	33
Kujala untreated	TR, PBM	60	11	72
Topinoja untreated	PE, PP, PBM	64	93	157
Hollola untreated	PE, PS, PET, PU	27	n.d.	27
Hollola treated	PE, PS	12	n.d.	12

Sample name	Identified microplastic	47 µm filter Approximate maximum total mass in mg*	411 µm filter Approximate maximum total mass in mg*	Approximate maximum total mass in mg*
Analytical Method		**2.4.2.2**	**2.4.2.1**	**2.4.1**
Korvenmäki, LSJH untreated	PE, PVC, PS, PET, PA, PU, PMMA	11	n.d.	11
Anonymous 1 untreated	PET, PU	2	23	25
Anonymous 1 treated	PE, PS, Bitumen	5	2	7
Böler untreated	PE, PET, Tire	9	10	19
Böler treated	Tire	29	122	151
Gjerdrum untreated	Negative**, PE	483**	15	498**
Anonymous 2 untreated	PET, PMMA	23	9	32
Blank tap water	TR	8	11	19

Note: * Total mass including recalcitrant inorganic or non-plastic organic material, which could not be removed by the chemical pre-treatment.

** 50 µm sample reacted heavily with chemicals in pre-treatment, producing precipitation residues. Large amounts of remaining insoluble inorganic residues impaired the screening for microplastics. An additional investigation into the nature of reaction products revealed prevailing minerals containing iron and silica. Double-checking with the landfill operator did not reveal any chemical anomalies in the sample which might suggest what caused the reaction with pre-treatment agents. The leachate from the sample point is normally rich in iron but this is not uncommon for MSW-landfill leachates.

Generally, negligible numbers of microplastic particles of the size 5,000–500 µm were detected. Rather, particles below 500 µm and approximately closer to 50 µm were detected on the 411 µm screens. Consequently, a partition in particle size fractions is not meaningful and the results are displayed as sum of particles > 50 µm subsequently. Microplastics (including TR and PMB) where detected in all samples as well as the blank sample where TR particles where indicated. The identified polymers are displayed in the table below in descending order.

Table 4: Abundance of polymers in landfill leachates (n.d. = none detect)

Counts	Polymer
8	PE
6	PS, TR
5	PET, PU
4	PMB
2	PMMA, PP
1	PA, PVC
n.d.	POM, PC

3.2 Analytical results from quantification of microplastic

The table below summarizes results from the 2nd analytical step; quantification of microplastic particles in samples with positive identification in the screening step. The quantification method does not support quantification of PMB and TR, as described above. Consequently, these are excluded from the analysis.

Table 5: Analytical results from microplastic quantification for particles 5,000 µm > x > 50 µm

Sample name	PE	PP	PVC	PS	PET	PA	PU	PMMA	Total count	Count/l
Alfnes untreated	132	25		17			25		199	4.51
Filfhold New Cell treated	15								15	0.06
Filfhold New Cell untreated	14			2	4		40		60	0.20
Filfhold Old Cell untreated										0
Kujala untreated										0
Topinoja untreated	13	6							19	0.16
Hollola treated	4			2					6	0.03
Hollola untreated	41			16	8		73		138	1.97
Korvenmäki, LSJH untreated	19		15	17	25	4	8	4	88	1.10

Sample name	PE	PP	PVC	PS	PET	PA	PU	PMMA	Total count	Count/l
Anonymous 1 treated	25			35					60	0.32
Anonymous 1 untreated					9		9		18	0.30
Böler treated										0
Böler untreated	22				4				26	1.3
Gjerdrum untreated*	10								10	1.00
Anonymous 2 untreated	6				51			26	57	1.40
Blank tap water										0
Sum of particles	301	31	15	89	101	4	155	30		

Note: * Subsample 50 µm reacted heavily with chemicals in pre-treatment, large amounts of remaining insoluble inorganic residues impaired the screening for microplastics.

