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Microplastic Particle Loads Discharged by Swiss Wastewater Treatment Plants

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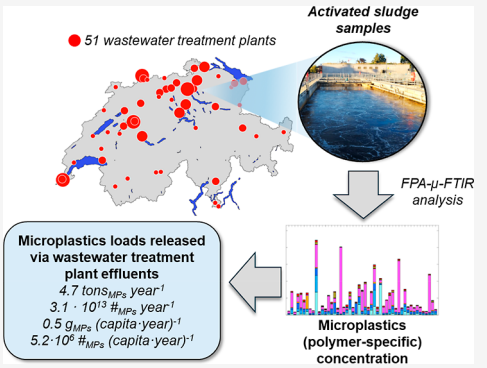
ABSTRACT: The contribution of wastewater treatment plants (WWTP) to the loads of microplastic particles (MP) entering surface waters is debated intensively. We, therefore, quantified MP (>20 μm) in activated sludge from 51 Swiss WWTP to estimate the MP loads discharged through WWTP into surface waters. MP were identified by Fourier transform infrared spectroscopy. We used surrogate standards as a quality assurance/quality control measure and conducted an uncertainty assessment across the whole analytical chain. The MP number concentrations in sludge per gram total suspended solids (TSS) ranged from 900 to 22,000 MP $gTSS^{-1}$, resulting in calculated mass concentrations between 10 and 1700 μg MP $gTSS^{-1}$. The MP number size distribution was dominated by MP < 100 μm and was well-described by a power law function. Assuming similar removal efficiencies for TSS and MP during the activated sludge process in combination with the MP concentrations in the activated sludge and the TSS contents in WWTP effluents allowed us to assess the loads of MP released by WWTP to surface waters. The loads across Switzerland were estimated to be 3.1×10^{13} MP yr^{-1} corresponding to 4.7 t MP yr^{-1}, resulting in per capita emissions of 5.2×10^{6} MP (capita $yr)^{-1}$ corresponding to 0.5 g MP (capita $yr)^{-1}$.



KEYWORDS: *QA/QC, sludge, FTIR, surrogate standards*

1. INTRODUCTION

Microplastics (MPs) are defined as plastic particles with sizes ranging from 1 μm to 1 or 5 mm, either manufactured in this size range or formed through the degradation and fragmentation of larger plastic items.[1,2] Due to the large mass of plastics produced (431 Mt in 2024[3]), their widespread use, and insufficient waste management, MPs have been detected in nearly all environmental compartments, with still poorly understood impacts on ecosystems.[4]

A frequently discussed pathway for MPs into the environment is through the effluent of wastewater treatment plants (WWTPs).[5−9] Most of the MPs emitted by households, by industries, and from road runoff (combined sewer systems) are transported along sewer pipes and eventually reach a WWTP. During treatment, approximately 95 to 99.9% of MPs in the influent wastewater are removed from the wastewater stream and accumulate in the sewage sludge.[8−12] A small fraction of the MPs, however, pass through WWTPs and are discharged into surface waters.

Several studies therefore proposed WWTPs as important contributors of MPs to surface waters.[6,13−15] However, reported MP concentrations in WWTP effluents vary by up to 6 orders of magnitude,[16] hampering a robust assessment of the WWTP-derived MP loads in surface waters. The large variability of MPs in WWTP effluents likely reflects both actual variations in MP concentration and methodological and analytical uncertainties in MP quantification. In support of the latter, harmonized sampling procedures and standardized sample processing protocols for MP analysis are largely lacking, and quality assurance/quality control (QA/QC) measures are still poorly established in the field of MP research.[16,17]

Accurate quantification of MPs in the wastewater effluent is critical to compare MP inputs to surface waters through WWTP effluents with diffuse sources, such as atmospheric deposition.[18,19] High diffuse inputs are supported by mass-flow analysis of MPs in Switzerland, which estimated comparable inputs of MPs into surface waters from atmospheric deposition and WWTP effluents.[20] However, quantitative, experimental data on MP concentrations in different natural and engineered systems are required to assess to what extent results from modeling studies match those of experimental data.

Most commonly, MPs in WWTP effluents are collected by filtering large volumes, often exceeding 500 L.[16] This

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procedure is logistically demanding and prone to frequent filter clogging, which increases the risk of sample contamination and MP loss during filter exchange. A promising and much simpler alternative to direct effluent filtration is to estimate the concentration of MPs in the WWTP effluent based on the MP contents of activated sludge samples and the total suspended solids (TSS) in the effluent. This makes use of the previously established fact that during biological treatment, MPs efficiently form heteroagglomerates with sludge flocs, and a fraction of these flocs pass through the WWTP and are released into the receiving waters. As a result, the ratio between MPs and TSS (e.g., sludge flocs) in activated sludge is similar to the respective ratio in the WWTP effluent.[21,22] Emissions of MPs from the WWTP effluent can, thus, be derived from the TSS concentrations in the WWTP effluent, which is routinely measured, in combination with the experimentally determined concentration of MPs in the activated sludge.

This approach offers two major advantages compared to the filtration of the effluent water: (1) activated sludge reactors are continuously stirred systems, resulting in an even distribution of MPs in the activated sludge. Grab-sampling sludge, therefore, integrates the variability in influent MP concentrations, whereas capturing variability in TSS and MP concentrations in WWTP effluents requires flow-proportional composite sampling. (2) MP concentrations in activated sludge are approximately 600 times higher than in the WWTP effluent, as estimated from the approximately 600 times higher TSS content in sludge than in the effluent (e.g., 5 mg TSS L^{-1} and 3 g TSS L^{-1}, respectively). Therefore, considerably smaller sample volumes are needed, which substantially reduces logistic demands for sampling and simplifies sample processing.

The goal of this work was, therefore, to determine MP contents in activated sludge samples and to estimate the loads of MPs (>20 μm) discharged by WWTP effluents in Switzerland. For that purpose, we quantified the MP contents in activated sludge samples from 51 WWTPs using focal plane array−μ-Fourier Transform Infrared (FPA-μ-FTIR) spectroscopy. Based on these data, in combination with the routinely monitored TSS contents in WWTP effluents and the volumetric flow rates of the individual WWTP, and assuming a similar ratio between MPs and TSS in the activated sludge and in the WWTP effluent, we estimated average number- and mass-based emissions of MPs from WWTP effluents. These emissions were then scaled to Switzerland. The rigorous use of surrogate standards allowed us to derive robust uncertainty estimates spanning the entire analytical chain.

