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## Evaluating the consumption of chemical products and articles as proxies for diffuse emissions to the environment†

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In this study we have evaluated the use of consumption of manufactured products (chemical products and articles) in the EU as proxies for diffuse emissions of chemicals to the environment. The content of chemical products is relatively well known. However, the content of articles (products defined by their shape rather than their composition) is less known and currently has to be estimated from chemicals that are known to occur in a small set of materials, such as plastics, that are part of the articles. Using trade and production data from Eurostat in combination with product composition data from a database on chemical content in materials (the Commodity Guide), we were able to calculate trends in the apparent consumption and in-use stocks for 768 chemicals in the EU for the period 2003–2016. The results showed that changes in the apparent consumption of these chemicals over time are smaller than in the consumption of corresponding products in which the chemicals are present. In general, our results suggest that little change in chemical consumption has occurred over the timespan studied, partly due to the financial crisis in 2008 which led to a sudden drop in the consumption, and partly due to the fact that each of the chemicals studied is present in a wide variety of products. Estimated in-use stocks of chemicals show an increasing trend over time, indicating that the mass of chemicals in articles in the EU, that could potentially be released to the environment, is increasing. The quantitative results from this study are associated with large uncertainties due to limitations of the available data. These limitations are highlighted in this study and further underline the current lack of transparency on chemicals in articles. Recommendations on how to address these limitations are also discussed.

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### Environmental significance

Emission data of chemicals can serve as a proxy for environmental concentrations which are useful to identify (i) chemicals of concern (for screening purposes), and (ii) changes in environmental trends of chemical contamination. As a large fraction of the emissions originates from manufactured products, it is important to quantify the consumption and in-use stocks of these products over time. There are however large gaps in our knowledge regarding the chemical composition of products which need to be addressed before emissions can be estimated in an accurate way.

## Introduction

The discovery of hazardous and persistent organic pollutants in the environment in the 1960's became the catalyst that started the development of management strategies and legislation in many countries to reduce the risk to human and environmental health caused by chemicals.<sup>1</sup> Since then, the production of chemicals has increased 60-fold by mass between the years 1950 and 2000, and global sales of chemicals have more than

doubled in 2004–2014.<sup>2</sup> A consequence of the ubiquitous use of chemicals in our modern society is that practically all human and wildlife populations and environmental compartments, no matter how remote or pristine, hold measurable amounts of chemicals originating from human activities.<sup>3,4</sup>

Assessing and understanding the effects on environmental and human health caused by chemicals is an immense challenge for society, as chemicals in the environment occur as complex mixtures of an unknown but broad range of chemicals with diverse properties and potential for negative effects. Even a seemingly simple question such as “is the threat from chemical pollution increasing or decreasing?” is difficult to answer. Yet, assessments of chemical or ecological status, or their equivalents, are key in European legislation and policy such as the European Water Framework Directive,<sup>5</sup> the Marine Strategy Framework Directive,<sup>6</sup> the European Ground water

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Directive,<sup>7</sup> the EU strategy for a non-toxic environment currently under development<sup>2</sup> and agreements for *e.g.* the Baltic Sea (HELCOM).<sup>8</sup>

The current strategy that society has taken to determine the environmental status with respect to contaminants is to identify a limited number of priority chemicals and assess their concentrations in the environment *versus* their environmental quality standard (*i.e.* set toxicity thresholds) on an individual-chemical basis. A significant drawback of this approach is that the selected priority substances are not necessarily good predictors of other toxic chemicals that may be present in the assessed compartment. In addition, combination effects are not addressed. Effect-based indicators are also used, but suffer from the drawback that the chemicals causing the effect are not easily identified and there are difficulties in defining the baseline. Assessment methodologies are under constant development to identify (more) relevant indicators to improve the success of chemical management to protect the environment and human health.<sup>9–11</sup>

Environmental monitoring as a part of chemical management includes analysis of chemicals in the environment, in humans, and observations of effects. Chemical analysis is costly and time demanding, and it is impossible to analyse all chemicals in use (many thousands). Thus, prioritizations are necessary. In Baltic Sea fish, for example, 105 substances or substance groups were analysed during the years 2000 and 2012, and the most frequently analysed chemicals were well known hazardous and regulated substances such as PCBs and dioxins.<sup>12,13</sup> Monitoring of substances, also of those that are already regulated, are of importance as time series of chemical concentrations in the environment, apart from indicating risk caused by chemical pollutants, are linked to chemicals management and can demonstrate effects of actions taken to reduce emissions of specific chemicals.<sup>14</sup> A biased focus on legacy pollutants in environmental monitoring can, however, result in a decreased potential to identify risks posed by chemicals that have an ongoing production and distribution to the environment. Monitoring efforts can also be limited for certain compounds, *e.g.* highly polar organic chemicals that are persistent and mobile, due to a lack of analytical methods to analyse them.<sup>15</sup>

Effect based monitoring includes joint effects of all chemicals present, but multiple confounding factors in the environment obscure the interpretation of status and time trends.<sup>16–18</sup>

A more indirect way to estimate environmental concentrations and to prioritize chemicals for further in-depth analysis and monitoring is to use emission data for chemicals. However, accurate data on emissions is missing for the majority of produced chemicals, and estimates from production volume and use have uncertainties spanning over several orders of magnitude,<sup>19–22</sup> due to lack of publicly available data from industry. A recent study however exploited confidential business data on use characteristics and total tonnage placed on the EU market, which is provided in REACH registration dossiers and available to EU Member State Competent Authorities, to rank substances with high intrinsic potential to contaminate aqueous environments.<sup>23</sup> Direct measurements of emissions are

limited to point sources. Emissions from industrial facilities in 39 countries (including non-European OECD members such as USA, Canada, Japan, Korea and Chile) are reported in the European Pollutant Release and Transfer Register (E-PRTR) with yearly data since 2011, but it only covers 91 pollutants,<sup>24</sup> out of the many thousands of potentially emitted chemicals. Diffuse emissions are, however, in many cases larger than those originating from point sources.<sup>25</sup> Diffuse emissions of chemicals into the environment originate from our use of thousands of chemical products (chemicals or mixtures thereof, such as pesticides, paints, cosmetics) and articles, which refers to products defined by their shape rather than composition,<sup>26</sup> such as furniture, electronic equipment and clothes. In this paper, we use the term “products” to encompass both chemical products and articles.

