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Microplastic Types in the Wastewater System—A Comparison of Material Flow-Based Source Estimates and the Measurement-Based Load to a Wastewater Treatment Plant

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Abstract: Microplastics are omnipresent in the environment, and wastewater treatment plants (WWTPs) have been highlighted as a transport pathway. The aim of this study is to contribute to increased understanding of microplastic sources in wastewater and test the possibilities of source tracking. Previous research has focused either on estimating microplastic contribution from various sources or on quantifying occurrence based on measurements. In this paper, these two approaches are compared. Microplastic types detected in the influent to a WWTP in Sweden are compared with estimations of sources in the WWTP catchment area. The total load from the identified sources was estimated to 1.9–14 tonnes/year, and the measurement-based load was 4.2 tonnes/year. In general, there was a good agreement between the two approaches; microplastic types with large shares at the inlet also had large contributions in the source estimates. An exception was cellulose acetate, which was not found at the inlet despite a large theoretical contribution. Many uncertainties remain, which lead to large intervals for the source estimates. The comparison can give an indication into which part of the intervals is most likely. Investigating more WWTPs with different characteristics and including particle morphology will further increase the understanding of sources that contribute to the presence of microplastics in wastewater.

Keywords: FTIR; polymer; substance flow analysis; source tracking; stormwater

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1. Introduction

Plastic is a versatile material that is widely used in society, but its use is connected to pollution problems [1]. In addition to large plastic items, so-called microplastics (plastic particles <5 mm) have been identified as a water pollution problem of growing concern [2,3]. Microplastics have been reported in both marine [4] and freshwater environments [5]. They are also found in urban wastewater and stormwater [6–8], and elevated levels have been reported outside of wastewater treatment plants (WWTPs) [9]. The research on microplastics has grown rapidly and made important conceptual as well as empirical progress. However, microplastics research is still in its infancy, and research results are often uncertain and sometimes contradictory. This makes it difficult to develop a sufficiently reliable overview of sources and pathways that can be used as a basis for comprehensive and efficient abatement strategies.

Earlier studies have, based on measurements, quantified the occurrence of microplastics in different environmental compartments, urban flows, or sources (see, e.g., [4,5,10] for reviews). There are also studies that have estimated the contributions from different sources (e.g., [11–13]). These source-quantification estimates are subject to large uncertainties, and a comparison to measurement-based estimates is challenging. One challenge is that whereas sources are often estimated in mass, measurement-based studies often report the results

in number of microplastic particles. Another aspect that complicates a comparison is that all synthetic polymers may not be considered microplastics. Some synthetic polymers are not solid (such as polymer gels), or they can be soluble [14]. This may cause problems with regard to source estimates, for example, as it is not certain that polymers present in products are microplastics [15].

In a review about the possibilities for source allocation of microplastics in freshwater, Fahrenfeld et al. [16] suggest that analyses of morphology (e.g., fibres, microbeads, and fragments) and polymer types (e.g., polypropylene (PP), polyethylene (PE), and polyamide (PA)) can offer ways forward. The morphology aspect has been used to compare microplastic fibres in wastewater influent with the estimated emissions from laundry, with good conformity [17]. A similar source-tracking approach has also been used for contributions of heavy metals from stormwater and wastewater to a WWTP, where the conformity differed largely between the metals studied [18].

The aim of this study is to contribute to an improved understanding and verification of sources of microplastics in urban waters by comparing source estimates with measurement-based estimates. Specifically, estimates based on measurements at the inlet of the Sjölunda WWTP in Malmö, Sweden, are compared with estimates of microplastic sources for the wastewater and stormwater system connected to the WWTP. The purpose of this comparison is to see how well these approaches correspond in terms of microplastic types and to test the possibilities of using polymers for source tracking. Micro-FTIR imaging was used for the identification and estimation of the mass of the microplastics at the inlet of the WWTP. The source estimates are based on production and consumption in the study region and emission factors from studies of microplastic releases. The term "microplastic types" is used to distinguish between synthetic polymers and microplastics in this article to describe synthetic polymer types that are also solid, below 5 mm in size, and insoluble in water.

2. Methods

2.1. Study Approach

This study builds on the principles of substance flow analysis (SFA), which is often used to develop a systematic overview of substances within a defined system [19]. A key step of SFA is to define the system and the system components [20], which are defined in terms of space, time, and substances [19]. The geographical boundaries are set to the area connected to the Sjölunda WWTP, which consists of most of the city of Malmö in southernmost Sweden and four neighbouring municipalities. The sampling at the Sjölunda WWTP was carried out in 2017, and therefore, data from the year 2017 were, as far as possible, used for the source estimates as well.

Regarding substance definition, the estimation of sources was limited to microplastics that are possible to identify with the applied FTIR analysis [21]. FTIR analysis cannot detect microplastics from car tyres, and as the granulate used in artificial turfs often is made up of old car tyres, neither of these are included in this study. Further, the mass estimates in the measured samples are limited to plastic particles between 10 and 500 µm.

