

# Global life cycle releases of engineered nanomaterials

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Received: 13 February 2013 / Accepted: 3 May 2013 / Published online: 16 May 2013  
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**Abstract** Engineered nanomaterials (ENMs) are now becoming a significant fraction of the material flows in the global economy. We are already reaping the benefits of improved energy efficiency, material use reduction, and better performance in many existing and new applications that have been enabled by these technological advances. As ENMs pervade the global economy, however, it becomes important to understand their environmental implications. As a first step, we combined ENM market information and material flow modeling to produce the first global assessment of the likely ENM emissions to the environment and landfills. The top ten most produced ENMs by mass were analyzed in a dozen major applications. Emissions during the manufacturing, use, and disposal stages were estimated, including intermediate steps through wastewater treatment plants and waste incineration plants. In 2010, silica, titania, alumina, and iron and zinc oxides dominate the ENM market in terms of mass flow through the global economy, used mostly in coatings/paints/

pigments, electronics and optics, cosmetics, energy and environmental applications, and as catalysts. We estimate that 63–91 % of over 260,000–309,000 metric tons of global ENM production in 2010 ended up in landfills, with the balance released into soils (8–28 %), water bodies (0.4–7 %), and atmosphere (0.1–1.5 %). While there are considerable uncertainties in the estimates, the framework for estimating emissions can be easily improved as better data become available. The material flow estimates can be used to quantify emissions at the local level, as inputs for fate and transport models to estimate concentrations in different environmental compartments.

**Keywords** Life cycle · Emissions · TiO<sub>2</sub> · SiO<sub>2</sub> · ZnO · CeO<sub>2</sub> · Al<sub>2</sub>O<sub>3</sub> · Nano-Ag · Nano-Cu · Nano-Fe · CNTs · Nanoclays

## Introduction

While some engineered nanomaterials (ENMs) have been used for several decades, their volume of production and diversity of applications have grown extensively over the past decade and continue to grow rapidly. ENMs are currently used in many applications including agriculture (Lopez-Moreno et al. 2010; Peralta-Videa et al. 2011; Gruère 2012; Khot et al. 2012; Zhao et al. 2012); aerogels (Bigall et al. 2009); aerospace (Baur and Silverman 2007); automotive (Presting and König 2003; Salonitis et al. 2010;

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**Electronic supplementary material** The online version of this article (doi:10.1007/s11051-013-1692-4) contains supplementary material, which is available to authorized users.

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Coelho et al. 2012); catalysts (Zhou et al. 2011); coatings, paints, and pigments (Khanna 2008; Dhoke et al. 2009; Gopalakrishnan et al. 2011); composites (Borchardt 2003; Khanna and Bakshi 2009; Sahoo et al. 2010; Petrov and Georgiev 2012); construction (Lee et al. 2010); cosmetics (Musee 2011; Sabitha et al. 2012; Singh and Nanda 2012); electronics and optics (Alda et al. 2005; Avasthi et al. 2007; Song et al. 2012; Subramanian and Takhee 2012); energy (Serrano et al. 2009); environmental remediation (Dionysiou 2004; Khin et al. 2012); filtration and purification (Savage and Diallo 2005; Dhakras 2011); food products (Weiss et al. 2006; Blasco and Picó 2011); medical (Farokhzad and Langer 2006; Boisseau and Loubaton 2011); packaging (Silvestre et al. 2011); paper and board (Kharisov and Kharissova 2010); plastics; security (Marín and Merkoçi 2012); sensors (Ding et al. 2010, Duncan et al. 2012; Su et al. 2012; Tan et al. 2012); and textiles (Qian and Hinestroza 2004; Wong et al. 2006), and research is underway on many new applications.

Although there are many benefits to using ENMs in these various applications, there is concern that their environmental implications are not fully understood. In addition to determining their potential toxicity (Shvedova et al. 2005; Klaine et al. 2008; Navarro et al. 2008; Xia et al. 2008; Gao et al. 2009; Lanone et al. 2009; Damoiseaux et al. 2011; Peralta-Videa et al. 2011; Thomas et al. 2011), there is a need to estimate the concentrations of specific ENMs in different environmental media as well as the likelihood of exposure to humans and ecosystem receptors. Estimating environmental concentration and exposure of ENMs requires information on potential emissions during the life cycle of the ENMs (i.e., manufacturing, incorporation of ENMs into intermediate and final products, use of product containing ENMs, disposal, or recycling), as well as the subsequent fate and transport calculations (Keller et al. 2010; Liu and Cohen 2012; Lowry et al. 2012a) taking into account the dispersal of the ENMs in different media and their transformations (e.g., dissolution, aggregation, oxidation, reduction, sulfidation, etc.).

As a necessary first step, this study aims at providing a broad-brush, global picture of ENM emissions from a wide range of major ENM applications during their life cycles. The result of this study can be used as an input to site-specific fate and transport modeling studies to predict concentrations.

## Prior approaches to modeling ENM emissions

Thus far, research on the subject of ENM release and resulting environmental concentrations has fallen into one of three main categories: (1) experimental studies designed to measure the release of specific ENMs from various products; (2) studies focused on the development of application-specific ENM release models; and (3) research focused on the development of models that account for the entire life cycle release of ENMs from various applications. Application-specific experiments and models provide the basis for constructing emission scenarios, but are usually limited to the use phase. Thus, a life cycle approach that combines all potential applications and release scenarios is more appropriate for determining the levels of particular ENMs entering the environment and for predicting initial environmental concentrations.

Experimental release studies provide the most concrete basis for estimating emissions. For example, Kaegi et al. (2008) used analytical electron microscopy and bulk chemical analysis to assess TiO<sub>2</sub> release from exterior facades into surface waters. During the experiments, runoff was collected from new and old painted building facades and analyzed. They estimated the amount of synthetic TiO<sub>2</sub> in runoff to be about  $3.5 \times 10^7$  TiO<sub>2</sub> particles/L (for particles <100 nm in diameter) and concluded that TiO<sub>2</sub> particles from exterior facades may represent a significant release into surface waters. In an effort to better understand the release of TiO<sub>2</sub> from consumer textiles during the washing process, Windler et al. (2012) quantified the amount and form of Ti released from six different textiles during commercial and laboratory washing. They found that, aside from one textile that was an outlier, only about 0.01–0.06 % of the Ti present in the textile was released and that a majority of it was in an aggregate form. However, in one case 3.4 % of the Ti present in the textile was released, with a significant amount in the nanoform. A concentration of 1.5–15 µg/L of Ti in wastewater was estimated due to the release from textiles. Benn and Westerhoff (2008) investigated the quantity and form of silver released from commercial textiles into wastewater. They analyzed the adsorption characteristics of the released silver in order to determine its potential fate in the wastewater treatment plants (WWTPs) and the associated quantity of silver that may be present in WWTP effluent and biosolids. From 1 to 100 % of

silver was released from socks during the washing process, thus determining that the leaching rate was more dependent upon the manufacturing process than the amount of silver present within the sock.

