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Microplastics en route: Field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota

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ABSTRACT

Environmental contamination by plastic particles, also known as 'microplastics', brings synthetic materials that are non-degradable and biologically incompatible into contact with ecosystems. In this paper we present concentration data for this emerging contaminant in wastewater treatment plants (WWTPs) and freshwater and marine systems, reflecting the routes via which these particles can travel and the ecosystems they potentially impact along their path. Raw sewage influents, effluents and sewage sludge from seven municipal WWTPs in the Netherlands contained mean particle concentrations of 68–910 L^{-1}, 51–81 L^{-1} and 510–760 kg^{-1} wet weight (ww), respectively (particle sizes between 10 and 5000 μm). Even after treatment, wastewater constitutes a source of microplastic pollution of surface waters, and via biosolids applications in farming and forestry, plastic retained in sewage sludge can be transferred to terrestrial environments. The WWTPs investigated here had a mean microplastics retention efficiency of 72% (s.d. 61%) in the sewage sludge. In the receiving waters of treated and untreated wastewaters, we detected high microplastic levels in riverine suspended particulate matter (1400–4900 kg^{-1} dry weight (dw)) from the Rhine and Meuse rivers. Amsterdam canal water sampled at different urban locations contained microplastic concentrations (48–187 L^{-1}), similar to those observed in wastewater that is emitted from sewage treatment facilities in the area. At least partial settling of the particles occurs in freshwater as well, as indicated by microplastics in urban canal sediments (<68 to 10,500 particles kg^{-1} dw). Microplastics in suspension in the water column have the potential to be discharged into the sea with other riverine suspended particulates. We report microplastic concentrations from 100 up to 3600 particles kg^{-1} dry sediment collected at 15 locations along the Dutch North Sea coast. The high microplastic enrichment in marine sediments compared to most literature data for seawater at the surface supports the hypothesis of a seabed sink for these materials. Marine species are heavily exposed to plastic particles. Body residues between 10 and 100 particles g^{-1} dw were measured in benthic macroinvertebrate species inhabiting the Dutch North Sea coast: filter-feeding mussels and oysters (species for human consumption) as well as other consumers in the marine food chain.

1. Introduction

There is growing evidence that plastic particles are accumulating in the marine environment and that they pose a threat to marine ecosystems and have significant potential for adverse human health and socio-economic impacts (e.g. Thompson et al., 2004; Moore, 2008; Ivar do Sul and Costa, 2014; Oosterhuis et al. 2014). Tiny, solid, water insoluble and persistent, these plastic particles have been termed 'microplastics', which by current definitions include particles up to 5 mm in size, and down to the nano-sized particle range (≤999 nm) as well (Arthur et al., 2009; MSFD Technical Subgroup on Marine Litter, 2013; GESAMP, 2015). Microplastics are in the lower range of plastic item size categories present in the marine environment. Plastics themselves are a huge category of synthetic materials, in which polymers and other chemicals are compounded in diverse combinations to create a wide range of plastic materials; close to 5000 grades of plastic materials were on the market at time of writing (see industry material information database www.campusplastics.com).

Microplastics have been detected worldwide within every marine habitat including the water column, and beach, subtidal and deep sea sediments, Arctic ice (Barnes et al., 2009; Van Franeker et al., 2011; Browne et al., 2011; Cole et al., 2011; Claessens et al., 2011; Liebezeit and Dubaish, 2012; Van Cauwenberghe et al., 2013; Obbard et al., 2014) and more recently fresh water bodies (Faure et al., 2012; Eriksen et al., 2013; Free et al., 2014; Sanchez et al., 2014; Wagner et al., 2014; Driedger et al., 2015), and food and drinks for human consumption (Bouwmeester et al., 2015). An increasing number of marine

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organisms are known to ingest microplastics voluntarily or through contaminated prey (Cole et al., 2011; Wright et al., 2013; Besseling et al. 2013; Watts et al. 2014; GESAMP, 2015).

Complete mineralization of polymers in plastic in the environment is a process estimated to take on the order of hundreds of years (Andrady, 2011), long outlasting most of the chemical additives in the materials which have much faster mineralization kinetics (Andrady, 2015). Plastic materials including particulates may be emitted to seas and oceans via rivers, sewage discharge, land run-off, via ships at sea, spillage of pre-production pellets, plastic granulates and powders, or created by the weathering and fragmentation of macro-sized litter already emitted to the environment (Gregory and Andrady, 2003; Barnes et al., 2009; Andrady, 2011; GESAMP, 2015). Most plastic particulates found in the environment are created by weathering and fragmentation of plastic litter of any size ('secondary' microplastics), however manufactured particulates such as nurdles, granulates, powders, microbeads and nanoparticles made of plastic ('primary' microplastics) may also be represented in the total microlitter fraction.

It is important to understand which matrices and environments are contaminated with plastic particles because exposure implies potential risks of trophic transfer (Farrell and Nelson 2013; Setälä et al. 2014) and adverse ecosystem and health effects (Tanaka et al., 2013; Wright et al. 2013; GESAMP 2015).

In this study we screened the concentrations of microplastics in both marine and freshwater matrices, from wastewater treatment plants (WWTPs), rivers and canals to North Sea biota and sediments. These data help fill microplastic data gaps regarding WWTPs and freshwater environments and can be used to guide future monitoring programs and to facilitate decision making and other political processes.

