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MODELLING THE EMISSIONS OF MICRO- AND MACROPLASTICS TO THE ENVIRONMENT

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1 Table of contents

2 Summary

Since plastic can be found in almost any location sampled worldwide, plastic pollution has become a burning topic in public media and scientific research. Plastic can be found in many shapes, colours and sizes in the environment, from microscopic particles to metre-sized litter objects. Plastic has been reported in sediments, soils, water and air, and in both marine and terrestrial environments. Remote locations like deserts, mountains and arctic waters are not an exception. The strong spatial and temporal variations of plastic concentrations depending on the proximity to emission sources and the dynamics of transport processes make quantifying the impact of plastic on ecosystems difficult. Modelling can be used as a complementary approach to measurements to tackle this question from a larger perspective. In this thesis, the goal was to assess the quantities of plastic emitted to the environment while distinguishing two categories of size: pieces smaller than 5 mm called microplastic, and the remaining larger pieces called macroplastic. Different types of polymers were distinguished so that differences in material density and toxicity could be used as parameters for further work. The focus was on Swiss freshwaters and soils, but emissions to air were also included. The general research goal was subdivided into three steps of modelling. First the life-cycle flows of plastics were studied, i.e. the flows through production, manufacturing, use, waste management and recycling. In a second step, the emissions from each life-cycle stage were examined and emissions were quantified based on available data. In a third and last step, the emission flows were regionalized by considering geographical datasets as proxies for the spatial distribution of emission flows, and total emission maps were created.

The life-cycle modelling provided insight into consumption patterns and preferred waste management options for Switzerland and Europe. Seven different polymers were considered, chosen for their popularity and frequency of reporting in environmental samples: low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), expanded polystyrene (EPS), polyvinylchloride (PVC) and polyethylene terephthalate (PET). Probabilistic Material Flow Analysis (PMFA) was used as method to model the life-cycle flows, since it permits to include parameter uncertainty systematically in the model. Each parameter is described as a probability distribution based on the uncertainty attributed to each data from literature. The results obtained are then also probability distributions reflecting the expected confidence in the results. Out of the seven polymers considered, PP was the most consumed in Switzerland at 19.5±2.1 kg/capita/year (mean ± standard deviation), followed by PET at 16.0±1.2 kg/cap/a, HDPE at 14.8±1.7 kg/cap/a, LDPE at 13.5±1.7 kg/cap/a and PVC at 12.4±1.5 kg/cap/a. The least consumed polymers within our scope were PS at 6.74±0.74 kg/cap/a consumed and EPS at 3.8±0.48 kg/cap/a. A very similar behaviour is observed for Europe. Textile

products account for 42±3% of the consumption of PET and 22±4% of PP in Europe. Most of the plastic is incinerated in Switzerland, at 89.1±1.6 % of EPS, 88.2±2.1 % of PS, 84.8±2.7 % of PVC, 78.8±2.2 % of PP, 77.0±3.1 % of HDPE, 76.2±3.1 % of LDPE and 74.3±2.4 % of PET. Large amounts of plastic are as well landfilled in Europe. These results provided a basis for the next modelling step describing the emission pathways but may also be used for further exposure modelling studies, including for example human or environmental exposure to plastic additives.

The emission modelling described the emissions starting from the source process in the life-cycle until the final environmental compartment for Switzerland. Direct emissions flowed directly from the initial source in the life-cycle, and indirect emissions flowed through intermediate compartments representing technical or virtual processes. The emission flows were modelled using PMFA as for the life-cycle flows. The largest environmental emissions were obtained for PET at 200±120 g/cap/a, followed by PP at 126±43 g/cap/a, HDPE at 98±50 g/cap/a, LDPE at 94±34 g/cap/a, PVC at 65±36 g/cap/a, PS at 24±13 g/cap/a, and finally EPS at 16±12 g/cap/a. These figures include emissions to soil, freshwater and air, as macroplastic and microplastic. The majority of these emissions were released to soil as macroplastic. Combining all polymers together, 540±140g/cap/a were modelled to be emitted to soil as macroplastic and 73±14g/cap/a as microplastic, 13.3±4.9g/cap/a to freshwater as macroplastic and 1.8±1.1g/cap as microplastic. Littering of consumer packaging was the leading emission pathway for both soil and freshwater. Other important microplastic emissions to soil were found for construction, agriculture, production and manufacturing, waste management and recycling, although significant uncertainties on the specific emission flows remain. Waste management and recycling, textiles and personal care products were shown to release most of the microplastic to freshwater. This analysis revealed which mitigation policies may have a higher importance and also where the most important data gaps can be found in the life-cycle.

In the final modelling step, the emission flows to soil, water and air were regionalized for Switzerland using 11 geographical datasets on land-use statistics, traffic and population densities, wastewater treatment plants and combined sewer overflows. These geographical datasets were used as proxies to regionalize the 61 final emission flows obtained from the second modelling step. Each of the 61 emission flows was combined with an appropriate geographical dataset to produce high resolution maps of the emission flows in Switzerland. Separate maps were generated for microplastic and macroplastic emissions, for emissions to soil, water and air, and for the mean and quantiles of the modelled emission flows. Most of the emissions are found in

areas with high human activity according to this analysis, which corresponds to the Swiss plateau and the main Alpine valleys. Different patterns could be observed depending on the polymer because of their varying uses and emission flows. The median local emissions of macroplastic to soil ranged from 0.0006 to 0.06 kg/ha/a. More than 50% of the raster cells for microplastic to soil displayed no emission flows at all, but could reach locally much higher values at for example 12.7 kg/ha/a in the case of HDPE. Maps of emissions were created for freshwater as well, including 20167 river segments and 210 lake polygons. Between 0.062 kg/km/a and 1.5 kg/km/a macroplastic were emitted along river segments depending on the polymer and between 0.0025 kg/km/a to 0.11 kg/km/a microplastic.

In total, 134 compartments and 402 flows were implemented in the flow modelling for each polymer considered, which permitted to highlight the most important life-cycle and emission pathways. The regionalization provided the first step towards a fully developed fate model describing the dynamics of the emitted plastic. Further research including a temporal dimension in the emission model would be necessary to predict long-term trends in emissions. The potential accumulation of plastic in environmental sinks could then as well be predicted if the models developed here were linked to a fate model. Relevant fate processes are for example transport in water, runoff from soil to water, sedimentation and resuspension, aggregation of microplastic, degradation and fragmentation of macroplastic into microplastic. This scientific progress will help designing risk assessment and life-cycle assessment frameworks which can then take into account the impact of plastic in different settings.

3 Zusammenfassung

Da Kunststoff an nahezu jedem Ort der Welt zu finden ist, ist die Kunststoffverschmutzung zu einem brennenden Thema sowohl in den öffentlichen Medien als auch in der wissenschaftlichen Forschung geworden. Plastik kann in vielen Formen, Farben und Grössen in der Umwelt gefunden werden, von mikroskopischen Partikeln bis hin zu metergrossen Müllobjekten. Plastik wurde in Sedimenten, Böden, Wasser und Luft sowie in maritimen und terrestrischen Systemen nachgewiesen. Abgelegene Orte wie Wüsten, Berge und arktische Gewässer sind dabei keine Ausnahme. Die starken räumlichen und zeitlichen Schwankungen von Kunststoffkonzentrationen, die von der Nähe zu Emissionsquellen und der Dynamik der Transportprozesse abhängen, erschweren eine Quantifizierung der Auswirkung von Kunststoff auf Ökosysteme. Modellierung kann als komplementärer Ansatz zu Messungen verwendet werden, um diese Frage aus einer grösseren Perspektive anzugehen. Diese Dissertation hat als Ziel, die Menge an freigesetztem Kunststoff zu bestimmen, und unterscheidet dabei zwischen zwei Grössenklassen: Teilchen kleiner als 5 mm sind als Mikroplastik bekannt und grössere Teile als Makroplastik. Es wurden verschiedene Arten von Polymeren unterschieden, so dass materialspezifische Dichte und Toxizität als Parameter in späteren Studien benutzt werden können. Der Fokus lag auf Schweizer Süsswasser und Böden, doch wurde auch die Freisetzungen in die Luft berücksichtigt. Das generelle Forschungsziel wurde in drei Modellierungsschritte unterteilt. Zuerst wurden die Stoffflüsse über den ganzen Kunststofflebenszyklus untersucht, d. h. die Flüsse durch Produktion, Verarbeitung, Verbrauch, Abfallbehandlung und Recycling. In einem zweiten Schritt wurden alle Lebenszyklusprozesse untersucht und die Emissionen in die Umwelt auf Basis der verfügbaren Daten quantifiziert. In einem dritten und letzten Schritt wurden die Emissionsströme unter Berücksichtigung geografischer Datensätze als Proxies für die räumliche Verteilung regionalisiert und Gesamtemissionskarten erstellt.

Die Lebenszyklusmodellierung lieferte Einblicke zum Verbrauch und der bevorzugten Entsorgungsoptionen für die Schweiz und Europa. Es wurden sieben verschiedene Polymere in Betracht gezogen, die aufgrund ihrer Beliebtheit und Häufigkeit ihres Auftretens in Umweltproben ausgewählt wurden: Polyethylen mit niedriger Dichte (LDPE), Polyethylen mit hoher Dichte (HDPE), Polypropylen (PP), Polystyrol (PS), expandiertes Polystyrol (EPS), Polyvinylchlorid (PVC) und Polyethylenterephthalat (PET). Zur Modellierung der Lebenszyklusflüsse wurde die probabilistische Materialflussanalyse (PMFA) angewendet, da sie eine systematische Einbeziehung der Parameterunsicherheit in das Modell ermöglicht. Jeder Parameter wurde als Wahrscheinlichkeitsverteilung beschrieben, basierend auf der Unsicherheit, welcher den jeweiligen Daten aus der Literatur zugeordnet wurde. Die Ergebnisse entsprechen dann Wahrscheinlichkeitsver-

teilungen, die das erwartete Vertrauen in die Ergebnisse widerspiegeln. Von den sieben untersuchten Polymeren wurde PP mit 19.5 ± 2.1 kg/Kopf/Jahr (Mittelwert ± Standardabweichung) am meisten in der Schweiz konsumiert, gefolgt von PET mit 16.0 ± 1.2 kg/Kopf/Jahr, HDPE mit 14.8 ± 1.7 kg/Kopf/Jahr, LDPE bei 13.5 ± 1.7 kg/ Kopf/Jahr und PVC bei 12.4 ± 1.5 kg/Kopf/Jahr. Die am wenigsten verbrauchten Polymere waren PS mit 6.74 ± 0.74 kg/Kopf/Jahr und EPS mit 3.80 ± 0.48 kg/Kopf/Jahr. Ein sehr ähnliches Verhalten wurde für Europa beobachtet. Textilprodukte machen in Europa 42 ± 3% des PET-Verbrauchs und 22 ± 4% des PP-Verbrauchs aus. Der grösste Teil des Kunststoffs wird in der Schweiz verbrannt, mit 89.1 ± 1.6% EPS, 88.2 ± 2.1% PS, 84.8 ± 2.7% PVC, 78.8 ± 2.2% PP, 77.0 ± 3.1% HDPE, 76.2 ± 3.1% LDPE und 74.3 ± 2.4% PET. In Europa werden grosse Mengen Kunststoff auch deponiert. Diese Ergebnisse bildeten eine Grundlage für den nächsten Modellierungsschritt zur Beschreibung der Emissionspfade, können jedoch auch für weitere Studien zur Modellierung der Exposition verwendet werden, z. B. für die Exposition von Menschen oder der Umwelt gegenüber Kunststoffadditiven.

Die Emissionsmodellierung beschreibt die Emissionen während des ganzen Lebenszyklus bis zum endgültigen Umweltkompartiment für die Schweiz. Die Emissionsflüsse wurden wie für die Lebenszyklusflüsse mit PMFA modelliert. Direkte Emissionen fliessen von der ursprünglichen Quelle im Lebenszyklus ohne Umwege in die Umwelt, indirekte Emissionen durch Zwischenkompartimente, die technische oder virtuelle Prozesse darstellen. Die grössten Freisetzungen wurden für PET mit 200 ± 120 g/Kopf/Jahr ermittelt, gefolgt von PP mit 126 ± 43 g/Kopf/Jahr, HDPE mit 98 ± 50 g/Kopf/Jahr und LDPE mit 94 ± 34 g/Kopf/Jahr, PVC mit 65 ± 36 g/Kopf/Jahr, PS mit 24 ± 13 g/Kopf/Jahr und schliesslich EPS mit 16 ± 12 g/Kopf/Jahr. Diese Zahlen umfassen Emissionen in Boden, Süsswasser und Luft sowie in Form von Makroplastik und Mikroplastik. Der Grossteil dieser Emissionen wurde als Makroplastik in den Boden freigesetzt. Wenn alle Polymere gleichzeitig betrachtet sind, wurden 540 ± 140 g/Kopf/Jahr als Makroplastik und 73 ± 14 g/Kopf/Jahr als Mikroplastik in den Boden abgegeben, 13.3 ± 4.9 g/Kopf/Jahr als Makroplastik in das Süsswasser und 1.8 ± 1.1 g/Kopf/Jahr als Mikroplastik. Das Littering von Verbraucherverpackungen war sowohl für Boden als auch für Süsswasser der grösste Freisetzungsweg. Wichtige Mikroplastikemissionen in den Boden wurden für Bau, Landwirtschaft, Produktion und Herstellung, Abfallwirtschaft und Recycling festgestellt, obwohl grosse Unsicherheiten bei den spezifischen Emissionsströmen bestehen. Gemäss dieser Modellierung geben Abfallwirtschaft und Recycling, Textilien und Körperpflegeprodukte den grössten Teil des Mikroplastiks an Süsswasser ab. Diese Analyse kann als Grundlage für Entscheidungen dienen, welche Massnahmen einen höheren Beitrag zur Minderung leisten könnten und wo sich die wichtigsten Datenlücken im Lebenszyklus befinden.

Im letzten Modellierungsschritt wurden die Emissionsströme in Boden, Wasser und Luft mithilfe von 11 geografischen Datensätzen zu Landnutzungsstatistik, Verkehrs- und Bevölkerungsdichte, Abwasserreinigungsanlagen und Regenüberläufen für die Schweiz regionalisiert. Diese geografischen Datensätze wurden als Proxy für die Regionalisierung der 61 endgültigen Emissionsflüsse aus dem zweiten Modellierungsschritt verwendet. Jeder der 61 Emissionsflüsse wurde mit einem entsprechenden geografischen Datensatz kombiniert, um Karten der Emissionsflüsse in der Schweiz zu erstellen. Es wurden separate Karten für Mikroplastik- und Makroplastikemissionen, für Emissionen in Boden, Wasser und Luft sowie für den Mittelwert und die Quantile der modellierten Emissionsflüsse erstellt. Diese Analyse ergab, dass der grösste Teil der Emissionen in Gebieten mit hoher menschlicher Aktivität zu finden ist. Diese Gebiete entsprechen dem Schweizer Mittelland und den Hauptalpentälern. Je nach Polymer können unterschiedliche Muster beobachtet werden, da diese Polymere unterschiedlich eingesetzt werden und unterschiedliche Emissionsströme aufweisen. Die mittleren lokalen Emissionen von Makroplastik auf den Boden lagen zwischen 0.0006 und 0.06 kg/ha/a. Mehr als 50% der Rasterzellen für Mikroplastik und Boden zeigten überhaupt keine Emissionsströme, konnten jedoch lokal hohe Werte erreichen, zum Beispiel bei HDPE bis zu 12.7 kg/ha/a. Emissionskarten wurden auch für Süsswasser erstellt, einschliesslich 20’167 Flussabschnitten und 210 See-Polygonen. Entlang von Flussabschnitten wurden je nach Polymer zwischen 0.062 kg/km/a und 1.5 kg/km/a Makroplastik und zwischen 0.0025 kg/km/a und 0.11 kg/km/a Mikroplastik freigesetzt.

Insgesamt wurden 134 Kompartimente und 402 Flüsse in die Materialflussanalyse für jedes der sieben Polymer implementiert, wodurch die wichtigsten Lebenszyklus- und Emissionspfade hervorgehoben werden konnten. Die Regionalisierung war der erste Schritt zu einem voll entwickelten Schicksalsmodell, welches die Dynamik des emittierten Kunststoffs beschreiben kann. Um langfristige Trends der Emissionen vorherzusagen zu können, wäre die Berücksichtigung der zeitlichen Dimension im Emissionsmodell erforderlich. Wenn die hier entwickelten Modelle mit einem Schicksalsmodell verknüpft werden, kann die Anreicherung von Kunststoff in Umweltsenken ebenfalls vorhergesagt werden. Relevante Schicksalsprozesse sind beispielsweise Transport in Wasser, Erosion von Boden ins Wasser, Sedimentation und Resuspension, Aggregation von Mikroplastik, Abbau und Fragmentierung von Makroplastik in Mikroplastik. Die Kopplung der in diesem Projekt entwickelten Material und Freisetzungsmodelle mit Umweltverhaltensmodellen würde es dann ermöglichen, verbesserte Risikoabschätzungen und Ökobilanzen für Kunststoffverschmutzung zu entwickeln, mit welchen dann die Auswirkungen von Kunststoff in verschiedenen Ökosysteme ermittelt werden könnten.

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5 Introduction

Many materials which are not naturally present in nature have been emitted to ecosystems by humans, thus potentially altering the equilibria present in these ecosystems. Several aspects will influence the magnitude of the impacts of such materials, one of which is their persistence in the environment or their accumulation potential. One such material that is being found ubiquitously in ecosystems worldwide is plastic, which is a material that has continuously increased in popularity over the last decades, among others because of its resistance to biodegradability. In the absence of processes that can degrade and remove plastic from the environment, it is bound to accumulate, and therefore, potentially cause a permanent change in ecosystems.

5.1 A brief history of plastic pollution

Plastic particles were first reported in marine environments in the early 1970s in the North Sea and the Sargasso Sea[1]. Many reports of larger plastic litter on beaches or at the sea surface occurred around a similar time[1]. At the time, it was already suggested that the "increasing production of plastics, combined with present waste-disposal practices, will undoubtedly lead to increases in the concentration of these particles"[2]. Many of these particles had a pellet shape about 0.25 to 0.5 cm in diameter, which was already recognized as an indicator of losses from producers or manufacturers[3]. The presence of larger plastic litter at the time was mostly not considered an environmental problem, but a "nuisance"[4] or an "eyesore"[5]. Plastic pollution in terrestrial contexts took among others the form of open dumpsites, but little information is available to this day[6,7].

An increase in public attention to the marine litter issue followed the discovery of the "Great Garbage Patch" in the early 2000s[1,8], where plastic particles and larger plastic litter accumulate in the oceanic gyres. In 2002, José D.B. Derraik wrote that "the threat of plastics to the marine environment has been ignored for a long time, and its seriousness has been only recently recognised"[5]. The portraying of the garbage patches as an "seventh continent" sparked the public's attention in later years[9]. An increase in attention to the plastic particles was as well noticed, especially since a scientific article[10] coined the term "microplastic" in year 2004 to describe plastic particles (The terminology "microplastic" for plastic particles smaller than 5 mm will be used henceforward, and "macroplastic" will be used to refer to larger plastic litter). As a result, research on plastic pollution intensified, and consequently the reports of plastic in the environment. It is now known that microplastic is present in the environment from the South pole[11] to

the North pole[12]. Macroplastic was reported in the deep sea with a concentration of 17-335 items/km^2 at depths between around 1000-6000 m below the sea surface[13]. A plastic bag was even reported in the Mariana Trench at 10898 m below the sea surface.

Figure 1: Photographs of animals interacting with plastic litter. Image copyrights: *(Left)* Justin Hofman, *(Right)* Neal Guevara and Greenpeace. Used with permission.

The first report of microplastic pollution in freshwater environments occurred in 2005 in California and was published as a non-peer reviewed report[14]. Microplastic in freshwater was increasingly reported in later years[15], both in densely populated areas[16] and remote locations[17]. Extensive research has been performed in the Great lakes in North America[18] and in European rivers[19,20] and lakes[21,22]. Research on microplastic in Asia is as well emerging[23,24]. A few accounts of microplastic in African freshwaters were as well published[25]. Recently, reports of microplastic pollution in soil samples were also published[26–28]. Microplastic was as well reported in air deposition samples, both in densely populated areas[29] to remote locations[30].

The current attention from public and media has initiated movements of citizen science, improving our knowledge on the amount of macroplastic in freshwater environments. The occurrence of macroplastic litter in Switzerland has been documented through two endeavours. The first one was an initiative from an association called Hammerdirt who conducted regular surveys on the shores of Swiss lakes and used the OSPAR protocol to document the litter found[31]. The second initiative produced the Swiss Litter Report[32] where more than 150 citizen scientists picked up trash in 112 locations all over Switzerland on a monthly basis over a year. Macroplastic in terrestrial environments has been documented in urban settings[33,34], along roadsides[34,35] and in rural areas[28,34,35], but the extent of this pollution is not well characterized.

The sheer amount of plastic present in the environment increases the likelihood of encounters between organisms and plastic particles or objects which has been shown to have varying consequences.

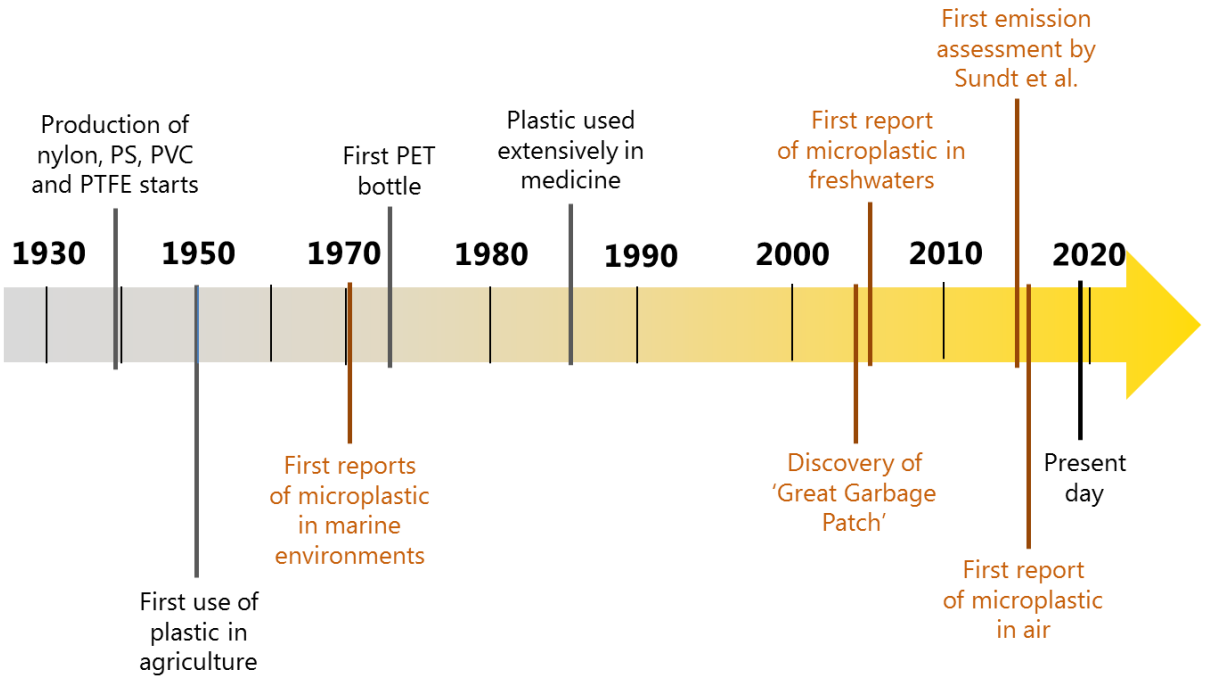

Figure 2: Timeline of important steps in plastic pollution documentation and plastic innovation. Most of the information regarding plastic innovation comes from four scientific articles[36–39]. Acronyms: PS (polystyrene), PVC (polyvinyl chloride), PTFE (polytetrafluoroethylene) and PET (polyethylene terephthalate). The emission assessment referred to is Sundt et al.[40].

5.2 The fish who mistook plastic for food and other hazard tales

Aside from the aesthetical inconvenience of plastic litter and the economic consequences on tourism, there are issues concerning impacts on wildlife and ecosystems. Fish, birds, mammals and reptiles, among others, that had ingested or were entangled in macroplastic have been reported numerous times[5]. The hazard of plastic debris on organisms in marine environments is thought to be primarily due to deleterious effects caused by the ingestion of plastic debris or the entanglement in plastic debris[5]. Some sea birds have been reported to selectively feed on microplastic, which has been linked to a worse physical condition due to poorer feeding and many other marine animals have been found dead with plastic in their guts[5]. Some evidence of endocrine disruption due to chemicals associated with microplastic has been presented, although the source of the chemicals could not be proved[41]. Other adverse effects at the level of ecosystems have also been reported as organisms can travel longer distances when

attached to floating plastic litter[42]. In freshwater, plastic was found in the gut of fish[22] and high concentrations of hazardous chemicals have as well been discovered in plastic litter items along Lake Geneva[43]. Among other heavy metals, 23% of the samples analysed contained lead and 2.5% contained mercury. The impact of this contamination is still unclear but there is also evidence that ingesting plastic can lead to a transfer of some pollutants to internal tissues in marine organisms[41]. The hazard linked to macroplastic in terrestrial environments has been anecdotally documented, as for example in the case of camels in the desert[44] or deer in Japan[45] eating plastic bags.

A large variety of organisms can ingest microplastic: from molluscs to mammals, plants and microorganisms to cite only a few[46]. The environmental and health risks associated with microplastics are not entirely known. A recent review summarized that microplastic of a size of 1-500 mm may cause negative effects on animals, microplastic smaller than 6 µm may have an impact on plants, and microplastic smaller than 100 nm may even be harmful to microbiota[46]. Suggested health effects for microplastics comprise the leakage of plastic additives in organisms, leakage of Persistent Organic Pollutants (POPs) that were taken up in the environment, and effects primarily due to the size of the microparticles[47]. The bioaccumulation of microplastics in biota, and the trophic transfer along the food chain have already been discussed[48]. In smaller organisms, intake of microplastics can lead to a blockage of the digestive tract and a reduced food intake. The role of microplastics as transportation hubs for microorganisms has also been suggested[48].

5.3 The origin of plastic pollution

All of the plastic found in the environment originates from human activities, and with a material as commonly used as plastic, many different sources of environmental emissions are possible and fractional releases from a single process may already lead to large quantities emitted. The characteristics of the plastic collected in the environment can give some insight into the origin of plastic debris.

Different types of microplastic have been documented in the environment. They are most of the time classified as either fragments, fibres, spheres, pellets, lines, sheets, flakes or foam[49]. In essence, these are representative of three types of sources of microplastic. There is first the microplastic that is emitted in the same form as it is used. It can be emitted from activities where plastic is used as small pieces or particles, for example used in personal care and cosmetic products (PCCP) or pre-production pellets used in manufacturing processes. These can

be referred to as "primary microplastics", but considering the differences in nomenclature existing[50], they will be referred to as "manufactured microplastics" in this chapter. Then, there is the microplastic that is emitted from the wear of products during their use phase, for example clothing undergoing washing and thus releasing fibres in the wastewater, or products exposed to environmental conditions outdoors and thereby releasing fragments. These microplastics will be referred to as "wear microplastics" in this chapter. The last type of source of microplastic occurs when macroplastic already present in the environment, fragments into smaller pieces. Macroplastic can be lost to the environment by accidental spillage and inappropriate disposal and will fragment into microplastic at different rates depending on the environmental conditions[51]. This last category will be referred to as "degradation microplastics" in this chapter. Most of the microplastic identified in the environment belongs in the category corresponding to degradation microplastics as fragments, film and foam[15] but wear microplastic is also very common with fibres released from textiles. Fewer manufactured microplastics are found in environmental samples overall[15]. This indicates that the degradation of macroplastic is a very important contributor to microplastic burdens, but also that emissions have multiple origins.

Figure 3: Examples of litter pieces which can be found in the environment with different likelihoods. All pictures were taken by the author or by Maciej Kawecki-Wenger. Likeliest attribution: A (packaging), B (household item), C (microplastic), D (fishing net or technical textile), E (bicycle), F (drone), G (EPS microplastic around a renovation site), H (technical textile), I (litter consisting mostly of packaging), J (plastic bag or film), K (tampon applicator), L (microplastic originating from the sawing of pipes in a residential area), M (shotgun wad), N (cotton swab), O (EPS fragment), P (vinyl disk).

When macroplastic is identified in the environment, it becomes clear that some products are more recurrent than others, indicating that some emission sources may be more significant. A huge variety of litter items can be found in the environment by an attentive observer (Figure 3). Plastic packaging and films are among the most common, but more exotic types of plastic litter can occasionally be found, such as warm cushions to heat one's fingers during winter (Figure 3B) or vinyl disks (Figure 3P). Single-use plastics were the most abundant items during monthly visual observations in the Rhone river in France[52]. The data from the Swiss Litter Report suggests that a large fraction of the macroplastic litter found on shores and riversides in Switzerland originate from take away consumption, industry, construction or demolition sites and personal hygiene products[32]. And according to a report from Hammerdirt, expanded polystyrene, cigarette butts, plastic fragments, snack wrappers, cotton swabs and plastic sheeting and

films are the six most commonly found types of litter around Lake Geneva[31]. The composition of macroplastic at the bottom of lakes and rivers is likely to be different in composition, as is the case in marine environments[53]. In a study on the littering behaviour of people in different contexts, the composition of pre-existing litter in soil environments was examined[34]. Of the 130 sites observed, only two had no litter visible at the beginning of the study. The most frequently observed litter items in that study were cigarette butts (82% of the sites) and paper (67%). Almost half as many sites had plastic litter visible (45% of sites had food wrappers and 33% miscellaneous plastic). Most of the plastic reported corresponded to packaging. In a littering study conducted in 5 cities in Switzerland, 18-84% of the littered objects originated from on-the-go consumption, i.e. beverage packaging and takeaway[33]. In a litter study conducted in the US, litter samples were collected along roads, interchanges and in public areas in 1998-1999[35]. Beverage containers represented the largest mass of the litter in most site types with 14.6-31.4%. Other types of packaging were as well an important fraction, with takeaway representing 1.9-11.3% across all types of sites, other food and beverage packaging representing 1.0-2.7% and non-food packaging comprising 1.1-3.8%. It has also been observed that the composition of plastic debris roughly follows the distribution of plastics used for packaging[54,55]. In addition, automotive pieces represented 3.6-17.2% of the total litter mass collected, which was suggested to be connected to dumping. A surprising amount of tires was as well found along highways, at 24.7% of the total mass of the litter.

All of these items and particles originate from human emissions and may originate from different stages in the plastic life-cycle. A description of some characteristics of the flows of plastic through society will help having an overall picture of the plastic flows.

5.4 The greatest plastic flow on Earth

It is often said that the same properties that make the use of plastics so appealing to use, are the reason that plastic can be found everywhere in the environment[5]. Long-range transport is enabled by their low density and extended residence times in the environment are possible because of their very limited biodegradability. Both of these properties and countless others make plastic a very attractive material for innovation and modern lifestyles. The low density of plastics makes them interesting for packaging purposes, and permits to reduce energy consumption for both transportation and manufacturing[36,56]. Their resistance to biodegradability makes them also very convenient for all kinds of weathering exposed constructions or for packaging. For instance, window frames built from PVC require much less maintenance than wooden window frames and plastic packaging of food enables to reduce food waste by up to

4% to 30% depending on the product[57]. In Chinese agriculture, the use of plastic mulch films increased the crop yield by around 30% while reducing water use by at least 30%[58]. The European plastic production has increased from 0.35 million metric tonnes (Mt) in 1950[59] to 64.4 Mt in 2018[60] (Figure 4A) in an ever increasing range of applications. Around 7800 Mt of plastic are estimated to have been manufactured globally from year 1950 to year 2015[61]. Many desired material properties can be obtained by choosing different polymers and tuning their properties with additives. Plastics are used in packaging (39.7%), building and construction (19.8%), automotive (10.1%), electrical and electronic (6.2%), household, leisure and sports (4.1%) and agriculture (3.4%) in Europe[60] (Figure 4B).

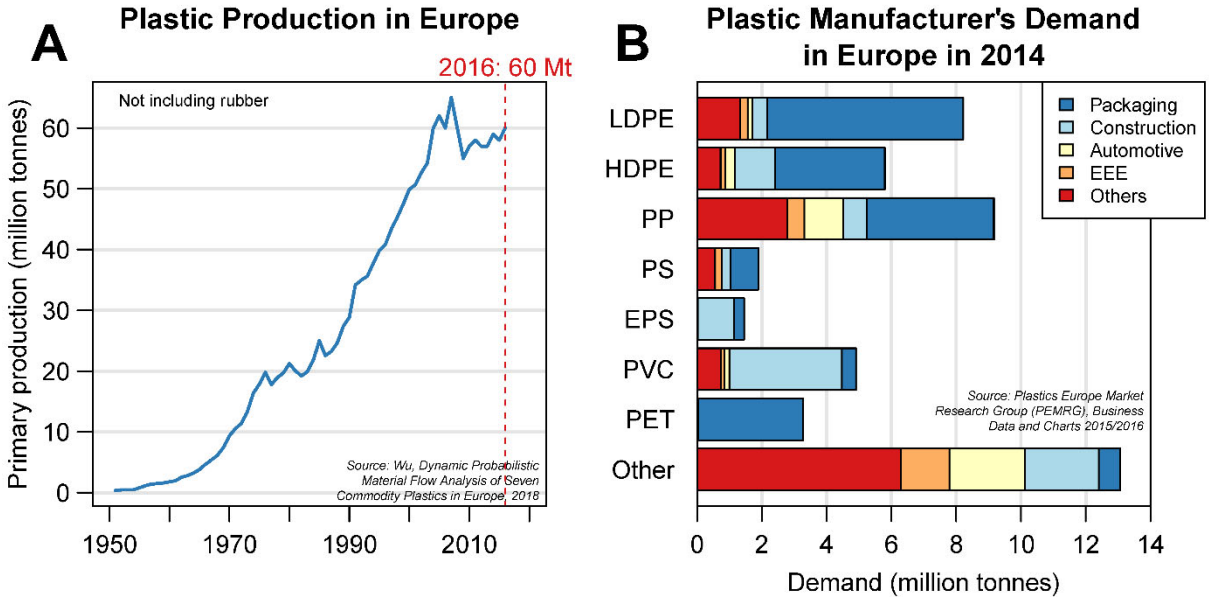

Figure 4: Plastic market evolution and most important polymers and uses in Europe, excluding fibre and textile applications.

In addition to the previously mentioned figures, plastic is used extensively in fibre and textile applications, and synthetic fibres have gained a lot of importance in recent years. The global production of polyester has increased from 2 Mt in 1980 to 31 Mt in 2015[62]. Man-made fibre production had already surpassed natural fibre production around year 2000[62]. The European textile market is largely dominated by clothing, followed by household textiles and technical textiles[63]. When including fibre production, global plastic production estimates rise to about 388 Mt for 2015[64].

The plastic and synthetic textiles consumption is expected to continue to increase worldwide with the population growth and increased access to modern technologies and lifestyles[56,65]. The increase in macroplastic occurrence in oceans between 1957 and 2016 has been linked to the increase in global plastic production[66]. It is becoming urgent to assess the extent of this pollution and its impact on ecosystems.

5.5 The plastic-haunted world: Modelling as an environmental exposure assessment tool

In order to quantify the risk posed by plastic in the environment, in-depth understanding of the extent and characteristics of plastic pollution needs to be gained. The extent of this contamination has been measured numerous times in diverse environments. Microplastic concentrations in freshwater can vary by 10 orders of magnitude depending on the proximity to emission sources and other parameters[15]. Microplastic concentrations have been shown to increase after a rain event[29,67], due to wind patterns[68] and increased vertical mixing[69], or because of converging water currents[18]. Sampling on different river banks can also yield different concentrations[20]. During samplings on beaches and river banks[32], more macro-litter was found around lakes at 123 items/100m^2 than along rivers at 38 items/100m^2 (including non-plastic materials). More litter can be found in freshwater close to urban areas at 103 items/100m^2 than in rural areas at 53 items/100m^2, but the density also varied by a factor of 2 depending on the season.

This plastic pollution is made up of different synthetic materials. Floating macroplastic has been reported to mainly consist of polypropylene (PP) (35.2% ± 21.6%), polyethylene (PE) (26.0% ± 14.6%), polyethylene terephthalate (PET) (20.7% ± 20.1%) and polystyrene (PS) (10.8% ± 12.3%) in one study on the Seine River[54], but depends on the location and time of sampling. PVC is less often measured in the environment, even though its presence has also been reported[55]. In a literature review on microplastic and macroplastic in freshwater and marine environments, PE, PP and PS were the most common polymers on beaches, at the water surface and in sediments[70]. Low density polymers dominate the water surface in marine environments, whereas denser polymers are prevalent at the sea bottom[71]. A similar behaviour can be expected in freshwater environments as well and could contribute to the low occurrence of PVC litter in freshwater surface and beach samples.

Fewer studies on the issue of plastic in terrestrial environments are available. An average concentration of 0.34±0.36 particles per kilogram of dry soil of microplastic was found in agricul-

tural soil in Germany[28]. Microplastic was found in 26 of 29 Swiss floodplain soils sites at concentrations of up to 55.5 mg/kg, including remote locations[27]. Soil that had undergone 5 subsequent applications of sludge in Chile had a median of 3.5 particles per gram of dry soil with a majority of fibres[72]. In the US litter study[35], 23g of litter were found to be generated along highways per 1000 km driven per year and 54g/1000km driven/y along inter-regional routes. In recreational areas 527 kg per hectare of high use area per year of litter was generated, and 67 kg/ha of high use area in state parks. High use areas corresponded to parking lots, camping grounds and trailheads, the whole parks were not inspected. In agricultural soil in Germany, around 0.066 kg/ha of plastic was documented where no mulching and greenhouses are used[28]. In Chinese agricultural soils with intense plastic mulch use, up to 50-260 kg/ha of residue can be found[58]. Around 6-35 plastic bags/km^2 and 39-63 balloon clusters/km^2 were found in the Sonoran Desert in the USA[73]. The balloon cluster density exceeded the density of local desert tortoises and rattlesnakes.

Microplastic air deposition was documented to be 2-355 particles/m^2/day around Paris in France[74], on average 275 particles/m^2/day around Hamburg in Germany or 365±69 particles/m^2/d in the French Pyreenees[30], and might be influenced by precipitation[74] and wind patterns[30]. The influence of the population density on the total burden remains unclear because of conflicting results[75].

Because of the high spatial and temporal variability in plastic concentrations in freshwater, soil and air, it can be difficult to obtain a large scale view on the extent of the pollution through measurements alone. A complementing approach can be obtained using modelling to obtain larger scale information. One such approach is to first model the emissions of plastic from anthropogenic activities and then to model the physical and chemical processes occurring to the emitted plastic in different environmental compartments.

An increasing amount of modelling studies aiming at describing the export of plastic to marine environments have been published in recent years[76–79]. Jambeck et al. predicted macroplastic emissions of about 4.8 to 12.7 Mt/y from populations living within 50 km of the coastline to the ocean[76]. These predictions were based on mismanaged waste estimates according to a country by country economic classification[80] and an assumption of 2% littering. This study estimated that the largest emitters of macroplastic to oceans are China and South-East Asia. More recently, Lebreton et al. estimated that 1.15-2.41 Mt/y of macroplastic enters the oceans from rivers, with the 20 most polluting rivers accounting for 67% of the total released plastic[79].

Siegfried et al.[77] quantified microplastic inputs into European rivers and the corresponding release to the sea from four types of sources: personal care products, household dust, washing of textiles and tyre and road wear particles. They estimated an export of microplastic from rivers to sea of between 0 and 192 kg per km^2 of drainage basin per year for the year 2000. Van Wijnen et al. built on this model to develop a global model for the export of microplastic from rivers to the oceans including the three types of sources of microplastic[78]. According to these results, macroplastic is the first source of microplastic in all regions, with tyre wear and textiles washing making up most of the remaining emissions. They estimated that around 47000 t/y of microplastic were exported to the oceans globally and also concluded that region specific analyses are required to account for the differing expected changes in waste management and sanitation[78]. In a more recent global plastic release study, a total release of 9.2 Mt was modelled for year 2015, of which 67% corresponded to macroplastic and 33% to microplastic[64].

The first regional assessments of microplastic emissions were conducted by several national environmental agencies and revolved mostly around personal care products, washing of textiles and tyre wear, although many other smaller flows were as well included[40,81–83]. Macroplastic emissions were mostly not included in these studies because of the scarcity of data on the mismanagement of waste and the fragmentation rate of macroplastic into microplastic. These studies nevertheless provided first estimates of some emission flows which are still used in more recent peer-reviewed publications.

First fate models for microplastic have already been published. A combined input and fate model was developed to describe the emissions and whereabouts of tyre and road wear particles in the Seine watershed in France[84]. Around 1.8 kg/capita/year of particles were released in this watershed, with 18% released to freshwater and 2% finally released to marine water through the estuary. Besseling et al. also developed their own hydrological fate model for microplastic in a river including processes such as fluvial transport, sedimentation and resuspension, aggregation, degradation and burial[85]. Alternative approaches were also developed to model the export of macroplastic to the sea by using data from booms collecting litter along a river[86]. Within their parametrization, differences in polymer density did not affect the fate of the particles significantly, but differences in particle size were dominant.

These two types of modelling can provide Predicted Environmental Concentrations (PEC), which are essential for estimating the exposure of ecosystems to plastic. A last modelling step could then assess the uptake and bioaccumulation of plastic in organisms depending on ambient concentrations.

5.6 Research needs for people in a hurry

While a vast amount of research has already been performed regarding the extent of plastic pollution (34862 documents on the Scopus database with the search "(TITLE-ABS-KEY (micro-plastic) OR TITLE-ABS-KEY (plastic)) AND (TITLE-ABS-KEY (environment) OR TITLE-ABS-KEY (pollution))" as of the 28th of October 2019), the impact of plastic on ecosystems is still unclear[87–89]. Existing risk assessments predict no to low risk in marine and freshwater environments[90,91], but spatial variability may cause regions to present a higher risk. A deeper understanding of the spatial distribution of emissions and the dynamics of fate processes will help assessing the impact of plastic on all ecosystems. Few approaches have considered microplastic and macroplastic simultaneously, although the two issues are closely linked. In order to estimate the quantity of microplastic released in the environment, the emissions of macroplastic and its fate also need to be taken into account.

Moreover, more insight into the composition of plastic pollution in the different environments is necessary, as it may influence the toxicity and the fate of the plastic. Since different polymers have distinct applications of choice[92], different emission patterns can be expected. Secondly, the density of the polymers may influence the transport of plastic in aquatic environments[70,93]. Last, differences in toxicity might be expected either for the different polymers themselves[91,94] or the additives included in the polymer matrix during manufacturing which may be measured from environmental samples[43]. For all these reasons, a distinction between polymers is necessary for exposure assessments.

In this thesis, we focus on emission modelling to contribute to assessing the exposure to individual polymers in freshwater and terrestrial environments. In order to document the emission flows of microplastic and macroplastic in a systematic and comparable manner, the method called Material Flow Analysis (MFA) is used. Each emission stage is modelled individually and the pathways until the receiving environmental compartments are described. The MFA tool used for this thesis is called Probabilistic MFA (PMFA), and uses probability distributions for all parameters as a means to take into account data uncertainty and propagate it to the final results.

5.7 The grand design[‡]

The goal of this thesis is to provide a first step to calculating PECs for plastic in Switzerland by modelling the emissions pathways of plastic, and thus contribute to the exposure assessment of macroplastic and microplastic in the environment.

The research questions of the thesis are:

1. What are the life-cycle flows of chosen polymers in Europe and Switzerland?
2. What are the environmental flows of macroplastic and microplastic in Switzerland for the same polymers?
3. What is the spatial distribution of these emissions?

Given the above research questions, the goals of the project are to:

1. Quantify the life-cycle of chosen polymers materials in Europe and Switzerland,
2. Quantify the losses of macroplastic and microplastic to the environment from all life-cycle stages for Switzerland,
3. Regionalize the model for Switzerland.

For this thesis, Probabilistic Material Flow Analysis (PMFA) was used to predict polymer flows. The proposed plastics were polyethylene (PE), polyethylene terephthalate (PET), polystyrene (PS), polyvinyl chloride (PVC) and polypropylene (PP), following the pattern of some of the most commonly measured plastics in the environment as aforementioned[54].

5.8 The first three publications

The following publications are included in this thesis:

1. Kawecki, D.; Scheeder, P.; Nowack, B. Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe. Environ. Sci. Technol. 2018, 52, 9874–9888.
2. Kawecki, D.; Nowack, B. Polymer-specific modelling of the environmental emissions of seven commodity plastics as macro- and microplastics. Environ. Sci. Technol. 2019, 53, 9664-9676.

[‡] It is probably noteworthy at this point to mention that all the titles in this chapter refer to popular or historically important science books, and that to be consistent with this so-called Easter egg, some titles needed to sound more presumptuous than initially planned. Although, in the defence of this title, one could say that every researcher should believe in their research plan, or if that is not possible, make a better one.

3. Kawecki, D.; Nowack, B. A proxy-based approach to predict spatially resolved emissions of macro- and microplastic to the environment. Manuscript ready for submission.

The supporting information of these three papers is presented as annex.

5.9 References

(1) Ryan, P. G. A Brief History of Marine Litter Research. In *Marine Anthropogenic Litter*; Bergmann, M., Gutow, L., Klages, M., Eds.; Springer, 2015; pp 57–72.

(2) Carpenter, E. J.; Smith Jr, K. L. Plastics on the Sargasso Sea Surface. *Science (80-.).* **1972**, *175* (4027), 1240.

(3) Carpenter, E. J. Polystyrene Spherules in Coastal Waters. *Science (80-.).* **1972**, *178* (4062), 749–750.

(4) Bakker, P. J.; Frieling, T. Environmental Problems of Plastics. [MILIEU-ASPECTEN VAN KUNSTSTOFFEN.]. *Plastica* **1972**, *25* (12), 572–579.

(5) Derraik, J. G. . The Pollution of the Marine Environment by Plastic Debris: A Review. *Mar. Pollut. Bull.* **2002**, *44* (9), 842–852.

(6) Agence de l'Environnement et de la Maîtrise de l'Énergie ADEME. *La Loi Sur Les Déchets a 40 Ans !*; 2015.

(7) Bauen, A. *Erhebung Verwertung Deponiegas Über Fackelanlagen in Der Schweiz 1990 Bis 2014*; 2016.

(8) Moore, C. J.; Moore, S. L.; Leecaster, M. K.; Weisberg, S. B. A Comparison of Plastic and Plankton in the North Pacific Central Gyre. *Mar. Pollut. Bull.* **2001**, *42* (12), 1297–1300.

(9) Veolia Group. The "the seventh continent of plastic" https://www.livingcircular.veolia.com/en/eco-citizen/seventh-continent-plastic (accessed Oct 14, 2019).

(10) Thompson, R. C.; Olsen, Y.; Mitchell, R. P.; Davis, A.; Rowland, S. J.; John, A. W. G.; McGonigle, D.; Russell, A. E. Lost at Sea: Where Is All the Plastic? *Science* **2004**, *304* (5672), 838.

(11) Cincinelli, A.; Scopetani, C.; Chelazzi, D.; Lombardini, E.; Martellini, T.; Katsoyiannis, A.; Fossi, M. C.; Corsolini, S. Microplastic in the Surface Waters of the Ross Sea (Antarctica): Occurrence, Distribution and Characterization by FTIR. *Chemosphere* **2017**, *175*, 391–400.

(12) Peeken, I.; Primpke, S.; Beyer, B.; Gütermann, J.; Katlein, C.; Krumpen, T.; Bergmann, M.; Hehemann, L.; Gerdts, G. Arctic Sea Ice Is an Important Temporal Sink and Means of Transport for Microplastic. *Nat. Commun.* **2018**, *9* (1), 1505.

(13) Chiba, S.; Saito, H.; Fletcher, R.; Yogi, T.; Kayo, M.; Miyagi, S.; Ogido, M.; Fujikura, K. Human Footprint in the Abyss: 30 Year Records of Deep-Sea Plastic Debris. *Mar. Policy* **2018**, No. March, 0–1.

(14) Wagner, M.; Scherer, C.; Alvarez-Muñoz, D.; Brennholt, N.; Bourrain, X.; Buchinger, S.; Fries, E.; Grosbois, C.; Klasmeier, J.; Marti, T.; et al. Microplastics in Freshwater Ecosystems: What We Know and What We Need to Know. *Environ. Sci. Eur.* **2014**, *26* (1), 12.

(15) Koelmans, A. A.; Mohamed Nor, N. H.; Hermsen, E.; Kooi, M.; Mintenig, S. M.; De France, J. Microplastics in Freshwaters and Drinking Water: Critical Review and Assessment of Data Quality. *Water Res.* **2019**, *155*, 410–422.

(16) Morritt, D.; Stefanoudis, P. V.; Pearce, D.; Crimmen, O. A.; Clark, P. F. Plastic in the Thames: A River Runs through It. *Mar. Pollut. Bull.* **2014**, *78* (1–2), 196–200.

(17) Free, C. M.; Jensen, O. P.; Mason, S. A.; Eriksen, M.; Williamson, N. J.; Boldgiv, B. High-Levels of Microplastic Pollution in a Large, Remote, Mountain Lake. *Mar. Pollut. Bull.* **2014**, *85* (1), 156–163.

(18) Eriksen, M.; Mason, S.; Wilson, S.; Box, C.; Zellers, A.; Edwards, W.; Farley, H.; Amato, S. Microplastic Pollution in the Surface Waters of the Laurentian Great Lakes. *Mar. Pollut. Bull.* **2013**, *77* (1–2), 177–182.

(19) Lechner, A.; Keckeis, H.; Lumesberger-Loisl, F.; Zens, B.; Krusch, R.; Tritthart, M.; Glas, M.; Schludermann, E. The Danube so Colourful: A Potpourri of Plastic Litter Outnumbers Fish Larvae in Europe's Second Largest River. *Environ. Pollut.* **2014**, *188*, 177–181.

(20) Mani, T.; Hauk, A.; Walter, U.; Burkhardt-Holm, P. Microplastics Profile along the Rhine River. *Sci. Rep.* **2016**, *5* (1), 17988.

(21) Sighicelli, M.; Pietrelli, L.; Lecce, F.; Iannilli, V.; Falconieri, M.; Coscia, L.; Di Vito, S.; Nuglio, S.; Zampetti, G. Microplastic Pollution in the Surface Waters of Italian Subalpine Lakes. *Environ. Pollut.* **2018**, *236*, 645–651.

(22) Faure, F.; Demars, C.; Wieser, O.; Kunz, M.; De Alencastro, L. F. Plastic Pollution in Swiss Surface Waters: Nature and Concentrations, Interaction with Pollutants. *Environ. Chem.* **2015**, *12* (5), 582–591.

(23) Wang, W.; Ndungu, A. W.; Li, Z.; Wang, J. Microplastics Pollution in Inland Freshwaters of China: A Case Study in Urban Surface Waters of Wuhan, China. *Sci. Total Environ.* **2017**, *575*, 1369–1374.

(24) Xiong, X.; Zhang, K.; Chen, X.; Shi, H.; Luo, Z.; Wu, C. Sources and Distribution of Microplastics in China's Largest Inland Lake – Qinghai Lake. *Environ. Pollut.* **2018**, *235*, 899–906.

(25) Biginagwa, F. J.; Mayoma, B. S.; Shashoua, Y.; Syberg, K.; Khan, F. R. First Evidence of Microplastics in the African Great Lakes: Recovery from Lake Victoria Nile Perch and Nile

Tilapia. *J. Great Lakes Res.* **2016**, *42* (1), 146–149.

(26) Hurley, R. R.; Nizzetto, L. Fate and Occurrence of Micro(Nano)Plastics in Soils: Knowledge Gaps and Possible Risks. *Curr. Opin. Environ. Sci. Heal.* **2018**, *1*, 6–11.

(27) Scheurer, M.; Bigalke, M. Microplastics in Swiss Floodplain Soils. *Environ. Sci. Technol.* **2018**, *52* (6), 3591–3598.

(28) Piehl, S.; Leibner, A.; Löder, M. G. J.; Dris, R.; Bogner, C.; Laforsch, C. Identification and Quantification of Macro- and Microplastics on an Agricultural Farmland. *Sci. Rep.* **2018**, *8* (1), 17950.

(29) Dris, R.; Gasperi, J.; Rocher, V.; Saad, M.; Renault, N.; Tassin, B. Microplastic Contamination in an Urban Area: A Case Study in Greater Paris. *Environ. Chem.* **2015**, *12* (5), 592–599.

(30) Allen, S.; Allen, D.; Phoenix, V. R.; Le Roux, G.; Durántez Jiménez, P.; Simonneau, A.; Binet, S.; Galop, D. Atmospheric Transport and Deposition of Microplastics in a Remote Mountain Catchment. *Nat. Geosci.* **2019**, *12* (May).

(31) Erismann, R.; Erismann, S. *In(Beach-Litter-Density): A Comparison of Marine and Freshwater Litter Densities*; 2016.

(32) Blarer, P.; Kull, G. *Swiss Litter Report*; 2018.

(33) Heeb, J.; Ableidinger, M.; Berger, T.; Hoffelner, W. *Littering – Ein Schweizer Problem ? Eine Vergleichsstudie Schweiz-Europa*.

(34) Schultz, P. W.; Bator, R. J.; Large, L. B.; Bruni, C. M.; Tabanico, J. J.; Brown Large, L.; Bruni, C. M.; Tabanico, J. J. Littering in Context. *Environ. Behav.* **2013**, *45* (1), 35–59.

(35) Washington State Department of Ecology. *Washington State Litter Study Volume Two – Litter Generation and Composition Report*; 2000.

(36) Andrady, A. L.; Neal, M. A. Applications and Societal Benefits of Plastics. *Philos. Trans. R. Soc. Lond. B. Biol. Sci.* **2009**, *364* (1526), 1977–1984.

(37) Zalasiewicz, J.; Waters, C. N.; Ivar do Sul, J. A.; Corcoran, P. L.; Barnosky, A. D.; Cearreta, A.; Edgeworth, M.; Gałuszka, A.; Jeandel, C.; Leinfelder, R.; et al. The Geological Cycle of Plastics and Their Use as a Stratigraphic Indicator of the Anthropocene. *Anthropocene* **2015**, *13*, 4–17.

(38) Kasirajan, S.; Ngouajio, M. Polyethylene and Biodegradable Mulches for Agricultural Applications: A Review. *Agron. Sustain. Dev.* **2012**, *32* (2), 501–529.

(39) Hodges, S. Hospitals as Factories of Medical Garbage. *Anthropol. Med.* **2017**, *24* (3), 319–333.

(40) Sundt, P.; Schulze, P.-E.; Syversen, F. *Sources of Microplastic Pollution to the Marine Environment*; 2014.

(41) Gallo, F.; Fossi, C.; Weber, R.; Santillo, D.; Sousa, J.; Ingram, I.; Nadal, A.; Romano, D. Marine Litter Plastics and Microplastics and Their Toxic Chemicals Components: The Need for Urgent Preventive Measures. *Environ. Sci. Eur.* **2018**, *30* (1).

(42) Kiessling, T.; Gutow, L.; Thiel, M. Marine Litter as Habitat and Dispersal Vector. In *Marine Anthropogenic Litter*; Bergmann, M., Gutow, L., Klages, M., Eds.; 2015.

(43) Filella, M.; Turner, A. Observational Study Unveils the Extensive Presence of Hazardous Elements in Beached Plastics from Lake Geneva. *Front. Environ. Sci.* **2018**, *6* (February), 1–8.

(44) Plastic Soup Foundation. Camels Continue to Die of Plastic in the Desert https://www.plasticsoupfoundation.org/en/2018/01/camels-continue-to-die-of-plastic-in-the-dessert/ (accessed Sep 23, 2019).

(45) BBC News. Nine Nara deer in Japan die after eating plastic bags https://www.bbc.com/news/world-asia-48941494 (accessed Sep 26, 2019).

(46) Zhang, S.; Wang, J.; Liu, X.; Qu, F.; Wang, X.; Wang, X.; Li, Y.; Sun, Y. Microplastics in the Environment: A Review of Analytical Methods, Distribution, and Biological Effects. *TrAC - Trends Anal. Chem.* **2019**, *111*, 62–72.

(47) Eerkes-Medrano, D.; Thompson, R. C.; Aldridge, D. C. Microplastics in Freshwater Systems: A Review of the Emerging Threats, Identification of Knowledge Gaps and Prioritisation of Research Needs. *Water Res.* **2015**, *75*, 63–82.

(48) Wright, S. L.; Thompson, R. C.; Galloway, T. S. The Physical Impacts of Microplastics on Marine Organisms: A Review. *Environ. Pollut.* **2013**, *178*, 483–492.

(49) Kooi, M.; Besseling, E.; Kroeze, C.; van Wenzel, A. P.; Koelmans, A. A. Modeling the Fate and Transport of Plastic Debris in Freshwaters: Review and Guidance. In *Freshwater Microplastics*; Wagner, M., Lambert, S., Eds.; The Handbook of Environmental Chemistry; Springer International Publishing: Cham, 2018; Vol. 58.

(50) Hartmann, N. B.; Hüffer, T.; Thompson, R. C.; Hassellöv, M.; Verschoor, A.; Daugaard, A. E.; Rist, S.; Karlsson, T.; Brennholt, N.; Cole, M.; et al. Are We Speaking the Same Language? Recommendations for a Definition and Categorization Framework for Plastic Debris. *Environ. Sci. Technol.* **2019**, *53* (3), 1039–1047.

(51) Gewert, B.; Plassmann, M. M.; MacLeod, M. Pathways for Degradation of Plastic Polymers Floating in the Marine Environment. *Environ. Sci. Process. Impacts* **2015**, *17* (9), 1513–1521.

(52) Castro-Jiménez, J.; González-Fernández, D.; Fornier, M.; Schmidt, N.; Sempéré, R. Macro-Litter in Surface Waters from the Rhone River: Plastic Pollution and Flows to the NW Mediterranean Sea. *Mar. Pollut. Bull.* **2019**, *146* (May), 60–66.

(53) Erni-Cassola, G.; Zadjelovic, V.; Gibson, M. I.; Christie-Oleza, J. A. Distribution of Plastic

Polymer Types in the Marine Environment; A Meta-Analysis. *J. Hazard. Mater.* **2019**, *369* (February), 691–698.

(54) Gasperi, J.; Dris, R.; Bonin, T.; Rocher, V.; Tassin, B. Assessment of Floating Plastic Debris in Surface Water along the Seine River. *Environ. Pollut.* **2014**, *195*, 163–166.

(55) Sadri, S. S.; Thompson, R. C. On the Quantity and Composition of Floating Plastic Debris Entering and Leaving the Tamar Estuary, Southwest England. *Mar. Pollut. Bull.* **2014**, *81* (1), 55–60.

(56) World Economic Forum; Ellen MacArthur Foundation; Company, M. &. *The New Plastics Economy: Rethinking the Future of Plastics*; 2016.

(57) Aspalter, K.; Denkstatt GmbH. *How Packaging Contributes to Food Waste Prevention*; 2014.

(58) Liu, E. K.; He, W. Q.; Yan, C. R. 'White Revolution' to 'White Pollution'—Agricultural Plastic Film Mulch in China. *Environ. Res. Lett.* **2014**, *9* (9), 091001.

(59) PlasticsEurope. *Plastics – the Facts 2013: An Analysis of European Latest Plastics Production, Demand and Waste Data*; 2013.

(60) PlasticsEurope. *Plastics – The Facts 2018*; 2018.

(61) Geyer, R.; Jambeck, J. R.; Law, K. L. Production, Use, and Fate of All Plastics Ever Made. *Sci. Adv.* **2017**, *3* (7), 25–29.

(62) Qin, M. Y. Global Fibres Overview. *Synth. Fibres Raw Mater. Comm. Meet. APIC 2014* **2014**, *2014* (May), 27.

(63) Beton, A.; Dias, D.; Farrant, L.; Gibon, T.; Le Guern, Y.; Desaxce, M.; Perwuelzz, A.; Cordella, M.; Dodd, N. *Environmental Improvement Potential of Textiles (IMPRO Textiles)*; 2014.

(64) Ryberg, M. W.; Hauschild, M. Z.; Wang, F.; Averous-Monnery, S.; Laurent, A. Global Environmental Losses of Plastics across Their Value Chains. *Resour. Conserv. Recycl.* **2019**, *151* (March), 104459.

(65) Ellen Mac Arthur Foundation. *A New Textiles Economy: Redesigning Fashion' Future*; 2017.

(66) Ostle, C.; Thompson, R. C.; Broughton, D.; Gregory, L.; Wootton, M.; Johns, D. G. The Rise in Ocean Plastics Evidenced from a 60-Year Time Series. *Nat. Commun.* **2019**, *10* (1), 8–13.

(67) Fischer, E. K.; Paglialonga, L.; Czech, E.; Tamminga, M. Microplastic Pollution in Lakes and Lake Shoreline Sediments - A Case Study on Lake Bolsena and Lake Chiusi (Central Italy). *Environ. Pollut.* **2016**, *213*, 648–657.

(68) Browne, M. A.; Galloway, T. S.; Thompson, R. C. Spatial Patterns of Plastic Debris along Estuarine Shorelines. *Environ. Sci. Technol.* **2010**, *44* (9), 3404–3409.

(69) Goldstein, M. C.; Titmus, A. J.; Ford, M. Scales of Spatial Heterogeneity of Plastic Marine Debris in the Northeast Pacific Ocean. *PLoS One* **2013**, *8* (11).

(70) Schwarz, A. E.; Ligthart, T. N.; Boukris, E.; van Harmelen, T. Sources, Transport, and Accumulation of Different Types of Plastic Litter in Aquatic Environments: A Review Study. *Mar. Pollut. Bull.* **2019**, *143* (March), 92–100.

(71) Erni-Cassola, G.; Zadjelovic, V.; Gibson, M. I.; Christie-Oleza, J. A. Distribution of Plastic Polymer Types in the Marine Environment; A Meta-Analysis. *J. Hazard. Mater.* **2019**, *369*, 691–698.

(72) Corradini, F.; Meza, P.; Eguiluz, R.; Casado, F.; Huerta-Lwanga, E.; Geissen, V. Evidence of Microplastic Accumulation in Agricultural Soils from Sewage Sludge Disposal. *Sci. Total Environ.* **2019**, *671*, 411–420.

(73) Zylstra, E. R. Accumulation of Wind-Dispersed Trash in Desert Environments. *J. Arid Environ.* **2013**, *89*, 13–15.

(74) Dris, R.; Gasperi, J.; Saad, M.; Mirande, C.; Tassin, B. Synthetic Fibers in Atmospheric Fallout: A Source of Microplastics in the Environment? *Mar. Pollut. Bull.* **2016**, *104* (1–2), 290–293.

(75) Klein, M.; Fischer, E. K. Microplastic Abundance in Atmospheric Deposition within the Metropolitan Area of Hamburg, Germany. *Sci. Total Environ.* **2019**, *685*, 96–103.

(76) Jambeck, J. R.; Geyer, R.; Wilcox, C.; Siegler, T. R.; Perryman, M.; Andrady, A.; Narayan, R.; Law, K. L. Plastic Waste Inputs from Land into the Ocean. *Science (80-.).* **2015**, *347* (6223), 1655–1732.

(77) Siegfried, M.; Koelmans, A. A.; Besseling, E.; Kroeze, C. Export of Microplastics from Land to Sea. A Modelling Approach. *Water Res.* **2017**, *127*, 249–257.

(78) van Wijnen, J.; Ragas, A. M. J.; Kroeze, C. Modelling Global River Export of Microplastics to the Marine Environment: Sources and Future Trends. *Sci. Total Environ.* **2019**, *673*, 392–401.

(79) Lebreton, L. C. M.; Zwet, J. Van Der; Damsteeg, J.; Slat, B.; Andrady, A.; Reisser, J. River Plastic Emissions to the World's Oceans. *Nat. Commun.* **2017**, *8*, 1–10.

(80) Hoornweg, D.; Bhada-Tata, P. *What a Waste: A Global Review of Solid Waste Management*; 2012.

(81) Essel, R.; Engel, L.; Carus, M.; Ahrens, R. H. *Sources of Microplastics Relevant to Marine Protection in Germany*; 2015.

(82) Lassen, C.; Hansen, S. F.; Magnusson, K.; Norén, F.; Bloch Hartmann, N. I.; Jensen, P. R.; Nielsen, T. G.; Brinch, A. *Microplastics: Occurrence, Effects and Sources of Releases to the Environment in Denmark*; The Danish Environmental Protection Agency, 2015.

(83) Magnusson, K.; Eliasson, K.; Fråne, A.; Haikonen, K.; Hultén, J.; Olshammar, M.; Stadmark, J.; Voisin, A. *Swedish Sources and Pathways for Microplastics to the Marine Environment. A Review of Existing Data*; 2016.

(84) Unice, K. M.; Weeber, M. P.; Abramson, M. M.; Reid, R. C. D.; van Gils, J. A. G.; Markus, A. A.; Vethaak, A. D.; Panko, J. M. Characterizing Export of Land-Based Microplastics to the Estuary - Part I: Application of Integrated Geospatial Microplastic Transport Models to Assess Tire and Road Wear Particles in the Seine Watershed. *Sci. Total Environ.* **2019**, *646*, 1639–1649.

(85) Besseling, E.; Quik, J. T. K.; Sun, M.; Koelmans, A. A. Fate of Nano- and Microplastic in Freshwater Systems: A Modeling Study. *Environ. Pollut.* **2017**, *220*, 540–548.

(86) Tramoy, R.; Gasperi, J.; Dris, R.; Colasse, L.; Fisson, C.; Sananes, S.; Rocher, V.; Tassin, B. Assessment of the Plastic Inputs From the Seine Basin to the Sea Using Statistical and Field Approaches. *Front. Mar. Sci.* **2019**, *6* (April), 1–10.

(87) Hale, R. C. Are the Risks from Microplastics Truly Trivial? *Environ. Sci. Technol.* **2018**, *52* (3), 931–931.

(88) Burton, G. A. Stressor Exposures Determine Risk: So, Why Do Fellow Scientists Continue To Focus on Superficial Microplastics Risk? *Environ. Sci. Technol.* **2017**, *51* (23), 13515–13516.

(89) Backhaus, T.; Wagner, M. Microplastics in the Environment: Much Ado about Nothing ? A Debate. *PeerJPreprints* **2018**, No. May 2018, 0–5.

(90) Everaert, G.; Van Cauwenberghe, L.; De Rijcke, M.; Koelmans, A. A.; Mees, J.; Vandegehuchte, M.; Janssen, C. R. Risk Assessment of Microplastics in the Ocean: Modelling Approach and First Conclusions. *Environ. Pollut.* **2018**, *242*, 1930–1938.

(91) Adam, V.; Yang, T.; Nowack, B. Toward an Ecotoxicological Risk Assessment of Microplastics: Comparison of Available Hazard and Exposure Data in Freshwaters. *Environ. Toxicol. Chem.* **2019**, *38* (2), 436–447.

(92) Applied Market Information Ltd. *AMI's 2015 EUROPEAN PLASTICS INDUSTRY REPORT*; 2015.

(93) Hoellein, T. J.; Shogren, A. J.; Tank, J. L.; Risteca, P.; Kelly, J. J. Microplastic Deposition Velocity in Streams Follows Patterns for Naturally Occurring Allochthonous Particles. *Sci. Rep.* **2019**, *9* (1), 1–11.

(94) Zimmermann, L.; Dierkes, G.; Ternes, T. A.; Völker, C.; Wagner, M. Benchmarking the in Vitro Toxicity and Chemical Composition of Plastic Consumer Products. *Environ. Sci. Technol.* **2019**, *53* (19), 11467–11477.

6 Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe

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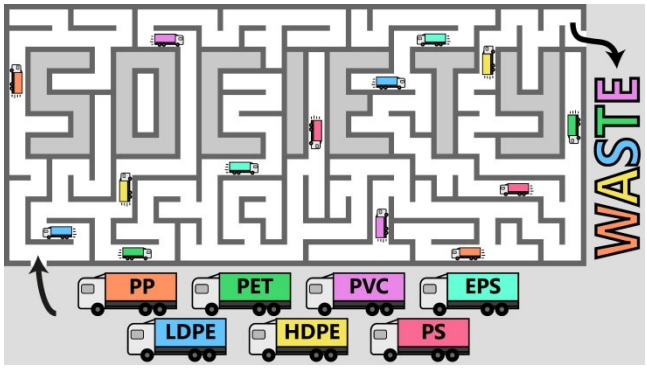
6.1 Abstract

The omnipresence of plastics in our lives and their ever increasing application range continuously raise the requirements for the monitoring of environmental and health impacts related to both plastics and their additives. In this work we present a static probabilistic material flow analysis of seven polymers through the European and Swiss anthroposphere to provide a strong basis for exposure assessments of polymer-related impacts, which necessitates that the plastic flows from production to use and finally to waste management are well understood. We consider seven different polymers, chosen for their popularity and application variety: low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), expanded polystyrene (EPS), polyvinylchloride (PVC) and polyethylene terephthalate (PET). We include synthetic textile products and consider trade flows at various stages of the life-cycle, and thus achieve a complete overview of the consumption for these polymers. In Europe, the order of consumption is PP>LDPE>PET>HDPE>PVC>PS>EPS. Textile products account for 42±3% of the consumption of PET and 22±4% of PP. Incineration is the major waste management method for HDPE, PS and EPS. No significant difference between landfilling and incineration for the remaining polymers is found. The highest recycling share is found for PVC. These results can serve as basis for a detailed assessment of exposure pathways of plastics or their additives in the environment or exposure of additives on human health.

6.2 TOC-Art:



6.3 Introduction

The European plastic production has increased by a factor of 160 between 1960 and 2010[1], and the use of plastics is expected to continue to rise in the future[2] in a wide range of applications[3]. Many different polymers can be used for specific applications, as for example polypropylene (PP), low-density polyethylene (LDPE), high-density polyethylene (HDPE), polyvinyl chloride (PVC), polyurethane (PUR), polystyrene (PS) and polyethylene terephthalate (PET), which together represent around 80% the total manufacturer's polymer demand in Europe[4]. Synthetic textiles are also increasingly used in Europe, with a total European production of man-made fibres of 3.7 Mt in 2011, largely dominated by polyester (29% of the European production) and PP (25%)[5]. Additives such as plasticizers or flame retardants permit to adjust the properties of these materials for different applications, and are as a result, widely used[6].

Many questions regarding the environmental impacts of plastics and the health and environmental impacts of additives are being raised[7]. Between 1.15 Mt[8] and 12.7 Mt[9] of plastic are estimated to end up in the marine environment from land-based sources each year, either as macro-litter or microplastic particles. Microplastic is found in diverse forms, from fragments, to pellets, flakes and fibres[10], suggesting the importance of both plastic-related sources and synthetic textiles. Polyethylene, PP and PS microplastics tend to be most frequently sampled in environmental samples, along with frequent mention of PVC, polyamide and PET[11–14]. The resistance to biodegradability of commonly used plastics gives them a very long residence time in the environment, making them potential Persistent Organic Pollutants (POPs)[15]. Multiple problems are caused by such pollutions, as for example chemical toxicities due to residual monomers, plastic additives or adsorbed POPs[16], with the additional threat of accumulation of microplastic along the trophic chain[16]. Moreover, it has been suggested that plastic additives have effects on human endocrine, reproductive and developmental functions[17] and could also

be involved in other human health problems as for instance asthma[18], due to their leaching from products[19]. All the mentioned hazards may be strongly polymer dependent, as the monomers and additives used vary from polymer to polymer and leaching of chemicals may depend on the chemistry of the material[20].

Depending on the life-cycle and the specific applications of a material, release pathways can be very different[21]. Knowledge on the full life cycle of the material can therefore improve our understanding of its releases and the releases of corresponding additives. In order to give a strong basis for an exposure assessment of polymer-related impacts, the polymer flows from production to use and finally to waste management need to be well understood with a high degree of detail. Describing the life cycles of different polymers separately will furthermore enable taking into account polymer-specific pathways and toxicities into a risk assessment.

Material Flow Analysis (MFA) is one approach of modelling the flows of materials through the anthroposphere and it has been used for many types of materials[22]. Many MFA studies on plastic flows in society have been published without distinguishing individual polymers[2,23,32–34,24–31]. Hitherto, few studies have focussed on individual polymers. Most polymer-specific MFA studies have focussed on PVC, because of strong debates regarding its possible health impacts[35]. The most recent PVC flow analysis concentrated on Europe[36] and modelled the in-use stock of various applications generated from 1960 to 2012. Other PVC-specific MFAs have focussed on Sweden[35], Japan[37] and China[38]. The remaining polymer-specific MFAs have focussed on PET because of its potential for closed-loop recycling, once for the USA[39] and once for a Columbian city[40]. In both studies, the attention given to the consumption stage was limited. No polymer flow studies were found that cover other commodity polymers. In Switzerland, one study described specific polymer flows as a part of the total description of the Swiss waste management system, including other materials such as metals and glass[41], and one study described the flows of packaging in Austria while distinguishing polymers[42]. Consulting companies provide information on production, manufacturer's demand and/or waste management options for plastic in Europe[1,4,43–45], Germany[46–50], or Switzerland[23], or for PVC in Germany[51], without using the systematic approach of MFA and without including textile products. PlasticsEurope[43] provides estimates of the plastic manufacturer's demand by country, or for individual polymers for Europe excluding textile applications as part of their yearly market analyses for Europe. AMI[45] provides polymer production data for Europe, polymer manufacturer's demand data by country, along with polymer application data for Europe. Nevertheless, a complete overview of the life-cycles of the individual polymers cannot be obtained from these reports, as polymer-specific flows are only sporadically addressed.

This overview shows that despite the omnipresence of polymers in our society and the world-wide discussion on plastic waste in the environment, little is known about the polymer-specific mass flows through our society. The aim of this study is therefore to fill this gap and quantify the material flows for seven commodity polymers in Europe and Switzerland using a static Probabilistic MFA (PMFA). Using a static model enables to provide a detailed overview of the consumed products as well as the proportions of plastic undergoing different waste management practices without experiencing any effect from the steady-state assumption. The ongoing debates regarding the impacts of plastics on the environment and human health have prompted reactions from policy makers[52,53]. In order to support decision-making, an aim of the present research is to lay the base for a flow assessment of different polymers to provide a basis for discussions between scientists and regulators.

6.4 Methods

6.4.1 Materials considered

The popularity of use[4] and the frequency of presence in the environment[11,12] were the two criteria to decide what polymers were considered in this study: LDPE, HDPE, PP PS, EPS, PVC and PET. PS and EPS are considered separately even though their chemical compositions are identical, as they have very different applications and physical properties. The copolymers of PP are covered in the masses reported, following conventions from market reports. Ultra-High Molecular Weight Polyethylene (UHMWPE) and Medium Density Polyethylene (MDPE) are included in the masses reported for HDPE. Linear LDPE (LLDPE) is considered in LDPE. In the reported masses for PET, polyester fibres are also included, as polyester fibres consist to a large extent of PET fibres[54]. The contribution of additives to the total mass of goods was removed to the best of our abilities for consistency with available production data. Throughout the text, individual plastics will be referred to as polymer, while all polymers together will be referred to as plastic.

6.4.2 PMFA

The PMFA method used in this study has been described in detail elsewhere[55] and has already been applied to several materials[56,57]. It relies on repeated sampling of Bayesian probabilities defining the inflows into the system and the transfer coefficients (TCs) used to describe the partitioning of the mass in a process[22]. For each iteration of the Monte-Carlo simulation, two mathematical objects are sampled from the chosen Bayesian distributions: an input vector

where the mass inflow across the system boundary to each compartment is described, and a TC matrix which is based on the defined TCs. The input vector describes the starting mass in every compartment of the system and is used to describe the primary production input as well as trade inflows. These two mathematical objects are then used to solve a matrix equation 10^6 times, yielding Bayesian distributions of the masses contained in each compartment. We assume the system is in steady state and neglect stocks in this assessment. Since the flows are calculated on the basis of an input vector and a transfer coefficient matrix only, no masses are equilibrated along the system to obtain missing information as is of use in other standard MFA methods. As a result, solely the masses or flows after the consumption stage are affected, for which only the proportions will be presented as results. More information on this can be found in the discussion.

The shape of the Bayesian distributions depends on data availability[58] and quality[25]. Triangular, trapezoidal or step distributions may be chosen depending on the number of data points available. An uncertainty is associated to each data point following a method introduced for MFA uncertainties[25], based on a pedigree matrix with 5 different data quality indicators for geographical, temporal and material representativeness as well as completeness and source reliability. TC distributions are truncated between 0 and 1 to insure a mass balance in the system, while input mass distributions are truncated below 0. Conservation of mass is also insured by constraining TCs leaving one compartment to sum up to 1, either by normalizing them, or defining one flow as the remaining share. The normalization step does not notably affect the distribution shape, as long as the chosen distributions are compatible. More details on the method are given in the Supporting Information (SI). Data from peer-reviewed publications, databases and reports were used to find the appropriate parameters for the definition of the Bayesian distributions. Details on the method are given in the Supporting Information (SI).

6.4.3 Model structure, system boundary and assumptions

Europe and Switzerland are independently modelled for year 2014. Europe is defined as EU28 in this study, as most of the data refers to EU28. Processes were aggregated according to the available data. A description of the compartments and flows of the system is shown in a generic flow chart valid for all the seven polymers (Figure 5). The system consists of five stages: production, manufacturing, consumption, waste collection and waste treatment. Two compartments included in the flowchart are not part of the system but describe the flows in and out of the system: trade and elimination.

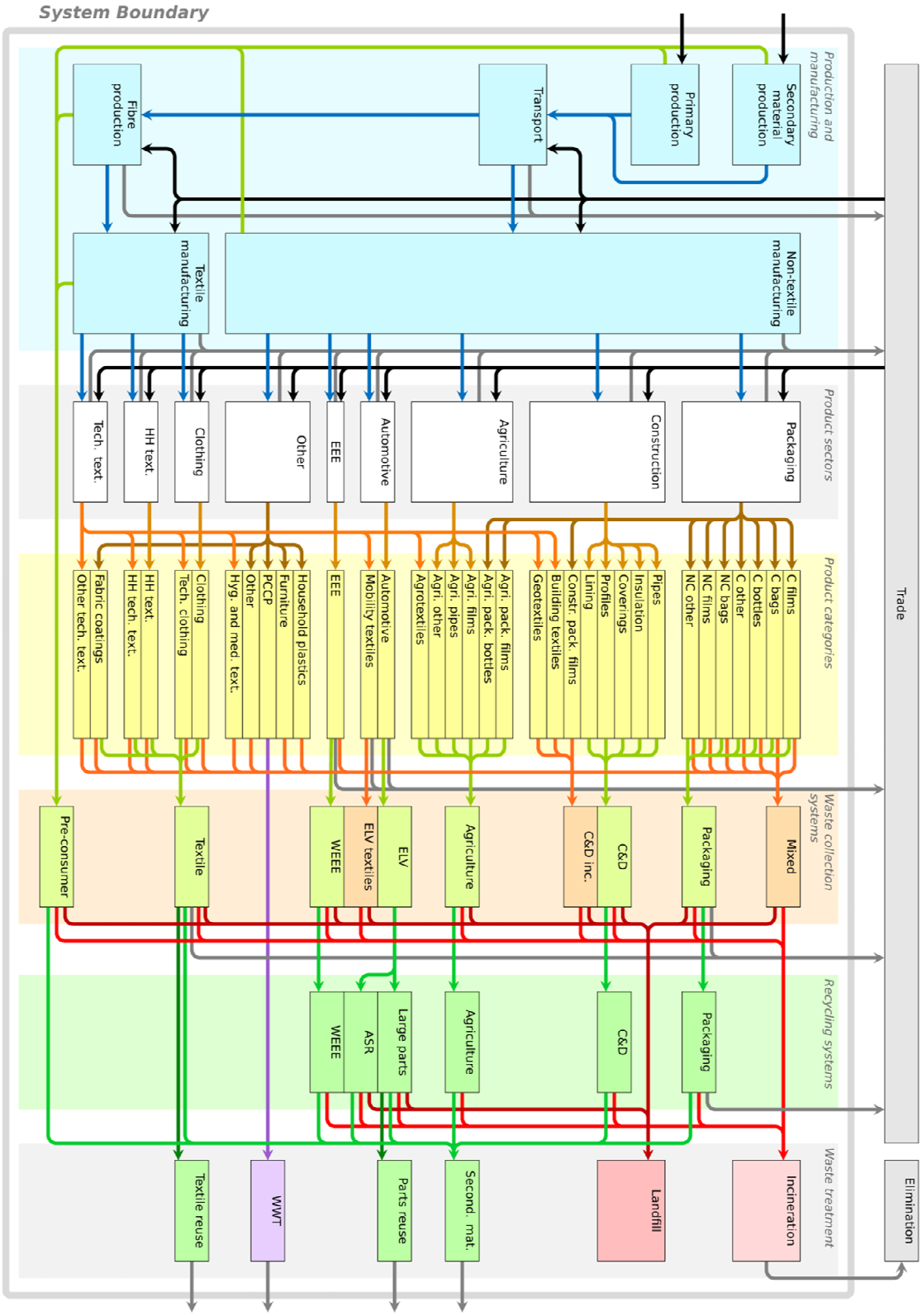


Figure 5: General structure of the material-flow model. The system is used for all the polymers, but some compartments or flows are zero for some polymers. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hy-

giene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), Second. mat. (Secondary material), WWT (Wastewater Treatment).

The production and manufacturing stage comprises six processes: primary production, secondary material production, transport, fibre production, non-textile and textile manufacturing. Polymers are produced from raw materials in primary production and from reclaimed material in secondary material production. The bulk of the material from these two processes flows to transport which is the hub of the trade of plastic in primary forms. From there, the material can be processed into plastic products in non-textile manufacturing, or can be processed into filaments and yarns in fibre production. Filaments and yarns are further processed into textile products in textile manufacturing. A small part of the plastic in the different production and manufacturing processes is lost as waste and flows to pre-consumer waste collection for further treatment.

Between manufacturing and the product categories, an intermediate stage is implemented to facilitate the calculation of import and export flows before the actual consumption. This stage describes the various product sectors: packaging, construction, automotive, electrical and electronic equipment (EEE), agriculture, clothing, household textiles, technical textiles and other plastic products. A sector may be further divided in up to ten different product categories to better describe the flows in and out of consumption. No import of goods is modelled at this level, since all the trade into consumption is included in the previous step. Export from the consumption stage is only defined for some specific product categories as second-hand products.

The plastic contained in the different product categories then flows to End-of-Life (EoL) compartments: waste collection, recycling and waste treatment. EoL products can be collected in a specific waste collection system or as mixed waste before entering recycling or waste management. Most packaging applications are either collected separately or collected as mixed waste. Construction products are collected in Construction and Demolition (C&D) waste. Packaging applications for the construction sector are collected in incinerable C&D collection. Agricultural applications and agricultural packaging are modelled to be collected in a separate waste stream called agricultural waste collection. Automotive plastics are collected in End-of-Life Vehicle (ELV) collection, while EEE flows to Waste of Electrical and Electronic Equipment (WEEE) collection and mixed waste. Clothing and household textile applications are either separately collected or disposed of in mixed waste. Technical textiles flow to various waste collection systems: construction textiles and geotextiles flow to incinerable C&D collection, agrotextiles to agriculture plastic collection, mobility textiles to ELV textiles, hygiene and medical textiles to

mixed waste, technical clothing and technical household textiles to textile collection and mixed waste, other technical textiles to mixed waste. Applications in the *Other* sector have various destinations: mixed waste or textile collection for fabric coatings, Waste Water Treatment (WWT) for Personal Care and Cosmetic Products (PCCP), and mixed waste for the remaining applications.

Most waste collection compartments have a corresponding recycling compartment, except for mixed waste, incinerable C&D waste and ELV textiles which flow to incineration plants and landfills, and except for collected textiles and pre-consumer waste which are directly reused, incinerated, landfilled or exported. Every other waste collection system has a flow connection to the corresponding recycling process, along with incineration plants and landfills for the share of products sorted out of the waste stream. In the case of ELV, two distinct recycling processes are modelled: the recycling of large auto-motive parts and the recycling of Automotive Shredder Residue (ASR).

EoL treatment is described by four processes: recycling and reuse, landfill, incineration and WWT. No distinction is made between incineration with or without energy recovery. A portion of each collection and recycling system flows to landfill and incineration, while the rest either flows to recycling and reuse or to export. Trade flows at this level are only modelled for collected packaging and textiles. The fate of plastic after the landfill, WWT and recycling and reuse processes is not further considered in this study.

