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**Transfer dynamic of macroplastics in estuaries – New insights from the Seine estuary:
Part 1. Long term dynamic based on date-prints on stranded debris**

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Abstract

Rivers are a major pathway for plastics between lands and the ocean. At the land-ocean interface, estuaries make the transfer dynamic of plastics complex and nonlinear. That is why very little is known about this dynamic. In this respect, a specific marker (i.e. Microlax packaging) showing date-prints was systematically investigated in different riverbanks of the Seine estuary to identify the share of “old” and “recent” litter transiting through the estuary toward the ocean. Up to 70% of Microlax were “old” plastic items probably related to the meandering dynamic of the river over large time and space scales, and hydrodynamic conditions (tides) at smaller scales. This contributes together to increase the residence time of plastics into the estuary up to decades with almost endless transport, deposit and remobilization cycles. Consequently, the Seine estuary may function as a “microplastic factory” resulting from the fragmentation of macroplastics into microplastics well before they reach the ocean.

Keywords: Residence time, Microlax, Use-By Dates, Sources, Pathways.

Introduction

The accumulation of plastic litter is widely documented worldwide in marine, coastal, estuarine and fresh water environments (Axelsson and van Sebille, 2017; Schneider et al., 2018; Schwarz et al., 2019). Plastic litter is mostly exported into the ocean through rivers (Lebreton et al., 2017; Schmidt et al., 2017) and plastic items larger than 5 mm in particular constitute a global threat for sea birds, fishes and marine mammals after ingestion or entanglement (Barreiros and Raykov, 2014; Kühn et al., 2015; Wilcox et al., 2015; Thiel et al., 2018). However, very little is known about the transfer dynamic of plastics through rivers and in particular through estuaries. Estuaries are at the land-ocean interface and are very specific hydrosystems characterized by alternate flow directions and the fresh and salt water mixing related to tidal dynamic (Perillo, 1995; Vermeiren et al., 2016). Because of the tides, water depth in estuaries changes and banks are alternatively submerged and uncovered, which enables the buildup of a foreshore made of natural and anthropogenic debris, including plastics, depending on their buoyancy. Estuaries are also subject to flood events related to intense rains and runoff in the catchment which causing increase of water level and the submersion of floodplains. This leads to the transfer of debris to areas far from the main channel, even disconnected ones. Other variables like vertical mixing, debris densities, or wind influence their dynamics, with greater accumulation of plastic on shorelines at downwind sites (Brown et al., 1991; Browne et al., 2010; Vermeiren et al., 2016).

Because of their specific behavior, estuaries may greatly influence the plastic debris dynamics. However, the origin, fate and typical residence time of plastics in those environments remain poorly understood, and up to now it is not clear whether estuaries function as net sources or sinks of plastic litter (Sadri and Thompson, 2014; Naidoo et al., 2015; Vermeiren et al., 2016; Schwarz et al., 2019; Krelling and Turra, 2019). More than sources or sinks, estuaries are pathways with a retention capacity of plastics and the question would be: “are estuaries fast or slow pathways for plastics?” In turn, the understanding of the transfer dynamic of macroplastics from land to the ocean remains unclear, although it is of crucial interest to settle efficient mitigation policies to monitor and reduce macroplastic pollution (Tencati et al., 2016; Axelsson and van Sebille, 2017; European Commission, 2018). Estimating the age of plastics found in the environment would help to understand whether plastics are just leaked to the environment and linearly transferred from upstream to downstream, or whether they are non-linearly transferred remaining in the environment for years. Usual methods for assessing the age of polymers in the environment (e.g. the analysis of FTIR spectra) are inefficient in estuaries as degradation processes (UV exposition,

hydrolysis, biodegradation) depend on transport and deposit conditions (Edge et al., 1991; Allen et al., 1994; Müller et al., 2001; Ioakeimidis et al., 2016; Weinstein et al., 2016)

An alternative approach is to analyze the age of debris on the basis of dates printed or engraved on them. For example, Sander (2016) mapped plastic items showing date-prints to reconstruct the timing and extend of overwash deposition on the shorelines of the Danish Walden Sea. He demonstrated that date-prints on stranded items can be used as indicators for deposition dynamic of litter on shorelines. Other authors (Johnson and Merrell, 1988; Williams and Simmons, 1996; Ivar do Sul et al., 2014; Krelling and Turra, 2019) used tagged litter deliberately released in the environment to assess the residence time of debris in the environment and their transport dynamic. They showed that debris retention varied much more with habitats and item behaviors – according to hydrodynamic conditions, vegetation height and density, beach or riverbank morphology –, than with seasons, with the exception of extreme events like storms, high tides or flood events.

