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Predicting microplastic masses in river networks with high spatial resolution at country level

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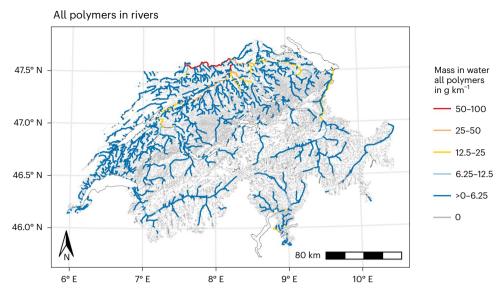


Microplastics are a ubiquitous contaminant of natural waters, and a lot of field monitoring is currently performed. However, what is missing so far is a general understanding how emissions of microplastics are linked to environmental exposure, especially on larger geographic scales such as countries. Here we coupled a high-resolution microplastic release model with a fate model in rivers and lakes and parameterized it for Switzerland on a country scale to predict masses of microplastics in each river section for seven different polymers. The results show that catchment characteristics, for example, distribution of releases within the catchment, location and size of lakes or river connections, are as important as polymer properties such as density. There is no simple linear function of microplastic retention within a catchment in dependency of river length to the outlet. Instead, we found that different catchments cover a wide range of retained fractions for microplastics. Consequently, we argue that the availability and use of spatially distributed release data and performing modelling on high spatial resolution is of importance when estimating concentrations of microplastics in large areas such as countries.

Across the globe we are facing an increasing emission of plastics into the environment¹. Consequently, plastics, and in particular microplastics, are present across all water bodies in every corner of the world: from large to small lakes, from rivers to ground water and from remote glaciers to deep-ocean sediments²⁻⁷. Most microplastics found in the environment have in common that they were probably not emitted at the sampling site, but were transported there, with rivers being one of the main transport pathways $^{8-13}$. This raises the question of how far microplastics can be transported in rivers and how they vary across catchments, countries or continents with different landscapes and land use. Measurements of microplastics provide snapshots of concentrations at specific locations using a variety of different sampling and measurement methods^{14,15}. Nevertheless, for a more profound understanding of transport processes, a higher temporal and spatial resolution of measurements would be desirable but remains challenging or almost infeasible due to various reasons such as inconsistent measurement quality or comparability across different sampling studies as well as time and funding constraints 16,17.

To overcome measurement limitations, several models of (micro) plastic transport in water have been developed 18-22. However, highspatial-resolution plastic transport models for large areas such as countries or continents are currently available only for macroplastic transport across the oceans (for example, refs. 18,23). Large-area microplastic models for fresh waters simulating the transport of microplastic along river and lake networks are lacking. The only existing freshwater models for microplastic transport cover single catchments with a single river without tributaries in the order of 1,000 km maxi $mum\,river\,length^{19-21}, or\,a\,few\,large\,rivers^{22}, or\,are\,only\,estimations\,on$ small catchment scales without consideration of single rivers²⁴. Additionally, the existing river-based microplastic transport models, that is, nanoDUFLOW¹⁹, INCA-microplastics²¹, Full Multi²⁰ or NICE²², are demanding highly accurate hydrological and particle-specific input

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 $\textbf{Fig. 1} | \textbf{Spatial distribution of the microplastic masses suspended in rivers of Switzerland in g} \quad km^{-1} \\ \textbf{for the sum of the seven studied polymers.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography, swisstopo.} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\ \textbf{The map is based on data by Swiss Federal Office of Topography} \\$

data (for example, particle size, sediment characteristics, water depth, river width and critical shear stress), which highly restricts their application to rivers and catchment for which all these data are available. For any larger areas, for instance in the order of countries or continents, any application is highly challenging. Additionally, all four models were also so far not broadly applied for research questions related to microplastic transport in a variety of different rivers.

The aim of this paper was therefore to develop a new model that could be broadly applied to predict microplastic fate and transport for large geographical areas such as countries with a high spatial resolution. In this Article, we integrated existing fate modelling approaches for microplastics (that is, nanoDUFLOW or the Full Multi model) and a spatially resolved release model with a large-scale hydrological model for whole countries. We applied the model in a case study to Switzerland by predicting the transported masses of seven different polymers (expanded polystyrene (EPS), polypropylene (PP), lowdensity polyethylene (LDPE), high-density polyethylene (HDPE), polystyrene (PS), polyvinyl chloride (PVC) and polyethylene terephthalate (PET)) across all rivers and lakes in Switzerland, using available emission data as input²⁵. Using this case study, we wanted to explore how microplastic transport, environmental concentration and retention differ among rivers and catchments on a country scale caused by the presences of lakes of different sizes and by various catchment characteristics. We hypothesize that focusing on polymer and sediment properties only as done in the past overlooked important drivers of microplastic transport.

Microplastic pollution in Switzerland

We modelled the entire river network of Switzerland as well as three catchments of focus (Rhine, Rhône and Doubs) by connecting each river segment or lake with the corresponding downstream river or lake. The influence of retention by lakes and rivers was assessed by applying different scenarios: no retention (S_0) , retention only in lakes (S_{lake}) , retention only in the 15 biggest lakes in Switzerland (S_{lake1S}) or retention in lakes and rivers (S_{all}) .

