

Development of a monitoring and data analysis method for microplastic particles in Dutch marine sediments

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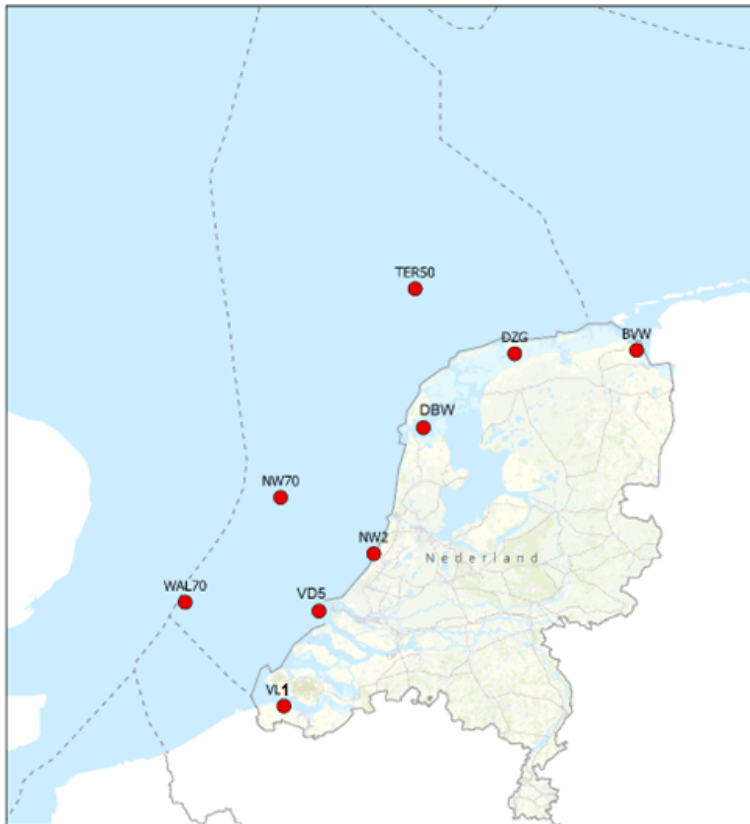
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Dutch monitoring locations for microplastic particles in marine sediments. See Figure 1 for the location names.

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SUMMARY

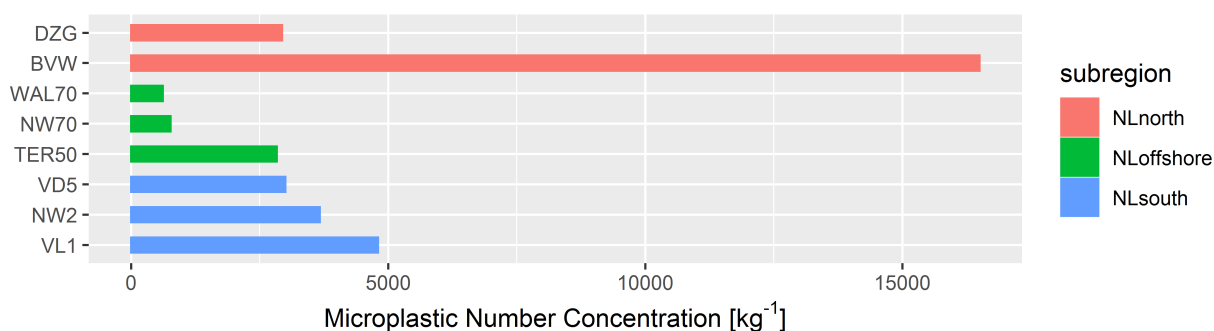
Background and sampling

The Dutch Ministry of Infrastructure and Water Management requires monitoring data for Dutch marine sediments produced with a method specified by the EU Marine Strategy Framework Directive (MSFD, Galgani et al., 2023) and the Oslo Paris convention (OSPAR, CEMP guidelines). This information is needed for national information on the status and trends of microplastic pollution, and for reporting to the EU and OSPAR.

Sampling of marine sediment was performed by Rijkswaterstaat CIV in February 2023. At eight locations in the Dutch coastal zone and offshore, three samples were taken per location.

Key Results

Indicative Microplastic Number Concentrations (MNC) for 2023 (three samples per location) are shown below (see location map on front page).



A relatively high MNC was found at the location Bocht van Watum (BVW) in the Ems-Dollard estuary; which can be explained by the local very high sedimentation rate and organic carbon content. As expected, the offshore locations show lower MNCs, because they are more distant from coastal inputs. Note that no conclusions can be drawn based on these data, because three years of baseline data (N=9 per location), produced with a finalized and validated method, are needed to present results with sufficient confidence. However, we propose that these initial data provide a useful indicative picture of microplastics (MPs) in Dutch marine sediment. A clear indicative relation (N=8) was found between the sediment organic carbon (OC) content and the MNC; illustrating the importance of sedimentation rate, possibly increased by biofouling.

An indicative top-list of polymers (particle number based) found in Dutch marine sediments are shown in the Table below.

region	period	polymer_type	Location PNC*, median, [kg ⁻¹]	N** locations	fraction [%]
NL	2023-2023	Rubber	1142	8	32.8
NL	2023-2023	polyethylene based	1034	8	29.7
NL	2023-2023	polypropylene based	409	8	11.7
NL	2023-2023	cellulose based	330	4	9.5
NL	2023-2023	polystyrene based	209	4	6.0
NL	2023-2023	acrylates/polyurethanes/varnish	134	8	3.8
NL	2023-2023	polyvinylchloride	100	4	2.9
NL	2023-2023	polyamide based	33	5	0.9
NL	2023-2023	polyester based	17	4	0.5

*) Polymer Number Concentration kg⁻¹ dry sediment

***) Number of locations where the specific polymer was found

Method validation

We used the siMPle software (v.1.1.β) for the processing of the μFTIR-FPA files. siMPle produces microplastic hitlists, which were exported and further analysed using a tailor-made script (siMPleR). The siMPle results include for each microplastic particle the following parameters: the maximum, minimum and Feret (sieve) dimensions; the area (ecotoxicologically relevant), the coordinates on the filter, the number of (aggregated) pixels, the polymer type and the Hit Quality Index (HQI). The basic data analysed were the polymer type, max. dimension and max. HQI score.

This siMPleR script was designed to analyse large numbers of microplastic records correctly, efficiently and reproducibly.

We have validated, based on a power relation between particle length (max. dimension) and count, that 50 μm is the correct lower reporting limit for MPs to use with this method.

We applied Polymer-specific Limits of Detection (PLODs), which are more correct to use when complete polymer identifications are available, as in this study. First indicative PLODs indicate low blank levels for a few polymers. For rubber, an indicative PLOD of 52 kg⁻¹ sediment d.w. (N=4) was calculated. This PLOD is well below the reported MNCs for rubber (Annex 4).

A recovery of 94% of polyethylene particles (125-150 μm) was obtained (EU requirement: ≥80%). An additional simple extraction model indicates that for 83% of the replicate samples ≥80% of the sample microplastics was recovered.

The sample amounts used (10 g for muddy sediments and 20 g for sandy sediments) in most cases provided a substantial amount of MPs (>50-100) when three replicates are combined.

The repeatability of the analysis, expressed as the median standard error (SE) of the Microplastic Number Concentrations (N=3 per location) is 26%. This SE for relatively small samples is an improvement compared to earlier results from Bauerlein et al. (2023, Fig. 3) for larger samples. This SE would probably be lower (estimated 15%) if 9 replicates per location would have been measured.

Recommendations

Quality control of the Anodiscs (see Annex 3) and removal of the duplicate results (Table 4) showed that the digestion may be improved further for specific samples, which e.g. contain very high organic carbon content or shell fragments. This improved digestion may result in higher MP recoveries for these specific samples.

We recommend to test the use of μFTIR using silicon filters in transmission mode. These filters enable to measure more complete FTIR spectra, including the selective fingerprint range (700-1850 cm⁻¹; Renner et al., 2019). This will increase the confidence of microplastic identifications and may lead to more identifications of MPs of with maximum score (max. HQI) ≥0.6.

We tested and optimised the settings of the siMPle software (version 1.1.β). It appeared that these settings have a large effect on the number of MPs detected. We therefore recommend using standardised siMPle Pearson weight settings (0/1/1; c.f. Primpke et al., 2020), a generic first probability threshold value of 0.5, and a post-selection of HQI ≥0.6, to provide a microplastic results with optimised sensitivity.

Some additional optimisation tests of the siMPle settings will be performed in 2024.

It is recommended to use polymer reference spectra measured on the same machine as the sample spectra. This may further increase the Hit Quality Indices of identified microplastics.

1. INTRODUCTION

1.1 Definition of Microplastics

Marine Microlitter is defined as a size subcategory of Marine Litter, which is, according to Commission Decision 2017/848/EU and UNEP, “any persistent, manufactured or processed solid material discarded, disposed of or abandoned in the marine and coastal environment”. In the EU guidance (Galgani et al., 2023), microlitter is limited to artificial polymers: “Microlitter particles consist of different materials as for example metal, glass or artificial polymers. This chapter focuses on the artificial polymer component of microlitter as descriptor 10 considers this material the most significant microlitter material in the environment.”

Microlitter is the size subcategory defined as “marine litter with a length of its maximum dimension below 5 mm”. Note that the minimum dimension of microlitter is not defined.

1.2 Information needs

The EU Marine Strategy Framework Directive (MSFD) requires basic information gathered by monitoring of microplastic and micro rubber particles in marine sediments. These information needs have been described in technical detail in the recent EU monitoring guidance (Galgani et al., 2023) and are described in more detail in 2.2.1. Two major requirements are recoveries of reference material of $\geq 80\%$ and to determine the Limit of Detection (LOD). In addition, Netherlands has an international agreement with OSPAR to participate in the candidate indicator for microplastics (MPs) in marine sediments (OSPAR Microplastic expert group, 2023). These guidelines closely follow the EU guidance (recovery $\geq 80\%$, LOD definition), but have an additional demand for the sediment sampling depth (2-5 cm).

1.3 Dutch and UK monitoring context

The Netherlands has been investing in a suitable monitoring method for MPs in marine sediments since 2019 (Bäuerlein et al. 2023; Mintenig, 2021). Recently, a suitable sampling method was developed by RWS-CIV using boxcore sampling (RWS-CIV, 2022). The method development was continued in cooperation with the UK Cefas microplastic laboratory, leading to a pilot report (Bakir 2022) and this first method development report.

Cefas UK has substantial experience in monitoring of MPs in the North Sea, mostly with the Nile red coupled with a micro-FTIR method. However, in 2021 Cefas has started to implement μ FTIR with Focal Plane Array (FPA) detection (Bakir et al., 2023b).

1.4 Monitoring methods

The EU and OSPAR guidelines presented above provide the basic monitoring and validation methods that have to be used. We have applied a relatively simple method based on Tagg et al. (2015; see 2.1). Several additional validation methods have been applied to further improve the method and data quality (see 2.5).

Special attention has been given in this first project year to validation of the data analysis of μ FTIR data using the siMPle software (<https://simple-plastics.eu/>). The siMPle software appeared to be one of the best performing FTIR-data analysis packages (Cowger et al., 2021; Table 1). Moses et al. (2023) reported that the siMPle software reported substantially more MPs compared to the Purity software.

According to Renner et al. (2019), spectral data analysis is an underestimated subject in microplastic analysis, and can have a large effect on the microplastic results produced. This paper points to the relevance of the following subjects: (a) a good understanding of the Hit Quality Index (HQI) and the use of the most suitable calculation criterion (i.e., first derivative of the FTIR spectra); (b) the use of baseline (background) correction of the measured data, (c) the selectivity of different spectral IR

regions (advice to use the fingerprint region (700-1850 cm^{-1}), and (d) the use of reference spectra measured on the same machine as the samples. Our results confirm the importance of the siMPle settings used, and we propose an optimised set of settings (see 3.5.7).

1.5 Aims of this project

- a) to develop and validate a monitoring method for MPs in marine sediments based on FTIR spectrometry.
- b) to develop and apply a data analysis method to the MPs data produced in this project.

2. METHODS

2.1 Global method description

Monitoring parameters

The monitoring parameters are derived from the EU monitoring guidance (Galgani et al., 2023). These include: total number of MPs per kg sediment (dry weight); number of MPs per kg sediment (d.w.) per length class: <20 μm (optional), 20-49 μm (optional); 50-99 μm (optional); 100-299 μm (recommended); 300-999 μm (recommended), 1000-4999 μm (recommended).

Furthermore, the morphology classes: filaments, fragments, films, foams and pellets/granules/beads are recommended to report. Colour classes and transparency are optional.

Polymer identification is partly recommended (at least 10% or a minimum of 20 particles per sample should be validated with FTIR; Galgani et al., 2023) and is partly optional (not all particles have to be analysed using a spectroscopic method such as FTIR). However, Rijkswaterstaat has chosen for μFTIR measurement of all microparticles, to avoid high error rates (up to 60%) by visual microscopic identification (Kroon et al., 2018)

Only microplastic particle data are mandatory. Microplastic mass data are not requested for EU and OSPAR reporting, as motivated in more detail in the EU guidance (Galgani et al., 2023).

Particle size distribution (PSD) is at present not mandatory but under discussion for MSFD (Galgani et al., 2023). Organic carbon (OC) content is optional.

Sampling locations

The EU guidance advises the following selection criteria for locations: "Monitoring strategy for marine microlitter should consider sampling locations based on factors such as proximity to potential sources of litter and microlitter, including the contribution of riverine litter inputs, flow and/or sediment-deposition rates". In view of these criteria, we selected most of the monitoring locations close to the Dutch coast, in particular close to mouths of estuaries, major rivers and in the Wadden Sea (see Figure 1). In addition, we selected three locations in the shipping lanes in the Dutch offshore area.

Sampling

Sampling is performed each year in February; before benthic spawning (starting in March) which may give additional organic pollution of the sediment.

The sampling is organized by Rijkswaterstaat-CIV, using a finalized sampling protocol (RWS-CIV 2022). In short, this protocol is based on boxcore sampling (three samples per location), subsampling of the 2 cm top layer of the boxcore sample, and storage of this subsample in stainless steel jars of 1 L. Quality assurance is in place to minimise plastic contamination. Two field blank samples are taken at different locations each year.

