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REVIEW

Life-cycle assessment of engineered nanomaterials: a literature review of assessment status

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Abstract The potential environmental impacts of engineered nanomaterials (ENMs), and their engineered nanoparticles (ENPs), have, in recent years, been a cause of concern. Life-cycle assessment (LCA) is a highly qualified tool to assess products and systems and has an increasing extent been applied to ENMs. However, still only 29 case studies on LCA of ENMs have been published in journals and this article investigates these studies. Generally, data on production of ENMs as well as the coverage of the life cycle are limited. In particular, within use and disposal stages data are scarce due to many unknowns regarding the potential release and fate of ENMs/ENPs to and in the environment. This study investigates the sensitivity of case studies with respect to ecotoxicity impacts through a quantification of the potential ecotoxicity impacts to algae, daphnia and fish as a result of direct release of Ag and TiO_2 ENPs (mainly <200 nm in nominal diameter size) from various ENM products to the freshwater compartment. It was found that Ag and TiO_2 release, from 1 g Ag or TiO_2 ENM product, poses up to ca. 3.5 orders of magnitude higher ecotoxicity impact than the production of 1 g polymer (PP, PE and PET average) or 1 Wh of grid mix electricity from Scandinavia. ENMs from Ag had higher ecotoxic impact than those from TiO_2 and there was a linear regression between Ag ENM content in the considered products and the potential ecotoxicity impacts to the freshwater species, according to release of total Ag during use (mainly washing).

Keywords Life-cycle assessment · Engineered nanomaterials · Impact assessment · Sustainability · Environmental effects

Introduction

Nanometer, the metric unit of length of a billion of a metre (10^{-9}), has created a terminology for technologies that operate at the nanometer scale ranging from 0.1 to 1,000 nm, or 1 μm. Nanomaterials, also referred to as nanoobjects, by ISO (2008) and SCENIHR (2007), have been defined as materials with at least one dimension of 1–100 nm. According to ISO (2008), a nanoparticle needs to have all three dimensions within the 1–100 nm range. Nanomaterials and nanoparticles occur naturally in our surroundings as e.g. soil or salt particles and humans can be exposed to these on an everyday basis through air or food. Naturally occurring, or unintentionally produced,

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nanomaterials are known in general to cause little harm to humans (Buzea et al. 2007). Nanoparticles are also produced by activities as e.g. combustion. These anthropogenic particles have been an area of human health research for many years and are known to cause harm even though exact mechanisms are still being explored (Oberdörster et al. 2007). Engineered nanoparticles' (ENPs) behaviour and potential impact to environment and humans are on the other hand to a larger extent unknown (Buzea et al. 2007; IITB 2012).

Through the life cycle of engineered nanomaterials (ENMs), i.e. from production of ENMs through their use and final disposal, their environmental and health impacts vary. A main cause of variation is the environmental exposure as a result of release of ENPs. This in turn depends on how ENMs are processed from raw materials and incorporated into a nanoproduct (commercial product containing ENMs) and how this product is affected by the surroundings in its lifetime (from use to disposal stage). The exposure is also a function of how the ENPs behave in the environment, prior to having an effect on biota and humans. To include the whole life cycle, for comparison and assessment of products and systems, life cycle thinking has been introduced (Som et al. 2010; Klöpffer et al. 2007), and one holistic albeit rigorous tool for that is Life-Cycle Assessment (LCA) (Fig. 1).

