To what extent are wastewater treatment systems a gateway for microplastic particles in the aquatic and terrestrial environment?

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INTRODUCTION

Substantial amounts of microplastic particles are likely to be collected in the sewage system, mainly deriving from cleaning of synthetic clothing, and waste water disposal facilities have been identified as point sources of microplastic in the aquatic environment [1,2,3,4]. Today's waste water treatment plants (STP) are not designed to manage microplastics. The main objective of this study was to develop a standardized method to collect, quantify and characterize microplastic particles. This study aims at contributing to the knowledge about the possible significance of waste water treatment facilities as point sources of micro-plastic particles into food chains both in water and land. Special attention is paid to the sewage sludge. According to Scandinavian practice stabilized waste water sludge shall preferably be utilized on agricultural soil. However, more research is needed to understand the incidence, potential accumulation and biological adverse effects of micro-plastic in aquatic and terrestrial ecosystems [5,6]. We have implemented a pyrolysis-gas chromatography-mass spectrometry (Pyr-GC-MS) based methodology to waste water filtrates, sludge and plant material. Applied to waste water deriving from the Stavanger area, a typical Scandinavian urban settlement of about 250.000 inhabitants, the data obtained so far suggest a microplastic content (10-500 um) in the influx water of approximately 0.5 % of the water dry matter content, with polyvinylchloride, polystyrene and polyethylene terephthalate weighted the most recurrent polymer types. Application of the methodology to a wider range of WWTPs will bear out whether similar figures generally holds in a Scandinavian context.

MATERIALS AND METHODS

The study was carried out in two sewage treatment plants (StP) applying different cleaning systems. The North Jæren StP (NJ StP) collects household wasterwater and consists of bar screening, grit removal, pre-aeration, primary sedimentation, chemical-physical treatment and secondary sedimentation while the Grødaland StP (GR StP) collects industrial discharges and consists of bar screening, grit removal, pre-aeration, primary sedimentation, activated sludge treatment, secondary sedimentation and a tertiary filtration. Samples were taken by filtration of influent and effluent waste water, and also from the water right before each of the main sewage treatment steps (fig. 1). Tests were performed on the two sewage treatment plants on both sludge and inlet/outlet water samples. First efforts were made to reduce the quantity of organic matter mainly responsible to trap and cluster plastic particles as well as to interfere with the detection ability of the main used techniques.

An ad hoc engineered stainless-steel device able to filter large volume of wastewater and separate particles into two size dimensional classes: <450 μm and 450 μm < X < 10 μm was developed (LVMCD, fig. 2). The prototype was designed to hold two quickly interchangeable filtering modules being free from any source of plastic particles contamination. Total volume of filtered raw water was recorded during each of the sampling sessions while the trapped solid matter was submitted to analysis.

The obtained samples were submitted to a sequence of enzymatic reactions to selectively degrade the most relevant and predominant fractions of the organic matter i.e., cellulose, paper and fat material as well as proteins. The multistep detergent wash off and enzymatic degradation was followed by an alkali KOH-driven or oxidizing H2O2 driven procedures. Briefly, approx., 0,5 – 1,0 gram from each of the two LVMCD' s retaining filters was placed i borosilicate crucible filters with certified porosity of 10 μm and incubated under a sequence of different enzymes, alkali or oxidizing conditions (fig. 3). In the selection of optimal sample digestion parameters priority was given to digestion efficiency and time consumption. After every incubation the digestate was vacuumed off and the loss of material and volume recorded. Final sample obtained was finally quantitatively transferred and preconcentrated in a Ø 25 mm, 1,2 μm fiberglass GF/A filter. The filter was carefully folded in a tin cup, derivatized and submitted to pyrolysis. Similar approach was followed while analyzing the microplastics composition in sludge.

Pyrolysis GC was used to obtain structural information about macromolecules by carrying out a GC/MS analysis of their thermal degradation products. eight polymer types of the most common standard polymers PVC, HDPE, PET, PA66, PP, PS, PMMA and PC, were analyzed using Pyr-GC/MS to obtain a pyrogram database. Briefly, the sample is pyrolyzed at 550 C for 60s. The temperature of the transfer line is 350 C. In order to separate and detect pyrolysis products, the Multi-Shot Pyrolizer EGA/PY-3030D (Frontier Laboratories Ltd) is interfaced to a gas chromatograph (GCMS) Shimadzu QP2010 Ultra. Mass spectra of organic plastics and pyrolysis products were obtained by running the MS in full scan mode with a mass range between 10 and 600 amu. Polymers were identified according to their retention times and specific targeted masses. Pyrolysis products are further identified by consulting the F-search Ver.3.4 Search engine mass spectra library (Frontier Laboratories Ltd). A spiking solution obtained by mixing of the most environmentally occurring polymers was processed and incubated under the above reported digestion conditions in both raw water and sludge samples to investigate method's accuracy and recovery performances.

Data related to inputs (raw water) and outputs (sludge and wastewater) were normalized to the size of the STPs by dividing the concentrations of plastic particles in influent and effluent water with their population equivalents.