2. Methods

2.1. Sampling locations, matrices and procedures

The samples for microplastic analysis from marine and freshwater environments in the Netherlands included: wastewater influent, sludge and treated effluent, riverine suspended particulate matter (SPM), canal water, canal and marine sediments and marine biota (Table S1 lists all sample information). The sampling, which took place between 2012 and 2013, was performed at seven Dutch WWTPs, on the Rhine and Meuse Rivers in the Netherlands and Germany, the Amsterdam canals, three sites on the Dutch coast for marine biota, and for marine sediments at two estuaries, one site in the Wadden Sea and 12 other sampling points in the North Sea, off the Dutch coast (map of all locations Fig. S1). All glass jars for sampling were precleaned and rinsed with MilliQ analytical grade water prior to sampling to avoid background contamination. Precautions were taken during sampling to avoid sample contamination in the field (e.g. limiting sample material exposure to air, synthetic textiles and plastic materials).

2.1.1. WWTPs

The seven WWTPs sampled had various hydraulic capacities and receiving surface waters in the Netherlands. From Heenvliet (R7), Amstelveen (R3), Horstermeer (R5) and Blaricum (R4) samples of both influents and effluents were taken. Maintenance at the Amsterdam West facility (R2) prevented influent sampling, although effluents and sewage sludge were sampled. Effluent and sewage sludge were collected from Westpoort (R1) and effluent only was collected from Houtrust (R6). At the time of sampling, Heenvliet (R7) was experimenting with a membrane bioreactor (MBR), now no longer in use. All WWTP effluents, influents and sewage sludge samples were collected in glass jars (ca. 2 L for each sample) and stored in the dark until analysis.

2.1.2. Riverine SPM, canal water, freshwater and marine sediments

Riverine SPM was collected using a continuous centrifugation system on the Rivers Meuse (P1) and Rhine (P2, P3) and stored wet (to avoid microplastic occlusion) in glass jars.

Grab samples of sediment were taken and homogenized to make a single sample for analysis (1 L for each sample) and stored in the dark at 4 °C until analysis.

At six sites (A1–A6) on canals in the City of Amsterdam, 2 L surface water samples were collected in glass bottles. At each of the same six sites, two or more Van Veen grab samples of sediment were taken and homogenized to make a single sample (1 L) for analysis.

Marine sediments were collected with a Van Veen grab at 15 locations (S1–S15) at sea as part of the Dutch national water quality monitoring program (MWTL). Each sample consisted of a homogenized pool of five individual sediment grab samples (top 10 cm). All sediment samples were stored in glass sample jars in the dark at 4 °C until analysis. Dry weight of sediments and SPM was determined gravimetrically after freeze-drying ca. 5 g of each sample until a constant weight.

2.1.3. Biota

Biological specimens were collected from artificial rocky shores in three littoral zone locations along the Dutch coast in March 2013: Eastern Scheldt outside Neeltje Jans (ES); Rhine estuary, Port of Rotterdam (RE), and the coast near Ter Heijde (TNS). Five benthic species were selected for the screening survey: common shore crab (*Carcinus maenas*); sand hopper (*Gammarus* spp.); periwinkle (*Littorina littorea*); blue mussel (*Mytilus edulis*) and Pacific oyster (*Crassostrea gigas*). All individuals per species and per location were pooled prior to analysis. Whole organisms were preserved either frozen or in 70% isopropanol and stored cool and dark until analysis. The microplastics were measured in soft tissues of the biota (including gut contents) and in whole bodies of the sand hoppers. The microplastics measured in this way represent what is transferred when such prey organisms are consumed whole: the contaminant body residue of the prey is consumed as a whole (including stomach contents) by the predator.

2.2. Microplastic extraction and analysis

2.2.1. Particle filtration and analysis

Microplastics were extracted from the sample matrices (details below), and the final step involved filtration over 0.7 μm glass filters (Whatman GF/F). Each extract was examined using light microscopy to count particles and record shape (fibres, spheres and foils and total microplastics were recorded), as well as the size category: >300 μm (i.e. 300–5000 μm) or <300 μm (i.e. between ca. 10 and 300 μm). The larger size category corresponds to particle sizes commonly targeted in seawater surface microplastic sampling. Concentrations were expressed as number of particles per unit mass or volume of the sample matrix.

To confirm the particles counted were plastic and not another material, Fourier transform infrared (FTIR) analysis was performed on a subsample of particles from sediment and biota samples representing 6% of the total number of particles counted in the study as a whole. The samples were filtered over Al_2O_3 filters (Whatman Anodisc 25, pore size 0.2 μm). Microscopic particles visually identified as plastic were analysed using an integrated Bruker LUMOS FTIR microscope operated in transmission mode. 64 scans were used, corrected for 64 background scans with the resolution set at 4 cm^{-1}. Polymers were identified using the Bruker library containing approximately 26,000 material types.

2.2.2. Wastewater treatment plant effluents and influents, canal water

All influent and effluent samples were well mixed immediately prior to taking aliquots (100 g) from each for extraction, since microplastics are not in solution and most can be assumed not to be neutrally buoyant. Sodium chloride (NaCl) was added to the saturation point (1.2 kg NaCl L^{-1}). Whole water samples from each canal site were