Urban wastewater pollution sources are primarily related to households or enterprises that are connected to the sewer system. In 2017, 337,000 persons were connected to the WWTP, and approximately 40 million m^3 of water was treated [22]. In combined sewer systems, wastewater and stormwater are collected in the same pipe, which means that diffuse pollution from urban activities, transported by stormwater, will be added to the wastewater reaching the WWTP. In 2017, the Sjölunda WWTP received 12 million m^3 inflow and infiltration [22]. The combined sewers cover an area of approximately 1990 hectares (ha). About 0.3 million m^3 did not reach the plant due to combined sewer overflows (CSOs).

2.2. Data Collection and Analysis

The data collection consisted of two parts: measurements at the WWTP and a literature review of studies that address microplastic sources. The literature was used for the identification and estimation of sources of the microplastics to the WWTP.

2.2.1. Measurements at the Inlet of the WWTP

At the Sjölunda WWTP, inlet samples were taken during two periods in 2017: March and December. In each period, a 24 h flow-proportional sample was taken (5–10 L) for four days. The sampling was performed in dry weather, and the samples were collected in 5 L blue cap bottles (the lid was Teflon coated) after the wastewater had passed a primary screen of 3 mm. A yearly load was derived by extrapolating the measured values from the sampling.

The wastewater samples from each period were pooled and thoroughly mixed, and two litres from each period were used for the subsequent analysis. The two litres were first filtered onto a 10 µm mesh stainless steel filter. The particles captured on the filter were then removed by 15 min of sonication in Milli-Q. Afterwards, a sodium dodecyl sulphate solution was added to the Milli-Q suspended particles to a final concentration of 1% and left for 48 h at 50 degree Celsius (rpm 100). From this point, the microplastic extraction protocol described by Liu et al. [7] was followed; however, no density separation was needed. In short, the extraction method includes enzymatic treatment with cellulolytic enzymes (Cellubrix L and Vicsozyme, both from Sigma-Aldrich) and then proteolytic enzymes (Alcalase, Novozymes) and a wet Fenton oxidation, all to remove other materials that are not microplastic particles. The remaining particulates were separated by wet sieving (Retsch AS 200 vibratory sieve shaker) into size fractions suitable for infrared analysis. Particles above 500 µm were manually evaluated under a stereomicroscope (Stereo Discovery V8, Zeiss, Oberkochen, Germany), and the potential plastic particles were analysed by ATR (Cary 630 FTIR Spectrometer with a germanium internal reflectance element, Agilent Technologies).

µFTIR imaging was used to examine the particles of size 10–500 µm, the applied settings and scanning method is described by Simon et al. [23], and the data were analysed using the software MPhunter, an automatic software developed by Aalborg University, Denmark, and Alfred Wegener Institute, Germany [7]. In MPhunter, mass estimations were made by measuring the major and minor dimensions, estimating the thickness, and multiplying the estimated volume by the particle density [7]. There was no recovery test performed for this microplastic extraction method, but the method applied is similar to that reported for water in Olesen et al. [8]; therefore, the recovery rate of 96% is thought to be valid for these measurements as well. Mass estimates were only performed for particles sized 10–500 µm, and therefore, only this size range is included in the comparison in this study. As mentioned above, particles above 500 µm were also analysed, but only a few particles were found [24].

To minimise contamination in the laboratory, equipment of plastic origin was avoided, and all equipment were cleaned three times with filtered Milli-Q water prior to use. Filters were muffled at 500 °C, and all samples were covered by aluminium foil whenever possible. Only cotton laboratory coats were used, and the work was done in a fume hood if possible. The air of the FTIR scanning room was also filtered continuously. In addition, three blanks were taken to assess potential contamination that might occur despite the precautions.

2.2.2. Identification of Sources and Estimations of Their Contribution

Swedish studies aimed at quantifying sources [12,25–27] were used as a starting point to identify potential sources. Furthermore, a literature review of scientific publications that quantified releases was performed to identify sources and obtain emission factors. Reports from authorities that presented quantitative information on emissions and/or polymer distribution were also used for the source estimations. Three groups of sources

were considered: households, enterprises, and stormwater added via the combined sewer systems (see Supplementary Materials Table S1).

Households

For households, emissions from laundry, personal care products (PCPs), cleaning products, paint, glue, and dust were estimated.

The emissions from laundry ($E_{Laundry}$) were calculated in the following way:

$$E_{Laundry} = T_{washed} \times \text{inhabitants} \times P_{textiles} \times EF \quad (1)$$

T_{washed} is the textiles washed in kg per capita per year. A person in Sweden washes approximately 1.5 cycles of laundry per week [28]. A typical washing machine load is estimated at 3–4 kg [29]. $P_{textiles}$ is the polymer distribution among textiles and was derived from the material distribution of imported and produced textiles in Sweden [30] (see Supplementary Materials). Materials that had less than 1% of the share were not included in the assessment.