Shown as a map (Fig. 1), our model highlights regions of higher-polluted rivers as well as the high number of rivers without expected pollution. All results represent a steady state of emissions in 2014 due to the underlying release model²⁶. Clearly, the mass of transported microplastics increases downstream towards the border of Switzerland with highest masses observed for the river Rhine close

to Basel. On the other hand, smaller rivers and rivers in remote and mountainous regions are less affected by direct microplastic pollution, which explains the high number of non-polluted rivers. Generally, the masses of microplastic accumulation reveal similar trends to masses of suspended microplastics for most of the rivers resulting in concentrated pollution along relative few river sections (see additional maps in Supplementary Figs. 10–15).

Furthermore, river segment pollution of different polymers is very similar when masses are normalized for each polymer by the maximum values per polymer. We observe a highly substantial correlation between all different polymers with a correlation coefficient of 0.9.

On the basis of the different scenarios (S_0 , S_{lake} , S_{lakel} and S_{all}), we predict that half of all input emissions are retained within Switzerland. In detail, we observe that 33% of all microplastics are retained in lakes (Fig. 2), of which 99% are retained in the 15 biggest lakes in Switzerland, although they correspond only to 7% of all lakes receiving microplastic pollution.

While lakes dominate the retention, rivers, on the other hand, are expected to retain only about 17% of all input microplastics. Here we observed a wide range from no retention for EPS up to 40% retention for PET (see colours in Fig. 2). Consequently, the reduction from $S_{\rm lake}$ to $S_{\rm all}$ in Fig. 2 is mainly caused by accumulation of PVC and PET in rivers since lighter polymers are less likely to accumulate in rivers. Hence, the accumulated mass of PET is one order of magnitude higher in rivers than PS accumulation, although the input emissions are slightly higher for PS. Overall, 50% of the plastic mass directly emitted into the water bodies is retained within Switzerland; however, differences in terms of masses among polymers are notable due to differences of retention described by sedimentation ($f_{\rm sed}$) and accumulation factors ($f_{\rm acc}$) (Methods).

Masses and sources of microplastics along the river lengths

Microplastic masses and retention vary not only among different polymers but also along catchments and the lengths of rivers. Here, sedimentation traps such as lakes are highly influencing microplastic masses in suspension but also in sediments. For the three studied catchments (Rhine, Rhône and Doubs) we observed outflowing microplastic masses that differ across three orders of magnitude with a total outflow (in suspension and as sediment transport) of about 4,565 kg per year microplastics leaving the Rhine catchment towards Germany, while

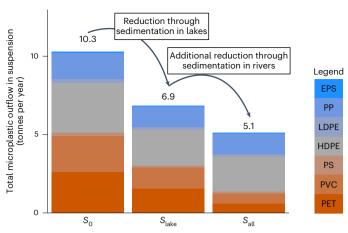


Fig. 2 | **Microplastic retention of all analysed polymers differentiated by different colours in entire Switzerland.** The scenarios S_0 , S_{lake} and S_{all} are different model runs that consider no sedimentation and accumulation (S_0) sedimentation and accumulation only in lakes (S_{lake}) and sedimentation and accumulation in lakes and rivers (S_{all}). S_0 equals the input emission to the system based on Kawecki and Nowack²⁶. The results represent a steady-state system for the year 2014.

300 kg per year and 61 kg per year are leaving the Rhône and Doubs catchment, respectively, towards France. (Fig. 3e–g). Together, these catchments cover about 88% of all microplastic outflows of Switzerland (compare with $S_{\rm all}$ in Fig. 2). The remaining 12% are shared among the outflow of Lago Maggiore (Ticino River) (6%; 327 kg per year) and along flow into 'unknown' downstream river segments (including smaller border crossing rivers, 2%), the Breggia River towards Italy (2%), the Inn River towards Austria (1%) and other smaller rivers.

Generally, rivers flowing through densely populated areas and rivers passing through cities are receiving high amounts of microplastic emissions. Hence, the Rhine at the catchment outlet in Basel is highly influenced by the tributary Aare and its tributaries Reuss and Limmat, which discharge the areas of some of Switzerland's biggest cities (that is, Bern, Zurich and Lucerne). On the basis of our modelling approach, about 2-9% of all microplastic masses leaving Switzerland the three rivers of focus are transported via sediment transport, with higher percentages for rivers with higher total microplastic transport (that is, Rhine).

Retention of microplastics along the river length

Due to the importance of lakes as retention sites, it is crucial to consider the location of lakes in relationship to the locations of the emissions. In Switzerland both biggest lakes with around $500 \, \text{km}^2$, Lake Geneva (Rhône catchment) and Lake Constance (Rhine catchment), are expected to reduce the microplastic mass through sedimentation to about one-third of the inflow masses (Fig. 3b,c). Nevertheless, for the Rhine catchment, the overall retention in lakes is less dominant than for the Rhône (Fig. 4d-f). This is because large mass flows originate from the Aare catchment, which flows into the Rhine only after Lake Constance (Fig. 3b). This is especially true for polymers that are generally less influenced by sedimentation in rivers (that is, EPS, PP, LDPE, HDPE and PS).

