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Modellierung der Umweltkonzentrationen von Makro- und Mikroplastik in der Schweiz

Schlussbericht des BAFU-Projektes

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Zusammenfassung

Das Ziel dieses Projektes war es, basierend auf einem existierenden Materialflussmodell für die Freisetzung von Plastik in die Umwelt, welches im BAFU-Projekt «Modellierung der Massenflüsse von Mikroplastik in die Umwelt in der Schweiz» entwickelt wurde, die Umweltkonzentrationen von Makro- und Mikroplastik in der Umwelt der Schweiz zu modellieren. Als Umweltkompartimente werden sowohl Gewässer als auch Böden berücksichtigt. Dafür wurde ein Fate-Modell entwickelt, welches es erlaubt, die Massenflüsse von Makro- und Mikroplastik in sämtlichen Flüssen und der Seen der Schweiz zu modellieren. Das vorhandene Freisetzungsmodell wurde dann mit dem Fate-Modell gekoppelt und für sieben verschiedene Polymere berechnet (LDPE, HDPE, PP, PET, PVC, PS und EPS). Als Resultat wurden Karten mit Mikroplastikkonzentrationen in sämtlichen Schweizer Flüssen und Seen erhalten. Es konnte gezeigt werden, dass die Polymerart, die Geographie des Einzugsgebietes und vor allem die Lage der Städte im Einzugsgebiet die Konzentrationen stark beeinflussen. Mit diesem kombinierten Modell können erstmals Konzentrationen von Mikroplastik in der Umwelt basierend auf der Verwendung von Plastik in der Gesellschaft vorhergesagt werden. Eine Validierung des Modells mit gemessenen Werten ist noch nicht möglich, da sowohl der Transport vom Boden ins Wasser als auch die Fragmentierung von Makroplastik zu Mikroplastik noch nicht im Modell enthalten ist. Unter diesen Einschränkungen sind die modellierten Konzentrationen bei Basel und Genf etwas tiefer als die gemessenen Werte, was jedoch durch diese noch nicht berücksichtigten Prozesse erklärt werden kann. Auch sind Messungen und Modellierungen mit gewissen Unsicherheiten behaftet, die ebenfalls berücksichtigt werden müssen.

Das Modell wurde anschliessend an die Modellierung von Makroplastik angepasst und erste Resultate basierend auf Szenarien wurden erhalten. Hier zeigt sich, dass der Rückhalt von Makroplastik im Gewässer deutlich höher als derjenige von Mikroplastik sein muss, um die Modellwerte an gemessene Mengen angleichen zu können.

Als letzter Schritt wird der Transport vom Boden ins Wasser ins Modell integriert. Da hierzu noch publizierte Daten zum Verhalten fehlen, wird nur mit Szenarien gearbeitet. Diese Modellierung, welche die oben genannten Modelle verbinden wird, erlaubt dann eine Gesamtbetrachtung der Massenflüsse von Makro- und Mikroplastik von der Freisetzung bis zu Umweltkonzentrationen. Dies wird es in einem weiteren Schritt erlauben, den Effekt von Massnahmen an der Quelle auf die Umweltkonzentrationen zu quantifizieren.

Ausgangslage

Im Projekt «Modellierung der Massenflüsse von Mikroplastik in die Umwelt in der Schweiz», welches vom BAFU finanziert wurde und welches von Mai 2016 bis Oktober 2019 lief, wurden die Grundlagen gelegt, um die Mengen und die Freisetzung von Plastik in die Umwelt abschätzen zu können. In diesem Projekt wurde für sieben wichtige Kunststoffe (LDPE, HDPE, PP, PET, PVC, PS und EPS) die Massenflüsse durch die Anthroposphäre quantifiziert. Dabei wurden in einem ersten Schritt Import, Produktion und Herstellung von Produkten berücksichtigt, in einem weiteren Schritt dann die Verteilung der Massenflüsse auf verschiedene Produktkategorien (Gebrauchsphase). Anschliessend wurden die Abfallsammlung, das Recycling und die Abfallbehandlung quantifiziert. Diese Berechnungen wurden in diesem Detaillierungsgrad weltweit das erste Mal durchgeführt und bilden die Grundlage, um die Flüsse von Plastik in die Umwelt abschätzen zu können. In einer zweiten Projekt-Phase wurde dann basierend auf diesem Flussmodell über den ganzen Lebenszyklus die möglichen Freisetzungen der gleichen sieben Kunststoffe in die Umwelt quantifiziert. Es wurden dabei sowohl die Makro- als auch die Mikroplastikflüsse bestimmt. In der dritten und letzten Projektphase wurden die Flüsse regionalisiert.

Offene Fragen

Im ersten BAFU-Projekt konnten die bis anhin detailliertesten Abschätzungen zur Freisetzung von Plastik in die Umwelt gemacht werden. Viele der verwendeten Daten zur Parametrisierung des Modells sind aber äusserst unsicher und viele beruhen auf Abschätzungen. Da ein Model basierend auf Wahrscheinlichkeiten verwendet wurde, konnten diese Unsicherheiten zum Teil berücksichtigt werden. Auch wenn diese Flüsse am Ende des Projektes regionalisiert vorliegen, so lassen sie doch keinen Rückschluss auf Umweltkonzentrationen zu, welche das eigentliche Ziel von Umweltmodellen sind. Damit die Umweltflüsse in Umweltkonzentrationen (PEC-Werte, Predicted Environmental Concentrations) umgewandelt werden können, muss auch das Umweltverhalten berücksichtigt werden. Wichtige Prozesse sind dabei der Transport von Makro- und Mikroplastik in der Umwelt. Während in der ersten BAFU-Studie die Materialflussmodellierung verwendet wurde, braucht es zur Bestimmung der PEC-Werte eine Kopplung von diesen Modellen mit solchen zur Berechnung des Umweltverhaltens

(Environmental fate modeling). Mit einem regionalen Umweltverhaltensmodell lassen sich dann regionale Umweltkonzentrationen bestimmen. Diese Werte können dann auch mit gemessenen Werten verglichen und das Modell eventuell sogar validiert werden.

Ziel des Projektes

Das Ziel dieses Projektes war es, basierend auf dem existierenden Materialflussmodell für die Freisetzung von Plastik in die Umwelt die Umweltkonzentrationen für Makro- und Mikroplastik in der Umwelt der Schweiz zu modellieren. Als Umweltkompartimente werden sowohl Gewässer als auch Böden berücksichtigt. Dabei wird das Stoffflussmodell mit einem zu entwickelnden Fate-Modell gekoppelt. Die Modellierung wird für die sieben im ersten Projekt betrachteten Polymere durchgeführt (LDPE, HDPE, PP, PET, PVC, PS und EPS).

Das Fate-Modell für Makro- und Mikroplastik wird regionalisiert sein, weshalb als Resultat des Projektes Karten der Schweiz mit den modellierten Umweltkonzentrationen erhalten werden. Die modellierten Konzentrationen und Polymerverteilungen können dann mit gemessenen Werten verglichen werden (Validierung des Modells).

Vorgehen

Die folgenden Schritte wurden bearbeitet, um das Ziel des Projektes zu erreichen.

1) Entwicklung eines Fate-Modells von Mikroplastik im Wasser

2) Entwicklung eines Fate-Modells von Makroplastik im Wasser

3) Kombination des Materialflussmodels mit den Fate Modellen und Berechnung von Umweltkonzentrationen von 7 Polymeren in Wasser von Makro- und Mikroplastik

4) Entwicklung eines Fate-Modells von Makro- und Mikroplastik auf dem Boden und vom Boden ins Wasser

5) Vergleich der modellierten PEC-Werte mit Messungen der verschiedenen Polymere in der Umwelt (Validierung)

Der Hauptteil des Projektes wird darin bestehen, Fate-Modelle für Mikro- und Makroplastik in Wasser und Boden zu formulieren und wenn möglich auch die Fragmentierung von Makro- in

Mikroplastik in den Modellen einzuschliessen. Das Modell soll an die Situation in der Schweiz angepasst werden und dazu dienen, geographisch aufgelöst das Verhalten von Mikroplastik in sämtlichen Schweizer Fliessgewässern zu modellieren. Zusätzlich muss ein Fate-Modell für Mikroplastik in Seen entwickelt werden, welches auf den gleichen Prozessen wie das Fluss-Modell basieren wird.

Bis anhin existieren keine Fate-Modelle für Plastik in Böden, welche aber wegen der Wichtigkeit der Freisetzung auf Böden ein wichtiger Teil der Fate-Modellierung sein werden. Hier ist vor allem der Transport von Mikroplastik im Boden und die Abschwemmung in Fliessgewässer wichtig. Ein wichtiges Modul des Modells wird die Fragmentierung von Makro- zu Mikroplastik sein. Auf diesem Thema findet zurzeit viel Forschung statt, so dass im Laufe des Projektes experimentelle Daten zur Verfügung stehen sollten, welche als Grundlage für das Modell dienen können.

Die Module werden in einer Art formuliert, dass sie geografisch aufgelöst modelliert werden können. Basierend auf der räumlichen Verteilung der Plastikquellen aus dem ersten BAFU-Projekt wird dann ein Gesamtmodell entwickelt, welches räumlich aufgelöst den Transport von Mikro- und Makroplastik in Boden und Wasser beschreibt. Für jeden Flussabschnitt und See werden so Konzentrationen von Mikro- und Makroplastik erhalten. Die Konzentrationen in Fluss- und Seesedimenten werden ebenfalls als Resultat des Modelles erhalten. Die Konzentrationen werden für jedes Polymer separat erhalten, können aber auch für alle zusammen ausgewertet werden (Gesamtbelastung).

In einem letzten Schritt des Projektes werden die vorhergesagten Konzentrationen mit gemessenen Werten verglichen. Eine strikte Validierung wird wahrscheinlich nicht möglich sein, da die Probenahme und Analytik von Plastik noch nicht so weit entwickelt sind, dass reproduzierbar und quantitativ sämtlicher Plastik und Mikroplastik an einem Ort gemessen werden können. Der Vergleich von Modell und Messungen kann aber dazu dienen, eine bessere Einschätzung der Exposition zu bekommen und kann entweder wertvollen Input für die analytische Seite geben.

Der grosse Vorteil des finalen Modells über einzelne Messungen in der Umwelt ist, dass eine Gesamtsicht der Plastikbelastung in der ganzen Schweiz erhalten wird. Diese ganzheitliche Betrachtung kann dann die Grundlage für genaue Risikoabschätzungen bilden. Mit den räumlich aufgelösten Konzentrationen können dann in einem nächsten Schritt Risikokarten für Plastik in der Umwelt erhalten werden.

Die Resultate des Modelles können auch dazu dienen, fundierte Aussagen zu Massnahmen gegen die Plastikbelastung in verschiedenen Gegenden zu machen. Mittels Szenarien können die Effekte von verschiedenen Massnahmen modelliert werden um abschätzen zu können, wie z.B. Verbote einzelner Produkte oder Änderungen im Verhalten die Gesamtbelastung der Umwelt beeinflussen.