Following those approaches, we propose a new method based on Use-By Dates (UBD) engraved on specific packages to assess the transport dynamics of plastic debris in estuaries, kind of like geologists use specific fossil assemblages to date and reconstruct paleoenvironments (Zalasiewicz et al., 2016). In this study, we focused on Microlax® packaging, which often show engraved UBD. It is a laxative with rectal administration belonging to the ATC-class A06AG11 (<http://base-donnees-publique.medicaments.gouv.fr/>). Its packaging originates mostly from sewer systems after toilet dumping and are transferred to the surface water system because of sewer system failure: misconnections on separate sewer systems, wastewater treatment plant bypass or combined sewer overflows during intense wet weather periods (Gasperi et al., 2014; Ellis and Butler, 2015; Rocher et al., 2017). The packaging design results in a very high buoyancy because of air entrapment inside. Consequently, they are frequently stranded on riverbanks in the Seine estuary among other plastic litter. The estuary of the Seine River is around 150 km long with an average annual water discharge of $\sim 500 \text{ m}^3/\text{s}$ in its upstream zone (<http://www.hydro.eaufrance.fr/presentation/procedure.php>) and a tidal range up to 7 m at the river mouth. According to its morphology and high tidal range, the Seine estuary is a tide dominated and well-mixed estuary with the mixing of fresh and salt water (Perillo, 1995; Vermeiren et al., 2016).

Several sites showing historical accumulation of plastic debris were punctually investigated together with one site regularly monitored to determine the distribution of UBD engraved on Microlax packaging. The comparison between the historical sites and the

monitored site over time aims (i) to identify the share of “old” and “recent” litter which compose the plastic load transiting through the estuary and (ii) to discuss whether the Seine estuary is a fast or slow pathway for plastic litter with consequences on undertaken solutions to reduce marine litter.

Material and methods

Targeted markers

Microlax were collected on riverbanks of the Seine estuary between March 2018 and April 2019. The packaging is made of polyethylene and did not change significantly since the 70'. Items are ~10 cm long with a pocket containing the medicine before administration and a cannula for rectal application. Throughout the manuscript, Microlax will refer to its packaging (see picture in Figure 1A).

The printed date corresponds to the “Use-By Date” (UBD), which is the limit of consumption of the medicine, i.e. 3 years. The oldest UBD recorded for Microlax is 1984, but the medicine received its marketing approval in 1977 ([https://ansm.sante.fr/searchengine/detail/\(cis\)/67367270](https://ansm.sante.fr/searchengine/detail/(cis)/67367270)). Thus, Microlax without any UBD corresponds to Microlax produced between 1977 and 1980, i.e. with a virtual UBD between 1980 and 1983. They were noted as “NA”. Few Microlax were too degraded to determine their UBD, or the UBD was erased. They were noted as “DIS” (for “Discarded”) into the database and were discarded for subsequent analyses. Microlax collected are considered as “recent” Microlax when their UBD range is: year of collection \leq UBD \leq year of collection + 3 years. The rest is considered as “old” Microlax. The overall data about Microlax are available here: Mendeley data, doi: 10.17632/4cp3dc8vrk.2