From the results displayed in tables 4 above the following can be derived:

- the blank sample and 3 leachates where free from quantifiable polymer particles;
- the number of identified polymers varies from 0 to 7;
- the most abundant identified polymer was PE (found in 10 of 15 leachate samples) with a total of 301 counts;
- polymers are found in the following order, measured as counts;
- PE > PU > PET > PS > PP > PMMA > PVC > PA;
- the total counts of quantifiable microplastic particles per sample vary from 0 to 199;
- overall, counts in treated samples are circa 1 or even 2 orders of magnitude lower than untreated samples (Filfhold New Cell, Hollola and Böler), with the exception of Anonymous 1 (see chapter 2 for discussion);
- there is little difference in the microplastic concentration of treated and untreated leachate from landfill "Anomymous 1".

4. Estimation of microplastic concentrations

In order to receive an approximate concentration of microplastics in landfill leachates (as in µg/l), in samples with quantified microplastic particles, i.e. excluding TR and PMB, the mass was estimated. The following assumptions about the density and morphology of the microplastic particles have to be made:

- uniform density of particles of the same polymer as follows (kg/l or Mg/m^3):
  - PA: 1,05
  - PE: 0,965
  - PET: 1,45
  - PMMA: 1,20
  - PP: 0,91
  - PS: 1,1
  - PU: 1,2
  - PVC: 1,58.
- uniform or average shape of particles;
- size of particles; assumed minimum and assumed maximum as well as assumed average particle volume according to the following:
  - minimum volume: Plates (height h = 0,5 µm; diameter d = 50 µm)
  - maximum volume: Globe (r=250 µm)[2]
  - average volume: Globe (r=50 µm).

[2] This is a theoretical maximum volume for particles not able to pass a filter of 500 µm. Most particle appeared to be closer to 50 µm in size.

In the tables below, resulting mass-concentration calculations and reduction ratios are displayed, respectively.

Table 6: Calculated results for microplastic concentrations in untreated samples with quantified microplastic particles (µg/l)

Sample name, unit	Min (µg/l)	Max (µg/l)	Average (µg/l)
Alfnes	4.7E-03	295.1	2.36
Filfhold New Cell	2.3E-04	15.4	0.12
Filfhold Old Cell	n.d.	n.d.	n.d.
Korvenmäki, LSJH	1.4E-06	94.6	0.76
Kujala	n.d.	n.d.	n.d.
Topinoja	1.5E-04	9.8	0.08
Anonymous 1	3.3E-04	22.3	0.17
Hollola	2.2E-03	146.2	1.17
Böler	1.3E-03	88.5	0.71
Gjerdrum*	9.5E-04	63.2	0.51
Anonymous 2 untreated	2.7E-03	181.5	1.45

Note: * Subsample 50 µm reacted heavily with chemicals in pre-treatment, large amounts of remaining insoluble inorganic residues impaired the screening for microplastics and is not included.

Table 7: Calculated results for microplastic concentrations in treated samples (µg/l)

Sample name, unit	Min (µg/l)	Max (µg/l)	Average (µg/l)
Filfhold New Cell	5.6E-05	3.7	0.03
Anonymous 1	3.2E-04	21.6	0.17
Hollola	2.8E-05	1.9	0.02
Böler	n.d.	n.d.	n.d.

Based on mass-concentration calculations, the reduction of (calculated) microplastic concentrations was derived. The reduction was calculated as follows:

(concentration in – concentration out) / concentration in.

Observe that, although microplastics concentrations appear to be reduced by all leachate treatments, in case of "Anonymous 1" the polymers in the treated leachate are not the same as in the untreated (see Table 5).

Table 8: Concentration reduction by leachate treatment based on calculated concentrations

Sample name, unit	Reduction, %
Filfhold New Cell	76
Anonymous 1	3
Hollola	99
Böler	100

5. Discussion

5.1 Occurrence of microplastics in landfill leachates

Regardless of the counts or masses of microplastics in the studied leachates, the evaluation of the abundance of positive identifications is an indication for either an abundance of plastic materials in landfills and/or of the mobility of those plastics. The number of occurrences of the plastic materials in falling order is as follows:

PE > PS = TR > PET = PU > PMB > PMMA=PP > PVC=PA > POM = PC (n.d.)