Fate Modell für Mikroplastik im Wasser

Basierend auf existierenden Modellen in der Literatur war davon auszugehen, dass es am meisten Informationen über den Transport von Mikroplastik in Gewässern geben würde. Deshalb wurde beschlossen mit diesem Teil zu starten. Ziel war es von vorne herein, ein möglichst erweiterbares Modell zu entwickeln, sodass weitere Prozesse wie z. B. Makroplastiktransport, Transport über Böden oder Fragmentierung in das Model später eingearbeitet werden können. Des Weiteren war der Anspruch, dass das Model einen weiten Anwendungsbereich finden kann.

Für Mikroplastik gibt es, Stand heute, vor allem Modelle, die einzelne Flüsse abdecken können. Ganze Netzwerke von Flüssen auf Bundesebene können jedoch noch nicht modelliert werden. Deshalb wurde ein Modell entwickelt, welches genau diesen Einsatzbereich hat: Mikroplastikfate auf der Skala der ganzen Schweiz inklusive aller Flüsse und Seen.

Beschreibung des Fate Modells

Da im Rahmen des Projektes keine Messungen oder Experimente gemacht wurden, basiert das Modell auf existierenden Modellen und Messungen, die in der Literatur vorhanden sind. Die Hauptkomponenten des Modells sind in Abbildung 1 festgehalten. Grundsätzlich unterscheiden wir in Anlehnung an andere existierende Modelle zwischen "in suspension", "in sediments" und "accumulation". Hierbei gilt, dass Mikroplastikemissionen zunächst der Suspension zugeteilt werden. Die Input Emissionen stammen aus dem Vorgängerprojekt des BAFU (Modelling the Flows of Plastics and Microplastics into the Environment) und sind in dem Paper von Kawecki und Nowack, 2020 (doi: 10.1016/j.scitotenv.2020.141137) veröffentlicht. Kawecki und Nowack (2020) modellieren die Mikroplastikemissionen von sieben verschiedenen Polymeren (EPS, PP, LDPE, HDPE, PS, PVC, PET) in die Umwelt basierend

auf einem Materialflussmodell. Die Emissionen sind wurden anhand von Proxies, wie z. B. Bevölkerungsdichte, Strassennetz etc., geographisch für die Schweiz modelliert.

Für unser hier vorgestelltes Modell, gehen wir davon aus, dass ein Teil des Mikroplastiks in der Suspension sedimentieren kann und somit temporär gespeichert werden. Aus dem temporären Speicher können Plastikmassen wieder resuspendiert werden oder es erfolgt eine Akkumulation, die in dem Modell als nicht umkehrbar gilt.

process	polymer	lakes	rivers
sedimentation of particles in flowing water in [s^{-1}] A = lake surface area in [km^2]	EPS	$0.5*(0.95*(1-e^{-0.004*A}))$	0
	PP	$0.75*(0.95*(1-e^{-0.004*A}))$	5.050×10^{-8}
	LDPE	$0.95*(1-e^{-0.004*A})$	2.565×10^{-7}
	HDPE	$0.95*(1-e^{-0.004*A})$	2.565×10^{-7}
	PS	$0.95*(1-e^{-0.004*A})$	1.115×10^{-6}
	PVC	$0.95*(1-e^{-0.012*A})$	1.499×10^{-5}
	PET	$0.95*(1-e^{-0.012*A})$	2.499×10^{-5}
resuspension	all	0%	3%
long term accumulation (burial)	all	everything	10% maximum rate: $1 \times 10^{-8}\ s^{-1}$

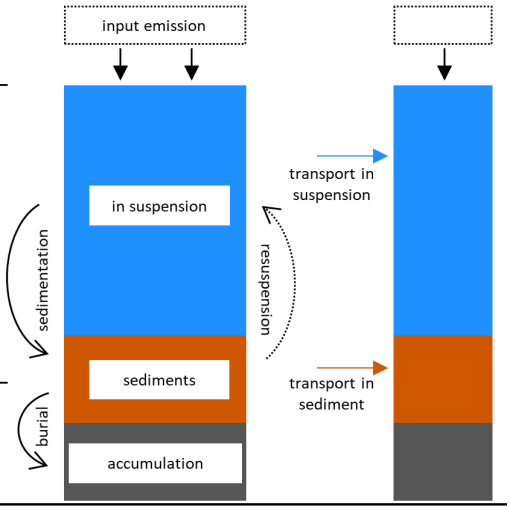

Abbildung 1: Konzeptionelle Darstellung der Modellparameter zum Modellieren des Verhaltens von Plastik in Flüssen und Seen. Zusätzlich sind die Faktoren für die Massenflüsse in Flüssen und Seen angegeben.

Um die Flüsse zwischen den verschiedenen Modellkompartimenten zu beschreiben, benutzen wir Faktoren. Die Faktoren wiederum können auf unbegrenzt komplexen Rechnungen basieren und ermöglichen damit eine möglichst grosse Brandweite an Anwendungen.

Für die Schweiz ist das Flussnetzwerk in ca. 600.000 Flusssegmente eingeteilt und wird durch ca. 17.000 Seen ergänzt, wobei jeder See als ein "Segment" berechnet wurde. Für Flüsse ist bei der Sedimentationsrate die mittlere Durchflusszeit in Sekunden des Flusssegments mit ausschlaggebend für die finalen Sedimentationsmassen, während bei Seen die Seeoberfläche als Proxy für die Sedimentationsmassen gilt (s. Abbildung 1). Somit wurde für jedes Segment individuelle Faktoren berechnet. Ausserdem werden die Faktoren zwischen den sieben oben genannten Polymeren unterschieden. Für Mikroplastik wurden die Faktoren primär aus dem nanoDUFLOW (Bessling et al. 2017, doi: 10.1016/j.envpol.2016.10.001) für Flüsse und aus verschiedenen Messungen für Seen abgeleitet.

Die wichtigste Aufgabe des Modelles ist es, dass die Plastikmassen logisch dem Flussverlauf flussabwärts folgen. Da das Modell Massen betrachtet, ist somit von einer Aufsummierung auszugehen, falls Plastik nicht durch Prozesse wie Akkumulation zurückgehalten wird. Weitere Details zum Modell finden sich im Anhang.

Zusammenfassung der Ergebnisse und Erkenntnisse über das Verhalten von Mikroplastik in Flüssen und Seen der Schweiz

Abbildung 2 zeigt eine Karte der modellierten Konzentrationen in den Schweizer Flüssen und Seen. Blau bezeichnet dabei die tiefsten Konzentrationen, rot die höchsten. Es zeigt sich deutlich, dass die höchsten Massen im Flusswasser im Rhein vor Basel erreicht werden (einzige rot markierten Flüsse in Abbildung 2a) bedingt durch die Aufsummierung der Massen (s. u. für weitere Informationen). Auch sind Effekte der Retention in Seen sichtbar, wie z. B. die Farbskala vor und nach dem Bodensee zeigt in Abbildung 2a. Auf die Effekte der Seen wird zusätzlich auch später noch genauer eingegangen.

Die Karten in Abbildung 2 zeigen auch deutlich, dass von den vielen berücksichtigten Flüssen nur ein Bruchteil mit Mikroplastikmassen belastet ist laut unseren Modellierungen. So erwarten wir für alle grau dargestellten Flüsse keine Belastungen durch direkte Mikroplastikeinleitungen in die Gewässer. Es sollte hierbei jedoch berücksichtigt werden, dass diffuse Einträge über die Luft noch nicht berücksichtigt sind. Durch diese können auch kleine Einträge in den abgelegenen Bergregionen erwartet werden, wie zahlreiche Studien gezeigt haben, die einzelne Mikroplastikfasern überall auf den entlegensten Teilen der Welt nachgewiesen haben. Generell gilt jedoch, dass hohe Belastungen dort zu erwarten sind, wo menschliche Aktivitäten und insbesondere Kläranlagen sich befinden.

Die Daten zur Akkumulation und Belastung der Seen (Abbildung 2b und c) sind im Gegensatz zu den Daten der Wasserbelastung weniger eindeutig zu interpretieren. Insbesondere für Seen können kleine Seen hohe Belastungen pro ha ausgesetzt sein. Hierbei gelten auch Staustufen, wie z. B. an der Aare, als Seen oder stehende Gewässer und können somit die logarithmische Skala in Abbildung 2c verzerren. Allgemein ist die Durchmischung von Mikroplastik in Seen noch wenig erforscht, weshalb wir uns für vereinfachte Darstellung basierend auf totalen Massen im See entschieden haben.

Allgemein gilt jedoch, dass zwischen den verschiedenen Polymeren eine starke positive Korrelation besteht. D. h. sind zum Beispiel hohe PET Werte zu erwarten, dann sind auch für die anderen Mikroplastikpolymere hohe Belastungen zu erwarten.

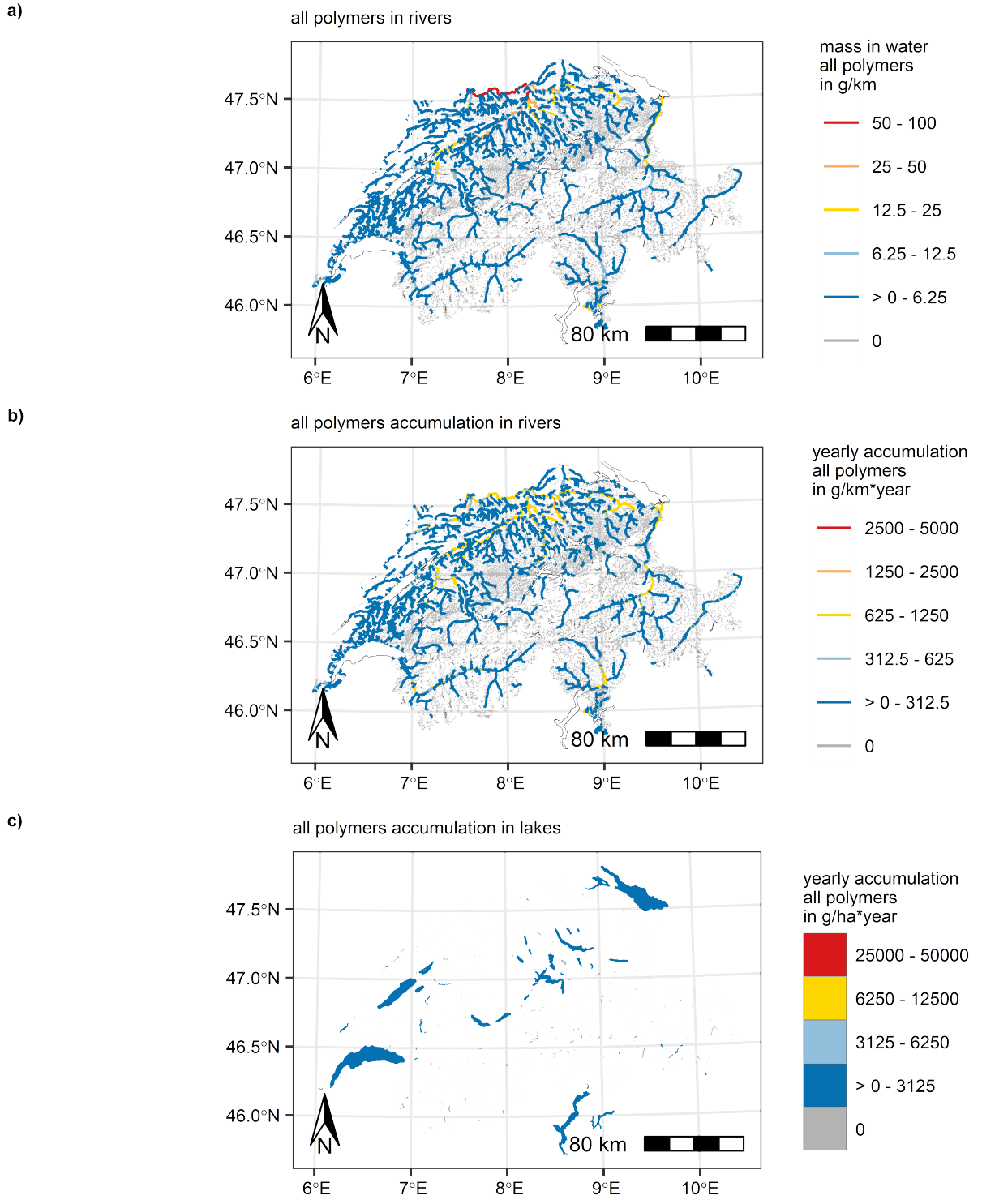

Abbildung 2: a) Karte der Schweiz mit den modellierten Mikroplastikkonzentrationen in g/km im Wasser; b) modellierten jährlichen Akkumulationen an Massen in Flüssen in g/km; c) Akkumulationen in Seen in g/ha. Bitte beachten, dass die Skalen logarithmisch sind.