2. MATERIALS AND METHODS

2.1. Reagents and Microplastic Standards

Hydrogen peroxide (H_2O_2 35%, Roth, order number: 9683.5), iron sulfate ($FeSO_4{\cdot}7H_2O$, Roth, Pro analysis, 2619776), sodium polytungstate (SPT; ThermoScientific, 652154-12-6), and protocatechuic acid (Sigma, 37580−25g-F) were used to perform the Fenton reaction and isolate the MPs from the sludge matrix. Rinsing of all reaction vessels and sampling tools before and after the analysis was performed with ethanol (Reuss Chemie, RC-FSP-018), acetone (Reuss Chemie, RC-ACT-018), and deionized (DI) water (Arium Pro, Sartorius). The DI water, ethanol, and the SPT solution were always filtered through a 0.22 μm pore-size polyvinylidene fluoride membrane (Durapore) before use. Stainless-steel meshes (pore size 15 μm and 47 mm diameter, Haver & Boecker OHG, Germany) and aluminum oxide filters (Anodisc, pore size 0.2 μm and 25 mm diameter, Whatman plc, UK) were used to separate the MPs from the liquid matrices. In addition, red polyethylene (PE) spheres (53−63 μm; Cospheric, CA, USA) were used for QA/QC.

2.2. Wastewater Treatment Plants

Activated sludge samples from 51 WWTP across Switzerland were collected (Figure S1). The WWTPs received variable shares of industrial and municipal wastewater and were connected to different types of sewer networks (i.e., combined and separate). The individual WWTP served between 3000 and 470,000 inhabitants, together accounting for 44% of the Swiss population (9,051,029 inhabitants as of 31.12.2024). All WWTPs included a biological treatment step, consisting of either the activated sludge process (n = 47), biofiltration units (n = 2), or a combination of both (n = 2). Notably, five of these WWTPs were equipped with a moving bed bioreactor (MBBR) as an additional treatment step to their activated sludge or biofiltration. The surface area of the WWTP catchments corresponds to 21% of the total area (water and land) of Switzerland (8569 km^2; Figure S1). Data about the catchments, sewer network, and effluent parameters of the individual WWTP included in this study were provided by the WWTP operators or the public authorities for the year 2024 and are summarized in Table S1.

2.3. Sample Collection

The sampling campaign was conducted over an 8-month period (April 2024 to December 2024). Activated sludge samples were collected in 1 L glass Schott bottles rinsed thoroughly with DI water and acetone prior to sample collection. To prevent contamination, aluminum foil was placed between the bottle neck and the plastic lid. Samples were taken from the aeration tank at every individual WWTP, where the sludge is continuously stirred and aerated, using a long sampling rod made of steel or plastic provided by the operator. In WWTPs employing biofiltration processes, backwash water from the biofilters was collected instead of activated sludge. All samples were collected by the same person following the procedure outlined above. The (filled) glass bottles were transported in a cooling box back to the laboratory, where sample processing started immediately upon arrival.

2.4. Sample Processing

The sludge in the glass bottles was first manually shaken to ensure representative subsampling. For each WWTP, a subsample containing ~200 mg of TSS (about 25−333 mL of sludge) was transferred to a 600 mL glass beaker for further treatment. While the subsample volume needed was estimated based on the TSS content of the sludge provided by the WWTP operators, the actual TSS concentration was determined in the laboratory to calculate the exact subsampled mass of TSS (Table S2). In addition, for one WWTP, three subsamples each containing 200 mg of TSS were collected from the same 1L bottle to evaluate the homogeneity of the sample.

The sample processing was adapted from Philipp et al. (2022).[23] First, surrogate standards were added to the 600 mL beakers containing the activated sludge subsample (Section 2.6). The organic matter contained in the sludge was digested using the Fenton reaction based on 30 mL of H_2O_2 (35%), 1 mL of $FeSO_4$ (2 mM), and 1 mL of protocatechuic acid (2 mM). The 600 mL glass beaker was incubated in a horizontal shaker at 40 °C for 24 h, after which an additional 20 mL of H_2O_2 was added, and the reaction was continued for another 48 h, totaling 3 days. The suspension was filtered through a 15 μm stainless-steel mesh to retain MPs as well as other residual particles. To separate the MP from inorganic particles, 30 mL of SPT solution (ρ = 1.6 g cm^{-3}) was added on the stainless-steel mesh, and the mesh was briefly sonicated to resuspend the particles. The suspension was transferred to a polypropylene (PP) centrifuge tube and centrifuged at 2900g for 10 min. The supernatant was filtered through the same 15 μm steel mesh, resuspended by sonication, and filtered onto a 25 mm Anodisc filter. The whole filter was imaged with an automated light microscope (VHX-7000, Keyence, Japan) before a subsample was investigated using infrared spectroscopy. More details about the procedure are provided in Section S3.

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2.5. Focal Plane Array–μ-Fourier Transform Infrared Spectroscopy

Hyperspectral FTIR data were recorded using a μ-FTIR imaging system consisting of a Cary 670 FTIR spectrometer coupled to a Cary 610 IR microscope (Agilent Technologies) and equipped with a FPA detector (64 × 64 elements) for automated large-area measurements. All analyses were performed using a 15× IR objective in the transmission mode, covering a wavenumber range of 3900−1250 cm^{-1} with a spectral resolution of 8 cm^{-1}. Spectra were acquired by coadding 24 scans per FPA frame, while the background (blank Anodisc filter) was measured using 64 scans. The pixel resolution of the FPA was 5.5 μm, resulting in a measurement area of 352 μm × 352 μm per FPA frame.