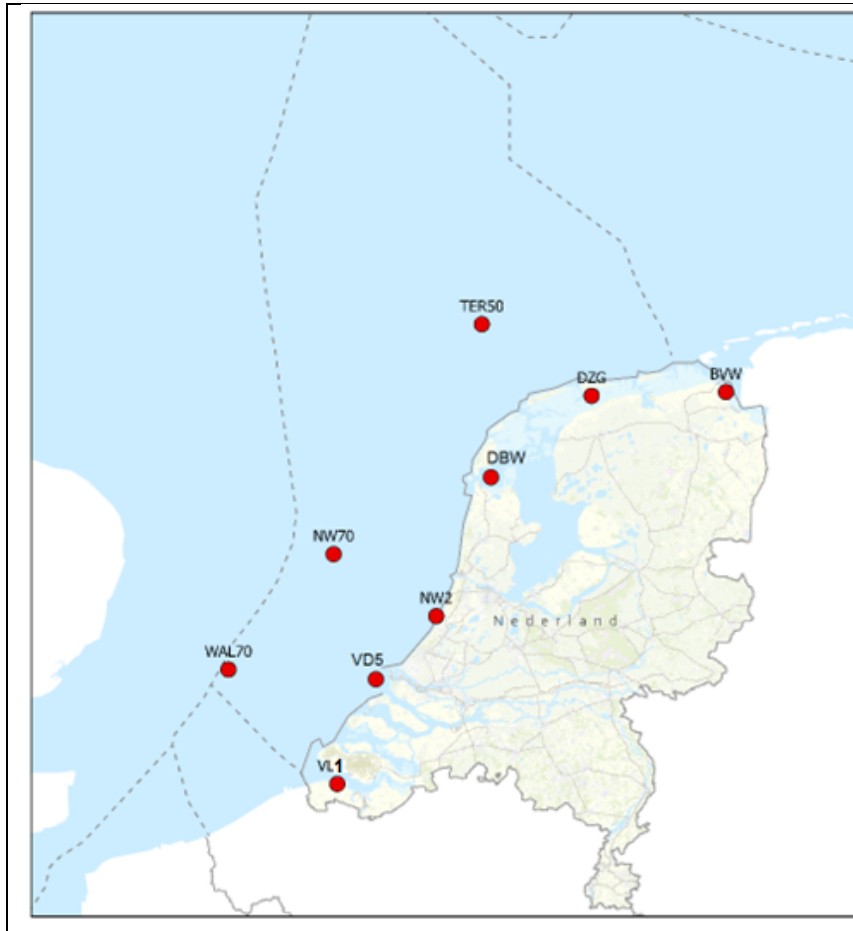


Figure 1: Dutch sediment sampling locations in the coastal zone (6) and offshore area (3). The location names are: Voorland1 (VL1), Voordelta 5 (VD5), Noordwijk2 (NW2), Doovebalg West (DBW, not sampled in 2023), Dantziggat (DZG), Bocht van Watum (BVW), Walcheren 70 (WAL70), Noordwijk 70 (NW70) and Terschelling 50 (TER50).

Detailed information on these locations and samples is presented in Annex 1.

Overview of analytical and data-analysis method

The basic method steps used in the Cefas/RWS method are:

1. Apply careful quality assurance during the analytical process
2. Three boxcore samples are taken at each sampling location and date (Bauerlein et al., 2023)
3. Combination and mixing of the three top layer subsamples (2 cm) per location-date
4. Drying of the combined samples at 40 °C
5. Characterize the combined samples with PSD and OC content
6. Take three subsamples (10-20 g; Table 2) of the composite sample in three separate centrifuge tubes
7. Digest the samples with hydrogen peroxide (H₂O₂) 30% across 48 hours
8. Dry the digested samples
9. Density extraction with ZnCl₂ 1.5 g mL⁻¹ (duplicate extraction); centrifugation of the extracts
10. Filtration of two extracts per sample on two separate Anodiscs. Using ethanol post-flushing
11. Drying of the Anodiscs
12. Measurement of the Anodiscs with μFTIR-FPA
13. Processing of the data files with the siMPle software and optimised settings
14. Quality control of the data files
15. Further data-analysis of the siMPle export files with the siMPleR script
16. Reporting of the validated data file and a concise monitoring report

The detailed analytical and data analysis method is described in the separate Standard Operating Protocol (Bakir et al., 2023; SOP 3091).

2.2 Sample characterization

The composite samples per location were characterized using PSD and OC analysis, respectively. Methods for PSA and OC are specified in Bakir (2023bb). Data for 2023 were combined with recent mud/OC data from Bakker for comparable locations and presented in a relation plot. Mud is defined as the sediment fraction <63 µm.

2.3 Method optimization

The digestion temperature was measured and optimised to stay below 40 °C (Galgani et al., 2023). The optimized procedure is: muddy dry samples were digested directly. For sandy samples, minimal wetting of the samples (with a water film on top of the sediment <1 mm) was used to reduce the digestion temperature <40 °C.

The digestion quality was evaluated by imaging and visually assessing filter coverage of each Anodisc (Annex 3).

Density separation was carried out on dried samples. Extraction of dried sediments was consistent with Cefas protocols and was also in accordance with NIVA processes (Alling et al., 2023). Extraction on dried sediments offer several practical advantages, including avoiding the dilution of the zinc chloride solution (density 1.5 g mL⁻¹). This minimally required density for MPs also provides a more effective separation of fine sediment particles by centrifugation. It remains however a possibility to use sample prewetting directly before density extraction to possibly increase the recovery (Filgueras et al., 2021).

FTIR scanning with 2 sample and background scans (instead of 1) was tested to possibly improve the signal-to noise-ratio ratios of the spectra (Loder et al., 2015).

2.4 Quality Assurance and Quality Control

2.4.1 Quality assurance (QA) contains the following main components:

- (a) creating and maintaining clean laboratory conditions, among others by daily laboratory cleaning and working in a laminar flow cabinet;
 - (b) using plastic-free laboratory clothing and equipment wherever possible;
 - (c) using microfiltration to remove plastic contamination from liquids used in the sample pretreatment processes.
 - (d) Removing polypropylene particles from the Anodisc support ring from the results file.
- Additional details on the Quality Assurance used can be found in SOP 3091 (Bakir et al., 2023a).

2.4.2 Quality control (QC) contains the following main components:

- (a) Take an image of the first sample extract on Anodisc and visually assess the quality of the digestion.
- (b) Exporting two siMPle particle maps containing all particles and only MPs, respectively. The map with all particles is used to assess if the cleanup of natural particles is sufficient.
- (c) Exporting two siMPle datafiles with all particles and only MPs, respectively. The file with all particles quantifies the number of co-extracted natural particles.
- (d) Removing duplicate records using the siMPleR script. See the script manual for more information (Walvoort et al., 2023).
- (e) Analyzing blank samples for both the field (atmospheric blanks) and the laboratory only.

2.5 Method validation

2.5.1 Lower length limit

The lower length reporting limit (LLL) was determined by combining all sample MP records from 2023 with a siMPle Max. (index) Score of ≥ 0.6 . The number of MPs per bin of 50 μm was counted and plotted against the mean length of the specific bin. By exception, a bin of 20-49 μm was used because 20 μm is the FTIR detection limit. A power function was plotted through the number of data points per bin (Bäuerlein et al., 2023). The lower particle length limit was set where the lower bin count clearly deviates from the power function, thus showing a clear decrease of recovery and FTIR detection.

2.5.2 Recovery of Polyethylene reference particles

The recovery of polyethylene (PE) fluorescent red spheres (125-150 μm ; Cospheric) was determined by spiking 30 PE particles to water (N=2), sandy sediment (N=2) and muddy sediment (N=2). The extraction was performed using the SOP 3091, including a post-flushing using filtered ethanol in the centrifuge tube and filtration funnel.

2.5.3 Recovery of sediment MPs

In addition to use of a PE reference material, estimations were made of the recovery of sediment MPs using the ratio R of the number of particles in the second and first extract. This ratio is used to define a simple exponential model, which is used to estimate the recovery of sample MPs in additional extraction steps, and to estimate total recovery of the first two extraction steps.

For example, if the first extraction results in 10 MPs and the second in 5 MPs, $R = 0.5$. This leads to estimations for the third extraction ($0.5 \cdot 5 = 2.5$) and for the fourth extraction ($0.5 \cdot 2.5 = 1.25$). The extrapolation is stopped when a value of approx. 1 is reached. In this example, the total estimated number of MPs = $10 + 5 + 2.5 + 1.25 = 18.75$. The estimated recovery of extractions 1 + 2 is then: $15 / 18.75 = 80\%$. Therefore, we aim for an R value ≤ 0.5 , to obtain an estimated recovery of sediment MPs of ≥ 80 .

2.5.4 Detection limits

Procedural laboratory blanks and field blanks were combined to calculate polymer specific detection limits (PLOD) for particles $\geq 50 \mu\text{m}$. The PLODs were calculated using the mean polymer blank values plus three times the standard deviation (Galgani et al., 2023).

See SOP 3091 for details on the blank determination methods.

Two field atmospheric blanks were obtained by opening an empty sample container, with 30 mL of RO water, during collection of the top layer of sediment from the boxcore sample (RWS-CIV, 2022). The resulting water samples were treated as a sediment sample.

2.5.5 Sample load

The linear sample load range was determined by analysing sample amounts of 5, 10, 15 and 20 g of very muddy (BVW) and sandy sediment (NW2).

The minimum sample amount needed for analysis can probably best be based on the desired minimum number of MPs per sample. The TGML guidance indicates that ≥ 20 particles per sample are needed (Galgani et al., 2023) and this guideline is used in this method to choose the sample load. In addition, we use an indicative maximum of 100 MPs per filter, to avoid overlapping of MP particles (Xu et al., 2022)

2.5.6 Measurement uncertainty

Measurement uncertainty is defined here as two times the standard deviation of the recovery of reference particles from a realistic sample matrix. We will test the calculation of the Measurement Uncertainty based on eight polyethylene recovery results (NEN 7779, 2018). These results will become available in 2024. In 2023 four results were determined (see 2.5.2).

2.5.7 siMPle validation

The siMPle software was used because it is one of the leading FTIR data analysis packages; it is well documented and validated (Primpke et al., 2018; 2020), freeware and used by many European laboratories. We used siMPle version 1.1.β, combined with the simple database version 1.0.1 (<https://simple-plastics.eu/>). This software version provides "Max. (Hit Quality Index, HQI) scores" which are essential in the selection and reporting of confident microplastic identifications.

The resulting microplastic hit lists are exported to CSV files and further analysed with the custom-made siMPleR script described below.

The use of default siMPle settings for the Pearson weight factors for original spectra (0), first derivative spectra (1) and second derivative spectra (1) were compared to the Pearson weight factors recommended by Bruker (1, 1, 0, respectively) for three samples (VL1, NW2, BVW).

The use of generic siMPle first probability threshold values of 0.50, 0.55 and 0.60, respectively; combined with second and third probability threshold values of 0.4 and 0.3; and combined with a post-selection of MP records with a max. HQI score ≥ 0.6 , was investigated. The aim of this test was to investigate if the use of a lower TV setting, at 0.5 and 0.55 which is still considered to be acceptable (Renner et al., 2019), results in a more sensitive detection and reporting of more MPs.

Duplicate records (with the same coordinates, polymer and max. score) are produced by siMPle now and then. The meaning of these duplicate records in the siMPle export files was investigated using siMPle particle plots for several examples.

2.6 Data post-processing using siMPleR

The polymer type names in siMPle export files are converted to polymer group names (e.g. polyethylene based) needed for EU reporting (Galgani et al., 2023).

We developed the siMPleR script (in R) for the post-processing of the large number of microplastic records produced by siMPle. This script basically performs the following steps (Walvoort et al., 2024):

- Removal of duplicate/triplicate records from the siMPle results files with the same X,Y coordinates, polymer type and max. score. The duplicate record with the largest length is maintained because it is regarded to be the most likely. This cleanup step is validated in this study (see 2.5.6)
- Unlikely polymers (polyetheretherketone, polycarbonate, polylactic acid) were selected in the ≥ 0.7 max. score range because they occur in less than 3 locations at a median based percentage $< 1\%$ (Table 6) These unlikely polymers were included in the script file "polymers-excluded.csv" and were excluded from the results in the max. score 0.60-0.69 range. This microplastic identification procedure is based on Leistenschneider et al. (2021), in which identifications with a HQI ≥ 0.7 are directly accepted; and hits in the HQI range 0.6-0.69 are subjected to manual inspection. Instead of manual inspection, the siMPleR script automatically removes unlikely polymers from the MP particle list in the 0.60-0.69 range. We propose that this method results in MPs identifications in the 0.60-0.69 range with sufficient confidence. In 2024, we plan to start using silicon filters, which will provide IR spectra including the fingerprint range (700-1850 cm^{-1}) and will further improve the confidence of the identifications.

- Analysis and reporting of the mean Microplastic Number Concentration (MNC) per location, grouped per subregion. Comparison of the mean and median values per location (3 replicates) show that these values are comparable (see Annex 6), and that there is no need to use median location values.
- Analysis and reporting of the mean Polymer Number Concentration (PNC) per location, grouped per subregion.
- Reporting of figures of MNC and selected polymers (using the polymers-figures.csv file).

Note: We grouped three locations in the South of NL (VLM, VDA5 and NW2; Figure 1), three locations in the North of the Netherlands (NL; BVW, DZG and BVW), and three locations in the offshore area (WAL70, NW70 and TER50) (see Figure 1). This limited spatial grouping provides a wider spatial picture (c.f. beach litter assessment) and prevents that too much is concluded from results for a single location.

2.7 Sample results

The following results are reported:

- 2.7.1 MNC per location (mean).
- 2.7.2 PNC: top list and concentrations per location.
- 2.7.3 A complete cleaned microplastic data table (duplicates and unlikely polymers removed). This table is suitable for additional data analysis at the sample level, database storage and OSPAR/EU reporting.
- 2.7.4 A table with the detailed total count results for the individual replicate samples and extracts.

3. RESULTS AND DISCUSSION

The paragraph numbering follows the numbering in the Method section.

3.2 Sample characterization

The relation between the mud content (<63 µm) and the OC content is shown in Figure 2.

The PSDs for all locations are presented in Annex 1 for illustration.

It appears that until a mud content of 64%, a linear relation exists between mud and OC content ($R^2 = 0.95$). Above 64% mud, the OC content increases more strongly. The conclusion is that mud and OC content are strongly related (showing a bimodal relation), and that it is sufficient to only measure the mud content and to estimate the OC content using this relation. Note however that above 64% mud OC estimation are less accurate due to fewer data points.

The mud content is not yet a parameter required for EU MSFD reporting. Since we now know the mud and OC content of the Dutch monitoring locations, it can be argued that it is not necessary any more to measure PSDs and OC for the Dutch monitoring samples in the future. On the other hand, monitoring the PSD and OC of sediment samples will enable the detection of potential variations of the sediment composition in the future.

