



Analysis of microplastic particles in Danish drinking water

Strand, Jakob; Feld, Louise; Murphy, Fionn; Mackevica, Aiga; Hartmann, Nanna B.

Publication date:
2018

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):
Strand, J., Feld, L., Murphy, F., Mackevica, A., & Hartmann, N. B. (2018). *Analysis of microplastic particles in Danish drinking water*. Aarhus University. Scientific Report from DCE – Danish Centre for Environment and Energy No. 291

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.



ANALYSIS OF MICROPLASTIC PARTICLES IN DANISH DRINKING WATER

Scientific Report from DCE - Danish Centre for Environment and Energy

No. 291

2018



AARHUS
UNIVERSITY

DCE - DANISH CENTRE FOR ENVIRONMENT AND ENERGY

[Blank page]

ANALYSIS OF MICROPLASTIC PARTICLES IN DANISH DRINKING WATER

Scientific Report from DCE – Danish Centre for Environment and Energy

No. 291

2018

Jakob Strand¹
Louise Feld¹
Fionn Murphy¹
Aiga Mackevica²
Nanna B. Hartmann²

¹ Aarhus University, Department of Bioscience

² Technical University of Denmark, Department of Environmental Engineering



AARHUS
UNIVERSITY

DCE – DANISH CENTRE FOR ENVIRONMENT AND ENERGY

Data sheet

Series title and no.: Scientific Report from DCE – Danish Centre for Environment and Energy No. 291

Title: Analysis of microplastic particles in Danish drinking water

Authors: Jakob Strand¹, Louise Feld¹, Fionn Murphy¹, Aiga Mackevica² & Nanna B. Hartmann²

Institutions: ¹Aarhus University, Department of Bioscience
²Technical University of Denmark, Department of Environmental Engineering

Publisher: Aarhus University, DCE – Danish Centre for Environment and Energy ©
URL: <http://dce.au.dk/en>

Year of publication: September 2018
Editing completed: September 2018

Referee: Mikkel Tamstorf
Quality assurance, DCE: Susanne Boutrup

Financial support: The Danish Environmental Protection Agency, Ministry of Environment and Food of Denmark

Please cite as: Strand, J., Feld, L., Murphy, F., Mackevica, A. & Hartmann, N.B. 2018. Analysis of microplastic particles in Danish drinking water. Aarhus University, DCE – Danish Centre for Environment and Energy, 34 pp. Scientific Report No. 291.
<http://dce2.au.dk/pub/SR291.pdf>

Reproduction permitted provided the source is explicitly acknowledged

Abstract: Contents and composition of microplastic (MP) particles have been analysed in 50 L samples of tap water collected from 17 households and working places across Denmark in 2018. In 16 out of 17 samples the concentration of MP-like particles > 100 µm were below the LoD, which were determined as 0.58 MP-like particles per L.

Keywords: Microplastic, drinking water, Denmark

Layout: Graphic Group, AU Silkeborg
Front page photo: Louise Feld

ISBN: 978-87-7156-358-0
ISSN (electronic): 2245-0203

Number of pages: 34

Internet version: The report is available in electronic format (pdf) at
<http://dce2.au.dk/pub/SR291.pdf>

Contents

Summary	5
Sammenfatning	6
1. Introduction	7
2. Sampling and methodology	9
2.1 Sampling sites	9
2.2 Description of sampling method	9
2.3 Cleaning and preparation of sampling equipment	10
2.4 Pre-treatment and extraction of filter samples for analysis	11
2.5 Visual quantification of MP-like particles by microscopy	12
2.6 Determination of limit of detection for MP-like particles	14
2.7 Chemical identification of MP-like particles by μ FT-IR analysis	14
3. Results and discussion	16
3.1 Detection of MP-particles in Anodisc samples	19
4. Conclusions	22
5. References	23
Appendix 1. Description of sampling sites	25
Appendix 2. Identification of cellulose-like particles by μFT-IR	27
Appendix 3. Identification of protein-like particles by μFT-IR	28
Appendix 4. Detection of MP-particles using Anodiscs	29
Appendix 5. Identification of MP-particles using Anodiscs	30

[Blank page]

Summary

Microplastic (MP) is a pervasive pollutant found in sediment, water and biota throughout the world. However, the extent to which humans are exposed to this contaminant is not well researched. Recently studies have indicated that MP is also present in tap water, bottled water and food destined for human consumption. Here, we investigate the presence of MP in drinking water from 17 sites around Denmark. 50 L of drinking water were sampled per site directly from taps through 10 μm stainless steel filters in a closed steel filter system to prevent contamination. The filters were examined visually by stereomicroscopy and the number and characteristics of all MP-like particles with sizes $> 100 \mu\text{m}$ were registered – i.e. particles which were either potential MP or particles of uncertain origin that might be MP. 124 (44 %) MP-like particles representatively selected among the particles from the filters were then manually transferred to ZnSe discs and the chemical composition identified using advanced $\mu\text{FT-IR}$ utilizing focal plane array (128 x 128) mapping with a pixel resolution of 5.5 μm .

Additionally, three tap water samples were passed directly through Anodisc filters (\varnothing 2.5 cm, pore size 0.2 μm) for additional chemical $\mu\text{FT-IR}$ analysis to detect potential MP $< 100 \mu\text{m}$. This analysis mapped particles in 10 % of the Anodisc filter area (corresponding to 10 % of all particles per sample), and the direct transfer process meant that any potential bias from visual picking of particles could be excluded.

The results from the visual assessment of filters showed that in average 15.6 MP-like particles $> 100 \mu\text{m}$ were observed per 50 L water sample. In 16 of the 17 samples, the number were below the limit of detection (29 per 50 L) and in the sample with the highest concentration, 30 MP-like particles per 50 L were observed. Identification by ZnSe discs showed that 3 % of the MP-like particles were verified as MP, whereas the majority consisted of cellulose-like material (76 %), and the rest were poor spectra (10 %), unknown (7 %) or protein-like (4 %). The types of MP particles detected in the tap water samples were polyethylene terephthalate, polypropylene and polystyrene. Using the Anodisc method, polyethylene terephthalate and polypropylene were also found, as well as acrylonitrile butadiene styrene and polyurethane.

However, due to the concentrations of MP below the limit of detection, no conclusions regarding the origin of MP from either tap water or sample contamination can be made. In contrast to previous American and Danish studies of MP in drinking water, which were highly publicised by the media, this study shows no significant concentrations of MP in Danish tap water. This result is in line with a recent study of Norwegian drinking water from 2018.

Sammenfatning

Forurening med mikroplastik (MP) findes udbredt i sedimenter, vand samt i dyr og planter over hele verden. I hvilken grad mennesker er eksponeret for denne forurening er dog ringe undersøgt. Nyere studier har indikeret, at MP også kan findes i vand fra vandhaner og fra flasker samt i mad til menneskelig indtagelse. I dette studie undersøger vi tilstedeværelsen af MP i drikkevand fra 17 forskellige steder i Danmark. 50 L drikkevand blev ved prøvetagning udtaget direkte fra vandhanerne gennem 10 µm rustfri stålfiltere i et lukket rustfrit filtersystem for at forhindre kontaminering. Filtrene blev derefter undersøgt visuelt med stereomikroskopi og antal samt karakteristika af alle MP-lignende partikler med størrelser > 100 µm blev registreret – dvs. partikler, der blev identificeret enten som potentiel MP eller partikler af ukendt materiale, som muligvis kunne være MP. 124 (44 %) MP-lignende partikler repræsentativt udvalgt blandt de fundne partikler blev derefter overført manuelt til ZnSe discs og deres kemiske komposition identificeret ved avanceret µFT-IR vha. focal plane array (128 x 128) mapping med en 'pixel resolution' på 5,5 µm. Derudover blev tre vandprøver filtreret direkte gennem Anodisc filtre for at detektere potentiel MP med størrelser < 100 µm. Ved denne analyse blev partikler i 10 % af Anodisc filterområde undersøgt (svarende til 10 % af alle partiklerne i en prøve). Den direkte overførsel til Anodisc gør, at analysen ikke er påvirket af potentiel bias, som følge af en visuel prævurdering af partiklerne.

Resultaterne af den visuelle vurdering af filtrene viste, at der i gennemsnit blev observeret 15,6 MP-lignende partikler > 100 µm per 50 L vandprøve. I 16 ud af 17 prøver var antallet under detektionsgrænsen (29 per 50 L) og i prøven med den højeste koncentration blev der observeret 30 MP-lignende partikler per 50 L. Identifikation vha. ZnSe discs viste, at 3 % af de MP-lignende partikler blev verificeret som MP, mens størstedelen bestod af cellulose-lignende materiale (76 %), og resten gav dårlige spektre (10 %), var ukendte (7 %) eller bestod af protein-lignende materiale (4 %). Typerne af MP, som blev detekteret i vandprøverne, var polyethylen terephthalat (PET), polypropylen (PP) og polystyren (PS). Ved Anodisc metoden blev der foruden PET og PP desuden også fundet acrylonitril butadien styren (ABS) og polyurethan (PU).

Da de fundne koncentrationer af MP er under detektionsgrænsen, kan det ikke konkluderes om fundene af MP stammer fra drikkevandet eller fra prøvekontaminering. I modsætning til tidligere amerikanske og danske studier af drikkevand, som fik stor presseomtale, finder dette studier ingen signifikante koncentrationer af MP i drikkevand fra danske vandhaner. Dette resultat svarer til resultaterne fra en nylig undersøgelse af norsk drikkevand fra 2018.

1. Introduction

In a previous study of tap water collected from countries around the world, widespread contamination with microplastic (MP) was reported by American researchers (Kosuth et al., 2017). Likewise, in a study performed by CphBusiness Laboratory and Environment, findings of MP in tap water, collected from Danish households around Copenhagen was reported (Cphbusiness, 2017). Both the American and the Danish study found that MP were widely present in tap water, with 83 % of 159 globally collected samples and all of the samples from the 16 households from Denmark containing MP, respectively. The concentrations of MP were somewhat variable, but with an average of 4.3 MP particles L⁻¹ (range 0 - 57 MP L⁻¹) in the global study and an average of 18 MP particles L⁻¹ (range 2 - 45 MP L⁻¹) in the Danish study (Cphbusiness, 2017; Kosuth et al., 2017).

The findings of MP in drinking water received considerable media attention, and were subject to a debate about the spread, amount and impact of MP pollution both globally and in Denmark. Furthermore, the interpretation of the data in terms of the reliability of the analyses, and whether the samples provided a representative measure, were up for discussion (Cphbusiness 2017; Kosuth et al., 2017). In order to address concerns that had arisen due to potential intake of MP via drinking water and the consequences to human health, the Danish Environmental Protection Agency (EPA) requested a scientific assessment of the validity of the published results. This assessment pinpointed some weaknesses of the studies, which amongst others included the use of small sampling volumes, use of sampling and filtration equipment that was made of plastic or filter paper, and lack of a valid verification that the microparticles identified as MP were actually MPs (Strand, 2017a; Strand, 2017b). To improve the quality and the reliability of the analysis, the Danish EPA therefore requested an optimization of the method of measurement, and based on this improved method, a new investigation of the extent of MP contamination of Danish tap water was conducted. The present report describes the results of this investigation.

The optimized method of measurement for MP in tap water has been developed at DCE – National Center for Environment and Energy, Department of Bioscience, Aarhus University (Strand, 2018), and met the recommendations for improvement of the previously published studies. In this method, sampling volume was increased to 50 L compared to 0.5 L and 1.0 L in the American and the Danish studies, respectively. By increasing the sample volume, the relative contribution of background contamination in the samples should be decreased. Secondly, a new design of sampling equipment has been produced in stainless steel, consisting of a closed column system with an inserted filter for entrapment of microparticles bigger than 10 µm. The system can be directly coupled to the water tap to prevent contamination during sampling, and can be heated up to 550 °C before use for removal of background contamination. Another important optimization includes the use of advanced Fourier transform infrared spectrometry (µFT-IR) analysis to verify that the potential MP particles in fact consist of synthetic plastic-polymers, and as such are true MP. Altogether, these proceedings enabled a reliable and sensitive method for sampling and analysis of MP in tap water, which has been applied in the present study of Danish drinking water.