A software developed in-house (described in Ashta et al. (2026)[24]) was used to automate the FPA-μ-FTIR measurements. The software enabled random selection and analysis of several nonoverlapping subsections of the filter surface, each subsection consisting of 8 × 8 FPA frames (=7.9 mm^2 per subsection). We prompted the software to select between 9 and 11 different subsections, corresponding to a total area of 28−34% of the filter surface. An individual height (z-coordinate) of the FPA-μ-FTIR stage was manually assigned to each filter subsection, and the stage was then automatically adjusted during the analysis, allowing optimal focal conditions for each subsection.

Infrared absorption spectra of individual MP particles were identified from the hyperspectral data using the software "MicroPlastics Finder" and classified into 21 polymer categories.[25] Operational parameters for the "MicroPlastics Finder" were selected based on expert knowledge, minimizing false positives and false negatives, as described in Ashta et al. (2026).[24] The selected values for the individual parameters are listed in Table S3. A minimum particle size of 11 pixels (equivalent to 333 μm^2 or to a 20 μm equivalent circular area diameter (ECD)) was applied to ensure sufficient spectral quality.

The individual MPs detected within the selected filter subsections were summed up to obtain the total number of MPs within the subsections ($n_{MP,sub}$). The total counts of all particles (plastic and nonplastic) were derived from (light) microscopy images on the whole filter (n_{tot}) and in the individual subsections (n_{sub}) (Table S4). To estimate the total number of MPs on the whole filter ($n_{MP,tot}$), we calculated the ratio of the number of MPs to the total number of particles within the subsections ($n_{MP,sub}/n_{sub}$) and multiplied this ratio by the total number of particles of the entire filter $\left(\frac{n_{MP,sub}}{n_{sub}} \cdot n_{tot}\right)$. This method of extrapolation was selected to account for the nonuniform particle distribution across the filter surface, and the associated extrapolation uncertainty was calculated following the methodology described in Schwaferts et al. (2021).[26]

2.6. Surrogate Standards and Recoveries

To estimate MP losses during the sample processing, red PE spheres of 53−63 μm diameter were used as surrogate standards, following the procedure described in Philipp et al. (2022).[23] In brief, between 100 and 250 red PE surrogate standard particles were deposited on a glass slide and counted using an automated light microscope (VHX-7000, Keyence, Japan). Thereafter, the surrogate standards were rinsed with ∼5 mL of ethanol into each individual 600 mL glass beaker containing a sludge sample, before any sample processing steps were initiated. Any surrogate standard particles remaining on the slide after rinsing were subtracted from the initial count.

Surrogate standard particles were quantified again after the entire sample treatment chain on Anodisc filters using both optical microscopy and FPA-μ-FTIR spectroscopy. The (sample-specific) optical recovery (R^{opt}) and the FPA-μ-FTIR recovery (R^{IR}) are defined according to eqs 1 and 2, respectively.

$$R^{opt} = \frac{n_{tot}^{opt}}{n_{spiked}} \tag{1}$$

$$R^{IR} = \frac{n_{subsample}^{IR}}{n_{subsample}^{opt}} \tag{2}$$

where n_{spiked} is the number of surrogate standard particles spiked to the samples and n_{tot}^{opt} and $n_{subsample}^{opt}$ are the numbers of surrogate standard particles recovered using the optical microscope on the whole filter and in the filter subsections, respectively. The number of surrogate standards identified in filter subsections based on FPA-μ-FTIR analysis is represented by $n_{subsample}^{IR}$. Note that the surrogate standards identified were eventually subtracted from the number of PE particles detected in the sludge samples. Further details on the identification of the surrogate standards based on light microscopy and IR images, as well as the number of surrogate standards spiked and recovered, are provided in the Supporting Information (Section S4 and Table S5). Importantly, we reported the recoveries but did not apply any corrections to the number of detected MPs.

2.7. Conversion of Particle Area to Particle Volume and Particle Mass

The image area of each MP particle obtained from MicroPlastics Finder was converted into a corresponding particle volume V, assuming ellipsoidal particle geometry and using eq 3 (Simon et al. (2018))[8]

$$V = \frac{4}{3}\pi \cdot \frac{M}{2} \cdot \frac{m}{2} \cdot \left(\frac{m}{M}\right)_{median} \cdot \frac{m}{2} \tag{3}$$

where M and m are the long and short axes of the MP projected areas, respectively, and $\left(\frac{m}{M}\right)_{median}$ is the median ratio of m and M of all MPs detected. The mass of individual MP was then calculated using the density of polymers found in the literature (Table S3).

2.8. Uncertainty Estimations

Sample collection and preparation and data generation and processing are associated with individual uncertainties that contribute to the total uncertainty of the results. Following the approach outlined by Ashta et al. (2026),[24] we derived the standard uncertainties regarding the spectral assignment (6%), the variability in surrogate standards recovery (standard deviation of R^{opt}, 13%), and the subsampling uncertainty (sample-specific, on average 11%) from our data set. In addition, we used an uncertainty linked to the reproducibility of FTIR measurements of 4% and an uncertainty associated with the uneven surface of the Anodisc filters of 24%, as suggested by Ashta et al. (2026).[24] These individual uncertainties were assumed to be independent from each other and random and were propagated to obtain the total uncertainty associated with the quantification of the number of MPs in respective samples (eq 4)

$$U = 2 \cdot u_{tot} = 2 \cdot \sqrt{\sum_{i=1}^{n} u_i^2} \tag{4}$$

where u_i and u_{tot} are the individual and total uncertainty on the number of particles, respectively. The total uncertainty u_{tot} was multiplied by a coverage factor of 2 (confidence level 95.5%) to obtain the expanded measurement uncertainty U. Furthermore, a systematic uncertainty of 50% was assigned for the conversion of the projected area of individual MPs in their respective volume or mass.[24] More details on the uncertainty quantification are provided in Section S10. Our reported uncertainties for the spectral assignment essentially addressing "false-positives" and "false negatives" were derived from a simplified sensitivity analysis. We are aware that this reflects a pragmatic approach. A more rigorous assessment, however, would require spike recovery experiments, for which reference materials are still largely lacking. Furthermore, size-dependent detection efficiencies may introduce additional uncertainties. However, as our sample preparation workflow included a filtration over a 20 μm stainless-steel mesh, we constrained the smallest detectable particle size to 20 μm (corresponding to 11 contiguous pixels). Manual inspection of selected spectra of 20 μm-sized MPs confirmed sufficient spectral quality for a spectral assignment. By adding surrogate standards to

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every individual sludge sample, we were able to determine the recoveries for every individual sample. Due to the same behavior of 20 μm and of 50 μm sized particles during sample processing (digestion, filtration, and resuspension), we expect similar recoveries also for the 20 μm particles, although direct experimental evidence is currently lacking to support this claim.

