The quest for the missing plastics: Large uncertainties in river plastic export into the sea

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Abstract

Plastic pollution in the natural environment is causing increasing concern at both the local and global scale. Understanding the dispersion of plastic through the environment is of key importance for the effective implementation of preventive measures and cleanup strategies. Over the past few years, various models have been developed to estimate the transport of plastics in rivers, using limited plastic observations in river systems. However, there is a large discrepancy between the amount of plastic being modelled to leave the river systems, and the amount of plastic that has been found in the seas and oceans. Here, we investigate one of the possible causes of this mismatch by performing an extensive uncertainty analysis of the riverine plastic export estimates. We examine the uncertainty from the homogenisation of observations, model parameter uncertainty, and underlying assumptions in models. To this end, we use the to-date most complete time-series of macroplastic observations (macroplastics have been found to contain most of the plastic mass transported by rivers), coming from three European rivers. The results show that model structure and parameter uncertainty causes up to four orders of magnitude, while the homogenisation of plastic observations introduces an additional three orders of magnitude uncertainty in the estimates. Additionally, most global models assume that variations in the plastic flux are primarily driven by river discharge. However, we show that correlations between river discharge (and other environmental drivers) and the plastic flux are never above 0.5, and strongly vary between catchments. Overall, we conclude that the yearly plastic load in rivers remains poorly constrained.

1 Introduction

Plastic pollution in rivers increasingly causes environmental concern at both local and global scale. It has been found to negatively impact ecosystems, increase flood risk by blocking hydraulic infrastructure, and cause damage to the human livelihoods in the vicinity of polluted rivers (van Emmerik and Schwarz, 2020). In addition, plastic emissions from rivers into the sea are assumed to be a major component of marine plastic pollution (Lebreton et al., 2017; Schmidt et al., 2017). However, there is currently a stark mismatch between the amount of plastic estimated to enter the ocean (Jambeck et al., 2015; Lebreton et al., 2017; Schmidt et al., 2017) and the amount of plastic observed in the open ocean (Weiss et al., 2021). Understanding the sources, sinks and transport mechanisms of plastic pollution is of key importance in both designing effective monitoring strategies (Vriend et al., 2020a), and developing pollution mitigation and cleanup campaigns (Lebreton et al., 2017).

Most of the currently available observations of plastic in rivers are isolated short-term measurements, taken at a single location in the river system (van Calcar and van Emmerik, 2019). Recently, observational campaigns have begun to sample rivers more extensively in space and time (e.g. Kiessling et al., 2019; van Emmerik et al., 2020; Schirinzi et al., 2020). This has led to the first consistent and continuous riverine plastic transport datasets, spanning up to one year of repeated measurements (González-Fernández et al., 2021). Although these data sources have proven to be valuable in creating data driven hypotheses on the modes of transport of plastic pollution (Roebroek et al., 2021b), they do not provide enough detail to fully describe annual plastic mass fluxes in individual rivers, and to estimate the total amount of plastic entering into the oceans each year. To answer such questions different modelling approaches have been proposed, ranging from simple extrapolation models (e.g. Castro-Jiménez et al., 2019), to regression models (e.g. Schmidt et al., 2017) and more complex probabilistic modelling approaches (Meijer et al., 2021).

Observing and modelling riverine plastic fluxes are currently associated with large uncertainties. As riverine plastic research is still an emerging field, measurement techniques differ substantially between observational campaigns, making the resulting data very hard to compare and combine to constrain model parameterisation. Current observations have a relatively coarse spatial and temporal resolution (González-Fernández et al., 2021), and current studies sample only a specific subset of the total riverine plastic flux. Often, observations focus only on floating plastic items larger than 2.5 cm when visually counting the floating objects for a given river stretch, or cover only part of the river width (e.g. Schöneich-Argent et al., 2020) and depth when taking samples or making observations (e.g. Broere et al., 2021). Furthermore, the output of the riverine plastic models is usually given in tonnes per year, but most observations are reported in the number of plastic items per unit of time or plastic item concentrations in water samples (van Calcar and van Emmerik, 2019; González-Fernández et al., 2021). Using observations to constrain models hence requires unit conversions that introduce additional uncertainty. Finally, statistical models contain several types of uncertainty by themselves, including parameter uncertainties and model structure uncertainty (represented in model performance). These various sources of uncertainty are not disentangled in most riverine plastic modelling studies and often not discussed in detail, which severely limits interpretability and study comparability, and risks misinformed policy decisions and mitigation action.

Here we discuss four modeling strategies that have been used to describe the yearly plastic flux in rivers. Two of these models were used to extrapolate isolated plastic flux observations within an individual river to a yearly flux, while the other two models were used to create a global estimate of the total amount of plastics reaching the oceans. To explore the uncertainties associated with the different modeling strategies, we use time-series of riverine floating macroplastic observations (macroplastics describe most of the plastic mass in rivers (Mai et al., 2020)) in three rivers (Rhône, France, and Llobregat and Besós, Spain). These time-series are in size comparable to the datasets used for the global modelling studies (Lebreton et al., 2017; Mai et al., 2020)). Using these time-series we set up models for each river individually, to systematically describe the uncertainties in the modelling of the riverine plastic flux, assessing the transport mechanisms hypotheses underpinning the modelling strategies, and evaluate the potential uncertainty in the reported global numbers.

2 Current modelling approaches

The four riverine plastic transport modelling approaches discussed in this study are of increasing complexity: the first category is a temporal extrapolation approach, category two and three are regression models, with an increasing complexity of input data, and the fourth category is the most complex, probabilistic modelling approach. The models and their data requirements are presented in Table 1 and are discussed in the paragraphs below.

M1: Temporal extrapolation

A common approach to estimate the annual plastic flux leaving a catchment is to directly extrapolate the plastic flux observations in a single catchment over time. This strategy has for example been used by González-Fernández et al. (2021) to estimate annual litter flux in individual European rivers. The advantage of this approach lies in its simplicity; no additional data are required to obtain an estimate, and no assumptions regarding relations between e.g. the plastic flux and environmental variables have to be made. The quality of the estimate thus depends on how well the sample distribution matches the unknown yearly plastic flux distribution. As most plastic is expected to be mobilised and transported under extreme conditions, and hence the plastic flux is likely not normally distributed (Roebroek et al., 2021a), a large number of samples is needed to accurately capture the true mean of the flux distribution.

Table 1: The four modelling categories defined here, with the data that are required to develop them.

	M1: Temporal extrapolation	**M2: Linear regression with environmental drivers**	**M3: Exponential regression with environmental drivers**	**M4: Spatial probabilistic modelling**
River plastic observations	x	x	x	x
Environmental variables (rain, discharge, wind speed etc.)	-	x	x	x
Land-based plastic pollution (e.g. concept of Mismanaged Plastic Waste)	-	-	x	x
River network	-	-	x	x
Land use	-	-	-	x

M2: Linear regression with environmental drivers

The second model category represents linear regression models which link plastic observations to environmental transport mechanisms, such as discharge, wind, and precipitation driven surface runoff (Schirinzi et al., 2020; Meijer et al., 2021). Developing a linear regression model for a particular river requires plastic load observations and matching hydro-meteorological observations. Such linear regression models have been applied in various studies (e.g. Wong et al., 2020). The advantage of regression models is that they connect the plastic flux to the physical transport mechanisms, thus allowing to predict the plastic mass flux in absence of plastic observations as long as observations of the environmental transport mechanisms are available. The prediction accuracy depends on the quality of the relationship between the plastic flux and the transport mechanisms. Additionally, the relationship between the hydrometeorological drivers and the plastic flux is not necessarily linear (Lebreton et al., 2017). In particular, using these regressions to predict the plastic flux during extreme hydrometeorological events is unlikely to perform well because extreme events (e.g. storms and floods) transport plastic through additional pathways such as through sewage overflows and mobilisation from floodplains (Castro-Jiménez et al., 2019; Roebroek et al., 2021a,b). Lastly, temporal regression models using environmental drivers alone attempt to explain the temporal variability in plastic transport exclusively with natural transport mechanisms, while temporal patterns of human littering and litter redistribution are ignored.