In the Rhône catchment Lake Geneva is located close to the outlet of the catchment and therefore retains high amounts of microplastics emitted upstream in the catchment as shown in Figs. 3c and 4b,e. However, just downstream of the lake, the City of Geneva is located, which emits roughly the microplastic masses retained in Lake Geneva. Consequently, the microplastic pollution in the Rhône River at the border with France is mainly driven by the input pollution downstream of Lake Geneva (that is, the City of Geneva). This applies in particular

to the more dense polymers (PVC and PET), since almost all pollution upstream from the lake will be retained in the lake, meaning that the pollution of PVC and PET at the border originates from sources more downstream than other polymers (Figs. 3c and 4b).

For the less dense polymers EPS, PP, LDPE, HDPE and PS, microplastic retention in lakes is much more important than retention in rivers, especially for the more lake-dominated Rhône catchment (Fig. 4). Only for the Doubs catchment with lakes of small surface areas is sedimentation in rivers more dominant for less dense polymers (Fig. 4f and Supplementary Figs. 6 and 7).

Discussion

As a first validation we compared our model with measurement data published in studies about microplastic concentrations in Swiss lakes and the rivers Rhine and Rhône^{4,27}. Both studies present microplastic particle numbers found at the water surface and collected with a trawl. For rivers, we transferred reported results to mass per second using masses per cubic metre and annual average discharge at sample locations. While Faure et al.²⁷ provided masses per cubic metre, we estimated masses for the particle numbers reported in the Rhine (2.5 particles m⁻³) by Mani et al.⁴ assuming either spherical microplastics (diameter 1 mm, volume 0.5 mm³) or microplastic fragments $(1 \text{ mm} \times 1 \text{ mm} \times 10 \text{ } \mu\text{m}, \text{volume } 0.01 \text{ mm}^3) \text{ and a density of } 1,000 \text{ kg m}^{-3}.$ The sizes for the sphere and fragment are based on a representative mass per particle using the concept of microplastic size distributions by Kooi and Koelmans²⁸ and Kooi et al.²⁹. For lakes we were not able to compare our model with measurement data due to missing process understanding for transforming the inflowing microplastic mass to lake concentrations and high measurement uncertainties. Faure et al.²⁷ found, for instance, 78-5,000 microplastic particles per square metre at the surface of Lake Geneva.

For the River Rhine at the border with Germany we estimated the microplastic outflow to be one magnitude lower or one magnitude higher than the measurements depending on our mass calculations: $0.13~g~s^{-1}$ compared with about $1.2~g~s^{-1}$ for our assumption of spheres or $0.02~g~s^{-1}$ for our assumption of fragments and based on the results of Mani et al. 4 . For the River Rhône at the border between France and Switzerland we underestimated outflowing microplastic masses. Here Faure et al. 27 measured $0.12~g~s^{-1}$ while our model estimated $0.009~g~s^{-1}$. For the outflow of Lake Geneva (about 20 km upstream of the border but before the influence of the City of Geneva), we predicted concentrations about one magnitude lower than at the border (Fig. 3f). A similar relationship of microplastic masses at these two locations was measured by Faure et al. 27 .

As another comparison between modelled and measured data we used the reported distributions of different polymers types. Here we are able to capture the dominance of polyethylene (HDPE and LDPE) and PP (Fig. 4) that was measured by Faure et al.²⁷ and Mani et al.⁴. However, comparing measurements for other polymers, for example, PS, remained challenging. Faure et al.²⁷ identified 12% of the analysed particles as PS of which most particles were EPS (unspecified percentages). However, for EPS, a high percentage of counted particles relates only to very low masses due to the very low density of EPS. Similarly, Mani et al.⁴ found PS to be the most abundant polymer by number without stating whether EPS was assigned to PS.

Our model considers only microplastic releases directly into water bodies as predicted by Kawecki and Nowack²⁶. Hence, lower modelled values should be expected since fragmentation of macroplastics to microplastics, atmospheric deposition, as well as transport from land to surface waters are not considered yet. Furthermore, we considered only releases within Switzerland that correspond to 77% and 67% of the total catchment area for the Rhine and Rhône catchment, respectively. Our modelled microplastic masses are expected to increase when fragmentation of macroplastics and emissions into soils followed by transfer from soil to water are implemented. On the

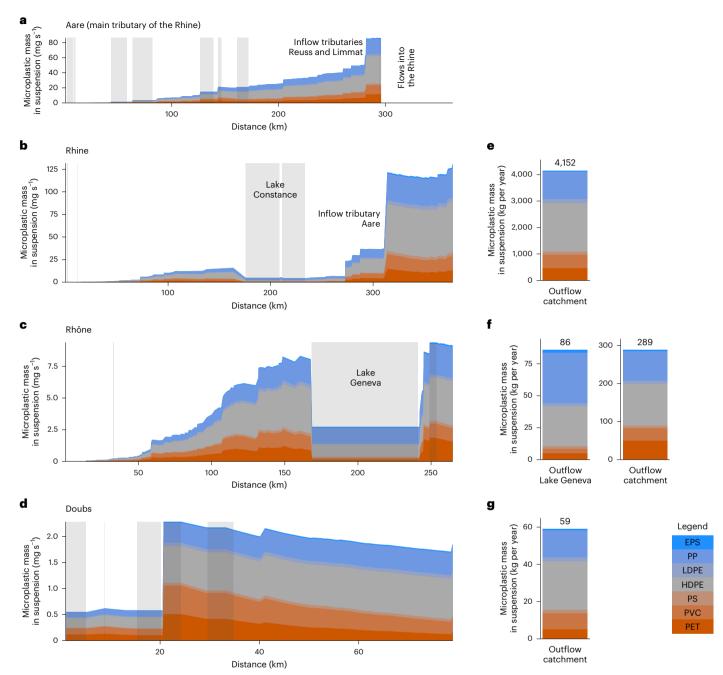


Fig. 3 | Masses of microplastic in selected Swiss rivers along the river lengths as well as total outflow of the catchment. a–d, Transport of microplastic masses in suspension along the main streams of the rivers Rhine (b), Aare (the main tributary of the Rhine) (a), Rhône (c) and Doubs (d). Blue to red shades indicate the different polymers as stacked values (see legend at bottom right), while grey vertical shades are symbolizing lakes. The microplastic masses can be understood as: 'how much microplastic is passing a river cross section per second', which