EF is the emission factor for microplastic fibres during laundering and was set to 33–399 mg/kg for polyester [31–35]. The reports on emissions of synthetic fibres during wash vary greatly between studies. Studies that only sampled the first cycle were excluded since several studies indicate a decrease in fibre release after the first wash [36–38]. Further, studies reporting a total fibre release, including fibres of natural origin (e.g., wool and cotton), and studies that used natural and synthetic fabric blends, were also excluded. Priority was given to studies that simulated real conditions in terms of loads, temperature, use of detergent, and cycle duration. Moreover, studies that reported the results in mass (either using gravimetric methods or mass calculations) were chosen.

The emission factor for polyester was used as a proxy for other synthetic materials. Most of the studies on emissions from laundry have studied polyester, but there are a few studies that have investigated other materials. PP was shown to release fewer fibres than polyester in one study [32], but within the interval for polyester described above. Acrylic was shown to release fewer fibres than polyester [36], but in other studies it released more [37,38]. A reason for this discrepancy can be the large differences between polyester fabrics. Carney Almroth et al. [39] showed that a knitted polyester released fewer fibres than acrylic and PA, but a polyester fleece released more.

For PCPs, the amount of microbeads used in PCPs on the European market in 2015 [15] was converted to a per capita share and multiplied by the number of people connected to the Sjölunda WWTP. This was assumed to be equal to the release to the wastewater system. The contribution from cleaning products was derived from an adaptation of [15] to Swedish conditions [40]. The polymer distribution was obtained from the content in PCPs and cleaning products reported by [15] (see Table S2). For PCPs that are not rinsed off, a polymer distribution was not found, and therefore, it is not included in the final comparison.

Contributions from household paint and glue were estimated by adapting national estimations [40] to the inhabitants connected to the Sjölunda WWTP. How much of the polymer content in paint and glue that can be considered microplastics is uncertain [40], and therefore, these sources were not included in the final assessment.

There is limited information about microplastics in dust. Synthetic fibres deposited in dust have been estimated for indoor environments in France [41], and the findings from that study were used as a basis for the estimations of microplastics in dust for the catchment of Sjölunda WWTP using the following equation:

$$Fibres_{dep.catchment} = Fibres_{dep.} \times A_{connected} \quad (2)$$

where $Fibres_{dep.}$ is the yearly deposition of fibres in dust per m^2 (derived from [41]) and $A_{connected}$ is the average living area in Malmö in 2017 [42] multiplied by the number of persons connected to the Sjölunda WWTP. The average living area in Malmö was assumed to be representative of the neighbouring municipalities as well.

Not all microplastics present in dust will be added to the wastewater. According to Ewers et al. [43], 95% of household dust is removed by vacuuming wooden floors and 75% on linoleum floors. The final wet-washing step removes 20% of the remaining dust, which is the part that ends up in the wastewater. The contribution of from dust to the Sjölunda WWTP was estimated using the following equation:

$$E_{WWTP} = (Fibres_{dep.catchment.} \times 0.05 \text{ or } 0.25) \times 0.2 \quad (3)$$

where E_{WWTP} is the number of fibres that are released per year, based on the assumption that 5–25% of the dust remains after vacuum cleaning and 20% is captured when wet-washing [43]. $A_{connected}$ is the average living area in Malmö in 2017 [42] multiplied by the number of persons connected to Sjölunda WWTP.

Not all fibres in dust are of synthetic origin. Dris et al. [41] estimated that 30% of the fibres were synthetic. PP dominated the dust samples, but the distribution among polymers was not reported. However, the polymers found could be attributed to fabrics present at the test site. This indicates that the polymer distribution is context dependent and, hence, not directly transferable to the Malmö case. Instead, the polymer distribution from textiles on the Swedish market [30] was used. The number of particles was converted to mass by using average dimensions of polyester fibres (length 0.1–0.8 mm; diameter 0.011–0.026 mm) obtained from laundry studies [32,34,36,44] and the densities of the polymers.

Enterprises

Information about potential industrial sources to the Sjölunda WWTP was obtained from EnvoMap (https://www.gemit.se/products/envomap, accessed on 11 May 2021). This is the management tool that the municipal water and wastewater utility that operates the Sjölunda WWTP uses to store information about enterprises connected to the plant. Information was also gathered from environmental engineers at the water and wastewater utility, the municipal environmental department, and by directly contacting specific enterprises. For enterprises, the estimations included the release from laundries, lacquer used in manufacturing, mechanical workshops using hand soap, and landfill leachate (see Table S2).

The estimates for release from laundries were performed in the same way as for households (see Equation (1)) but excluding inhabitants and using loads washed at the laundries for T_{washed}. There were 12 commercial laundries that released water to the Sjölunda WWTP and an additional 3 laundries where a release was unknown. Among the connected laundries, one company only washes by hand, and its release can, therefore, be assumed to be smaller than that for enterprises using washing machines due to less attrition. Another company washes carpets in a largely circular system with on-site sedimentation treatment, and therefore, the contribution was assumed negligible. There were, thus, 10 laundries that can be considered to have significant releases of microplastics to the WWTP. For two of these, it was possible to get data on their activity in terms of yearly washing loads. The other eight laundries were mostly small; two of them only had a single washing machine, and it was not possible to obtain data on their wash loads. They were assumed to be of the same size as the smallest of the two where data were available, which is probably at the higher end of the actual situation.