M3: Exponential regression with environmental drivers

To estimate the plastic flux in catchments with no observations, estimates of the integrated mismanaged plastic waste within catchments (plastic waste not entering an adequate waste management system) are included in regression models. In doing so, catchments with high discharge and high levels of mismanaged pollution are attributed the highest plastic flux in rivers. In the literature this method has been applied to obtain the first global estimates of the riverine plastic flux entering the oceans, such as Lebreton et al. (2017), Schmidt et al. (2017), and Mai et al. (2020). These methods allow estimates about the plastic flux in rivers for which no observational estimate is available. Additionally, including information on anthropogenic littering should result in a more realistic model. However, mismanaged plastic waste estimates are highly uncertain (Jambeck et al., 2015; Ryberg et al., 2019; Lau et al., 2020; Edelson et al., 2021), introducing additional uncertainty to the model. Also, as with the previously described model categories, the model relies on the assumption that the relation between discharge and litter flux holds for discharges that have not been observed. Under extreme discharge events this assumption does likely not hold.

M4: Spatial probabilistic modelling

The last modelling strategy is spatially probabilistic modelling, proposed by Meijer et al. (2021). It uses estimates of land-based plastic pollution similarly to the spatiotemporal regression models (but spatially discrete). In this case, rather than deriving statistical relationships between terrestrial plastic pollution, hydrometeorology and the riverine plastic flux, the model uses spatially resolved probabilities to estimate the fraction of terrestrial plastic waste that contributes to the plastic flux leaving the catchment. These probability maps are not based on empirical evidence, as the transport probabilities of plastic over land are currently not understood or quantified. Instead, the probabilities are statistically estimated based on the distance to the river network and land-use type. The advantage of this approach is that it takes the topography and vegetation cover into account, thus similar catchments with different land-use and different distances between plastic hotspots and the river channel do not yield the same final plastic flux. A limitation of this approach is that the temporal variability in plastic flux is attributed solely to natural processes, as no data are available on the temporal variability in littering and mismanaged waste input. Additionally, this modelling framework includes a much larger number of parameters than in the modeling approaches described above. As the current number of observations of the plastic flux are limited, constraining these models may currently be impossible.

3 Methods

Data used in this study

Floating plastic litter time series

We used data collected through visual observations within the RIMMEL (Riverine and Marine floating macrolitter Monitoring and Modelling of Environmental Loading) project from the European Commission-Joint Research Center (EC-JRC) (González-Fernández and Hanke, 2017). Visual observations were done from bridges and recorded through a dedicated mobile app. For each observation, all floating litter items within a defined observation track width were counted for a given duration. Items were tallied per specific category, based on the European Marine Strategy Framework Directive (MSFD) Master List of Categories of Litter Items (MSFD Technical Subgroup on Marine Litter, 2013). The minimum detectable item size depends on e.g. observation height, turbidity of the water, sun glare, and waves, and therefore only macrolitter items ¿2.5 cm were considered. Thanks to its simplicity, the visual counting method is nowadays widely used for continental and global assessments of (floating) litter assessments (e.g. van Calcar and van Emmerik, 2019; González-Fernández et al., 2021). Long-term observations however are still scarce, and therefore our study is limited to the three one-year datasets for the Rhône, Besos and Llobregat rivers (see Table 2). To compare the data of the different studies, they were standardized by converting them to hourly values, covering the full river width (if not sampled, linearly extrapolated). The observations are displayed against discharge in Figure 1.

Table 2: Riverine plastic flux observations used in this study

River	**Country**	**Location**	**Number of observations**	**Period**	**Reference**
Rhône	France	Trinquetaille bridge, Arles (43.678914, 4.623093)	16	September 2016 – September 2017	Castro-Jiménez et al. (2019)
Besós	Spain	Pont del Ferrocaril, (41.42161179, 2.22872734)	36	November 2016 – August 2017	Schirinzi et al. (2020)
Llobregat	Spain	Pont Nelson Mandela, (41.32198384, 2.114564929)	50	October 2016 – September 2017	Schirinzi et al. (2020)

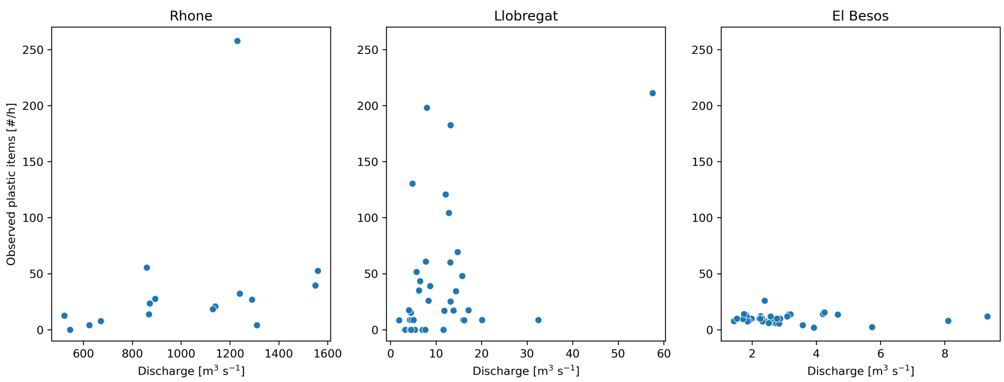

Figure 1: Plastic flux observations and their corresponding river discharge for the Rhône, Besós and Llobregat rivers. Note the difference in scales on the x-axis.

River plastic litter samples for mass conversion

To convert the floating litter time series to mass transport we used two datasets that quantified the mass of sampled floating litter items in the Rhine river, Netherlands (Vriend et al., 2020b), and the Saigon river, Vietnam (van Emmerik et al., 2019). The Rhine dataset includes 508 analyzed floating litter items collected from a litter trap in the Lekhaven, Rotterdam. The Saigon dataset includes 3057 analyzed floating litter items sampled using a 1 m by 0.5 m net, deployed from the Thu Thiem bridge in Ho Chi Minh City. In both studies, the mass, size and polymer category of the collected items were determined. For our analysis, only the items larger than 2.5 cm were used (n = 452 for the Rhine, n = 2123 for the Saigon), as this represents the smallest visually observable size reported in RIMMEL (González-Fernández and Hanke, 2017).

Hydrometeorological data

We used daily mean river discharge and wind speed, and daily total precipitation data from measurements stations closest to the litter observational sites. All data have been retrieved through the respective sources referred to in the original studies, see references in Table 2.

Catchment level mismanaged plastic waste estimates

To estimate the total Mismanaged Plastic Waste (MPW) leaked into each river basin we used the global MPW estimates from Lebreton and Andrady (2019). This dataset uses country-level waste management data and high-resolution data of population and gross domestic product to estimate MPW globally at 1 km resolution. We determined the total annual MPW leaked into the river basins using the HydroSHEDS catchment database (Lehner et al., 2008). This database includes Digital Elevation Model derived river networks and delineated catchments.

Land-cover classes

For the land-cover classes we use a global dataset at the same resolution as MPW (Tuanmu and Jetz, 2014). For each grid cell, the fractional contribution of each of the 17 land-cover classes is given as a percentage.

