ABSTRACT: Microplastic research, initially focusing on marine environments, left freshwater ecosystems largely unexplored. Freshwaters are also vulnerable to microplastics and are likely the largest microplastic supplier to the ocean. However, microplastic sources, transport pathways, and fluxes at the catchment level remain to be quantified, compromising efficient actions toward mitigation and remediation. Here we show that 70–90% of microplastics reaching Norway’s largest lake, originating primarily from urban waste mismanagement and sludge application on crops, continue their journey toward the ocean without being buried. Indeed, our microplastic budget for the catchment shows that out of the 35.9 tons (7.4–119.4 t) of microplastics annually released into the lake, only 3.5 tons (1.3–8.8 t) are settling to the lake bottom. The spatial and vertical microplastic distribution and diversity in lake sediments, the socio-economic modeling of plastic fluxes and spatial information on land use and potential plastic sources all point toward urban and agricultural areas as emission hotspots of increasing importance. We conclude that the degree to which lake sediments represent a net microplastic sink is likely influenced by the nature of microplastics the lake receives, and ultimately on their origin.

KEYWORDS: microplastics, lake sediment, catchment, land use, plastic sources

INTRODUCTION

Originally recognized as a problem in the marine environment, increasing amounts of microplastics are now also reported in terrestrial environmental samples around the world. Developing methods for microplastic source identification is a priority to enable actions toward mitigation and remediation. However, the sources of these items often lie on land where most of the plastic is used, rather than in marine and coastal ecosystems. Freshwater systems can act as transporters and/or sinks for terrestrial microplastics. Investigations into freshwater systems have been receiving increased attention in the past few years, although most of the focus is on reporting microplastic distribution. Hence, source tracking methods are currently limited and the factors influencing the behavior of freshwaters toward being transporters versus sinks are ill-known. Therefore, microplastic research efforts need to move upstream and improve catchment-level approaches to be able to identify the main sources and quantify freshwater microplastic fluxes and stocks.

Quantifying plastic release into surface waters is challenging due to data sparsity and difficulties attributing microplastics to their initial sources. Considering the standard waste mismanagement rate of 2%, and a release rate of 30% for mismanaged waste from coastal areas, we estimate that 106 tons of plastics could be annually released into Norway’s largest lake, Lake Mjøsa (5300 t of plastic waste annually processed in the catchment). However, precise release pathways are influenced by land use with emission hotspots as highly populated areas and industrial and agricultural sites. In addition, the amount of microplastics deposited on the Lake bottom or that are transported downstream remain to be quantified.

Several studies investigated spatial variability in microplastic distribution in the Great Lakes and modeled the propagation of floating plastics. While these approaches are relevant for understanding plastic flows within large lakes, spatial information from the catchment were not included in these assessments limiting their usefulness for identifying the sources and release pathways.

The distribution of sediment contaminants can yield significant spatial and temporal information on pollution, although, in the case of plastics, a non-negligible fraction may not sink to the bottom due to their low density. Nevertheless, lake sediments provide reliable natural archives for estimating historical contamination of heavy metals and organic
contaminants but their use to describe microplastics pollution has been limited so far.

Here we present an original approach to investigate microplastic stocks and fluxes and apply it to a Nordic catchment-lake ecosystem, Lake Mjøsa. We quantify the different plastic morphologies identified along sediment cores collected at 20 sites and explain their spatial distribution with an innovative spatial data set. We report spatial information on land use, wastewater treatment plant (WWTP), urban and industrial services for the whole catchment as well as for delineated subcatchments where higher microplastic concentrations are reported. We further provide a first microplastic budget for the catchment where top-down estimates of microplastic emissions from the modeling of socio-economic activities are corroborated with sediment inventories.