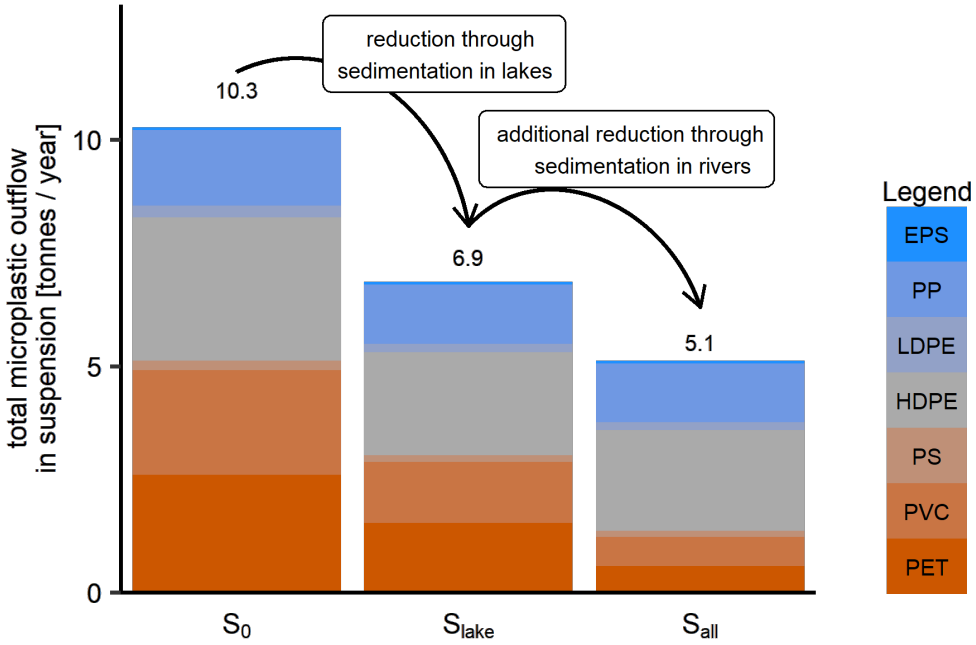

Abbildung 3: Ausfliessende Mikroplastikmassen aus der Schweiz für verschiedene Szenarien. S_0 beschreibt keinen angenommenen Rückhalt (Inputemissionen), S_{lake} beschreibt Rückhalt nur in Seen und S_{all} umfasst Annahmen zum Rückhalt in Seen und Flüssen.

Abbildung 3 zeigt die Ergebnisse der schweizweiten Prozesse bezüglich Mikroplastiktransport laut unserer Modellierungen und ist somit eine Zusammenfassung aller in Abbildung 2 dargestellter Flüsse. S_0 gilt als Szenario ohne Retentionsprozesse und somit als Inputemissionen in das System. Bedingt durch die Modellierungsprozesse ist der Input bis jetzt, auf direkte Emissionen in die Gewässer beschränkt und umfasst somit vor allem Kläranlageneinleitungen und zu geringeren Anteilen Littering. S_{lake} und S_{all} beschreiben die zu erwartenden Mikroplastikmassen, die die Schweiz durch Flüsse verlassen würden, wenn nur Retention in Seen (S_{lake}) oder Flüssen und Seen (S_{all}) berücksichtigt werden. Die wichtigste Information ist, dass ca. 50% der Mikroplastikemissionen in die Gewässer und Seen in der Schweiz zurückgehalten werden (vergleiche die Szenarien in Abbildung 3). Hierbei sind die Seen deutlich wichtiger als die Flüsse für die gesamte Retention. Dies gilt jedoch nicht für alle Polymere gleichermassen, wie die Farbskala in Abbildung 3 zeigt. So sind Seen vor allem für den Rückhalt von leichteren Polymeren (alle ausser PVC und PET) von entscheidender Bedeutung, während PVC und PET auch in Flüssen vermehrt zurückgehalten wird.

In Bezug auf Massen kann beobachtet werden, dass trotz Rückhalt von Mikroplastik, die Mikroplastikmassen mit der Flusslänge zunehmen wie auch schon in Abbildung 2 gezeigt. Beispielhaft sind in Abbildung 4 die Massentransporte für die Flüsse Rhein (inkl. Zufluss Aare),

Rhone und Doubs dargestellt. Ebenfalls werden in Abbildung 4 die Einflüsse der Seen (graue Schattierung) sichtbar indem nach grösseren Seen deutlich geringere Frachten zu erwarten sind als vor den Seen.

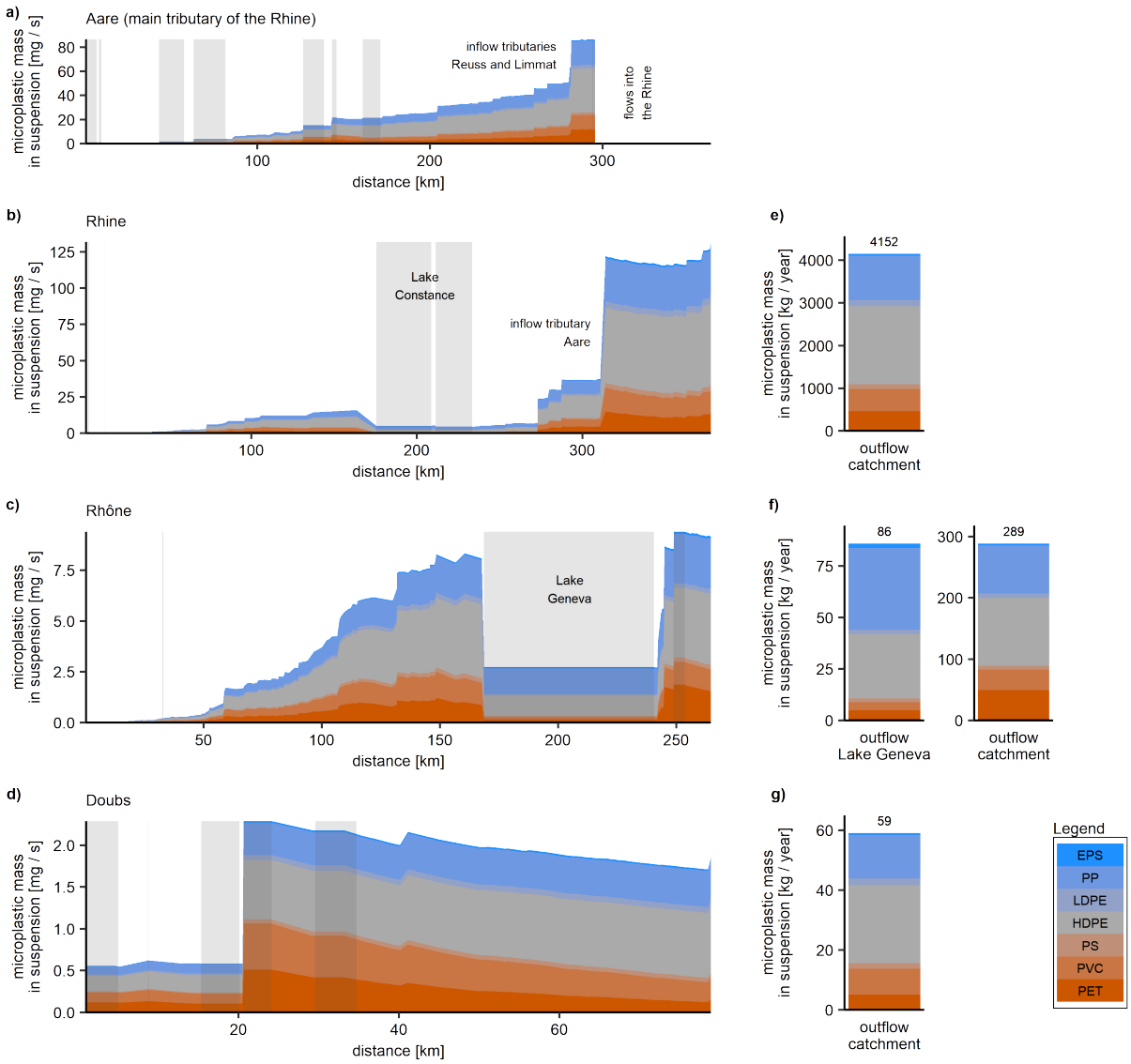

Abbildung 4: Massentransporte für Mikroplastik in den Flüssen Aare, Rhein, Rhone und Doubs. Zusätzlich sind noch die Jahresausflüsse am Grenzübertritt der Flüsse dargestellt. Km 0 ist die Quelle und die maximale Km Zahl entspricht dem Grenzübertritt zu den Nachbarländern der Schweiz.

Abbildung 4 zeigt ebenfalls, wie unterschiedlich der Massenfluss in verschiedenen Einzugsgebieten und Flussverläufen sein kann. Allgemein konnten wir feststellen, dass die Unterschiede zwischen den verschiedenen Einzugsgebieten in etwa ähnlich gross sind, wie die Unterschiede, die wir auf Grund von unterschiedlichen Polymereigenschaften feststellen konnten. Dies ist insoweit wichtig, weil in bisherigen Modellen Polymereigenschaften, und

insbesondere die Dichte, die verschiedene Sedimentationsgeschwindigkeiten verursacht, als Hauptkriterium verwendet wurde, wie viel Mikroplastik von den Landflächen in die Ozeane exportiert wird. Genauere Informationen sind dem angehängten Preprint zu entnehmen.

Für die Schweiz bedeutet dies, dass insbesondere die Orte zum "Mikroplastikexport" durch die Flüsse beitragen, die flussabwärts von grossen Seen gelegen sind. Das sind mit Ausnahme von Bern alle grösseren Städte der Schweiz aber insbesondere die grenznahen Städte Genf und Basel. Schlussfolgernd kann gesagt werden, dass das Retentionspotential durch Seen in der Schweiz geringer ausfällt als zu erwarten wäre, da die Emissionsorte entsprechend flussabwärts gelegen sind.