LCA of nanoproducts addresses all environmental impacts, but even though different studies have measured release from different products (e.g. Geranio et al. 2009; Künninger et al. 2010; Lorenz et al. 2012; Benn et al. 2010), there is currently no method available to quantitatively assess the environmental impacts deriving from ENM release. In chemicals risk assessment (RA), it is possible to identify the (eco) toxicological risk of a substance (provided the data are available) through calculating, measuring or modelling the exposure concentration and relates this to the effect concentrations observed in laboratory studies (Olsen et al. 2001; Grieger et al. 2012). LCA on the other hand is a relative impact assessment method (ISO 2006). The object of the assessment is a defined functional unit (quantified performance of a product system) making comparison between product systems possible but at the same time making it impossible to make risk estimates. Furthermore, a wide range of potential environmental impacts are taken into account. In theory, LCA can be applied to nanoproducts, but in practice the potential impacts of ENPs cannot be adequately assessed. A main challenge is that the physico-chemical characteristics of ENPs are not adequately understood in relation to environmental behaviour and effects, a challenge that is also faced by RA (Bauer et al. 2008; Grieger et al. 2012; Flemström et al. 2004).

Currently, a few reviews have been published on LCA of ENM products (Gavankar et al. 2012; Hischier and Walser 2012; Meyer et al. 2009; Upadhyayula et al. 2012). They addressed some of the observed barriers that need to be overcome in order to strengthen LCAs of ENMs (in particular, adequate and comprehensive life-cycle inventory data, especially for manufacturing and use, and the lack of data for nanospecific fate, transport and toxicity effects). Additionally, Grieger et al. (2012) reviewed the complementary use of RA and LCA, concluding that

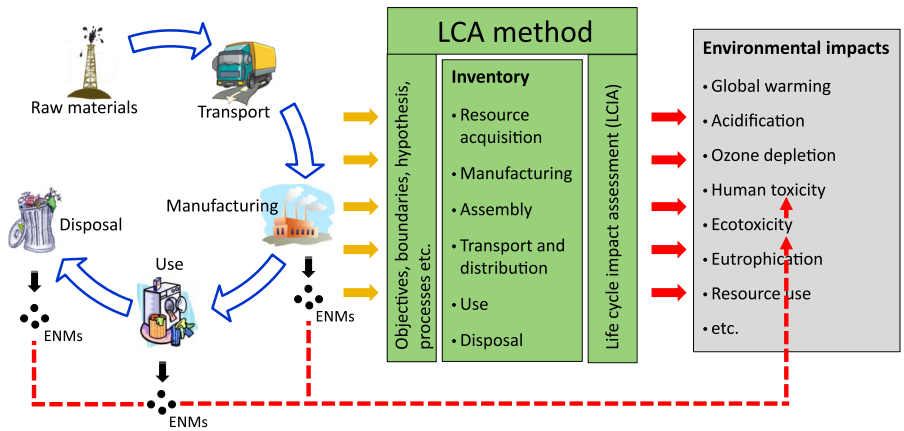

Fig. 1 LCA framework and ENM release related impacts, based on USEtox™ (2012)

the approach is not mature for practical application but holds the potential to improve environmental assessment of ENMs. The reviews concluded that many studies performed until now are quite limited in their Life-Cycle Impact Assessment (LCIA). In comparison to the already performed reviews, this paper emphasises gaps of LCAs of ENM products with a focus on challenges for LCIA, mainly related to the potential toxicological impacts resulting from release of ENPs to freshwater.

Objectives of this paper

LCA of ENM products is still a rather new and developing application area for LCA and in this article we will review published scientific articles on LCAs of ENM products and systems (defined with 'nano' term as a prerequisite, not size). A particular focus is to identify and explain the challenges for LCIA, mainly those related to the ecotoxicological impact categories and to ENP behaviour in the environment. The potential release of ENPs and ENMs in the life cycle is also addressed, since this is considered central in the discussions of impacts from ENMs (e.g. Bauer et al. 2008). The main outcome of the review is an overview of the possibilities and limitations of LCA within this field, as well as of the challenges related to modelling of ENP behaviour in the freshwater environment. Finally, a sensitivity analysis is performed on Ag and TiO_2 ENM products in order to estimate the potential ecotoxicological impacts of ENPs on freshwater species (algae, daphnia and fish)—an aspect that is missing in current approaches of LCIA of ENMs.

