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## Macro-litter in surface waters from the Rhone River: Plastic pollution and loading to the NW Mediterranean Sea

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### ABSTRACT

We present here the first estimates of floating macro-litter in surface waters from the Rhone River, based on monthly visual observations during 1-year period (2016–2017). Plastic represented 77% of the identified items, confirming its predominance in riverine floating litter. Fragments (2.5–50 cm) and Single Use Plastics (i.e. bags, bottles and cover/packaging) were among the most abundant items. Frequent non-plastic floating litter were paper items such as packaging material and newspapers, and metal items (mostly cans), representing 14% and 5% of total litter, respectively. A lower-end estimate resulted in ~223,000 plastic items (~0.7 t of plastic) transported annually by the Rhone surface waters to the Gulf of Lion (NW Mediterranean Sea). Floating macro-plastics are only a fraction of the total plastic export by the Rhone. Our study highlights the current discrepancy between field observations and theoretical estimations. Improvements are needed to harmonize data collection methodologies for field studies and model validation.

### 1. Introduction

There is no doubt today that plastic waste generated by the world's populations is accumulating in our seas and oceans at large amounts. Numerous marine species including fishes, seabirds, marine mammals and turtles have been reported to either ingest plastic or get entangled in marine debris (Kühn et al., 2015; Gall and Thompson, 2015; Rummel et al., 2016). Coastal populations were estimated to pollute the global ocean with 4.8 to  $12.7 \times 10^6$  tonnes (t) of plastic waste in 2010 (Jambeck et al., 2015). Rivers have been described as efficient “highways” playing a major role in the land-to-ocean litter transport of plastic waste, not only delivering materials generated in coastal areas, but also transporting waste generated further inland in the catchment. Recent modelling estimations point to 0.5 to  $2.7 \times 10^6$   $\text{ty}^{-1}$  of plastic waste loaded to the seas and oceans of the world through inland water courses (Lebreton et al., 2017; Schmidt et al., 2017). These estimations are mostly based on the calculation of the amount of mismanaged plastic waste (MMPW), including a limited number of field observations and, in most cases, considering mostly the smaller plastic fragments like micro-plastics (< 5 mm). The occurrence of larger plastic fragments, called macro-plastics, in river waters and their loading to coastal areas are largely uncharacterized. This could be in part the result of a lack of

consistent methodologies for sampling/monitoring larger plastic fragments, as recently pointed out (González et al., 2016; van Emmerik et al., 2018). In addition, the scientific community has paid a stronger attention to micro-plastics, even if macro-plastics are important secondary sources of micro-plastics due to degradation/fragmentation processes (Gewert et al., 2015). Moreover, the majority (87%) of the existing studies dealing with plastic pollution focused on the marine environment and freshwater ecosystems have certainly been less investigated (Blettler et al., 2018). A limited number of studies reporting on loads of macro-plastics in rivers are available, like those in the Saigon River (Vietnam) (Lahens et al., 2018; van Emmerik et al., 2018). In Europe, macro-plastic measurements have been published for the Seine (France) (Gasperi et al., 2014; Tramoy et al., 2019), Thames (UK) (Morritt et al., 2014) and Tiber (Italy) (Crosti et al., 2018) rivers. However, no previous field data on macro-plastics occurrence and loading exist for large European rivers draining into the Mediterranean Sea, a semi-enclosed marine environment reported as one of the major hotspots of plastic contamination in the World (Eriksen et al., 2014; Cózar et al., 2015).

We present here results on macro-litter occurrence, with focus on macro-plastics, in surface waters from the Rhone River (Switzerland/France). This river is the major watercourse flowing into the Western