**MATERIALS AND METHODS**

**Study Site.** Mjøsa is a glacially excavated deep lake, the largest lake in Norway and one of the deepest in Europe with a volume of 56 km$^3$ (Figure 1a; Table S1 of the Supporting Information, SI). Its catchment (17 028 km$^2$) includes large mountain areas in the north, and forest, urban, and agricultural areas east and west of the lake. Most of the urban, industrial, and agricultural lands are located along the lake shoreline around the central part of the lake (Figure 1a). The river Gudbrandsdalslågen is the main tributary into Lake Mjøsa, draining an area of 13 000 km$^2$, i.e., 78% of the catchment, with a higher proportion of natural land types than the areas around the lake (Figure 1a). Mjøsa is a drinking water source for approximately 70 000 people through the municipal and private water supply, as well as industry.

The 20 sampling sites within Lake Mjøsa (Figure 1c; Table S2) were chosen to be representative of a range of diverse lacustrine environments with some close to potential sources of microplastics, as well as reference points in Lake Mjøsa’s natural sediment accumulation areas. Several of the selected stations have been previously described regarding location, depth, sediment dating, and content of certain pollutants. The river Gudbrandsdalslågen is the main tributary into Lake Mjøsa, draining an area of 13 000 km$^2$, i.e., 78% of the catchment, with a higher proportion of natural land types than the areas around the lake (Figure 1a). Mjøsa is a drinking water source for approximately 70 000 people through the municipal and private water supply, as well as industry.

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Microplastics Sampling and Analyses. Sediment sampling was conducted between August 6th and 9th, 2018. Cores were collected from each of the 20 locations using a Kajak-Brinkhurst sediment corer with an internal diameter of 8.5 cm. In the deepest parts of the lake a Van Veen grab was used and a core was taken once on the deck. Each sediment core was divided into 1 cm slices in the field, within 30 s for each core slice. On return to the laboratory, each core slice was freeze-dried for further processing by density separation with sodium iodide (NaI, 1.7 g cm$^{-3}$), in some instances organic matter removal had to be included using Fenton’s reagent. Samples were passed through a 75 μm sieve to remove smaller particles. The retained material was then rinsed onto filter papers (GF/D, 47 mm, pore size 2.7 μm).

All particles found in each sample were analyzed by visual identification and measured along their longest and shortest dimension followed by chemical confirmation of the polymer material. Suspected microplastics were analyzed using a PerkinElmer Spotlight 400 μFT-IR in transmission mode. A diamond compression cell (DC-2 Diasqueeze) was used to improve spectral quality. Background scans were taken each time the compression cell was reloaded onto the instrument (circa every 1–4 suspected microplastic particles). All spectra were compared with a series of commercial (PerkinElmer ATR Polymers library, STJapan Polymers ATR library), the BASEMAN library$^{25,26}$ and in-house libraries (including reference polymers, different textile materials, and potential sources of laboratory contamination) and manually inspected to confirm the match. Only 3% of the suspected microplastics were rejected after FT-IR analysis because of no polymer match. Procedural contamination was monitored throughout the sampling and analysis with the use of a series of blank samples to allow for results corrections based on presence of plastics in blanks. Only 1 out of 62 blanks contained one particle of pink polypropylene. Since this microplastic type was absent in our samples, no bank correction was performed. Data processing method (75 μm) was extended using a 10 × 10 m$^2$ Digital Elevation Model publicly available from høyedata.no which was reclassified to 25 × 25 m$^2$ for computational efficiency. The reclassified DEM was then processed with the TauDEM package$^{30}$ in order to fill depressions and then obtain the flow direction raster (all water from one cell flows to the lowest adjacent one). The NVE river network$^{31}$ was burned into the DEM with filled depressions. The consolidated DEM was then used to derive the catchments which were further used in a GIS application (QGIS v.3.6.0) to retrieve land cover data.