To estimate diffuse emissions of chemicals, it is necessary to quantify the release of chemicals from products. Within the European Union (EU), there are several regulations that address the chemical content of products. The composition of chemical products is fairly well known for cosmetics (including personal care products) since full disclosure of ingredients (although not mass fraction) is required in the European legislation.<sup>27</sup> Other chemical products fall under the EU regulation on chemicals<sup>26</sup> and the classification, labelling and packaging of substances and mixtures (CLP; European equivalent of the Globally Harmonized System of Classification and Labelling of Chemicals, GHS). The CLP requires product labels informing of chemicals classified as hazardous, and information about hazardous chemical content in safety data sheets when present in volumes exceeding 1% of the product.<sup>28</sup> Specific labelling is required for biocides, paint and varnishes and detergents when containing certain chemicals according to product specific regulations.<sup>29–32</sup> Such data on chemical composition of chemical products has been exploited in various studies estimating in particular indoor and human exposure.<sup>33–35</sup>

Emissions of chemicals from articles such as clothes and mobile phones, on the other hand, are difficult to estimate as no requirements on labelling of chemical content in articles exists, and information on composition is therefore lacking. Suppliers of articles containing chemicals that are listed as “Substances of Very High Concern” (SVHC) under REACH at concentrations exceeding 0.1% on a mass basis, need to inform downstream professional users or retailers of at least the names of these compounds (the same information should be available for private consumers upon request within 45 days).<sup>26,36</sup> This information is currently not yet available in a compiled form, but efforts to collect this information is ongoing. A recent amendment to the EU Waste framework directive for example requires suppliers to notify ECHA of all products containing SVHC's and have ECHA to make this information available to waste treatment operators and upon request also to the general public.<sup>37</sup>

Emissions of chemicals from articles, in particular to indoor-environments, have been quantified for a limited range of chemicals in specific articles or material categories. These studies employ either measured emission factors or models requiring chemical and/or article-specific parameters,



including material–ambient phase diffusion and partitioning coefficients.<sup>38–42</sup> A more generic model based on molecular structure has been developed to predict emissions of additives from polymers (evaluated for two phthalates in vinyl-flooring).<sup>43</sup> High-throughput estimates of emissions from articles are hampered by the fact that information on service life is required and models for multiple layer products are lacking.<sup>42,44</sup>

The scarce information on chemicals in articles is not entirely a consequence of lacking regulatory requirements. Complex supply chains with multiple sub-suppliers for many articles make information transfer challenging.<sup>45</sup> EU legislation requires only that information on chemical content is transferred to end-users for chemicals considered to be substances of very high concern<sup>26</sup> and for confirming the absence of certain regulated substances in specific articles.<sup>46,47</sup> However, the compliance with these requirements is frequently found to be violated.<sup>48,49</sup>

Taken together, weak regulatory requirements and violated compliance with those regulations result in patchy and uncertain data on emissions of chemicals from articles and products. In fact, available data on chemical content of products and articles is often coming from national environmental agencies that screen consumer products for a certain set of hazardous chemicals as part of regulation enforcement. The Swedish Chemicals Agency has developed a tool, The Commodity Guide, which to our knowledge is the only public database combining data on typical material composition of manufactured products identified by product codes employed in Eurostat, with typical chemical composition of major material categories.<sup>50</sup>

While data availability on chemical emissions and environmental concentrations is poor, economic data on production and trade is more plentiful (*e.g.* Eurostat). Consumption or production data has therefore been suggested as proxy for chemical emissions to the environment, *e.g.* by relating carbon dioxide emissions to consumption or industrial chemical emissions to components of final demand (finished products).<sup>51,52</sup> In this study, we combine trade data from Eurostat with chemical composition data in the Commodity guide to assess changes in mass flows of chemicals in products used within the EU (28 countries) during 2003–2016. Our aim is to explore and test the potential of this approach to aid in prioritization of chemicals for further screening and chemical

analysis. We discuss the calculated time trends for apparent product consumption, the subsequent chemical consumption and in-use stocks of products and chemicals that have built up over time. Data gaps and challenges remaining to develop consumption-based indicators of chemical emissions to the environment are highlighted.

## Material and methods

The work presented in this paper results from combining datasets from the European Union's Statistical Agency Eurostat and the Swedish Chemicals Agency. The data processing was done in KNIME (3.4.2 Knime gmbH, Konstanz, Germany) in combination with R (3.4.0 R Core Team 2017) and MariaDB (10.2.6 2000, 2017, Oracle, MariaDB Corporation Ab and others).

A detailed overview of the workflow is discussed below and illustrated in Fig. 1. In brief: production and trade data was taken from Eurostat's database EUROPROMS and combined with material and chemical data from the Swedish Chemicals Agency's Commodity Guide.

### Trade and production data in Eurostat

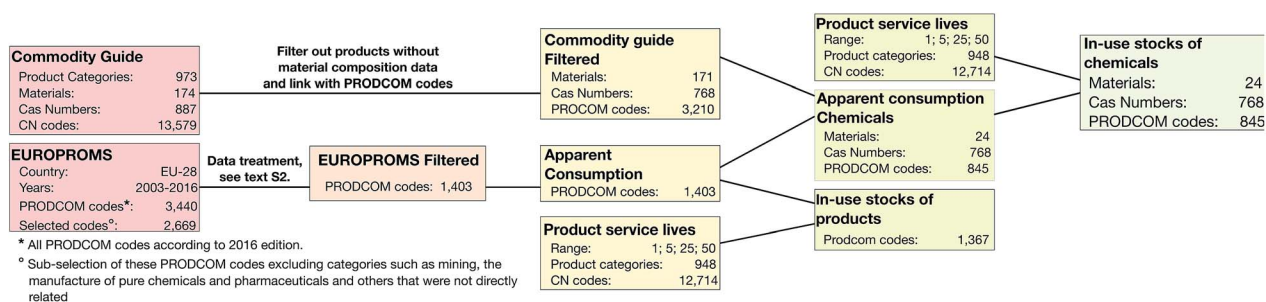
The apparent European consumption of products was calculated according to eqn (1):<sup>53</sup>

$$\text{Apparent EU consumption} = \text{production in EU} + \text{import into EU} - \text{export out of EU} \quad (1)$$

Data on imports, exports and production in the EU was available from the EUROSTAT EUROPROMS database and was given as monetary value in combination with a physical unit (*e.g.* mass, volume, pieces *etc.*).

EUROPROMS combines trade data (import and export) from COMEXT (an EU database for statistics on international trade in goods, listed under 8-digit Common Nomenclature, CN codes) with production data from PRODCOM (an EU database for industrial production statistics, listed under 8-digit PRODCOM codes).

Combining the two databases is possible as many of the product categories in PRODCOM and COMEXT can easily be converted into each other using conversion tables.



**Fig. 1** Overview of the data-treatment workflow applied in this study, illustrating how the data was combined and what data is remaining after each step. In summary: production and trade data was taken from Eurostat's database EUROPROMS and combined with material and chemical data from the Swedish Chemicals Agency's Commodity Guide. Product service lives were then used to calculate the in-use stocks from the yearly consumption.



