

# Integrated dynamic probabilistic material flow analysis of engineered materials in all European countries

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## ABSTRACT

Uncertainties remain regarding the potential environmental risks of engineered nanomaterials, reflecting missing information on both the exposure and the hazard sides. Probabilistic material flow analysis (PMFA) is a useful exposure assessment tool that maps the flows of a substance through its lifecycle towards the environment, taking into account the uncertainties associated with the input data. In the last years, several refinements have been made to the original PMFA method, increasing its complexity with respect to systems dynamics, fate during recycling and reprocessing and forms of release. In this work, an integrated dynamic probabilistic material flow analysis (IDPMFA) was developed that combines all separate advancements of the method in one overarching software code. The new method was used to assess the forms in which nano-Ag, nano-TiO<sub>2</sub> and nano-ZnO are released into air, soils and surface water. Each European country (EU28, Norway and Switzerland) was studied from the year 2000 to the year 2020. The present model includes new assessments of the forms in which nano-ZnO is released into the environment and of the flows out of reprocessing (last step of recycling) of nano-Ag, nano-TiO<sub>2</sub> and nano-ZnO towards both technical and environmental compartments. The forms of ZnO released to different compartments vary greatly with different proportions between pristine, dissolved, matrix-embedded and transformed forms. The same applies for the forms of the other ENMs released after reprocessing, where different processes result in very different distributions between the various forms. The country-specific assessment showed that it is mainly the different solid waste treatment schemes that influence the distribution to final environmental sinks. Overall, the results of IDPMFA show the great importance of considering the full life cycle of nanoproducts including the different stages of recycling, the differences between countries, and the forms of the released materials. The results from the integrated model will provide useful input information for environmental fate models and for environmental risk assessments.

## 1. Introduction

Although many engineered nanomaterials (ENMs) such as TiO<sub>2</sub>, ZnO and Ag have been studied extensively over the last years (Kühnel et al., 2018; Lead et al., 2018), uncertainties remain regarding the risks they could pose towards ecosystems. Data exist on the biological effects of their pristine (as-produced) forms (see for example the reviews on aquatic environments by Skjolding et al. (2016) and Canesi and Corsi (2016), or the review on terrestrial plants by Laxma Reddy et al. (2016)), but it remains difficult to quantify the concentrations in which ENMs are released to and are present in the environment (Laborda et al., 2016; Lead et al., 2018). Recent advances in analytical methods enabled a few studies to detect ENMs in natural waters (Gondikas et al., 2014; Gondikas et al., 2018; Li et al., 2016; Vogt et al., 2019). Soils are even less

studied and, to our knowledge, no quantification of the engineered compared to the total nano-sized fraction in natural soil samples has been performed so far, although detection of nanoparticulate TiO<sub>2</sub> in sludge-treated soils (Yang et al., 2014) and soils (Yi et al., 2020) has been reported. Therefore, models remain necessary to quantify environmental concentrations of ENMs.

Material flow analysis (MFA) proved useful for such a purpose, as it allows tracking the flows of materials from their production throughout their lifecycle (production, use and end-of-life) towards environmental compartments (air, water and soil). Release models of this type take as input two types of data: the amount of material produced and transfer coefficients describing the fractions of material flowing from one compartment to the others. MFAs rely heavily on literature data and experimental work. MFA models for ENMs were first published in 2007

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and 2008 (Boxall et al., 2007; Mueller and Nowack, 2008) and much progress has been achieved in the following years. Keller and co-workers for example developed MFAs to model ENM releases at the global scale and for the USA, the United Kingdom and China (Lazareva and Keller, 2014; Song et al., 2017), while Ding et al. (2019) performed the MFA of nano-Ag in the environment of Hunan, China and Suzuki et al. (2018) modelled the flows of ENMs from the Japanese construction sector. Gottschalk et al. (2009) laid the foundations of probabilistic MFA (PMFA) for ENMs in Europe. The authors assessed the uncertainties associated with production volumes and transfer coefficients, resulting in probability distributions associated with each flow within the life-cycle and towards the environment. Several updates and further developments of this model were then performed. The dynamics of production, manufacturing and consumption were important additions to the model to be able to assess the stocks of ENMs and their dynamics of release. For example, ENMs in electronics or construction do not reach their end-of-life within the year they are produced, but later. Moreover, some compartments, such as landfills and soils, were modelled as sinks to enable the quantification of ENM accumulation, thereby avoiding the underestimation of their concentrations (Bornhöft et al., 2016; Sun et al., 2017, 2016). The dynamics of waste management is another important aspect to consider, as over the last 20 years, more and more material was recycled across Europe, increasing the potential for ENMs to enter back into the market (Rajkovic et al., 2020). Modelling production amounts and waste management also needs to be country-specific to consider large discrepancies (Adam and Nowack, 2017; Kuenen et al., 2020). For example, while Switzerland or Denmark incinerate all their mixed waste, landfilling constitutes a very large part of waste management in other countries such as in Romania, for example.

Another major aspect of ENM emission modelling consists in tracking their transformations within their lifecycle, in the aim of understanding the forms in which they are released to the environment (Adam et al., 2018). Based on information available in the scientific literature, the authors defined five forms of release: pristine, matrix-embedded, chemically transformed, dissolved and product-embedded. A form-specific PMFA was developed by Wigger and Nowack (2019) by using information on particle functionality to separate the generic ENM flows into different crystal forms of the same materials, e.g. anatase and rutile TiO<sub>2</sub>. The importance of considering the forms of release was recently stressed in a review about the key principles of environmental exposure assessment of nanomaterials (Svendsen et al., 2020).

These updates of the MFA first built by Gottschalk et al. (2009) are valuable, but scattered among different versions of the model. Moreover, not all versions of the model consider all flows of ENMs out of recycling. After waste collection, recycling can be divided in two steps: 1) sorting and 2) reprocessing of materials. Caballero-Guzman et al. (2015) used MFA to study the fate of ENMs within the Swiss recycling system, but did not assess their flows from recycling to environmental compartments. Rajkovic et al. (2020) did assess ENM flows out of sorting, but considered reprocessing as a sink, ignoring flows out of these processes. Yet, these authors showed that significant proportions of nano-waste went through reprocessing of their host material.

The goal of this study is therefore to integrate all updates of the model originally built by Gottschalk et al. (2009) into one model, as well as adding flows out of reprocessing, therefore providing an Integrated Dynamic Probabilistic Material Flow Analysis (IDPMFA). The modelling is done for each European country (EU28, Norway and Switzerland) from 2000 to 2020 and is applied to nano-Ag, nano-TiO<sub>2</sub> and nano-ZnO. The main novelty of the IDPMFA model is that it combines all the sub-models that were built previously and detailed in Table S17 into one coherent model. Additionally, the study covers three specific aspects that were not modelled before: i) the assessment of the forms of release of nano-ZnO through its lifecycle, ii) the assessment of the forms of release of nano-Ag and nano-TiO<sub>2</sub> out of reprocessing and iii) the assessment of the total flows of nano-Ag, nano-TiO<sub>2</sub> and nano-ZnO out of

reprocessing.