Land Cover Data. Land use data has been retrieved from a Jupyterhub intersecting the catchment and subcatchment polygons with the land use data from CORINE landcover from 2000 to 2018 (Copernicus European program of Earth observation). The various CORINE land classes have been reclassified as shown in Table S3. The total length of urban shoreline and the shortest distance from each sampling station to the nearest upstream town (>2000 inhabitants) was manually computed. Municipalities at least partly located in the catchment were identified; however, to construct a representative data set, only municipalities for which at least 50% of their territory was located within the catchment were included. The total surface area represented by the selected municipalities was only 7% larger than that of the catchment. Missing or added areas were mostly natural land types and all urban settlements with more than 2000 inhabitants were included. Official data on plastic industries, waste sorting facilities, waste production and management, population estimates, wastewater volume, driving distances and road length from the selected municipalities were then downloaded from Statistics Norway. The average water outflow was reported for each WWTP as provided in Snilsberg et al. (2005).$^{32}$

Plastic industries were normalized according to the number of employees in the company, e.g., 1 and 0.5 normalized plastic industry are equivalent to a company of more than 100
employees and 50 to 99 employees, respectively. Plastic industries were then associated with the “Industrial and commercial unit” polygon found within the municipality boundary. In only one case, several industrial polygons were located within the municipality boundary. We therefore performed a web search for the main plastic industries in this municipality to confirm their location.

We identified five urban and industrial zones close to sites 2, 4, 7, 11, and 13 of which only three included plastic industries (close to sites 2, 4 and 13; Figure 1c). Subcatchments S3 and S5 are the most agricultural, with 28% and 45% agricultural areas, respectively. Most importantly, about 90% of their agricultural land is used for intensive agriculture (>75% of cultivation; Figure 1b). Some other subcatchments (S2, S4, S6, S7, and S8) also have a significant fraction of agricultural land (15 to 20%), but most of it falls into land types with <75% of cultivated land and considerable natural attributes. Other potential sources of microplastics, i.e., mines, airports, harbors, large recycling facilities, industrial laundry, and WWTP, represented in Figure 1c, also show a higher density close to sites 7 and 13, within the two largest urban centers, i.e., Gjevik and Hamar.

**Top-Down Plastic Flux Estimates.** The plastic fluxes for the Mjøsa catchment were estimated following a top-down approach with statistics on socio-economic activities within the catchment similar to Boucher et al. (2019). This procedure included the estimation of: (i) the magnitude of different sources in the plastic consumption and production processes; (ii) the losses to the environment; and (iii) the releases into surface waters and Lake Mjøsa.

Annual national and municipal data on waste production and management, population estimates, wastewater volume, plastic industries, driving distances, and road lengths were retrieved from Statistics Norway. Other data sources included PlasticsEurope and scientific literature (Tables S4 and S5).

**Step 1: Quantification of the Magnitude of Plastic Sources within the Catchment.** The plastic life-cycle assessment included three phases: production (including primary production and plastic conversion), use, and end-of-life. The production of plastic was downscaled from European and national statistics on plastic production and sales taking into account the number of plastic producers and their size (i.e., number of employees) within the watershed. The total amount of plastics in use was estimated from French and Swiss estimates from Boucher et al. (2019) and population within the catchment, and usage by market was downscaled from data given by PlasticsEurope 2019. Plastic end-of-life treatment was obtained from official statistics on household plastic waste and treatment from municipalities within the watershed and on plastic waste by sector at the national level.

**Step 2: Quantification of Environmental Losses.** Environmental losses included all microplastics released into the various compartments of the environment. Note that at this stage, some plastics, e.g., from household waste mismanagement, are still macroplastics but show a high potential for microplastic generation.

**Wastewater Treatment Plants.** Losses during wastewater treatment were calculated from data on microplastics removal efficiency from several wastewater treatment plants in Norway, including one within the watershed. Since microplastics from household laundry represent a significant fraction of those contained in wastewater influents, microplastic losses from textile were estimated considering generic washing habits per household and standard share of synthetic clothes and shredding rate reported in the literature.

**Agriculture.** Microplastic losses from agriculture included two main processes: (i) the weathering of mulching films and (ii) the application of sewage sludge. Most of microplastics originating from household and laundry, industrial processes (e.g., blasting and shredding of plastics), and from the decomposition of plastic surfaces (e.g., polymeric paints) will be conveyed to municipal effluents and mainly be collected by wastewater treatment plants. Existing wastewater treatment processes effectively removes most of the microplastics from water and retain them in the sludge. In Europe and North America about 50% of sewage sludge is used as a fertilizer in agriculture. In Norway, a total of 71.505 tons (dry weight) of sewage sludge has been applied to agriculture in 2018. Data on wastewater microplastic concentration and removal efficiency from several wastewater treatment plants in Norway, including one within the watershed, and wastewater volumes from municipalities within the catchment are used to estimate the amount of microplastic applied to agricultural soils. Additional estimates are derived from official municipal statistics on agricultural sludge application and typical microplastic content in sludge reported in the literature. Plastic losses from mulching were downscaled from national mulching surface area and typical rates of mulch application and mulch recovery reported in the literature.