Approximately 91% of all PRODCOM codes 7 (3440 out of 3789 codes) were found to correspond with CN codes in COMEXT (see also the file *ProdcomCodes\_Overview.csv* in the ESI†).

COMEXT data is collected from custom clearances while PRODCOM data originates from surveys sent out to a representative selection of producers. The matching of trade and production data in EUROPROMS to calculate the apparent consumption could therefore be biased for some products as the two datasets do not always share the same coverage.

### Treatment of trade and production data

Roughly 18% of all the yearly production values reported by individual member states in EUROPROMS were flagged as confidential (1995–2016, all countries), a procedure that protects individual companies from being identified in the statistics. One way to minimize data gaps due to confidential data is to use product categories aggregated at higher level such as 4-digit codes or 6-digit codes instead of 8-digits, but this comes at the cost of a lower resolution. Instead, data aggregated at EU-level was used here to limit the impact of missing or confidential data (*e.g.* exclusion of products from the data set due to discontinuous consumption time series). Data aggregated at EU-level is not confidential, although sometimes so-called rounded values are given, *i.e.* a range instead of a single value, see ESI Text S2.†

Data aggregated for the same 28 member states was available for years 2003–2016 (EU28). The focus of this study is on manufactured articles (*e.g.* clothes, furniture, electronic equipment, vehicles), building materials (*e.g.* ready-mixed concrete, cement), chemical products (*e.g.* cosmetics, paint, glue), and the packaging of food and beverages (see ESI Text S3† for details on how the mass of packaging material was estimated).

Data related to non-manufactured trade categories such as mining and the manufacture of pure chemicals and pharmaceuticals was excluded. In total, 2669 PRODCOM codes remained from the original database after excluding 771 PRODCOM codes covering products that were considered not of relevance to this study. An overview of all PRODCOM codes can be found in the ESI.†

For the purpose of this study, several modifications had to be made to the EUROPROMS dataset. These are described in detail in ESI Text S2.†

In brief: (1) the rounded values (see first paragraph in this section) were only used if their range was 50% or less of the reported value, otherwise the data entry was replaced by interpolating between adjacent years, or the PRODCOM code was excluded. (2) Only PRODCOM codes that were consistent over time or could be converted from old to new code numbers over the years 2003–2016 were included. (3) Missing single entries in Eurostat were replaced by linear interpolation between adjacent years. (4) In contrast to COMEXT data, data in PRODCOM and therefore EUROPROMS does not have to be reported in a unit of mass. Eurostat's own unit conversion factors were therefore used to convert all values to units of mass (kg). When this was not possible, the PRODCOM code was excluded. (5) Outliers in the dataset of calculated consumption were identified by

calculating the change between any two successive years and then flagging the top 1% largest changes. In this way, peaks were identified (caused *e.g.* by erroneous decimal spacing or reported units), which could in some cases be corrected by linear interpolation. In other cases, the large change between two years was followed by entries with similar values (*i.e.* the consumption changed swiftly from one level to another and stayed there for several years), making it difficult to classify the change as an error. PRODCOM codes with this behaviour were discarded. (6) PRODCOM codes with a negative calculated consumption were also discarded. Although this “book keeping” phenomenon is possible in PRODCOM where both sold production and total production are recorded and previously produced stocks are used during a time period, EUROPROMS only includes sold production. Negative consumption values are therefore likely to be due to inconsistencies in how import, export and production data were reported as production and trade data come from different sources.<sup>53,54</sup>

To avoid the introduction of artificial time trends in the dataset, we chose to discard rather than keep data of uncertain quality. We also observed a risk of double counting product masses as it is difficult to separate intermediate and final products in Eurostat's trade data. Rubber tires for example can be sold directly to consumers but can also go into the production of cars. The consumption of tires (or other products with multiple end-uses) could therefore be overestimated. Mass flow diagrams for certain products where the supply chain is well known exist and these could potentially be used to remediate the issue (*e.g.* for tantalum flows<sup>55</sup>). However, it is challenging and outside the scope of this study, to derive these mass flows for all types of products covered by Eurostat. Other solutions such as the use of broad economic categories (BEC)<sup>56</sup> that classify products according to their end use (consumer, intermediate and capital goods) exist as well, but their use in combination with EUROPROMS data is not straight forward as BEC categories are more broadly defined than PRODCOM codes. It is therefore not possible to give an estimate as to what extent the trade and production data in the filtered dataset is affected by double counting.

### Chemical and material composition of products

The Commodity Guide developed by the Swedish Chemicals Agency reports the material compositions of products as described in a Danish EPA report from 1995 (ref. 57) (updated in 2004 for motor vehicles<sup>50</sup>) based on a large survey among manufacturers. The chemical composition data of four types of materials – plastics, rubbers, cellulose fibers and textiles – are currently included in the database and originate from reports and handbooks published by the Swedish Chemicals Agency.<sup>50</sup>

The Commodity Guide contains data on 973 product categories, covering 174 materials and 887 distinct CAS numbers. For 768 out of 887 chemicals, there was quantitative data regarding their concentration in certain materials. These 768 chemicals have been included in this study.

This group of chemicals covers 20 different chemical functions and is dominated by chemicals that are used as dyes, flame retardants, pigments and stabilising agents (see Fig. S1†).



A large fraction of the chemicals has also been assigned multiple functions (*e.g.* both flame retardant and filler). Of the 768 chemicals, 61 are listed on the SIN-List,<sup>58</sup> 28 of which are also identified as SVHCs under REACH. In addition, 55 out of 768 have recently been identified as potential persistent mobile organic chemicals (PMOCs), a group of highly polar chemicals that is currently overlooked by most legislation as they do not bio-accumulate and can be difficult to analyse.<sup>23</sup>

For an overview of all chemicals covered, we refer the reader to the csv file *Chemical\_Trends\_Classes.csv*, which can be found in the ESI.†

The materials included in the database also cover the materials that make up the packaging.<sup>50</sup> Only 24 of the materials, however, are linked to information on chemical composition. In total, the product categories in the Commodity Guide cover 13 579 CN numbers which can be converted into 3210 PRODCOM codes (year 2016 edition) using the conversion tables available from EUROSTAT's metadata webpage RAMON.<sup>59</sup> Chemical composition data is available for 53% of these codes. Some of the data in the Commodity Guide is likely outdated, as the reports and handbooks were published between 1991 and 2006. Nevertheless, we here make the initial assumption that the composition of products has been constant over time.

All data in this study regarding material fractions and chemical content is based on the information in the Commodity Guide.