## 2. Material and methods

### 2.1. Collection of transfer coefficients

One model was developed for each country and each ENM studied. All country- and ENM-specific sub-models were built on the same base-model: they are divided in compartments following the ENM lifecycle: production, manufacturing, use, wastewater management, solid waste management, and environmental compartments (air, surface water and soil). To perform the IDPMFA, several input parameters were needed, which were either taken from the previous most recent ENM MFAs or collected from the literature (Fig. 1). The model is applicable to all ENMs. Here, nano-Ag, -TiO<sub>2</sub> and -ZnO are used as case studies. To apply the model to other ENMs, the input parameters (production amounts, transfer coefficients and associated probability distributions) need to be assessed specifically to the ENM under study.

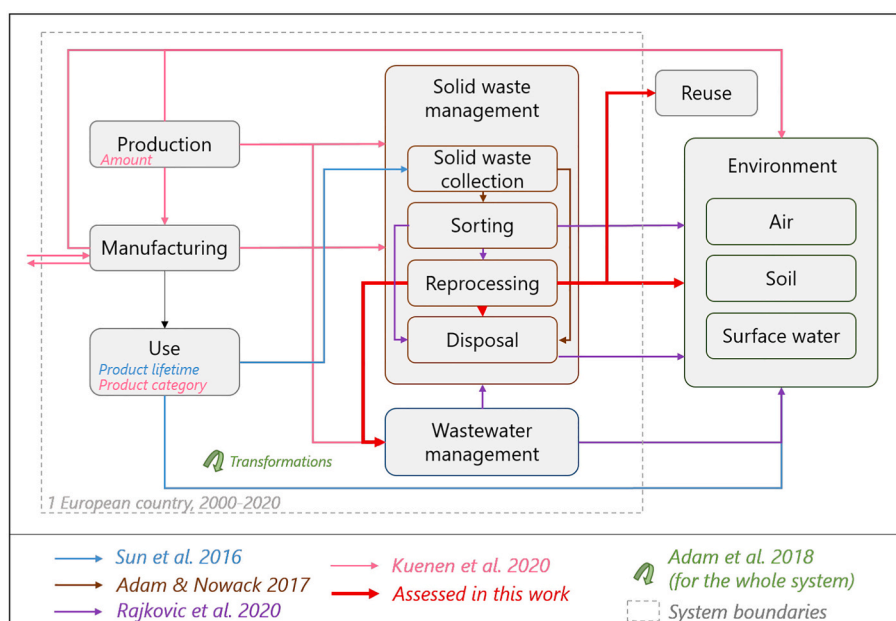
Production amounts were taken from Kuenen et al. (2020), as well as all transfer coefficients into and from production and manufacturing. The masses of ENMs in manufacturing and consumption are allocated to product categories for a more detailed tracking of their flows (see Supplementary Information B). Those were also taken from Kuenen et al. (2020). Product lifetimes during use were assessed in Sun et al. (2016) and used as such in the present models.

Transfer coefficients from use to wastewater, surface water, air, soils and solid waste were taken from Sun et al. (2016). Compartments and flows within and out of wastewater management refined in Rajkovic et al. (2020) were used in the present model. Wastewater management compartments include sewer system, on-site treatment such as septic tanks, wastewater treatment plant and sludge treatment. Regarding solid waste management, the first step of solid waste collection as mixed waste (going to landfill and incineration) or to sorting was modelled as in Adam and Nowack (2017). Flows from sorting were taken from Rajkovic et al. (2020), while transfer coefficients out of reprocessing were collected from the literature. Details are given in the Supporting Information A and B. Reprocessing processes and associated transfer coefficients were assumed to be constant from the year 2000 to year 2020. It is worth noting that the recycling considered here is that of the materials (such as plastic or metal) in which the ENMs are embedded during product manufacturing. The recycling of ENMs themselves is not considered, only their associated flows during the recycling of the materials in which they occur.

The data collection of transfer coefficients for reprocessing was organised according to the materials in which the ENMs are embedded: the reprocessing systems of plastics, textiles, metals, glass and minerals were studied separately, as detailed in Section 1 of the Supplementary Information A.

The forms in which the ENMs flow from one compartment to another were also considered in the present model. For compartments other than reprocessing, transformations of nano-Ag and nano-TiO<sub>2</sub> were reported as they were assessed in Adam et al. (2018), while transformations of all ENMs in reprocessing and those of nano-ZnO all along its lifecycle are assessed for the first time in the present work. Five forms of ENMs were defined according to Adam et al. (2018):

- Pristine: Particles released as they were inserted in the product. They are not embedded in a matrix nor transformed. Single, aggregated and agglomerated pristine particles are all in this category.
- Matrix-embedded: ENMs embedded in a solid matrix. The matrix particle can be a piece of polymer, paint or cement, for example.
- Transformed: ENMs which have been subject to chemical reactions, for example sulfidation, leading to the formation of new particles.
- Dissolved: Any dissolved species released from an ENM.
- Product-embedded: ENMs still contained in a complete nanoparticle. Releases to water occurring during use of liquid or gel nanoparticle



**Fig. 1.** System studied. Colours of arrows and text represent the models in which the corresponding data were assessed. The dashed grey line represents the system boundaries: each European country from 2000 to 2020. Processes occurring in the environment and in reuse are out of the scope of this study and out of the system boundaries.

(e.g. sunscreens) were not considered as being product-embedded but pristine, as the liquid product would disperse in water.

To account for these forms, each ENM total mass flow was divided into five flows (or fewer if the number of relevant forms was smaller), so each form was tracked all along the ENM lifecycle, from production to release into the environment. The reasoning and references used to assess the transformations of nano-ZnO along its lifecycle are detailed in Section 2 of the Supplementary Information A.

## 2.2. Assessment of uncertainties

Uncertainties associated with the production amounts and transfer coefficients of ENMs were considered in the model by using probability distributions. Probability distributions associated with the flows taken from previous models were used as such from those models. Probability distributions associated with transfer coefficients within and out of reprocessing were built specifically for this integrated model, based on the quality of the data collected. A pedigree matrix was developed to reflect the quality criteria relevant to the system studied (Table S1, Supporting Information A). From this pedigree matrix, a score was determined for each criterium. For each data point, quality scores were aggregated into a coefficient of variation (Laner et al., 2015), used to build either triangular or trapezoidal probability distributions (see Section 3 of the Supplementary information A for details). Triangular distributions were used when only one transfer coefficient could be obtained, while trapezoidal distributions were used when two transfer coefficients were available. In both cases, the obtained transfer coefficients were used as the modes of the distributions, while the coefficients of variation were used to widen the probability distributions to a more realistic range (Figs. S6 and S8). Uniform distributions were also used when only minimum and maximum values of transfer coefficients were given (Fig. S7). Regarding the forms in which nano-ZnO flow from one compartment to another, specific probability distributions were defined, as described in Section 2 of the Supplementary information A.

Transfer coefficients, associated probability distributions and the references used to define them are summarised in Supplementary Information B. Details regarding the transfer coefficients used for

reprocessing of plastics, textiles, metals, glass and minerals are available in Tables S1 to S12 in Supplementary Information A.

