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(Micro) plastic fluxes and stocks in Lake Geneva basin

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ABSTRACT

High amounts of macro and microplastic have been reported in rivers, lakes and seas. However, links between the observed pollution and their sources remain unclear. This study aims to clarify these links in the Lake Geneva basin by analysing each step of the local plastic life cycle.

Two distinct approaches have been compared: (i) a top-down approach based on modelling socio-economic activities, plastic losses and releases into the lake, and, (ii) a bottom-up approach based on extrapolating plastic flows into the lake based on field measurements from 6 different pathways.

The two approaches yield results with similar orders of magnitude and provided a first estimation of the plastic flow from land to Lake Geneva in the order of magnitude of 55 tons year^{-1}.

Preliminary mass balance of plastic in Lake Geneva indicates that the vast majority of plastic may be deposited into the sediments.

1. Introduction

Plastics are used worldwide, with consumption rates increasing steadily since the 1950s [1]. In 2016, 335 million tons of plastic were produced globally, 60 million tons of which were produced in Europe [2]. Some of these plastics are inadequately managed and end up in the environment either in the form of mismanaged waste [3] or directly from the life cycle of some products such as tyre and road painting abrasion [4,5], textile washing [6], and cosmetics [7] through shedding, erosion or intentionally dispersed microplastics. For a review of the contribution of these different sources through a global inventory of plastic flows leaking to the oceans, see Refs. [8,9].

This ever-increasing volume of plastic entering oceans, rivers and lakes is a major concern due to the potential environmental impacts on biodiversity and ecosystems, as well as impacts on human health [10,11]. These impacts are caused by different effects such as entanglement, ingestion and toxicity [12,13] and accumulation in the food web [13,14]. As such, plastic pollution has become an increasingly pertinent issue requiring a better understanding of its sources, fate and pathways [15].

Mismanaged plastic waste generally consists of macroplastics, i.e. plastic above 5 mm diameter that may in turn be fragmented into smaller pieces (secondary microplastics), once exposed to environmental conditions [16]. These secondary microplastics are complemented by the so-called primary microplastics originating from different sources, and defined as plastic entering oceans or waterways already smaller than 5 mm [8,17,18]. Secondary microplastics and well as primary microplastics arising from textiles are described as more abundant in densely urbanized areas [17,19], whereas some forms of primary microplastics (e.g. plastic pellets) are more often found in regions where industries are located [20,21].

However, the precise sources and pathways of plastic pollution as well as the precise quantities and fate of plastic accumulated remain uncertain. When it comes to the quantities of plastic entering or accumulated in the oceans, the literature reveals contrasting data. Two streams of research co-exist: one based on modelling inputs and one based on field measurements. However, these two approaches currently do not match and yield results which differ at a scale of several orders of magnitude. On the one hand, global model estimates of the yearly input of plastic into the ocean range from 9.5 million tons per year [8] to 12.2 million tons per year [9]. On the other hand, measurement-based global

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estimates of the amount of plastic floating in the oceans are in the order of 250’000 to 300’000 tons [22,23], almost two orders of magnitude below the predictions of annual inputs from the models. This “missing plastic” is a matter of debate in the scientific community and it is unknown whether the plastic may sink and hence accumulate in the deep sea and not be measured by surface sampling [24,25], and/or may be accumulated in the food web [26]. Another hypothesis to bear in mind is that contemporary methods are not suitable for the detection of very small particles or particles maintaining certain characteristics, thus biasing the comparisons. A better insight on this discrepancy would require comparison of both approaches (sampling vs modelling) on a smaller scale, such as a lake with a well-defined watershed.

We therefore propose in this research to focus on Lake Geneva (known as Lac Léman in French). Recent studies have demonstrated significant plastic pollution in Swiss waters and in this lake in particular [27–29]. The watershed has a surface of 7999 km^2 and a population of approximatively 1 million people (~10% French and ~90% Swiss) [30]. The main outlet is through the Rhone River located at the south western part of the lake in Geneva, with an average flow of 250 m^3 s^{-1} [31]. Although Switzerland is believed to have a more efficient litter management system than other countries, a recent publication discovered high plastic contamination with hazardous substances, essentially non-authorized metals, suggesting that plastics remain in the lake for an extended period of time [58]. Indeed, research focussing on plastic contamination of freshwater has been increasing recently showing high contamination of densely inhabited and developed areas [32], as well as more remote lakes without adequate waste management facilities [33]. For a review, see article published by Wu et al. [32].

The present research aims to determine the respective contribution of different sources in the plastic pollution of Lake Geneva (Switzerland and France) and to compare results from modelling (top-down) and field studies (bottom-up). By articulating a first estimation of the annual input and stocks of plastic in Lake Geneva, it brings a novel insight and understanding on the fate and pathways of plastic, and explores methodological gaps.

2. Methods

The study methodology is based on two distinct streams of work: (1) a top-down approach, consisting of modelling plastic fluxes into Lake Geneva based on inquiries of socio-economic activities; and (2) a bottom-up approach consisting of estimating the plastic fluxes based on field measurements.

An assessment of plastic stocks accumulated in the different compartments areas of Lake Geneva was also performed based on compilation of concentrations reported in literature for surface water, sediments and accumulation on the shoreline.