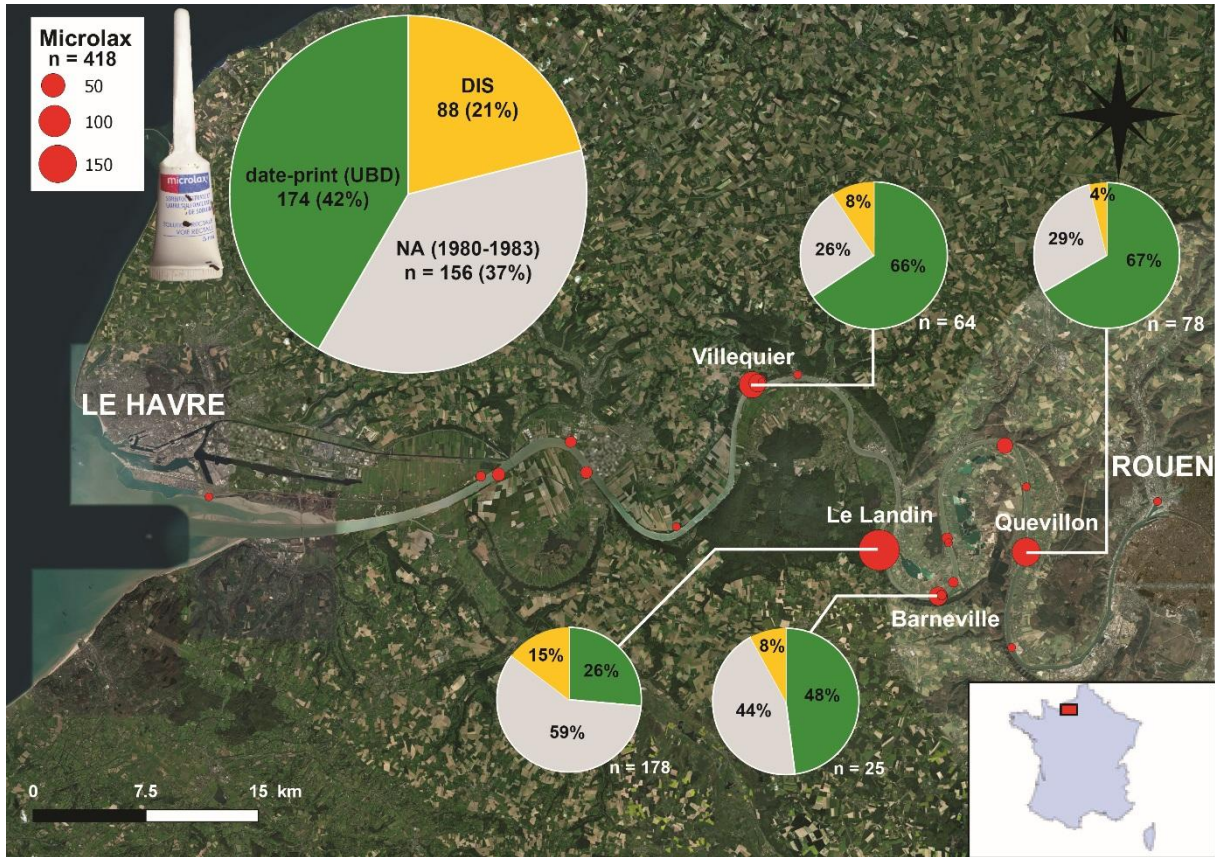
Sites of collection

Four sites were investigated: Quevillon, Barneville and Le Landin showing historical accumulation of plastic litter and Villequier, a site regularly cleaned by the non-lucrative landscaping company Nautaul'un (See sup. Data 1 for GPS coordinates). Microlax were collected once in the historical sites, which had never been cleaned before to our knowledge except for the largest debris, in April (Le Landin and Barneville) and July 2018 (Quevillon). Each site was visually inspected and Microlax were systematically collected by hands in the whole area. In Villequier, Microlax were collected on a regular basis by Nautaul'un during 31 cleaning campaigns, approximately once each 15 days between March 2018 and April

2019. Few other *Microlox* were identified in the estuary by participative sciences through an interactive map on <https://oceanplastictracker.com/#/> (**Figure 1A**).

Quevillon is an ancient dredging deposit chamber of 300 m long and 10 m width located on the right side of the estuary, created between 1979 and 1982 and abandoned between 1985 and 1989 (See aerial images in sup data 1). The chamber is now filled with organic debris and plastic litter. It is separated from the river system by a smooth levee with high vegetation and several gutters connecting the chamber to the river during high tides or flood events. Barneville is an active floodplain of 3 hectares located on the left side of the river, covered by vegetation and bordered by a chalk cliff toward the land. No levees prevent exchanges between the area and the river system, but large trunks and successive sedimentary ridges can act as temporary barriers to litter deposits. Le Landin is a small area of ~1 000 m² located on the left side of the river, bordered by an abrupt levee made of coarse sand and pebbles on its east and by a towpath toward the land on its west. This configuration makes the area a sink relatively disconnected from the river system but flooded by capillarity when water level gets higher. However, the high levee prevents most plastic debris to escape from the area. Finally, Villequier is a large and smooth foreshore bordered by an abrupt river bank of more than 1 km long on the right side of the river which forms a vegetated seawall. This site is under a strong tidal influence with daily exchange of debris between the river and the foreshore. Exchanges are facilitated by the absence of levees and large amounts of litter are daily deposited in and removed from Villequier. Aerial images of Quevillon and Le Landin are provided in supplementary data.