Prompted by the high likelihood of release of ENMs into wastewater during the use phase of certain consumer products, Kiser et al. (2009) conducted a study to quantify Ti concentrations in a full-scale municipal WWTP, characterize the morphology and composition of Ti-based solids in consumer products plus wastewater effluents and biosolids, and quantify Ti concentrations in laboratory-scale treatment reactors and the sorption capacity of wastewater biomass for TiO<sub>2</sub> nanoparticles. The measured distribution of TiO<sub>2</sub> revealed the likely pathways and quantities of ENM entry from WWTP into the environment. The conclusions of the study supported the assertion that a majority of Ti will associate with biomass and be present in finished biosolids, while a relevant portion will remain in the WWTP effluent that is released into the environment. The presence of TiO<sub>2</sub> in biosolids presents another likely route of environmental exposure through the application of the treated biomass to agricultural land and disposal through landfilling or incineration.

O'Brien and Cummins (2010) modeled the release of TiO<sub>2</sub> from exterior paints, Ag from food packaging, and CeO<sub>2</sub> from exhaust emissions into surface waters and the atmosphere. Assuming a 5–10 % market penetration in Ireland, probability distributions for the predicted concentrations of the selected ENMs in air and surface water were constructed.

Several studies have attempted to develop a theoretical framework for quantifying the release of ENMs from specific applications. Boxall et al. (2007) assessed likely release scenarios of ENMs based on product use and estimated ENM concentrations in products in applications including cosmetics, paints and coatings, catalysts and lubricants, water treatment, food and food packaging, human and veterinary medicines, and plant protection products. The authors used simple algorithms to compute the predicted concentrations of ENMs in air, soil, and water for Ag, AlO<sub>3</sub>, Au, CeO<sub>2</sub>, fullerenes, hydroxyapatite, latex, organo-silica, SiO<sub>2</sub>, TiO<sub>2</sub>, and ZnO.

Blaser et al. (2008) estimated exposure to the aquatic environment from Ag released from plastic and textile applications in Europe. A mass flow analysis was conducted and emissions were estimated, along with an

evaluation of toxicity, in order to present a risk characterization for nanosilver. The mass flow model estimated low, medium, and high levels of nanosilver release. The study found that a majority of silver present within WWTP influent is removed and incorporated into sewage sludge. However, the resulting modeled releases into surface water are highly sensitive to the assumed effectiveness of the WWTP treatment.

Based on a developing body of knowledge of application-specific ENM release, some researchers have begun investigating the life cycle emissions of ENMs into the environment in order to generate estimates of their environmental concentrations. This approach is designed to account for all possible ENM emissions, considering all applications and life cycle stages of the products. Mueller and Nowack (2008) used a life cycle approach to model the release and resulting environmental concentrations of ENMs in the Swiss environment. Using several assumptions about the production and use of nanoparticles in Switzerland, they modeled expected concentrations of Ag, TiO<sub>2</sub>, and carbon nanotubes (CNTs) in water, air, and soil for that region. The study took into account the share of total ENM production allocated among product categories and uses and used assumptions on how the ENMs were incorporated into the products to determine their likely release scenarios. Due to the lack of application-specific data, the authors allocated total ENM production between product categories using inventory information from the Woodrow Wilson Institute and the results of web-based product research. Using realistic and a high release scenario, based on literature values and expert estimations, they were able to characterize the potential risk of these three nanoparticles. The primary flow of ENMs was determined to take place between waste incineration plants (WIPs) and landfills due to high rates of removal of ENMs during wastewater treatment and the subsequent incineration of sludge. The authors recognized several areas for continued research including the behavior of nanoparticles in the environment as in WIP and WWTP systems; emissions from production; and transformation, degradation, and bioaccumulation of nanoparticles.

Expanding on the previous study, Gottschalk et al. (2009) created a probabilistic model using Monte Carlo and Markov chain Monte Carlo simulations to account for the significant variations and uncertainties in the model input parameters. The model was applied

to generate probability distributions for the concentrations of  $\text{TiO}_2$  in Swiss soil, air, sediment, and surface waters (Gottschalk et al. 2010a). This modeling approach was applied to model the environmental concentrations of  $\text{TiO}_2$ , ZnO, Ag, CNTs, and fullerenes for the United States, Switzerland, and Europe. Worldwide ENM production estimates were used, and production volumes were allocated to each region based on population size. Differences in some of the model parameters between the countries were accounted for, such as percentage storm water overflow and differences in the management of sewage sludge. The results of the study showed that  $\text{TiO}_2$  is expected to have the highest concentration in all environmental compartments due to its higher worldwide production volumes and applications.

Using market research information on ENM production volumes and applications, and peer-reviewed literature for emission estimates, in this study we estimate potential releases of ENMs into the environmental compartments of air, surface water, and soil, plus landfills as a final stage for a fraction of the ENMs. Intermediary compartments considered were WWTPs and WIPs. Ten ENMs were evaluated based on their production volumes and the potential for environmental and human health risk. While these estimates are based on limited current knowledge and extrapolations from focused studies to global scenarios, they provide a framework for refining predictions of emissions and initial environmental concentrations for air, water, and soils. This information can then be linked to ENM fate and transport models (Liu and Cohen 2012) to determine potential exposure concentrations for local or regional conditions. One important difference in our approach is the reliance on market studies to better estimate ENM production and application volumes. This differs from previous studies, for example, Gottschalk et al. (2010b), which relied on unspecified sources for their global ENM production, and a qualitative interpretation of commercially available ENM-containing products. Other production estimates have relied on patent information, number of employees, and other proxies (Hendren et al. 2011).

## Methods

Building on the studies reviewed in the previous section, this section presents the method of estimating

ENM emissions employed in our study. Global production rates of the selected ENMs were based on a market study (Future\_Markets 2012). Even though the market study is based on manufacturer information gathered through surveys and interviews, in many cases there is a substantial range between the minimum and maximum of estimated production rates. The market study was also used to estimate the fraction of global annual production directed to specific applications for a given ENM. Of the applications discussed in the market study, those which were considered to be either insignificant or have limited pathways to the environment during the use phase (e.g., academia and research centers, aerogels, communications, rheological modifiers, security, and sensors) were omitted from the analysis.

### Basic assumptions

We assumed that all ENMs produced will be eventually released into the environment or end up in landfills at a certain point in time. It is quite likely that a large fraction of these ENMs will transform (e.g., aggregate to larger particles, dissolve, oxidize or be reduced, and acquire coatings) once they are released, and this in part is taken into consideration in our estimate of partitioning between WWTP liquid effluent and sludges. We track the original mass of ENMs through the world economy, including their transformed products, but cannot at this stage specify the transformation products beyond their initial release. In other words, long-term accumulation of ENM as stock in the economy (excluding landfill) is assumed to be minimal. Our estimates of emissions are time-integrated: Emissions in near-term and long-term are aggregated altogether. Under these assumptions, a mass balance approach is used to estimate ENM material flows.