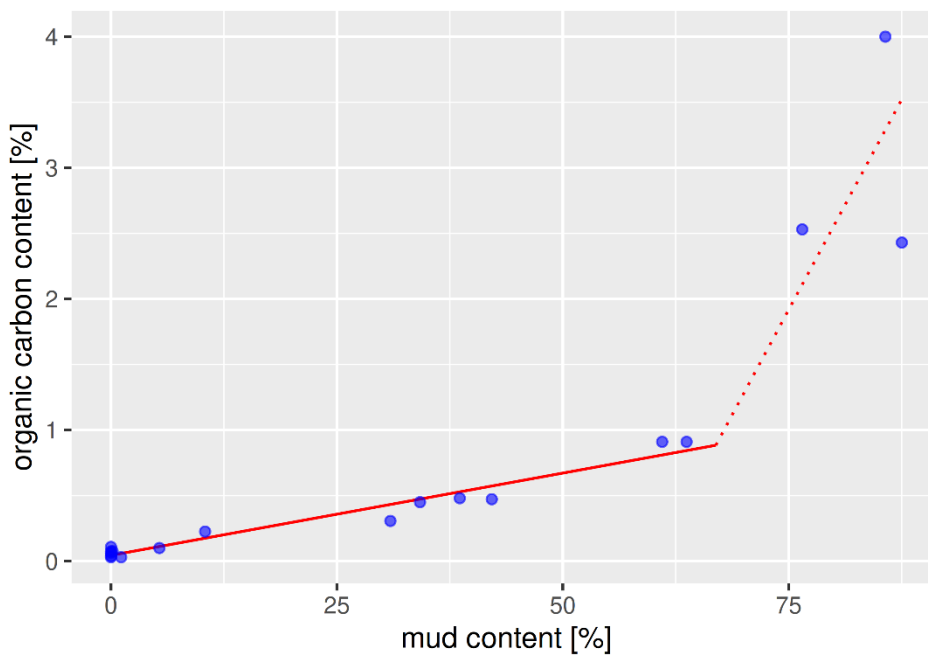


Figure 2: Relation between the mud content and organic carbon (OC) content of Dutch marine samples. The model formula of the left part of the curve is: $OC\% = 0.044 + 0.0125 * mud\%$ ($R^2 = 0.96$).

3.3 Method optimization

3.3.1 Digestion temperature

The temperature plots during sample digestion are shown in Figure 3.

The results showed that the temperature of muddy sediments remain below 40 °C during digestion, while the temperature of sandy sediments increased slightly above 40 °C during digestion. This can be explained by the better conduction and dissipation of thermal energy in the finer muddy sediments, compared to the coarser sandy material. To prevent this effect in sandy sediments, we use prewetting of sandy sediment, with a small layer of water (<1 mm) on the sand, before adding

the peroxide. This decreased the digestion temperature due to the improved heat dissipation. The slight dilution of the peroxide is not a problem due to the low OC content of sandy samples. Muddy samples are digested dry, because prewetting is not necessary and unlikely to improve the digestion of OC.

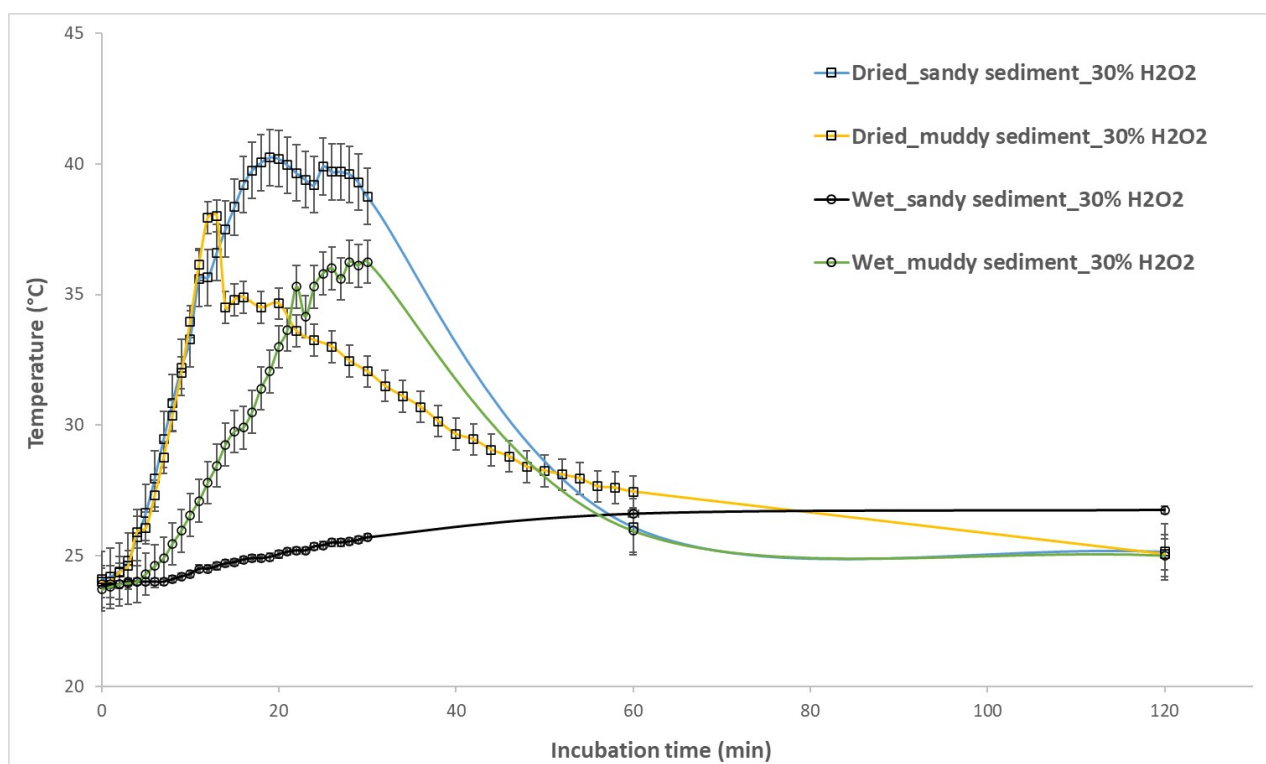


Figure 3: Temperature-time profiles during digestion of natural organic material in samples using 30% hydrogen peroxide (H_2O_2) and dry and wet sediment, respectively.

3.3.2 Digestion quality

The quality of the sample digestion is judged from pictures of every first Anodisc within a sample series (see Annex 2).

A question is if the digestion still should be improved further. The Anodisc images in many cases show a reasonably clean filter. This is caused by a combination of the relatively low sample amounts (10 g), the digestion and the separate filtration of the two extracts.

However, for Bocht van Watum (BVW), Voorland1 (VLIS*) and Terschelling50 (TER50) the sample cleanup can be improved.

For Bocht van Watum, an obvious option is to reduce the sample amount to 5 g, because the microplastic counts are large in this sample (see Table 4). In addition, it is proposed to test Fenton reagent on this sample, as used by NIVA (Hurley et al., 2018)

For Terschelling50, the 10 g of sample is needed to obtain the minimally needed MP count per sample (20), so an improved digestion seems to be necessary. It is proposed to test the addition of Fenton reagent for this sample (Hurley et al., 2018).

3.3.3 Dry or wet extraction

We used dry extraction in view of several practical advantages of using dry samples.

However, the results in Table 4 show that the recovered number of MPs in the first extract is not always high enough compared to the second extract. This may partly be caused by using dry samples for density extraction, as reported by Filgueras et al. (2021). However, this may also partly be caused by using an extraction liquid with insufficient density (1.5 g mL^{-1}), or by insufficient digestion

of organic material. However, we prefer to keep the extraction liquid density of 1.5 g mL^{-1} , because this limits the amount of co-extracted mud particles and makes it possible to centrifuge samples alone, without sieving. We want to refrain from sieving in the low size range ($20\text{-}50 \text{ }\mu\text{m}$), because we think that this may lead to losses, damage of MPs and further contamination. With the present method (without sieving), the recovery of MPs $\geq 50\mu\text{m}$ appears to be good (see 3.5.1).

We therefore plan to investigate this further by using (a) minimal prewetting of samples with ZnCl_2 (1 hour wetting time; 1 out of 3 replicates) prior to extraction and (b) by improving the digestion prior to extraction (see 3.3.2).

3.3.4 FTIR analysis

Loder et al. (2015) recommended the use of 6 co-added scans at a resolution of 8 cm^{-1} with 4×4 binning in the range $3600\text{-}1200 \text{ cm}^{-1}$ for the measurement of large areas using FPA (64×64) when analysing MPs in environmental samples. Due to the nature of the Lumos II FPA (32×32) used for this study, it was not feasible to use the settings recommended by Loder et al. (2015) for two main reasons: i) the size of the outputs was too large for storage and processing and ii) the extended analysis time (e.g. min of 7 hours for 2 scans) led to the warming up of the FPA detector and required frequent addition of liquid nitrogen which was not time and cost-effective. As a result, a single scan per measurement was selected. Other options could be explored in the future, especially with the use of more advanced chemical imaging systems and larger FPAs (i.e. 64×64 or 128×128).

This may further increase the number of MPs with a max. score ≥ 0.6 , by using e.g. 2 to 6 sample and background scans (Loder et al., 2015). However, a good balance needs to be met between speed and accuracy of analysis as well as operational costs which could lead to costly yearly monitoring programmes, especially for large scale mapping.

3.4 Quality Assurance and Quality Control

3.4.2 Quality control

- The first Anodisc of each sample series is imaged to assess the quality of the sample digestion (see Annex 2). The results of this QC indicate (see discussion in Annex 2) that for several samples some additional cleanup may be necessary. This will be tested in the 2024 project.
- Results for the field and laboratory blanks results are presented in Table 5. These results are also presented at the polymer level because polymer-specific detection limits are used. The blank results are discussed in more detail in paragraph 3.5.4.
- Detailed extraction results per sample and extract are presented in Table 4.

3.5 Method validation

3.5.1 Lower length limit

The power function obtained with the data is: $\text{bin count} = 3.004\text{e}+07 * (\text{mean MP length})^{-2.472}$

The bin count results, for all 2022 samples combined (Figure 4), show that below a particle length of $50 \text{ }\mu\text{m}$ (see red dot) the recovery of MPs clearly decreases below the modelled power plot. This deviates from theory, because it is known that at lower MP sizes the number of particles increases (Bäuerlein et al., 2023). This phenomenon can probably be explained by a known lower extraction efficiency of MPs $< 50 \text{ }\mu\text{m}$, combined with a decreased detectability of these MP sizes in a sample matrix. We therefore conclude from this plot that a lower length limit (LLL) of $50 \text{ }\mu\text{m}$ is valid to use for the reporting of MPs using this method. This LLL is also used by NIVA (Van Bavel et al. 2022).

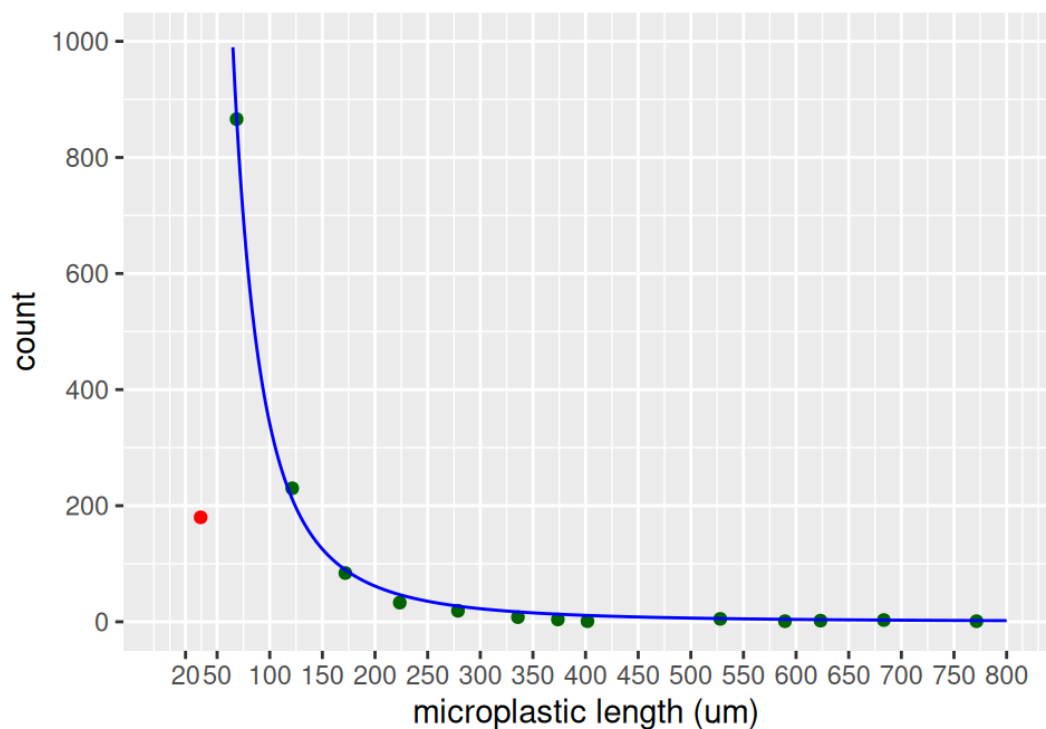


Figure 4: Particle length distribution for all 2023 microplastic and micro rubber data combined (Max. score ≥ 0.6). The fraction 20-49 μm (red dot) was excluded from the model. Parameters of the power function are: $b_0 = 3.004e+07$; $b_1 = -2.472$.

3.5.2 Recovery of polyethylene reference particles

Table 1: Recoveries of PE reference particles (125-150 μm).

Matrix	Recovery 1 [%]	Recovery 2 [%]	Mean recovery
Water	100	97	99
Sand	100	87	94
Mud	90	97	94

The recoveries for the PE reference material are $>90\%$; and are higher than the required 80% recovery (TGML, 2023). This is partly caused by the post-flushing with ethanol, combined with the excellent visibility of the fluorescent particles under blue light (420-470 nm).

3.5.3 Recovery of sediment MPs

In addition to the use of this reference material, we also modelled the recovery of real-life MP particles using the recovery data from the two extracts per sample. These extraction results are shown in Table 2.

Table 2: Microplastic counts at the location-replicate-extract level for the year 2023. All samples were taken in February 2023. The ratio of the two extracts per replicate is presented to estimate the total recovery. Ratios >0.5, which may indicate a recovery <80%, are marked.