Statistical analysis

Riverine plastic transport estimates approximate the full annual plastic flux of a river by sampling the plastic load of a subcomponent of the total flux (e.g. visually counting floating macroplastics over a section of the river width, or measuring the microplastic density in a water sample) and extrapolate these discrete observations of plastic items to a continuous plastic mass flux. In this study we quantify the uncertainty introduced by the item-to-mass conversion and the parameter uncertainty in the above identified models. To model the plastic flux from observed floating items, a linear extrapolation over the river width (if not sampled) is performed and in all models a component for depth extrapolation is included. The uncertainty introduced by this width and depth conversion is not analysed in our study (due to the lack of sufficient data) but reviewed in the discussion section.

Item-to-mass conversion

For item-to-mass conversion (m_p) much more suitable data has been obtained in recent years. To estimate the uncertainty of this conversion factor, we used data measured in the Rhine and Saigon rivers (as described in section 3.1). In order to compare these plastic mass and size measurements with floating plastic observations from bridges, we excluded items smaller than 2.5 cm, which is the minimum detectable item size of floating items from bridges as stated by Castro-Jiménez et al. (2019). To estimate the uncertainty of the item-to-mass conversion, we sampled the two datasets repeatedly with an increasing sample size and calculated the mass of each sample. The smallest sample size is 1 and the highest sample size is 250, which is the largest item count per hour that has been reported in the combined Rhône and Spain studies. For each sample size, we draw 1000 bootstrap samples (with replacement).

Parameter uncertainty in models

M1: Temporal extrapolation

In *temporal extrapolation* the annual plastic flux is estimated using the following equation:

$$M_{out} = dm_p T\bar{X} \tag{1}$$

with M_{out} being the annual plastic mass at the point of observation, $\bar{X}$ the average plastic count per hour and T the number of hours per year (8760, in a regular year), d a constant factor converting the surface flux to the whole flux and m_p is the item-to-mass conversion factor. Besides the uncertainty introduced with d and m_p, this formulation assumes that the average of the observed plastic count per hour $\bar{X}$ approximates the unknown true average hourly plastic flux. To estimate the accuracy of this method with the currently available data, we bootstrap the original sampling data of the three rivers separately 1000 times (retaining the original sample size) and analyse the resulting distribution of $\bar{X}$.

M2: Linear regression with environmental drivers

In *linear regression with environmental drivers*, we calculate the plastic flux from a linear combination of discharge, precipitation and wind speed for each catchment individually. As the effect of wind and precipitation on river plastic load is likely not instantaneous, we include experiments that use a time lag between the environmental driver and the plastic flux. To determine these time lags, we calculate the correlation between plastic observations and aggregated hydrometeorological fluxes (sum of precipitation and average of discharge and wind speed) over an increasing number of days (max 45). We use the time lag l that results in the highest correlation coefficient in the time lag experiments. We then use the following formulation of the regression models:

$$M_{out,t} = \beta_1 Q_{l_Q \to t} + \beta_2 P_{l_P \to t} + \beta_3 U_{l_U \to t} \tag{2}$$

where $\beta_1 Q_{l_Q \to t}$ is the average discharge over the period of l_Q days before the day of the plastic observation t, and l_Q is the optimal lag-time with respect to discharge. $\beta_2 P_{l_P \to t}$ represents the accumulated precipitation over the period l_P days before t and $\beta_3 U_{l_U \to t}$ the average wind speed over the period of l_U days before t. β_1, β_2 and β_3 are regression parameters. Combining this equation with equation 1, to define the regression parameters, results in:

$$dm_p X_t = \beta_1 Q_{l_Q \to t} + \beta_2 P_{l_P \to t} + \beta_3 U_{l_U \to t} \tag{3}$$

Subsequently, the plastic observations can be isolated:

$$X_t = \frac{\beta_1}{dm_p} Q_{l_Q \to t} + \frac{\beta_2}{dm_p} P_{l_P \to t} + \frac{\beta_3}{dm_p} U_{l_U \to t} \tag{4}$$

And simplified by generating :

$$X_t = \beta_1^* Q_{l_Q \to t} + \beta_2^* P_{l_P \to t} + \beta_3^* U_{l_U \to t} \tag{5}$$

We set up the regression in eight different ways, to isolate the effect of the different transport mechanisms on the plastic flux and to assess whether the model improves when including time lags between the environmental driver and the plastic flux. In the first four experiments we do not use time lags, i.e. we set all l_i parameters to 0. We first test the discharge individually, i.e. set the 5 and 6 parameters to zero, to isolate the effect of the discharge on the plastic flux. We then add precipitation and wind speed separately and in combination, resulting in four different regression models. (Experiments using precipitation or wind speed without discharge resulted in poor model performance and have hence been omitted in the subsequent analysis). As the effect of all these environmental drivers is likely not instantaneous, we then repeat the four experiments using a time lag as described above between the environmental driver and the plastic flux.

To quantify the parameter uncertainty of this model setup, the regression models are set up a 1000 times with bootstrapped data (retaining the original sample size). Subsequently, this ensemble is used to calculate the plastic flux for every day during the year, using the whole hydrometeorological dataset. For each regression model, the predicted daily plastic item fluxes are then averaged over the entire year, resulting in one estimate for the annually averaged plastic item flux per model. This method yields 1000 bootstrapped estimates of the annually averaged plastic item flux in each of the three catchments. This distribution is directly proportional to the distribution of the average plastic mass flux per hour, assuming constant d and m_p.

M3: Exponential regression with environmental drivers

This third modelling strategy is implemented in the global modelling studies of Lebreton et al. (2017), Schmidt et al. (2017), and Mai et al. (2020). All three studies use global land-based plastic pollution estimates to extend the regression models described above to be able to predict plastic fluxes in every global catchment. The proposed models differ slightly, Lebreton et al. (2017) use catchment integrated runoff data, while Mai and Schmidt use discharge directly, also different land-based plastic pollution estimates were used. Note that the pollution within a catchment is averaged for use in the regression equation, i.e. the spatial distribution of the plastic pollution within a catchment is ignored. All three models can be described with the following nonlinear equation:

$$M_{out,t} = \alpha (W Q_t)^{\beta} \tag{6}$$

where α and β are the parameters that need to be estimated, Q_t the discharge at a given time and W the integrated land-based plastic pollution in the catchment. β expresses the idea of exponentially increasing plastic flux as a function of discharge, while α is the linear scaling factor, linking discharge to plastic.

To isolate the uncertainty of this model formulation from the uncertainty of the land-based plastic pollution estimates, we assume the average plastic pollution within a catchment, W,to be constant over time and we derive the regression parameters for all three catchments individually. Firstly, $M_{out,t}$ is replaced by its approximation from equation 1 and the exponent is propagated to both the discharge and W:

$$dm_p X_t = \alpha W^\beta Q_t^\beta \tag{7}$$

Subsequently, the plastic observations (X_t) are isolated and all (assumed constant) unknowns are combined in a new regression parameter c:

$$X_t = \frac{\alpha W^\beta}{dm_p} Q_t^\beta = c Q_t^\beta \tag{8}$$

By taking the natural logarithm at both sides this equation becomes linear (assuming α and β positive), making it possible to perform linear regression to obtain the values of the parameters:

$$\ln(X_t) = \ln(c) + \beta \ln(Q_t) \tag{9}$$

To derive a measure of uncertainty caused by this model formulation, the regression model is set up 1000 times with bootstrapped data (retaining the original sample size) for each catchment. Similarly as in linear regression (M2), the parameter sets are then used to calculate the floating plastic item flux X_t for every day of the year during which the plastic samples were taken. The distribution resulting from averaging X_t over time is a measure for the uncertainty stemming from this model formulation, independent of the uncertainty in the estimates of land-based plastic pollution. Additionally, the R^2-value distribution of the model ensembles are discussed and compared between the three catchments. Lastly, to assess the validity of applying such a model at the global scale, the resulting distribution of β parameters for the three catchments (as resulting from the bootstrap analysis) are compared to each other. In the global modelling studies, a single β parameter for all global catchments was used, with a resulting value of slightly above 1 (Lebreton et al. (2017) 1.5, Mai et al. (2020) 1.12, Schmidt et al. (2017) 1.28). If this modelling assumption is valid, our analysis should yield similar numbers and should be consistent between catchments.