No distribution among synthetic materials washed was found for commercial laundries, and the same polymer distribution as for households was used. Empirical studies in Sweden suggest that shirts and workwear, as well as textiles from hospitals, hotels, and restaurants (e.g., sheets, towels, and table linen), are common textiles laundered at laundries in Sweden [26,45]. These textile types primarily consist of cotton, followed by polyester and, to a lesser extent, PA, PE, acrylic, polyurethane (PU), and polyvinylchloride (PVC) [30], which is similar to the distribution used for households.

The enterprises reported usage of chemicals to EnvoMap, and these chemical lists were scanned for polymers that could be microplastics. The list of polymers confirmed as microplastics in products presented in [15] was used to guide this scanning and was

limited to polymers that could be detected with the analytical method. If the enterprises had not provided a list for 2017, the closest year before 2017 was used. Microbeads in PCPs that are rinsed off were prohibited in Sweden in 2018 [46] and might, therefore, have been phased out by the enterprises that year.

Four enterprises used hand soap containing PU and PE. For one of these products, the PU content was not registered, and the product could not be found at the retailer, which is why it was assumed that the content was the same as for the other products containing PU. A yearly use (in L/year) and percentage PU or PE in the product was derived from the chemical lists and calculated to mass. The chemical lists provided by the connected enterprises are based on self-reporting. There is a risk that PCPs are underreported since it cannot be taken for granted that these products are viewed as a product type that needs reporting.

There were two industries, one car repair workshop and one beverage packaging manufacturer, that reported use of epoxy. Both used epoxy in lacquer, but the workshop did not report the amount used, and it could, therefore, not be estimated. While microbeads used in hand soap can be assumed to, in total, be released into the wastewater, this is not the case for lacquer as it is not rinsed off. The fraction that is lost due to overspray during application and ends up on the ground [47] was assumed to enter wastewater.

There is also a closed landfill in the catchment area of the Sjölunda WWTP where the leachate partly enters on-site treatment and partly enters the Sjölunda WWTP. Approximately 30,000 m^3 per year of untreated leachate enters the Sjölunda WWTP. This water has not been tested for microplastics. Based on reports on microplastic content in untreated leachate in Norway, Finland, and Iceland [48] and China [49], the load to the Sjölunda WWTP would be 2–70 g/year based on [48] and 300–400 g/year based on [49] which can be considered negligible.

There are a few potential industry-related sources connected to the Sjölunda WWTP where a contribution of microplastics is uncertain or unlikely (see Table S1). For the pharmaceutical production plant connected to the WWTP, it cannot be determined whether the polymer used in production can be considered microplastics. Further, considering that a spill will only occur if there is a malfunctioning in the process and that the plant has on-site treatment, the potential release from this source was considered negligible. Similarly, factories producing PCPs may be a source if they add microplastics to the products and if there is a malfunction in production or in the internal treatment, but this was also considered too uncertain to estimate on a yearly basis.

There is a plastic industry with process-related water connected to the Sjölunda WWTP. The potential release was assumed negligible as there is a pre-treatment facility that captures particles larger than 10 µm. Further, although the water had not been tested for microplastics, it was tested for monomers, which were not detected. There was also an enterprise that washed tyres with plastic beads as abrasive media, but there is limited knowledge about this type of source. The enterprise has an on-site device with the primary function of separating oil and heavy metals from the water before release to the WWTP, but how it affects microplastics is not known. Fibre release from swimwear during use in public baths has been suggested as a potential source of microplastics [26]. Approximately 3 g of solid material is estimated to be released from a bathing person [50]. However, this includes not only textile fibres from swimwear but also hair, skin, and soap residues and the proportions between them has, to the authors knowledge, not been determined. It is further uncertain how much of the solid material escapes the bathing facilities.

Stormwater

No detailed information about land use in the area with combined sewers was found, but these are often present in the old parts of cities [51]. Information about land use in Malmö was derived from Statistics Sweden, and the distribution between land use categories was assumed to be representative for the area that is connected to the WWTP.

The following equation was used to estimate the proportion of impervious area and road (land use types) for the area with combined sewers:

$$A_{\text{Land use types combined sewers}} = (A_{\text{Land use types Malmö}} / A_{\text{tot-Malmö}}) \times A_{\text{combined sewers}} \quad (4)$$

where $A_{\text{tot-Malmö}}$ is the total land area in Malmö and $A_{\text{combined sewers}}$ is the area of combined sewers connected to the Sjölunda WWTP.

The sources of microplastics in stormwater that were considered in this study were littering from cigarette filters, paint from road markings, and wear and removal of paint from outdoor surfaces.