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Mediterranean Sea with its 816 km encompassing a catchment area of  $\sim 95,000 \text{ km}^2$  (Eyrolle et al., 2012). The Rhone River delivers 2–10 Mt of sediments and  $\sim 50 \times 10^9 \text{ m}^3$  of freshwater annually and large amounts of nutrients and inorganic/organic carbon (Sempéré et al., 2000; Eyrolle et al., 2012). It is also a known source of classical anthropogenic organic compounds like polychlorinated biphenyls (PCBs) (Mourier et al., 2014; Poulhier et al., 2019), polycyclic aromatic hydrocarbons (PAHs) (Guigue et al., 2014), and dicarboxylic acids (Sempéré et al., 2018). Furthermore, the presence of the major families of organic plastic additives, like phthalates acid and organophosphate esters (PAE and OPE, respectively) and bisphenols in the Rhone surface waters have been very recently reported (Paluselli et al., 2018; Schmidt et al., n.d.). These important riverine inputs also transport unquantified amounts of plastic litter, spreading out in the entire Gulf of Lion, particularly after heavy rain events. Accumulation of plastic material has already been observed at the sea floor in the French Mediterranean coast including: (i) Marseille Bay, continental slopes and canyons from the Gulf of Lion in the NW Mediterranean Sea, with plastic bags accounting for 90% of the total debris on the floor of the continental shelf and in particular in the eastern canyons (Galgani et al., 1995, 1996, 2000); (ii) the Rhone River plume area and nearby canyons (Fabri et al., 2014). Additionally, recent reports investigated the presence of microplastics in Rhone River waters (Constant, 2018; Schmidt et al., 2018). However, no discrete data or monitoring on macro-plastics in the Rhone River is available to the best of our knowledge. The overall objective of this work was to provide the first estimate on the occurrence of floating macro-litter, with focus on plastic items, in the Rhone River and its loading to the Gulf of Lion area by applying an easy and cost-effective monitoring tool based on visual observation.

New knowledge on this topic can have an impact in the implementation of environmental regulations such as the European Marine Strategy Framework Directive (MSFD, 2008/56/EC), aimed at achieving Good Environmental Status in EU's marine waters, including the monitoring and assessment of marine litter under Descriptor 10; or the new European Strategy for Plastics in a Circular Economy (COM/2018/028 final), which has recently agreed on banning certain single use plastic (SUP) items ([http://europa.eu/rapid/press-release\\_IP-18-3927\\_en.htm](http://europa.eu/rapid/press-release_IP-18-3927_en.htm)).

## 2. Materials and methods

### 2.1. Observation site and data collection

The Rhone Delta is divided into two flows the “Petit Rhone” and the “Grand Rhone”, corresponding to around 10% and 90% of the water flow, respectively (Fig. 1A). Monthly monitoring was carried out from the Trinquetaille bridge ( $\sim 13 \text{ m}$  height) in Arles (Southern France, Fig. 1B) located on the “Grand Rhone” at around 40 km NW from the river mouth in the Gulf of Lion (NW Mediterranean Sea). Data was collected by visual observations of floating macro-plastics and documented with the European Commission-Joint Research Centre (EC-JRC) Floating Litter monitoring application (App) for mobile devices (González-Fernández and Hanke, 2017). The observations were performed within the framework of the project RIMMEL (Riverine and Marine floating macro-litter Monitoring and Modelling of Environmental Loading) from the EC-JRC. This strategy allowed the harmonized monitoring of macro-plastics, compatible with the MSFD.

The App used for the observations is aligned with the MSFD MasterList of litter categories included in the “Guidance of Monitoring of Marine Litter in European Seas” document (EC-JRC, 2013), containing the list of litter items established under the MSFD (González-Fernández and Hanke, 2017). Further, the App includes categories such as ‘non litter items’, ‘other litter items’ and also ‘other plastic items’ to register items that were not possible to identify or not listed in the MSFD MasterList. Non litter items were excluded for the following analysis.

A total of 16 observation sessions (generally around 30 min each) were performed from September 2016 to September 2017 (Table 1), considering an observation track width (OTW) of 65 m (i.e., the section at both sides from the observer) over a total river width of 150 m at the observation site (Fig. 1B). Although observations were aimed at monitoring litter  $> 2.5 \text{ cm}$ , identification of small items (2.5–5 cm) from a 13 m height could not be assured across the whole OTW selected, expanding 32.5 m left and right from the observation site, particularly at the edges (Fig. 1B). The distance from the observer to the edges of the OTW corresponds to 35 m, and therefore we define a more realistic minimum observable size for this study ranging from 2.5 cm at the vertical of the observer to  $\sim 7 \text{ cm}$  at the edges of the OTW. Considering this limitation, results reported herein probably include a bias in detecting large items vs. small items towards the edges of the OTW, underestimating the flux of smaller items, which also implies providing a conservative litter loading estimate. A dedicated study would be required to empirically ascertain the lower-end size of observable items across a given OTW.