can be directly related to the actual microplastic concentration in water when considering the discharge. 'Distance' shows the distance from the source of the river according to the GIS vector file. $\mathbf{e}-\mathbf{g}$, The stacked bar plots present yearly masses at the outflow (the Swiss border) of the corresponding catchments for the Rhine (\mathbf{e}), Rhône (\mathbf{f}) and Doubs (\mathbf{g}). Additionally, for the Rhône the masses at the outflow of the Lake Geneva are shown, which represents the masses before the city Geneva. Please note that total masses vary across orders of magnitude.

other hand, fragmentation to nanoplastics may decrease the masses of microplastics. A full microplastic model additionally needs a link to a macroplastic release and fate model as well as a model describing flows from soils to water. The general plastic release model by Kawecki and Nowack²⁶ as well as the the spatially resolved model of Kawecki and Nowack²⁵ include both macroplastics as well as releases into soils. The specific release information for a coupled model is therefore available, and as soon as fate models for macroplastics and soils are available, a full microplastic model can be obtained. This full model could also

result in an even better prediction of the polymer distribution in the environment. We speculate, for example, that PS is currently underrepresented in our mass flow modelling in water because emissions of PS/EPS into soils are much higher than direct input into waters because of its abundant use in construction²⁶. Including transport from soil into water may therefore explain the missing mass in our model. Finally, we argue that atmospheric deposition can be ignored in the overall picture due to small quantities and because most surface area in Switzerland is land (about 95% for Switzerland).

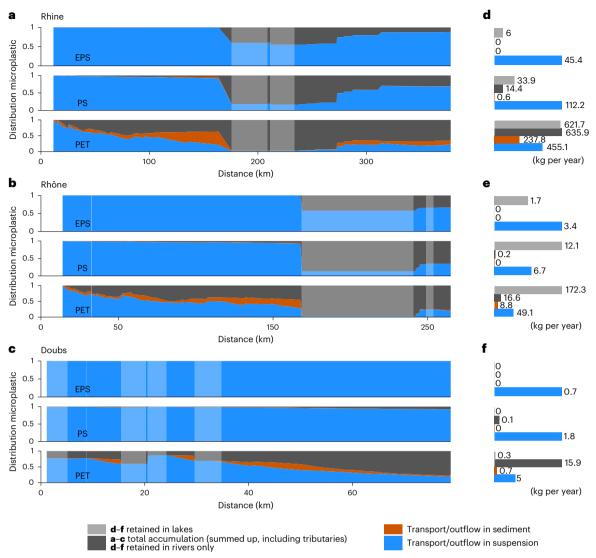


Fig. 4 | Comparing the distribution of microplastics among retained masses (in lakes or rivers) and transported masses (in suspension or sediments) for the rivers Rhine, Rhône and Doubs. \mathbf{a} - \mathbf{c} , Distribution between microplastics in suspension, in sediments and accumulated (long-term buried in sediments) along the main river for the three catchments of the Rhine (\mathbf{a}), Rhône (\mathbf{b}) and Doubs (\mathbf{c}) and for three selected polymers (EPS, PS and PET) that cover the range from very low density (EPS) to similar density to water (PS) until the most dense considered polymer (PET). Figures for all other polymers are shown in Supplementary Figs. 8 and 9. Values represent for any point the entire

upstream distribution including all tributaries. The distance corresponds to the accumulated lengths of the mapped river lengths (polylines), including distance through lakes, from the spring to the border of Switzerland. Shaded areas correspond to distance through lakes. Gaps at the beginning of the river correspond to non-existing plastic masses. \mathbf{d} - \mathbf{f} , Outflow masses and location of retention in kg per year for the entire catchment (Rhine (\mathbf{d}), Rhône (\mathbf{e}) and Doubs (\mathbf{f}) and the same polymers. Here we consider only the catchment area located in Switzerland. Please note that numbers vary across a few orders of magnitude among different polymers and catchments.

Regarding our modelling validation, we are aware that not including detailed hydrological or fluid mechanical parameters might seem to be a strong simplification of the model that might increase uncertainties on the level of the hydrological behaviour. However, as we have shown with our model, on a country scale other processes become more important such as catchment properties or location of pollution sites. Hence, any (small) changes in behaviour caused by polymer, river or sediment properties would modify our results only to a small extent. Furthermore, including parameters such as shear stress in models for microplastic behaviour in natural rivers remains challenging for mixed grain sizes and heterogeneous flow conditions as shown by many studies (for example, refs. 30–33). Although critical shear stresses for microplastics are available in the literature for some polymers³⁰, calculating the corresponding forces for each river segment on a catchment scale remains even more challenging because of the required input data

(for example, flow velocity, water depth, slope and so on). Hence, we argue that flow velocity is an appropriate first simplification to cover these processes. Similarly, Mani et al. *showed that flow velocity is proxy for microplastic sedimentation behaviour along Rhine and within single cross-sections. Furthermore, only this simplification makes it possible to model whole watersheds, which increases the usability of the model for future applications.