Life-cycle impact assessment and life-cycle assessment

LCA is a holistic, ISO 14040 series standardised, tool able to assess the potential environmental impacts of a product or a system throughout their entire life cycle (ISO 2006). Impact categories assessed include local (e.g. ecotoxicity), regional (e.g. acidification) and global (e.g. climate change, resource use) impacts. In LCIA, the impact potentials through the entire product life cycle must be considered in order to assess the overall environmental impact potentials (Wenzel et al. 1997). For each specific environmental impact category, contributing emissions are assessed using an environmental model to estimate the impact contribution compared to that of a reference amount of the substance. This relation is expressed as an equivalence factor, in LCIA terminology called a characterisation factor. The well-known example is CO_2-equivalents for climate change impacts. Most LCIA methods are based on the following principles for calculation of environmental impacts (Wenzel et al. 1997; Guinée et al. 2002; Jolliet et al. 2003; European Commission Joint Research Centre (ECJRC) 2010):

$$\mathrm{EP}(j) = \sum \mathrm{EP}(j)_i = \sum \left(\mathrm{Q}_i \cdot \mathrm{CF}(j)_i\right) \quad (1)$$

The emission of a substance (i) has the magnitude of Qi and the characterisation factor for that substance to the environmental impact category (j) is CF(j)i. From this, the emissions potential contribution to the environmental impact is calculated ($EP_{(j)i}$) and all emissions contributing to the impact category are summed ($EP_{(j)}$) (Wenzel et al. 1997).

A main challenge in LCA of ENMs is how to derive the characterisation factors (CF) for ecotoxicological impacts of ENMs. The widely recognised USEtox™ model for deriving CFs on (eco-)toxicity is based on the general impact assessment framework (Eq. 2) (Rosenbaum et al. 2008). It considers a fate factor (FF) which is equal to the compartment specific residence time (in days) of a chemical. The exposure factor (XF) considered is in days^{-1} for human toxicity and unitless for ecotoxicity by indicating the dissolved and bioavailable fraction that biota is exposed to. Effects considered are expressed in the effect factor (EF) in cases/kg intake (EF) or in PAF m^3/kg (PAF = potentially affected fraction). In ecotoxicology, XF can be neglected if the exposure is expected to be equal to the environmental concentration and the characterisation factor will then be FF × EF. The intake fraction (iF) for human toxicity expresses the fraction of released substance that will be taken in by humans. The derived characterisation factor (CF) is expressed in cases/kg emitted or in PAF m^3/kg emitted (Rosenbaum et al. 2008):

$$\mathrm{CF} = \mathrm{EF} \cdot \mathrm{XF} \cdot \mathrm{FF} = \mathrm{EF} \cdot \mathrm{iF} \quad (2)$$

The USEtox™ framework and calculation procedures could in theory be applied to derive CFs for ENMs. However, the USEtox™ framework is developed for organic chemicals and takes into account partitioning based primarily on octanol–water partitioning coefficient as well as degradation. So, for

example, when applied to metals, the model fails to account for multiple and interconverting metal species and the sensitivity of metal-species distribution to ambient chemistry (Gandhi et al. 2010). Such issues will be relevant for ENMs as well and in order to calculate CFs for ENMs, the USEtox™ model needs to be adapted to model the fate of ENMs and take the aggregation and dissolution of ENPs into consideration. However, ENM behaviour in the environment is still not well understood, and data are not easily available (Lowry et al. 2012; Quik et al. 2011).

LCA has, still only to a limited extent, been applied to products containing ENMs and in the following the few published studies are reviewed. Almost none of the studies have directly considered fate of released ENMs due to the challenges described in "Difficulties in toxicological impact characterisation of ENPs" Section, where focus is on the LCIA challenges related to the physiochemical characteristics of ENMs and their behaviour in the environment.