### Release during manufacturing

Although ENM emissions may be accounted for within regulatory [e.g., National Pollutant Discharge Elimination System (NPDES) permits, Resource Conservation and Recovery Act (RCRA) reports] and voluntary (e.g., Toxics Release Inventory) reporting in the United States, ENMs are not distinguished from their bulk counterparts within these documents. Gottschalk and Nowack (2011) estimated from 0 to 2 % release during manufacturing, with a distribution

**Table 1** Potential ENM release during manufacturing

	Total release during manufacturing (%)	To air (%)	To water prior to WWTP (%)	To landfill (%)
Low estimate	0.1	10	10	80
High estimate	2.0	40	40	20

to atmosphere, water that reaches a WWTP, and soil. The ENM release estimates in this study consider total emissions from 0.1 to 2 % of the total production rate, with a distribution to air, water, and landfill as indicated in Table 1. These release estimates are generic for all ENMs, given the lack of ENM-specific manufacturing information. Although there are some emission control devices for different processes related to the synthesis and handling of ENMs, there is considerable uncertainty as to the state and amount of ENMs that may be removed by such control devices. It is assumed that the upper and lower bound estimates of ENM emissions are equally applicable to both ENM manufacturing and manufacturing of ENM-containing products. One can expect that differences may arise based on the method of ENM synthesis and products into which ENMs are incorporated, but given the lack of monitoring of industrial emissions of ENMs and scant information on production processes, it would be difficult at this stage to account for such differences.

Release during use

Estimates for release during the use phase were based on the available studies for different nanomaterials and applications, as listed above (Boxall et al. 2007; Benn and Westerhoff 2008; Blaser et al. 2008; Kaegi et al. 2008; Mueller and Nowack 2008; Kiser et al. 2009; Gottschalk et al. 2010a; O’Brien and Cummins 2010; Windler et al. 2012). Due to the limited number of studies that have been done on releases from specific applications, some estimates are based on information from similar applications or studies on other nanomaterials. The estimated ENM releases during use are presented in Table 2, which provides a low and high estimate for each major application, as well as an estimate of the fraction released into the atmosphere, water flowing to a WWTP, or a release into soil from a discarded product. These release estimates are generic for all ENMs in a given application, since there is little or no ENM-specific data for each application. The balance of ENMs not

**Table 2** Estimates of ENM release into the environment during use

	Low estimate (%)	High estimate (%)	Release into air (%)	Release into WWTP (%)	Release into soil (%)
Academia and research	1	20	5	95	0
Aerospace	0	1	5	90	5
Automotive	0	1	5	90	5
Catalysts	1	5	5	90	5
Coatings, paints, and pigments <sup>a</sup>	10	90	1	60	35
Cosmetics <sup>b</sup>	75	95	1	90	1
Composites	1	10	5	5	90
Electronics and optics	1	5	5	5	90
Energy and environment	5	20	5	5	90
Filtration	10	90	0	95	5
Medical	5	25	5	90	5
Packaging	5	25	0	5	95
Paper and board	5	25	0	5	95
Plastics	5	25	1	5	94
Sensors	0.5	1	0	5	95
Textiles	10	95	5	95	0

<sup>a</sup> Assume 4 % to water bodies via storm water

<sup>b</sup> Assume 8 % to water bodies via swimming

released during manufacturing or use is considered to be discarded via traditional disposal practices. Many of these estimates will require in-depth studies of manufacturer and consumer practices to improve and refine the calculated releases.

### Release during disposal

Estimates of ENM release during disposal were generated based on results from several studies (Shafer et al. 1998; Mueller and Nowack 2008; Kiser et al. 2009), and the United Nations (2011) waste generation and disposal data (United Nations 2011), and 2011 world population data. Waste generation data were available for 71 countries, and waste incineration data were available for 61 countries. By dividing the world into eight regions and extrapolating the available per capita waste generation and incineration rates to the entire region, we estimated an average world municipal waste incineration rate of 19 %. By making assumptions about the disposal methods for each allocation, municipal waste treatment versus direct landfill disposal, we determined ranges of releases from various disposal treatments into the environment (Table 3), which are considered similar for all metal and metal oxide ENMs. The leaching of ENMs from landfills is not included in this estimate because landfill construction and lining systems vary globally and estimates of nanoparticle releases are not available.

Release of ENMs from various product disposal methods (e.g., into landfills, WIP, and WWTP) was estimated using constant multipliers for all ENMs and applications considered, absent ENM-specific data. Estimated rates of release from WIPs are 0.05–1 % into air, 1–50 % into slag, and ~50–98 % captured by filters (Mueller and Nowack 2008). It is assumed that WIP slag and filters are ultimately disposed in landfills. WWTPs are estimated to remove between 75 and 97 % of ENMs from the inflow waters (Kiser et al. 2009) and transfer them to biosolids. The biosolids are then disposed of through incineration, application to agricultural land, and landfills. To close the mass balance,

our estimates must add up to 100 %; the precision in the estimate on the release from WIP into filters reflects this, and not a higher certainty in these values. Low and high estimates in Table 3 are labeled based on potential emissions to air and water. Although the disposal of biosolids varies greatly, a range was established for each route based on reported common practices, thus providing a reasonable range for release estimates (DEFRA 2002; Peckenham 2005). ENMs which are not filtered out of the water by the treatment processes are ultimately released into the environment as effluent. Recycling was not accounted for in this analysis, as recycling methods are extremely varied across products and across regions and specific rates of recycling of ENM-enabled products have not been documented. Following the Mueller and Nowack (2008) approach, we considered slightly different estimates for CNTs (Table 4).

For each ENP, the following equations were used to determine the environmental releases into landfill, soil, water, and air:

$$M_W = \text{mass flow to water bodies} \\ = M_{\text{prod}} \left[ T_{t,e} * \left( F_{m,t} + \sum (F_{u,t,i} + F_{d,t,i}) \right) + F_{u,w} \right]$$

$$M_S = \text{mass flow to soil} \\ = M_{\text{prod}} \left[ \sum F_{u,s,i} + T_{t,b4} * T_{b,s} * F_t \right]$$

$$M_A = \text{mass flow to air} \\ = M_{\text{prod}} \left[ F_{m,a} + \sum (F_{u,a,i} + F_{d,a,i}) + T_{I,\text{air}} * F_{d,I} + T_{t,b} * T_{b,I} * T_{I,\text{air}} * F_t \right]$$

$$M_L = \text{mass flow to landfill} \\ = M_{\text{prod}} \left[ F_{m,L} + F_{d,L} + T_{I,L} * F_{d,I} + T_{t,b} * T_{b,I} * T_{I,L} * F_t + T_{t,b} * T_{b,L} * F_t \right]$$

where  $M_{\text{prod}}$  = production volume of a given ENM,  $T_{I,\text{slag}}$  = transfer from WIP to slag,  $T_{I,\text{filter}}$  = transfer from WIP to filter,  $T_{I,L}$  = total transfer from WIP to landfill =  $T_{I,\text{slag}} + T_{I,\text{filter}}$ ,  $T_{I,\text{air}}$  = transfer from WIP to

**Table 3** Potential ENM release from disposal systems

	Release from WIP into air (%)	Release from WIP into slag (%)	Release from WIP into filters (%)	Release from WWTP into effluent (%)	Release from WWTP into sludge (%)
Low estimate	0.05	50	49.95	3	97
High estimate	1	1	98	25	75