On the other hand, experimental findings and existing models suggest that sedimentation behaviour also depends on size and polymer density (for example, refs. 19,30). However, existing data are based on laboratory work and theoretical assessments, while measurements under natural conditions are largely missing. Thus, in existing modelling approaches or experiments, polymers less dense than water (EPS, PP, LDPE and HDPE) are either not considered or are assumed to remain floating on the water surface without any settling ^{19,20,30,34}.

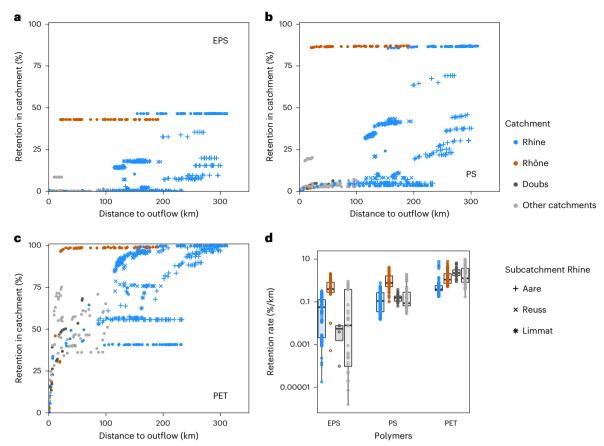


Fig. 5 | **Influence of catchments on microplastic retention through sedimentation and accumulation for the polymers EPS, PS and PET.** Shown is how much of the input emission microplastics is not leaving the catchment in suspension because they are either accumulated or transported in sediments (see also Fig. 4). We selected 502 river segments randomly and set the input emission to 100%, while all other river segments were initially set to 0 input emissions (number of points per catchment: Rhine, 349; Rhône, 58; Doubs, 21; other, 74). **a-c**, Shown are the retention and losses of EPS (**a**), PS (**b**) and PET (**c**)

into the sediment at the outlet of the catchment in relationship to the distance to the outlet according to the river polyline maps including distances through lakes. Colours and shapes show the catchment the sampling point is located in. Blue round points (Rhine catchment) are located in the Rhine catchment but not in any subcatchment listed. ${\bf d}$, Fraction retained per kilometre for a selection of Swiss catchments. Distances are equal to distances in ${\bf a}-{\bf c}$. The box plots represent median values (line) with lower and upper quartile (hinges) and 95% confidence interval (whiskers).

In contrast, these assumptions are clearly not supported by river sediment measurements that also found these polymers with density less dense than water (for example, refs. 35,36). Thus, further research would be beneficial to better derive sedimentation factors but also resuspension factors of microplastics. On the other hand, small change of the sedimentation or resuspension would only shift total plastic masses in different model compartments, but the overall picture would remain similar.

Also, our model is limited in the temporal resolution, which can affect the dynamics of plastic masses. Although we calculated in 1 s timesteps, we only provide yearly values because also the emissions obtained from Kawecki and Nowack²⁵ are given in yearly masses. Consequently, the effects of important flooding events³⁷ with temporally limited higher plastic mass transport are not represented in the current model, but could be implemented with a change of factors. Here also the modelling of lakes would need an adjustment according to residence time since reduction of plastic masses in lakes are modelled in one timestep (Supplementary Information Section 9).

Finally, the model focuses on masses while researchers interested in toxicity might be more interested in particle numbers, especially of the smaller size classes of microplastics, instead of overall masses. For this purpose, the model could be adjusted to specific size classes by calculating multiple size classes simultaneously similar to the nanoDUFLOW model¹⁹. However, this would first require the release data to be size specific in the microplastic range. The release data

that our model is based on only distinguish between micro- and macroplastic masses but do not further provide a size distribution of microplastics.

A main outcome of our modelling is that no simple linear function for microplastic retention as a function of river length to the outlet exists. Instead, we found that different catchments cover a wide range of retention for microplastics. Fig. 5 shows an analysis of the retained fraction towards the Swiss border of 502 randomly selected river segments. The Rhône catchment follows best a logarithmic function due to the influence of Lake Geneva (Fig. 5a–c); in the Rhine catchment the retention becomes more complex. Here each tributary contributes very differently to the overall microplastic transport in the river, which is also visible for the different subcatchment of the Rhine, that is, Aare, Reuss and Limmat, shown in Fig. 5a–c. Even within the subcatchment Aare (crosses) differences can clearly be observed Fig. 5a–c. Our findings are supported by other studies that show that even small to medium lakes might play an important role in microplastic retention (for example, refs. 13,38,39).

Consequently, we argue that using input data and performing modelling on high spatial resolution is of high importance when estimating transport of microplastics, including the additives contained in the plastics⁴⁰, or predict concentrations on the basis of release estimates in large areas such as countries. Finally, better spatial resolution will improve the accuracy of microplastic exported out of the system

(Fig. 5). Better and more accurate input data, on the other hand, will help to identify hotspots and composition of polymer masses in the system 41,42. Especially, modelling approaches using catchments as single units might overlook important spatial differences (for example, ref. 24). Hence, we can clearly show as an example that reducing input pollution downstream of the Lake Geneva (that is, through the City of Geneva) would be by far more effective than reducing pollution more upstream (Fig. 3).