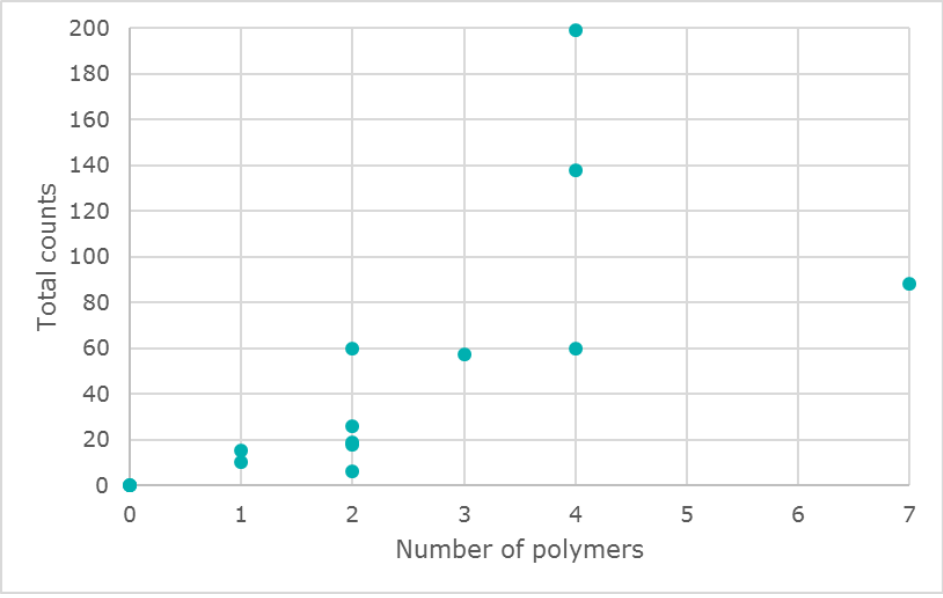
The abundance of polymer production is different from the above (total production year 2015 circa 270 Mtonnes, [4]), except for the most abundant polymer, PE:

PE > PP > PVC > PS > PA > PET > PUR > other

Although the order above reflects the recent distribution of produced polymers, historical data indicates that few changes have occurred since production started and increased [5].

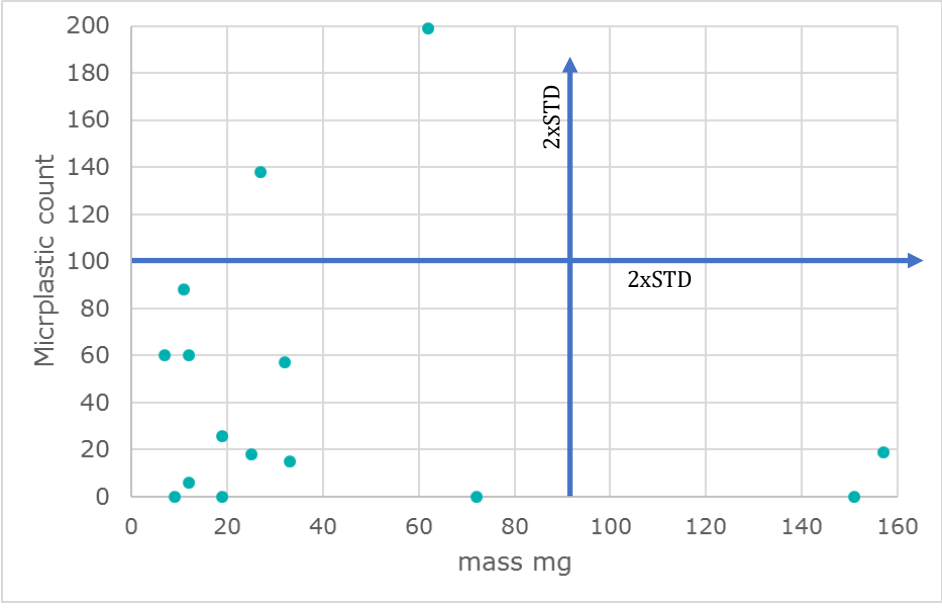
A look at the number of different polymers quantified against the counts does not indicate a strong relation of the two (see Figure 3 below, mind that TR and PMB detects are not included).

Figure 3: Sum of microplastic particle counts in landfill leachate samples vs. number of different polymers identified



The differences in total mass of residues after pre-treatment of samples and microplastic count might suggest a possible correlation (the more residual material the higher the count). By plotting the approximate total mass after sample pre-treatment against the particle count, a first impression can be gained whether such a correlation might exist (see Figure 4).