2.1. Top-down approach (modelling the losses and releases from 11 sources)

The modelling was based on a life-cycle perspective that can be separated into 3 main steps for plastics: production (including primary production and plastic conversion), use and end-of-life. At each of these stages, different sources of plastic may be responsible for emissions into the environment. Plastic enters the environment through diverse pathways, such as waste-water treatment plants (WWTP) and road runoff waters. The general procedure applied in this study, to link the pollution to a potential source, can therefore be split into 3 main steps consisting of: (1) estimating the plastic Fluxes (i.e. magnitude of the different sources in the plastic consumption and production processes); (2) estimating the Losses (from the plastic consumption and production processes); (3) estimating the Releases (into waterways).

The data and measurements collected for the calculation are given in Supplementary Information. For each source a range of values was reported in the form of a mean value plus higher/lower bounds (see Appendices 1, 2 and 3).

Step 1 Quantification of the magnitude of the plastic sources within the river basin (detailed calculations are provided in Appendix 1)

The quantification of plastic sources was different for the three stages of the plastic life cycle i.e. production, use and end-of-life.

The production of plastic was deduced as a function of the number of plastic industries and the quantity produced by those industries based on inquiries and visits to different factories, enterprises and administrations.

The consumption i.e. the amount of plastics used per person, was extrapolated from the density of the human population living in the river basin, for the different domains: packaging, construction, tyres, textiles, electronics and cosmetics.

Finally, the end-of-life treatment was obtained separately for the two countries (Switzerland and France). The percentage of waste separated, recycled and incinerated was derived from official statistics for Switzerland [34] and France [35], and was extrapolated directly for the basin based on its population.

Step 2 Loss estimations (detailed calculations are provided in Appendix 2)

Plastic industry: the losses in production were based on records from the plastic converting industry, as there is no primary plastic production taking place within the river basin. Losses were quantified based on the reported spills occurring during unloading of primary plastic as they are delivered to the industrial sites.

Packaging: for losses linked to packaging, the amount of waste cleaned by the community (officially or through private initiatives) was subtracted from the amount theoretically littered. Accordingly, what hasn't been picked up can be considered lost in nature. In Switzerland the proportion of waste collected on streets is 30% [36], 70% being collected in public bins. The percentage of plastic in littered waste is 11.6% [37].

The amount of plastics picked up (in urban and rural areas) by the community within the river basin was obtained from different cities. The amount theoretically littered in urban areas was deduced from a study by Heeb et al. [37]. The mass of plastics theoretically littered (or illegally tipped) in the countryside was calculated from information obtained by cooperatives or environmental associations such as COSEDEC (*Coopérative Romande de Sensibilisation à la gestion des déchets* - http://www.cosedec.ch). The fraction of littered plastic not collected was adjusted in order for the mean leakage quantity for this source to correspond to the plastic quantities deposited on urban lake banks and harbours, estimated at 10 tons year^{-1} from the data of the main Lake Geneva clean-up initiative (*Net’Léman*) [38].

Construction: plastic losses in this domain arise primarily during the building phase. Indeed, once plastics have been integrated into buildings, almost no losses occur until the deconstruction/demolition, a process not considered in this model. The estimation of plastic losses was thus based on construction sites and is comprised of two steps. Firstly, the proportion of different types of plastic evacuated during rain events was determined through sampling runoff water from a building yard, allowing to identify that expanded polystyrene (EPS) was the major source. Other types of plastics are also used in the construction domain. The three most used categories are polyvinyl chloride (PVC), polyethylene (PE) and (expanded) polystyrene (PS) [2]. PVC for example is used in pipes and window frames and polyethylene in other resistant plastics. EPS is principally used for insulation and undergoes a polishing step during the installation, which emits a lot of plastic particles into the environment [39,40]. Then, in a second step, the amount of plastic from the construction sector was extrapolated by focussing on EPS only. Even though this approach can be considered as an underestimation, the polishing of EPS represents the biggest loss, which justifies this procedure.

Automobiles/tyres: concerning the automobile industry, losses are resulting from the erosion of car tyres while driving. The rubber used is partially natural and partially synthetic. Synthetic rubber can be considered as plastic and the profile erosion results in a loss of plastic into the environment. Emission statistics published elsewhere were used for this study on a per km basis [5].

Road paintings: road markings are increasingly being made with paints containing Methyl methacrylate. The exact quantity of these paints applied in the river basin are poorly quantified. Thus, estimating the amounts of plastic loss arising from road paintings remain difficult and highly uncertain. By knowing the road surface in the river basin [41], the approximate proportion of the roads covered by paintings and the mass of painting applied per area, an estimation of the quantity of road painting was derived. Consequently, an estimation of the amount of plastic lost through that domain was produced, considering the predicted lifetime of road paint.

Textiles: losses from textile washing were quantified based on generic washing habits per household [42], share of synthetic textiles in the region and fibre shedding rates reported in the literature [6,43].