A)



B)

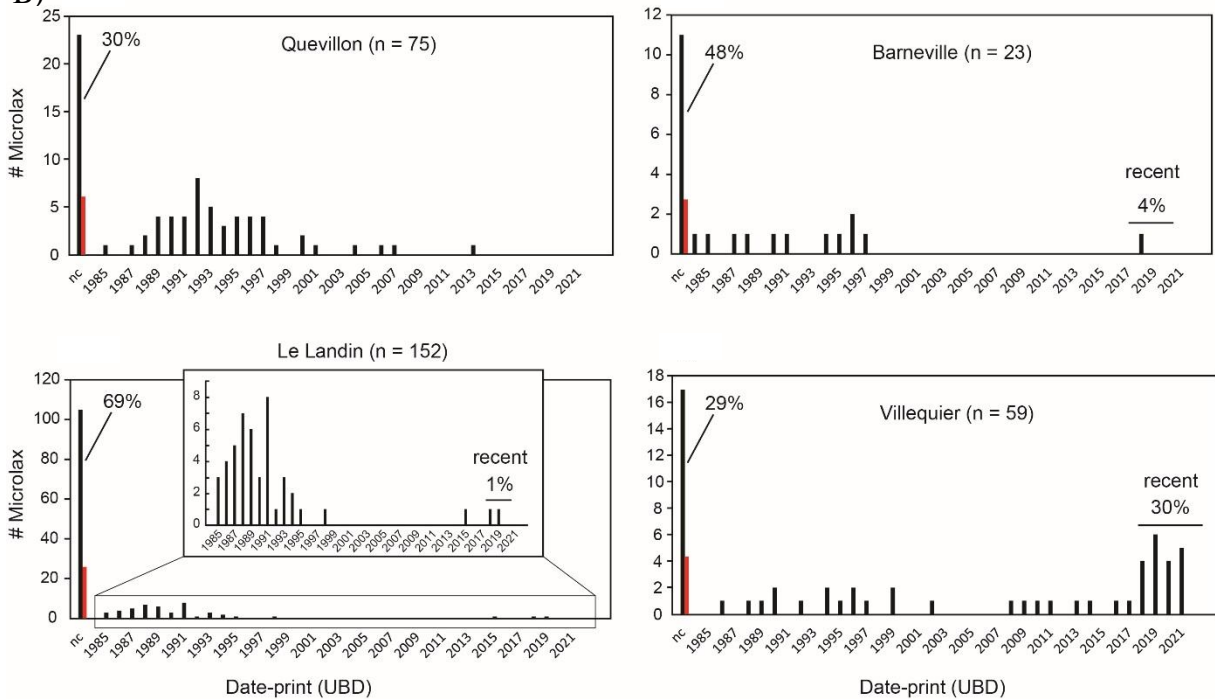


Figure 1: A) Distribution of the Microlax collected in the Seine estuary. For most of Microlax it was possible to assign a “Use-By Date” (UBD + “NA”). ”DIS”, for discarded, refers to Microlax without any assignable UBD. B) Distribution of the Microlax collected in the historical sites (one campaign) and in Villequier (31 campaigns). The red bar refers to the mean number of Microlax per year between 1977 and 1980.

The four main sites studied can be distinguished by their morphological behavior and historical evolution with differential river-floodplain connectivity. Over a small time scale, the connectivity is mitigated by tidal cycles, whereas over a greater time scale, it is mitigated by river meandering dynamic, river-riparian vegetation density and anthropic actions (Perillo, 1995; Perucca et al., 2007). Investigating relatively disconnected historical sites of accumulation and a site well connected to the main channel through the intertidal zone enables distinguishing how “old” litter and “recent” litter interact in an estuarine context.