**Construction.** Microplastic losses from construction primarily occurs during the building phase3 assuming that once plastics have been integrated into a building, losses are negligible and that during the demolition phase, waste is properly managed. Expanded polystyrene (EPS) represents the main losses from construction sites, and is used for its expanding property for insulation and typically undergoes a polishing step which emits plastic dust. Losses were estimated using typical EPS usage in construction, plastics used for construction in the catchment and typical EPS loss rate reported in the literature.

**Plastic Industry.** Losses in production were estimated from standard rates of losses during delivery of primary plastic within the watershed as a function of the total plastic produced in the watershed.

**Car and Lorry Tires.** Plastic emissions from tires have been calculated from emission rates estimated for Norway using official statistics on driving distances within the watershed for four categories of vehicles and standard synthetic rubber composition of tires.

**Road Marking.** Road paint is a major source of synthetic polymers such as methyl methacrylate. Road marking losses were quantified using official statistics on road length within the watershed and downsizing the amount of paint annually applied on Norwegian roads considering that old paint was not gathered prior to new paint application.

**Household Waste Mismanagement.** Even if the Norwegian waste management system is among the most efficient in the world, involuntary spill and illegal littering is still a probable and important source of microplastics for the environment. Tons of plastic waste, mainly from private use, are recurrently collected on lake beaches in Norway, including along Lake Mjøsa. Jambeck et al. (2015) considered a standard rate of 2% of waste mismanagement for plastics in occidental countries. Here, we take this 2% rate as the most pessimistic rate for microplastic losses and consider a rate, 1 order of magnitude lower, as a more realistic estimate for...
Each Sampling Site

Boucher et al. (2019) considered equestrian gear as a major source of microplastics for the environment, although, as Boucher et al. (2019) noted, it is likely not the dominant source. Most of these items are likely not the dominant source of microplastics, including balloons and everyday objects like household appliances, medical waste, toys, and household waste mismanagement. Note that a recent study showed that >99% of microplastics applied from biosolids were likely exported to the aquatic environment.47

Table 1. Sediment Microplastic Concentration ([MP]) and Richness (S), Diversity (H and D) and Evenness (E_H) Indexes at Each Sampling Site