2.9. Contamination Control, Blanks, and Critical Microplastic Particle Concentrations

Several measures were implemented to minimize contamination in the laboratory. Lab coats made of cotton were used, and sticky mats were placed in front of the lab doors to mitigate the introduction of dust. Prior to sample processing, 600 mL glass beakers were muffled at 550 °C for 8 h to eliminate potential plastic and organic residues. Plastic-free pressurized aluminum spray bottles were used to rinse stainless-steel meshes, glass vials, and centrifuge tubes with ethanol or DI water. Moreover, field blanks with DI water instead of sludge (n = 9) were included to estimate the contamination during the sample collection and processing in the laboratory (Section S5).

To distinguish between background contamination (i.e., MPs found in blank samples) and true MPs present in sludge samples, critical MP levels (L_C) were calculated following Ashta et al. (2026)[24]

$$L_C = \overline{x_{MPs,B}} + t_{n-1,1-\alpha} \cdot \sigma_{MPs,B} \quad (5)$$

where $\overline{x_{MPs,B}}$ and $\sigma_{MPs,B}$ are the mean and standard deviation of the number of MPs identified in the blank samples, respectively, and $t_{n-1,1-\alpha}$ is the 5% quantile of the one-sided t distribution given $n-1$ degrees of freedom, n being the number of blanks analyzed. The confidence level was set to α = 0.05 (95%), and therefore, $t_{8,0.95}$ = 1.860. Consequently, sludge samples with MP counts lower than L_C were excluded from the analysis.

2.10. Estimation of the Fluxes of Microplastics Released by Wastewater Treatment Plants in Surface Water

The MP (number) concentration in the activated sludge and the TSS contents in the sludge of the WWTPs were experimentally determined. Based on the similar ratio of MPs and TSS in the activated sludge and in the effluent of the WWTP,[21,22] the number (and the calculated mass) of MPs released by the effluent of the WWTP was calculated according to eq 6

$$\dot{m}_{MP,effluent}\left[\frac{\#\text{ or }\mu g}{yr}\right] = C_{MP,sludge}\left[\frac{\#\text{ or }\mu g}{mg_{TSS}}\right] \cdot Q\left[\frac{m^3}{yr}\right] \cdot C_{TSS,effluent}\left[\frac{mg_{TSS}}{m^3}\right] \quad (6)$$

where $\dot{m}_{MP,effluent}$ [$\#_{MPs}$ or μg_{MP} yr^{-1}] is the annual flux of MPs discharged by the WWTP through the effluent water, $C_{MP,sludge}$ [$\#_{MPs}$ or μg_{MP} mg TSS^{-1}] is the concentration of MPs in the activated sludge, Q is the annual volume of treated wastewater [m^3 yr^{-1}], and $C_{TSS,effluent}$ [mg TSS m^{-3}] is the average TSS concentration in the effluent of the WWTP (Table S1).

3. RESULTS AND DISCUSSION

3.1. Surrogate Standards Particle Recovery and Analytical Uncertainty

The surrogate standards recoveries by optical microscopy (R^{opt}), referring to the ratio of surrogate standard particles detected on light microscopy images of processed sludge samples relative to the spiked surrogate standard particles, were on average 68 ± 14% (Figures S2 and S3). These results are in line with results from other studies, in which recoveries were determined based on spiking experiments using MPs of ~50−60 μm.[27,28] In a few cases (n = 7), however, recoveries were below or equal to 50%, indicating substantial (real) losses during sample preparation or apparent losses during data processing. We included these data in our analysis but marked them accordingly. The considerable variability in surrogate standard particle recoveries highlights that preliminary method validation, performed independently of the study samples, is insufficient to guarantee MP recovery during real sample processing/analysis. Sample-specific surrogate standard particle addition, therefore, offers a pragmatic approach for routine QA/QC.

Infrared recoveries (R^{IR}), defined as the ratio between the number of surrogate standard particles detected by FPA-μ-FTIR and the number of surrogate standard particles detected optically (eq 2), were on average 90%, demonstrating a good agreement between optical and FTIR-based surrogate standard particle detection (Figures S4 and S5). The average diameters of the surrogate standard particles detected optically and by FPA-μ-FTIR were 57.5 and 52.7 μm (Figure S7), respectively, suggesting that the diameter of surrogate standards and of other environmental MPs is underestimated by approximately 10% when measured with FPA-μ-FTIR. Due to the cubic relationship between size and volume, this error extrapolates to a roughly 30% underestimation of total particle volume. The sample-specific total uncertainty on the MP counts for a given sludge sample ranged from 57 to 72% (Table S8). Accounting for a systematic 50% error in converting projected area to volume resulted in a MP mass uncertainty with a lower bound of 14% and an upper bound of 258% of the calculated mass (Table S9).

Analyses of triplicate sludge subsamples from one WWTP demonstrated consistent MP number concentrations (3100 ± 250 MPs $gTSS^{-1}$) and similar polymer-type distributions (Section S11), indicating good method precision with <10% variability across the triplicate, while calculated MP mass concentrations exhibited greater variations, averaging 239 ± 118 μgMPs $gTSS^{-1}$ (49% variability). This underscores that even with detailed and rigorous QA/QC, the MP concentrations in environmental samples are inherently associated with considerable uncertainties that must be carefully considered.

3.2. Microplastic Particles Number in Activated Sludge

FPA-μ-FTIR analysis of the processed sludge samples resulted in between 8 and 1436 MPs (>20 μm) detected within the subsampled areas of the individual filter (~30% of the total particles on the filter). Extrapolated to the entire filter surface based on the ratio of particles in the subsamples to the total particles count, this corresponded to 24 to 4208 MPs (Table S12). The critical level (L_C) calculated based on 9 blanks was 135 MPs for the total MP number on a whole filter. While the L_C was lower than the number of MPs detected for 96% of the samples (49 over 51), two sludge samples had a total number of MPs below the L_C, and their respective results were excluded from further analysis. Note that one of these two samples had a surrogate standard particles recovery of 36%, suggesting significant losses of environmental MPs during sample processing and highlighting again the importance of systematic surrogate standards addition.