Life-cycle assessment case studies of ENMs

In order to evaluate the current status of LCA application on ENMs and identify needs for improvement, a summary of published scientific articles containing LCA case studies performed on various products, components and systems containing ENMs are shown in Tables 1 and 2. The found case studies are divided into two tables addressing metal and their compounds, and carbon and composite materials case studies, respectively. The following interpretation addresses the strengths and weaknesses of these studies as well as the challenges still to be solved or even fully understood.

The identification of studies in Tables 1 and 2 was based on whether LCA was used. Some studies include life cycle thinking but do not directly claim to perform LCA. These have been included since they address one or several life-cycle stages, most often with a focus on energy consumption of ENM manufacturing.

Cradle-to-gate consideration

The case studies on metal, carbon and composite ENM products usually consider a cradle-to-gate LCA and focus on impacts caused by the energy-consuming manufacturing processes. The use and disposal stages, as well as the potential toxicity associated with the release of ENPs during the life cycle of the product, are most often neglected. Most studies in Tables 1 and 2 consider toxicity impacts, but only the case study from Walser et al. (2011) considers toxicity impacts from release of ENMs.

Some studies (Lloyd and Lave (2003), Lloyd et al. (2005), Babaizadeh and Hassan (2012), Manda et al. (2012), Roes et al. (2007), Steinfeldt et al. (2010a) and Walser et al. (2011)) consider the use and disposal stage, but the coverage of these stages is rather incomplete (excl. Walser et al. 2011). For example, Lloyd and Lave (2003) consider clay-polypropylene ENMs in car body panels and there they consider resource savings (fuel consumption) during the operation of a vehicle due to the lower weight of ENM, but no other supplementary materials are considered and thereby this study neglects the potential release of other agents in the use stage (Som et al. 2010). The disposal of ENMs is in general not properly dealt with due to lack of knowledge of whether end-of-life products would be landfilled, incinerated or recycled.

Functional unit

In order to perform a comparative LCA, the functional unit is central to ensure a comparable functionality of the two products. Studies as Joshi (2008), Kushnir and Sandén (2008), Grubb and Bakshi (2010) and others use a simplified functional unit relating just to the weight of the material, e.g. 1 kg of a certain material with ENMs. However, as also stated by Hischier and Walser (2012), a functional unit based on weight does not make sense in studies comparing ENMs with conventional materials, as functionality is not proportional with weight. If the use of ENMs results in improved functionality, this should be considered in the functional unit. ENM production tends to have high resource and energy use in the production stage (Lloyd and Lave 2003; Roes et al. 2007; Khanna et al. 2008), but an improvement in functionality can potentially justify the use of more resources in this stage in order to produce a better product with less environmental impact in the use stage.

For example, Roes et al. (2007) includes elasticity (Young modulus) and strength (tensile strength) in the functional unit when comparing PP/layered silicate nanocomposites with conventional PP, since the

Table 1 Summary of published LCAs of metal ENMs (nanoproducts, -components and systems). Arranged according to latest published