**Table 4** Potential release from disposal systems for CNTs

	Release from WIP into air (%)	Release from WIP into slag (%)	Release from WIP into filters (%)	Release from WWTP into effluent (%)	Release from WWTP into sludge (%)
Low estimate	0.03	75	24.97	3	97
High estimate	1	49.5	49.5	10	90

air,  $T_{t,b}$  = transfer from WWTP to biosolids,  $T_{t,e}$  = transfer from WWTP to effluent,  $T_{b,I}$  = biosolids incinerated in WIP,  $T_{b,s}$  = biosolids applied to agricultural soil,  $T_{b,L}$  = biosolids landfilled,  $F_{m,t}$  = fraction released into WWTP during manufacturing,  $F_{u,t,i}$  = fraction released into WWTP during use from the  $i$ th application,  $F_{d,t,i}$  = fraction released into WWTP during disposal from the  $i$ th application,  $F_t$  = total fraction released into WWTP during life cycle =  $F_{m,t} + \sum(F_{u,t,i} + F_{d,t,i})$ ,  $F_{u,w}$  = fraction released directly into water during use from the  $i$ th application,  $F_{d,l,i}$  = fraction released into WIP during disposal from the  $i$ th application,  $F_{m,L}$  = fraction released into landfill during manufacturing,  $F_{d,L,i}$  = fraction released into landfill during disposal from the  $i$ th application,  $F_{u,s,i}$  = fraction released into soil during use from the  $i$ th application,  $F_{m,a}$  = fraction released directly into air during manufacturing,  $F_{u,a,i}$  = fraction released directly into air during use from the  $i$ th application, and  $F_{d,a,i}$  = fraction released directly into air during disposal from the  $i$ th application.

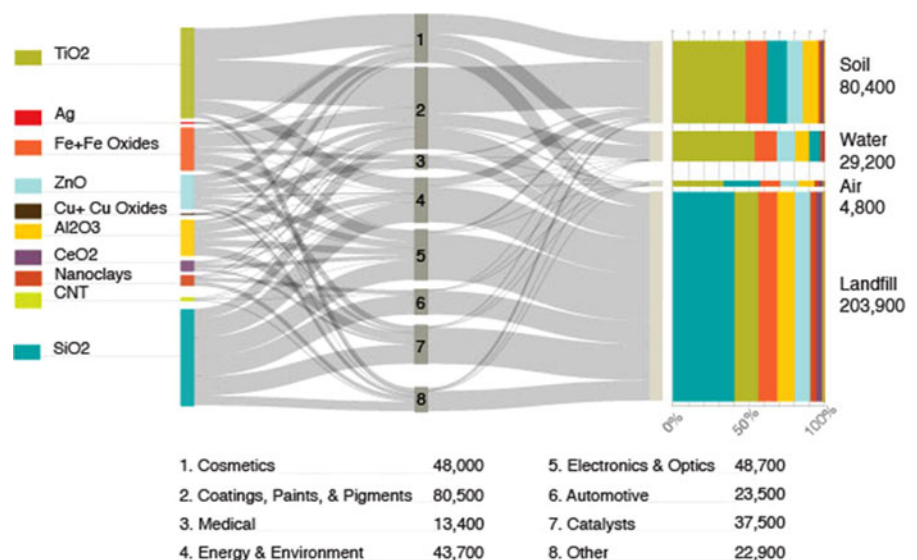
The summation terms ( $\Sigma$ ) integrate the releases over all applications for the use and disposal phases.

Finally, we use Sankey diagrams to visualize the pathways of ENMs, which are widely practiced in the field of industrial ecology in general, and in material flow analysis (MFA) in particular (Hashimoto and Moriguchi 2004; Schmidt 2008; Du and Graedel 2011; Suh and Yee 2011; Reck and Graedel 2012).

**Results**

The global flow of ENMs through the world economy is presented in Fig. 1 for the top 10 ENMs, considering high production estimates for 2010 and high emission assumptions throughout their life cycle. Intermediate flows through WWTPs and WIPs are not shown in Fig. 1 for the sake of simplicity, although they are considered in the calculations. The largest application area is coatings, paints, and pigments, followed by electronics and optics, cosmetics, and energy and the environment. The majority (63–91 %) of ENMs will likely be disposed in landfills, with a smaller but significant fraction going to soils (8–28 %), water (0.4–7 %), and the atmosphere (0.2–1.5 %). Silica

**Fig. 1** Global flow of ENMs in 2010 (metric tons/year) from manufacturing to applications and eventual disposal or release into the environment, considering the high range of production estimates and releases. Life cycle stages from *left* (production of ENMs) to *right* (final disposal or release)



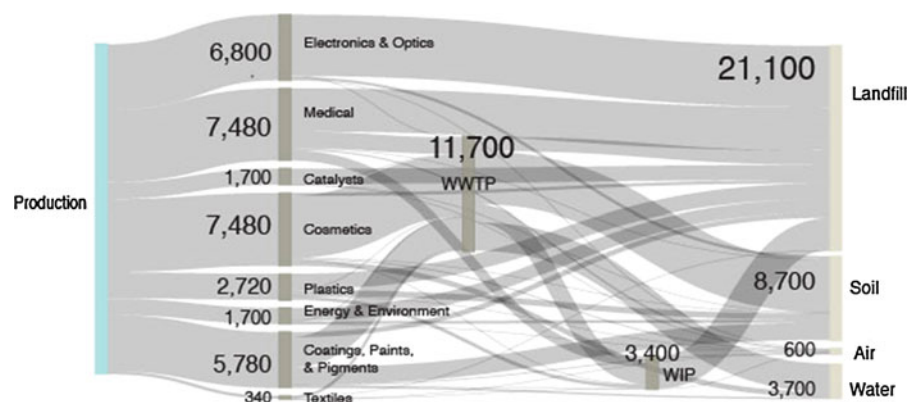
(SiO<sub>2</sub>), titania (TiO<sub>2</sub>), iron and zinc oxides, and alumina (Al<sub>2</sub>O<sub>3</sub>) dominate the ENM material flows at present. Of these, titania has the largest releases into soil, water, and air, followed by iron and zinc oxides. Titania is likely to remain as a particle, due to its very low solubility, although most likely not a nanoscale particle in the long term, unless it has been stabilized significantly. Silica, also rather insoluble, dominates disposal to landfills. Iron and zinc oxide ENMs are likely to dissolve, releasing Fe<sup>2+</sup>, Fe<sup>3+</sup> (Adeyeye 2013), and Zn<sup>2+</sup> (Zhou and Keller 2010; Fairbairn et al. 2011), but eventually re-precipitating, most likely as larger particles (Lowry et al. 2012b). For most of these materials, emissions to soils (22,000–80,400 metric tons/year) are based on the assumption that a fraction of the sludges from WWTP will be applied on soils, with a much smaller fraction of direct ENM application to soils (e.g., agricultural pesticides or fertilizers, environmental remediation). Emissions to water bodies, either from direct use (e.g., cosmetics) or indirectly through WWTP effluent, are estimated to be on the order of 1,100–29,200 metric tons/year worldwide. Many of these ENMs will likely settle out as larger aggregates in natural water bodies within hours to days (Keller et al. 2010), to be stored in sediments. Ongoing research attempts to determine the bioavailability of ENMs under these conditions. ENM emissions to the atmosphere are generally estimated to be quite low (590–4,800 metric tons/year), based on the assumptions that (a) releases during the manufacturing process are small; (b) few ENMs are directly released into the atmosphere during use; and (c) during incineration most of the air emissions are controlled. Based on expected fate and transport calculations (Liu and Cohen 2012), most of the

atmospheric releases will eventually deposit on land and water surfaces, although of course the risk of inhalation while in the atmosphere needs to be considered.