Agriculture: losses from agriculture were based on plastics used for the protection of crops. Plastics used in protected cultivation are exposed to weathering and represent a large proportion of plastics used in agriculture [44]. Professional opinion indicated that plastic losses occur only when plastic covers are forgotten in the field. After a certain amount of time, these plastics disintegrate and fragments are left in the environment. Therefore, depending on the amount of plastic lost annually in the field and the fraction of that plastic that degrades prior to removal, plastic losses were estimated.

Cosmetics: numerous cosmetic products such as scrubbing shower gels or toothpaste may contain plastic microbeads. The quantities used per capita in the region of interest were deduced from values reported elsewhere [8].

Sport & Hobbies: various hobbies and sports can potentially emit plastic into the environment. Equestrian sand riding areas for example often mix synthetic materials with the sand, which could be transported away through rain action. The estimation of plastic losses for this domain was based on the amount of synthetic material added annually. The contribution of artificial lawns for kindergarten and sport facilities remains unknown and were not quantified herein.

The release of balloons into the atmosphere is another problematic activity. An estimation has been drawn on the number of balloons released in the river basin and their mean weight. Evidently, it is possible that released balloons could leave the Geneva Lake watershed and land in another, but it was assumed that the same amount would enter the Geneva Lake watershed after having been released elsewhere.

Fishing activities also significantly contribute to plastic pollution in water bodies. Plastic losses of plastic from these activities were quantified in terms of plastics emitted by individual recreational anglers. Knowing the amount of recreational anglers in the river basin and the mass of plastic lost per angler (based on interviews with fishing association), an estimation of the total losses was produced.

Not considered: household appliances, medical waste, toys, furniture and domestic equipment haven't been considered due to their predicted low contribution to plastic losses.

Step 3 Release estimations (detailed calculations are provided in Appendix 2)

The release was modelled as the quantity of plastic loss reaching surface waters, the remaining quantities accumulating in other compartments of the environment such as soils or the atmosphere. Release can occur through different pathways as described in previous studies [8].

Mismanaged waste: mismanaged waste includes waste that is not collected e.g. resulting from littering. A fraction of mismanaged waste is prone to being released into the environment, but the precise quantities are difficult to predict. Jambeck et al. estimated a loss-rate of mismanaged waste ranging from 15 to 40%, with a mean value of 25% [3]. These values were used in the present study. The same release ratios were used for construction waste as they are more appropriate than those for road runoff pathways, with construction waste generally maintaining a much lower density than tyre and road wear particles.

Road runoff: tyre dust and road markings are released through road runoff pathways or aerial routes. Recent studies have modelled the fate and transport of tyre and road wear particles and demonstrated that the proportion of these particles reaching surface waters ranges from 6 to 18% [5,45], with the final proportion released into the ocean in the order of magnitude of 2% [45]. Based on these studies, and assuming similar characteristics of the watersheds, we used release rates between 2 and 18%, with mean value of 6%. Road runoff was used as a proxy for the plastic released from equestrian riding areas.

Sewage systems: we calculated the capture rate of microplastics in sewage systems based on two flows. (i) storm overflows are estimated at 3% of total sewage water for Lake Geneva watershed [46] (ii) the capture rate of treated water is estimated as 80–98% with a mean value of 92%, based on a study on Swiss WWTP [47].

Direct release: direct release applies for losses that occur directly within the waterways (e.g. fishing devices) and is 100%.

2.2. Bottom-up approach (extrapolating the releases from measured concentrations in 6 pathways) (detailed calculations provided in Appendix 3)

The Lake Geneva watershed covers the Swiss cantons of Valais, Vaud and Geneva and the French departments of Ain and Haute-Savoie. For this study we have identified 6 entry paths for plastic into the lake: (i) storm overflows, (ii) waste water effluents, (iii) urban runoff, (iv) river discharge (wet period), (v) river discharge (dry period) and (vi) atmospheric deposition.

Within this study, three urban runoffs and one storm overflow were sampled in wet periods (rainy day), and four waste water effluents were sampled directly (because of the prevalence of separated sewer system, the pluviometry of the sampling day is not expected to influence the plastic concentration in WWTP). Four rivers were also sampled, in wet and dry periods.

Sampling and analytical techniques were based on techniques previously described by the authors [27,48]. Effluents were sampled using a Manta trawl (300 μm mesh size). All samples were then sieved through different mesh sizes (>5 mm, 5 mm, 1 mm, 300 μm, <300 μm) to separate size classes of plastic. Small microplastics (<300 μm) were dried (60 °C during 24 h) before undergoing oxidation to remove residual organic matter (35% H_2O_2 with

0.05 M Fe ^{II} catalyst during 6 h). Some samples were analysed with FTIR for more specific identification of plastic polymers as defined elsewhere [49].

(i) *Storm overflows*: several samples were collected from the overflow at 5 different periods (Storm overflow of Capelard in Lausanne). The samples of storm overflow have been considered representative of the whole watershed and extrapolated based on the estimated yearly storm overflow discharge to the lake [30].

(ii) *Waste-water effluents:* four WWTP (Morges, Lully, Bex and Vidy), with slightly different treatment options, were sampled to obtain a representative picture of the whole drainage basin and extrapolated based on volume of water treated.