Finally, we would like to address some suggestions on how to improve future measurement campaigns to enable a better coupling with modelling. Using some principal hydrological considerations could increase dramatically the value of sampling. In particular, we suggest to time two measurements in the same river according to the river flow velocity to measure the 'same' parcel of water, which would allow to much better estimate retention within the river. Additionally, we would like to see measurements of reference segments in rivers, that is, inflow and outflow measurements. As reference segments we imagine a river stretch in which changes of microplastic pollution can be directly attributed to certain properties; for example, a natural segment without input emissions would represent an average sedimentation behaviour. These types of reference segments could then be used for further modelling.

Conclusions

Our work presents a first country-wide modelling approach with focus on microplastic masses using high spatial resolution and including polymers less dense than water. As a main driver of plastic retention we identified in our model lakes that, depending on the polymer, retain up to ten times more mass of microplastics than rivers. Although lakes may be very important for microplastic retention, the effect of retention by dams is not fully confirmed yet⁴³ and detailed distribution modelling of plastic within lakes remains challenging. We highly encourage future studies to investigate the role of lakes and dams in microplastic retention including different lake sizes, depths, dam constructions and hydrological conditions, for example, flooding events and lake stratification.

By using a simplified approach, the model can be adapted for many different regions with options to focus on different plastic sizes or to increase the detail of transportation processes. By applying the model to Switzerland we were able to identify hotspots of microplastic pollution and critical emission sites. Such a model can therefore provide policy-makers a tool to better validate, rate and understand the effects of different policies. Thus, the model could be used the explore different theoretical scenarios, such as changes in plastic pollution into the freshwater environment on point source perspective or country scale.

Finally, we would like to emphasize that more measurement campaigns are needed that contribute to a process-based understanding of microplastic transport and distribution in catchments or countries. We should shift our resources from just presenting another proof of the presence of microplastics towards measurements that provide a process understanding. Generally, considering hydrological parameters during measurement campaigns similar to nutrient measurements in surface waters and the choice of sampling locations useful for further modelling would be beneficial.

Methods

The following section will guide through the different states of the model and the geographical and hydrological data needed. The case-study-related information is listed at the end of this section. We also provide a simple example river network (Supplementary Information Section 11) to explore the model functionality.

Geographical and hydrological data

The basic information needed is a digital map of the river network and the lakes in the study area. Typically, this information is available as vector dataset in the format of a shapefile or GeoPackage format

file. The digital river and lake maps should fulfil the criteria listed in Supplementary Information Section 2 to guarantee a logical river connectivity that will ensure a correct microplastic transport through the river network. Information about river segments and lakes is presented in the river network file. The exact requirements and names are listed in Supplementary Information Section 2.

Microplastic release

With a river and lake network separated into individual river and lake segments, the input microplastic emissions should be known for each river and lake segment as masses per second. Such a release model with a high spatial resolution is available by Kawecki and Nowack²⁵. This model is based on a material flow analysis of plastics through society⁴⁴ that was then coupled with a release model to estimate emission flows into the environment, including among others point sources of waste water treatment outflows as well as diffuse sources²⁶. To allocate the emissions in a next step to water bodies, Kawecki and Nowack²⁵ used geographical proxies such as population density, land use, construction sites or traffic density to geographically distribute the total masses. The final data were yearly emissions of microplastics into each river segment and lake differentiated among the seven polymers.

Modelled states of microplastics

In the model, microplastic masses were considered in three different states for each polymer: in suspension, in sediments and in deep sediments (accumulation) (Fig. 6). While accumulation in deep sediments is a final sink, microplastics in suspension and in sediments are allowed to migrate downstream with the river current. By default, all input emissions were first assigned to suspension before allowed to sediment (Fig. 6). For plastics in the sediment three pathways were possible: they can be buried in deep sediments and accumulate, they can resuspend into the suspended state or they can be transported with the sediment to the following downstream sediment container of the river or lake.

The model used microplastic masses instead of particle numbers because microplastic release models based on material flow analysis provide only mass flows. We simplified our model to one particle size class with the focus on masses and mass flows instead of particle numbers. While measurements of the full size spectrum of microplastic particles show that small sizes dominate the particle number distribution^{28,29}, the dominance is reversed for masses using the same number distribution. Hence, in the particle number distributions first presented by Kooi and Koelmans²⁸ and further improved by Kooi et al. ²⁹ the bigger microplastic particles (in the millimetre range) are in terms of masses the most important size class although the likelihood of occurrence is the lowest. For spheres or fragments, a representative microplastic particle mass is about 1 mm in size based on the average size (longest site) and weighted by occurrence probability distribution^{28,29}.