6.4.4 Input and transfer coefficients

Amounts of produced polymers in primary forms were obtained from a market report for EU28+2 (defined as European Union with Norway and Switzerland)[45] and from the Eurostat database[59] for EU28. We assume that the error induced by the different geographic systems is negligible, as the plastic production industry is not very strong in Norway and inexistent in Switzerland for the polymers considered[45]. The corresponding production values can be found in the SI (see Table S4).

Trade statistics can be obtained from the Eurostat[60] database for Europe and from the Swiss-Impex[61] database for Switzerland, for a wide amount of goods characterized by codes using the Harmonized System. Only net imports are considered (Imports – Exports). If the net trade is positive, it is used as parameter to create the corresponding Bayesian distribution from which the input vector will be sampled. If the net trade is negative, the mean mass contained in the compartment is compared to the outflow mass, and a Bayesian distribution is constructed

around it. Additional information on trade can be obtained from reports describing the shares of goods on the market that were imported or locally produced, or the shares of waste management options for specific products. For some less known trade flows as packaging, a separate calculation needed to be performed which is explained in detail in the SI. A detailed description of the all the production values and tradeflows is given in the SI.

The parameters used to create the TC distributions were taken from several studies including market reports, national reports and peer-reviewed literature. Data that was specific to the chosen geographical system, year and polymer was preferred but could not be found for all parameters. For the parameters for which no system-specific data could be found, proxy data from other systems was used, while accordingly adapting the uncertainty attributed to these parameters. Since over 600 TC distributions needed to be defined in total for the two different geographical systems and seven polymers, a detailed description is given in the SI. An overview of the literature used to create the TC and input distributions for Europe can be seen in Figure 6.

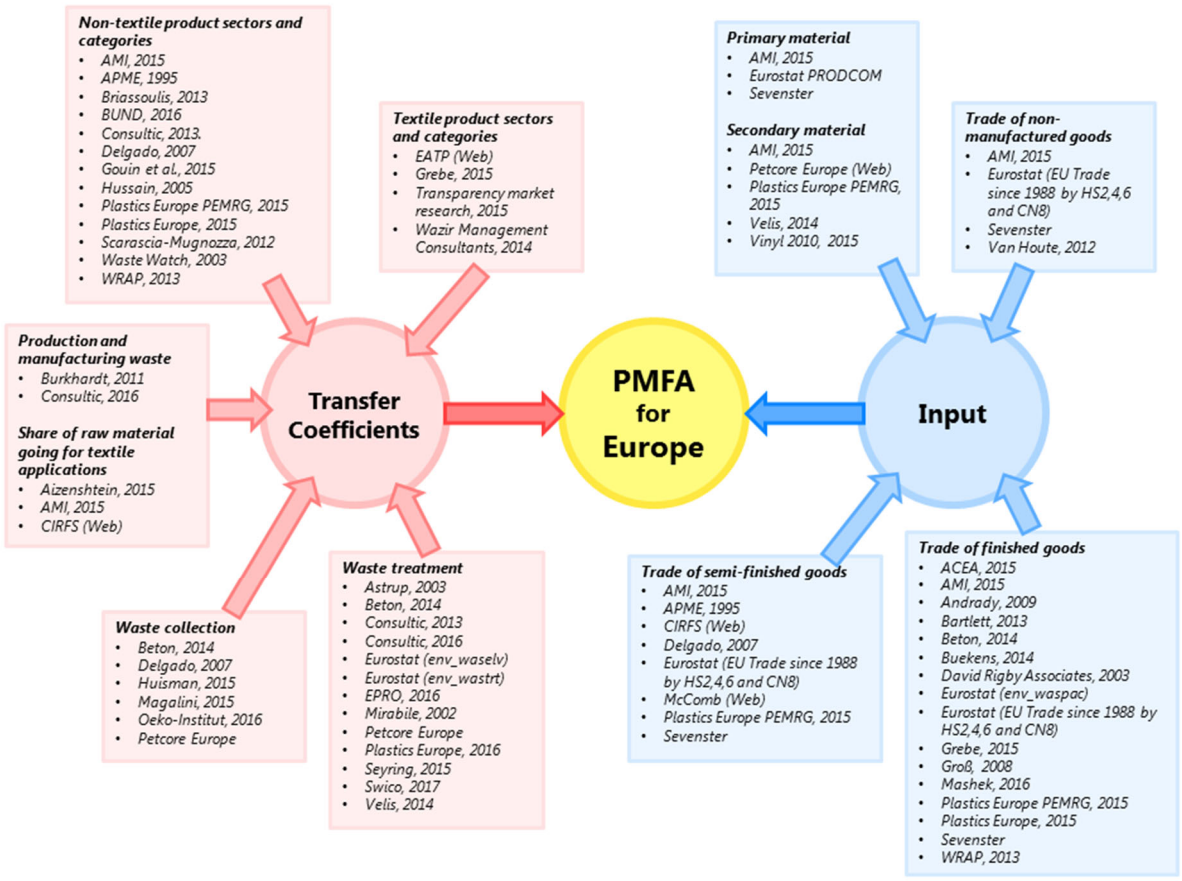


Figure 6: Summary of the data sources used for a quantification of the model parameters. A key to the references is available in Table S15. A similar figure for Switzerland, as well as the key to the references can be found in Figure S26 and Table S16.

6.5 Results

Different types of probability distributions and metrics can be analysed from the simulations. Four types of probability distributions are shown as examples in Figure 7 for the seven materials. The first example is the distribution of an input distribution into the model, in this particular case the European polymer production (Figure 7A). In this case, all of the distributions adopt a trapezoid shape since two different data sources were used, although this behaviour is not visible for PS and EPS. The largest input into production is clearly for PP, followed by LDPE, PVC and HDPE. The largest spread is found for PVC as the difference in produced mass between the two references is the largest for this polymer. PET, PS and EPS have the lowest production amounts. The second example illustrates a TC distribution with the fraction of plastic in primary

forms that is used for fibre production in Europe (Figure 7B). Only three polymers are represented here, as there are no modelled fibre and textiles production for the other polymers. The third plot presents a resulting mass in a compartment using the example of mixed waste collection (Figure 7C). It is already apparent that the distributions are smoother compared to the production distributions, as they are the result of many different probabilistic parameters combined. HDPE, LDPE and PP have conserved similar proportions between the production distribution and the mixed waste collection, but the other polymers do not follow the same repartition. EPS and PVC have a much lower mass than what could be expected compared to HDPE, LDPE and PP because of the large fraction of these polymers that is used in construction applications and that follows different waste streams. On the contrary, the mass of PET is much larger than the production volume, due to the large amounts of PET imported into Europe as textiles and various plastic goods. The fourth and last plot shows an aggregated mass over several compartments representing our estimate of the European polymer consumption (Figure 7D). This mass is an aggregate over the 35 product categories shown in yellow in Figure 5. It shows that the polymer most used is PP, followed by LDPE, PET, HDPE, PVC, PS and EPS.

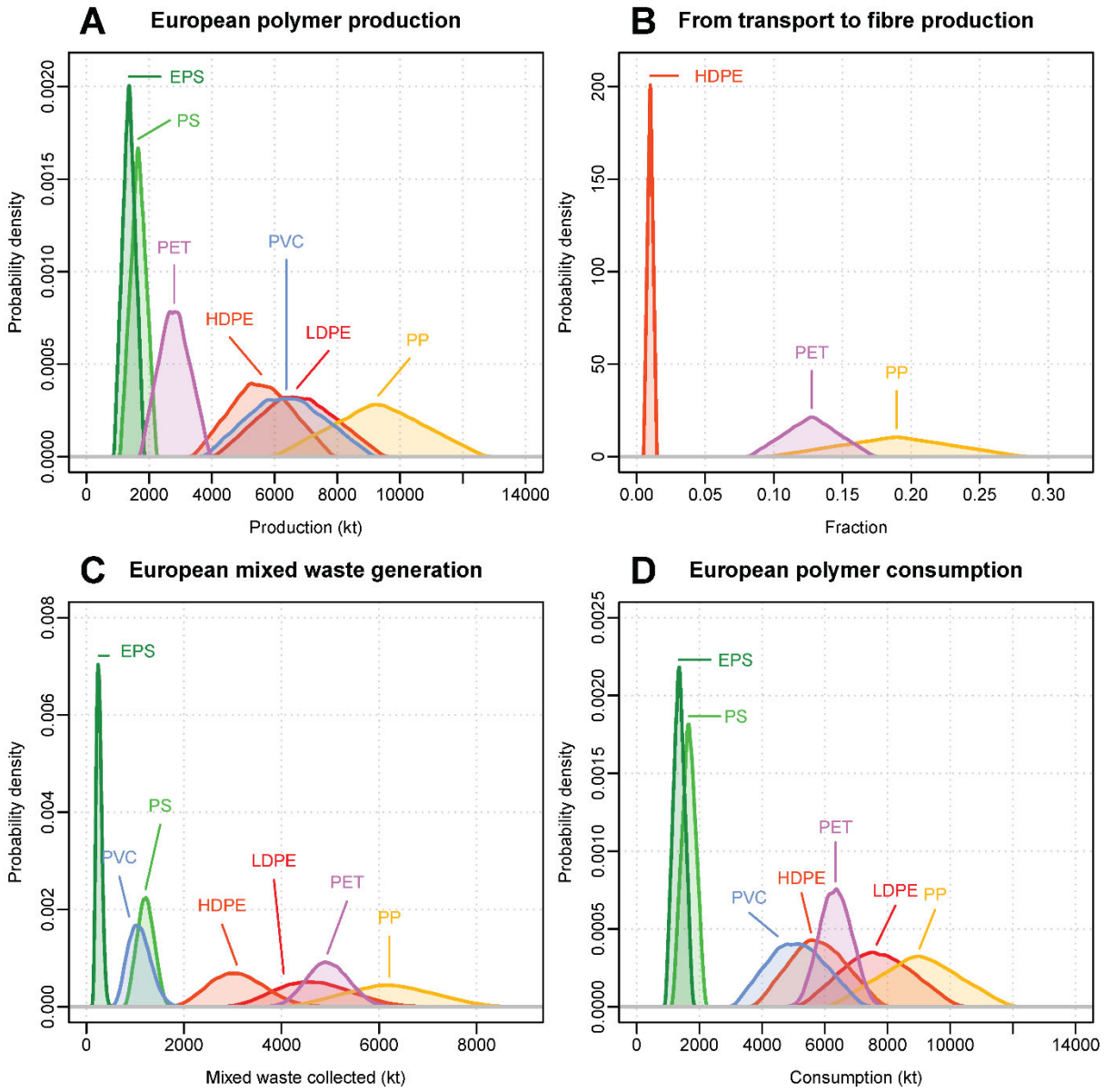


Figure 7: Selected probability distributions from the results of the Monte-Carlo simulation. Examples of (A) input, (B) transfer coefficient, (C) mass in a compartment and (D) aggregated mass over several compartments are shown.

Simplified flow diagrams are shown for all seven polymers in Europe in Figure 8. In these diagrams, the processes production, manufacturing, separate waste collection, recycling and recycling and reuse are shown as aggregates over several compartments visible in Figure 5, with the purpose of improving readability. Furthermore, the product sectors are not presented in the simplified flow diagrams and the product categories are also not shown individually, but regrouped to improve legibility. Flows between these aggregated processes were aggregated so that no mass flow is neglected. Corresponding diagrams for Switzerland can be found in the

SI (Figure S27-Figure S33). The complete flow diagrams with all flows shown separately are given in the SI in Figure S38-Figure S51. Mean masses are reported for flows and compartments along with the standard deviation of the probability distribution. The sum of flows into a compartment and the actual mass inside a compartment might not coincide due to rounding, as the mean values were rounded to the first significant number of the standard deviation[62].

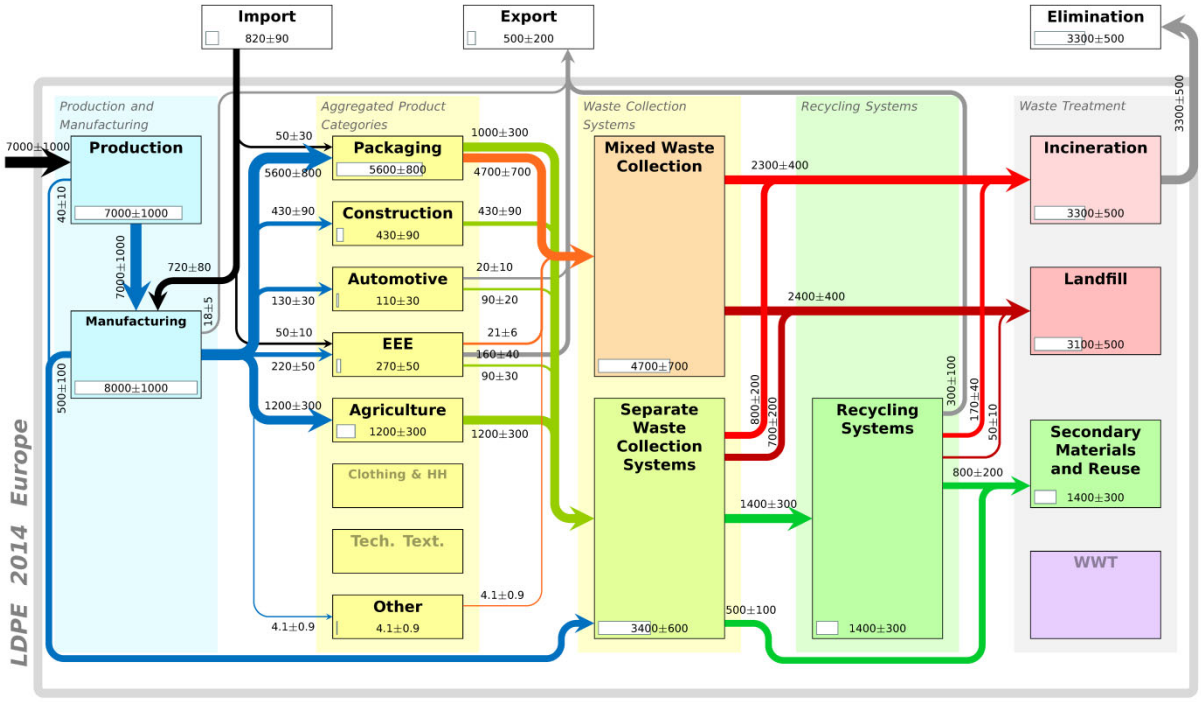


Figure 8: Simplified flow diagrams for the seven thermoplastics in Europe. All units are in thousand metric tonnes (kt). In each diagram, the system it is valid for is inscribed in the lower left corner. The masses reported for the flows and compartments are rounded to the precision of the given standard deviation. The sums of flows might therefore not coincide with the reported masses. The width of the flow arrows is larger for larger masses, and the white bars in the compartments are longer for larger masses. Colours were used to help visualizing the flow diagrams. The simplified flows for Switzerland are also given in Figure S27-Figure S33 of the SI. The complete flow diagrams with all flows shown separately are given in Figure S38-Figure S44 of the SI. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

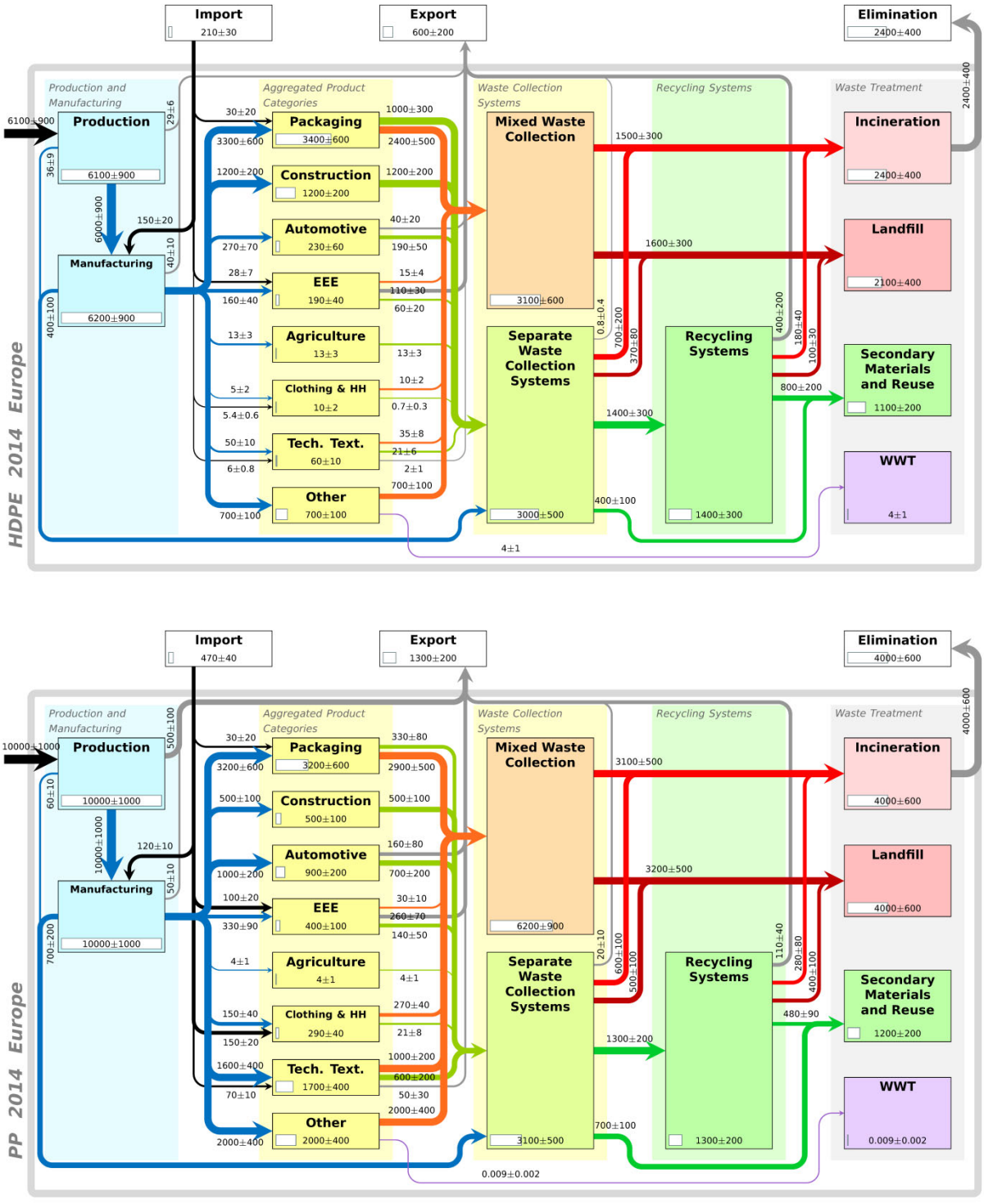


Figure 8 (continued)

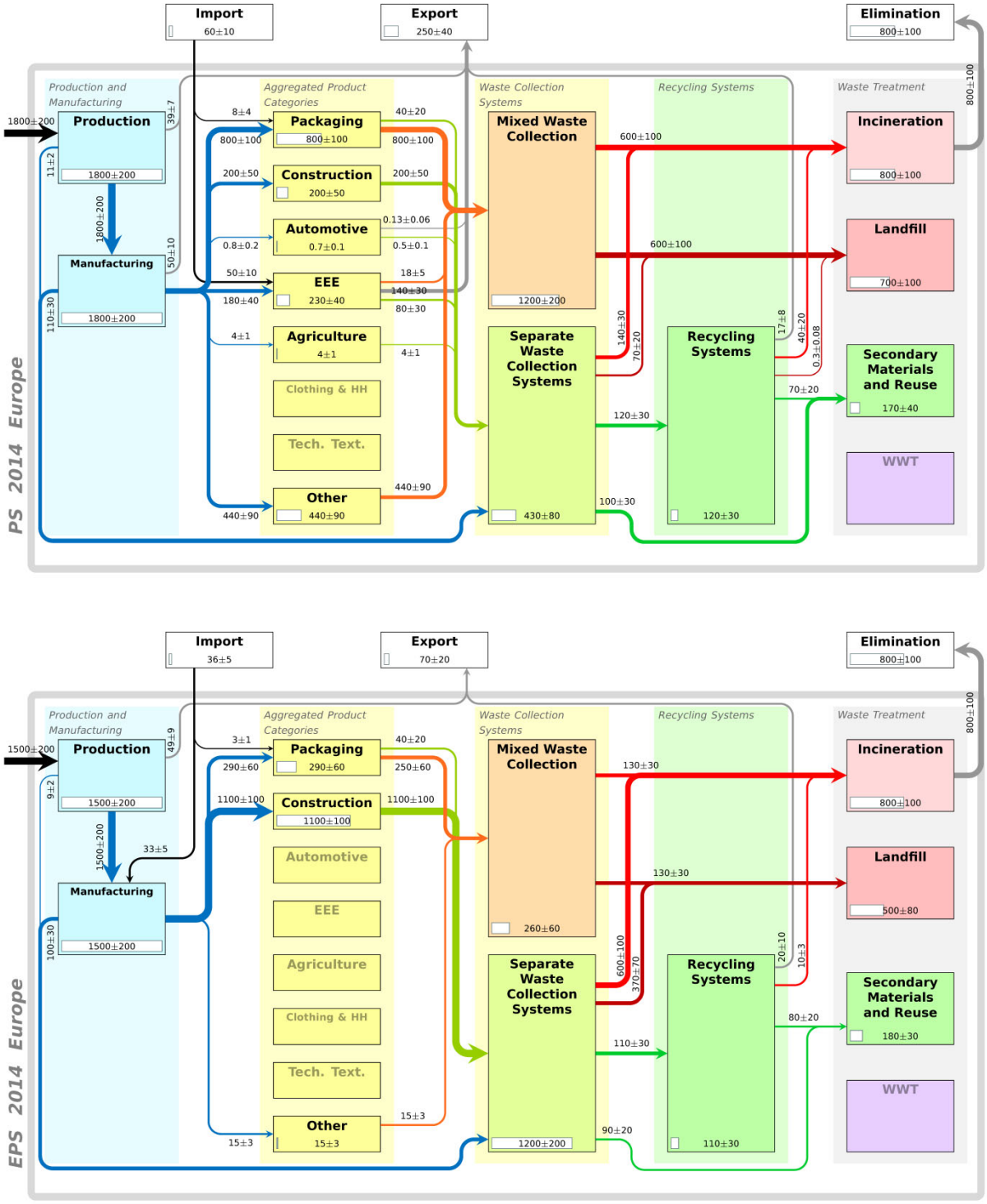


Figure 8 (continued)

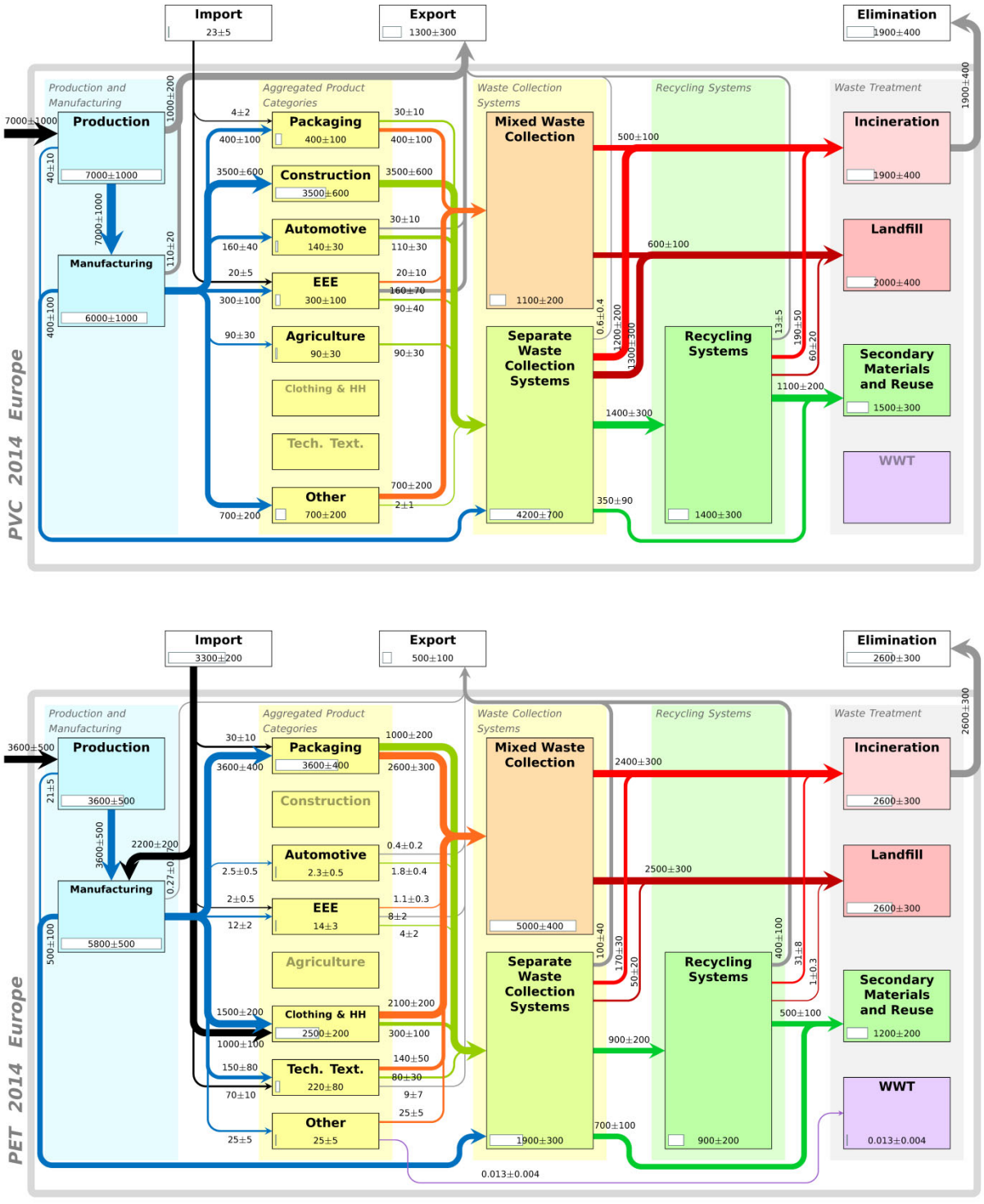


Figure 8 (continued)

In Europe, production is the dominating input for all polymers modelled. 5600±900 kt of HDPE are produced in Europe from raw materials, 500±100 kt from recycled material, 150±20 kt are imported as preliminary products and only 70±20 kt as finished products. This means that 89±2% of the plastic in circulation was produced locally. Similarly, 81±3% of LDPE, 88±3% of

PP, 88±2% of PS, 90±2% of EPS, 96±1% of PVC of the input comes from the local plastic production (Figure S38-Figure S44). For PET, only 40±4% of the input stems from the production stage, due to the large imports of textiles, preliminary products and the re-use of recycled material. The vast majority of the produced plastic remains in the system and only little is exported if we consider net exports: 0.5±0.1% of HDPE, 0±0% of LDPE, 5.3±0.7% of PP, 2.2±0.3% of PS, 3.3±0.5% of EPS, 15±2% of PVC and 0±0% of PET. As could be expected from the parameters used, the waste produced at the production stage is very low.

A visual comparison of the consumption is given in Figure 9 for all product categories and polymers. The leading applications are found in packaging and construction applications, which are the two main application sectors for plastics. The most consumed applications for these polymers are non-consumer films, other consumer packaging, construction pipes, consumer bottles, consumer bags and other non-consumer packaging. LDPE dominates most of the film applications, as for instance non-consumer films, including agricultural packaging films and construction packaging films, non-consumer bags, and agricultural films. PET and PP share most of the textile applications, with PP dominating for most of the technical textile applications, and PET for the remaining applications. A similar overview of the Swiss consumption can be found in Figure S35. Small differences can be found between the Swiss and the European consumptions. One main cause for this is the smaller proportion of LDPE imported compared to the remaining polymers.

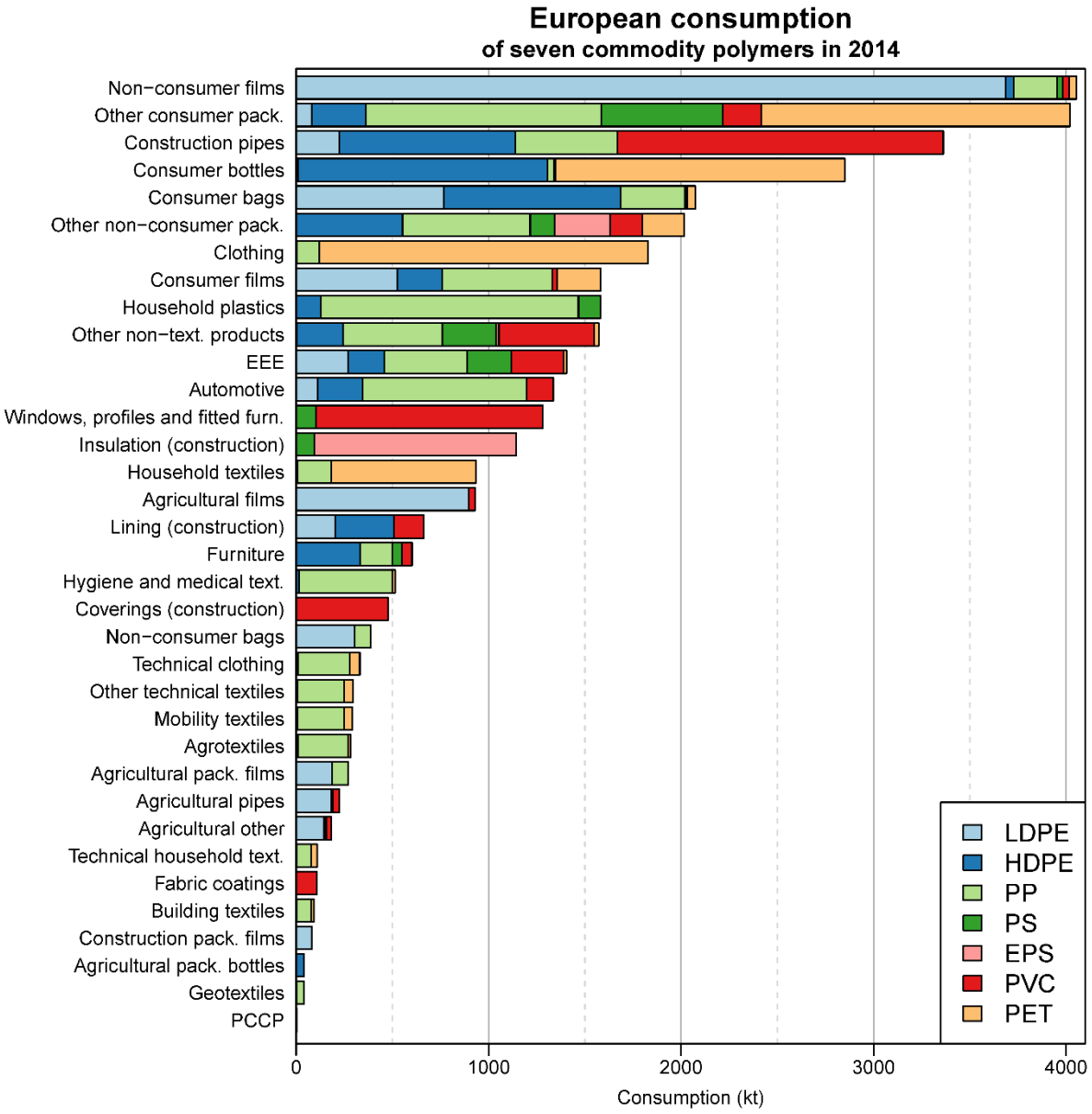


Figure 9: Overview of the European polymer consumption in 2014 for the seven polymers considered. Abbreviations: pack. (packaging), furn. (furniture), EEE (Electrical and Electronic Equipment), text. (textiles), PCCP (Personal Care and Cosmetic Products). The corresponding data for Switzerland are shown in Figure S35 of the SI.

In the simplified flow diagrams (Figure 8), the waste collection systems have been aggregated to improve the readability of the flows. In total, nine different separate waste collection systems are being modelled (Figure 5). 42±4% of LDPE waste is modelled to be collected separately (Figure S38), for the most part as agricultural plastics, but also as packaging pre-consumer and construction waste. Similarly, 49±5% of HDPE waste is modelled to be collected separately (Figure S39), mostly as packaging and construction waste. Of the PP waste generated, 33±3%

is collected separately (Figure S40) with the leading waste streams being technical textiles, pre-consumer waste, construction and automotive plastics. Of all considered polymers, PS has the lowest separate collection rate with only 26±3% (Figure S41), mainly from construction and pre-consumer waste. 82±3% of EPS (Figure S42) and 79±3% of end-of-life PVC (Figure S43) are collected separately, of which the vast majority comes from construction applications. Last, only 28±3% of PET is collected separately (Figure S44) with the main waste streams being packaging, textiles and pre-consumer waste.

Figure 10 shows the final compartments for the seven polymers by application sector for both Europe and Switzerland. The possible final compartments are: landfill, incineration, reuse, export and WWT. Applications for which no information is shown are for polymers that are not used in that specific sector. In Europe in most cases, the prevailing waste treatment options are incineration and landfills. Most incineration and landfilling are observed for technical textiles, household textiles and *Other* products, where recycling and reuse is very low or inexistent. Packaging also has a high share of incineration and landfilling depending on the material. The highest recycle and reuse shares are accomplished for construction and agriculture applications. PVC in particular has the highest rate of recycling and reuse (Figure S34) partly due to the higher uniformity of applications and purity of used material, and to the commitment of the PVC industry with the VinylPlus programme[63]. Most packaging material recycling and reuse occurs for PET and HDPE, less so for LDPE and EPS, and even less for the remaining three. For construction plastic, the highest recycling and reuse rate occurs for PVC, followed by LDPE, HDPE and PP. The recycling and reuse of PS and EPS in construction is the least significant out of all polymers. There is no difference between the recycling and reuse of the different polymers used in automotive applications since this sector was not described in more detail with product categories. We can expect the recycling and reuse of different polymers in the automotive sector to actually vary more, as the reuse of car parts should most likely be more important for polymers that are used in large car parts as for example PP, than for PS and PET which are used in smaller and less homogeneous parts[64]. Recycling and reuse of textiles is important for clothing applications but less so for household applications, and even less so for technical textiles. The small portion of recycling and reuse observed in Figure 10 for technical textiles is due to technical clothing and technical household textiles, which are assumed to follow the same recycling and reuse rates as consumer clothing and household textiles, and to mobility textiles which are exported along with second-hand cars. Export is a significant outflow for EEE and automotive applications, less so for packaging and clothing and barely significant for the remaining applications. The WWT option only exists for the product category PCCP in the *Other* sector. The share of this waste treatment option nevertheless disappears compared

to other options, as this application has a very low share of the consumption of PET, HDPE and PP (Figure 9).

The differences between the waste treatment options in Europe and Switzerland are largely due to the landfill ban for incinerable waste enforced in Switzerland in the early 2000s[65]. The fraction of waste which is landfilled in Europe is instead incinerated in Switzerland. Another notable difference between the two systems is the more prevailing export of second-hand vehicles and textiles out of Switzerland. Contrastingly, a lower export rate of EEE is observed in Switzerland, due to the absence of consideration of illegal trade of EEE, as was the case for Europe[66].

Figure 10: Ultimate compartment of the plastic shown for the nine application sectors and seven polymers. The bars missing are for polymers that are not used in that specific application. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

6.6 Discussion

Of all materials considered here and to the best of our knowledge, only the life-cycles of PVC and PET have already been separately described in MFA studies. Of these two, only PVC has previously been studied for Europe[36], which means that the life-cycles of the remaining polymers LDPE, HDPE, PP, PS, EPS and PET in Europe had not yet been studied separately. The remaining polymers have been modelled along with PVC, PET and other plastic polymers *en bloc* in a few studies on a national scale[23,24,33,34,25–32]. The life-cycle as described in this study provides a high level of detail comprising the production, manufacturing, consumption, waste collection and recycling stages, while including trade flows at all stages of the system. Some consumption data can be obtained in market reports[4,67] with little information regarding the kind of demand calculated, the assumptions and the calculation steps, and excluding the share of consumption of textile products. Our study makes up for these shortcomings by providing a transparent calculation of the total polymer life cycle, including trade flows and the textile life-cycle for the chosen polymers.

6.6.1 Probabilistic approach

Bayesian probabilities are a mathematical construction used to communicate reasonable expectations for a specific proposition[68]. These Bayesian distributions permit to incorporate both parameter uncertainty and variability into the model by decreasing the confidence accordingly. In this case, parameter uncertainty is the major driver for lowering the confidence in the results because of the parametrization using data proxies. The largest distribution spreads originate from parameters which were rated as more uncertain using the Pedigree matrix. These are found for the recycling practices of end-of-life vehicles in Europe which are based on incomplete data sets from Eurostat, the textile product sectors for PET and the technical textiles product categories which were based on global data, the collection rates of textiles and packaging after consumption which are based on estimations for specific applications or older predictions, and the construction product categories which are based on older data. The relative uncertainty on the modelled masses, defined as the standard deviation divided by the mean of the mass distributions, is a direct result of the uncertainties on the used parameters (Figure 11). It is visible that construction and technical textiles product categories have a larger relative uncertainty than the remaining applications, as the data used is older or for a different geographical unit. Nevertheless, some masses still do appear with a low uncertainty within them, for example for insulation made of EPS, because it is the only application in construction for this polymer and its uncertainty only originates from the flows upstream. Data variability is expected to play a minor role, as all flows are averaged over a whole geographical entity for a specific year.

Quantitative information on the relative uncertainty can be found in the SI for both Europe and Switzerland. On another note, as a normalization of the TCs of the compartments takes place, the spreads of the distributions of some TCs are notably affected. As the uncertainties are independently attributed to the flows, but these flows remain coupled, incompatibilities between the resulting distributions arise and they are deformed accordingly. This is not considered to be an issue, but only a mathematical repartition of the uncertainty on coupled flows.

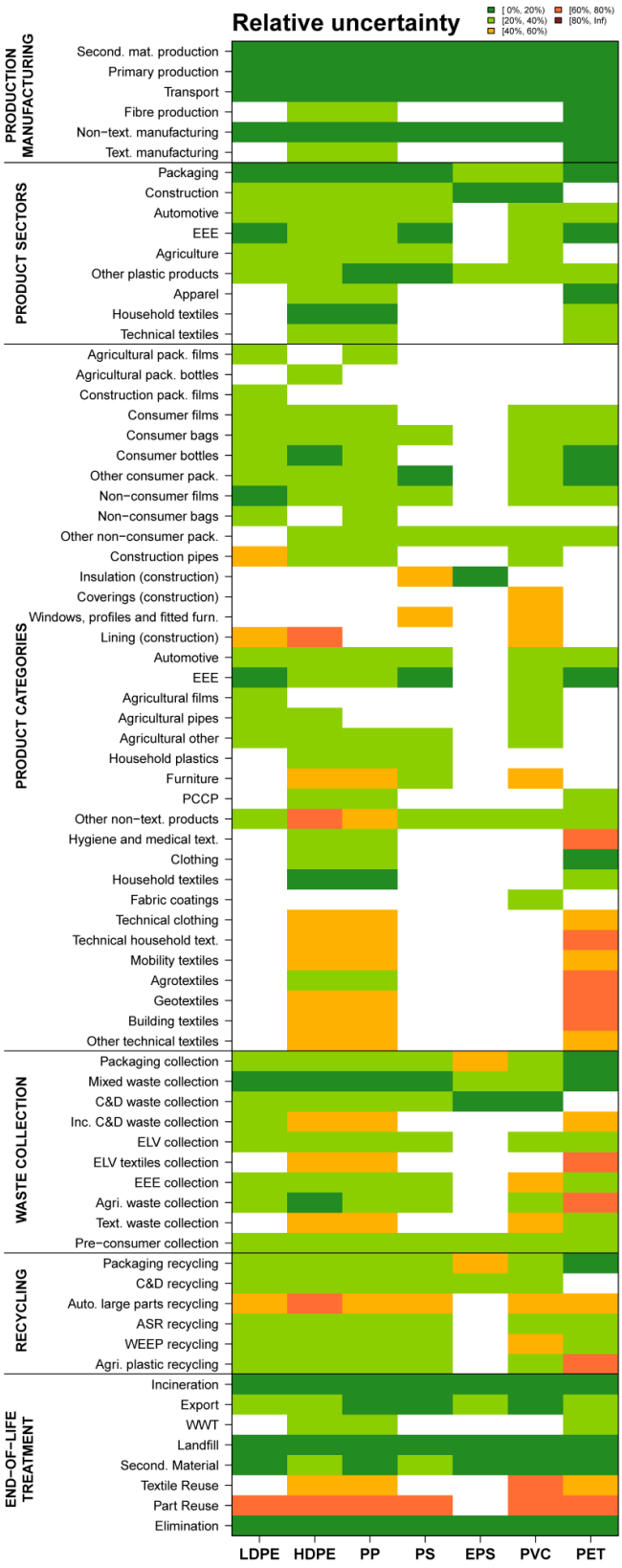


Figure 11: Heat map of the relative uncertainties (standard deviation divided by the distribution mean) associated to each compartment's mass for Europe in 2014. A lower uncertainty is shown in green, while a larger uncertainty is shown in red. White cells correspond to compartments with zero mass. A more detailed version with the numerical values is given in Figure S37 of the SI, also including the respective values for Switzerland.

6.6.2 Steady-state assumption

A static approach to the modelling is sufficient in order to predict the quantities produced and consumed, and to predict the proportions of waste management options in Europe and Switzerland by product. Nevertheless, due to the lack of lifetimes in this model, the predicted waste amounts generated may be subject to an additional uncertainty. Nevertheless, the lack of lifetimes does not affect any stage before consumption, since the method relies only on an input vector and a transfer coefficient matrix, without any balancing of masses consumed and discarded. Previous stages of the life-cycle remain unaffected. Moreover, as approximately 40% of plastic products are said to have a lifetime shorter than 1 month[69], the influence of the steady-state assumption on waste generation is considered limited. Deviations might only be expected for long-lived and very long-lived products, such as EEE (average lifetime of 8 years[70]), automotive (13 years[70]) and construction plastics (35 years[70]). A dynamic MFA approach would permit to have a description of the societal stocks and would yield better predictions of the amount of waste generated for long-lived applications[71,72]. To illustrate the effects of the steady-state assumption, our results for PVC in Europe can be compared to the results of a study from Ciacci et al.[36] who have modelled the PVC life cycle dynamically from 1960 to 2012. In Ciacci et al., more attention was given to the production and manufacturing stages while our study goes in more detail on the consumption and waste management stages. Comparing the waste generated per capita predicted by both models can give insight into the error induced by the absence of lifetimes and stocks in our model. The dynamic approach of Ciacci et al. predicts a waste generation of 0.9 kg/cap of packaging PVC in 2012, 0.4 kg/cap of EEE PVC, 0.3 kg/cap of transportation PVC and 2.0 kg/cap of construction PVC. Our model on the other hand predicts 0.9±0.2 kg/cap of packaging PVC, 0.5±0.2 kg/cap of EEE PVC, 0.27±0.07 kg/cap of automotive PVC, 7.0±1.3 kg/cap of construction PVC[73]. Our results are in accordance with the results from the dynamic model for all applications except for construction plastics where our prediction is more than three times higher than the waste predicted when including lifetimes. This means that our model can predict accurate waste outputs for short-lived and long-lived applications, but not for very long-lived applications such as construction plastics. For products where the market has not yet reached saturation or equilibrium the stock will keep increasing and lifetimes are essential when assessing the waste generated by very long-lived applications. A dynamic MFA constitutes therefore a necessary next step in the flow assessment but is requiring many times the amount of historic data on production, use and flows between compartments. Nevertheless, the presented results on the detailed consumption and proportions of waste management options are not affected by this assumption and do not depend on the inclusion of a dynamic aspect.

6.6.3 Trade

Trade flows were included at every stage of the life cycle including consumption. Only net trade was considered in this study to model the flows of traded goods. A similar approach was followed in other MFA studies[25,36] as it permits to model trade flows with less difficulties than describing import and export explicitly. The importance of the calculation of trade flows for consumption varies depending on the consumption sector considered. The most important trade flow to consider seems to be for textiles for both Europe and Switzerland. Indeed, our calculations show that 38-41% of clothing, 40-62% of household textiles and 4-40% of technical textiles are imported from abroad in Europe, depending on the polymer, and even more in Switzerland (Figure S38-Figure S51). EEE and automotive trade are also quite important to consider in both systems, but especially so for Switzerland, where up to 62% may be imported, in this case for automotive LDPE. Besides, the trade of packaging along with other goods would need more knowledge about the amount and type of packaging required for the packing and transportation of goods. A coarse description of this flow was achieved based on available data. This flow was expected to play an important role for the total packaging consumption in both Europe and Switzerland. Indeed, in Switzerland, this import flows accounts for 12-27% of the consumption of packaging and is thus relevant to consider. For Europe however, this import flows only accounts for around 1% of the polymer consumption in this sector, regardless of the polymer considered. More research on this topic would permit to test this result further. The poor data availability for some of these trade flows limits their accuracy (Table S5). For instance, splitting traded sheets into packaging, construction or agriculture would require additional information on the composition of the good which is currently not available. For this reason, a separate description of construction, agriculture and other plastic products trade was not undertaken. A better knowledge of the composition of traded goods by application and polymer would improve the whole trade description.

6.6.4 Environmental implications

Knowledge about the flows of polymers through the anthroposphere provides the first step towards an assessment of the further flows into the environment. Since emissions of polymers may occur at all stages of its life-cycle, a quantitative knowledge of their life-cycles can help predict the potential for plastic pollution. First indications on this potential can be gained from this model, by comparing process importance in the flow diagrams. Applications with large masses in circulation could be responsible for much larger emissions. In order to rank applications by emission magnitude, release factors need to be assessed in a next step, as well as the full pathways from initial release to final emission into the environment. The material flow

model presented in this work will form the basis to model the intended and unintended emissions of polymers to the environment by quantifying their release. The results presented here could also be used as basis for the exposure assessments of additives which are known or suspected to have an impact on health and pollution. On another note, these results may be helpful for identifying priorities to reach polymer recycling targets by highlighting larger potentials by application. These results also highlight the relevance of a landfill ban in order to recover plastics more efficiently.

6.7 Supporting Information

Description of the method used, of the transfer coefficients and their derivation, data used for production, calculations of trade flows, additional figures for Europe and Switzerland. Excel sheet containing all the data used for the MFA calculation with sources and uncertainty. Excel sheet containing the proportions of polymer included in each considered trade category.

6.8 Acknowledgements

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6.9 References

(1) PlasticsEurope. *Plastics – the Facts 2013: An Analysis of European Latest Plastics Production, Demand and Waste Data*; 2013; http://staging-plasticseurope.idloom.com/application/files/7815/1689/9295/2013plastics_the_facts_PubOct2013.pdf.

(2) Ellen MacArthur Foundation. *The New Plastics Economy: Rethinking the Future of Plastics*; 2016; https://www.ellenmacarthurfoundation.org/assets/downloads/EllenMacArthurFoundation_TheNewPlasticsEconomy_Pages.pdf.

(3) Thompson, R. C.; Swan, S. H.; Moore, C. J.; vom Saal, F. S. Our Plastic Age. *Philos. Trans. R. Soc. Lond. B. Biol. Sci.* **2009**, *364* (1526), 1973–1976.

(4) PlasticsEurope. *Plastics - The Facts 2015: An Analysis of European Latest Plastics Production, Demand and Waste Data*; 2015; http://staging-plasticseurope.idloom.com/download_file/view/478/179.

(5) Van Houte, F. Polyester Fibres in Europe: Developments and Trends. In *9th China International Polyester and Intermediates Forum*; European Man-made Fibres

Association, 2012; pp 1–28.

(6) Pritchard, G. *Plastics Additives, An A-Z Reference*; Brewis, D., Briggs, D., Eds.; Springer, 1998; http://link.springer.com/10.1007/978-94-011-5862-6_18.

(7) Koelmans, A. A.; Besseling, E.; Foekema, E.; Kooi, M.; Mintenig, S.; Ossendorp, B. C.; Redondo-Hasselerharm, P. E.; Verschoor, A.; van Wezel, A. P.; Scheffer, M. Risks of Plastic Debris: Unravelling Fact, Opinion, Perception, and Belief. *Environ. Sci. Technol.* **2017**, *51* (20), 11513–11519.

(8) Lebreton, L. C. M.; Zwet, J. Van Der; Damsteeg, J.; Slat, B.; Andrady, A.; Reisser, J. River Plastic Emissions to the World's Oceans. *Nat. Commun.* **2017**, *8*, 1–10.

(9) Jambeck, J. R.; Geyer, R.; Wilcox, C.; Siegler, T. R.; Perryman, M.; Andrady, A.; Narayan, R.; Law, K. L. Plastic Waste Inputs from Land into the Ocean. *Clim. Chang. 2014 Impacts, Adapt. Vulnerability Part B Reg. Asp. Work. Gr. II Contrib. to Fifth Assess. Rep. Intergov. Panel Clim. Chang.* **2015**, *347* (6223), 1655–1734.

(10) Eerkes-Medrano, D.; Thompson, R. C.; Aldridge, D. C. Microplastics in Freshwater Systems: A Review of the Emerging Threats, Identification of Knowledge Gaps and Prioritisation of Research Needs. *Water Res.* **2015**, *75*, 63–82.

(11) Gasperi, J.; Dris, R.; Bonin, T.; Rocher, V.; Tassin, B. Assessment of Floating Plastic Debris in Surface Water along the Seine River. *Environ. Pollut.* **2014**, *195*, 163–166.

(12) Sadri, S. S.; Thompson, R. C. On the Quantity and Composition of Floating Plastic Debris Entering and Leaving the Tamar Estuary, Southwest England. *Mar. Pollut. Bull.* **2014**, *81* (1), 55–60.

(13) Hendrickson, E.; Minor, E. C.; Schreiner, K. Microplastic Abundance and Composition in Western Lake Superior As Determined via Microscopy, Pyr-GC/MS, and FTIR. *Environ. Sci. Technol.* **2018**, *52* (4), 1787–1796.

(14) Klein, S.; Worch, E.; Knepper, T. P. Occurrence and Spatial Distribution of Microplastics in River Shore Sediments of the Rhine-Main Area in Germany. *Environ. Sci. Technol.* **2015**, *49* (10), 6070–6076.

(15) Worm, B.; Lotze, H. K.; Jubinville, I.; Wilcox, C.; Jambeck, J. Plastic as a Persistent Marine Pollutant. *Annu. Rev. Environ. Resour.* **2017**, *42* (1), 1–26.

(16) Wright, S. L.; Thompson, R. C.; Galloway, T. S. The Physical Impacts of Microplastics on Marine Organisms: A Review. *Environ. Pollut.* **2013**, *178*, 483–492.

(17) Meeker, J. D.; Sathyanarayana, S.; Swan, S. H. Phthalates and Other Additives in Plastics: Human Exposure and Associated Health Outcomes. *Philos. Trans. R. Soc. London B Biol. Sci.* **2009**, *364* (1526), 2097–2113.

(18) Villberg, K.; Mussalo-Rauhamaa, H.; Haahtela, T.; Saarela, K. Prevalence of Plastic Additives in Indoor Air Related to Newly Diagnosed Asthma. *Indoor Built Environ.* **2008**,

17 (5), 455–459.

(19) Rahman, M.; Brazel, C. S. The Plasticizer Market: An Assessment of Traditional Plasticizers and Research Trends to Meet New Challenges. *Prog. Polym. Sci.* **2004**, *29* (12), 1223–1248.

(20) Lambert, S.; Scherer, C.; Wagner, M. Ecotoxicity Testing of Microplastics: Considering the Heterogeneity of Physicochemical Properties. *Integr. Environ. Assess. Manag.* **2017**, *13* (3), 470–475.

(21) Som, C.; Berges, M.; Chaudhry, Q.; Dusinska, M.; Fernandes, T. F.; Olsen, S. I.; Nowack, B. The Importance of Life Cycle Concepts for the Development of Safe Nanoproducts. *Toxicology* **2010**, *269* (2–3), 160–169.

(22) Brunner, P. H.; Rechberger, H. *Practical Handbook of Material Flow Analysis*; Lewis Publishers, 2004; http://thecitywasteproject.files.wordpress.com/2013/03/practical_handbook-of-material-flow-analysis.pdf.

(23) Schelker, R.; Geisselhardt, P. *Projekt „Kunststoff-Verwertung Schweiz ": Bericht Module 1 Und 2*; 2011; https://www.bafu.admin.ch/dam/bafu/fr/dokumente/abfall/externe-studien-berichte/projekt_kunststoff-verwertungschweiz.pdf.download.pdf/projekt_kunststoff-verwertungschweiz.pdf.

(24) Rewlutthum, K. Evaluation of Plastic Waste Management in Thailand Using Material Flow Analysis, Ph.D. Dissertation, Asian Institute of Technology, School of Environment, Resources and Development, 2013.

(25) Laner, D.; Feketitsch, J.; Rechberger, H.; Fellner, J. A Novel Approach to Characterize Data Uncertainty in Material Flow Analysis and Its Application to Plastics Flows in Austria. *J. Ind. Ecol.* **2016**, *20* (5), 1050–1063.

(26) Waste Watch. *Plastics in the UK Economy: A Guide to Polymer Use and the Opportunities for Recycling*; 2003; http://legis.wisconsin.gov/lc/committees/study/2010/SUP/files/1 Plastics in the UK economy .pdf.

(27) Salmons, R.; Mocca, E. *Material Flow Analysis and Value Chain Analysis for the UK Plastics Sector*; 2010; http://www.psi.org.uk/pdf/2011/plastics_VCA_final.pdf.

(28) Joosten, L. A. J.; Hekkert, M. P.; Worrell, E. Assessment of the Plastic Flows in The Netherlands Using STREAMS. *Resour. Conserv. Recycl.* **2000**, *30* (2), 135–161.

(29) Patel, M. K.; Jochem, E.; Radgen, P.; Worrell, E. Plastics Streams in Germany—an Analysis of Production, Consumption and Waste Generation. *Resour. Conserv. Recycl.* **1998**, *24* (3–4), 191–215.

(30) Fehringer, R.; Brunner, P. H. *Kunststoffflüsse Und Möglichkeiten Der Kunststoffverwertung in Österreich*; 1997;

http://www.umweltbundesamt.at/fileadmin/site/publikationen/M080.pdf.

(31) Kaps, R. Comparative Environmental Evaluation of Plastic Waste Management at National Level on Example of Polish and Austrian Systems, Ph.D. Dissertation, Poznan University of Economics, 2008.

(32) Bogucka, R.; Kosińska, I.; Brunner, P. H. Setting Priorities in Plastic Waste Management - Lessons Learned from Material Flow Analysis in Austria and Poland. *Polimery* **2008**, *53* (1), 55–59.

(33) Vujic, G. V.; Jovicic, N. M.; Babic, M. J.; Stanisavljevic, N. Z.; Batinic, B. M.; Pavlovic, A. R. Assessment of Plastic Flows and Stocks in Serbia Using Material Flow Analysis. *Therm. Sci.* **2010**, *14* (SUPPL.1), 89–96.

(34) Mutha, N. H.; Patel, M.; Premnath, V. Plastics Materials Flow Analysis for India. *Resour. Conserv. Recycl.* **2006**, *47* (3), 222–244.

(35) Tukker, A.; Kleijn, R.; Van Oers, L.; Smeets, E. R. W. Combining SFA and LCA: The Swedish PVC Analysis. *J. Ind. Ecol.* **1997**, *1* (4), 93–116.

(36) Ciacci, L.; Passarini, F.; Vassura, I. The European PVC Cycle: In-Use Stock and Flows. *Resour. Conserv. Recycl.* **2017**, *123*, 108–116.

(37) Nakamura, S.; Nakajima, K.; Yoshizawa, Y.; Matsubae-Yokoyama, K.; Nagasaka, T. Analyzing Polyvinyl Chloride in Japan with The Waste Input-Output Material Flow Analysis Model. *J. Ind. Ecol.* **2009**, *13* (5), 706–717.

(38) Zhou, Y.; Yang, N.; Hu, S. Industrial Metabolism of PVC in China: A Dynamic Material Flow Analysis. *Resour. Conserv. Recycl.* **2013**, *73*, 33–40.

(39) Kuczenski, B.; Geyer, R. Material Flow Analysis of Polyethylene Terephthalate in the US, 1996-2007. *Resour. Conserv. Recycl.* **2010**, *54* (12), 1161–1169.

(40) Rochat, D.; Binder, C. R.; Diaz, J.; Jolliet, O. Combining Material Flow Analysis, Life Cycle Assessment, and Multiattribute Utility Theory. *J. Ind. Ecol.* **2013**, *17* (5), n/a-n/a.

(41) Haupt, M.; Vadenbo, C.; Hellweg, S. Do We Have the Right Performance Indicators for the Circular Economy?: Insight into the Swiss Waste Management System. *J. Ind. Ecol.* **2017**, *21* (3), 615–627.

(42) Van Eygen, E.; Laner, D.; Fellner, J. Circular Economy of Plastic Packaging: Current Practice and Perspectives in Austria. *Waste Manag.* **2018**, *72*, 55–64.

(43) PlasticsEurope. *Plastics – the Facts 2014/2015: An Analysis of European Plastics Production, Demand and Waste Data*; 2015; https://www.plasticseurope.org/application/files/5515/1689/9220/2014plastics_the_facts_PubFeb2015.pdf.

(44) PlasticsEurope. *Plastics – the Facts 2012: An Analysis of European Plastics Production,*

Demand and Waste Data for 2011.; 2012; https://www.plasticseurope.org/en/resources/publications/109-plastics-facts-2012.

(45) Applied Market Information Ltd. *AMI's 2015 EUROPEAN PLASTICS INDUSTRY REPORT*; 2015.

(46) Consultic Marketing & Industrieberatung GmbH. *Produktion, Verarbeitung Und Verwertung von Kunststoffen in Deutschland 2011 - Kurzfassung*; 2012.

(47) Consultic Marketing & Industrieberatung GmbH. *Produktion, Verarbeitung Und Verwertung von Kunststoffen in Deutschland 2007 - Kurzfassung*; 2008.

(48) Consultic Marketing & Industrieberatung GmbH. Vom Land ins Meer – Modell zur Erfassung landbasierter Kunststoffabfälle http://www.bkv-gmbh.de/infothek/studien.html.

(49) Consultic Marketing & Industrieberatung GmbH. *Produktion, Verarbeitung Und Verwertung von Kunststoffen in Deutschland 2013 - Kurzfassung*; 2014.

(50) Consultic Marketing & Industrieberatung GmbH. Produktion, Verarbeitung und Verwertung von Kunststoffen in Deutschland 2015 - Kurzfassung http://www.kunststoffverpackungen.de/show.php?ID=5953.

(51) Consultic Marketing & Industrieberatung GmbH. *Analyse Der PVC Produktion, Verwertungsströme in Deutschland 2013*; 2013.

(52) LOI n° 2016-1087 du 8 août 2016 pour la reconquête de la biodiversité, de la nature et des paysages (1) https://www.legifrance.gouv.fr/eli/loi/2016/8/8/2016-1087/jo/texte (accessed Mar 20, 2018).

(53) European Commission. *A European Plastics Strategy For Plastics In A Circular Economy*; 2017; https://ec.europa.eu/commission/sites/beta-political/files/plastics-factsheet-people-environment_en.pdf.

(54) Aizenshtein, E. M. Global and Russian Output of Polyester Fibres in 2013. *Fibre Chem.* **2015**, *47* (1), 1–7.

(55) Gottschalk, F.; Scholz, R. W.; Nowack, B. Probabilistic Material Flow Modeling for Assessing the Environmental Exposure to Compounds: Methodology and an Application to Engineered Nano-TiO2 Particles. *Environ. Model. Softw.* **2010**, *25* (3), 320–332.

(56) Mueller, N. C.; Buha, J.; Wang, J.; Ulrich, A.; Nowack, B. Modeling the Flows of Engineered Nanomaterials during Waste Handling. *Environ. Sci. Process. Impacts* **2012**, *15* (1), 251–259.

(57) Caballero-Guzman, A.; Sun, T.; Nowack, B. Flows of Engineered Nanomaterials through the Recycling Process in Switzerland. *Waste Manag.* **2015**, *36*, 33–43.

(58) Gottschalk, F.; Nowack, B. A Probabilistic Method for Species Sensitivity Distributions Taking into Account the Inherent Uncertainty and Variability of Effects to Estimate Environmental Risk Health & Ecological Risk Assessment. *Integr. Environ. Assess. Manag.* **2012**, *9* (1), 79–86.

(59) Eurostat. Eurostat PRODCOM database http://ec.europa.eu/eurostat/web/prodcom/data/database#.

(60) Eurostat. EU trade since 1988 by HS2,4,6 and CN8 http://ec.europa.eu/eurostat/web/international-trade-in-goods/data/database#.

(61) Swiss Federal Customs Administration FCA. SwissImpex Version 3.4.2-1 https://www.swiss-impex.admin.ch/index.xhtml (accessed Mar 31, 2017).

(62) Rechberger, H.; Cencic, O.; Frühwirth, R. Uncertainty in Material Flow Analysis. *J. Ind. Ecol.* **2014**, *18* (2), 159–160.

(63) Vinyl 2010. *Progress Report 2015*; 2015; https://vinylplus.eu/uploads/docs/VinylPlus_Progress_Report_2015_English.pdf.

(64) Szeteiová, K. Automotive Materials. *Inst. Prod. Technol. Sloval Univ. Technol. Bratislava* **2010**, 27–33.

(65) Kettler, R. Statistique Des Déchets 2000, Avec Données 2001 Sur La Planification Des UIOM. *Doc. Environ. Off. fédéral l'environnement, des forêts du paysage* **2001**, *152*, 95.

(66) Huisman, J.; Botezatu, I.; Herreras, L.; Liddane, M.; Hintsa, J.; Luda di Cortemiglia, V.; Leroy, P.; Vermeersch, E.; Mohanty, S.; van den Brink, S.; Ghenciu, B.; Dimitrova, D.; Nash, E.; Shryane, T., Wieting, M.; Kehoe, J.; Baldé, C.P.; Magalini, F.; Zanasi, A.; Ruini, F.; Männistö, T.; and Bonzio, A. *Countering WEEE Illegal Trade Summary Report*; Lyon, France, 2015; http://www.cwitproject.eu/wp-content/uploads/2015/08/CWIT-Final-Summary1.pdf.

(67) Simon, C.; Hupfer, C.; PlasticsEurope. *Business Data and Charts 2015/2016: Plastics Europe Market Research Group*; 2016.

(68) Cox, R. T. Probability, Frequency and Reasonable Expectation. *Am. J. Phys.* **1946**, *14* (1), 1–13.

(69) Achilias, D. S.; Roupakias, C.; Megalokonomos, P.; Lappas, A. A.; Antonakou, V. Chemical Recycling of Plastic Wastes Made from Polyethylene (LDPE and HDPE) and Polypropylene (PP). *J. Hazard. Mater.* **2007**, *149* (3), 536–542.

(70) Geyer, R.; Jambeck, J. R.; Law, K. L. Production, Use, and Fate of All Plastics Ever Made. *Sci. Adv.* **2017**, *3* (7), 25–29.

(71) Bornhöft, N. A.; Sun, T. Y.; Hilty, L. M.; Nowack, B. A Dynamic Probabilistic Material Flow Modeling Method. *Environ. Model. Softw.* **2016**, *76*, 69–80.

(72) Müller, E.; Hilty, L. M.; Widmer, R.; Schluep, M.; Faulstich, M. Modeling Metal Stocks and

Flows: A Review of Dynamic Material Flow Analysis Methods. *Environ. Sci. Technol.* **2014**, *48* (4), 2102–2113.

(73) Eurostat. Population change - Demographic balance and crude rates at national level [demo_gind] http://ec.europa.eu/eurostat/data/database?node_code=proj# (accessed Sep 27, 2017).

7 Polymer-specific modelling of the environmental emissions of seven commodity plastics as macro- and microplastics

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7.1 Abstract

Plastic has been identified as emerging contaminant in aquatic and terrestrial ecosystems. Uncertainties remain concerning the amounts present in the environment and on the main responsible sources. In this study, the emissions of macro- and microplastic have been mapped for seven polymers in Switzerland. The modelling is based on a complete analysis of the flows from production over use to end-of-life using probabilistic material flow analysis. We estimate that 94±34g/capita/year of low-density polyethylene, 98±50g/cap/a of high-density polyethylene, 126±43g/cap/a of polypropylene, 24±13g/cap/a of polystyrene, 16±12g/cap/a of expanded polystyrene, 65±36g/cap/a of polyvinyl chloride and 200±120g/cap/year of polyethylene terephthalate enter the Swiss environment. Combining all, 540±140g/cap/a are emitted to soil as macroplastic and 73±14g/cap/a as microplastic, 13.3±4.9g/cap/a to freshwater as macroplastic and 1.8±1.1g/cap as microplastic. The leading emission pathway is littering for both terrestrial and aquatic environments. Construction, agriculture and pre- and post-consumer processes cause important emissions for microplastics in soils and post-consumer processes, textiles and personal care products release most of microplastics to waters. Because mass flows to soils are predicted to be 40 times larger than to waters, more attention should be placed on this compartment. Our work also highlights the importance of referring to specific polymers instead of "plastics".

7.2 TOC-Art

7.3 Introduction

Microplastics (MP) have been reported in numerous freshwater systems in Europe[1–3], North America[4], Africa[5], Asia[6–8], both in densely populated areas[9] and remote systems[10]. The presence of MP in soil is not as well documented as in freshwater since accurate methods for measuring MP concentrations in soil are still being developed[11–13] however first estimates hint that burdens in soil may be considerable[14,15]. MP have been reported in soils where sludge has been applied[16] although also in areas with reduced direct human impacts[11]. Main sources of MP to soil have been suggested to be compost and sludge application onto land, mulching plastics, littering, street runoff and atmospheric deposition[17]. Emissions to outdoor and indoor air have so far only been marginally addressed[18].

The risk associated with MP is currently under debate[19–22] due to challenges arising in both hazard and exposure assessments. Current preliminary risk assessments predict little to no risk on average in marine[23] and freshwater[24] environments. In order for science to support actions from policy makers and citizens, a solid assessment of the present degree of pollution needs to be undertaken. MP emission assessments have been performed for Norway[25], Denmark[26], Germany[27,28], Sweden[29], Europe[30] and the whole world[31]. The receiving environmental compartment is sometimes not mentioned[25,28] or the focus is only on aquatic environments[26,27,29–31]. Existing estimates for soil burdens are only preliminary and call for more precise assessments[17,32]. MP originating from macroplastic fragmentation are often outside of the scope of published MP-release studies[26,29–31] or based on a rough estimate of the fraction of mismanaged waste[25,27]. In one instance, the method of quantifying release remains unclear[28]. Some published emission assessments have focussed entirely on the release of macroplastic to the oceans[33,34], likewise based on roughly estimated fractions of mismanaged waste[35]. To this day

large segments of the emission pathways are not well known[32], since data on specific release processes is scarce and subject to high variability.

To the best of our knowledge, no release model has systematically investigated the material composition of the emission flows. The distinction between different polymers for the exposure assessment is needed for several reasons. First, a risk assessment should ideally distinguish individual materials, since toxicities may be different depending on the material itself and the additives included. Since release pathways may also be very different depending on the life-cycle of the material[36], the exposure is also polymer dependent. Secondly, if fate models are to be developed, a difference between lower and higher material densities needs to be made. At last, for the implementation of the MP release into Life Cycle Inventories (LCI), a distinction of material and life-cycle stage is needed.

The goal of this study is therefore to quantify the emissions of different plastics based on a complete analysis of the life-cycle of all products containing the chosen materials. This modelling is undertaken for different polymers with an increased level of detail compared to existing studies and targets both macro- and microplastics. This allows us to compare in a complete picture the different emission sources of plastic to the environment, identify the polymers emitted and pinpoint possible points of action. The model focusses on emissions to water and soil environments without including fate processes such as fragmentation of macro- to microplastics. The modelling is based on probabilistic material flow analysis (PMFA)[37] which permits to account for the various uncertainties associated to the data sources. Moreover, by considering the whole life-cycle, all emissions may easily be compared to one another and no double accounting occurs. The modelling was done separately for seven of the most highly used thermoplastics. Switzerland was used as geographic region due to the high availability of data.

7.4 Method

7.4.1 Algorithm

Material Flow Analysis (MFA) is an established method for analysing the flows of materials through the anthroposphere in a systematic fashion[38,39]. The probabilistic MFA (PMFA) method used in this work builds upon MFA by including a systematic uncertainty analysis and propagation using Monte Carlo simulations[37]. This method has already been used for modelling the flows of polymers through the anthroposphere[40] as well as several other materials[41–43]. The basic principle of the modelling relies on two mathematical objects. The first object is a Transfer

Coefficient (TC) matrix, which describes all the flows from one process to another. The second object is an input vector containing all the external inputs to the defined system. The matrix and vector constitute a system of linear equations, which is solved using matrix inversion. This matrix equation is solved 10^5 times in a Monte-Carlo set-up, where every element is randomly taken from predefined probability distributions depending on data availability and quality[44,45]. A schematic description of the method is given in the supplementary information (Figure S52) and more details are available in the original publications[37,40]. For simplicity, most of the results will be presented as the mean and standard deviation of the distributions where the mean is rounded to the second significant digit of the standard deviation[46].

7.4.2 Materials

The polymers chosen for this analysis, are identical to the ones for which the flows within the anthroposphere were modelled in a previous study[40]: low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), expanded polystyrene (EPS), polyvinyl chloride (PVC) and polyethylene terephthalate (PET). These polymers were chosen based on their popularity of use[47] and the frequency at which they were reported in freshwaters[48,49]. No additives are included in the masses reported. More details on the material definition are available in the first study[40].

7.4.3 Flows considered

The basis for assessing the flows of the polymers to the environment is the modelled life-cycle of the polymers within the anthroposphere[40]. Some adjustments to the life-cycle description were necessary compared to the previous study to permit the modelling of emission flows. First of all, additional product categories were included to the 35 already defined in the previous study: wet wipes, sanitary pads, panty liners, tampons, tampon applicators, cotton swabs, disposable cutlery, straws and shotgun cartridges. Second, a distinction between pre-consumer waste generated by textile or plastic industries was made in order to model the emissions of pellets more accurately (Figure S67).

Building on the description of the life-cycle, emission flows were included describing the initial emission and the pathways followed until the final release into the environment (Figure S68-Figure S72 and Figure S62). Emissions of macroplastic and MP are tracked separately by creating a compartment for both sizes of plastic in our system. This permits to account for the different processes responsible for the MP release to freshwater and soils. MP emissions occur for either MP that is designed as particles for their use, as for example pre-production pellets

and MP in personal care and cosmetic products (PCCP), or for MP generated from the wear of products, for instance fibres shed from textiles (SI chapter 4). MP can also originate from the abiotic or biotic degradation of macroplastic which creates MP. In order to account for all types of sources, emission flows of both MP and macroplastic are considered. No fate processes are implemented in water and soil environments. A simple air deposition model is nevertheless implemented since particles do not stay in air for longer time periods. The emissions of MP and macroplastic will be presented separately throughout this study. MP includes all particles smaller than 5 mm, with no strict lower size threshold since the different data underlying the model may have very varying lower thresholds, and macroplastic includes all pieces larger than 5 mm.

A total of 168 flows were used to model the life-cycle of the polymers in Switzerland on the basis of year 2014, mostly based on the original study[40]. They represent the flows of plastic through production, manufacturing, use and end-of-life. In addition, 234 flows were built to model the emission pathways starting from the individual processes in the life-cycle. For both MP and macroplastic emissions, the emission pathways can either be direct or follow more intricate pathways by flowing through several technical compartments before reaching the environment. Both soil and freshwater environments are modelled as final environmental compartments, for which the emissions as MP and macroplastic are separately recorded. A further distinction between residential soil, natural soil, agricultural soil, road sides and subsurface soil is made. The definition of residential, natural and agricultural soil follow the classification of surface use in Switzerland[50]. Road sides are defined as the immediate surrounding of highways and smaller roads. The subsurface soil compartment was defined for the release of MP due to sewer exfiltration and the MP lost from geotextiles in use. Emissions from industrial activities are released to residential soil or road sides depending on the emission process, plastic and textiles from agricultural origin will be emitted to agricultural soil. MP in outdoor air was redistributed to soil or water environments applying statistics on the land use as a coarse deposition model.

Many parameters involved in the modelling of the emissions are identical for the seven polymers since the release mostly depends on the processes. The largest differences in parameters among the seven polymers are found in the life-cycle[40]. A short description of the release pathways considered is presented in the following subsections and in Table S18. A complete description of the literature and data used for the modelling of the release flows is presented in the Supporting Information (SI).

7.4.3.1 Emissions of MP due to wear processes

Most of the products in use undergo wear processes, which are implemented in varying levels of detail depending on the literature available. All textile applications are subject to wear in different proportions. Emissions of MP fibres (MPF) from clothing and household textiles by washing, drying and wearing are included. These estimates are based on the amounts of MPF released during one cycle of washing[51–55], drying[54] or wearing[56–58], the number of cycles during the whole lifetime of the products and other parameters which were required for the implementation of the original data. The technical textiles are subject to an assumed standard shedding rate[59]. The whole description for these and other emissions is available in the SI.

7.4.3.2 Intentionally mismanaged waste

Four distinct processes are modelled for the mismanaging of waste: littering, dumping, flushing and contamination of organic waste. A short description of the approach is given in this section. For more details, we refer to the SI.

Littering: Plastic products which are used away from home may be littered instead of discarded in bins[60]. In order to model this flow, several intermediate steps were introduced. Since only the products which are used away from home may be subject to this process, the consumption of products on-the-go was estimated. In a second step, a distinction was made between products used in residential or natural areas or in the car. Once the consumption of plastic in these three areas is known, a location-dependent littering probability is applied. The non-littered portion is collected with mixed waste. The final step is the modelling of the sweeping of the litter. A fraction of the litter is collected and flows to mixed waste, and the rest flows to soil, surface water and storm water. The products which may be subject to littering are consumer films, bags and bottles, other consumer packaging, cutlery and straws.

Dumping: Mismanaged waste also arises in a more premeditated manner than littering, with all the waste that is deliberately dumped outdoors[61]. This process is not limited to consumer products used on-the-go and occurs for any used good. This process is implemented for all product categories in the model except cars and mobility textiles. The litter arising through this process is redistributed to litter in residential or natural areas or road sides and then follows the same pathways as litter arising through littering.

Flushing: Sanitary waste is often flushed down the toilet instead of discarded in the waste bin[62,63]. A flushing probability is attributed to wet wipes, sanitary pads, panty liners, tampons, tampon applicators and cotton swabs, which are then collected along with wastewater.

Contamination of organic waste: Various plastic products and types are found in collected organic waste[64–66]. The following products may be collected inadvertently with organic waste: consumer films, consumer bags, consumer bottles, other consumer packaging, agricultural packaging films, agricultural bottles, agricultural pipes and films, other agricultural plastics, agrotextiles, household plastic, straws and cutlery. A certain fraction of this plastic is removed during the processing of the organic waste and the rest is distributed to agricultural and residential soil. The processing of the organic waste also causes some of the plastic to be fragmented into MP.

Unintentionally mismanaged waste

Some of the products found in the environment, such as plastic films from construction or industry may be lost inadvertently due to weather or during transportation. This release is separate of plastic that is dumped or littered. Such a release is modelled for non-consumer packaging (non-consumer bags, non-consumer films, other non-consumer packaging), construction plastics (construction packaging films, pipes, insulation, coverings, profiles, lining) and agricultural plastics (agricultural packaging films, agricultural packaging bottles), and automotive. This litter then follows the same steps as littered waste before it is ultimately considered as released to the environment.

7.5 Results

7.5.1 Emission pathways

An aggregated overview of the emission flows is presented for PP in Figure 12 for MP and macroplastic simultaneously, and for the six remaining polymers in Figure S56-Figure S61. Seven hot spots for plastic release in Switzerland appear:

1. On-the-go consumption: for PET, HDPE, PP, LDPE, PS and PVC,
2. Post-consumer processes: for PVC, HDPE, EPS, PP and PS,
3. Use of plastic in agriculture: for LDPE and PP,
4. Construction and demolition sites: for PVC, HDPE, EPS and PS,
5. Flushing of hygiene products: for HDPE, PET and PP,

6. Collection with organic waste: for PP, HDPE, LDPE, PET and PS,
7. Textiles: for PET and PP.

LDPE is typically used for producing films and is largely used in packaging, construction and agriculture. The largest emissions originate from these sectors, with consumer packaging leading by a large margin via littering with 1380±450 t (Figure S56). From there, most of the emitted plastic is swept again by the authorities. A large portion nonetheless reaches the environment, mostly on soils. Large amounts of LDPE are also collected along with organic waste: 880±240 t from consumer packaging and 400±170 t from agriculture, where the vast majority is sorted out again. HDPE has a very broad range of applications which results in many different kinds of emissions (Figure S57). The largest initial release from products is found for consumer packaging littered (3'500±1'100 t), construction (250±130 t) and hygiene products (210±180 t). PP also has a very large range of possible applications, and therefore many types of emissions (Figure 12). Littering of consumer packaging again has the first position for the initial emission with 2'430±730 t littered, followed by items discarded in organic waste and flushed hygiene products. Large amounts of litter also arise from construction (114±73 t) and agricultural activities (360±210 t to soil and 273±80 t to organic waste collection). PS is mostly used in packaging, so littering plays a very important role in its release with a total of 760±230 t littered (Figure S58). Large amounts of PS are also released from non-consumer activities related to packaging, construction and waste management. EPS and PVC are mostly used in construction, which is again reflected in the predicted emissions (Figure S59 and Figure S60). 240±130 t EPS and 690±260 t PVC litter arises from construction activities, of which a large fraction is swept. Large emissions of EPS and PVC are also caused by the collection of construction waste. PVC is also subject to littering to a large extent with 235±88 t littered every year. Lastly, for PET, there is a very clear domination of release through littering with 10'100±2'800 t before sweeping (Figure S61). Large amounts of PET MPF are also released from textile applications, even though the amount initially released from textiles is two orders of magnitude lower than the amount of PET littered.

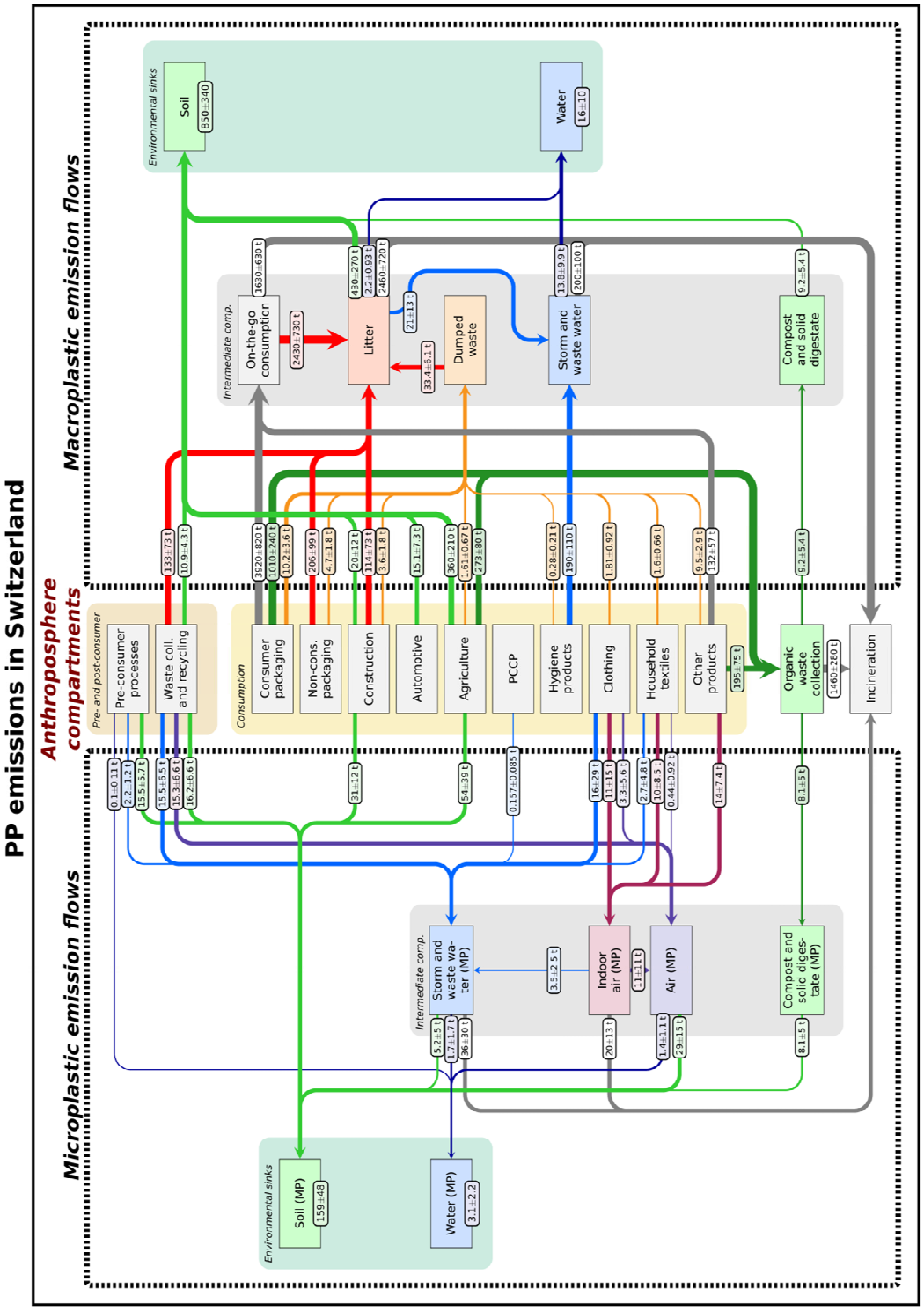

Figure 12: Aggregated emissions flows for PP in tonnes per year in Switzerland for macro- and microplastic in 2014. The aggregated flow distributions are represented rounded with their mean ± standard deviation. The life-cycle processes are aggregated for visualization purposes. The life-cycle flows (not shown) are calculated based on the existing life-cycle model[40] and generate the input into the aggregated compartments shown in the middle of the figure. The exact flow values for the individual compartments can be found in the table document of the SI. Equivalent flowcharts are available for the remaining polymers in Figure S56-Figure S61. The colour of the flows is representative of the receiving compartment, and the width of the flows qualitatively represents its magnitude. Abbreviations: waste collection (waste coll.), non-consumer packaging (non-cons. packaging), personal care and cosmetic product (PCCP).

For all mentioned polymers, one of the most preeminent emissions is almost always caused by littering. More details about the flows during littering are shown in Figure 13. The only parameters that are polymer-dependent for littering are the fractions of the packaging product categories used on-the-go, which were available for the seven materials[67]. Between 5% and 35% of the used consumer packaging applications are consumed away from home depending on the polymer and the product. Consumer bottles in particular, of which 35% are used away from home present a higher potential for littering. According to our estimates, 25% of this packaging used away from home are used in transportation, 10% in natural environments and the rest in residential areas. A total of 22'900±3'300 tonnes of litter of all seven polymers are generated along road sides, in residential and natural areas according to our model. The largest amount of litter generated arises for PET because of the high consumption of PET bottles and their high on-the-go consumption. As mentioned earlier, most of the litter arises in residential areas, followed by road sides and natural environments. Because of the different sweeping efficiencies considered, most litter remaining after sweeping is found along road sides and in natural environments. Quantitative information on the differences between the materials is given in the SI.