## 2.3. Implementation of the code

When all transfer coefficients and associated probability distributions were defined, the relevant parts of the different codes (described above) were merged into one code for each country, written in Python. The package built by Bornhöft et al. (2016) and refined by Rajkovic et al. (2020) was used, which consists in 5 modules. (1) *Components* defines the different classes of compartments and flows included in the model; (2) *Model* defines the methods that link the compartments and the flows; (3) *Simulator* includes the methods necessary to run the simulation and to extract the results. These three modules are common for all models, they are the base of the simulations. Two other modules are specific to each ENM and country (in total, 90 of each module): (4) *ENM\_Country\_Model* is where all system components and transfers are parametrized using the probability distributions defined from the literature and (5) *ENM\_Country\_Runner* is the module where the simulation is initiated and the results are processed, specifying the time periods and the number of simulation runs.

## 3. Results

In the following sections, first the form-specific model for ZnO is presented as contrary to TiO<sub>2</sub> and nano-Ag, a form-specific analysis has not yet been done for this ENM. The next section then presents an analysis of the flows out of reprocessing, which represents the last piece of information needed to fully track the flows of ENMs from cradle to grave. With these basic building blocks in place, the next Sections 3.3. and 3.4 are then presenting the full IDPMFA results. In Sections 3.1 to 3.4, for better readability, one country was selected to describe the flows. The means of all flows obtained for each country and each ENM are available in Supplementary Information C. The UK was selected to illustrate the results for two reasons. First, it is the country that was studied in most details within the NanoFASE project and thus a large amount of data is available. Second, it is representative of a waste management system where mixed waste and sludge go to both

incineration and landfill, as in most European countries and in contrast to e.g. the Netherlands or Switzerland where all mixed waste and sludge are incinerated (Rajkovic et al., 2020). Numbers shown in the figures are the means of the corresponding probability distributions calculated by the codes.

### 3.1. Forms of release of nano-ZnO

After use, 86% of nano-ZnO goes to wastewater, either directly (2063 t) during use of personal care products or indirectly (96 t), via sorting of the packaging waste in which part of these products remain (Fig. 2). Most transformations of nano-ZnO occur in wastewater management. Although 96% of nano-ZnO arrives to wastewater as pristine, it was assumed that all pristine material was sulfidised during the activated sludge stage (Brunetti et al., 2015), resulting in 96% transformed

in the sewer and 94% in on-site treatment. After the wastewater treatment plant (WWTP), 98% of nano-ZnO occurs in transformed forms, the remaining 2% staying protected by the solid matrix (e.g. textile or paint) in which they are embedded. 94% of what arrives to incineration is in WWTP sludge, explaining the very high proportion (96%) of transformed nano-ZnO leaving this compartment and arriving to landfill and reuse. Transformations of nano-ZnO during wastewater management also results in very high proportions of transformed forms in sludge-treated soil (99%) and in the subsurface (91%). In surface water however, a significant part of nano-ZnO occurs in pristine form (78%), because of the direct contact of this water with personal care products during bathing. Most of the transformed nano-ZnO in this environmental compartment (accounting for a quarter of the total amount) comes from wastewater management. Most of what arrives to air comes from the use of paints, lacquers and antibacterial products made of plastic or textile.

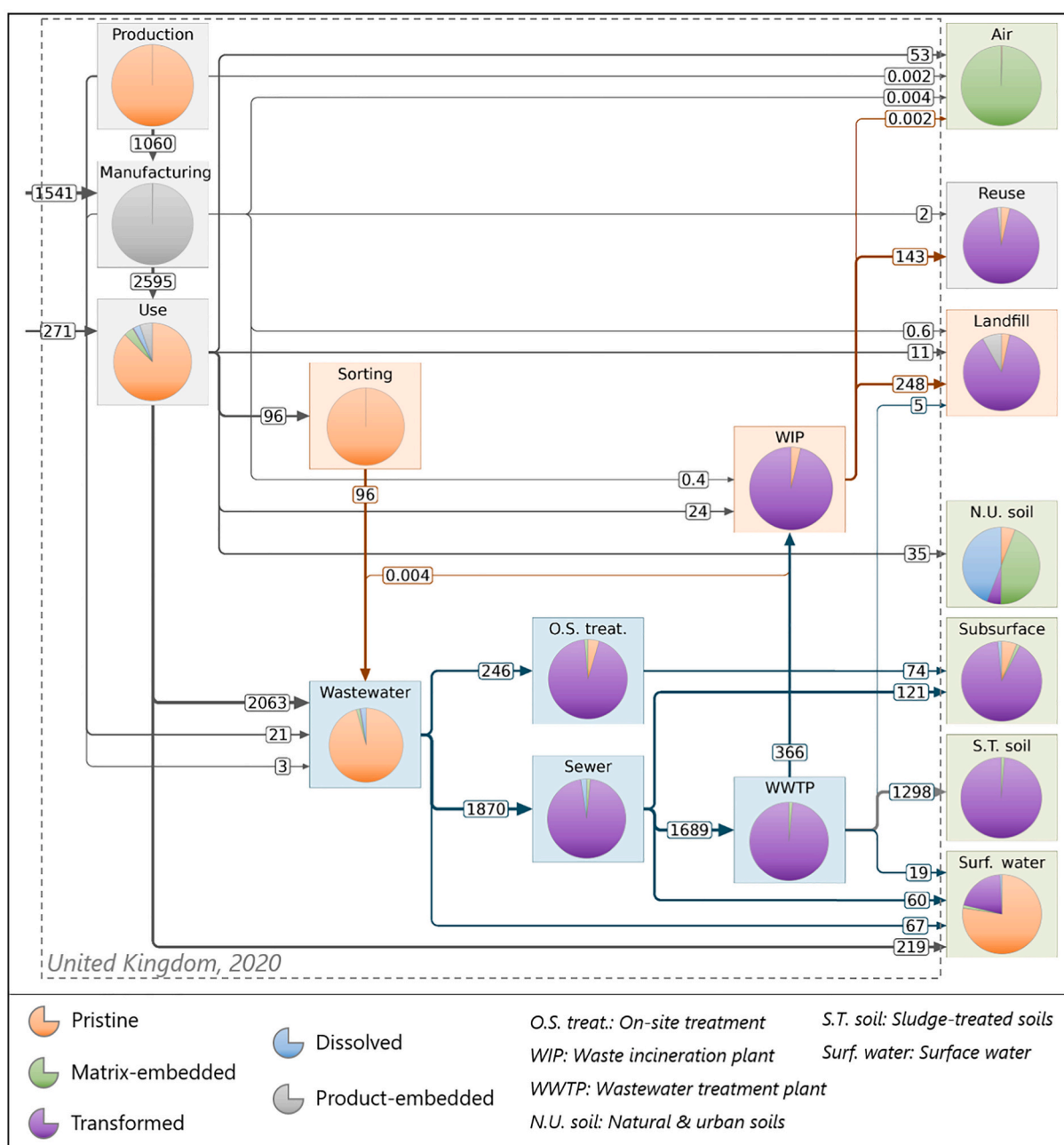


Fig. 2. Flow chart illustrating the flows and transformations of nano-ZnO in the United Kingdom in year 2020. Numbers are the means of the corresponding probability distributions, in tonnes.