Results and discussion

Between March 2018 and April 2019, 418 Microlax were recorded as stranded litter on the river banks. They were always found together with other plastic debris such as expanded polystyrene, cotton swabs, lollipop sticks, syringes and other unidentified plastic fragments that are common on riverbanks in the Seine estuary (Tramoy et al., 2019). Among the 418 Microlax collected, 281 were found in the historical sites of Quevillon (n = 78), Barneville (n = 25) and Le Landin (n = 178), and 64 were found in Villequier (**Figure 1A**). In total, 156 (37%) did not show any UBD (“NA”, i.e. virtual UBD between 1980 and 1983) and 88 (21%) were discarded from analyses because no UBDs were assignable.

In Quevillon, 30% of the Microlax were as old as 1977-1980 (“NA”). Microlax as old as 1977-1980 reach 48% of the Microlax collected in Barneville and even 69% in Le Landin (**Figure 1B**). When excluding only the “recent” Microlax, i.e. UBD post 2018/2019, the share of “old” Microlax in historical sites reaches 96% to 100%. Those “old” Microlax in historical sites constitute an ancient stock of litter ready to be released again into the river. Except in Le Landin, the mean number of Microlax per year between 1977 and 1980 is not very different from the other UBDs (**Figure 1B**). In contrast, 30% of Microlax in Villequier showed “recent” UBDs. Notice that a sampling bias cannot be ruled out because a very high amount of “old” Microlax was found in one historical site (Le Landin). Nevertheless, Microlax reflect past and actual inputs of litter into the Seine estuary that potentially remains for years.

Sources and pathways of Microlax

A common pattern in the distribution of UBDs is observed in the four study sites with more Microlax showing UBD from the 1980’ and 1990’ than from the 2000’ and 2010’ (Figure 1B). This pattern may come from the source and pathway of this waste. The presence of Microlax is more likely related to sewerage failures after toilet dumping. Toilet dumping is

a common behavior in every kind of population for decades and results in significant quantities of solid matter into the sewerage (Friedler et al., 1996). On catchments with combined system, overflows occur during wet weather periods (Rocher et al., 2017). According to Ellis and Butler (2015), the rate of misconnections to the surface water system typically reaches 3-4% in Europe and US, with possible local hot spots of 20-30%. Thus, on catchments with separate sewer systems, misconnections enable the transfer of wastewater and litter to the rain water system and directly to the receiving hydrosystem.

The *Agence française de sécurité sanitaire des produits de santé* (Afssaps, French drug administration) states that the sales of “laxatives” from the A06 ATC-class, the one Microlax belongs to, remain relatively unchanged between 1998 and 2008 (afssaps, 2010). Thus, the prevalence of “old” Microlax in the estuary results most probably from differential rates of leakage to the environment over time, than from changes in consumption. This is supported by a significant improvement in the sewerage over the whole catchment and a decrease in raw wastewater release since the 1970’ (**Figure 2**; Rocher et al., 2017). In addition, misconnections to the sewerage system (not taken into account in the Figure 2) have been tackled by successive laws in 2006 and 2010 leading to a mandatory diagnostic of the connection conformity before a sell of property is being approved (<https://www.legifrance.gouv.fr/>; LOI n°2006-1772 of the 30th of December, 2006; LOI n°2010-788 of the 12th of July, 2010).