Figure 2 presents an example of the global material flows for ZnO ENMs, considering the maximum production and emission rate estimates. Estimated global annual production as of 2010 is >30,000 metric tons, with most of ZnO going to applications in medicine, cosmetics, electronics and optics, and coatings, paints, and pigment products. Emissions during the manufacturing phase are estimated to be on the order 32–680 tons/year. For the use phase, the highest predicted emissions are from the use of ZnO ENMs in cosmetics, which are expected to pass through the WWTP and then onto biosolids and effluent. Overall estimated emissions are 90–578 tons/year to the atmosphere, 170–2,985 tons/year to receiving water bodies, and 3,100–9,283 tons/year to soils. We estimate that 21,153–28,171 tons/year are disposed in landfills, given the high solubility of ZnO nanoparticles, a significant fraction, if not all these ENMs are expected to end as Zn(II) compounds.

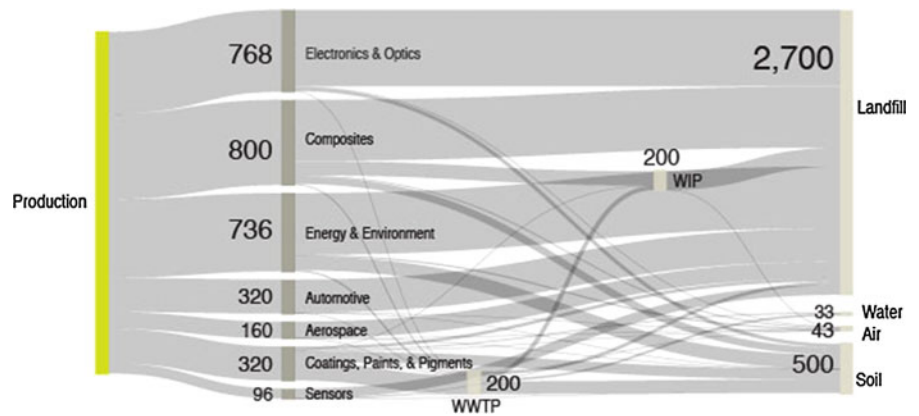
While the majority of ZnO ENMs will likely be disposed via WWTPs and WIPs, the expected pathway for CNTs through the world economy is significantly different (Fig. 3). Only a small fraction is currently expected to pass through WWTPs (6.0 %) or WIPs (5.5 %), with most of the disposal going directly to landfills, due to the nature of the applications. Disposal in landfills represents 83–98 % of the material flow, with only 2.2–14.8 % going to soil, 0.1–1.4 % to air, and 0.01–0.6 % to water. Subsequent transport from air or soils may lead to additional CNTs in water. Similar material flows were prepared for eight other ENMs (Figs. 4, 5, 6, 7, 8, 9, 10, 11).

**Fig. 2** Global material flows for ZnO ENMs (metric tons/year) in 2010, assuming maximum production and emissions rates

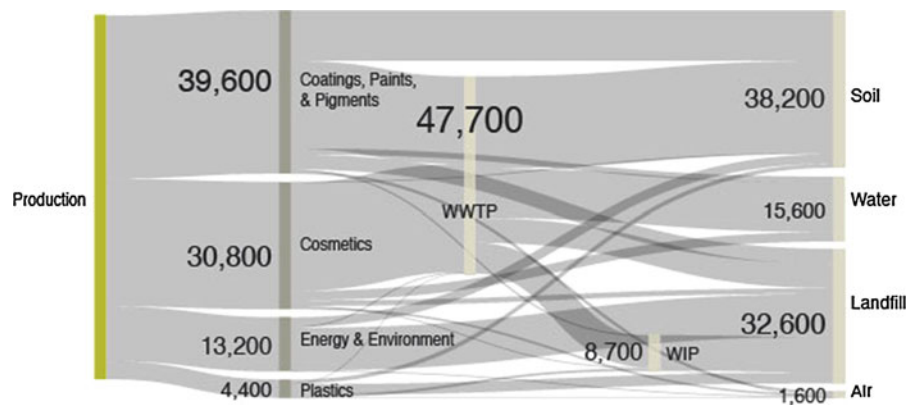




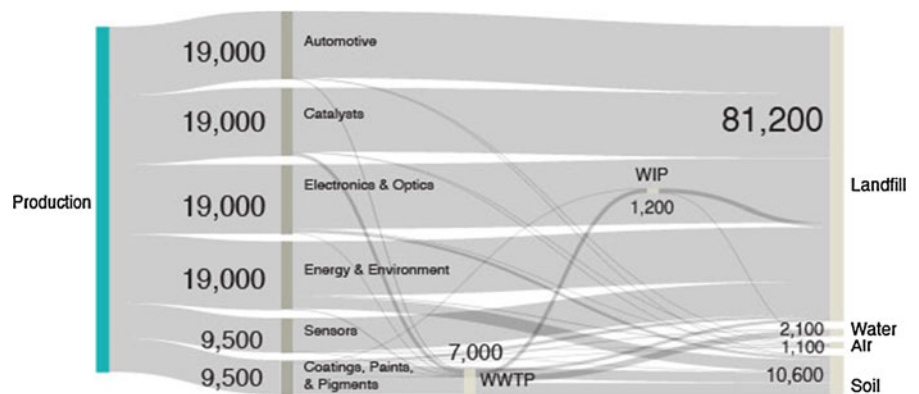
**Fig. 3** Global material flows for CNTs (metric tons/year) in 2010, assuming maximum production and emissions rates



**Fig. 4** Global material flows for TiO<sub>2</sub> (metric tons/year) in 2010, assuming maximum production and emissions rates



**Fig. 5** Global material flows for SiO<sub>2</sub> (metric tons/year) in 2010, assuming maximum production and emissions rates