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Babaizadeh and Hassan (2012)/ Uncoated and titanium dioxide (TiO_2) coated glass for residential windows	Cradle-to-grave	Multi-attribute Decision Analysis with Building for environmental and economic sustainability (BEES) and TRACI, Life cycle cost (LCC) is also included	Not specified	Manufacturing: Grubb and Bakshi (2010)	DOE (1994), Worrell et al. (2008), FIRE (2011), U.S. EPA (1990), DOE (2011), JIS (2004), Hassan (2010), Ballari et al. (2010), Yu et al. (2006), BLS (2011)	No	Global warming, acidification potential, eutrophication potential, fossil fuel depletion, indoor air quality, water intake, criteria air pollutants, human health, smog formation potential, ozone depletion potential, ecological toxicity, economic performance	The LCA combined with economic score for the construction materials shows that the most cost-efficient and environmentally friendly material for use in the building is the TiO_2 nano-coated glass, according to most of the impact categories. Likewise, in the weighted environmental and economic impacts, the TiO_2 nano-coated windows perform better when considering the raw material extraction to recycling boundary
Manda et al. (2012)/ Comparison of printing and writing paper production with new coatings (micro or nano TiO_2) and different pulp types with conventional approaches	Cradle-to-grave	CED (Cumulative energy demand), GHG emissions with IPCC 2007 GWP method, ReCiPe single score (mid- and end-point) method	Simapro 2007	Manufacturing: Grubb and Bakshi (2010), Cheng and Kelly (2010)	Ecoinvent (unspecified version), producers and suppliers (unspecified), Laurijssen et al. (2010), EC (2001), Reimann (2006)	No	Energy requirements, global warming potential (100 years), fossil depletion, metal depletion, natural land transformation, urban land occupation, agricultural land occupation, terrestrial ecotoxicity, climate change ecosystems, particulate matter formation, human toxicity, ozone depletion, climate change human health	Nanoparticle based paper shows the highest savings, e.g. in recovered paper fibre, and the lowest environmental impacts. The consequential LCA modelling approach yields the highest savings for nano and secondly for the micro-sized TiO_2 paper, compared to the conventional paper

Table 1 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Walser et al. (2011)/ Nanosilver t-shirts and conventional t-shirts (with/ without biocide triclosan)	Cradle-to-grave	USES-LCA	Not specified	Manufacturing: Kammler et al. (2001), Giessmann (2002), Hegemann et al. (2009), Körner and Hegemann (2008) Use: Geranio et al. (2009), Kumar and Münstedt (2005)	Ecoinvent (v2.2), Wernet et al. (2009), Rüdenauer et al. (2006), Saouter et al. (2002), Koehler and Wildbolz (2009), NLV (2010), Stamminger (2007), Capello et al. (2007), Seyler et al. (2005)	Yes	Climate footprint, seawater toxicity and freshwater toxicity	The annual climate footprint for T-shirt use in Switzerland is lower in the nano than in the non-nano scenarios (including triclosan treated polyester T-shirts). Because the additional burden of the nanosilver T-shirt production process is compensated with lower washing frequency. However, this is only the case for the flame spray pyrolysis technology production. Additional analyses indicate that a nanosilver T-shirt produced with commercial plasma polymerisation with silver co-sputtering process will never perform better than regular T-shirts in terms of climate footprint, even if consumers decrease the impact of the use phase by reducing washing temperature, or washing and tumbling frequencies
Meyer et al. (2010)/ Socks with and without Ag nanoparticles	Cradle-to-gate	TRACI 2.0 v-3.01	Simapro 7	Not specified	Hilliard (1998), U.S. Census Bureau (2009), Home Depot (2009), Demesne (2009), Durfee and Tomlinson (2001), Consumersearch (2009), Ecoinvent (unspecified version)	No	Global warming, acidification, carcinogenics, non-carcinogenics, respiratory effects, eutrophication, ozone depletion, ecotoxicity	Nano-Ag socks cause higher environmental impact across all categories, due to the energy required for production of nano-Ag. The liquid flame spray production method for producing the particles has the highest impact due to its use of hydrogen and oxygen as fuel gases