Location	Replicate	Sample amount [g]	Extract1 count	Extract2 count	extract2/extract1	Total count
BVW	1	10	75	75	1	150
BVW	2	10	72	11	0.15	83
BVW	3	10	178	84	0.47	262
DZG	1	10	37	0	0	37
DZG	2	10	14	3	0.21	17
DZG	3	10	29	5	0.17	34
NW70	1	20	11	5	0.45	16
NW70	2	20	15	3	0.2	18
NW70	3	20	11	1	0.09	12
TER50	1	10	15	4	0.27	19
TER50	2	10	43	4	0.09	47
TER50	3	10	16	3	0.19	19
WAL70	1	20	11	5	0.45	16
WAL70	2	20	15	3	0.2	18
WAL70	3	20	3	0	0	3
NW2	1	20	93	2	0.02	95
NW2	2	20	30	15	0.5	45
NW2	3	20	45	35	0.78	80
VD5	1	10	42	0	0	42
VD5	2	10	27	9	0.33	36
VD5	3	10	12	0	0	12
VL1	1	10	27	21	0.78	48
VL1	2	10	27	31	1.15	58
VL1	3	10	31	7	0.23	38

It appears that for 83% of the replicates the desired ratio $E2/E1 \leq 0.5$ is obtained which we regard as an acceptable result. The samples for which $E2/E1$ is >0.5 are indicated in yellow in Table 4. We consider it desirable to obtain ratios ≤ 0.5 , which correspond to an estimated recovery $\geq 80\%$, for $\geq 90\%$ of the samples. For BVW, we will reduce the sample amount to 5 g to improve the ratios. For VL1, we have little margin to reduce the sample amount to 5 g, because this would reduce the total MP count to around the minimum of 20 per sample. Therefore, we will try to improve the digestion of the Voorland1 samples.

The use of silicone filters, combined with an improved digestion, will hopefully increase the number of positive identifications with Max. Score ≥ 0.6 . The use of triplicate samples per location-year (see Table 4) has an additional advantage that the mean values become more robust than using duplicate samples; and can be used effectively at the annual level (see annex 6).

3.5.4 Limits of Detection

The blank samples (field and laboratory) appeared to be relatively clean, compared to the related sample amounts. The calculated polymer-specific LODs (PLODs) are presented in Table 3 below.

Table 3: Indicative PLODs for the polymers found in blank samples.

Polymer type	LOD [kg ⁻¹]	In blank samples:
Acrylates/polyurethanes/varnish	35	NW2
Polyethylene based	25	NW2
Polyester based	15	LAB1
Rubber	52	NW2, TER50, LAB1

For these four polymers, concentrations found in the sediment samples (see Figure 5) were above the indicative PLODs. However, at least eight blank results are needed to provide sufficiently reliable PLODs.

The use of field and laboratory blanks is yet to be evaluated when more results are available.

At present, the blank values do not indicate that the LODs are limiting in the reporting of polymer concentrations.

We propose that, if full spectroscopic data are available, polymer-specific detection limits are more correct to use than LODs based on total counts. For example, if a specific polymer would never occur in blank samples, it would be incorrect to apply an LOD based on MNCs (all polymers combined) to this specific polymer.

3.5.5 Sample load

It appeared that for BVW (muddiest sample) a maximum sample amount of 10 g could be used. Using 10 g of sample, the mean number of MPs (N=27) was approx. two times as high as for 5 g of sample (N=11). This illustrates the expected linear relation between sample load and the number of MPs. At 15 g, the Anodisc filter clogged rapidly and extracted sample could not be filtered completely. We have demonstrated in Table 1 that even a sample amount of 5 g is enough for BVW, in view of its high MNCs.

For NW2 (sandy sample) 20 g of sample could be used, which is the physical maximum amount that fits well into the centrifuge extraction tube. However, the results in Table 1 suggest that 10-15 g of this sediment may give better R values, while still providing sufficient MPs in a triplicate analysis (>100).

3.5.6 Measurement uncertainty

We do not have already enough recovery data for polyethylene (N = 8 for sediment needed; 4 results available) to calculate the measurement uncertainty. We will finalize the necessary dataset in 2024.

3.5.7 siMPLe validation

The results for the comparison of the siMPLe default and Bruker settings (suggested during machine installation) are presented in Table 4 below.

Table 4: Comparison of microplastic counts using the default and Bruker siMPle weight settings.

Sample*	Weight settings	TV setting	N total ≥ 0.60	N 50-99	N 100-299	N ≥ 300
NW2_R1_E1	default	0.6	83	57	25	1
NW2_R1_E1	Bruker	0.6	14	9	4	1
VL1_R1_E1	default	0.6	24	17	7	0
VL1_R1_E1	Bruker	0.6	15	11	4	0
BVW_R1_E1	default	0.6	71	46	22	3
BVW_R1_E1	Bruker	0.6	18	10	6	2

*) for each sample, replicate 1 and extract 1 are used.

The conclusion from these results is that using the default siMPle weight settings 2 to 6 times more MPs are obtained in the three test files than when using the Bruker weight settings. These results show the importance of using optimised and standardised siMPle settings to increase MP detection and to reduce the number of false negatives.

These default siMPle settings were already recommended by Pimpke et al., (2020). These results also demonstrate the need to standardise the siMPle settings used within OSPAR and Europe, in case this software is used. In a wider sense, we propose to adopt siMPle software as the standard microplastic software in OSPAR and Europe in cases for which FPA analysis is being applied. This will most likely improve the comparability of the microplastic results from different chemical imaging systems.

Using a siMPle generic threshold value of 0.55 or 0.50 led to 5 to 10% more MPs being found after selection with a Max score ≥ 0.60 . This suggests that at a TV setting of 0.50 the siMPleR analyses the data more precisely than at a TV setting of 0.60. The additional duplicate records that are produced at a lower TV setting produces are removed by the siMPleR script, so this is not a problem for the correctness of the final MP list. For comparison, it was reported that the siMPleR software produces substantially more MP hits compared to the Purity software (Moses et al., 2023), which shows that the siMPle software is more sensitive. The method used here appears to be useful to further increase the sensitivity of the siMPle MP identification process.

Table 5: number of microplastic counts (Max. HQI score ≥ 0.6 , max. dim $> 50 \mu\text{m}$) as related to the siMPle TV setting used.

Sample	Weight settings	TV setting	N total	Comment
NW2_R1_E1	default	0.50	93	More duplicate records compared to 0.6
NW2_R1_E1	default	0.55	110	More duplicate records compared to 0.6. The sample load seems to be too high.
NW2_R1_E1	default	0.60	83	
VL1_R1_E1	default	0.50	27	No duplicates
VL1_R1_E1	default	0.55	24	No duplicates
VL1_R1_E1	default	0.60	24	No duplicates
BVW_R1_E1	default	0.50	76	More duplicates. Probably due to large number of MPs on Anodisc
BVW_R1_E1	default	0.55	74	Less duplicates compared to TV 0.50
BVW_R1_E1	default	0.60	71	Much less MPs $< 50\mu\text{m}$ found compared to TV 0.50. This illustrates the reduced sensitivity for MP detection.

An example of pixel plots of two duplicate records are shown in Figure 5.



Figure 5: pixel plots of duplicate records, produced by siMPle, for a rubber particle, composed of 5 pixels (left) and 6 pixels (right), respectively.

This example, and four other examples (not presented), clearly show that the duplicate particles represent the same particle, with a difference of one or two particles. The cause of the production of these duplicate records is not clear. Maybe these duplicate pixels plots are produced by duplicate scanning of the same particle by the siMPle software. Anyway, the tested pixel plots (N=5) all show that it is valid that the siMPleR script selects only one of these duplicate records for further analysis.

3.6 Data analysis

As a first step in the production of sample results, we combined all microplastic records from all samples with a length $>50 \mu\text{m}$ and Max. HQI score ≥ 0.7 (confident identifications) and calculated their total count and percentage (see Annex A). Using this overview table of polymers, we concluded which polymers are unlikely to occur ($<1\%$ on <3 locations). We therefore removed polycarbonate, polyetheretherketone and polylactic acid from the polymer list in the max. score range 0.60-0.69, because these polymers are less likely to be correct.

3.7 Sample results

3.7.1 Microplastic number concentrations

The MNCs (number of particles kg^{-1} d.w. sediment) at each location, and grouped per subregion, are shown in Figure 6 and Table 5, respectively.

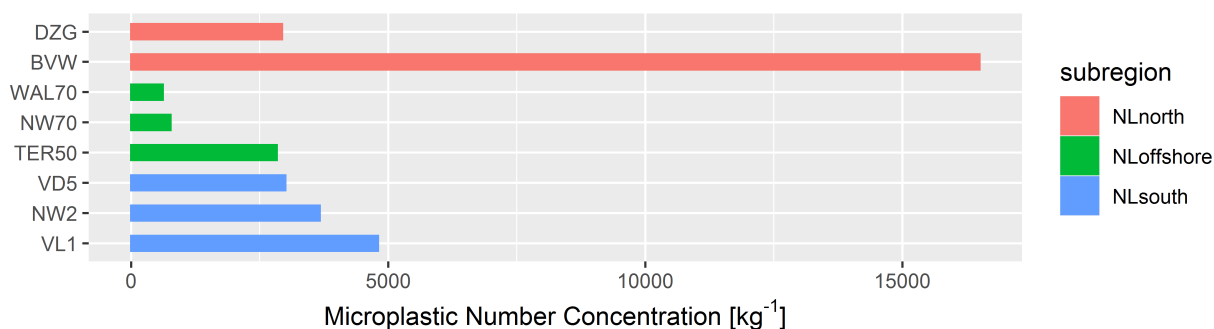


Figure 6: Mean Microplastic Number Concentrations [kg^{-1} d.w. sediment] for locations, grouped per subregion.

It appears from Figure 6 that the MNC in Bocht van Watum (Ems-Dollard estuary) is by far the highest in the Dutch marine waters. Furthermore, it appears that the offshore locations have lower

MNCs compared to the southern coastal locations. This is not unexpected, because of the larger distance to estuarine and riverine sources. In 2024, one additional northern coastal location (Doovebalg West) will be sampled and measured. Note that no conclusions can be drawn based on these data, because three years of data (N=9 per location), produced with a finalized and validated method, are needed to present with sufficient confidence (NEN7777, 2012). However, we propose that these first data provide a useful indicative picture of MPs in Dutch marine sediment. For LOD values, see paragraph 3.5.4. The MNC results are presented in more detail in Table 5.

The MNCs reported in Lorenz et al. (2019, Fig. 1) for the Dutch coast are in general substantially lower than the coastal concentrations reported in Table 5. This may be explained by the good recoveries and/or more sensitive data analysis settings of the method reported here. The results from this study in the length range $\geq 100 \mu\text{m}$ are approximately two times as high as the results of Bauerlein et al. (2023) for the locations NW2 and VLIS (close to VL1), and much higher for the location BVW. This comparison suggests that the method reported here is more sensitive than the method used by Bauerlein et al. (2023), although increases in environmental MP concentrations may also play a role.

Table 6: Indicative Microplastic Number Concentration (MNC) results for MPs $\geq 50 \mu\text{m}$ and with max. score ≥ 0.6 . Results are rounded to three digits.

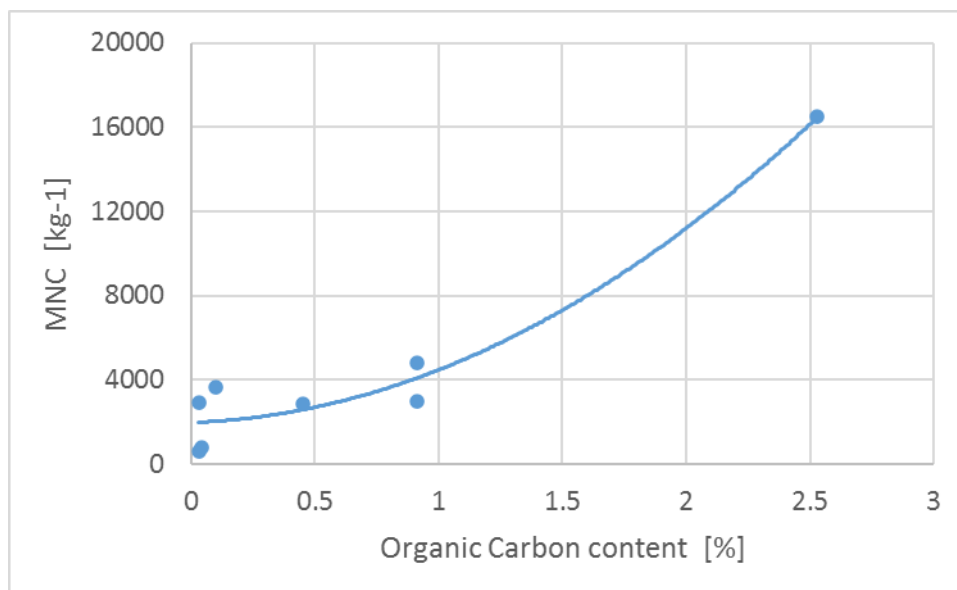
Subregion	Location code	Period	MNC_location [kg^{-1}]	SE [kg^{-1}]	N samples	N MPs
Nlnorth	DZG	2023-2023	2930	623	3	88
Nlnorth	BVW	2023-2023	16500	5220	3	495
NLoffshore	WAL70	2023-2023	617	235	3	37
NLoffshore	NW70	2023-2023	767	88	3	46
NLoffshore	TER50	2023-2023	2830	933	3	85
NLsouth	VD5	2023-2023	3000	917	3	90
NLsouth	NW2	2023-2023	3670	741	3	220
NLsouth	VL1	2023-2023	4800	577	3	144

We expect that three years of data with a finalized and validated method will be necessary to produce a valid status assessment of MNCs per location. In that situation, the use t-tests will be possible to determine if the differences of MNCs at different locations are significant.

We calculated the correlation between the OC and mud content, respectively, and the MNC of the eight samples (Figure 5). This indicates a clear relation between OC content and MNC ($R^2 = 0.95$; but largely determined by the high value (leverage effect)). This relation can be explained by the known relation of MNC and sediment deposition rates, and possibly increased by biofouling with OC. The correlation coefficient with the mud content ($< 63 \mu\text{m}$) is much poorer ($R^2 = 0.48$). This result suggests that the OC content, also in view of its relevance for digestion, is a relevant sediment parameter to estimate the MP content, at least within a limited spatial region such as the Netherlands. This finding will be corroborated with more results in the coming years.