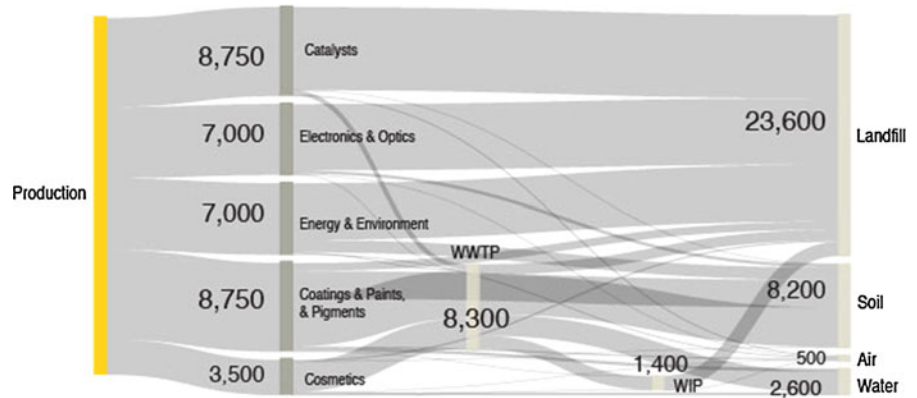


**Analysis of key ENM applications with potentially important environmental implications**

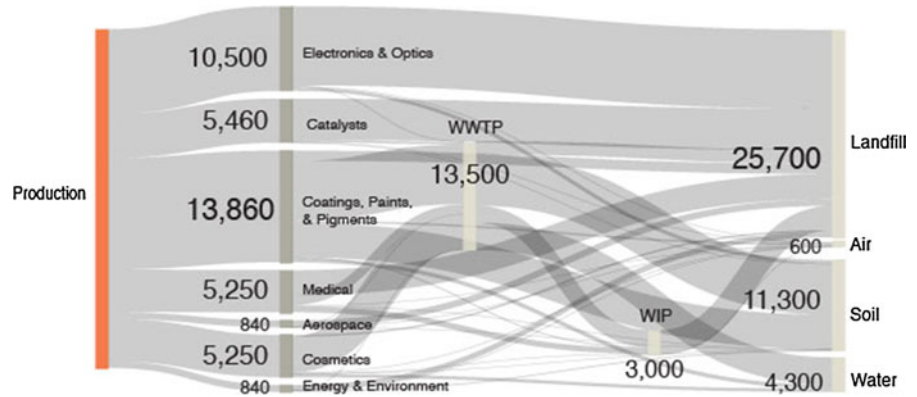
The estimated releases into various environmental compartments are useful not only for quantifying mass loadings but also for identifying applications with potentially important environmental implications (Fig. 12). The low estimate considers the lower

production estimate as of 2010 and the low release assumptions during manufacturing, use, and disposal. Under this scenario, most of the ENMs end up in landfills, at a higher amount than under the high production and emission scenario. Currently, coatings, paints, and pigments are not only the most important application in terms of overall use, but have the highest likelihood of being released into soil and water. While

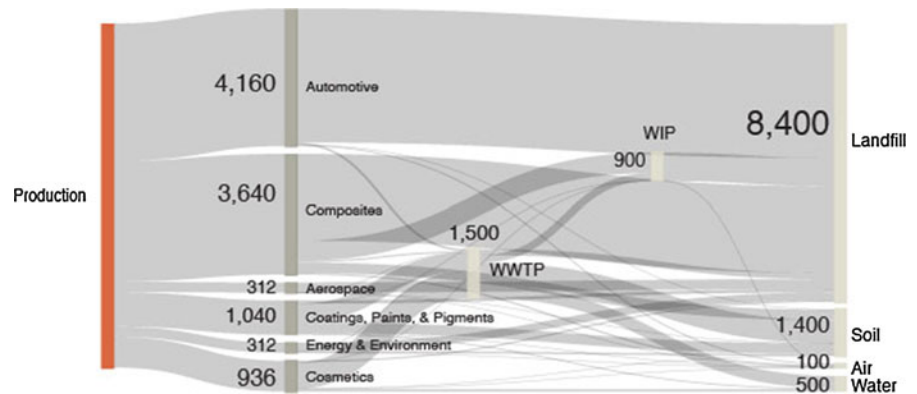
**Fig. 6** Global material flows for Al<sub>2</sub>O<sub>3</sub> (metric tons/year) in 2010, assuming maximum production and emissions rates



**Fig. 7** Global material flows for Fe and Fe oxides (metric tons/year) in 2010, assuming maximum production and emissions rates



**Fig. 8** Global material flows for nanoclays (metric tons/year) in 2010, assuming maximum production and emissions rates

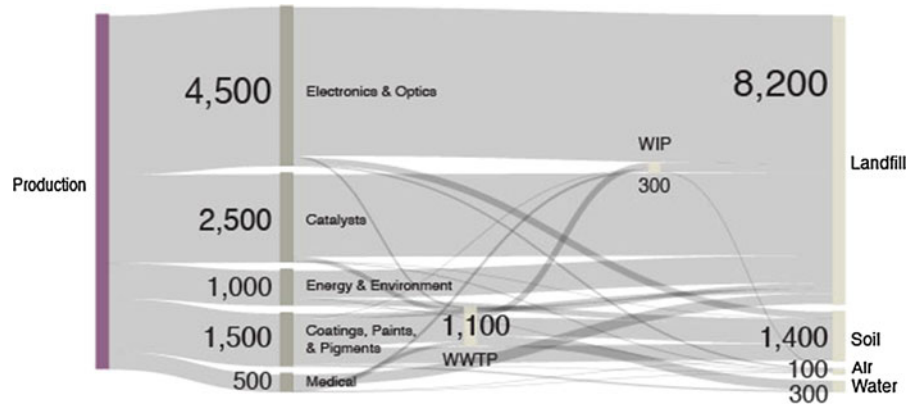


electronics and optics represent a major application, these ENM-enabled products are more likely to be disposed in landfills, with a much lower release into soil and water. ENMs in cosmetics also are released into soils and waters in significant amounts, mostly via WWTP effluent to surface waters and sludge applied to land. The balance of the sludge is directed to landfills. The balance of the applications contributes a small fraction to the potential release of ENMs into soils, water, and air.

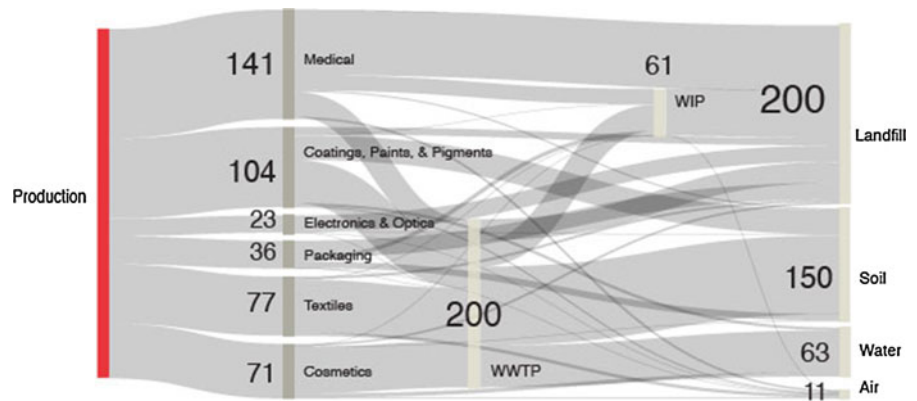
Coatings, paints, and pigments

ENMs are utilized in coatings, paints, and pigments for their wear-resistant, transparent, and UV-resistant properties, among others. Specific examples include the use of Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> ENMs to protect steel (Dhoke et al. 2009) and other materials (Baglioni and Giorgi 2006; Kaiser et al. 2013). ENMs may be released into the environment from weathering,