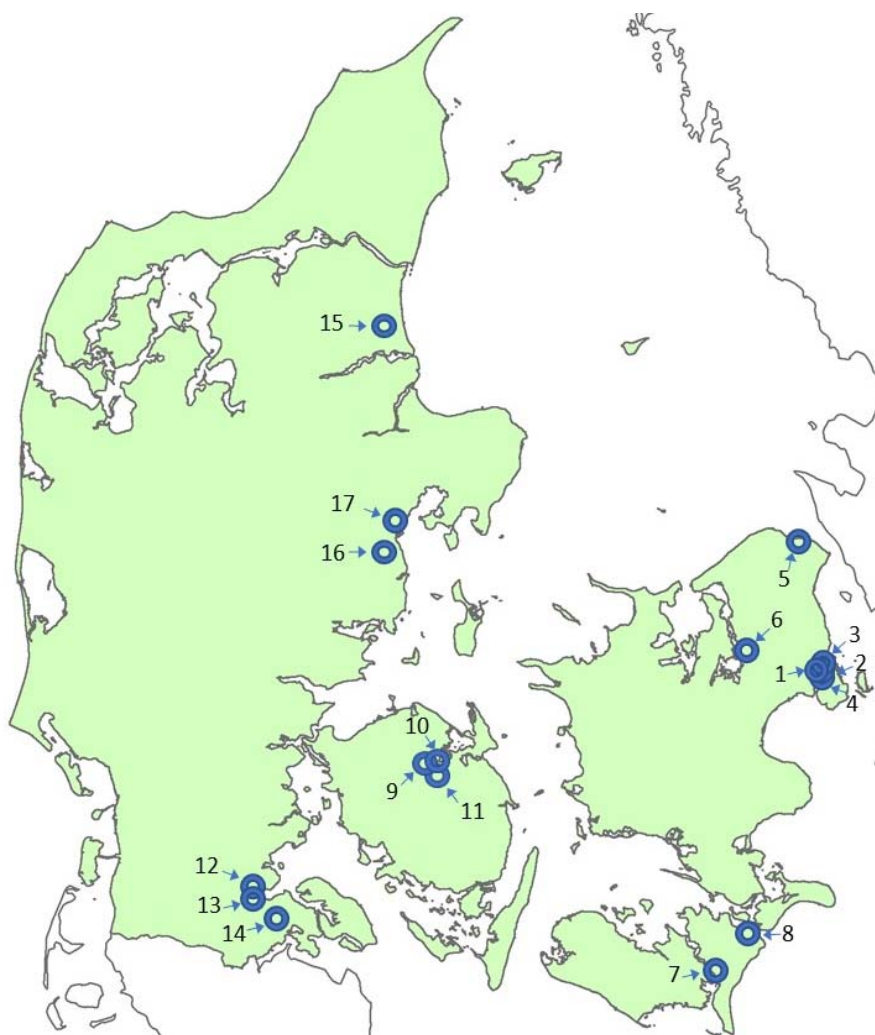
During spring 2018, samples of tap water were collected from 17 different locations spread across Denmark. The locations were chosen to broadly represent Danish drinking water with inclusion of different water abstraction areas, age, type and material of water supply networks and piping, and private households as well as private and public work places. The filter samples have all been brought to the laboratory at Aarhus University, and stereomicroscopy and visual characterization and quantification of all potential MP particles > 100 μm have been performed. A subset (44 %) of representatively selected particles have been subject to $\mu\text{FT-IR}$ analysis for verification of MP and identification of the specific plastic polymer types. In addition, three locations were selected for a more comprehensive analysis of smaller MP with a size-range down to 10 μm , using Anodisc filters and subsequent $\mu\text{FT-IR}$ analysis.

2. Sampling and methodology

2.1 Sampling sites

Samples of tap water were collected from 17 different sites in Denmark, in the period April-June 2018 (Fig. 1). The sampling sites represents a broad geographical origin and cover different water abstraction areas, as well as a large diversity in age and material of the water supply networks and piping. The majority of samples were collected from taps used for drinking water on a daily basis, either in private households (n = 9), in private work places (n = 3) or in private or public institutions (n = 5). For more detailed information of sampling sites, see *Appendix 1*.

Figure 1. Map of the 17 sites selected for analysis of microplastic in Danish drinking water.



2.2 Description of sampling method

Sampling of tap water was performed by direct coupling of a stainless steel column mounted with a stainless steel filter to the tap (Fig. 2), as previously described in the method developed for analysis of MP in tap water (Strand, 2018). A small modification was made to the equipment by exchange of the 20 μm filters with new steel filter nets with a finer pore size and an absolute filtering ability of 11-12 μm .



Figure 2. Tap water sampling. Deployment of the stainless steel sampling column to the drinking water tap for collection of 50 L water samples.

Before sampling was initiated, a short flushing of the tap outlet was performed for about 2 minutes. The sampling equipment was deployed to the tap using a small pipe with an appropriate thread, which was coupled directly to the sampling column with a fitting. During sampling, the taps were opened to full water pressure, and the flow through the sampling column was adjusted to 2.0 L per minute, while the residual water was led directly to the sink through an outlet tube. For each sample, a volume of 50 L was passed through the sampling column and potential MP in the water were retained in the steel filter for quantification and chemical analysis.

The sampling columns were sealed with tinfoil to avoid contamination upon sampling, and brought to the laboratory for assessment of the filters by direct stereomicroscopy, and for chemical analysis of representative potential MP particles by Fourier transform infrared spectrometry (μ FT-IR). In addition, duplicate samples were obtained from a subset of three sampling sites in order to complement the stereomicroscopy assessment with an extended chemical μ FT-IR analysis on Anodisc filters. The Anodisc analysis includes particles $< 100 \mu\text{m}$, which were disregarded in the visual stereomicroscopy assessment, since they are too small to be reliably characterized by this method.

2.3 Cleaning and preparation of sampling equipment

All equipment used for sampling or for treatment of samples consisted of stainless steel or glass, whenever possible. This was chosen to ensure optimal cleaning and reduction of background contaminants. All individual parts were cleaned before usage by washing in a dish washer and baking at $450 \text{ }^\circ\text{C}$ for at least 2 h to evaporate potential contaminants. A few parts of the equipment consisted of or contained rubber material that could not be baked. This was the case for the O-rings that were used for tightening on each side of the steel filter within the sampling column, and for the pipe and fittings (also containing O-rings) used to connect the sampling column to the drinking water tap. These parts were manually cleaned first in a 5 % detergent (filtrated $5 \mu\text{m}$

glass filter) and then in distilled water (filtrated 5 μm) before use. The sampling equipment was assembled in a LAF bench, and was sealed with tinfoil until use on the sampling site. This allowed quick deployment of the gear on site and thus minimized handling in the open air and potential contamination of the sample. In the laboratory used for handling of the samples, an air filtration system was installed (Dustbox Typ 1000, Möcklingshoff, Germany) to reduce airborne contamination.

2.4 Pre-treatment and extraction of filter samples for analysis

The procedure used for pre-treatment and extraction of filter samples for quantification and characterization of microparticles was modified and simplified from the previously suggested method for analysis of MP in tap water (Strand, 2018). The primary changes in the protocol included an initial treatment of the sample with acetic acid to dissolve inorganic particles of e.g. CaCO_3 and a direct flushing of the steel column to collect the microparticles directly onto the steel filter. The previously described protocol was based on extraction of the sample to a mixed cellulose ester (MCE) filter by inversion of the sample column system and release of the microparticles from the steel filter by back-flushing. In the present study, pre-treatment and extraction of the sample was done by firstly adding acetic acid (32 %, 5 μm filtrated) to the sample column and allowing treatment to continue for an hour to dissolve inorganic particles. Subsequently, potential MP particles attached within the sides of the column were carefully flushed onto the steel filter by addition of 500 ml ethanol (5 μm filtrated) to the column sides while turning manually. The sample column was subsequently disconnected, and the steel filter was carefully transferred to a clean glass petri dish and the lid immediately closed to avoid contamination from the air.

The recovery of MP was investigated for the previous method with back-flushing, and extraction efficiency of > 90 % was found for 4 out of 5 MP reference particles (three different sized spheres of PE-material, fragments of PP and fibers of PET) (Strand, 2018). Only the PET fibers showed a lower recovery of 75 % with the back-flushing protocol. In the present study, we tested the recovery of PET fibers with the modified protocol, and we found a recovery of 90 %. In short, 100 green fluorescent PET fibers were added to a steel filter and the filter was assembled within the sample column. The column was mounted directly to a water tap and 60 L of filtrated water was flushed through the column – initially with the tap on full water pressure of 4-5 L per min (high water swirling) and subsequently with a flow of 2 L per min. Subsequently, the column was disconnected from the tap, and extraction of the filter was performed as described above. By inspection with stereomicroscopy, 90 fluorescent fibers were observed on the steel filter, 5 fibers were retained in the column and could be washed out upon disconnection of the system, 1 fiber was observed on the rubber-membrane (O-ring) and the remaining 4 fibers could not be accounted for.

In general, we believe that the modified protocol represented an improvement to the method, by excluding the potential risk of MP entrapped and retained in the steel filter after back-flushing, and thus resulting in a higher recovery.

For the three samples collected for Anodisc analysis, backwashing of the sample column was necessary in order to release the microparticles from the sample filter and collect them on the Anodisc filter (0.2 μm , diameter 25 mm including a polypropylene support ring, GE Healthcare). For these samples, a

pre-treatment with acetic acid was first performed, as described above, and then flushed briefly with pre-filtered water to wash away the acetic acid. An Anodisc filter was mounted in a mini stainless steel column (25 mm diameter), which was then coupled directly to the sample column, and the system was subsequently inverted. Back-flushing with 4×25 ml of a 5 % detergent solution (5 μm filtrated) followed by approximately 20 ml pre-filtered water and 20 ml ethanol (5 μm filtrated) was performed during vacuum suction. The Anodisc filter was carefully removed from the column with forceps and placed in a clean petri dish until analysis.

2.5 Visual quantification of MP-like particles by microscopy

All filters were visually examined with a stereomicroscope (magnification 10-135 \times) for quantification and characterization of MP-like particles. Characterization was done according to recommendations from the Joint Research Center for description of potential MP in aquatic environments (JRC, 2013), i.e. in relation to the type (fibre, fragment, film etc.), color and size of the particles. In general, the use of this approach is recommended only for particles > 100 μm , as the visual distinguishing and characterization of smaller particles < 100 μm is too indeterminate. Hence, a quantification of all MP-like (categorized as either potential or uncertain) MP particles > 100 μm were performed.

The quantification and characterization of MP-like particles by stereomicroscopy relies on a subjective discrimination, as the filters contained a range of different particle types that were not MP. Particles that were regarded as potential MP were MP-like fibers with strong colors such as blue, pink or black (see examples in Fig. 3A-D). Furthermore, all fragments and films with characteristics of MP (see examples in Fig. 3F-I), were also counted as potential MPs. The most dominant group of particles that were regarded as uncertain MPs were fibers or fiber-like filaments with white to transparent colors (see example Fig. 3E). Other fragments or films with MP-like but less characteristic features than the particles grouped as potential MPs, were also recorded as uncertain MPs. Particles that were disregarded in the quantification were all white to transparent fragments with characteristics of either organic or inorganic material (e.g. salts, crystalline material, quartz, skin) (see examples Fig. 3J-L). In addition, all protein-like, rust-like and metallic-like particles (see examples Fig. 3M-O) were also disregarded.