Notably, eight out of nine blanks had low contamination levels (21−48 MPs per filter), whereas one blank contained 170 MPs, exclusively PP (Table S6). We hypothesize that this elevated PP count occurred randomly, potentially resulting from the use of PP centrifuge tubes used during sample preparation, and may explain the exceptionally high PP contents in a few samples (further details and discussion in Section S24).

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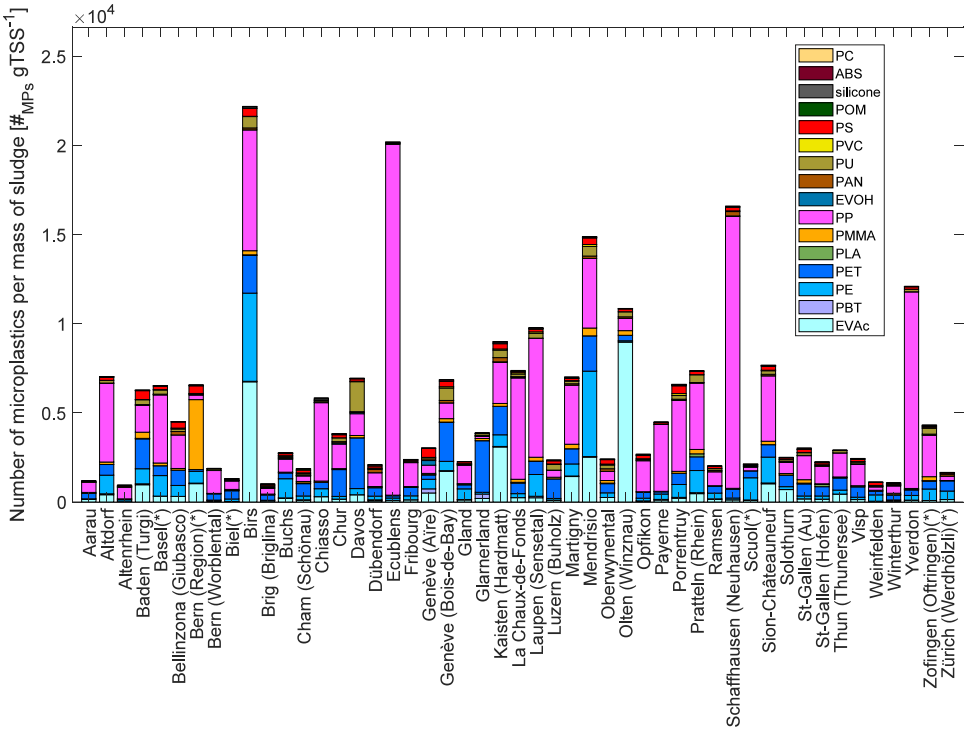

Figure 1. Number of microplastic particles (MP) normalized to total suspended solids (TSS) of the activated sludge samples from the individual wastewater treatment plants. Samples with surrogate standards recoveries below 50% are indicated with (*). Abbreviations for all polymers are provided in the Supporting Information (Table S3).

The total number of MPs detected was normalized by the mass of TSS in each subsample (~200 mg TSS), revealing concentrations ranging from 900 to 22,000 MP $gTSS^{-1}$ (Figures 1 and S14 and Table S13). These concentrations align with or exceed the upper range of literature values for biological or activated sludge (approximately 1000 to 10,000 MP $gTSS^{-1}$).[29−31] We suggest that methodological and analytical differences likely explain why the MP number concentrations obtained in this study fall within the upper range of reported values. Most previous studies used optical microscopy, attenuated total reflectance (ATR)—FTIR, or single-point FTIR techniques to identify MPs. These approaches, however, require selecting individual particles based on optical microscopy images, which inherently introduces a bias toward larger or visually distinctive MPs (e.g., colored particles or fibers). In contrast, the FPA-μ-FTIR technology used in this study allowed recording hyperspectral data of approximately 30% of the total particles count on the filter and of the filter surface (~80 mm^2) without prior selection of suspect particles, thereby ensuring the detection of MPs with equal efficiency regardless of MP color or size.[32,33]

The observed variation across the investigated WWTP was approximately 1 to 2 orders of magnitude and can originate from spatial and temporal variability and from methodological uncertainty. Because the sampling campaign extended over eight months and one sample per WWTP was analyzed, it is challenging to determine if the range of MP concentrations we report reflects temporal (intra-WWTP) or spatial (inter-WWTP) variations in MP concentrations in sludge. We argue that short-term temporal variations in sludge (daily or weekly) are minimal because the solids retention time in activated sludge systems spans several days to weeks, thus dampening potential fluctuations that may occur in the influent concentration of MPs. While long-term variations may exist, such as seasonal differences in the behavior of the population, clear data about these potential variations are currently lacking.[34] Overall, we expect temporal variations in the sludge to be limited, and therefore, a single sample is considered a robust snapshot of the concentrations of MPs in WWTPs over the studied period. Because the observed variation in MP concentrations between WWTPs in our study exceeds the quantified measurement uncertainties (57−72%), we conclude that these differences indicate true spatial variations across the individual catchments. This underscores that reporting quantified uncertainties is essential for a meaningful interpretation of the results and to enable reliable comparisons across studies. Unfortunately, such detailed uncertainties are rarely reported in MP studies. On the basis of the robust method and QA/QC used in this study and the representative selection of WWTPs across Switzerland in terms of catchments, sewer networks, and treatment processes, we speculate that the general variability in MP concentrations among WWTPs is approximately 1 or 2 orders of magnitude. Conversely, the variability ranging from 4 to 5 orders of magnitude reported in the literature[30,31] mostly reflects methodological discrepancies rather than true spatial or temporal differences.