(iii) *Urban runoff:* three urban runoffs were chosen to cover areas of different levels of urbanisation and none of the samples analysed were collected during the first flush of a rainfall event (Storm overflows from Chemin du Temple and Avenue de Gottettaz in Lausanne, and Penthaz on the A1 highway, during a rain event – 3 samples each).

(iv) *River discharge (wet period)*: by sampling 4 rivers during wet periods and knowing the average yearly rainy days, the amount of plastics carried away by the water could be estimated (by subtracting the urban runoff and the storm overflow due to their contribution during rain events). Plastic discharge during wet period is expected to be representative of both surface runoff (littered plastic and plastic on river banks that is flushed out during these events) and possibly the remobilisation of plastics stored in river sediments [50].

(v) *River discharge (dry period)*: similar to the surface runoff, the direct emissions could be estimated by using the concentrations of the rivers, obtained through sampling. Indeed, the supply from the rivers during dry periods takes the direct reject into account. Therefore, by removing the waste water effluents from the supply of the rivers during dry periods, an estimation of the direct reject could be obtained. Four rivers, among Lake Geneva's most important, were sampled: the Rhone, the Venoge, the Aubonne and the Vuachère.

(vi) *Atmospheric fallout:* the mass of plastic entering the environment through atmospheric fallout was measured directly and extrapolated across the watershed using relationships from a study in the city of Paris [51]. Data was collected from three locations, urban (Lausanne), peri-urban (St Prex) and rural (Ballen) areas, using 42 cm funnels. Dry and wet deposition was measured over 50 days [52]. Plastic supply is assumed to be dependent on population density, allowing for extrapolation across the Lake Geneva river basin.

2.3. Stock assessment in surface water, sediments and the shoreline

To complement the flow approaches presented above and to allow the creation of a gross mass balance of plastic in Lake Geneva watershed, an estimation of the stocks of plastic in the different compartments (e.g. surface, sediments) was also carried out.

One such assessment focused on plastic debris in the benthic sediments of the lake. It was based on 12 samples taken in 2015 between 44 and 309 m depth, reporting plastic concentrations ranging from 0.1 to 1.3 g m^{-2}, with a mean value of 1.0 g m^{-2} (these values were re-calculated based on a sample volume of 1L collected on the 2.5 first centimetres of sediments) [53]. By multiplying these concentrations to the area of Lake Geneva (580 km^2), the order of magnitude of the quantity of plastic stored in sediments can be extrapolated.

The concentration of surface particles and the flow of plastic out of Lake Geneva at the Rhone river was obtained from data published elsewhere [27,48].

Shoreline plastic stock was extrapolated from the data published by Net'Léman [38].

3. Results

3.1. Quantifying the mass of plastics contained within Lake Geneva's watershed

Plastic production in the Lake Geneva basin equates to 173,000 tons $year^{-1}$, mainly led by plastic converting industries as no primary plastic production is undertaken. Part of this production is exported and other plastic is imported for use in the area. Plastic usage in the area reaches close to 135,000 tons $year^{-1}$ with main contributors being packaging and the construction industry (Fig. 2). Agriculture, sports and hobbies are all contained in “others”, alongside with the medical domain, tools, furniture and domestic equipment. This high contribution in terms of mass consumption does not signify major losses or releases.

As shown in Fig. 2, the plastic mass balance for the Lake Geneva basin does not reach an equilibrium. Total production is larger than

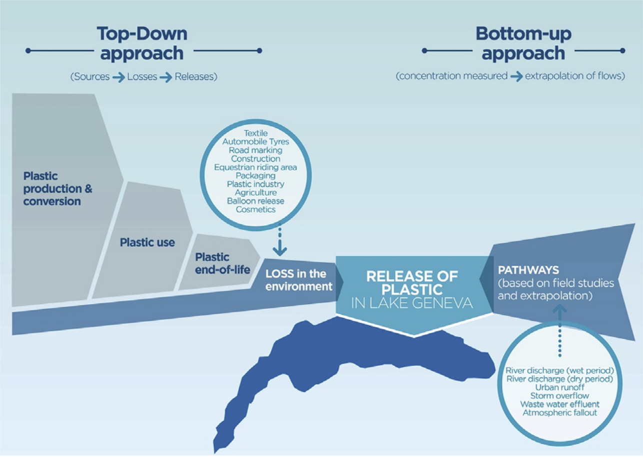

Fig. 1. Concept of the modelling used throughout the study - comparison of a top-down approach based on modelling of activities and a bottom-up approach based on field measurements and extrapolation.

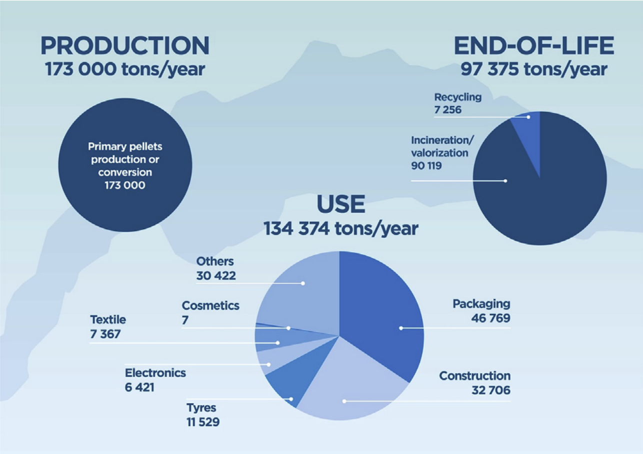

Fig. 2. Production, use and end-of-life of plastic in the Lake Geneva basin.

consumption while consumption is even larger than end of life treatment. This difference is due to the import and export of plastic materials from the watershed – not represented in the model.