From each sample, a representative number of potential or uncertain MP-particles were selected for validation by $\mu\text{FT-IR}$ analysis. These particles were manually picked from the stainless steel filter and transferred with forceps to a ZnSe disc for analysis. During the transferring process, the lid had to be opened, and the sample filter was subject to potential air contamination. In order to minimize the interference of air contaminants on the study results, the quantification and characterization of microparticles was performed first in a complete inspection of the filter with the lid closed, and the transfer of representative MP-like particles for $\mu\text{FT-IR}$ analysis was done in a second round. The MP-like particles selected for analysis was as far as possible matched to the specific particles observed during the initial microscopy, however particularly for white/transparent fibers, this matching was difficult and particles introduced from the air may to a minor extent, have been included in the assessment.

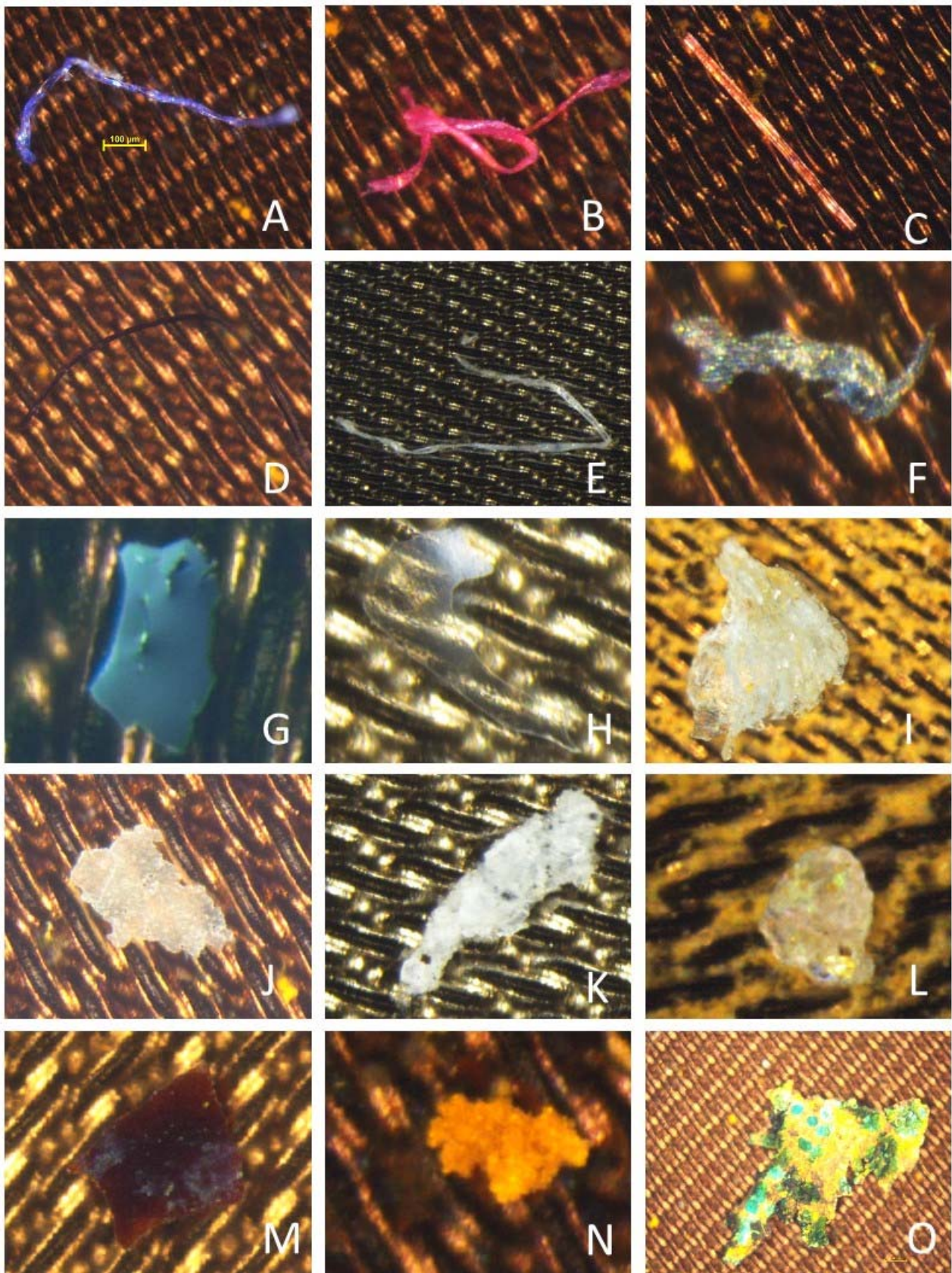


Figure 3. Stereomicroscopy and classification of characteristic particles > 100 μm observed in the water samples. The pictures show examples of particles characterized as potential MP-like fibers (A-D); uncertain MP-like fibers (E); potential MP-like fragments (F,G,I) and potential MP-like films (H). Particles of inorganic and organic material resembling salts (J-K), quartz (L), protein-like (M), rust (N) or metallic particles (O) were disregarded as MP.

2.6 Determination of limit of detection for MP-like particles

In the five blank samples an average of 13.2 (range 5-29) MP-like particles > 100 µm were observed (Table 1). A fraction of these particles (32 out of 66, corresponding to 48 %) were also validated by µFT-IR and amongst these, four particles were verified as MP, consisting of polymethyl methacrylate (PMMA), PET and two fibers of ethylene-vinyl acetate (EVA) (Table 2). Hence, a low level of background contamination exists in the samples, and we determined the limit of detection (LoD) for MP-like particles in the 50 L water samples to be 29 MP-like particles (mean blank + 1.645 × SD for blank) (Armbruster & Pry, 2008), corresponding to 0.58 MP-like particles per L water.

Table 1. Quantification of MP-like particles > 100 µm in blank samples. From each of the 5 blank samples, the total number of MP-like particles as well as the specific numbers of MP-like fibers, fragments and films are shown. The counts are obtained by visual stereomicroscopy and are shown as total numbers per sample - in brackets, the respective numbers of potential and uncertain particles are shown. A subset of 48 % of all MP-like particles were representatively selected from the samples for verification by µFT-IR and results of the identification are shown. PMMA is polymethyl methacrylate, PET is polyethylene terephthalate and EVA is ethylene-vinyl acetate.

Blank sample	Total MP-like particles	MP-like fibres	MP-like fragments	MP-like films	Identification of MP-like particles using µFT-IR A subset of representative particles from each sample have been selected for validation
	Number in sample Total (potential/ uncertain)	Number in sample Total (potential/ uncertain)	Number in sample Total (potential/ uncertain)	Number in sample Total (potential/ uncertain)	
Blank 1	5 (2/3)	4 (1/3)	1 (1/0)	0 (0/0)	PMMA 1 (1/0) Cellulose-like 1 (1/0)
Blank 2	9 (1/8)	7 (1/6)	0 (0/0)	2 (0/2)	PET 1 (0/1) Cellulose-like 6 (0/6) Unknown 2 (0/2)
Blank 3	9 (1/8)	9 (1/8)	0 (0/0)	0 (0/0)	Cellulose-like 5 (1/4) Protein-like 1 (0/1) Poor spectra 1 (0/1)
Blank 4	14 (1/13)	11 (0/11)	3 (1/2)	0 (0/0)	Cellulose-like 3 (0/3) Protein-like 1 (0/1)
Blank 5	29 (1/28)	26 (1/25)	0 (0/3)	0 (0/0)	EVA copolymer 1 (0/1) EVA 1 (0/1) Cellulose-like 5 (0/5) Protein-like 3 (0/3)
Average	13.2 (1.2/12)	11.4 (0.8/10.6)	1.4 (0.4/1)	0.4 (0/0.4)	6.6 (0.6/5.8)

2.7 Chemical identification of MP-like particles by µFT-IR analysis

Particles were identified in the tap water samples using advanced focal plane array (FPA) µFourier transform infrared spectroscopy (µFT-IR). µFT-IR allows for the identification of chemical bonds present in a sample and produces an IR spectrum of an analyzed sample. The resulting spectrum is unique to the specific type of plastic, which with the use of a reference library allows identification of plastic particles as well differentiating plastics from naturally occurring particles. An Agilent Technologies Carry 600 series µFT-IR spectrometer was used in Transmission mode at a resolution of 8 cm⁻¹. Before analysis, 64 background scans were taken to remove any background noise. Each sample had 32 scans taken for identification. A spectral range of 3800 to 875 cm⁻¹

was used for ZnSe samples (full spectrum, since ZnSe discs are IR-transparent) and a spectral range of 3800 to 1300 cm^{-1} for the Anodisc samples (reduced spectrum, since Anodisc filters absorb IR from 1300 cm^{-1}). The spectra were compared to a library of reference spectra to identify them. Particles on the Anodisc filter were analyzed using MP Hunter v 2.0 - a software designed specifically to identify MP in samples analyzed using FPA $\mu\text{FT-IR}$. The software has been developed by the Aalborg University research group lead by professor Jes Vollertsen, Denmark, in collaboration with Alfred Wegener Institute, Germany. This software can automate a significant portion of analysis particularly for complex samples. It can identify MP as well as measure dimensions of particles present in a given sample. Tap water samples that were filtered down on to Anodiscs were first analyzed by Aarhus University (AU), after which the mapping data was sent to The Technical University of Denmark (DTU) for a second individual analysis. At AU and DTU, two independent MP Hunter analyses were performed on the collected data to account for operator bias/operator differences etc. The results from the two separate analyses of the same data set were compared to determine any potential differences in the mapping results of MP particles. From the Anodisc filters, 10 % of the total sample was analyzed, by mapping four different plots summing up to 10 % of the filter area.

3. Results and discussion

In the tap water collected from the 17 different Danish locations, an average of 15.6 (range 4-30) MP-like particles with a size larger than 100 μm were observed, per 50 L sample. In water samples from 16 of the 17 different Danish locations analyzed, the number of MP-like particles $> 100 \mu\text{m}$ were below the LoD (29 MP-like particles per 50 L). It was only from location 8 that the level of MP-like particles were slightly above the LoD with 30 MP-like particles per 50 L sample, corresponding to 0.6 MP-like particles per L tap water (Table 2).

However, only a minor proportion of these particles were categorized as potential MP (on average 3.3 potential MP-like particles per 50 L sample), which with a larger certainty resembled MP, whereas most of the MP-like particles were categorized as uncertain MP (on average 12 uncertain MP-like particles per sample) (Table 2). In order to determine whether these potential and uncertain MP-like particles were in fact MP, chemical validation by $\mu\text{FT-IR}$ were performed. In total, 124 out of 281 (44 %) MP-like particles were validated by $\mu\text{FT-IR}$ analysis (Table 2). The results of the $\mu\text{FT-IR}$ analyses showed that four particles (3 % of all validated) from the water samples were confirmed as MP of the type polypropylene (PP) (1.6 %), polystyrene (PS) (0.8 %) and polyethylene terephthalate (PET) (0.8 %). Whereas the majority of particles were identified as cellulose-like (76 %), protein-like (4 %) or returned unknown (7 %) or the $\mu\text{FT-IR}$ spectra was too poor to identify (10 %) (Table 2).

The predominant type of MP-like particles observed were fibers (82 %). With a total of 231 MP-like fibers detected in the tap water samples, in comparison to only 39 and 11 MP-like fragments (14 %) and films (4 %), respectively (Table 2). All of the water samples contained MP-like fibers, and most of these were white to transparent and were categorized as uncertain MP-like fibers, whereas only a minor proportion of the fibers had strong colors like pink, blue or black and were categorized as potential MP-like fibers. Validation by $\mu\text{FT-IR}$ revealed that the majority of these fibers were not MP, but primarily were fibers consisting of cellulose-like material (see example in Appendix 2). Yet, in two tap water samples originating from site 7 and 16, two transparent fibers were verified as MP of the type PP and PET (Fig. 4C,D).