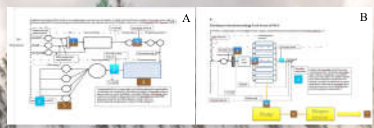

Fig.1 - Simplified scheme of the seleted StP of Grødland (A) and Nord-Jæren (B)

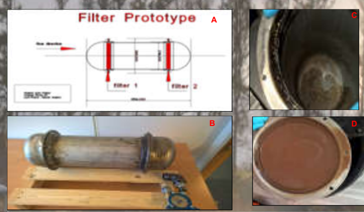

Fig.2 - (A) Large Volumes Microplastics Collector Device - LVMCD Prototype. Red arrows: position of the switchable stainless steel filters. (B) Casted LVMCD and plastic free pumping system. (C) sampling activities in the Grødaland STP Inlet.

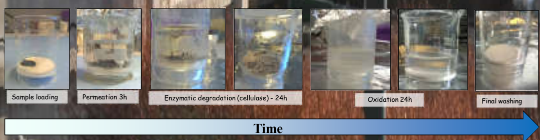

Fig.3 - Samples of processed raw water inlet submitted to the sequence of detergents, enzymatic and oxidation steps


Fig. 4 - Shimadzu Optima 2010 GCMS coupled with Lab Frontiers EG/PY 3030 two step pyrolizer and 48 well autosampler

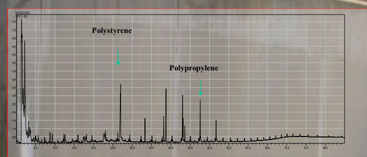

Fig.5 Sample of a pyrogram from processed raw water ilntet

RESULTS

Our findings point out that an incubation of SDS 5% (w/v) for 3h at 50°C followed by cellulase driven enzymatic activity at 45 °C in PBS (pH 5) overnight and subsequent incubation of H2O2 (30%) at 50°C yielded the best experimental condition to degrade the organic matter content trapped in the crucible filters (fig. x). The MPs that were observed by the loop during the course of these extraction and purification studies showing an irregular shape. Polyethylene, polystyrene and polyvinylchloride and polyester are most frequently found in the influent wastewater while the effluent's analysis pointed out a polyethylene and polypropylene enriched composition. Results are summarized in tab. 1.

Accoridng to these observations, microplastics removal was in range of 66-83% with higher removal rates estimated in NJ StP and a slightly lower removal efficiency in the GR StP (45-61%).

	NJ StP		Grød StP	
	μg/l			
	Inlet	Outlet	Inlet	Outlet
PVC	4,90 ± 1,20	0,80 ± 0,50	3,10 ± 0,50	1,10 ± 0,50
PS	30,40 ± 3,10	2,15 ± 0,10	10,60 ± 0,10	0,99 ± 0,10
PET	0,90 ± 0,10	0,20 ± 0,10	0,50 ± 0,10	0,20 ± 0,10
HDPE	10,90 ± 0,36	1,20 ± 0,20	3,11 ± 0,20	0,90 ± 0,25
PMMA	2,20 ± 1,00	< LoD	< LoD	< LoD
PP	1,11 ± 0,50	0,20 ± 0,10	0,90 ± 0,20	0,30 ± 0,20
PC	*	*	*	*
PA66	*	*	*	*

* levels discontinuosly observed in analyzed samples

Tab. 1 - Estimated levels of plastic particles in the the inlet/outlet woter of the two investigated sewage tretof the sewage treatment processes in the investigated StPs

DISCUSSION & CONCLUSIONS

This work provides the first insights about potential discharge of microplastic ranging from 10 to 450 μm into the aquatic and terrestrial environment. The pyrolysis-gas chromatography-mass spectrometry (Pyr-GC-MS) was selected as promising detection technique for the characterization microplastics in complex environmental matrices such as raw water inlet and sludge. The technique offers some key advantages like the ability to identify and estimate the levels of several polymers within a single chromatographic run and further info related to the co-occurring plastic additives composition can be added and it is in progress in our research activities. Furthermore, the technique shows a good combination of execution rapidity, analytical precision and sensitivity and it is not influenced by the plastic particle's dimensions such as the other detection techniques related to planar microscopy like μ-FTIR and μ-Raman. On the other hand, it is a destructive analysis; therefore, samples cannot be resubmitted to analysis in case of controversy. However, collecting the total ion chromatogram during the analysis makes possible to re-process the data, targeting polymer not initially considered in the research activity. The Pyr-GC-MS has limited capability to give estimation of plastic particles dimensions which is a fundamental info for ecotoxicologists, toxicologists and environmental risk assessors. Hence a combined approach integrating microscopy and gas chromatographic is suggested.

The early outcomes of this work provide new information on the role of wastewater treatment plants as a route for microplastics entering the sea. An efficient treatment process appears to decrease the concentrations of microplastics in the effluent but may increase the input to the terrestrial ecosystem. Further work is needed to orient the industrial technology toward an effective reduction of microplastic particles inputs into the environment thus improving their collection within critical steps in the sewage treatment process.

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