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## **Environmental Modeling**

# **Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe**

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# Probabilistic Material Flow Analysis of Seven Commodity Plastics in Europe

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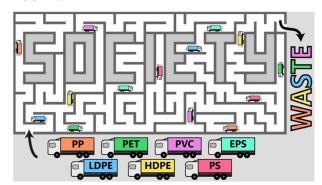
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#### 12 **TOC-Art:**



## **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 anthropospheres 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.

## Introduction

The European plastic production has increased by a factor of 160 between 1960 and 2010<sup>1</sup>, and the use of plastics is expected to continue to rise in the future<sup>2</sup> in a wide range of applications<sup>3</sup>. 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<sup>4</sup>. 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%)<sup>5</sup>. 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<sup>6</sup>.

Many questions regarding the environmental impacts of plastics and the health and environmental impacts of additives are being raised<sup>7</sup>. Between 1.15 Mt<sup>8</sup> and 12.7 Mt<sup>9</sup> 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<sup>10</sup>, 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<sup>11–14</sup>. 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)<sup>15</sup>. Multiple problems are caused by such pollutions, as for example chemical toxici-

ties due to residual monomers, plastic additives or adsorbed POPs<sup>16</sup>, with the additional threat of accumulation of microplastic along the trophic chain<sup>16</sup>. Moreover, it has been suggested that plastic additives have effects on human endocrine, reproductive and developmental functions<sup>17</sup> and could also be involved in other human health problems as for instance asthma<sup>18</sup>, due to their leaching from products<sup>19</sup>. 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<sup>20</sup>.

Depending on the life-cycle and the specific applications of a material, release pathways can be very different<sup>21</sup>. 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<sup>22</sup>. Many MFA studies on plastic flows in society have been published without distinguishing individual polymers<sup>2,23,32–34,24–31</sup>. 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<sup>35</sup>. The most recent PVC flow analysis concentrated on Europe<sup>36</sup> and modelled the in-use stock of various applications generated from 1960 to 2012. Other PVC-specific MFAs have focussed on Sweden<sup>35</sup>, Japan<sup>37</sup> and China<sup>38</sup>. The remaining polymer-specific MFAs have focussed on PET because of its potential for closed-loop recycling, once for the USA<sup>39</sup> and once for a Columbian city<sup>40</sup>. 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<sup>41</sup>, and one study described the flows of packaging in Austria while distinguishing polymers<sup>42</sup>. Consulting companies provide information on production, manufacturer's demand and/or waste management options for plastic in Europe<sup>1,4,43-45</sup>, Germany<sup>46-50</sup>, or Switzerland<sup>23</sup>, or for PVC in Germany<sup>51</sup>, without using the systematic approach of MFA and without including textile products. PlasticsEurope<sup>43</sup> 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<sup>45</sup> 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 worldwide 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 ef-

fect 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<sup>52,53</sup>. 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.

## Methods

#### **Materials considered**

The popularity of use<sup>4</sup> and the frequency of presence in the environment<sup>11,12</sup> 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<sup>54</sup>. 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.