MP-like fragments were observed in water samples from 12 out of 17 sites, and the number of MP-like fragments ranged from 0 to 7 MP-like fragments per sample (Table 2). Most of the fragments categorized as MP-like were either white/transparent or black, while the occurrence of fragments with strong colors in the water samples was very low. Two transparent fragments originating from location 8 and 15 were verified as MP consisting of PS and PP, respectively (Fig. 4A,B). A large proportion of the other fragments validated by $\mu\text{FT-IR}$ returned spectra of unknown identity or low quality, while some of the MP-like fragments were identified as protein-like (See example in Appendix 3). MP-like films were the least frequent type of MP-like particles observed in the water samples. In 6 of the 17 different sites, MP-like films were found in quantities of one to four particles per sample, whereas in the remaining 11 samples no MP-like films were observed (Table 2). None of the MP-like films validated by $\mu\text{FT-IR}$ were verified as MP.

In this study, the presence of MP > 100 μm was only verified in four tap water samples (from sites 7, 8, 15 and 16) (Table 2). Since not all potential or uncertain MP-like particles were validated by $\mu\text{FT-IR}$ (44 % validated), more than four water samples could potentially contain MP. Yet, as most of the MP-like particles quantified by visual stereomicroscopy were disregarded as MP by $\mu\text{FT-IR}$ (97 %), we thus estimate that approximately half of the tap water samples from the 17 locations actually contained MP. In all positive samples only a single particle of MP > 100 μm was verified and these findings were all below the LoD (Except from sample 8). Whether the observed MP-particles are derived from the tap water or occur due to contamination can therefore not be determined on the basis of this study.

The relatively few particles of MP detected in this study presented different characteristics in terms of plastic type and morphology. Amongst the visually characterized particles > 100 μm , transparent fibers of PET and PP and transparent fragments of PP and PS (Fig 4A-D) were observed in the water samples. In addition, some smaller fragments of PP, ABS and PU were also observed by $\mu\text{FT-IR}$ (Appendix 4). The origin of these particles can be multiple and are hence difficult to deduce. As indicated in a previous study in Denmark, it is highly unlikely that MPs are present in groundwater (Mackevica & Hartmann, 2018). One possible source of MP contamination of tap water could be from the network pipes or local pipes supplying water from the water works to the end user. In this study, information about the water supply network was collected for all the sampling sites. However, from this information only plastic types of PE or PVC were used for piping, and thus the pipes seem not to be the source for the observed MP particles. Contamination could also potentially occur further downstream the water supply network during processes at the waterworks or in the groundwater reservoirs, or further upstream during passage of in-house VVS installations or fittings. However, as only a single site exceeded the LoD, the source of detected MP in this study may equally well derive from contamination of the sampling equipment or atmospheric contamination from the multiple sources of plastic items in our surroundings (Wesch et al., 2017).

The concentrations of MP found in the present study are in line with a recent study of MP in Norwegian drinking water (Uhl & Svendsen, 2018). In that study, the concentration of MP particles at the end user were below the limit of quantification of 1 MP-particle per L in 19 of 24 samples (79 %) and ranged from 2.0 to 6 MP-particles per L in the remaining 5 samples. In comparison, we found that 16 out of 17 Danish tap water samples (94 %) were below the LoD of 0.58 MP-like particles per L, as determined by visual stereomicroscopy of the blank samples. In the Norwegian study only MP with a size greater than 100 μm were measured, and the results are thus directly comparable to the size-fraction determined in this study. However, the method used for detection and verification of MP in the Norwegian study is not described. Hence, whether the particles determined as MP have been verified to be true MP or also encompass MP-like particles of cellulose-like or other material, is not known.

Compared to previous studies of Danish drinking water (Cphbusiness, 2017) where 18 MP-particles per L water was reported, and of global drinking water where on average 4,3 MP-particles per L was reported (Kosuth et al., 2017), this study finds a very low level of < 0.58 MP-particles per L in 94 % of the investigated Danish tap water samples.

Table 2. Quantification of MP-like particles > 100 µm in 50 L tap water samples. From each of the 17 Danish sampling sites, the total number of MP-like particles as well as the specific numbers of MP-like fibers, fragments and films are shown. The counts are obtained by visual stereomicroscopy and are shown as total numbers per 50 L sample - in brackets, the respective numbers of potential and uncertain particles are shown. A subset of 44 % of all MP-like particles were representatively selected from the samples for verification by µFT-IR and results of the identification are shown. From site 1, two water samples (a, b) were obtained from two different sampling days. PP is polypropylene, PS is polystyrene and PET is polyethylene terephthalate.

Sampling site	Total MP-like particles	MP-like fibres	MP-like fragments	MP-like films	Identification of MP-like particles using µFT-IR A subset of representative particles from each sample have been selected for validation
	Number in 50 L Total (potential/ uncertain)	Number in 50 L Total (potential/ uncertain)	Number in 50 L Total (potential/ uncertain)	Number in 50 L Total (potential/ uncertain)	
1 a + b	29 (6/23)	25 (4/21)	3 (2/1)	1 (0/1)	Cellulose-like 9 (2/7)
	18 (6/12)	13 (1/12)	5 (5/0)	0 (0/0)	Cellulose-like 5 (1/4) Poor spectra 5 (5/0)
2	16 (4/12)	14 (3/11)	1 (1/0)	1 (0/1)	Cellulose-like 9 (2/7)
3	12 (1/11)	12 (1/11)	0 (0/0)	0 (0/0)	Cellulose-like 5 (1/4)
4	13 (5/8)	8 (1/7)	1 (1/0)	4 (3/1)	Cellulose-like 3 (1/2) Protein-like 2 (1/1) Unknown 1 (0/1) Poor spectra 1 (1/0)
5	7 (1/6)	6 (1/5)	1 (0/1)	0 (0/0)	Cellulose-like 4 (1/3) Poor spectra 1 (0/1)
6	11 (4/17)	19 (2/17)	2 (2/0)	0 (0/0)	Cellulose-like 2 (0/2) Unknown 1 (1/0)
7	10 (1/9)	10 (1/9)	0 (0/0)	0 (0/0)	PP 1 (0/1) Cellulose-like 7 (1/6)
8	30 (10/20)	20 (5/15)	7 (4/3)	3 (1/2)	PS 1 (1/0) Cellulose-like 4 (2/2) Protein-like 1 (1/0) Unknown 1 (1/0) Poor spectra 1 (0/1)
9	12 (1/11)	10 (0/10)	1 (1/0)	1 (0/1)	Cellulose-like 4 (0/4) Unknown 2 (1/1)
10	13 (5/8)	13 (5/8)	0 (0/0)	1 (0/0)	Cellulose 7 (3/4) Poor spectra 1 (0/1)
11	11 (2/9)	10 (2/8)	0 (0/0)	0 (0/1)	Cellulose-like 6 (2/4)
12	27 (1/26)	20 (1/19)	7 (0/7)	0 (0/0)	Cellulose-like 8 (1/7) Unknown 1 (0/1)
13	9 (0/9)	9 (0/9)	0 (0/0)	0 (0/0)	Cellulose-like 3 (0/3) Protein-like 1 (0/1) unknown 1 (0/1) poor spectra 2 (0/2)
14	4 (0/4)	4 (0/4)	0 (0/0)	0 (0/0)	Cellulose-like 4 (0/4)
15	19 (6/13)	14 (1/13)	5 (5/0)	0 (0/0)	PP 1 (1/0) Cellulose-like 4 (0/4)
16	17 (6/11)	12 (1/11)	5 (5/0)	0 (0/0)	PET 1 (0/1) Cellulose-like 6 (1/5) unknown 2 (2/0)

17	13 (1/12)	12 (0/12)	1 (1/0)	0 (0/0)	Cellulose-like 4 (0/4) Protein-like 1 (1/0) Poor spectra 1 (0/1)
Average	15.3 (3.3/12)	12.8 (1.6/11)	2.2 (1.5/0.7)	0.6 (0.2/0.4)	6.9 (1.9/5)
Total	281 (60/221)	231 (29/202)	39 (27/12)	11 (4/7)	124 (34/90): PP 2 (1/1) PS 1 (1/0) PET 1 (0/1) Cellulose-like 94 (18/76) Protein-like 5 (3/2) Unknown 9 (5/4) poor spectra 12 (6/6)

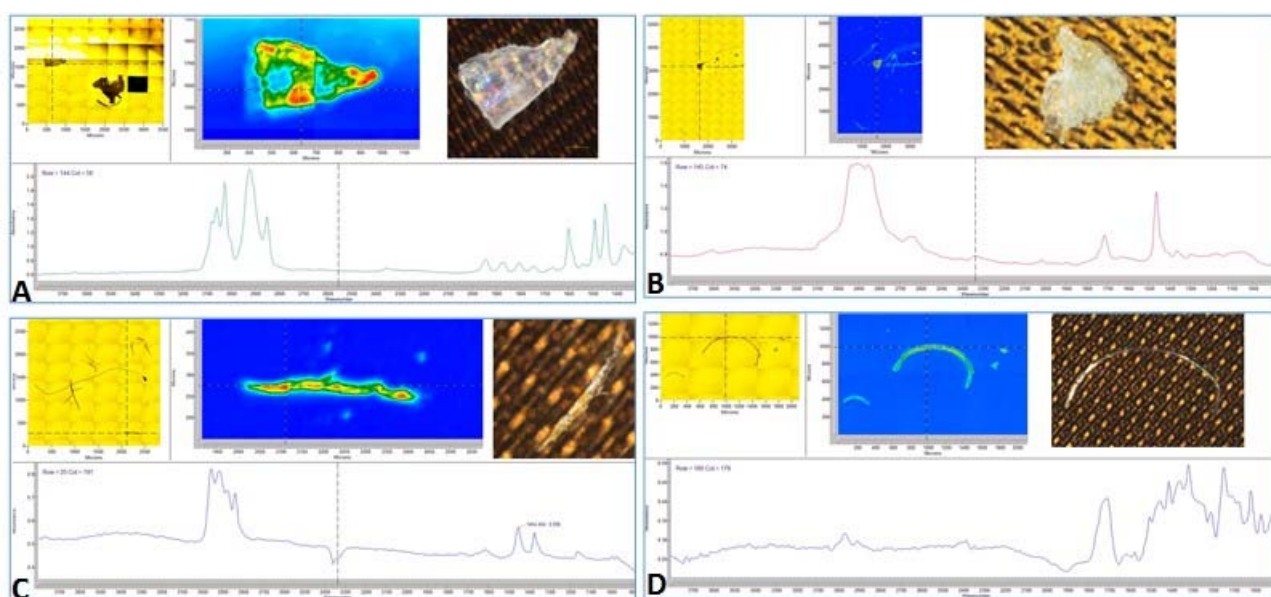


Figure 4. Verification and identification of MP-particles > 100 µm by µFT-IR. Specific µFT-IR spectres and corresponding photos from microscopy are shown for; A) a transparent fragment (~600 µm) from site 8 characterized as a potential MP-fragment by visual inspection and identified as polystyrene; B) a transparent fragment (~400 µm) from site 15 characterized as a potential MP-fragment and identified as polypropylene; C) a transparent fiber (~450 µm) from site 7 characterized as an uncertain MP-fiber and identified as polypropylene; D) a transparent fiber (~1500 µm) from site 16 characterized as an uncertain MP-fiber and identified as polyethylene terephthalate.