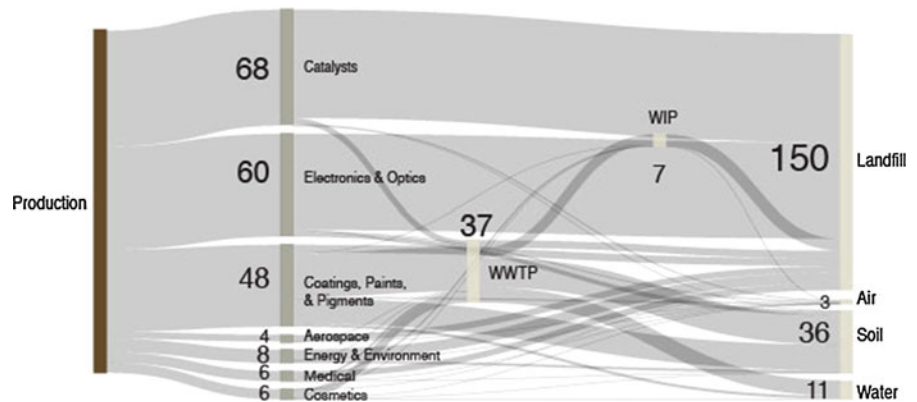
**Fig. 9** Global material flows for CeO<sub>2</sub> (metric tons/year) in 2010, assuming maximum production and emissions rates



**Fig. 10** Global material flows for Ag (metric tons/year) in 2010, assuming maximum production and emissions rates



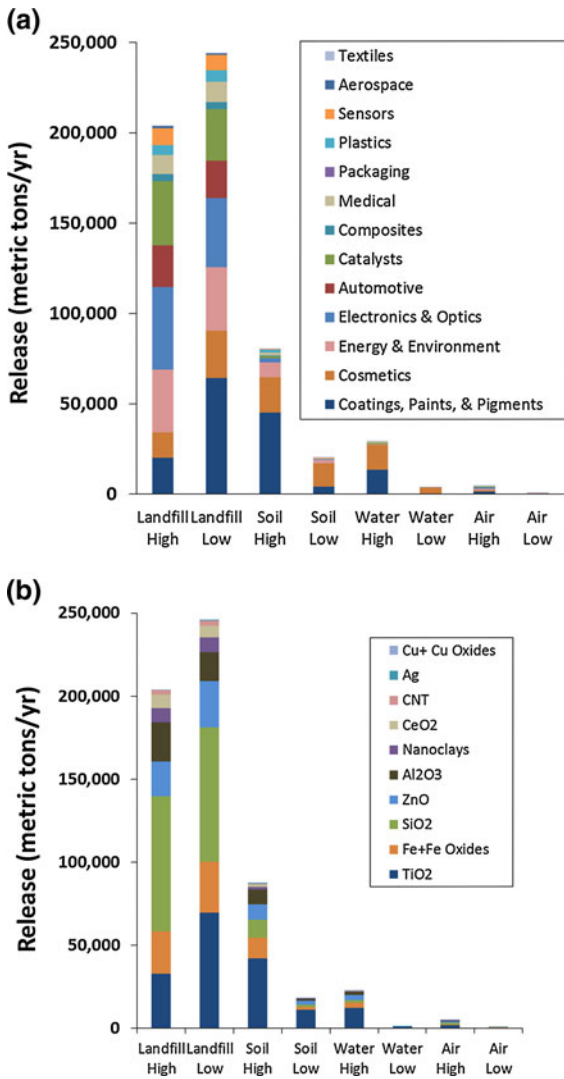
**Fig. 11** Global material flows for Cu and Cu oxides (metric tons/year) in 2010, assuming maximum production and emissions rates



specifically into surface waters and soil. On aggregate, 64,000–81,000 metric tons of ENMs were used in these applications in 2010, with over 34,000 tons/year for TiO<sub>2</sub> and nearly 10,000 tons/year for SiO<sub>2</sub> used in coatings, paints, and pigments. Using modeled estimates from Mueller and Nowack (2008) for releases during the use and disposal phases as well as estimates from other studies for releases during use (Hsu and

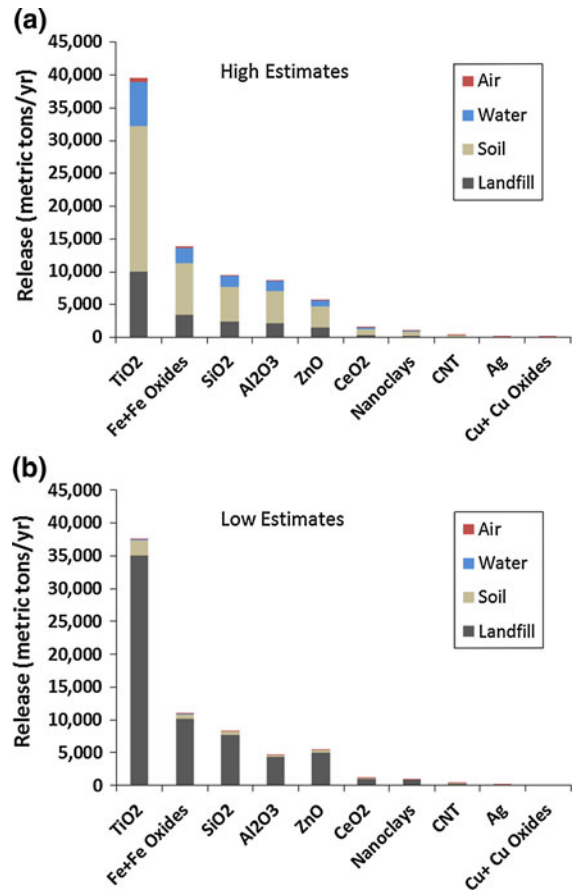
Chen 2007; Hassellöv and Kaegi 2009), we determined high and low estimates for the release of nanoparticles from coatings, paints, and pigments (Table 2).

For the lower production estimate and with low release assumptions, disposal in landfill dominates the emissions (nearly 93%), with the balance mostly ending on soils (nearly 7%), and only 0.2% emitted



**Fig. 12** Estimated global releases of ENMs into different compartments, by application, based on 2010 production estimates by **a** application; and **b** ENM type. *Low* indicates low production and release assumption estimates, and *high* refers to high production and release assumptions

to water bodies and 0.1 % to the atmosphere. In contrast, when the high release assumptions and production rates are considered (Fig. 13), a significant fraction may end up in soils (62 %), with a smaller fraction going to landfills (24 %). A significant fraction (12 %) of the initial release from washing and weathering of the coatings and paints is transported by storm water to soils and surface waters. Depending on local storm water management, varying amounts of storm flow will be directed to WWTPs, where 75–97 % of the nanoparticles will be removed