In order to perform an assessment of the litter and plastic loading and their temporal variability, river flow ( $\text{m}^3\text{s}^{-1}$ ) time series were extracted from “Banque Hydro” (<http://www.hydro.eaufrance.fr/>), whereas daily precipitation measurements (mm), wind speed ( $\text{m}\text{s}^{-1}$ ) and direction were obtained from Météo-France (<https://donneespubliques.meteofrance.fr/>). These data correspond to the closest measuring stations to the observation site: Tarascon,  $\sim 16 \text{ km}$  upstream of Arles and, Tour-du-Valant, around 25 km south from the observation site, for precipitation and wind data, respectively (Fig. 1A and Table 1).

### 2.2. Loading estimation

The macro-litter loading was calculated for each session as number of observed anthropogenic litter items (Table S1, SI) divided by observation time, and then extrapolated from OTW (65 m) to river width at the sampling site (150 m) (Fig. 1B), obtaining  $\text{items}\cdot\text{h}^{-1}$ . Mean and median loadings were calculated for all observation sessions ( $n = 16$ ). The annual load was estimated as: median loading ( $\text{items}\cdot\text{h}^{-1}$ )  $\times 24 \text{ h} \times 365 \text{ d}$ , providing  $\text{items}\cdot\text{y}^{-1}$ . The median was selected against the mean, minimizing the influence of extreme litter events for a conservative estimate. This estimate was based on the assumption that floating litter is homogeneously distributed on the cross-section of the river and the datasets are representative for the litter loading variability along the year.

## 3. Results and discussion

### 3.1. Macro-litter abundance and plastic loading

Items belonging to twenty-two macro-litter categories were observed in surface waters from the Rhone River during the monitoring period, summing up a total of 181 items for 8.2 h of cumulative observation (Fig. 2).

Plastic items, including pieces, bags, sheets, bottles, and cover/packaging, represented around 77% of the floating macro-litter in Rhone River waters confirming the predominance of plastic pollution, in agreement with the top items list identified by RIMMEL across Europe (González-Fernández et al., 2018). Paper items, like packaging material and newspapers, and metal items (mostly cans), represented 14% and 5% of total floating macro-litter in Rhone Rivers waters, respectively, and were the most abundant non-plastic floating items (Fig. 2B).

Litter composition presented herein differed to a certain extent from top items lists found in the marine environment. Depending on the materials and the differential environmental conditions in rivers and marine areas, some items may get degraded and fragmented, while others may get ballasted or lose their floatability at different rates.



**Fig. 1.** Map showing the Rhone River from its source upstream Lake Geneva to its estuary in the Gulf of Lion (NW Mediterranean Sea), the position of the river flow (Tarascon) and precipitation and wind (Tour-du-Valant) measuring stations, location of Arles (A) in the “Grand Rhone”, and detail of the observation site (Trinquetaille bridge) (B).

**Table 1**

Details of the observations performed with corresponding observation time (min) and river flow ( $\text{m}^3\text{s}^{-1}$ ) as registered in Tarascon; and precipitation (mm), wind speed ( $\text{m}\text{s}^{-1}$ ) and wind direction (WD, degrees), as registered in Tour-du-Valant (Météo-France station). \*  $360^\circ = \text{N}$ ,  $180^\circ = \text{S}$ ,  $90^\circ = \text{E}$ ,  $270^\circ = \text{W}$ .

Observation	Date	Starting time	Obs time (min)	River flow ( $\text{m}^3\text{s}^{-1}$ )	Precipitation (mm)	WS ( $\text{m}\text{s}^{-1}$ )	WD (degree)*
Obs-1	16/09/2016	11:11:50	30	868	1.0	3.7	159
Obs-2	04/10/2016	09:27:32	17	545	0.0	3.3	350
Obs-3	08/11/2016	15:10:41	34	1310	0.0	4.1	318
Obs-4	06/12/2016	11:01:16	36	1290	0.0	3.0	184
Obs-5	19/01/2017	11:48:28	33	1140	0.0	3.8	346
Obs-6	15/02/2017	10:30:21	35	1550	0.0	2.9	100
Obs-7	28/03/2017	09:14:44	34	1560	0.0	2.4	158
Obs-8	12/04/2017	13:00:00	30	894	0.0	2.8	214
Obs-9	05/05/2017	11:20:09	32	1230	0.0	3.0	173
Obs-10	06/05/2017	10:37:47	15	1130	11.3	3.7	280
Obs-11	23/05/2017	09:29:37	30	1240	0.2	3.5	268
Obs-12	22/06/2017	11:00:55	35	872	0.0	2.7	126
Obs-13	10/07/2017	09:29:16	36	671	0.0	2.6	245
Obs-14	12/07/2017	10:25:41	30	859	0.0	3.9	236
Obs-15	24/08/2017	11:52:45	33	522	0.0	3.6	125
Obs-16	25/09/2017	10:16:56	34	625	0.0	2.0	171