Table 1 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Moign et al. (2010)/ Three plasma spraying technologies for manufacturing of yttria-stabilised-zirconia nano-structured coating	Manufacturing	EDIP 2003	Simapro 7	Manufacturing: Yamagata et al. (2005), Riondel (1998)	Ecoinvent v2.0, Berghaus et al. (2007), Etchart-Salas (2007), Shan et al. (2010), Shan et al. (2007)	No	Global warming, ozone depletion, terrestrial eutrophication, aquatic eutrophication, ozone formation, human toxicity, ecotoxicity, hazardous waste, slags/ ashes, bulk waste, radioactive waste, resources, etc. (Presented as single score)	From the three used plasma spray technologies, each using powder, suspension or solution as feedstock to manufacture zirconia coating. It was showed that the solution precursor technology had the lowest environmental impacts. The thermal spray process (coating deposition on products) is the impact hotspot and 70–80 % of impacts derive from the use of electricity for spraying
Grubb and Bakshi (2010)/Nano TiO_2 production and conventional material production (steel, aluminium, polysilicon etc.)	Cradle-to-gate	Eco-indicator 99	Not specified	Manufacturing: Duyvesteyn et al. (2000a, 2000b, 2000c), Verhulst et al. (2002, 2003, 2006a, b)	Eidgenössische Technische Hochschule Zürich (ETH), European data sets and electricity for U.S. mix – not further specified	No	Carcinogenics, resp. organics, resp. inorganics, climate change, radiation, ozone layer, ecotoxicity, acidification/ eutrophication, land use, minerals, fossil fuels, energy use, resource use, non-renewable fuel use	The Altair hydrochloride process for producing TiO_2 nanoparticles has lower energy requirements than the traditional materials, such as steel and aluminium. The main impacts from producing TiO_2 comes from ilmenite mining and steam generation, while in terms of normalisation the fossil fuel category has the largest normalised damage impact due to steam generation and methane combustion

Table 1 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Sengül and Theis (2010)/Quantum dot photovoltaic (QDPV) module compared to other PV modules (silicon and thin film PV's) and energy sources	Cradle-to-gate	IMPACT 2002+	Simapro 7	Manufacturing: Sengül and Theis (2009) (mainly)	Ecoinvent database (from 2007), Roscheisen and Pichler (2006), Ha et al. (2005), Groner et al. (2004), Chilson (2008), Kandabarow (2006), Hung and Tang (1999), Cetinkunt (2007, 2008), (Fthenakis 2008, Fthenakis et al. 2008), Koroneos et al. (2006), Sheats (2004) and (Sheats et al. 2007), Kato et al. (2001), Gur et al. (2005), Scher (2006), Wickboldt (2008), Nanosolar Inc (2008), Moeller and Coe-Sullivan (2006)	No	Energy use, global warming, aquatic acidification potential, heavy metal emission, energy payback time	The study shows that CdSe QDPV modules would exhibit energy return and environmental emission levels better than the other types of PV solar systems (existing and novel PV types), with exception of heavy metals emissions. It should be noted that the study compares relatively mature technologies with emerging ones like CdSe QDPV, which in reality are apt to change. In general QDPV modules are performing better in all impact categories than carbon based energy sources, but have a longer energy payback time than wind- and hydropower and also a higher global warming potential
Bauer et al. (2008)/ Comparison of different physical vapour deposition (PVD) metal nano-coating processes (TiN, TiAlN, Ti + TiAlN)	Cradle-to-gate	CED (Cumulative energy demand) & CML 2001	Umberto	Manufacturing: Healy et al. (2006), Hwang et al. (2005)	Ecoinvent (unspecified version), (Umberto, GaBi and Team data libraries mentioned)	No	Energy demand, aquatic ecotoxicity, ionising radiation, photo-oxidant formation, ozone depletion, human toxicity, climate change, eutrophication, acidification, depletion of resources	The CED increases with the energy consumption of each PVD process and can be related like this: TiN < TiAlN < Ti + TiAlN. Metal production of titanium and aluminium accounts for more than 75 % of the total amount of the global warming potential - the consumption of metals is of essential importance. As Ti + TiAlN coating needs explicitly less metal than TiAlN coating, the potential environmental impacts are lower in the categories, climate change, resource depletion and acidification compared to TiAlN coating