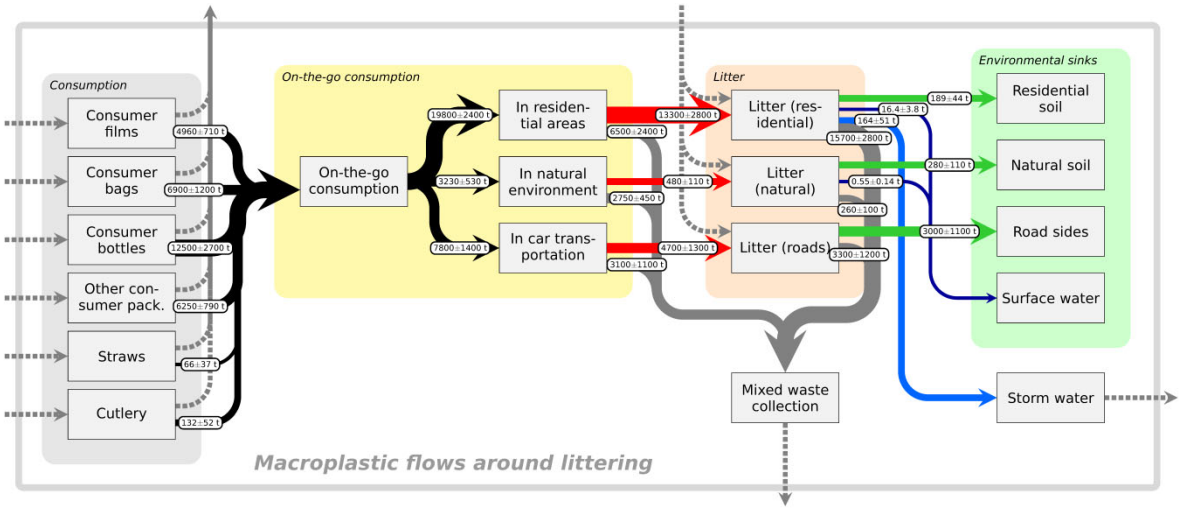

Figure 13: Detailed pathways for all flows connected to littering in Switzerland in tonnes per year, summed over all polymers. The colour of the flows is representative of the receiving compartment. Dashed flows represent existing flows for which the data is not represented. The flow distributions are represented rounded with their mean ± standard deviation.

The second most important release for most polymers is caused by mismanaged agricultural products. In total, 800±280 tonnes of the seven polymers are directly emitted to agricultural soil by burying after use. Most of this plastic consists of PP with 360±210 tonnes and LDPE with

340±180 tonnes buried, followed by 41±36 tonnes PET, 29±22 tonnes PVC, 24±12 tonnes HDPE and 2.2±2.1 tonnes PS.

Other important indirect emissions flows are caused by storm water and wastewater management (Figure S62). 600±300 t macroplastic and 210±150 t MP are emitted to wastewater every year (Figure S63). All of the macroplastic released to wastewater are hygiene articles flushed instead of appropriately discarded. Macroplastic can only escape WWTPs through combined sewer overflows (CSOs), except for cotton swabs which may escape secondary treatment. Nevertheless, cotton swabs are amongst the least flushed items in terms of mass along with cleaning cloths (Figure S63). Much larger masses of wet wipes and feminine hygiene products are flushed, however since 100% of the large macroplastic are retained in WWTPs overall, the emission of such items through the WWTP becomes negligible. The emissions caused by CSOs cause the largest emissions of these products with a total of 10.6±7.2 of all polymers and products combined. Less MP enter WWTPs than macroplastic, yet the emissions to surface water are almost as important. The largest MP releases to wastewater are caused by PCCP and clothing. Smaller releases originate from post-consumer processes, technical clothing and household textiles. The largest amounts of MP escape through CSOs with 6.2±4.7 t, and significant amounts also leave from tertiary treatment with 1.4±4.4 t. The largest emissions caused by wastewater nevertheless flow to sub-surface soils through exfiltration from sewers with around 23±19 t.

An important MP release pathway to soil and surface water environments occurs through air deposition. 32±35 tonnes of PET, 30±15 tonnes of PP, 11.7±6.2 tonnes of HDPE, 9.3±4.9 tonnes of PVC, 8.0±3.4 tonnes of PS, 3.3±1.4 tonnes of LDPE and 2.42±0.60 tonnes of EPS are transported through outdoor air and deposit onto soil or surface water (Figure 12 and Figure S56-Figure S61). PET emissions to outdoor air are dominated by emissions from indoor air of MPF from clothing and household textiles. Around 33% of the MP emitted to indoor air is released to outdoor air through air exchanges between the two environments in our model. The largest emissions to indoor air are caused by MPF shed from clothing and household textiles, which also contribute to the largest emissions to outdoor air after post-consumer processes. PP emissions to outdoor air are dominated by emissions from waste collection and recycling with 15.3±6.6 t, more specifically from automotive shredder residue and waste electrical and electronic plastic which are often shredded in the open. Large amounts of PP MP are also emitted to outdoor air from indoor environments with 11±11 t. Similarly for HDPE, PS, PVC and LDPE,

MP emissions to outdoor air are primarily caused by the recycling processes of cars and electronics. For PS and EPS, emissions from construction and demolition activities are also very relevant with 3.4±2.5 t PS and 2.42±0.60 t EPS.

An overview of the final emissions caused by the individual products is given in Figure 14, with aggregated emissions from pre- and post-consumer processes. The largest emissions to soil occur for consumer packaging as macroplastic through littering and from waste management and recycling. Large amounts of macroplastic are also released by burying of agricultural goods reaching their end-of-life. The next largest emission to soil is caused by cutting of construction pipes and agricultural films fragmenting during use, both generating MP. The total macroplastic emissions to soil sum up to 4400±1200 tonnes and the total MP emissions to 600±110 tonnes. Similarly, more macroplastic is emitted to freshwater than MP, with 109±41 tonnes of macroplastic and 14.9±8.8 tonnes of MP. Consumer packaging littered is the leading emission of macroplastic also for surface waters. Smaller yet relevant emissions are caused by MP released from recycling processes. Emissions of macroplastic by industrial activities, non-consumer packaging and construction sites are also relevant. Hygiene articles flushed and collected with wastewater also figure in the largest emissions of macroplastic to surface waters. Other MP emissions to surface water are caused primarily by clothing and PCCP, followed by fabric coatings, pre-consumer processes and household textiles.

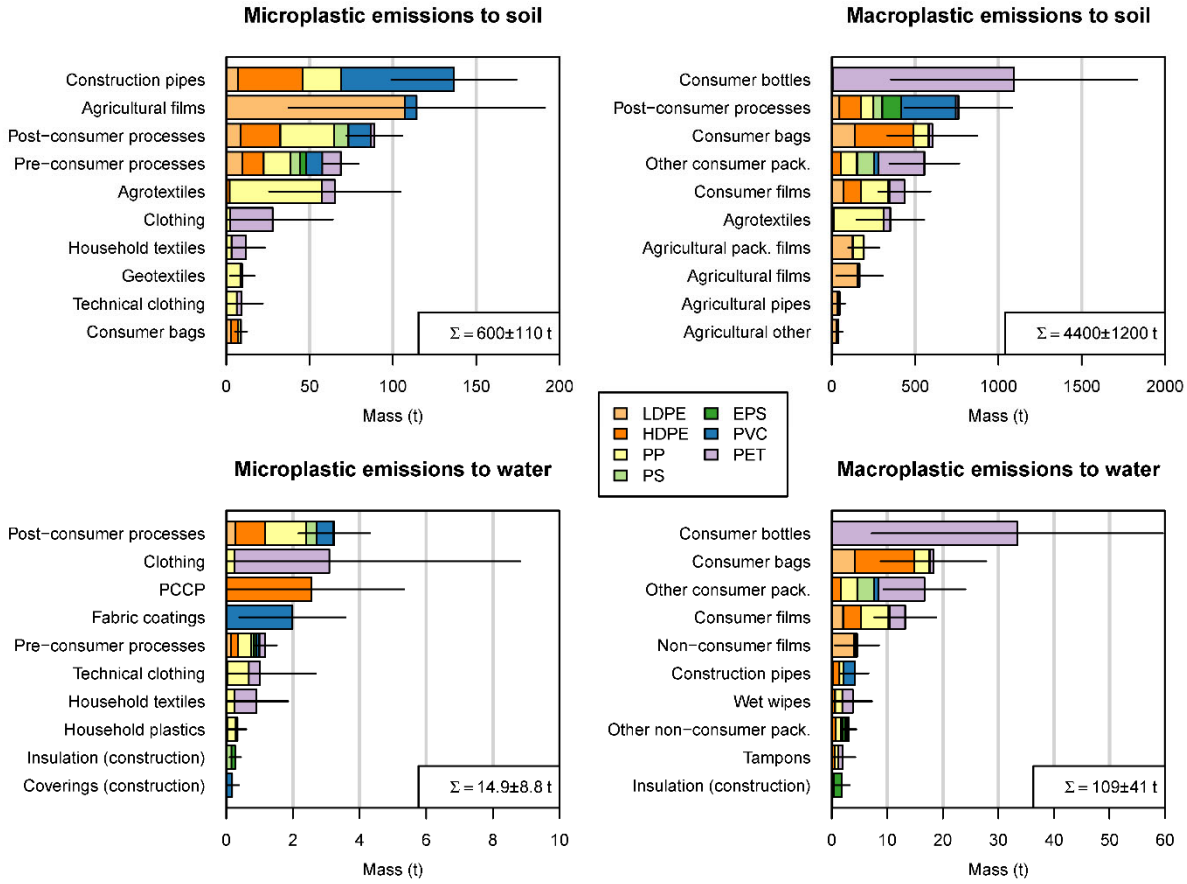

Figure 14: Macroplastic and MP emissions to soil and water in tonnes per year for Switzerland and their material composition. Only the ten largest contributions are shown. The contributions are shown by type of process, either product categories or pre- and post-consumer processes. The total release is shown in the lower right corner as mean ± standard deviation, rounded to two significant digits of the standard deviation.

7.5.2 Receiving environmental compartments

The distribution of the release among environmental compartments varies as a function of the polymer and the type of emission (Figure 15). The main reason for this variation is due to the different applications of each polymer. LDPE and PP MP are mostly emitted to agricultural soil due to the use of LDPE films and PP agrotextiles in agriculture. Around 80% of LDPE MP emissions flow to agricultural soil, and 45% of PP MP. The emissions of LDPE MP to other compartments are much lower in comparison. Around 30% of the PP MP is emitted to residential soil and only around 10% to natural soil. HDPE MP is mostly emitted to residential soil with 50% of the emissions because of non-consumer activities, followed by subsurface soil with around 30% because of the assumed wear of HDPE pipes in use. Around half of PS MP is emitted to residential soil because of non-consumer activities. Around two thirds of EPS and half of PVC MP emissions land in residential soil, because of pre-consumer processes in the case of EPS and construction activities for PVC. A large amount of PVC MP also ends in subsurface soil because

of the wear of pipes during use. PET MP is mostly emitted to agricultural soil (around 36%) through air deposition, wear of agrotextiles, and compost and digestate application onto land. All polymers combined, MP is mainly emitted to agricultural soil, followed by residential soil and subsurface soil (Figure S64).

Macroplastic of all polymers is found mostly on road sides (Figure 15) because of littered consumer packaging, and lost construction and demolition waste. An exception to this exists for LDPE and PP for which around half of the macroplastic is emitted to agricultural soil, caused by burying of agricultural products. All polymers combined, road side pollution constitutes 67% of all the macroplastic emissions (Figure S64). Agricultural and residential soils then contribute to around 24% of the emissions together. Only 6.3% of all the macroplastic emitted flows to natural soil, and 2.4% to surface water. It thus appears that for both MP and macroplastic emissions, most of the burdens are initially emitted to soils.

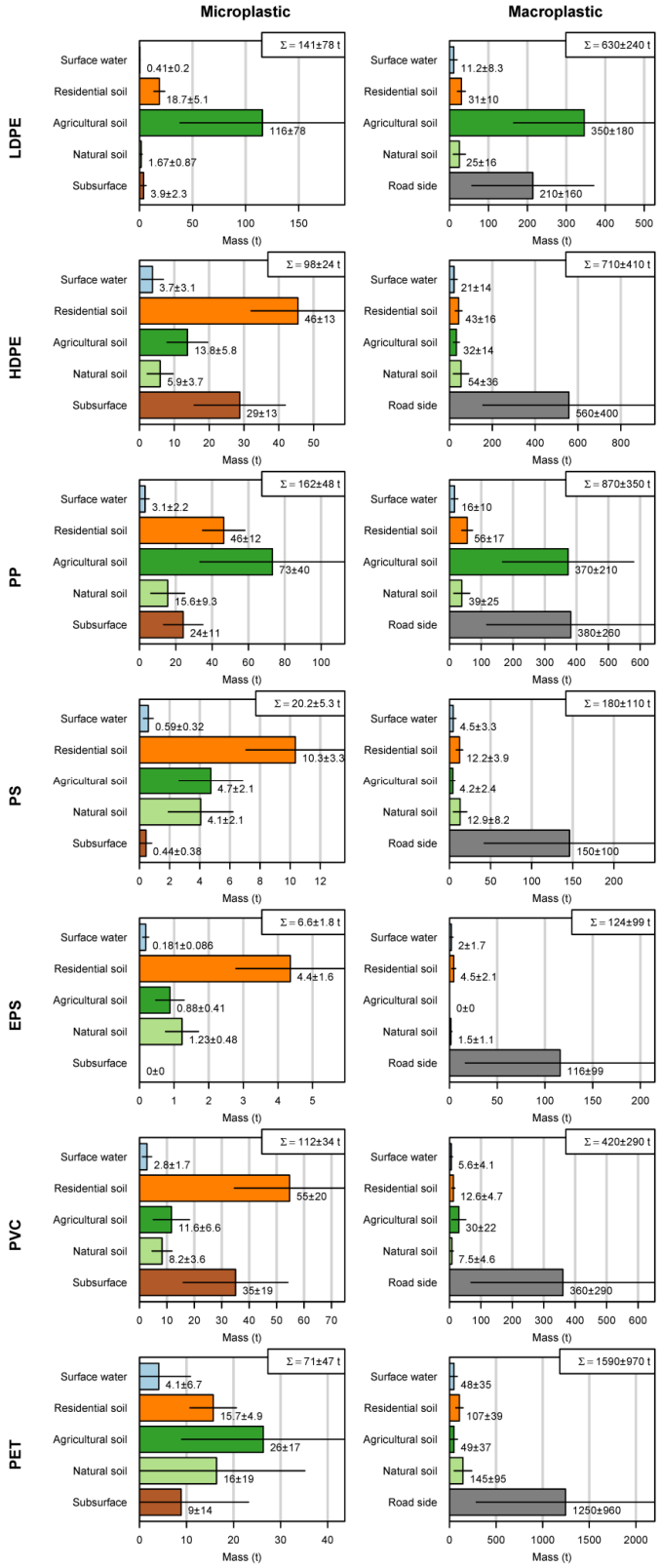

Figure 15: Comparison of the total emissions by polymer, type of emission (MP or macroplastic) and final compartment in tonnes per year for Switzerland. The total amount of polymer released as MP or macroplastic is displayed in the upper right corner as mean ± standard deviation, rounded to two significant digits of the standard deviation.

7.5.3 Emission factors

According to our model, 0.59±0.16 % of the mass of polymer consumed in Switzerland is released to the environment when neglecting emissions occurring outside of the consumption phase. This overall emission factor is different for the seven polymers: 0.64±0.23 % of LDPE, 0.53±0.28 % of HDPE, 0.57±0.19 % of PP, 0.24±0.14 % of PS, 0.037±0.012 % of EPS, 0.177±0.047 % of PVC and 1.25±0.73 % of PET.

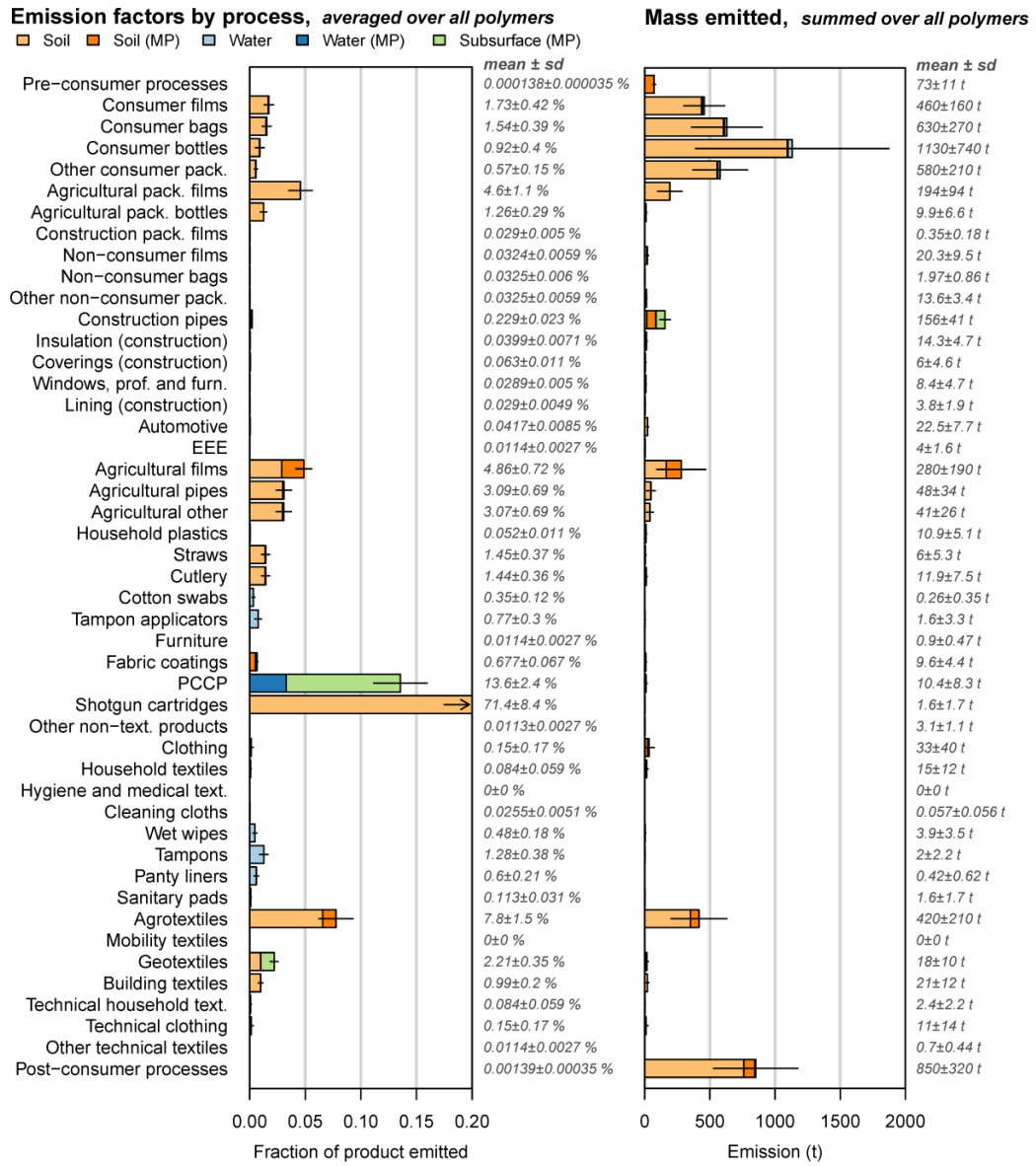

Figure 16: Emission factors (left) and total mass released in tonnes per year (right) by product and receiving environmental compartment for Switzerland. The emission factors are averaged over the seven different polymers and the emission masses are summed over the seven polymers. For "pre-consumer processes" and "post-consumer processes", a weighted average over the emissions from the sub-processes was calculated, using the mass in the sub-processes as weight. All displayed numbers are shown as mean ± standard deviation, rounded to two significant digits of the standard deviation. Abbreviations: Electrical and Electronic Equipment (EEE), Personal Care and Cosmetic Products (PCCP), textile (text.)

In Figure 16, the emission factors and the mass of the products emitted are compared for all product categories considered in the model and pre- and post-consumer processes. Consumer packaging is responsible for the largest macroplastic emissions to soil, followed by post-consumer processes, agricultural applications, construction pipes and pre-consumer processes. The remaining emissions are much smaller compared to these products. The largest emission factors are found for shotgun cartridges with 71.4±8.4 % of the cartridges consumed released directly into the environment during use. The second largest emission factor is found for PCCPs with 13.6±2.4 % of the total mass consumed being released, of which a large amount is released to subsurface soil through exfiltration from sewers. Agricultural products also have large emission factors, ranging from 7.8±1.5 % for agrotextiles to 1.26±0.29 % for bottles. Other products with large emission factors are consumer packaging, hygiene products, straws and cutlery.

7.6 Discussion

7.6.1 Overall release of polymers to the environment

Several assessments of macro- and microplastic emissions have already been published. Nevertheless, the results from different studies cannot easily be compared because of the differences in scope. More often than not, the largest emission flow in previous assessments originates from tyre wear[25,26,29,30] which consists of other polymers than those considered here. The materials included in the present study were chosen according to the prevalence of the polymer types measured in freshwater[40]. This issue highlights the importance to move away from referring to "plastics" to specific polymers.

For Europe, it was previously assumed that approximately 6-10% of the plastic produced was emitted to the oceans[27]. On a global scale, estimates of the amount of mismanaged waste reaching the oceans was estimated to be 2-5%[33]. The plastic litter input into the oceans from rivers was estimated based on the amount of mismanaged waste[34] by country around the world. Globally, 32% of all the plastic waste generated was modelled as mismanaged and after calibration with measurements in rivers, it was estimated that 1-2% of the plastic waste generated globally reached the oceans[34]. Such broad estimates are good first estimates, yet need to be improved by more accurate and specific numbers. Our results clearly show that using a generic release estimate for all existing plastic products is not feasible, as the life-cycle of the different applications is very different. Moreover, large regional differences may be expected,

depending on the regional waste management practices and global emission rates will not be representative of the emissions of one country.

A comparison by emission type can be done for the most important flows in this model, starting with the largest emission source of plastic to the environment according to the present study: littering. Previous estimates of quantities of plastic released due to littering use littering rates of around 2%[33]. This value combines the different parameters modelled in our study: on-the-go consumption, littering probability and sweeping efficiency. According to our results, 1.2±0.5 % of the products consumed on-the-go is released to the environment due to littering. However, the previous studies applied the littering rate to ***all*** plastic applications which leads to a large overestimation of the amount of plastic pollution caused by littering since only products used on-the-go may be littered.

Little information can be found on other emission flows in literature. Emissions from agriculture and construction which may both be very relevant are omitted from existing plastic emission assessments.

7.6.2 Per capita polymer emissions

In order to allow a comparison with other published plastic release estimates, we converted the released masses into per capita flows using a population of 8.19 million[68] in Switzerland for 2014. In total, 94±34 g/cap of LDPE enter the Swiss environment per year, aggregated over all environmental compartments and considering both MP and macroplastic. The corresponding numbers for the other polymers are 98±50 g/cap of HDPE, 126±43 g/cap of PP, 24±13 g/cap of PS, 16±12 g/cap of EPS, 65±36 g/cap of PVC and 200±120 g/cap of PET. All seven polymers combined, 630±150 g/cap of plastic enters the environment per year. Modelled emissions to soil are much higher than emissions to freshwater for all polymers: 610±150 g/cap to soil and 15.1±5.1 g/cap to water. 1.8±1.1 g/cap enter waters as MP and 13.3±4.9 g/cap as macroplastic. For soils the corresponding flows are 73±14 g/cap as MP and 540±140 g/cap as macroplastic. The difference in MP and macroplastic emissions is primarily due to the prevalence of emissions from consumer packaging for most polymers. We predict that soils receive 40-times more plastic (both macro- and microplastics) than waters. This means that much more focus should be placed on this compartment, both in relation to future research as well as for regulators.

Previous estimates of plastic emissions range from 8 g/cap[69] for MP added intentionally in products to 5400 g/cap[28] for many types of emissions. Comparing all of these studies together, our estimates are much smaller, especially for MP. It should however be noted that depending

on the study, tyre wear represents between 26[27] to 79[29]% of the total MP emissions. Publishing release estimates while distinguishing materials involved will therefore be central to assessing the impacts of synthetic microparticles. Tyre wear particles may be linked to entirely different environmental issues and risks[70] than particles consisting of non-rubber plastic[23,24]. Paints and coatings also represent large portions of the previously published estimates. Removing all the emission sources which are not included in the present study from previously published estimates, one obtains emissions of 129 g/cap[25], 3.6-95 g/cap[26], 4.7-32.25 g/cap[31]or 108 g/cap[30] for MP reaching aquatic environments or 858 g/cap[28] emitted without specification of the receiving compartments. Our estimate of MP reaching surface waters is with 1.8±1.1 g/cap lower than all these estimates. This highlights the importance of moving away from generic release estimates to more product- and polymer-specific assessments and also shows that using global values will not capture release flows in an industrialized country with an efficient waste management system.

7.6.3 Comparison with measured emissions

Although many measurements of MP in freshwater exist, it is difficult to use them for a comparison with our model results as we predict releases and not environmental exposure. Fate processes occurring after the emissions are not part of the model, which is very important in order to predict actual concentrations. The macroplastic and MP will be affected by sedimentation, runoff, fluvial transport, fragmentation and degradation. The presented data may serve as basis for the creation of such fate models[71] yet cannot predict environmental concentrations.

The order of magnitude of the modelled emissions can be put into perspective using the estimated amount of plastic litter on Swiss lakes and river shores obtained from the Swiss Litter Report by sampling of litter[72]. According to this study, around 276 t/year of plastic can be found along shores in Switzerland, assuming that the litter collections are independent for an interval of one month[73]. Our model estimates 109±41 t/year of macroplastic released to water, so the same order of magnitude than a value obtained from upscaling from a few sites to whole Switzerland.

For a few of the processes in our model a comparison with measured values can be made. The high amount of sanitary items in surface waters predicted by our model is correlated to the prevalence of such products in litter inventories from field studies. According to the Swiss Litter Report[72], one cotton swab is found on every 100 m^2 of shores in Switzerland per month. A rough estimation of the number of cotton bud sticks on river and lake sides can be made by

using the length of the hydrographic network of Switzerland and the length of the shorelines of the lakes[74]. Assuming a shore is 1-5 m broad, one cotton swab may be found every 20-100 m of shore every month, resulting in 2'900'000-40'000'000 cotton swabs per year. A total of 2'300'000 cotton swabs are emitted per year to surface waters in Switzerland according to our model, using the mass of a single cotton swab (see SI), which lies on the lower range of the estimate based on the Swiss Litter Report.

A comparison can also be made with the total amount of sanitary textiles collected on screenings in four different WWTPs in France, which were reported to range from 400-1700 g/cap/year[75]. The present model predicts that a total of 73±37 g/cap/year of macroplastic enter the WWTPs in Switzerland. This value encompasses only the seven polymers considered in the model which make up 6-73% of the composition of the sanitary product (see SI). This comparison tends to show that the amount of sanitary items flushed is not overestimated.

Emissions of MP through wastewater can be compared using measurements of MP burdens in wastewater in 28 WWTPs in the canton of Zurich[76]. From these measurements, it was estimated that 600 g/day of MP enter surface waters from the Canton of Zurich alone. Scaling up to Switzerland results in 1.2 tonnes of MP[77]. A total release of 1.5±4.6 tonnes MP through WWTPs from the seven polymers considered was estimated within this study, which is in good agreement with the estimate obtained from the measurements.

The predicted emissions to air can also be put into perspective with other approaches. According to our model, 98±47 tonnes of MP are emitted to outdoor air per year, originating to a large extent from post-consumer processes due to dust generated from shredding in recycling sites and from textile wear. This value can be compared to the reported atmospheric deposition of MPF in Paris[78]. Between 3 and 10 tonnes of fibres were estimated to deposit on the Parisian agglomeration every year, of which around 29% were synthetic[78]. By scaling to the area and the Swiss population, we can estimate that 0.57-1.97 t/year are depositing on soil and water environments. Two key uncertainties are worth mentioning: the shedding rate which can be applied to various types of textiles and the air exchange rate between indoor and outdoor air. The estimated dust mass that is deposited in households in Switzerland can be compared to dust deposition rates as known from literature[79–81] which are in the range of 1.1-2.4 g/m^2/year. The useful floor space in Switzerland is estimated to be 0.62 billion m^2 [82], resulting in approximately 680-1500 t/y of indoor dust deposited in Switzerland. The synthetic fibre content in dust was reported to be 1-5%[83] by volume in a household or 1.5%[84] by mass in an office. Con-

sequently, 6.8-34 tonnes of synthetic fibres would be a possible range of the amount of synthetic fibres depositing in indoor environments in Switzerland every year compared to a model prediction of 109±69 tonnes MP. The discrepancy of the two estimates by a factor of 3-16 may suggest that our evaluation of the fibre shedding processes may be overestimated. However, it is smaller than the discrepancy in the estimates of MP in outdoor air presented earlier. Very large uncertainties are attached to the dust generation rate and the useful floor space to which the dust generation rate is applied. The shedding rate of MPF used in the presented model may depend on parameters such as the activity performed, the type of garment, the cleanliness of the clothes and the ambient relative humidity[58]. Because of the lack of literature on emissions of fibres caused by wear and drying, their description was simplified. More research on the mechanics underlying these shedding processes as well as the various uses of textiles would clarify the importance of MPF shed indoors for the MP present in outdoor air.

7.6.4 Assessment of model uncertainty

The PMFA method enables to consider the uncertainty associated to each parameter used in the model. This uncertainty is then directly reflected in the final results which may have very varying relative uncertainties (Figure S66). These differences in confidence in the predicted results can easily be explained by considering the distribution of the parameters influencing it. The relative uncertainty on the mass contained in compartments ranges from 7 to 538 %, with a median relative uncertainty of 39 %. Low relative uncertainties are found for results depending on well-known parameters, and higher uncertainties are given for less confident predictions. In this manner, no false confidence in the results is given from the model. For example, a very high relative uncertainty is attributed to the amount of MP found in tertiary treatment in WWTPs, with a value of around 300% for most polymers and as high as 538% for PET. For all polymers, a strong influence on this final relative uncertainty comes from the high variability in removal efficiencies for the different treatment stages in the WWTP (see additional table document in the SI). The removal efficiencies during primary treatment range from 50%[85] to 98%[86], and during secondary treatment from 7%[86] to 81%[76]. In the case of PET, there is an important additional uncertainty coming from the release of fibres from textiles (Figure S73) where data scarcity and variability leads these parameters to have very long tails in their distributions and a high skewness. This leads the calculated standard deviation to reach very high values. This is reflected in the high relative uncertainty for the amount of MP released from clothing with 4.8±5.8 tonnes (Figure 16) and for the final amount of PET MP found in surface

waters with 4.1±6.7 tonnes (Figure 15). To conclude, while it can be expected to have considerable uncertainty in a few parameters considered in the model, this uncertainty is then reflected and presented in the confidence attributed to the results.

An additional uncertainty may come from the processes considered in the model itself. To reduce this uncertainty as much as possible, a high degree of detail was chosen for the modelling. Nevertheless, specific emission flows may be missing from the inventory if the corresponding processes are not part of the system. It is noteworthy to mention that fishing products were not included in the study, since the fishing industry has a low importance in Switzerland. However, if these results were to be applied to another country, regional differences should be taken into account. Other plastic emissions may be expected if their origin was not included in the modelled life-cycle, but we expect this to be of low importance for Switzerland, considering the low mass contained in the process "other products".

7.6.5 Policy recommendations

Comparing the emission factors and the mass of the product emitted gives insight into the efficiency of mitigation options. According to the total masses, consumer packaging would need to be examined first to reduce the exposure to plastic in the environment. A reduction of emission for these products would have the most impact on the total mass of plastic emitted. However, if one considers that only around 1% of the consumer packaging used causes such large emissions and that littering is a widespread phenomenon, mitigation seems more difficult. The next largest emission is found for post-consumer processes, for which very small emission factors are responsible. Most of these emissions are caused by construction sites and construction waste collection. Similarly as for consumer packaging, mitigation might be difficult. Agricultural products also cause considerable emissions and have large emission factors, making a possible mitigation easier. For products with great emission factors, there is a large potential for improvement on an individual scale. The products with the biggest emission factors are shotgun cartridges and PCCP, yet only 0.22±0.24 tonnes of shotgun cartridges and 1.5±1.2 tonnes of PCCP reach the environment. A large fraction of the PCCP released is flowing to subsurface soil due to exfiltration from sewers. When compared to the total 5300±1100 tonnes emitted yearly in Switzerland, reducing these emissions seems less urgent than the other ones.

From our analysis of the polymer flows to the environment, we can conclude that the following initiatives would be the most effective solutions to reduce plastic pollution:

1. Increasing the attention given to MP in soils where the largest flows are occurring. There is currently more information available on MP in waters likely because of the easier analytics and the experience gained in the oceans yet the modelling results clearly advocate for a much larger focus on soil.
2. Many of the largest emissions of macroplastic to soil and water are caused by single-use plastics (Figure 14). An action plan such as the one proposed by the European Parliament[87] may improve the situation, since it is meant to cover single-use plastics such as for example takeaway packaging and sanitary products.
3. Reducing littering and/or improving sweeping may have a large impact on the total environmental burden. Large efforts are already made in that direction, through education, campaigns and cleaning, and even though the efficiency of campaigns is debated, an improvement of the situation has been suggested over the last couple of years in Switzerland[88].
4. Improving the waste management practices in construction and agriculture would reduce the emissions caused by these sectors, which are only second to consumer packaging. It should nevertheless be highlighted that the emission flows from agriculture are very uncertain and not specific to Switzerland due to a lack of data and that the emission flows from construction are based on local data. More research into this area should be encouraged.
5. For further improvement, many measures aiming at incremental improvement may be suggested, for example reducing the occurrence of combined sewer overflows. Inhibiting shedding of MP fibres from textiles caused by washing, wearing and drying would reduce one of the largest emission of MP in surface waters and should be looked into more in detail. Similarly reducing the use of PCCPs would reduce emissions of MP in surface waters. For this specific product, media coverage and public attention have led companies to reduce their use of some polymers already[89]. Legal restriction of use of intentionally added MP in products has also been proposed[90].

7.7 Supporting information

1. PDF document: Additional figures for the method and the results, detailed description of parameters and assumptions underlying the model,
2. Parameter table: data used for the model and associated uncertainty,
3. Results table: Information on the flows resulting from the calculation for each polymer: mean, SD, quantiles.

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7.9 References

(1) Klein, S.; Worch, E.; Knepper, T. P. Occurrence and Spatial Distribution of Microplastics in River Shore Sediments of the Rhine-Main Area in Germany. *Environ. Sci. Technol.* **2015**, *49* (10), 6070–6076.

(2) Dris, R.; Gasperi, J.; Rocher, V.; Saad, M.; Renault, N.; Tassin, B. Microplastic Contamination in an Urban Area: A Case Study in Greater Paris. *Environ. Chem.* **2015**, *12* (5), 592–599.

(3) Imhof, H. K.; Ivleva, N. P.; Schmid, J.; Niessner, R.; Laforsch, C. Contamination of Beach Sediments of a Subalpine Lake with Microplastic Particles. *Curr. Biol.* **2013**, *23* (19), R867–R868.

(4) Eriksen, M.; Mason, S.; Wilson, S.; Box, C.; Zellers, A.; Edwards, W.; Farley, H.; Amato, S. Microplastic Pollution in the Surface Waters of the Laurentian Great Lakes. *Mar. Pollut. Bull.* **2013**, *77* (1–2), 177–182.

(5) Biginagwa, F. J.; Mayoma, B. S.; Shashoua, Y.; Syberg, K.; Khan, F. R. First Evidence of Microplastics in the African Great Lakes: Recovery from Lake Victoria Nile Perch and Nile Tilapia. *J. Great Lakes Res.* **2016**, *42* (1), 146–149.

(6) Zhang, W.; Zhang, S.; Wang, J.; Wang, Y.; Mu, J.; Wang, P.; Lin, X.; Ma, D. Microplastic Pollution in the Surface Waters of the Bohai Sea, China. *Environ. Pollut.* **2017**, *231*, 541–548.

(7) Zhang, K.; Gong, W.; Lv, J.; Xiong, X.; Wu, C. Accumulation of Floating Microplastics behind the Three Gorges Dam. *Environ. Pollut.* **2015**, *204*, 117–123.

(8) Sruthy, S.; Ramasamy, E. V. Microplastic Pollution in Vembanad Lake, Kerala, India: The First Report of Microplastics in Lake and Estuarine Sediments in India. *Environ. Pollut.* **2017**, *222*, 315–322.

(9) Dris, R.; Gasperi, J.; Rocher, V.; Tassin, B. Synthetic and Non-Synthetic Anthropogenic Fibers in a River under the Impact of Paris Megacity: Sampling Methodological Aspects and Flux Estimations. *Sci. Total Environ.* **2018**, *618*, 157–164.

(10) Free, C. M.; Jensen, O. P.; Mason, S. A.; Eriksen, M.; Williamson, N. J.; Boldgiv, B. High-Levels of Microplastic Pollution in a Large, Remote, Mountain Lake. *Mar. Pollut. Bull.* **2014**, *85* (1), 156–163.

(11) Scheurer, M.; Bigalke, M. Microplastics in Swiss Floodplain Soils. *Environ. Sci. Technol.* **2018**, *52* (6), 3591–3598.

(12) Rillig, M. C. Microplastic in Terrestrial Ecosystems and the Soil? *Environ. Sci. Technol.* **2012**, *46* (12), 6453–6454.

(13) Ng, E.-L.; Huerta Lwanga, E.; Eldridge, S. M.; Johnston, P.; Hu, H.-W.; Geissen, V.; Chen, D. An Overview of Microplastic and Nanoplastic Pollution in Agroecosystems. *Sci. Total Environ.* **2018**, *627*, 1377–1388.

(14) de Souza Machado, A. A.; Kloas, W.; Zarfl, C.; Hempel, S.; Rillig, M. C. Microplastics as an Emerging Threat to Terrestrial Ecosystems. *Glob. Chang. Biol.* **2017**, No. September.

(15) Nizzetto, L.; Futter, M.; Langaas, S. Are Agricultural Soils Dumps for Microplastics of Urban Origin? *Environ. Sci. Technol.* **2016**, *50* (20), 10777–10779.

(16) Zubris, K. A. V; Richards, B. K. Synthetic Fibers as an Indicator of Land Application of Sludge. *Environ. Pollut.* **2005**, *138* (2), 201–211.

(17) Bläsing, M.; Amelung, W. Plastics in Soil: Analytical Methods and Possible Sources. *Sci. Total Environ.* **2018**, *612*, 422–435.

(18) Gasperi, J.; Wright, S. L.; Dris, R.; Collard, F.; Mandin, C.; Guerrouache, M.; Langlois, V.; Kelly, F. J.; Tassin, B. Microplastics in Air: Are We Breathing It In? *Curr. Opin. Environ. Sci. Heal.* **2018**, *1*, 1–5.

(19) Hale, R. C. Are the Risks from Microplastics Truly Trivial? *Environ. Sci. Technol.* **2018**, *52* (3), 931–931.

(20) Burton, G. A. Stressor Exposures Determine Risk: So, Why Do Fellow Scientists Continue To Focus on Superficial Microplastics Risk? *Environ. Sci. Technol.* **2017**, *51* (23), 13515–13516.

(21) Backhaus, T.; Wagner, M. Microplastics in the Environment: Much Ado about Nothing ? A Debate. *PeerJPreprints* **2018**, No. May 2018, 0–5.

(22) Koelmans, A. A.; Besseling, E.; Foekema, E.; Kooi, M.; Mintenig, S.; Ossendorp, B. C.; Redondo-Hasselerharm, P. E.; Verschoor, A.; van Wezel, A. P.; Scheffer, M. Risks of Plastic Debris: Unravelling Fact, Opinion, Perception, and Belief. *Environ. Sci. Technol.* **2017**, *51* (20), 11513–11519.

(23) Everaert, G.; Van Cauwenberghe, L.; De Rijcke, M.; Koelmans, A. A.; Mees, J.; Vandegehuchte, M.; Janssen, C. R. Risk Assessment of Microplastics in the Ocean: Modelling Approach and First Conclusions. *Environ. Pollut.* **2018**, *242*, 1930–1938.

(24) Adam, V.; Yang, T.; Nowack, B. Toward an Ecotoxicological Risk Assessment of Microplastics: Comparison of Available Hazard and Exposure Data in Freshwaters. *Environ. Toxicol. Chem.* **2019**, *38* (2), 436–447.

(25) Sundt, P.; Schulze, P.-E.; Syversen, F. *Sources of Microplastic Pollution to the Marine Environment*; 2014;

https://www.miljodirektoratet.no/globalassets/publikasjoner/M321/M321.pdf.

(26) Lassen, C.; Hansen, S. F.; Magnusson, K.; Norén, F.; Bloch Hartmann, N. I.; Jensen, P. R.; Nielsen, T. G.; Brinch, A. *Microplastics: Occurrence, Effects and Sources of Releases to the Environment in Denmark*; The Danish Environmental Protection Agency, 2015; http://orbit.dtu.dk/files/118180844/Lassen_et_al._2015.pdf.

(27) Essel, R.; Engel, L.; Carus, M.; Ahrens, R. H. *Sources of Microplastics Relevant to Marine Protection in Germany*; 2015; https://www.umweltbundesamt.de/sites/default/files/medien/378/publikationen/texte_64_2015_sources_of_microplastics_relevant_to_marine_protection_1.pdf.

(28) Bertling, J.; Bertling, R.; Hamann, L. *Kunststoffe in Der Umwelt : Mikro- Und Makroplastik. Ursachen, Mengen, Umweltschicksale, Wirkungen, Lösungsansätze, Empfehlungen. Kurzfassung Der Konsortialstudie.*; Oberhausen, 2018; https://www.umsicht.fraunhofer.de/content/dam/umsicht/de/dokumente/publikationen/2018/kunststoffe-id-umwelt-konsortialstudie-mikroplastik.pdf.

(29) Magnusson, K.; Eliasson, K.; Fråne, A.; Haikonen, K.; Hultén, J.; Olshammar, M.; Stadmark, J.; Voisin, A. *Swedish Sources and Pathways for Microplastics to the Marine Environment. A Review of Existing Data*; 2016; https://www.naturvardsverket.se/upload/miljoarbete-i-samhallet/miljoarbete-i-sverige/regeringsuppdrag/2016/mikroplaster/swedish-sources-and-pathways-for-microplastics-to-marine environment-ivl-c183.pdf.

(30) Hann, S.; Sherrington, C.; Jamieson, O.; Hickman, M.; Kershaw, P.; Bapasola, A.; Cole, G. *Investigating Options for Reducing Releases in the Aquatic Environment of Microplastics Emitted by (but Not Intentionally Added in) Products Final Report*; 2018; http://www.eumicroplastics.com/eumpwp/wp-content/uploads/investigating-options-eunomia-draft-report-v4-main-report-public.pdf.

(31) Boucher, J.; Friot, D. *Primary Microplastics in the Oceans : A Global Evaluation of Sources*; 2017; https://portals.iucn.org/library/sites/library/files/documents/2017-002.pdf.

(32) Horton, A. A.; Walton, A.; Spurgeon, D. J.; Lahive, E.; Svendsen, C. Microplastics in Freshwater and Terrestrial Environments: Evaluating the Current Understanding to Identify the Knowledge Gaps and Future Research Priorities. *Sci. Total Environ.* **2017**, *586*, 127–141.

(33) Jambeck, J. R.; Geyer, R.; Wilcox, C.; Siegler, T. R.; Perryman, M.; Andrady, A.; Narayan, R.; Law, K. L. Plastic Waste Inputs from Land into the Ocean. *Clim. Chang. 2014 Impacts, Adapt. Vulnerability* **2015**, *347* (6223), 1655–1732.

(34) Lebreton, L. C. M.; Zwet, J. Van Der; Damsteeg, J.; Slat, B.; Andrady, A.; Reisser, J. River Plastic Emissions to the World's Oceans. *Nat. Commun.* **2017**, *8*, 1–10.

(35) Hoornweg, D.; Bhada-Tata, P. *What a Waste: A Global Review of Solid Waste Management*; 2012; www.worldbank.org/urban.

(36) Som, C.; Berges, M.; Chaudhry, Q.; Dusinska, M.; Fernandes, T. F.; Olsen, S. I.; Nowack, B. The Importance of Life Cycle Concepts for the Development of Safe Nanoproducts. *Toxicology* **2010**, *269* (2–3), 160–169.

(37) Gottschalk, F.; Scholz, R. W.; Nowack, B. Probabilistic Material Flow Modeling for Assessing the Environmental Exposure to Compounds: Methodology and an Application to Engineered Nano-TiO2 Particles. *Environ. Model. Softw.* **2010**, *25* (3), 320–332.

(38) Brunner, P. H.; Rechberger, H. *Practical Handbook of Material Flow Analysis*; Lewis Publishers, 2004; http://thecitywasteproject.files.wordpress.com/2013/03/practical_handbook-of-material-flow-analysis.pdf.

(39) Baccini, P.; Brunner, P. H. *Metabolism of the Anthroposphere: Analysis, Evaluation, Design*, 2nd Editio.; MIT Press, 2012; www.jstor.org/stable/j.ctt5vjrm9.

(40) Kawecki, D.; Scheeder, P.; Nowack, B. Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe. *Environ. Sci. Technol.* **2018**, *52*, 9874–9888.

(41) Gottschalk, F.; Sonderer, T.; Scholz, R. W.; Nowack, B. Modeled Environmental Concentrations of Engineered Nanomaterials (TiO2, ZnO, Ag, CNT, Fullerenes) for Different Regions. *Environ. Sci. Technol.* **2009**, *43* (24), 9216–9222.

(42) Adam, V.; Nowack, B. European Country-Specific Probabilistic Assessment of Nanomaterial Flows towards Landfilling, Incineration and Recycling. *Environ. Sci. Nano* **2017**, *4* (10), 1961–1973.

(43) Mahapatra, I.; Sun, T. Y.; Clark, J. R. A.; Dobson, P. J.; Hungerbuehler, K.; Owen, R.; Nowack, B.; Lead, J. Probabilistic Modelling of Prospective Environmental Concentrations of Gold Nanoparticles from Medical Applications as a Basis for Risk Assessment. *J. Nanobiotechnology* **2015**, *13* (1), 1–14.

(44) Laner, D.; Feketitsch, J.; Rechberger, H.; Fellner, J. A Novel Approach to Characterize Data Uncertainty in Material Flow Analysis and Its Application to Plastics Flows in Austria. *J. Ind. Ecol.* **2016**, *20* (5), 1050–1063.

(45) Gottschalk, F.; Nowack, B. A Probabilistic Method for Species Sensitivity Distributions Taking into Account the Inherent Uncertainty and Variability of Effects to Estimate Environmental Risk Health & Ecological Risk Assessment. *Integr. Environ. Assess. Manag.* **2012**, *9* (1), 79–86.

(46) Rechberger, H.; Cencic, O.; Frühwirth, R. Uncertainty in Material Flow Analysis. *J. Ind. Ecol.* **2014**, *18* (2), 159–160.

(47) PlasticsEurope. *Plastics - The Facts 2015: An Analysis of European Latest Plastics Production, Demand and Waste Data*; 2015; http://staging-plasticseurope.idloom.com/download_file/view/478/179.

(48) Gasperi, J.; Dris, R.; Bonin, T.; Rocher, V.; Tassin, B. Assessment of Floating Plastic Debris in Surface Water along the Seine River. *Environ. Pollut.* **2014**, *195*, 163–166.

(49) Sadri, S. S.; Thompson, R. C. On the Quantity and Composition of Floating Plastic Debris Entering and Leaving the Tamar Estuary, Southwest England. *Mar. Pollut. Bull.* **2014**, *81* (1), 55–60.

(50) Schubarth, C.; Weibel, F. *Die Bodennutzung in Der Schweiz. Resultate Der Arealstatistik*; 2013; Vol. 002–0901; https://www.bfs.admin.ch/bfsstatic/dam/assets/348986/master.

(51) Hernandez, E.; Nowack, B.; Mitrano, D. M. Polyester Textiles as a Source of Microplastics from Households: A Mechanistic Study to Understand Microfiber Release During Washing. *Environ. Sci. Technol.* **2017**, *51* (12), 7036–7046.

(52) Sillanpää, M.; Sainio, P. Release of Polyester and Cotton Fibers from Textiles in Machine Washings. *Environ. Sci. Pollut. Res.* **2017**, *24* (23), 19313–19321.

(53) Hartline, N. L.; Bruce, N. J.; Karba, S. N.; Ruff, E. O.; Sonar, S. U.; Holden, P. A. Microfiber Masses Recovered from Conventional Machine Washing of New or Aged Garments. *Environ. Sci. Technol.* **2016**, *50* (21), 11532–11538.

(54) Pirc, U.; Vidmar, M.; Mozer, A.; Kržan, A. Emissions of Microplastic Fibers from Microfiber Fleece during Domestic Washing. *Environ. Sci. Pollut. Res.* **2016**, No. 2014.

(55) De Falco, F.; Gullo, M. P.; Gentile, G.; Di Pace, E.; Cocca, M.; Gelabert, L.; Brouta-Agnésa, M.; Rovira, A.; Escudero, R.; Villalba, R.; Mossotti, R.; Montarsolo, A.; Gavignano, S.; Tonin, C.; Avella, M. Evaluation of Microplastic Release Caused by Textile Washing Processes of Synthetic Fabrics. *Environ. Pollut.* **2018**, *236* (XXX), 916–925.

(56) Byrne, M. A.; Kearns, G.; McKenna, C. A Test Chamber Study of the Contribution of Human Body Surfaces to the "Personal Cloud." In *Indoor Air 2002, 9th International conference on Indoor Air Quality and Climate*; Monterey, California, 2002.

(57) European Communities. *European Union Risk Assessment Report: Hexabromocyclododecane CAS-No.: 25637-99-4, EINECS- No.: 247-148-4. Final Report*; 2008; https://echa.europa.eu/documents/10162/661bff17-dc0a-4475-9758-40bdd6198f82.

(58) Yoon, Y. H.; Brimblecombe, P. Clothing as a Source of Fibres within Museums. *J. Cult. Herit.* **2000**, *1* (4), 445–454.

(59) Vigo, T. L.; Lamb, G. E. R.; Kepka, S.; Miller, B. Abrasion and Lint Loss Properties of Fabrics Containing Crosslinked Polyethylene Glycol. *Text. Res. J.* **1991**, *61* (3), 169–176.

(60) Keizer, K.; Lindenberg, S.; Steg, L. The Spreading of Disorder. *Science (80-.).* **2008**, *322* (5908), 1681–1685.

(61) Department for Environment Food & Rural Affairs. *Fly-Tipping Statistics for England,*

2016/17; 2017; https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/652958/Flytipping_201617_statistical_release_FINAL.pdf.

(62) Friedler, E.; Brown, D. M.; Butler, D. A Study of WC Derived Sewer Solids. *Water Sci. Technol.* **1996**, *33* (9), 17–24.

(63) Spence, K. J.; Digman, C.; Balmforth, D.; Houldsworth, J.; Saul, A.; Meadowcroft, J. Gross Solids from Combined Sewers in Dry Weather and Storms, Elucidating Production, Storage and Social Factors. *Urban Water J.* **2016**, *13* (8), 773–789.

(64) Goldberg, L. Plastic in Organic Waste in Switzerland : Material Flows and Consequences, University of Zurich, 2018.

(65) Weithmann, N.; Möller, J. N.; Löder, M. G. J.; Piehl, S.; Laforsch, C.; Freitag, R. Organic Fertilizer as a Vehicle for the Entry of Microplastic into the Environment. *Sci. Adv.* **2018**, *4*, 1–8.

(66) Faure, F.; De Alencastro, L. F. *Recherche de Fragments de Plastique Dans Les Composts et Digestats Industriels*; 2016.

(67) WRAP. *Report: Plastic Packaging Composition 2011*; 2013; http://www.wrap.org.uk/sites/files/wrap/Plastics Composition 2011 Report.pdf.

(68) Eurostat. Population change - Demographic balance and crude rates at national level [demo_gind] http://ec.europa.eu/eurostat/product?code=demo_gind&language=en&mode=view (accessed Apr 2, 2018).

(69) Scudo, A.; Liebmann, B.; Corden, C.; Tyrer, D.; Kreissig, J.; Warwick, O. *Intentionally Added Microplastics in Products*; 2017; http://ec.europa.eu/environment/chemicals/reach/pdf/39168 Intentionally added microplastics - Final report 20171020.pdf.

(70) Wik, A.; Dave, G. Occurrence and Effects of Tire Wear Particles in the Environment – A Critical Review and an Initial Risk Assessment. *Environ. Pollut.* **2009**, *157* (1), 1–11.

(71) Besseling, E.; Quik, J. T. K.; Sun, M.; Koelmans, A. A. Fate of Nano- and Microplastic in Freshwater Systems: A Modeling Study. *Environ. Pollut.* **2017**, *220*, 540–548.

(72) Blarer, P.; Kull, G. *Swiss Litter Report*; 2018; www.stoppp.org/swiss-litter-report.

(73) Erismann, R.; Erismann, S. In(Beach-Litter-Density): Annex B. **2016**, 1–23.

(74) Federal Office for the Environment FOEN. The Swiss hydrographic network: Network analyses https://www.bafu.admin.ch/dam/bafu/en/dokumente/hydrologie/fachinfo-daten/auswertungen_zumgewaessernetz.xlsx.download.xlsx/network_analyses.xlsx (accessed Dec 3, 2018).

(75) Le Hyaric, R.; Canler, J. P.; Barillon, B.; Naquin, P.; Gourdon, R. Characterization of Screenings from Three Municipal Wastewater Treatment Plants in the Region Rhône-Alpes. *Water Sci. Technol.* **2009**, *60* (2), 525–531.

(76) Cabernard, L.; Durisch-Kaiser, E.; Vogel, J.-C.; Rensch, D.; Niederhauser, P.; AWEL Gewässerschutz. Mikroplastik in Abwasser Und Gewässern. *Aqua Gas* **2016**, *7* (8), 78–85.

(77) Office Fédéral de la Statistique OFS. Bilan de la population résidante permanente par canton, 1991-2017 https://www.bfs.admin.ch/bfsstatic/dam/assets/5886171/master.

(78) Dris, R.; Gasperi, J.; Saad, M.; Mirande, C.; Tassin, B. Synthetic Fibers in Atmospheric Fallout: A Source of Microplastics in the Environment? *Mar. Pollut. Bull.* **2016**, *104* (1–2), 290–293.

(79) Raunemaa, T.; Kulmala, M.; Saari, H.; Olin, M.; Kulmala, M. H. Indoor Air Aerosol Model: Transport Indoors and Deposition of Fine and Coarse Particles. *Aerosol Sci. Technol.* **1989**, *11* (1), 11–25.

(80) Edwards, R. D.; Yurkow, E. J.; Lioy, P. J. Seasonal Deposition of Housedusts onto Household Surfaces. *Sci. Total Environ.* **1998**, No. 224, 69–80.

(81) Ruotsalainen, R.; Rönnberg, R.; Säteri, J.; Majanen, A.; Seppänen, O.; Jaakkola, J. J. K. Indoor Climate and the Performance of Ventilation in Finnish Residences. *Indoor Air* **1992**, *2* (3), 137–145.

(82) Economidou, M.; Atanasiu, B.; Despret, C.; Maio, J.; Nolte, I.; Rapf, O.; Laustsen, J.; Ruyssevelt, P.; Staniaszek, D.; Strong, D.; Zinetti, S. *Europe's Buildings Under the Microscope*; 2011; http://bpie.eu/wp-content/uploads/2015/10/HR_EU_B_under_microscope_study.pdf.

(83) Webster, T. F.; Harrad, S.; Millette, J. R.; Holbrook, R. D.; Davis, J. M.; Stapleton, H. M.; Allen, J. G.; McClean, M. D.; Ibarra, C.; Abdallah, M. A.; Covaci, A. Identifying Transfer Mechanisms and Sources of Decabromodiphenyl Ether (BDE 209) in Indoor Environments Using Environmental Forensic Microscopy. *Environ. Sci. Technol.* **2009**, *43* (9), 3067–3072.

(84) Mølhave, L.; Schneider, T.; Kjærgaard, S. K.; Larsen, L.; Norn, S.; Jørgensen, O. House Dust in Seven Danish Offices. *Atmos. Environ.* **2000**, *34* (28), 4767–4779.

(85) Talvitie, J.; Heinonen, M.; Pääkkönen, J.-P.; Vahtera, E.; Mikola, A.; Setälä, O.; Vahala, R. Do Wastewater Treatment Plants Act as a Potential Point Source of Microplastics? Preliminary Study in the Coastal Gulf of Finland , Baltic Sea. *Water Sci. Technol.* **2015**, *72* (9), 1495–1504.

(86) Talvitie, J.; Mikola, A.; Setälä, O.; Heinonen, M.; Koistinen, A. How Well Is Microlitter Purified from Wastewater? A Detailed Study on the Stepwise Removal of Microlitter in a Tertiary Level Wastewater Treatment Plant. *Water Res.* **2017**, *109*, 164–172.

(87) European Parliament. *P8_TA-PROV(2019)0305 Reduction of the Impact of Certain Plastic Products on the Environment*; 2019; https://www.europarl.europa.eu/doceo/document/TA-8-2019-0305_EN.pdf.

(88) IGSU. *Recul Du Littering Dans Toutes Les Régions Du Pays*; 2017; https://www.igsu.ch/files/fr_2017-12-05_mm_littering-index.pdf.

(89) Cosmetics Europe. Cosmetics Europe survey shows an 82% reduction in use of plastic microbeads https://www.cosmeticseurope.eu/news-events/reduction-use-plastic-microbeads (accessed Aug 8, 2017).

(90) European Chemicals Agency. *Annex to the Annex XV Restriction Report: Proposal for a Restriction*; 2019; https://echa.europa.eu/documents/10162/decfbb48-7240-9aa2-d0fa-7f18582fce36.

8 A proxy-based approach to predict spatially resolved emissions of macro- and microplastic to the environment

This chapter is a manuscript that has been submitted to a scientific journal for publication.

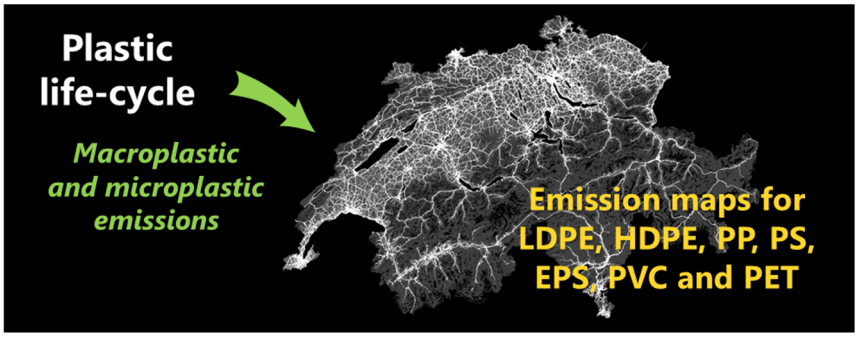
Authors: Kawecki, D.; Nowack, B.

8.1 Abstract

Large disparities on micro- and macroplastic concentrations are to be expected between residential, natural and agricultural areas, since specific uses of plastic will determine the magnitude of the corresponding emissions and consequently, the total burden and its material composition. Identifying regions and environmental compartments with potentially higher than average microplastic fluxes is crucial to assess the possible risk in different settings. The aim of this work was to develop a method to regionalize emissions of macroplastic and microplastic for soil, freshwater and air. The emissions were regionalized using geographical datasets on land-use statistics, traffic and population densities, wastewater treatment plants and combined sewer overflows as proxies. Each emission flow was attributed to a proxy depending on the characteristics of the emission. High resolution maps (for soil and air raster maps with a resolution of 100x100 m and for water vector maps at a scale of 1:25 000) of the emissions were then generated for micro- and macroplastic. Using emission data available for Switzerland for seven commonly used polymers (LDPE, HDPE, PP, PS, EPS, PVC and PET), a total of 35 maps were created using the mean emission flows, and a further 35 maps were created for the 5^{th} and 95^{th} percentiles each. Most of the emissions can be found in areas with high human activity on the Swiss plateau and the alpine valleys, but the influence of the different proxies varies for each polymer. The median local emission rate of macroplastic on soil varies from 0.0006 to 0.06 kg/ha/a depending on the location, whereas no emission flows are predicted for more than 50% of the raster cells for microplastic regardless of the polymer, but the maxima can reach up to 12.7 kg/ha/a in the case of HDPE and lower values for the other polymers. The average emission rate of macroplastic along river segments ranges between 0.062 kg/km/a and 1.5 kg/km/a depending on the polymer. For microplastic, the average emission rate varies from 0.0025 kg/km/a to 0.11 kg/km/a. The analysis reveals that a significant deviation is expected if the population density is used as only proxy with a high resolution. The correlation between the population density and the predicted emissions is only r = 0.16-0.23 for a cell size of 100x100 m and goes up to r = 0.86-0.88 for a resolution of 10 km for polymers not significantly

used in agriculture, however an r of only 0.56-0.68 is observed for those polymers used a lot in agriculture such as HDPE and PP. The emission maps obtained in this work can serve as input to regionalized fate models for macro- and microplastics.

8.2 TOC-Art



8.3 Introduction

Plastic can be found in nearly every environmental sample in marine and continental environments and is present either as small particles called microplastic (MP) or as larger debris often referred to as macroplastic. Most of the MP research has focussed on the marine environment but research in freshwater environments has intensified[1] and MP have been reported in a wide range of concentrations all over the world[2,3]. First estimates of MP pollution in soils suggest that it may be considerable as well[4,5] but this compartment was up to now much less studied than freshwaters[1,6]. Macroplastic litter is intimately connected to the MP issue since macroplastic may eventually fragment into MP under environmental conditions but is in comparison to MP much less studied in terrestrial contexts[7–12]. Macroplastic pollution in soils has, to our knowledge, only been examined in the context of citizen science studies focussing on littering[13] or roadsides[14]. Online databases have been created to enable the recording of litter items in any environment by citizen scientists[15].

The sources and pathways of plastic to the environment are increasingly known[16–18] although large uncertainties remain. The largest sources are mismanaged macroplastic and rubber wear particles[17,19] (if rubber particles are to be considered MP[20]), but they are strongly dependent on the polymer considered[16]. Most of the macroplastic emissions originate from mismanagement of waste through littering, dumping and other types of improper disposal of consumer

goods and, in a European context, waste from construction sites and agriculture[16]. Large uncertainties remain for example regarding the cleaning efficiencies of macroplastic in rural environments and along roadsides[16].

The concentration of MP in freshwater has been suggested to depend on the location[1] but as well on the time of sampling since weather[21–24] and currents have been shown to influence it[25,26]. On the other hand, the concentration of MP in small streams was shown to be of the same order of magnitude as for large rivers[27], suggesting that the input into small streams may be more relevant than previously expected. Conflicting results on the influence of the population density and combined sewer overflows (CSOs) on MP concentrations were reported[27,28]. The spatial variation and dynamics of MP in freshwater is therefore yet to be understood fully. Macroplastic debris in freshwater may be correlated to the population density[29] but few studies have yet investigated the relationship between the burdens in soil and freshwaters. MP contents in soil may be strongly location-dependent because the different emission routes may dominate individual locations[30,31].

Plastic transport may be very different depending on the density of the material[32,33] and the emissions are strongly affected by the life-cycle of the product[16]. It is not yet clear if a difference in toxicity can be expected for the different polymers themselves[34,35] but it is reasonable to assume variations in additive toxicity depending on the material considered. It is therefore necessary to provide polymer-specific assessments about their possible fate and risk.

Spatially resolved models have recently emerged as a solution for understanding the distribution and variation of pollutants across ecosystems[36,37]. These tools typically contain two distinct parts: a first model describing the input into environmental systems and a second model for the fate processes in the environment. The emission flows of commodity plastics to the environment in Switzerland have been published recently[16], considering Switzerland as a whole region and neglecting any local variations. The next step towards predicted environmental concentrations requires a regionalization of these emission flows, which can then in a later step, be used as input for fate models to obtain a range of possible predicted environmental concentrations.

The aim of the present study was to develop a method to quantify the spatially resolved releases of MP and macroplastic to water, soil and air based on a regional polymer-specific release model. The model was then applied to predict the spatially resolved releases of seven different polymers to the Swiss environment.

8.4 Method

8.4.1 Emission flows

The emission flows of seven polymers into the environment in Switzerland were estimated in an earlier study[16] and are regionalized using eleven geographical proxies. These emission flows were modelled using probabilistic material flow analysis. A total of 134 processes model the life-cycle and the emission flows, interconnected with a total of 402 flows of which 234 are for modelling emission. The 61 final release flows to surface water, soil and air were considered within this study –releases to subsurface soil were excluded. The polymers considered were low-density and high-density polyethylene (LDPE and HDPE, respectively), polypropylene (PP), polystyrene (PS), expanded PS (EPS), polyvinyl chloride (PVC) and polyethylene terephthalate (PET) which are the plastic materials most commonly reported in freshwater environments[2]. The emission flows are available as probability distributions[16] (Figure S74), of which only the mean and the 5th and 95th percentiles will be considered in the current work.

8.4.2 Proxies

Eleven different geographical proxies were used to regionalize each of the 61 emission flows (Table S36). Proxies were chosen so that they should be representative of the mechanism underlying each emission flow. For soil and air emissions, six raster maps were created using official statistics. For emissions to water, five more proxies were created. All raster maps were available with a 100 m resolution. The vector maps had a precision of 1:25000.

8.4.2.1 Population density

A map of the population density was used for all nine emissions occurring in residential areas (Table S36) as for example the emission of litter to residential soil or the emissions of fibres from clothing to outdoor air. For this, the total permanent residing population in 2014 was obtained from an online available geographical dataset (geodataset) collected by the Swiss Federal Statistical Office (FSO)[38]. The geocoding of the residing population in this geodataset was primarily performed using the localization of buildings intended for habitation and the federal registry for buildings and habitations. For data protection purposes, values below 3 inhabitants/ha are displayed as 3 inhabitants/ha for the freely available geodataset which was used in the present study.

8.4.2.2 Land use statistics

Land use statistics from the FSO were used to create proxy maps for industrial, agricultural and natural areas. The original geodataset was created based on aerial photographs verified with other available datasets and point verifications[39]. The available land use statistics were divided into 17 classes for the geodataset developed for 2004-2009.

Two classes were considered for the creation of the industry proxy: industrial and artisanal areas (corresponding to class 1) and special infrastructure areas (4). This proxy was used for the 19 emissions caused by the manufacturing of plastic goods, and the collection and recycling of end-of-life plastic (Table S36).

Similarly, two classes were considered for the creation of the agriculture proxy: fruit arboriculture, viticulture and horticulture (6), and arable land (7). The agriculture proxy was used for all 12 emission flows related to agricultural activities as for example losses of MP from agricultural films and pipes to agricultural soil, or the application of compost containing plastic to land (Table S36).

The last proxy developed based on land use statistics aimed at describing all areas which are of natural character. Six different classes were considered for this proxy: natural prairies and pastureland (8), forest (10), shrubland (11), other woods (12), unproductive vegetation (15) and surfaces without vegetation (16). The proxy for forests and other natural areas was used for the three emission flows occurring on rural soil as for example littering or the release of shotgun cartridges to soil (Table S36).

8.4.2.3 New buildings

A map of the areas with newly constructed buildings was used for the four emission flows from construction and demolition sites as for example the release of MP from construction pipes to soil or MP from insulation to outdoor air (Table S36). The statistic on buildings and habitations was used for this, from which the number of new buildings for habitation built between 2006 and 2010 were considered[40]. The geocoding of the buildings in the original geodataset was based on the federal registry for buildings and habitations and completed with additional information when necessary.

8.4.2.4 Traffic

The traffic density in Switzerland was used to regionalize the emissions from littering during transport and from the accidental release of plastic from vehicles (Table S36). The traffic density was obtained from sonBASE, a model developed to predict noise levels induced by traffic in Switzerland[41]. The model is based on the road network from Openstreetmap, combined with an agent-based model and hourly measurements of traffic density at 1954 locations. The modeled average number of vehicles per day per road segment was used to create the geodataset. All road segments corresponding to tunnels were previously removed.

8.4.2.5 Area surrounding water bodies

The properties of the areas surrounding water bodies needed to be characterized in a geodataset to be able to distinguish emissions to water occurring around residential or natural areas. Emissions to water in natural environments can occur from littering or shotgun cartridges (Table S36). Emissions to water in residential environments only occur from littering.

Geographical information on the water bodies in Switzerland was obtained from a database from the Swiss Federal Office for the Environment (FOEN) containing the river network, drainage basins and lakes at a precision of 1:25000[42]. The geodataset contains 20167 river segments and 210 polygons for the lakes. The average river segment length is 1.6 km but can vary from 11 m to 18 km. Polygons representing the portions of lakes outside of Switzerland are available in the dataset and will be displayed as grey polygons in the results section, except for the Italian part of Lago Maggiore, which is not present in the initial dataset. The first proxy was calculated by counting the number of inhabitants in a radius of 500 m around each water body. The second proxy was built by counting the number of cells corresponding to forests and other natural environments in a radius of 500 m around the water body. The calculation of the number of cells or inhabitants surrounding a water body was performed in R using the *extract* function from the R package *raster* and using a buffer of 500 m.

8.4.2.6 Wastewater treatment plants and combined sewer overflows

Emissions occurring from wastewater treatment plants (WWTPs) were regionalized using WWTP locations and the number of inhabitants connected. The level of treatment of the WWTPs enabled to differentiate between emissions from secondary and tertiary treatment. The WWTP geodataset was combined from two separate datasets available from the FOEN. Starting from the first dataset[43], the WWTP identification number, location or address, and the number

of inhabitants connected to the WWTP could be obtained for 759 WWTPs. Two out of 759 WWTPs in the dataset did not have any information regarding the number of inhabitants connected and were excluded from the dataset. For 73 of these WWTPs, no coordinates were present in the dataset. The available postal address was then automatically translated into coordinates using OSM Nominatim[44] in an R script. The treatment type of the WWTP was obtained from a second dataset[45]. Out of all the 759 WWTPs in the primary dataset, 37 are absent from the treatment dataset and 212 are unknown but present in the treatment dataset. For all of these WWTPs, we assume that the highest treatment stage corresponds to secondary treatment.

Emissions occurring from combined sewer overflows (CSO) were regionalized using CSO locations and volumes discharged[46]. The volume discharged is calculated from the published dataset by multiplying the wastewater influent to the WWTP and the time the CSOs were active.

The point emissions were attributed to the closest water body. The nearest water body to the point emission was identified using the *st_nearest_feature* and *st_distance* functions in the *sf* package in R.

8.4.3 Regionalization

For emissions to air and soil, the geodatasets were converted to rasters using the function *rasterFromXYZ* from the *raster* package in R, while making sure that the extent of the different rasters was identical. The traffic geodataset which consisted of vectors was then projected on a raster object with the same resolution and extent as the remaining geodatasets in QGIS. The native resolution of 100 m of the original rasters is kept for the calculation. The raster maps presented in this article are aggregated to a resolution of 1 km for visualization purposes. The final emission maps use the coordinate system EPSG 2056 (also called CH1903+ LV95). Vector data using the coordinate system CH1903 LV03 were converted by using the built-in functions in the *sf* and *rgdal* packages in R for vector data. The rasters based on the land use geodataset and the point sources to water (WWTP and CSO) were converted by adding 2000000 m in the x direction and 1000000 m in the y direction. This manual coordinate transformation leads to minor deformations on the edges of Switzerland of up to 1.6 m[47]. This was preferred over a reprojection of the rasters in a new coordinate system where the data itself may undergo smoothing.

The emission flows are regionalized as follows:

$$\boldsymbol{f}_r = \frac{\boldsymbol{P}}{\sum_{i,j} \boldsymbol{P}_{ij}} \cdot f \,,$$

where $\boldsymbol{f}_r$ represents the regionalized emission flow, $\boldsymbol{P}$ the proxy used for the regionalization and f the emission flow. $\boldsymbol{f}_r$ and $\boldsymbol{P}$ can either be a matrix describing the individual cells of the raster or a vector describing the river segments or lake polygons. All regionalized emission flows can then be summed to obtain the total emission maps of a specific polymer in soil, water or air, as macroplastic or MP:

$$\boldsymbol{f}_{\text{total}} = \sum_r \boldsymbol{f}_r \,.$$

All operations were performed in R using the *sf* and *raster* packages. The scripts are available online.

8.5 Results

8.5.1 Summed maps of the emissions

The final emissions maps are a linear combination of the proxy maps with varying weights depending on the emission flows (Figure 17). Depending on the polymer, specific proxies dominate the final emission map, as for example the traffic proxy for macroplastic emissions to soil for all polymers except LDPE and PP for which the agriculture proxy is also essential. The MP emissions to soil are mostly influenced by industry (in particular PS and EPS), agriculture (LDPE, PP and PET) and new buildings (HDPE and PVC). Both MP and macroplastic emissions to water are to a large extent influenced by CSOs at around at least 80% of the weight. The population around water bodies accounts for most of the remaining emissions of macroplastic, the WWTPs for MP. The emissions to air depend on three proxies only: industry, population and new buildings, which are each very relevant for at least one polymer: LDPE is exclusively influenced by industry, EPS by new buildings and PET almost exclusively by the population proxy.

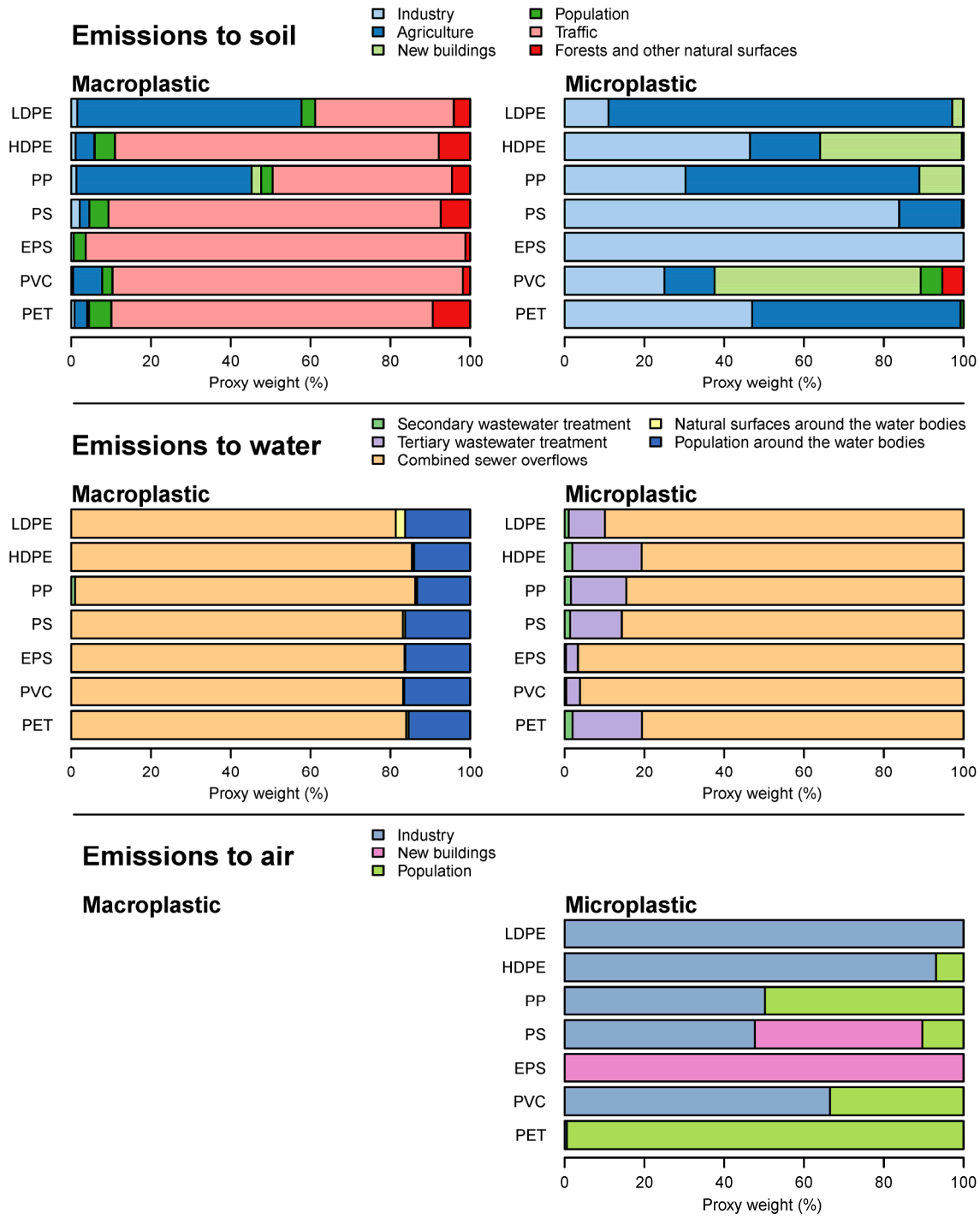


Figure 17: Weight attributed to the different proxies for emissions to soil, water and air, as macroplastic and microplastic. The weights are calculated as proportions of the emission flows attributed to the respective proxies.

The example of PET macroplastic emissions to soil is considered in more detail to illustrate the regionalization process. Six different proxies contribute to it: traffic, forests and other natural

environments, population, agriculture, industry and new buildings (Figure 18). 81% of the emissions of PET macroplastic are regionalized using the traffic network as proxy because of littering along roads, which is reflected in the final result with the traffic network standing out the most. The emissions of PET related to this proxy can reach high levels, with up to 13 kg/ha/a emitted. 9% of the emissions of PET macroplastic are regionalized using the forests and other natural environments mostly due to littering in natural environments, giving a low emission background of around at most 0.1 kg/ha/a in the Alps. The remaining proxies contribute to features which are less prominent.

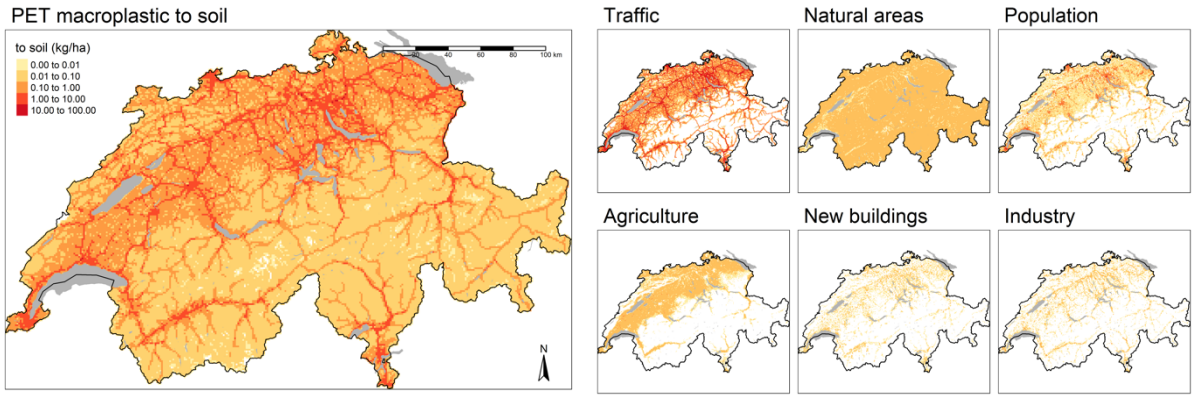


Figure 18: Total emissions of PET macroplastic to soil (left) with the maps created from the six different proxies (right): traffic, forests and other natural areas, population, agriculture, new buildings and industry. The smaller maps are added to obtain the total map of PET macroplastic emissions.

Once the contributions of all proxies are added, one obtains maps of the modelled emissions per material, environmental compartment and size of plastic (Figure 18, Figure 19, Figure 20 and Figures S77-S81). Maps of the 5% and 95% quantiles of the emission probability distributions were as well generated and are available upon request. In most maps, a large fraction of the emissions takes place in the Swiss plateau, which is the region extending from Lake Constance in the Northeast to Lake Geneva in the Southwest where most of the human activities are concentrated in Switzerland. The main valleys in the Alps are also easily recognizable in most maps.

The macroplastic emission maps (Figure 19 for PS and LDPE, Figure 18 for PET and Figure S77 for all polymers) can be coarsely categorized into two types, the first one in which the traffic network is the most preeminent feature and the second one where the whole area of the Swiss plateau appears as the main feature. This is in accordance with the weights used where most

polymers are largely defined by the traffic and agriculture proxies (Figure 17). LDPE and PP are the two polymers for which agriculture plays an important role, and for which the Swiss plateau appears as a band of higher emissions. The local emission rates of LDPE are within 0-0.01-20 kg/ha/a (minimum-median-maximum) and the local emission rates of PP are within 0-0.02-36 kg/ha/a. The remaining polymers are mostly emitted along roadsides with emissions reaching a maximum for PET at 0-0.06-117 kg/ha/a. All of these polymers are used a lot in packaging and are littered along roadsides. In addition, accidental releases of construction material also cause large emissions to roadsides for PS (0-0.01-25 kg/ha/a), PVC (0-0.002-34 kg/ha/a) and EPS (0-0.001-11 kg/ha/a). The emission background visible in the mountainous regions is caused by the emissions in forests and natural soils. The largest emission flow associated to this proxy is littering in natural environments. PET, PP and HDPE present higher emission backgrounds in the Alps of at most 0.1 kg/ha/a emitted depending on the location. LDPE, PS, EPS and PVC display lower emission rates of at most 0.01 kg/ha/a.

Similarly, MP emission maps can be categorized into three types (Figure 19 for LDPE and EPS and Figure S78 for all polymers), the first one in which urban centres are central, the second one in which agriculture is more important and the third one in which the emission background over the Alps is larger. LDPE (0-0-2.6 kg/ha/a min-median-max), HDPE (0-0-13 kg/ha/a), PP (0-0-7.7 kg/ha/a) and PET (0-0-0.33 kg/ha/a) display a strong influence of the agriculture sector. On the other hand, PS (0-0-0.34 kg/ha/a), EPS (0-0-0.1 kg/ha/a) and PVC (0-0.001-22 kg/ha/a) are less influenced by the agriculture sector. Most of the MP emissions of all polymers are restricted to areas of high human activity, i.e. the Swiss plateau and the valleys. The higher emission background for PVC over the Alp region is caused by the wear of products outdoors producing MP.

The emissions of MP to air are restricted to areas with high human activity (Figure 19 for PET and EPS and Figure S81 for all polymers). The regionalization of the emissions to air depends exclusively on the proxies industry, population and new buildings (Figure 17), which are all concentrated on the Swiss plateau and the valleys (Figure S75). The urban centres are more accentuated for PET, PP and PVC. The industry and new building proxies are in regions of high human activity but further away from areas with high population densities. The largest local emission rates occur for PET in urban centres, and range within 0-0-22 kg/ha/a overall (min-median-max). PP follows similar emission rates at 0-0-11 kg/ha/a. The remaining polymers have lower emission rates: LDPE (0-0-0.08 kg/ha/a), HDPE (0-0-0.6 kg/ha/a), PS (0-0-3.8 kg/ha/a), EPS (0-0-1.6 kg/ha/a) and PVC (0-0-2.2 kg/ha/a).

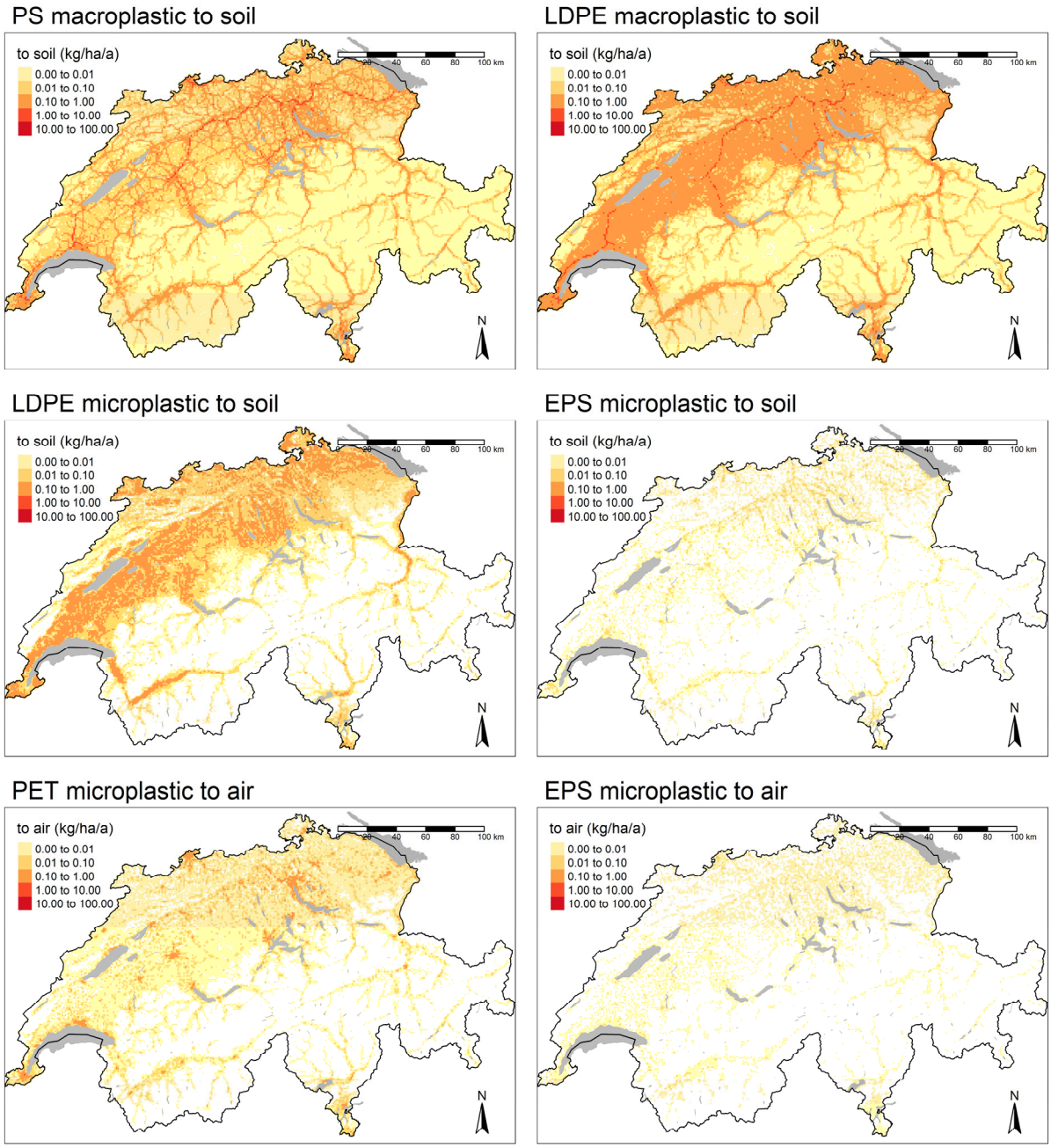


Figure 19: Maps of the modelled emissions of PS and LDPE macroplastic to soil, of LDPE and EPS MP to soil and of PET and EPS MP to air. Equivalent maps for HDPE, PP, PS, EPS and PVC can be found in the SI in figures S77-S78 and figure S81.