3.1 Detection of MP-particles in Anodisc samples

Duplicate samples of tap water were collected from 3 of the 17 different sample locations (site 1, 6 and 8) in order to make Anodisc analyses of MP-particles, including the smaller size fraction of 10-100 µm particles. For each of the three Anodisc samples, 10 % of the total Anodisc sample area was analyzed, in two individual analyses carried out at AU and at DTU. The findings from both AU and DTU were quite similar, indicating that MPhunter software is suitable for MP identification with minimal differences between independent users. In total, five MP-particles were identified in the three water samples, with one MP-particle from site 1 (PP), four MP-particles from site 6 (PU, 2 × PET and acrylonitrile butadiene (ABS)) and no MP-particles from site 8 (Table 3, details on size and verification of MP particles in Appendix 4+5). All identified MP-particles were fragments and no MP-like fibers were observed. A

single MP particle was larger than 100 μm (a fragment of ABS, $138 \times 69.5 \mu\text{m}$) and three particles were in the size-range of 20 – 100 μm , whereas one MP-particle was < 20 μm (Appendix 4). In addition to the well-categorized plastic-types that were found, a number of MP-like particles with resemblance to rubber was observed (Table 3). These MP-like particles were all recognized as a reasonably good match to the reference PE spectrum by the MP Hunter software, with the MPhunter settings used at AU but not at DTU. However, a closer manual examination of the $\mu\text{FT-IR}$ spectra and comparison with a more extensive reference spectra library indicated that these fragments were likely composed of rubber (Appendix 5E). This was carried out using KnowitAll® Informatics System 2017, which compares sample spectra with an extensive library of reference plastic developed by AU containing over a hundred spectra such as plastics, rubbers, natural material as well as many others. The fragments, which were also present in both of the blank samples might originate from the rubber O-rings used in the sampling gear. However, many of the questionable spectra were difficult to identify even by using larger spectral libraries, since all the spectra collected on Anodisc samples had a cutoff at 1300 cm^{-1} , which means that the fingerprint region of the spectrum is not available. In order to avoid this issue and improve the recognition of MP particles in the samples, other substrates can potentially be used such as silicon filters, which are IR-transparent (Kappler et al., 2015).

The finding of only five MP-particles by using Anodiscs for a broad-range particle size screening (> 10 μm) in three tap water samples indicates a very low level of MP in the tap water. A concentration of 0.2, 0.8 and 0.0 MP-particles per L water was estimated for the three analyzed samples, respectively (Table 3), which is in the same range as the LoD. In two blank samples, a total of three MP-particles were detected, which results in the same average concentration of 0.3 MP-particles per L water, as found for the tap water samples (Table 4). That other types of plastic polymers (i.e. PP, ABS and PU) were found in tap water samples than in the blank samples, although only as single particles, can potentially indicate that they originate from the sampled tap water. The same statement cannot be made for the PET particles, because a PET particle also was found in one of the blank samples. As the support ring from the Anodisc consisted of PP, the PP particle that was found in the water sample from site 1 could potentially originate from here, though it was not found in any of the other samples. However, the finding of only 1-2 particles of each of the identified plastic polymer types per sample, underlines that any firm conclusions of the origin of the plastic polymer particles must be treated with caution.

Table 3. Number and ID of MP-particles (> 10 μm) in tap water detected by Anodisc analysis. Three sampling sites (1, 8 and 6) were randomly selected for Anodisc analysis, which were independent of pre-selection by stereomicroscopy and also included analysis of smaller particle-sizes. 10% of the Anodiscs area were analysed, corresponding to 10 % of each of the water samples. ¹Particles identified by AU. ²particles identified by DTU. *only one of the two PET-particles were identified by DTU. PU is polyurethane, PET is polyethylene terephthalate, PP is polypropylene, PA is polyamide, ABS is acrylonitrile butadiene.

Site	PU	PET	PP	PA	ABS	Rubber	Total MP-particles identified (rubber)	Estimated MP-particles in 50 L	Estimated MP-particles in 1 L
1	0	0	1 ^{1,2}	0	0	0	1 (0)	10 (0)	0.2 (0)
6	1 ²	2 ^{1,2*}	0	0	1 ^{1,2}	0	4 (0)	40 (0)	0.8 (0)
8	0	0	0	0	0	2 ¹	0 (2)	0 (20)	0.0 (0.4)
Total	1	2	1	0	1	2	5 (2)	16.7 (6.7)	0.3 (0.1)

Table 4. Number and ID of MP-particles (> 10 µm) in blank samples detected by Anodisc analysis. Two blank samples were prepared identically to the tap water samples for Anodisc analysis. In the analyses, 10% of the Anodiscs area were analysed, corresponding to 10 % of each of the blank samples. ¹Particles identified by AU. ²particles identified by DTU. PU is polyurethane, PET is polyethylene terephthalate, PP is polypropylene, PA is polyamide, ABS is acrylonitrile butadiene.

Blank	PU	PET	PP	PA	ABS	Rubber	Total MP-particles identified (rubber)	Estimated MP-particles in 50 L	Estimated MP-particles in 1 L
1	0	0	0	1 ²	0	3 ¹	1 (3)	10 (30)	0.2 (0.6)
2	0	1 ^{1,2}	0	1 ²	0	9 ¹	2 (9)	20 (90)	0.4 (1.8)
Total	0	1	0	2	0	12	3 (12)	15 (60)	0.3 (1.2)

4. Conclusions

The analyses of microplastic in tap water performed on samples taken from 17 different locations in Denmark showed that:

- On average, the total number of visually determined MP-like particles > 100 µm were 15.6 (range 4-30) per 50 L sample. In 16 out of 17 (94 %) tap water samples, the amount of MP-like particles were below the limit of detection, which were 29 particles per 50 L sample, corresponding to 0.58 MP-like particles per L tap water.
- Fibres, i.e. elongated filaments, were the dominating type of MP-like particles, contributing with 82 % followed by fragments (14 %) and films (4 %). Most fibres (202 out of 231) were white to transparent, and were categorized as “uncertain” MP-like particles, as they were suspected mainly to consist of cellulose-like material, µFT-IR analysis confirmed that this was in fact the case.
- µFT-IR analysis was used to identify 44 % of the visually characterized MP-like particles. The identification showed that 3 % (4 out of 124) of the MP-like particles could be verified as MP, whereas the majority consisted of cellulose-like material (76 %), and the rest provided poor spectra (10 %), unknown spectra (7 %) or were protein-like material (4 %).
- The presence of MP was verified by µFT-IR in tap water samples from 6 of the 17 different locations, using either ZnSe-discs or Anodisc analysis. However, as not all MP-like particles detected by stereomicroscopy were validated (44 %) by µFT-IR, more samples could potentially contain MP, though in concentrations below the limit of detection.
- The specific plastic polymer types, which were verified as MP in the tap water samples were PET, PP, PS, ABS and PU. In the blank samples, MP particles of PET, PMMA, PA and EVA were observed. Since only 1-2 of each type of plastic polymers were observed in each of the tap water samples with verified MP content, no firm conclusions about the origin of the MP from tap water or contamination can therefore be made.
- This study finds that 94 % of Danish tap water samples contain MP below the Limit of detection of 0.58 MP particles per L water. This is significantly lower levels than reported in previous American and Danish studies from 2017, which were highly publicised by the media, but in line with the levels reported from a study of Norwegian drinking water in 2018.

5. References

Armbruster D. A. & Pry T. (2008). Limit of blank, limit of detection and limit of quantification. Clin Biochem Rev. 29(Suppl 1): S49-S52.

Cphbusiness (2017). Miljøteknologer har fundet mikroplast i dansk drikkevand. Pressemeldelse fra Cphbusiness. <https://www.cphbusiness.dk/om-cphbusiness/nyheder/2017/09/miljoeteknologer-har-fundet-mikroplast-i-dansk-drikkevand>

JRC (2013). A guidance document within the common implementation strategy for the marine strategy framework directive MSFD technical subgroup on marine litter, JRC scientific and policy reports, Joint Research Centre (JRC) report EUR 26113 EN. <http://publications.jrc.ec.europa.eu/repository/bitstream/JRC83985/lb-na-26113-en-n.pdf>

Käppler, A., Windrich, F., Löder, M.G., Malanin, M., Fischer, D., Labrenz, M., Eichhorn, K.J. and Voit, B., 2015. Identification of microplastics by FTIR and Raman microscopy: a novel silicon filter substrate opens the important spectral range below 1300 cm⁻¹ for FTIR transmission measurements. Analytical and bioanalytical chemistry, 407(22), pp.6791-6801.

Kosuth M., Wattenberg E.V., Mason S.A., Tyree C., & Morrison D. (2017). Synthetic polymer contamination in global drinking water, Orb media, Final report May 16, 2017. https://orbmedia.org/stories/Invisibles_final_report

Mackevica A., Hartmann NB. (2018). Mikroplast i grundvand - En vurdering af potentialet for forekomst af mikroplast i dansk grundvand. Notat til Miljøstyrelsen, April 2018. <https://mst.dk/media/148257/bilag-3-notat-mikroplast-i-grundvand.pdf>

Strand J. (2017a). Udtalelse om validiteten af målemetode anvendt i amerikansk undersøgelse, der viser mikroplast i drikkevand verden over. Faglig redegørelse fra DCE, 21. september, 2017. http://dce.au.dk/fileadmin/dce.au.dk/Udgivelser/Notater_2017/Notat_om_amerikansk_undersogelse_af_MP_i_drikkevand_globalt_21sep17fin.pdf

Strand J. (2017b) Udtalelse om validiteten af målemetode anvendt i dansk undersøgelse, der viser mikroplast i drikkevand fra taphaneprøver i 16 husestande i Københavnsområdet. Faglig redegørelse fra DCE, 21. september, 2017. http://dce.au.dk/fileadmin/dce.au.dk/Udgivelser/Notater_2017/Notat_om_MP_analyser_af_dansk_drikkevand_21sep17fin.pdf

Strand J. (2018). Forslag til målemetode til brug for undersøgelser af mikroplast i taphanevand. Notat fra DCE – Nationalt Center for Miljø og Energi, 22. januar 2018. http://dce.au.dk/fileadmin/dce.au.dk/Udgivelser/Notater_2018/Notat_DCE_Maalemetode_mikroplast_i_taphanevand.pdf

Uhl W. & Svendsen C. (2018). Mapping microplastics in Norwegian drinking water. A summary of results and evaluation of suspected health risks. https://norskvann.no/files/docs/MP_in_DW_-_executive_summary.pdf

Wesch C., Elert AM., Wörner M., Braun U., Klein U., Paulus M. (2017). Assuring quality in microplastic monitoring: About the value of clean-air devices as essentials for verified data. *Scientific Reports* 7(1): 5424.