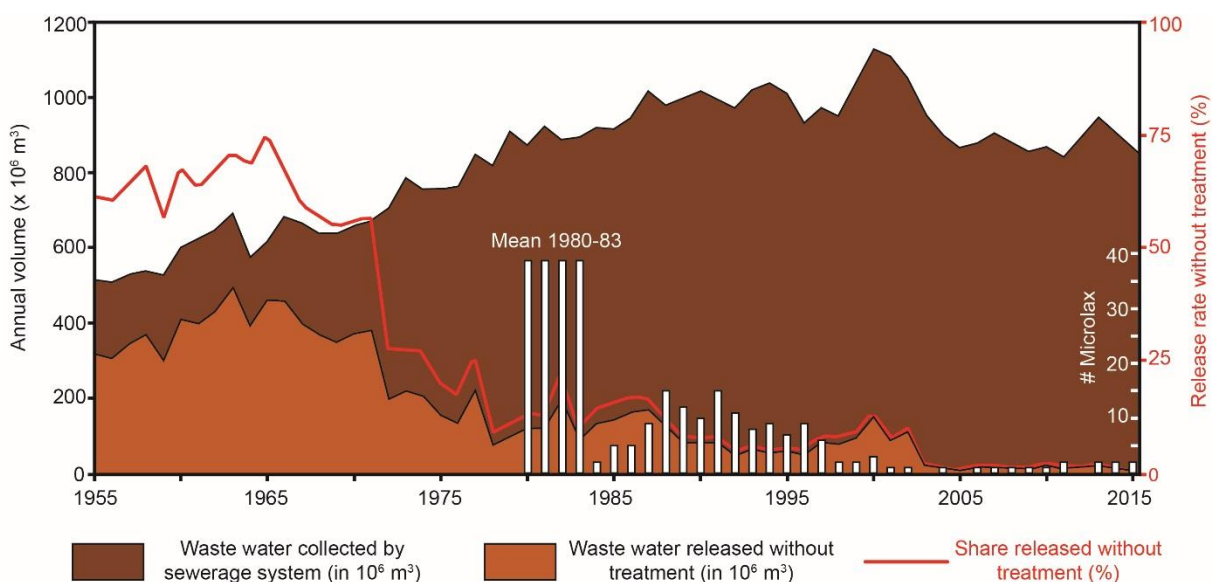


Figure 2: Wastewater collected and released without treatment for Paris megacity (modified from Rocher et al., 2017), with the amount of Microlax found in the whole estuary classified per UBD (n =330). The mean amount of Microlax (1980-1983) corresponds to the total

amount of Microlax without UBD (“NA”) divided by the number of years covering the production period of those Microlax (1977 to 1980).

Accumulation of Microlax in the historical sites

The key factor driving the UBD distribution of Microlax in each site might be the river-floodplain connectivity related to complex interactions between river meandering dynamic, river-riparian vegetation density and anthropic actions (Perucca et al., 2007). Aerial images from Quevillon in 1978, 1982, 1985 and 1989, (see sup. Data 1) show that the dredging deposit chamber did not exist before 1978-1982 and has been abandoned between 1985 and 1989. During this time period, litter started to accumulate after successive flood events until the site was relatively disconnected from the river system at the end of the 90’ thanks to the development of vegetation. This may explain the UBD distribution in this site, with an over representation of Microlax from the end of the 1980’ and the 1990’, when compared to the other sites (**Figure 1B**). Recently (2011-2015), the riverbank has been eroded toward the south and a part of the litter may have escaped to the river during high flood tides. In Le Landin, aerial images show the relatively natural evolution of this site back to the 1960’, with a probable maximum of connectivity to the river system (i.e., less dense vegetation) reached between 1973 and 1989 (see sup. Data 1). This time period also corresponds to the most numerous Microlax accumulated there (**Figure 1B**). The distribution of UBDs may thus be related to the morphological dynamic of riverbanks that controls connectivity into the floodplain river system. At the estuary scale, the meander dynamic alternatively connects and disconnects the riparian area from the river system which acts over time as a sink of “recent” litter or a source of “old” litter.

Transfer dynamic of floating macroplastics based on Microlax indicator

Among the 64 Microlax found in Villequier, 5 Microlax without assigned UBD were removed, 70% of the rest showed “old” UBD (**Figure 3**). This suggests that “old” plastics from historical zones of accumulation are frequently remobilized and deposited again from riverbanks to riverbanks, including Villequier. In contrast to historical sites, organic and plastic debris are easily deposited in Villequier with tides because there is no obstacles between the river and the gentle slope of its riverbank. Plastic debris deposited on this site are thus a sample of the whole plastic load flowing in the Seine estuary. Considering Microlax as a representative floating plastic debris, it can provide information about the transfer dynamic of the floating plastic load in the estuary.