### Apparent consumption of materials and chemicals

The apparent consumption of materials was calculated according to eqn (2) and the apparent consumption of chemicals according to eqn (3):

$$\text{App. cons. (material } x) = \sum (\text{app. cons. PRODCOM}_y \times \text{material fraction}_{x \text{ in } y} (\%)) \quad (2)$$

$$\text{App. cons. (chemical } x) = \sum (\text{app. cons. material}_y \times \text{chemical fraction}_{x \text{ in } y} (\%)) \quad (3)$$

It was assumed that each of the 768 CAS codes represents a unique chemical or unique group of chemicals. No additional analysis was done to merge existing CAS codes that represent the same chemicals as this procedure itself might lead to new errors in the database (*e.g.* mismatches in online databases when looking up unique structural identifiers such as InCHI codes or SMILES<sup>60</sup>). Note however that the uncertainty introduced by ambiguous CAS numbers is small compared to the uncertainty and variability in chemical content of the various products.

### Product service lives and in-use stocks of products and chemicals

The in-use stocks of products and chemicals in the EU were calculated by summing up the consumption over the years, while taking product service lives into account. PRODCOM codes were linked to one of four service life alternatives (1, 5, 25, 50 years) estimated by experts at the Swedish Chemicals Agency (Personal communication, Stellan Fischer).

Estimated service lives were in general very short for beverages (packaging only), food products (packaging only), tobacco products and paper products (1 year) and long ( $\geq 25$  years) for 2-digit categories such as machinery and equipment, furniture, fabricated metal products, electrical equipment, computer and electronic equipment, rubber and plastic products, other transport equipment and non-metallic mineral products. The eight remaining 2-digit categories had more evenly distributed service lives (see ESI†). The estimated service lives for certain products are quite high (*e.g.* laptops, cell phones and flat screen TV's all have service lives of 25 years), which is likely an over-estimation. As the longest service lives were longer than the time period focused on in this study, the consumption of products in 1950 to 2003 were estimated to serve as the basis for the initial in-use stock, adjusted for respective service life per category (see ESI Text S4† for more details).

### Evaluating changes in consumption and in-use stocks over time

Significantly increasing or decreasing trends in the time series were identified using Mann–Kendal rank correlation tests ( $\alpha = 5\%$ , 2-sided), using the package “Kendall” in R.<sup>61</sup> The magnitude of change was estimated by calculating the ratio of the average consumption in the last three years (2014–2016) and in the first three years (2003–2005) of data. The three-year average was used to reduce the influence of temporary shifts in consumption.

## Results and discussion

### Quality and consistency of trade data

There were several obstacles to overcome in order to analyse the change in consumption of manufactured products in the EU and chemicals therein. Of the 2669 PRODCOM codes that were initially selected for this study (*i.e.* excluding non-manufactured trade categories such as mining and the manufacture of pure chemicals and pharmaceuticals), only half (1403) remained for inclusion in the time trend analysis after filtering and correcting/removing outliers (see Table S1†). This dataset is from here on referred to as the “Filtered dataset” and forms the basis for the results presented below. Most PRODCOM codes were excluded during the unit conversion step, where 778 codes were discarded since no mass data was available and conversion from their respective unit into kilograms was not possible. Note that all chemicals in the Commodity guide linked to any material data could also be linked to the PRODCOM codes in the filtered dataset.

Most of the PRODCOM codes in the filtered dataset could be linked to material composition data listed in the Commodity Guide (97% of all codes, representing 99% of the mass). In contrast, only 60% of these PRODCOM codes could be linked to any chemical data. The average mass of materials with chemical data made up just 8% of the total mass of materials. Note that roughly 35% (in 2016) of the total consumed mass of materials consisted of materials such as solid wood and mineral material (*e.g.* stone) where chemical additives are often lacking. However, for the major fraction of the consumed mass (57%), no information on chemical content was available.



## Consumption and in-use stocks of chemical products and articles

The estimated consumption of products aggregated at the main product category level (2-digit level) was largely influenced by the financial crisis in 2008, with a large decrease in total consumed mass after the peak in 2007 for the majority of the product categories (see Fig. 2). In many instances, the consumption was still below peak values almost 10 years after the crisis. Changes between the average consumption of products in the first and last three years were less than a factor two for all 2-digit product categories. The size of the total in-use stock of each product group varied greatly depending on expected service life (see Fig. 2). Some products have “accumulated” in society because of long service lives and increasing demand. A long service life means a considerable lag time between changes in consumption and changes in in-use stocks, *i.e.* a buffer towards temporary shifts in consumption such as during the financial crisis.

The total yearly consumption was dominated by the consumption of “non-metallic mineral products” such as concrete, cement, building blocks, bricks and tiles, which contributed roughly 60% of the total mass. Second largest was “food products” (packaging only) making up *ca.* 7% of the total mass. Other 2-digit categories following in order of decreasing consumed mass were: paper products, rubber & plastics, metal products, and chemical products (see Fig. 2 and S3† for more detail). Due to their long service life and large mass, products in the category non-metallic mineral consumption also made up

the largest contribution to the total in-use stock of products in Europe (85%), followed by the groups: fabricated metal products (3.7%), rubber & plastics (2.9%), motor vehicles (2.9%) and electrical equipment (1.6%) (see Fig. 2 and S6† for more detail).

Our results are consistent with Eurostat’s analysis of Europe’s consumption of raw materials, which shows that the total amount of materials directly consumed in the economy by businesses for economic production and by households, the domestic material consumption (DMC), has decreased in Europe during the 2000s.<sup>62</sup> This measure is largely influenced by consumption of non-metallic minerals and fossil fuels. DMC was 15.6 tonnes per capita in 2000 and dropped to 13 in 2016.<sup>62</sup> Changes in Europe’s DMC have been driven mainly by increased wealth (GDP) since the 1970’s, and only to a smaller extent by population increase.<sup>63</sup> The declining DMC in Europe is a result of increased resource efficiency in the region (disregarding the dependence on imported materials) in combination with the temporal decline in GDP during the financial crisis.<sup>64</sup> A decoupling of GDP from DMC has been observed since 2009,<sup>62</sup> possibly due to off-shoring of material extraction and shifting towards a service economy.<sup>63</sup> The worldwide material extraction, both in absolute terms and per capita is, however, steadily increasing particularly in Asia.<sup>64</sup>