| sites | [MP] (microplastic g⁻¹) | S | H | D | E_H |
|-------|------------------------|---|---|---|-----|
|       | 0–1 cm | 1–2 cm | 2–3 cm | 3–4 cm | 7–8 cm | average per site | 0–1 cm | 1–2 cm | 2–3 cm | 3–4 cm | 7–8 cm | average per site |
| urban |     |       |       |       |       | 0.14 |       |       |       |       |       |       | 0.07 |       |       |       |       |       | 0.07 | 3 | 0.95 | 2.3 | 0.86 |
| 2     | 0.78 | 0.14  | 0.13  | 0.03  | 0.23  | 0.26 | 9 | 1.84 | 5.0 | 0.84 |
| 4     | 2.54 | 0.36  | 0.85  | 0.10  | 0.04  | 0.78 | 6 | 1.46 | 3.5 | 0.82 |
| 7     | 0.91 | 0.45  | 0.63  | 0.03  | 0.41  | 0.41 | 10 | 1.83 | 3.9 | 0.80 |
| 8     | 1.18 | 1.44  | 0.20  | 0.38  | 0.18  | 0.68 | 11 | 2.05 | 6.0 | 0.85 |
| 12    | 0.58 | 0.06  | 0.67  | 0.08  | 0.28  | 0.28 | 7 | 1.91 | 6.5 | 0.98 |
| 13    | 0.93 | 0.07  | 0.29  | 0.09  | 0.03  | 0.28 | 8 | 1.80 | 4.6 | 0.86 |
| 14    | 1.15 | 1.25  | 2.03  | 2.17  | 0.71  | 1.46 | 11 | 1.94 | 5.6 | 0.81 |
| 15    | 0.75 | 0.93  | 0.92  | 0.52  | 0.52  | 0.52 | 6 | 1.39 | 3.0 | 0.78 |
| 16    | 0.86 | 0.08  | 0.96  | 0.10  | 0.40  | 0.40 | 7 | 1.85 | 5.8 | 0.95 |
| 10    | 0.20 | 0.37  | 0.18  | 0.41  | 0.06  | 0.25 | 8 | 1.87 | 5.7 | 0.90 |
| 11    | 0.14 | 0.08  | 0.12  |       |       | 0.07 | 3 | 0.95 | 2.3 | 0.86 |
| 6     | 0.05 | 0.10  | 0.06  | 0.04  | 0.04  | 0.04 | 4 | 1.33 | 3.6 | 0.96 |
| 10    | 0.24 | 0.20  | 0.07  |       |       | 0.10 | 3 | 1.05 | 2.8 | 0.96 |
| 11    | 0.13 | 0.24  | 0.24  | 0.12  | 0.12  | 0.12 | 4 | 1.24 | 3.0 | 0.90 |
| 12    | 0.19 |       |       | 0.04  | 0.04  | 0.04 | 1 | 0.00 | 1.0 |       |
| 17    | 0.04 |       |       | 0.01  | 0.01  | 0.01 | 1 | 0.00 | 1.0 |       |
| 18    | 0.13 |       |       | 0.03  | 0.03  | 0.03 | 1 | 0.00 | 1.0 |       |
| 19    | 0.33 | 0.14  | 0.46  | 0.18  | 0.18  | 0.18 | 5 | 1.55 | 4.5 | 0.96 |
| 20    | 0.05 | 0.15  |       | 0.04  | 0.04  | 0.04 | 3 | 1.04 | 2.7 | 0.95 |
| average per site | 0.54 | 0.29  | 0.37  | 0.22  | 0.08 |       |       |       |       |       |       |       |

"Sites showing average sediment microplastic concentrations greater than 0.20 microplastic g⁻¹. All these sites, except one (site 16) which is located in an agriculture influenced area, are located in high urban influenced areas (Figure 1c). Site 11 is located near an urban zone (Figure 1c) but does not display patterns regarding microplastic sediment concentration, diversity, evenness, and richness that are similar to the other urban-influenced sites. It was therefore considered as a natural site.

Microplastic losses from waste mismanagement. These rates are typically used for macroplastic losses, attributing them to microplastics comes with the assumption that the amount of plastic present in the environment is at steady state, i.e., that the amount of macroplastics losses equals microplastic generation through degradation. We applied these rates to annual amounts of household plastic waste generated within the watershed, i.e., mainly food packaging. While the underlying assumption that macroplastics stocks in the environment are at steady state (losses equal microplastic generation) cannot be supported by our data set, it helps taking into account microplastics generated from illegal littering, which would have been ignored otherwise.

This analysis excluded microplastics from cosmetics, recreational products, medical waste, toys, household appliances, and furniture. Most of these items are likely not the dominant sources of microplastics for the environment, although, as Boucher et al. (2019) considered equestrian gear as a major source, as well as balloons and fishing gear. Altogether, we consider that the top-down environmental loss estimates are conservative given the nonexhaustive list of sources.

Step 3: Release Pathways to Surface Waters. Once in the environment, microplastics can reach surface waters through several pathways described in detail elsewhere. Mismanaged Waste. Leaching of mismanaged waste, e.g., resulting from littering or forgotten plastics in agriculture, to adjacent surface water has been previously estimated at 15–40%, with a mean value of 25%. We used these values for microplastic releases from agriculture, construction, the plastics industry, and household waste mismanagement. Note that a recent study showed that >99% of microplastics applied from biosolids were likely exported to the aquatic environment. Given that, to our knowledge, only one study has specifically looked at microplastic agricultural runoff, and they argue that release rates are likely influenced by local properties and weather events, we used the mismanaged waste release rate for agriculture microplastic releases, although it is likely conservative.