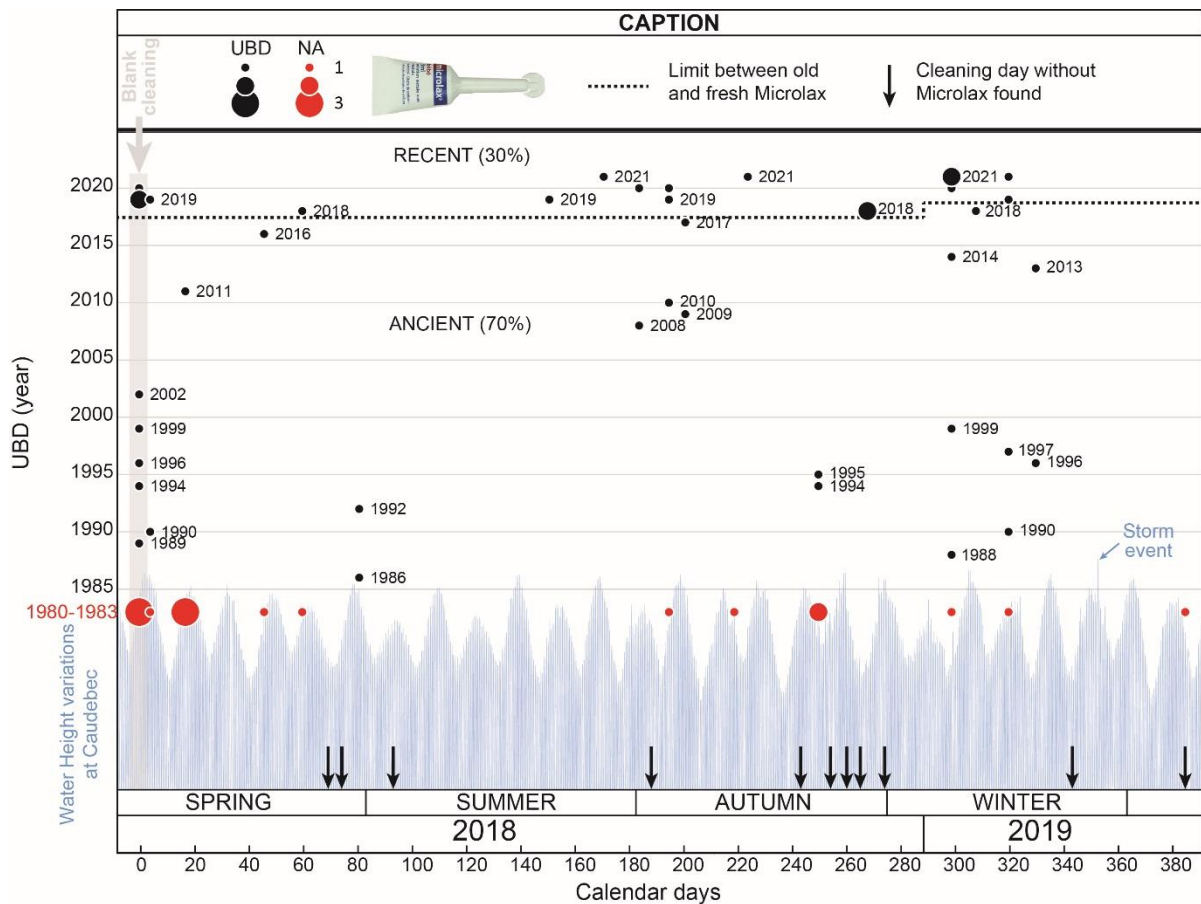


Figure 3: Distribution of the UBD engraved on Microlax relative to cleaning days on riverbanks in Villequier. The calendar day “0” corresponds to the 23th of March 2018.

According to Microlax collected in Villequier, up to 70% of the floating plastic debris may have spent more than 1 year in the estuary (**Figure 3**). As suggested by Brown et al. (1991), during flood tides, currents are faster mid-channel than on the edges because of friction with the riverbanks. This forms a flood tide convergence zone leading to downward currents, which subsequently move over the channel bed towards the channel edges. Then, retroactive currents form at the surface from the riverbanks to mid channel. Those currents collect floating plastic debris from the riverbanks and transport them to mid-channel. Because they do not sink, those plastics are then exported upstream. According to Vermeiren et al. (2016), the relative balance of the strength of the upstream transport, versus the strength of the downstream transport by fresh water outflow, determines whether partially and well-mixed estuaries are a net source or sink of floating plastics. Considering estuaries as pathways with a power of retention for floating plastics (Ivar do Sul et al., 2014), this assumption implies estuaries function as fast or slow pathways between land and ocean. The high proportion of

“old” litter like “old” Microlax collected in Villequier might point to the second option for the Seine estuary with the exception of periods of high discharges (Williams and Simmons, 1996; Krelling and Turra, 2019).