### Time trend analysis of consumption and in-use stocks

**Direction and magnitude of change.** The results of the Mann–Kendall analysis of time trends in consumption of

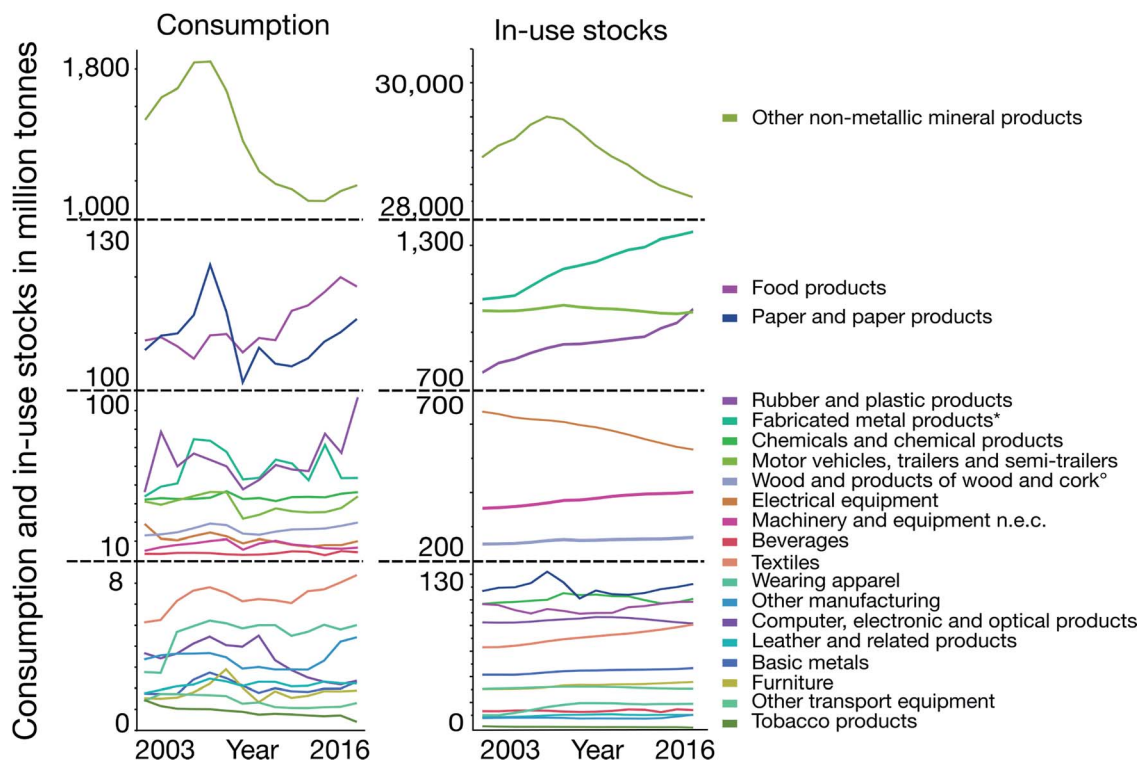
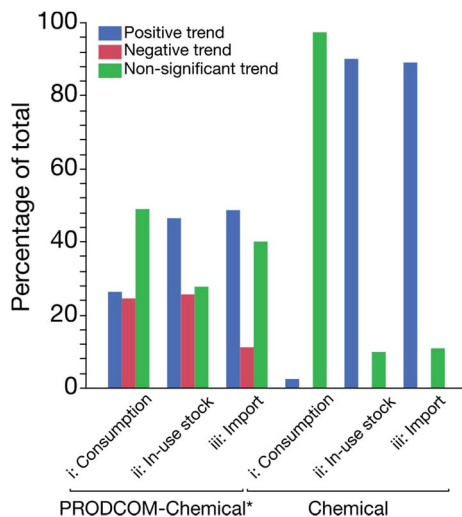


Fig. 2 Calculated consumption and in-use stocks over time for the PRODCOM codes in the filtered dataset, aggregated at the 2-digit level. \*Fabricated metal products, except machinery and equipment, °wood and of products of wood and cork, except furniture; manufacture of articles of straw and plaiting materials. Please note the broken y-axis. Individual plots can be found in Fig. S6† and the raw data can be found in Table S2.†





**Fig. 3** Results of the Mann–Kendall analysis assessing the change over time (2003–2016) in (i) the consumed mass per year, (ii) the estimated mass of the in-use stocks and (iii) the imported mass for products and chemicals. To test for significance  $\alpha$  was set at 0.05. \*The category PRODCOM-Chemical refers to the PRODCOM codes for which chemical data was available. The category chemical refers to the 768 chemicals that were linked to these PRODCOM codes.

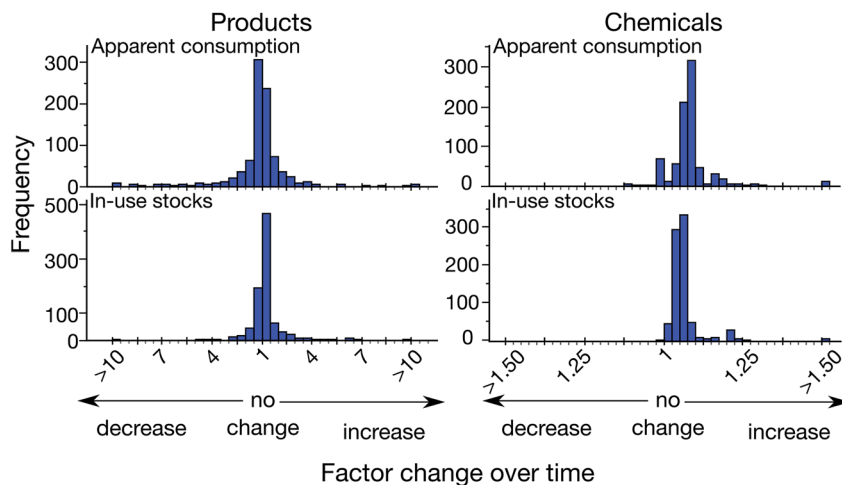
products (8-digit PRODCOM codes) and chemicals are shown in Fig. 3. We focus our analysis on the changes over time rather than absolute masses due to the large uncertainty and variability in chemical mass fractions of the materials recorded in the Commodity guide. Note that information about chemical content was not available for all product categories, *e.g.* the non-metallic mineral products which dominate the total consumed mass of products. Results of the analysis based on the subset of PRODCOM codes for which there was chemical data were, however, similar to that of all PRODCOM codes in the filtered dataset (see Fig. S4,† categories “PRODCOM” and “PRODCOM-Chemicals”). We therefore focus the discussion on results based

on codes with chemical data, as these data underlie the estimated trends for individual chemicals (Fig. 3).

For roughly half of the PRODCOM codes, no significant trend over time for the apparent consumption was observed. Of the remaining codes, the number of codes displaying significant increases (25%) or decreases (27%) were close to equal (see Fig. 3). The patterns shifted when looking at the in-use stocks, where the largest fraction of PRODCOM codes showed significantly increasing trends. This difference was due to the buffering effect of the stocks from previous years, which reduced the impact of the general reduction in consumption during the economic crisis in 2008.