Therefore, the litter composition in rivers may change compared to beaches and marine waters. Additionally, the marine environment is subject to sea-based litter sources, often related to fisheries, introducing items that are not present or limited in rivers. Despite differences among floating items observed in the Rhone River and top items found in beaches and floating at sea in the Mediterranean region, there is coherence in the high percentage of plastic items composing the marine litter (Table S2, SI). Furthermore, these top items lists included SUP items such as bottles or bags within the most frequent items (Addamo et al., 2017; Arcangeli et al., 2018; Di-Meglio and Campana, 2017).

Overall, the most abundant items observed in the Rhone River were plastic pieces in the size range 2.5–50 cm, influenced by a peak in litter

flux observed on 5th May 2017 (Fig. 3). No significant correlations were found with environmental variables such as river flow, rain or wind (meridional and zonal components), using Pearson correlation coefficients for  $n = 16$  observations and p-level 0.05 (Table S3, SI). We acknowledge that our data set is limited and higher frequency observations may reveal some significant correlations. Predominance of plastic pieces could indicate: (i) a significant fragmentation of the larger items most commonly found such as bags, cover/packaging and other plastic materials flowing down the river; (ii) and/or the preferential direct inputs of plastic fragments in the catchment area. Additionally, small fragments are expected to have a more efficient transport in the river waters, while larger items may get trapped in the riverine banks and