The dominant polymer types identified in the samples in the order of decreasing abundance were PP (42%), polyethylene terephthalate (PET; 19%), PE (12%), ethylene vinyl acetate (EVAc; 10%), polystyrene (PS; 4%), poly(methyl methacrylate) (PMMA; 4%), and polyurethane (PU; 4%). PP, PET, PE, and EVAc were detected in all samples, whereas PS, PMMA, and PU were identified in 98, 96, and 92% of the samples, respectively (Figures S15 and S16). Some pairs of polymer types, such as PE and EVAc or PET and polybutylene terephthalate (PBT), have similar FTIR spectra, which may lead to misclassifications of respective MP classes by

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automated spectral matching algorithms. Interpretation of the MP abundances of these specific polymer classes, especially with respect to specific source profiles, may thus be challenging and associated with considerable uncertainties. However, total MP number counts will remain the same, and as the densities of PE and EVAc and of PET and PBT are similar, changes of the calculated masses will also be negligible. Note that polytetrafluoroethylene (PTFE) was not included in our study as the database behind the MicroPlastics Finder does not contain spectra of PTFE.

The shape of the MPs was classified into two categories: fibers (aspect ratio ≥ 3) and nonfibers (aspect ratio <3), according to Hartmann et al. (2019).[1] With a share of 92%, nonfibrous morphologies dominated the MP shapes, which contrasts with the literature about WWTP sludge and effluents where fibers are often the most common shape.[9,30,35] This may result, again, from a bias of visual preselection of suspect particles through light microscopy prior to chemical identification. This process favors fibers, which are distinct and easily recognizable, while overlooking smaller or nonfibers that are less visually prominent but detectable through automated spectroscopic mapping such as FPA-μ-FTIR. The polymer types PET and polyacrylonitrile (PAN) showed the highest fractions of fibers (30−40%) (Figure S19), consistent with the common use of these two polymer types in synthetic textiles and clothes. High release rates of fibers from textiles into wastewater during laundry have been reported.[36]

The number concentrations of MP obtained from the sludge samples were compared with the catchment characteristics of the respective WWTP. Correlation coefficients between total MP concentrations and the type of sewer network (e.g., combined or separate) or the presence of industries in the catchment were consistently below 0.1 (Wilcoxon rank-sum test, p-values >0.05). However, silicone and PU were more frequently detected in WWTPs dominantly treating industrial wastewater (p-values <0.05). Because the absolute counts of these polymers, particularly silicone, were very low, the interpretation of these trends remains challenging.

This study also included WWTP using MBBR and biofilters (Table S1), treatment schemes that often rely on biofilm carriers made of PP, PE, or PS. It has been speculated that aeration- and mixing-induced turbulence may result in MP formation from the carriers caused by collisions among carriers and between carriers and reactor walls.[37] In one sample collected from a WWTP using plastic carriers (WWTP Schaffhausen (Neuhausen)), the concentration of PP was much higher than for other WWTPs. ATR-FTIR analysis of the carriers collected from the respective WWTP (Figures S17 and S18) confirmed that the carriers were made of PP, suggesting that the abrasion of the carriers released substantial amounts of MPs. Another example is the WWTP in Bern (region), which uses PS beads as biofilm supports. In the sample collected from the WWTP in Bern (region), we observed an exceptionally high number concentration of PMMA. We speculate that the high number concentration of PMMA indeed represents abraded PS particles and that the apparent mismatch of the two polymer types reflects a systematic misclassification by the spectral matching software. In contrast, in sludge samples from other WWTPs using plastic carriers (Gland, Scuol, Aarau), the MP concentration remained within the range observed for most WWTPs, in terms of both the absolute concentration and the fractional contribution of the carrier polymer type. The different release of MPs from biofilm carriers may be explained by the age and different types of the plastic supports: in Schaffhausen, the MBBR process has been installed since the early 2000s, whereas Gland's MBBR was implemented in 2020.

3.3. Microplastic Particle Size Distribution in Activated Sludge

The size distribution of MP observed in sludge samples was calculated based on the ECD of all MPs identified (n = 15,593, Figure 2). The results demonstrate the prevalence of MP

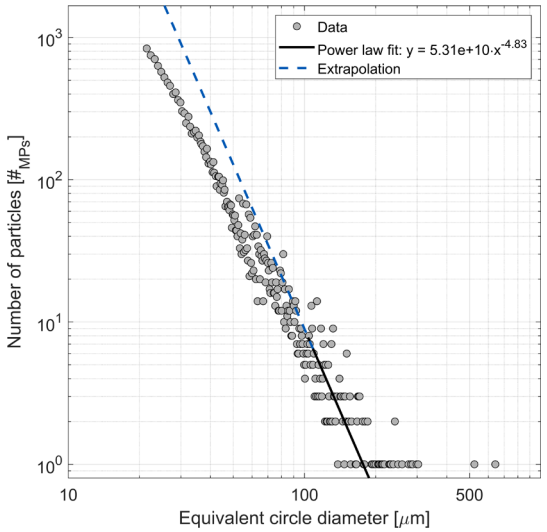

Figure 2. Particle size distribution of the detected microplastic particles (MP). The solid black line represents the power law function fitted to the MP numbers larger than 104 μm (equivalent circle diameter corresponding to the onset of the power law behavior, determined following Clauset et al. (2009)[38]). The dashed blue line corresponds to the extrapolation of the power law function for MPs smaller than 104 μm.

particles <100 μm, which accounted for 98% of all MPs quantified, whereas only 285 MPs (2%) were larger, with the maximal size recorded at 645 μm. Using the statistical testing described in Clauset et al. (2009), the onset of a power law behavior was identified at 103.5 μm. Above this threshold, the size distribution followed a power law ($n(d_{ECD}) = \beta \cdot d_{ECD}^{-4.83}$), with parameters assessed via maximum-likelihood estimation. However, when this power law function is extrapolated to MP sizes smaller than approximately 70−80 μm, the observed abundance of MP particles is lower than predicted by the power law function (Figure 2). This discrepancy can be explained in two ways. First, it could result from analytical discrimination against detection of small MPs, such as selective losses of small MPs during sample processing, or from a less efficient detection by the FPA-μ-FTIR because the particle sizes approach the instrument's size detection limit (20 μm).[39] Second, since a power law behavior reflects environmental fragmentation processes,[40,41] the deviation observed for small MPs may imply a change in the fragmentation regime, where the generation of small MPs becomes increasingly difficult. It may thus indicate that the mode of a (log) normal distribution (i.e., the particle size for which the frequency is the highest) is approached.