The magnitude of change was evaluated by comparing the average mass in 2014–2016 with that in 2003–2005 (see Fig. 4). The median change in yearly product consumption between the first and last three years was a factor 1.34, which is similar to the increase in GDP during this time period (see Fig. S2†). For almost 80% of the product groups, the change was less than a factor of two throughout the assessed time period. For 17% of the products, the change in consumption between the first and last three years was less than 10%. Note that the seven PRODCOM codes with mass changes over time larger than a factor 10 contributed only 0.004% of the total consumed mass. The median change for in-use stocks of all product groups was a factor 1.20, which is lower than that for the apparent consumption of products and lower than the change in GDP over the same period of time.

The change in consumption of products over time leads to changes in the apparent consumption of chemicals. However, the increase in consumption of chemicals over time was significant for only 20 out of the 768 chemicals included in this study. These 20 substances were listed as “softener (plasticizer)” or “stabilizing agent” in the Commodity guide, and included a range of phthalates and organophosphate plasticizers/flame retardants and phenolic benzotriazoles (a class of ultraviolet-light absorbers), some of which are identified as SVHCs (see



**Fig. 4** The factor change of the average consumption and in-use stock over time. Bars indicate the number of products (left panels) and chemicals (right panels) within each bin. Note that a value of 1 means no change. And a factor 4 for example indicates a change by a factor 4. Bins for the products (8 digit PRODCOM codes) are 0.5 units wide, those for chemicals are 0.025 units wide. The factor change was calculated by comparing the average mass consumed/in stock of the last 3 years (2014–2016) compared to that of the first three years (2003–2005).



the file `Chemical_Trends_Classes.csv` in the ESI†). In stark contrast, the in-use stocks of chemicals in Europe were found to be increasing for 694 out of 768 chemicals, despite the financial crisis in 2008 and the small number of chemicals for which there were significant increases in the yearly consumption as a consequence of the service lives of many products being longer than one year. This difference in change over time between apparent consumption of chemicals in products on the one hand, and in-use stocks of chemicals, on the other hand, highlights the importance of information on products' service lives for estimating diffuse chemical emissions. Note that a long service life implies a strong buffering effect, dampening the between year variability and hence increasing the number of statistically significant trends. However, although 90% of the chemicals in the in-use stock showed significantly increasing trends (see Fig. 3), the magnitude of these changes was in general very small (see Fig. 4). The median change in yearly chemical consumption and in use stocks between the first and last three years (a factor 1.08 and 1.05, respectively) were considerably smaller than the changes of the individual product groups, and also smaller than the total changes for the main product categories (see Fig. 2).

These results indicate that the mass of the total chemical reservoir in in-use products has not increased to the same degree as consumption and in-use stocks of manufactured products during the studied time period. The trade data suggests that the size of the in-use stock of some important product categories with high content of specialty chemicals such as electrical equipment, motor vehicles and chemical products has been constant or even declining in recent years. Still, both annual consumption and the estimated in-use stock of rubber and plastic products has increased in the recent years (Fig. 2). Due to lack of time resolved data on product composition, this analysis does not consider neither substitution of chemicals (*e.g.* due to regulations or development in material design) nor transitions to non-chemical solutions. It can be assumed, however, that *e.g.* banned chemicals are likely replaced with chemicals with the similar function and it can be assumed that the estimated changes in total consumption and in-use stock of chemical mass presented here are less uncertain than the exact identity of the chemicals.

### Widespread use of chemicals in many types of products and products containing many chemicals

**Number of products per chemical.** The small changes over time in consumption and in-use stocks of chemicals in products observed can be attributed partly to a widespread use of many chemicals in a large number of products. On average, each chemical in this study was potentially present in 442 product groups, ranging between 47 and 834 PRODCOM numbers per CAS in the Commodity guide database (see the file `Chemical_Consumption_InUseStocks.csv` in the ESI† for the number of PRODCOM codes associated with each number in the dataset). Some of the products were consumed more and some consumed less during the investigated time period (see Fig. 3), and hence changes over time in chemical consumption

cancelled out to some extent. The large number of products per chemical in the Commodity guide can be explained by the fact that the materials covered in this database are generic and therefore widespread in their use. In fact, the 768 chemicals and 24 materials for which there was chemical information made up only 130 unique combinations of chemicals and materials. Many of the chemicals therefore shared the same trends as they appear in the same set of materials. We also observed that in general, chemicals associated with the largest number of PRODCOM codes displayed consumption time trends that were similar to the general trends of the consumption of all products (see Fig. S7†).

In reality, the range of uses is however often limited for many substances.<sup>65</sup> The relatively large number of low production volume chemicals as compared to the fewer high production volume chemicals registered under REACH is, for example, an indication of this circumstance (see Fig. 5).

In a recent study applying suspect screening to evaluate the chemical content of 100 consumer products from 20 categories (such as carpets, sunscreen, paint, plastic toys *etc.*), it was found that the majority of tentatively identified chemicals (1,602) were found only in a single product, and only 43 chemicals (including a range of organic solvents and phthalates) were present in more than 25 products.<sup>66</sup> Another study explored the "chemical space of consumer products" using data reported in safety data sheets (SDSs) from products available at Walmart, a large US retailer.<sup>34</sup> The authors used cluster analysis to associate the chemicals with products in various product categories and noted that the chemicals in the dataset could be divided into three categories: ubiquitous chemicals with multiple uses and high exposure potential, multi-category-chemicals and use-category-specific chemicals present in a narrower range of products and thus with lower exposure potential. For example, chemicals such as water, ethanol, glycerol, titanium dioxide, 1,2-propanediol are used in a wide range of products, but in fact only 5% of the *ca.* 1600 chemicals in the dataset were present in more than 100 different products. However, in the product

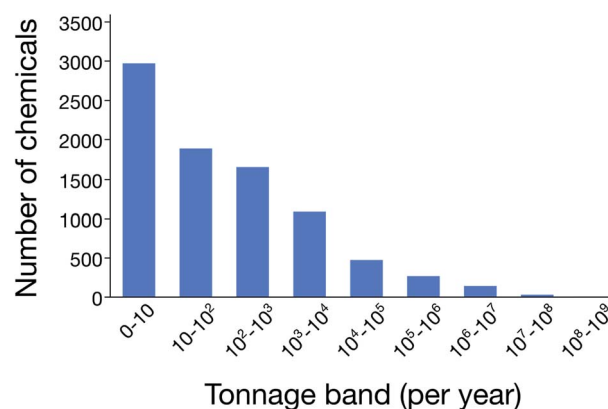


Fig. 5 Data on the number of chemicals by tonnage in the EU, based on data on registered substances under REACH. The data was accessed on the 31<sup>st</sup> of May, 2018 and contained 20 140 unique substances and information from 80 183 dossiers. Only data with a well-defined tonnage-band was included.