Figure 5: Indicative relation between the sediment organic carbon content and the Microplastic Number Concentration (MNC) for eight marine sediment locations.

Model formula: $y = 1959 + 400x + 2108x^2$ ($R^2 = 0.95$).



3.7.2 Polymer number concentrations

The top list of most common polymers in Dutch marine sediments, based on mean numbers for all sample data of 2023, is presented in Table 7. The polymer type concentrations per location are presented in Annex 4.

Table 7. Polymers found in Dutch marine sediment samples, sorted on decreasing PNC. Note that this polymer list is indicative; and more data obtained with a larger FTIR scanning window (including the full fingerprint range) will be produced in 2024.

region	period	polymer_type	Location pnc, median, [kg ⁻¹]	N locations	median [%]
NL	2023-2023	rubber	1142	8	32.8
NL	2023-2023	polyethylene based	1034	8	29.7
NL	2023-2023	polypropylene based	409	8	11.7
NL	2023-2023	cellulose based	330	4	9.5
NL	2023-2023	polystyrene based	209	4	6.0
NL	2023-2023	acrylates/polyurethanes/varnish	134	8	3.8
NL	2023-2023	polyvinylchloride	100	4	2.9
NL	2023-2023	polyamide based	33	5	0.9
NL	2023-2023	polyetheretherketone	33	1	0.9
NL	2023-2023	polycarbonate based	25	2	0.7
NL	2023-2023	polyester based	17	4	0.5
NL	2023-2023	polylactic acid	17	1	0.5

Note that in Table 6 polymers which were only found once or twice at a median location <1% (marked yellow), are removed from the results in the max. index score range 0.60-0.69 because they are less likely to occur and may be false positives within this range.

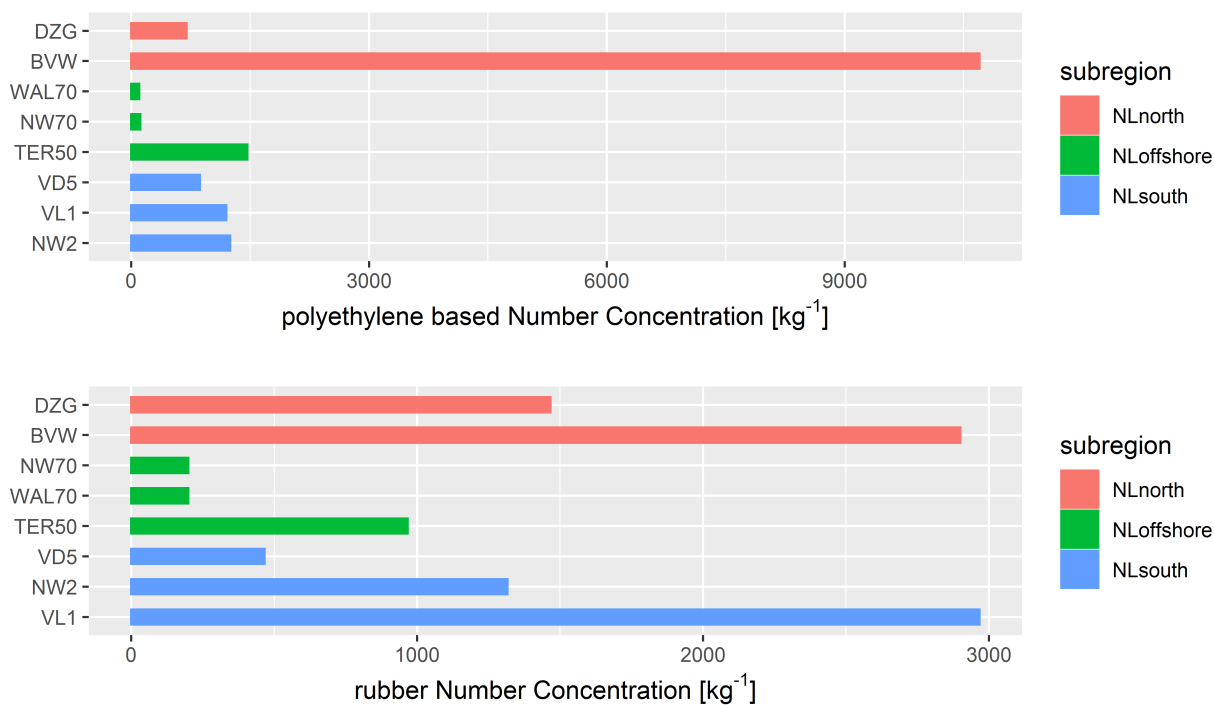
The siMPle software reported relatively larger numbers of chlorinated polyethylene in the 2023 samples. Although this polymer has also been reported in North Sea sediment (Lorentz et al., 2019) we think that this may partly be misidentifications due to a partial lack of spectral information in the fingerprint range (700-1250 cm^{-1}) due to absorption of the Anodisc filter. The study from More et al. (2022) indicated that there are two characteristic IR peaks in the fingerprint region which discriminate polyethylene from chlorinated polyethylene. We therefore reported these MPs in this report as Polyethylene based, in line with the TGML guidelines (Galgani et al., 2023). Furthermore, we will try to implement the use of silicon filters in 2024, which provide full spectral data in the fingerprint range (Martinez et al., 2023). These upcoming data are expected to enable more reliable identifications of polyethylene and chlorinated polyethylene, respectively.

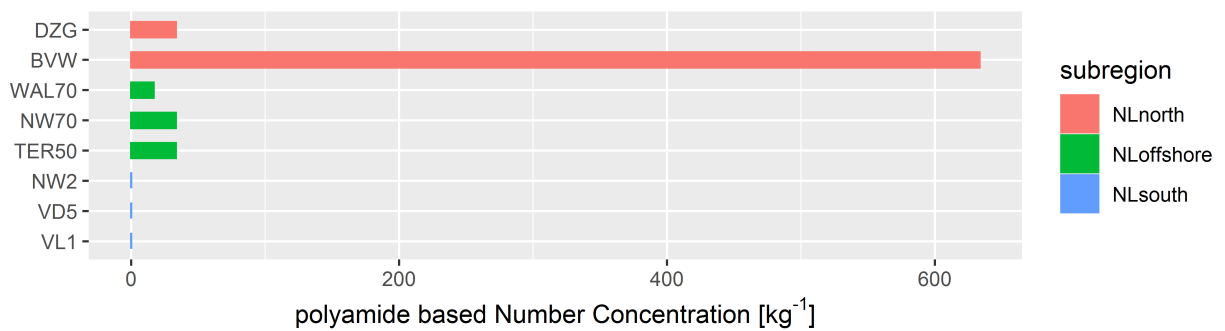
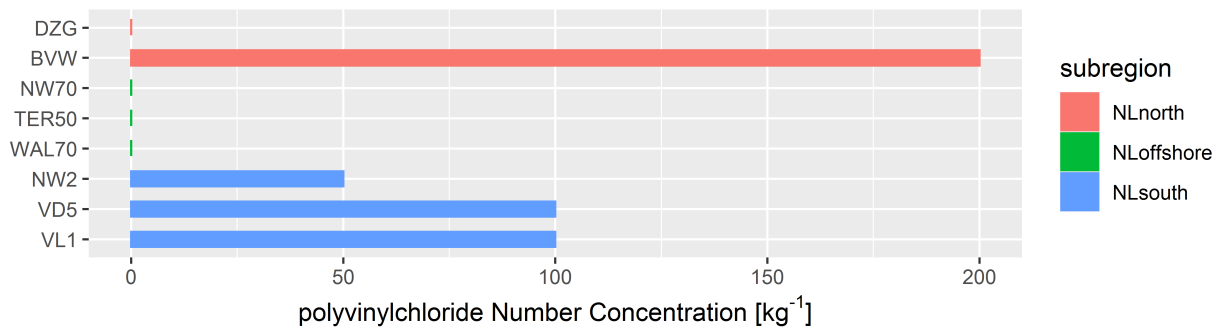
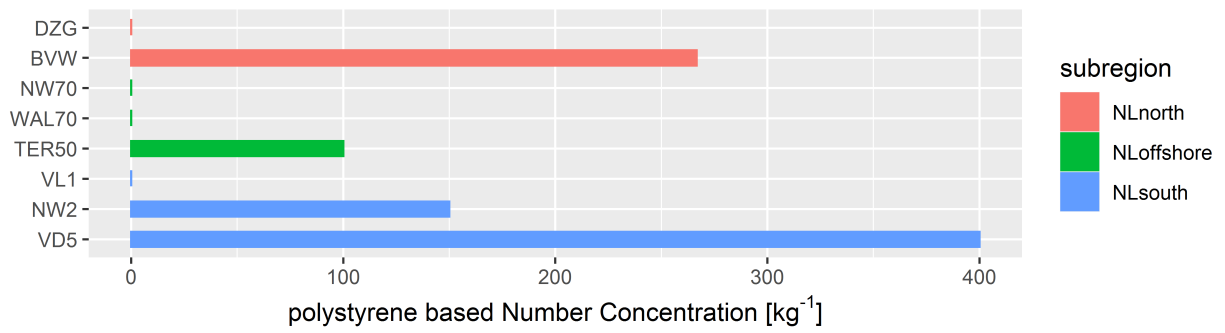
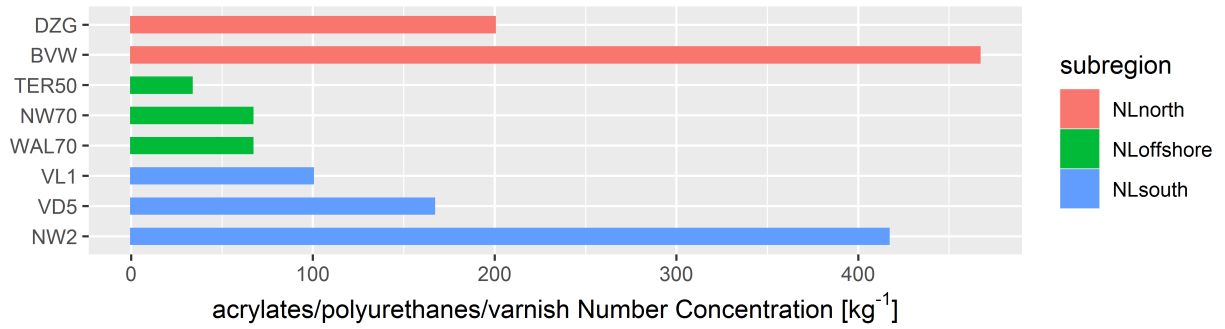
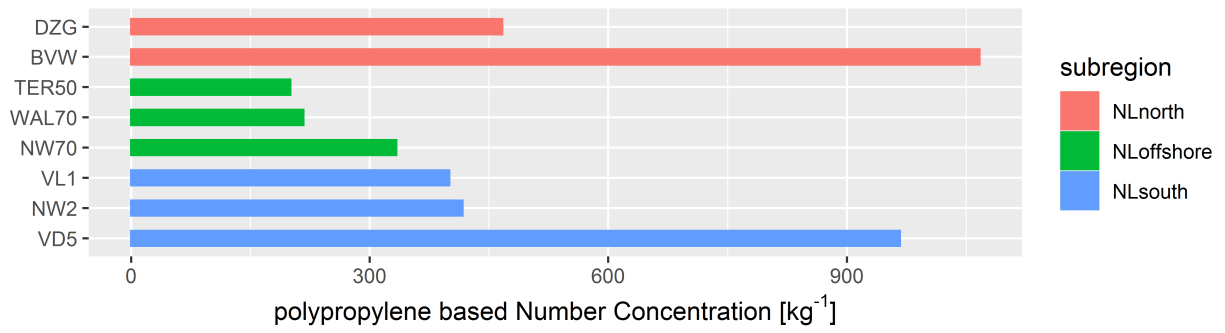
The siMPle software reported many MPs identified as rubber type 3 (EPDM, ethylene propylene diene monomer rubber). This is in line with the literature (Lorenz et al., 2019; Mintenig et al., 2020; Bauerlein et al., 2023). However, we suspect that a part of these identifications can be Styrene Butadiene Rubber (SBR) which is the main rubber type in Dutch rivers (MinVenW, 2023). In view of the density range of these SBR particles (1.13-2.2 g/cm^3 ; Baensch et al., 2020), and demonstrated by Valling et al. (2023), density extraction can extract this rubber type. The use of silicon filters, which allow to scan the IR fingerprint region completely, will provide evidence if SBR rubber can be identified using the present method.

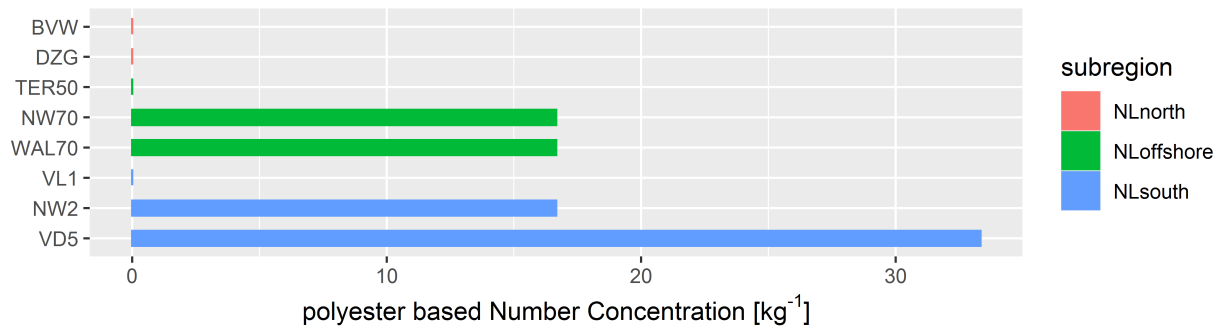
In addition, pyrolysis-GC-MS appears to be used regularly to analyse tyre wear particles (TWP), especially of large rubber particles which may not be IR transparent or reflectant due to the black carbon content (Galgani et al., 2023; Alling et al., 2023). These large rubber particles dominate the total rubber mass (Alling et al., 2023). Small tyre wear particles may be below the detection limit of pyrolysis-GC-MS (Valling et al., 2023) and may still be analyzed using μFTIR (see above).

Plots for each polymer type are shown in Figure 7.

Figure 7: Indicative polymer Number Concentrations (kg^{-1} sediment dry weight) for the seven most occurring polymers in Dutch marine sediments; based on 24 samples from 2023.







We propose that these indicative PNC plots are informative to investigate the spatial distribution of specific polymers. Location differences per polymer are often observed. However, three years of data with a finalised validated monitoring method are needed to draw reliable conclusions about polymer particle concentrations and spatial distribution in Dutch marine sediments. In that situation, the use t-tests will be possible to determine if the differences of PNCs at different locations are significant.