Da die Modellierung massenorientiert erfolgte, wurden Grössenordnungen zweitrangig behandelt. Es können jedoch verschiedene Grössenklassen getrennt betrachtet werden, falls dies gewollt ist. Für genauere Informationen und weitere Ergebnisse sei auf den Preprint des Papers zum Modell im Anhang verwiesen.

Transport von Makroplastik

Der Transport von Makroplastik wurde bisher vor allem im Ocean betrachten. Hier seien auf viele verschiedene Arbeiten von "The Ocean Clean-up" verwiesen. Für Flüsse sind in den letzten Jahren zunächst erste Monitoring Studien veröffentlich worden, unter anderem auch für die Schweiz, wie z. B. der Hammerdirt Datensatz. Basierend auf vorhandenen Datensätzen (z. B. Hammerdirt und Arbeiten von Wageningen Research and University, WUR) konnten wir mit unseren Modellierungen zeigen, dass unter mittleren Abflussverhältnissen der Rückhalt sehr viel höher sein muss als für Mikroplastik. Auf Grundlage einer bisher nicht veröffentlichten Studie der WUR gehen wir davon aus, dass auf einem ca. 10 km langen Flussabschnitt mit teilweisem Uferbewuchs ca. 75% des Makroplastikinputs zurückgehalten werden. Wenn wir diesen Wert als Faktor in das oben beschriebene Model einsetzten, scheinen die Werte in der Grössenordnung mit Messungen (WUR, noch nicht veröffentlicht) zu liegen.

Abbildung 5 zeigt, wie in einem ersten Versuch der Verlauf des Rheines aussehen würde, wenn wir die oben beschriebenen Annahmen anwenden. Die Abbildung ist somit mit Abbildung 4 vergleichbar und die Farbskala ist ebenfalls die gleiche. Für Makroplastik ist jedoch davon auszugehen, dass die Polymerarten nicht so wichtig sind, wie für Mikroplastik. Vielmehr werden verschiedene Produkte entscheidend sein, die sich dann evtl. wieder verschiedenen

Polymeren zuordnen lassen können. Beispielhaft sei z. B. die PET-Flasche genannt, die aus PET besteht, wohin gegen Verpackungen z. B. aus HDPE, LDPE oder PP etc. bestehen könnten. Für verschiedene Produkte könnten dann unterschiedliche Faktoren (s. Methoden) in Betracht gezogen werden.

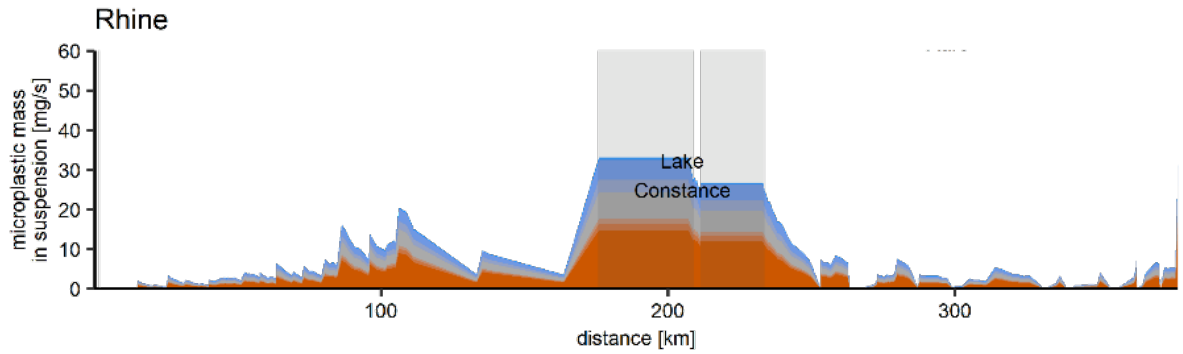

Abbildung 5: Erste Modellierungen für Makroplastikmassen im Rhein. Die Abbildung ist Vergleichbar mit Abbildung 4 für Mikroplastik. Die Seen (hier der Bodensee) sind im Modell noch nicht berücksichtigt worden.

Zur Zeit sind wir mit der konkreten Modellierung von Makroplastiktransport beschäftigt, wobei wir das vorhandene Modell erweitern wollen. Sicher lässt sich sagen, dass es unteren mittleren Abflusswerten einen hohen Makroplastikrückhalt geben muss, da die zu erwartenden Inputemissionen sonst die Messwerte um viele Magnituden überschreiten. So kann es keine so starke "Aufsummierung" der Massen geben, wie wir sie für Mikroplastik erwarten. Mit genaueren Ergebnissen kann im Frühjahr 2023 gerechnet werden.

Plastik Transport vom Boden ins Wasser

Der Transport von Mikro- oder Makroplastik über Böden ist erst zu einem kleinem Teil erforscht. Für Makroplastik konnten bis jetzt unveröffentlichte Daten aus Wageningen (WUR) zeigen, dass Wind oder Regen in Zusammenhang mit Hangneigung für einen Transport sorgen könnten. Allerdings scheinen dafür glatte Oberflächen unabdingbar zu sein, falls nicht Wasser als Transportmedium betrachtet wird. Somit gehen wir in einer ersten Annahme davon aus, dass der Überlandtransport über grosse Distanzen eher unbedeutend ist, wenn wir von bewachsenen Landschaften (Bäume, Büsche, Grass) ausgehen.

Ähnlich wenig Daten sind für Mikroplastiktransport über Land verfügbar. Eine erste Studie auf grösserer Feldskala (Rehm et al., 2021, doi: 10.1016/j.scitotenv.2021.148774) zeigt, dass vertikaler Transport höher als horizontaler Transport sein könnte. Wiederum war eine

Hangneigung Voraussetzung, damit die Mikroplastikpartikel im Oberflächenabfluss transportiert werden konnten.

Zusammenfassend lässt sich sagen, dass der weiträumige Transport über Land von Plastik hin zu den Gewässern eher unwahrscheinlich ist. Eine Ausnahme könnte hierbei der Transport durch die Luft sein. Als weiteren wichtigen Faktor sehen wir den Anschluss von urbanen Gebieten und Strassen an die Kanalisation. Durch die Kanalisation können Plastikmassen vereinfacht in Richtung Gewässer transportiert werden, insbesondere durch Trennwasserkanalisationen ohne Kläranlagenanschluss. Zusätzlich sind auf / neben Strassen und in bebauten Gebieten die höchsten Plastikemissionen zu erwarten (s. Kawecki und Nowack, 2020).

Da im Moment die für das Modell nötigen Parameter noch nicht bestimmt werden können, wurde entschieden, ein Boden-Land gekoppeltes Model nur mittels Szenarien zu rechnen. Die wird sowohl für Mikro- als auch für Makroplastik gemacht. Die potentiellen Einträge werden nach Landnutzungskategorien unterteilt ermittelt. In diesen Szenarien nehmen wir an, dass Plastik nur bis zu einem gewissen Abstand von Gewässern ins Wasser gelangt und berechnen dann mit dem Makroplastikmodell die Konzentrationen im Wasser. Dies erlaubt es, den Einfluss des Transportes von Plastik über Land auf die Menge im Wasser zu quantifizieren und abzuschätzen, wie relevant dieser Prozess auf die Menge Plastik im Wasser sein kann.

Ausblick

Durch den Abbruch der Dissertation von Johannes Schorr nach 4 Monaten und die Zeit bis zur Findung eines neuen Doktoranden ist es zu Verzögerungen im Zeitplan gekommen. Das Projekt hat daher die Dissertation von David Mennekes, welcher das Projekt übernommen hat, nur zu einem Teil finanziert. Nach Abschluss des BAFU-Projektes wird das Projekt aus Eigenmitteln weiter finanziert, um die noch offenen Arbeiten abzuschliessen. Der Abschluss der Dissertation ist auf Ende Sommer 2023 geplant, weshalb dann auch die letzten Resultate des BAFU-Projektes verfügbar sein werden.

Anhang

Preprint der Publikation «Modelling Microplastic Transport in River Networks with High Spatial Resolution at Country-level»

Modelling Microplastic Transport in River Networks with High Spatial Resolution at Country-level

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Abstract

Microplastics are an 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. In this work we coupled a high-resolution microplastic release model with an aquatic fate model and parameterized it on a country-scale. This model reveals that for transport and fate of seven different polymers on a country-scale, catchment characteristics (e.g. lake sizes and location, river connections) are as important as polymer properties such as density. For Switzerland, out of 10 tons microplastics directly emitted into water bodies, up to 80 % are retained within the country for the more dense polymers (i.e. PVC, PET) while retention rates for light polymers are much lower (0.2-33 %). Finally, the model predicts microplastic masses in each river section of the whole country.

Keywords: keyword01, keyword02

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1. Introduction

Across the globe we are facing an increasing emission of plastics into the environment (Geyer et al., 2017). 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 sediments (Ambrosini et al., 2019; Eriksen et al., 2013; Mani et al., 2016; Rochman, 2018; Samandra et al., 2022; Van Cauwenberghe et al., 2013). Most microplastic found in the environment have in common that they were likely not emitted on the sampling site, but were transported there, with rivers being one of the main way of transport (Correa-Araneda et al., 2022; Eriksen et al., 2014; Lechthaler et al., 2020; Meijer et al., 2021; Nizzetto et al., 2016; Watkins et al., 2019). Consequently, this raises the question how far microplastic can be transported in rivers and how they vary across catchments, countries or continents with different landscapes and land use. Measurements of microplastics were performed all around the globe, providing snapshots of concentrations at specific locations using a variety of different sampling and measurement methods (Bellasi et al., 2020; Vivekanand et al., 2021). Nevertheless, for a more profound understanding of transport processes, a higher temporal and spatial resolution of measurements would be desirable but remains challenging due to various reasons such as inconsistent measurement quality or comparability across different sampling studies as well as time consumption for sampling (Conkle et al., 2018; Silva et al., 2018).

To overcome measurement limitations, models of (micro)plastic transport have been developed (Besseling et al., 2017; Domercq et al., 2022; Norling, 2020; Van Sebille et al., 2020). Plastic transport models of large areas such as countries or continents are currently only available for macroplastic transport across the oceans (e. g. Van Sebille et al., 2020, 2015). Large area microplastic models for fresh waters simulating the transport of (micro-)plastic along river and lake networks are lacking. The only existing fresh water models for microplastic transport cover single catchments with a single river without tributaries in the order of 1000 km maximum river length (Besseling et al., 2017; Domercq et al., 2022; Norling, 2020) or are only estimations on small catchment scales without consideration of single rivers (Siegfried et al., 2017).

The existing river based microplastic transport models, i. e. nanoDUFLOW (Besseling et al., 2017), Full Multi (Domercq et al., 2022) or INCA-microplastics (Norling, 2020), include transport, sedimentation and accumulation of microplastics along different river segments. Pollution is commonly modelled as numbers of microplastic particles transported downstream from one river segment to the following one. To determine sedimentation (retention) of microplastic particles, the models consider different particle sizes, shapes and polymer-dependent critical shear stresses as well as forces generated by the fluid. Furthermore, these models partly consider (hetero-)aggregation and biofouling as influencing factors for sedimentation behaviour. Addi-

tionally, different particle states (e. g. in suspension, in sediment or deep-sediment burial) are implemented. In order to calculate the model parameters, all three existing models require highly accurate input data about hydrological properties, e. g. slope, river width, water depths, as well as particle-dependent properties of the microplastics and other particles present in suspension and the bed sediments, e. g. concentration, diameter or shape.