Figure 4: Approximate total mass in leachate samples after pre-treatment vs. microplastic count (with 2 times standard deviations, STD, indicated as arrows)



As can be derived from the graph above, an obvious or even weak relationship does not seem to exist.

Also shown in Figure 4, some values stick out, i.e. they exceed two times the respective standard deviation:

- the microplastic counts of leachate from Hollala and Alfnes landfills at 138 and 199 respectively, compared to 2xSTD of 112 counts;
- the approximate total mass in samples in leachate from Böler (treated) and Topinoja (untreated) at 151 and 157 mg, respectively, compared to 2xSTD of 94 mg.

5.2 Potential differences of microplastics in leachates with regards to types of waste

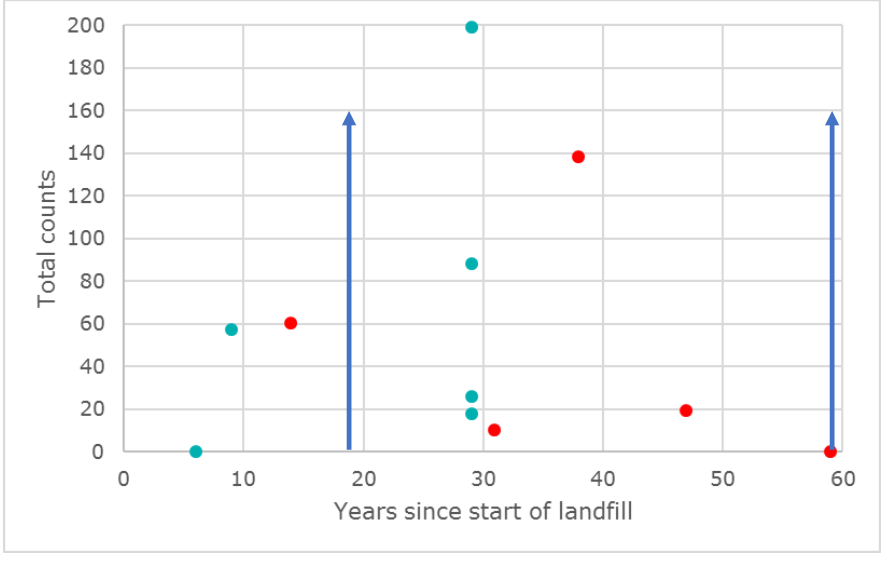
Most landfills receive(d) both industrial and municipal solid waste, as is common with municipally owned or operated landfills serving a local or regional area.

Two operators indicated that the landfill in question only received municipal waste or industrial waste, respectively: Kujala and Topinoja in Finland (MSW) and Anonymous 1 (IW). The counts for the leachates from these landfills are well below the average of 56 counts for untreated leachates at 0, 19 and 18 particles, respectively.

5.3 Potential differences of microplastics in leachates with regards to landfill age

In the figure below, the year the landfills were started are plotted against the microplastic counts in untreated leachate samples.

Figure 5: Years since start of landfill operation vs. microplastic count



Note: Closed landfills are represented by red dots, arrows indicate substantial changes in plastic production and waste management.

Figure 3 does not give a clear indication whether the period in which a landfill has been operative has an impact on microplastic emissions. Theoretically, one might expect the following three distinct anomalies through time, as indicated by arrows in Figure 3:

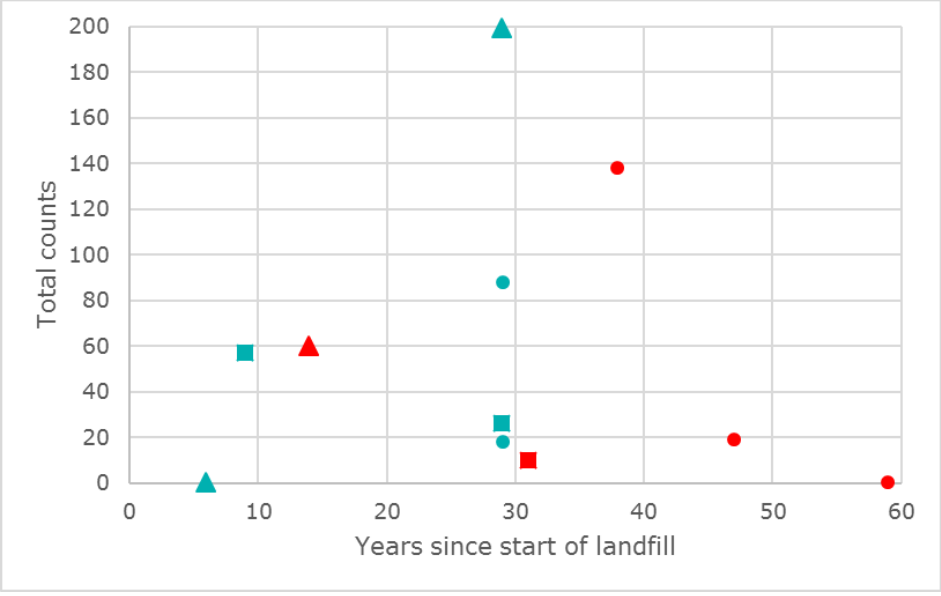
- the exponential use, and disposing off, plastics after 1960;
- the advent of the polluter pays principle and extended producer responsibility (turn of last century);
- the banning of organic waste from landfills by law (starting turn of last century).

The collected data does not give an indication of these developments and regulations having an effect on microplastic emissions in landfill leachates.

5.4 Potential differences between countries

Differences in industrial activity, habits, laws- and regulations, GDP etc. are likely to have an influence on both amounts of waste going to landfills and the kind of waste to landfill that might lead to microplastic emissions in the leachates. Figure 6 below depicts the same information as in Figure 5, but with the country of origin for each landfill indicated.

Figure 6: Years since start of landfill operation vs. microplastic count with country indicated



Note: Triangle=Iceland, box=Norway, dot=Finland; closed landfills are represented in red.

Leachate samples from landfills in Iceland exhibit both non-detect and the highest count. Microplastic counts in leachate samples from Norway are close to or below average of 57. In order to be able to draw any conclusions, however, many more results from different leachates from each country are needed.

5.5 Landfill leachate as a source of microplastics

Generally, landfills might be a source of microplastics to aquatic ecosystems. In the table below, minimum and maximum annual loads of microplastic particles are displayed (loads calculated with the estimated minimum and maximum concentrations, see Tables 6 and 6).

Table 9: Calculated annual load of microplastics from studied landfills

Landfill, leachate treatment	Volume (m^3 per a)	"Minimum" annual load (g)	"Maximum" annual load (kg)	Recipient
Alfnes untreated	600,000	2.81	177.1	Sea
Filfhold Old Cell untreated	40,077	n.d.	n.d.	Wetland
Filfhold New Cell untreated	30,294	113	0.1	Sand bed
Filfhold New Cell treated	no data	n.d.	n.d.	Wetland
Korvenmäki, LSJH untreated	46,472	0.07	4.4	WWTP – Uskela river
Gjerdrum untreated	12,500	0.01	0.8	Open ditch – river Børterelva
Anonymous 1 untreated	no data	no data	no data	To flocculation, filtration
Anonymous 1 treated	no data	no data	no data	WWTP
Anonymous 2 untreated	50,000	0.14	9.1	WWTP – one of Norway's larger lakes
Böler treated	no data	n.d.	n.d.	WWTP – river Nitelva
Böler untreated	120,000	0.16	10.6	To SBR
Hollala treated	10,758	3.05E-04	0.02	WWTP to river Porvoonjoki
Hollala untreated	4,815	0.01	0.70	To soil filtration
Average		14.5	25.3	

Note: With the assumption of maximum particle volume according to the concentration calculations above and last available data for volume.

As can be derived from the load ranges in the table above, there is a considerable variation both between landfills and in minimal and maximal loads. The latter is, of course, due the different assumptions of prevailing size of the identified particles. The present study indicates that most particles are in the lower range, close to 50 µm. Looking at the maximum loads calculated in the kg to tonnes range, one might get the impression that landfills are a large source of microplastics to the environment. In order

to assess this, the calculated results have to be put into context of other verified or suspected sources of microplastics.

In the table below, the summarized analytical results are put into perspective to results from other published studies, both on treated landfill leachate and other sources or pathways. Observe that the scope, sampling methods and analytical techniques (if employed) differ widely between studies. Consequently, the loads emitted by different sources should be regarded a preliminary indication. As the particle size cut-off differs considerably between studies, results have not been recalculated to mass-concentrations. Consequently, units for "Annual load" differ in the table below.