4. Conclusions

4.1 The microplastic monitoring method in this report is an advanced development stage and is approaching to meet the specifications from the TGML guidelines. However, a few method aspects can still be improved, as described below.

The dual extraction/dual Anodisc method is very useful to optimize the sample amount and for routine QC of the recovery.

The siMPle software, with standardised weight settings (Primpke et al., 2020) and using a generic threshold value of 0.5; complemented with the custom-made siMPleR script which selects MPs ≥ 50 μm and with HQI ≥ 0.6 ; appears to be effective and efficient in the detection and reporting of MPs. The Pearson weight settings for the original spectra, and for the first and second derivative spectra, appear to have a strong effect on the number of MPs found and must be standardised to 0 for the original Pearson value, 1 for the Pearson 1st derivative and 1 for the Pearson 2nd derivative (Primpke et al., 2020).

4.2 The digestion of samples can be improved for some locations, for OC or shell fragments, respectively.

4.3 The recovery of fluorescent reference polyethylene particles from muddy and sandy samples, using fluorescence microscopy, is 94%. The post-flushing with ethanol appeared to be effective in increasing the recoveries and was made a part of the SOP.

However, we have additionally estimated the recoveries of sample MPs. We estimated this by using a simple theoretical power model and the results of the two extractions per sample. We conclude from these estimations that the recovery of the first extraction is not always as high as probably needed. This may be related to an insufficient digestion, or maybe due to insufficient prewetting time of the sediment sample before extraction.

4.4 We conclude from the microplastic binning power plot that a lower length limit (LLL) of 50 μm is valid to use for the reporting of microplastics using this method.

4.5 The field and laboratory conditions appeared to be relatively clean, leading to low blank concentrations and indicative LODs of specific polymers.

4.6 The data analysis using the siMPleR script provided a top list of nine polymer types commonly found in Dutch marine sediments. In the PNC plots, many differences between polymer concentrations at different locations are observed. However, these results are at present indicative, because three years of data with a finalized and validated method are needed to report baseline microplastic results with sufficient confidence.

With three years of data, t-tests will be possible to investigate if significant differences between MNC and PNC values of locations exist.

4.7 The spectral range which is possible to use with Anodiscs (1250-3600 cm^{-1}) disregards a part of the selective IR fingerprint range (700-1850 cm^{-1} ; Renner et al., 2019). As a result, several polymer identifications (rubber types, chlorinated polyethylene, polyester) are not yet considered to be sufficiently reliable.

5. Recommendations

5.1 It is planned to test the use of silicon filters instead of Anodisc filters, because silicon filters are transparent in the IR fingerprint region (700-1850 cm^{-1}) (personal communication Jakob Strand; Martinez et al., 2023). These filters are likely to give more reliable identifications for polymers, for example for several rubber types, chlorinated polyethylene and polyester. In addition, more MP identifications with a match index score ≥ 0.6 are expected, which would increase the number of MPs that can be reported.

It will be tested if only the fingerprint region (700-1850⁻¹) can be used, as recommended by Renner et al. (2019). This scanning of a smaller range may also improve the S/N-ratio and/or reduce the scan time.

Alternatively, maybe the middle IR region (2750-1850 cm^{-1}) could be skipped during μFTIR scanning or siMPle analysis, and the two informative IR-regions can be maintained.

5.2 It is recommended to develop and use a decision tree for the sample preparation in 2024 as follows:

- Adjust the dry sample weight to a minimally needed number of MPs (20) or to avoid overloading of the filter (e.g. max 100 MPs). For example, offshore locations need 20 g of sample; while for the highly polluted estuarine location Bocht van Watum 5 g is probably sufficient.
- Apply digestion of shell fragments if needed. This need can be judged visually or maybe a qualitative test (1 g + acetic acid 10%) can be used (Alling et al., 2023). The measurement of Calcium or Carbonate content is not expected to be a selective test.
- Apply, if needed for specific samples, the Fenton reagent for improved digestion. A need for this can be judged using images of the filters (see Annex 3).

5.3 In order to improve the recovery of the first extraction, it is recommended to test the prewetting of samples using 1 hour prewetting time for 1 out of 3 replicates for each sample. This may improve the recovery of the first extract (Filgueras et al., 2021).

5.4 We are hesitant to add a sieving step of 300 or 500 μm . Because although this may improve the identification of a few large particles in this range and may reduce the clouding of the 50-300 μm particles, it may be that damage and losses of MPs are induced due to the forcing of MPs through the 300 μm sieve. Therefore, we do not recommend to add this step, unless the advantage can clearly be demonstrated.

5.5 To investigate the need to add reference μFTIR spectra, measured on the same machine as the samples, to the siMPle library. This may improve the Max. HQI Scores and the resulting number of identifications with Max. Score ≥ 0.6 (personal communication Jakob Strand).

5.6 It is recommended to test in siMPle to only use the first spectrum derivative, because the second derivative performs poorly according to Renner et al. (2019, Figure 1). At present, a combination of the first and second spectrum derivative is used.

5.7 It is recommended to do some more manual validation on alternative polymer identifications for a specific MP particle with a lower HQI compared to the reported hit (Renner et al., 2019). This will provide additional validation on the selectivity of the MP identifications.

5.8 To test the morphology classification method of Kooi and Koelmans (2019) in 2024.

<https://pubs.acs.org/doi/10.1021/acs.estlett.9b00379>

5.9 To investigate if epoxy resins from wind farms can be detected using our method (RIVM report, in Dutch).

5.10 Investigate if there is an alternative for the polypropylene support ring of the filter, because this ring can introduce blank contamination with polypropylene particles.

5.11 It is recommended to organize a technical review of the next Cefas-RWS microplastic report by an external EU microplastic expert.

6. References

- Alling, V. et al., 2023. Monitoring of microplastics in the Norwegian environment (MIKRONOR). NIVA, Report no. 7922-2023.
- Bakir, A., 2022. Monitoring of microplastics in Dutch marine sediments: a pilot study. Report, CEFAS.
- Bakir, A., van Loon, W., 2023a. Standard Operating Procedure 3091: Sample preparation and analysis of microplastics in marine sediments.
- Bakir A., Doran D., Silburn B., Russell J., Archer-Rand S., Barry J., Maes T., Limpenny C., Mason C., Barber J., Nicolaus E.E. M., 2023b. A spatial and temporal assessment of microplastics in seafloor sediments: A case study for the UK. *Front. Mar. Sci.* 9, pp 20
- Bäuerlein P.S., Erich M.W., van Loon W.M.G.M., Mintening S. M., Koelmans A.A. (2023) A monitoring and data analysis method for MPs in marine sediments. *Marine Environmental Research*, 183, 105804.
- British Plastics Federation (2023a) Ethylene Vinyl Acetate EVA [Ethylene Vinyl Acetate EVA \(bpf.co.uk\)](https://bpf.co.uk)
- British Plastics Federation (2023b) Nylons (Polyamide) [Nylons \(Polyamide\) \(bpf.co.uk\)](https://bpf.co.uk)
- British Plastics Federation (2023c) Polyurethane cast elastomers (EP) [Polyurethane cast elastomers \(EP\) \(bpf.co.uk\)](https://bpf.co.uk)
- British Plastics Federation (2023d) Polycarbonate (PC) [Polycarbonate \(PC\) \(bpf.co.uk\)](https://bpf.co.uk)
- British Plastics Federation (2023e) Unsaturated Polyester UP [Unsaturated Polyester UP \(bpf.co.uk\)](https://bpf.co.uk)
- British Plastics Federation (2023f) Polyethylene (High Density) HDPE [Polyethylene \(High Density\) HDPE \(bpf.co.uk\)](https://bpf.co.uk)
- British Plastics Federation (2023g) Polyethylene (Low Density) LDPE, LLDPE [Polyethylene \(Low Density\) LDPE, LLDPE \(bpf.co.uk\)](https://bpf.co.uk)
- British Plastics Federation (2023h) Polypropylene (PP) [Polypropylene \(PP\) \(bpf.co.uk\)](https://bpf.co.uk)
- British Plastics Federation (2023i) Polyvinyl chloride (PVC) [Polyvinyl Chloride PVC \(bpf.co.uk\)](https://bpf.co.uk)
- Bronzo, L., Lusher, A. L., Schøyen, M., & Morigi, C. (2021). Accumulation and distribution of microplastics in coastal sediments from the inner Oslofjord, Norway. *Marine pollution bulletin*, 173, 113076.
- Castro-Aguirre E., Iñiguez-Franco F., Samsudin H., Fang X., Auras R. (2016) Poly(lactic acid)—Mass production, processing, industrial applications, and end of life. *Advanced Drug Delivery Reviews*, 107, 333-366.
- Cowger, W., Steinmetz, Z., Gray, A., Munno, K., Lynch, J., Hapich, H., ... & Herodotou, O. (2021). Microplastic spectral classification needs an open source community: open specy to the rescue!. *Analytical Chemistry*, 93(21), 7543-7548.
- Deopura B.L., Alagirusamy R., Joshi M., Gupta B., 2008. Polyesters and polyamides. *Technology and Engineering*, pp 1–632.
- Erni-Cassola, G., Zadjelovic, V., Gibson, M. I., & Christie-Oleza, J. A. (2019). Distribution of plastic polymer types in the marine environment; A meta-analysis. *Journal of hazardous materials*, 369, 691-698.
- Fellows, P.J., 2022 in *Food Processing Technology* (Fifth Edition).
- Filgueiras, A. V., Gago, J., García, I., León, V. M., & Viñas, L. (2021). Plackett Burman design for microplastics quantification in marine sediments. *Marine Pollution Bulletin*, 162, 111841.
- Gaylarde C.C., Neto J.A.B., da Fonseca E.M., 2021. Paint fragments as polluting MPs: a brief review. *Marine Pollution Bulletin* 162, 111847.
- F. Galgani et al., 2023. Guidance on the monitoring of marine litter in European seas. JRC report. <https://mcc.jrc.ec.europa.eu/main/dev.py?N=41&O=466>
- Hof, M., Bakker, J., & Spanbroek, N. (2023). Beoordeling mogelijke risico's van chemische stoffen en plastic deeltjes van windturbines op zee. *RIVM rapport 2022-0241*.

Hurley, R. R., Lusher, A. L., Olsen, M., & Nizzetto, L. (2018). Validation of a method for extracting microplastics from complex, organic-rich, environmental matrices. *Environmental science & technology*, 52(13), 7409-7417.

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Kooi, M. and Koelmans, A.A. (2019) simplifying microplastic continuous probability distributions size shape density. *Environ. Sci. Technol. Lett.*, 6, 9, 551–557

Kroon, F., Motti, C., Talbot, S., Sobral, P., & Puotinen, M. (2018). A workflow for improving estimates of microplastic contamination in marine waters: A case study from North-Western Australia. *Environmental Pollution*, 238, 26-38.

Kukkola, A. T., Senior, G., Maes, T., Silburn, B., Bakir, A., Kröger, S., & Mayes, A. G. (2022). A large-scale study of microplastic abundance in sediment cores from the UK continental shelf and slope. *Marine Pollution Bulletin*, 178, 113554.

Lacroix, C., André, S., & van Loon, W. (2023). Abundance, composition and trends of beach litter. *The 2023 Quality Status Report for the North-East Atlantic*.

Leistenschneider, C., Burkhardt-Holm, P., Mani, T., Primpke, S., Taubner, H., & Gerdts, G. (2021). Microplastics in the Weddell Sea (Antarctica): a forensic approach for discrimination between environmental and vessel-induced microplastics. *Environmental Science & Technology*, 55(23), 15900-15911.

LeNoir, Richard T. in *Global Legislation for Food Contact Materials (Second Edition)*, 2022

Löder, M. G. J., Kuczera, M., Mintenig, S., Lorenz, C., & Gerdts, G. (2015). Focal plane array detector-based micro-Fourier-transform infrared imaging for the analysis of microplastics in environmental samples. *Environmental Chemistry*, 12(5), 563-581.

Lorenz, C., Roscher, L., Meyer, M. S., Hildebrandt, L., Prume, J., Löder, M. G., ... & Gerdts, G. (2019). Spatial distribution of microplastics in sediments and surface waters of the southern North Sea. *Environmental Pollution*, 252, 1719-1729.

Martínez, K. P., da Silva, V. H., Andersen, T. J., Posth, N. R., & Strand, J. (2023). Improved separation and quantification method for microplastic analysis in sediment: A fine-grained matrix from Arctic Greenland. *Marine Pollution Bulletin*, 196, 115574.

Mattsson, K., de Lima, J. A., Wilkinson, T., Järskog, I., Ekstrand, E., Sköld, Y. A., ... & Hassellöv, M. (2023). Tyre and road wear particles from source to sea. *Microplastics and Nanoplastics*, 3(1), 14.

McCoy K.A., Hodgson D.J., Clark P.F., Morrirt D., 2020. The effects of wet wipe pollution on the Asian clam, *Corbicula fluminea* (Mollusca: Bivalvia) in the River Thames, London. *Environmental Pollution* 264, 114577

Ministerie van Infrastructuur en Waterstaat, 2023. Op weg naar microplastics monitoring in rivieren. Deel 3: pilotmetingen in oppervlaktewater, waterbodem en rivieroevers. Rapport.

Mintenig, S., 2021. A novel protocol for the analysis of microplastics in marine sediments: Method review. Report M.363, Wageningen University and Research.

More, V. K., Krishnaswamy, S., Ponnani, K., & Singh, M. K. (2022). Co-chlorination of low-density polyethylene in paraffin: An intensified green process alternative to conventional solvent-based chlorination. *Green Processing and Synthesis*, 11(1), 842-856.

Moses, S. R., Roscher, L., Primpke, S., Hufnagl, B., Löder, M. G., Gerdts, G., & Laforsch, C. (2023). Comparison of two rapid automated analysis tools for large FTIR microplastic datasets. *Analytical and Bioanalytical Chemistry*, 1-13.

Munno, K., Helm, P. A., Jackson, D. A., Rochman, C., & Sims, A. (2018). Impacts of temperature and selected chemical digestion methods on microplastic particles. *Environmental toxicology and chemistry*, 37(1), 91-98.

NEN 7777, 2012. Environment and food - Performance characteristics of measurement methods. Dutch standard, in English.

NEN 7779, 2018. Environment, food and feed - Measurement uncertainty. Dutch standard, in English.

Ospar, 2017_assessment document of land-based inputs of microplastics in the marine environment OSPAR MPEG, 2023. Guidelines for the monitoring of microlitter (including microplastics) in seafloor sediments for the OSPAR Maritime Area.