Given the high demand of input data for these existing models, their application is restricted 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. To obtain accurate hydrological input data for calculating critical shear stress or related forces is almost impossible, which makes these models not applicable at this level. For example, the hydrological base-model for nanoDUFLOW is highly sensitive to hydrological input data and does not allow for moderate steep slopes since it was developed for the Netherlands. The three models described above were so far not broadly applied for research questions related to microplastic transport.

Any fate model on a large scale also requires information on microplastic releases. The three models described above are mostly applied to point sources without considering a spatially distributed emissions model (e.g. Besseling et al., 2017; Drummond et al., 2022). A first country-wide high-resolution macro- and microplastic release model for soil and water was developed recently and was first applied to Switzerland (Kawecki and Nowack, 2020). This model distributes the environmental releases which were predicted by a material-flow analysis coupled to an emissions model, (Kawecki and Nowack, 2019) by using proxies for release processes. Such a country-wide release model needs to be available for any country-wide fate model and forms the basis for the current work.

The aim of this paper was therefore to develop a new model to predict microplastic fate and transport for large geographical areas such as countries with a high spatial resolution (single river sections). We integrated existing fate modelling approaches for microplastics (i. e. nanoDUFLOW or the Full Multi model) and a spatially-resolved release model with a large-scale hydrological model for whole countries. We apply the developed model in a case study to Switzerland by calculating the transported mass of seven different polymers (EPS, PP, LDPE, HDPE, PS, PVC, PET) across all rivers and lakes in Switzerland. Using this case study, we aim are able to identify how important plastic transport in rivers on a country-scale is compared to removal in lakes and how regional differences between catchments within a country affect microplastic concentrations at a certain location.

2. 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. Also, we provide a simple river network (see SI) to explore the model independently.

2.1. 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 data set in the format of a shapefile or GeoPackage format file. The digital river and lake maps should fulfil the criteria listed in the SI to guarantee a logical river connectivity which will ensure a correct microplastic transport through the river network. Information about river segments and lakes are presented in the river network file. The exact requirements and names are listed in the SI.

2.2. 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 Kawecki and Nowack (2020). This model is based on a material flow analysis of plastics through society (Kawecki et al., 2018) which 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 sources (Kawecki and Nowack, 2019). To allocate the emissions in a next step to water bodies, Kawecki and Nowack (2020) used geographical proxies such as population density, land use, area of construction or traffic density to geographically distribute the total masses. The final data are yearly emissions into each river segment and lake differentiated along the seven polymers.

2.3. Modelled states of microplastics

In the model, microplastic masses are considered in three different states for each polymer individually: in suspension, in sediments and in deep sediments (accumulation) (Figure 1). 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 are first assigned to suspension before allowed to sediment (Figure 1). For plastics in the sediment three possible pathways are possible: they can be buried in deep-sediments and accumulate, they resuspend into the suspended state or they are transported with the sediment to the following downstream sediment container of the river or lake. Further information is given

process	polymer	lakes	rivers
sedimentation of particles in flowing water in [s^{-1}]	EPS	$0.5*(0.95*(1-e^{-0.004*A}))$	0
	PP	$0.75*(0.95*(1-e^{-0.004*A}))$	5.050×10^{-8}
	LDPE	$0.95*(1-e^{-0.004*A})$	2.565×10^{-7}
	HDPE	$0.95*(1-e^{-0.004*A})$	2.565×10^{-7}
A = lake surface area in [km^2]	PS	$0.95*(1-e^{-0.004*A})$	1.115×10^{-6}
	PVC	$0.95*(1-e^{-0.012*A})$	1.499×10^{-5}
	PET	$0.95*(1-e^{-0.012*A})$	2.499×10^{-5}
resuspension	all	0%	3%
long term accumulation (burial)	all	everything	10% maximum rate: 1×10^{-8} s^{-1}

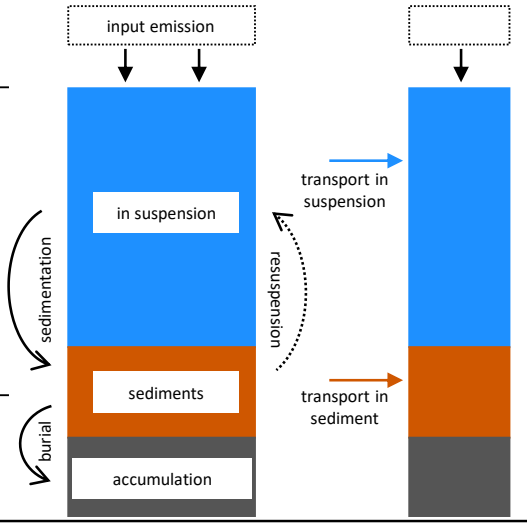

Figure 1: Conceptional model setup per each river segment including processes within one segment and transport processes to the next segment. Additionally, parameters to derive sedimentation and long term sedimentation (burial) are listed. The parameterisation is different for rivers and lakes and depends on the microplastic polymer.

below, explaining each state more in detail.

The model uses microplastic masses instead of particle numbers as microplastic release models based on material flow analysis only provide mass flows. We simplified our model to one particle size class with the focus on masses and mass flows instead of particle numbers. Thus, bigger particles are relatively more mass dominant than many small particles. For instance one spherical particle with 3 mm diameter represents the mass of one billion very small particles in the order of a few µm diameter. However, results in masses can be approximately transferred to size distributions by assuming a distribution of size classes.

2.4. Transport in suspension (advection)

Transport in suspension is 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 erosion (Waldschläger and Schüttrumpf, 2019b).

We calculated microplastic transport and input emissions as mass per second assuming steady state for a one second time step 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 calculate the mass of plastic emissions which will be transported to the next river section down stream per second. Please, refer to the SI for further

explanation and an example. For lakes we assumed a steady state system per second which means that the inflow mass from upstream plus the input emissions on site per second equal to the outflow mass per second. To derive the plastic mass per river segment, the mass per second is multiplied with the time the water needs to flow through the corresponding section (see the SI for possible ways to derive average flow velocities).

2.5. Fate processes

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

Following the pathways in Figure 1 we derive sedimentation factors (f_{sed}) as a first step of microplastics reduction from masses in suspension. Sedimented microplastic masses then can be transported to deep sediments and accumulated according to the corresponding factor f_{acc}. We applied both factors, f_{sed} and f_{acc}, for each river segment and lake and each polymer individually based on available literature data. Removal, e. g. 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 are estimated based on the modelling results by Besseling et al. (2017), Siegfried et al. (2017) and (Domercq et al., 2022) and measurement results (e. .g Klein et al., 2015; Yan et al., 2022). While existing modelling results suggest that spherical particles bigger than around 1 mm sediment immediately after entering the waters (e. g. Besseling et al., 2017; Domercq et al., 2022), measurement studies show that the dominant size class in the environment, including rivers, are 0.5 to 5 mm in size (Conkle et al., 2018; Klein et al., 2015; Laermanns et al., 2021; Osorio et al., 2021). Estimating retention factors remained challenging due to the lack of existing data and contradictory data of measurement and existing modelling results. Here, we estimate f_{sed} in perspective to the modelling results of Besseling et al. (2017) which suggest that the two most dense polymers (PVC, PET) would be almost fully retained over the 40 km long modeled river while retention for less dense polymers would be lower respectively (see SI for further information). Consequently, values for polymers more dense than water are similar to the 5 µm size class in Besseling et al. (2017) without directly referring this size class. For polymers less dense than water which were not considered in existing models we used literature data to estimate f_{sed} in relationship with the heavier polymers.

Finally, f_{sed} was derived for an entire river segment based on the travel time (L in s) through the river segment and the sedimentation rate extracted from Besseling et al. (2017) (Figure 1, SI). Furthermore, we used a negative compound interest approach shown in eq. (1) to assure that microplastics lost in the beginning of

the segment cannot be lost afterwards. For rivers f_{sed} is calculated as followed:

$$f_{sed,\ river} = 1(1 - k_s)^L \tag{1}$$

where k_s is the sedimentation factor per second derived from the results of Besseling et al. (2017) and given in Figure 1. L is the average travel time in seconds through a river segment calculated by eq. (2).

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

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 correspond with longer residence time in a river segment which causes higher plastic retention in the river segment for eq. (1).

For f_{acc} we assumed 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. (2022). However, the maximum f_{acc} was set to $1 \times 10^{-8}\,s^{-1}$ which is between values used by Domercq et al. (2022) and Besseling et al. (2017) who based their long term sedimentation rates on Praetorius et al. (2012) and Koelmans et al. (2009) correspondingly.

To derive fate processes in lakes, we aimed for a single f_{sed} per lake. However, we found only very few studies describing a mass balance of microplastic fluxes through lakes to derive f_{sed} or f_{acc} for lakes. Based on available data points (see SI) we fitted a logarithmic curve relating plastic sedimentation yield (k_l) with lake surface area (A) in km^2. 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 (see SI). For lakes, f_{sed} was calculated as followed:

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

The asymptotic maximum plastic loss C was set to 95 % (0.95) and k_l was varied with polymer type. Across all polymers we aimed for an approximately 90 % loss rate for Lake Geneva ($A = 500\,km^2$) with $k_l = 0.005$ which is a retention rate based on a modelling results by Boucher et al. (2019).

To account for differences between polymers we varied k_l based on Yang et al. (2022) who summarized multiple studies of sediment analysis. Thus, polymers notable less dense than water (i.e. PP) were found to be less abounded in sediments compared with particles in suspension while for polymers more dense than water the ratio was found to be opposite (Yang et al., 2022). For the analyzed polymer PE (here differentiated

into LDPE and HDPE) no trends were found (Yang et al., 2022). Hence, for the polymers EPS, PP, LDPE, HDPE and PS we used $k_l = 0.004$ (Figure 1). For EPS and PP eq. (2) was multiplied with 0.5 and 0.75 correspondingly to decrease f_{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_l (0.012). All equations are shown in Figure 1. Further information about factors and available studies are presented in the SI. We assume that all plastics in the sediment will be accumulated ($f_{acc} = 1$).

2.6. Resuspension

Microplastic resuspended (f_{resus}) from the sediment compartment are added to the microplastic in suspension. We allowed 3 % (Figure 1) of plastics in the sediment to resuspend since Praetorius et al. (2012), and consequently Domercq et al. (2022), 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_{sed} describes the entire microplastic retention per lake including potential resuspension influences.

2.7. Transport in sediments

Based on our steady state assumption, velocity of microplastic transport in sediments is only important 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_{acc}, f_{resus}). In our model, sediment transport velocity is 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.

2.8. Code

The code is freely available on ... So far the model is written in `R` and uses `QGIS` functions through the `R` package `qgisprocess`.