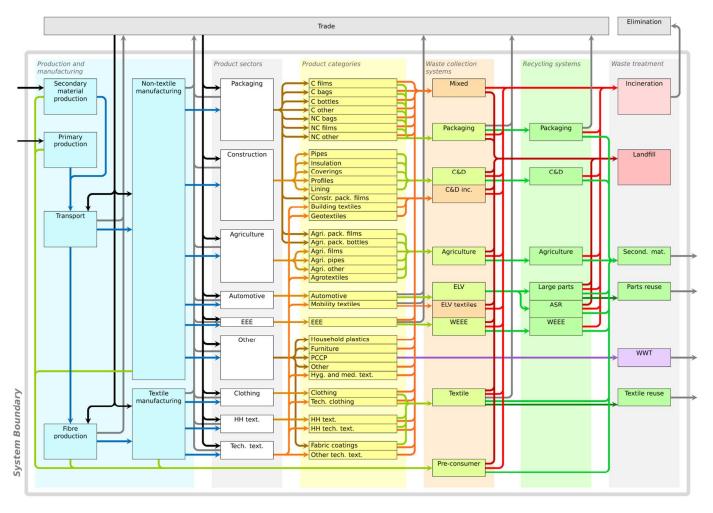
#### PMFA

The PMFA method used in this study has been described in detail elsewhere<sup>55</sup> and has already been applied to several materials<sup>56,57</sup>. 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<sup>22</sup>. 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<sup>6</sup> 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<sup>58</sup> and quality<sup>25</sup>. 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<sup>25</sup>, 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).

## 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 1). 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.



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Figure 1: 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. (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), 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 automotive 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.

## Input and transfer coefficients

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- 197 Amounts of produced polymers in primary forms were obtained from a market report for EU28+2 (de-
- 198 fined as European Union with Norway and Switzerland)<sup>45</sup> and from the Eurostat database<sup>59</sup> for EU28.
- 199 We assume that the error induced by the different geographic systems is negligible, as the plastic pro-
- 200 duction industry is not very strong in Norway and inexistent in Switzerland for the polymers consid-
- 201 ered<sup>45</sup>. The corresponding production values can be found in the SI (see table S2).
- Trade statistics can be obtained from the Eurostat<sup>60</sup> database for Europe and from the Swiss-Impex<sup>61</sup>
- 203 database for Switzerland, for a wide amount of goods characterized by codes using the Harmonized
- 204 System. Only net imports are considered (Imports Exports). If the net trade is positive, it is used as
- 205 parameter to create the corresponding Bayesian distribution from which the input vector will be sam-
- 206 pled. If the net trade is negative, the mean mass contained in the compartment is compared to the
- 207 outflow mass, and a Bayesian distribution is constructed around it. Additional information on trade can
- 208 be obtained from reports describing the shares of goods on the market that were imported or locally
- 209 produced, or the shares of waste management options for specific products. For some less known
- 210 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.
- 212 The parameters used to create the TC distributions were taken from several studies including market
- 213 reports, national reports and peer-reviewed literature. Data that was specific to the chosen geograph-
- 214 ical system, year and polymer was preferred but could not be found for all parameters. For the param-
- eters for which no system-specific data could be found, proxy data from other systems was used, while
- 216 accordingly adapting the uncertainty attributed to these parameters. Since over 600 TC distributions
- 217 needed to be defined in total for the two different geographical systems and seven polymers, a de-
- 218 tailed description is given in the SI. An overview of the literature used to create the TC and input distri-
- butions for Europe can be seen in Figure 2.

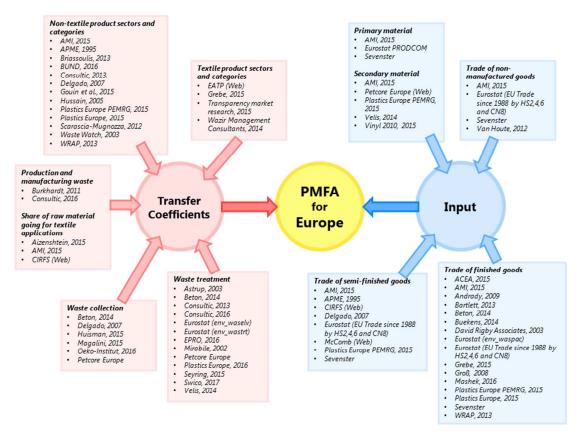


Figure 2: Summary of the data sources used for a quantification of the model parameters. A key to the references is available in Table S15. A similar figure for Switzerland, as well as the key to the references can be found in Figure S4 and Table S16.

#### 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 3 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 3A). 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 3B). 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 3C). 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 3D). This mass is an aggregate over the 35 product categories shown in yellow in Figure 1. It shows that the polymer most used is PP, followed by LDPE, PET, HDPE, PVC, PS and EPS.