M4: Spatial probabilistic modelling

The *probabilistic modelling* approach has been proposed by Meijer et al. (2021). It defines the plastic flux leaving the catchment at any given time as a function of spatially distributed probabilities, combined with data on land-based plastic pollution (as in *exponential regression with environmental drivers*, but here without averaging the plastic pollution within a catchment).

To calculate the plastic flux at the river mouth, the probability of plastic emission in each grid cell is multiplied with the respective land-based plastic pollution within the grid cell:

$$M_{out,t} = \sum_{i=0}^{n} c P(E)_{i,t} W_i \tag{10}$$

Here $P(E)_{i,t}$ is the probability of the plastic in grid-cell i reaching the river mouth at time t(the probability of emission), c a parameter that summarizes the accumulation of pollution over time and clean-up of the mismanaged plastic waste, as a single catchment wide constant) and W_i is the land-based plastic waste in grid-cell i. n is the number of grid-cells in the catchment. $P(E)_{i,t}$ is calculated by intersecting the following probability maps: 1) probability of mobilisation ($P(M)$), 2) probability of reaching the river network ($P(R)$) and 3) probability of reaching the river mouth ($P(O)$). The different probabilities are assumed independent here, as they include different drivers. In practice this assumption might not hold under extreme weather conditions.

$$P(E)_{i,t} = P(M \cap R \cap O)_{i,t} = P(M)_{i,t} * P(R)_{i,t} * P(O)_{i,t} \tag{11}$$

The first probability map, $P(M)_i$, defines the probability of plastic mobilisation at a given grid cell. The grid cells are assumed to be composed of fractions with different land-use, which have different mobilisation probabilities. This is expressed as follows:

$$P(M)_i = \vec{LU}_i \cdot \vec{f}^T = \vec{LU}_i \cdot [f_{forest}, f_{agriculture}, f_{build-up}, f_{barren}, f_{water}] \tag{12}$$

Where $\vec{LU}_i$ is the land-use classification vector (containing fractions of the land-use types) and $\vec{f}$ is a vector describing the likelihood of plastic mobilisation for each land-use type. The likelihood of plastic mobilization within open water (f_{water}) is assumed to be 1, the values for the remaining probabilities f_{forest}, $f_{agriculture}$, $f_{build-up}$, f_{barren} are estimated in the optimization process.

The second probability map expresses the likelihood of plastic to arrive in the river ($P(R)_{i,t}$), which we assume to be inversely proportional to the distance of the river network for each grid cell. For simplicity we assume that transport only occurs through wind and that the probability of litter reaching the river network decreases exponentially with the distance to the river:

$$P(R)_{i,t} = \exp\left(\frac{-\alpha\delta_{RN,i}}{U_t}\right) \tag{13}$$

With $delta_{RN,i}$ is the distance between the grid cell (i) to the closest point on the river network, U_t wind speed at time t, and α a proportionality constant that is estimated in the parameter optimization. The third probability map describes the likelihood of litter to be transported towards the river mouth. In a similar fashion as the previous calculations this can be defined as being inversely proportional to the distance between each grid cell and the river mouth. Here we will use an identical exponential formulation but use discharge:

$$P(O)_{i,t} = \exp\left(\frac{-\beta\delta_{RM,i}}{Q_t}\right) \tag{14}$$

Where $\delta_{RM,i}$ expresses the distance between the grid cell (i) and the river mouth, Q_t the average discharge and β a proportionality constant.

To be able to constrain this model setup, equation 10 is combined with equation 1, and the floating plastic observations are isolated. Subsequently, the conversion constants are grouped together in one parameter q:

$$X_t = \frac{c}{dm_p} \sum_{i=0}^{n} P(E)_{i,t} W_i = q \sum_{i=0}^{n} P(E)_{i,t} W_i \tag{15}$$

The final equation includes 7 parameters (f_{forest}, $f_{agriculture}$, $f_{build-up}$, f_{barren}, α, β and q) which are optimized using the trust region reflective algorithm as implemented in the python package scipy (https://scipy.org/).

To assess the parameter uncertainty of this model setup, we examine the point estimate and standard deviation of the optimised parameters and their propagation to the resulting probability maps.

4 Results

Observational uncertainty

We used observations from the Rhine and Saigon rivers to constrain the error introduced by converting items to mass. Figure 2 shows how the uncertainty in conversion is dependent on the sample size. In general, larger samples are associated with lower uncertainty. However, even with 250 samples the item-to-mass conversion error is still substantial. With sample sizes of the magnitude as found in the Llobregat, Besós and Rhône rivers, this conversion is subject to a possible 2 to 3 orders of magnitude of uncertainty when assuming an item-mass distribution comparable to the Rhine and Saigon rivers.

Model uncertainty

M1: Temporal extrapolation

The only non-parametric method for modelling the riverine plastic flux is extrapolating the observations over time. Figure 3 shows the results for a 1000 times bootstrapping analysis of this modelling strategy for the Rhône, Llobregat and Besós rivers. The resulting data based uncertainty of this modelling strategy is proportional to the variance in the original data (see Figure 1), with the Rhône showing the largest spread in predictions, while the Besós has a very low spread. Altogether, the predictions show 1 to 2 orders of magnitude lower spread than caused by the item-to-mass conversion, but remain significant.

M2: Linear regression with environmental drivers

Linear regression models that predict the plastic flux based on hydrometeorological fluxes are widely used. The underlying hypothesis is that the hydrometeorological fluxes (precipitation, wind speed and discharge) are the main drivers of plastic propagating through the river system. As the effect of all these environmental drivers is likely not instantaneous, we include lags between the environmental driver and the plastic flux in our analysis. Figure 4 shows the correlations between precipitation, wind speed and discharge and the plastic flux observation for different time lags. For the time lags we use aggregates of the environmental drivers, sum for precipitation and average for discharge and wind speed (see Methods).

The three rivers differ strongly in terms of the correlations between the plastic flux and the environmental drivers. Precipitation is positively correlated with the plastic flux for all time lags in the Llobregat and Rhône, but negatively correlated in the Besós river. The time lag resulting in the highest correlation is 8-9 days for Llobregat and Rhône but 25 days for the Besós. Correlation between discharge and plastic observations is positive (or approximately zero) for all rivers and all time lags, but never higher than 0.5. Interestingly, the correlation coefficient for the Rhône and Llobregat rivers decrease with increasing time lag, while the Besós river displays a peak at around a 7-day lag. The wind speed - plastic flux correlations show a more peaky