2.9. 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 km^2, including multiple lakes up to about 500 km^2 surface area and multiple thousands of different rivers which all flow out of the country due to topography. For input microplastic emission data into the environment we used modeled data by Kawecki and Nowack

(2020). 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 year^{-1} for the year 2014 (Kawecki and Nowack, 2019).

For the results we focus 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 km^2 (always Swiss´ area only), 9210 km^2 and 372 km^2 for the Rhine, Rhône and Doubs while 3.4 %,5.3 %, 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 only available 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 Faure et al. (2015); Mani et al. (2016). Please, refer to the SI for further information and maps of the catchments of focus and the Swiss river network.

3. Results and Discussion

The model was able to connect each river segment with the corresponding downstream river segment. If multiple river features flow into one feature the plastic masses were summed for the following downstream segment. Similar for lakes, all inflowing mass plus the emissions into the lake itself were assigned to the outlet after applying sedimentation factors.

3.1. Microplastic pollution in Switzerland

For Switzerland we analyzed the three catchments of focus as well as the retention by lakes and rivers. This is possible by using different scenarios such as allowing no retention (S_0), allowing retention only in lakes (S_{lake}) or only in the 15 biggest lakes in Switzerland (S_{lake15}) and including retention in lakes and rivers (S_{all}). Shown as a map (Figure 2), our model highlights regions of higher polluted rivers as well as the high number of rivers without expected pollution. 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 effected 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. Only some slower flowing rivers receive relatively high accumulation masses (compare Figure 2 with SI). As a

result, microplastic pollution in suspension (Figure 2) as well as microplastic accumulation are concentrated along relative few river sections (see additional maps in SI).

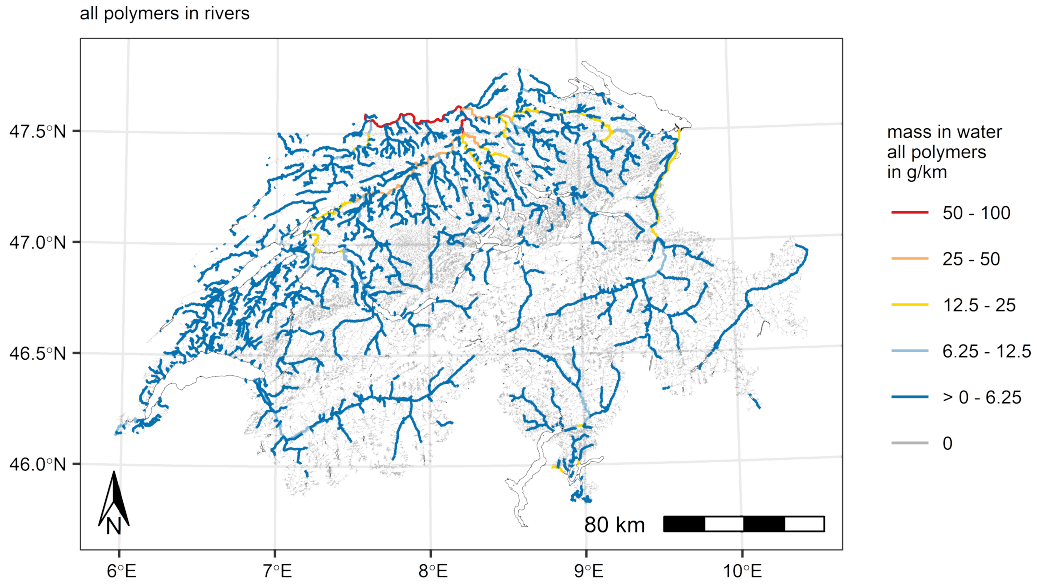

Figure 2: Spatial distribution of microplastic in rivers shown as microplastic masses in suspension for rivers.

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 significant correlation between all different polymers with a correlation coefficient of 0.9.

Using the scenarios S_0, S_{lake}, S_{lake15} and S_{all}, we observe that 33 % of all microplastics are retained in lakes (Figure 3) of which 99 % are retained in the 15 biggest lakes in Switzerland although they correspond only to 7 % of all lakes receiving microplastic pollution. Additionally, microplastic sedimentation in relationship to lake size show that most smaller lakes (i. e. smaller than 0.1 km^2) have much lower microplastic accumulation per area than bigger lakes. Thus, the 20 lakes with the highest accumulation per area are all bigger than 0.1 km^2 (see SI).

Rivers, on the other hand, retain only about 17 % of all input microplastic when retention in lakes and rivers are considered. Here, we observed a wide range from no retention for EPS up to 40 % retention for PET (see colors in Figure 3). Consequently, the reduction from S_{lake} to S_{all} in Figure 3 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 f_{sed} and f_{acc}.

It should be stated that accumulation of microplastics in lakes slightly decreased when microplastic sedimentation in rivers was considered due to less microplastic masses in the modeled system (compare S_{lake} with S_{all}).

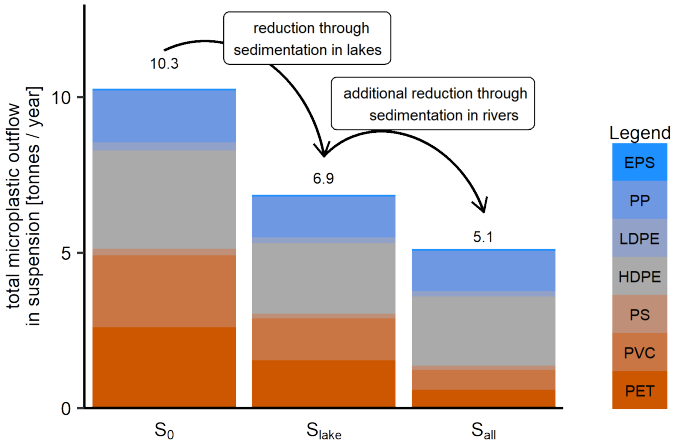

Figure 3: Microplastic retention of all analysed polymers differentiated by different colors in entire Switzerland. The scenarios S_0, S_{lake}, S_{all} are different model runs which consider no sedimentation and accumulation (S_0) sedimentation and accumulation only in lakes (S_{lake}) and sedimentation and accumulation in lakes and rivers (S_{all}). S_0 equals to input emission to the system.

3.2. Masses and sources of microplastics along the river lengths

Microplastic masses and retention do not only vary among different polymers but along catchments and the lengths of rivers, too. Here, sedimentation traps, such as lakes, are highly influencing microplastic masses in suspension but also in sediments. For the three catchments of focus (Rhine, Rhône and Doubs) we observe outflowing microplastic masses which differ across three order of magnitudes from about 60 kg year^{-1} for the Doubs river to over 4000 kg year^{-1} for the Rhine which corresponds to 81 % of all microplastics leaving Switzerland (Figure 4 e), f), g)). Together, these catchments cover about 88 % of all microplastic outflows of Switzerland (compare with S_{all} in Figure 3). The remaining 12 % are shared among the outflow of Lago Maggiore (Ticino River) (6 %; 327 kg year^{-1}) and along flow into "unknown" (including smaller border crossing rivers, 2 %), the Breggia River towards Italy (2 %), the Inn River towards Austria (1 %) and other smaller rivers.

We estimate that in total (in suspension and as sediment transport) about 4565 kg year^{-1} microplastics are leaving the Rhine catchment towards Germany while 300 kg year^{-1} and 61 kg year^{-1} are leaving the Rhône and Doubs catchment respectively towards France. About 2 to 9 % of all microplastic masses leaving Switzerland

through the three rivers of focus are transported via sediment transport with higher percentages for rivers with higher total microplastic transport (i. e. Rhine).

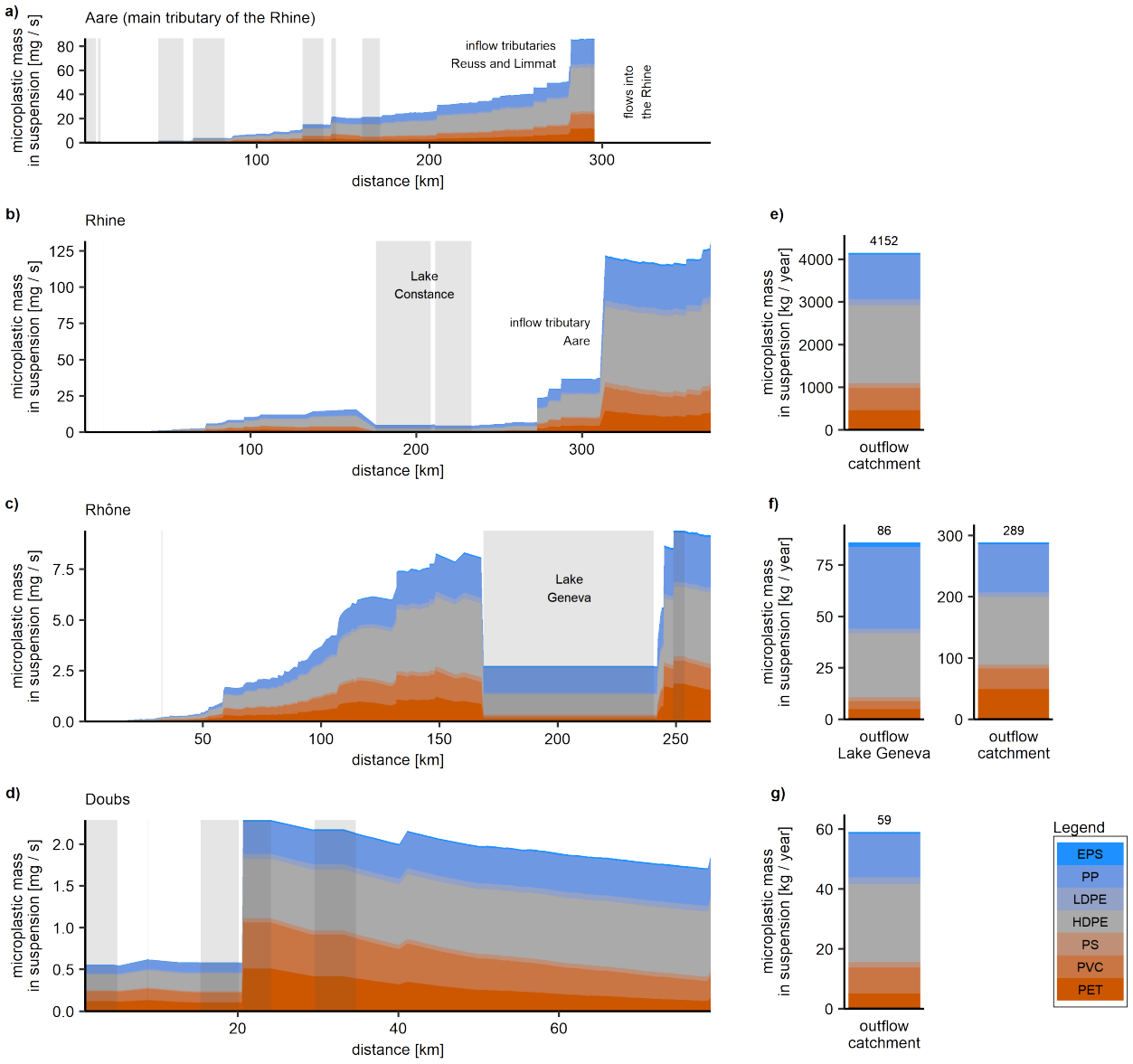

Figure 4: a) - d): Transport of microplastic masses along the main stream of the rivers Rhine, Aare (main tributary of the Rhine), Rhône and Doubs.Blue to red shades show the different polymers as stacked values while grey shades are symbolizing lakes. The microplastic masses can be understood as: "how much microplastic is passing a river cross section per second" which can be directly related to actual microplastic concentration in water when considering the discharge. Distance shows the distance from spring according to the GIS vector file.
e) - g): The stacked barplots present yearly masses at the outflow, the Swiss border, of the corresponding catchments. 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 magnitudes.