Road Runoff. Microplastic emissions from tire dust and road paint are mainly released to surface waters through road runoff. Previous studies have estimated that 2–18% of these particles reached the surface waters. We considered a mean value of 6%.

Sewage Systems. We used reported microplastic capture rates for wastewater treatment plant in Norway including one in the watershed. Microplastic removal efficiency was reported to be as high as 90 to 99% for these WWTP with a mean value of 95%. This release pathway is likely to provide a conservative estimate since we neglected any microplastic release caused by storm overflow.

Direct Release. This pathway was only applied to WWTP effluents, as microplastic capture rates were already applied during the calculation of the associated environmental losses (step 2).

Microplastic Stock in Lake Mjøsa Sediments. To estimate the total mass of microplastic in the top 5 cm of the sediment column in L. Mjøsa, two cases have been considered: (i) sediment located within 2 km downstream of urban areas, defined as high urban influence areas; and (ii) sediment located in the rest of the lake bottom defined as natural areas. Mean sediment microplastic concentration ([MP]) in high urban influence areas has been modeled with the following equation:
higher concentrations than the other sites (Mann–Whitney rank sum test, \( P < 0.001 \)). These sites are referred to as urban sites hereafter, while the remaining are so-called natural sites.

This finding is in line with recent freshwater studies also reporting higher microplastic concentrations closer to urban areas.\(^49,50\) Most of the cores show increasing concentrations with decreasing sediment depth although local trends may differ. Variability within the reported cores arise from differences from one layer to another. These differences can be caused by changes in plastic inputs to the lake, but they also likely reflect local depositional and erosional processes. Our data set and the absence of high-resolution bathymetric data does not enable us to resolve any of these processes. Hence, we thereafter focus on the microplastic concentrations averaged for each site and spatial and temporal patterns that are supported by several stations.

A total of 13 polymers were found (Figure S2) with contrasting microplastic diversity across sites, but consistent patterns among urban and natural sites. Microplastic polymers were dominated by acrylic and polyester representing nearly 50%, followed by polyethylene, polypropylene, polyethylene terephthalate, polystyrene, viscose, and polyamide, while synthetic rubber, poly methyl methacrylate, polyurethane, polyvinyl chloride, and polycarbonate were quasi absent (Figure S2). The polymer density of sediment microplastics ranged from 0.9 to 1.5 g cm\(^{-3}\) with an average of 1.15 g cm\(^{-3}\). Only two polymers were less dense than water: polyethylene and polypropylene. In total, fibers accounted for 50%, fragments 49% and beads 1%. The urban sites, showing higher mean concentrations (>0.25 microplastic g\(^{-1}\)), also displayed higher richness (\( S \)) and evenness (\( E_H \)) indexes as well as lower evenness (\( E_D \)) and diversity (\( H \)) indexes (Figure 2). All these indexes are consistent with typical concentrations reported from freshwater sediments.\(^49,50\) Half of the selected sites, located in urban (and/or agriculturally) influenced areas (Table 1 and Figure 1c), showed mean concentrations above 0.20 microplastic g\(^{-1}\) and significantly higher concentrations than the other sites (Mann–Whitney rank sum test, \( P < 0.001 \)). These sites are referred to as urban sites hereafter, while the remaining are so-called natural sites.

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and richness are expected closer to the source. In addition, higher evenness at more distal natural sites also supports this interpretation since these sites receive microplastics following transport and redistribution during storm events. These random-like transport processes have the effect of dispatching microplastic across space, homogenizing their spatial distribution.