Therefore, nano-ZnO is mainly present in matrix-embedded forms in this compartment. All nano-ZnO occurring in natural and urban soils come from the use of paints and lacquers. As experimental data was lacking for this specific scenario, it was assumed, based on what was defined for nano-Ag in Adam et al. (2018) that most nano-ZnO could be dissolved on the weathered building surface or arriving as matrix-embedded to the soils, also considering that smaller portions could stay pristine or be transformed (Supplementary Information B).

### 3.2. Flows associated with reprocessing

Details regarding reprocessing transfer coefficients and associated probability distributions are available in Tables S2 to S13. Aggregated mass flows are given in Fig. 3. It is noteworthy that results from the present models do not show nano-ZnO in reprocessing during the time period 2000–2020, since the only product category which would be relevant is paints & lacquers and was assumed not to reach construction & demolition waste in this time period (Sun et al., 2016).

Nano-Ag flows to reprocessing mainly embedded in metal waste (270 kg in the UK in 2020, Fig. 3A) from printed-circuit boards and waste from electronic appliances. In metal reprocessing, 99.98% of

nano-Ag is melted, where it is transformed. This explains the high proportion of transformed forms in reuse (98%), landfill (99.6%) and air (100%), which are the sink compartments in which ENMs end after metal reprocessing (Fig. S4). Plastic waste represents the second type of waste in which nano-Ag occurs in reprocessing (4 kg), generated in waste of electrical and electronic equipment (WEEE). Plastic and textile reprocessing both imply flows of nano-Ag to wastewater, where it is mainly dissolved (83%).

Reuse is the main compartment to which nano-TiO<sub>2</sub> flows after reprocessing (2.59 t in the UK in 2020, Fig. 3A). There, it occurs in transformed forms (48%) and in product-embedded forms (52%). Nano-TiO<sub>2</sub> in reuse comes from plastic, textile, metal and glass reprocessing. Glass melting represents the highest source of transformed nano-TiO<sub>2</sub> in reuse (0.76 t), while plastic granulation is the main contributor to product-embedded nano-TiO<sub>2</sub> in reuse (0.87 t, Supplementary Information C2). Flows from reprocessing to landfill mainly occur in metal slag (0.50 t in the UK in 2020), with low contributions of plastic granulation (0.08 t) and textile baling (0.0004 t). This results in 57% of nano-TiO<sub>2</sub> occurring as product-embedded and 43% in transformed forms in landfill. Nano-TiO<sub>2</sub> flows to air from plastic, textile, metal shredding and glass crushing. As a result, all releases from reprocessing to this

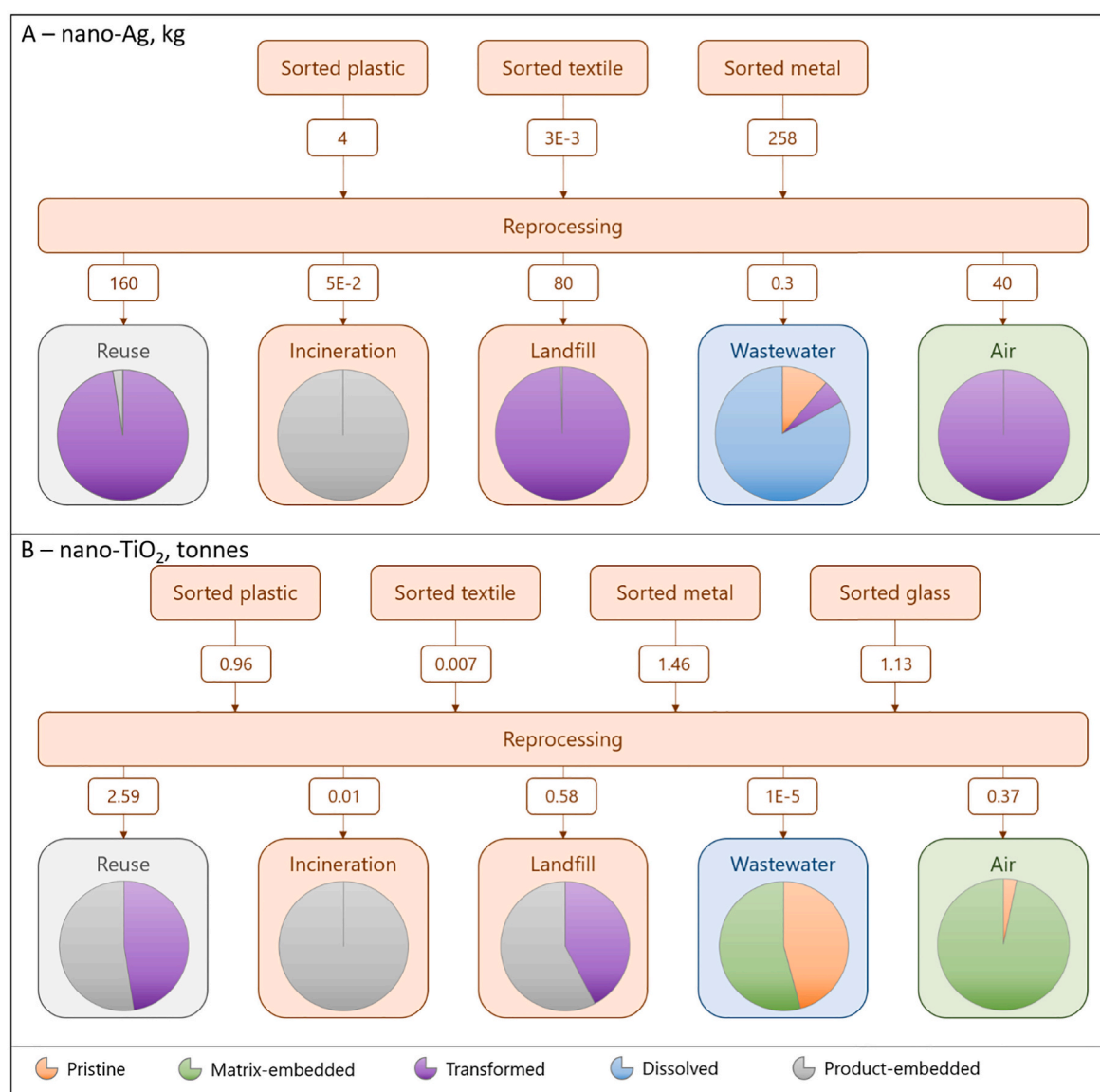


Fig. 3. Flows of nano-Ag (A, in kg) and nano-TiO<sub>2</sub> (B, in tonnes) in reprocessing systems (means of probability distributions, United Kingdom, 2020).

compartment were considered matrix-embedded. As for nano-TiO<sub>2</sub> arriving to wastewater, they come exclusively from textile washing, resulting in 46% pristine and 54% matrix-embedded.