Emission maps were also created for macroplastic and MP emissions to water (Figure 20, Figures S79-S80). PP macroplastic and PET MP emissions are shown in Figure 20 with a zoomed map inset for each. Emissions to river segments are reported in kg/km/a and emissions to lakes in kg/ha/a. Most of the emissions are situated in areas with high human activity. The mean emission rate of macroplastic along river segments ranges between 0.062 kg/km/a for EPS and

1.5 kg/km/a for PET. For MP, this emission rate varies from 0.0025 kg/km/a for EPS to 0.11 kg/km/a for HDPE. In lakes, the average emission rates of macroplastic range from 0.0025 kg/ha/a for EPS to 0.061 kg/ha/a for PET. Similarly, between 0.0001 kg/ha/a of EPS and 0.004 kg/ha/a of HDPE are emitted as MP to lakes on average. The map insets presented in Figure 20 show a more detailed view of the water bodies modelled. CSOs have the largest weight for the final result for macroplastic consisting of any polymers at around 85% of the final result (Figure 17) and the population around water bodies accounts for around 15%. Similarly, for MP, CSOs correspond to around 80-95% of the final result and the main remaining fraction is caused by emissions through tertiary wastewater treatment. Since the CSO proxy has a similar weight for macroplastic and MP emissions of all polymers, the emission maps display similar tendencies within the river network and lakes. It is also clearly apparent from the map insets that there is no continuity of emission magnitudes along a river; each segment is independently calculated from the connected ones.

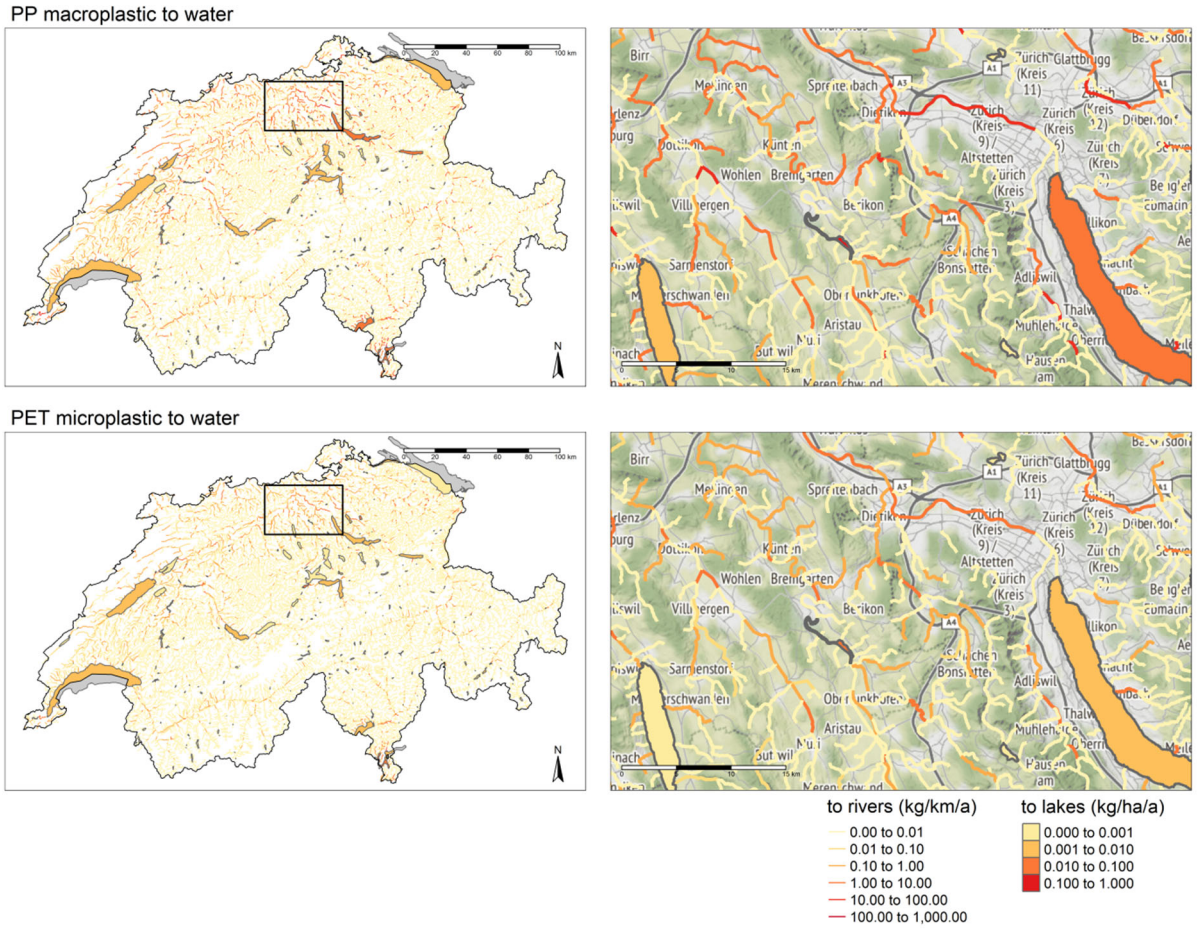


Figure 20: Maps of the modelled emissions of PP macroplastic and PET microplastic to water. Emission maps for macroplastic and MP are presented in Figures S79 and S80. The map insets on the right correspond to the area in the rectangles in the main maps. Map tiles by Stamen Design, under CC BY 3.0. Data by OpenStreetMap, under ODbL.

8.5.2 Correlation analysis

The population density has already been used as only proxy to regionalize emissions of plastic globally[36]. It has never been investigated how well the population density can be used as single proxy for plastic emissions. The correlation coefficient of the emissions to soil and air with the population density was calculated as a function of the plastic material and the raster resolution (Figure 21). The rasters of emissions of microplastic to air, macroplastic and microplastic to soil were added for this analysis. The correlation coefficient increases with the cell size, reaching better correlation coefficients for a cell size of 10 km for most polymers. The best correlation is attained for HDPE, PS, EPS, PVC and PET at r = 0.86-0.88. A much worse correlation is found for PP at r = 0.68 and LDPE at r = 0.56. These two polymers are used extensively in agriculture and the regional distribution of agricultural activities does not coincide well with the population density. For these two polymers, the use of the population density as only proxy may cause large errors. For cell sizes equal to or below 1 km, much lower correlation coefficients (r < 0.5) are obtained.

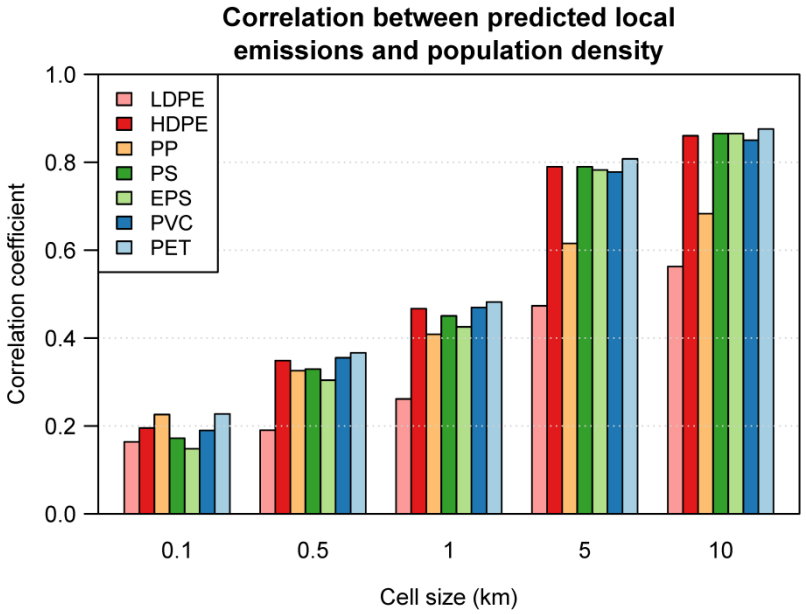


Figure 21: Correlation coefficient of the raster of the emissions to soil and water with the population density raster. The correlation coefficients are grouped by raster resolution on the x-axis and the seven colours correspond to the seven polymers.

8.5.3 Influence of the input data

Large variations in local emission rates can be observed for the emission maps created using the mean, the 5th or 95th percentiles of the initial emission flow distributions (Figure 22). In these density diagrams, the frequency of occurrence of the emissions is shown on a logarithmic scale. The differences observed between the three scenarios are caused by the data used to model the emission flows [16]. The three spectra of emissions calculated with the mean and the quantiles have very similar shapes, with a few narrow peaks and smaller plateaus in the case of the emissions to soil and air. The spectra of emissions to rivers and lakes are much smoother in comparison, which is due to the fact that emissions to a single water body may originate from several point sources and a 500 m wide buffer used around the water body. Nevertheless, the three different spectra are separated by at least one order of magnitude for most environmental compartments, polymers and plastic sizes.

These density diagrams also reveal that there is no continuum in the emissions modelled for soil and air. This behaviour is mostly a consequence of the use of land-use statistics as proxies, since this geodataset contains categorical data. As a result, for a mean emission flow of 62 t for PP macroplastic regionalized using the agriculture proxy, and 458038 cells where agriculture is present, one obtains an average emission of 62000/458038 = 0.14 kg/ha/a, which corresponds to the largest peak in the red spectrum of the PP MP plot in Figure 22. The smaller peak in the same graph situated around 1 kg/ha/a can be explained by new buildings and industry. Each of the 27057 cells corresponding to new buildings obtained a local emission rate of 0.7 kg/ha/a, and each of the 41006 cells corresponding to industry obtained a local emission rate of 0.78 kg/ha/a. The remaining features are caused by the population proxy, which is not a categorical dataset and thus doesn't create unique narrow peaks. Similarly, EPS MP emissions to soil also display a unique narrow peak of emissions, since only the industry proxy contributes to this emission. The spectrum for HDPE macroplastic emissions to soil is more intricate since 83% of the emissions are attributed to the traffic network which is not categorical. The two largest narrow peaks visible can be attributed to natural areas and agriculture. The PVC MP emissions to air depend only on the population proxy, which generates the first peak on the left and the plateau until 10^{-2}, and on the industry proxy which generates the second peak at 0.15 kg/ha/a. The first peak caused by the population proxy can be attributed to the high number of cells with 3 inhabitants/ha which is the minimum in the geodataset for privacy purposes. A large number of cells or vector elements remain zero after the regionalization. The lowest share of cells with zero emissions is attained for macroplastic emissions to soil at 20%, since

the littering to natural environments is spread out over the mountainous regions. For the remaining emission maps, the share of elements with zero emissions varies from 20% to 91%.

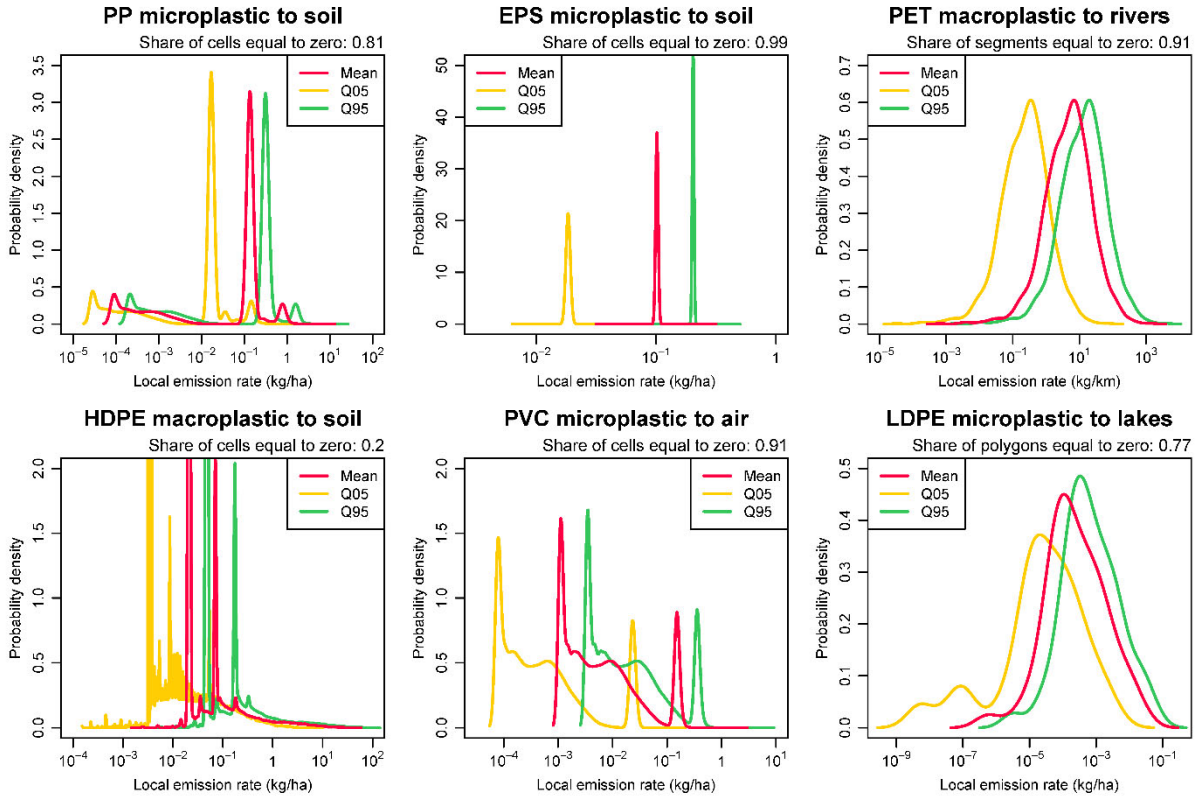


Figure 22: Variability of the local emission rates of polymers to environmental compartments. The density functions are calculated using the *density* function from the *stats* package in R, after removal of the zero values. The fraction of cells or vector elements with zero values is shown below the title in each graph. The emission rates calculated using the mean of the emission flow distributions is shown in red, and the emission rates calculated using the quantiles are shown in yellow and green.

8.6 Discussion

This study proposes a new approach to regionalize plastic emission flows in a specific region, by attributing emission flows to an appropriate geodataset used as proxy. A total of 35 maps were created using the mean emission flows, and a further 35 maps were created for the 5th and 95th percentiles each. These maps represent the modelled geographical repartition of previously modelled emission flows. These results reveal that the local emission rates can vary by at least two orders of magnitude depending on the polymer and receiving environmental compartment. The influence of the main applications of each polymer is as well visible, especially for polymers largely used in agriculture. The population density has already been used as only proxy to regionalize emission flows in previous studies[36]. Our analysis shows that using the

population density as only proxy will lead to large deviations in the spatial distribution of the plastic emissions at high resolution, in particular for polymers with major applications in agriculture as for example LDPE and PP. Regionalization of emission flows should therefore either consider more proxies than only the population density or use low resolutions. Deviations will still occur at a resolution of 10 km for all polymers but especially for LDPE and PP.

The model presented in this study relies on two types of data: the emission flow probability distributions and the map proxies. The uncertainty and validity of the model predicting the emission flows for Switzerland was already discussed in the original study[16]. In order to account for the uncertainty of the emission flows, maps using the 5th and 95th percentiles of the emission distributions were as well produced. Another type of uncertainty arises from the use of proxies to regionalize the emission flows. These proxies were chosen so that they should be representative of the behaviour underlying the emissions. In the density plots displayed in Figure 22, narrow peaks in the local emission rates are observed, which are a consequence of the limited degree of detail in the categorical geodatasets. Since four out of the six raster proxies used for soil and air emissions rely on land-use statistics, emissions attributed to these proxies can only take a single value, which leads to a multitude of raster cells showing the same emissions. It is reasonable to assume that a certain variation occurs within such an emission type following variations in use intensity and use type as for example different crops in the case of agriculture, or varying levels of use of natural areas. Moreover, no differences between different parts of the country with respect to the emission flows are included. It should also be noted that only the emissions from Switzerland are included, which is important especially when considering the magnitude of the emissions to water bodies where a contribution from neighbouring countries takes place such as for Lake Constance (from Germany and Austria) and Lake Geneva (from France).

While MP concentrations in freshwater are available from an array of studies, very few measurements of emission flows are available which could be used to validate the results of the here presented model. The only Swiss-specific emission data that is available is from a study examining the MP release from 28 WWTPs[48]. The MP release has been estimated using the MP concentration in the effluent and the average dry weather discharge rate. When comparing these estimates to our predictions, one finds that our predictions are a factor of around two times higher. A correlation coefficient of $r = 0.78$ was found between the two datasets, with a root mean squared error (RMSE) of 15 kg/a (compared to a minimum of 0.66 kg/a and maximum of 93 kg/a in our predictions). Most of the error can be attributed to one single WWTP that treats around 45% of the water out of the 28 WWTPs considered for this comparison.

Considering the multitude of uncertainty sources influencing both estimates, we consider the agreement between measured and predicted values is an indication that the implementation of the wastewater emission part of the model is capturing the actual emissions quite well.

If emission maps on the scale of a city for example would be of particular interest, some adjustments in the proxies would be necessary. For example, considering a map of the human activity instead of the residence would add accuracy to the regionalization of littering and fibre emissions through wear in residential and natural areas. For natural areas, online databases for hiking trails and barbecue locations for example could be used to improve the precision of the proxy. The behaviour behind littering is known to be strongly dependent on the surroundings[49]. For a study of the amount littered at the scale of a city, other model types may be more appropriate as for example agent-based modelling[50] distinguishing the quality of the surroundings[51].

It should be kept in mind that the present analysis is valid for Switzerland, and directly depends on the modelled life-cycle of the polymers used in Switzerland[52]. Large variations in the uses of plastic in other countries might lead to a proxy having a much larger weight than the remaining proxies, for example in the case of agriculture[53], and thus leading to a different geographical repartition of the plastic emissions. For example, the agricultural sector can be more or less pronounced depending on the country, with varying plastic uses depending on the crops cultivated[53]. In a country with a strong agricultural sector, the validity of the population density as only proxy would be even lower. It should however be noted that the importance of traffic density and developed land use for plastic litter incidence was already demonstrated in a study in Iowa[54], which argues in favour of the population density as single proxy. Further studies could shed some light on the validity of this assumption.

The maps provided in this work represent emissions and should not be confused with concentration maps. The regionalization of emissions is a first step towards a fate model for macro- and microplastic. Based on the results of this study, a fate model could be adapted to account for all the processes occurring in the environment such as fragmentation, fluvial transport, run-off and sedimentation to name but a few[33,55,56]. Since the fate of different plastics in freshwater environments depends on polymer density and product application[32], a distinction between different polymers is essential as provided in this work. Littering has been suggested as the main plastic source in several instances[10,16], so the fragmentation of macroplastic to MP would

need to be implemented. Such a fate model would then permit to predict environmental concentrations in soil and water of macroplastic and MP, which in turn could enable to perform regionalized risk assessments.

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8.8 Supporting information

Additional figures and tables are available as Supporting Information. The generated dataset is available upon request.

8.9 Model availability

The R scripts written to perform the calculations are available at:

https://github.com/empa-tsl/plastic-reg

8.10 References

(1) Eerkes-Medrano, D.; Thompson, R. C.; Aldridge, D. C. Microplastics in Freshwater Systems: A Review of the Emerging Threats, Identification of Knowledge Gaps and Prioritisation of Research Needs. *Water Res.* **2015**, *75*, 63–82.

(2) Koelmans, A. A.; Mohamed Nor, N. H.; Hermsen, E.; Kooi, M.; Mintenig, S. M.; De France, J. Microplastics in Freshwaters and Drinking Water: Critical Review and Assessment of Data Quality. *Water Res.* **2019**, *155*, 410–422.

(3) Rios Mendoza, L. M.; Balcer, M. Microplastics in Freshwater Environments: A Review of Quantification Assessment. *TrAC - Trends Anal. Chem.* **2018**, *113*, 402–408.

(4) de Souza Machado, A. A.; Kloas, W.; Zarfl, C.; Hempel, S.; Rillig, M. C. Microplastics as an Emerging Threat to Terrestrial Ecosystems. *Glob. Chang. Biol.* **2017**, No. September.

(5) Nizzetto, L.; Futter, M.; Langaas, S. Are Agricultural Soils Dumps for Microplastics of Urban Origin? *Environ. Sci. Technol.* **2016**, *50* (20), 10777–10779.

(6) Horton, A. A.; Walton, A.; Spurgeon, D. J.; Lahive, E.; Svendsen, C. Microplastics in Freshwater and Terrestrial Environments: Evaluating the Current Understanding to Identify the Knowledge Gaps and Future Research Priorities. *Sci. Total Environ.* **2017**, *586*, 127–141.

(7) Gasperi, J.; Dris, R.; Bonin, T.; Rocher, V.; Tassin, B. Assessment of Floating Plastic Debris in Surface Water along the Seine River. *Environ. Pollut.* **2014**, *195*, 163–166.

(8) Castro-Jiménez, J.; González-Fernández, D.; Fornier, M.; Schmidt, N.; Sempéré, R. Macro-Litter in Surface Waters from the Rhone River: Plastic Pollution and Flows to the NW Mediterranean Sea. *Mar. Pollut. Bull.* **2019**, *146* (May), 60–66.

(9) Bauer-Civiello, A.; Critchell, K.; Hoogenboom, M.; Hamann, M. Input of Plastic Debris in an Urban Tropical River System. *Mar. Pollut. Bull.* **2019**, *144* (April), 235–242.

(10) Kiessling, T.; Knickmeier, K.; Kruse, K.; Brennecke, D.; Nauendorf, A.; Thiel, M. Plastic Pirates Sample Litter at Rivers in Germany – Riverside Litter and Litter Sources Estimated by Schoolchildren. *Environ. Pollut.* **2019**, *245*, 545–557.

(11) Rech, S.; Macaya-Caquilpan, V.; Pantoja, J. F.; Rivadeneira, M. M.; Jofre Madariaga, D.; Thiel, M. Rivers as a Source of Marine Litter - A Study from the SE Pacific. *Mar. Pollut. Bull.* **2014**, *82* (1–2), 66–75.

(12) Morritt, D.; Stefanoudis, P. V.; Pearce, D.; Crimmen, O. A.; Clark, P. F. Plastic in the Thames: A River Runs through It. *Mar. Pollut. Bull.* **2014**, *78* (1–2), 196–200.

(13) Schultz, P. W.; Bator, R. J.; Large, L. B.; Bruni, C. M.; Tabanico, J. J.; Brown Large, L.; Bruni, C. M.; Tabanico, J. J. Littering in Context. *Environ. Behav.* **2013**, *45* (1), 35–59.

(14) Cascadia Consulting Group Inc. *Washington 2004 State Litter Study: Litter Generation and Composition Report*; 2005.

(15) Litterati LLC. Litterati Global Map https://map.litterati.org/globalmap/ (accessed Sep 19, 2019).

(16) Kawecki, D.; Nowack, B. Polymer-Specific Modeling of the Environmental Emissions of Seven Commodity Plastics As Macro- and Microplastics. *Environ. Sci. Technol.* **2019**, *53* (16), 9664–9676.

(17) Essel, R.; Engel, L.; Carus, M.; Ahrens, R. H. *Sources of Microplastics Relevant to Marine Protection in Germany*; 2015.

(18) Ryberg, M. W.; Hauschild, M. Z.; Wang, F.; Averous-Monnery, S.; Laurent, A. Global Environmental Losses of Plastics across Their Value Chains. *Resour. Conserv. Recycl.* **2019**, *151* (March), 104459.

(19) Sieber, R.; Kawecki, D.; Nowack, B. Dynamic Probabilistic Material Flow Analysis of Rubber Release from Tires into the Environment. *Environ. Pollut.* **2019**, 113573.

(20) Hartmann, N. B.; Hüffer, T.; Thompson, R. C.; Hassellöv, M.; Verschoor, A.; Daugaard, A. E.; Rist, S.; Karlsson, T.; Brennholt, N.; Cole, M.; et al. Are We Speaking the Same Language? Recommendations for a Definition and Categorization Framework for Plastic Debris. *Environ. Sci. Technol.* **2019**, *53* (3), 1039–1047.

(21) Fischer, E. K.; Paglialonga, L.; Czech, E.; Tamminga, M. Microplastic Pollution in Lakes and Lake Shoreline Sediments - A Case Study on Lake Bolsena and Lake Chiusi (Central Italy). *Environ. Pollut.* **2016**, *213*, 648–657.

(22) Dris, R.; Gasperi, J.; Rocher, V.; Saad, M.; Renault, N.; Tassin, B. Microplastic Contamination in an Urban Area: A Case Study in Greater Paris. *Environ. Chem.* **2015**, *12* (5), 592–599.

(23) Browne, M. A.; Galloway, T. S.; Thompson, R. C. Spatial Patterns of Plastic Debris along Estuarine Shorelines. *Environ. Sci. Technol.* **2010**, *44* (9), 3404–3409.

(24) Goldstein, M. C.; Titmus, A. J.; Ford, M. Scales of Spatial Heterogeneity of Plastic Marine Debris in the Northeast Pacific Ocean. *PLoS One* **2013**, *8* (11).

(25) Eriksen, M.; Mason, S.; Wilson, S.; Box, C.; Zellers, A.; Edwards, W.; Farley, H.; Amato, S. Microplastic Pollution in the Surface Waters of the Laurentian Great Lakes. *Mar. Pollut. Bull.* **2013**, *77* (1–2), 177–182.

(26) Mani, T.; Hauk, A.; Walter, U.; Burkhardt-Holm, P. Microplastics Profile along the Rhine River. *Sci. Rep.* **2016**, *5* (1), 17988.

(27) Dikareva, N.; Simon, K. S. Microplastic Pollution in Streams Spanning an Urbanisation Gradient. *Environ. Pollut.* **2019**, *250*, 292–299.

(28) Wagner, S.; Klöckner, P.; Stier, B.; Römer, M.; Seiwert, B.; Reemtsma, T.; Schmidt, C. Relationship between Discharge and River Plastic Concentrations in a Rural and an Urban Catchment. *Environ. Sci. Technol.* **2019**, *53* (17), 10082–10091.

(29) Battulga, B.; Kawahigashi, M.; Oyuntsetseg, B. Distribution and Composition of Plastic Debris along the River Shore in the Selenga River Basin in Mongolia. *Environ. Sci. Pollut. Res.* **2019**, 14059–14072.

(30) Zubris, K. A. V; Richards, B. K. Synthetic Fibers as an Indicator of Land Application of Sludge. *Environ. Pollut.* **2005**, *138* (2), 201–211.

(31) Scheurer, M.; Bigalke, M. Microplastics in Swiss Floodplain Soils. *Environ. Sci. Technol.* **2018**, *52* (6), 3591–3598.

(32) Schwarz, A. E.; Ligthart, T. N.; Boukris, E.; van Harmelen, T. Sources, Transport, and Accumulation of Different Types of Plastic Litter in Aquatic Environments: A Review Study. *Mar. Pollut. Bull.* **2019**, *143* (March), 92–100.

(33) Hoellein, T. J.; Shogren, A. J.; Tank, J. L.; Risteca, P.; Kelly, J. J. Microplastic Deposition Velocity in Streams Follows Patterns for Naturally Occurring Allochthonous Particles. *Sci.*

Rep. **2019**, *9* (1), 1–11.

(34) Adam, V.; Yang, T.; Nowack, B. Toward an Ecotoxicological Risk Assessment of Microplastics: Comparison of Available Hazard and Exposure Data in Freshwaters. *Environ. Toxicol. Chem.* **2019**, *38* (2), 436–447.

(35) Zimmermann, L.; Dierkes, G.; Ternes, T. A.; Völker, C.; Wagner, M. Benchmarking the in Vitro Toxicity and Chemical Composition of Plastic Consumer Products. *Environ. Sci. Technol.* **2019**, *53* (19), 11467–11477.

(36) Lebreton, L. C. M.; Zwet, J. Van Der; Damsteeg, J.; Slat, B.; Andrady, A.; Reisser, J. River Plastic Emissions to the World's Oceans. *Nat. Commun.* **2017**, *8*, 1–10.

(37) Unice, K. M.; Weeber, M. P.; Abramson, M. M.; Reid, R. C. D.; van Gils, J. A. G.; Markus, A. A.; Vethaak, A. D.; Panko, J. M. Characterizing Export of Land-Based Microplastics to the Estuary - Part I: Application of Integrated Geospatial Microplastic Transport Models to Assess Tire and Road Wear Particles in the Seine Watershed. *Sci. Total Environ.* **2019**, *646*, 1639–1649.

(38) Swiss Federal Statistical Office. Statistique de la population et des ménages (STATPOP) dès 2010 https://www.bfs.admin.ch/bfs/fr/home/services/geostat/geodonnees-statistique-federale/batiments-logements-menages-personnes/population-menages-depuis-2010.html.

(39) Swiss Federal Statistical Office. Statistique de la superficie selon nomenclature 2004 – Standard https://www.bfs.admin.ch/bfs/fr/home/services/geostat/geodonnees-statistique-federale/sol-utilisation-couverture/statistique-suisse-superficie.html.

(40) Swiss Federal Statistical Office. Statistique des bâtiments et des logements (StatBL) https://www.bfs.admin.ch/bfs/fr/home/services/geostat/geodonnees-statistique-federale/batiments-logements-menages-personnes/batiments-logements-des-2010.html.

(41) Senozon AG im Auftrag des Bundesamts für Umwelt BAFU. *SonBASE – Verkehrsdaten Schweiz 2015*; 2017.

(42) Federal Office for the Environment FOEN. Géodonnées sur la subdivision de la Suisse en bassins versant https://www.bafu.admin.ch/bafu/fr/home/themes/eaux/etat/cartes/geodonnees-sur-la-subdivision-de-la-suisse-en-bassins-versant.html.

(43) Federal Office for the Environment FOEN. *Adresses Des Stations d'épuration Avec Mention de Leur Capacité de Traitement*; 2017.

(44) OpenStreetMap Foundation. OpenStreetMap https://www.openstreetmap.org/copyright.

(45) Federal Office for the Environment FOEN. VSA Kennzahlen. *Obtained upon Req.* **2011**.

(46) Mutzner, L.; Staufer, P.; Ort, C. Model-Based Screening for Critical Wet-Weather Discharges Related to Micropollutants from Urban Areas. *Water Res.* **2016**, *104*, 547–557.

(47) Federal Office of Topography swisstopo. *Formulas and Constants for the Calculation of the Swiss Conformal Cylindrical Projection and for the Transformation between Coordinate Systems*; 2016.

(48) Cabernard, L.; Durisch-Kaiser, E.; Vogel, J.-C.; Rensch, D.; Niederhauser, P.; AWEL Gewässerschutz. Mikroplastik in Abwasser Und Gewässern. *Aqua Gas* **2016**, *7* (8), 78–85.

(49) Keizer, K.; Lindenberg, S.; Steg, L. The Spreading of Disorder. *Science (80-.).* **2008**, *322* (5908), 1681–1685.

(50) Rangoni, R.; Jager, W. Social Dynamics of Littering and Adaptive Cleaning Strategies Explored Using Agent-Based Modelling. *J. Artif. Soc. Soc. Simul.* **2017**, *20* (2).

(51) Weaver, R. Littering in Context(s): Using a Quasi-Natural Experiment to Explore Geographic Influences on Antisocial Behavior. *Appl. Geogr.* **2015**, *57*, 142–153.

(52) Kawecki, D.; Scheeder, P.; Nowack, B. Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe. *Environ. Sci. Technol.* **2018**, *52*, 9874–9888.

(53) Scarascia-Mugnozza, G.; Sica, C.; Russo, G. Plastic Materials in European Agriculture: Actual Use and Perspectives. *J. Agric. Eng.* **2012**, *42* (3), 15–28.

(54) Cowger, W.; Gray, A. B.; Schultz, R. C. Anthropogenic Litter Cleanups in Iowa Riparian Areas Reveal the Importance of Near-Stream and Watershed Scale Land Use. *Environ. Pollut.* **2019**.

(55) Besseling, E.; Quik, J. T. K.; Sun, M.; Koelmans, A. A. Fate of Nano- and Microplastic in Freshwater Systems: A Modeling Study. *Environ. Pollut.* **2017**, *220*, 540–548.

(56) Williams, A. T.; Simmons, S. L. Movement Patterns of Riverine Litter. *Water. Air. Soil Pollut.* **1997**, *98* (1–2), 119–139.

9 Conclusion

This thesis was divided into three steps. The first step aimed at creating a model for the life-cycle of specific polymers, so that environmental emissions could be modelled from each process in a second step. In a third and final step, the final emission flows obtained were regionalized to obtain spatially-differentiated emissions.

9.1 On the presented results

The life-cycle modelled comprised 84 compartments (including the modifications of the life-cycle in the second publication) connected with 168 flows. This high number of compartments was achieved because of the level of detail required for the consumption stage, in which 45 product categories were modelled, including textile applications which are usually not considered in plastic life-cycle analyses. Since, the consumption stage was critical to be able to predict emission flows, trade also needed to be included in stages before and at the consumption stage, including for in-use packaging and semi-finished goods. More in-depth research about this aspect is still needed to verify the assumptions used. The models for LDPE, HDPE, PP, PS, EPS, PVC and PET used an identical framework and are therefore fully comparable. A total of 110000±14000 t of LDPE, 121000±14000 t of HDPE, 160000±17000 t of PP, 55200±6000 t of PS, 31200±3900 t of EPS, 101000±13000 t of PVC and 131200±9900 t of PET were consumed in Switzerland in 2014. Most of the plastic consumed was in the form of packaging and construction plastics. The vast majority of this plastic is then incinerated in Switzerland, at 76.2±3.1 % of LDPE, 77.0±3.1 % of HDPE, 78.8±2.2 % of PP, 88.2±2.1 % of PS, 89.1±1.6 % of EPS, 84.8±2.7 % of PVC and 74.3±2.4 % of PET.

A total of 50 compartments and 234 flows were added to this to model the emission flows. Of these compartments, 11 represent environmental compartments and 39 technical or virtual processes which are on the way from the primary emission to the final release. The emissions modelling of polymer flows revealed that littering is very likely to be the most important pathway of plastic to the environment in Switzerland. The amounts littered were explicitly modelled, with other mismanagement waste flows modelled separately. To the best of the author's knowledge, no previous study had distinguished the different mechanisms underlying the mismanagement of waste, which is important to design appropriate policies since the behavioural patterns underlying littering, dumping and accidental releases are very different. Other important emission pathways include accidental releases of litter from industry, construction sites and agricultural activities. The majority of plastic emissions occur in the shape of macroplastic

at 87.5±3.3 % of the total modelled emissions. This macroplastic may then fragment into microplastic in the environment depending on the weathering conditions. All polymers combined, a total of 630±150 grams per capita are emitted yearly in Switzerland. Of this total, 610±150 grams/capita/year are initially emitted to soil and 15.1±5.1 g/cap/y to water. This means that soils receive around 40-times more plastic as initial emissions than waters according to these results, which calls for more research on plastic pollution in soil environments and on the transport processes between soil and water. Most of the environmental emissions can be attributed to PET at 200±120 g/cap/y, followed by PP at 126±43 g/cap/y, HDPE at 98±50 g/cap/y, LDPE at 94±34 g/cap/y, PVC at 65±36 g/cap/y, PS at 24±13 g/cap/y, and finally EPS at 16±12 g/cap/y.

For the first two articles published as part of this thesis, the method called Probabilistic Material Flow Analysis (PMFA) was used to model the flows of polymers through society and to the environment. This method had already been applied to model the environmental emissions of nanomaterials[1,2]. The Bayesian probability distributions used in this method permit to incorporate the data uncertainty into the model and reflect it in the uncertainty of the final results. The expected confidence in the results can then be presented in a transparent manner. More often than not, models such as the one presented in this thesis are limited by the amount of data available. The data availability was expected to be higher for polymer flows than for nanomaterial flows as in previous works[2,3], it was nevertheless surprising to see that a lot of information was either not made available to the public or researchers, or non-existing. The uncertainty attributed to the individual parameters in the model was based on a Pedigree matrix approach adapted for MFA models[4], which permits to quantify the uncertainty of the original source based on a set of criteria. The advantage of this approach is its total transparency to the reader, but like the former approaches, the definition of these probability distributions has not been validated. As a result, the interpretation of the probability distributions is entirely Bayesian[5], i.e. the distributions reflect the confidence in the result as proposed by the model based on the original data.

The regionalization model combined eleven different geodatasets with the final emission flows modelled from the second publication. Emission maps were created using the mean, 5th and 95th percentiles of the emission flows of microplastic and macroplastic to soil, water and air. The results showed that the local emission rates can vary by at least two orders of magnitude over Switzerland. Higher emissions were concentrated in areas with high human activity, i.e. urban areas and agricultural areas. The influence of the main applications of each polymer can be recognized in the maps obtained, for example for polymers largely used in agriculture or

packaging which can be littered along roadsides. It was also demonstrated that using the population density as only proxy with a high spatial resolution may lead to large deviations in the emission maps and that this behaviour was more present for polymers used extensively in agriculture.

A comparison of the final results of the model with measurements would be ideal in order to validate it. It is unfortunately mostly impossible to find measurements of emission flows, since most of them consist of single unsystematic release events. A comparison may be possible for specific flows, as for example WWTP effluent as was done in the second publication. Yet a comparison with environmental samples would only be possible with a fully developed fate model using the regionalized dataset.

9.2 Outlook

The research conducted for this thesis may be further developed in several directions. Some of these aspects are already under scrutiny, sometimes even based upon the here presented work.

A framework for describing the emission flows in a consistent manner was developed as part of the second study presented in this thesis[6], but many of the modelled emission flows may profit from an improved description. This is especially important for emission flows responsible for large emissions according to this article. A more detailed study on the sweeping rate of litter on roadsides and in natural environments would improve the accuracy and precision of the estimates of litter remaining in these environments. Little information is available on the emissions from agricultural activities and construction and demolition sites. The estimates in this study needed to rely on coarse estimates and would profit greatly from in-depth studies. It is further relevant to note that more research on the releases of microplastic to indoor air should be carried out, since the shedding of fibres through other types of wear than washing is unclear for all types of textiles. It is for example unclear if household textiles as carpets and curtains act more as a source or as a sink of household dust in indoor environments[7]. Such questions are important for assessing the human exposure through inhalation and ingestion of household dust, and are also related to environmental exposure through housing ventilation systems.

Another direction in which the model could be improved is the inclusion of a temporal dimension. A tool for Dynamic Probabilistic Material Flow Analysis (DPMFA) is available[8] and could be used to estimate the total amount of plastic released since the 1950s which would be

needed for modelling the current concentrations of microplastic originating from the fragmentation of macroplastic. Including a temporal dimension in the life-cycle model would as well enable to estimate the magnitude of in-use stocks of plastic and future trends in waste generation. The absence of temporal dimension was discussed in the first publication, showing that the impact was restricted to products with very long lifetimes as construction plastics. For this reason, absolute masses of waste generation were avoided in the first publication, but the proportions of end-of-life options per sector could still be calculated. In the second publication revolving around the emissions, the impact of this assumption is also limited, since construction plastics are not the leading source. Nevertheless, such an extension is necessary to be able to predict long-term emission trends. The first steps of the research in this direction were conducted during a master thesis and an internship in parallel to this thesis[9,10], in which the life-cycles of the same seven polymers were modelled from 1950 to 2017. These results could not yet be finalized as scientific publications but work is ongoing. Knowing how much waste will be produced and what plastic quality can be expected is key for setting appropriate recycling targets and increasing the circularity of the plastic supply chain[11]. Additional information on the purity of the waste streams would be necessary for that purpose as well, including information on the additive content in the streams to determine what the possible applications for recycled plastic are.

Last, more plastic materials could also be considered for further modelling studies. In this thesis, only seven commonly used thermoplastics were considered, but many more other synthetic materials exist which could be of interest. For instance, polyurethane was not included in this project, but it represented 7.5% of the European manufacturer's demand in 2017[12] and might cause important emissions as well. Moreover, other polymers which are commonly mentioned as sources of microplastic were not included, as for example microplastic from paint and tyres. Some research in this direction was already performed in parallel to this thesis in the same research group for the case of tires[13]. This model used the DPMFA method mention earlier and used a similar modelling approach, but is not included in this thesis. Another study also recently published emission flows with a global focus for 23 different categories of polymers or application types[14].

The overall goal of this thesis was to provide a first step towards a calculation of Predicted Environmental Concentrations (PECs). The natural next step for this goal is to develop a fate model including all processes occurring in and between soil, water and air for microplastic and macroplastic. First fate models describing the dynamics of microplastic in freshwater exist[15,16], enabling to model the retention of microplastic in a river network. Further research needs to

be conducted on the fate processes influencing microplastic in soil and air and the transport processes between soil and these compartments. Macroplastic fate models for both freshwater and soil are missing and would need to be developed as well. A fully developed fate model could then use the emission maps created in the third publication as input and calculate PECs based on gathered knowledge on fate processes such as runoff, deposition, sedimentation, aggregation and fragmentation for example. Calculating PECs would as well permit a validation of the complete modelling approach presented in this thesis. As mentioned earlier, the emission flows can only be compared to measured data in a few rare instances. Most of the emission flows are too spread out in space and time to be measured, and a comparison is as a consequence impossible. Yet, PECs can be compared to measured environmental concentrations, which can hopefully improve our knowledge of the emission flows and their spatial distribution.

Finally, in order to provide guidance on appropriate mitigation measures, the inclusion of plastic emissions in Life Cycle Inventories (LCI) could provide the missing step for plastic products Life Cycle Assessments (LCA). Emission factors may be derived from the results of the second publication which could be used to assess the impact of plastic compared to alternatives. Similarly, the PECs as predicted from a fate model would permit to develop a spatially resolved risk assessment, and thus give insight into the impact of plastic in different environments.

9.3 References

(1) Gottschalk, F.; Scholz, R. W.; Nowack, B. Probabilistic Material Flow Modeling for Assessing the Environmental Exposure to Compounds: Methodology and an Application to Engineered Nano-TiO2 Particles. *Environ. Model. Softw.* **2010**, *25* (3), 320–332.

(2) Caballero-Guzman, A.; Sun, T.; Nowack, B. Flows of Engineered Nanomaterials through the Recycling Process in Switzerland. *Waste Manag.* **2015**, *36*, 33–43.

(3) Wang, Y.; Kalinina, A.; Sun, T.; Nowack, B. Probabilistic Modeling of the Flows and Environmental Risks of Nano-Silica. *Sci. Total Environ.* **2016**, *545–546*, 67–76.

(4) Laner, D.; Rechberger, H.; Astrup, T. Systematic Evaluation of Uncertainty in Material Flow Analysis. *J. Ind. Ecol.* **2014**, *18* (6), 859–870.

(5) Cox, R. T. Probability, Frequency and Reasonable Expectation. *Am. J. Phys.* **1946**, *14* (1), 1–13.

(6) Kawecki, D.; Nowack, B. Polymer-Specific Modeling of the Environmental Emissions of Seven Commodity Plastics As Macro- and Microplastics. *Environ. Sci. Technol.* **2019**, *53* (16), 9664–9676.

(7) Dris, R.; Gasperi, J.; Mirande, C.; Mandin, C.; Guerrouache, M.; Langlois, V.; Tassin, B. A

First Overview of Textile Fibers, Including Microplastics, in Indoor and Outdoor Environments. *Environ. Pollut.* **2017**, *221*, 453–458.

(8) Bornhöft, N. A.; Sun, T. Y.; Hilty, L. M.; Nowack, B. A Dynamic Probabilistic Material Flow Modeling Method. *Environ. Model. Softw.* **2016**, *76*, 69–80.

(9) Flükiger, P. Dynamic Probabilistic Material Flow Analysis of Seven Commodity Plastics in Switzerland, ETH Zürich, 2019.

(10) Wu, Q. Dynamic Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe, ETH Zurich, 2018.

(11) European Commission. *A European Plastics Strategy For Plastics In A Circular Economy*; 2017.

(12) PlasticsEurope. *Plastics – the Facts 2017: An Analysis of European Plastics Production, Demand and Waste Data*; 2017.

(13) Sieber, R. Microplastic Release from Tires into the Environment - Dynamic Probabilistic Material Flow Analysis of Tire Wear Particles in Switzerland, ETH Zurich, 2019.

(14) Ryberg, M. W.; Hauschild, M. Z.; Wang, F.; Averous-Monnery, S.; Laurent, A. Global Environmental Losses of Plastics across Their Value Chains. *Resour. Conserv. Recycl.* **2019**, *151* (March), 104459.

(15) Unice, K. M.; Weeber, M. P.; Abramson, M. M.; Reid, R. C. D.; van Gils, J. A. G.; Markus, A. A.; Vethaak, A. D.; Panko, J. M. Characterizing Export of Land-Based Microplastics to the Estuary - Part I: Application of Integrated Geospatial Microplastic Transport Models to Assess Tire and Road Wear Particles in the Seine Watershed. *Sci. Total Environ.* **2019**, *646*, 1639–1649.

(16) Besseling, E.; Quik, J. T. K.; Sun, M.; Koelmans, A. A. Fate of Nano- and Microplastic in Freshwater Systems: A Modeling Study. *Environ. Pollut.* **2017**, *220*, 540–548.

APPENDICES

10 Supporting Information for: Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe

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10.1 Details on the method

10.1.1 PMFA theory

Let there be a system containing n processes, of which m are not sinks. The flow from process i to process j is defined using a transfer coefficient (TC). This TC is defined as the ratio of the outflowing mass to the mass contained in the process:

$$TC_{ij} = \frac{m_{ij}}{M_i}.$$

All the transfer coefficients needed to describe the flows in the system can be gathered into a matrix as follows:

$$A = \begin{pmatrix} \mathbf{1} & \cdots & -TC_{m1} & 0 & \cdots & 0 \\ \vdots & \ddots & \vdots & \vdots & \vdots & \vdots \\ -TC_{1m} & \cdots & \mathbf{1} & 0 & \cdots & 0 \\ -TC_{1m+1} & \cdots & -TC_{mm+1} & \mathbf{1} & \cdots & 0 \\ \vdots & \cdots & \vdots & \vdots & \ddots & \vdots \\ -TC_{1n} & \cdots & -TC_{mn} & 0 & \cdots & \mathbf{1} \end{pmatrix},$$

where the TCs in column i describe the outgoing flows from process i to processes $1, \dots, n$. The columns until m contain non-zero values for TCs. The remaining columns correspond to sinks, from which no outflows are defined.

The Probabilistic Material Flow Analysis (PMFA) method relies on solving a matrix equation iteratively. The matrix equation reads:

$$A \cdot X = I$$

where $A \in (\mathbb{R}^n \times \mathbb{R}^n)$ is a matrix describing the flows between n processes as described above, $I \in \mathbb{R}^n$ is a vector describing the initial material input into n processes, and $X \in \mathbb{R}^n$ describes the final mass in n processes. This equation system is solved a number of times in a Monte-Carlo simulation, where each parameter is sampled from chosen Bayesian distributions.

10.1.2 Uncertainty attribution

The distribution spread for a specific parameter is determined via a semi-quantitative approach inspired by another MFA study[1]. This approach is favoured over other methodologies described for Life Cycle Inventory data[2], as it permits to produce a continuous range of uncertainties. Five different data quality indicators are used to represent the different uncertainty sources[1,3]. The Data Quality Indicator Scores (DQIS) can take a value between 1 and 4, with high values corresponding to poorer data quality (Table S1).

Table S1: Pedigree matrix with 5 data quality indicators and 4 data quality levels.

Data quality	**Very good**	**Good**	**Poor**	**Very poor**
DQIS	**1**	**2**	**3**	**4**
Geographical representativeness	Same region *(EU28 and EU28+2 qualify)*	Socioeconomically similar region *i.e. Europe vs. Switzerland*	Socioeconomically different region *i.e. USA vs. Europe*	Socioeconomically very different region *i.e. World vs. Europe*
Temporal representativeness	2014	2009-2013, 2015	2004-2008	Prior to 2004
Material representativeness	Same polymer	Same polymer datum corrected with data for all polymers	Data for a different polymer, or for plastic as a whole, or including similar materials	Including non-similar materials
Completeness	Includes all relevant processes/flows	Includes main processes/flows	Partially including main processes/flows	Important processes/flows are missing
Source reliability	Official report Peer reviewed documentation	Market reports and other reports Public database	Qualified estimate	Non-qualified estimate

The DQIS are transformed into Coefficients of Variation (CV) with an exponential function, as it better reflects the qualitative evaluation of the data quality indicators listed above (Table S1) than a linear function. All parameters are assumed to have high sensitivity to the DQIS (for details, see the original article[1]). The CVs are parametrized differently for the source reliability indicator and the other indicators, to allow for a non-zero uncertainty even for good source reliabilities:

$$\mathrm{CV}_{rel} = 1.5 \cdot e^{1.105 \cdot \mathrm{DQIS}}$$
$$\mathrm{CV}_{other} = 1.5 \cdot e^{1.105 \cdot (\mathrm{DQIS}-1)}.$$

The total CV of a datum is then calculated from the individual CVs using the variance additivity rule:

$$\mathrm{CV}_{tot} = \sqrt{{\mathrm{CV}_{geo}}^2 + {\mathrm{CV}_{temp}}^2 + {\mathrm{CV}_{mat}}^2 + {\mathrm{CV}_{tech}}^2 + {\mathrm{CV}_{rel}}^2}.$$

The described methodology is valid for normal distributions. As we prefer to use triangular distributions and other distributions based on triangular distributions (see next section for a complete description), we would like to obtain information on the maximal spread of the distribution, which is the required information to create a symmetric triangular distribution.

Knowing that the variance of a triangular distribution can be expressed as follows:

$$\sigma^2 = \frac{a^2 + b^2 + c^2 - ab - ac - bc}{18},$$

where a, b and c are the minimum, maximum and modal values of the triangular distribution. With the following constraint, the symmetry of the triangular distributions is enforced:

$$b = 2c - a\,.$$

The standard deviation can then be rewritten:

$$\sigma = \frac{1}{\sqrt{6}}(a - c)$$

Knowing that the CV is defined as the standard deviation divided by the mean, the distance from the mean to one extreme value of the triangular distribution can be expressed as such:

$$\frac{a - c}{c} = \sqrt{6} \cdot \frac{\sigma}{c} \cong 2.45 \cdot \mathrm{CV}_{tot}$$

This final expression is the one used to compute the distributions' spread. A comparison of a normal and a triangular distribution is shown in Figure S23.

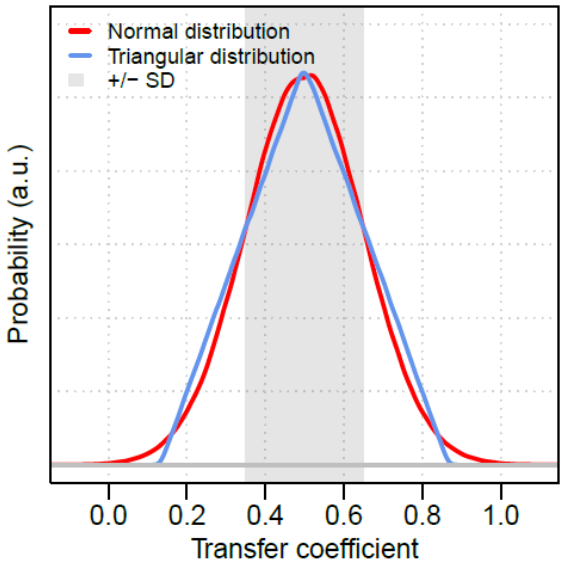

Figure S23: Comparison of a normal and triangular distribution which have the same parameters: $\mu = 0.5$, $CV_{tot} = 0.3$**. The triangular distribution's spread was calculated from** CV_{tot} **as described above.**

10.1.3 Probability distributions

Depending on the amount of data available for a specific parameter in the model, different probability distributions are used to model the data (Figure S24):

1. If a single data point is available, the distribution chosen is a triangular distribution (Figure S24a) centred on the data point, with a maximal spread defined by the pedigree matrix approach explained in the previous chapter.
2. If two data points are available, they are combined in a trapezoidal distribution (Figure S24b), where the lower and upper tails are defined using the CVs associated to the two data points.
3. If three or more data points are available, they are combined together in what we call a step function. Uniform distributions are built between two consecutive data points. Equal weights are attributed to each uniform distribution. The upper and lower tails of the step distribution are built using the CVs of variation for the extreme data points, while ensuring semi-continuity with the adjacent uniform distribution.

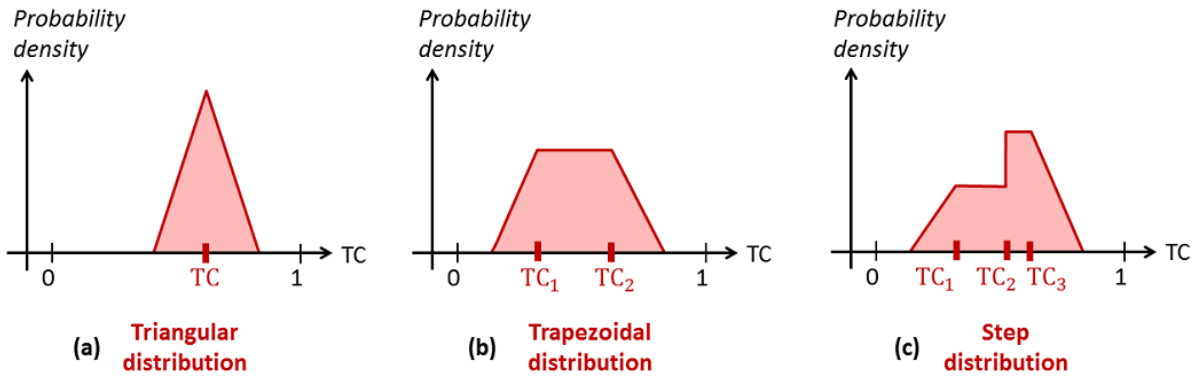

Figure S24: Schematic representation of the distributions chosen depending on the number of available data points: (a) for one, (b) for two, (c) for more than two.

The approach used, in which a distribution's spread is defined by a central value and a CV, may lead to values that are unphysical. For example, obtaining transfer coefficients (TCs) smaller than 0 or larger than 1 would violate mass conservation. In such cases, the approach chosen is to truncate the distribution at the threshold values (Figure S25).

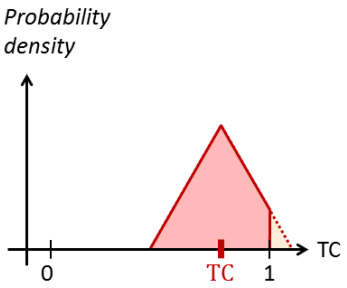

Figure S25: If the spread chosen for a data point would lead the distribution to have values larger than what is physically realistic, a simple truncation is applied to the distribution.

10.1.4 Algorithm: Data preparation and calculation

The following describes the order in which operations are performed, in order to set up the model, calculate export flows, normalizing the system and running the simulation.

1. **Import of parameters:** The Bayesian distributions for the TCs and the inflows into the system are generated.

2. **Normalization of the TC distributions:** The sum of the TC distributions from a specific process is fixed to 1.
3. **Calculation of export:** The mean TCs for export flows are calculated, by solving the equation system with the mean values of all parameter distributions. The TC distributions are constructed using the calculated mean TCs and the uncertainty attributed to the export flow. This stage includes a renormalization process.
4. **Calculation of special flows:**
    a. Simultaneously, the TC for PET from *Transport* to *Fibre Production* is calculated. This calculation is done after the export calculation for the production and manufacturing stage. Since no export of PET fibres and filaments occurs at this life-cycle stage, this TC can be calculated here. As both TCs out of *Transport* are calculated simultaneously, no renormalization is required.
    b. At the end of the export calculation, the TC distributions from *Other* to *PCCP* are separately calculated. The fraction attributed to *PCCP* is removed from the *Other* product category, so no renormalization is required.
5. **Monte Carlo Simulation:** The matrix equation is solved iteratively 10^6 times.

10.2 Details on transfer coefficients used

In the following description, the distinction between TC distribution and parameter used to generate the distribution will not be made, in order to improve readability. Nevertheless, each time a TC or single number is mentioned, the parameter used to generate the Bayesian distributions is meant.

10.2.1 Production and manufacturing

A certain amount of waste is generated during each production and manufacturing process. The plastic waste generated during the primary production and non-textile manufacturing phases can be estimated comparing the production and non-textile manufacturing waste mass produced in Germany in 2015[4]. One obtains that 0.59% waste arises during production and 6.63% during non-textile manufacturing. We assume that similar amounts are discarded for all the polymers in Europe or Switzerland. We also assume that 0.59% waste occurs during pelletizing from recycled plastic. The amounts of waste produced during fibre production and

textile manufacturing were taken from a report on nano-textiles[5] for both Europe and Switzerland for all polymers. It is estimated that 3% waste arises during fibre production and 10% during textile manufacturing.

From transport, plastic in primary forms can flow to textile and non-textile manufacturing. The fibre market is significant for three of the seven polymers considered: PET, PP and HDPE (in order of significance). Other fibres are neglected in this study, as they have a very narrow application range[6] and are not mentioned anywhere in market reports on plastic applications. The share of PET in primary forms used in fibre production or non-textile manufacturing is estimated by comparing the total mass of PET in primary forms available to the mass of PET fibres produced in Europe in 2014. The mass of PET fibres produced in the EU28 in 2014 can be estimated from the known polyester production in the EU28 and Turkey in 2014 which amounts to 1'030 kt[7]. 448 kt of polyester were produced in Turkey in 2013[8], a change of 5% to the previous year's figure. Assuming the growth from 2013 to 2014 is identical, one finds that 470 kt of polyester were produced in Turkey in 2014. As a consequence, the European polyester production amounted to 560 kt in 2014, to the best of our knowledge. The TC is automatically calculated for the value of 560 kt considering the 3% of waste produced during fibre production. For HDPE and PP, the TC are estimated from the market shares of fibres for Europe in 2015[9]: 20% for PP and 1% for HDPE. These numbers are very close to the ones suggested by a British study for the UK[10]. The Swiss total plastic demand for fibre production is slightly lower than in Europe: 4% instead of 5%[9], but since this difference is small, we assume that the same fibre market shares by polymer can be used for Switzerland and for Europe.

The size of the non-textile product sectors by polymer in Europe are obtained from a report from PlasticsEurope[11]. This report was favoured over other studies, as it was the only one found that uses the same sectors for all polymers, and thus facilitates the comparison amongst them. Nevertheless, their product sector data is in agreement with other studies that use varying sectors for different polymers[9,10]. The size of the agriculture sector needed to be calculated separately (see section on agriculture product categories). In the case of Switzerland, different data sets are available: Swiss sector data by polymer based on German data for 2005[12] or Swiss sector data all polymers combined for 2015[9] or for a 5-year average[13,14]. As the first data set is already older, and seeing that the German plastic markets in 2005 and 2015 are quite different[9,12], we estimate that using Swiss sectors all polymers combined to recalibrate the European sector by polymer decomposition is more appropriate than using the older German data.

The size of the textile sectors clothing, household textiles and technical textiles for both Europe and Switzerland are taken from two reports. For PET, the figures are taken from a report giving the global polyester fibre segmentation for 2012[15]. The sector sizes for PP are taken from a web-based resource describing the applications of PP textiles in the EU28 and Turkey in 2013[16]. The HDPE textile sectors were assumed to be identical to the ones of PP, since HDPE and PP fibres are commonly referred to as polyolefin fibres and very little data can be found on HDPE fibre applications alone.

10.2.2 Packaging product categories

The packaging product categories are constructed from a bottom-up assessment of the plastic packaging market in the UK for 2011[17]. This study provides shares of different applications for the different polymers, distinguishing consumer and non-consumer applications. A distinction is also made in the non-consumer applications between packaging for agricultural products, construction products or other. The reported 68% of consumer packaging out of the total packaging mass is in good agreement to the estimated 65-75%[18], 64%[19], and 73%[20] in other studies.

10.2.3 Construction product categories

The construction product categories are taken from a report that covers Western Europe in 1995[21].

10.2.4 Automotive product categories

The automotive sector is not split into more categories.

10.2.5 EEE product categories

The EEE sector is not split into more categories.

10.2.6 Agriculture product categories

The agriculture product categories are constructed using information from several studies[11,18,22–26]. The agriculture sector is estimated to constitute 3.4% of all plastic manufacturers' demand in Europe in 2014[22]. Since this sector is very diverse, little data can be found on the repartition among polymers and applications for Europe. This repartition has to be estimated

according to data available for different systems (Western Europe and Italy). The chosen product categories are films, pipes and tubings and other applications.

Films are said to account for 71[23]-74%[18] of the total agriculture sector, not including agricultural packaging in Western Europe. The mean value of these two estimates is used: 72.5%.

Agricultural films are constituted of LDPE, PVC and EVA[24]. Since LDPE films are said to account for 60% of all plastic in agriculture[25], 12.5% are left for both PVC and EVA films. We assume that PVC and EVA are consumed in equal amounts in the different film applications (greenhouse, tunnel, mulching, silage and other coverings) when they are mentioned in the Italian agricultural plastic consumption data[24]. We obtain that 2.05% of plastic in agriculture are PVC films, and 10.45% are EVA films, which are not considered further.

As next step, knowing that LDPE constitutes 81% of the Western Europe consumption in agricultural plastics[23,26], we can attribute the missing 21% of LDPE to pipes and other applications. The repartition between the two follows the same method as for the films. We obtain that 11.8% of agricultural plastics are LDPE pipes, and 9.2% are other LDPE applications.

Similarly, knowing that the PVC agriculture sector constitutes 1.9%[26] of the total PVC German manufacturer's demand, and knowing that PVC represents 10.27% of the total plastic manufacturer's demand in Europe[11], we can estimate that 5.7% of agricultural plastics consist of PVC.

$$\frac{m(\mathrm{PVC_{Agri}})}{m(\mathrm{Plastic_{Agri}})} = \left[\frac{m(\mathrm{PVC_{Agri}})}{m(\mathrm{PVC})}\right] \cdot \frac{\left[\frac{m(\mathrm{PVC})}{m(\mathrm{Plastic})}\right]}{\left[\frac{m(\mathrm{Plastic_{Agri}})}{m(\mathrm{Plastic})}\right]} = 1.9\% \cdot \frac{10.27\%}{3.4\%} \approx 5.7\%$$

Knowing from above that 2.05% of PVC are used in films, 3.65% are left for pipes and other applications. Following the same approach as above with the Italian consumption data, we obtain that 2.15% are included in pipes, and 1.5% in other applications.

The total remaining unknown proportions of pipes and other applications amounts to 2.85% of the total sector. Aside from LDPE and PVC, agricultural pipes are constituted of HDPE and other polymers not considered here, while other applications (fibre covers, nets, sheets, pots, twine) consist of HDPE, PP, PS and other non-considered polymers[24]. The repartition is estimated in the same way using the Italian consumption data.

Table S2: Composition of the agriculture sector by polymer and application

	LDPE	HDPE	PP	PS	EPS	PVC	PET	Other	***Total***
Films	60%	0	0	0	0	2.05%	0	10.45%	*72.5%*
Pipes	11.8%	0.4%	0	0	0	2.15%	0	0.8%	*15.15%*
Other	9.2%	0.4%	0.35%	0.3%	0	1.5%	0	0.55%	*12.3%*
Total	*81%*	*0.8%*	*0.35%*	*0.3%*	*0*	*5.7%*	*0*	*11.8%*	***100%***

The agricultural product categories can be obtained by normalizing this repartition by the total share of the polymer in the agricultural sector. It is also possible to calculate the size of the agriculture sector by polymer using the fact that 3.4% of plastics are agricultural plastics, and the share of one polymer out of all the plastics.

$$\frac{m(\text{Polymer}_{\text{Agri}})}{m(\text{Polymer})} = \left[\frac{m(\text{Polymer}_{\text{Agri}})}{m(\text{Plastic}_{\text{Agri}})}\right] \cdot \frac{\left[\frac{m(\text{Plastic}_{\text{Agri}})}{m(\text{Plastic})}\right]}{\left[\frac{m(\text{Polymer})}{m(\text{Plastic})}\right]}.$$

A similar approach is used for the Swiss agricultural product sector, using two data points for the share of agricultural plastics out of all plastics: the same 3.4% as above and 0.3% from a Swiss study[13].

Table S3: Fraction of each polymer used in agriculture

System for which it is used	LDPE	HDPE	PP	PS	EPS	PVC	PET	Other
Europe, Switzerland	16.01%	0.23%	0.06%	0.25%	0%	1.89%	0%	1.48%
Switzerland	1.48%	0.02%	0.01%	0.02%	0%	0.11%	0%	0.15%

10.2.7 Clothing product categories

The clothing sector is not further split into product categories as it has a limited diversity in terms of use and waste treatment.

10.2.8 Household textile product categories

The household textile sectors is also not further split into product categories,

10.2.9 Technical textile product categories

Technical textiles are commonly split into 12 application fields[27], of which only some will be described separately here: technical clothing (26% of the global sales in 2010[28]), mobility textiles (23%), agrotextiles (7%), building textiles (7%), technical household textiles (7%), hygiene and medical textiles (7%), geotextiles (1%) and other technical textiles (22%). The data used for these categories is mostly taken from a global perspective[28], but also from more specific information on PP[16]. The HDPE product categories were assumed to be identical to the ones of PP.

10.2.10 Other product categories

The *Other* product sector was divided into product categories following available data. The applications that were separately mentioned for at least one polymer are included here: fabric coatings[9], household items[9], cosmetics[29], and furniture[9,10]. For polymers where one of these categories is not explicitly mentioned in the literature, it can be that the *Other* category contains shares of the previously mentioned product categories. An additional product category for plastic used as an ingredient in Personal Care and Cosmetic Products (PCCP) was created based on the reported mass[30] of polyethylene and plastic used in Europe and Switzerland as microbeads in cosmetic applications. It is assumed here that all of the polyethylene used is HDPE, because of its lower softness than LDPE. We also assume that 0.2% of this reported plastic mass is PP and 0.3% is PET, following proportions of mentioned polymers in a database of plastic in cosmetic products on the market[31]. The remaining portion consists mostly of polyacrylates which are not further considered here. We assume that the same product categories hold for Switzerland, even though some large Swiss companies had renounced to using polyethylene in cosmetics in 2014 (Private communication with Ulrich Eicken, 17.11.2016).

10.2.11 Waste collection

Packaging waste is modelled to either be collected in mixed waste or separately. The European packaging collection rates chosen are estimates for 2015 made in 2005[18], except in the case of PET bottles, where the collection rate is known[32]. The Swiss packaging collection rates are taken from a study reporting recycling rates[33] corrected with the amounts lost during sorting that will be described below, except for PET bottles where the collection rate is published by the Federal Office for the Environment[34]. For both Europe and Switzerland, agricultural and construction packaging are assumed to have a 100% collection rate, as their waste streams are separate from the rest. Waste from construction applications are entirely collected separately, no stream flows to mixed waste. Automotive applications are also entirely collected, but 19%

of the European ELVs[35] and 54% of the Swiss ELVs[36] are directly exported as second hand cars. In Europe in 2012, it was estimated that 68.25%[37] of WEEE were collected separately, 7.94%[37] ended in mixed waste and the remaining 23.81[37]-37%[38] were exported. In Switzerland in 2006, 2.71% of WEEE was collected as mixed waste, 9.99% was exported as second-hand products, and the rest was collected in the separate waste stream for WEEE[39]. Agricultural applications are assumed to be entirely collected in their own waste stream, and thus have a collection rate of 100%. Household plastics, furniture, hygiene and medical textiles and other applications are assumed to be exclusively collected in mixed waste. Cosmetic microplastics are assumed to be entirely collected as waste water in WWTPs.

Of clothing textiles, 15-20% are separately collected in Europe[40] and 34% in Switzerland[41]. The same parameters are used for fabric coatings and technical clothing. We assume that the household and technical household textiles have a collection rate that is 10% of the one for clothing. Agrotextiles, mobility textiles, geotextiles, building textiles are assumed to be entirely collected in their respective separate waste streams. The remaining technical textiles are assumed to be only collected as mixed waste.

10.2.12 Waste management

The plastic in mixed waste streams in Europe is assumed to follow the proportions between the fraction disposed onto or into land and the incinerated fraction of the mixed ordinary waste stream in Eurostat for EU28 and Norway[42]. We obtain that 50.95% are landfilled and 49.05% are incinerated. In Switzerland, everything is incinerated[43].

Of all the plastic collected in packaging collection, we assume that 10% are sorted out and incinerated, except for PET where 5.56% are sorted out in Europe[32] and 7% are sorted out in Switzerland (Private communication PET Recycling Schweiz, 02.12.2016). In the case of Switzerland, 9.43% of PET is exported at this stage[44], and we assume that 10% of the other polymers are also exported.

The recycling rates of construction products can be obtained by polymer for Europe[45], and by application for Switzerland[33]. 42% of the incinerable waste from C&D in Europe is assumed to be landfilled following the proportions landfilling and incineration of C&D waste[45].

The proportions of the plastic dismantled in cars or shredded in Europe can be estimated using available data on management options for large plastic pieces and the shredder light fraction (or fluff) for many different European countries[46]. As data for some countries is missing, an

aggregate over EU28 or EU27 is not provided in the database. The data used here are an aggregate of the data available for all countries from 2010 to 2014, to find a balance between recent and comprehensive data. To compare the fraction of plastic dismantled from the vehicles and shredded, we compare the two total masses, assuming that fluff contains 41% plastic[47]. We find that 1.50% of the plastics are removed before shredding. In Switzerland, 3% of ELV plastic undergoes recycling and the rest is incinerated[33]. Knowing that no recycling of ASR plastics is undertaken[48], we can attribute the 3% of ELV plastic that is recycled to the recycling of large parts[33].

Of all the WEEP collected in Europe, 2.1-6%[49] are incinerated, nothing is landfilled, and the rest is flowing to WEEP recycling. In Switzerland, 64% of WEEP is conveyed to WEEP recycling[50] and the rest is incinerated.

Of the plastic waste generated by the agriculture sector in EU28+2, 28% was recycled in 2014, while 31.1% were incinerated and 40.9% were sent to landfills[19]. Different recycling rates can be found for agricultural plastic applications in Switzerland: 12%[33], 13.33-26.67%[51] or 20%[52]. The remaining fraction is incinerated.

Collected textiles in Europe can be reused as such immediately (10%), exported for reuse (30%), recycled as rags (50%), landfilled (3%) or incinerated (7%)[40]. In Switzerland, 0.4% are reused, 97.7% are exported, 1.9% are incinerated[44].

Waste management options for pre-consumer waste plastic are assumed to follow given trends for Germany[4]: 89.19% flows to reuse, 0.44% to landfill and 11.10% to incineration. The landfilled fraction is assumed to be incinerated in the case of Switzerland.

10.2.13 Recycling

For both Europe and Switzerland, 7.6% of the mass of plastic in recycling is assumed to be lost, except for PET where 3% are lost[53]. 46% of the plastic packaging in recycling in Europe is exported[54] (or 60% according to Furfari 2016 as cited by Brouwer[55]), and the rest is reused in Europe. In Switzerland, 16% of recycled PET is exported[44]. We assume that 15% of the other recycled polymers are exported as well.

In C&D recycling, as the plastic stream is estimated to be inhomogeneous, 10% are estimated to be lost[53]. We assume no fraction is landfilled.

The recycling of ELV large parts in Europe is expected to follow Eurostat data[46]: 72% material reuse, 9% parts reuse, 15% landfilling and 4% incineration. Regarding ASR plastics in Europe, since the most common identified materials contained in fluff are plastics[47,56,57], we assume that the waste management options described in the database can be applied to plastics in fluff as well: 17% material reuse, 56% landfilling and 26% incineration[46]. The same data for ELV large parts recycling is reused for Switzerland, assuming that the fraction landfilled in Europe is incinerated instead. As already mentioned earlier, no recycling of ASR plastic takes place in Switzerland[48].

Of the WEEPs in recycling, we assume that 51% are recycled[50], which is the regulation limit in Switzerland for a process to count as recycling of plastics in WEEE recycling.

During the recycling process of agriculture plastics, we assume that 10% of the material is lost and incinerated, as the stream is expected to be contaminated[53].

10.2.14 Incineration

Due to the high temperatures in a Waste Incineration Plant, all plastics incinerated are modelled to leave the system[58].

10.3 Production

Amounts of produced polymer in primary forms were obtained from a market report for EU28+2[9] and from the Eurostat database[59] for EU28 (Table S4). We assume that the error induced by the different geographic systems is negligible, as the plastic production industry is not very strong in Norway and inexistent in Switzerland for the polymers considered[9].

Table S4: Plastic production by polymer in Europe in 2014 from two different sources. All masses are in kt. Comments can be found in the SI.

	LDPE	HDPE	PP	PS	EPS	PVC	PET
EU28+2, 2014[9]	7071	5170	9090	1668	1371	5737	2610
EU28, 2014[59]	6237	5862	9475	1641	1341	6814	2937

10.4 Input of recycled material

10.4.1 Europe

The amount of reintegrated recycled material in Europe can be estimated knowing that 7657 kt of plastic was recycled in 2014[60] and that 46% of that amount is exported abroad[54]. We find that 3522 kt of plastic was reintroduced onto the European market in 2014. Moreover, 481 kt of PVC[61] and 1700 kt of PET[32] were recycled in 2014. Assuming again that 46% of that amount is exported abroad[54], we find that 221 kt of PVC and 782 kt of PET are reused in Europe. The remaining 2519 kt are distributed among the remaining polymers following the demand for virgin polymer in Europe in 2014[9]. The resulting inputs of recycled material are 724 kt for LDPE, 491 kt for HDPE, 762 kt for PP, 163 kt for PS and 116 kt for EPS.

10.4.2 Switzerland

The amount of reinjected recycled material in Switzerland can be estimated using the demand for plastics[9] and the amount of recycled material used in manufacturing plants in Switzerland in 2014[13]. Assuming that the demand figure only comprises virgin plastic, the virgin plastic demand was approximately 502 kt in 2014. In manufacturing plants, the used material was 90% virgin material, 5% recycled plastic and 5% internal recycling[13]. We thus estimate an input of 28 kt of recycled plastic to the Swiss manufacturers. Knowing that 26 kt of recycled PET is reused in Switzerland (since 37 kt[34] are recycled and 29%[44] are exported), it would mean that around 2 kt are left for the remaining polymers. Nevertheless, the uncertainty associated to these 2 kt is deemed to be quite high, and since this amount is negligible compared to other input flows from trade, we prefer to neglect it.

10.5 Trade

A qualitative assessment of the data availability of relevant trade flows is given below (Table S5). Trade data is available from online databases: Eurostat for Europe[62] and Swiss-Impex for Switzerland[63].

Table S5: Different trade flows and the data availability for each trade flow.

Trade level	Description of flow	Data availability
Production	Plastic in primary forms (pellets, powder, fibres)	Very high
Manufacturing	Half-finished products (films, sheets, fabrics…)	High
Consumption	Finished products consisting only of plastic (and additives)	Medium
	Finished products where plastic is one part of the product:	Low
	Packaging with a finished product	Very low
End of Life	End of life products	Medium
	Plastic scrap	Medium
	Plastic in waste	Low

10.5.1 Trade of plastic in primary forms

Plastic is traded in various forms at the production and manufacturing level: as plastic in primary forms at the level of transport or as fibres and yarns from or to fibre production. Except when mentioned in the good's description, it is assumed that no additives are involved.

10.5.1.1 Plastic in primary forms

The polymer composition of a category is mentioned in its description.

Table S6: Codes and descriptions of the traded plastic in primary forms.

Code	Description	Material
390110	Polyethylene with a specific gravity of < 0,94, in primary forms	LDPE
390120	Polyethylene with a specific gravity of >= 0,94, in primary forms	HDPE
390210	Polypropylene, in primary forms	PP
390230	Propylene copolymers, in primary forms	PP
390311	Expansible polystyrene, in primary forms	EPS
390319	Polystyrene, in primary forms (excl. expansible)	PS
390410	Poly"vinyl chloride", in primary forms, not mixed with any other substances	PVC
390421	Non-plasticised poly"vinyl chloride", in primary forms, mixed with other substances	70% PVC (Private com. A. Sevenster)
390422	Plasticised poly"vinyl chloride", in primary forms, mixed with other substances	70% PVC (Private com. A. Sevenster)
390760	Poly"ethylene terephthalate", in primary forms	PET

10.5.1.2 Fibres and yarns

The subheadings of the codes given in Table S7 are the codes considered for the fibres traded at the manufacturing level. Filament tows, sewing threads, staple fibres, yarns are considered here. The waste of fibres is not considered, as well as woven fabrics which are part of the trade at the manufacturing level. The subheadings themselves and their allocation to the different polymers are not shown due to space scarcity. The goods were attributed to the different polymers following various assumptions[9,64]:

1. If the subheading description clearly states what material it consists of, no further correction is made.
2. For subheadings where the material composition is "synthetic", the proportions between man-made fibres produced in Europe[64] is used to allocate the traded mass to a material. In these proportions, the artificial fibre content is subtracted to ensure a consistent calculation.
3. For subheadings containing more than 85% mass of another synthetic material, we assume that 10% of the mass follows the proportions of man-made fibres mentioned earlier[64].
4. For subheadings containing less than 85% mass of another synthetic material, we assume that 40% of the mass follows the proportions of man-made fibres mentioned earlier[64].
5. For subheadings containing more than 85% mass of PET or PP, we assume it contains 90% mass of this material on average. The remaining 10% is allocated following the proportions of man-made fibres mentioned earlier[64] excluding the material explicitly mentioned in the subheading.
6. For subheadings containing less than 85% mass of PET or PP, we assume it contains 60% mass of this material on average. The remaining 40% is allocated following the proportions of man-made fibres mentioned earlier[64] excluding the material explicitly mentioned in the subheading.
7. If the subheading excludes one or more types of material, similar calculations are made to allocate the mass to relevant materials, following the proportions of man-made fibres mentioned earlier[64].
8. Since HDPE is never separately mentioned, for all subheadings which are reported as synthetic, the European fibre demands of PP and HDPE[9] are used to calculate the allocation to HDPE.

Table S7: Codes and descriptions of the traded fibres and yarns.

Code	Description	Material
54	Man-made filaments; strip and the like of man-made textile materials (subheadings)	PET, PP, HDPE
55	Man-made staple fibres (subheadings)	PET, PP, HDPE

10.5.2 Trade at the manufacturing level

The subheadings of the codes given in Table S8 are the goods considered for the fabrics traded at the manufacturing level. Woven, pile, knitted and crocheted fabrics, lace and embroideries are considered. The waste of fibres is not considered, as well as filament tows, sewing threads, staple fibres, yarns which are part of the trade at the fibre production level. The subheadings themselves and their allocation to the different polymers are not shown due to space scarcity. The goods were attributed to the different polymers following the same method as for fibres any yarns (see above). Additional assumptions are:

1. If the subheading may contain other materials than man-made fibres, the share of man-made fibres out of all textiles is estimated from the CIRFS website[65], citing that 82% of the fibres produced in Europe are man-made.

Table S8: Codes and descriptions of the goods considered for the calculation of traded fabrics.

Code	Description	Material
54	Man-made filaments; strip and the like of man-made textile materials (subheadings)	PET, PP, HDPE
55	Man-made staple fibres (subheadings)	PET, PP, HDPE
58	Special woven fabrics; tufted textile fabrics; lace; tapestries; trimmings; embroidery	PET, PP, HDPE
60	Knitted or crocheted fabrics	PET, PP, HDPE

The subheadings of the codes given in Table S9 are the codes considered for the semi-finished goods traded at the manufacturing level. The subheadings themselves are not shown due to space scarcity. The goods were attributed to the different polymers following various assumptions depending on the good considered[9,11,18,21,66].

Table S9: Codes and descriptions of the goods considered for the calculation of traded semi-finished plastic products.

Code	Description	Material allocation
3917	Tubes, pipes and hoses, and fittings therefor, e.g. joints, elbows, flanges, of plastics	[21]
3918	Floor coverings of plastics, whether or not self-adhesive, in rolls or in the form of tiles; wall or ceiling coverings of plastics, in rolls with a width of >= 45 cm, consisting of a layer of plastics fixed permanently on a backing of any material other than paper, the face side of which is grained, embossed, coloured, design-printed or otherwise decorated	[21] Assuming 30% additives in PVC (Private com. A. Sevenster)
3919	Self-adhesive plates, sheets, film, foil, tape, strip and other flat shapes, of plastics, whether or not in rolls (excl. floor, wall and ceiling coverings of heading 3918)	[9]
3920	Plates, sheets, film, foil and strip, of non-cellular plastics, not reinforced, laminated, supported or similarly combined with other materials, without backing, unworked or merely surface-worked or merely cut into squares or rectangles (excl. self-adhesive, and floor, wall and ceiling coverings of heading 3918)	[9] Assuming 30% additives in PVC (Private com. A. Sevenster)
3921	Plates, sheets, film, foil and strip, of plastics, reinforced, laminated, supported or similarly combined with other materials, or of cellular plastic, unworked or merely surface-worked or merely cut into squares or rectangles (excl. self-adhesive, and floor, wall and ceiling coverings of heading 3918)	[9] Assuming 30% additives in PVC (Private com. A. Sevenster)
3922	Baths, shower-baths, sinks, washbasins, bidets, lavatory pans, seats and covers, flushing cisterns and similar sanitary ware, of plastics	[9]
3923	Articles for the conveyance or packaging of goods, of plastics; stoppers, lids, caps and other closures, of plastics	[9,11,18]
3924	Tableware, kitchenware, other household articles and toilet articles, of plastics (excl. baths, shower-baths, washbasins, bidets, lavatory pans, seats and covers, flushing cisterns and similar sanitary ware)	[66] (based on mention frequency)
3925	Builders' ware of plastics, n.e.s.	[21] Assuming 30% additives in PVC (Private com. A. Sevenster)
3926	Articles of plastics and articles of other materials of heading 3901 to 3914, n.e.s.	[66] (based on mention frequency)

10.5.3 Trade at the consumption level

Finished products are exchanged from the corresponding product sector in consumption.

10.5.3.1 Packaging

A certain mass of packaging is traded along with nearly any traded good. This mass is not reported in trade databases, which is why this flow is seldom mentioned in plastic packaging flow analyses. Nonetheless, a rough estimate of packaging trade can be found knowing that for a product packed in plastic, 1-3% of its mass corresponds to the mass of primary packaging necessary[67] and that 19% of packaging used consists of plastic[68]. This contains the implicit

assumption that for products with non-plastic packaging, the same mass of packaging is used. We further assume that the same additional amount per product is used as secondary and tertiary packaging. Considering these different assumptions, 0.38-1.14% of the mass of traded packaged goods corresponds to plastic packaging. Trade categories containing finished consumer products and other relevant products were included in the calculation. Examples of products that were not included are live animals and plants and products from the mining industry. A detailed list of the included codes is given in Table S10. The validity of these assumptions was tested by applying them to the UK in 2011, where a study estimated the amount of packaging that was imported with finished goods as being 506 kt[17]. With a predicted 152-456 kt of imported plastic packaging for the UK in the same year from our method, we can conclude that this way of calculating predicts an appropriate order of magnitude but still underestimates the amount of plastic packaging traded. The same approach predicts 66-197 kt of imports for the EU28 in 2014, and 31-94 kt for Switzerland. The surprising difference between the net imports of packaging in Europe and in the UK is due to the products considered. If all trade codes are considered, the EU has a net import of around 10^6 kt in 2014, while the UK has around 10^5 kt net imports for 2011. For the trade codes considered in this calculation, the tendency is reversed, and the net imports are larger for the UK in 2011 with 45'000 kt, whereas 18'000 kt of goods were imported in the EU28 in 2014. These quantities are distributed between polymers using knowledge on the polymer demand for packaging applications in Europe[11].