The contribution from cigarette filters was estimated by multiplying the number of cigarette filters present in urban areas per m^2 by the amount of impervious area connected to the Sjölunda WWTP from Equation (4) and the filter weight. Even if cigarette filters are also littered on impervious surfaces, it was assumed that they are only washed away and, hence, enter stormwater from impervious surfaces. It was further assumed that all cigarette filters are washed away within a year. Cigarette filters are the most common litter in urban areas in Sweden, but other plastic materials are littered as well. However, that contribution was deemed too uncertain to estimate.

The contribution from road markings was estimated in a similar manner as that of cigarette filters. The estimations of amount of road in the catchment area of the Sjölunda WWTP from Equation (4) was multiplied by the amount of paint used on roads in Sweden [12]. The yearly application was assumed to be equal to release. Verschoor et al. [52] suggest that a large share of paint from buildings ends up in soil and not in water. This led to that the estimated load from road markings being treated in two different ways, assuming that either it all ended up in the stormwater or most of it ended up in the soil.

Wear of painted surfaces and removal of the old paint layer were estimated in two different ways: one based on painted surfaces [52] and one based on per capita paint consumption in Sweden [12], where the first approach showed slightly lower values than the second approach (see Table S2). As polymer distribution from paint was not possible to obtain, these estimates were not used in the final assessment.

Atmospheric deposition contributes with microplastics to the urban area [53,54] and may reach the WWTP from the combined sewer system. Sampling of microplastics from atmospheric fallout has been conducted in Malmö [55]. Styrene-butadiene rubber, fibres, and fragments were reported, with the rubber being most abundant. The samples were only visually analysed, which is why the distribution among polymers is not known. In central London, fibres accounted for over 90% of the deposition [54]. Acrylic was the most common fibre type. Due to the seemingly contextual aspects of atmospheric deposition, as well as the impact of wind speed and direction, and the risk of double-counting sources, atmospheric deposition has not been included in the source estimations.

3. Results and Discussion

3.1. Comparison of Total Load

The measurement-based load at the inlet was 4.2 tonnes/year, while the total load of the source estimations was 1.9–14 tonnes/year when only the sources where the distribution among polymers was known and only the polymers that can be detected with the applied analytical method were included. The measurement-based estimate is, thus, at the lower end of the source estimates. In this study, release to the wastewater system is compared to the WWTP inlet. Microplastics may also be lost on the way from the source to the WWTP. For the Sjölunda WWTP, 0.3 million m^3 of water did not reach the plant in 2017 due to CSOs, corresponding to approximately 15–110 kg/year of the total estimated release of microplastics. Further, the amount of microplastics captured in, for example, pipe sediments is not known.

According to the source estimates, the largest contribution of microplastic comes from households, followed by that from stormwater and enterprises (Figure 1). In terms of the total flow of water to the plant in 2017, 69% was wastewater (of which 74% was from

households) and the rest was inflow and infiltration [22]. In contrast to the findings in this study, Tumlin and Bertholds [56] found that the majority of the microplastics at the inlet originated from sources that were not households or household-related enterprises. One influencing factor for this discrepancy can be that at this WWTP, over 50% of the water at the inlet is inflow and infiltration, while it was around 30% for the Sjölunda WWTP in 2017. Another explanation is that the stormwater sources in the source estimates for this study are underestimated, as there were several sources that were not possible to quantify.