Table 10: Approximated emitted microplastic loads from potential sources/pathways

Source/pathway	Particle size range	Count/l	Annual Load	Reference/comment
Landfills this study	50–5,000 µm	0–4.5	0–177 kg* per landfill	Untreated and treated leachate
Ø this study	50–5,000 µm	0–4.5	15 g–25 kg* per landfill	On average
Landfills in Sweden	≥ 100 µm	0–2.7	0–170 kg per year for Swedish landfills	Treated leachate (estimation untreated <2,4 tons), [6]
Raw Sewage Sweden	>20 µm	20–80	$2{,}6 \cdot 10^{12}$ particles	[7]
Effluent large WWTP Sweden	>20 µm	10–100	$2{,}6 \cdot 10^{11}$ particles	[7]
Traffic in Sweden	>100 µm	n.d.	7,670 tons per year	[3]
Air deposition in Paris	100–5,000 µm		3–10 tons fibres	[8]
Artificial turfs in Sweden	>100 µm	n.d.	1,640–2,460 tons	[3]
Players on artificial turfs in Norway		2 ml	65 tons	2 ml per game and player [9]

Note: * Observe that, due to identified particles being closer to 50 than 500 µm, loads are more likely to be in the gram than kg range.

** Average ranges with estimated minimum and maximum calculated loads.

In short, the comparison of calculated annual loads of microplastics from landfill leachates with other verified or suspected sources shows the following:

- untreated landfill leachate is a source of microplastic to recipients;
- annual calculated loads of microplastics were in the same order of magnitude as previously published in a study on microplastics in treated landfill leachates in Sweden, when the lower estimate is used (observe that the studies employed different assumptions to calculate annual loads);
- compared to other sources such as untreated and treated sewage, artificial turfs and road traffic, leachate appears to be a less relevant source.

5.6 Limitations and discussion of potential sources of error

Not least due to the lack of standardization of sampling and analysis of microplastics, certain sources of error can, at present, not be ruled out. These are summarized and commented below:

- Landfill leachate variability; although especially older MSW-landfills have developed a relatively stable chemical and biological regime leading to, in most cases, predictable leachate quality, variations do occur and concentration variations of one or several standard deviations from the mean are not uncommon. The variation of (micro)particles and of factors influencing the emission of these have, to the knowledge of the authors, not been subject of comprehensive studies before. Potential variations of microplastic concentration in leachate, e.g. seasonal variations or the influence of heavy rainfall events, are not accounted for in this study. The same reasoning goes, naturally, for the representativeness of the included landfills in the Nordic countries as a whole;
- The sampling and analytical methods are not standardized, and no duplicate or triplicate samples have been taken, analysed and cross-referenced yet. As a result, the error margin and detection limits are not verifiable;
- Sampling leachate microplastics by filtration of leachate in the field results in smaller leachate sampling volumes for samples rich in particles. Sampling a fixed amount of leachate at one occasion, e.g. 1 litre, with subsequent filtration in the

laboratory would most likely have given different results. In that case, the chance of "catching" microplastics would have been much lower;

- If microplastic particle concentrations are in fact correlated to particle concentrations of non-plastic matter in leachates, the sampling method chosen for this study should give a relatively representative picture of microplastic counts in landfill leachates;
- Sampling pre-treatment; as described earlier and also obvious from the discrepancy between the theoretical total mass of microplastic in samples and the calculated concentrations, chemical pre-treatment does not remove all non-targeted substances, which might interfere with the analysis;
- Although similarities of different tire materials and different polymer modified bitumen are likely to create similar and coherent FT-IR patterns, respectively, it cannot be ruled out that microplastic particles of significantly different blends of both materials than those used as references might go undetected in leachate samples (there is, according to a producer of polymer modified bitumen, an almost unlimited number of blends that can be used with different fractions of polymers). Likewise, a false positive, cannot be ruled out to 100%. Non-target matter might obscure or otherwise influence the exact analysis of microplastics. The difference between calculated and approximate total mass in samples emphasizes the need to actually identify the materials in the samples in order to determine the occurrence of microplastics or the need to improve sample pre-treatment;
- Although the polymers included in the screening by far represent the major part of plastics used in the technosphere (>80%), it cannot be ruled out that microplastics of non-targeted polymers were present in the samples;
- The filter equipment was cleaned between samples, and treated samples were taken prior to untreated. Still, cross-contamination regarding tire or material that has been identified as tire residue, for example, cannot be ruled out, which is indicated by the tire residue in the blank sample;
- The choice of lower size limit of 50 µm (actually 47 µm) is likely to exclude numerous smaller particles;