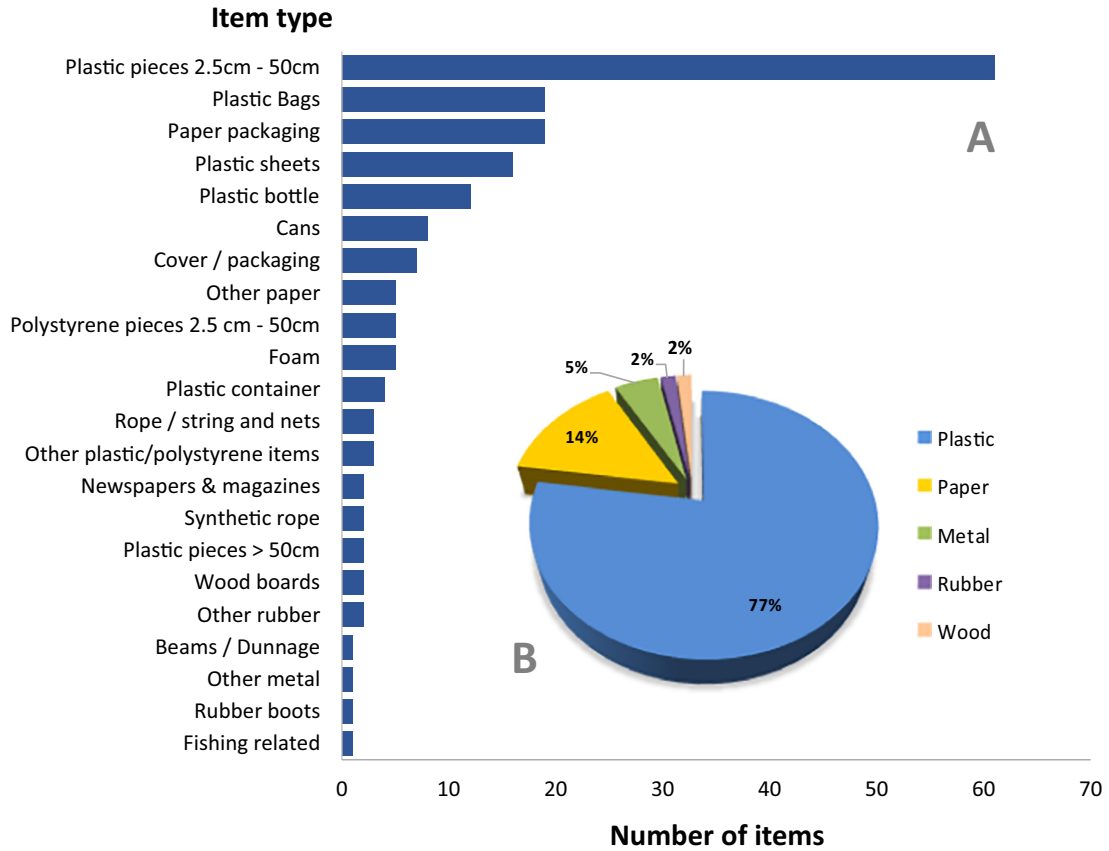


Fig. 2. Number and type of macro-litter items found in surface waters from the Rhone River (A) and relative abundance of macro-litter items grouped by material type (B).

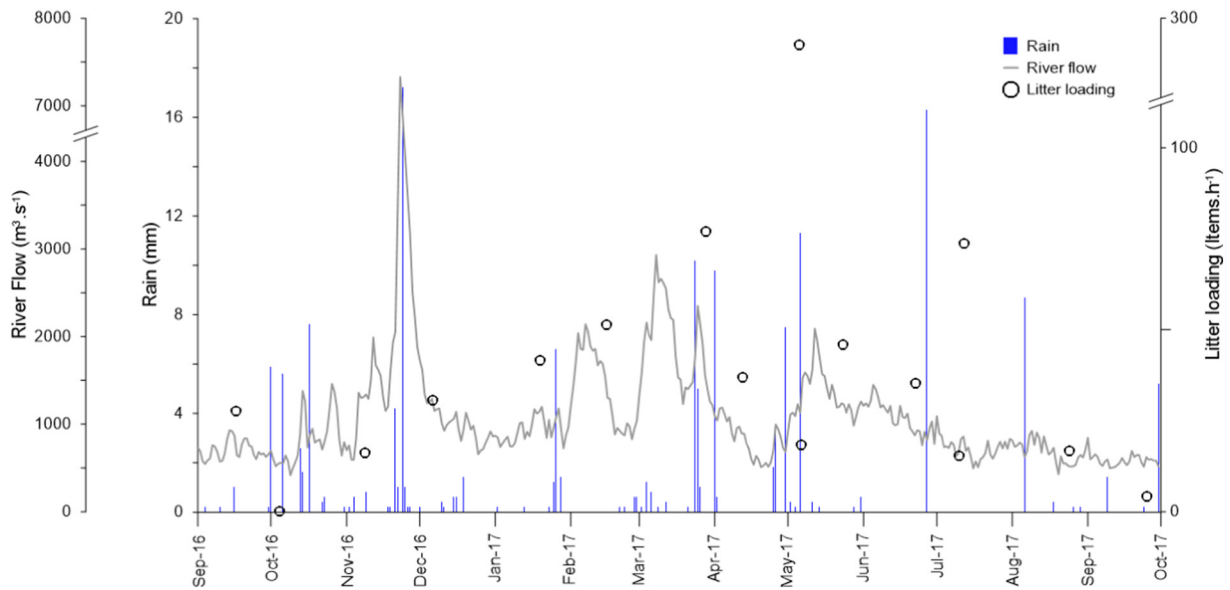


Fig. 3. Temporal variation of litter loading ( $\text{items}\cdot\text{h}^{-1}$ ), annual river flow ( $\text{m}^3\cdot\text{s}^{-1}$ ) and precipitation (mm) in the observation period from September 2016 to September 2017. These variables are separately displayed on a complementary figure presented in SI (Fig. S2).

vegetation for longer periods (Williams and Simmons, 1996).

Fig. 3 shows the estimated litter loading along with river flow and precipitation at Tarascon during the observation period. The estimated litter loading values (whole river cross-section) exhibited a high variability, ranging from none to  $293 \text{ items}\cdot\text{h}^{-1}$ , with a mean value of  $\sim 50 \pm 69 \text{ SD items}\cdot\text{h}^{-1}$  ( $\sim 33 \text{ items}\cdot\text{h}^{-1}$  median value). Plastic items reached  $226 \text{ items}\cdot\text{h}^{-1}$  with a mean macro-plastic loading of