Table S10: Codes and descriptions of the goods considered for the calculation of traded filled packaging.

Code	Description
02	Meat and edible meat offal
03	Fish and crustaceans, molluscs and other aquatic invertebrates
04	Dairy produce; birds' eggs; natural honey; edible products of animal origin, not elsewhere specified or included
05	Products of animal origin, not elsewhere specified or included
07	Edible vegetables and certain roots and tubers
08	Edible fruit and nuts; peel of citrus fruit or melons
09	Coffee, tea, maté and spices
10	Cereals
11	Products of the milling industry; malt; starches; inulin; wheat gluten
12	Oil seeds and oleaginous fruits; miscellaneous grains, seeds and fruit; industrial or medicinal plants; straw and fodder
15	Animal or vegetable fats and oils and their cleavage products; prepared edible fats; animal or vegetable waxes
16	Preparations of meat, of fish or of crustaceans, molluscs or other aquatic invertebrates

Code	Description
17	Sugars and sugar confectionery
18	Cocoa and cocoa preparations
19	Preparations of cereals, flour, starch or milk; pastrycooks' products
20	Preparations of vegetables, fruit, nuts or other parts of plants
21	Miscellaneous edible preparations
22	Beverages, spirits and vinegar
23	Residues and waste from the food industries; prepared animal fodder
24	Tobacco and manufactured tobacco substitutes
28	Inorganic chemicals; organic or inorganic compounds of precious metals, of rare-earth metals, of radioactive elements or of isotopes
29	Organic chemicals
30	Pharmaceutical products
31	Fertilisers
32	Tanning or dyeing extracts; tannins and their derivatives; dyes, pigments and other colouring matter; paints and varnishes; putty and other mastics; inks
33	Essential oils and resinoids; perfumery, cosmetic or toilet preparations
34	Soap, organic surface-active agents, washing preparations, lubricating preparations, artificial waxes, prepared waxes, polishing or scouring preparations, candles and similar articles, moddeling paste
35	Albuminoidal substances; modified starches; glues; enzymes
37	Photographic or cinematographic goods
38	Miscellaneous chemical products
39	Plastics and articles thereof
40	Rubber and articles thereof
4417	Tools, tool bodies, tool handles, broom or brush bodies and handles, of wood; boot or shoe lasts and shoetrees, of wood (excl. forms used in the manufacture of hats, forms of heading 8480, other machines and machine components, of wood)
4418	Builders' joinery and carpentry, of wood, incl. cellular wood panels, assembled flooring panels, shingles and shakes, of wood (excl. plywood panelling, blocks, strips and friezes for parquet flooring, not assembled, and pre
4419	Tableware and kitchenware, of wood (excl. interior fittings, ornaments, cooperage products, tableware and kitchenware components of wood, brushes, brooms and hand sieves)
48	Paper and paperboard; articles of paper pulp, of paper or of paperboard
49	Printed books, newspapers, pictures and other products of the printing industry; manuscripts, typescripts and plans
57	Carpets and other textile floor coverings
60	Knitted or crocheted fabrics
61	Articles of apparel and clothing accessories, knitted or crocheted
62	Articles of apparel and clothing accessories, not knitted or crocheted
63	Other made-up textile articles; sets; worn clothing and worn textile articles; rags
64	Footwear, gaiters and the like; parts of such articles
65	Headgear and parts thereof
66	Umbrellas, sun umbrellas, walking sticks, seat-sticks, whips, riding crops and parts thereof
67	Prepared feathers and down and articles made of feathers or of down; artificial flowers; articles of human hair
69	Ceramic products
70	Glass and glassware

Code	Description
82	Tools, implements, cutlery, spoons and forks, of base metal; parts thereof of base metal
85	Electrical machinery and equipment and parts thereof; sound recorders and reproducers, television image and sound recorders and reproducers, and parts and accessories of such articles
91	Clocks and watches and parts thereof
94	Furniture; bedding, mattresses, mattress supports, cushions and similar stuffed furnishings; lamps and lighting fittings, not elsewhere specified or included; illuminated signs, illuminated nameplates
95	Toys, games and sports requisites; parts and accessories thereof

10.5.3.2 Automotive

In order to estimate the magnitude of trade flows in the automotive sector, the traded vehicle amounts were compared to the produced vehicle amounts. In 2014, 17.2 million vehicles were manufactured in the EU28[69], while 2.6 million were imported and 6.0 million exported[69]. For a plastic demand in the automotive sector of 4110.8 kt in 2014[22], and assuming that the same proportion of plastic is included in manufactured, imported and exported vehicles, we obtain a plastic import of 628.5 kt and export of 1446.3 kt. The repartition of this plastic mass to different polymers is done following the European automotive plastic demand among polymers[11]. In Switzerland in 2014, 642 kt of vehicles were imported and 251 kt exported[63]. The exported vehicles are assumed to be still operational end-of-life vehicles and as a consequence, not included in the calculation here. Assuming 10% of a vehicle's mass is plastic[70] and that the repartition of this plastic mass to different polymers follows the European automotive polymer demand[11], we can calculate the mass of plastic imported and exported in Switzerland. Similar fractions of additives are used for automotive plastics as for EEE.

10.5.3.3 EEE

The trade of EEE can be described with data from Eurostat[62] and the Swiss-Impex database[63]. The chosen codes are given in Table S11. We assume that plastics make up 23% of the mass of electrical and electronic goods[71]. This plastic mass is then shared between the polymers following the manufacturers' demand by polymer in that sector[11]. These masses of polymer may still contain a fraction of additives. Little information is available regarding the average quantities of additives contained in products. Nevertheless, in order to remove this contribution, we make the following assumptions: LDPE, HDPE, PP and PET contain 25 % additives

(based on the knowledge that polyolefins may contain 20-30% halogen-free flame retardants[72]), PS contains 6% flame retardants in EEE applications[72] (based on the knowledge that HIPS may contain 5-7% flame retardants[72]), PVC contains 30% plasticizers and other additives for both flexible and rigid applications (Private communication with A. Sevenster).

Table S11: Codes and descriptions of the goods considered for the calculation of traded EEE plastic.

Code	Description
85	Electrical machinery and equipment and parts thereof; sound recorders and reproducers, television image and sound recorders and reproducers, and parts and accessories of such articles
9405	Lamps and lighting fittings, incl. searchlights and spotlights, and parts thereof, n.e.s; illuminated signs, illuminated nameplates and the like having a permanently fixed light source, and parts thereof, n.e.s.
9504	Video game consoles and machines, articles for funfair, table or parlour games, including pintables, billiards, special tables for casino games and automatic bowling alley equipment

10.5.3.4 Clothing

Three different codes in the trade databases are attributed to clothing and footwear (Table S12). The PET and PP shares in clothing are readily available[40]. The share of HDPE is estimated by comparing the European PP and HDPE demands for fibre production[9]: 169.6 kt for PP and 6.29 kt for HDPE. The average composition of footwear was taken from a British study combined with the textile data previously mentioned[40,73].

Table S12: Codes and descriptions of the goods considered for the calculation of traded clothing.

Code	Description
61	Articles of apparel and clothing accessories, knitted or crocheted
62	Articles of apparel and clothing accessories, not knitted or crocheted
64	Footwear, gaiters and the like; parts of such articles

10.5.3.5 Household textiles

6 different codes are attributed to household textiles (Table S13). The masses of PET and PP were calculated using the average composition of household textiles[40]. The mass of HDPE is again estimated by comparing the European PP and HDPE demands for fibre production[9].

Table S13: Codes and descriptions of the goods considered for the calculation of traded household textiles.

Code	Description
57	Carpets and other textile floor coverings
6301	Blankets and travelling rugs of all types of textile materials (excl. table covers, bedspreads and articles of bedding and similar furnishing of heading 9404)
6302	Bedlinen, table linen, toilet linen and kitchen linen of all types of textile materials (excl. floorcloths, polishing cloths, dishcloths and dusters)
6303	Curtains, incl. drapes, and interior blinds; curtain or bed valances of all types of textile materials (excl. awnings and sunblinds)
6304	Articles for interior furnishing, of all types of textile materials (excl. blankets and travelling rugs, bed-linen, table linen, toilet linen, kitchen linen, curtains, incl. drapes, interior blinds, curtain or bed valances, lampshades and articles of heading 9404)

10.5.3.6 Technical textiles

Several HS codes can be attributed to technical textiles, and their trade can be estimated assuming a share of 25% PET (Global value for 2010), 23% PP (Global value for 2000) and 2% HDPE (Difference of a global value for 2010 (PP+HDPE) and 2000 (PP))[28,74]. Coated applications were assumed to have 50% coating material.

Table S14: Codes and descriptions of the goods considered for the calculation of traded technical textiles.

Code	Description
56	Wadding, felt and nonwovens; special yarns; twine, cordage, ropes and cables and articles thereof
59	Impregnated, coated, covered or laminated textile fabrics; textile articles of a kind suitable for industrial use
6305	Sacks and bags, of a kind used for the packing of goods, of all types of textile materials
6306	Tarpaulins, awnings and sunblinds; tents; sails for boats, sailboards or landcraft; camping goods of all types of textile materials (excl. flat protective coverings of light woven fabrics; umbrella and play tents; rucksacks, napsacks and similar containers; sleeping bags, mattresses and pillows, incl. their fillings)

10.6 Summary of data sources

Table S15: List of references used for Europe in 2014.

Use	Short reference	Full reference
Production and manufacturing waste	*Burkhardt, 2011*	5
	Consultic, 2016	4
Share of raw material going for textile applications	*Aizenshtein, 2015*	8
	AMI, 2015	9

Use	Short reference	Full reference
	CIRFS (Web)	7
Non-textile product sectors and categories	*AMI, 2015*	9
	APME, 1995	21
	Briassoulis, 2013	25
	BUND, 2016	31
	Consultic, 2013.	26
	Delgado, 2007	18
	Gouin et al., 2015	30
	Hussain, 2005	23
	PlasticsEurope PEMRG, 2015	11
	PlasticsEurope, 2015	22
	Scarascia-Mugnozza, 2012	24
	Waste Watch, 2003	10
	WRAP, 2013	17
Textile product sectors and categories	*EATP (Web)*	16
	Grebe, 2015	28
	Transparency market research, 2015	75
	Wazir Management Consultants, 2014	15
Waste collection	*Beton, 2014*	40
	Delgado, 2007	18
	Huisman, 2015	37
	Magalini, 2015	38
	Oeko-Institut, 2016	35
	Petcore Europe	32
Waste treatment	*Astrup, 2003*	53
	Beton, 2014	40
	Consultic, 2013	26
	Consultic, 2016	4
	Eurostat (env_waselv)	46
	Eurostat (env_wastrt)	42
	EPRO, 2016	19
	Mirabile, 2002	47
	Petcore Europe	32
	PlasticsEurope, 2016	45
	Seyring, 2015	49
	Swico, 2017	50
	Velis, 2014	54
Primary material	*AMI, 2015*	9
	Eurostat PRODCOM	59
	Sevenster	Private com.
Secondary material	*AMI, 2015*	18
	Petcore Europe (Web)	32
	PlasticsEurope PEMRG, 2015	11
	Velis, 2014	54
	Vinyl 2010, 2015	61
Trade of non-manufactured goods	*AMI, 2015*	9
	Eurostat (EU Trade since 1988 by HS2,4,6 and CN8)	62
	Sevenster	Private com.
	Van Houte, 2012	64
Trade of semi-finished goods	*AMI, 2015*	9
	APME, 1995	21
	CIRFS (Web)	65

Use	Short reference	Full reference
	Delgado, 2007	18
	Eurostat (EU Trade since 1988 by HS2,4,6 and CN8)	62
	McComb (Web)	66
	PlasticsEurope PEMRG, 2015	11
	Sevenster	Private com.
Trade of finished goods	*ACEA, 2015*	69
	AMI, 2015	9
	Andrady, 2009	67
	Bartlett, 2013	73
	Beton, 2014	40
	Buekens, 2014	71
	David Rigby Associates, 2003	74
	Eurostat (env_waspac)	68
	Eurostat (EU Trade since 1988 by HS2,4,6 and CN8)	62
	Grebe, 2015	28
	Groß, 2008	72
	Mashek, 2016	70
	PlasticsEurope PEMRG, 2015	11
	PlasticsEurope, 2015	22
	Sevenster	Private com.
	WRAP, 2013	17

Table S16: List of references used for Switzerland in 2014.

Use	Short reference	Full reference
Production and manufacturing waste	*Burkhardt, 2011*	5
	Consultic, 2016	4
Share of raw material going for textile applications	*Aizenshtein, 2015*	8
	AMI, 2015	9
	CIRFS (Web)	7
Non-textile product sectors and categories	*AMI, 2015*	9
	APME, 1995	21
	Briassoulis, 2013	25
	BUND, 2016	31
	Consultic, 2013	26
	Delgado, 2007	18
	Gouin et al., 2015	30
	Hussain, 2005	23
	Meyer, 2015	13
	PlasticsEurope PEMRG, 2015	11
	Scarascia-Mugnozza, 2012	24
	Waste Watch, 2003	10
	WRAP, 2013	17
Textile product sectors and categories	*EATP (Web)*	16
	Grebe, 2015	28
	Transparency market research, 2015	75
	Wazir Management Consultants, 2014	15
Waste collection	*BAFU, 2015*	34
	Dettli, 2015	41
	Müller, 2010	39
	SARS, 2015	36
	Schelker, 2011	33
Waste treatment	*Astrup, 2003*	53
	Consultic, 2013	26

Use	Short reference	Full reference
	Haupt, 2016	44
	Müller, 2016	51
	PET Recycling Schweiz	Private com.
	Restrepo, 2017	48
	Schelker, 2011	33
	Swico, 2017	50
	Wälti, 2016	43
Primary material	*AMI, 2015*	9
Secondary material	*AMI, 2015*	9
	BAFU, 2015	34
	Haupt, 2016	44
	Meyer, 2015	13
Trade of non-manufactured goods	*CIRFS (Web)*	65
	Sevenster	Private com.
	Swiss-Impex	63
	Van Houte, 2012	64
Trade of semi-finished goods	*AMI, 2015*	9
	APME, 1995	21
	CIRFS (Web)	65
	Delgado, 2007	18
	McComb (Web)	66
	PlasticsEurope PEMRG, 2015	11
	Sevenster	Private com.
	Swiss-Impex	63
Trade of finished goods	*Andrady, 2009*	67
	AMI, 2015	9
	Bartlett, 2013	73
	Beton, 2014	40
	Buekens, 2014	71
	David Rigby Associates, 2003	74
	Eurostat (env_waspac)	68
	Grebe, 2015	28
	Groß, 2008	72
	Mashek, 2016	70
	PlasticsEurope PEMRG, 2015	11
	Sevenster	Private com.
	Swiss-Impex	63
	WRAP, 2013	17

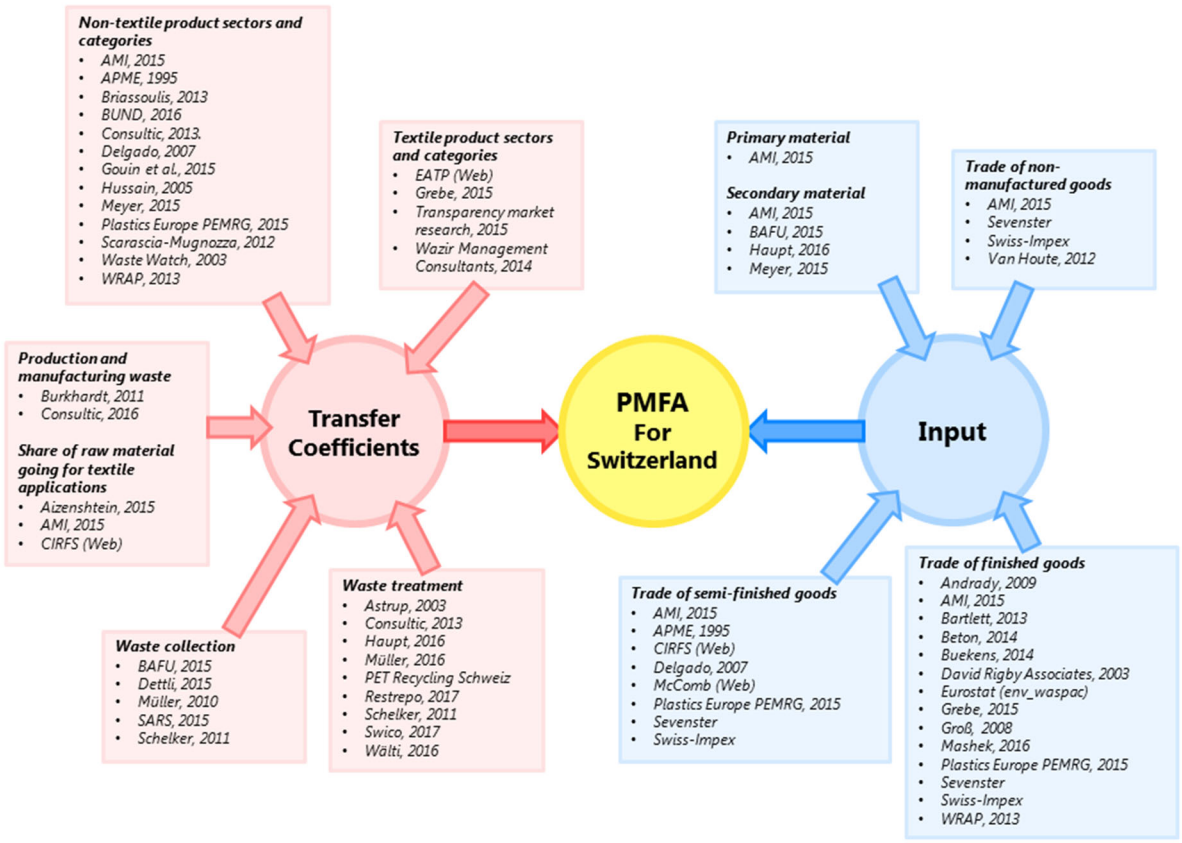

Figure S26: Summary of the data sources used for a quantification of the model parameters. A key to the references is available in Table S16.

10.7 Additional figures

10.7.1 Simplified flow diagrams for Switzerland

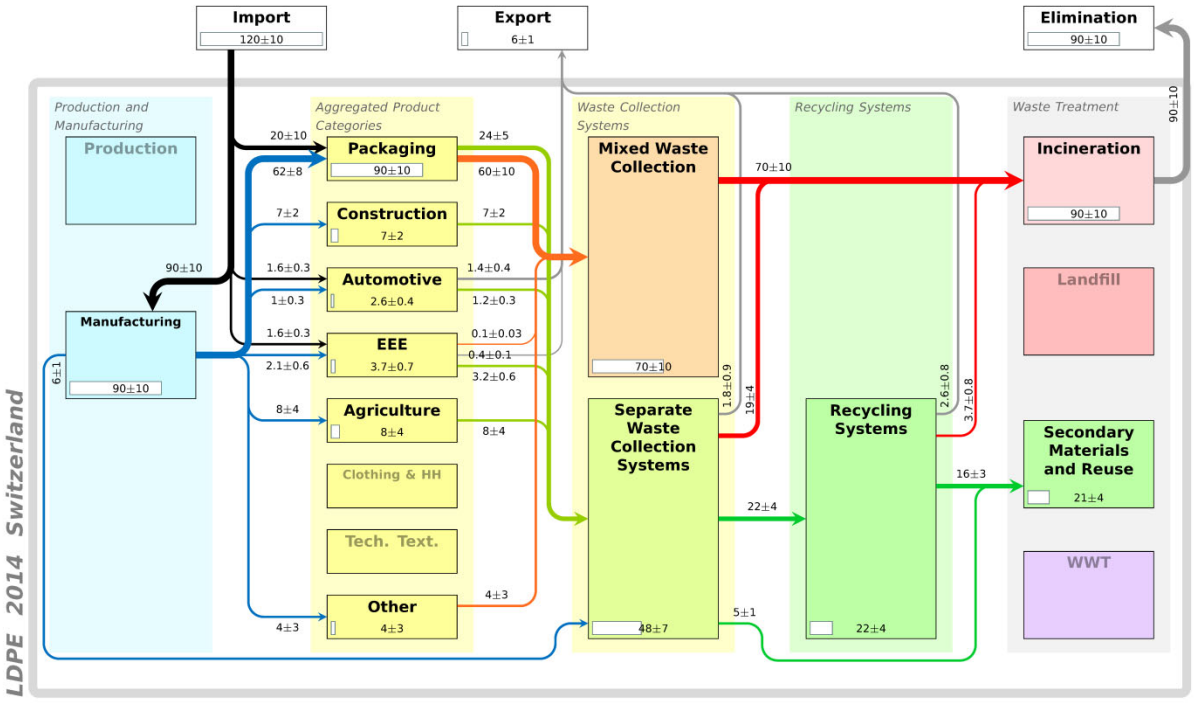

Figure S27: Simplified flow diagrams for LDPE in Switzerland in 2014. All units are in thousand metric tonnes (kt). The masses reported for the flows and compartments are rounded to the precision of the given standard deviation. The sums of flows might therefore not coincide with the reported masses. The width of the flow arrows is larger for larger masses, and the compartment bars are longer for larger masses. Colours were used to help visualizing the flow diagrams. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

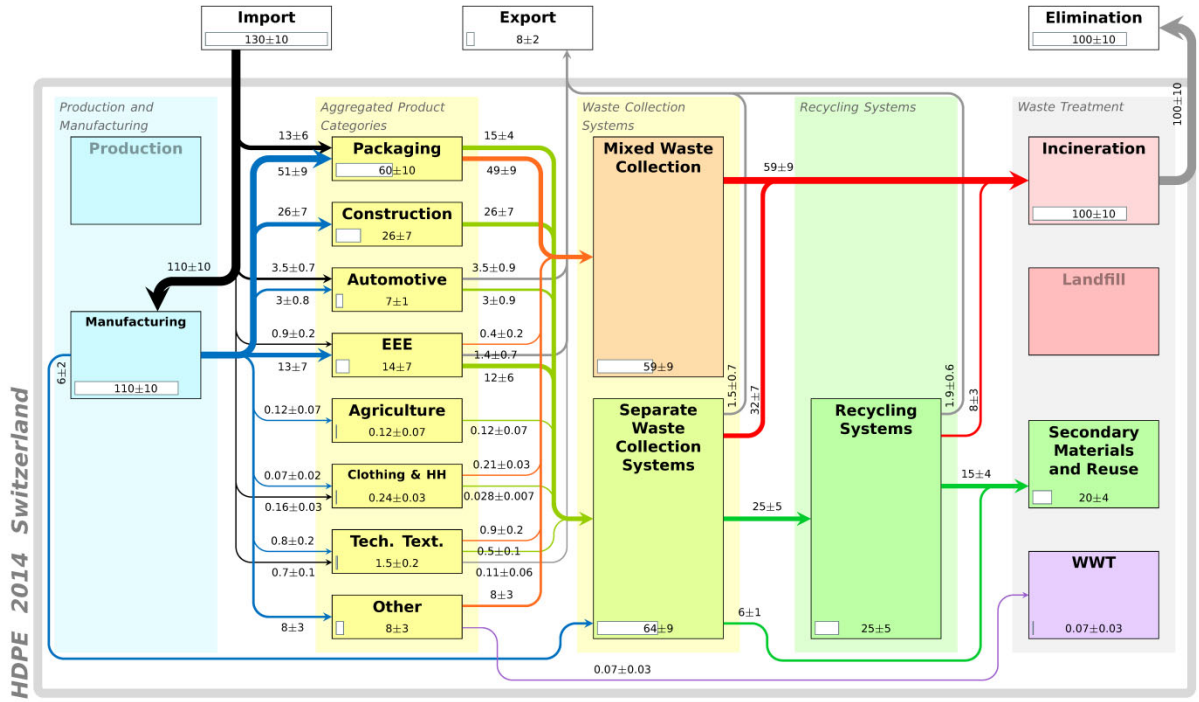

Figure S28: Simplified flow diagrams for HDPE in Switzerland in 2014. All units are in thousand metric tonnes (kt). The masses reported for the flows and compartments are rounded to the precision of the given standard deviation. The sums of flows might therefore not coincide with the reported masses. The width of the flow arrows is larger for larger masses, and the compartment bars are longer for larger masses. Colours were used to help visualizing the flow diagrams. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

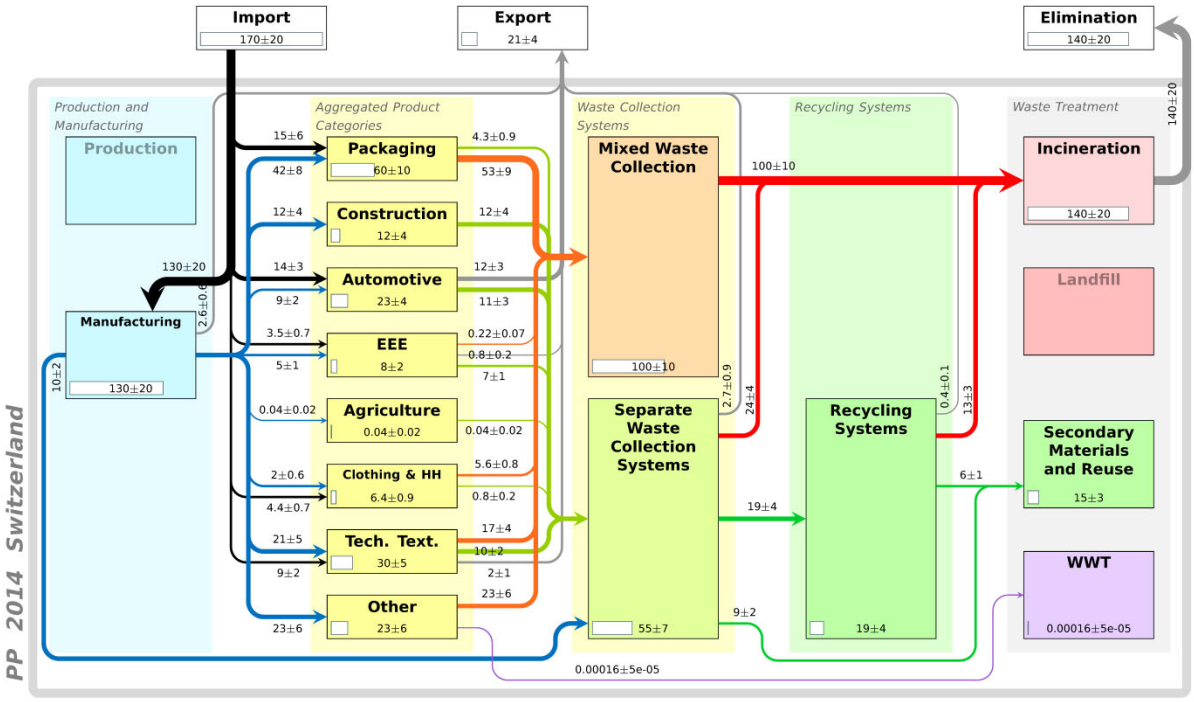

Figure S29: Simplified flow diagrams for PP in Switzerland in 2014. All units are in thousand metric tonnes (kt). The masses reported for the flows and compartments are rounded to the precision of the given standard deviation. The sums of flows might therefore not coincide with the reported masses. The width of the flow arrows is larger for larger masses, and the compartment bars are longer for larger masses. Colours were used to help visualizing the flow diagrams. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

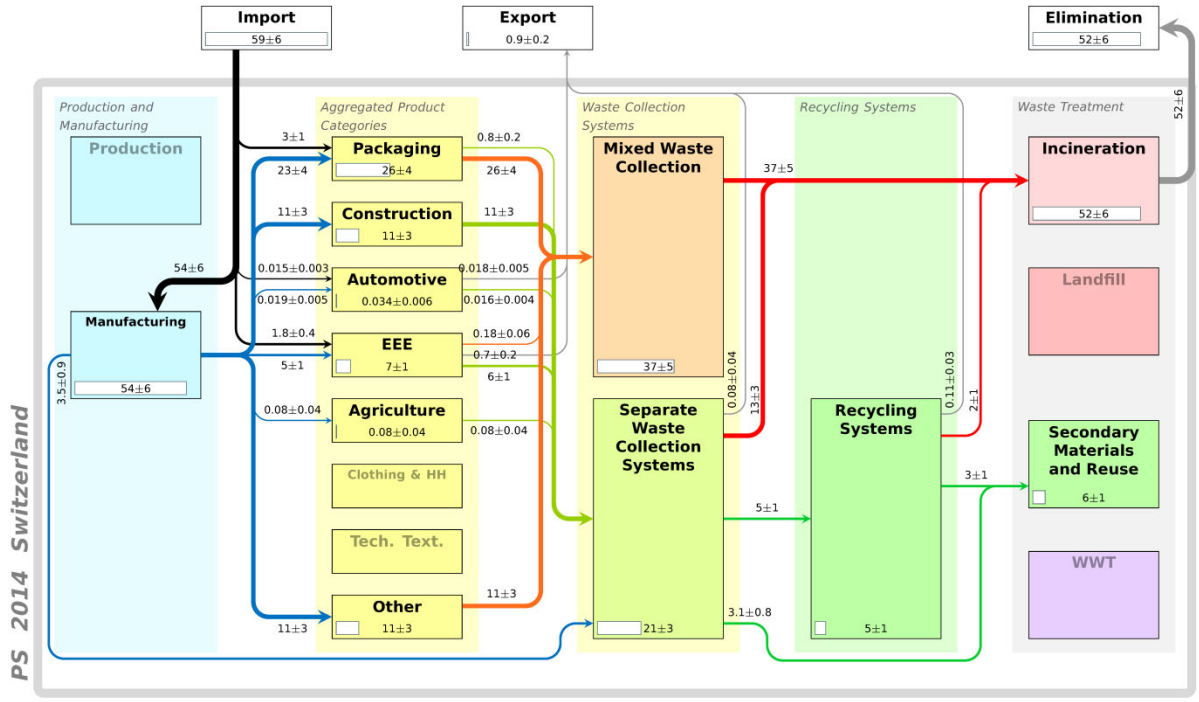

Figure S30: Simplified flow diagrams for PS in Switzerland in 2014. All units are in thousand metric tonnes (kt). The masses reported for the flows and compartments are rounded to the precision of the given standard deviation. The sums of flows might therefore not coincide with the reported masses. The width of the flow arrows is larger for larger masses, and the compartment bars are longer for larger masses. Colours were used to help visualizing the flow diagrams. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

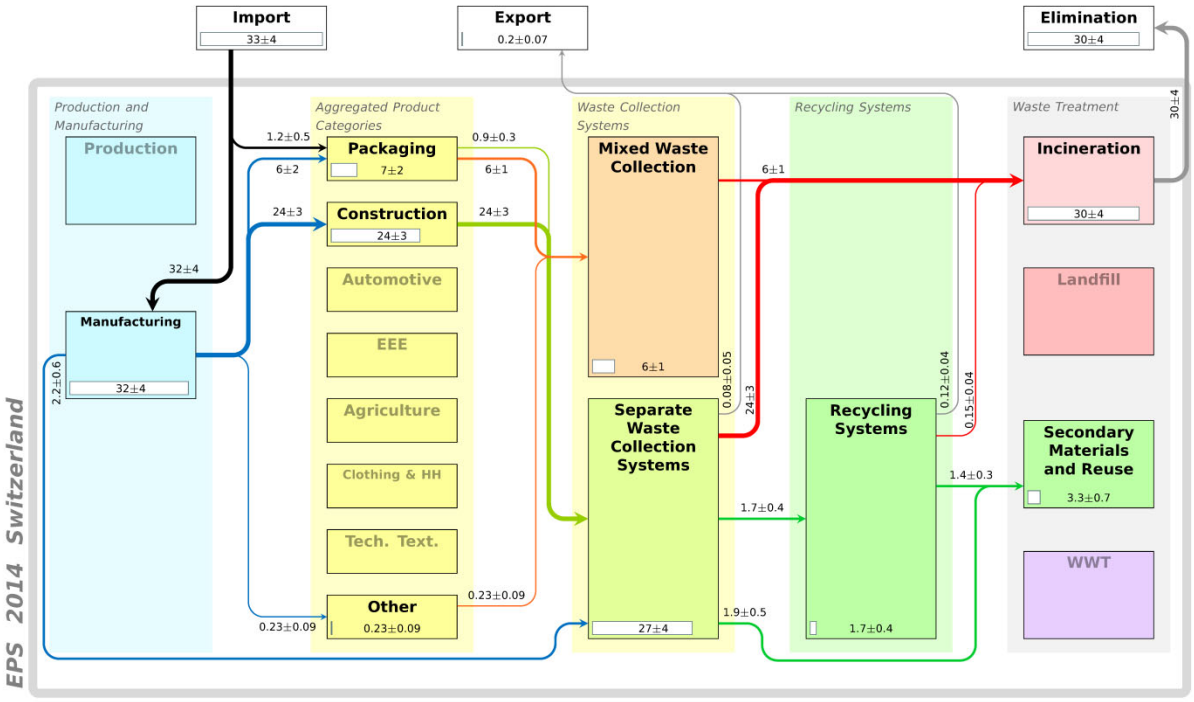

Figure S31: Simplified flow diagrams for EPS in Switzerland in 2014. All units are in thousand metric tonnes (kt). The masses reported for the flows and compartments are rounded to the precision of the given standard deviation. The sums of flows might therefore not coincide with the reported masses. The width of the flow arrows is larger for larger masses, and the compartment bars are longer for larger masses. Colours were used to help visualizing the flow diagrams. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

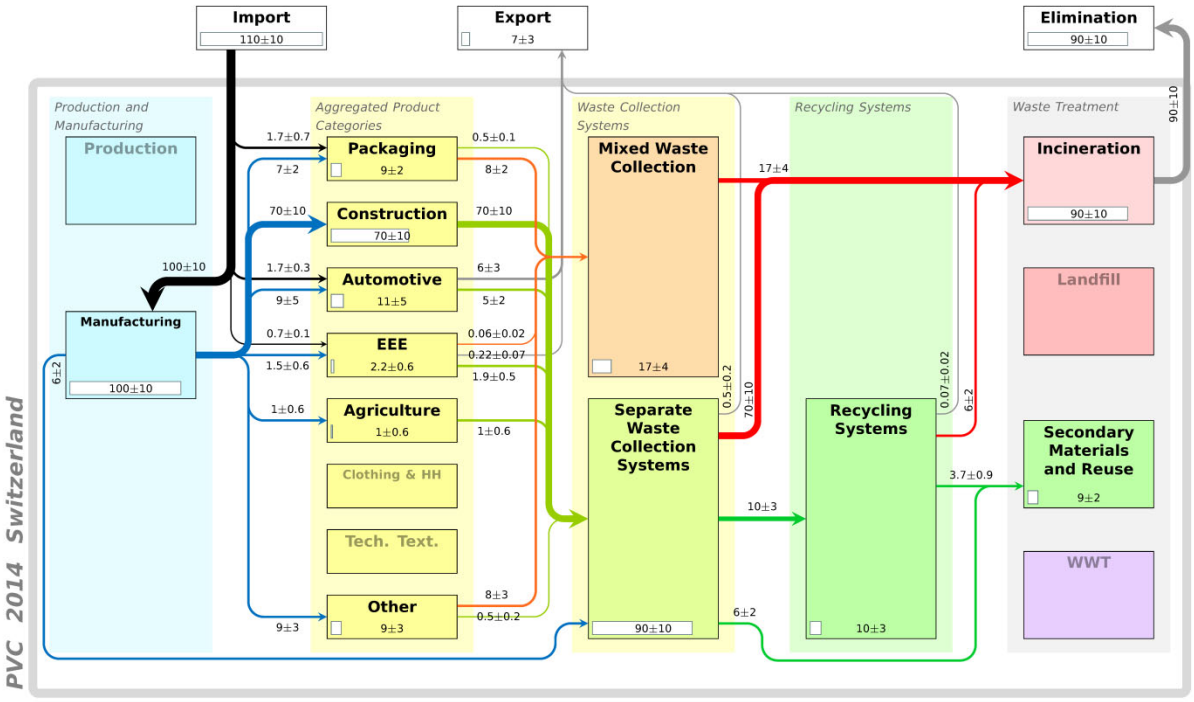

Figure S32: Simplified flow diagrams for PVC in Switzerland in 2014. All units are in thousand metric tonnes (kt). The masses reported for the flows and compartments are rounded to the precision of the given standard deviation. The sums of flows might therefore not coincide with the reported masses. The width of the flow arrows is larger for larger masses, and the compartment bars are longer for larger masses. Colours were used to help visualizing the flow diagrams. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

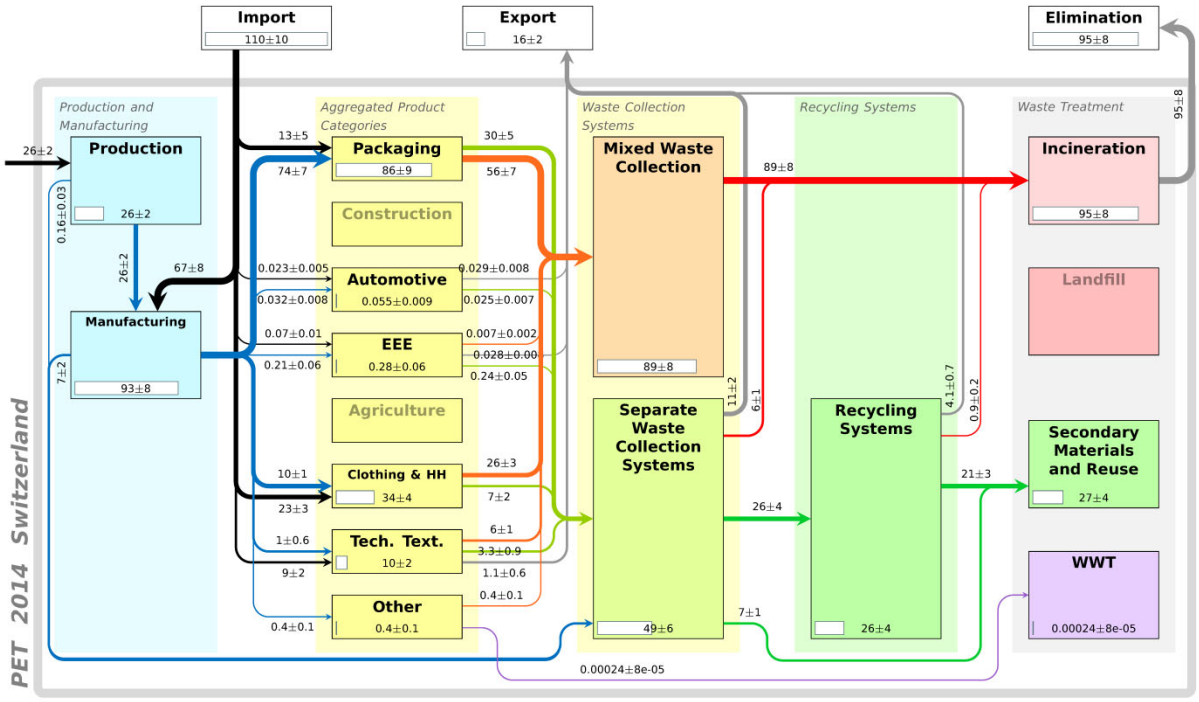

Figure S33: Simplified flow diagrams for PET in Switzerland in 2014. All units are in thousand metric tonnes (kt). The masses reported for the flows and compartments are rounded to the precision of the given standard deviation. The sums of flows might therefore not coincide with the reported masses. The width of the flow arrows is larger for larger masses, and the compartment bars are longer for larger masses. Colours were used to help visualizing the flow diagrams. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

10.7.2 Bar plots for Europe

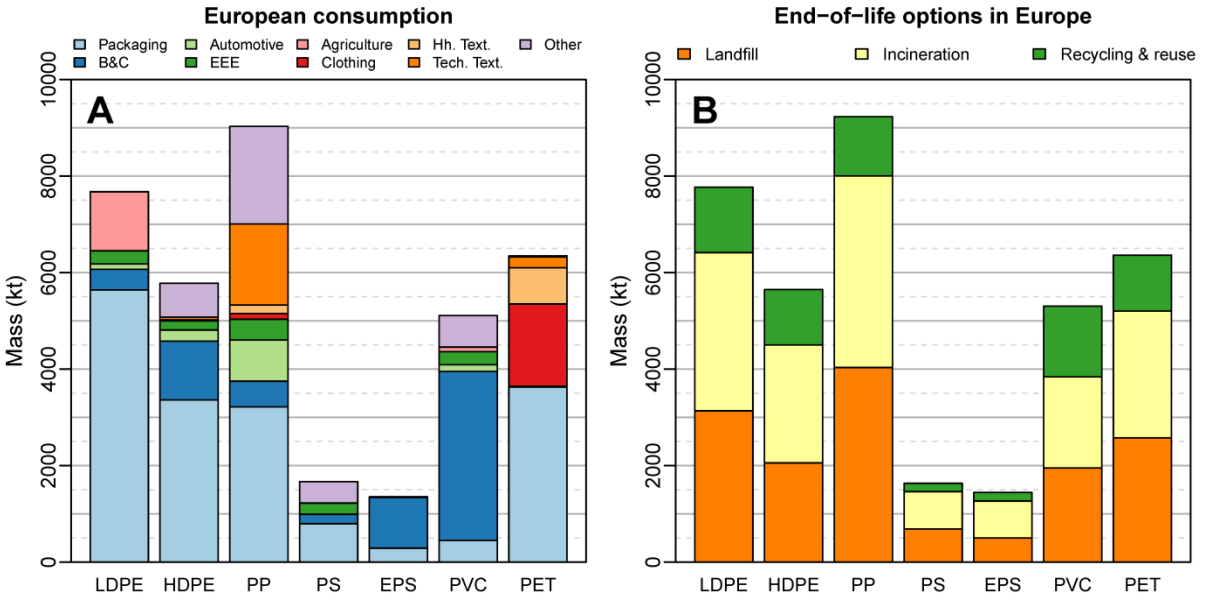

Figure S34: (A) Bar plots of the product sectors at the consumption stage by polymer. (B) Waste management options by polymer. The total height of the bars between the two plots do not coincide because of the influence of export flows from consumption, waste collection and recycling systems, as well as of the pre-consumer waste which does not flow through consumption (see Figure 4 for the magnitude of these flows).

10.7.3 Bar plots for Switzerland

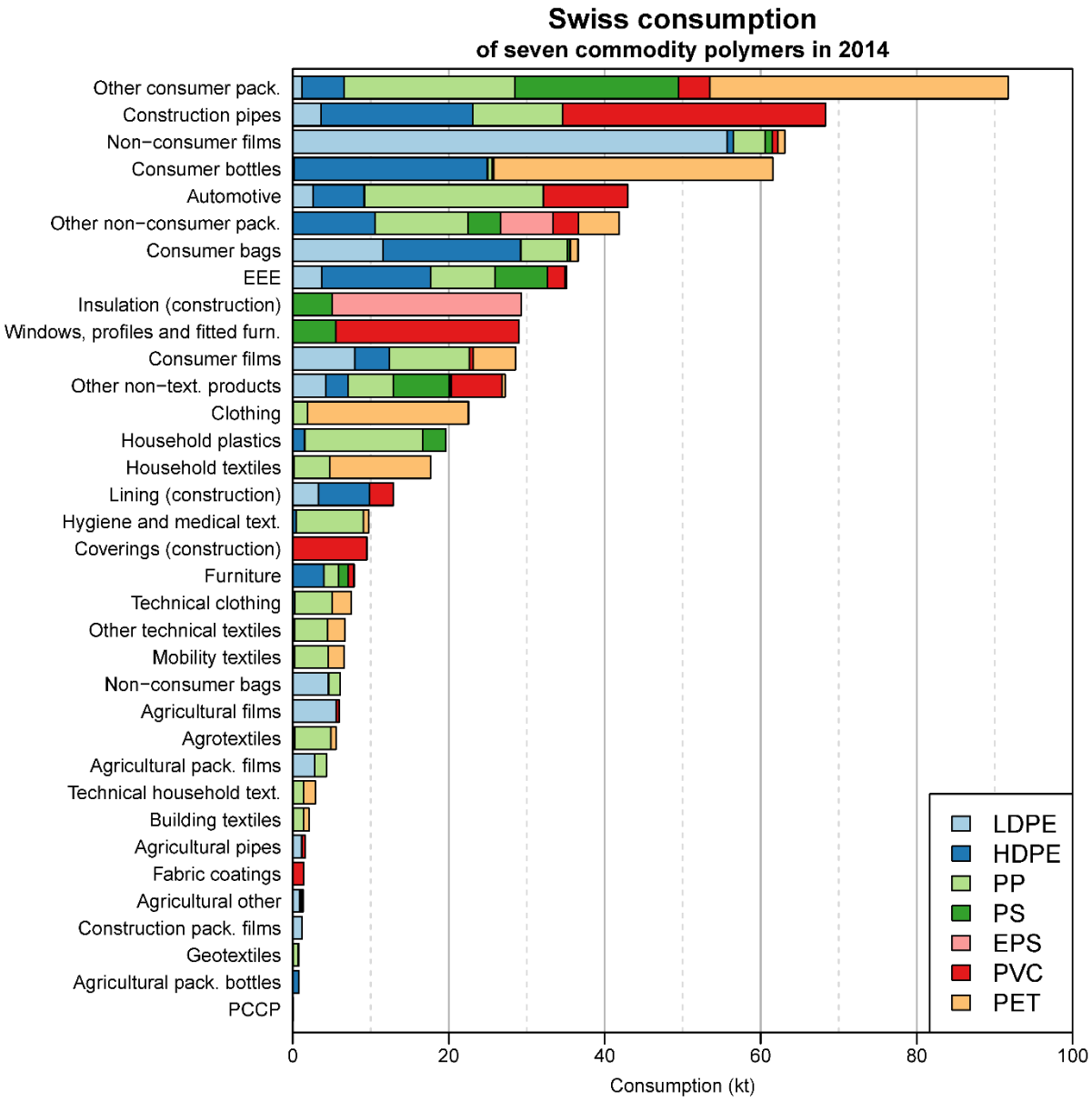

Figure S35: Overview of the Swiss polymer consumption in 2014 for the seven polymers considered. Abbreviations: pack. (packaging), furn. (furniture), EEE (Electrical and Electronic Equipment), text. (textiles), PCCP (Personal Care and Cosmetic Products).

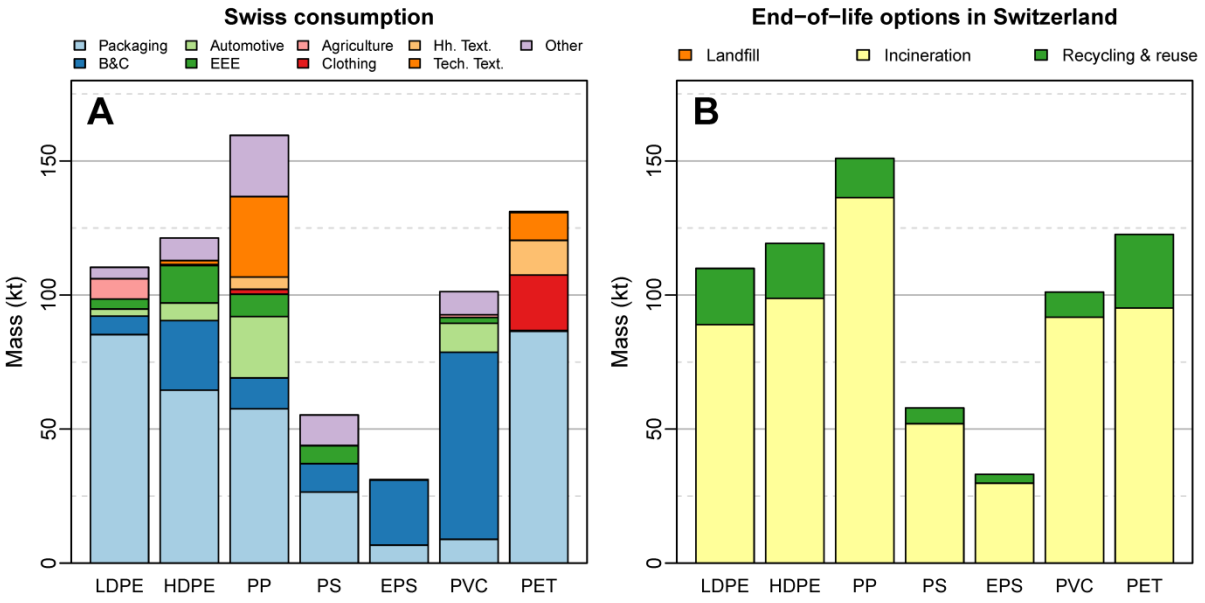

Figure S36: (A): Bar plots of the product sectors at the consumption stage by polymer. (B) Waste management options by polymer. The total height of the bars between the two plots do not coincide because of the influence of export flows from consumption, waste collection and recycling systems, as well as of the pre-consumer waste which does not flow through consumption (see Figure S27- Figure S33 for the magnitude of these flows).

10.7.4 Relative uncertainty

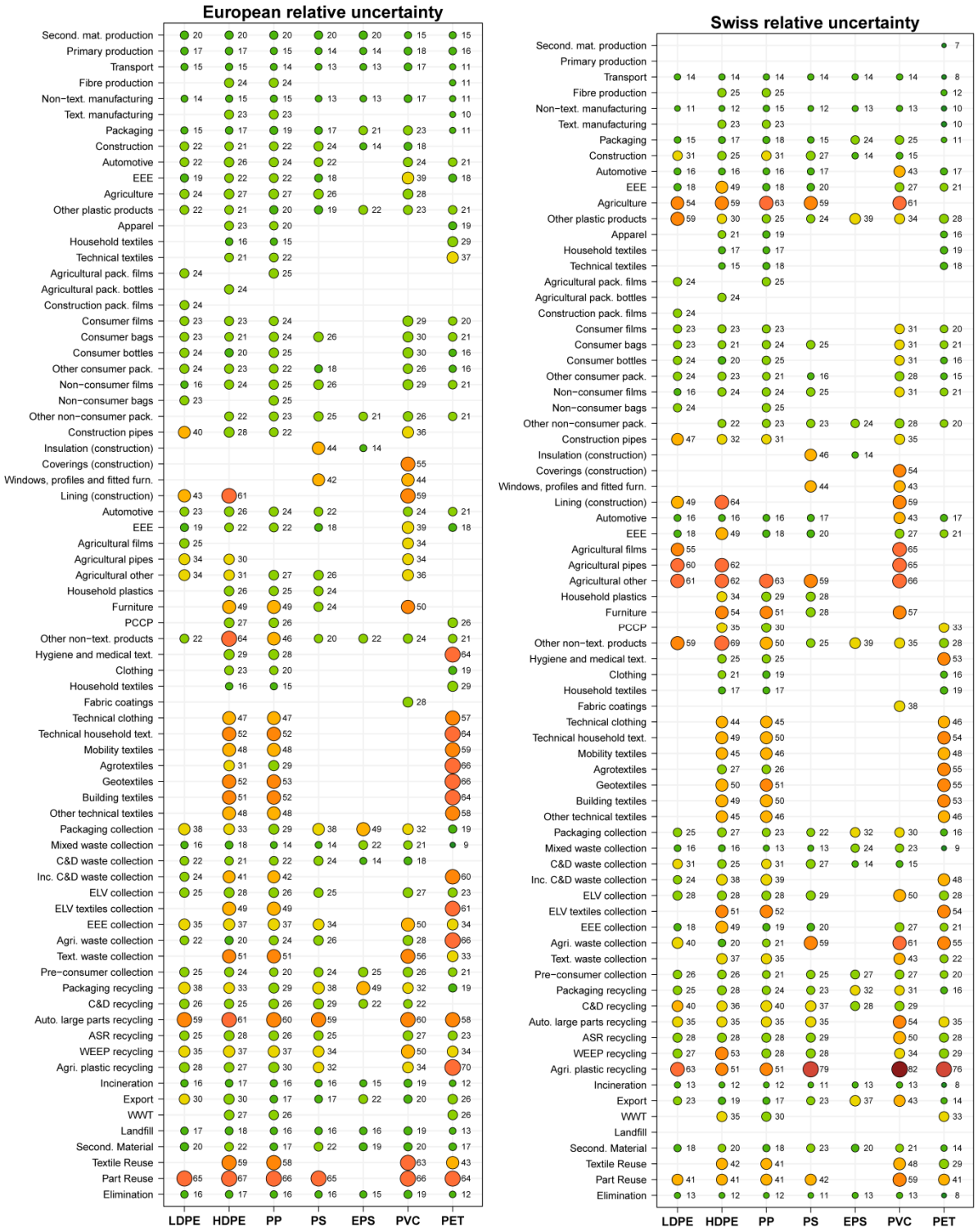

Figure S37: Bubble chart of the relative uncertainties associated to each compartment's mass. A lower uncertainty is shown in green, while a larger uncertainty is shown in red. The relative uncertainty is reported in percent next to each bubble. Missing bubbles correspond to compartments with zero mass.

10.7.5 Complete flow diagrams for Europe

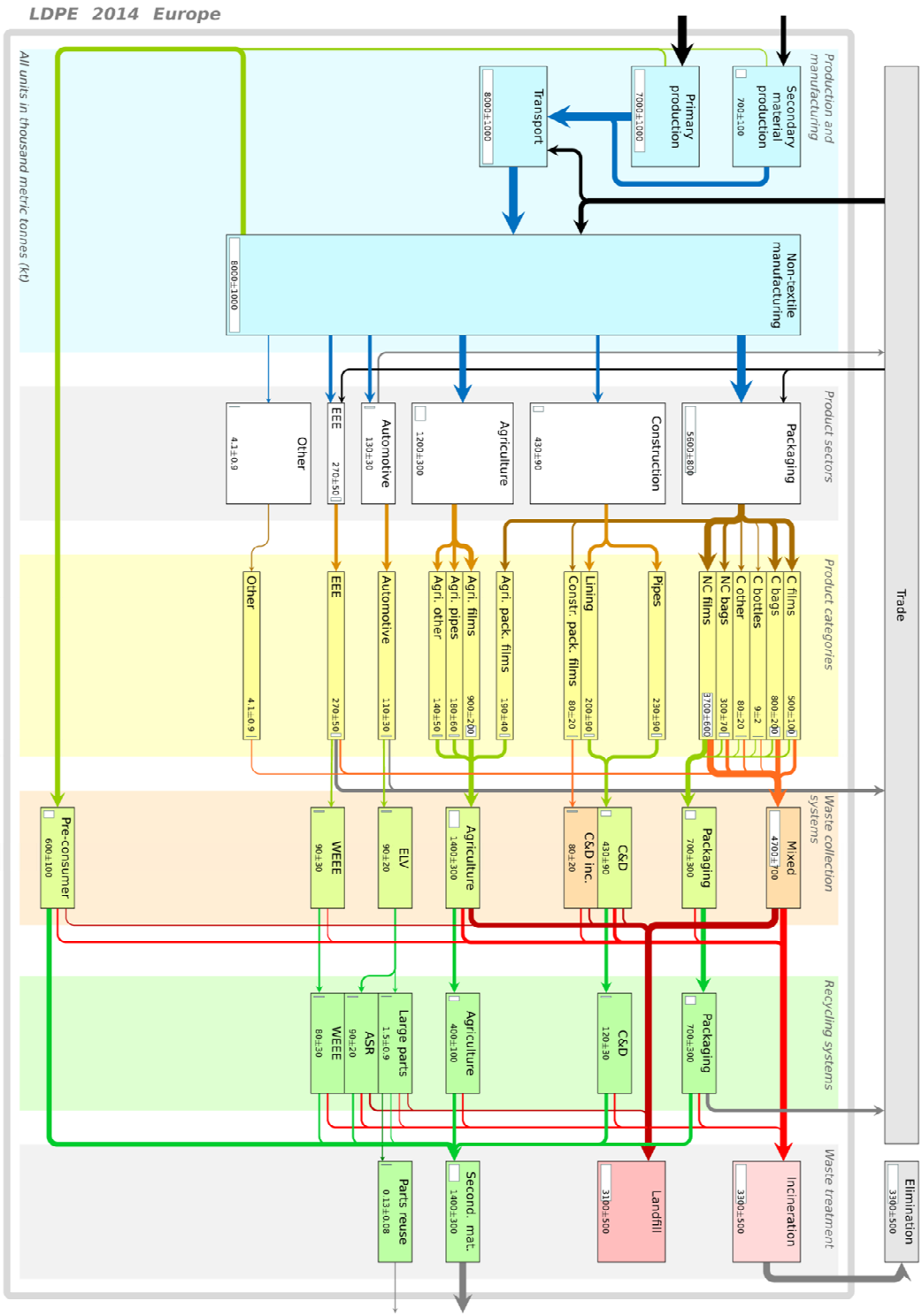

Figure S38: Flow diagram for LDPE in Europe. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

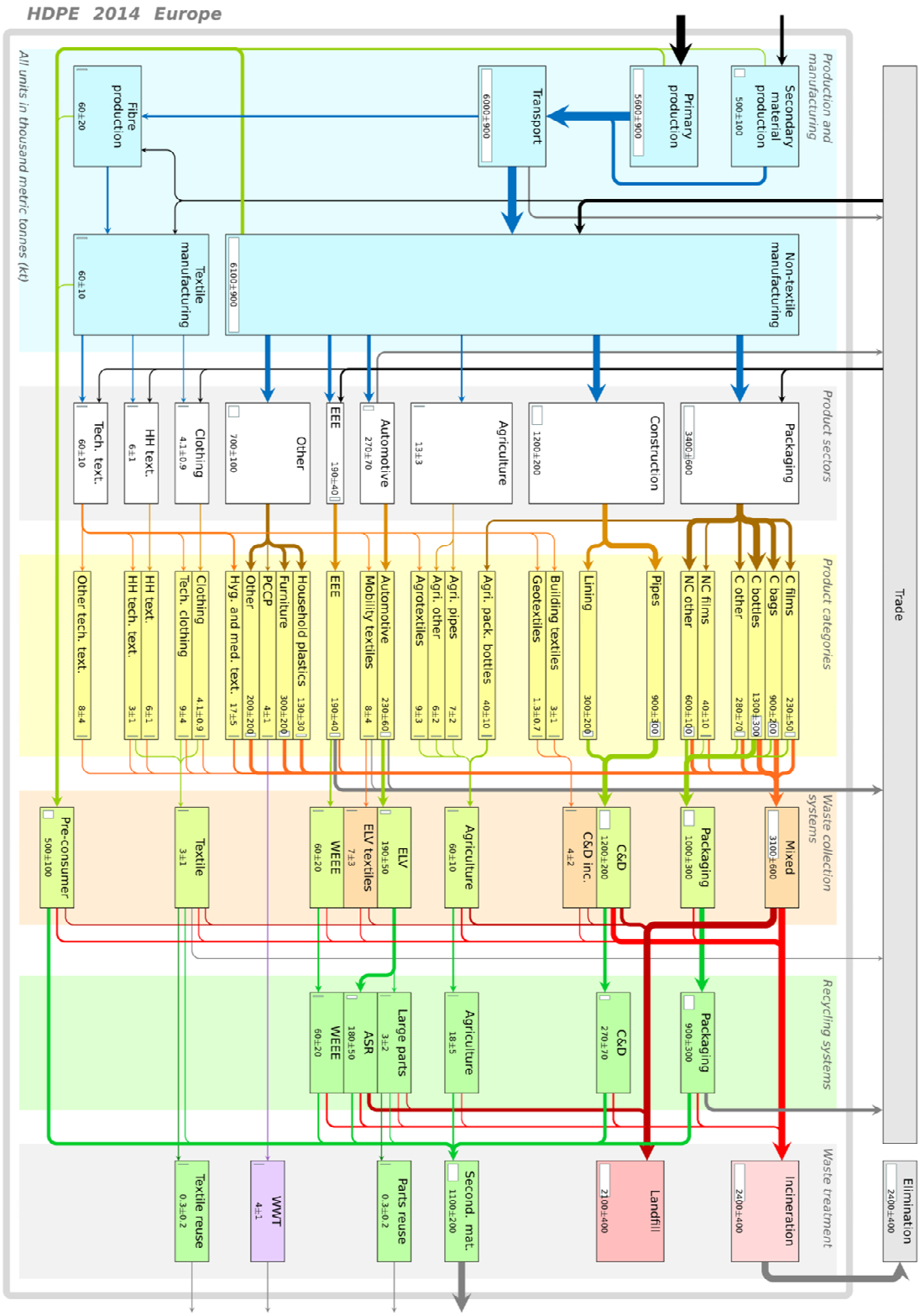

Figure S39: Flow diagram for HDPE in Europe. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

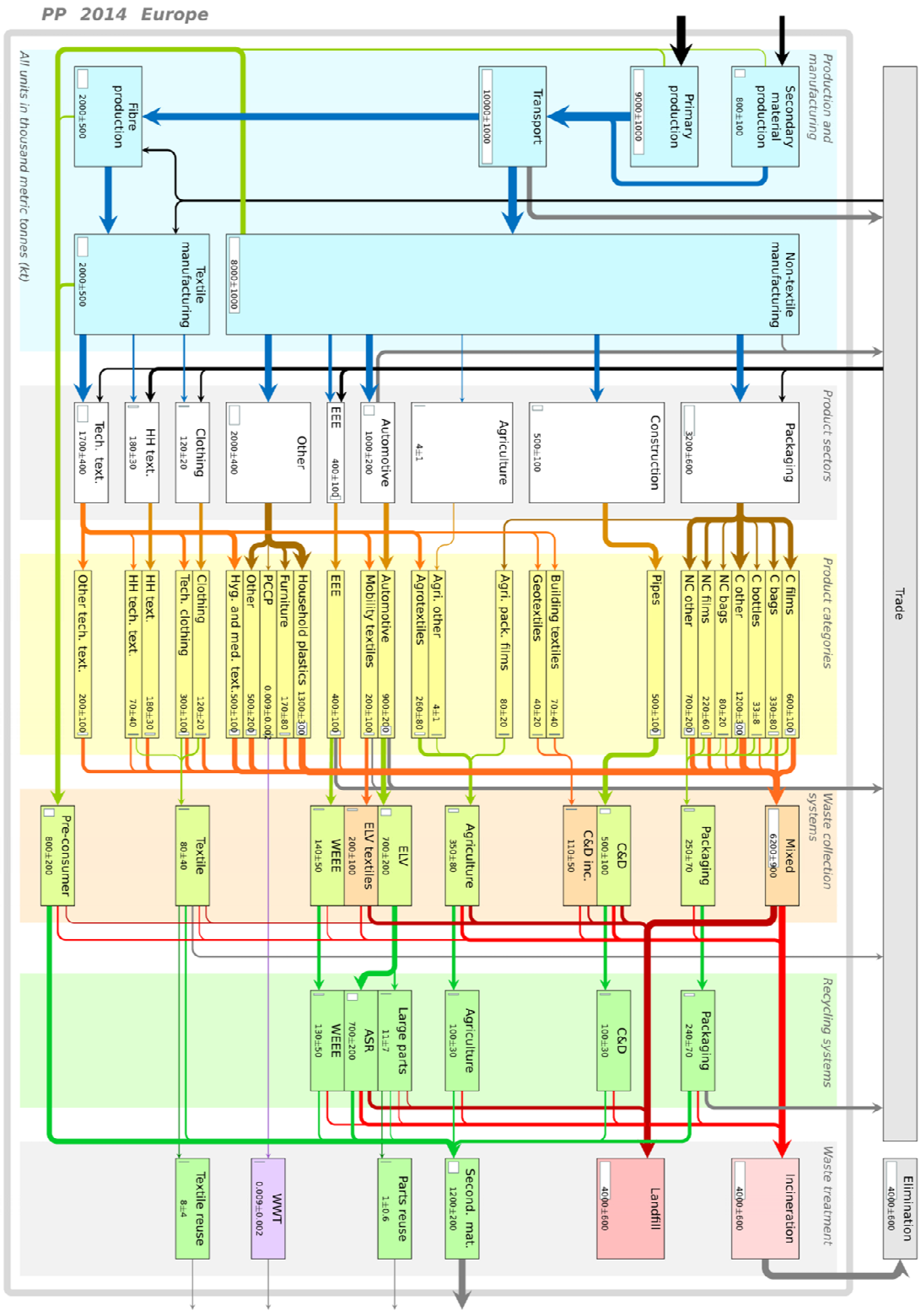

Figure S40: Flow diagram for PP in Europe. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

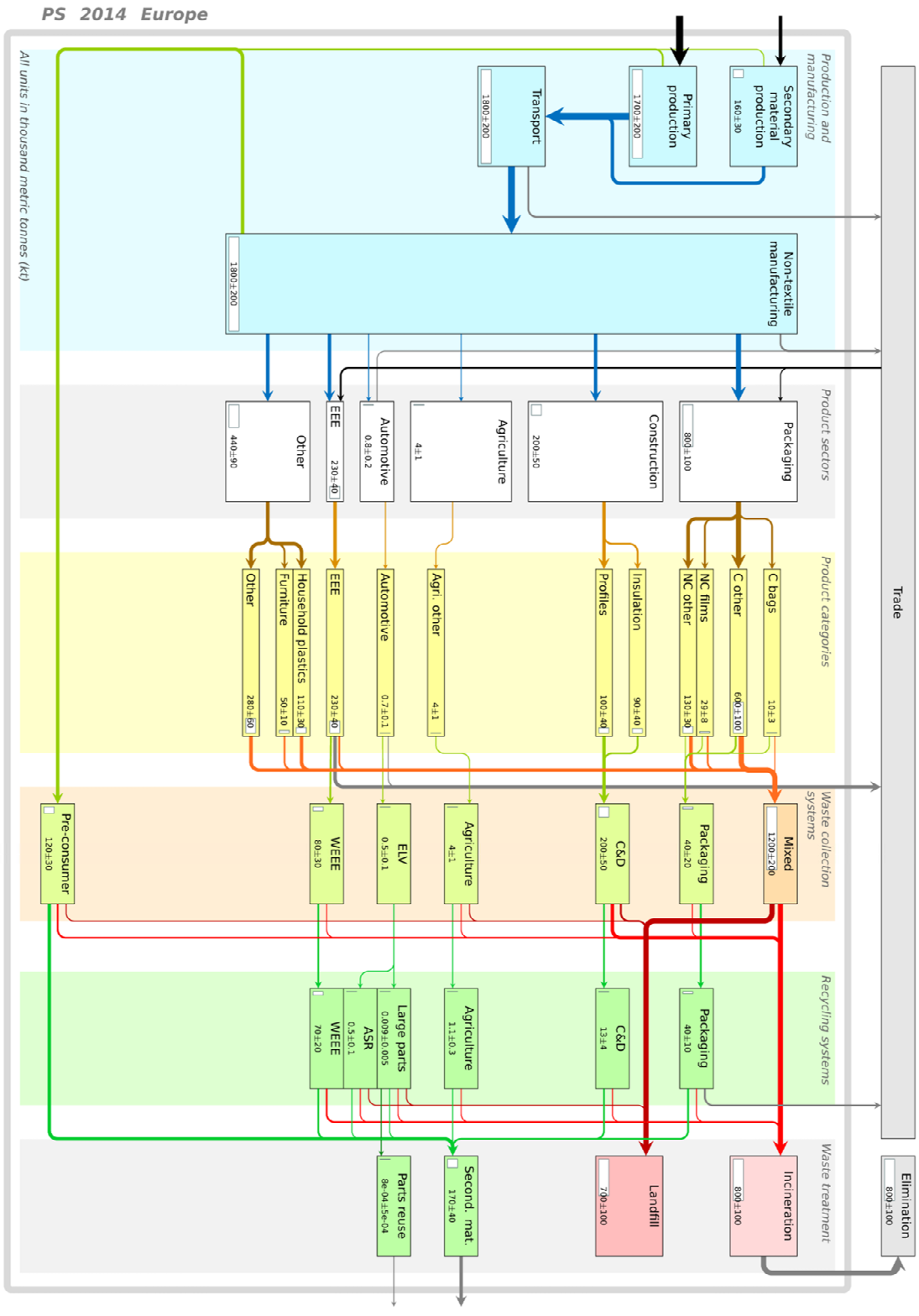

Figure S41: Flow diagram for PS in Europe. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

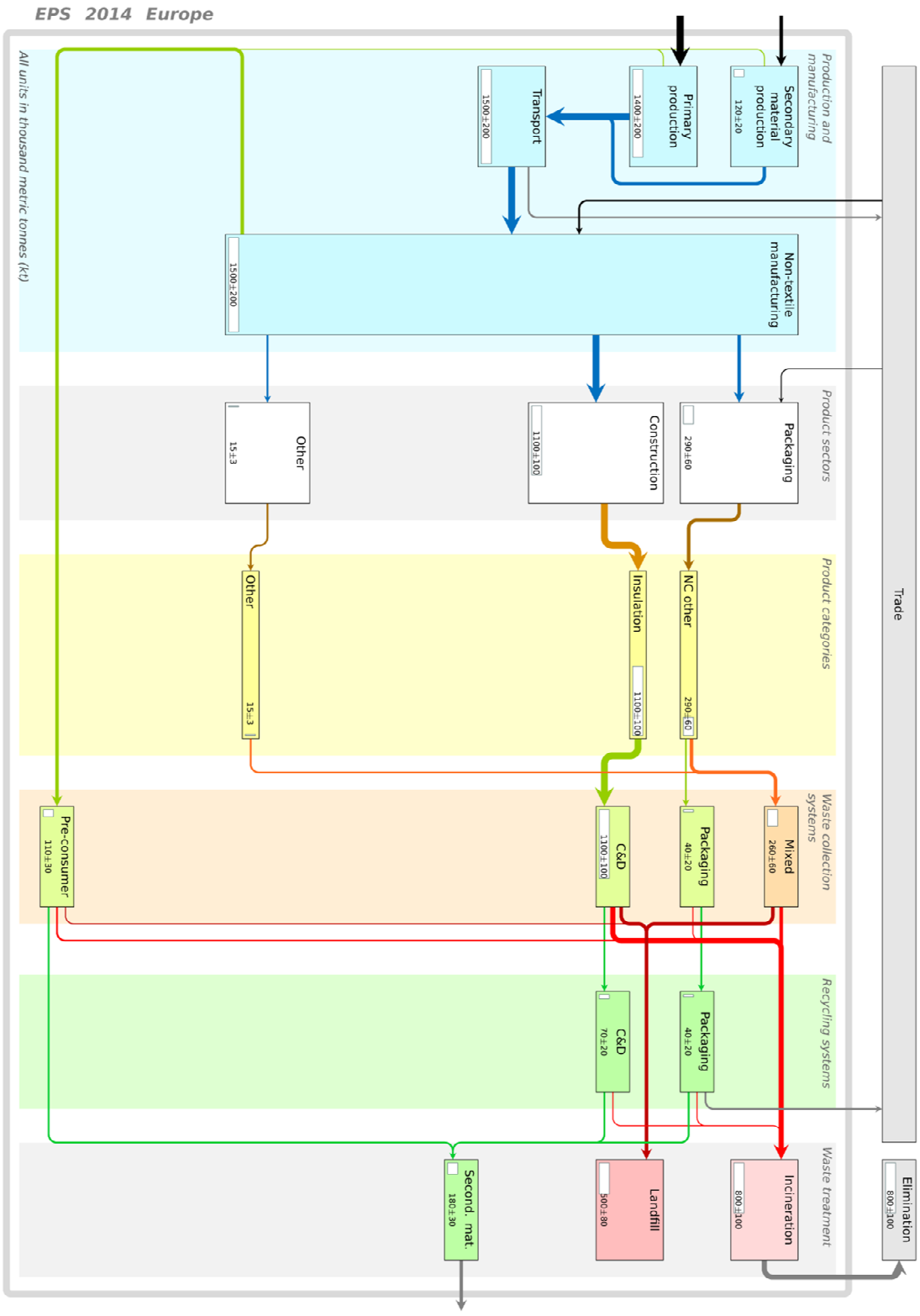

Figure S42: Flow diagram for EPS in Europe. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

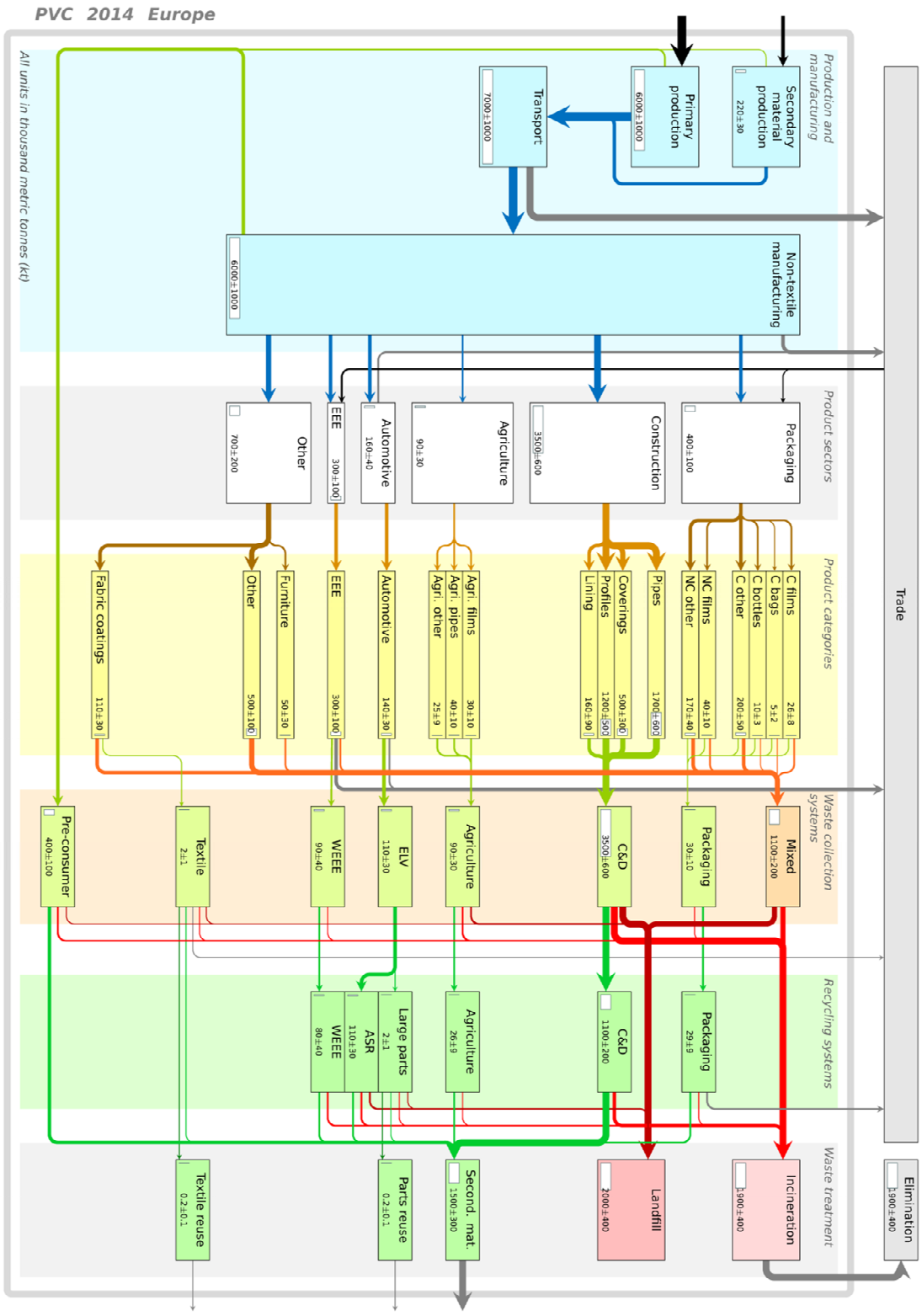

Figure S43: Flow diagram for PVC in Europe. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

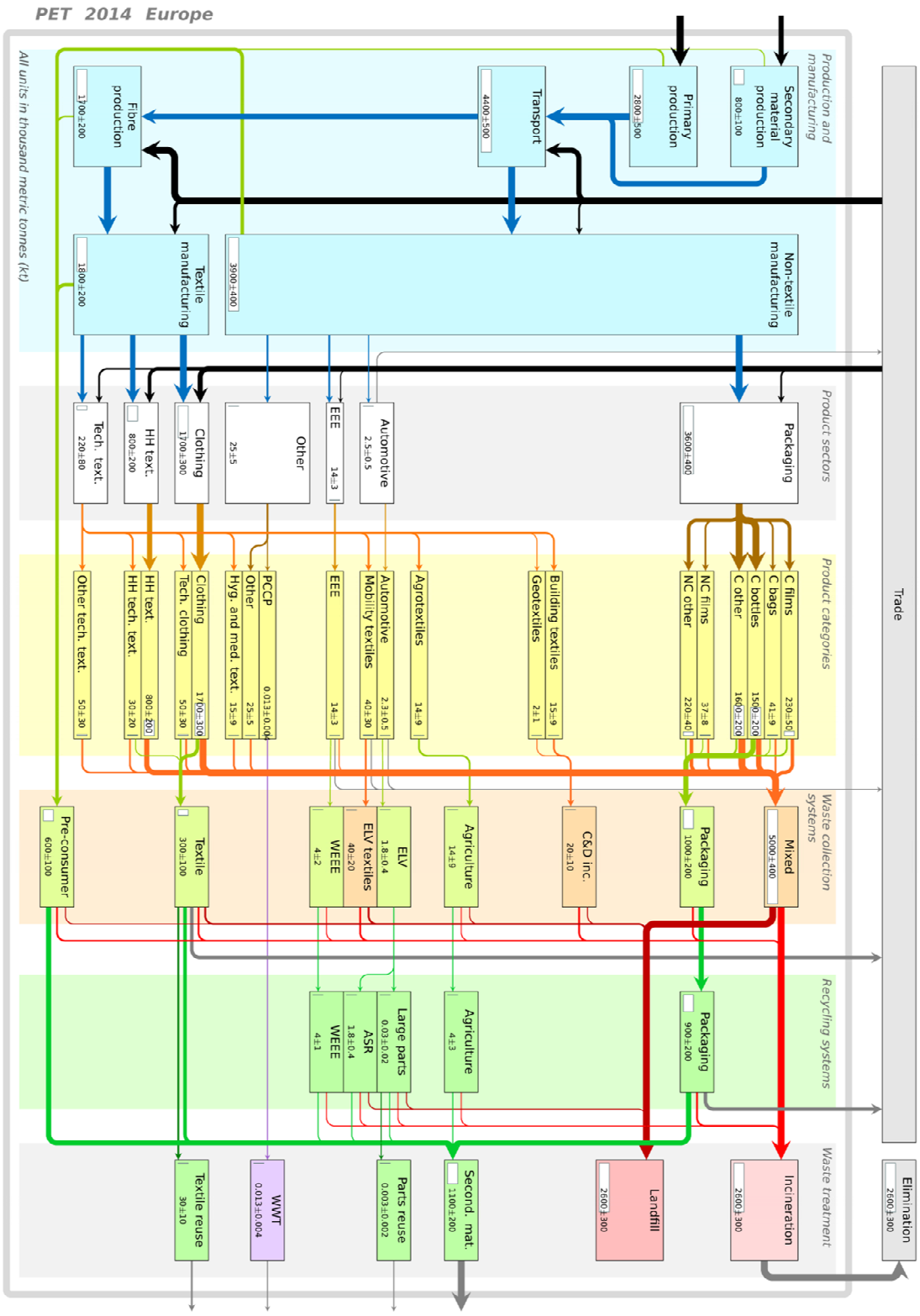

Figure S44: Flow diagram for PET in Europe. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

10.7.6 Complete flow diagrams for Switzerland

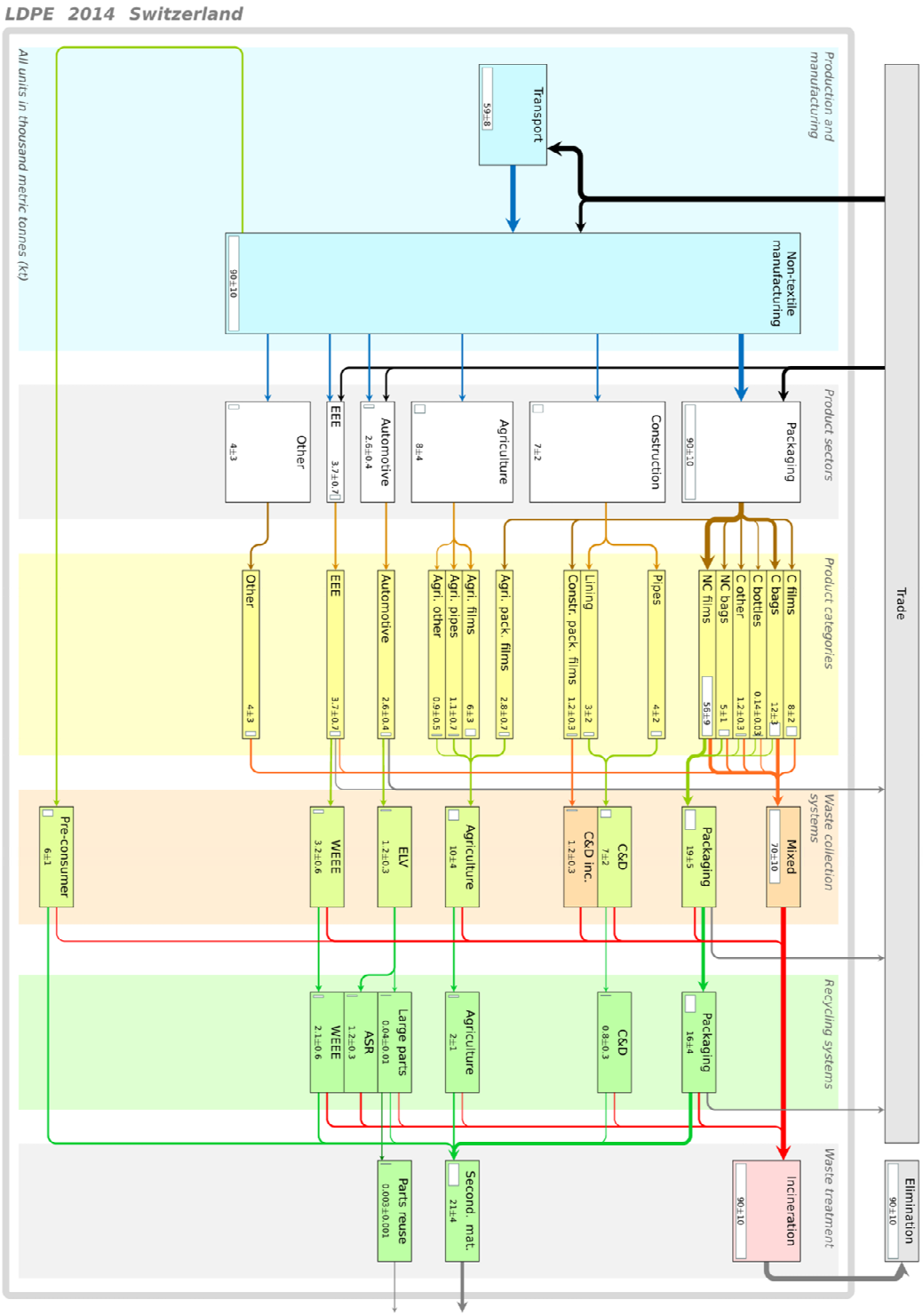

Figure S45: Flow diagram for LDPE in Switzerland. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