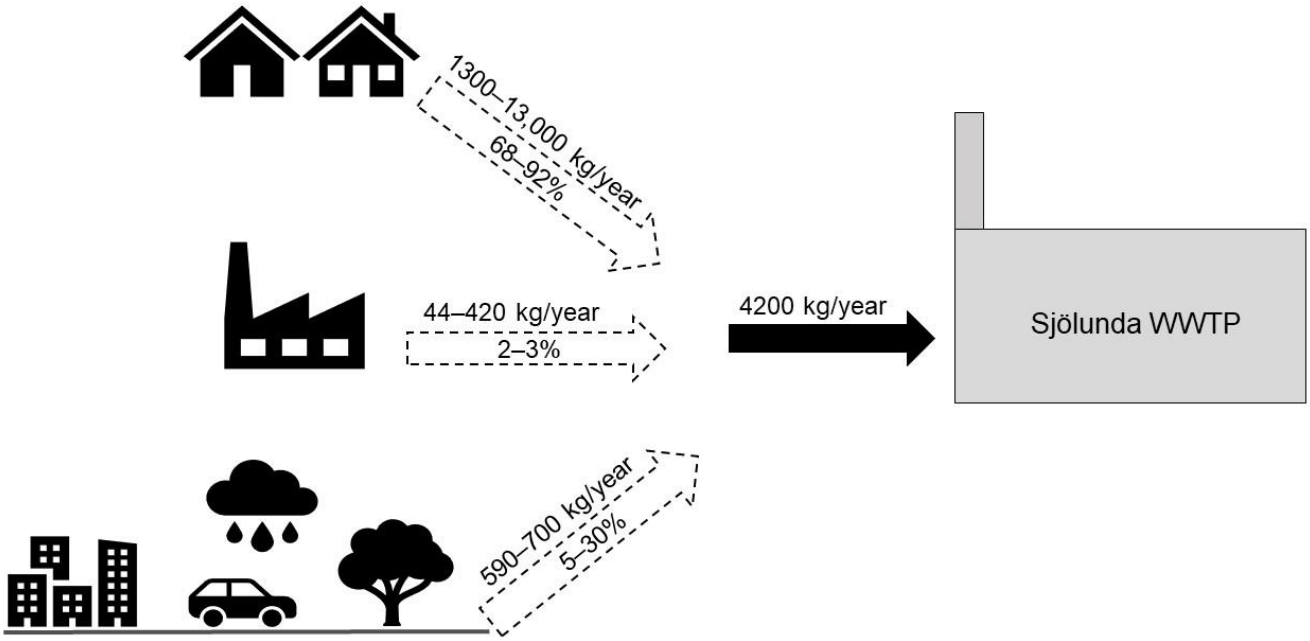

Figure 1. Overview of the contribution from the three main groups of sources to the wastewater treatment plant (WWTP) (households, enterprises/industries, and stormwater) and the measurement-based load at the inlet of the Sjölunda WWTP. The dashed arrows represent the source estimates, and the filled arrow shows the measurement-based load (rounded numbers).

Enterprises contribute with less microplastics than their share of the wastewater coming to the Sjölunda WWTP. Some of the potential sources from enterprises were considered too uncertain to estimate, and the self-reporting system may underestimate the contribution from this group. Furthermore, potential releases of sanitary water from enterprises were not assessed. Nevertheless, the relative importance of households as emitters is increasing for several substances, which is likely an effect of the many efforts that have been taken to control emissions from industries in Sweden [57].

Laundry was the largest source of microplastics and was responsible for 40–87% of the total load to the Sjölunda WWTP (see Supplementary Materials Table S3). Although laundry is also an industrial activity, households were estimated to be responsible for 96–97% of the total contribution from laundry. The large interval for households in Figure 1 can be attributed to the large differences in reported emissions from laundry, which resulted in a large range for the emission factor (33–399 mg/kg). The significance of laundry as a source has been highlighted by other authors [17,58]. As the treatment processes at the WWTP do not seem to retain fibres sufficiently [10], the emissions from laundry may need to be handled upstream. Cesa et al. [37] suggest that a filter, three pre-washings, and detergent will reduce the fibre load by over 50%. However, the impact of these measures is uncertain. Other studies have reported an increase in fibre release when detergent is used [32,34,59] and that subsequent washings did not impact the fibre release [34].

The second largest source was PCPs that are rinsed off, which contributed with approximately 500 kg/year. PCPs can be used in both households and enterprises, but the large majority came from households in this study. As a ban on the use of microbeads in products that are rinsed off was introduced in 2018, this source should have been

significantly reduced. However, there may still be emissions from the PCPs that are not regulated. For these products, the amount that will end up in wastewater is more uncertain as the microplastics can also be added to the solid waste.

Littered cigarette filters that will end up at the Sjölunda WWTP via the combined sewers also contributed with approximately 500 kg/year, and this was the largest estimated source for stormwater. However, it should be noted that the FTIR analysis does not detect car tyre particles, and therefore, sources that have previously been identified as large in stormwater (tyre wear and artificial turfs) [12] were not included in this analysis.

The comparison of total load can give an indication into how well the overall estimates match, but to give increased insight into the distribution among sources, a more detailed comparison is needed. In the next section, the different types of microplastics in source estimates and measurement-based estimates are compared.

3.2. Comparison of Microplastic Types

In total, 11 microplastic types were discovered when investigating the theoretical contribution to the Sjölunda WWTP. For 8 of the microplastic types, the measurement-based loads were within the interval for the source estimates (Table 1). For epoxy, PA, PE, and PVC, the measurement-based loads were at the lower end of the source estimates, and for polyester, PP, and PU, the measurement-based loads were closer to the middle of the intervals.

Table 1. Loads of microplastic types based on the source estimates to the wastewater treatment plant (WWTP) and the yearly loads based on the extrapolation of measured values at the inlet of the Sjölunda WWTP (rounded numbers). EVA = ethylene-vinyl acetate, PA = polyamide, PE = polyethylene, PLA = polylactic acid, PP = polypropylene, PU = polyurethane, PVC = polyvinylchloride.

Microplastic Type	Source Estimates (kg/year)	Measurement-Based Load (kg/year)
Acrylic	110–1780	90
Cellulose acetate	500	-
Epoxy	10–40	10
EVA	50–110	10
PA	120–1430	270
PE	230–1090	250
PLA	1	-
Polyester	240–4110	1680
PP	130–2210	810
PU	460–1770	880
PVC	80–1350	90

Polyester was the microplastic type with the largest share at the inlet, followed by PU and PP (Table 1). PE and PP are often observed at the inlet of WWTPs and were found in almost all studies in the review by Ngo et al. [10]. However, the dominating type at the inlet varies largely between different studies. Acrylate was the most common type at Danish WWTPs [23], while alkyd was most common at a WWTP in Scotland [60], and PA at a WWTP in China [61]. Similar to this study, polyester dominated at a Finnish WWTP [62]. There are likely both general sources and sources that depend on the context of the specific WWTP. While household sources probably are similar to other areas with a similar standard, the number and types of connected enterprises can differ. Further, stormwater sources are likely similar for most WWTPs in cities, but their contribution is affected by context-specific aspects, for example, the extent of combined sewers and land use in the catchment.