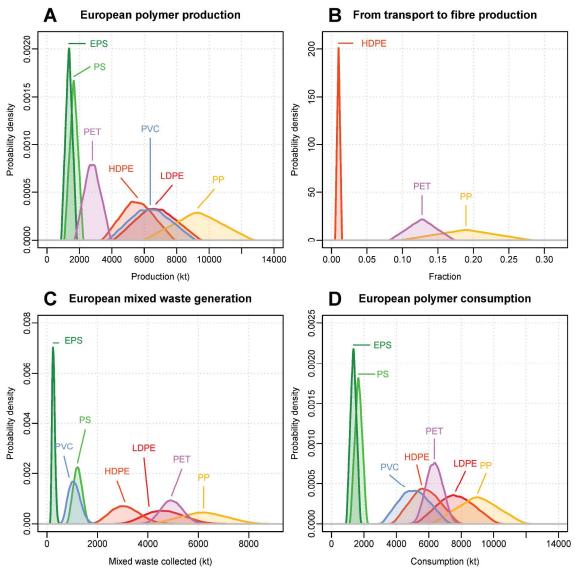
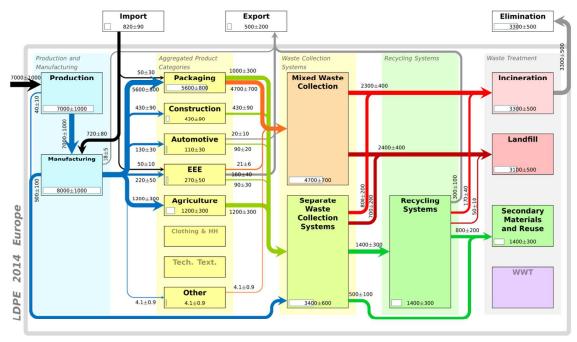


Figure 3: Selected probability distributions from the results of the Monte-Carlo simulation. Examples of (A) input, (B) transfer coefficient, (C) mass in a compartment and (D) aggregated mass over several compartments are shown.

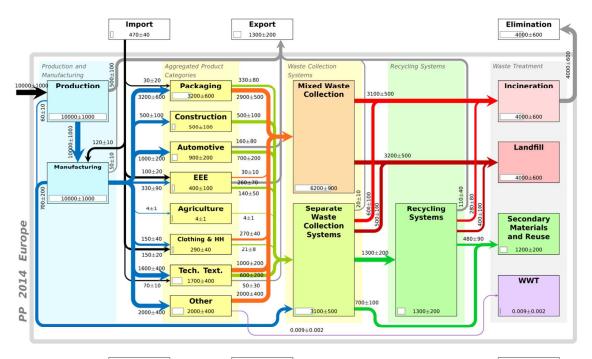
Simplified flow diagrams are shown for all seven polymers in Europe in Figure 4. In these diagrams, the processes production, manufacturing, separate waste collection, recycling and recycling and reuse are

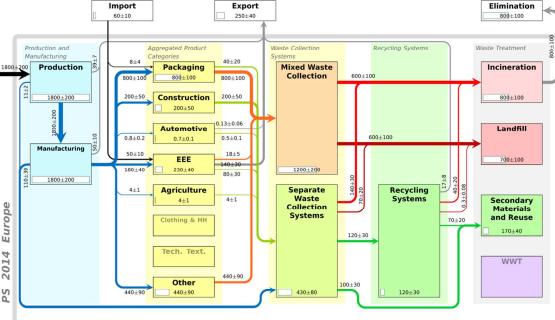
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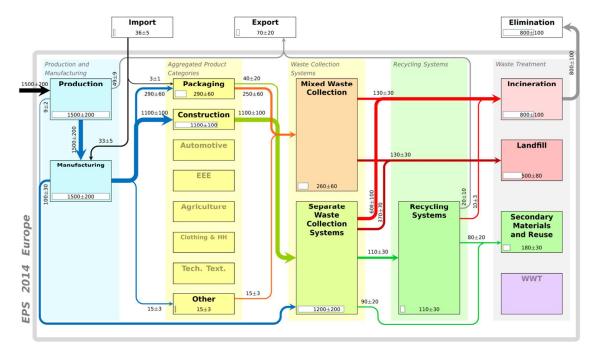
shown as aggregates over several compartments visible in Figure 1, with the purpose of improving readability. Furthermore, the product sectors are not presented in the simplified flow diagrams and the product categories are also not shown individually, but regrouped to improve legibility. Flows between these aggregated processes were aggregated so that no mass flow is neglected. Corresponding diagrams for Switzerland can be found in the SI (Figures S5-S11). The complete flow diagrams with all flows shown separately are given in the SI in Figures S16-S29. 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<sup>62</sup>.