The power function was also a plausible model for the size distributions of seven individual polymer types, with α ranging from −2.71 (PAN) to −4.84 (PET) (Table S14). As studies

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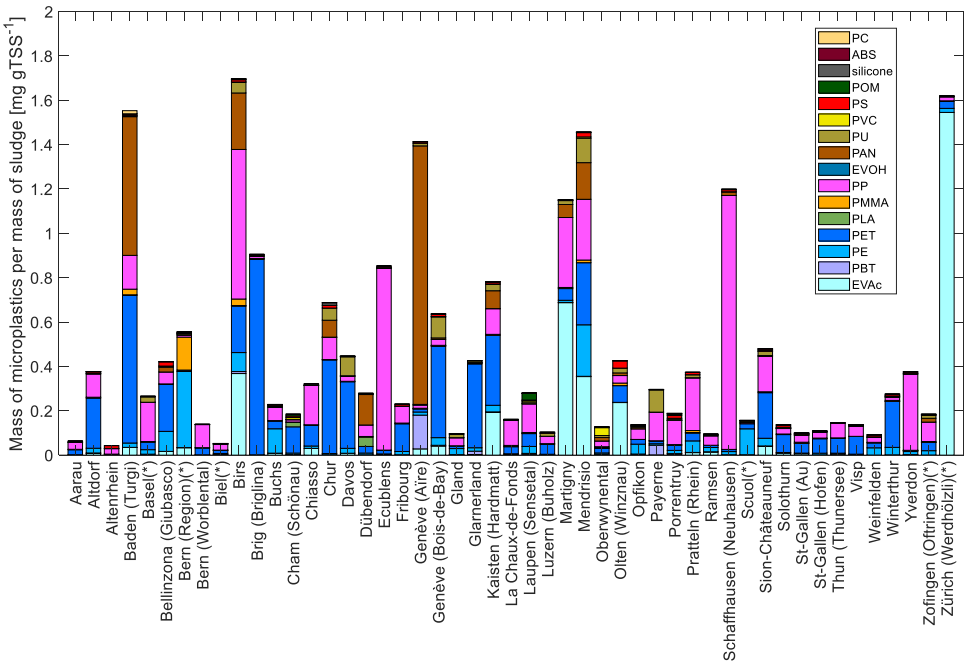

Figure 3. Calculated mass concentration of microplastic particles (MP) per total suspended solids (TSS) of the activated sludge samples collected from the different wastewater treatment plants. Samples with surrogate standards recoveries below 50% are indicated with (*). Abbreviations for all polymers are provided in the Supporting Information (Table S3).

fitting a power law function to MP size distributions in sludge are lacking, we compared our results with the literature describing MP size distributions in WWTP effluents. Several studies considering MPs (100 μm−5000 μm) in WWTP effluent report α ranging from −1.81 to −2.54[40,42,43] which is lower than the α values obtained from our study. This indicates that the proportion of small MPs relative to large MPs is considerably higher in our data set than in previously reported data sets and may reflect a systematic bias toward larger particles in previous reports, which mostly relied on (manual) ATR−FTIR or single-point μ-FTIR measurements. Alternatively, the MP particle size distribution in sludge may differ from that of WWTP effluents. However, the limited published data regarding the MP size distribution in sludge makes it difficult to test this hypothesis.

3.4. Mass Concentration of Microplastics in Activated Sludge

The mass concentration of MPs in sludge, estimated by converting the 2D projected area of each MP into a volume assuming an ellipsoidal geometry (Section 2.7), ranged from 10 μg_{MP} $gTSS^{-1}$ to 1.7 mg_{MP} $gTSS^{-1}$, with most concentrations falling between 50 and 100 μg_{MP} $gTSS^{-1}$ (Figure 3 and Table S16 and Figure S20). This corresponds to 0.005 to 0.01% of the TSS of the activated sludge being microplastic. Most important polymer types by mass were PET and PP (Figures S21 and S22). In a few sludge samples, we observed exceptionally high mass concentrations of MPs. These high mass concentrations were caused by a few very large MPs, which dominated the total mass in the respective samples.

The average mass concentration of MPs in activated sludge samples agrees with previous studies reporting values ranging from 6 to 240 μg_{MP} g^{-1} in activated sludge (dry weight).[12,29] However, higher concentrations in the range of 0.1−10 mg_{MP} g^{-1} sludge (dry weight) have been reported in two studies that relied on mass-based analytical methods (i.e., Pyrolysis−gas chromatography/mass spectrometry (Py-GC/MS)).[44,45] These higher values may reflect that Py-GC/MS analyses are susceptible to an overquantification related to interferences caused by naturally occurring substances.[46−48]

3.5. Estimation of Microplastics Loads Emitted by Wastewater Treatment Plant Effluents

Previous studies suggested similar ratios between MPs and TSS in the activated sludge and between MPs and TSS in the effluent of the WWTP.[21,22] This, in combination with the concentration of TSS in the effluent of the WWTP, the volume of treated wastewater, and the experimentally determined MP contents in activated sludge, allowed us to estimate the fluxes of MPs in WWTP effluents, expressed in terms of MP number and mass (Figure S23 and Table S17).

Since the WWTPs included in this study collectively treat the wastewater from 44% of the Swiss population, and assuming that the MPs emitted scale proportionally with population, the total number and mass of MPs discharged by all Swiss WWTPs to surface waters amounts to approximately 3.1×10^{13} MPs yr^{-1} and 4.7 t MPs yr^{-1}, respectively. Considering the uncertainties discussed in Section 3.1 and reported in Tables S8 and S9, the estimated emission ranges (95.5% confidence interval) are 1.4×10^{13} to 5.5×10^{13} MPs yr^{-1} and 0.9−11.4 t MPs yr^{-1}. Normalizing these MP loads by the volumetric flow rates of the respective WWTP yields estimated concentrations of MP in the treated wastewater ranging from 0.7 to 118 MPs L^{-1} and 0.03 to 16 μg MPs L^{-1} (Figure S24). Spanning over more than 2 orders of magnitude, these values are comparable to the available literature reporting concentrations of 0.01−3.8 μg MPs L^{-1}.[7,31,49] The estimated concentrations in WWTP effluents in this study reflect the variations in both MP concentrations in sludge (1−2 orders of magnitude) and in TSS concentration in WWTP effluents (1−2 orders of magnitude, Table S1). Since MPs in WWTP effluents are attached to the suspended solids, the TSS

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concentration variability plays an important role in the fluctuations in MPs in effluents. This likely explains why the MP concentrations in WWTP effluents are more variable than in sludge. Accounting for TSS variability in WWTP effluents is thus essential when quantifying MPs in WWTP effluents, which further highlights the advantage of sampling sludge over effluent to estimate MP emissions.