categories with more articles (function defined by their shape rather than by their chemical composition) represented, such as electronics, toys, sports & outdoor items, more than 25% of the chemicals were present in more than 100 products and most chemicals occurred in 10–100 products.<sup>34</sup> Hence, based on these previous findings, relatively few chemicals can be expected to have the widespread use as assumed for the subset included in the Commodity guide. Those chemicals that do are often produced in large volumes and time trends in their diffuse emissions from consumption of products are likely to be governed by the general consumption trends of a wide range of products. It also means that the ubiquitous chemicals (such as titanium dioxide, 1,2-propanediol, methyl 4-hydroxybenzoate, propane, propyl 4-hydroxybenzoate<sup>34</sup>) probably exhibit rather similar changes in consumption over time, as long as the composition of the products does not change over time.

### Number of chemicals per product

Whereas the majority of chemicals on the market are likely present in a narrow range of products, each product may in turn contain a large number of chemical components. The consumption of a single product group can therefore dictate the time trends for a large number of product-specific chemicals. Each PRODCOM code in our dataset contained on average 403 chemicals, ranging between 13 and 755 chemicals. The high number of chemicals per PRODCOM code is consistent with other reports of chemicals potentially present in plastics, *e.g.* a recent compilation of hazardous substances known to be used in plastic materials, such as monomers, plasticizers, flame retardants, UV-stabilizers, corrosion inhibitors, pigments *etc.*, where 300–400 chemicals were listed as possibly present in a limited number of plastic types.<sup>67</sup> A review of chemicals in paper products reported that analytical measurements published in the scientific literature identified *ca.* 350 chemicals in paper products.<sup>68</sup> It is difficult to determine how many chemicals are commonly present in a single product. Chemical analyses are typically conducted for a specific chemical class and a limited range of products. For example, a study of liquid household products manufactured in Korea identified 9 to 113 volatile organic compounds (VOCs) in each of the 42 products assessed, and several of the target analytes appeared in many products (*e.g.* up to 32 products containing limonene).<sup>69</sup> It is also noteworthy that a higher number of chemicals are present in products than specified on labels or in safety data sheets (SDS), as not all ingredients need to be disclosed to the public according to US and EU regulations,<sup>26,70</sup> which is also evident from previous analysis of consumer products.<sup>66</sup> For example, a study assessing VOCs in fragranced consumer products found 133 different VOCs in 25 products (including identified hazardous substances such as 1,4-dioxane, methylene chloride, and acetaldehyde), on average 17 VOCs per product, with only one of these substances (ethanol) listed on any product label and two (ethanol and 2-butoxyethanol) on any SDS.<sup>70</sup>

The fact that many chemicals are unique to specific product groups highlights the need for more data on chemical content of products. The aforementioned studies are mainly based on

data from chemical products. A better mapping of chemical content in articles from various types of product groups is particularly needed. The current assessment of the available data highlights that compilations of typical chemical content of various materials need to be complemented by product specific chemical information.

### Change in mass of imported products over time

While time resolved composition data of manufactured products is currently not available, the origin of products consumed in Europe may give an indication of the content of hazardous or non-registered chemicals emitted from these products. For almost half of the PRODCOM codes in our dataset, the imported mass from non-European countries increased over time, with a median change of a factor 1.72 between the average import in the first and last three years. All the chemicals in the dataset were (*via* products) imported in masses that were lower than those incorporated in products produced in the EU (by a factor 1.1 to 40.2). However, the imported mass of chemicals was found to be increasing significantly for 684 out of the 768 chemicals, with a median change of a factor 1.68 between the average import in the first and last three years. Of these 684 chemicals, 56 were chemicals listed on the SIN list, 27 are substances of very high concern under REACH (SVHCs) and 50 have been found to be potential PMOCs. Hence, while the consumption of most chemicals *via* products in the EU is currently dominated by EU production, the EU is increasingly importing more chemicals *via* products from abroad.

This increased import of chemicals in imported products could imply increased amounts of hazardous substances consumed *via* these products if chemical regulations and regulation compliance in the country of origin are less strict than in the EU. A recent study by ECHA demonstrates that on average 18% of tested products on the European market were non-compliant with REACH regulations.<sup>71</sup> Non-compliance was observed for 10, 17 and 39% of the products originating from the EU/EEA region, China and products whose origin could not be identified, respectively. This indicates that the import of articles and chemical products may be an important source of potentially hazardous chemicals.

### Major knowledge gaps

Material and in particular chemical composition of products remains a major knowledge gap hampering the use of time resolved trade data to estimate diffuse emissions of chemicals from manufactured products in use. The Commodity guide, although partly outdated, is one of few available compilations of this type of information. Another extensive database is the Chemical and Product Categories (CPCAT) database from the U.S. Environmental Protection Agency.<sup>35</sup> CPCAT specifies chemical composition of individual (mainly chemical) products, and can be useful to predict the composition and use pattern for various chemicals, and if available also for articles.<sup>72–74</sup> Due to difficulties in linking the chemical use descriptions in CPCAT to PRODCOM codes we could not include this data in the current study. In addition to the risk of



adverse chemical exposure *via* consumer products and building materials, the ambition to create a circular economy with increased recycling of in particular plastics and paper materials has raised concern about the recirculation of hazardous substances in recycled materials. In response to this, inventories have been made for specific chemical categories (*e.g.* phthalates, chemicals listed as hazardous substances) present in various material and/or product categories (*e.g.* paper, plastics and liquid household products).<sup>67,68,75</sup>

The multiple potential objectives of data compilations such as CPCAT and the Commodity Guide raise questions regarding what kind of data can be used for different purposes. It is reasonable to aim for collecting data on typical chemical content of various material categories, as is done in the Commodity Guide, rather than individual products. On the other hand, this strategy requires detailed statistics on the composition of articles and materials, as different materials and chemicals can be used to reach the same functionality.<sup>2</sup> The ranges of possible chemical and material mass fractions in the Commodity Guide are often broad (*e.g.* a chair can be made of 0–100% plastics) and thus highlights the need for information on *e.g.* weight fraction distributions similar to those predicted for ingredients in personal care products.<sup>73</sup> Product-specific composition data for a representative subset of articles would facilitate the derivation of this type of information.