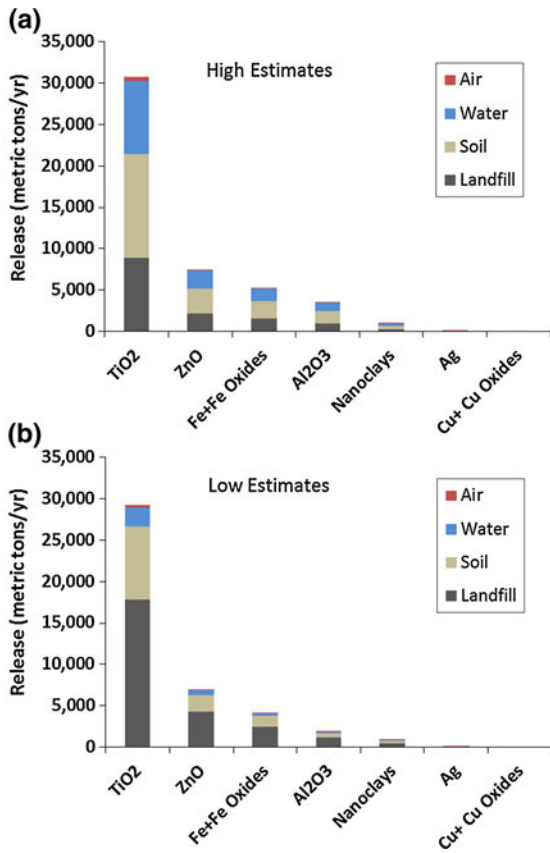


**Fig. 13** Final disposal in landfills and emissions to soil, water, and air for ENMs used in coatings, paints, and pigments, considering **a** maximum global production and high release estimates, and **b** minimum global production and low release estimates

from the water and transferred to sludge. Air emissions from this application are expected to be low (1.8 %). There is over an order of magnitude difference between the high and low estimates, indicating a need for further research to reduce the uncertainties. These results are qualitatively consistent with previous findings that this application of ENMs is likely to contribute significantly to environmental releases (Kaegi et al. 2008; Mueller and Nowack 2008).

### Cosmetics

Use of nanoparticles in cosmetic products is of particular interest as this application has a high potential for human exposure, as well as the tendency for these materials to be designed for short use periods



**Fig. 14** Final disposal in landfills and emissions to soil, water, and air for ENMs used in cosmetics, assuming **a** maximum global production and high release estimates, and **b** minimum global production and low release estimates

and daily disposal to WWTPs through washing (Mu and Sprando 2010; Henkler et al. 2012). Nearly 16 % of ENMs are used in cosmetic applications worldwide. The main nanoparticles used in this application are TiO<sub>2</sub>, ZnO, iron oxides, and Al<sub>2</sub>O<sub>3</sub> (Fig. 14). Mueller and Nowack (2008) estimated that 95 % of nanoparticles used in cosmetics are released during use. We used this as the high estimate and 75 % release for the low-end estimate. Although most of the initial emissions are to water, the ENMs will end up in soils (33–44 %), with 2.2–23 % potentially discharged to water bodies after WWTP treatment and only 0.8–1.9 % emitted to air.

### Textiles

While estimates of ENM usage in textiles for 2010 are only between 380 and 420 tons, it is a rapidly

emerging application. Promoted properties that ENMs bring to textiles include self-sterilization, UV protection, wear resistance, fire resistance, and increased mechanical strength (Qian and Hinestroza 2004; Wong et al. 2006). One reason for added scrutiny is that ENMs in this application come in close proximity to human contact (e.g., hospital pillows and blankets, socks, sports attire) and have the potential for high exposure to the environment. According to the market study, only Ag and ZnO ENMs are used significantly in textiles at present. The majority of these ENMs are expected to end in landfills (30–95 %), soils (4–42 %), and surface waters (0.3–22 %). Studies estimating the release from textiles during washing report results ranging from nearly zero release to nearly 100 % of the contained nanoparticles to be released into wash water (Geranio et al. 2009; Kiser et al. 2009; Windler et al. 2012). Due to this wide range, our release estimates are highly uncertain and deserve further research. In this study, the low estimate for release during use is based on the assumption by Mueller and Nowack (2008) that 5 % is released into both air and WWTP, for a total of 10 % release. This estimate is specific to nanoparticles, as it is recognized that there may be an additional release in ionic form. The high-end estimate for release during use is intended to recognize the potential for high release rates, while also making a reasonable estimate considering that a significant portion of the released materials will no longer be in nanoparticle form (Benn and Westerhoff 2008; Geranio et al. 2009). The distribution between WIP (25 %) and landfill (75 %) was based on a detailed study of textile life cycle (Domina and Koch 1997).

### Conclusions

Our study estimates global life cycle ENM emissions as of 2010, covering the ten most significant types of ENMs in over a dozen main applications. Our results provide a first global assessment of the magnitude of ENM releases into the environment. Currently, around 63–91 % of ENMs are eventually disposed in landfills. While a better understanding of their potential to be transported in leachate is needed, for well-constructed, secure landfills, this may be the final repository for most ENMs. Emissions to soils represent up to about a quarter of the material flows, mostly from disposal of

biosolids to land, highlighting the importance of understanding the implications for agriculture. Releases into water are only around 0.4–7 % of total global production. Air emissions represent the smallest flow to the environment, with an estimate of less than 1.5 % of production even considering the highest production and release estimates. TiO<sub>2</sub> is by far the most significant ENM in terms of exposure, based on estimated releases and use in the dominant applications. As exemplified by ZnO and CNTs, wide differences in emission patterns should be expected for different ENMs, given their applications. Additional differences will likely emerge as a better understanding of their release mechanisms becomes available.

Although it is difficult to directly compare our results to previous estimates of ENM releases quantitatively, due to differences in scope (global vs. regional or single country, types of ENMs), our results are qualitatively in line with those found for Switzerland or the EU (Gottschalk et al. 2009; Gottschalk and Nowack 2011), with differences for expected flow to WWTP, WIP, and landfill, which reflect different waste management practices around the world.

Our results show that despite the diversity of ENMs in production and applications, a few key applications dominate ENM releases into the environment: Coatings, paints and pigments, and cosmetics combined mediate around 42 % of total global ENM flows and likely currently contribute 82–87 % of total ENM emissions to soil and 89–97 % to water. This has policy implications in considering application-oriented regulatory measures—as opposed to facility-oriented, to manage ENM emissions effectively.

Our results also show that WWTPs are an important intermediate pathway for some ENMs to soil and water. From 17 to 34 % of ENMs likely pass through WWTP, which leads to 3–25 % of releases into water bodies via treated effluent and 44–47 % of emissions to soils via biosolids. Therefore, understanding the behavior and transformation in WWTPs is essential for accurate estimation of those ENM emissions.

The information generated in this assessment can be used to prioritize future research. For instance, the results of our study can help prioritize measurement experiments on ENM emissions from key applications, toxicity studies of ENM products with higher likelihood of exposure, and possible future regulations based on the applications and pathways (e.g.,

wastewater and sludges) that are most likely to impact human health and the environment. The emission estimates can also be the input into fate and transport models to predict environmental concentrations of ENMs.

It was our attempt to bracket the uncertainties in our estimate using high and low estimates to the extent that current data availability and knowledge permit us to do so. Nevertheless, there are significant uncertainties in ENM production and emission rates. However, our estimates can be easily updated and refined as more and better information becomes available.

We used a time-integrated, mass balance approach in our estimates. Assessing ENM stock accumulation in the economy, transformation of ENMs during the use and waste management phases of life cycle and time-dependent estimate of ENM emissions are important aspects for future research.

**Acknowledgments** This material is based upon work supported by the National Science Foundation (NSF) and the US Environmental Protection Agency (USEPA) under Cooperative Agreement Number DBI-0830117. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of NSF or USEPA. This work has not been subjected to USEPA review, and no official endorsement should be inferred. The authors thank Michael Bostok for the d3.js script used to produce the Sankey diagrams.

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