Transport in suspension (advection)

Transport in suspension was mainly influenced by river flow velocity since advection (transport with fluid) is much more important than transport through diffusion. Furthermore, the flow velocity influences the interaction of microplastics with the river sediment, such as the probability of sedimentation or erosion³⁰. We calculated microplastic transport and input emissions as mass per second assuming steady state for a one second timestep with input emissions being equal to all output emissions. By doing so, transport velocity in sediments or in suspension becomes negligible. In other words, we calculated the mass of plastic emissions that will be transported to the next river section downstream per second. Please, refer to Supplementary Information for further explanation and an example. For lakes we assumed a steadystate system per second, which means that the inflowing mass from upstream plus the input emissions directly into the lake per second is equal to the outflow mass per second. To derive the microplastic mass per river segment, the mass per second was multiplied with the

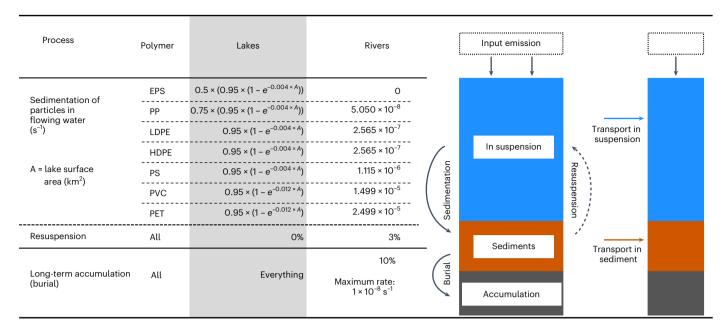


Fig. 6 | Conceptional model setup per each river segment including processes within one segment and transport processes to the next segment. Additionally, parameters to derive sedimentation and long-term sedimentation (burial) are listed. The parameterization is different for rivers and lakes and depends on the microplastic polymer.

time the water needs to flow through the corresponding section (for possible ways to derive average flow velocities, see Supplementary Information Section 5).

Fate processes

Considering only advection can be interpreted as maximum possible microplastic flow, which from here on will be referred to as scenario 0 (S_0). To model retention processes such as sedimentation, including burial into deep sediments, we used factors that were multiplied with the masses in each segment and each state.

Following the pathways in Fig. 6 we derived sedimentation factors ($f_{\rm sed}$) as a first step of microplastic reduction from masses in suspension. Sedimented microplastic masses then can be transported to deep sediments and accumulated according to the corresponding factor $f_{\rm acc}$. We applied both factors, $f_{\rm sed}$ and $f_{\rm acc}$, for each river segment and lake and each polymer individually on the basis of available literature data. Removal, for example, through cleaning or pick-ups, can be ignored for microplastics, and no other removal process was included in the current model.

Sedimentation factors (f_{sed}) for rivers were estimated on the basis of the modelling results by Besseling et al. 19, Siegfried et al. 24 and Domercq et al.²⁰ and measurement results (for example, refs. 45,46). While existing modelling results suggest that spherical particles bigger than around 0.2 mm sediment immediately after entering the waters (for example, refs. 19,20), measurement studies show that the dominant size class in the environment, including rivers, are 0.5-5 mm in size^{16,35,46,47}. Estimating retention factors remained challenging due to the lack of existing data and contradictory data of measurements and published modelling results. We estimated $f_{\rm sed}$ on the basis of the modelling results of Besseling et al. 19 using the 5 µm size class. We used this size class because it showed medium sedimentation rates with differences among the different polymers. Using the results of larger size classes, that is, the representative 2 mm, would have resulted in full microplastic retention through sedimentation within a very short distance according to Besseling et al. 19. However, this is contradictory with the monitoring results presented above. Besseling et al. 19 found for the 5 µm size class that the two most dense polymers (PVC and PET) would be almost fully retained over the 40-km-long modelled river, while retention for less dense polymers would be proportionally lower based on polymer density (for further information, see Supplementary Information Sections 7 and 8). For polymers less dense than water that were not considered in existing models, we used literature data mentioned above to estimate $f_{\rm sed}$ in relationship with the heavier polymers used by Besseling et al. ¹⁹.

Finally, $f_{\rm sed}$ was derived for an entire river segment on the basis of the travel time (L in s) through the river segment and the sedimentation rate extracted from Besseling et al.¹⁹ (Fig. 6 and Supplementary Information Section 7). Furthermore, we used a negative compound interest approach shown in equation (1) to assure that microplastics lost in the beginning of the segment cannot be lost afterwards. For rivers, $f_{\rm sed}$ is calculated as follows:

$$f_{\text{sed, river}} = 1 - \left(1 - k_{\text{s}}\right)^{L} \tag{1}$$

where k_s is the sedimentation factor per second derived from the results of Besseling et al. ¹⁹ and given in Fig. 6. L is the average travel time in seconds through a river segment calculated by equation (2).

$$L = \frac{l}{\nu} \tag{2}$$

Here, l is the river segment length in m and v is the average flow velocity in the river segment (in m s $^{-1}$). Consequently, higher L corresponds with longer residence time in a river segment, which causes higher plastic retention in the river segment for equation (1).

For $f_{\rm acc}$ we used 10% as a default value, meaning that 10% of the microplastics in the sediment will be buried in rivers across all polymers. For the more dense polymers PVC and PET, this assumption is in alignment with findings by Drummond et al. ⁴⁸. However, the maximum $f_{\rm acc}$ was set to 1×10^{-8} s⁻¹, which is between values used by Domercq et al. ²⁰ and Besseling et al. ¹⁹ who based their long-term sedimentation rates on Praetorius et al. ⁴⁹ and Koelmans et al. ⁵⁰.