$\sim 37 \pm 61 \text{ SD items}\cdot\text{h}^{-1}$  ( $\sim 26 \text{ items}\cdot\text{h}^{-1}$  median value). This large variability could have been induced by the important temporal variations in the river flow, with values ranging from around  $400$  to  $7300 \text{ m}^3\cdot\text{s}^{-1}$  for the observation period (median discharge of  $910 \text{ m}^3\cdot\text{s}^{-1}$ ). However, in spite of this variability a general positive trend between river water discharge and litter loading can be observed (Fig. 3). Indeed, a linear regression analysis showed a positive

statistically significant correlation when excluding the litter peak of 5th May 2017 ( $r^2 = 0.36$ ,  $\alpha = 0.05$ ,  $p$ -value = 0.018) (Fig. S1). The litter peaks of 5th May, and 12th July 2017 were registered under relatively low river flow (1230 and 860  $\text{m}^3\text{s}^{-1}$ ) and no precipitation (Table 1). Although these conditions cannot fully explain the high litter fluxes observed, several rain events took place before these two sampling events. First, in the period 25th–30th April 2017 and then on 27th June 2017 with  $\sim 13$  mm and  $\sim 16$  mm of cumulative precipitation registered, respectively (Fig. 3). These precipitation events could have facilitated new litter inputs to the watercourse and remobilized items already deposited on the river banks upstream. In addition, factors such as flow regulation by opening dams, storm overflow and runoff during rain events upstream in the basin may have an important effect, flushing litter accumulated in the basin by pulses within very short periods.

In order to gain understanding on short-term variability (e.g. after rain events), specific studies with more frequent observations are needed. No observation was performed on 23rd November 2016, when the river flow exhibited its highest value, most probably due to various consecutive days of rains in the basin, including  $\sim 22$  mm of cumulative precipitation registered in Arles area for the period 21st–24th November 2016. The macro-litter observations were performed either earlier in November or almost two weeks later on 6th December 2016 (Table 1), under no rain conditions and a river flow of  $\sim 1300 \text{m}^3\text{s}^{-1}$ , missing the flow peak.

An estimation of the total loading of the Rhone River to the NW Mediterranean Sea points to  $\sim 223,000$  items of floating macro-plastics annually. Assuming an item-to-mass conversion factor of 3.2 g per item based on macro-plastic sampling in Saigon river (Vietnam) (van Emmerik et al., 2018),  $\sim 0.7 \text{ty}^{-1}$  of macro-plastics could be transported by the Rhone surface waters into the Gulf of Lion.

We consider our plastic export from the Rhone River to the Mediterranean Sea as a lower-end estimate. First, it is based on floating macro-plastic only and the use of the median value for the calculation, which excludes the influence of extreme events. Second, the debris lying and/or flowing in the water sub-surface and the entire water column are not taken into account. River sediments, reported as final repository of marine litter (Galgani et al., 2000), have not been considered either. Given that the bottom nepheloid layer in front of the Rhone River Estuary comprises a large amount of particulate material transported and re-suspended by the Rhone (Sempéré et al., 1994), it is very likely that significant amounts of plastic debris are also exported with the sediment from the river bottom into the NW Mediterranean. In addition, an underestimation of the smallest plastic items, particularly at the observation track edges is acknowledged. Further, even if we did not observe significant accumulation areas in our site during the sampling events, an accumulation of macro-plastics could have happened upstream, and these waste deposits could be remobilized under certain circumstances, like heavy rains, leading to plastic waste pulses in surface waters punctually increasing the loading to coastal areas and having an important short-term impact. This phenomenon should be further investigated along the year.

In spite of the high uncertainties associated with this calculation, it represents the first estimate of floating macro-plastic inputs from the Rhone River to the NW Mediterranean Sea based on real observations. Thus, we cannot compare our results with other field observations of macro-plastics in this area. However, a few studies (field and modelling estimations) performed during the last years have investigated the micro-plastic export by the Rhone River (Table 2). Interestingly, our estimate is within the range of the micro-plastic loading derived from samples collected at the same site (Arles, France) and at the beginning of our observation period (Schmidt et al., 2018; Constant, 2018) (Table 2). The total floating plastic waste, as macro- and micro-plastics “traveling” at the same time in the Rhone surface waters, could reach up to  $8.5 \text{ty}^{-1}$ , considering our estimate and the higher-end estimate of micro-plastics from Schmidt et al. (2018). However, this estimation