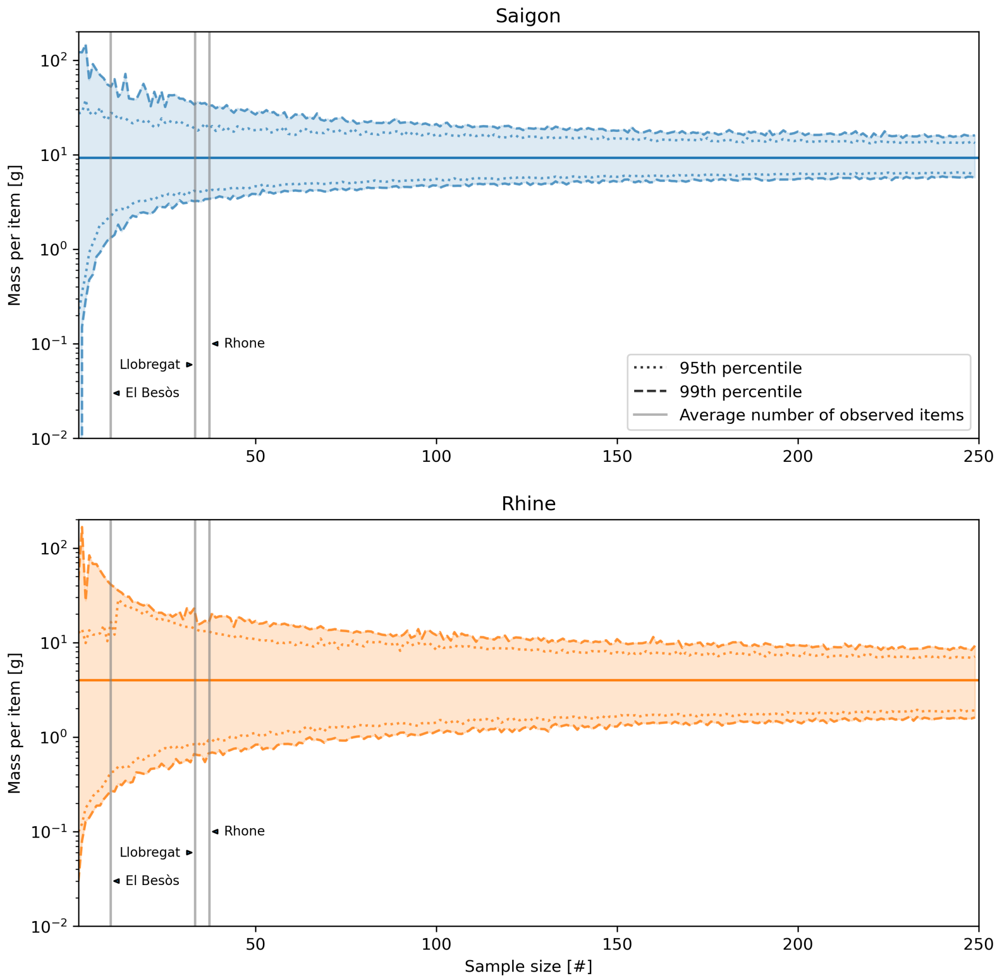

Figure 2: Uncertainty in item-to-mass conversion. Estimated plastic mass as a function of sample size for the Saigon and Rhine rivers. The horizontal lines display the mean mass of all items in the two datasets (2123 and 452 items for Saigon and Rhine, respectively), which would be used as the item-mass conversion factor. The colored area above and below these lines display the area between the 1st and 99th percentile confidence interval from a bootstrap analysis. The uncertainty range (99th percentile divided by the 1st percentile) from mass conversion for the Rhône, Llobregat and Besós is 24, 34 and 151, respectively, when assuming a similar item-mass distribution to the Rhine. The vertical lines depict the average number of items found in the Rhône Llobregat and Besós rivers per monitoring session.

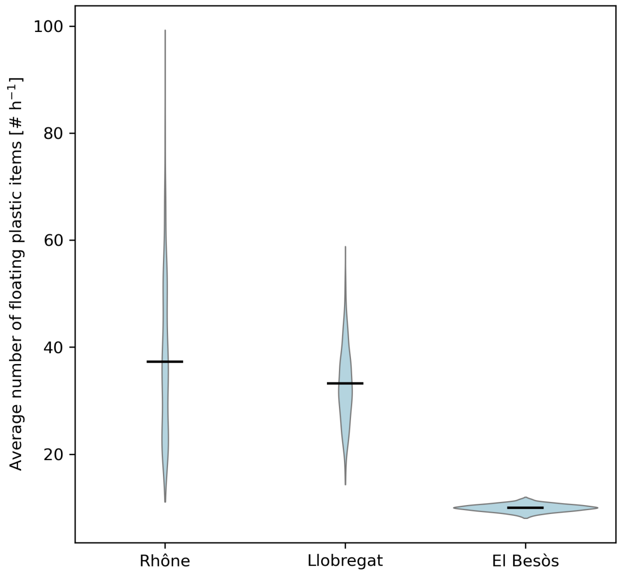

Figure 3: Bootstrapped uncertainty of the average floating plastic flux (X) using simple extrapolation. The violins represent the estimated average floating plastic flux of a 1000 bootstraps (of the original data size) of the plastic observations of the respective rivers. The black horizontal lines display the mean of the observations for the respective rivers.

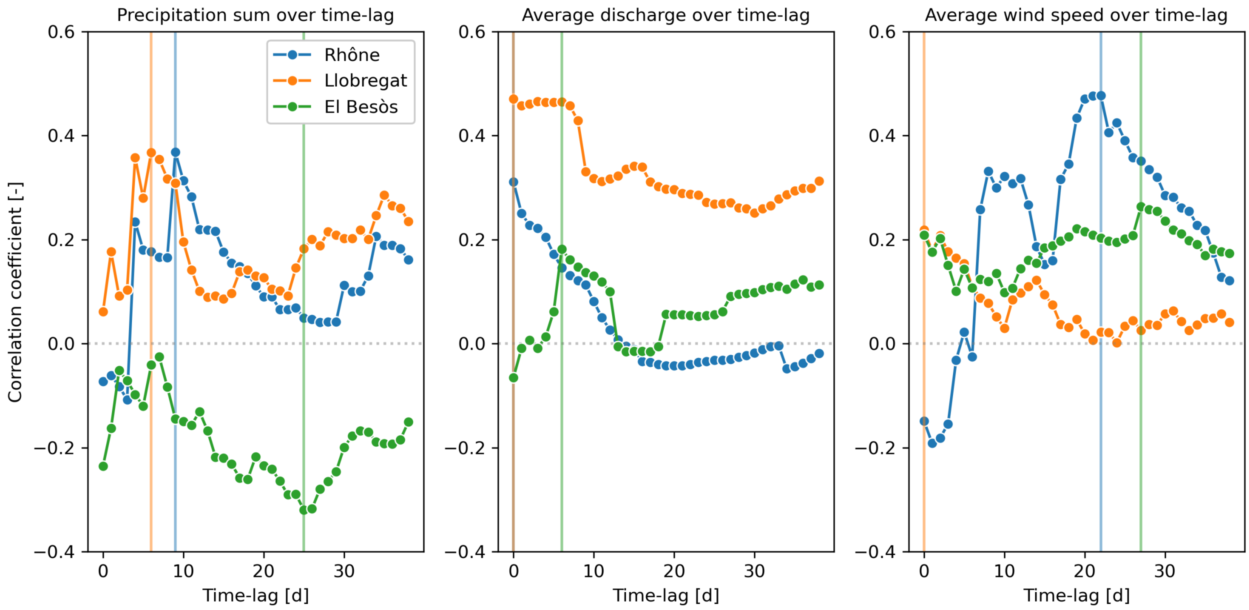

Figure 4: Pearson correlation between floating macroplastic observations and hydrometeorological fluxes under different time-lags. The vertical lines depict the highest absolute correlation values per river-flux pair.

pattern, with maximum correlation values observed between 20 and 30 days for the Rhône and Besós rivers, while the Llobregat displays a decreasing correlation trend, with the highest value observed without time lag.