### 3.3. Country-specific releases

The forms of release of nano-Ag and nano-TiO<sub>2</sub> were also assessed for the UK in this work (Figs. S9 and S10). Detailed results are described in Adam et al. (2018) for Europe taken as a whole. Here, country-specific releases were assessed for all countries separately (Fig. 4, Supplementary Information C). Solid waste management is the part of the system studied that varies most among countries. Consequently, flows of ENMs to reuse, landfill and sludge-treated soil vary with high amplitudes. Where flows of nano-Ag to sludge-treated soil decrease, they increase towards landfill, both directly from solid waste collection and via incineration. For example, in Switzerland, no solid waste collected from households goes to landfills. The high share of landfilling in this country (44%) corresponds to the ash from waste incineration plants. The proportion of nano-Ag going to reuse depends directly on the percentages of solid waste, especially electronic and electrical waste, either going to sorting (and subsequently to reuse) or disposed of with mixed waste after use. The lowest percentage of reuse of nano-Ag is 12% (Romania), the highest is 41% (The Netherlands, Table S14). Flows to the technical compartments also vary strongly for nano-TiO<sub>2</sub>. While in Cyprus, there is no release to reuse and very little to landfill (3%), most of the releases

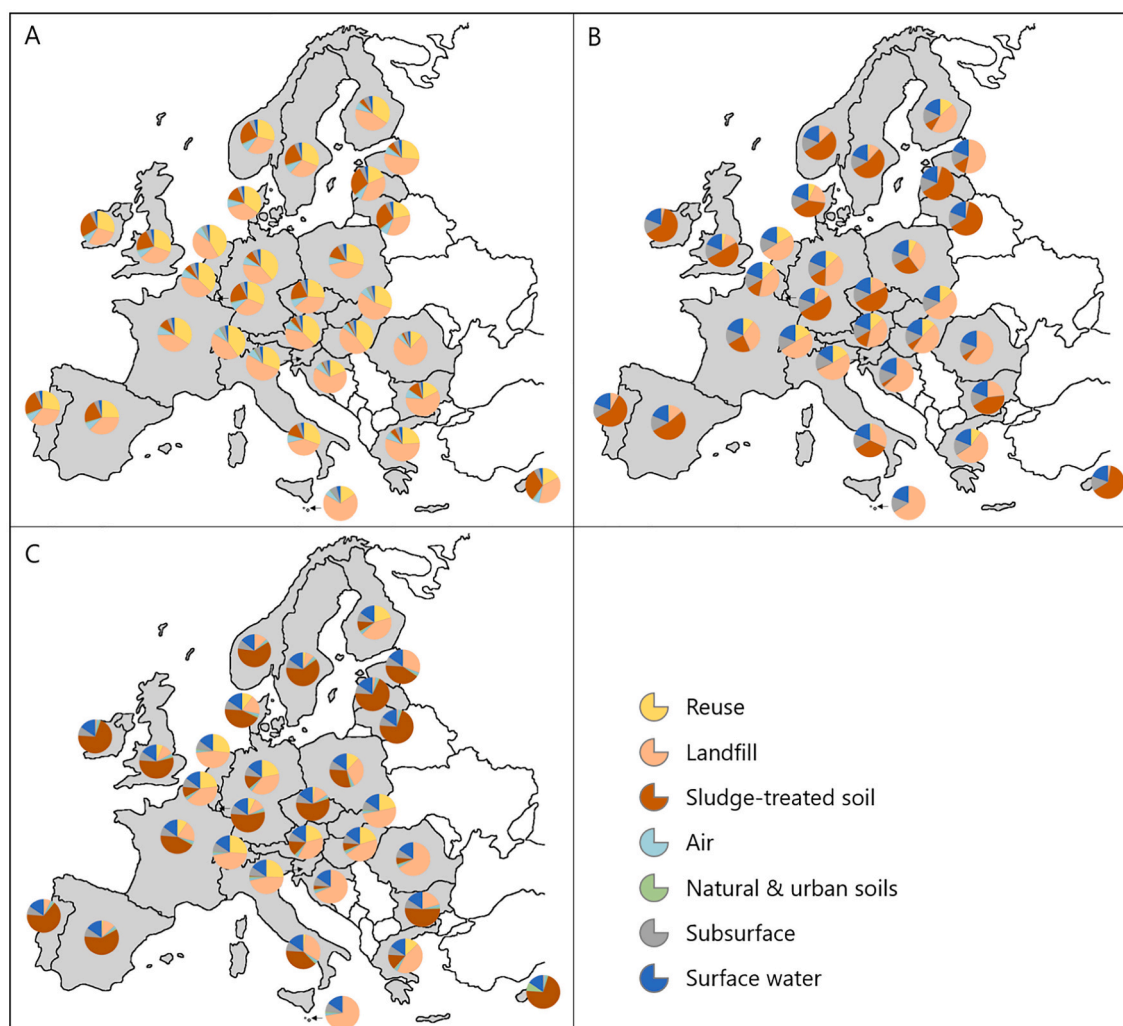
go to sludge-treated soil (63%, Table S15). On the opposite, sludge-treated soil in Slovakia does not receive any nano-TiO<sub>2</sub>, 14% of nano-TiO<sub>2</sub> goes to reuse and 53% to landfill. Nano-ZnO is present in even higher shares than nano-TiO<sub>2</sub> in personal care products, so its shares going to technical compartment depend even more on sludge treatment and vary even more among countries. For example, flows to sludge-treated soils vary from 0% of total releases (for example in Switzerland, the Netherlands and Malta) to 71% (Cyprus, Ireland and Lithuania, Table S16). It is worth highlighting that all nano-ZnO going to reuse comes from the ash of waste incineration plants.

While nano-TiO<sub>2</sub> enters the environment mostly in pristine forms (Fig. S14), nano-Ag and nano-ZnO are subject to more transformations and high shares of these ENMs arrive as dissolved and transformed forms (Figs. 2 and S9).