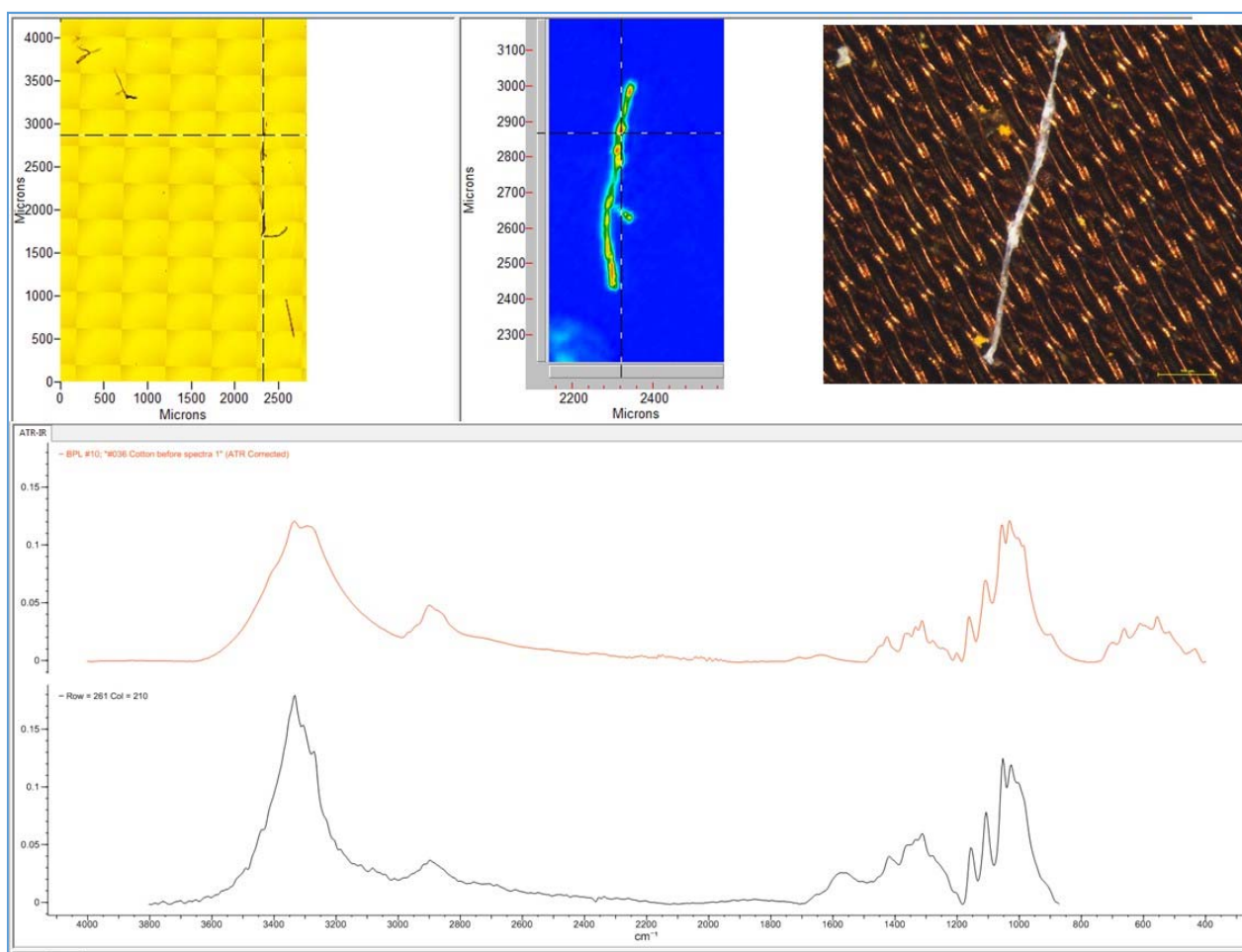
Appendix 1. Description of sampling sites

Site	Address (Postal code)	Date of sampling (2018)	Waterwork/ Water abstraction area	Description of water supply network (material, age, length)	Type of building
1	Knabrostræde (1210 Kbh. K)	11 April and 1 June	Marbjerg, Lejre, Slangerup, Søndersø, Regnemark, Thorsbro (Infrequently from Islevbro).	Local water pipes: PE 90 mm, from 2004. Supply network: Mix of old cast iron pipes and newer PE pipes.	Small shop, older building/ installations
2	Eriksgade (1708 Kbh. V)	11 April	Regnemark, Thorsbro, Slangerup, Søndersø (Infrequently from Islevbro, Lejre, Mar- bjerg).	Local water pipes: Cast iron 100 mm, from 1890. Supply network: Mix of old cast iron pipes and bonna pipes as well as newer PE and PEH pipes.	Apartment build- ing (private hous- ing)
3	Blegdamsvej (2200 Kbh N)	2 May	Søndersø, Slangerup, Lejre, Marbjerg and Is- levbro (Infrequently from Regnemark).	Local water pipes: Cast iron, 300 mm, from 1877 (Blegdamsvej) + PE 160 mm from 2015 (Tagensvej) + private lat- erals. Supply network: Mix of old cast iron pipes and newer PE pipes.	Newer building/ installations
4	Ernie Wilkins Vej (2450 Kbh S.)	2 May	Regnemark (primarily) and Thorsbro	Local water pipes: PE 160 mm, from 2007. Supply network: Mix of old and new cast iron pipes, Bonna pipes from 1970's and newer PEH pipes.	Apartment build- ing (private hous- ing), Newer building/ installations
5	Saunte Bygade (3100 Hornbæk)	4 April	Hellebæk	Supply network: PVC, from 1950-1970, DfW: 5.6 km	Hotel
6	Kolibrivej (4040 Jyllinge)	28 May	Værebros	New supply network (PE) during the last 7 years. Raw water pipes were changed in 2017. New water meters and new wells (where available). Hence, it corre- sponds almost to a new water works.	Single-family house
7	Østre Allé (4800 Nykøbing F)	8 June	Nykøbing F	Supply network: Primarily PE, from 2013. DfW: 1.9 km	Single-family house
8	Ore Strandpark (4850 Stubbekø- bing)	8 June	Ore	Supply network: PVC, from 1963. DfW: 800 m	Single-family summer house
9	Villestofteheg-net (5210 Odense NV)	26 April	Holmehaveværket	Supply network: Concrete, from mid 70s (12.5 km). Ductile iron, from 1980-1990 (9 km). Mix of PVC (from 1971) and PE (from 1990) (5 km).	Terrace house
10	Lindehaven (5270 Odense)	26 April	Lunde	Ductile iron, from 1997 (4 km). PVC, from 1965 (3.3 km). Local water pipes: PE, from 2017 (300 m).	Single-family house
11	Dagmargade 6000 Odense C)	26 April	Hovedværket	Supply network: Concrete, from 1948 (200 m). Ductile iron, from 1987 (4 km). Local water pipes: PE, from 1996 (240 m).	Apartment build- ing (private hous- ing)

12	Hjelmallé (6200 Aabenrå)	19 April	Farversmølle/ Lind- snakke	Laterals: PVC, 110 mm (242 m). Supply network: Eternit 150 mm (530 m)	Public institution (Sports facility)
13	Lundsberg Industri- vej (6200 Aabenrå)	19 April	Farversmølle	Laterals: PVC, from 1981. Supply network: PVC 200 mm. DfW: 3.6 km, passing through Sønderskov cistern	Public institution (Educational in- stitution)
14	Kværsløkke (6300 Gråsten)	19 April	Farversmølle	Long supply network. Mix between PVC 75 mm (from the 1970s) and PE (newer)	Small company
15	Aavangen (9575 Terndrup)	16 May	Terndrup	Supply network: depending on water flow, mix of PVC/PEL (PE) with some parts in eternit. Shortest DfW: 1.1 km; 160 mm PVC (430 m) 110 mm PVC (180 m) 75 mm PVC (450 m) 40 PVC (70 m), from 1990-2010	Public institution
16	Gunnar Clausens vej (8260 Viby J.)	16 May	Østerbyværket	Supply network: two options, depending on water flow: 225 mm PVC (1982) (600 m), 315 mm PE (2007) (3.5 km), 225 mm PVC (1971) (300 m) 300 mm Eternit (1972-78) (1300 m), 315 mm PE (2014) (200 m), 300 mm Eternit (1972) (750 m), 315 mm PE (1999) (400 m), Mix of PE (500 m) and PVC (450 m)	Medium-size company
17	Kristtornvej (8200 Aarhus N.)	16 May	Truelsbjergværket	Supply network: 400 mm Eternit (1974) (3.1 km) 400 mm PVC (1989) (700m) 400 mm Eternit (1963-74) (1.2 km) Mix of Eternit (400 m), PE (800 m), PVC (1.6 km) (1967-2014)	Double-family house

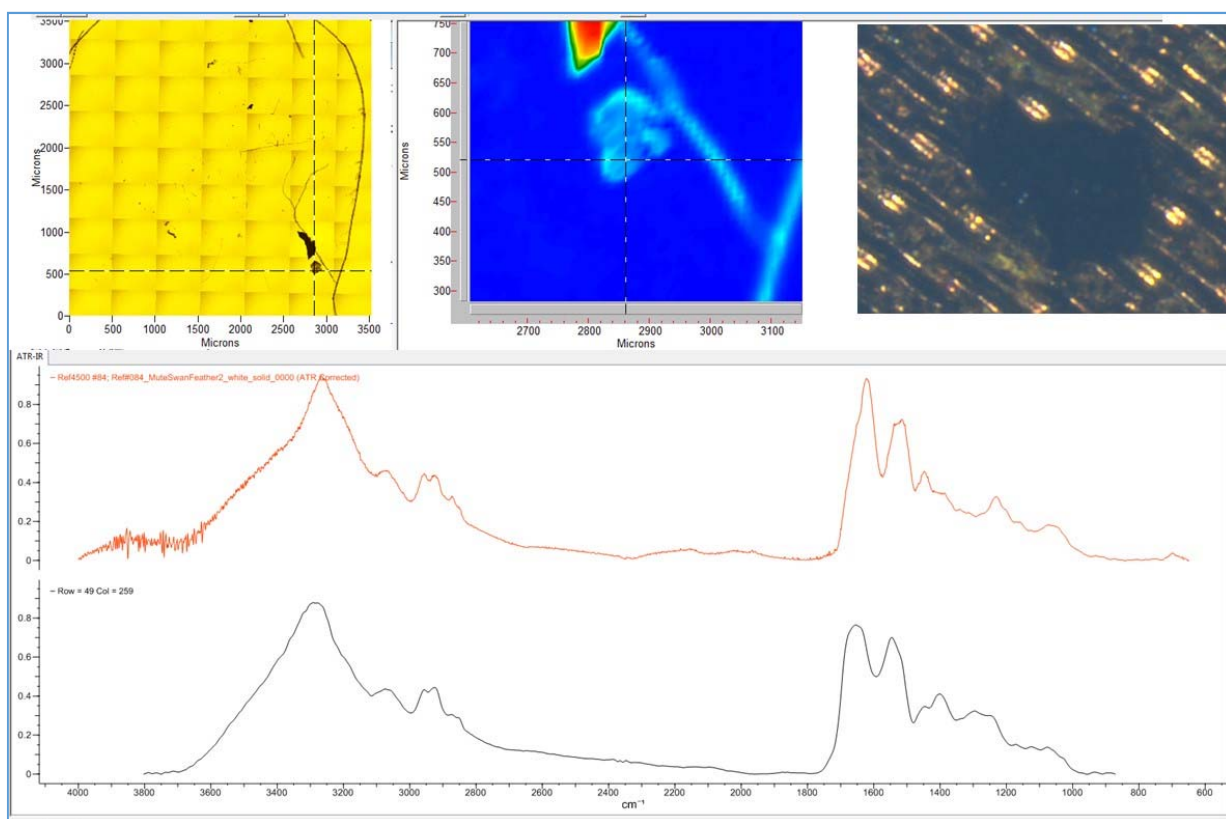
Abbreviations: Distance from water works (DfW).

Appendix 2. Identification of cellulose-like particles by μ FT-IR



Appendix 2. Example of an MP-like particle validated by μ FT-IR as cellulose-like material. The microscopy-photo of a fiber classified as an uncertain MP-like fiber is shown (upper right) together with the corresponding photo of the ZnSe-disc and heat-map of the same fiber from the μ FT-IR analysis (upper left and upper middle, respectively). The spectra shows in black the μ FT-IR spectra of the analyzed fiber and in orange the matching spectra of cotton from the MP Hunter reference database.