- The quantification of rubber and bitumen is likely to have led to a higher count of microplastic particles, as these appeared to be widespread in the included landfill leachate samples (if they are not due to cross-contamination or false positives);
- Particularly regarding microplastic particles from PE and PVC, but also others, it cannot be ruled out that these stem from landfill drainage and leachate treatment and collection systems rather than from the landfilled waste itself. This could, for example, explain the low reduction ratio of the landfill leachate treatment at landfill "Anonymous 1", and the occurrence of microplastic particles in the size range of 500–50 µm. As bag filtration units with a mesh size of 25 µm are used at "Anonymous 1", either the equipment itself emits microplastics, or it is bypassed.

6. Possible actions to reduce emissions of microplastics from landfills

This study, to the knowledge of the authors, is the most comprehensive investigation of microplastics in landfill leachates so far, and the selected landfills are regarded to be "typical" landfills (as far as there is such a thing as a typical landfill). Still, data has been gathered only at 3 to 4 landfills per country. Due to the lack of other, directly comparable studies, any conclusion on the necessity and possibilities to reduce microplastic emissions from landfills have to be preliminary and carefully drawn.

The generally lower counts of microplastic particles in treated leachate samples suggest, however, that local treatment of landfill leachate is a potentially viable option to reduce the emission of microplastics from landfills to either WWTPs or surface water recipients.

With or without pre-treatment, treating landfill leachate in municipal sewage treatment plants is a wide spread treatment option. As shown elsewhere [6], only a fraction of incoming microplastic particles to the WWTP reach the recipient. This is particularly true for larger particles (>300 µm). The fate of particles below 20 µm is less known, and as long as no treatment option with a well-defined cut-off in the micrometre range is used, e.g. membrane techniques, microplastic particles emitted with landfill leachate reaching the aquatic environment directly or indirectly cannot be ruled out.

What is more, both at on-site leachate treatment plants and at WWTP, microplastics are unlikely to be degraded, but rather retained in the (sewage) sludge. They might, via soil amendment or unviable treatment options, reach the environment still.

The following actions with regard to microplastics in landfill leachates are suggested:

- improved understanding on the variability of microplastics in landfill leachates;
- improved quantification methods especially for TR and PMB;

- investigate the concentrations and loads of smaller microplastic particles in landfill leachates (<50 µm, but rather <20 µm);
- investigate the emissions of microplastics from other waste management activities such as plastic recycling and littering;
- concentrate efforts of monitoring microplastic emissions from landfills via leachate to landfills without leachate treatment;
- gather and review upcoming knowledge of the effects of smaller particles to aquatic organisms and relate these to landfill leachate concentrations;
- investigate the importance of microplastics as a vector for priority pollutants despite relatively low particle counts.

Conclusions

Based on the results from this study, the underlying hypothesis of landfills generally being a significant source of microplastics is put in doubt. There seems to be a large variation in microplastic counts in landfill leachates, as well as a large variation in number and type of polymer. The counts and calculated annual loads, however, are small compared to other sources or pathways such as untreated and treated sewage.

Landfill leachate treatment seems to be able to significantly reduce the counts of microplastics in landfill leachate. Tire rubber or polymer modified bitumen particles occurred regularly in samples, either due to the abundance of these materials or cross-contamination. With regard to the particular sources contributing to microplastics in landfill leachates, drainage material and treatment processes involving plastic materials might contribute to microplastics concentration.

Future work should focus on the following:

- standardization of leachate sampling techniques with regard to landfill leachates;
- improved quantification methods especially regarding TR and PMB;
- generation of a more extensive dataset including information on variability;
- improved understanding of the reduction related to different leachate treatment methods;
- elucidation of counts of microplastics between 50 and 1 µm, possibly even in the nanometer range;
- elucidation of effects of microplastics as potential carriers of priority pollutants from landfills;
- include landfills from Sweden and Denmark.