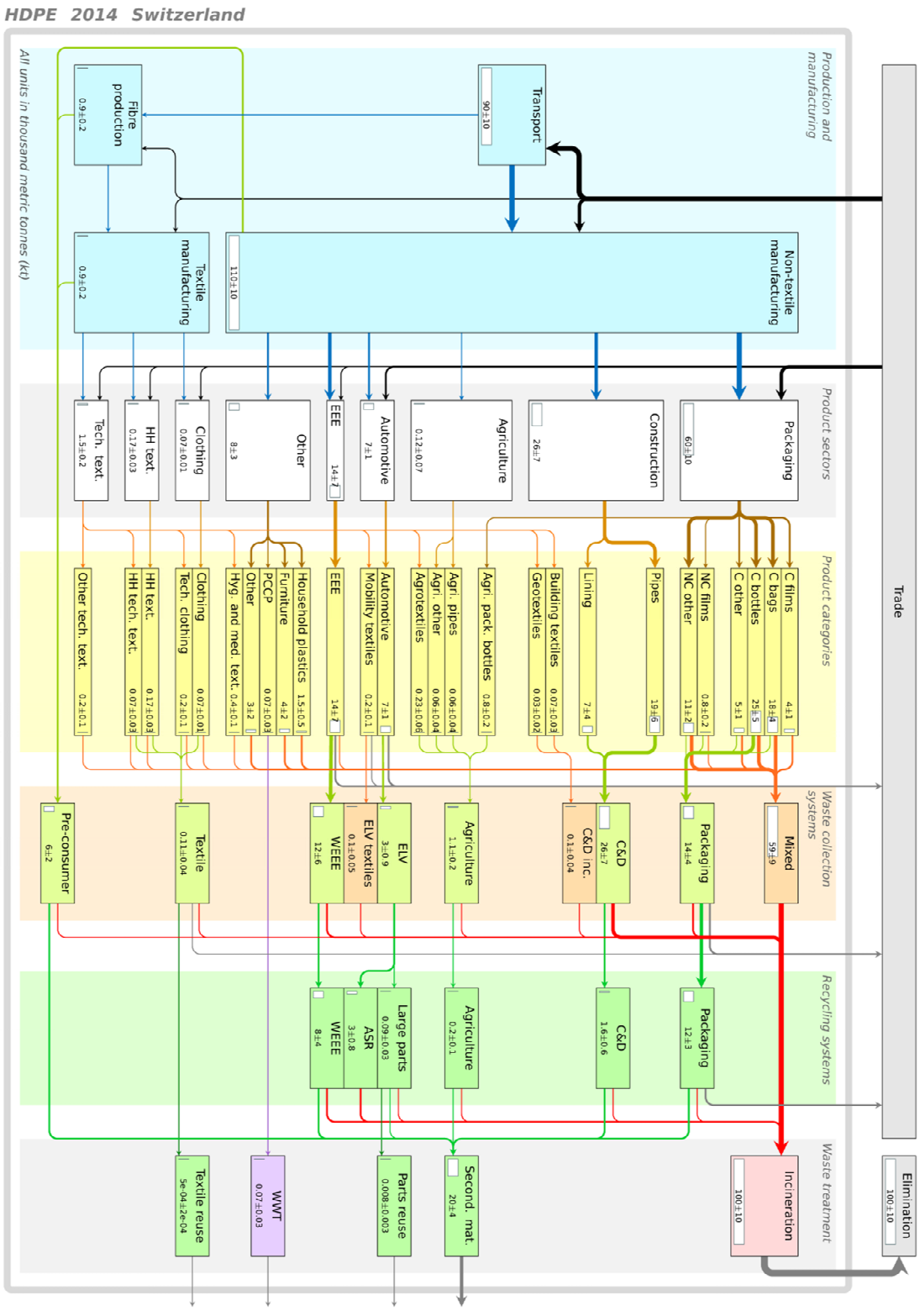

Figure S46: Flow diagram for HDPE in Switzerland. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

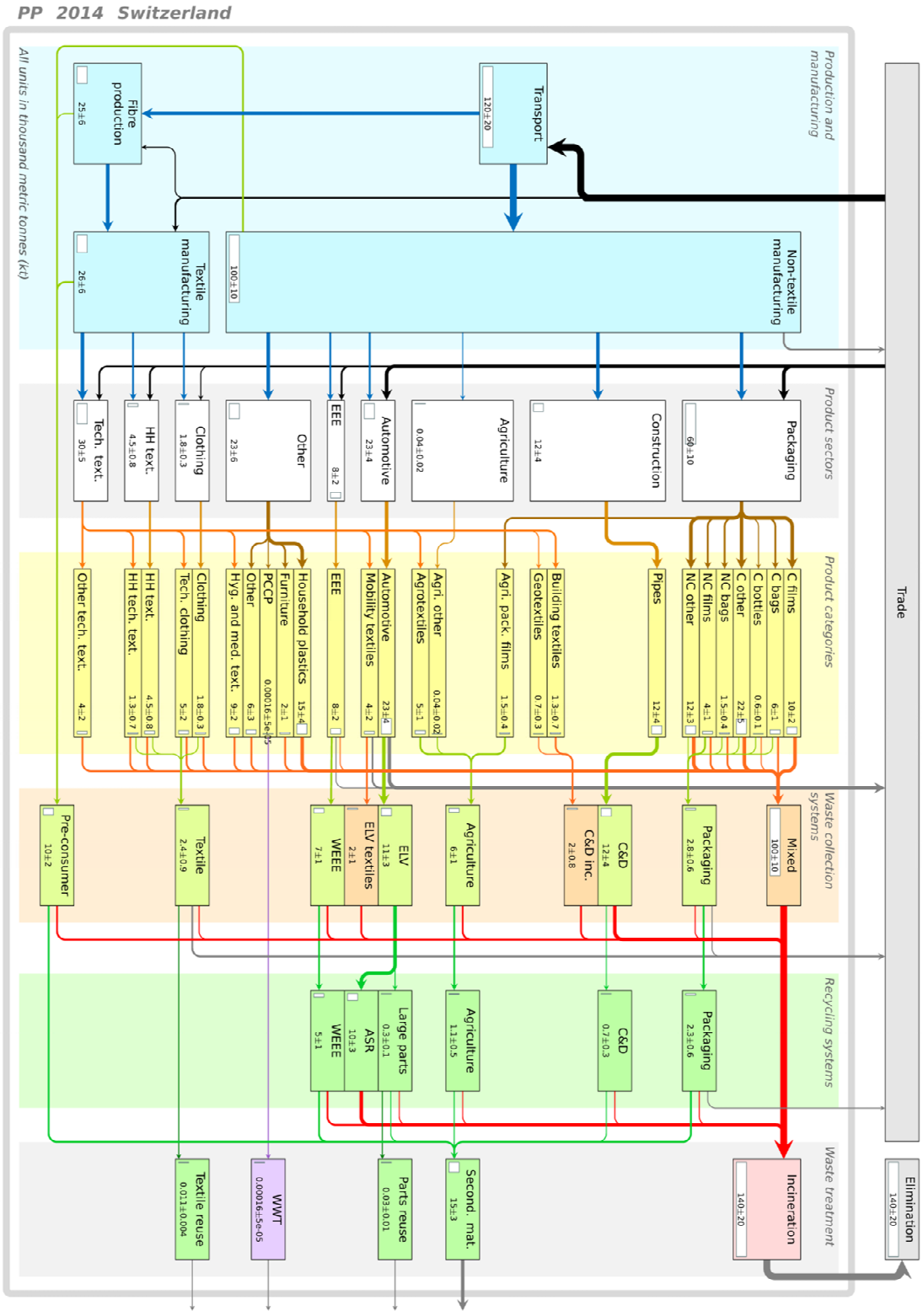

Figure S47: Flow diagram for PP in Switzerland. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

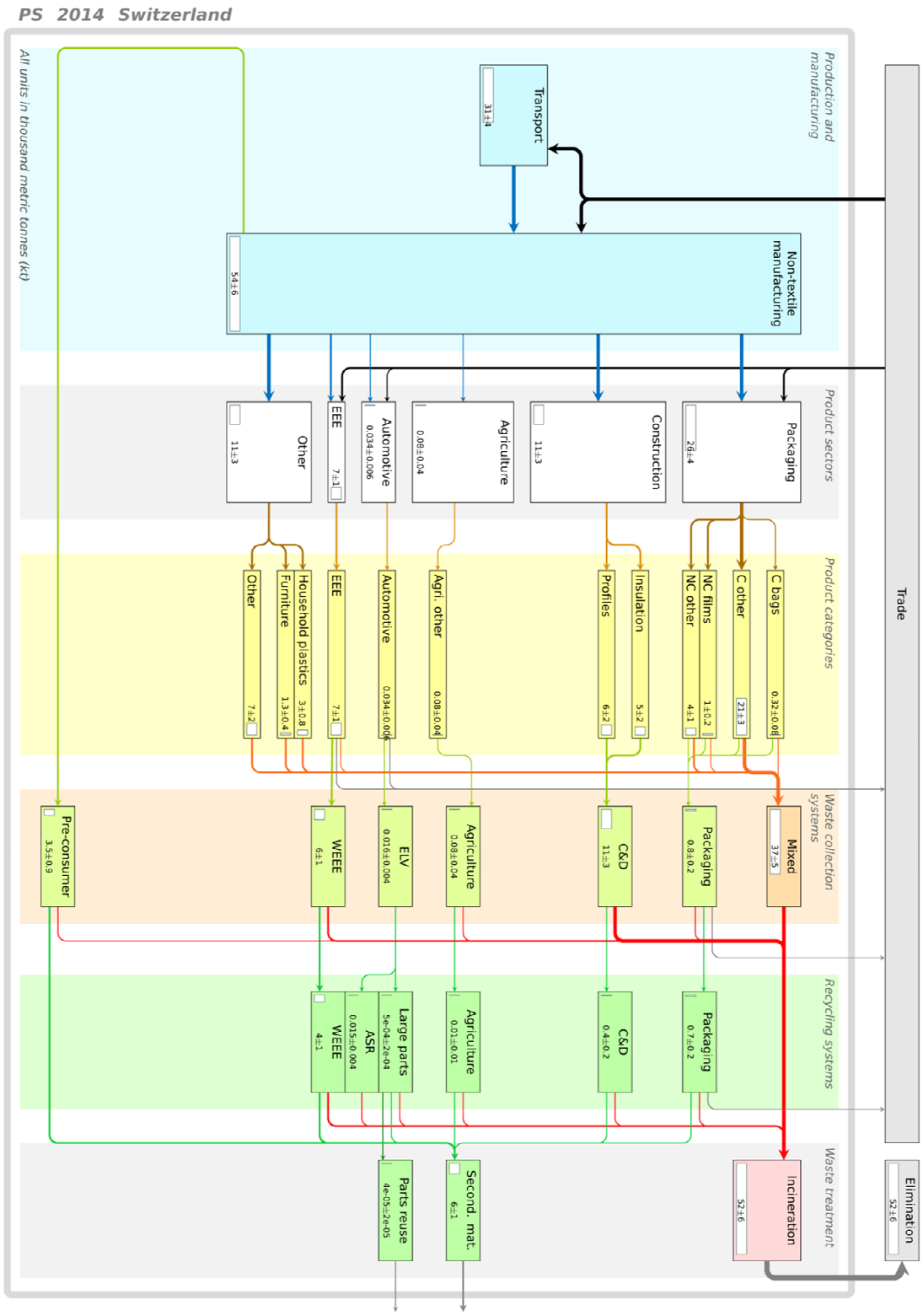

Figure S48: Flow diagram for PS in Switzerland. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

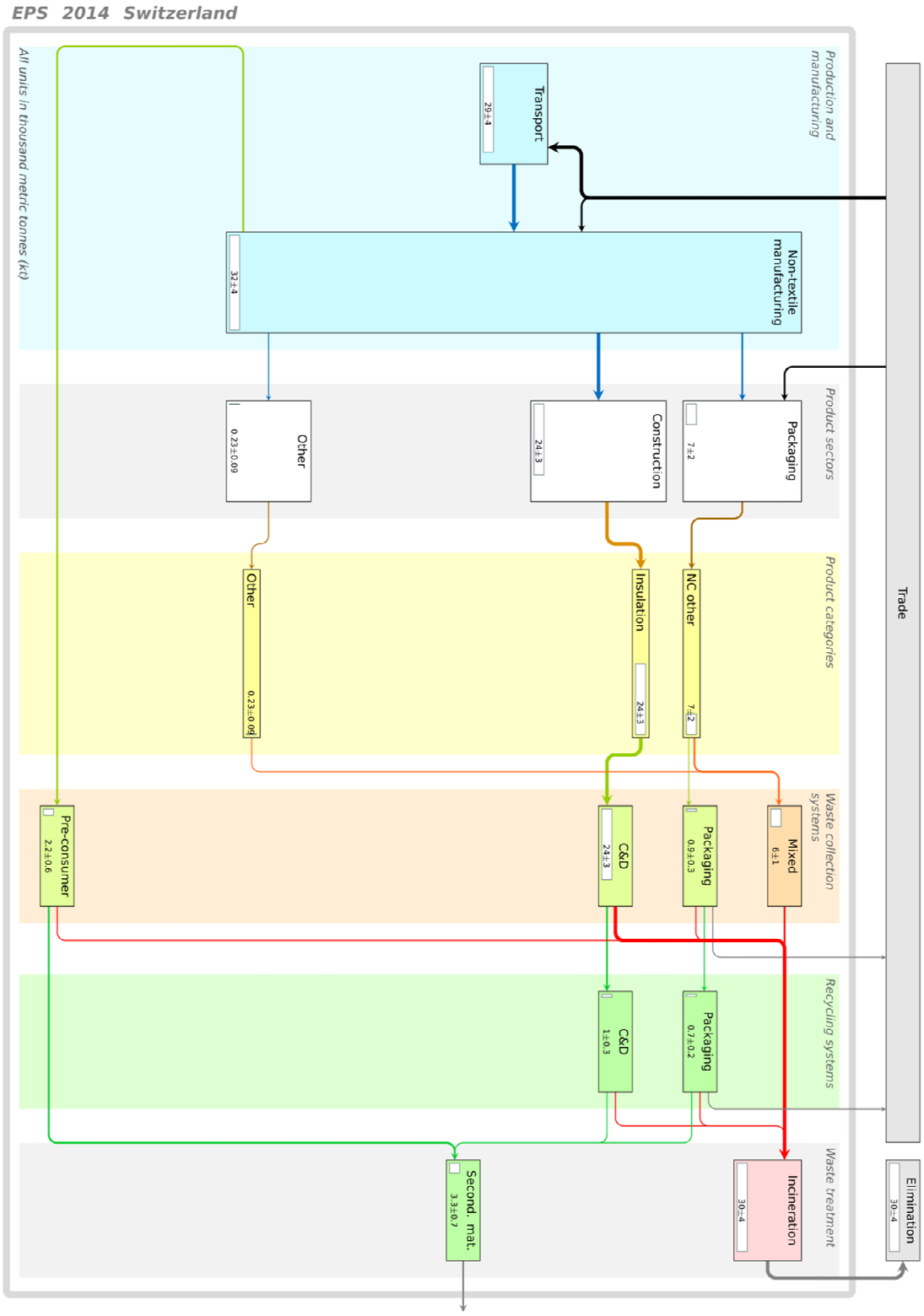

Figure S49: Flow diagram for EPS in Switzerland. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

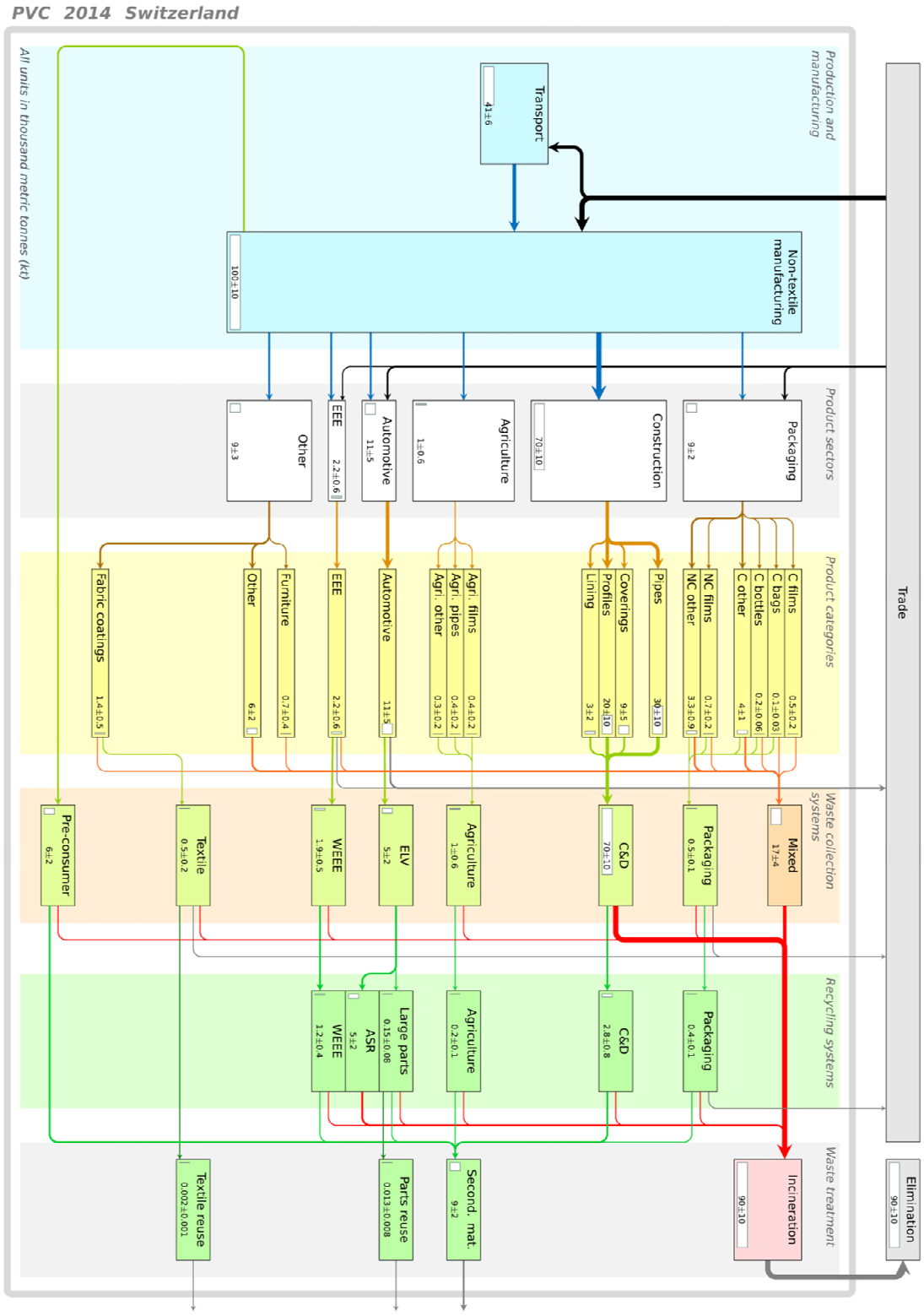

Figure S50: Flow diagram for PVC in Switzerland. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

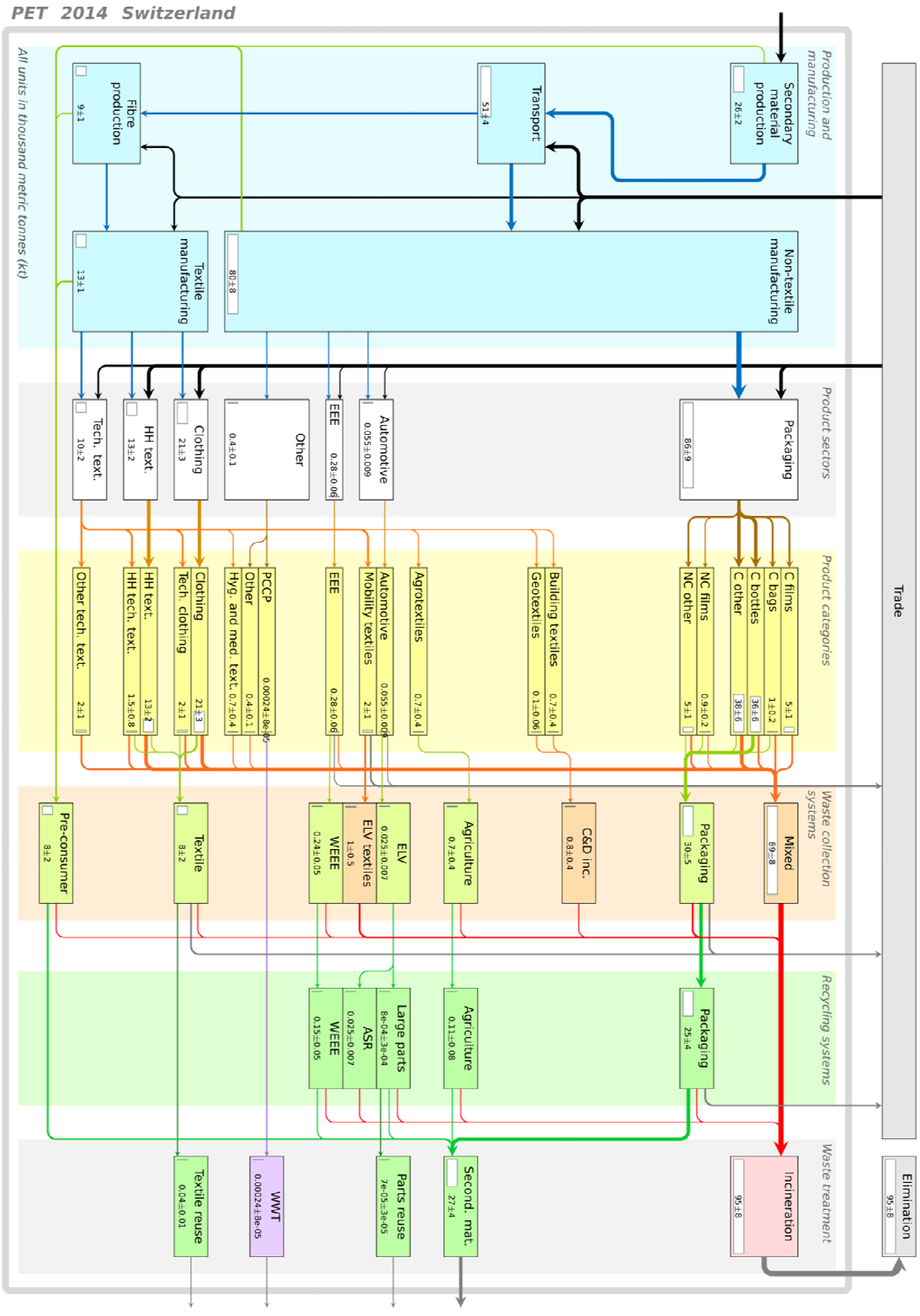

Figure S51: Flow diagram for PET in Switzerland. All units are in thousand metric tonnes (kt). The masses are rounded to the precision of the given standard deviation. Abbreviations: EEE (Electrical and Electronic Equipment), HH (Household), Tech. (Technical), Text. (Textiles), C (Consumer), NC (Non-Consumer), Hyg. and med. (Hygiene and medical), C&D (Construction and Demolition), ELV (End-of-Life Vehicles), WEEE (Waste of Electrical and Electronic Equipment), ASR (Automotive Shredder Residue), WEEP (Waste of Electrical and Electronic Plastics), WWTP (Waste Water Treatment Plant).

10.8 References

(1) Laner, D.; Feketitsch, J.; Rechberger, H.; Fellner, J. A Novel Approach to Characterize Data Uncertainty in Material Flow Analysis and Its Application to Plastics Flows in Austria. *J. Ind. Ecol.* **2016**, *20* (5), 1050–1063.

(2) European Commission -- Joint Research Centre -- Institute for Environment and Sustainability. *International Reference Life Cycle Data System (ILCD) Handbook -- General Guide for Life Cycle Assessment -- Detailed Guidance*; 2010; http://lct.jrc.ec.europa.eu/pdf-directory/ILCD-Handbook-General-guide-for-LCA-DETAIL-online-12March2010.pdf.

(3) Weidema, B. P.; Wesnaes, M. S. Data Quality Management for Life Cycle Inventories-an Example of Using Data Quality Indicators. *J. Clean. Prod.* **1996**, *4* (3–4), 167–174.

(4) Consultic Marketing & Industrieberatung GmbH. *Produktion, Verarbeitung Und Verwertung von Kunststoffen in Deutschland 2015 - Kurzfassung*; 2016.

(5) Burkhardt, M.; Englert, A.; Iten, R.; Schärer, S. *Entsorgung Nanosilberhaltiger Abfälle in Der Textilindustrie - Massenflüsse Und Behandlungsverfahren*; 2011; https://www.researchgate.net/profile/M_Burkhardt/publication/268804219_Entsorgung_nanosilberhaltiger_Abfalle_in_der_Textilindustrie_Massenflusse_und_Behandlungsverfahren/links/54763f420cf29afed6141ff2/Entsorgung-nanosilberhaltiger-Abfaelle-in-der-Textilin.

(6) Cook, J. G. Polyvinyl Derivatives. In *Handbook of Textile Fibres. Vol. II - Man-Made Fibres*; 2001; pp 392–535.

(7) European Man-made Fibres Asociation (CIRFS). Polyester in Europe http://www.cirfs.org/KeyStatistics/PolyesterinEurope.aspx (accessed Jul 26, 2017).

(8) Aizenshtein, E. M. Global and Russian Output of Polyester Fibres in 2013. *Fibre Chem.* **2015**, *47* (1), 1–7.

(9) Applied Market Information Ltd. *AMI's 2015 EUROPEAN PLASTICS INDUSTRY REPORT*; 2015.

(10) Waste Watch. *Plastics in the UK Economy: A Guide to Polymer Use and the Opportunities for Recycling*; 2003; http://legis.wisconsin.gov/lc/committees/study/2010/SUP/files/1 Plastics in the UK economy .pdf.

(11) Simon, C.; Hupfer, C.; PlasticsEurope. *Business Data and Charts 2015/2016: Plastics Europe Market Research Group*; 2016.

(12) Schelker, R.; Geisselhardt, P. *Erhebung Der Kunststoff Mengenströme in Der Schweiz (Schwerpunkt Polyolefine): „Stoff-Strom-Atlas Kunststoffe Schweiz"*; 2008.

(13) Meyer, U. F. *Wirtschaftsdaten 2014*; 2015; http://www.swiss-

plastics.ch/de/themen/daten-fakten/.

(14) Röschli, K. *Wirtschaftsdaten 2015*; 2016; http://www.swiss-plastics.ch/de/themen/daten-fakten/.

(15) Wazir Management Consultants. ROLE OF INDIAN TEXTILE AND APPAREL INDUSTRY IN CHANGING GLOBAL SUPPLY-DEMAND SCENARIO. In *International Conference and Exposition on Textiles & Apparel*; 2014.

(16) European Association for Textile Polyolefins (EATP). INDUSTRY DATA European Union (EU28) and Turkey - 2013 http://www.eatp.org/industry-data/ (accessed Jul 25, 2017).

(17) WRAP. *Report: Plastic Packaging Composition 2011*; 2013; http://www.wrap.org.uk/sites/files/wrap/Plastics Composition 2011 Report.pdf.

(18) Delgado, C.; Barruetabeña, L.; Salas, O. *Assessment of the Environmental Advantages and Drawbacks of Existing and Emerging Polymers Recovery Processes*; Wolf, O., Ed.; European Comission, 2007; http://ftp.jrc.es/EURdoc/eur22939en.pdf.

(19) European Association of Plastics Recycling and Recovery Organisations. *Plastic Packaging Waste Statistics 2014*; 2016; http://www.epro-plasticsrecycling.org/pages/75/epro_statistics.

(20) ARGUS; ACR; Carl Bro as. *European Packaging Waste Management Systems - Main Report*; 2001; http://ec.europa.eu/environment/waste/studies/packaging/epwms.pdf.

(21) APME. *Plastics: A Material of Choice in Building and Construction - Plastics Consumption and Recovery in Western Europe 1995*; 1995.

(22) PlasticsEurope. *Plastics - The Facts 2015: An Analysis of European Latest Plastics Production, Demand and Waste Data*; 2015; http://staging-plasticseurope.idloom.com/download_file/view/478/179.

(23) Hussain, I.; Hamid, H. Plastics in Agriculture. In *Plastics and Environment*; Andrady, A. L., Ed.; John Wiley and Sons, 2005; pp 185–209.

(24) Scarascia-Mugnozza, G.; Sica, C.; Russo, G. Plastic Materials in European Agriculture: Actual Use and Perspectives. *J. Agric. Eng.* **2012**, *42* (3), 15–28.

(25) Briassoulis, D.; Babou, E.; Hiskakis, M.; Scarascia, G.; Picuno, P.; Guarde, D.; Dejean, C. Review, Mapping and Analysis of the Agricultural Plastic Waste Generation and Consolidation in Europe. *Waste Manag. Res.* **2013**, *31* (12), 1262–1278.

(26) Consultic Marketing & Industrieberatung GmbH. *Analyse Der PVC Produktion, Verwertungsströme in Deutschland 2013*; 2013.

(27) Gries, T.; Veit, D.; Wulfhorst, B. Technical Textiles. In *Textile Technology*; 1995; Vol. 15, pp 26–33.

(28) Grebe, J. *Technical Textiles - Prospects and Challenges*; 2015.

(29) UNEP. *Plastic In Cosmetics: Fact Sheet*; 2015.

(30) Gouin, T.; Avalos, J.; Brunning, I.; Brzuska, K.; De Graaf, J.; Kaumanns, J.; Koning, T.; Meyberg, M.; Rettinger, K.; Schlatter, H.; et al. Use of Micro-Plastic Beads in Cosmetic Products in Europe and Their Estimated Emissions to the North Sea Environment. *SOFW J.* **2015**, *141*, 40–46.

(31) BUND. *BUND-Einkaufsratgeber: Mikroplastik – Die Unsichtbare Gefahr*; 2016.

(32) Petcore Europe. Over 66 billion PET bottles recycled in Europe in 2014 https://petcore-europe.prezly.com/over-66-billion-pet-bottles-recycled-in-europe-in-2014 (accessed Jul 26, 2017).

(33) Schelker, R.; Geisselhardt, P. Welche Fraktionen - Hauptkunststoffe: Fraktionen Aus Industrie + Gewerbe (I + G) Sowie Haushaltungen (HH). Zusätzliche Information zum Kunststoffatlas Schweiz 2011.

(34) Bundesamt für Umwelt. Abfallmengen Und Recycling 2014 Im Überblick. **2015**, 1.

(35) Oeko-Institut e.V. – Institute for Applied Ecology. *Assessment of the Implementation of Directive 2000 / 53 / EC on End-of Life Vehicles (the ELV Directive) with Emphasis on the End-of Life Vehicles of Unknown Whereabouts. Assessment of Current Situation of ELVs of Unknown Whereabouts – Preliminary Resul*; 2016.

(36) Foundation Auto Recycling Switzerland (SARS). *Annual Report 2014*; 2015.

(37) Huisman, J.; Botezatu, I.; Herreras, L.; Liddane, M.; Hintsa, J.; Luda di Cortemiglia, V.; Leroy, P.; Vermeersch, E.; Mohanty, S.; van den Brink, S.; et al. *Countering WEEE Illegal Trade Summary Report*; Lyon, France, 2015; http://www.cwitproject.eu/wp-content/uploads/2015/08/CWIT-Final-Summary1.pdf.

(38) Magalini, F.; Wang, F.; Huisman, J.; Kuehr, R.; Baldé, K.; van Straalen, V.; Hestin, M.; Lecerf, L.; Sayman, U.; Akpulat, O. *Study on Collection Rates of Waste Electrical and Electronic Equipment (WEEE)*; 2015; http://ec.europa.eu/environment/waste/weee/pdf/Final_Report_Art7_publication.pdf.

(39) Müller, E.; Widmer, R. *Materialflüsse Der Elektrischen Und Elektronischen Geräte in Der Schweiz*; 2010; https://www.bafu.admin.ch/dam/bafu/de/dokumente/abfall/fachinfo-daten/materialfluesse_vonelektrischenundelektronischengeraetenindersch.pdf.downloa d.pdf/materialfluesse_vonelektrischenundelektronischengeraetenindersch.pdf.

(40) Beton, A.; Dias, D.; Farrant, L.; Gibon, T.; Le Guern, Y.; Desaxce, M.; Perwuelzz, A.; Cordella, M.; Dodd, N. *Environmental Improvement Potential of Textiles (IMPRO Textiles)*; 2014; http://publications.jrc.ec.europa.eu/repository/bitstream/JRC85895/impro textiles_final report edited_pubsy web.pdf.

(41) Dettli, R.; Fasko, R.; Frei, U.; Habermacher, F. *Transformation Der Abfallverwertung in Der Schweiz Für Eine Hohe Und Zeitlich Optimierte Energieausnutzung*; 2014; http://www.bfe.admin.ch/php/modules/publikationen/stream.php?extlang=de&name=de_106736550.pdf.

(42) Eurostat. Treatment of waste by waste category, hazardousness and waste operations [env_wastrt] http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=env_wastrt&lang=fr (accessed May 29, 2017).

(43) Wälti, C.; Almeida, J. *Ent-Sorgen? Abfall in Der Schweiz Illustriert*; 2016; https://www.bafu.admin.ch/dam/bafu/de/dokumente/abfall/uz-umwelt-zustand/ent-sorgen_-_abfallinderschweizillustriert.pdf.download.pdf/ent-sorgen_-_abfallinderschweizillustriert.pdf.

(44) Haupt, M.; Vadenbo, C.; Hellweg, S. Do We Have the Right Performance Indicators for the Circular Economy?: Insight into the Swiss Waste Management System. *J. Ind. Ecol.* **2017**, *21* (3), 615–627.

(45) PlasticsEurope. *Overview Plastic Waste from Building & Construction by Polymer and by Recycling, Energy Recovery and Disposal - Building and Construction Post Consumer Waste Generation 2014 (Europe EU28+2)*; 2016.

(46) Eurostat. End-of-life vehicles by waste operations [env_waselv] http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=env_waselv&lang=en (accessed May 30, 2017).

(47) Mirabile, D.; Pistelli, M. I.; Marchesini, M.; Falciani, R.; Chiappelli, L. Thermal Valorisation of Automobile Shredder Residue: Injection in Blast Furnace. *Waste Manag.* **2002**, *22* (8), 841–851.

(48) Restrepo, E.; Løvik, A. N.; Wäger, P.; Widmer, R.; Lonka, R.; Müller, D. B. Stocks, Flows, and Distribution of Critical Metals in Embedded Electronics in Passenger Vehicles. *Environ. Sci. Technol.* **2017**, *51* (3), 1129–1139.

(49) Seyring, N.; Kling, M.; Weissenbacher, J.; Hestin, M.; Lecerf, L.; Magalini, F.; Khetriwal, D. S.; Kuehr, R. *Study on WEEE Recovery Targets, Preparation for Re-Use Targets and on the Method for Calculation of the Recovery Targets. Final Report*; 2015; http://ec.europa.eu/environment/waste/weee/pdf/16. Final report_approved.pdf.

(50) Swico; SENS; SLRS. *Technical Report 2017*; 2017.

(51) Müller, A. Situationsanalyse Der Entsorgung Und Verwertung von Kunststoffabfällen Aus Der Landwirtschaft, Berner Fachhochschule, 2016.

(52) Schelker, R.; Geisselhardt, P. *Projekt „Kunststoff-Verwertung Schweiz ": Bericht Module 1 Und 2*; 2011; https://www.bafu.admin.ch/dam/bafu/fr/dokumente/abfall/externe-studien-berichte/projekt_kunststoff-

verwertungschweiz.pdf.download.pdf/projekt_kunststoff-verwertungschweiz.pdf.

(53) Astrup, T.; Fruergaard, T.; Christensen, T. H. Recycling of Plastic: Accounting of Greenhouse Gases and Global Warming Contributions. *Waste Manag. Res.* **2009**, *27*, 763–772.

(54) Velis, C. *Global Recycling Markets - Plastic Waste: A Story for One Player – China*; 2014; https://www.iswa.org/fileadmin/galleries/Task_Forces/TFGWM_Report_GRM_Plastic_China_LR.pdf.

(55) Brouwer, M. T.; Thoden van Velzen, E. U.; Augustinus, A.; Soethoudt, H.; De Meester, S.; Ragaert, K. Predictive Model for the Dutch Post-Consumer Plastic Packaging Recycling System and Implications for the Circular Economy. *Waste Manag.* **2017**, *71*, 62–85.

(56) Santini, A.; Passarini, F.; Vassura, I.; Serrano, D.; Dufour, J.; Morselli, L. Auto Shredder Residue Recycling: Mechanical Separation and Pyrolysis. *Waste Manag.* **2012**, *32* (5), 852–858.

(57) Morselli, L.; Santini, A.; Passarini, F.; Vassura, I. Automotive Shredder Residue (ASR) Characterization for a Valuable Management. *Waste Manag.* **2010**, *30* (11), 2228–2234.

(58) Cafe, T. Physical Constants for Investigators http://www.tcforensic.com.au/docs/article10.html (accessed Oct 3, 2017).

(59) Eurostat. Eurostat PRODCOM database http://ec.europa.eu/eurostat/web/prodcom/data/database#.

(60) Simon, C.; Hupfer, C. *Business Data and Charts 2014/2015 Plastics Europe Market Research Group (PEMRG)*; 2015.

(61) Vinyl 2010. *Progress Report 2015*; 2015; https://vinylplus.eu/uploads/docs/VinylPlus_Progress_Report_2015_English.pdf.

(62) Eurostat. EU trade since 1988 by HS2,4,6 and CN8 http://ec.europa.eu/eurostat/web/international-trade-in-goods/data/database#.

(63) Swiss Federal Customs Administration FCA. SwissImpex Version 3.4.2-1 https://www.swiss-impex.admin.ch/index.xhtml (accessed Mar 31, 2017).

(64) Van Houte, F. Polyester Fibres in Europe: Developments and Trends. In *9th China International Polyester and Intermediates Forum*; European Man-made Fibres Association, 2012; pp 1–28.

(65) European Man-made Fibres Asociation (CIRFS). About Man-made Fibres http://www.cirfs.org/ManmadeFibres/AboutManmadeFibres.aspx (accessed Aug 24, 2017).

(66) McComb, G. Plastics in Everyday Household Articles

http://www.robotoid.com/appnotes/construction-plastics-in-household-items.html (accessed Aug 8, 2017).

(67) Andrady, A. L.; Neal, M. A. Applications and Societal Benefits of Plastics. *Philos. Trans. R. Soc. Lond. B. Biol. Sci.* **2009**, *364* (1526), 1977–1984.

(68) Packaging waste by waste operations and waste flow [env_waspac] http://ec.europa.eu/eurostat/product?code=env_waspac&language=en&mode=view (accessed Aug 9, 2017).

(69) ACEA. *The Automobile Industry Pocket Guide 2015-2016*; 2015; http://www.acea.be/uploads/press_releases_files/POCKET_GUIDE_2015-2016.pdf.

(70) Mashek, W.; Holmes, K.; Martin, K. *Automotive Recycling: Devalued Is Now Revalued.*; 2016.

(71) Buekens, A.; Yang, J. Recycling of WEEE Plastics: A Review. *J. Mater. Cycles Waste Manag.* **2014**, *16* (3), 415–434.

(72) Groß, R.; Bunke, D.; Gensche, C.-O.; Zangl, S.; Manhart, A. Study on Hazardous Substances in Electrical and Electronic Equipment, Not Regulated by the RoHS Directive. **2008**, *49* (070307), 273.

(73) Bartlett, C.; McGill, I.; Willis, P. *Textiles Flow and Market Development Opportunities in the UK*; 2013; http://www.wrap.org.uk/sustainable-textiles/scap/re-use-and-recycling/textiles-reports-and-trials.

(74) David Rigby Associates (DRA). Mixed Outlook for Synthetic Polymers in Technical Textile Markets. *Glob. Fibers Feed. Rep.* **2003**, No. 018, 10–13.

(75) Transparency Market Research. Technical Textiles Market to Reach US$160.38 Billion owing to Innovative Product Development http://www.transparencymarketresearch.com/pressrelease/technical-textiles-market.htm (accessed Feb 22, 2017).

11 Supporting Information for: Polymer-specific modelling of the environmental emissions of seven commodity plastics as macro- and microplastics

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- Number of pages in supporting information: 62
- Number of figures: 22
- Number of tables: 19

11.1 Method: Additional information and figures

The method used in present study can be subdivided into four main parts: data collection, probability distribution creation, modelling and results analysis (Figure S52).

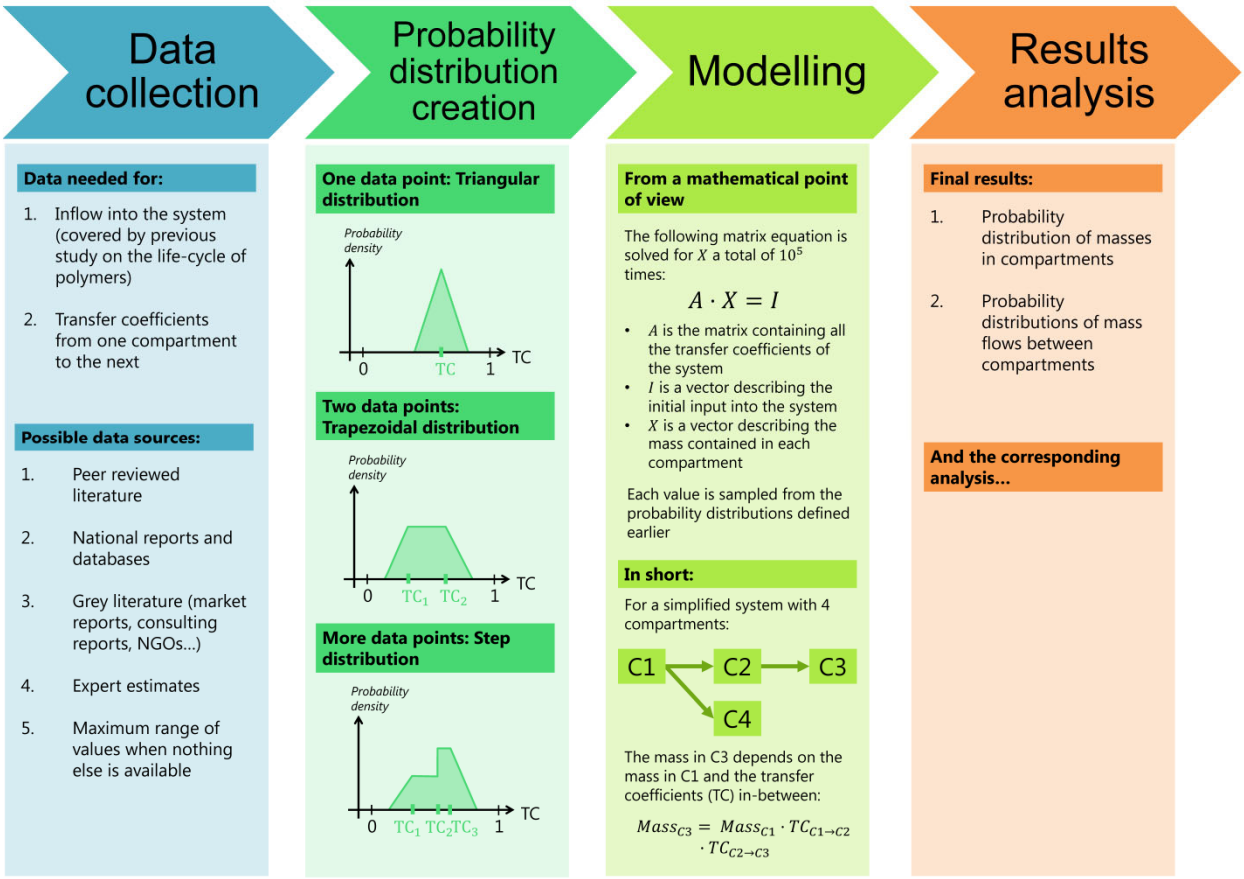

Figure S52: Overview of the approach followed in this study, divided into four main parts: data collection, probability distribution creation, modelling, data analysis.

Most of the following information in this chapter is an excerpt from the supplementary information from the previous publication on the life-cycle of plastics[1].

11.1.1 PMFA theory

Let there be a system containing n processes, of which m are not sinks. The flow from process i to process j is defined using a transfer coefficient (TC). This TC is defined as the ratio of the outflowing mass to the mass contained in the process:

$$TC_{ij} = \frac{m_{ij}}{M_i}.$$

All the transfer coefficients needed to describe the flows in the system can be gathered into a matrix as follows:

$$A = \begin{pmatrix} \mathbf{1} & \cdots & -TC_{m1} & 0 & \cdots & 0 \\ \vdots & \ddots & \vdots & \vdots & \vdots & \vdots \\ -TC_{1m} & \cdots & \mathbf{1} & 0 & \cdots & 0 \\ -TC_{1m+1} & \cdots & -TC_{mm+1} & \mathbf{1} & \cdots & 0 \\ \vdots & \cdots & \vdots & \vdots & \ddots & \vdots \\ -TC_{1n} & \cdots & -TC_{mn} & 0 & \cdots & \mathbf{1} \end{pmatrix},$$

where the TCs in column i describe the outgoing flows from process i to processes $1, \ldots, n$. The columns until m contain non-zero values for TCs. The remaining columns correspond to sinks, from which no outflows are defined.

The Probabilistic Material Flow Analysis (PMFA) method relies on solving a matrix equation iteratively. The matrix equation reads:

$$A \cdot X = I$$

where $A \in (\mathbb{R}^n \times \mathbb{R}^n)$ is a matrix describing the flows between n processes as described above, $I \in \mathbb{R}^n$ is a vector describing the initial material input into n processes, and $X \in \mathbb{R}^n$ describes the final mass in n processes. This equation system is solved a number of times in a Monte-Carlo simulation, where each parameter is sampled from chosen Bayesian distributions.

11.1.2 Uncertainty attribution

The distribution spread for a specific parameter is determined via a semi-quantitative approach inspired by another MFA study[2]. This approach is favoured over other methodologies described for Life Cycle Inventory data[3], as it permits to produce a continuous range of uncertainties. Five different data quality indicators are used to represent the different uncertainty sources[2,4]. The Data Quality Indicator Scores (DQIS) can take a value between 1 and 4, with high values corresponding to poorer data quality (Table S17).

Table S17: Pedigree matrix with 5 data quality indicators and 4 data quality levels.

Data quality	Very good	Good	Poor	Very poor
DQIS	**1**	**2**	**3**	**4**
Geographical representativeness	Same region *(EU28 and EU28+2 qualify)*	Socioeconomically similar region *i.e. Europe vs. Switzerland*	Socioeconomically different region *i.e. USA vs. Europe*	Socioeconomically very different region *i.e. World vs. Europe*
Temporal representativeness	2014	2009-2013, 2015	2004-2008	Prior to 2004
Material representativeness	Same polymer	Same polymer datum corrected with data for all polymers	Data for a different polymer, or for plastic as a whole, or including similar materials	Including non-similar materials
Completeness	Includes all relevant processes/flows	Includes main processes/flows	Partially including main processes/flows	Important processes/flows are missing
Source reliability	Official report Peer reviewed documentation	Market reports and other reports Public database	Qualified estimate	Non-qualified estimate

The DQIS are transformed into Coefficients of Variation (CV) with an exponential function, as it better reflects the qualitative evaluation of the data quality indicators listed above (Table S17) than a linear function. All parameters are assumed to have high sensitivity to the DQIS (for details, see the original article[2]). The CVs are parametrized differently for the source reliability indicator and the other indicators, to allow for a non-zero uncertainty even for good source reliabilities:

$$\mathrm{CV}_{rel} = 1.5 \cdot e^{1.105 \cdot \mathrm{DQIS}}$$
$$\mathrm{CV}_{other} = 1.5 \cdot e^{1.105 \cdot (\mathrm{DQIS}-1)}.$$

The total CV of a datum is then calculated from the individual CVs using the variance additivity rule:

$$\mathrm{CV}_{tot} = \sqrt{{\mathrm{CV}_{geo}}^2 + {\mathrm{CV}_{temp}}^2 + {\mathrm{CV}_{mat}}^2 + {\mathrm{CV}_{tech}}^2 + {\mathrm{CV}_{rel}}^2}.$$

The described methodology is valid for normal distributions. As we prefer to use triangular distributions and other distributions based on triangular distributions (see next section for a complete description), we would like to obtain information on the maximal spread of the distribution, which is the required information to create a symmetric triangular distribution.

Knowing that the variance of a triangular distribution can be expressed as follows:

$$\sigma^2 = \frac{a^2 + b^2 + c^2 - ab - ac - bc}{18},$$

where a, b and c are the minimum, maximum and modal values of the triangular distribution. With the following constraint, the symmetry of the triangular distributions is enforced:

$$b = 2c - a\,.$$

The standard deviation can then be rewritten:

$$\sigma = \frac{1}{\sqrt{6}}(a - c)$$

Knowing that the CV is defined as the standard deviation divided by the mean, the distance from the mean to one extreme value of the triangular distribution can be expressed as such:

$$\frac{a - c}{c} = \sqrt{6} \cdot \frac{\sigma}{c} \cong 2.45 \cdot \mathrm{CV}_{tot}$$

This final expression is the one used to compute the distributions' spread. A comparison of a normal and a triangular distribution is shown in Figure S23.

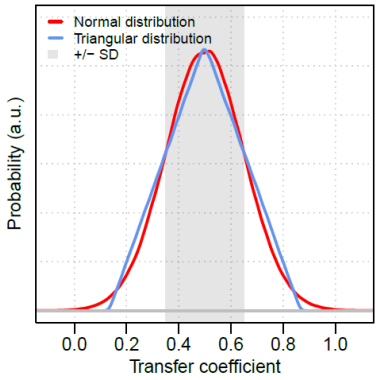

Figure S53: Comparison of a normal and triangular distribution which have the same parameters: $\mu = 0.5$, $\mathrm{CV}_{tot} = 0.3$. The triangular distribution's spread was calculated from CV_{tot} as described above.

11.1.3 Probability distributions

Depending on the amount of data available for a specific parameter in the model, different probability distributions are used to model the data (Figure S54):

4. If a single data point is available, the distribution chosen is a triangular distribution (Figure S54a) centred on the data point, with a maximal spread defined by the pedigree matrix approach explained in the previous chapter.

5. If two data points are available, they are combined in a trapezoidal distribution (Figure S54b), where the lower and upper tails are defined using the CVs associated to the two data points.
6. If three or more data points are available, they are combined together in what we call a step function. To model this type of distribution, the latest version available of that code was used[5].

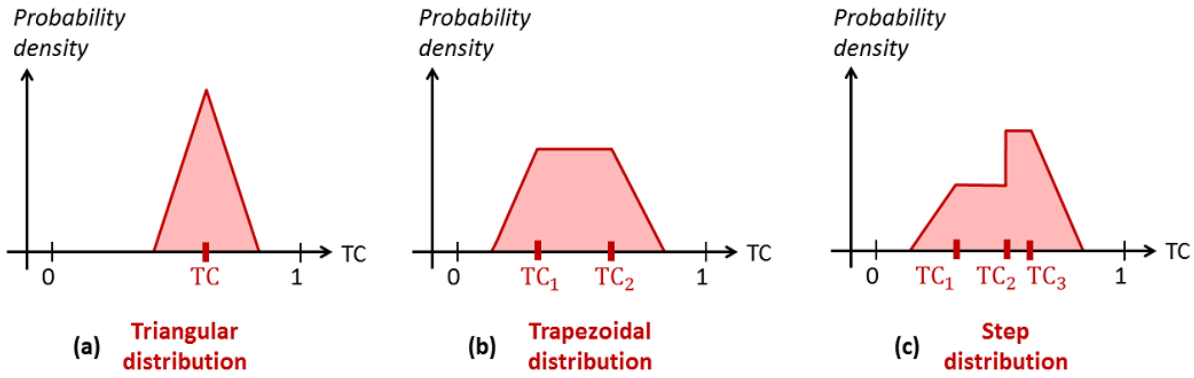

Figure S54: Schematic representation of the distributions chosen depending on the number of available data points: (a) for one, (b) for two, (c) for more than two.

The approach used, in which a distribution's spread is defined by a central value and a CV, may lead to values that are unphysical. For example, obtaining transfer coefficients (TCs) smaller than 0 or larger than 1 would violate mass conservation. In such cases, the approach chosen is to truncate the distribution at the threshold values (Figure S55).

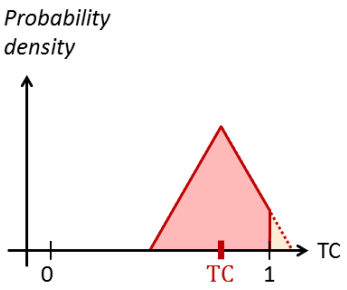

Figure S55: If the spread chosen for a data point would lead the distribution to have values larger than what is physically realistic, a simple truncation is applied to the distribution.

11.1.4 Algorithm: Data preparation and calculation

The following describes the order in which operations are performed to set up the model, calculate export flows, integrate emission flows, normalize the system and run the simulation.

6. **Import of the life-cycle parameters:** The Bayesian distributions for the TCs and the inflows into the system are generated for the life-cycle as calculated in the previous study[1]. The TCs are then normalized and the export flows are calculated. As in the previous study, some special flows are calculated at this moment: the consumption of Personal Care and Cosmetic Products (PCCP). In addition, the consumption of hygiene products and shotgun cartridges is now implemented at this stage.
7. **Implementation of the release flows:** The TCs corresponding to emission flows are included at this moment, while renormalizing the other non-emission flows. For example, if 30% of the mass in *X* is emitted from compartment *X*, only 70% may flow from the summed life-cycle flows.
8. **Special flows are calculated (for a detailed description, see the chapter on the description of the emission flows):**
    a. The TCs from *Wastewater secondary treatment (MP)* to *Surface Water (MP)* and *Wastewater secondary treatment (MP)* to *Wastewater tertiary treatment (MP)* are calculated considering the step distribution from *Wastewater secondary treatment (MP)* to *Sludge (MP)*.
    b. Similarly, the TCs for the rural sweeping efficiency are calculated separately in order to define rest flows appropriately.
    c. The TCs for the release of fibres from textiles are defined using many different parameters, to which an uncertainty is given using the Pedigree matrix for uncertainty propagation. A detailed description is given in a later chapter of this document.
    d. The TCs for the calculation of consumption of straws and cutlery are separately calculated because they rely on the TCs used for their release.
    e. The release of plastic parts from cars is also calculated in reverse.
    f. The release of plastic through organic waste collection is calculated using the amount known to be found in collected organic waste, and attributed to consumer product categories.
9. **Monte Carlo Simulation:** The matrix equation is solved iteratively 10^5 times.

11.2 Results: Additional figures

11.2.1 Aggregated emission flows

Similar flowcharts as in Figure 1 were created for all remaining polymers.

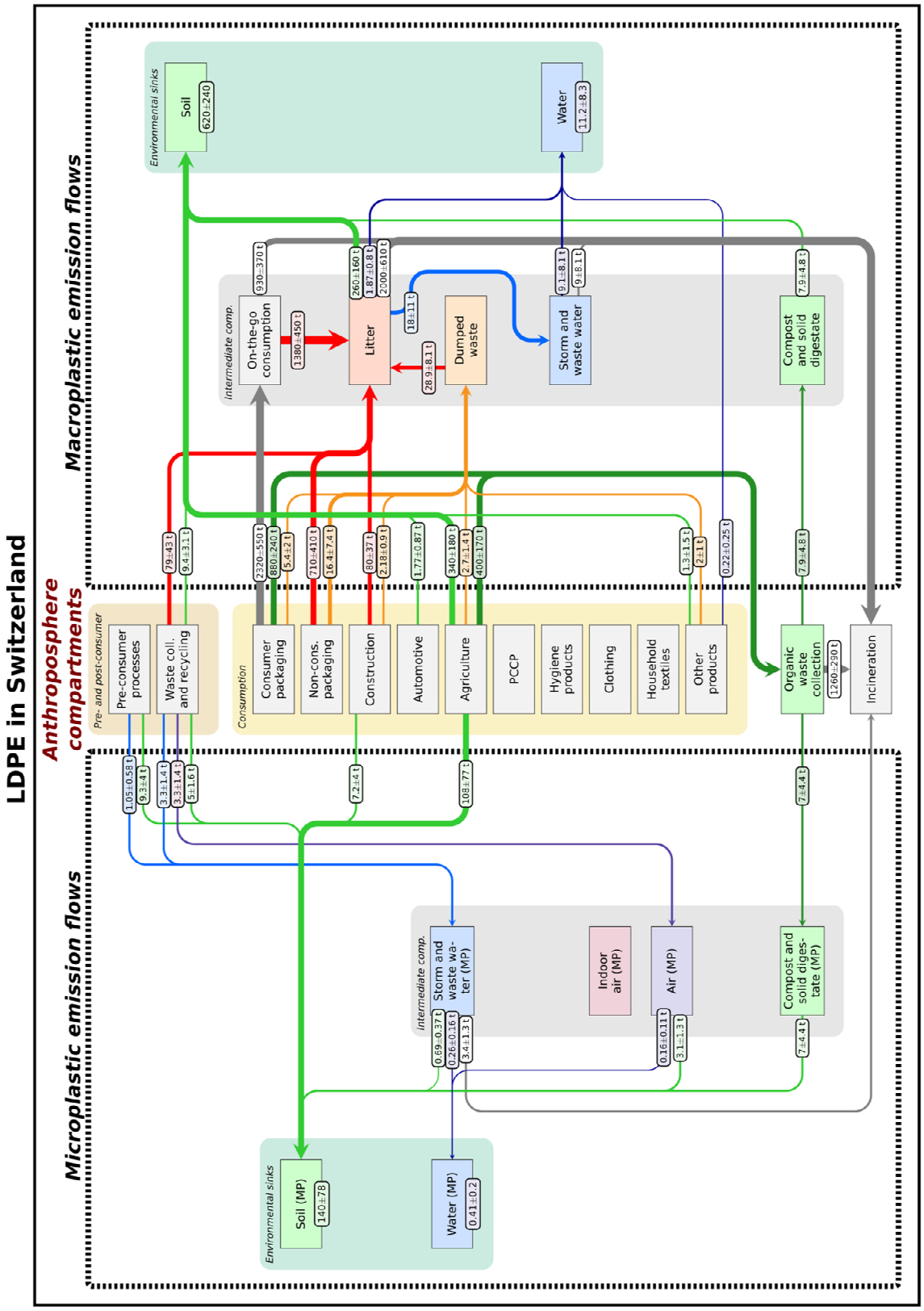

Figure S56: Aggregated emission flows in tonnes/year for LDPE in Switzerland for 2014. The colour of the flows is representative of the receiving compartment, and the width of the flows qualitatively represents its magnitude. Dashed arrows represent the flows between the different life cycle stages for which no data is shown. Abbreviations: waste collection (waste coll.), non-consumer packaging (non-cons. packaging), personal care and cosmetic product (PCCP).

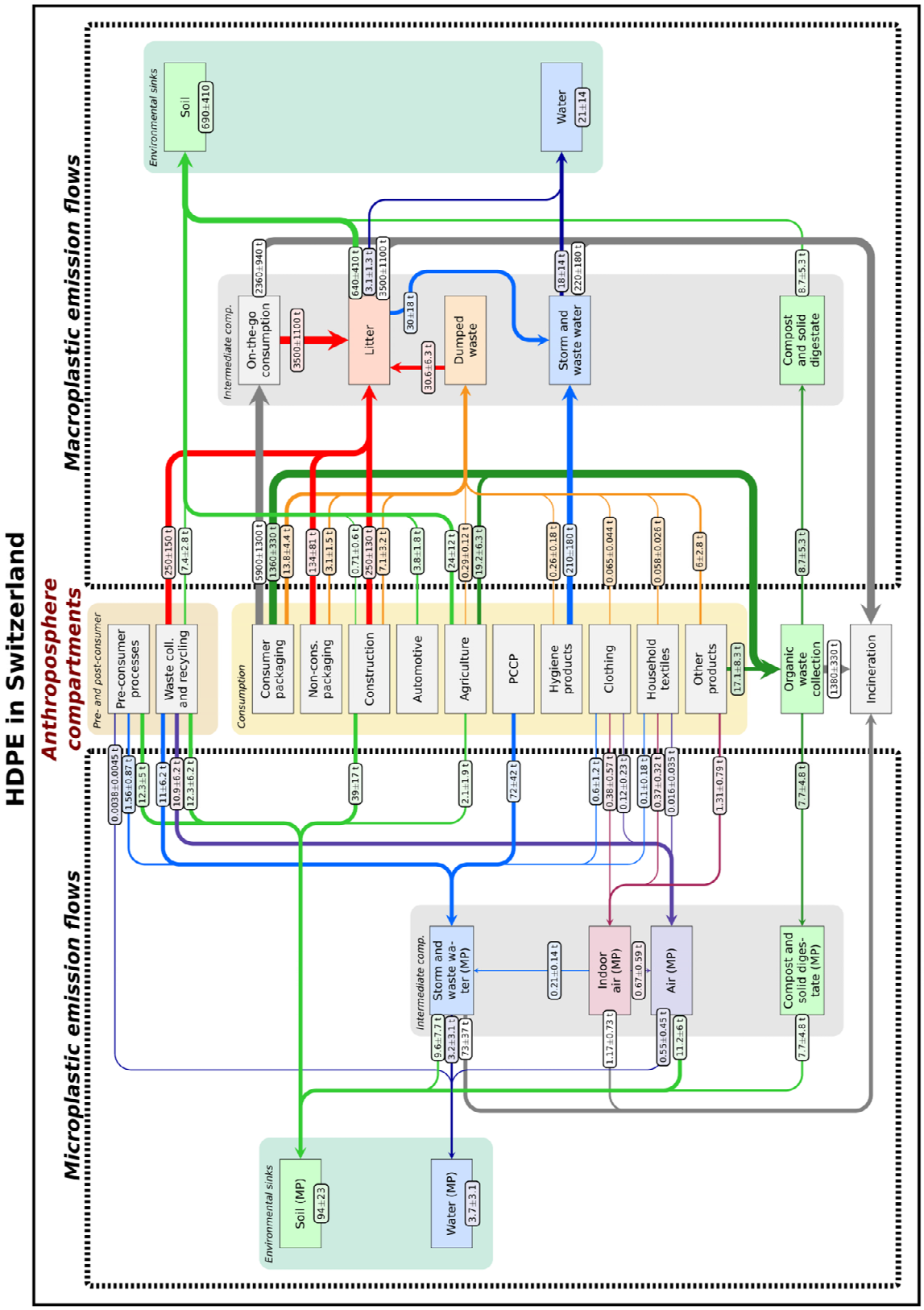

Figure S57: Aggregated emission flows in tonnes/ year for HDPE in Switzerland for 2014. The colour of the flows is representative of the receiving compartment, and the width of the flows qualitatively represents its magnitude. Dashed arrows represent the flows between the different life cycle stages for which no data is shown. Abbreviations: waste collection (waste coll.), non-consumer packaging (non-cons. packaging), personal care and cosmetic product (PCCP).

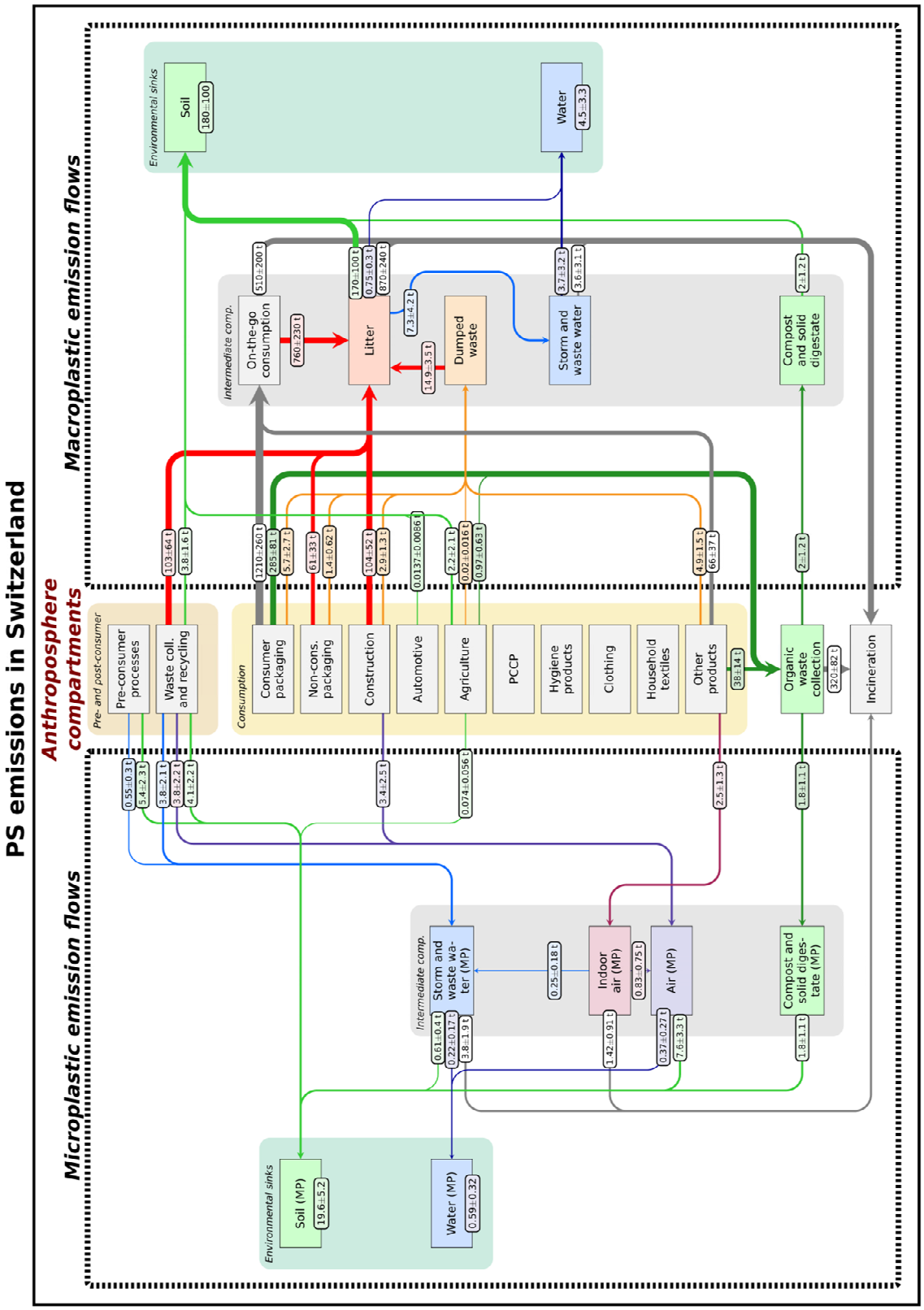

Figure S58: Aggregated emission flows in tonnes/year for PS in Switzerland for 2014. The colour of the flows is representative of the receiving compartment, and the width of the flows qualitatively represents its magnitude. Dashed arrows represent the flows between the different life cycle stages for which no data is shown. Abbreviations: waste collection (waste coll.), non-consumer packaging (non-cons. packaging), personal care and cosmetic product (PCCP).

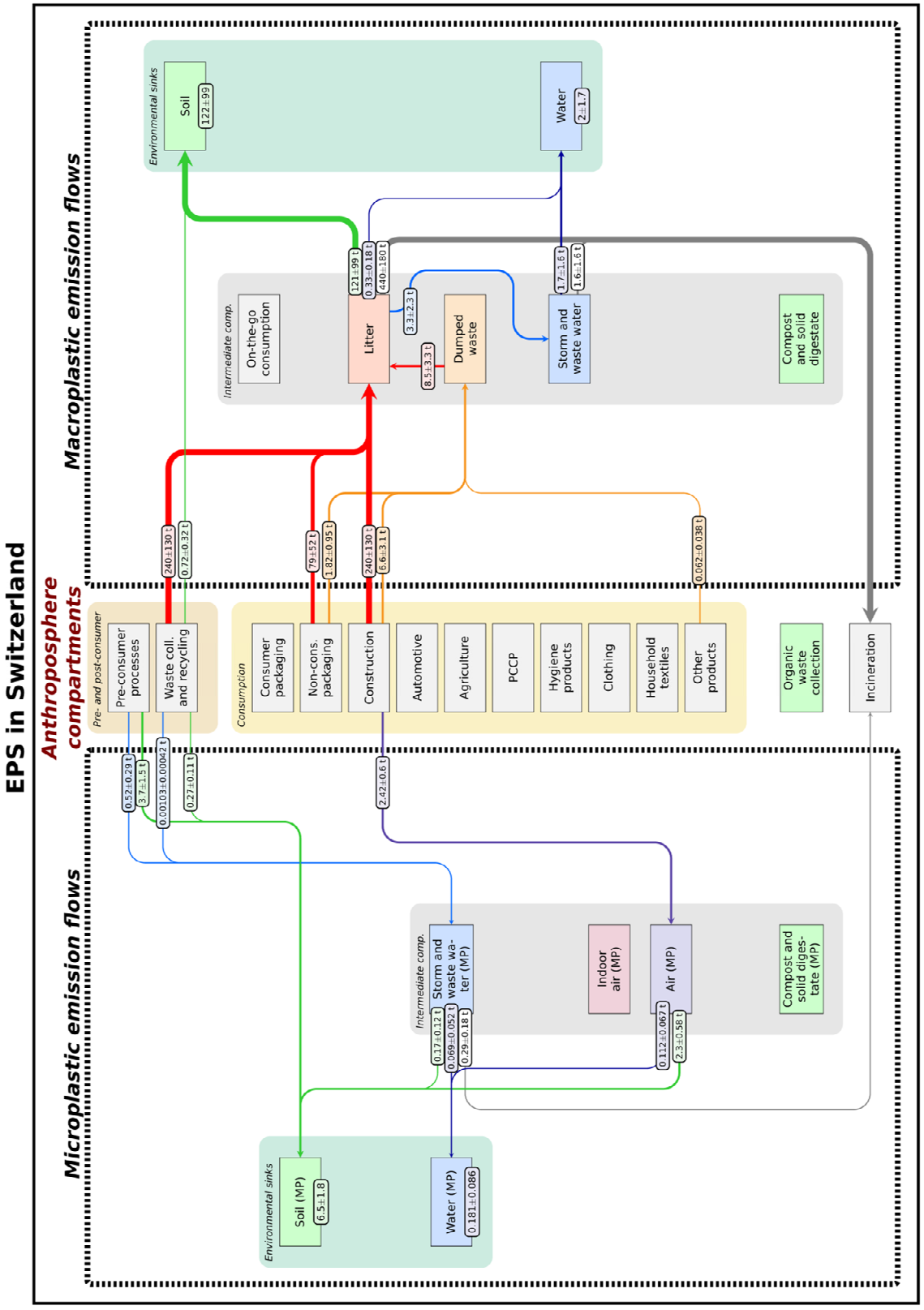

Figure S59: Aggregated emission flows in tonnes/year for EPS in Switzerland for 2014. The colour of the flows is representative of the receiving compartment, and the width of the flows qualitatively represents its magnitude. Dashed arrows represent the flows between the different life cycle stages for which no data is shown. Abbreviations: waste collection (waste coll.), non-consumer packaging (non-cons. packaging), personal care and cosmetic product (PCCP).

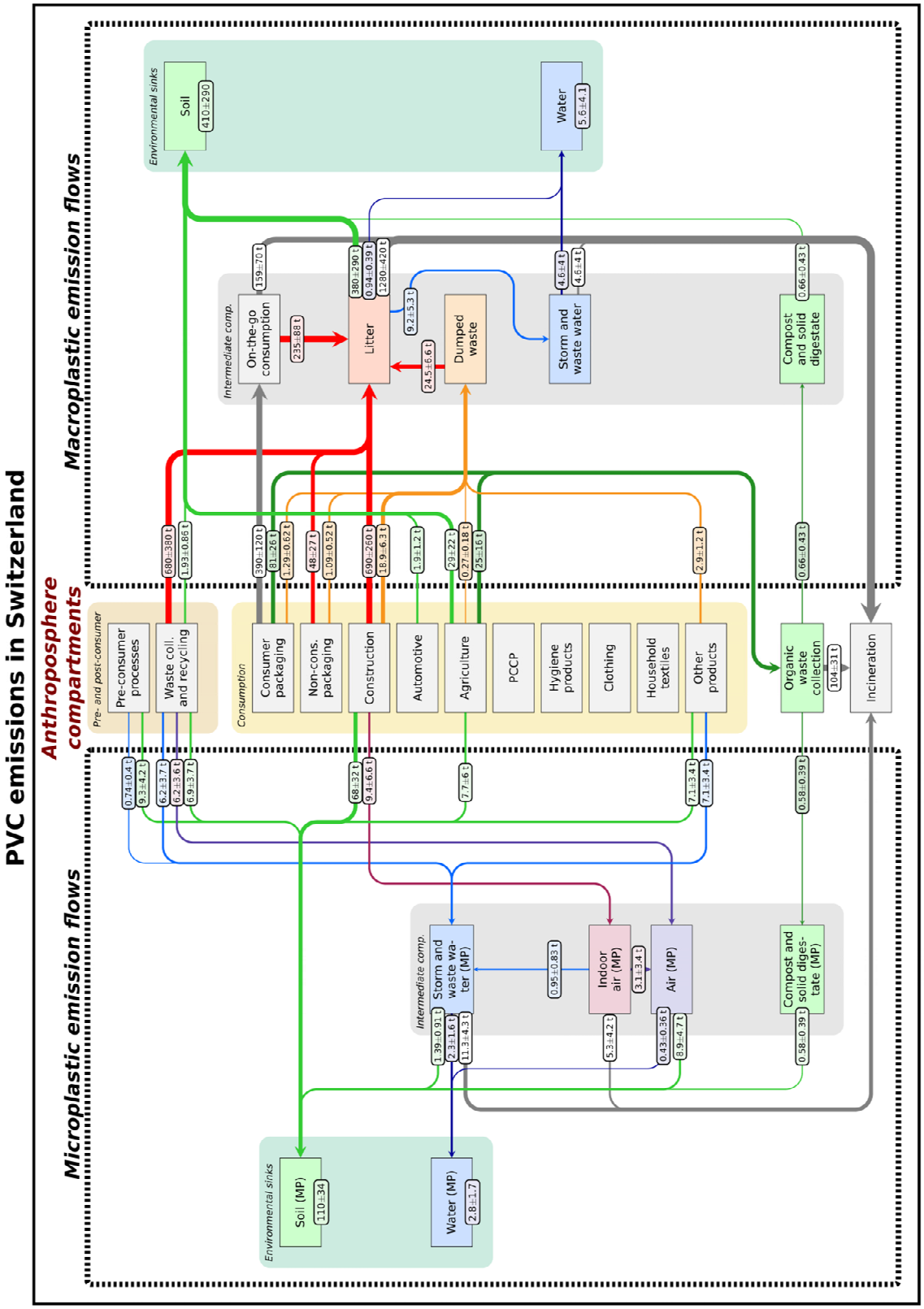

Figure S60: Aggregated emission flows in tonnes/year for PVC in Switzerland for 2014. The colour of the flows is representative of the receiving compartment, and the width of the flows qualitatively represents its magnitude. Dashed arrows represent the flows between the different life cycle stages for which no data is shown. Abbreviations: waste collection (waste coll.), non-consumer packaging (non-cons. packaging), personal care and cosmetic product (PCCP).

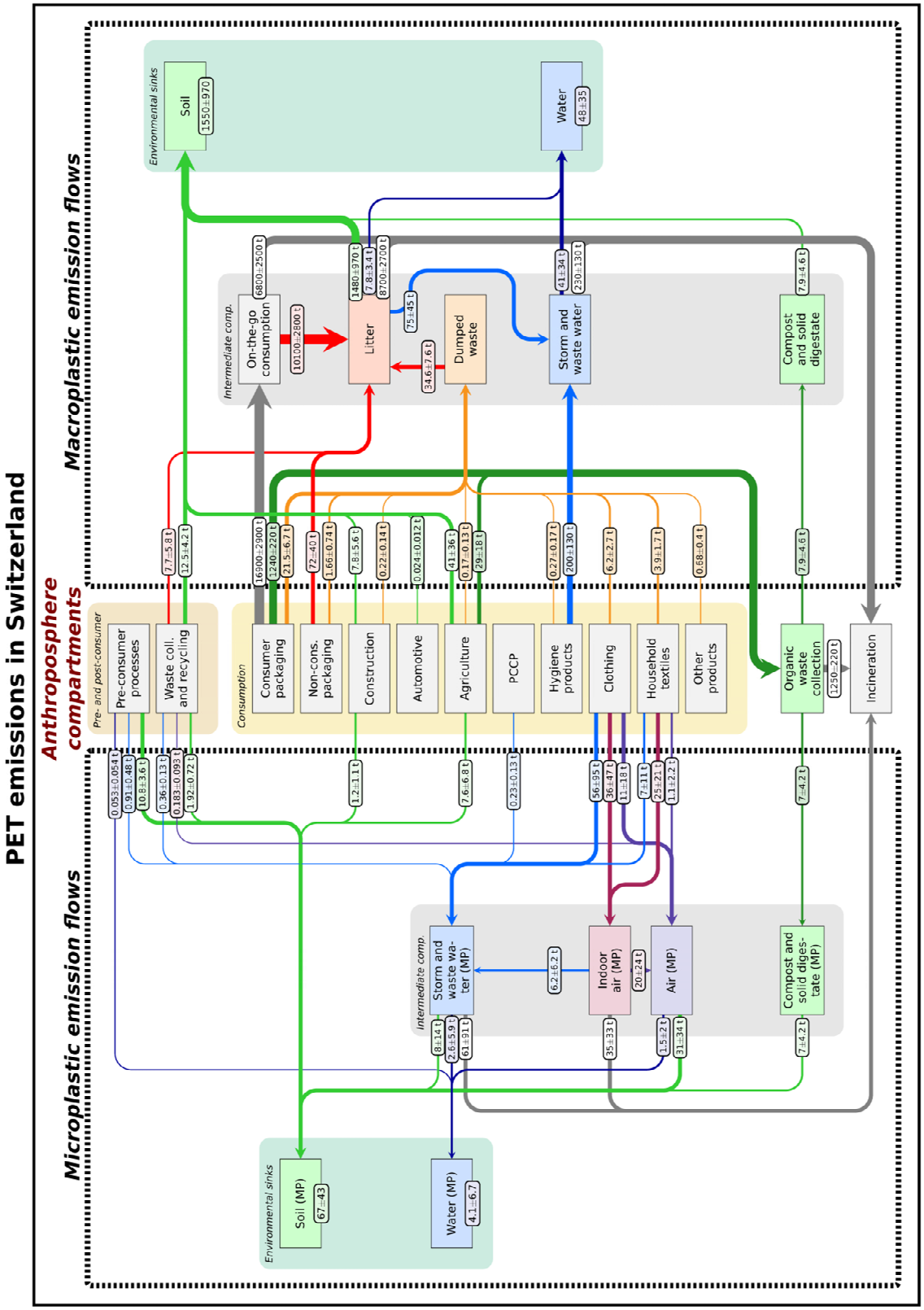

Figure S61: Aggregated emission flows in tonnes/year for PET in Switzerland for 2014. The colour of the flows is representative of the receiving compartment, and the width of the flows qualitatively represents its magnitude. Dashed arrows represent the flows between the different life cycle stages for which no data is shown. Abbreviations: waste collection (waste coll.), non-consumer packaging (non-cons. packaging), personal care and cosmetic product (PCCP).

11.2.2 Other additional figures

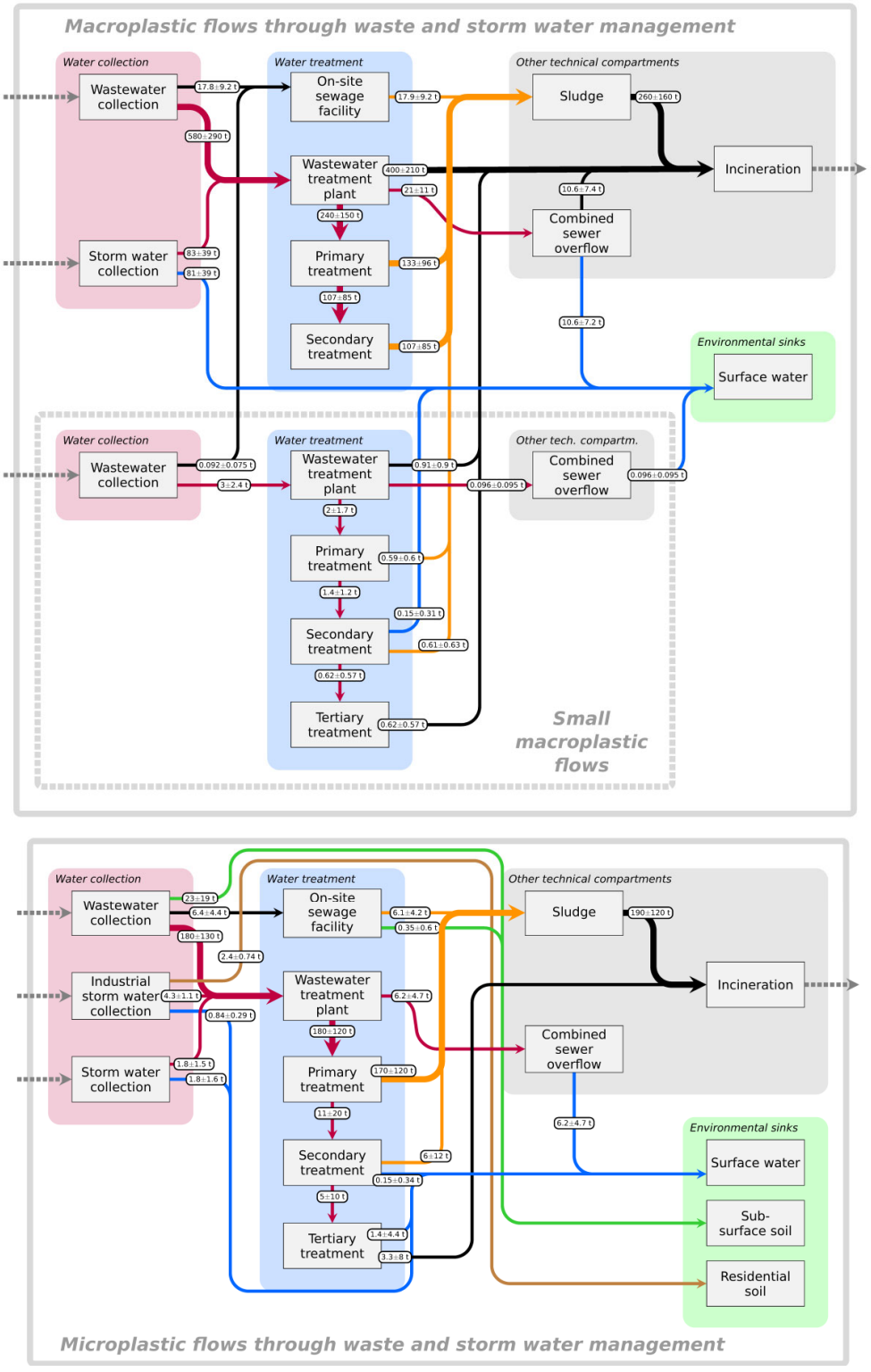

Figure S62: Detailed pathways for all flows connected to wastewater and storm water management, summed over all polymers in tonnes/year. The colour of the flows is representative of the receiving compartment. Dashed flows represent flows for which the data is not represented, but that exist in the model. A distinction is made between three sizes of plastic: large macroplastic, small macroplastic and MP.