For acrylic and EVA, the measurement-based estimations were smaller than the calculated interval, but the differences were small. Regarding acrylic, this is surprising as there are indications that the source estimations are underestimated. Laundry and dust were the two identified sources of acrylic. Polyester was used as a proxy for fibre release, but there

are indications that acrylic sheds more fibres than polyester [37,38]. In addition, acrylic is used in paints, which were not included in the source estimations.

Polylactic acid (PLA) and cellulose acetate were not found in the influent. According to the source estimates, PLA would contribute with approximately 1 kg/year to the WWTP. This is a small quantity compared to the other microplastic types, which is why it is not surprising that it was not measured. Cellulose acetate was estimated to contribute with approximately 500 kg/year to the WWTP. A possible explanation for the disparity is that the cigarette filters may not be easily fragmented in urban areas and can, therefore, be captured at, for example, the primary screen at the WWTP. Previous research has shown a slow degradation of cigarette filters, especially in the absence of soil [63]. The primary screen at the Sjölunda WWTP is 3 mm [22]. Smaller items can also stick to other materials captured on the screen.

Cellulose acetate has not been reported in other studies of inlets at WWTPs either [10,23,60] but has been measured in small quantities in household effluent [56]. Cellulose acetate is, to a small extent, used in PCPs. For the Sjölunda WWTP, the contribution from this source was estimated to be less than a kg per year.

Most of the cellulose acetate in the Sjölunda WWTP was estimated to come from the littering of cigarette filters in the urban area. Laboratory experiments have shown cigarette filters to release approximately 100 fibres/day (<0.2 mm) in water when subject to agitation and UV light [64]. However, cellulose acetate does not seem to occur in stormwater [7] or in surface water [65,66]. Cellulose acetate has a higher density than water and can be expected to sink, but it does not seem prevalent in stormwater sediments [8,21] or in marine [67] or freshwater [68] sediments. It has been reported in small quantities in sewage sludge [56].

If cellulose acetate is excluded from the analysis, there is an agreement between the measured and the estimated source proportions, meaning that the microplastic types that had a large share at the inlet also had a large share in the source estimates (Figure 2). Five microplastic types stand for over 90% of the microplastics in the influent. These five microplastic types stood for 76–83% of the source estimates. The microplastic type that primarily differed (when cellulose acetate was excluded) was acrylic.