3.2. *Loss estimations based on top-down modelling*

The loss of plastic in the environment from activities occurring in Lake Geneva watershed is estimated at 288–888 tons year^{-1}, with a mean value of 610 tons year^{-1}. As shown in Fig. 3, this loss is dominated by tyre dust, with 508 tons released into the environment on an annual basis (254–671 tons year^{-1}). The contribution of tyres is an order of magnitude higher than other sources, but consistent with published literature showing loss rates ranging between 0.23 and 4.7 kg pers^{-1} year^{-1} [5,45,54] for natural and synthetic rubber together. Uncertainty on the magnitude of this loss is quite narrow due to straight availability of traffic statistics for the region and robust loss rates (confirmed by measuring the weight of a used versus new tyre). Textiles that are often cited as the prevalent source of microplastic pollution in developed countries, also contribute to the loss [8,9,56] in a significant manner, but with high uncertainty resulting from less well defined loss rates and a higher variety of garments and washing parameters. Interestingly, construction material and plastic released from equestrian riding areas are important sources too. The contribution of plastic turfs from equestrian riding areas has also been stressed by other authors [57] as a significant source of microplastic in developed countries. Losses from fishing activities and balloon release are 2 orders of magnitude below the equestrian input and 4 orders of magnitude below the loss from tyres. Note that if this is true at a global level, at the individual level these practices can contribute to the individual footprint in a very significant manner i.e. in the order of several tenth of a gram per year.

The loss from packaging seems to be the second largest contributing domain to plastic pollution. Since the estimation of packaging has been based on littering, it can be deduced that human attitudes to waste disposal maintain an important influence on the quantities of plastic released into the environment (39 tons year^{-1} for the Lake Geneva watershed). The uncertainty on estimating the magnitude of the leakage from this last source is also very high due to the inherently behavioural and poorly documented leakage pathway.

A recent study by Fillella & Turner (2018) shows that beached plastics on Lake Geneva shores are a heterogeneous assortment of primary and secondary plastics and foams, coupled with a plastic pool that is dominated by polyolefins and with a relatively low abundance of higher density materials like PVC. Unlike marine studies, they found no primary production pellets and very little filamentous commercial fishing waste in the collected material, a proportion which supports our findings [58].

These results should be considered with caution as some domains were not exhaustively analysed. Some studies in Northern countries covered by snow in winter, for example Sweden, indicate a very high contribution of artificial lawns to plastic losses [57].

3.3. *Release estimations based on top-down modelling*

The estimation of releases to Lake Geneva was based on applying release rates to the different losses, dependent on the pathways: road runoff, sewage water, mismanaged waste and direct release.

The release of plastic into the environment resulting from the different losses described earlier has been estimated to range from 8 to 193 tons year^{-1}, with a mean value of 49 tons year^{-1}. Again, the release is dominated by the contribution of tyre dust, with 30 tons being discarded into the lake on a yearly basis, assuming a 6% release of tyre and road wear particles [5,45].

The total release is in the order of magnitude of 0.03% when compared to the quantity of plastic used in the basin (134,374 tons year^{-1}). This ratio is very low when compared to figures reported at world level by Jambeck et al. [3] with an estimated 8 million tons released out of 275 million tons of plastic produced worldwide in 2010 (2.9%). This relatively low leakage is likely due to efficient waste management systems across Switzerland and France. The leakage from plastic waste and packaging is expected to mainly result from littering, which is a behavioural phenomenon for which the quantities of plastic leakage are difficult to assess. Jambeck et al. [3] used a 2% littering ratio for packaging waste, which is 2 orders of magnitude higher than the ratio that we yield for the lake Geneva area i.e. 0.1% (this estimation corresponding to our higher range 43/46769 tons year^{-1}). This low leakage rate from mismanaged waste would still correspond to a 10 g pers^{-1} year^{-1} input of plastic across the Lake Geneva watershed or 20 g pers^{-1} year^{-1} for the higher estimation, quantities respectively close to the weight of 3 grocery bags or two water bottles.

The contribution of the different sources to the release of plastic are presented in Fig. 4, with the four highest contributions being the tyres, the construction sector, the textile industry and the packaging industry. These 4 main sources account for 93% of the total (87–94%). Emissions from the textile industry are characterised by high uncertainty mainly arising from the uncertainty in fibre shedding rates (the loss) and the capture rate in waste water treatment plants.

Tyre dust releases are also bound to uncertainties due to the release ratios of road runoff to surface water and the lack of reported measurements to calibrate transfer models [45,59]. However, the high proportion of separated sewer systems in Switzerland (rain water is not mixed with grey and black waters) calls for high release rate from road runoff waters as also evidenced by our measurements (see below bottom-up approach).