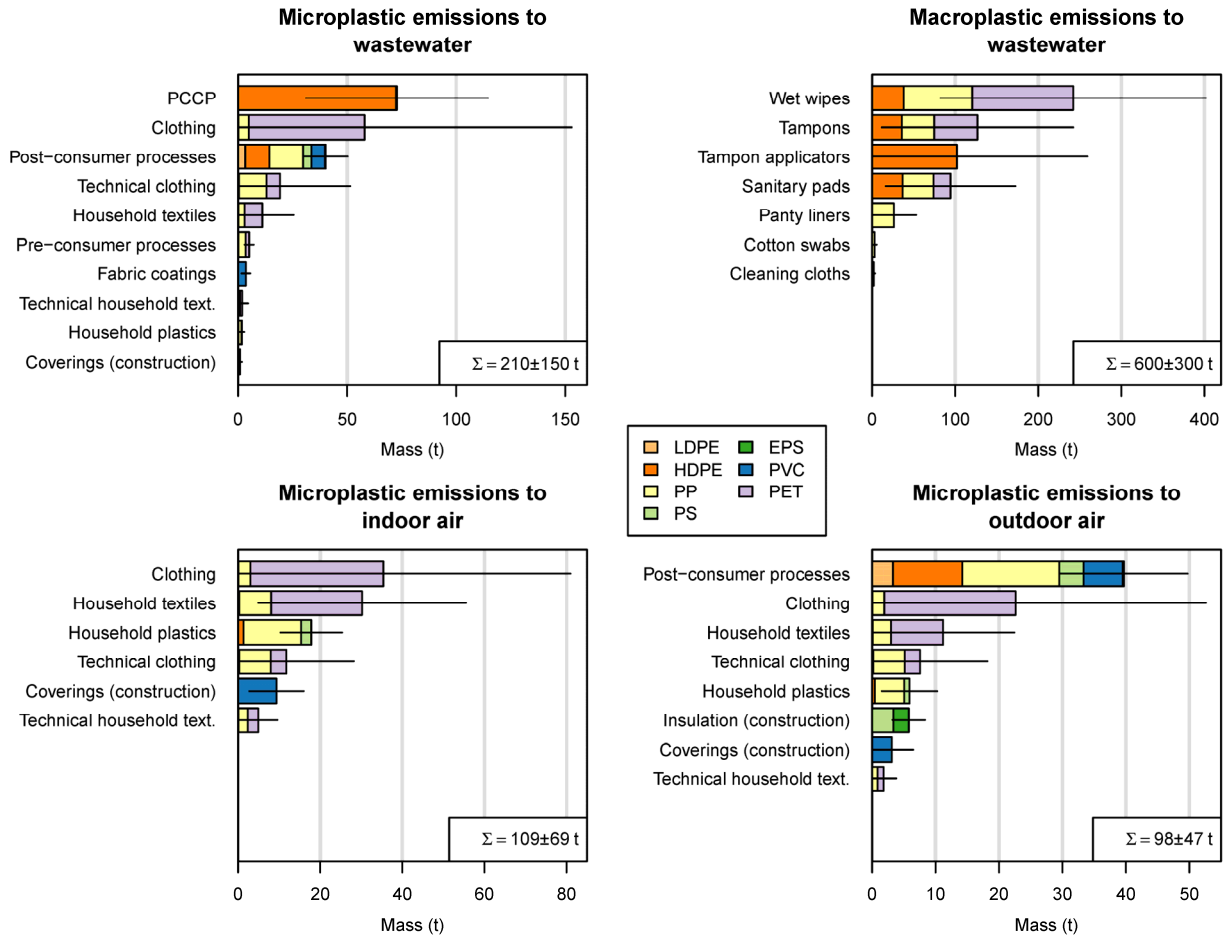

Figure S63: Products responsible for the emissions to indoor air, outdoor air, and wastewater. Only MP is shown for indoor and outdoor air. These two quantities are not independent since a fraction of the burden in indoor air is further transmitted to outdoor air. For plastic emissions to wastewater, MP and macroplastic emissions are shown. These emitted quantities exclude the emissions from the pre- and post-consumer processes.

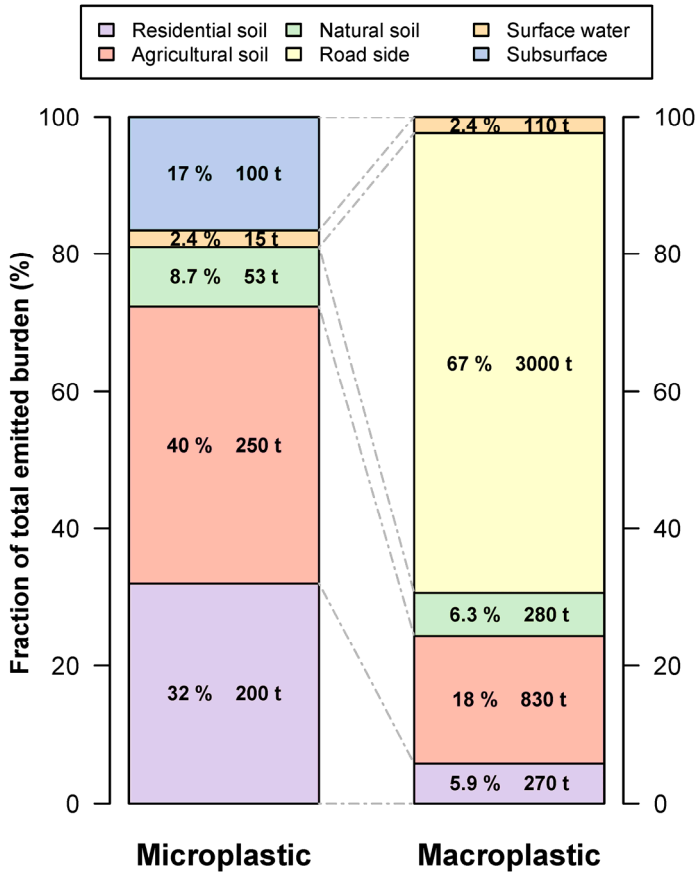

Figure S64: Comparison of the burdens of each environmental compartment for both MP and macroplastic emissions.

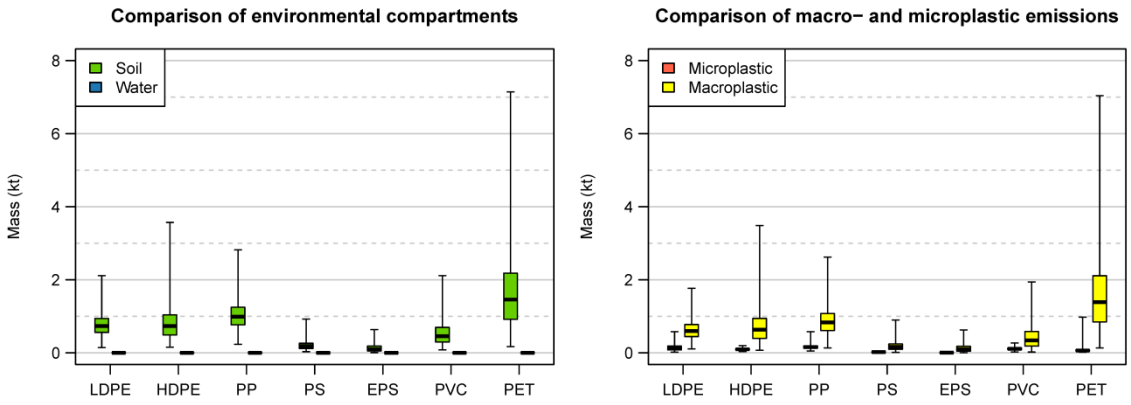

Figure S65: Box and whisker plots of the aggregated masses of emitted plastic into soil and water (A) and of the aggregated masses of emitted MP and macroplastic (B). The ends of the whiskers extend to the extrema of the probability distributions.

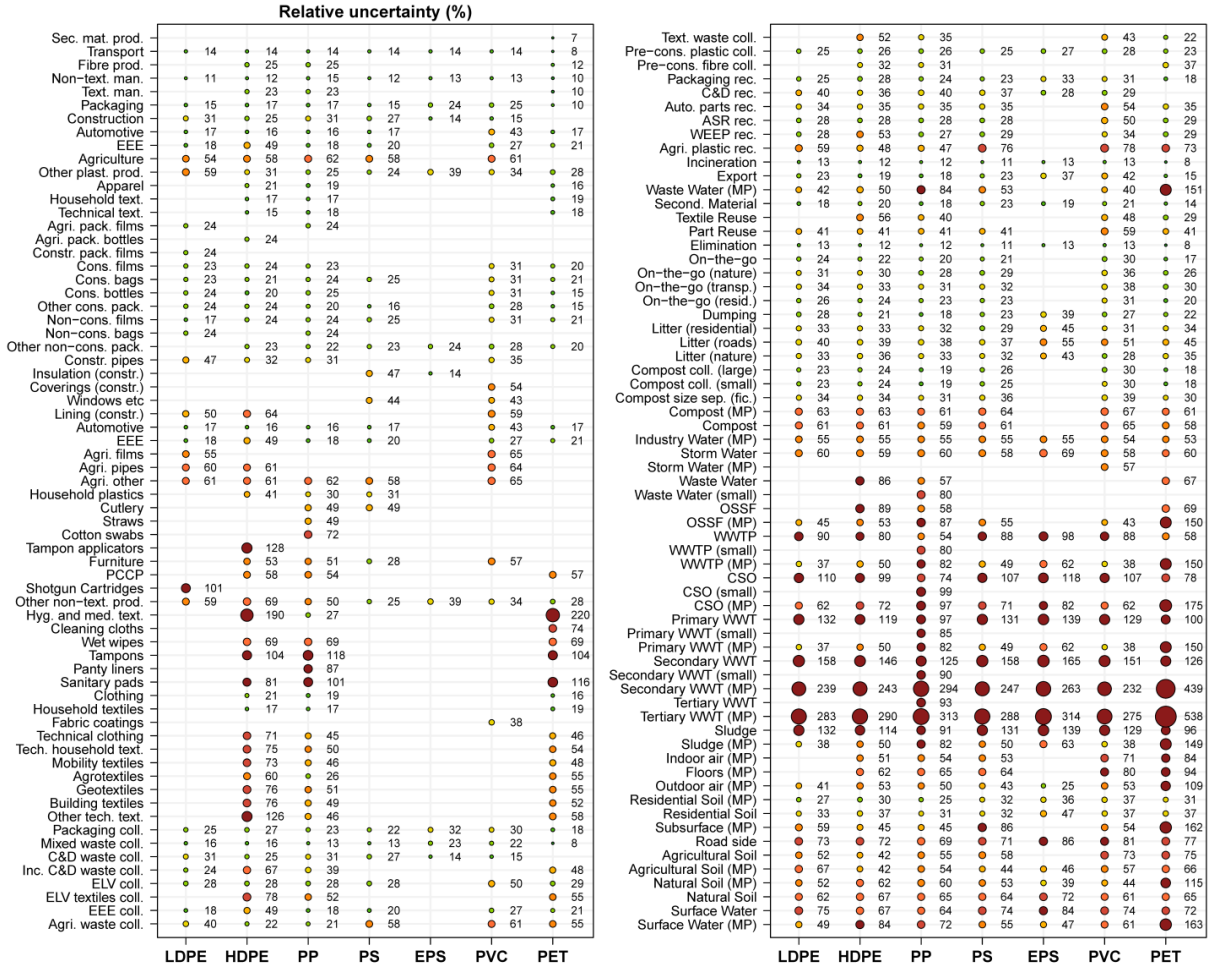

Figure S66: Overview of the relative uncertainty on the mass contained in each compartment in the system. The relative uncertainty is calculated by dividing the standard deviation of the distribution by its mean, and is displayed in percent.

11.3 Description of the emission flows

11.3.1 Overview of the modelled emission flows

Detailed flowcharts are presented in this section to give an overview of the flows included in the model. Table S18 also presents an overview of all emission flows modelled by size of plastic (macroplastic or MP) and type of emission (direct or indirect).

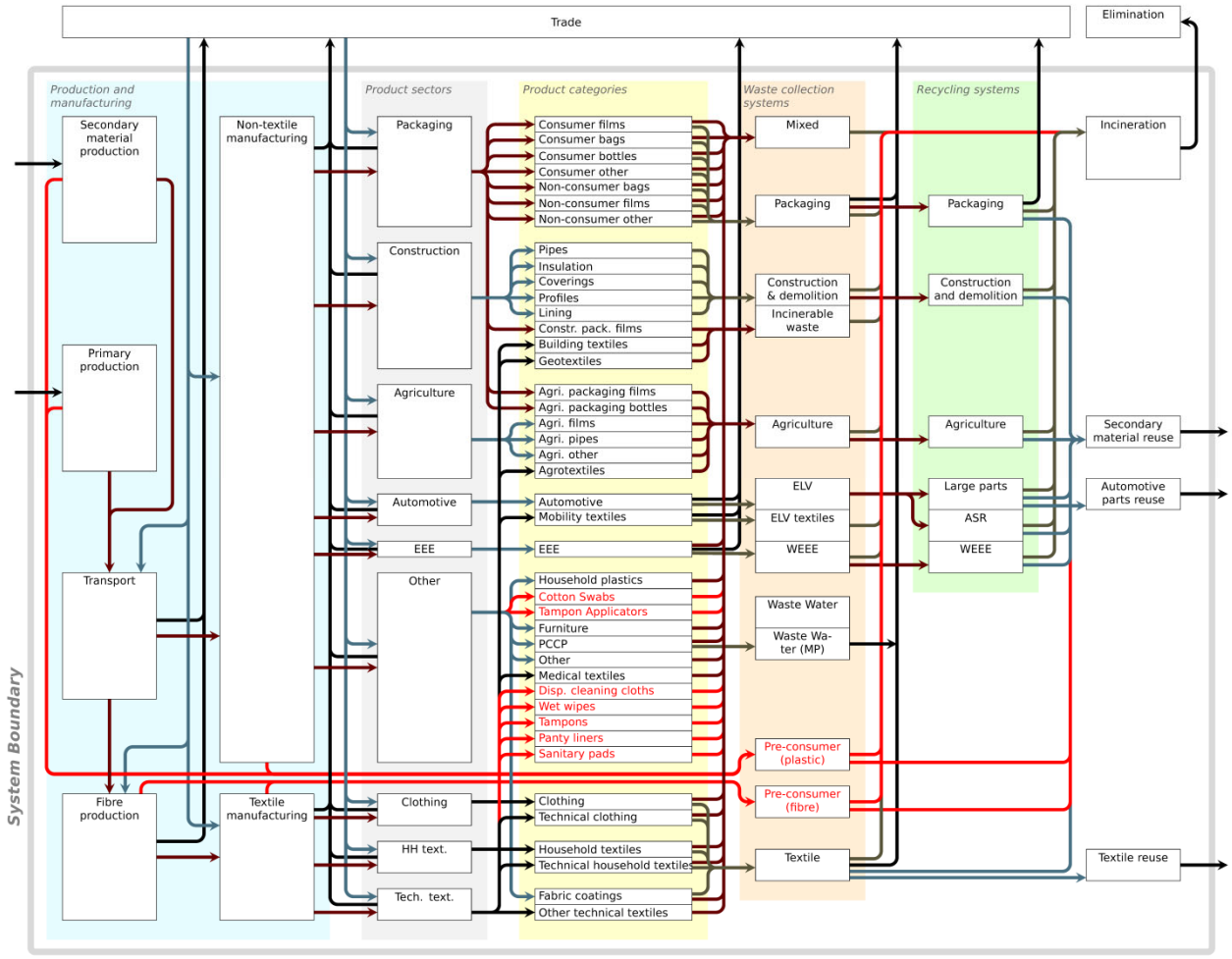

Figure S67: Overview of the flows modelled as part of the life-cycle. Flows and compartments shown in red are flows which were added or modified compared to the previous study on the flows in the anthroposphere.

Table S18: Overview of the release pathways from each compartment modelled in the life-cycle. Emissions are split into direct and indirect emissions, and into emissions occurring as MP or macroplastic.

Process or product category	MP		Macroplastic	
	Direct emissions	Indirect emissions	Direct emissions	Indirect emissions
Virgin material production **Recycled material production** **Non-textile manufacturing** **Fibre production** **Pellet and plastic waste collection** **Packaging recycling** **Agricultural plastic recycling** **Construction and demolition plastic recycling**	Pellets, flakes and dust to residential soil	Pellets, flakes and dust to industrial storm water	-	-
Transport	Pellets to residential soil	Pellets to industrial storm water	-	-
Textile manufacturing	-	Fibres to wastewater	-	-
Fibre and textile waste collection	Fibres to residential soil	Fibres to wastewater	-	-
Packaging collection **Mixed waste collection** **Agricultural plastic collection** **Textile collection**	-	-	Waste to residential soil	-
Construction and demolition plastic collection **Construction and demolition incinerable collection**	-	-	-	Waste to road sides as litter
ELV collection **ELV textiles collection** **WEEE collection** **Mobility textiles**	-	-	-	-
ASR recycling **WEEP recycling**	Dust to residential soil and outdoor air	Dust to wastewater	-	-
Consumer films **Consumer bags** **Consumer bottles** **Other consumer packaging** **Straws** **Cutlery**	-	-	-	Littering Dumping Collected with organic waste
Construction packaging films **Non-consumer bags** **Non-consumer bottles** **Other non-consumer films** **Windows, profiles and fitted furniture** **Lining**	-	-	-	Dumping Release to residential litter
Pipes	Wear to residential soil and subsurface soil	-	-	Dumping Release to residential litter
Insulation	Dust and particles to outdoor air during construction	-	-	Dumping Release to residential litter
Coverings	Wear to indoor air	-	-	Dumping Release to residential litter
Geotextiles	Wear to subsurface soil	-	Release to residential soil	Dumping
Building textiles	-	-	Release to residential soil	Dumping
Automotive	-	-	Release to road sides	-
EEE	-	-	-	Dumping
Agricultural packaging films **Agricultural packaging bottles**	-	-	Burying in agricultural soil	Dumping Collected with organic waste
Agricultural films **Agricultural pipes** **Other agricultural plastics** **Agrotextiles**	Wear to agricultural soil	-	Burying in agricultural soil	Dumping Collected with organic waste
Household plastics	Wear to indoor air	-	-	Dumping Collected with organic waste
Cotton swabs **Tampon applicators** **Disposable cleaning cloths** **Wet wipes** **Tampons** **Panty liners** **Sanitary pads**	-	-	-	Dumping Flushing
Fabric coatings	Wear to residential and natural soil	Wear to wastewater and storm water collection	-	Dumping
Shotgun cartridges	-	-	Release to surface water and natural soil	Dumping
Clothing **Technical clothing** **Household textiles** **Technical household textiles**	Wear to outdoor and indoor air	Wear to wastewater collection	-	Dumping
Furniture **Other technical textiles** **Other plastic products**	-	-	-	Dumping

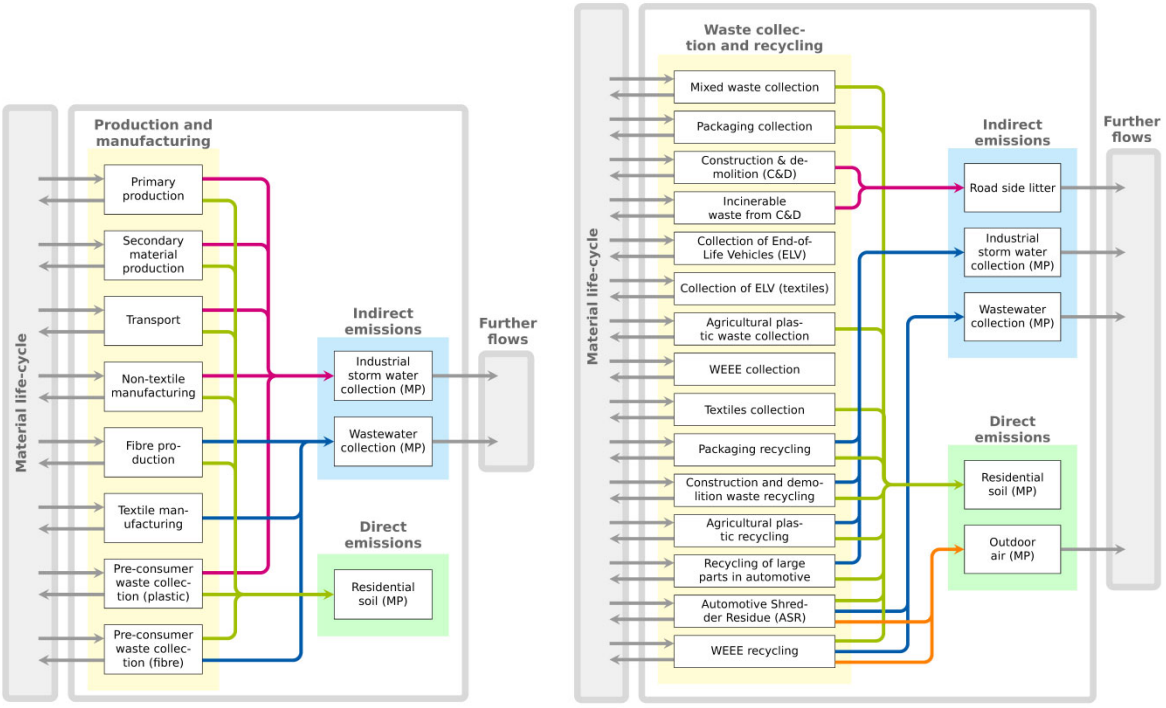

Figure S68: Emission flows from production and manufacturing (left) and waste collection and recycling (right).

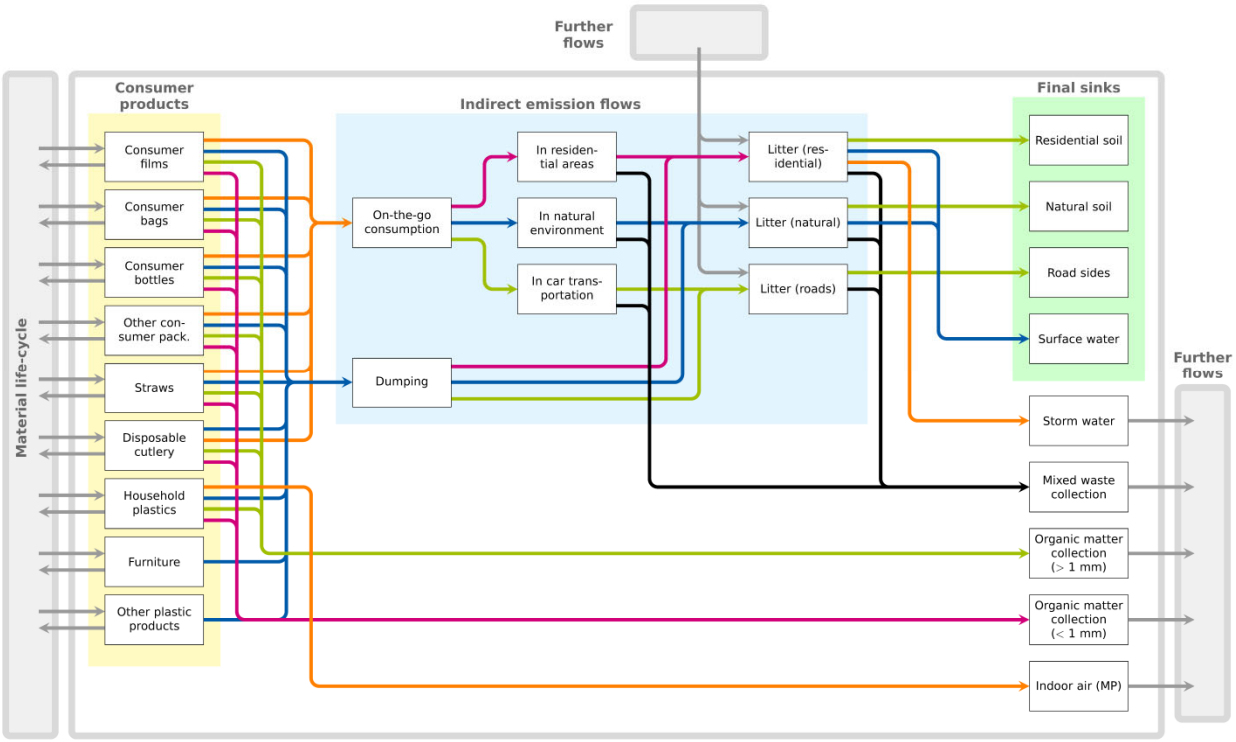

Figure S69: Emission flows from consumer products subject to littering and collected with organic matter.

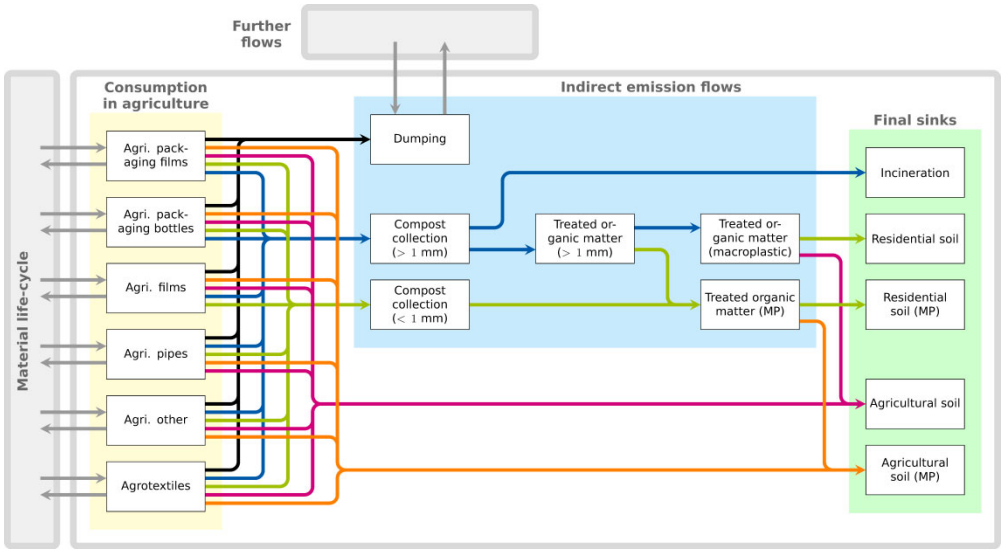

Figure S70: Emission flows from agricultural products.

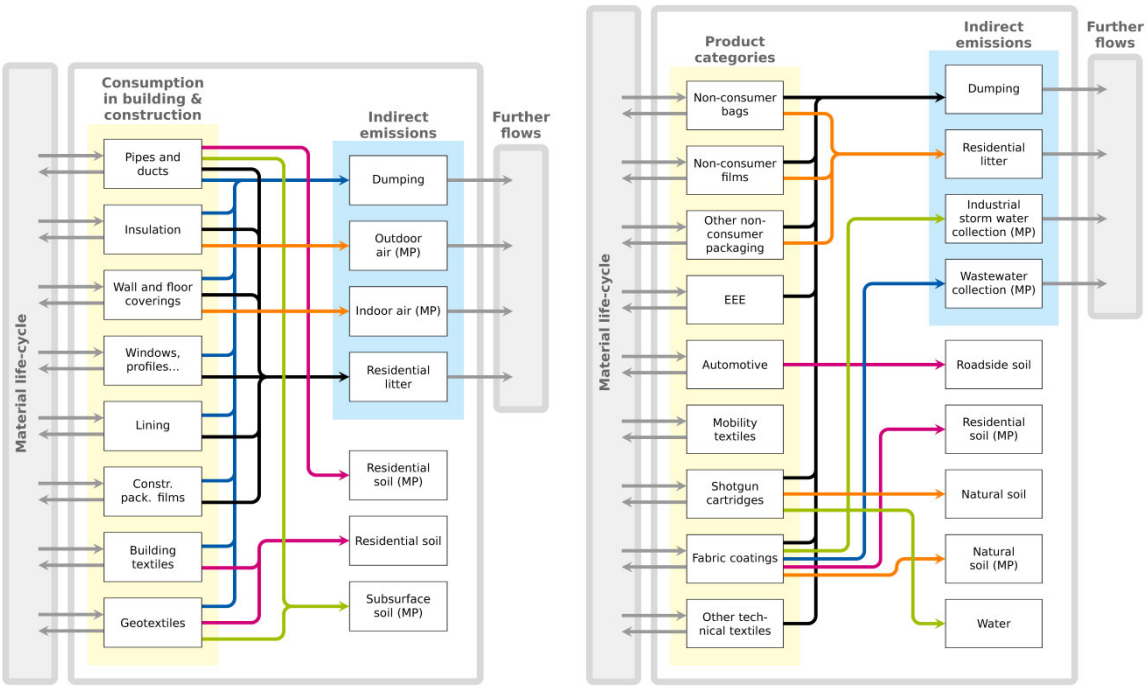

Figure S71: Emission flows from building and construction products (left) and other product categories (right).

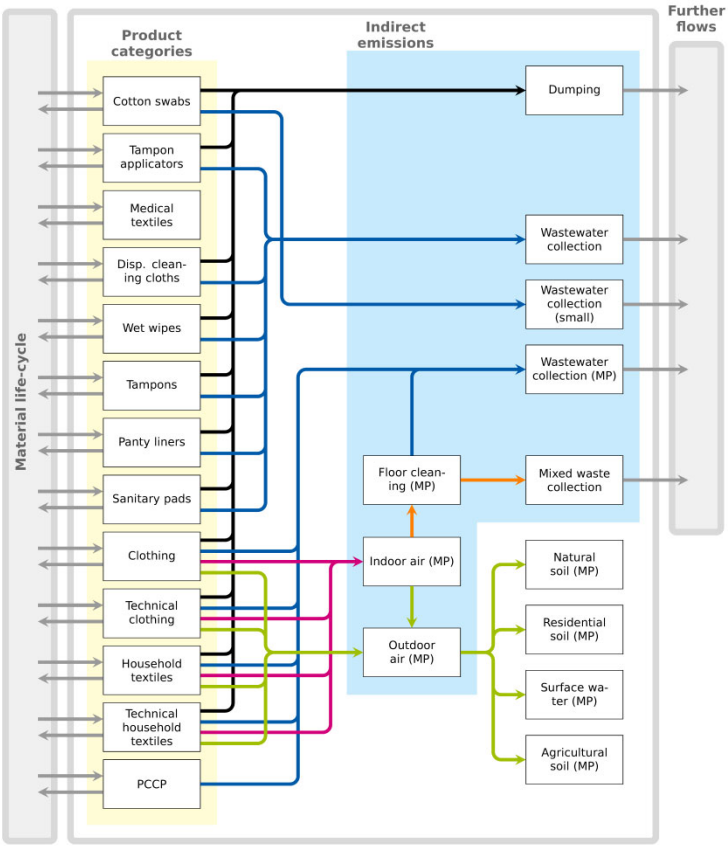

Figure S72: Emission flows from consumer products which may be released to wastewater. The modelled flows through wastewater treatment are shown in Figure S62.

11.3.2 Littering

Littering is defined as the discarding of a product outside of a waste bin, at the moment and place where the product reaches its end-of-life. Littering differs from dumping, as the primary motivation for dumping is to avoid the constraint of appropriate disposal which can be costly or time consuming, whereas littering is a spontaneous decision. The composition of waste littered differs from the composition of waste dumped. Littered waste is composed of waste generated on-the-go, whereas dumped waste is generated away from the disposal site.

In order to model the littering process, it is split into several steps:

1. First the fraction of products at risk consumed on-the-go is estimated, because goods need to be used outside of homes or offices in order to be littered outdoors.
2. These products used on-the-go are further separated into three types of on-the-go consumption: in transportation, natural and residential areas. This step is necessary for

later steps of the littering modelling which are dependent on the location of the littering.

3. Then, a littering probability describing the littering behaviour for an item reaching its end-of-life comes into play. This probability is estimated separately for a use in cars, in natural and in residential areas. The fraction not littered is collected as mixed waste.
4. Finally, the littered fraction is either swept or collected by municipalities. This fraction also depends on the location of the process.

11.3.2.1 Inclusion of additional product categories for the description of littering

No information on the consumption of plastic straws and plastic cutlery is included in the previous study[1]. In order to include them in this assessment, a first estimation of their consumption needs to be performed. The annual consumption of straws in the EU28 is equal to 36.4 billion units[6]. Using the European and Swiss population, we estimate that 587 million straws are used yearly in Switzerland. In order to estimate the composition and average mass of a straw, 14 different types of plastic straws available for purchase online were considered. Only plastic straws for which the total mass of the product and number of straws was given were considered. Based on this survey, a straw weighs on average 0.7g. All of these products were plastic straws. The polymer composition was not mentioned in all instances, but when it was, the straws were composed of PP. We therefore assume that PP is the material of choice for the manufacturing of plastic straws. It can therefore be concluded that 411 tonnes of PP straws are consumed yearly in Switzerland.

In a survey conducted in several municipalities in Switzerland, 1134 g of cutlery were found, compared to 29025 g of PET bottles[7]. Using this information, we estimate the consumption of cutlery based on the amount of PET bottles litter arising next to roads, in residential and natural areas and knowing the transfer coefficients from on-the-go consumption to litter. The composition of the cutlery is based on the availability of products to purchase online. On the first website, 1399 entries corresponded to PP and 1072 to PS. On the second website, 6055 entries corresponded to PP and 6193 to PS. Taking the average of the two gives us 53% PP and 47% PS.

11.3.2.2 On-the-go consumption of products at risk

The fraction of packaging consumed on-the-go can be found in a report on the plastic packaging composition for the UK[8] (Table S19), from which the packaging product categories (PC)

were also derived. This fraction is different for different product categories and materials. The non-consumer product categories are not shown, as they are not consumed on-the-go.

Table S19: Fraction of packaging categories that is consumed on the go[8] for the chosen packaging PC.

Product category	LDPE	HDPE	PP	PS	EPS	PVC	PET
Consumer films	0.1017	0.2857	0.1884	0.2000	0.2000	0.2000	0.2000
Consumer bags	0.1395	0.2411	0.1750	0.2222	0.2222	0.0000	0.2222
Consumer bottles	0.0000	0.0000	0.0000	0.3544	0.3544	0.0000	0.3544
Other consumer packaging	0.0000	0.1176	0.0500	0.0550	0.0550	0.0750	0.0823

For straws and cutlery, the fraction consumed on-the-go is estimated using the share of on-the-go consumption out of all consumer packaging in the UK[8]: 16.18%.

11.3.2.3 Repartition of on-the-go consumption in transportation, natural and residential areas

A distinction is made between the on-the-go consumption in residential and natural environments as well as littering from vehicles. The fraction of on-the-go products consumed in vehicles is estimated based on the fraction of fast-food restaurants equipped with a drive-in in Switzerland[9]. Information on market shares of goods sold through drive-ins could not be obtained from fast-food restaurants, so the fraction of restaurants equipped is used as a proxy. The remaining fraction of on-the-go products is then consumed either in residential or natural environments. The distinction between both is made considering the frequency of hiking, going on an excursion or picnicking outdoors. Of all Swiss inhabitants, 39% of Swiss inhabitants go hiking, on an excursion or picnicking outdoors at least once a week, 33% go 1-3 times per month, 25% occasionally, and the remaining 4% never go[10]. Assuming that 4 times a year is representative of the term "occasionally", we obtain that the average Swiss goes hiking or picnicking outdoors 29.2 times a year. Since the median duration of a hike is 3 hours[11] and assuming this duration is representative of a picnic or excursion, we obtain the average Swiss spends 87.6 hours outside of residential areas during one year. Knowing that 7% of a person's time is spent outdoors[12], we obtain that a person spends on average 613 hours outdoors. Comparing this value with the previously mentioned duration, we obtain that the average Swiss inhabitant spends 14% of his time outdoors away from cities.

11.3.2.4 Chosen littering probability distribution

The littering rate varies drastically with location and context[13–21]. It is therefore fairly difficult to estimate a littering probability distribution based on these available numbers without knowing what weight to attribute them. For residential areas, the littering rates reported for two cities in Switzerland are considered, where the quantity of swept litter compared to the quantity of waste contained in bins arising during one year was studied (Table S20).

Table S20: Littering rates derived from city data.

City	Total waste collected in public areas	Total litter collected	Waste from households discarded in public bins	Littering rate (after removal of household waste)
Geneva[22]	5'000 tonnes	4'320 tonnes	-	85 %
Bern[23]	4'000 tonnes	1'600 tonnes	800 tonnes	50 %

The listed littering rates do not include litter that would not have been swept by the authorities, but considering how high these littering rates are, and how efficient the cleaning is thought to be[23], we consider this to be of no influence. Since these littering rates were measured in urban settings, a correction factor is applied to derive the littering rate for transportation. Schutz *et al*[15] reported littering rates of 23% in urban, 18% in suburban and 15% in rural locations, with an average of 17% over all locations. We use a scaling factor of $A_{\text{transport}} = \frac{17\%}{0.5 \cdot (23\% + 18\%)} = 0.83$ for transportation assuming it is midway between urban and rural. For natural environments, the previously mentioned littering rate of 15% in rural areas is used.

11.3.2.5 Sweeping efficiency

In residential areas in Switzerland, sweeping is performed according to the needs. The collection of litter in central locations more prone to littering occurs several times a day, while cleaning may only occur once a week in suburban areas. It is generally considered that almost all of the litter is collected on streets. However, this is only valid for accessible waste. Litter which has been hidden on purpose or discarded in waterways will not be collected on a regular basis. In the study from Schultz *et al.*[15] 1.2% of the littered items were wedged away into small spaces where they will no longer be seen. We assume that this fraction remains on residential soil, however good the sweeping may be. The same study also reports that around 0.1% of the littered items were directly discarded into rivers or lakes, which gives us the amount of litter flowing to surface water. 1% of the litter in residential areas is modelled to flow to storm water,

which would be a worst case scenario as expected from the city cleaning authorities. The rest is modelled to be collected and sent to incineration.

The situation is similar for road sides: the sweeping frequency depends on the estimated need. Highways entrances and exits are more often cleaned than other parts of the road network. The fraction of the litter collected is difficult to estimate, so we describe it using a best case and worst case scenario: between 10% and 90%.

Similarly as for the residential litter, 0.1% of the litter in natural environments is modelled to be discarded in surface water. The sweeping distribution is similarly described with a distribution between 10 and 90%. The rest remains on soil.

11.3.3 Dumping

Dumping is the premeditated release of waste to the environment with the aim of avoiding the constraints associated with properly disposing of waste. In most cantons in Switzerland, there is a fee for each waste bag collected, and waste can be dumped or discarded in public bins to avoid this cost. In order to give an order of magnitude of the amount dumped in Switzerland, we assume that it is 100 times more likely for a person to discard their own waste in public bins than to dump it elsewhere. Knowing that 28'734 tonnes of household waste were generated in 2014 in the city of Bern[24] and that 800 tonnes of household waste were found in public bins (Table S20), we conclude that 0.027 % of the waste may be dumped outdoors.

Dumping may occur in different settings. The type of location of dumping events has been published for reported waste in England[25]. Assuming the dumping behaviour is similar in Switzerland and England, we can reuse the repartition between settings to redistribute dumped waste to different compartments Table S21). It results that 16% of the dumped waste is dumped in residential areas, 51% on roads and 33% in natural settings.

Table S21: Location of reported dumped waste in England in 2014-2015[25] and allocation to the compartments modelled in this study.

Land type	Dumping incidents	Corresponding compartment in this study	Proportion of waste dumped (%)
Highway	440'000	Road side litter	51
Council land	150'000	Natural soil	17
Footpath/bridleway	140'000	Natural soil	16
Back Alleyway	110'000	Residential soil	13
Other	30'000	-	-
Private/residential	20'000	Residential soil	2
Commercial/industrial	10'000	Residential soil	1

11.3.4 Flushing

Flushing is another important pathway for inappropriate disposal of products. During the September beach cleaning campaign of the Marine Conservation Society made in 2015 in the UK, 6.3% of the items found were sewage related[26]. Cotton swabs were the 6th items found most frequently on British beaches[26] and similarly the 5th most frequent item on Swiss freshwater beaches[27]. Between 2013 and 2014, a 50% increase in the number of wet wipes found on British beaches was observed, and between 2014 and 2015, a further 31% increase was observed[26]. These numbers highlight the prevalence of sanitary products in litter. Parts of this litter may reach the environment through CSOs, and parts through WWTPs. Cotton swabs in particular may cross WWTPs in some instances, as was already suggested in 1990 (Huntingdon 1990 as cited by Williams et al.[28]) and 1996[29].

The procedure for modelling the flushing process is split into several steps:

1. Identification of the products at risk
2. Estimation of their material composition
3. Calculation of their consumption
4. Calculation of flushing probabilities for each product
5. Identification of the retention efficiency of WWTP and CSOs

11.3.4.1 Identification of the products at risk

Most products that can end up being discarded through flushing are products related to personal care and hygiene. Friedler et al. [29] undertook an assessment of the frequency and composition of waste discarded in private WCs based on a survey completed by 137 households

during one week in the UK in July 1994. Their results show that the products most often flushed are tampons (22.9% of the items flushed in their study), wet wipes (14.4%), tissue paper (11.8%), cat faeces (7.1%), dental floss (4.4%) and 24 more other categories. The items reported which may contain plastic are shown in Table S22 with their flushing frequencies.

Table S22: Percentage of households reporting the flushing of items in the WC for selected items[29]. Only the products which may contain plastic are shown.

Item	Households reporting flushing of item (%)	Attribution to product category
Tampon	10.9	Hygiene and medical textiles
Wet wipes	5.8	Hygiene and medical textiles
Other - non specified	5.1	Unknown
Dental floss	3.6	Hygiene and medical textiles
Panty liner	2.9	Hygiene and medical textiles
Tampon applicator	2.9	Hygiene and medical plastics
Cotton swabs	2.2	Hygiene and medical plastics
Bandage	0.7	Hygiene and medical textiles
Disposable cleaning cloth	0.7	Technical household textiles
Sanitary pads	0	Hygiene and medical textiles

In a more recent study, Spence et al.[30] have sampled the gross solids incoming from the sewers of several neighbourhoods in Sheffield in the UK using a mesh bag of mesh size 6*4 mm. After identification and weighing of the individual items sampled, they found between 0.019-0.126 wipes/pers/day, 0.010-0.035 tampons/pers/day, 0.003-0.016 panty liners/pers/day, 0.004-0.006 cotton swabs/pers/day, 0.002-0.019 sanitary pads/pers/day and finally 0.002-0.003 sanitary pad shells/pers/day depending on the catchment type. Other types of products may have been sampled but are not reported.

Based on these two papers, the considered products for the estimation of plastic emissions through flushing are:

1. Tampons
2. Wet wipes
3. Floss
4. Panty liners

5. Tampon applicators
6. Cotton swabs
7. Bandages
8. Disposable cleaning cloths
9. Sanitary pads

11.3.4.2 Estimation of the material composition of flushed products

Little literature is available on the topic of material composition of sanitary items. Manufacturers are often not willing to share quantitative information. To overcome the data scarcity for some of these products, a survey was performed in three retailers in Switzerland. From this information, the average material composition could be estimated (Table S23).

Table S23: Calculation of the composition of sanitary items.

Item	Composition	Item mass (g)	Sources and comments
Tampon	PET: 4-5 % HDPE: 2-4 % PP: 0-5 %	2.0-2.7 g	Two different tampons were taken apart, and each fraction was weighted. The composition of each component was given on the packaging or on the website of the brand. Tampon 1: - Absorbent core: 2.48 g (viscose) - Thin fabric around the core: 0.12 g (polyester, polyethylene) - String: 0.10 g (polyester, cotton) Tampon 2: - Absorbent core: 1.56 g (cotton, rayon) - Thin fabric around the core: 0.31g (rayon, polyester, polypropylene, polyethylene) - String: 0.08 g (cotton, polyester, polypropylene) We assume that the different materials stated per component are used equally in the component, in the absence of better data. What is stated as polyethylene is attributed to HDPE because it is fibre material.
Wet wipes	PET: 25 % PP: 17 % HDPE: 8 %	1.4-1.7 g	Estimated following the frequency of mention of different materials related to wet wipes in different web resources[31–33].
Dental floss	-	-	Floss is commonly made of Nylon or Teflon[34]. Further quantification was not necessary, since these materials are not considered here.
Panty liner	PP: 35 %	2 g	Using the same approach as for the tampons, the composition of the panty liners could be obtained based on the stated composition of one product and the masses of individual components as weighted in the laboratory. - Upper veil: 0.27 g (PP) - Absorber: 1.3 g (cellulose) - Bottom veil: 0.43 g (PP)
Tampon applicator	HDPE: 44 %	2.3 g	Estimated following the information given on the packaging of 14 available products. When the information was not available, the product was purchased in order to determine it in the lab. Of all tampons available, 9 were sold with applicator, of which 4 were made of plastic. Only 2 products stated the type of plastic of the plastic applicators, which was polyethylene.
Cotton swabs	PP: 73%	0.15 g	Out of 11 different products available from the retailers, 8 had a stick made of PP and 3 of compressed paper.
Bandage	-	-	The composition of bandages was very difficult to come by, as it is not stated on the packaging and no other resources were available. Since this

Item	Composition	Item mass (g)	Sources and comments
			product that is not most at risk (low flushing behaviour, low consumption), it was removed from our assessment.
Disposable cleaning cloth	PET: 20 %	7.7-13.5 g	Two different types of disposable cleaning cloths were purchased. Their stated consumption was: cellulose and EVA for the first product, and cellulose, acrylic and polyester for the second. We assume that the different materials are used in the absence of better data.
Sanitary pads	PET: 0-14 % PP: 11-21 % HDPE: 15-23 %	3.3-5.1 g	Two different sanitary pads were taken apart, and each fraction was weighted. The composition of each component was given on the packaging or on the website of the brand. Sanitary pad 1: - Surface veil: 0.56 g (polypropylene) - Intermediate layer: 0.85 g (cellulose) - Absorbing core: 2.87 g (cellulose) - Bottom film: 0.76 g (polyethylene) Sanitary pad 2: - Surface veil: 0.46 g (polyolefin) - Absorbing core: 2.33 g (cellulose, polyolefin, absorbing polymer, rayon, polyester) - Bottom film: 0.51 g (polyolefin) We assume that the different materials stated per component are used equally in the component. What is stated as polyethylene is attributed to HDPE and what is stated as polyolefin is attributed to PP or HDPE following the materials stated for the first product when possible. For the absorbing core, the material is equally attributed to HDPE and PP.

11.3.4.3 Consumption of flushed products

In a second step, the consumption of the hygiene products needs to be known, since the basis for this framework which was published earlier[1] does not contain a separate description of hygiene products used. For most products, we adopt a "bottom-up" approach to estimating their consumption: we start at the consumer level and scale up the data found (Table S24).

Table S24: Approach for the calculation of the consumption of hygiene products.

Item	Sources and comments
Tampon	The number of users of feminine hygiene products needs to be first determined, before the consumption of tampons can be estimated. This number depends on the total population of the considered region, on the fraction of the population that is female, and the fraction of the female population that have menstruation. For 2014, the Swiss population to 8.19 million[35]of which 51% were women[36]. We estimate the fraction of the female population that experience menstruation based on the proportion of the population aged 12-54. In 2014, 57.9 % of all Swiss inhabitants were aged 12-54[36]. Branch et al[37] used a survey conducted in years 2001-2004 in the US to connect feminine hygiene practices with phthalate exposure. According to this study, the fraction of participants reporting use of tampons in the past month amounts to 42%. Some participants may have used several feminine hygiene products, but these numbers are not provided. All considered participants were aged 20-49. A woman using tampon is thought to use 18 tampons per month on average[38].
Wet wipes	Statistics on the use of wet wipes in Europe could be found in a web resource citing Euromonitor, in which the number of wet wipes used in Europe 2014 was available[39]. According to this resource, the Europeans used 60 billion wet wipes in 2014, of which a large majority were baby wipes, and the rest were cosmetic, general and intimate wet wipes. An equivalent number for Switzerland can be obtained by downscaling this number using the European and Swiss populations[35]: 1 billion wet wipes.
Panty liner	Panty liners are another feminine hygiene product that can be flushed down the drain. Using the Euromonitor data available via Chalabi[40], we know that women in the US used on average 69 panty liners per year. Using the number of women using feminine hygiene products, one obtains the number of panty liners used in Switzerland in 2014.
Tampon applicator	Of all tampons available on the market, a certain fraction is available with a tampon applicator. This part of the product can also be discarded down-the-drain, and needs to be separately estimated. The share of tampons sold with applicator was estimated based on the number of tampons products available with or without applicator in four different retailers in Switzerland. From this sampling, it results that out of 44 types of tampon products available, 14 were sold with applicator, which results in approximately 35% of the tampon sold with applicators.
Cotton swabs	The frequency of use of cotton swabs is estimated to be 81 uses per person per year, based on a survey performed at Riga University among students[41].
Disposable cleaning cloth	No data on the consumption of disposable cleaning cloths was found. Nonetheless, an estimate was built assuming that two such cleaning cloths are used per month per household. Switzerland has an average household size of 2.2[42] and a population of 8.19 million in 2014[35].
Sanitary towels	According to Branch et al 2015[37], 62% of women aged 20-49 may be using sanitary pads. Using the same number of feminine hygiene products per year per user as for tampons[38], one obtains an estimate of the number of sanitary pads used in Switzerland for 2014.

11.3.4.4 Flushing probabilities

The flushing frequency can be estimated from Friedler[29] and Spence[30] (Table S25 and Table S26) by comparing the reported amounts flushed to the estimated consumption for each product. The data published by Spence et al. is more recent and may be therefore more reliable to model the current state of the flushing behaviour. Unfortunately, some of the data could not be considered, as the total flushed amount of tampons as reported by that study exceeds the amount of consumed tampons as we estimate it. Since there may be a difference in the habits of the inhabitants of the two regions, the flushing probability of tampons was not estimated using this dataset. As a result, both datasets were used to estimate the flushing probabilities, as they completed each other well and may have contributed to give a better idea of the variability of this process.

Table S25: Calculation of the flushing probability as derived from Friedler[29]. Grey text corresponds to data that was not considered in this study.

Item	Fraction of households reporting flushing of item[29]	Fraction of households using the item	Flushing probability
Tampon	0.109	0.119[36,37]	0.914
Wet wipes	0.058	0.891[43]	0.065
Dental floss	0.036	0.368[44]	0.098
Panty liner	0.029	0.176[36,37]	0.165
Tampon applicator	0.029	0.038[36,37]	0.764
Cotton swabs	0.022	0.691[41]	0.032
Bandage	0.007	1	0.007
Disposable cleaning cloth	0.007	0.75 (assumption)	0.009

Table S26: Calculation of the flushing probability using the data obtained by sampling from the sewers by Spence et al.[30]. Grey text corresponds to data that was not considered in this study.

Product	Average number flushed per day	Number flushed per day per user	Number used per day per user	Flushing probability
Wipes	194.67	0.15	0.32[43]	0.460
Sanitary pads	8.67	0.04[37]	0.60[38]	0.065
Tampon	110.00	0.73[37]	0.60[38]	1.222
Cotton swabs	14.67	0.01	0.22[41]	0.050
Sanitary pad shells	7.33	0.03[37]	0.60[38]	0.055
Tampon applicators	1.33	0.03[37]	0.60[38]	0.042
Panty liners	22.00	0.10[37]	0.19[40]	0.525
Sanitary packaging	7.67	0.02[37]	0.60[38]	0.036

11.3.5 Flows through composting and anaerobic digestion of organic waste

In recent years, growing attention has been given to the problem of contaminants in organic waste[45], in the case of plastics not only because of the pollution it causes, but also for the decreased perceived value of compost and digestate as secondary resources. We base our estimates on the samples collected by the Swiss inspection association for composting and digestion plants[46]. In this study, the plastic content larger than 1 mm was quantified for a total of 139 samples of compost and solid digestate. The resulting analysis provided an estimate of approximately 80 tonnes of plastic reaching soil through these pathways. Assuming that around 99% of the plastic is retained during the sorting process[47], a total of 8000 tonnes of plastic larger than 1 mm are collected with organic waste in Switzerland for one year.

In order to also have an estimate of the amount of smaller microplastic in the organic matter treated, the data published by Faure et al.[48] were considered. In that study, seven compost and

digestate samples provided by the Swiss association of waste treatment plants managers were analysed. The samples were separated in 5 size classes, and the particles were visually identified, extracted, counted and weighted. 8%, 2%, 51%, 14%, 2%, 9% and 20% of the mass of plastic pieces larger than 300 µm was smaller than 1 mm. With an average of 15% mass, this can be added to the estimated output of plastic to organic waste collection: 14 tonnes.

Both of these amounts need to be shared among the different polymers considered and other polymers. The polymers considered in this study make up 72.8% of the plastic manufacturer's demand in 2014[49] which gives us a mass of 5824 tonnes for the pieces larger than 1 mm, and 10 tonnes for the smaller fraction. Out of all the larger plastic sorted from the 139 samples, 61% of the mass sorted were films and 39% hard plastics[46]. The following products are considered as products at risk for emission of films in compost and digestate: consumer films, consumer bags, agricultural packaging films, agricultural films and agrotextiles. For hard plastics, the following products are considered: consumer bottles, other consumer packaging, agricultural packaging bottles, agricultural pipes, other agricultural products, household plastics, straws and cutlery. The mean masses contained in these compartments are used as weight to attribute the emissions to the different products and polymers. A TC is then calculated using these emissions.

As already mentioned, 99% of the plastic larger than 1 mm are sorted out of the organic waste. The rest needs to be split into smaller and larger than 5 mm to follow the chosen size definition of MP. For this, the mass fraction of plastic pieces larger than 5 mm[48] out of all pieces larger than 1 mm, is considered. 97% of the plastic in the finished compost and digestate is applied onto agricultural soils and 3% on residential soils.

11.3.6 Other emission flows

11.3.6.1 Waste collection and recycling

We assume a release of 0.01% for all collection processes except:

- Pellet waste collection, for which the emissions are discussed along with emissions from the production of pellets in the following section.
- Construction and demolition waste collection, both recyclable and incinerable fractions. This specific release will be addressed in the section on construction plastics.

Various emissions can occur during recycling. For packaging recycling, construction and demolition waste recycling, automotive part recycling and agricultural plastic recycling, we refer to the section on MP releases from technical processes. Recycling processes involving a shredding stage generate large amounts of dust, which can cause emissions to outdoor air. Plastic from automotive and EEE applications undergoes shredding to some extent in Switzerland. We assume a release rate of 0.1%.

11.3.6.2 Pellet, chips, flakes and powder emissions during production, manufacturing and recycling

Losses of pellets, chips, flakes and powdered primary material can occur at any stage of the value chain[50]: during loading, unloading and storage at production, compounding, conversion (manufacturing), transport, trading and recycling sites, due to careless handling at any moment or due to losses of containers in the seas. These pellets may then be found in larger numbers in proximity to production facilities[51]. Pellet spills from producers, transport and processors have most recently been studied in 2016 for the UK[52] with the aim of estimating the quantity of pre-production pellets that are lost each year in the UK by reviewing previous research and speaking with industry representatives and stakeholders. The loss rates were estimated to be around 0.001-0.01% for four different points in the value chain: production, transport, processing and waste management. This amount can be shared between soil and storm drains using the reported amounts of pellets in drains and on site. For production sites, around 0.04% of this amount flows through storm drains and the rest is left on residential soil.-For storage and transport, around 25% of the pellets were reported in drains. Flakes or powders were not considered separately.

The only available estimate of emissions of plastic that may be applied specifically to powder originates from an emission scenario document on plastics additives by the OECD[53]. Their worst case scenario of dust generation is the following: 0.5% emissions for particles smaller than 40 µm and 0.1% for particles larger than 40 µm. These numbers may vary depending on the process and density of the material. We consider that these scenarios are quite pessimistic, and preferred to use the same losses for powders as for pellets.

More release may be expected from recycling plants than production plants due to the use of recycled bags for transportation which are more likely to tear and release fragments. Nevertheless, in the absence of better data, the numbers for pellet release are used. These release

rates are used for packaging recycling, construction and demolition waste recycling, automotive part recycling and agricultural plastic recycling.

In order to model the emissions of MP from textile production and manufacturing to wastewater, the release of fibres during the first washing cycle in release studies is used:

- Hernandez et al.[54] did not find a different MPF release during the first wash cycles compared to subsequent cycles: 0.025 mg MPF / g textile.
- Pirc et al.[55] found releases ranging 0.008 % to 0.021 % of the mass of the washed textiles during the first cycle.

Airborne emissions of fibres from the textile industry cannot be excluded. A study[56] reveals that workers in textile manufacturing plants are at a higher risk to develop respiratory symptoms than a control group. In the absence of data, and considering the small importance of this industry for Switzerland, this emission pathway was omitted from the model.

11.3.6.3 Construction and demolition sites, buildings in use

Macroplastic can be lost from a construction or demolition site or during the transportation of the collected waste. The emissions of collected construction and demolition waste are quantified based on an estimation received by private communication with Laure Müller (Canton de Genève) on 4/8/2017. Around 200-250 kg of incinerable construction and demolition waste is collected every week on the 4km before the incineration plant, coming from both the sites and the waste transportation. This corresponds to 10.4-13.0 metric tonnes collected in the 4 km around the incineration plant per year. Assuming that the waste is uniformly distributed in a 4 km circle around the incineration plant, we can upscale this number to the whole canton of Geneva using an area of 50 km^2 around the incineration plant, and of 282 km^2 for the Canton of Geneva. This means that 58-73 t are collected per year next to roads and in fields in the canton of Geneva. We assume that half of this amount is lost during transport of the waste: 29-36 t and the rest are lost during construction or demolition. Based on the used collection rate for roads of 10-90% (see section on littering), the previous estimate can be converted into the amount of waste lost in Geneva each year: 32-360 t. Knowing that the total incinerable construction and demolition waste around Geneva in 2014 amounts to 24'532 metric tonnes[57], we obtain that 0.13-1.47% of the plastic waste collected is lost and 0.13-1.47% of the plastic used on construction sites is released each year.

MP can also be generated during construction and demolition activities. The release of EPS and XPS microparticles during professional use is covered by a risk assessment report for the European Union on HBCDD[58]. According to these estimations, 5.0 g XPS-particles per tonne XPS are released by sawing. For EPS, 100 g particles per tonne EPS are estimated to be emitted by sawing and other cutting processes as a worst case scenario. The release during private use is considered small compared to emissions from professional use. The release of EPS during deconstruction was also considered in the risk assessment report for HBCDD[58], where they tried to mimic the particle generation when EPS boards are broken. This resulted in an emission factor of 90 g EPS-particles/tonne EPS, which corresponds to 0.009% release during deconstruction aimed at recycling. For other types of demolition, not knowing how often an EPS board may be broken, the release factor of 0.1% was used.

Emissions of MP during use of building plastics may occur. We model such an emission for applications subject to wear during use:

- Pipes: Release of 0.1% to residential soil and 0.1% to subsurface soil
- Wall and floor coverings: 0.1% to indoor air.

11.3.6.4 Agriculture

A survey made among French agriculture firms shows what waste management practices[59] are preferred. Different options are given: collection in specific channels, by specialist firms, by recycling centres, in mixed waste collection and other waste management practices. We assume that other waste management practices can be divided evenly into stockpiling, burying, burning and remaining options (Table S27). The buried fraction is flowing to agricultural soil as macroplastic. This estimate constitutes a worst case scenario for this release. A best case scenario in which nothing is buried is also considered.

Table S27: Waste management practices based on the survey carried out by Agreste[59].

Product category	Number of exploitations using other waste management practices	Fraction of exploitations using waste other management practices	Assumed fraction being buried
Agricultural packaging films	43787	0.236	0.059
Agricultural packaging bottles	14191	0.065	0.016
Agricultural films	23057	0.146	0.037
Agricultural pipes	30420	0.151	0.038
Other agricultural products	30420	0.151	0.038
Agrotextiles	73101	0.341	0.085

Additional releases of agricultural plastics are possible through wear. While mulching plastics are a commonly cited source of plastic to soil, little quantitative data could be found on that release process. The literature available focuses on comparing visual cues of degradation between different materials, depending on location and exposure time. Changes in visible bare soil area or mulch film area are used to assess the state of degradation of the mulch film.

1. The percentage of bare soil area of PE plastic mulch films, after 100 days exposition to environmental conditions in Spain, was estimated visually with several graphics processing methods. Bare soil areas of 17.7±4.5 %, 19.6±4.5 %, 18.9±4.9 % and 37.8±7.6 % were obtained. While this method may be useful for comparing the performance of different materials, quantifying plastic emissions based on this data remains uncertain, since the film may be stretched at first, and folded once cracks have developed[60].
2. The mass loss of plastic mulching films was also measured in an older study[61], where different materials were buried in a net, and the mass after 2 years of exposure was measured, after cleaning of the material. On the four locations studied in France, no degradation was observed for PE. It should nevertheless be noted, that this study focussed on material performance and did not look into the fragments of plastic possibly released.
3. The degradation of mulch films made from 7 different materials was studied using qualitative degradation indicators ranging from 1 to 9[62]. Above soil degradation was assessed using area coverage of the soil, where 1 represents 0% coverage and 9 represents 100% coverage. In-soil degradation was estimated by the percentage of dematerialisation of the film. PE mulch films obtained a score of 8.5 above soil across all locations and seasons studied. Assuming the qualitative score can be translated into a quantitative score with a linear equivalence, we obtain a degradation of 6.25%. PE mulch films also obtained a degradation score of 8.6 in soil, which translates into 5% degradation

The precision of these results should not be overestimated when applying this knowledge to MP release from plastic mulching films, because of the large uncertainties in the measurement process as well as the quantities measured. Similarly for greenhouse films, no information could be obtained. We assume that for a film with 1 m width, 1 cm is lost on both sides when it reaches its end of life. This results in an overall loss of 2% of plastic as microplastic. No MP release from agricultural packaging is included in the model. For agricultural pipes and other agricultural applications, we assume a release of 0.1% MP to agricultural soil.

11.3.6.5 Automotive plastics

The litter quantity and composition along roadsides has been studied in Washington state in 2004[63]. The mass of automotive parts emitted per distance driven can be calculated for different types of roads from these results and be adapted for Switzerland using data on distances driven by person and day with a car. For Switzerland, this number is reported to be 23.8 km/pers/day[64]. Considering the Swiss[65] populations, one finds that between 20 t and 66 t of automotive plastic can be found on roadsides in Switzerland. As data on distances driven by person and day are not available for the different types of roads reported in the litter composition study[64], only the lowest and highest emission rates of automotive parts are retained. These emitted masses are attributed to the different polymers following the automotive polymer demand in Europe[66] (Table S28). In order to define an appropriate TC for this emission, these masses are then compared to the masses of consumed automotive parts per polymer. This release is assumed to correspond to the amount of litter found on road sides after sweeping, and thus flows directly to the final road side compartment.

Table S28: Masses emitted from the different polymers with the corresponding uncertainty.

Material	Value (kt)	Data Quality Indicator Scores (DQIS)					Spread
		Geo	Temp	Mat	Tech	Rel	
LDPE	0.00068	3	3	3	1	2	0.67
	0.00224	3	3	3	1	2	0.67
HDPE	0.00146	3	3	3	1	2	0.67
	0.00481	3	3	3	1	2	0.67
PP	0.00579	3	3	3	1	2	0.67
	0.01914	3	3	3	1	2	0.67
PS	0	3	3	3	1	2	0.67
	0.00002	3	3	3	1	2	0.67
EPS	0	3	3	3	1	2	0.67
	0	3	3	3	1	2	0.67
PVC	0.00074	3	3	3	1	2	0.67
	0.00243	3	3	3	1	2	0.67
PET	0.00001	3	3	3	1	2	0.67
	0.00003	3	3	3	1	2	0.67

11.3.6.6 Shotgun cartridges during hunting

Shotgun cartridges which are used for specific types of game are responsible for emissions occurring directly in natural habitats[67]. In Switzerland, shotguns are used for "*Niederjagd*" and also roe deer hunting in many cantons.

The consumption of shotgun cartridges is estimated based on the number of animals killed in 2014 with a shotgun. The number of animals killed per year is available online by type of animal[68]. Some uncertainty exists concerning the fraction of these animals which were hunted using shotguns, especially for roe deer. A worst case scenario is built in which we assume that all the small game and roe deer were shot using shotguns. Considering the magnitude of this release flow compared to other ones, assuming a worst case scenario will not inflate the total emissions. 107'147 animals were reported to be killed in 2014 in Switzerland[68]. We assume that the number of shots per kill can be described by a distribution built on the following numbers: 1 (strict minimum of shots needed to kill an animal), 3.6, 2.63 and 4.05 (number of shots for water birds in Denmark as reviewed by Kanstrup et al.[67]).

Most of the plastic used for this application is said to be LDPE[67]. We assume that these shotgun cartridges are exclusively made of LDPE for our worst case scenario. Approximately 6 g of plastic are used per cartridge: 4 g for the shell and 2 g for the wad.

The shells and the wads may end up in nature in different proportions. An anonymous hunter from Switzerland suggested the following proportions:

- All of the wads end up in nature,
- Between half and more than ¾ of the shell end up in nature. The rest of the shell is normally taken home.

We therefore define a total release rate of: $\frac{4\ \mathrm{g}}{4\ \mathrm{g}+2\ \mathrm{g}} \cdot 0.75 + \frac{2\ \mathrm{g}}{4\ \mathrm{g}+2\ \mathrm{g}} \cdot 1 = 0.825$.

The plastic pieces may be emitted to soil and surface waters depending on the type of game shot. To estimate the proportions of the emitted plastic to each compartment, the proportion of killed water birds with respect to the total number of killed animals was used[68]: 12.13%. All parameters used are shown in Table S29 with their attributed uncertainty.

Table S29: Overview of the parameters used for the calculation of the emissions from hunting activities.

Parameter	*Description*	Value	*Source*	Data Quality Indicator Scores (DQIS)					Spread
				Geo	Temp	Mat	Tech	Rel	
Kills	*Number of kills with a shotgun*	107147		1	1	1	2	3	1.02
Number of shots per kill	*Strict minimum of necessary shots to make a kill*	1	*Fixed*	1	1	1	1	1	0.13
	Estimated number of shots per kill	3.6	[67]	2	3	1	2	1	0.39
	Estimated number of shots per kill	2.63	[67]	2	3	1	2	1	0.39
	Estimated number of shots per kill	4.05	[67]	2	3	1	2	1	0.39
Plastic mass per shot	*Mass of plastic per shot in g*	6	*Private communication anonymous hunter 21.11.2018*	1	1	1	2	3	1.02

11.3.6.7 Packaging

Besides littering, dumping and disposal in organic waste, packaging may reach the environment through alternative pathways. Non-consumer packaging may be inadvertently lost. We assume that this emission follows the same pattern as plastic lost during transportation of construction and demolition plastic.

Schymanski et al.[69] have reported MP concentrations in drinking water from plastic bottles, glass bottles and beverage cartons. Since no significant difference between the concentrations from the water in glass bottles and the other containers could be found in the study, assuming that the quantity of MP created during use of packaging is negligible would be reasonable. Nevertheless, to make sure that no relevant flows are overlooked, a worst case scenario was calculated assuming that all the PET particles measured originate from the bottle. Schymanski et al.[69] report a mean concentration of 14±14 particles/L MP in single-use bottles. 59% of these particles were PET, and 70% had a size below 20 µm. As a very conservative estimate, we assume that the particles are spherical and have a mean diameter of 50 µm. We further assume a 1L bottle mass of 10g[70] and a PET density[71] of 1.38 g/cm^3. We obtain an order of magnitude of the mass fraction released during use of 10^{-9}. Considering the very low order of magnitude of these flows, which constitutes by far a worst case scenario, this possible pathway is omitted from the MFA.

11.3.6.8 Personal care and cosmetic products (PCCP)

5% of the cosmetic product stays in the container and is discarded with mixed waste and the rest is modelled to be collected with wastewater[72].

11.3.6.9 Fabric coatings

No quantitative data regarding the wear of PVC coatings for textiles could be obtained. A total release of 1% as MP is modelled, split between the four compartments: residential soil, natural soil, wastewater collection and storm water collection.

11.3.6.10 Clothing

MPF can be shed from textile products by different processes during use. Washing is often mentioned as the main or only source of MP fibres (MPF) to the environment, but drying and wear of textiles are likely to play an important role as well[73,74]. In the absence of detailed data on some of these flows, a number of assumptions have to be made. The uncertainty associated to these assumptions has been adapted accordingly, in order to highlight the parameters for which there is a more urgent need for data.

The description of some emission processes depends on many different variables. In order to account for the uncertainty and variability associated to these variables, each parameter was transformed into a distribution using the method based on the semi-quantitative Pedigree matrix and assembled into a triangular, trapezoidal or step distribution, depending on the amount of different data points. Considering the little amount of data available, a separate description could not be made for PET, PP and HDPE.

Washing

The fraction of MPF emitted to wastewater by washing $TC_{\mathrm{Washing,WW}}$ is defined as the mass fraction released by all wash cycles undergone during the lifetime of the product. As the model we consider is static, the release accumulated over the lifetime of a product is aggregated to fit into one year. To calculate it, the mass fraction of fibres released during one wash cycle and the number of washes during the average lifetime of a product need to be known.

Information on the mass fraction released during a wash cycle can be obtained from recent studies on MPFs release from washing.

Laboratory studies:

- Hernandez et al.[54] report a fibre mass release of 0.01 % of the textile's mass for PET fabrics in laboratory washing conditions with DI water only and showed the importance of the presence of detergent for the fibre shedding.

- De Falco[75] report a fibre mass release between 0.057 g/kg and 0.399 g/kg for three different types of fabrics washed with liquid or powder detergent in a laboratory model. The three types of fabrics considered were plain weave PES and PP and double knit jersey PES. The washing experiments were performed according to an ISO method used for testing the colour fastness of textiles. The wash water was filtered through polyvinylidene fluoride filters with a pore size of 5 µm. As a washing performed with this method corresponds to approximately 5 washes in a standard consumer washing machine, the mass releases used here were divided by 5.

Real conditions:

- Sillanpää et al.[76] measured the release of MPFs from 100% PET and 100% cotton garments during 5 consecutive wash cycles with detergent. In the case of polyester fabrics, the amount of fibres released stabilised around the 3rd wash. The mass release percentages for four different garments and during any of these 3 last wash cycles was measured to be between 0.022 and 0.054% for front-load machines.
- Hartline et al.[77] measured the release of fibres from new and mechanically aged polyester garments without detergent. The fractional fibre mass release ranges between 0.008 and 0.177% per washing with front-load machines, based on two simultaneous filtrations with filter mesh sizes of 20 and 333µm.
- Pirc et al.[55] conducted 10 successive washing cycles of garments with and without detergent and softener. Their results show that fibre emissions stabilize around 0.0013% after seven washing cycles and do not depend much on the detergent and softener added.
- Liebezeit et al.[78] cite own fibre release experiments from polyester garments with no information as to the method chosen. In the absence of method description, these parameters were left out of our analysis. Additional studies[79,80] reported the number or mass of fibres released by washing one piece of garment. Since no information regarding the mass of the garments is given, appropriate TCs cannot be derived and these studies are not considered.

The number of washes of a garment during its lifetime can be estimated knowing the average number of times a garment is worn, and after how many wear cycles it is washed. The average number of times a new garment is worn is estimated to be around 98 times in EU-28 in 2014[81]. The number of wears between washes for a pair of jeans is estimated to be around 2.5 for the UK and France[82]. According to these two numbers, a garment is washed 39 times during its

lifetime. A final estimate was made in a report from the Danish EPA[83], where based on various statistics, they predict an average number of washes for a textile product of 19. The release of MPF to wastewater collection shed by washing of clothing is defined as follows:

$$R_{\text{Washing,WW}} = F_{\text{Washing,one cycle}} \cdot N_{\text{washing}} ,$$

where $F_{\text{Washing,one cycle}}$ is the distribution representing the mass fraction of fibres released during one wash cycle, and N_{washing} is the distribution representing the number of washes during the average lifetime of a product.

Drying

Drying of garments can also cause emissions of fibres[55,73]. The resulting flows can be modelled in a similar fashion to the emission of fibres caused by washing, while considering the differences caused by different drying processes. We consider the three following drying processes: tumble drying, indoor cloth-line drying or outdoor cloth-line drying. For every washing cycle, the frequency of tumble-drying is estimated to be 25%[84]. The frequencies of indoor and outdoor cloth-line drying can be estimated based on survey data for the UK for household textiles[85]. The frequency of indoor cloth-line drying is estimated to be 34%, and the frequency of outdoor cloth-line drying is estimated to be 41%.

MPFs shed by tumble drying are mostly collected on a filter, and subsequently disposed of with mixed waste. The possible remaining fractions may be released to indoor and outdoor air, by the dryer and the handling of the filter, and to waterways through the evacuation of water and by handling of the filter. The corresponding release is defined as the mass fraction released by tumble-drying during all drying cycles undergone during the lifetime of the product. They depend on the frequency of use of a tumble-dryer during the lifetime of the textile product considered and on the mass fraction of the textiles released during one cycle of tumble-drying. The total mass collected on the filter was measured by Pirc et al[55], they obtained a release of 0.0045 % of the mass of the garment. To describe the fraction released to air, we assume that $f_{\text{to indoor air}}$ = 0-1% of this mass can escape the filter while handling it. We also assume that a fraction corresponding to $f_{\text{to WW}}$ = 0-1% of this mass is released to wastewater.

We can therefore write the release caused by tumble-drying as:

$$_{\text{TD,Textiles,indoor air}} = F_{\text{TD,one cycle}} \cdot f_{\text{TD}} \cdot N_{\text{washing}} \cdot f_{\text{to indoor air}} ,$$

$$R_{\text{TD,Textiles,WW}} = F_{\text{TD,one cycle}} \cdot f_{\text{TD}} \cdot N_{\text{washing}} \cdot f_{\text{to WW}}$$

where $F_{\mathrm{TD,one\ cycle}}$ is the distribution describing the mass fraction emitted during one tumble-drying cycle, f_{TD} is the distribution describing the frequency of tumble drying, and N_{washing} is the distribution representing the number of washes during the average lifetime of a product.

MPFs emitted during indoor cloth-line drying are released to indoor air:

$$R_{\mathrm{ICLD,Textiles,Indoor\ Air}} = F_{\mathrm{Washing,one\ cycle}} \cdot F_{\mathrm{CLD\ vs\ washing}} \cdot f_{\mathrm{ICLD}} \cdot N_{\mathrm{washing}} ,$$

where $F_{\mathrm{Washing,one\ cycle}}$ is the distribution representing the mass fraction released during one cycle of washing as calculated above, $F_{\mathrm{CLD\ vs\ washing}}$ is a factor describing the relative mass shedding from cloth-line drying versus washing and f_{ICLD} is the distribution for the frequency of indoor cloth-line drying. In the absence of information regarding the magnitude of the fibre release during drying, we construct a distribution with two extreme scenarios: either no fibres are released or half the amount is released as during washing.

MPFs shed by outdoor cloth-line drying are modelled to be entirely released to outdoor air. The fractional release of fibres during one drying cycle is assumed to be identical to the indoor case:

$$R_{\mathrm{Drying,Textiles,Outdoor\ Air}} = F_{\mathrm{ICLD,one\ cycle}} \cdot f_{\mathrm{OCLD}} \cdot N_{\mathrm{washing}} ,$$

where $F_{\mathrm{ICLD,one\ cycle}}$ is the distribution representing the mass fraction released during one cycle of indoor drying and f_{OCLD} is the distribution for the frequency of outdoor cloth-line drying.

Wear

Very little literature regarding the magnitude of the emissions of fibres through wear are available. Estimates for shedding through wear for clothing applications could be constructed using three different sources from different fields.

A first estimate relies on a study mentioned a risk assessment report for the European Union[58], where the release through wear was investigated using a Martindale abrasion test. The tests were performed on one single cotton fabric with 7.7% flame retardant content as coating. Since cotton is known to wear faster than synthetic materials, this estimate constitutes a worst-case scenario. This estimate yields a shedding rate of 0.86%.

The second approach uses data published as conference proceeding in 2002[86], reporting fibre concentrations generated by volunteers performing an activity in a test chamber during one minute. Unfortunately, only a limited amount of information is available from the description

of the experiment, as for example the baseline fibre concentration between experiments and the activity performed by the volunteer. The particle concentration is reported by size category, and various assumptions are made in order to include this data into our model. Only the two volunteers who were wearing synthetic clothing during the test experiments are considered here.

Table S30: Reported particle concentrations[86] by size category for the two volunteers who were wearing synthetic clothes.

Particle concentration C_i (m^{-3} min^{-1})	<0.5µm	0.5-1 µm	1.0-2.0 µm	2.0-3.0 µm	3.0-5.0µm	5.0-10.0 µm
Clothing 1 (upper body: acrylic; lower body: polyester trousers)	0	97000	43000	41000	44000	97000
Clothing 2 (upper body: polyester/cotton; lower body: denim jeans)	60000	34000	13000	7000	5000	8000

Assuming:

- an average length of fibres for each size category l_i : 0.3 µm, 0.75 µm, 1.5 µm, 2.5 µm, 4.0 µm, 7.5 µm.
- a linear density of fibres $\rho_{lin} = 2.5\ \mathrm{dtex} = 2.5 \cdot 10^{-10} \mathrm{g/\mu m}$[55,87],
- that the dust concentration was not quite homogeneous in the test chamber, we use half of the chamber volume $V = \frac{1.1\ \mathrm{m} \cdot 1.47\ \mathrm{m} \cdot 2.6\ \mathrm{m}}{2} = 2.10\ \mathrm{m}^3$ over which the concentrations were sampled,
- an average mass of worn clothing $m_{clothing} = 1.0\ \mathrm{kg/pers}$, following suggestions for mass correction of body weight during clinical tests[88],
- an active time of a person $t_{active} = 0.76\ \mathrm{h/day}$, based on the average number of steps per person per day in Switzerland of 5512[89], and average step length of 0.7 m and an average walking speed of 1.4 m/s[90]
- an average number of times a textile is worn $N_{wear} = 98\ \mathrm{day}$[81] which was already introduced earlier in the paragraph about washing related emissions.

The shedding rate can then be expressed as:

$$\mathrm{Wear} = \left(\sum_i C_i \cdot 60 \cdot l_i \cdot \rho_{lin} \right) \cdot \frac{V}{m_{clothing}} \cdot t_{active} \cdot N_{wear} \ .$$

We obtain shedding rates of 0.27% and 0.04% for the two different volunteers considered.

The third estimate is based on a publication relating to dust generation in museums[91]. Several methods were used to generate dust, and several parameters were tested. We here use the

measurements that were performed for clean polyester clothes, by shaking a piece of garment in a closed test chamber, which generated 1.5 mg of particles in 30 minutes. This corresponds to a fibre shedding rate of $G = 0.003\ \mathrm{g/h/garment}$. Assuming:

- a garment mass of $m_{\mathrm{clothing}} = 500\ \mathrm{g/pers}$[88] (assuming half of a person's clothing mass)
- an assumed active time of $t_{\mathrm{active}} = 0.76\ \mathrm{h/day}$
- the average number of times a textile is worn $N_{\mathrm{wear}} = 98\ \mathrm{day}$[81]

we obtain a shedding rate of:

$$F_{\mathrm{Wear}} = \frac{G}{m_{\mathrm{clothing}}} \cdot t_{\mathrm{active}} \cdot N_{\mathrm{wear}} = 0.04\ \%$$

Clothing MPFs shed by wear can be released either indoors or outdoors, depending on where they are used. Knowing that 7% of a person's time is spent outdoors (excluding the time spent in transportation)[12], the destination of MPFs shed through wear to outdoor air can be approximated :

$$R_{\mathrm{Wear,Clothing,Outdoor\ Air}} = F_{\mathrm{Wear}} \cdot t_{\mathrm{Outdoor}}\ ,$$

where F_{Wear} is the distribution representing the total mass fraction lost during the lifetime of the product based on the four different estimates using the three explained approaches, and t_{Outdoor} is the distribution corresponding to the time fraction spent outdoors. The remaining fraction is shed indoors:

$$R_{\mathrm{Wear,Clothing,Indoor\ Air}} = F_{\mathrm{Wear}} \cdot (1 - t_{\mathrm{Outdoor}})$$

The validity of the studies used for the estimation of the fibres shed through wearing can be debated, because of the lack of blanks and some gaps in the method description. Nevertheless, considering the large variability in all measurements shown, one can assume that blank values may be as high as the lowest reported values, which are below the values used here. Moreover, no other data could be found that may be used for this estimation. Conducting more research in this area would be very relevant to both environmental and human exposure.

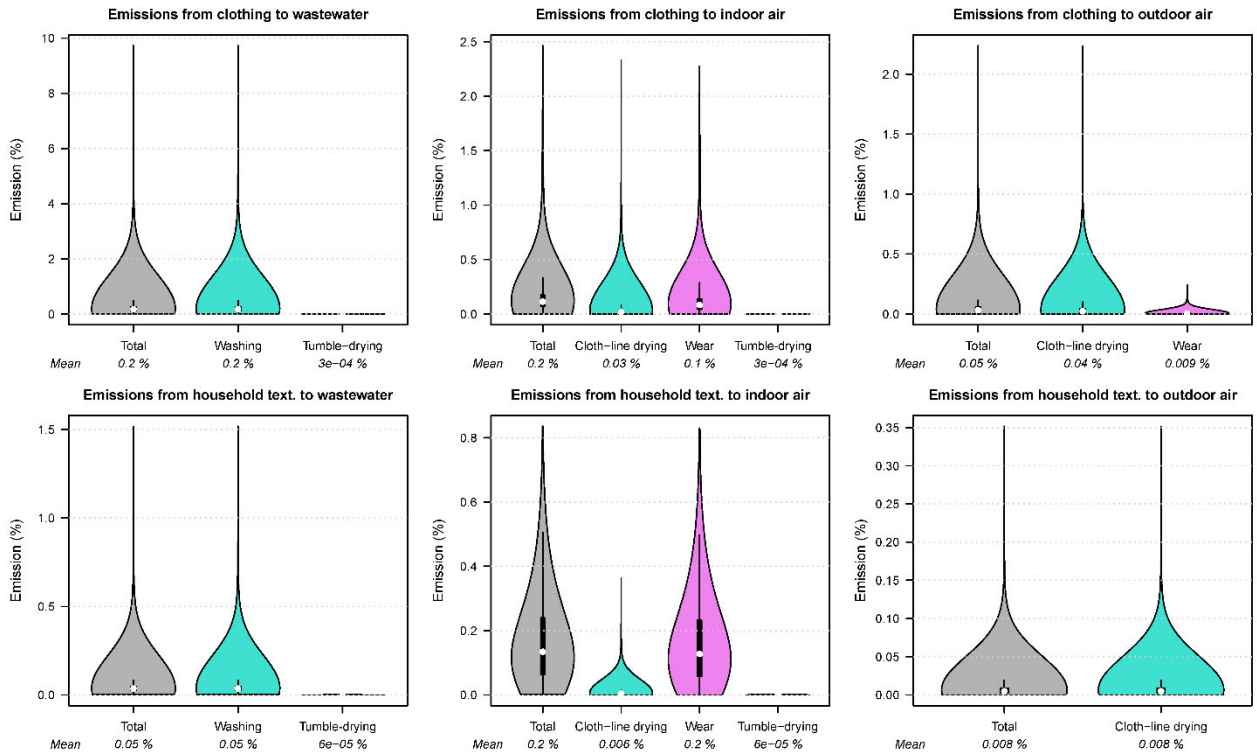

Figure S73: Resulting transfer coefficient distributions for clothing and household textiles. The contributions of the different shedding processes are indicated with the coloured violin plots.

11.3.6.11 Household textiles

Household textiles are subject to the same emission processes as clothing, but with different magnitudes. In order to estimate how often household textiles are washed, information from a survey on household textiles habits in the UK performed by WRAP[85] was used. Several questions were asked in this study as part of the survey, for different kinds of items. Based on this information, we estimated the average number of washes per year, the average number of items owned, the average mass of one item and the lifetime of the items (Table S31). The number of washes during the lifetime of the product is then calculated as following:

$$N_{\text{washes}} = \sum_{\text{item}} P_{\text{item}} \cdot \text{f}_{\text{washing,item}} \cdot \tau_{\text{item}} \,,$$

where P_{item} is the mass proportion of the individual items, $\text{f}_{\text{washing,item}}$ is the washing frequency of the item and τ_{item} is the lifetime of the item. P_{item} is calculated using the number of items owned and the item mass (Table S31) to obtain a repartition of the total household textiles into different items according to their mass. $\text{f}_{\text{washing,item}}$ is calculated using the number of washes per year per item and the number of items owned (Table S31). We finally obtain that on average, household textiles are washed 5.9 times in their lifetimes.

Table S31: Parameters used for the estimation of the average number of washes of household textiles during their lifetimes.

Parameter	Washes per year	Number owned	Assumed mass in grams	Lifetime in years
Source	*Figure 7 from* [85]	*Appendix 2, question 2 from* [85]	*Own assumptions*	*Figure 14 from* [85]
Tea towels	44	10.07	30	1.88
Towels	43	12.63	60	2.54
Bed linen	38	25.90	800	2.78
Bedding	18	12.14	1500	5.74
Mattress covers	16	3.50	800	2.55
Cushion covers	10	8.08	100	3.13
Cushions	5	7.30	200	3.42
Curtains	5	7.85	1000	4.16

The drying related emissions are modelled in the same way as for clothing. The fibre shedding rate through wear is assumed to follow the same shedding rate as in the first two approaches for clothing applications: 1.3%[92] and 0.86%[58], further assuming that only 11% of the fabric is subject to high wear[58]. As the remaining assumptions made are oriented for clothing textiles, they cannot be further used. Household textiles are modelled to be entirely emitted to indoor air.

11.3.6.12 Technical textiles

Fibres shed from the remaining textile applications are modelled differently depending on the product category. For most of the applications, two scenarios are used to estimate the release:

- a best case scenario in which nothing is shed: 0%
- a worst case scenario based on abrasion measurements: 1.3%[92].

Different receiving compartments are modelled depending on the application (Table S32).