Generally, microplastic masses in rivers are increasing with distance from the spring. Especially, 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 (i. e. Bern, Zurich and Lucerne). For the Rhône catchment, on the other side, the city of Geneva located just downstream of the Lake Geneva, is the main emission sources of microplastics leaving the Rhône towards France.

3.3. Retention of microplastics along the river length

Retention of microplastics is mostly influenced by lakes. For further analysis, this section focuses on the three polymers EPS, PS and PET which cover the range of very low density (EPS) to similar density to water (PS) until the most dense analyzed polymer (PET). Figures for all other polymers are shown in the SI.

Due to the importance of lakes, it is crucial to consider the location of lakes in relationship to the locations of the emissions. In Switzerland both biggest lakes with around $500\,\mathrm{km}^2$, Lake Geneva (Rhône catchment) and Lake Constance (Rhine catchment), reduce the microplastic mass through sedimentation to about one third of the inflow masses (Figure 4 b) and c)). Nevertheless, for the Rhine catchment overall retention in lakes is less dominant than for the Rhône (Figure 5 d) - f)). This is because large mass flows originate from the Aare catchment which flows into the Rhine only after Lake Constance (Figure 4 b)). This is especially true for polymers which are generally less influenced by sedimentation in rivers (i. e. EPS, PP, LDPE, HDPE and PS).

In the Rhône catchment the 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 Figure 4 c) and Figure 5 b), 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 (i. e. the city of Geneva). This applies in particular to the more dense polymers (PVC and PET), since almost all pollution upstream 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 (Figure 4 c), Figure 5 b)).

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 (Figure 5). Only for the Doubs catchment with lakes of small surface areas, sedimentation in rivers is more dominant for less dense polymers (Figure 5 f) and SI).

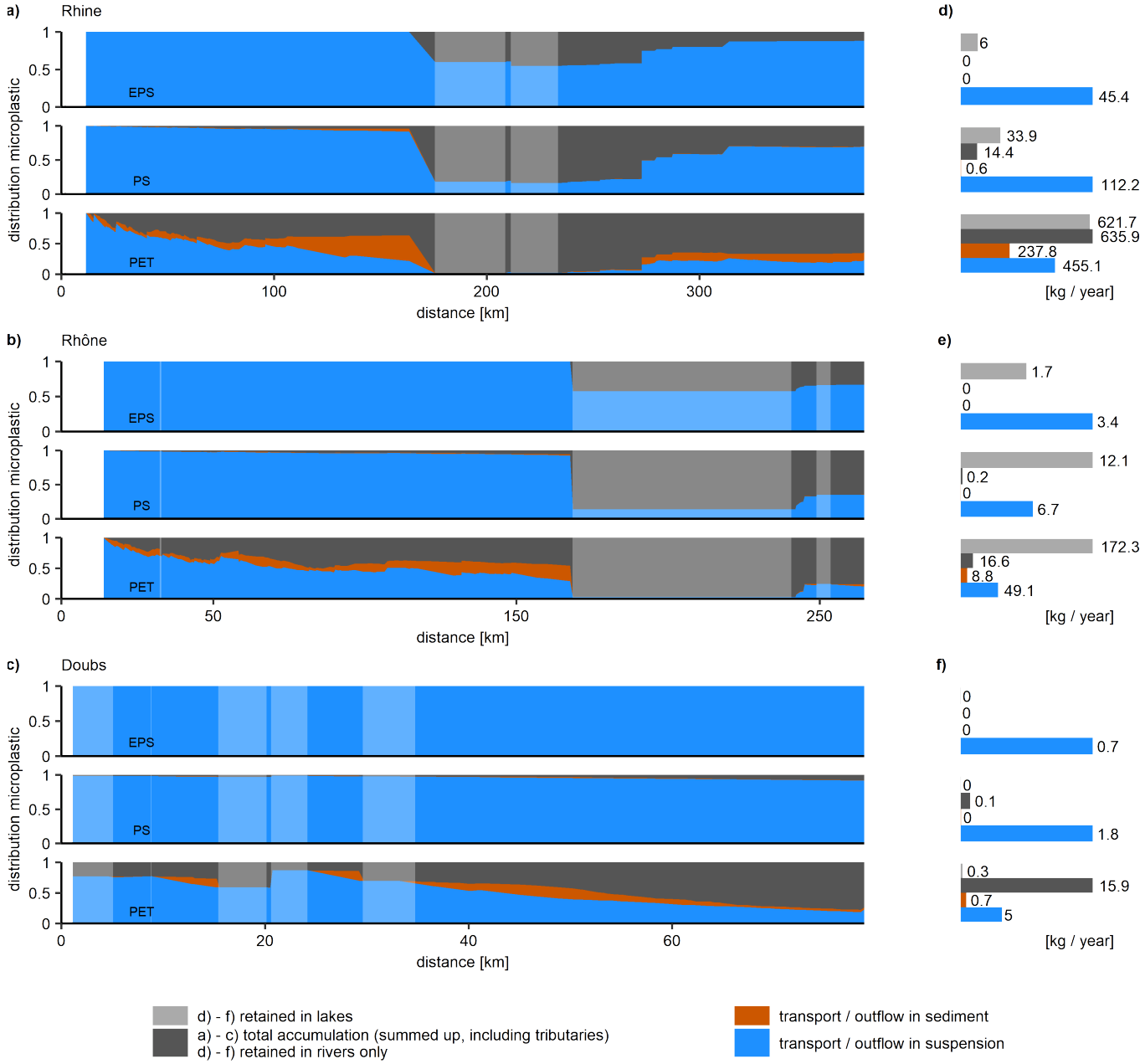

Figure 5: Left side a) - c): Distribution between microplastic in suspension, in sediment and accumulated along the main river for the three catchments of focus, Rhine, Rhône and Doubs and for three selected polymers (EPS, PS and PET). Values represent for any point the entire upstream distribution including all tributaries. The distance corresponds to the accumulated lengths of mapped river polylines from the spring to the border of Switzerland. Shaded areas correspond to lake areas. Gaps in the beginning of the river correspond to non-existing plastic masses.
Right side d) - f): Outflow masses and location of retention in kg per year for the entire catchment (Rhine, Rhône and Doubs) and the same polymers. Here, we only consider the catchment area located in Switzerland.
Please note that numbers variate across a few orders of magnitude among different polymers and catchments.

3.4. Importance of catchment differences

Given the geographical river and lake maps for Switzerland, we analyzed the pathways of emission from 500 randomly selected mapped river segments. To analyse retention rates of the segment, we set the input emission for the selected segment to $1\,s^{-1}$, while all other segments received no input emissions. This was repeated one by one for each 500 randomly selected segment. We found that different catchments cover a wide range retention rates for microplastics (Figure 6). There is no simple linear function of retention rate as dependency of river flow lengths to the outlet. Instead, the Rhône catchment mimics best a logarithmic function due to the influence of the Lake Geneva (Figure 6 a) - c)), while for the Rhine catchment the retention becomes more complex. Here, each tributary contributes very differently to the overall microplastic transport in the river which is best visible for the sub-catchment of the Rhine shown in Figure 6 a) - c). Even within the sub-catchment Aare (crosses) differences can clearly be observed Figure 6 a) - c).

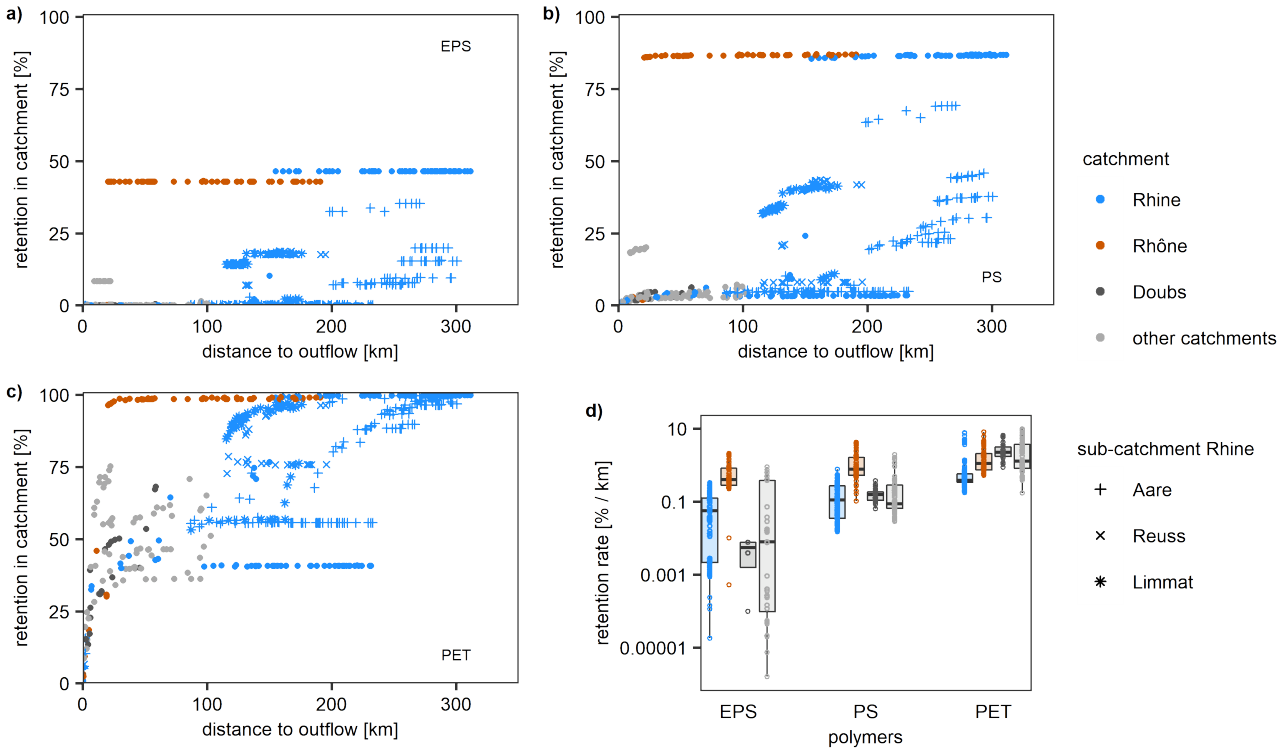

Figure 6: Influence of catchments on retention through sedimentation and accumulation for the polymers EPS, PS and PET and the catchments in Switzerland. In other words, how much microplastic in suspension is lost to sediments or accumulation (see also Figure 5). We selected 500 river segments randomly and set the input emission to 100 % while all other river segments were initially set to 0 input emissions.
a) - c): Shown are the retention and loss into the sediment at the outlet of the catchment in relationship to the distance to the outlet according to the polyline maps. Colors and shapes show the catchment the sampling point is located. Blue round points (Rhine catchment) are located in the Rhine catchment but not in any sub-catchment listed.
d): Retention rate per kilometer for a selection of Swiss catchments. Distances are equal to distances in a) - c).