**Temporal Trends in Sediment Microplastic Concentrations in Lake Mjøsa.** Figure 2e,f displays the sediment microplastic diversity, richness, and evenness indexes for urban, natural, and all sites, as well as the sediment microplastic concentrations from the 1980s to 2017 for the three groups of sites. The Shannon diversity index, the richness and concentration significantly increased at the urban sites since the 1980s. Further, the concentration for all sites showed significant increasing trends, while no significant trends were associated with the natural sites. Hence, the increase in the accumulation rate of microplastic over time in the Mjøsa sediments (+1.2 ± 0.4%) seems to mainly be caused by increased deposition at the urban sites (+2.2 ± 0.7%).

**Urban Areas as the Main Sources of Microplastics.** Figure 3 shows that there is a clear decreasing trend in sediment microplastic concentration as the distance from the nearest upstream city. Depth-average [MP] at all sites as a function of distance to the upstream nearest town. Gray dashed lines connect stations along a littoral-distal transect, e.g., stations 13, 14, and 15. Note the systematic decrease in [MP] along these transects.

**Modeled Plastics Environmental Losses and Releases into Surface Water.** Our socio-economic top-down modeling reveals that about 10.1 kt of plastic are annually introduced into the market in Mjøsa catchment, while between 15.2 and 27.6 kt are estimated to be currently in use and only 9.0 kt discarded (Figure 4; Table S5). The imbalance between production and end-of-life fluxes can be explained by the increasing use of plastics as well as their increasing lifetime, or by unaccounted export fluxes.

Total microplastic losses in the watershed are 308 (235–441) t yr⁻¹ with microplastics from tire abrasion, agriculture (mainly from wastewater sludge), laundry, household waste, and road markings as the main contributors (Figure 4; Table S5). Total releases into the catchment waters are 36 (7–119) t yr⁻¹ with mismanaged waste being the main release pathway, mainly through the application of sludge on agricultural soils (16.3 t yr⁻¹) and household waste littering (5.9 t yr⁻¹). Releases from road runoff (5.5 t yr⁻¹), including microplastic from tire and road paint, and from WWTP (3.8 t yr⁻¹) represent about 20% and 12% of the total, respectively. Microplastic releases, totalling 160 g inhabitant⁻¹ yr⁻¹, are more than twice larger than those reported for Lake Geneva. However, about half of these releases are due to sludge application, a practice that has been banned in Switzerland since 2006.⁵⁷

Release estimates from tires and road marking are robust since the tire weight before and after use, the amount of road paint that is applied and their release pathway are well constrained.⁴⁰,⁴¹ Other fluxes, such as releases from sludge application on agricultural land or from household waste, have an intermediate level of uncertainty since initial stocks/fluxes are based on official municipal statistics but some losses and releases rates are taken from the literature, e.g., littering rate of 2%⁵ and 0.1%³ for household wastes. These uncertainties did not compromise the consistency of our budget since we used conservative release and loss rates. In particular, despite the use of a conservative release rate, both independent methods used to estimate microplastic releases from sludge application showed that this flux was the largest (Table S5).

Besides corroborating the importance of urban areas as microplastic sources, the top-down modeling highlights agricultural practices as potential considerable microplastic emission pathways as recently pointed out.⁷ Indeed, sludge application is the most important release within Mjøsa catchment (Figure 4) which is consistent with the fact that sites located at the outlet of most agricultural subcatchments, e.g., sites 13–16, show the highest sediment microplastic concentrations when normalized to the distance to the nearest town (Figure 3).