Table S32: Emissions of MPF from the various technical textile applications

Application	Shedding rate	Receiving compartment	Comment
Building textiles	None		Little wear is expected for these applications
Geotextiles	Standard	Subsurface soil	Geotextiles are subject to different types of degradation depending on the material and its location[93]. Unfortunately, no information concerning the loss of mass during their lifetimes is available, probably partially due to the large variety in applications and their lifetimes.
Agrotextiles	Standard	Agricultural soil	
Mobility textiles	None		Little wear is expected for these applications
Hygiene and medical textiles	None		As these applications are short-lived, MPF emissions are neglected.
Technical clothing	Same as for clothing		
Technical household textiles	Same as for household textiles		
Other technical textiles	None		Little wear is expected for these applications

11.3.6.13 Parameters for emissions from textiles

Table S33: Parameters used for the probability distributions of the release of MP from textiles.

Parameter	*Description*	Value	*Source*	Data Quality Indicator Score (DQIS)					Spread
				Geo	Temp	Mat	Tech	Rel	
$N_{\text{washing,clothing}}$	*Number of washing cycles performed in the lifetime of a product (for clothing)*	39	[81,82]	2	2	3	2	2	0.51
		19	[83]	2	2	3	3	2	0.60
$N_{\text{washing,household}}$	*Number of washing cycles performed in the lifetime of a product (for household textiles)*	5.9	[85]	2	2	3	3	2	0.60
$F_{\text{Washing,one cycle}}$	*Fraction of textiles released during one cycle of washing*	0.0001000	[54]	2	2	3	2	1	0.40
		0.0002200	[76]	2	2	3	2	1	0.40
		0.0005400	[76]	2	2	3	2	1	0.40
		0.0017700	[94]	2	2	3	2	1	0.40
		0.0000800	*[94]*	2	2	3	2	1	0.40
		0.0000130	[55]	2	2	3	2	1	0.40
		0.0000798	*[75]*	2	2	3	2	1	0.40
		0.0000114	*[75]*	2	2	3	2	1	0.40
$F_{\text{TD,one cycle}}$	*Fraction of textiles released during one cycle of tumble-drying*	0.0000455	*[55]*	2	2	3	3	1	0.51
$F_{\text{CLD vs washing}}$	*Ratio of release from cloth-line drying versus washing*	0	*Assumption*	2	3	3	3	3	1.17
		0.5	*Assumption*	2	3	3	3	3	1.17
F_{Wear}	*Fraction of clothing released by wear during the lifetime of the product*	0.0027	*See description*	2	4	3	3	3	1.51
		0.0004	*See description*	2	4	3	3	3	1.51
		0.0004	*See description*	2	4	3	3	3	1.51
		0.0086	[58]	2	4	4	3	3	1.79
F_{Wear}	*Fraction of other textiles released by wear during the lifetime of the product*	0.0130	[92]	2	4	3	3	3	1.51
		0.0086	*[58]*	2	4	4	3	3	1.79
$f_{\text{surface worn}}$	*Fraction of the surface worn for household textiles*	0.1100	*[58]*	2	4	4	3	3	1.79
f_{TD}	*Factor describing frequency of tumble-drying out of all washes*	0.25	[84]	2	3	3	1	2	0.59
f_{ICLD}	*Factor describing frequency of indoor cloth-line drying out of all washes*	0.34	[85]	2	2	3	1	2	0.50
f_{OCLD}	*Factor describing frequency of outdoor cloth-line drying out of all washes*	0.41	*[85]*	2	2	3	1	2	0.50
t_{Outdoor}	*Fraction of time spent outdoors*	0.07	[12]	3	4	1	1	1	1.07
$f_{\text{to indoor air}}$	*Fraction of dust on filter that escapes to air*	0	*Assumption*	3	3	3	3	3	1.21
		0.01	*Assumption*	3	3	3	3	3	1.21
$f_{\text{to WW}}$	*Fraction of dust on filter that escapes to wastewater*	0	*Assumption*	3	3	3	3	3	1.21
		0.01	*Assumption*	3	3	3	3	3	1.21

11.3.6.14 Remaining products

Household plastics subject to wear may also cause releases of MP. We model a release of 0.1% to indoor air.

The remaining plastic applications are also subject to wear and tear, which may also result in abraded MP particles. Nevertheless, in the absence of better data, these possible pathways have been neglected.

11.3.7 Storm water and wastewater management

The wastewater and storm water collection and treatment process is described using a total of 10 processes for MP and 14 processes for macroplastic. Different systems exist for the collection and treatment of storm water. In combined sewers, the storm water is collected along with wastewater and treated in the wastewater treatment plants (WWTP). In a separate sewer system, the storm water is collected separately and discharged to surface water with or without treatment. Estimating what fraction of the storm water in Switzerland is collected in a separate or mixed system is a very challenging task for two reasons. First of all, large differences exist in Switzerland from one municipality to another, with sometimes both collection systems being used for a shared area. And moreover, estimating how much storm water will be collected in a basin is very variable. Because of these uncertainties, the flows of storm water to surface water or WWTPs are modelled using maximally spread distributions ranging from 0 to 100% for both MP and macroplastics present in storm water. Industrial storm water is mostly infiltrated in soil and partially released to surface water and WWTPs.

From wastewater collection, the plastic can flow to WWTPs or on-site sewage facilities (OSSF). MP can also exfiltrate from the sewers to subsurface soil[95]. WWTPs are described using three processes: primary, secondary and tertiary treatment. The degree of treatment of wastewater in Switzerland and the retention efficiencies of each treatment stage are used to model the flows between the treatment stages, surface water and sludge. The retention efficiencies for MP are obtained from literature[96–103]. For macroplastics, different assumptions were made based on communications with professionals. Raw wastewater can also be directly discharged after collection through combined sewer overflows (CSO)[104]. For macroplastic, a distinction between large and small macroplastic is made for a more accurate modelling of the retention of the plastic by sieves. Only cotton swabs are considered small macroplastic. All the generated sludge is incinerated in Switzerland[105].

11.3.7.1 Storm water collection

In the absence of better information, the amount of storm water being collected in a combined collection system is described by a uniform distribution between 0 and 100%. The same description applies for macro- and microplastic.

11.3.7.2 Wastewater collection

97% of the inhabitants are nowadays connected to a wastewater collection system in Switzerland[106]. We model the remaining 3% to be connected to on-site sewage facilities (OSSF) for both micro- and macroplastics.

The rest of the wastewater is transported to WWTPs using the sewer network, during which exfiltration may occur. The literature studying sewer leakage was reviewed by Rutsch[95]. The percentages of wastewater lost during dry weather flow are taken into account to model the amount of MP leaking from the sewers to sub-surface soil. No macroplastic is modelled to escape through this pathway.

11.3.7.3 Industrial storm water collection

The management of used water in Switzerland depends on the degree of pollution of the water[107]. Unpolluted water should be left to infiltrate in soil in first priority. If the local conditions prevent such an infiltration, the used water may be collected and treated in a WWTP, or discharged in surface waters. Polluted water is required to be treated before being released. Since each situation is considered individually before construction and no common database exists, and since older sites have been subject to different regulations, it is not possible to know what percentage of the storm water from industrial sites is infiltrated, discharged in surface waters or treated in a WWTP. A rough estimate was made which was considered as reasonable from an expert from the Swiss Water Association[108]:

- To surface water: 10%
- To residential soil: 30%
- To WWTPs: 60%.

11.3.7.4 Wastewater treatment

All the wastewater treated in Switzerland undergoes at least mechanical and biological treatment. Approximately 96.8% of the wastewater treated undergoes some additional processes

as phosphorus elimination, ammonium elimination, denitrification, and filtration. The following processes are modelled for the treatment of the wastewater:

1. **Entrance in the WWTP:** This process represents the influent in the WWTP.
    a. **Macroplastic:**
        i. 80% of the macroplastic is collected on screens and discarded
        ii. An estimate of the fraction of the household wastewater flowing through CSOs in Switzerland can be obtained from two different sources. The first one is a modelling study describing the emissions of micropollutants through wet weather discharges[104]. From the modelled data, it is possible to calculate the fraction of the time during which overflows are active. The frequency and duration of the events is preferred to the fraction of wastewater discharged, since mostly plastic originating from wastewater collection is flowing to wastewater treatment. The second source originally comes from an expert estimation[109]. Both sources give an estimate of 3% for the amount of pollutant escaping to surface water.
        iii. The rest of the macroplastic continues to primary treatment
    b. **Small Macroplastic:**
        i. 50% of the macroplastic is collected on screens and discarded
        ii. 3% of the macroplastic is discharged during CSO events[104,109]
        iii. The rest of the macroplastic continues to primary treatment
    c. **MP:**
        i. No MP is collected on the screens
        ii. 3% of the MP is discharged during CSO events[104,109]
        iii. The rest of the MP continues to primary treatment
2. **Primary treatment:** This process corresponds to the mechanical treatment in the WWTP. Since all the wastewater in Switzerland undergoes at least primary and secondary treatment, no outflows to surface water are modelled from this process.
    a. **Macroplastic:**
        i. 60% of the macroplastic is retained in the sludge. This number is based on a communication with professionals from the field, estimating that between the screens and the primary treatment, 90% of the macroplastic should be retained.
        ii. The rest then flows to secondary treatment.
    b. **Small macroplastic:**

   i. Similarly as for larger macroplastics, we base the fraction of plastic retained in the sludge at this stage on an assumed quantity retained overall. For small macroplastics, this overall quantity is set to 70%, which gives 42% of the small macroplastics in primary treatment to be caught in the sludge.
   ii. The rest then flows to secondary treatment.

c. **MP:**
   i. The MP retention during primary treatment is modelled using data from literature: 50%[96], 78%[97], 92%[98], 97%[99], 98%[99], 69%[100]. The retained fraction is caught in the sludge.
   ii. The rest then flows to secondary treatment.

3. **Secondary treatment:** This process corresponds to the biological treatment.
   a. **Macroplastics:**
      i. We assume that all the remaining large macroplastic is caught in the sludge.
   b. **Small macroplastics:**
      i. We assume that only 90% of the small macroplastic is caught in the sludge.
      ii. 3.22% of the rest then flows to surface water, following proportions of the wastewater being treated only with mechanical and biological treatment.
      iii. 96.78% of the rest flows to tertiary treatment.
   c. **MP:**
      i. The MP retention rate during secondary treatment was obtained from literature: 73%[96], 7%[99], 20%[99], 29%[100], 81%[101]. The retained fraction is caught in the sludge.
      ii. 3.22% of the rest then flows to surface water, as for the small macroplastics.
      iii. 9.78% of the rest flows to tertiary treatment.
4. **Tertiary treatment:** Any further processing is considered to be tertiary treatment. At this stage, all the macroplastic present is small, so no distinction is made between the sizes anymore.
   a. **Large and small macroplastics:**
      i. All the macroplastic is modelled to be retained and discarded.
   b. **MP:**

   i. The MP retention rate is taken from literature: 84%[96], 97%[102], 0%[99], 61%[101], 40%[100], 100%[110].
   ii. The rest flows to surface water.

In addition to the treatment processes, several other processes were included in the model, some of which were already mentioned above:

5. **Combined sewer overflows (CSO):** When the capacity of the WWTP is reached during a storm, raw sewage can be discharged to waterways.
   a. **Macroplastics:**
      i. In the absence of better data, a uniform distribution from 0 to 1 is assumed for the fraction of macroplastic which may be retained on screens.
      ii. The rest flows to surface water.
   b. **Small macroplastics:**
      i. Similarly as for large macroplastics, a broad distribution is assumed for small macroplastics. Because of their smaller sizes, we assume a distribution ranging from 0 to 0.5 for the fraction retained on screens.
      ii. The rest flows to surface water.
   c. **MP:**
      i. All of the MP is released to surface water.
6. **On-site sewage facilities (OSSF):**
   a. **Macroplastics:**
      i. All of the plastic is retained in the sludge.
   b. **MP:**
      i. The same retention of MP in the sludge as for the primary treatment in the WWTP is assumed.
      ii. The rest is released to sub-surface soil.
7. **Sludge:** All of the sludge used in Switzerland is incinerated since year 2006[105].

The distinction between macroplastics and small macroplastics in many processes is made in order to model different retention efficiencies for cotton swabs compared to larger objects which are less likely to cross WWTPs.

11.3.8 Release of microplastic from indoor and outdoor air

11.3.8.1 Indoor air MP

Household MP can be transferred to outdoor air through ventilation or can deposit on surfaces[111]. A rate can be attributed to each process, the combination of the two then gives an estimation of the fraction of particles escaping through ventilation or depositing on surfaces. The ventilation air exchange rate N by which indoor air is replaced with clean air can be approximated using the median exchange rate of 509 calculations performed in three US urban areas[112]: $N = 0.71\,\mathrm{h}^{-1}$. Existing measurements of the deposition rate by which particles are removed from indoor air N_e were reviewed in detail[111]. These air-exchange rates depend on a variety of parameters, of which one of the most important might be the particle size. Because of the high amount of fibres longer than 50 µm reported in indoor environments[73], the air exchange rates for sizes smaller than 2.5 µm were not considered. The amount of particles escaping through ventilation is then calculated as follows: $F_{\mathrm{Ventilation}} = \frac{N}{N_e+N}$. We obtain four estimates for the fraction of particles escaping to outdoor air $F_{\mathrm{Ventilation}} = (52\%\,;\,19\%\,;\,11\%\,;\,25\%)$.

All of the dust that has deposited on floors and furniture is assumed to be cleaned, either ending in mixed waste collection or in wastewater collection. The proportions between the two options are calculated using statistics on cleaning habits as collected from a telephone survey[113] (Table S34). Dry-mopping, sweeping and vacuum cleaning are activities that collect the dust and transfer it to mixed waste collection. On the other hand, dust can be transferred to wastewater through wet-mopping. The reported frequencies of these activities are compared for all populations combined, and we finally obtain that 15% of the dust collected flows to wastewater collection.

Table S34: Average frequency per month of various cleaning activities[113].

Activity	Parents with young children		Older adults	
	Women	Men	Women	Men
Dry-mop hard floors	7.20	3.50	5.43	4.88
Wet-mop hard floors	6.44	4.62	4.53	4.84
Sweep hard floors	14.19	9.88	8.55	8.61
Vacuum hard floors	7.26	6.77	5.15	4.51
Vacuum carpets	9.03	8.90	4.73	6.25

11.3.8.2 Outdoor air MP

A crude estimation of the fate of microplastic in outdoor air is given. All particles are modelled to deposit on the surface, and the repartition between environmental compartments is estimated based on land-use statistics[114,115].

Table S35: Transfer coefficients used for the definition of the probability distributions for the transfer from outdoor air to other environmental compartments.

Compartment	Transfer coefficient[114]	Comment
Residential soil	0.075	Residential areas
Natural soil	0.523	Including unproductive vegetation areas, bare areas, tree-covered areas and glaciers
Agricultural soil	0.359	Agricultural areas
Surface water	0.043	Including standing water and flowing water bodies

11.4 References

(1) Kawecki, D.; Scheeder, P.; Nowack, B. Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe. *Environ. Sci. Technol.* **2018**, *52*, 9874–9888.

(2) Laner, D.; Feketitsch, J.; Rechberger, H.; Fellner, J. A Novel Approach to Characterize Data Uncertainty in Material Flow Analysis and Its Application to Plastics Flows in Austria. *J. Ind. Ecol.* **2016**, *20* (5), 1050–1063.

(3) European Commission -- Joint Research Centre -- Institute for Environment and Sustainability. *International Reference Life Cycle Data System (ILCD) Handbook -- General Guide for Life Cycle Assessment -- Detailed Guidance*; 2010; http://lct.jrc.ec.europa.eu/pdf-directory/ILCD-Handbook-General-guide-for-LCA-DETAIL-online-12March2010.pdf.

(4) Weidema, B. P.; Wesnaes, M. S. Data Quality Management for Life Cycle Inventories-an Example of Using Data Quality Indicators. *J. Clean. Prod.* **1996**, *4* (3–4), 167–174.

(5) Wigger, H.; Kawecki, D.; Nowack, B.; Adam, V. Systematic Consideration of Parameter Uncertainty and Variability in Probabilistic Species Sensitivity Distributions. *Submitted* **2019**.

(6) Seas at Risk. *Single-Use Plastics and the Marine Environment*; 2017; http://metamag.org/2017/10/27/billions-of-throw-away-plastic-items-responsible-for-half-of-ocean-plastic-pollution-in-europe-research-finds/.

(7) Berger, T.; Duch, A. F.; Ricato, M.; Sommerhalder, M.; Henzen, C.; Steimer, N.; Locher, P.; Erzinger, F.; Monteil, M. *Le Littering a Un Coût: Coût Du Nettoyage Par Fractions de Déchets En Suisse*; 2011; https://www.bafu.admin.ch/dam/bafu/fr/dokumente/abfall/uw-umwelt-wissen/littering_kostet.pdf.download.pdf/le_littering_a_uncout.pdf.

(8) WRAP. *Report: Plastic Packaging Composition 2011*; 2013; http://www.wrap.org.uk/sites/files/wrap/Plastics Composition 2011 Report.pdf.

(9) McDonald's Suisse. Corporate Responsibility Report 2015 McDonald's Suisse https://www.mcdonalds.ch/mcd/317/150511_mcd_csr_report_2015_fr.pdf.

(10) Bundesamt für Statistik BFS. *Das Kultur- Und Freizeitverhalten in Der Schweiz: Erste Ergebnisse Der Erhebung 2014*; 2016; https://www.bfs.admin.ch/bfsstatic/dam/assets/349943/master.

(11) Lamprecht, M.; Fischer, A.; Stamm, H. *Sport Schweiz 2014: Factsheets Sportarten*; 2015; https://www.baspo.admin.ch/content/baspo-internet/de/dokumentation/publikationen/sport-schweiz-2014/_jcr_content/contentPar/downloadlist_copy_co_1675948713/downloadItems/571_1464186255775.download/sport_schweiz2014_factsheets_d.pdf.

(12) Klepeis, N. E.; Nelson, W. C.; Ott, W. R.; Robinson, J. P.; Tsang, A. M.; Switzer, P.; Behar, J. V; Hern, C.; Engelmann, W. H. The National Human Activity Pattern Survey (NHAPS): A Resource for Assessing Exposure to Environmental Pollutants. *J. Expo. Anal. Environ. Epidemiol.* **2001**, *11* (September 1998), 231–252.

(13) Finnie, W. C. Field Experiments in Litter Control. *Environ. Behav.* **1973**, *5* (2), 123–144.

(14) Schultz, P. W.; Bator, R. J.; Large, L. B.; Bruni, C. M.; Tabanico, J. J.; Brown Large, L.; Bruni, C. M.; Tabanico, J. J. Littering in Context. *Environ. Behav.* **2013**, *45* (1), 35–59.

(15) P. Wesley Schultz; Brown Large, L.; Tabanico, J.; Bruni, C.; Bator, R. *Littering Behavior in America: Results of a National Study*; 2009; https://www.kab.org/sites/default/files/News%26Info_Research_LitteringBehaviorinAmerica_2009Report_Final.pdf.

(16) Sibley, C. G.; Liu, J. H. Differentiating Active and Passive Littering: A Two-Stage Process Model of Littering Behavior in Public Spaces. *Environment* **2003**, *35* (3), 415–433.

(17) Heeb, J.; Ableidinger, M.; Berger, T.; Hoffelner, W. *Littering – Ein Schweizer Problem ? Eine Vergleichsstudie Schweiz-Europa*.

(18) Cialdini, R. B.; Reno, R. R.; Kallgren, C. A. A Focus Theory of Normative Conduct : Recycling the Concept of Norms to Reduce Littering in Public Places. *J. Pers. Soc. Psychol.* **1990**, *58* (6), 1015–1026.

(19) Bateson, M.; Callow, L.; Holmes, J. R.; Redmond Roche, M. L.; Nettle, D. Do Images of

"watching Eyes" Induce Behaviour That Is More pro-Social or More Normative? A Field Experiment on Littering. *PLoS One* **2013**, *8* (12), 1–9.

(20) Weaver, R. Littering in Context(s): Using a Quasi-Natural Experiment to Explore Geographic Influences on Antisocial Behavior. *Appl. Geogr.* **2015**, *57*, 142–153.

(21) Keizer, K.; Lindenberg, S.; Steg, L. The Spreading of Disorder. *Science (80-.).* **2008**, *322* (5908), 1681–1685.

(22) *Private Communication Ville de Genève, 02.05.2018*.

(23) *Private Communication Entsorgung+Recycling Stadt Bern, 10.08.2018*.

(24) Stadt Bern. *Abfallentsorgung in Tonnen Seit 2000*; 2017; http://www.bern.ch/themen/stadt-recht-und-politik/bern-in-zahlen/katost/02rauumw/downloads/t-02-7-040-abfallentsorgung-in-tonnen-seit-2000.pdf.

(25) Department for Environment Food & Rural Affairs. *Fly-Tipping Statistics for England, 2016/17*; 2017; https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/652958/Flytipping_201617_statistical_release_FINAL.pdf.

(26) Marine Conservation Society. *Great British Beach Clean 2015 Report*; 2016; https://www.mcsuk.org/downloads/gbbc/2016/487-2016 Beachwatch GBBC Summary 16pp A5 WEB Spreads.pdf.

(27) Erismann, R.; Erismann, S. *In(Beach-Litter-Density): A Comparison of Marine and Freshwater Litter Densities*; 2016.

(28) Williams, A. T.; Simmons, S. L. The Degradation of Plastic Litter in Rivers: Implications for Beaches. *J. Coast. Conserv.* **1996**, *2* (1), 63–72.

(29) Friedler, E.; Brown, D. M.; Butler, D. A Study of WC Derived Sewer Solids. *Water Sci. Technol.* **1996**, *33* (9), 17–24.

(30) Spence, K. J.; Digman, C.; Balmforth, D.; Houldsworth, J.; Saul, A.; Meadowcroft, J. Gross Solids from Combined Sewers in Dry Weather and Storms, Elucidating Production, Storage and Social Factors. *Urban Water J.* **2016**, *13* (8), 773–789.

(31) Warren Nonwovens Ltd. Wet & Dry Wipes Nonwoven Wipes http://www.warren-nonwovens.co.uk/wet-and-dry-wipe-nonwoven-materials.html (accessed Apr 5, 2018).

(32) Sahu, P. B. Wipes: What, Where, Why & How? https://www.nonwovens-industry.com/issues/2012-01/view_features/wipes-what-where-why-how (accessed Apr 5, 2018).

(33) Schueller, R. Baby wipes http://www.madehow.com/Volume-6/Baby-Wipes.html

(accessed Apr 5, 2018).

(34) Bard Hall, S. How dental floss is made - making, used, product, Raw Materials http://www.madehow.com/Volume-2/Dental-Floss.html (accessed Apr 6, 2018).

(35) Eurostat. Population change - Demographic balance and crude rates at national level [demo_gind] http://ec.europa.eu/eurostat/product?code=demo_gind&language=en&mode=view (accessed Apr 2, 2018).

(36) Eurostat. Population: Structure indicators [demo_pjanind] http://ec.europa.eu/eurostat/product?code=demo_pjanind&language=en&mode=view (accessed Apr 2, 2018).

(37) Branch, F.; Woodruff, T. J.; Mitro, S. D.; Zota, A. R. Vaginal Douching and Racial/Ethnic Disparities in Phthalates Exposures among Reproductive-Aged Women: National Health and Nutrition Examination Survey 2001-2004. *Environ. Heal. A Glob. Access Sci. Source* **2015**, *14* (1), 1–8.

(38) Schwartz, J. L.; Creinin, M. D.; Pymar, H. C. The Trimonthly Combination Oral Contraceptive Regimen: Is It Cost Effective? *Contraception* **1999**, *60* (5), 263–267.

(39) Engqvist, H. *Consumer & Healthcare Trends in European Wipes Markets World of Wipes 2013 A Tour to Europe – a Multi Facetted Region*; 2013; http://www.inda.org/BIO/wow2013_579_PPT.pdf.

(40) Chalabi, M. How Many Women Don't Use Tampons? https://fivethirtyeight.com/features/how-many-women-dont-use-tampons/ (accessed Apr 2, 2018).

(41) Veidere, L.; Arbidāne, A.; Grīnberga, I.; Borovika, A.; Skrule, J.; Karantajers, M.; Karganova, K.; Gusta, A.; Muižniece, I.; Mičule, D.; et al. The Impact of Cotton Swab Use On The External Auditory Canal https://www.slideshare.net/LindaVeidere/the-impact-of-cotton-swab-use-on-the-external-auditory-canal (accessed Apr 2, 2018).

(42) United Nations. Household Size and Composition Around the World 2017. **2017**.

(43) Statista. U.S. population : How many times have you used pre-moist wipes / cloths in the last days ? https://www.statista.com/statistics/276845/us-households-usage-frequency-of-pre-moist-wipes-and-cloths/ (accessed Apr 3, 2018).

(44) Macgregor, I. D. M.; Balding, J. W.; Regis, D. Flossing Behaviour in English Adolescents. *J. Clin. Periodontol.* **1998**, *25* (4), 291–296.

(45) Kupper, T.; Fuchs, J. G. *Compost et Digestat En Suisse. Étude N°1: Micropolluants Organiques Dans Le Compost et Le Digestat; Etude N°2: Influences Des Composts et Des Digestats Sur l'environnement, La Fertilité Des Sols et La Santé Des Plantes.*; Berne, 2007; Vol. 0743; www.environnement-suisse.ch/uw-0743-f.

(46) Schleiss, K. *Bericht Zur Analyse von Fremdstoffen in Kompost Und Festem Gärgut Der Kompostier- Und Vergärungsanlagen in Der Schweiz Gemäss ChemRRV*; 2017; https://www.mpsecure.ch/cvis/public/pdf/2017-12-22_Bericht_Fremdstoffanalysen_Auftrag_BAFU.pdf.

(47) Goldberg, L. Plastic in Organic Waste in Switzerland : Material Flows and Consequences, University of Zurich, 2018.

(48) Faure, F.; De Alencastro, L. F. *Recherche de Fragments de Plastique Dans Les Composts et Digestats Industriels*; 2016.

(49) PlasticsEurope. *Plastics - The Facts 2015: An Analysis of European Latest Plastics Production, Demand and Waste Data*; 2015; http://staging-plasticseurope.idloom.com/download_file/view/478/179.

(50) PlasticsEurope. PlasticsEurope Operation Clean Sweep® http://www.opcleansweep.eu/wp-content/uploads/2017/09/OCS_Report2017.pdf.

(51) Karlsson, T. M.; Arneborg, L.; Broström, G.; Almroth, B. C.; Gipperth, L.; Hassellöv, M. The Unaccountability Case of Plastic Pellet Pollution. *Mar. Pollut. Bull.* **2018**, *129* (1), 52–60.

(52) Cole, G.; Sherrington, C. *Study to Quantify Pellet Emissions in the UK*; 2016; https://www.eunomia.co.uk/reports-tools/study-to-quantify-pellet-emissions-in-the-uk/.

(53) OECD. Emission Scenario Document on Coating Industry (Paints, Lacquers and Varnishes). *Ser. Emiss. Scenar. Doc.* **2009**, No. 22, 1–201.

(54) Hernandez, E.; Nowack, B.; Mitrano, D. M. Polyester Textiles as a Source of Microplastics from Households: A Mechanistic Study to Understand Microfiber Release During Washing. *Environ. Sci. Technol.* **2017**, *51* (12), 7036–7046.

(55) Pirc, U.; Vidmar, M.; Mozer, A.; Kržan, A. Emissions of Microplastic Fibers from Microfiber Fleece during Domestic Washing. *Environ. Sci. Pollut. Res.* **2016**, No. 2014.

(56) Atis, S.; Tutluoglu, B.; Levent, E.; Ozturk, C.; Tunaci, A.; Sahin, K.; Saral, A.; Oktay, I.; Kanik, A.; Nemery, B. The Respiratory Effects of Occupational Polypropylene Flock Exposure. *Eur. Respir. J.* **2005**, *25* (1), 110–117.

(57) République et canton de Genève. *Inventaire 2016 Des Déchets Du Canton de Genève - Déchets Urbains, Déchets Industriels, Déchets de Chantier*; 2017; https://www.ge.ch/document/dechets-statistiques-dechets-urbains-industriels-chantier.

(58) European Communities. *European Union Risk Assessment Report: Hexabromocyclododecane CAS-No.: 25637-99-4, EINECS- No.: 247-148-4. Final Report*; 2008; https://echa.europa.eu/documents/10162/661bff17-dc0a-4475-9758-40bdd6198f82.

(59) Agreste. *Chiffres-Clés : 61 Hectares En Moyenne Par Exploitation*; 2017; http://agreste.agriculture.gouv.fr/IMG/xls/ESEA_2013_Tableaux_1bssef.xls.

(60) Moreno, M. M.; González-Mora, S.; Villena, J.; Campos, J. A.; Moreno, C. Deterioration Pattern of Six Biodegradable, Potentially Low-Environmental Impact Mulches in Field Conditions. *J. Environ. Manage.* **2017**, *200*, 490–501.

(61) Calmon, A.; Guillaume, S.; Bellon-Maurel, V.; Feuilloley, P.; Silvestre1, F. Evaluation of Material Biodegradability in Real Conditions-Development of a Burial Test and an Analysis Methodology Based on Numerical Vision. *J. Environ. Polym. Degrad.* **1999**, *7* (3).

(62) Martín-Closas, L.; Costa, J.; Cirujeda, A.; Aibar, J.; Zaragoza, C.; Pardo, A.; Suso, M. L.; Moreno, M. M.; Moreno, C.; Lahoz, I.; et al. Above-Soil and in-Soil Degradation of Oxo- and Bio-Degradable Mulches: A Qualitative Approach. *Soil Res.* **2016**, *54* (2), 225.

(63) Cascadia Consulting Group Inc. *Washington 2004 State Litter Study: Litter Generation and Composition Report*; 2005; https://fortress.wa.gov/ecy/publications/summarypages/0507029.html.

(64) Office fédéral de la statistique OFS; Office fédéral du développement territorial ARE. *Mobilité de La Population En 2015*; 2017; https://www.bfs.admin.ch/bfs/fr/home/statistiques/catalogues-banques-donnees.assetdetail.2545887.html.

(65) Office Fédéral de la Statistique OFS. *La Population de La Suisse 2015*; 2015; http://www.bfs.admin.ch/bfs/portal/fr/index/themen/01/22/publ.Document.125302.pdf.

(66) Simon, C.; Hupfer, C. *Business Data and Charts 2014/2015 Plastics Europe Market Research Group (PEMRG)*; 2015.

(67) Kanstrup, N.; Balsby, T. J. S. Plastic Litter from Shotgun Ammunition on Danish Coastlines – Amounts and Provenance. *Environ. Pollut.* **2018**, *237*, 601–610.

(68) Abteilung Arten Ökosysteme und Landschaften - Waldbiodiversität Sektion Wildtiere und Waldbiodiversität. Eidgenössische Jagdstatistik https://www.uzh.ch/wild/ssl-dir/jagdstatistik/?page=wildtiere&dt=1&gr=1&tr=1&th=1&ca=CH&co=CH&caco=1&ys=2009&ye=2016&lang=de (accessed Nov 21, 2018).

(69) Schymanski, D.; Goldbeck, C.; Humpf, H. U.; Fürst, P. Analysis of Microplastics in Water by Micro-Raman Spectroscopy: Release of Plastic Particles from Different Packaging into Mineral Water. *Water Res.* **2018**, *129*, 154–162.

(70) PETRA PET Resin Association. *DID YOU KNOW? Little-Known Facts about PET Plastic*; 2015; http://www.petresin.org/pdf/DidYouKnow.pdf.

(71) IFA Institut für Arbeitsschutz der Deutschen Gesetzlichen Unfallversicherung. GESTIS Substance database http://www.dguv.de/ifa/gestis/gestis-stoffdatenbank/index-2.jsp

(accessed Feb 6, 2018).

(72) Wang, Y.; Kalinina, A.; Sun, T.; Nowack, B. Probabilistic Modeling of the Flows and Environmental Risks of Nano-Silica. *Sci. Total Environ.* **2016**, *545–546*, 67–76.

(73) Dris, R.; Gasperi, J.; Mirande, C.; Mandin, C.; Guerrouache, M.; Langlois, V.; Tassin, B. A First Overview of Textile Fibers, Including Microplastics, in Indoor and Outdoor Environments. *Environ. Pollut.* **2017**, *221*, 453–458.

(74) Carr, S. A. Sources and Dispersive Modes of Micro-Fibers in the Environment. *Integr. Environ. Assess. Manag.* **2017**, *13* (3), 466–469.

(75) De Falco, F.; Gullo, M. P.; Gentile, G.; Di Pace, E.; Cocca, M.; Gelabert, L.; Brouta-Agnésa, M.; Rovira, A.; Escudero, R.; Villalba, R.; et al. Evaluation of Microplastic Release Caused by Textile Washing Processes of Synthetic Fabrics. *Environ. Pollut.* **2018**, *236* (XXX), 916–925.

(76) Sillanpää, M.; Sainio, P. Release of Polyester and Cotton Fibers from Textiles in Machine Washings. *Environ. Sci. Pollut. Res.* **2017**.

(77) Hartline, N. L.; Bruce, N. J.; Karba, S. N.; Ruff, E. O.; Sonar, S. U.; Holden, P. A. Microfiber Masses Recovered From Conventional Machine Washing of New or Aged Garments. *Environ. Sci. Technol.* **2016**, *50* (21), 11532−11538.

(78) Liebezeit, G.; Dubaish, F. Microplastics in Beaches of the East Frisian Islands Spiekeroog and Kachelotplate. *Bull. Environ. Contam. Toxicol.* **2012**, *89* (1), 213–217.

(79) Browne, M. A.; Crump, P.; Niven, S. J.; Teuten, E.; Tonkin, A.; Galloway, T.; Thompson, R. Accumulation of Microplastic on Shorelines Woldwide: Sources and Sinks. *Environ. Sci. Technol.* **2011**, *45* (21), 9175–9179.

(80) Napper, I. E.; Thompson, R. C. Release of Synthetic Microplastic Plastic Fibres from Domestic Washing Machines: Effects of Fabric Type and Washing Conditions. *Mar. Pollut. Bull.* **2016**, *112* (1–2), 39–45.

(81) Ellen Mac Arthur Foundation. *A New Textiles Economy: Redesigning Fashion' Future*; 2017; https://www.ellenmacarthurfoundation.org/assets/downloads/publications/A-New-Textiles-Economy_Full-Report.pdf.

(82) Levi Strauss & CO. *The Life Cycle Of A Jean*; 2015; http://levistrauss.com/wp-content/uploads/2015/03/Full-LCA-Results-Deck-FINAL.pdf.

(83) Lassen, C.; Hansen, S. F.; Magnusson, K.; Norén, F.; Bloch Hartmann, N. I.; Jensen, P. R.; Nielsen, T. G.; Brinch, A. *Microplastics: Occurrence, Effects and Sources of Releases to the Environment in Denmark*; The Danish Environmental Protection Agency, 2015; http://orbit.dtu.dk/files/118180844/Lassen_et_al._2015.pdf.

(84) Beton, A.; Dias, D.; Farrant, L.; Gibon, T.; Le Guern, Y.; Desaxce, M.; Perwuelzz, A.; Cordella,

M.; Dodd, N. *Environmental Improvement Potential of Textiles (IMPRO Textiles)*; 2014; http://publications.jrc.ec.europa.eu/repository/bitstream/JRC85895/impro textiles_final report edited_pubsy web.pdf.

(85) Giorgi, S. *Non-Clothing Textiles : Consumer Purchasing, Use and Discard Practices*; 2015; http://www.wrap.org.uk/content/household-textiles?utm_source=WRAP+Newsletters&utm_campaign=9dc79d0f28-SCAP+ezine+December+2015&utm_medium=email&utm_term=0_165af891aa-9dc79d0f28-4622653%5Cnhttp://www.wrap.org.uk/sites/files/wrap/REC601-001 NCT Final Report.

(86) Byrne, M. A.; Kearns, G.; McKenna, C. A Test Chamber Study of the Contribution of Human Body Surfaces to the "Personal Cloud." In *Indoor Air 2002, 9th International conference on Indoor Air Quality and Climate*; Monterey, California, 2002.

(87) Stark, M. Microplastics and Textile Fibers – A Critical Review. *Melliand Int.* **2017**, *22* (3), 135–139.

(88) Whigham, L. D.; Schoeller, D. A.; Johnson, L. K.; Atkinson, R. L. Effect of Clothing Weight on Body Weight. *Int. J. Obes.* **2013**, *37* (1), 160–161.

(89) Althoff, T.; Sosič, R.; Hicks, J. L.; King, A. C.; Delp, S. L.; Leskovec, J. Large-Scale Physical Activity Data Reveal Worldwide Activity Inequality. *Nature* **2017**, *547* (7663), 336–339.

(90) Browning, R. C.; Baker, E. A.; Herron, J. A.; Kram, R. Effects of Obesity and Sex on the Energetic Cost and Preferred Speed of Walking. *J. Appl. Physiol.* **2006**, *100* (2), 390–398.

(91) Yoon, Y. H.; Brimblecombe, P. Clothing as a Source of Fibres within Museums. *J. Cult. Herit.* **2000**, *1* (4), 445–454.

(92) Vigo, T. L.; Lamb, G. E. R.; Kepka, S.; Miller, B. Abrasion and Lint Loss Properties of Fabrics Containing Crosslinked Polyethylene Glycol. *Text. Res. J.* **1991**, *61* (3), 169–176.

(93) Wiewel, B. V.; Lamoree, M. Geotextile Composition, Application and Ecotoxicology — A Review. *J. Hazard. Mater.* **2016**, *317*, 640–655.

(94) Hartline, N. L.; Bruce, N. J.; Karba, S. N.; Ruff, E. O.; Sonar, S. U.; Holden, P. A. Microfiber Masses Recovered from Conventional Machine Washing of New or Aged Garments. *Environ. Sci. Technol.* **2016**, *50* (21), 11532–11538.

(95) Rutsch, M.; Rieckermann, J.; Krebs, P. Quantification of Sewer Leakage: A Review. *Water Sci. Technol.* **2006**, *54* (6–7), 135–144.

(96) Talvitie, J.; Heinonen, M.; Pääkkönen, J.-P.; Vahtera, E.; Mikola, A.; Setälä, O.; Vahala, R. Do Wastewater Treatment Plants Act as a Potential Point Source of Microplastics? Preliminary Study in the Coastal Gulf of Finland , Baltic Sea. *Water Sci. Technol.* **2015**, *72* (9), 1495–1504.

(97) Murphy, F.; Ewins, C.; Carbonnier, F.; Quinn, B. Wastewater Treatment Works (WwTW) as a Source of Microplastics in the Aquatic Environment. *Environ. Sci. Technol.* **2016**, acs.est.5b05416.

(98) Talvitie, J.; Heinonen, M. *Preliminary Study on Synthetic Microfibers and Particles at a Municipal Waste Water Treatment Plant*; 2014; http://www.helcom.fi/Documents/HELCOM at work/Projects/BASE/Microplastics final report Base_final.pdf.

(99) Talvitie, J.; Mikola, A.; Setälä, O.; Heinonen, M.; Koistinen, A. How Well Is Microlitter Purified from Wastewater? A Detailed Study on the Stepwise Removal of Microlitter in a Tertiary Level Wastewater Treatment Plant. *Water Res.* **2017**, *109*, 164–172.

(100) Ziajahromi, S.; Neale, P. A.; Rintoul, L.; Leusch, F. D. L. Wastewater Treatment Plants as a Pathway for Microplastics: Development of a New Approach to Sample Wastewater-Based Microplastics. *Water Res.* **2017**, *112*, 93–99.

(101) Cabernard, L.; Durisch-Kaiser, E.; Vogel, J.-C.; Rensch, D.; Niederhauser, P.; AWEL Gewässerschutz. Mikroplastik in Abwasser Und Gewässern. *Aqua Gas* **2016**, *7* (8), 78–85.

(102) Mintenig, S. M.; Int-Veen, I.; Löder, M. G. J.; Primpke, S.; Gerdts, G. Identification of Microplastic in Effluents of Waste Water Treatment Plants Using Focal Plane Array-Based Micro-Fourier-Transform Infrared Imaging. *Water Res.* **2017**, *108*, 365–372.

(103) Carr, S. A.; Liu, J.; Tesoro, A. G. Transport and Fate of Microplastic Particles in Wastewater Treatment Plants. *Water Res.* **2016**, *91*, 174–182.

(104) Mutzner, L.; Staufer, P.; Ort, C. Model-Based Screening for Critical Wet-Weather Discharges Related to Micropollutants from Urban Areas. *Water Res.* **2016**, *104*, 547–557.

(105) Laube, A.; Vonplon, A. Élimination Des Boues d'épuration En Suisse - Recensement Des Quantités. *Doc. Environ. Off. fédéral l'environnement, des forêts du paysage* **2004**, *181*, 47.

(106) Dominguez, D.; Diggelmann, V.; Binggeli, S. *Élimination Des Composés Traces Organiques Dans Les Stations D'Épuration*; 2016; https://www.bafu.admin.ch/dam/bafu/fr/dokumente/wasser/uv-umwelt-vollzug/elimination_von_organischenspurenstoffenbeiabwasseranlagen.pdf.download.pdf/elimination_des_composestracesorganiquesdanslesstationsdepuratio.pdf.

(107) Kaufmann, P.; Herren, H. M. *Wohin Mit Dem Regenwasser - Beispiele Aus Der Praxis: Versickern Lassen, Zurückhalten, Oberflächlich Ableiten*; 2000; https://www.zg.ch/behoerden/baudirektion/amt-fuer-umwelt/a-bis-z-publikationen/merkblaetter/wasser/wohin-mit-dem-regenwasser-beispiele-aus-der-praxis.pdf/view.

(108) *Private Communication Swiss Water Association VSA, 22.01.2019*.

(109) Sun, T. Y.; Gottschalk, F.; Hungerbühler, K.; Nowack, B. Comprehensive Probabilistic Modelling of Environmental Emissions of Engineered Nanomaterials. *Environ. Pollut.* **2014**, *185*, 69–76.

(110) Carr, S. A.; Liu, J.; Tesoro, A. G. Transport and Fate of Microplastic Particles in Wastewater Treatment Plants. *Water Res.* **2016**, *91*, 174–182.

(111) Schneider, T. Dust and Fibers as a Cause of Indoor Environment Problems. *Scand. J. Work. Environ. Health* **2008**, No. 4, 10–17.

(112) Yamamoto, N.; Shendell, D. G.; Winer, A. M.; Zhang, J. Residential Air Exchange Rates in Three Major US Metropolitan Areas: Results from the Relationship among Indoor, Outdoor, and Personal Air Study 1999-2001. *Indoor Air* **2010**, *20* (1), 85–90.

(113) Moran, R. E.; Bennett, D. H.; Tancredi, D. J.; Wu, X. M.; Ritz, B.; Hertz-Picciotto, I. Frequency and Longitudinal Trends of Household Care Product Use. *Atmos. Environ.* **2012**, *55*, 417–424.

(114) Schubarth, C.; Weibel, F. *Die Bodennutzung in Der Schweiz. Resultate Der Arealstatistik*; 2013; Vol. 002–0901; https://www.bfs.admin.ch/bfsstatic/dam/assets/348986/master.

(115) Eurostat. Land cover overview by NUTS 2 regions [lan_lcv_ovw] http://ec.europa.eu/eurostat/product?code=lan_lcv_ovw&language=en&mode=view (accessed Jun 29, 2018).

12 Supporting Information for: A proxy-based approach to predict spatially resolved emissions of macro- and microplastics to the environment

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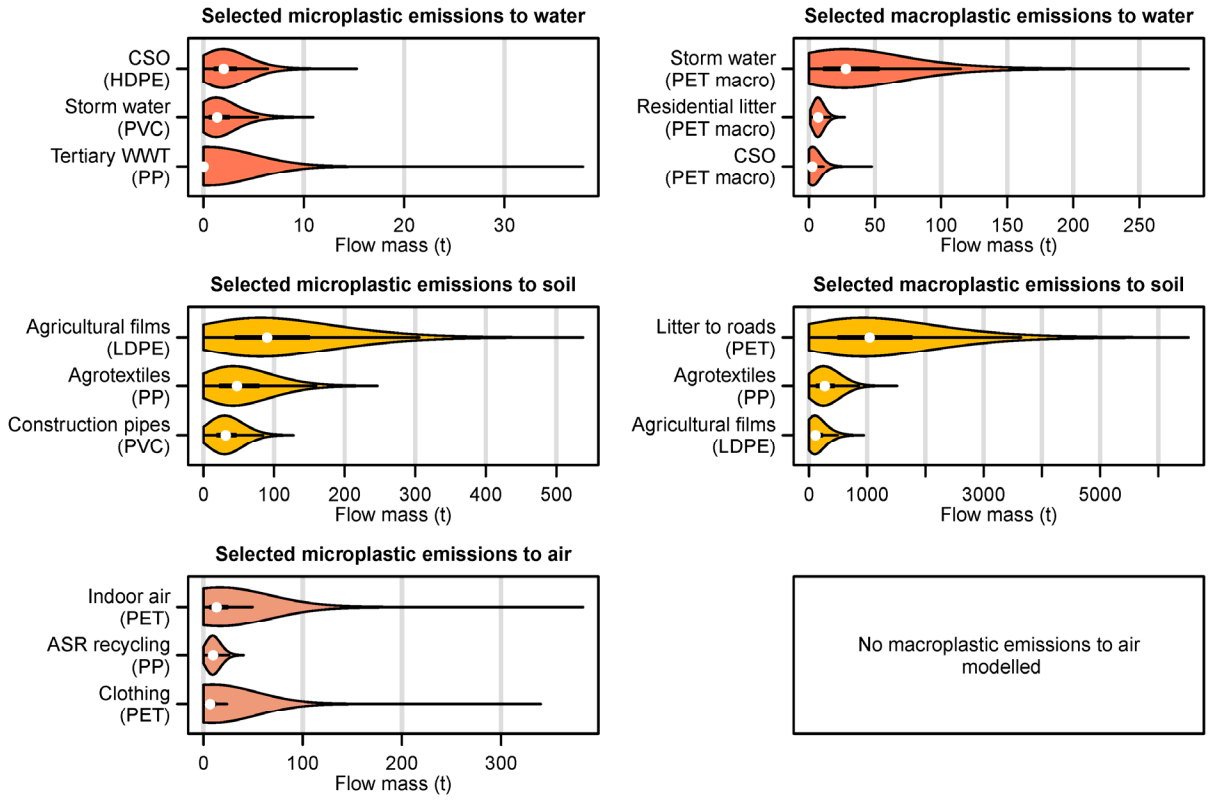

Figure S74: Examples of emission flow distributions to water, soil and air for specific polymers as macroplastic and microplastic. The type of polymer is given in brackets under the source name. The data presented in the violin plots originate from Kawecki and Nowack (2019)[1].

Table S36: Description of proxies used and the flows which were regionalized using these proxies.

Constructed geographical dataset	Dataset description	Reference for original data	Flows regionalized using this dataset
Population	Total permanent residing population in 2014	Statistique de la population et des ménages (STATPOP) dès 2010, Géodonnées de la statistique fédérale, Office fédéral de la statistique OFS	Litter (residential) to Residential soil Compost to Residential soil Fabric coatings to Residential soil (MP) Compost (MP) to Residential soil (MP) Clothing to Outdoor air (MP) Household textiles to Outdoor air (MP) Technical clothing to Outdoor air (MP) Technical household text. to Outdoor air (MP) Indoor air (MP) to Outdoor air (MP)
Newly built buildings	Buildings for habitation built between 2006 and 2010	Statistique des bâtiments et des logements (StatBL), Géodonnées de la statistique fédérale, Office fédéral de la statistique OFS	Geotextiles to Residential soil Building textiles to Residential soil Construction pipes to Residential soil (MP) Insulation (construction) to Outdoor air (MP)
Industry	Land use statistics divided into 17 classes for 2004/09. Considered classes for industry are : (1) industrial and artisanal area and (4) special infrastructure areas.	Statistique de la superficie selon nomenclature 2004 – Standard, Géodonnées de la statistique fédérale, Office fédéral de la statistique OFS	Packaging collection to Residential soil Mixed waste collection to Residential soil Agri. waste collection to Residential soil Text. waste collection to Residential soil Second. mat. production to Residential soil (MP) Transport to Residential soil (MP) Fibre production to Residential soil (MP) Non-text. manufacturing to Residential soil (MP) Pre-cons. plastic collection to Residential soil (MP) Pre-cons. fibre collection to Residential soil (MP) Packaging recycling to Residential soil (MP) C&D recycling to Residential soil (MP) Auto. large parts recycling to Residential soil (MP) ASR recycling to Residential soil (MP) WEEP recycling to Residential soil (MP) Agri. plastic recycling to Residential soil (MP) Industry water (MP) to Residential soil (MP) ASR recycling to Outdoor air (MP) WEEP recycling to Outdoor air (MP)
Agriculture	Land use statistics divided into 17 classes for 2004/09. Considered classes for agriculture are : (6) Fruit arboriculture, viticulture, horticulture and (7) Arable land.	Statistique de la superficie selon nomenclature 2004 – Standard, Géodonnées de la statistique fédérale, Office fédéral de la statistique OFS	Agricultural pack. films to Agricultural soil Agricultural pack. bottles to Agricultural soil Agricultural films to Agricultural soil Agricultural pipes to Agricultural soil Agricultural other to Agricultural soil Agrotextiles to Agricultural soil Compost to Agricultural soil Agricultural films to Agricultural soil (MP) Agricultural pipes to Agricultural soil (MP)

Constructed geographical dataset	Dataset description	Reference for original data	Flows regionalized using this dataset
			Agricultural other to Agricultural soil (MP) Agrotextiles to Agricultural soil (MP) Compost (MP) to Agricultural soil (MP)
Forest and other natural surfaces	Land use statistics divided into 17 classes for 2004/09. Considered classes for agriculture are : (8) Natural prairies, pastureland, (10) Forest, (11) Shrubland, (12) Other woods, (15) Unproductive vegetation, (16) Surfaces without vegetation	Statistique de la superficie selon nomenclature 2004 – Standard, Géodonnées de la statistique fédérale, Office fédéral de la statistique OFS	Shotgun cartridges to Natural soil Litter (nature) to Natural soil Fabric coatings to Natural soil (MP)
Water in natural environments	Cumulated number of cells attributed to a natural environment (see forest and other natural surfaces) in a distance of 500 m around the river segment or lake	[1] Statistique de la superficie selon nomenclature 2004 – Standard, Géodonnées de la statistique fédérale, Office fédéral de la statistique OFS. [2] Federal Office for the Environment FOEN. Géodonnées sur la subdivision de la Suisse en bassins versant.	Shotgun cartridges to Surface water Litter (nature) to Surface water
Water in residential environments	Cumulated population (see population) in a distance of 500 m around the river segment or lake	[1] Statistique de la population et des ménages (STATPOP) dès 2010, Géodonnées de la statistique fédérale, Office fédéral de la statistique OFS. [2] Federal Office for the Environment FOEN. Géodonnées sur la subdivision de la Suisse en bassins versant.	Litter (residential) to Surface water
Traffic	Modelled average number of vehicles per day on road segment transformed to raster. All road segments corresponding to tunnels were removed from the dataset.	sonBASE – Verkehrsdaten Schweiz 2015, Senozon AG 2017 im Auftrag des Bundesamts für Umwelt BAFUs	Automotive to Road side Litter (roads) to Road side

Constructed geographical dataset	**Dataset description**	**Reference for original data**	**Flows regionalized using this dataset**
WWTP: secondary treatment	Data recombined from several datasets, see description in article	[1] Federal Office for the Environment FOEN. Adresses Des Stations d'épuration Avec Mention de Leur Capacité de Traitement; 2017. [2] OSM Nominatim, OpenStreetMap Foundation. OpenStreetMap [3] Federal Office for the Environment FOEN. VSA Kennzahlen. Obtained upon Request, 2011.	Secondary WWT (small macroplastic) to Surface water Secondary WWT (MP) to Surface water (MP)
WWTP: tertiary treatment	Data recombined from several datasets, see description in article	[1] Federal Office for the Environment FOEN. Adresses Des Stations d'épuration Avec Mention de Leur Capacité de Traitement; 2017. [2] OSM Nominatim, OpenStreetMap Foundation. OpenStreetMap [3] Federal Office for the Environment FOEN. VSA Kennzahlen. Obtained upon Request, 2011.	Tertiary WWT (MP) to Surface water (MP)
CSO	CSO locations assumed to be at the WWTP or the lowest point of the largest water body	Mutzner et al. 2016[2]	Storm water to Surface water CSO to Surface water CSO (small macroplastic) to Surface water Industry water (MP) to Surface water (MP) Storm water (MP) to Surface water (MP) CSO (MP) to Surface water (MP)

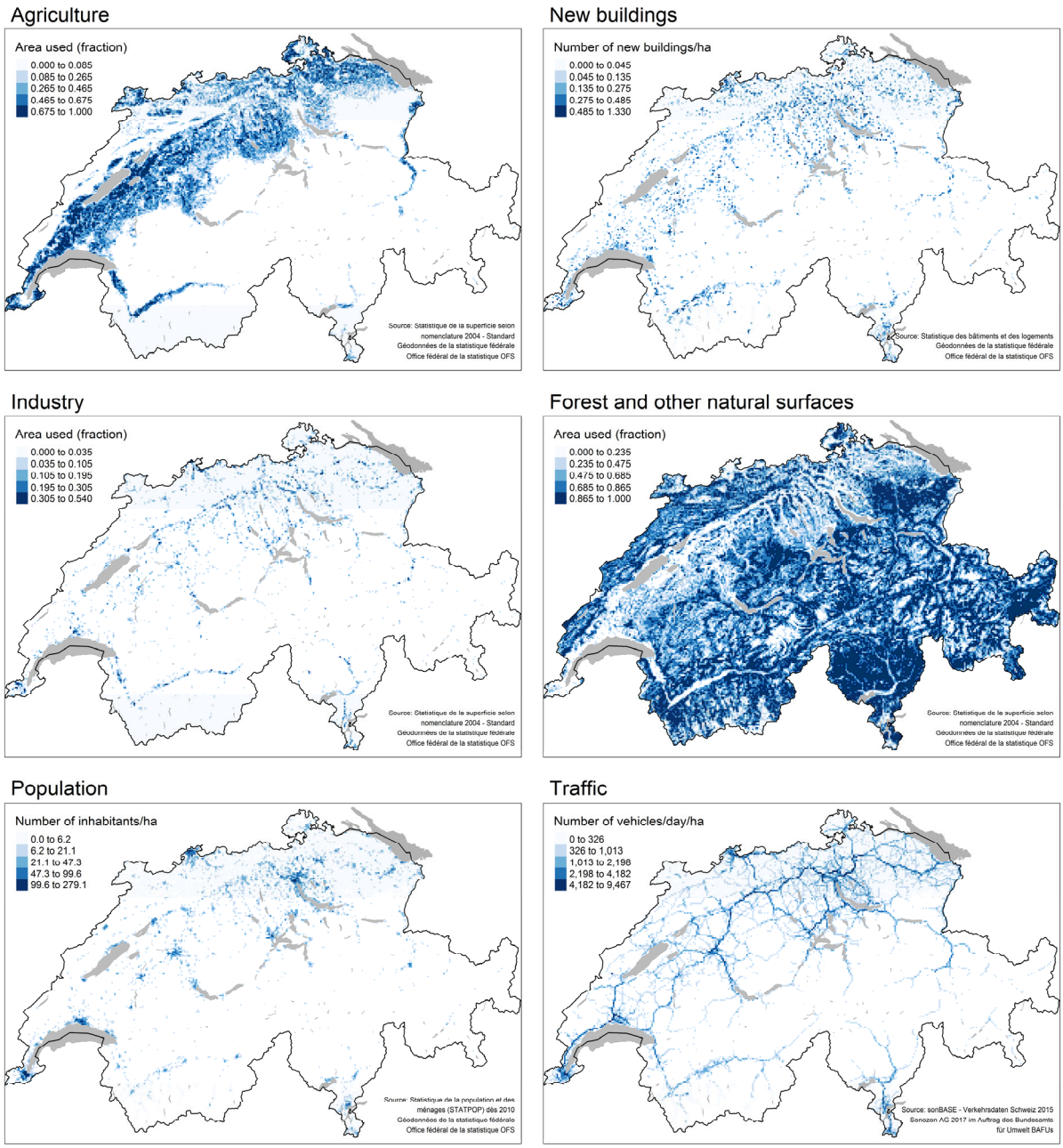

Figure S75: Proxies used for the regionalization of the flows to soil and air.

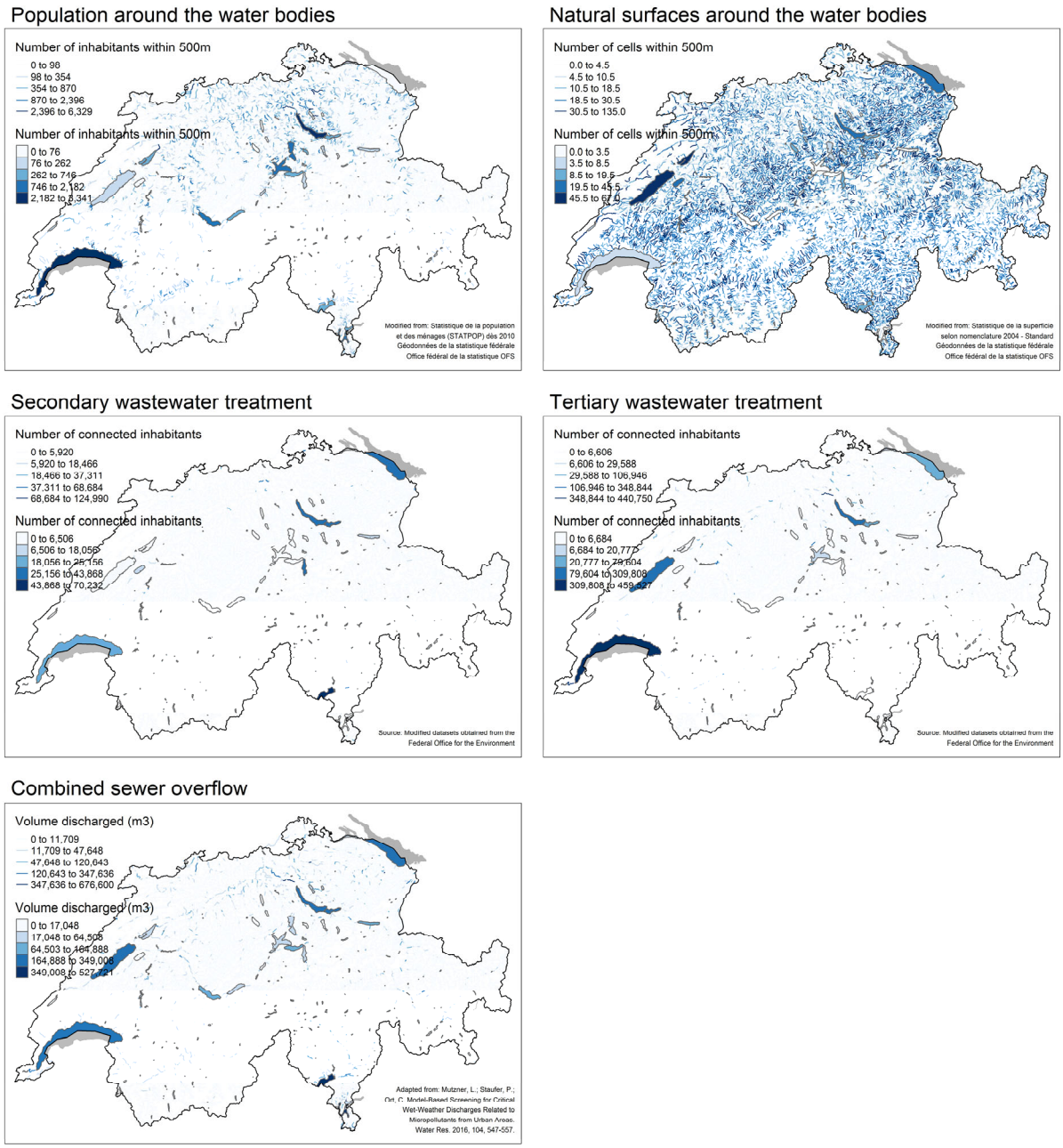

Figure S76: Proxies used for the regionalization of the flows to water.

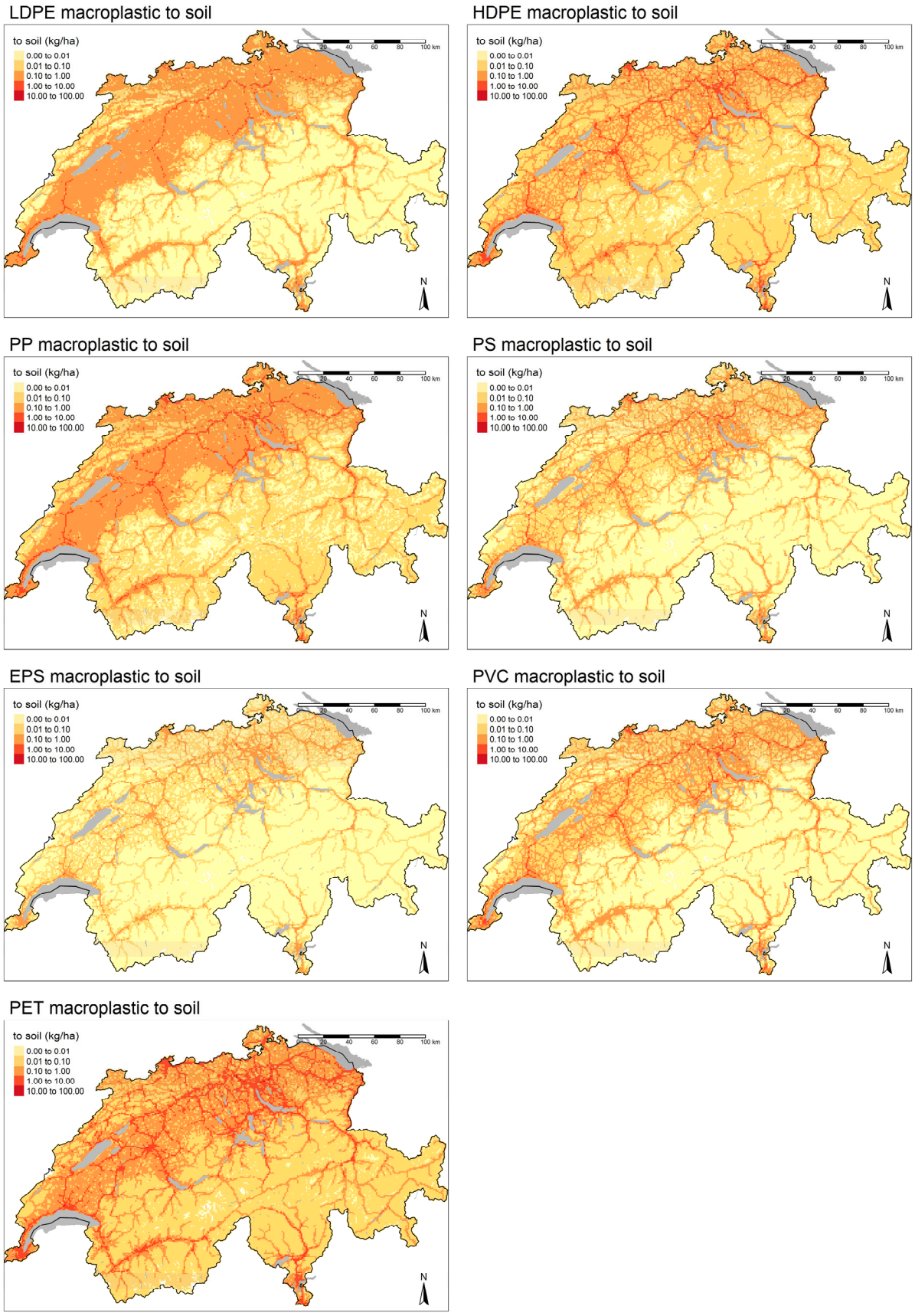

Figure S77: Maps of the emissions of macroplastic to soil.

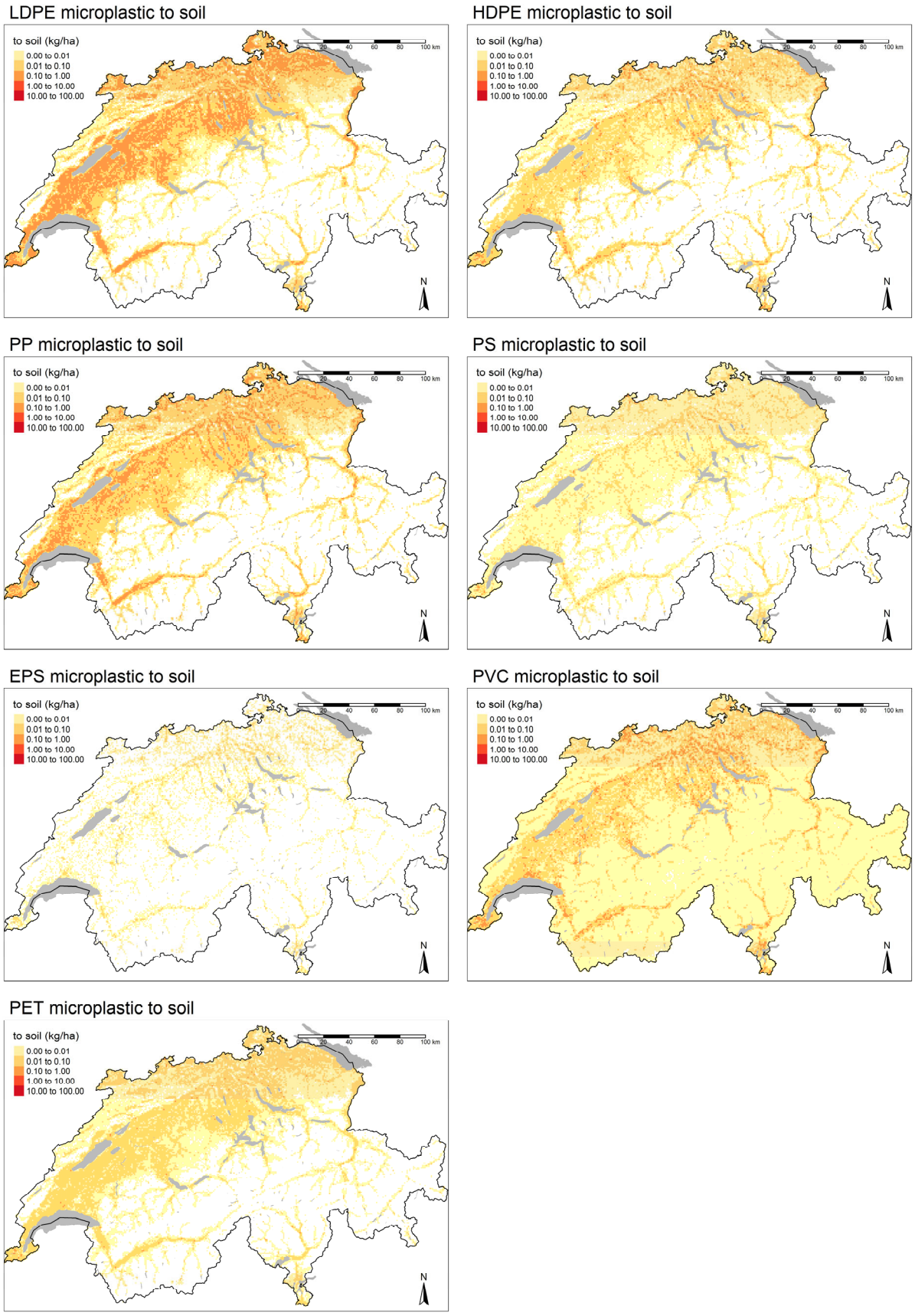

Figure S78: Maps of the emissions of microplastic to soil.

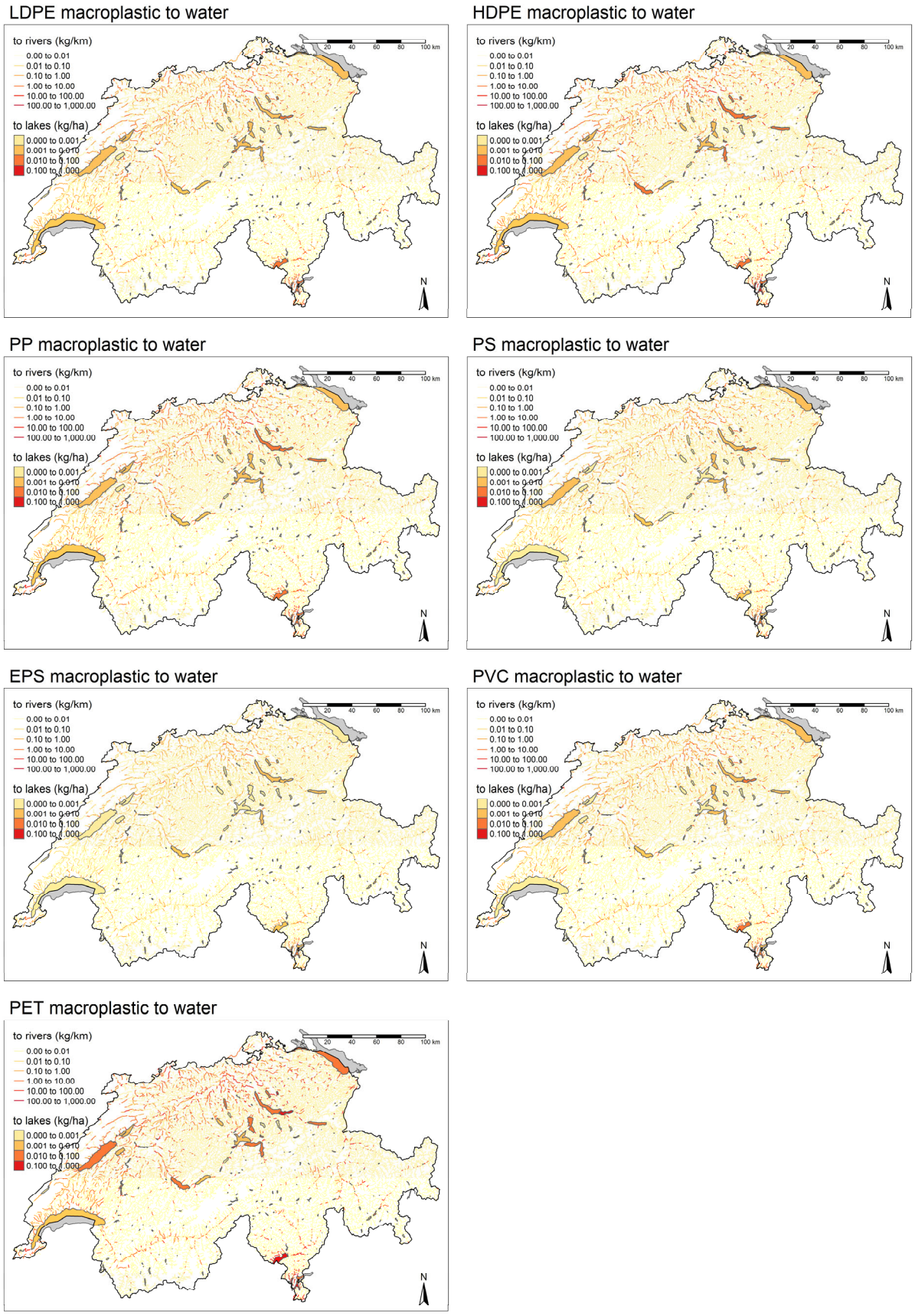

Figure S79: Maps of the emissions of macroplastic to water.

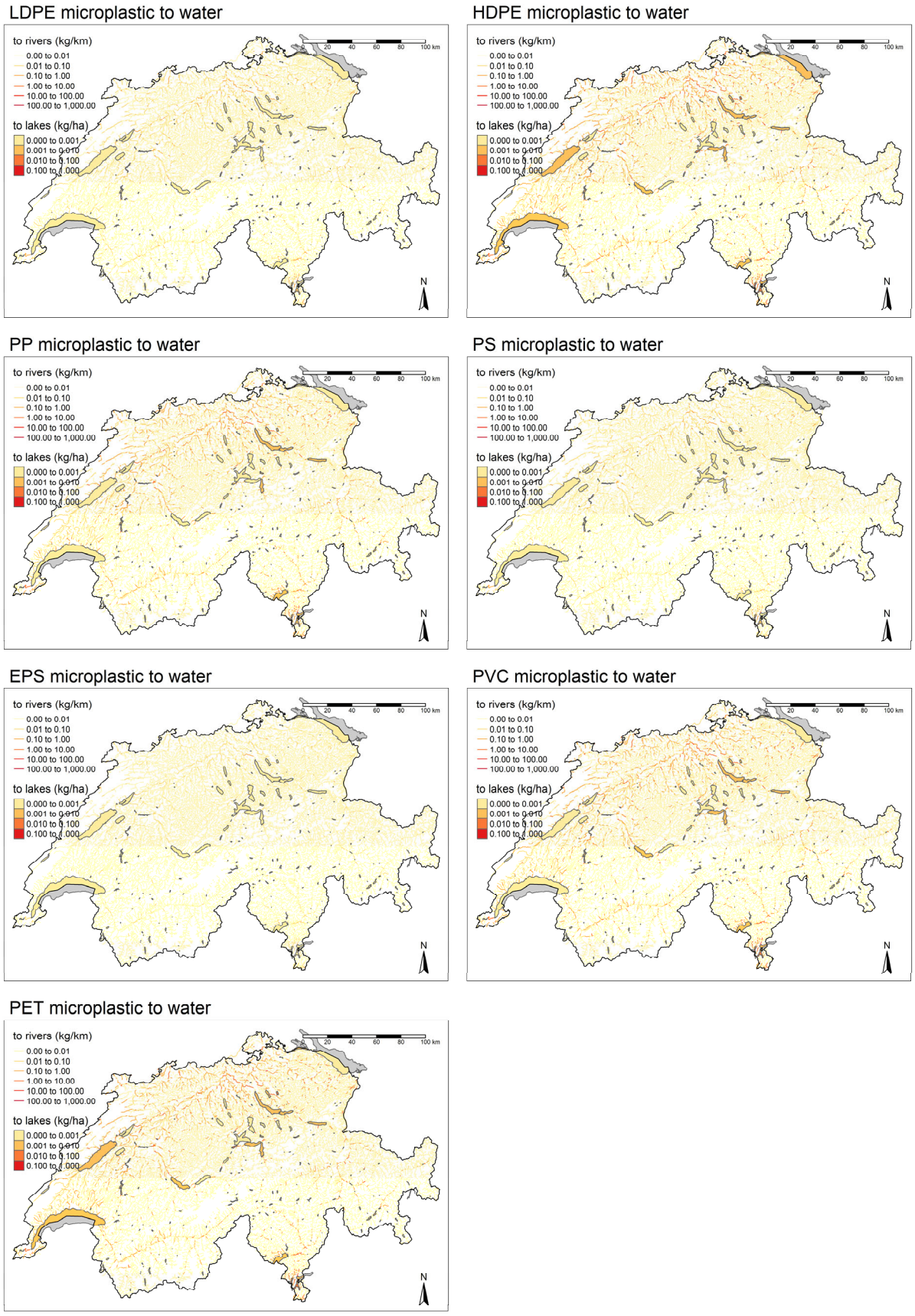

Figure S80: Maps of the emissions of microplastic to water.

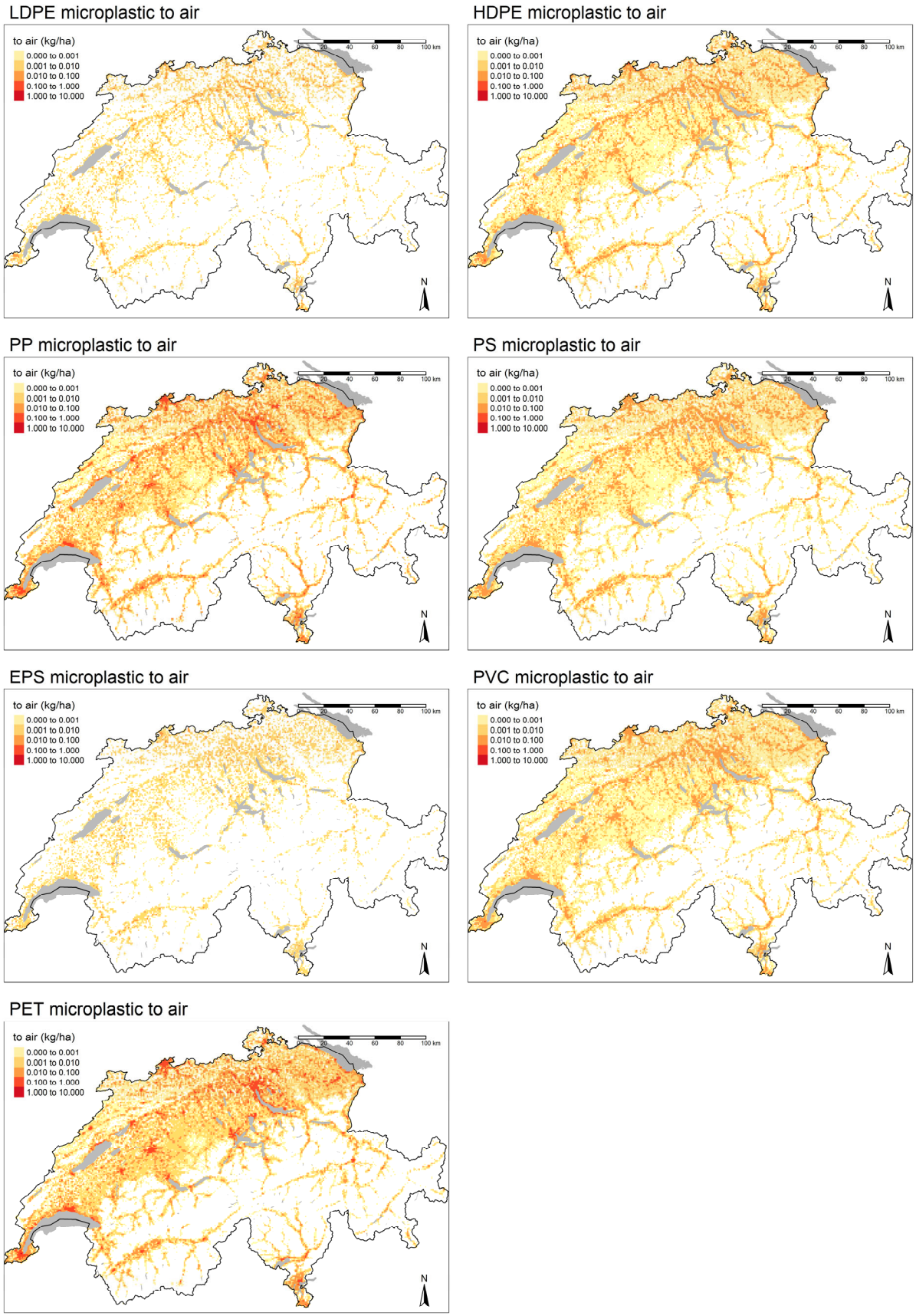

Figure S81: Maps of the emissions of microplastic to air.

12.1 References

(1) Kawecki, D.; Nowack, B. Polymer-Specific Modeling of the Environmental Emissions of Seven Commodity Plastics As Macro- and Microplastics. *Environ. Sci. Technol.* **2019**, *53* (16), 9664–9676.

(2) Mutzner, L.; Staufer, P.; Ort, C. Model-Based Screening for Critical Wet-Weather Discharges Related to Micropollutants from Urban Areas. *Water Res.* **2016**, *104*, 547–557.

13 Curriculum Vitae

Education

2016-2019	PhD candidate at ETH Zürich, Switzerland
2013-2015	Master of Sciences in Physics at Uppsala University, Sweden *Meteorology, climate, molecular physics and chemistry*
2010-2013	Bachelor of Sciences in Physics at Fribourg University, Switzerland *Physics, mathematics, chemistry*
2006-2009	High school in Ferney-Voltaire, France

14 List of publications, presentations and posters

14.1 Peer-reviewed publications

Sieber, R; **Kawecki, D.**; Nowack, B. Dynamic probabilistic material flow analysis of rubber release from tires into the environment. Environ. Poll. 2020, 113573 (258).

Wigger, H.; **Kawecki, D.**; Nowack, B.; Adam, V. Systematic Consideration of Parameter Uncertainty and Variability in Probabilistic Species Sensitivity Distributions. Integr. Environ. Assess. Manag. 2019.

Kawecki, D.; Nowack, B. Polymer-Specific Modeling of the Environmental Emissions of Seven Commodity Plastics As Macro- and Microplastics. Environ. Sci. Technol. 2019, 53 (16), 9664–9676.

Kawecki, D.; Scheeder, P.; Nowack, B. Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe. Environ. Sci. Technol. 2018, 52, 9874–9888.

Werner, J.; Persson, I.; Björneholm, O.; **Kawecki, D**.; Saak, C.; Walz, M.; Ekholm, V.; Unger, I.; Valtl, C.; Caleman, C.; Örhwall, G. and Prisle, N. Shifted equilibria of organic acids and bases in the aqueous surface region. Physical Chemistry Chemical Physics. 2018, 20, 36, 23281-23293.

14.2 Non peer-reviewed publications

Kalberer, A.; **Kawecki-Wenger, D**; Bucheli, T. D. Plastikströme in der Schweizer Landwirtschaft und ihr Risikopotenzial für Böden. Agrarforschung Schweiz. 2019, 10 (11–12), 416–423.

14.3 Reports

Kalberer, A.; **Kawecki-Wenger, D**; Bucheli, T. D. Plastik in der Landwirtschaft. Stand des Wissens und Handlungsempfehlungen für die landwirtschaftliche Forschung, Praxis, Industrie und Behörden. Agroscope Science. 2019, 89.

Wenger, D. Acid-Base Behaviour of Short-Chain Fatty Acids on Aqueous Surfaces: Using X-Ray Photoelectron Spectroscopy, Nuclear Magnetic Resonance and Molecular Dynamics. Master thesis. Uppsala University, Disciplinary Domain of Science and Technology, Physics, Department of Physics and Astronomy. 2015.

Wenger, D. Acid and Base Behaviour in Water Clusters. Student thesis. Uppsala University, Disciplinary Domain of Science and Technology, Physics, Department of Physics and Astronomy. 2015.

14.4 Conference presentations

Kawecki-Wenger, D; Nowack, B. Spatially resolved environmental emissions of seven commodity plastics in Switzerland. SETAC Helsinki. 25.05.2019.

Kawecki-Wenger, D; Nowack, B. Environmental flows of macro- and microplastics for seven different polymers using Material Flow Analysis. Microplastics 2018 Conference. Monte Verita. 29.10.2018.

Kawecki-Wenger, D; Nowack, B. Modelling of the environmental release of macro- and microplastics for seven different polymers. SETAC Rome. 15.05.2018.

14.5 Other scientific presentations

Kawecki-Wenger, D; Nowack, B. Modelling plastic flows into the environment, RFA NAREP + Energy Colloquium, Empa Dübendorf. 01.10.2018.

Kawecki-Wenger, D; Nowack, B. Modelling of the emissions of macro- and microplastics for seven different polymers. Ecological Systems Design, ETH Zürich, 10.04.2018.

Wenger, D; Nowack, B. Modelling the flows of micro- and macroplastics to the environment, Empa PhD Symposium. Empa Akademie, Dübendorf,. 13.11.2017.

14.6 Presentations to lay audiences

Kawecki-Wenger, D; Nowack, B. Modellierung der Plastikflüsse in die Umwelt. Verein Risiko & Sicherheit, Herbstanlass. 25.10.2018.

Wenger, D; Nowack, B. Flux de plastique vers l'environnement: Microplastiques et macroplastiques - Vue d'ensemble des sources principales. TecOrbe, Yverdon-les-Bains, 01.11.2017.

14.7 Posters

Kawecki-Wenger, D; Nowack, B. Modelling the environmental release of macro- and microplastics for seven different polymers, Empa PhD Symposium. Empa Akademie, Dübendorf,. 06.11.2018.

Wenger, D; Scheeder, P; Nowack, B. Mapping the flows of seven plastics through society: A first step towards building an environmental release model for plastics. SETAC Brussels. 01.05.2017.

Wenger, D.; Mitrano, D. M.; Nowack, B. Modelling the Flows of Plastics and Microplastics to the Environment. Wasserchemische Gesellschaft, 7th Late Summer Workshop, Microplastics in the aquatic environment. 25-28.09.2016