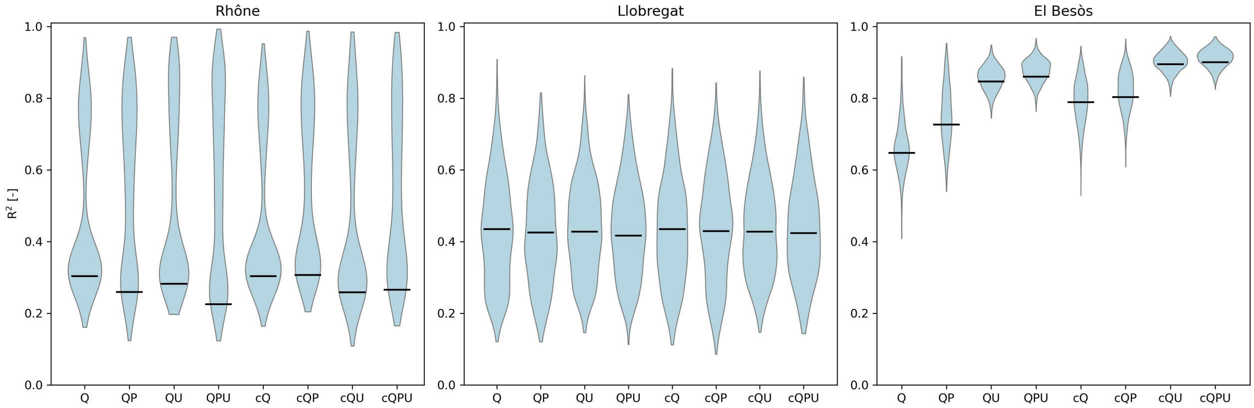

Figure 5: Distribution of the adjusted-R^2 values corresponding to the linear regression models linking hydrometeorological fluxes (Q - discharge, P - precipitation, U - wind speed; without lag-time: Q ; with lag time: cQ) to the plastic flux. The black horizontal lines display the adjusted-R^2 of the models with the original data.

Using the optimal lag times for the hydrometeorological fluxes defined above, the model is next applied on the bootstrapped data. Figure 5 shows adjusted-R^2 value of the different model configurations for the three rivers. Plastic observations in the Rhône river can be predicted with adjusted-R^2 of around 0.3, in the Llobregat river around 0.45 and plastic in the Besós is predicted with an adjusted-R^2 value of over 0.6 for all tested combinations of environmental drivers. Only the Besós model shows an increased R^2 with increased model complexity, while the Rhône model even decreases in performance when including more than discharge alone. In general, including a time-lag in the model does not seem to increase model performance. The distributions caused by bootstrapping the training data are very large for both the Rhône and Llobregat, ranging from just under 0.2 to close to 1, indicating a very high dependence of performance on the input data, which in general indicates a model structure poorly fitting the system. The distribution of the Besós models is much smaller, which can be explained by the very low variance in the input data. Fitting three parameters on almost homogeneous data very likely results in an overfitted model, making it hard to interpret the high R^2 values.

Figure 6, containing the predicted average hourly plastic flux, shows largely the same patterns. Including further parameters in addition to discharge does not change the distribution of the predictions for the Rhône and Llobregat, while the Besós sees a reduced variance in the predictions (and interestingly a slight increase of the mean predicted plastic flux). In all models and all catchments, including precipitation without lag-time substantially increases model output uncertainty. In comparison to temporal extrapolation of the data, the range and variance of the model predictions slightly decrease for the Rhône river, stay roughly equal for the Llobregat and increase for the Besós.

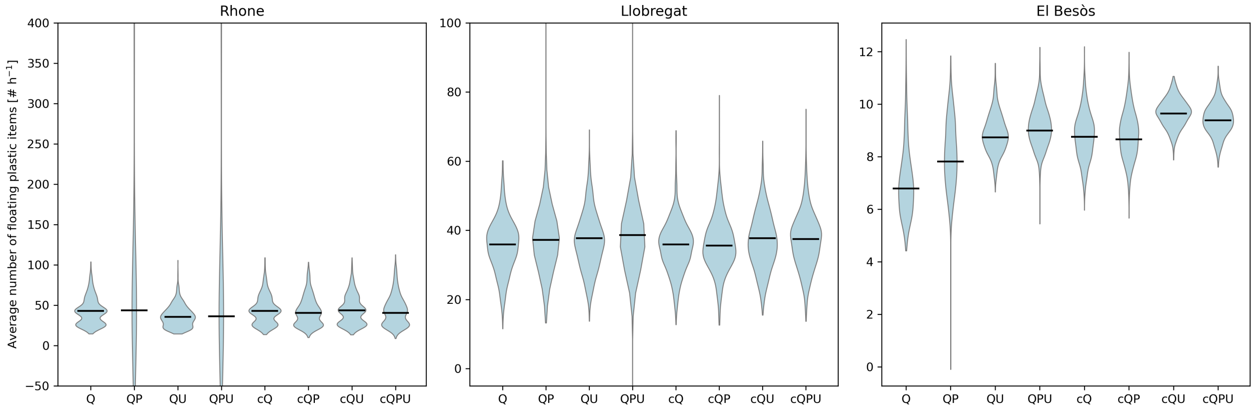

Figure 6: Distribution of the average hourly plastic item predictions from the models. The black horizontal lines display the predictions of the models trained on the original data.

M3: Exponential regression with environmental drivers

The plastic flux - discharge relation has been used to model the global plastic output into the ocean for all catchments (Lebreton et al., 2017; Schmidt et al., 2017; Mai et al., 2020). This method uses plastic waste estimates within the catchment in combination with discharge to derive the plastic flux. In our uncertainty and performance analysis, the waste component of the model is abstracted away in the model parameters as it is constant within the individual catchments. This approach is used to focus on the uncertainty of the model structure, instead of the combined uncertainty of waste estimates and model parameters.

The model performance of the *exponential regression with environmental drivers* model (adjusted-R^2) is displayed in Figure 7A. Average performance in comparison with temporal regression is approximately equal for the Rhône river, while being substantially lower for both the Llobregat and Besós. The distribution of adjusted-R^2 values of the Rhône models does not show the bimodality present in temporal regression, but the range increases to fill the whole domain. Again, this indicates a model structure that is very dependent on input data, raising the question how well the model structure represents the system. The Besós river has especially low adjusted-R^2 values. This is due to the model structure and the very low variance in the plastic flux observations. The model shows that the gradient over the discharge is almost zero, with the intersect describing the data (see following paragraphs). This results in a very low adjusted-R^2.

The averaged hourly plastic flux predictions of the bootstrapped models are displayed in figure 7B. Interestingly, the model predictions for both the Rhône and Llobregat rivers are substantially lower than both temporal extrapolation and linear regression. For the Besós, exponential regression models show very similar results to temporal extrapolation models, again due to the fact that the model structure is almost exclusively determined by the intercept, which lies very close to the mean of the data. Figure 7C shows the exponential parameter of the model configurations. As described above, the Besós river has an exponent very close to zero, which eliminates discharge effect on the result. Llobregat has an exponent of slightly above 1, making it very similar to the linear regression models described in the previous section. The increased performance and lower variance in the results can be attributed to the inclusion of an intercept in the model, which seems to help