From a spatiotemporal point of view, the site of Villequier integrates large and small scales of time and space related to (i) the meandering dynamic of the river at large scales, which connects/disconnects historical sites from the river system (Perucca et al., 2007) and (ii) the hydrodynamic and meteorological conditions like tides and wind at smaller scales (Browne et al., 2010; Sadri and Thompson, 2014). The result is a much more extended residence time of plastics in estuaries than previously thought with models showing annual plastic export into the ocean through rivers (Lebreton et al., 2017; Schmidt et al., 2017). Few studies already suggested a high residence time of plastics in estuaries, favored by tidal regimes, habitats, and leading to macroplastic fragmentation (Browne et al., 2010; Ivar do Sul et al., 2014; Sadri and Thompson, 2014; Fok and Cheung, 2015; Naidoo et al., 2015). In our study, we bring major evidences showing a long term dynamic of plastic transport in the Seine estuary with a high residence time, up to decades, related to complex processes of deposition/remobilization. However, there is still some remaining questions about the link between the short term dynamic and the long term dynamic of plastic transport in the Seine estuaries.

As previously mentioned, tidal cycles and winds are thought to be the major drivers of this short term dynamic (Browne et al., 2010; Sadri and Thompson, 2014), but date-prints plastics are unable to track their real influence because it is impossible to know precisely what happened before they were collected. Were they deposited somewhere for a long time before being remobilized and stranded again at their site of collection? Or, were they traveling from riverbanks to riverbanks each day with tidal cycles? Nevertheless, Weinstein et al. (2016) showed that polyethylene, polypropylene, and extruded polystyrene started to decompose into microplastics in few weeks under tidal influence. Extending the residence time of plastics under those environmental conditions may thus increase the probability of plastic fragmentation (Browne et al., 2010). Consequently, rivers, and estuaries in particular, may function as a “microplastic factory” resulting from the fragmentation of macroplastics after weeks to years of transport/deposit cycles on riverbanks. Although, there is still a lack of evidence linking the concentration in microplastics in water and the fragmentation of macroplastics, unidentified fragments of plastics are one of the main contributors of the plastic pollution on riverbanks in the Seine estuary (Tramoy et al., 2019).

Conclusion and perspectives

Date-prints items (Microlax) enabled to identify the presence of plastics in the Seine estuary as old as the late 70' relatively frequently. We suggest this is the case for many of the plastics found in the estuary based on the comparison between historical sites of accumulation and another shoreline regularly cleaned during 1 year. This method is a good way to overcome the difficulties to accurately date plastics found into the environment. It also provides useful insights on how the receiving environment functions over different time and space scales. Based on Microlax, it is suggested that the distribution of "old" plastics over large scales of time and space are controlled by (i) their decreasing input in the Seine estuary related to higher sewerage efficiency and (ii) the meandering dynamic of the river which drives the connectivity of the floodplain riversystem. Over smaller time and space scales, the distribution of the items may be controlled by the hydrodynamic and meteorological conditions of the estuary.

In this study, we focused on one type of item, i.e. the Microlax, to constrain sources and pathways of those items overtime. Extending the use of date-prints with other well identified items would enhance our understanding of the transfer dynamic of plastics into the ocean. A good marker is like a good fossil for geologists: (i) a short life (most of UBDs are short), (ii) a good spatial distribution to enable geographic comparisons (Microlax or similar is used worldwide) and (iii) a high abundance. For example, fruit drink pouches like CapriSun packaging would be a good candidate because they often show date-prints (but not engraved) and their consumption strongly increased last years, related to on-the-go consumption. This kind of reporting could then be reproduced in other estuaries and rivers with the suitable markers to determine a ratio "old"/"recent" plastic litter for each area. It would help to better understand which factors make estuaries a fast or slow pathway for plastics into the ocean to tackle the problem accordingly. In the case of slow pathways like in the Seine estuary, shoreline cleaning would be a good curative solution to avoid the export of macroplastics into the ocean before they get fragmented into microplastics. Those cleanings might also be an efficient tool to evaluate public prevention policies by monitoring the amount and type of litter collected overtime.

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