The average observed retention rate of microplastics in suspension was $0.51\,\%\,\mathrm{km}^{-1}$ (median: $0.24\,\%\,\mathrm{km}^{-1}$, standard deviation: $1.04\,\%\,\mathrm{km}^{-1}$). Based on all values, and presented in median and standard deviation, we found a right skewed distribution of retention rate values with maximum values of $9.87\,\%\,\mathrm{km}^{-1}$. High values are clearly caused by the Rhône catchment (Figure 6 and Figure 5). Overall, differences of average values among catchments are equivalent to differences among the three different polymers EPS, PS, PET. Consequently, we argue that accounting for different polymers is equally important than accounting for different catchments.

3.5. Main drivers of microplastic fate on a country scale

Our modelling clearly reveals the importance of including a higher spatial resolution, as shown by differences in masses (Figure 4) but also in a the catchment analysis presented in Figure 6. Furthermore, we highlight the importance of understanding the decrease of microplastic through retention effects of lakes as well as the increase of microplastic through pollution sites or tributaries.

Consequently, we argue that using input data and performing modelling on high spatial resolution is of high importance when estimating transport of microplastics in large areas such as countries. Finally, better spatial resolution will improve the accuracy of microplastic exportation out of the system (Figure 6). Better and more accurate input data on the other hand, will help to identify hotspots and composition of polymer masses in the system (Ballent et al., 2016; He et al., 2020).

The main driver of plastic retention in our model are lakes which, depending on the polymer, retain microplastics up to the order of one magnitude higher than rivers. Based on our findings lakes might retain up to 2/3 of inflowing microplastics which is supported by other studies also highlighting the importance of small lakes or dams (e. g. Eibes and Gabel, 2022; Watkins et al., 2019). Although lakes may be very important for microplastic retention, the effect of retention by dams is not fully confirmed yet (Weideman et al., 2019).

3.6. Model validation

To compare our model with measurement data, we used published studies about microplastic concentrations in Swiss lakes and the rivers Rhine and Rhône (Faure et al., 2015; Mani et al., 2016). Both studies present microplastic particle numbers found at the water surfaces and collected with a trawl. We transferred reported reported results to mass per second using masses per m^3 and annual average discharge at sample locations. While Faure et al. (2015) provided masses per m^3, we estimated masses for the particle numbers reported in the Rhine by Mani et al. (2016) assuming either spherical microplastics (diameter: 1 mm) or

microplastic fragments (1 mm*1 mm*10 µm).

For the river Rhine at the border with Germany we estimated the microplastic outflow by the order of one to two magnitude lower than the measurements depending on our mass calculations ($0.13\,g\,s^{-1}$ compared with about $1\,g\,s^{-1}$ for spheres or $20\,g\,s^{-1}$ for fragments based on the results of Mani et al. (2016)). We found similar differences for the river Rhône at the border between France and Switzerland. Here, Faure et al. (2015) 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 Geneva) we predicted values of almost one magnitude lower than at the border (Figure 4 f)). A similar relationship of microplastic masses was measured by Faure et al. (2015).

It should be stated that for all measurements the concentration of microplastic fluctuated across a river sections. Also, samples for the two measurements in the Rhône river were taken on different days (Faure et al., 2015; Mani et al., 2016). Additionally, our model does only consider microplastic releases directly into water bodies. Hence, lower values should be expected since fragmentation of macroplastics to microplastics as well as transport from land to surface waters are not considered, yet. Furthermore, we only consider the catchment area within Switzerland which correspond to 77 % and 67 % of the total catchment area for the Rhine and Rhône catchment respectively. Summarized, our modelled microplastic masses might increase when fragmentation of macroplastics and emission into soils are implemented. On the other hand, fragmentation to nanoplastics decreases the masses of microplastics.

As second validation measure we used the measured distribution of different polymers types. Here, we are able to capture the dominance of polyethylene (HDPE and LDPE) and PP (Figure 5) which was measured by Faure et al. (2015); Mani et al. (2016). However, comparing measurements for other polymers, e. g. PS remained challenging. Faure et al. (2015) identified 12 % of the analyzed particles as PS of which most particles were EPS (unspecified percentages). However, for EPS, a high percentages of counted particles relates only to very low masses due to the very low density of EPS. Similarly, Mani et al. (2016) found PS to be the most abounded polymer by number without stating whether EPS was assigned to PS. Summarizing the share of measured polymers, we speculate that PS is currently underrepresented in our outflow modeling, but we assume that modelled PS in rivers will increase once the contamination on soils is connected with the rivers. The reasons is that PS is much more emitted onto soils than directly into waters because of its high use in construction (Kawecki and Nowack, 2019).

Validating our model with measurement data from Swiss lakes remained challenging. First, measurements show high variability of plastic abundance across different lake sections and depths which introduces high

uncertainties for estimating plastic masses in lakes. Faure et al. (2015) found for instance 78 to 5000 microplastic particles per m^2 at lake surface of lake Geneva excluding variances for different depths. Second, we were not able to transfer microplastic mass inflows into the lakes per second to representative numbers for the entire lakes. For both modelled masses and measurements, high uncertainties would be added based on assumption of effected lake volume (i. e. effect of stratification) and the linked lake residence time.

3.7. Model parameterisation and limitation

Not including important hydrological or fluid mechanical parameters might seem to be a strong simplification of the model, however, on country scale other processes become more important. For instance, we showed that catchment properties have a similar influence on microplastic transport as different polymer properties in our model. Hence, any (small) changes based on polymer properties would modify our results only to a small extent. Furthermore, including parameters such as shear stress remains challenging for microplastics in natural rivers with mixed grain sizes and heterogenic flow conditions as shown by many studies (e. g. Shields, 1936; Waldschläger and Schüttrumpf, 2019b, 2020; Wilcock, 1988). Additionally, different flow conditions along a river cross section influence microplastic settling behaviour (Mani et al., 2019). Although critical shear stresses for microplastics are available in literature for some polymers (Waldschläger and Schüttrumpf, 2019b) calculating the corresponding forces for each river segment remains even more challenging because of required input data (e. g. flow velocity, water depths, slope etc.). Hence, we argue that flow velocity is an appropriate simplification to cover these processes which is supported by findings along the Rhine (Mani et al., 2016).

Also, mechanical parameters are closely connected with microplastic particle sizes and density. Here, experimental findings and existing models suggest that sedimentation behaviour depends on sizes and polymer density (e. g. Besseling et al., 2017; Waldschläger and Schüttrumpf, 2019b). However, existing data is based on laboratory work and theoretical assessments and measurements under natural conditions are largely missing. Additionally, the assumptions used in existing microplastic transport models for rivers are in conflict with some of the measurements. For instance polymers with a density of $1000\,\mathrm{kg\,m^{-3}}$ are assumed not to sediment according to existing models and polymers less dense than $1000\,\mathrm{kg\,m^{-3}}$ (EPS, PP, LDPE, HDPE) are not considered at all in these mechanistic models (Besseling et al., 2017; Domercq et al., 2022). Similarly, experimental laboratory studies suggest that only microplastics made of polymers denser than water are sedimenting in water (Waldschläger and Schüttrumpf, 2019a,b). In contrast, these findings are clearly not supported by river sediment measurements which also found less dense polymers in their samples (e. g. Mani et al., 2019; Osorio et al., 2021).

Summarized, we aimed for a model simplification, especially for size and shape, for instance by using representative mean values. Ideally, our model factors are derived from property (mass) distributions which should be preferred instead of multiple (size or shape) classes as also suggested by Kooi and Koelmans (2019). Finally, this work is the first modelling approach with focus on masses using high spatial resolution including polymers less dense than water. Focusing on masses instead of particle size classes also means that biofouling or (hetero-)aggregation becomes less relevant since it is only of importance for specific polymer densities (slightly less dense than water) and very small particle sizes (Besseling et al., 2017; Kaiser et al., 2017; Kooi et al., 2017; Van Melkebeke et al., 2020).

Another challenge is how to describe microplastic transport in lakes. For a detailed perspective on microplastic transport across lakes it would be desirable to better implement mixing behaviour in lakes, currents and lake water retention. We also would appreciate more monitoring studies of overall microplastic retention in lakes accounting of different lake sizes (area or volume) to improve the overall understanding. Facing all these unknowns using a single removal factor per like is the best solution so far.

Similar simplification apply for microplastics in sediments. In a simplified steady state system, as used in our modelling, input should equal to output minus buried and resuspended material. However, if masses of microplastic in sediments per segment are of interest, sediment transport velocities are of high importance which then can easily be modification modified in our model.

Finally, we suggest to use simplified models on country scale until increasing data availability might enhance more advanced modelling approaches. Therefore, more detailed equations and approaches can be implemented in our open source available model. This can be done best by integrating more advanced processes or data for deriving the factors (f_{sed}, f_{acc}, f_{resus}). Also, the model focuses on masses while researchers interested in toxicity might be more interested in 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 (Besseling et al., 2017).

3.8. Outlook, application and future steps

We strongly believe that our model can increase our understanding of microplastic distribution in the environment when analysing large areas of interest such as country or continental scale. A better understanding of the spatial distribution becomes especially important when trying to reduce microplastic pollution in oceans or downstream of emission sites. Through our model we can clearly show as an example that reducing input pollution downstream of the Lake Geneva (i. e. through the city Geneva) would be by far more effective than reducing pollution more upstream (Figure 4). We argue that policymakers could use findings like this

to better validate, rate and understand the effect of different policies.

In our model we aimed for simple input data sets to i) be able to apply the model on the country scale and ii) give other users the opportunity to reuse our model in other areas of interest. With a simple vector shapefile being the most important input file beside of the input emissions, other users could create their own shapefile in a open source geoinformation software (e. g. `QGIS`). Implementing simplified assumptions for sedimentation factors reveals already a basic understanding of the analyzed system. Also, the relative small computational power needed for the model makes it accessible to a broad field of users.

Based on our modelling results we would like to emphasize that more measurement campaigns are needed that contribute to a broad understanding of microplastic transport and distribution in catchments or countries. We should shift our resources from studies just presenting a proof of plastic presence, which is probably given anyways, towards measurements that provide a process understanding. Hence, measurements similar to Faure et al. (2015) who measured upstream and downstream of the city Geneva the Rhône river should be performed with transport processes in mind. Basic improvements would be to measure in alignment with the river current in order to measure the "same water" every time. Generally, considering hydrological parameters during measurement campaigns similar to nutrient measurements in surface waters would be beneficial.

Finally, 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, e. g. flooding events and lake stratification. We clearly show the importance of to better understanding of lake retention and it can be assumed that different depths and shapes of lakes influencing parameters of microplastic retention.

CRediT authorship contribution statement

David Mennekes: Methodology, Investigation, Visualization, Writing -original draft, review and editing.

Bernd Nowack: Methodology, Supervision, Funding acquisition, Writing - review and editing.

Declaration of competing interest

The authors declare that they have no conflict of interest.

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