Appendix 3. Identification of protein-like particles by μ FT-IR



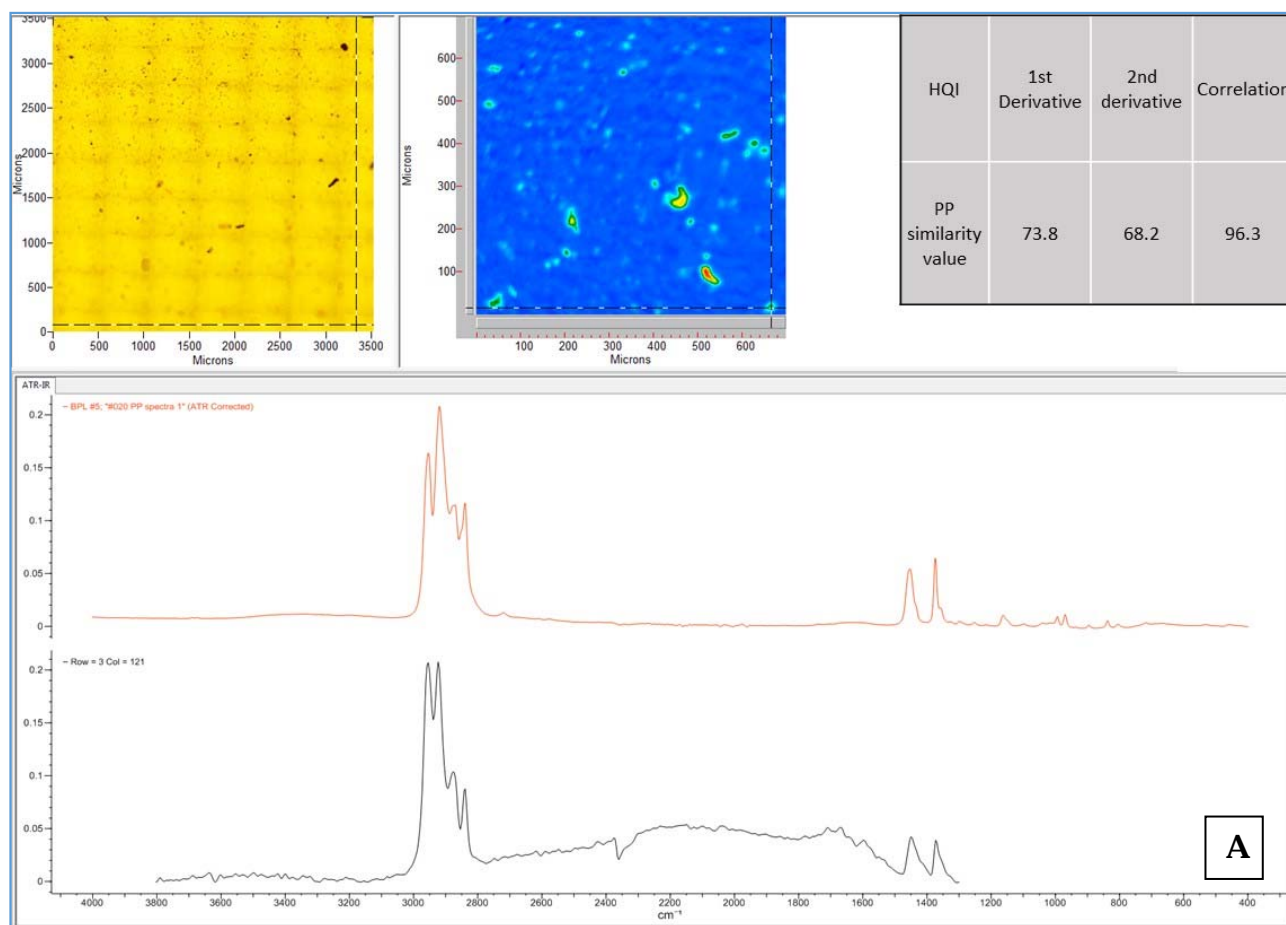
Appendix 3. Example of an MP-like particle validated by μ FT-IR as protein-like material. The microscopy-photo of a fragment classified as an uncertain MP-like fragment is shown (upper right) together with the corresponding photo of the ZnSe-disc and heat-map of the same fragment from the μ FT-IR analysis (upper left and upper middle, respectively). The spectra shows in black the μ FT-IR spectra of the analyzed fragment and in orange the matching spectra from the MP Hunter reference database of swan feather (only a few protein-based spectra are present in the database, and hence a close match of the origin of the protein material is not possible).

Appendix 4. Detection of MP-particles using Anodiscs

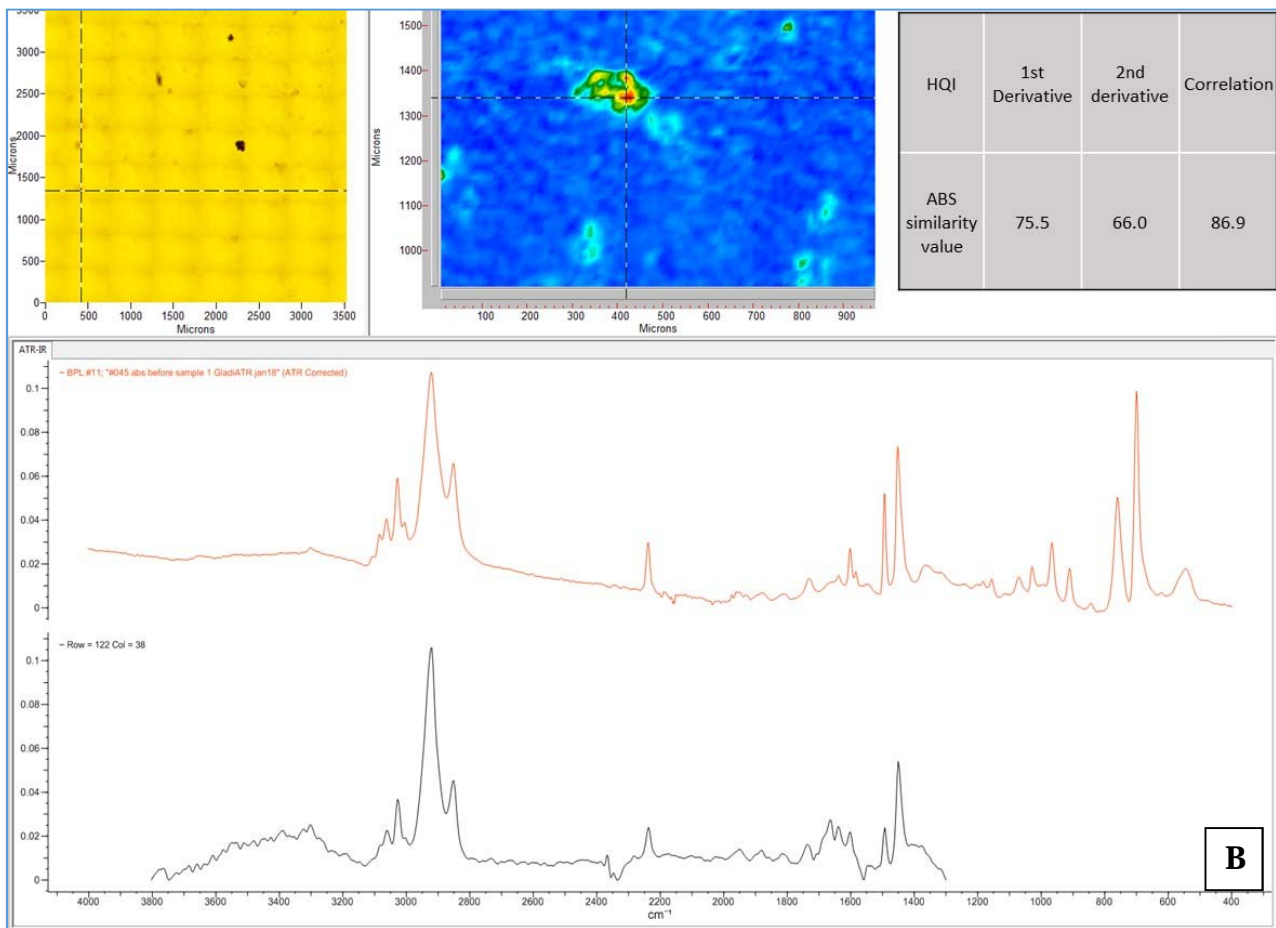
Appendix 4. Microplastic and their physical parameters identified in Anodisc samples. The table shows the complete list of all particles identified as either MP-particles or rubber in the three tap water samples (site 1, 6 and 8) and in the two blank samples subjected to Anodisc analysis. The ID and the physical parameters are shown for each particle detected. ¹Particles identified by AU. ²particles identified by DTU. PU is polyurethane, PET is polyethylene terephthalate, PP is polypropylene, PA is polyamide, ABS is acrylonitrile butadiene.

Site	ID	Area μm^2	Length μm	Width μm	Vol. μm^3	Mass ng	Size Class
1	PP ^{1,2}	303	25.3	15.2	1840	2.097	20-100
6	PET ¹	61	11	7	169	0.193	< 20
6	PU ²	696	40.3	22	6120	7.894	20-100
6	PET ^{1,2}	3418	78	55.8	76315	86.999	20-100
6	ABS ^{1,2}	7532	138	69.5	209332	238.639	> 100-300
8	Rubber ¹	333	30.1	14.1	1874	2.136	20-100
8	Rubber ¹	454	30.1	19.2	3484	3.972	20-100
Blank 1	PA ²	635	35.1	23	5852	6.555	20-100
Blank 1	Rubber ¹	1422	53.8	33.7	19152	21.833	20-100
Blank 1	Rubber ¹	666	37.6	22.6	6004	6.844	20-100
Blank 1	Rubber ¹	1634	54.7	38	24847	28.325	20-100
Blank 2	PA ²	847	38.3	28.2	9549	10.695	20-100
Blank 2	PET ^{1,2}	121	13.3	11.6	562	0.64	< 20
Blank 2	Rubber ¹	424	30.1	17.9	3035	3.46	20-100
Blank 2	Rubber ¹	363	28.2	16.4	2382	2.715	20-100
Blank 2	Rubber ¹	393	33.5	14.9	2348	2.677	20-100
Blank 2	Rubber ¹	635	40.7	19.9	5048	5.754	20-100
Blank 2	Rubber ¹	333	28.2	15	2001	2.281	20-100
Blank 2	Rubber ¹	454	30.1	19.2	3484	3.972	20-100
Blank 2	Rubber ¹	303	22.9	16.8	2036	2.321	20-100
Blank 2	Rubber ¹	303	25.3	15.2	1840	2.097	20-100
Blank 2	Rubber ¹	151	17.8	10.8	655	0.746	< 20

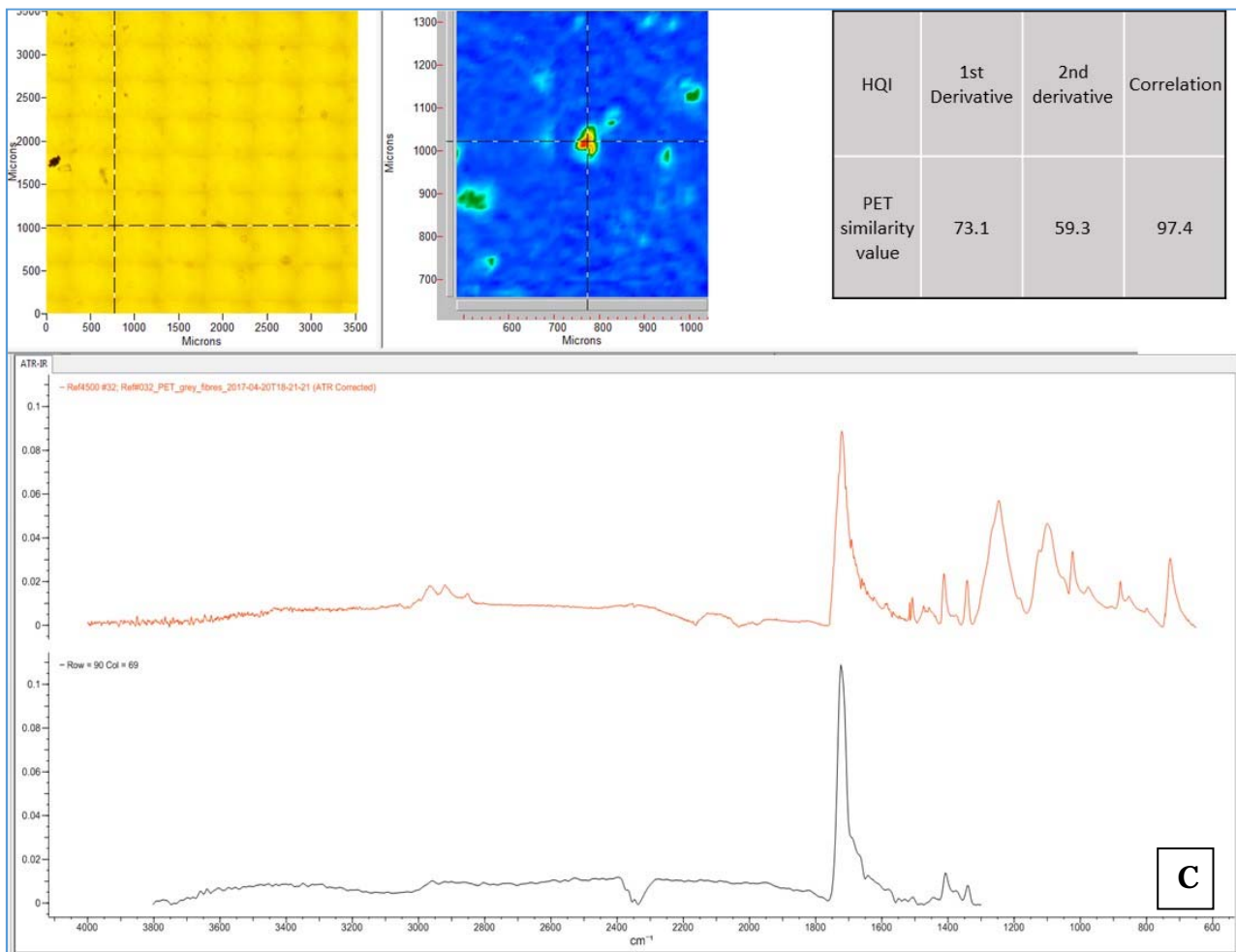
Appendix 5. Identification of MP-particles using Anodiscs