contrasts with the higher flow of plastics (mostly micro-plastics) measured in the Swiss part of the Rhone River (Faure et al., 2015), pointing to different loads of floating plastics upstream and downstream of the Rhone. Besides, our macro-plastic estimate are 1–4 orders of magnitude lower than the available modelling calculations of plastic waste export from the Rhone River, varying from 7 to  $\sim 1450 \text{ty}^{-1}$  (Table 2) (Schmidt et al., 2017; Siegfried et al., 2017; Liubartseva et al., 2018). In contrast, our estimates are in the range or slightly higher than the modelling inputs calculated for the Rhone River downstream dams ( $0.01$ – $0.14 \text{ty}^{-1}$ ) (Lebreton et al., 2017), consistent with the hypothesis of higher loads of floating plastic upstream in the river basin, probably influenced by the existence of different litter sources and river transport conditions, including retention in dams. With the exception of the estimations from Lebreton et al. (2017), there is an important discrepancy between our export estimates derived from direct observation of floating items (plastic fragments  $\geq 2.5$  cm) and the existing theoretical estimations, which are mostly based on data from plastic production and waste in a given river catchment or geographical area, and based on scarce micro-plastics field data. The use of micro-plastic data can hardly be representative for the macro-plastics pool because of the sampling conditions, e.g. short-term deployment of  $\sim 20$  min with 0.5–0.9 m width and 0.1–0.6 m height opening manta-nets (González et al., 2016). The available data also show that existing results from models, which have been partially or not at all validated with field observations due to the lack of data in most of the cases, can vary substantially among them, up to 5 orders of magnitude (Table 2).

Our estimate is lower compared to the  $27 \text{ty}^{-1}$  (mean value) of floating plastic debris reported for another French river (i.e., the Seine), based on items collection during April–July 2014 by using floating debris-retention booms (Gasperi et al., 2014) and even lower compared to a second study based on a longer data collection period in the same river ( $110$ – $170 \text{ty}^{-1}$ ) (Tramoy et al., 2019). However, about 16 million people live in the Seine catchment area ( $\sim 73,000 \text{km}^2$ ), including the megacity of Paris, whereas the population for the Rhone catchment area ( $\sim 95,000 \text{km}^2$ ), represents  $\sim 10$  million people (Schmidt et al., 2017), and therefore a lower loading is expected in the latter. In addition, the fact that the retention booms used in these studies were all located on the river margins, supposedly macro-plastic accumulation areas as pointed out by van Emmerik et al. (2018), could also explain part of this variability. These facts illustrate one more time the importance of the data collection methodologies and the complexity in the comparison of existing data.

### 3.2. Environmental implications

The primary production in the NW Mediterranean Sea has been reported as largely affected by the Rhone River inputs (mostly by the large sedimentary material and nutrients inputs), also playing an important role in the marine ecosystem functioning in the entire Gulf of Lion due to the effective dispersion in the area (Raimbault and Durrieu de Madron, 2003; Durrieu de Madron et al., 2008). However, the effects of plastic waste in this ecosystem are largely unknown. Our work indicates that significant amounts of plastic waste may be delivered annually to the NW Mediterranean Sea from the Rhone River, and that floating macro-plastic loads are most probably only a part of the total plastic waste entering this ecosystem. Floating macro litter inputs from rivers include persistent and fragmented plastics that can have a longer term availability to marine animals and possibility to cause harm (Werner et al., 2016). Once they reach the marine environment, they can get stranded on beaches, sink in shallow bottoms, or travel long distances in the open sea, being accessible to a wide range of marine animals (Kühn et al., 2015). In addition to the possible “physical damages” (e.g. ingestion and entanglement) caused to the aquatic organisms, these fragments of different polymers could also represent new sources of dissolved organic carbon (DOC) (Romera-Castillo et al., 2018; Paluselli et al., 2019) and organic additives like PAEs and OPES

**Table 2**

Compilation of existing plastic export estimations from the Rhone River to the NW Mediterranean Sea. Field studies are on surface waters only.