**Microplastic Lake Retention.** Only about 10% of the microplastics emitted settles into Lake Mjøsa sediments (Figure 4), making the lake more of a pipe than a sink, in contrast to the interpretation for Lake Geneva.⁵ Our sediment microplastic stock estimate is likely conservative since size fractions <75 μm were not collected. Also, urban-influenced sediments showing higher sediment microplastic concentration might cover a larger proportion of the lake bottom than estimated here. However, this underestimation is likely not several fold ensuring that most of the plastics emitted in the catchment is not buried in Lake Mjøsa sediments. While the fate of those microplastics cannot be completely resolved, we can say with confidence that they are still available for further downstream transport or remobilization, potentially all the way
to the ocean. The important contributions of sludge application (about 50%) and tire abrasion (about 15%) to microplastic releases in Mjøsa can partly explain the contrasted behavior of L. Mjøsa and Geneva. First, most of microplastics found in sludge from eight Norwegian WWTPs, including the largest one in Mjøsa catchment, are expected to float after remobilization since those are beads and fragments of low density (<1 g cm\(^{-3}\)).\(^{34}\) In Switzerland, by contrast, sludge application was banned in the 2000s.\(^{57}\) Second, microplastics emitted from tires, e.g., Styrene Butadene Rubber,\(^{42}\) are associated with higher density components resulting in composite particles of very high density (1.5–2.2 g cm\(^{-3}\)).\(^{48,58,59}\) This interpretation is also consistent with the quasi-absence of synthetic rubber in our sediment samples (Figure S1). Furthermore, the dominant presence of high density (1.2–1.5 g cm\(^{-3}\)) synthetic textile fibers in our sediment samples agrees well with synthetic textile fibers being the most abundant high-density microplastic in sludge.\(^{34}\) In contrast, fragments and films of polylethylene, polyethylene terephthalate or polyvinyl-chloride were the dominant microplastics found in Lake Geneva sediments.\(^{62}\)

Microplastic deposition follows a similar exponential increase in Santa Barbara basin (doubling time of about 15 years)\(^{63}\) and Lake Mjøsa sediments (doubling time of ~12 years; Figure 4), and shows significant correlations with increases in Global plastic production and local population in both cases\(^{63}\) (Figure 4). These relationships suggest a tight coupling between microplastic sedimentation and global production as well as household plastic usage and waste. This coupling is also highlighted in the microplastic budget for Mjøsa catchment where microplastics from municipal sewage sludge and from household mismanaged waste are the main environmental releases. However, the proportions of terrestrial microplastics being transported downstream or buried in freshwater sediments may differ from one catchment to another. Indeed, Lake Geneva was argued to represent a considerable sink for terrestrial microplastics,\(^3\) whereas Lake Mjøsa primarily acts as a transporter of microplastics which potentially reach the ocean (Figure 4). As discussed above, the microplastic emission pathways differ among both, Geneva and Mjøsa, catchments resulting in contrasted microplastic sedimentation processes. Hence, the proportion of terrestrial microplastics being trapped in freshwater sediments seems to be partly controlled by the nature of the local microplastics, and by extension, by their origin.

### ASSOCIATED CONTENT

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c08443.

Supporting note on sediment dating; Figures S1 and S2 and Tables S1–S6 (PDF)
Microplastic data set (XLSX)

■ AUTHOR INFORMATION

Corresponding Author
Francois Clayér — Norwegian Institute for Water Research (NIVA), 0349 Oslo, Norway; orcid.org/0000-0003-6939-400X; Email: francois.clayé@niva.no

Authors
Morten Jartun — Norwegian Institute for Water Research (NIVA), 0349 Oslo, Norway
Nina T. Buenaventura — Norwegian Institute for Water Research (NIVA), 0349 Oslo, Norway
Jose-Luis Guerrero — Norwegian Institute for Water Research (NIVA), 0349 Oslo, Norway
Amy Lusher — Norwegian Institute for Water Research (NIVA), 0349 Oslo, Norway; orcid.org/0000-0003-0539-2974

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.0c08443

Author Contributions
F.C., A.L., and M.J. conceptualized the project. A.L. and N.T.B. analyzed the samples for microplastics, and performed the FTIR analysis and preliminary data analysis. F.C. performed the GIS analysis, statistical analyses, wrote the original draft of the manuscript, and drafted the figures. J.L.G. developed the catchment delineation tool, assisted in the GIS analysis, and reviewed the manuscript. A.L. edited and reviewed the manuscript. M.J. provided project funding, managed the project, and reviewed the manuscript.

Funding
This study was funded by the Norwegian Environment Agency (Miljødirektoratet)

Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We thank David P. Eidsvoll, Jan-Erik Thrane, and Asle Økslensrud for laboratory and field work assistance. We are also grateful to Rachel Hurley, Heleen de Wit, Sondre Meland, and Elizabeth Redland for stimulating discussions and revisions of a preliminary version of this article.

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