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Sammanfattning

Nordiska Avfallsgruppen (NAG) och Havgruppen (HAV) under Nordiska Ministerrådet har gett ÅF Infrastructure AB i uppdrag att undersöka förekomsten av mikroplast partiklar i lakvatten från deponier i de Nordiska länderna Finland, Island och Norge. I ramen för undersökningen definieras mikroplastpartiklar som partiklar "av petroleumbaserade polymerer med antropogent ursprung i storlek mellan 5 mm och 50 µm (mikrometer), samt icke-syntetiska polymerer såsom gummi från däck och polymer modifierat bitumen".

Totalt har 11 deponier inkluderats i denna studie. Vid tre deponier har utöver obehandlat även behandlat lakvatten provtagits och analyserats, samt ett referensprov (kranvatten vid en av deponierna). Proverna togs som blandprover med hjälp av en rostfri pump ansluten till en rostfri filteranläggning utrustad med tre olika filter (5 mm, 411 µm och 47 µm). I Finland och Norge togs proverna av ÅF anställda, på Island av Resource International.

Alla filter skickades till laboratoriet vid Österrikes miljömyndighet (Umweltbundesamt) för analys i två steg. I det första steget förbehandlades proverna för att avlägsna ovidkommande organiskt och icke-organiskt material, samt undersöka förekomsten av de 10 vanligaste plastpolymererna, däckpartiklar samt partiklar av polymermodifierad bitumen i storleksklasser över 500 och över 50 µm. I det andra steget kvantifierades mikroplastpartiklar av de polymerer som har identifierats i steg 1 med hjälp av FT-IR spektroskopi.

Resultaten av studien är som följer:

- I alla prover hittades åtminstone en typ av mikroplast. I tre fall enbart däckgummi eller polymermodifierad bitumen.
- Två obehandlade och ett behandlat lakvattenprov visade inga mikroplastpartiklar förutom däckgummi eller polymermodifierad bitumen.

- I det undersökta intervallet visade sig vara väldigt få om ens några mikroplastpartiklar större än 500 µm, troligen var merparten av partiklarna i storleksordningen 100–50 µm.
- Behandlat lakvatten visade sig ha betydligt färre mikroplastpartiklar (bortsett från ett prov).
- Enbart däckgummi, hittades i referensprovet (kranvatten tagen på plats på en deponi), som kan vara ett tecken på kontaminering som beror på filtrerings- och provtagningsmetodiken.

Jämfört med andra identifierade källor eller transportvägar av mikroplaster indikerar resultaten att deponier troligen är av mindre betydelse för mikroplastemissioner för de undersökta storleksintervallerna. Även relativt enkla behandlingsmetoder för lakvatten såsom utjämning och filtrering verkar kunna reducera mikroplasthalter i lakvatten.

Eftersom partiklar mindre än 50 µm inte har varit del av denna undersökning, bör dessa inkluderas i kompletterande arbeten. Utöver det bör – när teknikutvecklingen möjliggör det – en noggrann kvantifiering av däckpartiklar och polymer modifierat bitumen ingå. Att mikroplastpartiklar kan agerar som vektorer för prioriterade, förorenande ämnen såsom mjukgörare, flamskyddsmedel etc. bör tas hänsyn till i kommande studier. Då bör även deponier i Danmark och Sverige inkluderas.

Nordic Council of Ministers
Nordens Hus
Ved Stranden 18
DK-1061 Copenhagen K
www.norden.org

Microplastics in Landfill Leachates in the Nordic Countries

This report summarizes results from sampling and analysis of microplastics in leachates from a total of 11 landfills in Finland, Iceland and Norway. The study was commissioned by the Nordic Waste Group (NAG) and the Marine Group (HAV) under the Nordic Council of Ministers. Polymer particles with an upper size limit of 5 mm and a lower limit of 50 µm (micrometres), were included. Samples were analysed by FT-IR spectroscopy. Most samples tested positive for multiple microplastics. Compared to other quantified sources of microplastics such as raw or treated sewage, landfill leachates are likely to be a relatively small source of microplastic particles between 5000 and 50 µm. Variations in particle count extends, however, over several orders of magnitude. Variability and potential effects of microplastics in landfill leachates, including particles <50µm, should be focus of future studies.

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