3.4. *Quantification of plastic found in the environment (bottom-up approach)*

The yearly input of plastic into the lake is estimated to range from 34 to 83 tons year^{-1}, with a mean value of 59 tons year^{-1}. Fig. 5 shows the proportion of plastic supply from the six different entry paths, with the stronger contributions from road and urban runoff.

Modelled values (Figs. 3 and 4) and measured values (Fig. 5) were coherent. Results for the two approaches are in the same order of magnitude. This does not implicitly indicate that the

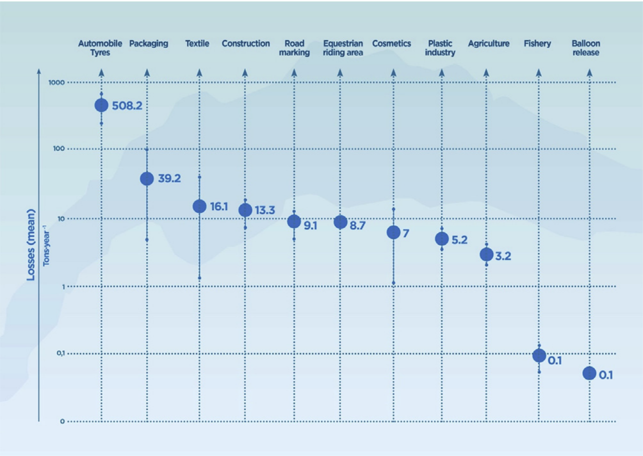

Fig. 3. LOSSES of plastic in the Lake Geneva watershed; contribution of the different sources (log10 scale).

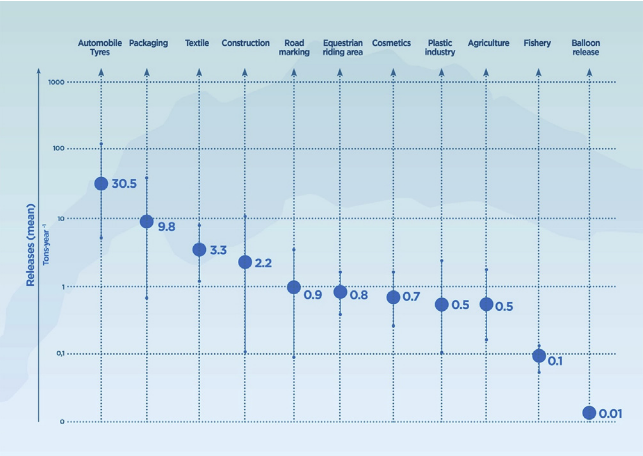

Fig. 4. RELEASES of plastic to the Lake Geneva; contribution of the different sources (log10 scale).

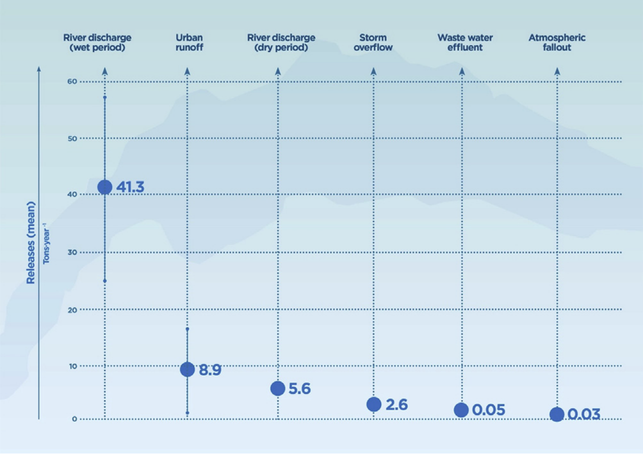

Fig. 5. Representation of the relative contribution of the different release pathways based on field measurement.

estimations are justified, but does imbue a certain level of reliability in modelling approaches.

Data from Fig. 5 indicates that the effluents of the WWTP appear to have a much lower contribution than almost all other pathways. This low contribution signifies that the waste water treatment plants are effective in reducing total plastic pollution. However, this deduction has to be taken with precaution as the majority of microplastics smaller than 300 μm could be emitted from the WWTP without being detected by the usual sampling procedures [60,61]. The contribution of storm overflows (3%) is important and should be considered as a hotspot to prevent any further contamination through the sewage system. Furthermore, it can be observed that atmospheric fallout does also provide a marginal contribution alongside the storm overflows. Clearly the major plastic supply to Lake Geneva comes from rain runoff (Fig. 5). Both river discharge and urban runoff together contribute to about 86% of total annual plastic flux, with the strongest contribution stemming from rivers in wet periods, as already demonstrated by other authors [50]. This dominant contribution indicates the importance of water in transporting plastics, with the main polymers recovered being PS, HDPE, LDPE and PP. Similar conclusions concerning the prevalence of runoff for transporting plastics have been made by Cheung et al. [62] (measuring a significant difference between the amount of plastics found on beaches during dry and wet periods) and Hurley et al. [50] (demonstrating the remobilisation and flushing of plastic particles from sediments during flooding events). It must be noted that similar field observations have also been made in Lake Geneva with higher concentrations measured in surface water in relation to rain events [27,48]. Furthermore, this prevalence of rain-runoff pathways observed in Fig. 5 is consistent with the results of the top-down modelling (Figs. 3 and 4) as the main release was described occurring through road runoff pathway i.e. the tyre dust and plastic from littering.