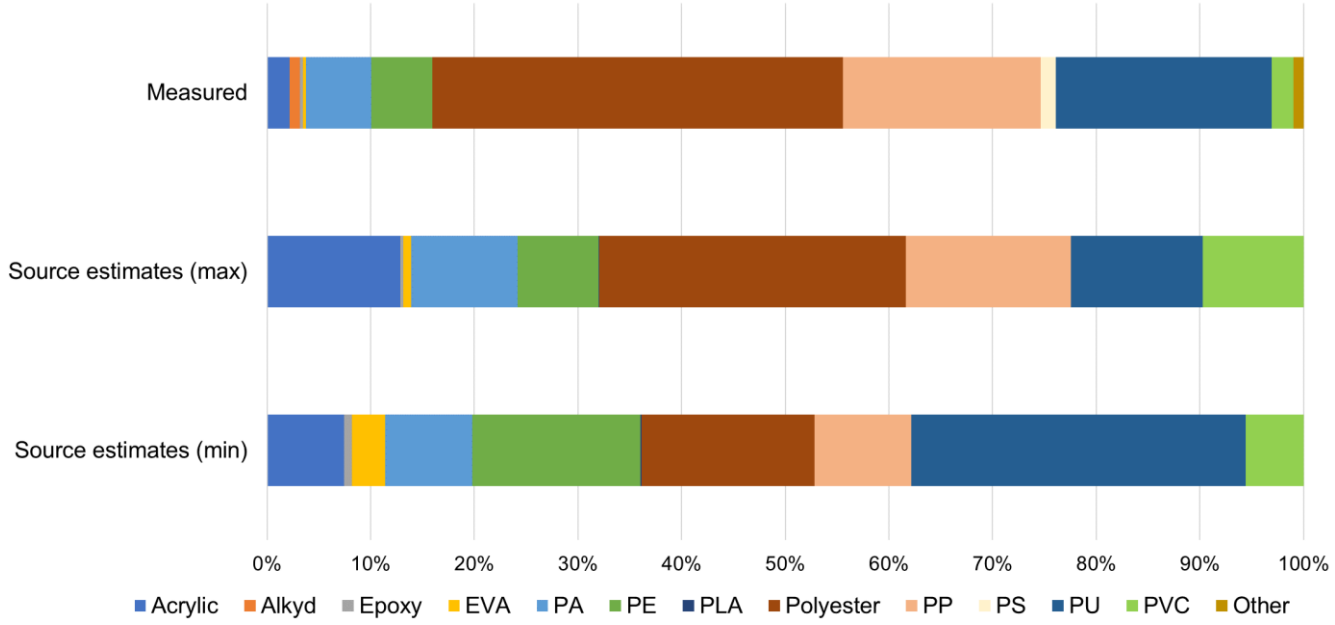

Figure 2. The distribution in percentage between microplastic types in the source estimates (minimum (min) and maximum (max) values) and the measurement-based load at the inlet of the Sjölunda wastewater treatment plant, excluding cellulose acetate. EVA = ethylene-vinyl acetate, PA = polyamide, PE = polyethylene, PLA = polylactic acid, PP = polypropylene, polystyrene = PS, PU = polyurethane, PVC = polyvinylchloride.

Polyester, which had the largest share at the inlet, came almost only (95 to >99%) from laundry in the source estimates (see Supplementary Materials Table S4). It was the same for PP. PE, PU, and PA had more sources. For these, laundry was also the largest source, but for PE PCPs also had a large contribution. For PU, cleaning products also contributed, in addition to laundry and PCPs. For PA, road marking also contributed in addition to laundry, while PCPs had a limited share. All five microplastic types had a small contribution from dust.

Two types of microplastics, alkyd and polystyrene (PS), contributed with more than 1% of the measurement-based yearly load but were not assessed in the source estimates. Alkyd is often used in paints [69], but paint was a source where polymer distribution was not known and, hence, was not included. Alkyd can also originate from painted surfaces at the WWTP [70]. PS is often used in food containers and utensils [1], and therefore, littering can be a source. Cigarette filters were the only type of litter that was included in this study. These results indicate that more knowledge is needed about these sources.

There are still many uncertainties related to microplastics, related both to measurements and source estimates. The measurement-based estimates can be underestimating the load of microplastics at inlet due to limitations with respect to sampling and analysis. The rather small sampling volume and time (a few days in March and December) might not be representative for the whole year since there are indications of seasonal variations of microplastics in influent [71]. The sampling was also performed in dry weather, which can impact the load of microplastics from stormwater. Moreover, not all microplastics present in the sample may be analysed. A 96% recovery was estimated for this study, and mass estimates were only performed on particles in the range 10–500 µm.

The many uncertainties related to sources of microplastics resulted in large intervals for the source estimates. Particularly, this study reveals the large uncertainties related to emission factors for textile fibres, which led to large intervals for laundry. Further, the limited understanding of how much of the polymer content in a product can be defined as microplastics meant that contribution from the rinsing of paint brushes and glue in households could not be estimated. There is also a lack of knowledge about the polymer types used in PCPs that are not rinsed off, as well as in outdoor paint; therefore, these sources could not be included in the comparison. Further, although littering and atmospheric deposition might be large sources in stormwater, these were challenging to estimate. The analytical method could also not detect microplastics from car tyres, so the impact from road traffic could not be included in the analysis. The potential impact of enterprises on microplastic emissions is also uncertain. As there is no legislation on release of microplastics and no standard method for sampling and analysis, there are no limit values and no systematised control established. Further, even if plastic is used by the enterprise, the impact this will have on the wastewater is dependent on how the plastic is used and the presence and function of on-site treatment.

4. Conclusions

In this study, a theoretical contribution of microplastic types from various sources to the Sjölunda WWTP, Malmö, Sweden, was compared with measurement-based estimates at the WWTP, and the results show that there is an agreement between the two approaches. In general, microplastic types with a large share in the measurement-based estimates also had a large share in the source estimations, with a few exceptions. Cellulose acetate had a significant theoretical contribution (approximately 500 kg/year), but no cellulose acetate was detected at the inlet. This indicates that there is a need to know more about the fate of cigarette filters. Alkyd and PS, on the other hand, were measured but not identified among the sources. This indicates that there is a need to understand more about the sources that may give rise to alkyd and PS, such as fragmentation and transport routes of urban litter, as well as the polymers used and microplastics derived from the use of paint. Polyester had the largest share at the inlet (40%), which was estimated to almost only come from laundry (95 to >99%), indicating that laundry is a significant source of microplastics in

wastewater. Similar to previous source-quantification studies, the large knowledge gaps resulted in large intervals. The comparison of microplastic types used in this study can give an indication into which part of the large interval is most likely. Investigating more WWTPs with different characteristics, such as different types of industries or completely separated pipe systems, as well as adding the morphology aspect, will further advance the understanding of sources of microplastics in wastewater.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/su13105404/s1, Table S1: Overview of sources of microplastics in wastewater and stormwater. Table S2: Estimated releases from the identified sources and polymer distributions. Table S3: Summary of source estimates divided by source type. Table S4: Summary of source estimates divided by microplastic types.

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