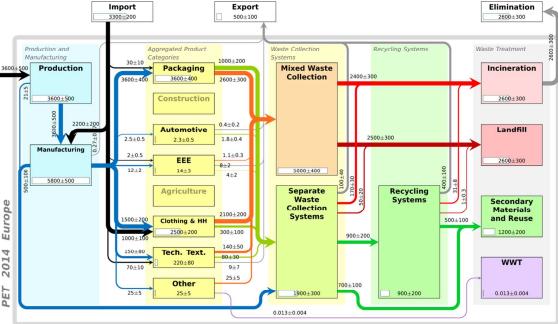


Elimination Import **Export** 600±200 2400±400 210±30 Aggregated Product Recycling Systems Waste Treatment Waste Collection Manufacturing 1000+300 6100 Production **Packaging Mixed Waste** Incineration Collection 3400±600 Construction 2400±400 1200±200 Landfill Automotive 15±4 EEE 6200±900 Agriculture Separate Recycling Systems Europe Waste Secondary Materials 13±3 13±3 Collection 10±2 and Reuse 800±200 Clothing & HH 5±2 10±2 1100±20 5.4±0.6 2014 35±8 Tech. Text wwt 2±1 HDPE Other 1400±300 4±1









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Figure 4: 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 Figures S5-S11 of the SI. The complete flow diagrams with all flows shown separately are given in Figures S16-29 of the SI. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

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In Europe, production is the dominating input for all polymers modelled.  $5600\pm900$  kt of HDPE are produced in Europe from raw materials,  $500\pm100$  kt from recycled material,  $150\pm20$  kt are imported as preliminary products and only  $70\pm20$  kt as finished products. This means that  $89\pm2\%$  of the plastic in circulation was produced locally. Similarly,  $81\pm3\%$  of LDPE,  $88\pm3\%$  of PP,  $88\pm2\%$  of PS,  $90\pm2\%$  of EPS,  $96\pm1\%$  of PVC of the input comes from the local plastic production (Figures S16-S22). For PET, only  $40\pm4\%$  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:  $0.5\pm0.1\%$  of HDPE,  $0\pm0\%$  of LDPE,  $5.3\pm0.7\%$  of PP,  $2.2\pm0.3\%$  of PS,  $3.3\pm0.5\%$  of EPS,  $15\pm2\%$  of PVC and  $0\pm0\%$  of PET. As could be expected from the parameters used, the waste produced at the production stage is very low.

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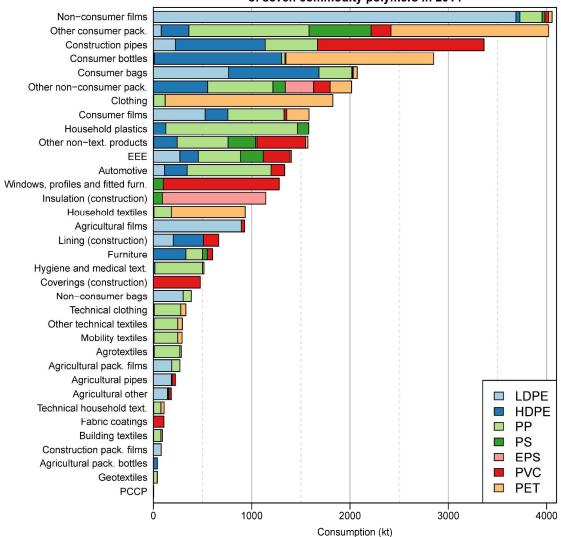
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A visual comparison of the consumption is given in Figure 5 for all product categories and polymers. The leading applications are found in packaging and construction applications, which are the two main application sectors for plastics. The most consumed 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 S13. 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.