Pfeiffer, F., & Fischer, E. K. (2020). Various digestion protocols within microplastic sample processing—evaluating the resistance of different synthetic polymers and the efficiency of biogenic organic matter destruction. *Frontiers in Environmental Science*, 8, 572424.

PlasticsEurope, 2022. Plastics the facts 2022 (online). [Plastics - the Facts 2022 • Plastics Europe](#)

Primpke, S., Lorenz, C., Rascher-Friesenhausen, R., & Gerdts, G. (2017). An automated approach for microplastics analysis using focal plane array (FPA) FTIR microscopy and image analysis. *Analytical Methods*, 9(9), 1499-1511.

Primpke S., Wirth M., Lorenz C. Gerdts G. (2018) Reference database design for the automated analysis of microplastic samples based on Fourier transform infrared (FTIR) spectroscopy. *Anal Bioanal Chem* 410, 5131–5141.

Primpke S., Cross R.K., Mintenig S.M., Simon M., Vianello A., Gerdts G., Vollertsen J. (2020) Toward the Systematic Identification of Microplastics in the Environment: Evaluation of a New Independent Software Tool (siMPle) for Spectroscopic Analysis Applied Spectroscopy, 74(9):1127-1138.

Primpke S, Christiansen SH, Cowger W, De Frond H, Deshpande A, Fischer M, Holland EB, Meyns M, O'Donnell BA, Ossmann BE, Pittroff M, Sarau G, Scholz-Böttcher BM, Wiggin KJ. Critical Assessment of Analytical Methods for the Harmonized and Cost-Efficient Analysis of Microplastics. *Appl Spectrosc*. 2020 Sep;74(9):1012-1047.

Puls J., Wilson S.A., Hölter D., 2011. Degradation of cellulose acetate-based materials: a review. *Journal of Polymers and the Environment* 19, 152 – 165

de Ruijter, V. N., Redondo-Hasselerharm, P. E., Gouin, T., & Koelmans, A. A. (2020). Quality criteria for microplastic effect studies in the context of risk assessment: a critical review. *Environmental Science & Technology*, 54(19), 11692-11705.

RWS-CIV, 2022. rsv_913-00-w100v2_Bemonstering_Sediment_Microplastics. <https://open.rijkswaterstaat.nl/overige-publicaties/2003/rijkswaterstaat-standaard-inwinning/staat-standaard-voor-de-inwinning-verwerking-en-uitgifte-van-chemische-gegevens>

Bakir, A. and Van Loon, W. Standard Operating Procedure 3091: Sample preparation and analysis of microplastics in marine sediments.

Tagg, A. S., Sapp, M., Harrison, J. P., & Ojeda, J. J. (2015). Identification and quantification of microplastics in wastewater using focal plane array-based reflectance micro-FT-IR imaging. *Analytical chemistry*, 87(12), 6032-6040.

Taib, NA.A.B., Rahman, M.R., Huda, D. et al. (2023) A review on poly lactic acid (PLA) as a biodegradable polymer. *Polym. Bull.* 80, 1179–1213.

Turner A., Ostle, C., Wootton, M. (2022) Occurrence and chemical characteristics of microplastic paint flakes in the North Atlantic Ocean. *Science of the Total Environment*, 806(1), 150375.

Walvoort, D. and van Loon, W., 2024. User manual for siMPler: data analysis of siMPle results files.

Webler T., Jakubowski K., 2022. Attitudes, beliefs, and behaviours about cigarette-butt littering among college-aged adults in the United States. *International Journal of Environmental Research and Public Health* 19, 8085

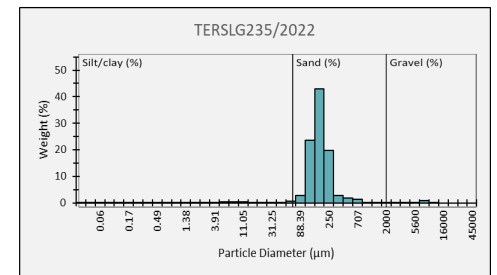
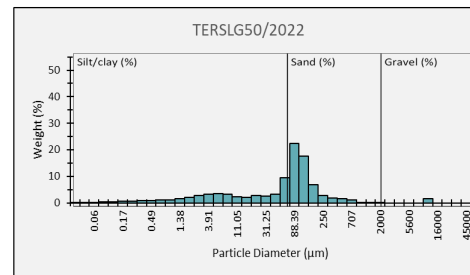
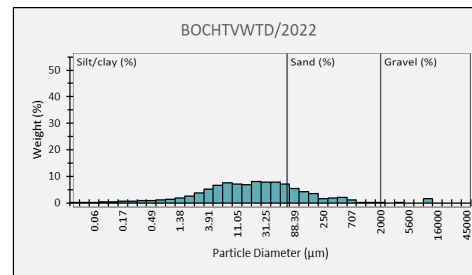
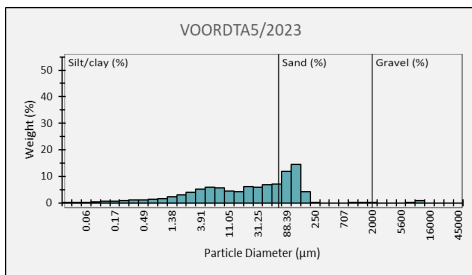
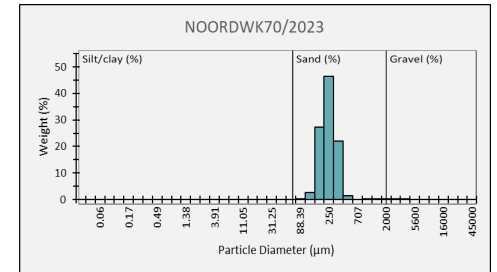
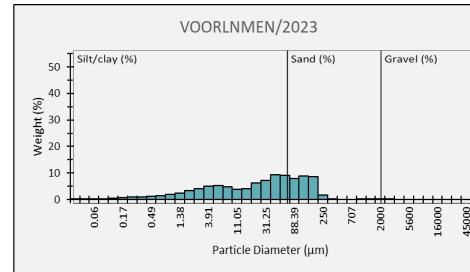
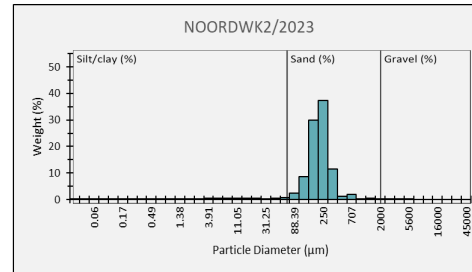
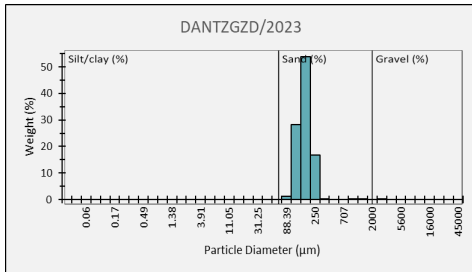
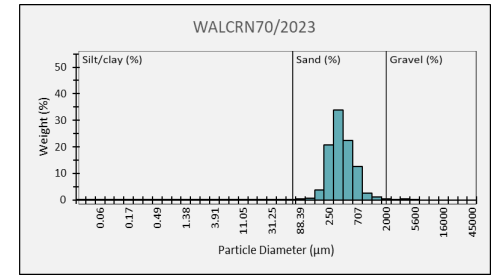
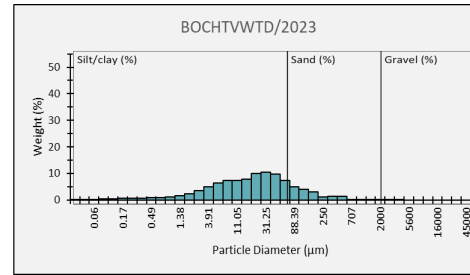
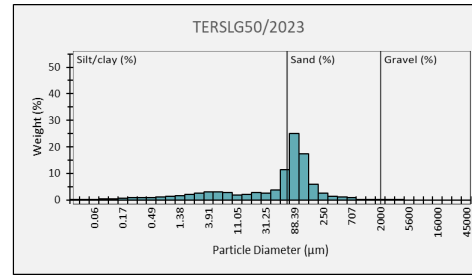
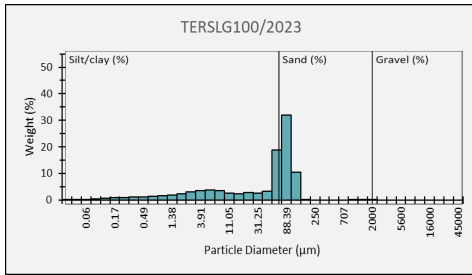
Wiesner Y., Hoffmann T., Range D., Altmann, K. (2023) Microplastics in sediments of the river Rhine—A workflow for preparation and analysis of sediment samples from aquatic river systems for monitoring purposes. *Applied Research* e202200125.

Xu, H., & Arakawa, H. (2022). Determining the appropriate number of particles on a filter to allow small microplastics to be analyzed by microscopy. *MethodsX*, 9, 101646.

Annex 1: Location and sample characteristics

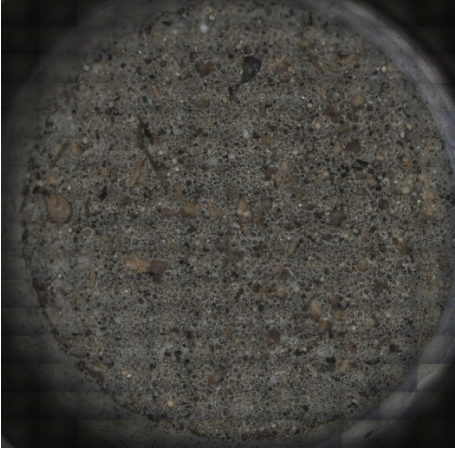
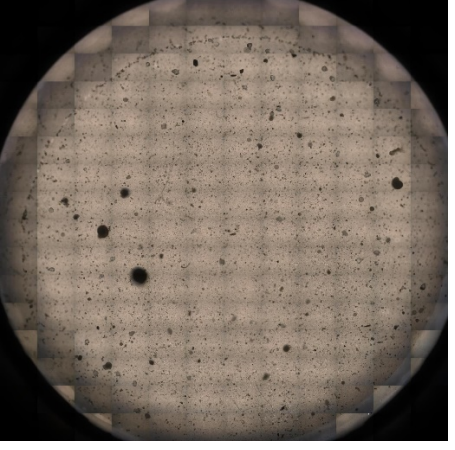
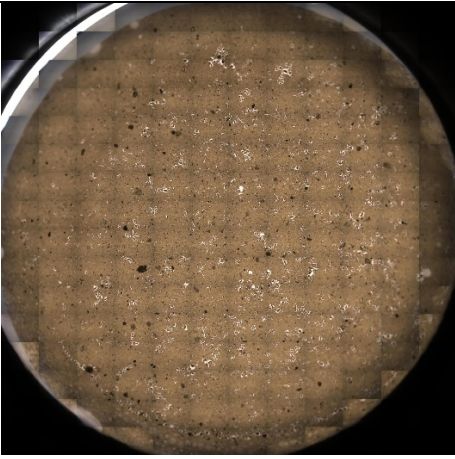
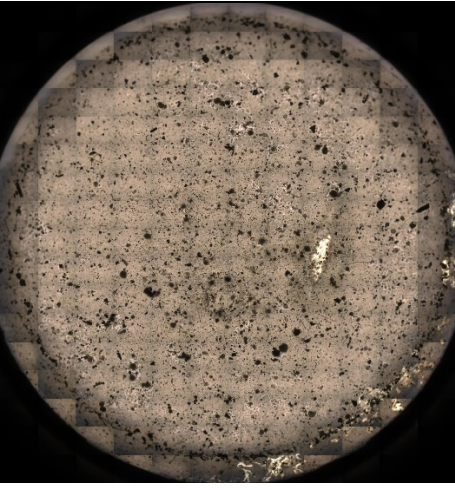
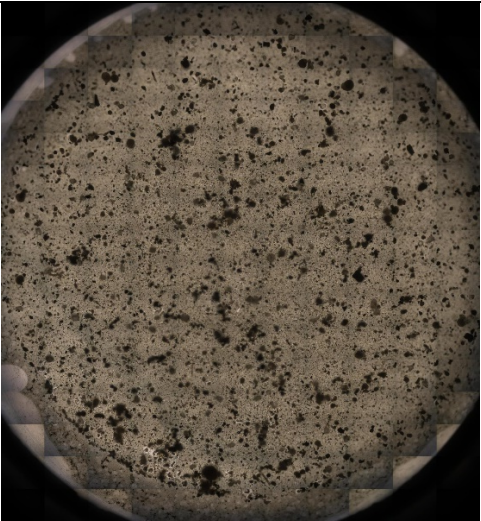
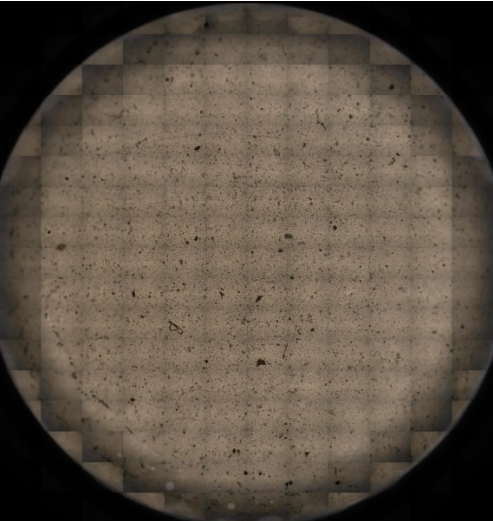
Location Code	Location name	Subregion	X coordinate (UTM31)	Y coordinate (UTM31)	Mud content [%]	Organic carbon content [%]
BVW	Bocht van Watum	NL North	757640	5924876	76.50	2.53
DZG	Dantzigat	NL North	680710	5920129	0.00	0.03
DBW	Doovebalg West	NL North	636211	5880087	na	Na
NW2	Noordwijk 2	NL South	595840	5790798	5.36	0.10
VD5	Voordelta 5	NL South	563119	5752696	61.0	0.91
VL1	Voorland 1	NL South	542327	5692735	63.70	0.91
TER50	Terschelling 50	NL offshore	616409	5959050	34.2	0.45
NW70	Noordwijk 70	NL offshore	535924	5824420	0.00	0.04
WAL70	Walcheren 70	NL offshore	477859	5756207	1.13	0.03

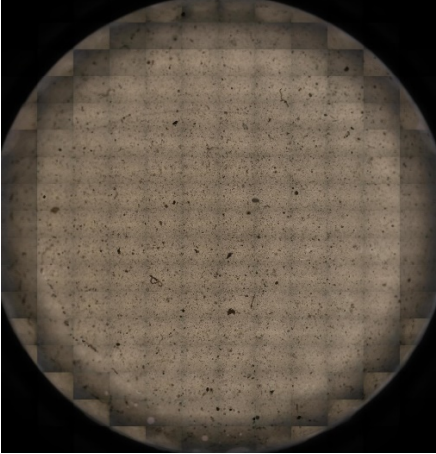
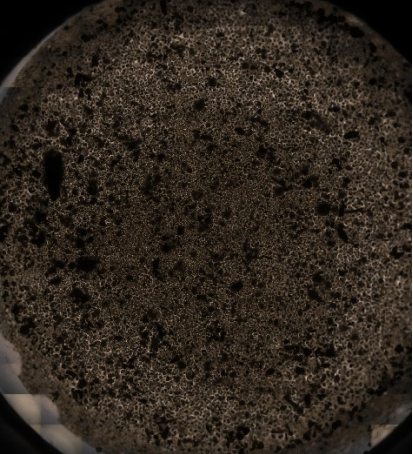
Annex 2: Particle Size Distributions of the Dutch composite location samples



Annex 3: Images of Anodiscs of samples from each monitoring location

Used for QC of the sample preparation.