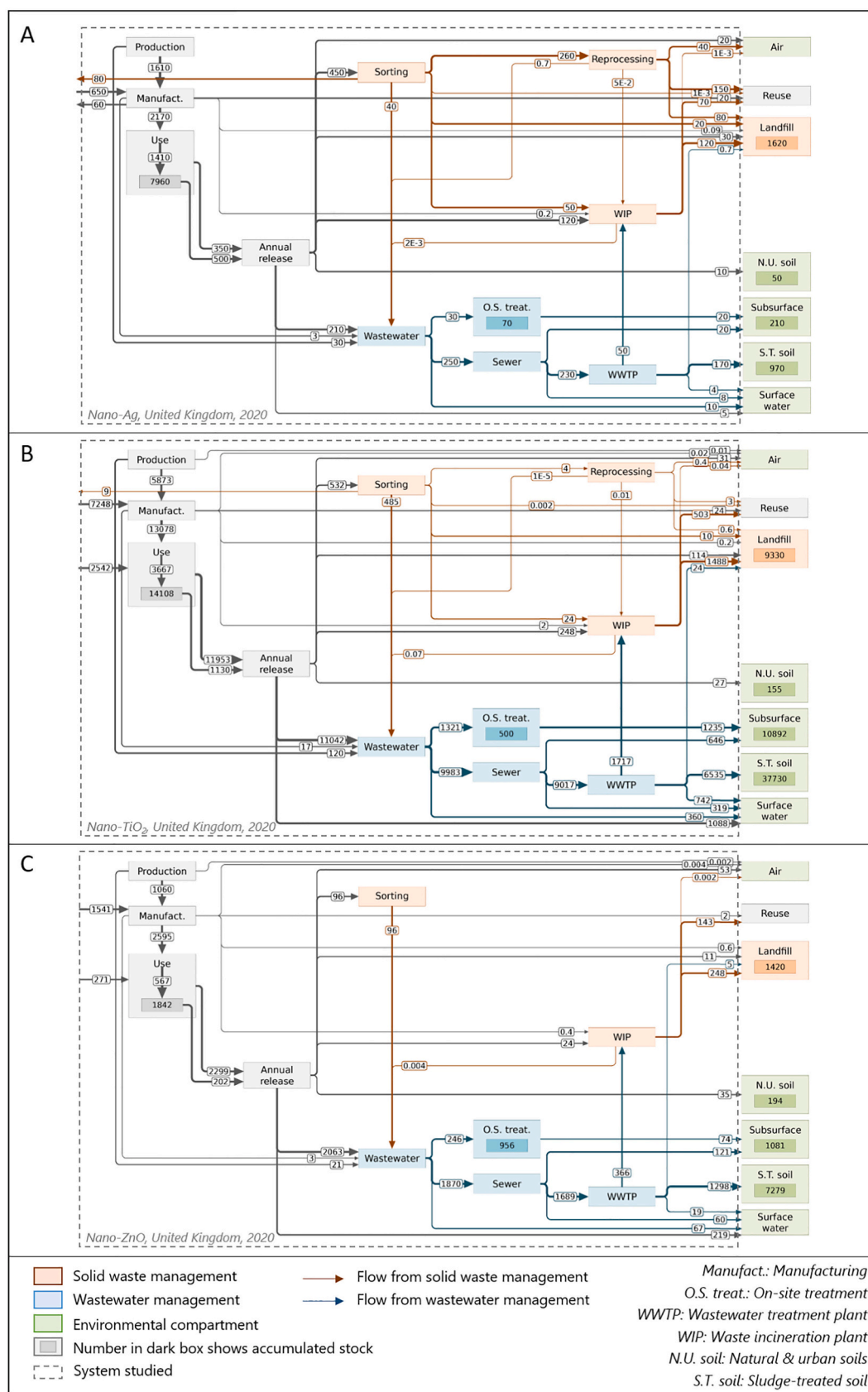
### 3.4. Overall IDPMFA results

#### 3.4.1. Description of pathways

In the UK in year 2020, most of the nano-Ag released after use goes to solid waste sorting (450 kg), mainly as waste printed circuit boards and other electronics (Fig. 5A). From there, a significant portion goes to metal reprocessing (260 kg) and reuse (150 kg), where it occurs at 98% in transformed forms. The reader is referred to Fig. S12 for probability distributions associated with selected flows of nano-Ag to solid waste management in the UK in 2020.



**Fig. 4.** Releases of nano-Ag (A), nano-TiO<sub>2</sub> (B) and nano-ZnO (C) to the final compartments of the model in each European country in 2020. Countries shaded in grey are those considered in the study. Fig. S11 includes maps with the names of each country.



**Fig. 5.** Flows of nano-Ag (A, in kg), nano-TiO<sub>2</sub> and nano-ZnO (both in tonnes) in the United Kingdom in 2020. Numbers shown are the means of the corresponding probability distributions. The thickness of the arrows depends on the order of magnitude of the flows. Darker boxes within a compartment represent sinks with the accumulated mass between 2000 and 2020.

The environmental compartment receiving the highest amount of nano-Ag in the UK in 2020 is sludge-treated soil. 170 kg of nano-Ag flows to this compartment, contributing to a total of 970 kg accumulated over the time period 2000–2020. The subsurface is another significant sink for nano-Ag, with 210 kg accumulated over the last 20 years.

After use, nano-TiO<sub>2</sub> mainly flows to wastewater (11,042 t in the UK in 2020, Fig. 5B), since the main product category in which it occurs is personal care products (76% of manufacturing, Kuenen et al., 2020). Nano-TiO<sub>2</sub> in wastewater mainly goes to sewer (9983 t), then WWTP (9017 t) and finally sludge-treated soils (6535 t). As a result, sludge-treated soils constitute the main environmental sink for these ENMs, with 37,730 t accumulated over the years. Nano-TiO<sub>2</sub> is also directly released to surface water during bathing (1088 t), to which misconnections, leakages and overflows add, leading to 2509 t of nano-TiO<sub>2</sub> released to surface water in the UK in 2020. The third environmental compartment in which nano-TiO<sub>2</sub> ends after flowing in wastewater is subsurface, where 1881 t arrive in 2020. Subsurface is the second highest sink for nano-TiO<sub>2</sub>, with 10,892 t accumulated from 2000 to 2020.

Regarding solid waste management, 532 t of nano-TiO<sub>2</sub> enter sorting in the UK in 2020, mainly in personal care products remaining in packaging waste. 91% of it (485 t) are washed away towards wastewater and contribute to the flows described above. 4 t only go to reprocessing, mainly with the metal and glass they coat. 3 t go back to the market via reuse, which represent 0.05% of what was produced that year.

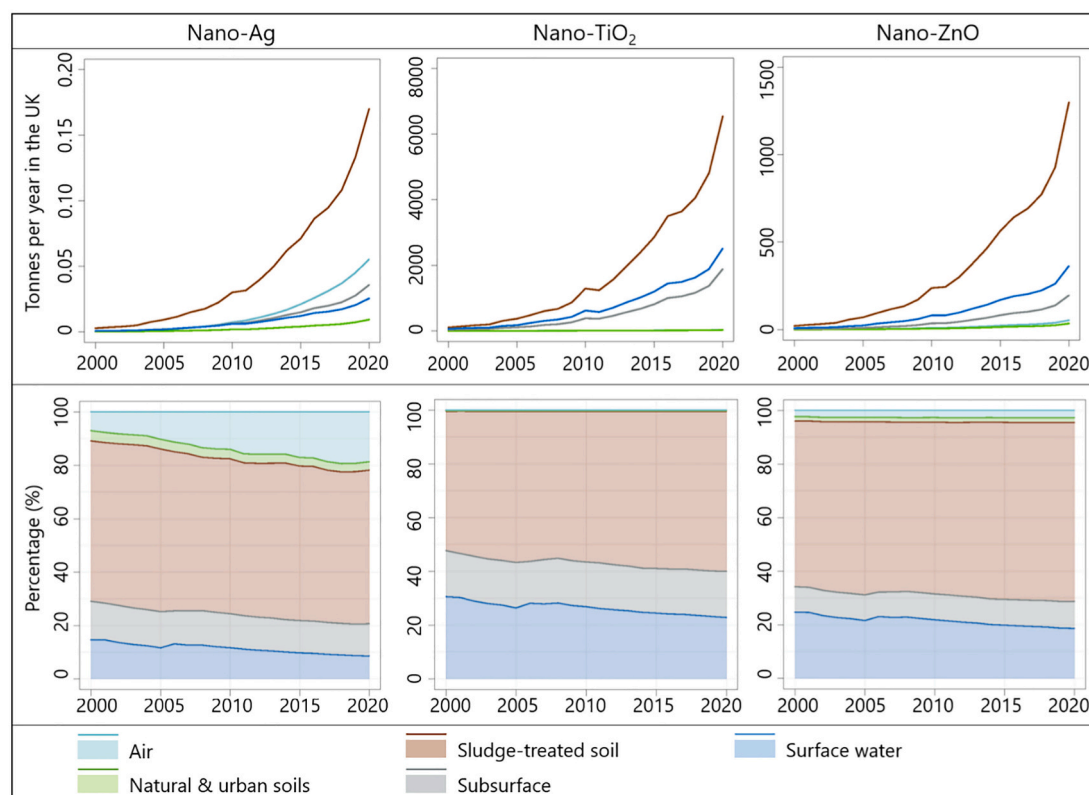
Nano-ZnO is present in three product categories (Kuenen et al., 2020): personal care products (85%), paints and lacquers (13%) and antibacterial products (2%). When in personal care products, nano-ZnO flows to sorting where it is washed away with the product remaining in the packaging (96 t in the UK in 2020). Paints and lacquers are applied on buildings. As buildings were assumed to have a mean lifetime of 80 years (Sun et al., 2016), nano-ZnO in these products do not reach end-of-