Efforts to compile databases of product composition, however, risk becoming a Sisyphean task as design and technology (*e.g.* introduction of LED TVs), choice of materials (*e.g.* increased use of plastics) combined with developments in both chemicals legislation and innovation in chemical design also alter the needs and preferences of chemicals in a complex way.<sup>34</sup> The lack of time resolved composition data makes it hard to detect changing applications of chemicals in products over time and their impacts on consumption and in-use stocks of chemicals. Currently, producers are not obliged to provide information on chemical content of articles (see Introduction). This lack of transparency on chemicals in products has been labelled an emerging policy issue under the UN Strategic Approach to International Chemicals Management (SAICM)<sup>76</sup> and has been discussed extensively in the recently published “Study for the strategy for a non-toxic environment” of the 7th Environment Action Programme by the EU Commission.<sup>2</sup>

The European commission is currently performing an assessment of the potential use of a tracking system to address challenges related to the presence of substances of very high concern in articles and product waste.<sup>48</sup> In a review on chemicals in products, Kogg and Thidell observed that while information on chemicals in products is available upstream of the supply chain (*i.e.* chemical suppliers), it is often lost before it can reach the relevant stakeholders.<sup>45</sup> The majority of systems currently in place to deal with the communication on chemicals in products rely on restricted substance lists that confirm the absence of certain chemicals of concern in a product rather than the chemicals that the product contains.<sup>45</sup> Few companies, let alone authorities or the general public, are therefore aware of which chemicals these products contain. Complex supply chains, a lack of legal pressure, the need to protect confidential

business information and the high costs of building and maintaining a system that tracks chemicals in products all contribute to the lack of available data on chemicals in products.<sup>2,45,77</sup>

In addition to the scarce information on chemical concentrations in manufactured products, estimations of chemical emissions from products is hampered by several other factors, including difficulties in deriving time dependent emission factors for various chemical-product combinations and expected service life of the products. Emissions from products are variable over time. Volatile organic compounds, for example in flooring materials, are often emitted at high rates initially but the loss slows down as the chemical concentration in material decreases.<sup>78</sup> Other types of additives can be lost at a more constant rate throughout the product lifetime.<sup>79</sup> Environmental factors also influence the emission rate.<sup>80</sup> Without this information, time trends in apparent consumption and in-use stocks have to be viewed as worst case situations, *i.e.* constant emissions proportional to initial chemical content are assumed.

## Conclusion

Trade data for manufactured products is relatively easy to access and diffuse emissions of chemicals from products are considered important sources of contaminants in the environment.<sup>25</sup> Therefore, changing consumption patterns over time are potential indicators for diffuse chemical pollution. In this study, we explored the current potential to develop such an indicator and identified obstacles hampering the use of consumption data to predict diffuse emissions of chemicals. Trade data available for the European market indicate an increased consumption of manufactured products over time (2003–2016) accompanied by a considerably less prominent increase in in-use stock of the chemicals included in the Commodity guide. For these chemicals, the estimated general increase in diffuse emissions from products consumed within the EU during this time period was smaller than the increase in consumption of products. The difference in estimated time trends for individual chemicals in the dataset were small compared to the large uncertainty in product composition data which thus hampered ranking of the chemicals based on emissions. The analysis highlighted the difference between chemicals with multiple or general functions, for which diffuse emissions commonly depend on a wide range of products from several categories, and chemicals with specific uses present in only a limited range of products. For the latter, product-specific information on chemical content, which is currently lacking, is necessary to estimate chemical emissions and identify substances with an increased use in society. The potential of using consumption patterns over time as indicators for diffuse chemical pollution and as a tool to aid in the prioritization of chemicals for screening and analysis in the environment can only be fully exploited if chemical content of individual products becomes available.

The practical issues identified in this study include the finding that the calculation of product consumption time trends was most limited by the range of products for which



there is data in a mass-based unit such as kg. Information regarding the typical end-use of the products in the Eurostat product categories would also be needed to avoid double counting of consumed masses. A more profound challenge highlighted in this study is the large gap in our knowledge of which chemicals are present in products. The Commodity Guide developed by the Swedish Chemicals Agency makes the essential link between chemical composition data and trade statistics but has limited use for the purpose of our study due to its focus on materials instead of actual product composition data. Currently, the existing databases on chemical content in products are biased towards chemicals that are already identified as problematic as these focus on chemical products (information derived from product labels or SDSs) or data generated in surveillance activities for restricted substances.<sup>2</sup> Chemical composition data of products is essential not just to estimate diffuse emissions and concentrations in the environment. This information also facilitates the transition to a circular economy with extensive material recycling, allows consumers to make informed choices when purchasing a product, facilitates preparedness for future legislations and helps companies to develop and use safer alternatives for chemicals in their products.<sup>2,45,77</sup>

To fill the knowledge gap regarding chemical composition of products, there is a need to develop systems for transferring chemical information along the supply chain and to specify what kind of information should be conveyed. An example of such a system exists within the car industry (International Material Data System) where all suppliers are required to report the chemical composition of their materials and components.<sup>45</sup> To protect confidential business information, 10% of the mass of each material or component may be labelled as confidential as long as it does not contain any restricted substances. In addition, suppliers select who is granted access to their data. Pilot projects have also been launched by SAICM under the Chemicals in Products programme that focus on defining and demonstrating best practices for information exchange on chemicals in four priority sectors, including textile products, electronics, building materials and toys.<sup>81</sup> It is conceivable that full access to the data could be granted to authorities responsible for identification of hazardous substances such as ECHA, although methodologies to disclose chemical content to research and the general public need to be developed to advance exposure science and fulfil the consumers' right-to-know. Furthermore, legal incentives for suppliers and companies to make use of these systems need to be enforced. Non-binding agreements such as the chemicals in products project by SAICM<sup>82</sup> exist and have potential but still lack participation of the private sector. Currently, the REACH regulation demands communication of information only regarding the presence of substances of very high concern along the supply chain and is limited to the name of the substance and not its concentration.<sup>36</sup> This right-to-know is so far, according to the relatively few assessments made, seldom levied by end-consumers,<sup>48</sup> possibly due to unawareness and the rather cumbersome procedure to request the data.

A remaining major challenge for today's chemicals management is to identify the chemicals with potential to threaten local or global environments and populations as highlighted in several publications.<sup>83–85</sup> In the light of these discussions, the focus on conveying information on only the rather limited range of already identified hazardous substances present in consumer products is not sufficiently pro-active. Previous studies attempting to rank chemicals based on exposure have identified emission estimates as a major knowledge gap.<sup>19–22</sup> The field of exposure science, and the protection of the environment and human health, can only advance if more data on use of chemicals in our society becomes available.

## Conflicts of interest

There are no conflicts to declare.

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