Geographical area	Period	Plastic size <sup>a</sup>	Type of study/sampling method	Export (t·y <sup>-1</sup> )	Reference
Rhone (Swiss part)	Oct. 2013	Micro-plastics	Field/Manta net	~208 <sup>b</sup>	Faure et al. (2015)
Rhone (Arles, France)	Mar., Apr. 2016	Micro-plastics	Field/Manta net	~0.07–7.8 <sup>c</sup>	Schmidt et al. (2018)
Rhone (Arles, France)	Oct. 2015–Oct. 2016	Micro-plastics	Field/Manta net	6.0	Constant (2018)
Rhone river basin	Year 2000	Micro-plastics	Numerical simulation	163	Schmidt et al. (2017)
Not specified	2013–2017	Micro-plastics	Numerical simulation	1454	Liubartseva et al. (2018)
Rhone river basin (downstream dams)	Not specified	Micro + macro-plastics	Numerical simulation	0.01–0.14	Lebreton et al. (2017)
Rhone river basin	Not specified	Micro + macro-plastics	Numerical simulation	7–16	Schmidt et al. (2017)
Rhone (Arles, France)	Sept. 2016–Sept. 2017	Macro-plastics	Field/visual observation	0.71	This study

<sup>a</sup> The microplastic sizes considered in the different studies may be different (see specific references for further details). We consider macro-plastics as fragments  $\geq 2.5$  cm for the purpose of this study.

<sup>b</sup> Estimated from the reported 570 kg·d<sup>-1</sup>.

<sup>c</sup> Estimated from the reported 0.20 and 21.32 kg·d<sup>-1</sup>.

(Paluselli et al., 2019). Indeed, a recent study reported from 5 to 54 t·y<sup>-1</sup> of organic plastic additives, including PAEs, OPEs and bisphenols, exported by Rhone surface waters to the Gulf of Lion (Schmidt et al., n.d.). The latter suggests that the potential additive leaching from the combined estimated export of 8.5 t·y<sup>-1</sup> for floating macro-plastics (this work) and micro-plastics (Schmidt et al., 2018) cannot explain the current levels of additives in the river dissolved water phase. However, the floating plastic waste seems to be only a fraction of the entire plastic pool in the water ecosystem, and the plastic stocks in the whole water column and sediments could contribute too to the “leaching source”. A dedicated study aiming at establishing a first mass balance of organic plastic additives in the NW Mediterranean Sea is needed. Such assessment should consider also the additive inputs not directly linked to the leaching from plastic debris in the water column. For example, the atmospheric deposition of OPEs has been confirmed as an important input pathway in coastal and open Mediterranean Sea (Castro-Jiménez et al., 2014, 2018). Up to 1.1 t·y<sup>-1</sup> of OPEs were estimated to be loaded from the atmosphere to the Gulf of Lion via dry deposition (Schmidt et al., n.d.). Finally, these inputs of macro-plastics and additives can contribute to the cocktail of pollutants that may affect the aquatic organisms and humans at a local/regional scale.

#### 4. Conclusions

The first estimation of floating macro-plastics input from the Rhone River to the NW Mediterranean Sea supports the hypothesis of large European rivers as important pathways for plastic litter to coastal areas. Our data suggest that floating macro-litter represents only a fraction of the total plastic waste transported into the marine environment through rivers. An important uncertainty is associated to these estimations due to the high temporal variability. Small plastic fragments in the range of 2.5 to 50 cm are the most frequent litter observed in surface waters. Further, Single Use Plastics (SUP), like bags, bottles and cover/packaging are among the most abundant items, highlighting the relevance of their restriction at EU level, through the EU Plastic Strategy. The results of this investigation can be used to design mitigation measures and monitor their effectiveness. Regarding banning of SUP, this study can indicate the most common items and flux/loading values found in large Mediterranean rivers, establishing a baseline for future monitoring. The generated data can also help in the plastic pollution management at river basin scale, supporting actions aimed at intercepting the plastic before it gets to the marine environment.

Our results highlight also the current discrepancy between field observations and theoretical estimations. It is clear that both field observations and modelling simulations aiming at quantifying the plastic waste export to coastal areas, particularly from large European rivers draining into the Mediterranean Sea, like the Rhone River, are still incomplete. Improvements are needed in order to harmonize data collection methodologies for field studies and model validation. Macro-to-

micro plastic transformation rates, and plastic item-to-mass conversion factors should be investigated for more accurate mass balances. Waterborne macro-plastics, traditionally described as a source of micro-plastics in rivers due to aging and fragmentation processes, should not be considered as the only source of the smallest plastic fragments. Other sources, like direct run-off and atmospheric inputs of micro-plastics, should be investigated. In addition, the role that waterborne plastic debris may play as contributing to the global stock of organic plastic additives in the water column should be further investigated also in relation to other potential sources of organic plastic additives like atmospheric deposition.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2019.05.067>.

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