# **European consumption** of seven commodity polymers in 2014



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Figure 5: 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 S13 of the SI.

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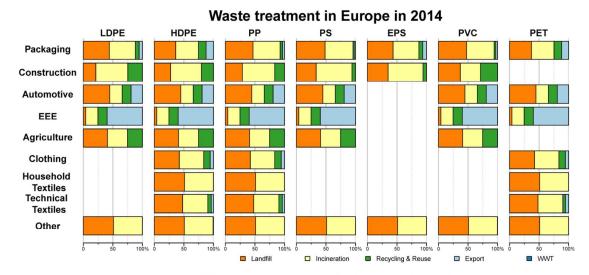
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In the simplified flow diagrams (Figure 4), the waste collection systems have been aggregated to improve the readability of the flows. In total, nine different separate waste collection systems are being modelled (Figure 1Error! Reference source not found.). 42±4% of LDPE waste is modelled to be collected separately (Figure S16), for the most part as agricultural plastics, but also as packaging preconsumer and construction waste. Similarly, 49±5% of HDPE waste is modelled to be collected separately (Figure S17), mostly as packaging and construction waste. Of the PP waste generated, 33±3% is collected separately (Figure S18) with the leading waste streams being technical textiles, pre-consumer 314 waste, construction and automotive plastics. Of all considered polymers, PS has the lowest separate 315 collection rate with only 26±3% (Figure S19), mainly from construction and pre-consumer waste. 316 82±3% of EPS (Figure S20) and 79±3% of end-of-life PVC (Figure S21) are collected separately, of 317 which the vast majority comes from construction applications. Last, only 28±3% of PET is collected 318 separately (Figure S22) with the main waste streams being packaging, textiles and pre-consumer 319 waste. 320 Figure 6 shows the final compartments for the seven polymers by application sector for both Europe 321 and Switzerland. The possible final compartments are: landfill, incineration, reuse, export and WWT. 322 Applications for which no information is shown are for polymers that are not used in that specific sec-323 tor. In Europe in most cases, the prevailing waste treatment options are incineration and landfills. Most 324 incineration and landfilling are observed for technical textiles, household textiles and Other products, 325 where recycling and reuse is very low or inexistent. Packaging also has a high share of incineration and 326 landfilling depending on the material. The highest recycle and reuse shares are accomplished for con-327 struction and agriculture applications. PVC in particular has the highest rate of recycling and reuse 328 (Figure S12) 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<sup>63</sup>. Most packaging material recycling 329 330 and reuse occurs for PET and HDPE, less so for LDPE and EPS, and even less for the remaining three. 331 For construction plastic, the highest recycling and reuse rate occurs for PVC, followed by LDPE, HDPE 332 and PP. The recycling and reuse of PS and EPS in construction is the least significant out of all poly-333 mers. There is no difference between the recycling and reuse of the different polymers used in auto-334 motive applications since this sector was not described in more detail with product categories. We can 335 expect the recycling and reuse of different polymers in the automotive sector to actually vary more, as 336 the reuse of car parts should most likely be more important for polymers that are used in large car 337 parts as for example PP, than for PS and PET which are used in smaller and less homogeneous parts<sup>64</sup>. 338 Recycling and reuse of textiles is important for clothing applications but less so for household applica-339 tions, and even less so for technical textiles. The small portion of recycling and reuse observed in Figure 6 for technical textiles is due to technical clothing and technical household textiles, which are as-340 341 sumed to follow the same recycling and reuse rates as consumer clothing and household textiles, and 342 to mobility textiles which are exported along with second-hand cars. Export is a significant outflow for 343 EEE and automotive applications, less so for packaging and clothing and barely significant for the re-344 maining applications. The WWT option only exists for the product category PCCP in the Other sector. 345 The share of this waste treatment option nevertheless disappears compared to other options, as this 346 application has a very low share of the consumption of PET, HDPE and PP (Figure 5). 347 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<sup>65</sup>. The fraction of waste 348 349 which is landfilled in Europe is instead incinerated in Switzerland. Another notable difference between 350 the two systems is the more prevailing export of second-hand vehicles and textiles out of Switzerland. 351 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<sup>66</sup>. 352