4. Discussion

4.1. Comparison of bottom-up and top-down approach

The novelty of this work consists in comparing two different approaches for estimating plastic input into Lake Geneva. Both approaches encompass large uncertainties but yield results in a similar order of magnitude - mean value of 49 (8–193) tons year^{-1} from modelling and 59 (34–83) tons year^{-1} from field measurements. Using data from both approaches, we estimate that 55 tons enter Lake Geneva every year, corresponding to a per capita input in the region of approximately 55 g pers^{-1} year^{-1}.

Caution must be applied when comparing the two approaches, due to methodological differences relating to plastic size distributions. The top-down approach is a global approach accounting for plastic and microplastics of all size, whereas the bottom-up approach is based on field measurements and thus only accounts for a restricted size distribution (>300 μm to <5 mm).

Tyre particles have a small size, mainly in the range 5–25 μm. This size range explains why no tyre particles were sampled with the Manta trawl (mesh size >300 μm). Furthermore, these particles have a higher density than water. Therefore, unless water is highly turbulent, surface sampling may not encapsulate such particles. Since these particles haven't been sampled in the context of this research, it isn't possible to compare them to the pollution measured in the environment. Tyre dust and textile fibres have subsequently not been accounted for in the bottom-up approach, while they represent the highest fraction of modelled plastic flow. Without tyre dust and textile contributions, the mean input into the lake obtained from modelling is 9 tons year^{-1}. This can be interpreted as an underestimation of other sources such as littering, or a real mismatch between models and measurements due to the low accuracy of the two approaches.

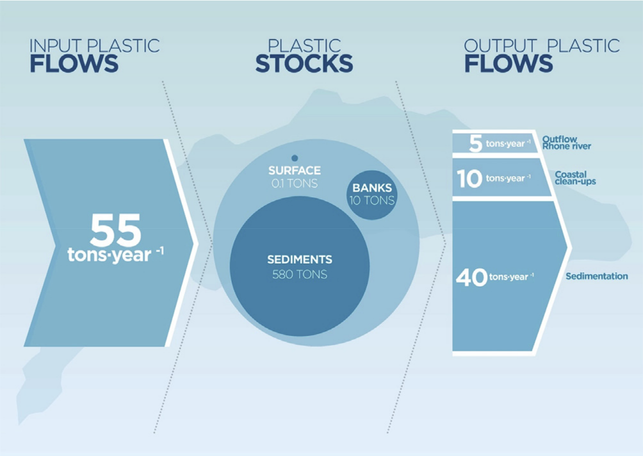

Fig. 6. Preliminary mass balance for plastic in Lake Geneva based on own measurements and other references. This diagram shows plastic flows and stocks for different compartments [27,38,48,53].

A comprehensive plastic mass-balance for the lake would help our understanding of plastic transfers in watersheds. An attempt to put together a first version of such a model is proposed in the next paragraph, by comparing the results from our research with other recent publications.

4.2. The fate of plastic in Lake Geneva — an attempt of mass balance

The estimated 55 tons of plastic entering Lake Geneva each year can potentially end up on the shores, in surface water, on the bottom of the lake, accumulated in biota or be evacuated by the single outflow, the Rhone River. Fig. 6 presents our current understanding of these flows and stocks.

Plastic floating at the surface of the lake has been measured in concentrations ranging from 46 ± 25/10 ± 11 g km^{-2} for microplastics and 44 ± 33/27 ± 23 g km^{-2} for macroplastics, respectively for Grand lac/Petit lac [27]. These particle concentrations are somehow lower than most marine plastic pollution reported at the surface, e.g. above 500 g km^{-2} average for the Mediterranean basin [63]. The lower buoyancy of plastic particles in freshwater and moderate plastic leakage within Lake Geneva basin as compared to some countries with less adapted water treatment facilities may explain some of this discrepancy. Attention must be paid to the fact that these measurements excluded coastal zones and samples taken after rain events. When extrapolated to the surface of the lake (580 km^2), the estimated stock is very low: 0.1 tons. As already reported by other authors in other regions, it seems that rain events have a strong influence on measures of surface concentrations [64]. In the same zones and under similar wind conditions, microplastics were 4.4 times more abundant in number and seven times heavier after large rain events, and 9 and 9.5 times respectively for macroplastics [27]. These estimates of the surface concentration and the emissary signify that the plastic debris lost from various sources are deposited elsewhere or is not measured by contemporary methods (e.g. due to fragmentation into smaller size fractions). Smaller plastics could be distributed throughout the water column (that has not been sampled at different depth in Lake Geneva). However, similar measurements at sea tend to show that most of the floating plastic lies within the first meters of the water column [65,66], a phenomenon that is expected to be intensified in freshwater environments as a result of the low buoyancy of particles, by opposition with salt water.