Bocht van Watum	Dantziggat
	
Noordwijk 2	Voordelta 5
	
Voorland 1	Walcheren 70
	

Noordwijk 70	Terschelling 50
	

Discussion:

- The cleanup of OC in the Anodisc filtrate of the sample from Bocht_van_Watum is insufficient. A sample weight of 5 g will be used in 2024.
- The filtrate of the sample from Dantziggat shows a few large “clouds” of large particles. A better digestion, or maybe sieving with 300 or 500 μm , may solve this.
- The filtrate of the Noordwijk 2 sample seems to show many smaller shell fragments. This will be confirmed. Additional pretreatment with acetic acid 10% will be considered.
- The cleanup of OC in the filtrate of the Terschelling 50 sample is insufficient. Additional OC removal will be tested.
- For the samples from Voordelta 5 and Voorland 1, a better digestion or sieving with 300 or 500 μm may cleanup the Anodisc filtrate.

Annex 4: Polymer Number Concentrations (PNC) results reported using EU types

Sorted first by polymer, then increasing concentration. Results are rounded to three digits.

subregion	location	period	polymer_type	pnc_location [g-1]	SE [g-1]	N samples	N MPs
NLoffshore	TER50	2023-2023	acrylates/polyurethanes/varnish	33	33	3	1
NLoffshore	NW70	2023-2023	acrylates/polyurethanes/varnish	67	44	3	4
NLoffshore	WAL70	2023-2023	acrylates/polyurethanes/varnish	67	44	3	4
NLsouth	VL1	2023-2023	acrylates/polyurethanes/varnish	100	58	3	3
NLsouth	VD5	2023-2023	acrylates/polyurethanes/varnish	167	67	3	5
NLnorth	DZG	2023-2023	acrylates/polyurethanes/varnish	200	58	3	6
NLsouth	NW2	2023-2023	acrylates/polyurethanes/varnish	417	130	3	25
NLnorth	BVW	2023-2023	acrylates/polyurethanes/varnish	467	233	3	14
NLsouth	NW2	2023-2023	cellulose based	17	17	3	1
NLoffshore	TER50	2023-2023	cellulose based	33	33	3	1
NLsouth	VL1	2023-2023	cellulose based	33	33	3	1
NLnorth	BVW	2023-2023	cellulose based	267	133	3	8
NLoffshore	WAL70	2023-2023	polyamide based	17	17	3	1
NLnorth	DZG	2023-2023	polyamide based	33	33	3	1
NLoffshore	NW70	2023-2023	polyamide based	33	17	3	2
NLoffshore	TER50	2023-2023	polyamide based	33	33	3	1
NLnorth	BVW	2023-2023	polyamide based	633	633	3	19
NLsouth	NW2	2023-2023	polycarbonate based	17	17	3	1
NLnorth	DZG	2023-2023	polycarbonate based	33	33	3	1
NLoffshore	NW70	2023-2023	polyester based	17	17	3	1
NLoffshore	WAL70	2023-2023	polyester based	17	17	3	1
NLsouth	NW2	2023-2023	polyester based	17	17	3	1
NLsouth	VD5	2023-2023	polyester based	33	33	3	1
NLnorth	DZG	2023-2023	polyetheretherketone	33	33	3	1
NLoffshore	WAL70	2023-2023	polyethylene based	100	76	3	6
NLoffshore	NW70	2023-2023	polyethylene based	117	67	3	7

NLnorth	DZG	2023-2023	polyethylene based	700	153	3	21
NLsouth	VD5	2023-2023	polyethylene based	867	437	3	26
NLsouth	VL1	2023-2023	polyethylene based	1200	306	3	36
NLsouth	NW2	2023-2023	polyethylene based	1250	400	3	75
NLoffshore	TER50	2023-2023	polyethylene based	1467	120	3	44
NLnorth	BVW	2023-2023	polyethylene based	10700	3190	3	321
NLsouth	NW2	2023-2023	polylactic acid	17	17	3	1
NLoffshore	TER50	2023-2023	polypropylene based	200	200	3	6
NLoffshore	WAL70	2023-2023	polypropylene based	217	148	3	13
NLoffshore	NW70	2023-2023	polypropylene based	333	101	3	20
NLsouth	VL1	2023-2023	polypropylene based	400	100	3	12
NLsouth	NW2	2023-2023	polypropylene based	417	88	3	25
NLnorth	DZG	2023-2023	polypropylene based	467	133	3	14
NLsouth	VD5	2023-2023	polypropylene based	967	291	3	29
NLnorth	BVW	2023-2023	polypropylene based	1.070	296	3	32
NLoffshore	TER50	2023-2023	polystyrene based	100	58	3	3
NLsouth	NW2	2023-2023	polystyrene based	150	29	3	9
NLnorth	BVW	2023-2023	polystyrene based	267	88	3	8
NLsouth	VD5	2023-2023	polystyrene based	400	153	3	12
NLsouth	NW2	2023-2023	polyvinylchloride	50	0	3	3
NLsouth	VD5	2023-2023	polyvinylchloride	100	58	3	3
NLsouth	VL1	2023-2023	polyvinylchloride	100	58	3	3
NLnorth	BVW	2023-2023	polyvinylchloride	200	153	3	6
NLoffshore	NW70	2023-2023	rubber	200	76	3	12
NLoffshore	WAL70	2023-2023	rubber	200	76	3	12
NLsouth	VD5	2023-2023	rubber	467	371	3	14
NLoffshore	TER50	2023-2023	rubber	967	517	3	29
NLsouth	NW2	2023-2023	rubber	1.320	335	3	79
NLnorth	DZG	2023-2023	rubber	1.470	291	3	44
NLnorth	BVW	2023-2023	rubber	2900	1480	3	87
NLsouth	VL1	2023-2023	Rubber	2970	318	3	89

Annex 5: Common applications of polymers

Polymer	Possible applications/sources and references
Acrylates/PU/ varnish	<ul style="list-style-type: none"> • Ship paints (Gaylarde et al., 2021) • PU cast elastomers: rinting rollers, solid tyres, wheels, shoe heels, car bumpers (BPF, 2023c) • Packaging, building & construction, cars, electronics, leisure & sports (PlasticsEurope, 2022).
Cellulose modified	<ul style="list-style-type: none"> • Cigarette butts (cellulose acetate: Puls et al., 2011; Webler and Jakubowski, 2022), wet wipes (Pampers), sanitary pads (Always)
Ethylene-vinyl-acetate	<ul style="list-style-type: none"> • Teats, handle grips, flexible tubing, record turntable mats, beer tubing, vacuum, cleaner hosing (BPF, 2023a) • Food contact applications (LeNoir, 2022) <ul style="list-style-type: none"> ◦ EVA that contains less than 5% vinyl acetate is used for deep-freeze applications (Fellows, 2022) ◦ Films with 6%–10% vinyl acetate are used in bag-in-box applications and milk pouches (Fellows, 2022) ◦ and above 10% vinyl acetate, the material is used as a hot-melt adhesive (Fellows, 2022).
Polyamide	<ul style="list-style-type: none"> • Textiles/apparel (Deopura et al.,2008). • Nylons: textiles, fishing line and carpets (BPF, 2023b). • Nylon films used for food packaging (BPF, 2023b). • Packaging, cars, electronics, leisure & sports (PlasticsEurope, 2022).
Polycarbonate	<ul style="list-style-type: none"> • Electrical and electronics, automotive, general industries, packaging (BPF, 2023d).
Polyester	<ul style="list-style-type: none"> • Wet wipes (McCoy et al., 2020; Pampers), • Sanitary pads (Always), • Textiles/apparel (Deopura et al.,2008) • Packaging, leisure & sports (PlasticsEurope, 2022)
Polyethylene	<ul style="list-style-type: none"> • HDPE: Chemical drums, jerricans, carboys, toys, picnic ware, household and kitchenware, cable insulation, carrier bags, food wrapping material (BPF, 2023f). • Packaging, building & construction, cars, electronics, leisure & sports (PlasticsEurope, 2022). • LDPE, LLDPE: Squeeze bottles, toys, carrier bags, high frequency insulation, chemical tank linings, heavy duty sacks, general packaging, gas and water pipes (BPF, 2023g). • Packaging, building & construction, cars, electronics, agriculture & gardening, leisure & sports (PlasticsEurope, 2022).
Polyethylene-chlorinated	<ul style="list-style-type: none"> • Cable sheathing and hose linings in the automotive industry. Used in production of seals, gaskets, and other flexible parts
Polylactose	<ul style="list-style-type: none"> • Agriculture, automotive and packaging (Taib et al., 2023) • Medical, textile, plasticulture and packaging industry (Castro-Aguirre et al., 2016)
Polypropylene	<ul style="list-style-type: none"> • Sanitary pads (Always; McCoy et al., 2020) • Buckets, bowls, crates, toys, medical components, washing machine drums, battery cases, bottle caps. Elastomer modified for bumpers, etc. Talc filled for additional stiffness at elevated temperatures - jug kettles, etc. OPP films for packaging (e.g. crisps, biscuits, etc.). Fibres for carpets, sports clothin (BPF, 2023h). • Packaging, building & construction, cars, electronics, agriculture & gardening, leisure & sports (PlasticsEurope, 2022)
Polystyrene	<ul style="list-style-type: none"> • Packaging, building & construction, electronics (PlasticsEurope, 2022).
Polyvinylchloride	<ul style="list-style-type: none"> • Packaging, building & construction, cars, electronics, agriculture & gardening, leisure & sports (PlasticsEurope, 2022). • Construction (i.e. doors, windows, pipes, cables, roofing flooring etc.), healthcare (i.e., blood bags, overshoes etc.), electronics, cars, sports (i.e., sportswear, stadiums etc.) (BPF, 2023i).
Rubber	Rubbers, automotive Styrene butadiene rubber (SBR), tyre wear Other rubbers Includes ethylene propylene diene monomer rubber (EPDM), silicone, nitrile rubbers and natural rubbers

Annex 6: Data distributions of replicate data

The relation between the mean and median results of the location replicates ($N = 8$) is presented in the Figure below. A linear relation is observed, with a slope of 1.06 ($R^2 = 0.98$). This slope of almost unity suggests that the replicate data are fairly normally distributed at the location level.

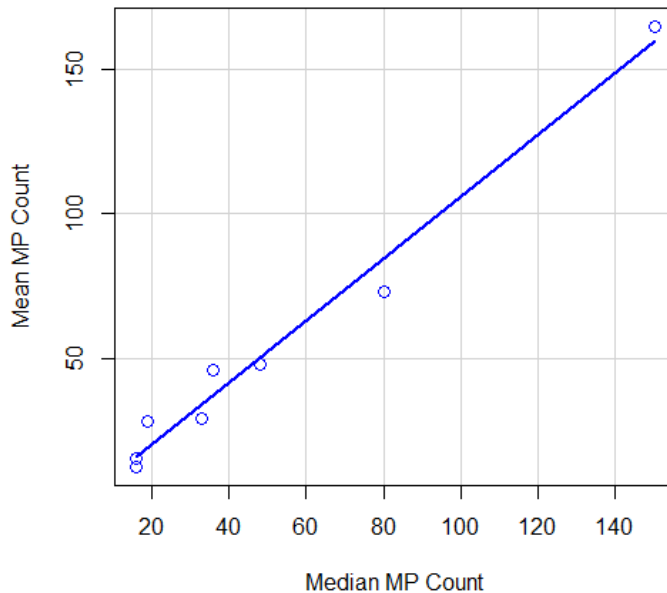


Figure: relation between the sample median and mean values ($N=8$).

The Total count distribution for all replicates of all locations combined are presented in the Figure below and clearly show a skewed distribution. The p-value of the Shapiro-Wilk test is very significant. This result shows that when location data are to be integrated to the subregional level, a median-based aggregation method should be used. This will avoid that the more extreme values would dominate the subregional averages. Therefore, for subregional aggregation median values of location means can best be used.

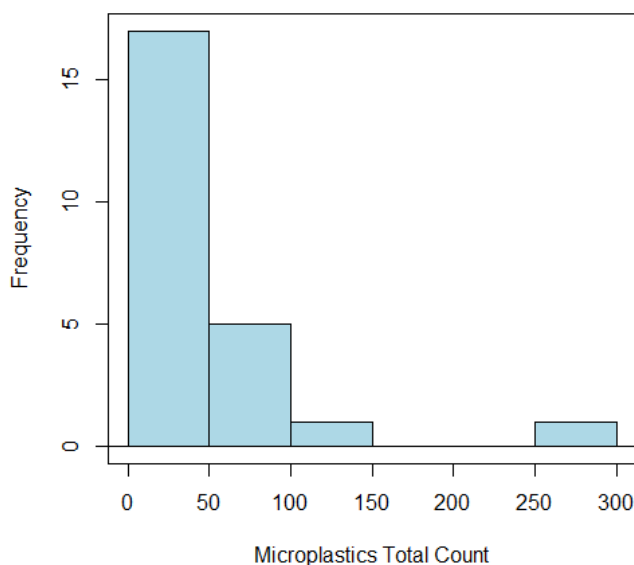


Figure: data distribution of the Total counts of all location replicates ($N=24$) combined.