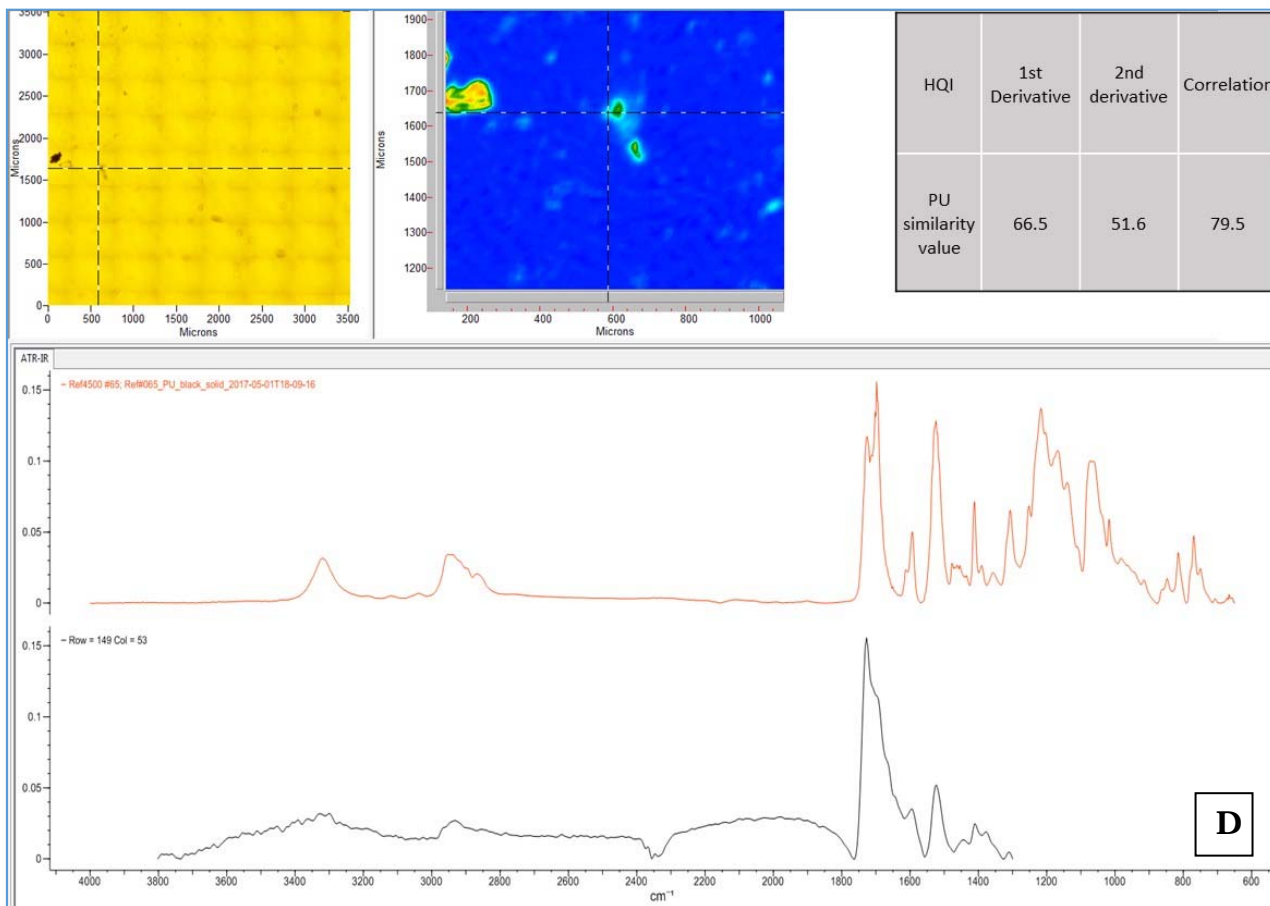
Appendix 5A. Example of a particle validated by Anodisc analysis as polypropylene (PP). The photos show the Anodisc and the corresponding heat-map of the PP-fragment from the μ FT-IR analysis (upper left and upper middle, respectively). The spectra shows in black the μ FT-IR spectra of the analyzed PP-fragment and in orange the matching spectra from the MP Hunter reference database of PP. The table shows the high-quality index (HQI) with the PP similarity values calculated by the MP Hunter software and giving a correlation of 96.3 % for the spectra match.



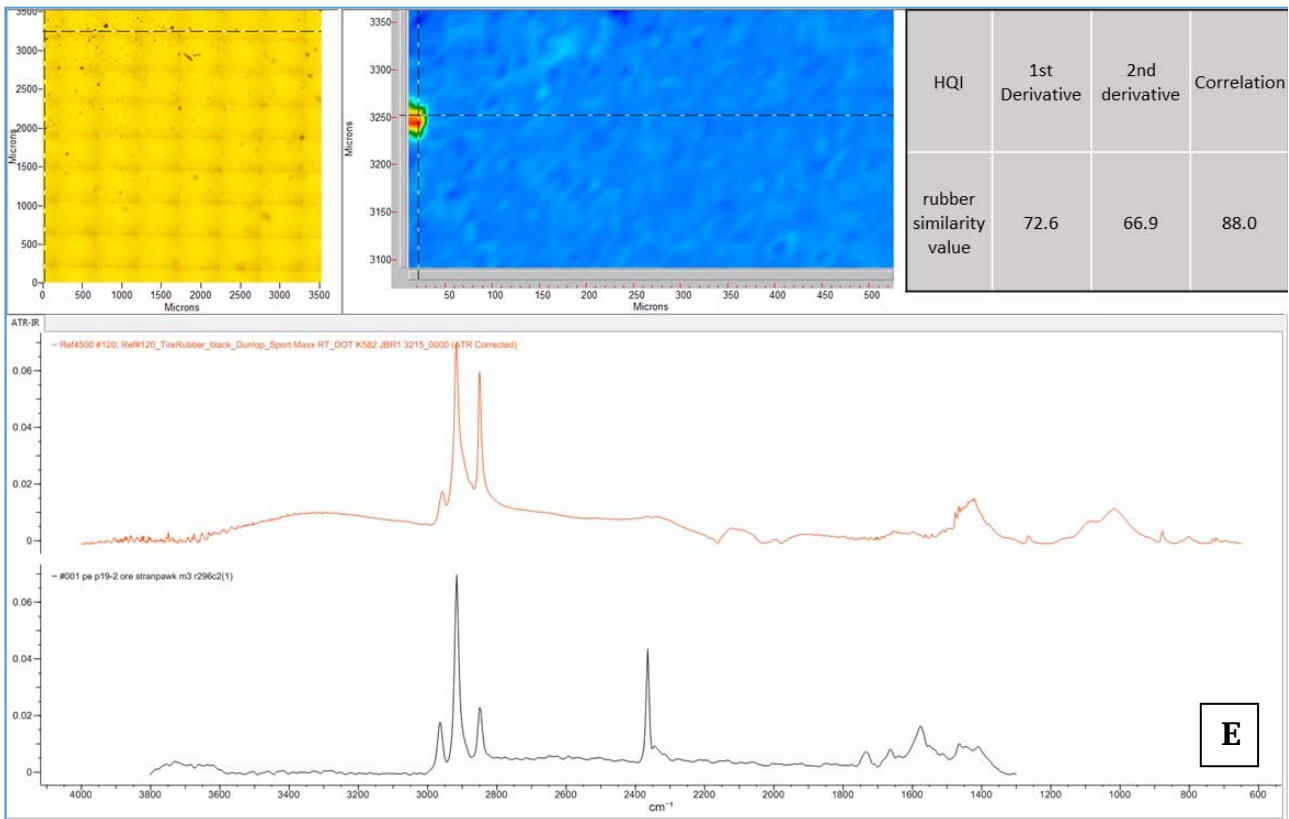
Appendix 5B. Example of a particle validated by Anodisc analysis as acrylonitrile butadiene (ABS). The photos show the Anodisc and the corresponding heat-map of the ABS-fragment from the μ FT-IR analysis (upper left and upper middle, respectively). The spectra shows in black the μ FT-IR spectra of the analyzed ABS-fragment and in orange the matching spectra from the MP Hunter reference database of ABS. The table shows the high-quality index (HQI) with the ABS similarity values calculated by the MP Hunter software and giving a correlation of 86.9 % for the spectra match.



Appendix 5C. Example of a particle validated by Anodisc analysis as polyethylene terephthalate (PET). The photos show the Anodisc and the corresponding heat-map of the PET-fragment from the μ FT-IR analysis (upper left and upper middle, respectively). The spectra shows in black the μ FT-IR spectra of the analyzed PET-fragment and in orange the matching spectra from the MP Hunter reference database of PET. The table shows the high-quality index (HQI) with the PET similarity values calculated by the MP Hunter software and giving a correlation of 97.4 % for the spectra match



Appendix 5D. Example of a particle validated by Anodisc analysis as polyurethane (PU). The photos show the Anodisc and the corresponding heat-map of the PU-fragment from the μ FT-IR analysis (upper left and upper middle, respectively). The spectra shows in black the μ FT-IR spectra of the analyzed PU-fragment and in orange the matching spectra from the MP Hunter reference database of PU. The table shows the high-quality index (HQI) with the PU similarity values calculated by the MP Hunter software and giving a correlation of 79.5 % for the spectra match.



Appendix 5E. Example of a particle validated by Anodisc analysis as rubber. The photos show the Anodisc and the corresponding heat-map of the rubber-fragment from the μ FT-IR analysis (upper left and upper middle, respectively). The spectra shows in black the μ FT-IR spectra of the analyzed rubber-fragment and in orange the matching spectra from the MP Hunter reference database of rubber. The table shows the high-quality index (HQI) with the rubber similarity values calculated by the MP Hunter software and giving a correlation of 88.0 % for the spectra match.

[Blank page]

ANALYSIS OF MICROPLASTIC PARTICLES IN DANISH DRINKING WATER

Contents and composition of microplastic (MP) particles have been analysed in 50 L samples of tap water collected from 17 households and working places across Denmark in 2018. In 16 out of 17 samples the concentration of MP-like particles > 100 µm were below the LoD, which were determined as 0.58 MP-like particles per L.