The Rhone emissary (Chancy, Geneva) has been sampled in previous studies and the estimated evacuation was calculated in the order of only 5 tons year^{-1} [27]. Even though this value appears to be an underestimation, because of the strong local turbulence, it can be concluded that far more plastics are entering the lake (55 tons year^{-1}) than leaving it. Thus plastic seems to accumulate in the lake itself. Interestingly, the Rhone upstream also conveys a significantly higher plastic flux than the outflow, supporting the hypothesis that the lake acts as a sink for plastic [27,48].

The most probable hypothesis, already demonstrated at sea [67] is that debris sink to the bottom of the lake, which can be an accumulation zone [24]. Since large amounts of plastic produced have densities higher than water, including tyre dust and road wear particles, it is readily possible that a significant proportion ends up stuck on the lake bed. Other studies on deposition of microparticles in the Great Lakes also indicate a strong deposition of plastic fibres [68,69], with the same being found in ocean sediment samples [70]. Furthermore, other less-dense plastics may also sink and end up on the bed when colonized by a biofilm or ingested/excreted into/by some planktonic organisms [71–73]. The order of magnitude of the quantity of plastic stored in sediments extrapolated from CIPEL measurements [53] ranges from 75 to 740 tons, with a mean value of 580 tons. This gross estimation when compared with the annual flux of plastic entering the lake (55 tons year^{-1}) and the outflow measure on the downstream Rhone river (5 tons year^{-1}) suggests that Lake Geneva acts a sink for plastic. The plastic most commonly found in sediments is mainly constituted of thin layer plastics, probably resulting from packaging and plastic bags [53]. The mass of other types of plastics such as fishing lines, pellets or foams seems to be insignificant in sediment samples [53]. Additionally, these estimates are based on sampling techniques which do not capture smaller particles thus not accounting for tyre dust with most of particles having size between 5 and 25 μm [5]. Interestingly, when compared to the average sedimentation rate of 2 mm year^{-1} [74,75], we can extrapolate that the 580 (75/740) tons accumulated on the first 2.5 cm of sediments result from a 12½ year period, and a 46 (7/59) ton year^{-1} flow. A value of 40 tons year^{-1} has been chosen for the model, which corresponds to 72% of the input plastic according to the model.

Accumulation in biota has been proven, with particles found in fish, however first measurements based on fish stocks do not build up significant quantities [48]. Nevertheless, even small quantities accumulated in the biota could have strong adverse environmental impacts and the magnitude of plastic storage in this compartment is not an indication of the severity of the phenomenon.

Lastly, a huge amount of plastics is collected by associations or private persons on beaches, ripraps or in recreational harbours. However, the precise quantity of this plastic withdrawal is not measured and must be extrapolated. One beach clean-up (Net'Léman and ASL | *Association pour la Sauvegarde du Léman*) organised every two years in 10 locations around the lake have reported (mainly recreational harbours) collecting 1.2 tons of plastic (including 90 kg PET bottles and 200 kg tyres) [38]. By extrapolating to the 70 main harbours of the lake this would equate to 8.4 tons of plastic without accounting for beaches. A value of 10 tons has been chosen as a potential quantity of the plastic stored on the lake's shoreline at a given time and removed by beaches and harbours clean-ups on regular basis. Typical concentrations of plastic found on Lake Geneva beaches are around 20 g m^{-2} [48], without a proper extrapolation to the lake being feasible as a result of the high heterogeneity of the shoreline. This value is consistent with figures published by other authors mentioning that 5% of all plastic leakage ends-up on beaches. Interestingly, if compared with local plastic packaging use in the watershed — 46'769 tons year^{-1} (Fig. 1), and considering a 25% release rate for littered plastic [3], the littering rate can be estimated having a value of 0.1%, which is about 20 fold lower than the percentage of littering generally used in literature to model plastic leakage, i.e. 2% [3].

5. Conclusions and outlook

The novelty of this work consists of having coupled different approaches for assessing plastic flows and stocks in a relatively small geographic area. Based on different approaches, we produced the first estimates of the annual input of plastic from land to Lake Geneva: in the order of magnitude of 55 tons year^{-1}. This plastic leakage (approx. 55 g. pers^{-1} year^{-1}) is low when compared with other areas of the world and presumably mainly stems from microplastics (tyre dust). The precise contribution of littering in this leakage is highly uncertain and debatable, with our finding showing a lower contribution of littering than usually reported in the literature. Methods to quantify littering in a regionalised manner are required to articulate more robust leakage figures both at regional and global scales, and for shaping sound mitigation strategies. In spite of a relatively low leakage rate for Lake Geneva, these larger plastic wastes (plastic from packaging and plastic released from the construction industry) should not be underestimated as they represent visible plastic waste commonly found on local beaches.

This research also presents evidence to support the hypothesis that Lake Geneva acts as a sink for plastic with an accumulation in bottom sediments. From a more global perspective, this suggests the potential role of big lakes as a barrier preventing more plastic from reaching the oceans.

The approach presented in this research would benefit from replication in different areas and with denser sampling data in order to increase robustness on both the results and some of the key parameters that are used to model plastic leakage (e.g. littering rates and release rates for different pathways). Ideally methods should be developed for measuring tyre dust and also to trace the smaller fraction of particles size. Developing sampling and measurement along sewage systems and urban infrastructure should enable better understanding of the leakage pathways and potential solutions.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.trac.2018.11.037.

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