To derive fate processes in lakes, we aimed for a single f_{sed} per lake. However, we found very few studies describing a mass balance

of microplastic fluxes through lakes to derive $f_{\rm sed}$ or $f_{\rm acc}$ for lakes. On the basis of available data points (Supplementary Table 6) we fitted a logarithmic curve relating plastic sedimentation yield (k_i) with lake surface area (A) in km². We used a logarithmic function to account that small lakes are found to retain microplastics proportionally higher than larger lakes in relationship to their surface area (Supplementary Fig. 3). For lakes, $f_{\rm sed}$ was calculated as follows:

$$f_{\text{sed, lake}} = C(1 - e^{-k_1 A})$$
 (3)

The asymptotic maximum plastic loss C was set to 95% (0.95), and k_i was varied with polymer type. Across all polymers we aimed for an approximately 90% loss rate for Lake Geneva ($A = 500 \text{ km}^2$) with $k_i = 0.005$, which is a retention rate based on modelling results by Boucher et al. ⁵¹.

To account for differences between polymers, we varied k_1 following results shown by Yang et al. ⁵² who summarized multiple studies of sediment analysis. Thus, polymers notably less dense than water (that is, PP) were found to be less abundant in sediments compared with particles in suspension, while for polymers more dense than water the ratio was found to be opposite ⁵². For the analysed polymer PE (here differentiated into LDPE and HDPE) no trends were found ⁵². Hence, for the polymers EPS, PP, LDPE, HDPE and PS we used k_1 = 0.004 (Fig. 6). For EPS and PP, equation (2) was multiplied by 0.5 and 0.75 correspondingly to decrease $f_{\rm sed}$ including maximum possible sedimentation rate C. Simultaneously, for PVC and PET we increased the sedimentation probability, especially for smaller lakes, by using a higher k_1 (0.012). All equations are shown in Fig. 6. Further information about factors and available studies is presented in the Supplementary Information. We assume that all plastics in the sediment will be accumulated ($f_{\rm acc}$ = 1).

Resuspension

Microplastics resuspended ($f_{\rm resus}$) from the sediment compartment are added to the microplastics in suspension. We allowed 3% (Fig. 6) of plastics in the sediment to resuspend since Praetorius et al.⁴⁹, and consequently Domercq et al.²⁰, used a resuspension rate of around one-third of their burial rate.

It should be stated that accumulation was considered first and therefore resuspension was impossible in few cases due to too little microplastics in the sediments. Resuspension for lakes was assumed to be 0 because $f_{\rm sed}$ describes the entire microplastic retention per lake including potential resuspension influences.

Transport in sediments

On the basis of our steady-state assumption, velocity of microplastic transport in sediments is important only for calculating microplastic masses temporally stored in one segment. The outflow of microplastics through sediments to the next segment, on the other hand, is equal to the input minus all factors reducing microplastic loads in the sediment state ($f_{\rm acc}$ and $f_{\rm resus}$). In our model, sediment transport velocity was equal to main river flow velocity. To calculate masses of microplastic in sediments per segment, the average travel time through a segment L can be adjusted to a slower, more realistic, sediment transport velocity.

Case study Switzerland

The case study presented is based on the Swiss river (Feature Class TLM_FLIESSGEWAESSER) and lake (Feature Class TLM_STEHENDES_GEWAESSER) network in scale 1:25,000 (swisstopo, swissTLM^{3D}, version 1.8, March 2020). Switzerland covers about 41,000 km², including multiple lakes up to about 500 km² surface area and multiple thousands of different rivers that all flow out of the country due to topography.

For input microplastic emission data into the environment we used modelled data by Kawecki and Nowack 25 . We updated the data to a newer river and lake map to provide microplastic emission data as masses for each of the over 600,000 river segments in Switzerland for the seven polymers EPS, PP, LDPE, HDPE, PS, PVC and PET. The total

emission of microplastics (the seven polymers mentioned above) into all water bodies was 15 tons per year for the year 2014 (ref. 26).

For the results we focused on the two dominant Swiss catchments of the Rhine (outlet: German border next to Basel) and the Rhône (outlet: French border next to Geneva) as well as the Doubs catchment for comparison. The three catchments cover an area of 27,981 km² (always Swiss area only), 9,210 km² and 372 km² for the Rhine, Rhône and Doubs, while 3.4%, 5.3% and 0.3% of the area are covered by lakes. Overall, our model covers 77%, 67% and 29% of the total Rhine, Rhône and Doubs catchment area of the outlet, respectively, since input emissions are available only for Switzerland. The Doubs catchment was selected because it does not include major lakes and is not a subcatchment of one of the two big catchments. Microplastic measurements are available for the Rhine and Rhône in the literature 4.27. Please refer to Supplementary Fig. 4 for further information and maps of the studied catchments and the Swiss river network.

Data availability

We provide an example river network to explore the model. Most raw data are publicly available at https://map.geo.admin.ch/ (swisstopo). The full dataset of processed data will be available upon request.

Code availability

The code is freely available at https://doi.org/10.5281/zenodo.7867010. So far the model is written in $\mathbb R$ and uses QGIS functions through the $\mathbb R$ package qgisprocess.

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D.M.: methodology, investigation, visualization, and writing—original draft, review and editing. B.N.: methodology, supervision, funding acquisition, and writing—review and editing.

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Additional information

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