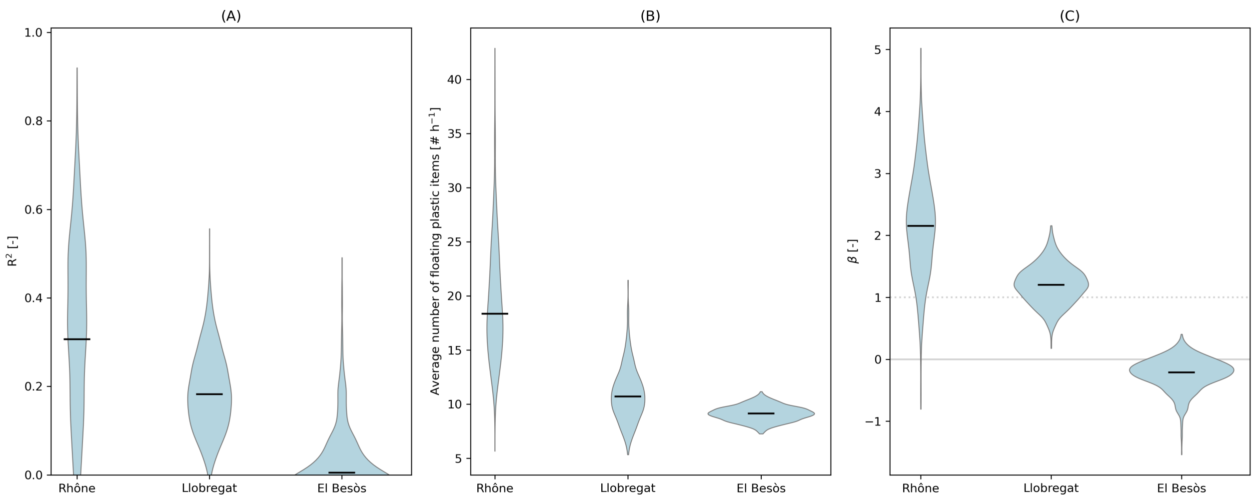

Figure 7: (A) distribution of the R^2 values corresponding to the exponential regression (with environmental drivers) models linking non-linear discharge to the plastic flux, (B) distribution of averaged hourly predictions from the exponential regression models (from the whole year of predictions) and (C) distribution of the exponential parameter of the exponential regression model configurations for the different rivers. The black lines describe the modelling results on the original data.

to explain the data. The Rhône river shows a much higher exponent, but the variance in the bootstrapped model in this parameter shows that the data poorly constrains the model.

The (globally constant) exponent in the models discussed in Mai et al. (2020) and Lebreton et al. (2017) lies slightly above 1, which seems to fit the Llobregat catchment. Calculating the global riverine plastic emissions following Lebreton et al. (2017), but replacing their fitted exponent (1.52) with the range of exponents found here (from -0.23 to 2.17), the global emissions would lie between 0.06 and 68 million tonnes per year (4 orders of magnitude difference). In contrast, Lebreton et al. (2017) reported that emissions lie between 1.15 and 2.15 million tonnes per year. It is likely that some rivers have higher and others lower exponents, possibly reducing this large range of possible values. Nonetheless, considering only the Yangtze, the river with highest emissions reported by Lebreton et al. (2017), the range of possible exponents leads to possible emission from the Yangtze between 3 tonnes and 32 million tonnes (23 times bigger than the global emissions estimates by Lebreton et al. (2017)). Similar uncertainties are to be expected in all exponential models.

M4: Spatial probabilistic modelling

Spatial probability modelling links information on land-based plastic waste and data on hydrometeorological fluxes to derive spatially explicit maps of plastic reaching the river mouth. After parameter optimization, maps of the probability of plastic mobilisation and annual average maps of the probabilities of plastic reaching the river network and reaching the river mouth are drawn. In contrast to the bootstrapped performance analysis done for the other model categories, here we analyse the point estimate and the uncertainty intervals of the probabilities resulting from the parameter optimisation. By generating the probability maps for the parameter combinations and parameter combinations shifted by their standard deviation it can be analysed how well the available data constrains the model configuration.

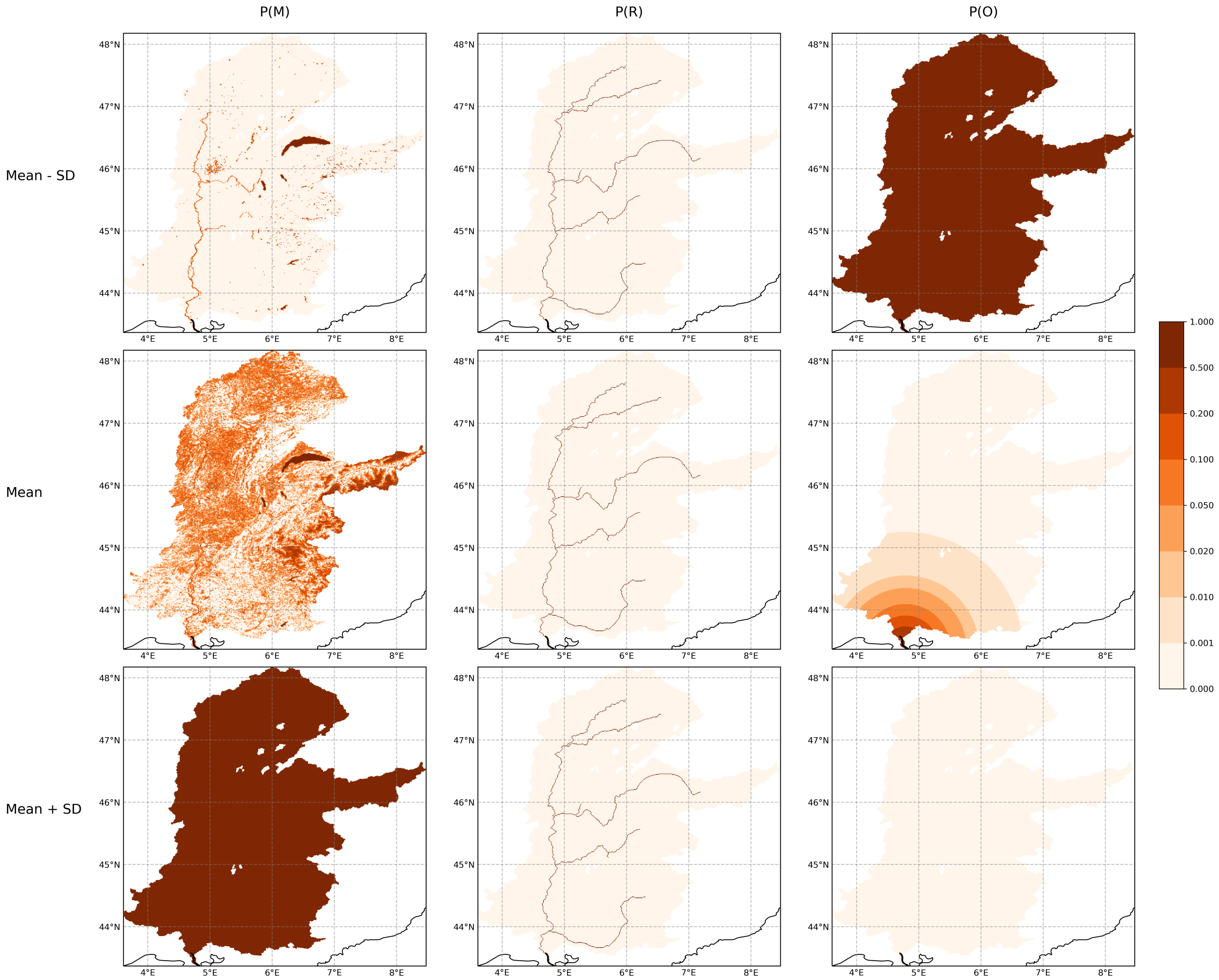

Figure 8: probability maps of mobilisation (P(M)), reaching the river network (P(R)) and reaching the river mouth (P(O)) for optimal parameters and parameters plus/minus one standard deviation. The plots represent the catchment of the Rhône river.

The probability maps are presented in figure 8, with the middle row showing the optimal parameter values, while the first and last row are created by subtracting and adding one standard deviation to the parameters, respectively, and rerunning the model. The maps show that at the distance of one standard deviation from the optimal parameters, both the probability of mobilisation P(M) and the probability of reaching the river mouth P(O) range from 0 to 100 percent, irrespective of location. This parameter range demonstrates that, with the currently available data, models of this complexity cannot be constrained. Interestingly, the probability of reaching the river network (P(R)) is approximately zero everywhere, except for the pixels containing the river network (representing a distance of at most 1 km), where the values are 1. This remains the case for both the optimum parameters as well as for the parameter shifted by their standard deviation. Although no concluding evidence, these results indicate that the footprint of river plastic lies very close to the river channel.