life in the time period considered in this model. Finally, antibacterial products were assumed to be small textile and plastic items disposed of with mixed waste. Therefore, in our model, nano-ZnO does not reach reprocessing (Fig. 5C). It mainly flows to wastewater (2063 t released after use), sewer (1870 t), WWTP (1689 t) and sludge-treated soil (1298 t). This compartment is again the main sink of ENMs, with 7279 t of nano-ZnO accumulated in the UK over the last 20 years. These ENMs also flow to surface water in significant amounts: 365 t are released there in 2020. In comparison, air and natural and urban soils receive low amounts of nano-ZnO, which then mainly come from the weathering of paints and lacquers. 53 t are released to air, while 35 t are released to natural and urban soils.

### 3.4.2. Releases to final compartments

Sludge-treated soils constitute the environmental compartment receiving the highest part of nano-Ag, nano-TiO<sub>2</sub> and nano-ZnO, not only in 2020, but over the whole time period considered in the models, from 2000 to 2020 (Fig. 6). The parts of nano-TiO<sub>2</sub> and nano-ZnO going to sludge-treated soils increase slightly over time (from 52% to 60% and from 62% to 67%, respectively), while the parts going to surface water tend to decrease (from 31% to 23% and from 25% to 19%, respectively). This is due to the improvement of wastewater management and treatment technologies over the years.

Nano-Ag shows a different trend, with the share of flows to surface water decreasing from 15% to 9% as shares of flows to air increasing from 7% to 19%. This is due to the fact that the main product category of this ENM is not personal care products as for nano-TiO<sub>2</sub> and nano-ZnO, but printed electronics. While personal care products are assumed to be completely used (and released) over the course of one year, printed circuit boards can stay in “stock” in households and offices for several years, going to sorting and reprocessing more and more as time goes. Since flows out of reprocessing are the main contributors to releases to air in 2020 (Fig. 5), flows to air increased to the extent shown in Fig. 6.



**Fig. 6.** Evolution of flows of nano-Ag, nano-TiO<sub>2</sub> and nano-ZnO towards environmental compartments in the United Kingdom from 2000 to 2020. The top row shows the absolute amounts, the bottom row the relative distribution.

The evolution over time of the forms in which the ENMs are released shows that although the amounts released increase over the years by several orders of magnitude, the proportions of the different forms remain quite stable (Fig. S13). Nano-Ag and nano-ZnO are mainly released in transformed forms (about 70% and 80%, respectively). The dissolved material represents 15% of released nano-Ag in 2000 and 9% in 2020. This decrease is roughly compensated by the increase of the matrix-embedded releases, from 9% in 2000 to 17% in 2020, and is explained by the role that reprocessing of printed circuit boards plays in releases to air as matrix-embedded, which overcame in time the dissolved releases from other types of products. The proportion of nano-Ag in forms that could still be available as ENM to the organisms (i.e. pristine and matrix-embedded) therefore increases from 16% in 2000 to 21% in 2020. On the opposite, almost all of the nano-TiO<sub>2</sub> released is in pristine and matrix-embedded forms (>99%).

#### 4. Discussion

With the current state of analytical methods for ENM detection in environmental samples, there is still only a limited number of measurements available (Gondikas et al., 2018; Loosli et al., 2019; Wagner et al., 2014). While these studies provide useful information on the presence, identity and concentration of nanoparticulate materials and in some cases even the unambiguous detection of engineered particles (Gondikas et al., 2014; Loosli et al., 2019), they do not allow to gain a complete picture of ENM exposure in the environment. To get quantitative data on environmental releases and exposure of ENMs, modelling is therefore still an indispensable tool (Nowack et al., 2015). Progress has been made both on modelling the environmental releases as well as the environmental fate. The release models are key as they provide the input to the fate models and quantify the amounts of ENM released by direct and indirect pathways into the environment. One family of release models has evolved out of the initial PMFA release model published by Gottschalk et al. (2009). Different aspects of the model have been improved over the years and new functionalities have been added but most of them remained separate. The current work now integrates all updates into one coherent modelling framework. Table S17 summarises the different aspects included in previous models and those integrated in the present model.

One major advancement includes the combination of the dynamic MFA model (DPMFA) (Bornhöft et al., 2016) with the updates made on the static model, mainly in the field of waste treatment. A dynamic assessment is needed in order to predict accumulated masses in sink compartments and to be able to model releases during the use of ENMs incorporated in products with a long lifetime, e.g. in construction or the automotive sector (Song et al., 2017). Inclusion of the dynamics therefore results in retarded release and this yields lower estimates of current environmental concentrations than static models. This becomes important for those ENMs that have a high share in long-lived applications such as CNT with their important use in polymer nanocomposites (Nowack et al., 2013) and nano-Ag in printed circuit boards. TiO<sub>2</sub> and ZnO on the other hand with very high shares in cosmetics with a relative fast turnover show less difference between a dynamic and a static assessment (Sun et al., 2016).

Dynamics not only refer to product lifetimes and the increase in ENM production and use over the years but also to changes in the waste treatment systems. There has been a constant improvement in wastewater treatment plants by addition of more and advanced treatment stages and this has an effect on the elimination rate during treatment, an aspect that has been included in the model updates by Rajkovic et al. (2020) and that is now added to the new IDPMFA. Whereas historic releases of ENMs from WWTP have no influence on the currently released amounts to freshwater, the historic releases are important to predict the amount currently stored in environmental sinks such as soils and sediments.

Especially with respect to waste treatment, the consideration of the

national scale is very important as different countries have a different evolution of the waste treatment system at different speeds. This aspect is contributed to the IDPMFA by the previous model of Adam and Nowack (2017). Especially with respect to the final treatment of solid waste, the two main options of landfilling or incineration result in greatly different final fate of the ENM, especially for those materials that are transformed during incineration such as carbon-based or oxidizable ENMs. A further refinement of the modelling can even consider a local scale, e.g. the situation around one single city (Parker and Keller, 2019). Kuenen et al. (2020) added another aspect to the model which is the regional variation in ENM production and manufacturing. There is only a very limited number of production sites of ENMs within Europe and release during synthesis of ENMs can only occur at these local hotspots.