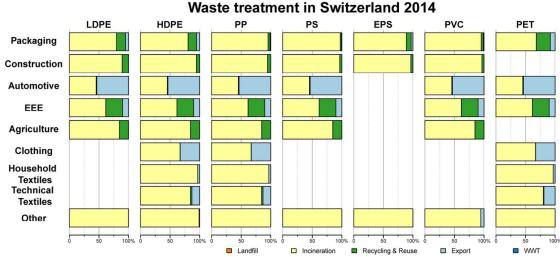


Figure 6: Ultimate compartment of the plastic shown for the nine application sectors and seven polymers. The bars missing are for polymers that are not used in that specific application. Abbreviations: EEE (Electrical and Electronic Equipment), WWT (Waste Water Treatment).

## **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<sup>36</sup>, 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<sup>23,24,33,34,25–32</sup>. 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<sup>4,67</sup> 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 shortcom-

ings by providing a transparent calculation of the total polymer life cycle, including trade flows and the textile life-cycle for the chosen polymers.

## Probabilistic approach

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Bayesian probabilities are a mathematical construction used to communicate reasonable expectations for a specific proposition<sup>68</sup>. 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 7). 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.

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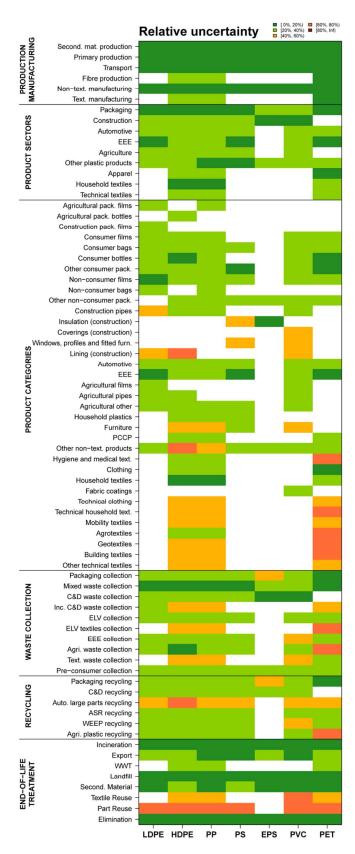


Figure 7: Heat map of the relative uncertainties (standard deviation divided by the distribution mean) associated to each compartment's mass for Europe in 2014. A lower uncertainty is shown in green, while a larger uncertainty is shown in red. White cells correspond to compartments with zero mass. A more de-

tailed version with the numerical values is given in Figure S15 of the SI, also including the respective values for Switzerland.

## Steady-state assumption

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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<sup>69</sup>, 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<sup>70</sup>), automotive (13 years<sup>70</sup>) and construction plastics (35 years<sup>70</sup>). 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.<sup>36</sup> 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<sup>73</sup>. 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.

#### Trade

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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<sup>25,36</sup> 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 (Figures S16-S29). 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.

#### **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

- 470 application. These results also highlight the relevance of a landfill ban in order to recover plastics more
- 471 efficiently.

# **Supporting Information**

- 473 Description of the method used, of the transfer coefficients and their derivation, data used for produc-
- 474 tion, 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 pro-
- 476 portions of polymer included in each considered trade category.

# 477 Acknowledgements

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