A mass-flow analysis for plastics and MPs (from 1 μm to 5 mm) in Switzerland conducted by Liu and Nowack (2025)[20] modeled the MP mass loads released by WWTP effluents. Considering the most prevalent six polymers and based on reported removal efficiencies of the WWTP, these authors calculated 6.9 t MPs yr^{-1} are emitted by WWTPs, which aligns well with our results. However, any conclusions based on this comparison should be made carefully as our study covers a different size range because (1) the WWTPs included in our study have bar screens strictly retaining MPs > 2 or 3 mm and (2) our analytical window only allows quantification of MPs down to 20 μm. Despite these differences, the mass fluxes, concentrations, size information, and power law dependence provided herein offer valuable data for refining future mass-flow models of MPs in wastewater systems.

To estimate per capita and per population-equivalent (P.E.) emissions, the quantities of MPs released were further normalized by the number of inhabitants and P.E. connected to the WWTP, respectively. On average, annual per capita emissions through WWTP effluents were 5.2×10^6 MPs (capita $yr)^{-1}$ or 0.5 g MPs (capita $yr)^{-1}$, and emissions per P.E. reached 5.9×10^6 MPs (P.E. $yr)^{-1}$ or 0.6 g MPs (P.E. $yr)^{-1}$ (Figure S25). While the per capita number emissions are slightly higher than the values reported in the literature (4.2×10^5 MPs (capita $yr)^{-1}$ to $0.6–1.2 \times 10^6$ MPs (capita $yr)^{-1}$[14,50]), the per capita mass emissions align well with other studies (0.0012–0.68 g MPs (capita $yr)^{-1}$[5,8,50,51]). The higher number in this study may reflect the more sensitive analytical methods used in this study (FPA-μ-FTIR), extending the analytical window to smaller particles. As the mass concentrations are dominated by the larger particles, a lower detection efficiency of smaller particles will only marginally affect the total MP mass.

One limitation of our approach for estimating MP loads emitted by WWTP effluents is that it relies on the assumption that the MP-to-TSS ratio in the activated sludge is similar to the MP-to-TSS ratio in the effluent of the WWTP. While lab-scale experiments largely support this assumption, deviations may occur for buoyant MPs, which undergo less efficient heteroagglomeration with sludge flocs.[22] Such buoyant MPs may therefore remain in suspension to some extent and pass through the WWTP. This would alter the MP-to-TSS ratio and would lead to an underestimation of the emissions of buoyant MPs by up to a factor of 4.[22] As buoyant polymer types (PE, EVAc, and PP) account for 33% of the estimated emitted MP loads, a 4-fold increase would lead to annual loads of approximately 9.5 tons of MPs discharged from WWTPs to surface waters. Overall, our approach serves as a robust proxy for estimating MP emissions through WWTP effluents and offers several advantages associated with sludge sampling. Nevertheless, full-scale validation may still be required to confirm similar MP-to-TSS ratios in activated sludge and in the WWTP effluent.

4. CONCLUSIONS

Based on the MP concentrations in activated sludge and the effluent TSS from 51 WWTPs in Switzerland, nation wide emissions of MPs (>20 μm) from WWTPs to surface waters were estimated to be 3.1×10^{13} MPs yr^{-1} or 4.7 t MPs yr^{-1}, translating to 0.5 g MPs (capita·$yr)^{-1}$. Upgrades of WWTPs to enhance the removal of organic micropollutants are ongoing in Switzerland since 2016 and are planned across the European Union by 2045 under the recast Urban Wastewater Treatment Directive (EU 2024/3019). Treatment approaches implemented in this upgrade—including activated carbon filtration and ozonation followed by sand filtration—further decrease TSS concentrations in the effluent and consequently decrease the MP loads discharged into surface waters. Based on effluent TSS concentrations reported from WWTPs, these treatments can lower the TSS in the effluents to approximately 1 mg TSS L^{-1}. Consequently, the Swiss-wide input of MPs from WWTP effluents to surface waters would decrease to approximately 6×10^{12} MPs yr^{-1} or 1 t MPs yr^{-1}, corresponding to roughly 0.1 g MPs (capita $yr)^{-1}$. Although the study has been conducted on Swiss WWTPs, the per capita emissions are transferable to other WWTPs using similar treatment schemes.

Future studies should compare the estimated current load (4.7 t yr^{-1}) and the predicted future load (1 t yr^{-1}) of MPs discharged from WWTPs into surface waters with data from MP monitoring programs of surface waters (e.g., rivers Rhine and Rhône, which largely drain Switzerland) once such data become available. This comparison would help assess the contribution of WWTP emissions to the total MP loads observed in surface waters. In addition, future work should provide MP input estimates to surface waters from other sources, such as atmospheric deposition and stormwater overflows. Accounting for these sources will advance a comprehensive MP budget and, thereby, help identify the most effective measures to lower MP inputs into surface waters.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestwater.6c00366.

Additional methodological details regarding sample processing, surrogate standards identification, blanks and contamination control, and data treatment figures and tables displaying WWTP characteristics, sample volume and mass, surrogate standard recoveries (optical and IR), blank analysis, uncertainties associated with particle counts and mass, triplicate analysis, biofilm carriers ATR-FTIR spectra, parameters of the power law function to fit particle size distribution, detailed MP and mass counts, and particles and mass loads emissions for all WWTPs (PDF)

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Notes

The authors declare no competing financial interest.

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