An important part of the IDPMFA is the detailed description of the fate of the ENM during all parts of the recycling process. As many ENMs are contained in products that have a very high share of separate collection and recycling, a detailed analysis of their flows during these processes is indispensable to get a full picture of all flows. Initial models such as PMFA or DPMFA considered flows into recycling but treated this compartment as a sink without further outflows. Caballero-Guzman et al. (2015) were the first to track the flows of two ENMs in the recycling system of one country (Switzerland) for some product categories. Some of the subsequent models included these flows that were available for Ag and TiO<sub>2</sub>. There are two main processes to consider: the sorting and the reprocessing step. The flows of ENMs during the sorting step were assessed by Rajkovic et al. (2020) for Europe for all product categories relevant for ENMs. In the present study, we now included the systematic assessment of the ENM flows out of reprocessing, therefore fully completing the mass flow assessment of ENMs in the technical compartments. The flows into the “sink” compartment “recycling” in previous models can now be fully attributed to subsequent compartments and only real sinks such as landfill, soils or sediments remain in the model. There is still one compartment in the model treated as a final sink although it is actually not: reuse of ENMs. No loop for reuse is included and therefore no transfer back into production and manufacturing. However, this is made on purpose because reused materials can be embedded in product categories different from those considered specific to ENMs – they are “polluting” secondary materials that are not part of the model because they do not represent nano-products.

A final aspect now also included in the full IDPMFA model are the forms of release. The large majority of all mass flows models, be it the ones based on the PMFA, e.g. Sun et al., (2014) and Gottschalk et al., (2015) or those from other research groups, e.g. Keller et al. (2013); Parker and Keller (2019); Song et al. (2017), only consider a generic ENM, not distinguishing between the different forms of release. Including form-specific transfer coefficients requires experimental data on each scenario occurring during the lifecycle. Since this data is very scarce and only recently became available, those earlier models were built based on a generic material. The use of probability distributions in the PMFA enables the consideration of qualitative data, as it makes it easier to translate uncertain knowledge into numbers. Adam et al. (2018) have indeed shown that for Ag and TiO<sub>2</sub>, considering the different forms of the ENM is extremely relevant as different forms can be released into different environmental compartments. The same behaviour is also found for ZnO that was assessed in the current work: air, soil and surface water show very different distributions of forms of release, caused by different processes during release and different technical compartments that are passed on the way to the environment. The passage through wastewater treatment is completely changing the speciation to a fully transformed (sulfidized) form while release into air is mostly in the matrix-embedded form caused by the abrasion processes. It is important to recognize that these forms represent the ones initially released into the environment and not the forms actually present there. In order to predict the form in an environmental compartment, environmental fate models would need to include the fate and

further transformation processes of matrix-embedded ENMs. The same applies of course also to the other forms, e.g. the pristine ZnO released into surface water. The distribution provided by our analysis represents also for pristine ZnO the initial form released into surface water, not the final form present in water. Transformation of ZnO, e.g. by dissolution or phase transformation into a thermodynamically more stable phase, e.g. ZnCO<sub>3</sub> or Zn-phosphate may occur and would need to be included in environmental fate models (Suhendra et al., 2020).

The IDPMFA modelling results provide in their entirety the most accurate estimation of ENM releases to the environment. However, as all release models for ENMs, its results cannot be validated (Nowack et al., 2015), as no data are available that could be used to estimate the mass of different forms of ENMs released into the environment of different countries. Similar to all other mass flow models for ENMs, its accuracy is mainly determined by the availability of production and product distribution data as well as the quantification of the amount and form of the released ENMs (Caballero-Guzman and Nowack, 2016).

Being a dynamic model, it is able to quantify the amount of ENMs deposited in final sinks such as soils and landfills. Most fate models such as Nanofate (Garner et al., 2017), SimpleBox4Nano (Meesters et al., 2014) or a water quality model that was amended with nano-specific features (Bouchard et al., 2017) have so far only targeted surface waters and did not consider fate in soils. Given the importance of the soil compartment for receiving a very large share of the ENM mass, having data available on the amount and form of ENMs added to soils over time is an indispensable input to all soil fate models as accumulation over time will be a very relevant issue to consider (Svendsen et al., 2020). The results from fate models can only be as good as the input data provided by MFA models and neglecting dynamic aspects and form-specific releases is greatly limiting the usefulness of the results from fate models.

As fate models are used to derive predicted environmental concentration (PEC values), they form the basis for all environmental risk assessments (Wigger et al., 2020). Simplified PEC values not including any fate processes were used in the past to perform environmental risk assessments, both based on static (Coll et al., 2016) as well as dynamic MFAs (Wang and Nowack, 2018). So far, none of the available risk assessments has considered the different forms of release but looking at the various distributions of the different forms released to the environmental compartments shows us the importance of assessing the hazard potential based on form. A sulfidized (transformed) form of nano-Ag is known to have very different toxicity than the pristine nano-Ag (Levard et al., 2013). The results of the IDPMFA performed in this work are therefore highly relevant for environmental risk assessment, as they can be used both as input into fate and uptake models as well as to understand the forms that need to be tested for ecotoxicity.

As all models, the IDPMFA in its current form has some limitations. Some parts of the model are not country-specific, e.g. the distribution of ENM to product categories, the wastewater management and the recycling system. Information on these systems could be added relatively straightforward as this information should be available on a country-specific basis and would then further allow to identify differences between European countries. Variations in nano-product use between countries is much more difficult to quantify as this information is even hard to obtain on an aggregated level on the scale of Europe.

One of the final sinks of the model, the landfills, are considered as final sink, comparable to all other existing release models for ENMs. However, release of ENMs from landfills by leaching or airborne emission may occur (Part et al., 2018). A nanoparticulate metal fraction has been observed in landfill leachates but it is normally considered to be of natural origin (Part et al., 2018), however, the presence of this fraction indicates that nanoparticles can to a certain extent be mobile in landfills. Engineered TiO<sub>2</sub> particles have been detected by electron microscopy in construction waste landfills but no quantification of the engineered fraction was possible (Kaegi et al., 2017). So far, no fate model of ENMs in landfills exists but further progress in this field may allow to couple the mass flows to the landfills with specific fate models to be developed

for ENMs in landfills.

## 5. Conclusions

The IDPMFA model constitutes the currently most advanced tool to predict the mass flows of ENM to the environment and include the released forms into the flow assessment. The IDPMFA will therefore be fully compatible with the requirements of advanced environmental fate models such as SimpleBox4Nano (Meesters et al., 2014) or NanoFATE (Garner et al., 2017) and risk assessment procedures that consider the different chemical/physical properties of the different forms that are released (Svendsen et al., 2020). As soon as fate models are used to derive PEC values the results of the models can only be as good as the input values (= environmental releases) which are provided by MFA models like the IDPMFA. The IDPMFA results can also be used for ecotoxicity assessment, as the environmental releases and their forms are important for uptake assessment and understanding the toxicity mechanisms at stake. This model therefore constitutes a very important piece for environmental risk assessment. The IDPMFA also aligns well with the recent advancement on the grouping of nanoforms that also includes life-cycle considerations and exposure (Stone et al., 2020).

## Model availability

The codes are available for download at DOI: <https://doi.org/10.5281/zenodo.4050159>.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

The Supporting Information contains a brief description of the newly evaluated reprocessing systems, the input data to determine the nano-ZnO transformations, the data quality assessment and probability distributions and additional results.

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