5 Discussion

Here, we conducted the first comparison of riverine plastic flux models, quantifying their robustness and comparing their underlying assumptions. We use three of the most extensive observational datasets for floating macroplastics in single rivers available to date. These datasets contain as many macroplastic observations per river (Rhône n=16, Llobregat n=50, Besós n=36) as the total number of observations that informed the global models of Lebreton et al. (2017)(macroplastics n=6, total plastic n=30), Schmidt et al. (2017) (macroplastics n=35, total plastic n=45) and Mai et al. (2020) (macroplastics n=2, total plastics n=80). We focused on macroplastics in this study, because in the most recent data compilations, macroplastics constitute more than 90 percent of total mass flux (Mai et al., 2020).

Uncertainty from observations

A substantial source of uncertainty revealed in our analysis results from the conversion of visual plastic item observations to the complete riverine plastic flux in mass. We find an average item-to-mass conversion factor between 5 and 10 grams per item (depending on the reference dataset), using the largest observational datasets of sampled riverine macroplastic items. The uncertainty in the conversion factor depends on the number of items found per observational round. It can reach three orders of magnitude if less than ten items are observed (Figure 2). More than half of the observations of the rivers studied here fall below this threshold (Rhône = 63%, Llobregat = 55%, Besós = 63%). Note that the conversion factors for macroplastics used in e.g. Lebreton et al. (2017); **?** are much lower, corresponding to the difference in sizes included in the analysis (this analysis focuses on items above 2.5 cm, while Lebreton and Mai include everything above 0.5 cm). Including this range to the analysis, however, would only increase the distance between minimum and maximum mass per item.Further uncertainty is caused by the extrapolation of these observations over the whole river width (if not sampled, such as in the Rhône) and river depth. Currently data availability is too limited to characterise this uncertainty further, but some initial findings indicate that they are not constant over time (Haberstroh et al., 2021).

Uncertainties from model structure

The second component of uncertainty quantified in this study is the uncertainty of model parameters. This reaches up to an order of magnitude for M1, M2 and M3. The model with the lowest prediction uncertainty depended on the selected catchment, with M2 being the most suitable for the Rhône, both M1 and M3 for the Besós, while for Llobregat all three models showed roughly equal results. However, the adjusted-R^2 values show a very large distribution (in the worst case almost covering the entire 0-1 range) for both M2 and M3 (cannot be calculated for M1) for the Rhône and Llobregat. This indicates that the predicted values are extremely dependent on the bootstrap, and the model setup shows very low robustness. All models seem to be working quite well for the Besós river, including M1, which can be explained by the comparatively low variance in the data (Figure 1). M3, the model setup used for the global modelling studies, surprisingly never outperformed M2. Additionally, M3 generates an exponential parameter ranging from -0.23 to 2.17.

Inserting this exponent into the model implemented in Lebreton et al. (2017) leads to global plastic emission estimates ranging between 0.06 and 68 million tonnes per year, a difference of four order of magnitude (a much larger range than reported by Lebreton et al. (2017). Our analysis of the last modelling strategy, the spatial probabilistic modelling, clearly displays the current limits of modeling. The standard deviations of the fitted model parameters are so large that the system is undetermined. With the currently available data it is not possible to constrain spatial probabilistic models and also more complex model setups (e.g. model describing fluxes between different compartments, retention and sinks).

Uncertainty from environmental drivers of plastic transport

Except for simple temporal extrapolation, all models explicitly use environmental drivers (in particular river discharge) as predictors of plastic transport. However, we show that the correlation between the plastic flux and river discharge, precipitation and wind speed is comparatively low, with values below 0.5 in all river basins and under a range of time lags (Figure 4). We hence conclude that river discharge, precipitation and wind speed are poor predictors of plastic transport, at least in the rivers that we analysed. Models using these environmental drivers might not even outperform simple temporal extrapolation models. Additionally, our results suggest that only a small area around the river network (about a 1 km zone) contributes to the riverine plastic flux (Figure 8). These results depend on terrestrial mismanaged plastic estimates, which may be significantly too high (Mai et al., 2020). Lower levels of terrestrial pollution would allow for further plastic transport over land within the models. However, if terrestrial plastic transport is indeed limited to very few kilometers, global regression models that consider the plastic pollution within the entire catchment are using a predictor that may be irrelevant for riverine plastic pollution. The amount, time and location of plastic entering the river potentially constitutes the biggest source of uncertainty in riverine plastic modelling, but is not quantifiable with the currently available data.

Uncertainties in the connection between river discharge and plastic flux

Despite the uncertainties discussed above, global modelling studies (e.g. Lebreton et al., 2017; Roebroek et al., 2021a; Mai et al., 2020) have unanimously found correlations between environmental pollution estimates, the average discharge of a river and isolated plastic flux observations. The accepted paradigm is that rivers with high discharge and flowing through catchments with high levels of pollution also carry a large amount of plastic items. Some studies additionally assume a temporal correlation between the discharge and the plastic flux. However, we show here that river discharge in and by itself does not necessarily predict the riverine plastic flux very well (see for example the Besós in Figure 1) and temporal correlations between discharge and plastic flux are low in all rivers analysed (see Figure 4). We therefore conclude that the driver of temporal variations in the plastic flux within rivers is currently not understood (and likely differs strongly between catchments), and we advise against using discharge to deduce the seasonality of plastic flux within a river. We acknowledge however that a correlation between plastic flux and discharge likely exists under extreme conditions. Obvious examples are when rivers temporarily dry up or emerge from the riverbed under flood conditions.

In terms of correlations between the annually averaged discharge of a river and the plastic flux, our results have only limited explanatory power as we only analyse three rivers. However, within those three rivers there is little connection between the average discharge and the plastic flux, the Rhone (average discharge 1093 $m^3 s^{-1}$, average 37 plastic items) carries a similar amount of plastic items as the Besós (average discharge 3 $m^3 s^{-1}$, average 10 plastic items, Figure 1). The correlations between plastic flux, environmental pollution estimates and discharge found in global studies are mostly based on microplastic observations which have been converted to macro- or total plastic mass estimates (for example, the Lebreton et al. (2017) study and the Mai et al. (2020) studies include only six and two macroplastic observations, respectively). As discussed above, these conversions are highly uncertain and the correlations presented in these global modeling studies may simply be unreliable. Another possible explanation may be that the amount of plastic a river carries is mainly determined by the environmental pollution level within the catchment. Lebreton et al. (2017), Schmidt et al. (2017) and Mai et al. (2020) use national statistics on the public waste management system and population density within the river catchment to estimate the environmental pollution levels. However, a recent comparison of global terrestrial plastic pollution estimates (Edelson et al., 2021) clearly show that environmental pollution levels are currently not well understood, either. We conclude that overall, the relation between environmental pollution, discharge and riverine plastic transport is very complex and only beginning to be understood. The current modelling approaches rely too much on the relationship between the river plastic flux and discharge, and cannot resolve the complexity of the system accurately.

Data and code availability statement

All data are openly available through the original references. Plastic and hydrometeorological data: Castro-Jiménez et al. (2019); Schirinzi et al. (2020). Mismanaged plastic waste estimates: Lebreton and Andrady (2019).

Author contributions

Conceptualization: CR; Methodology: CR; Formal Analysis: CR; Investigation: CR, CL, DGF, TvE; Visualization: CR; Writing–original draft: CR, CL, TvE; Writing–reviewing and editing: CR, CL, DGF, TvE.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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