Table 1 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Krishnan et al. (2008)/Life-cycle inventory of nano-scale semi-conductor manufacturing	Manufacturing	Economic input–output LCA (EIO-LCA), CED (Cumulative energy demand)	EIO model from Carnegie Mellon (2008)	Manufacturing: Details can be found in supporting information of Krishnan et al. (2008), though the origin of this data is not clear	Details can be found in supporting information of Krishnan et al. (2008), though the origin of this data is not clear	No	Energy use	Total primary energy requirement associated with upstream (chemicals and infrastructure) and semi-conductor manufacturing is at 14,100 MJ/wafer (high purity silicon substrate, which integrated circuits are manufactured on), with 7,100 MJ/wafer used for device fabrication and 2,900 MJ/wafer used for silicon wafer production. The results indicate that it may be important to further investigate the energy consumption of nanofabrication, as this study considers the specific processing energy within the manufacturing facility
Osterwalder et al. (2006)/Production of oxide nanoparticles by wet-chemistry or dry processes	Cradle-to-gate	CED (Cumulative energy demand)	–	Manufacturing: Defra (2004)	Ecoinvent v1.1, Althaus et al. (2003), FEA (2001), Defra (2004)	No	Energy requirements	Clear differences are seen in the energy requirements in the production of oxide nanoparticles (TiO_2, ZrO_2) using electricity-intensive plasma processes or chloride derived flame synthesis and liquid precipitation processes. The short process dry synthesis in general requires a larger amount of energy than the multi-step wet processes. The choice between these two processes is often determined by the product composition

Table 1 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Lloyd et al. (2005)/ Nanofabrication technique enabling precise control of nanoscale platinum-group metal particles in automotive catalysts	Cradle-to-grave	Economic input–output (and another unknown method)	GaBi 4	Manufacturing: Helmer (2002) (partially based on)	Amatayakul and Ramnäs (2001), Amatayakul (1999), Bartley et al. (1999), U.S. Environmental Protection Agency (1997), Jimenez et al. (1997), Andersen and Ballinger (1999), Stamatelos et al. (1998), Brisley et al. (1999), Hensel et al. (2000), Oh et al. (1993), Lucena et al. (2001), Chatterjee et al. (2001), Chafik et al. (1998), Braun et al. (2002), Burch et al. (1996), Santini (2003), Fiengo et al. (2004), Orbital Engine Corporation (1999), Lafyatis et al. (1999), Ekchian et al. (1999), Cornelius (2001), Extension production database (unspecified)	No	Energy use, resource use, global warming, hazardous waste, toxic release	The nanofabrication technique that enables precise control of the metal particles in catalysts should result in reduced loading levels of platinum-group metal. These reductions would then decrease energy consumption, improve environmental quality and contribute to sustainable resource usage. By application of this nanofabrication technique the environmental impacts would be lowered due to reduced mining and refining, as a function of less use of platinum in the manufacturing stage

Table 2 Summary of published LCAs of carbon and composite ENMs (nanoproducts, -components and systems). Arranged according to latest published

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Dahlben et al. (2013)/ Carbon nanotube semi-conductor switch production for cellular phone flash memory	Cradle-to-gate (incl. use stage improvements)	TRACI	Simapro 7	Manufacturing: Healy et al. (2008)	Ecoinvent v2.2	No	Global warming, acidification, carcinogenics, non-carcinogenics, respiratory effects, eutrophication, ozone depletion, ecotoxicity, smog formation, materials and energy inputs, speed of operation by applying CNT switch, projected mass of CNT for different disposal options	Two upstream processes are mainly responsible for the impacts, namely the electricity generation and the mining and refining of gold. Within these gold refining has the largest environmental burden, although only a small amount of gold is required per semi-conductor switch. The use of CNT switches would improve functionality, performance and cost-effectiveness of components such as memory storage devices. One of the concerns with LCA of CNTs is the shortage of consistent data for estimation of toxicity impacts of CNTs
Deorsola et al. (2012)/ Molybdenum sulphide (MoS_2) production for lubricant applications	Cradle-to-gate	CED (Cumulative energy demand) and carbon footprint (characterisation factors used from Frischknecht and Jungbluth, 2007)	Simapro 7	Manufacturing: Laboratory procedure, based on Aridi and Al-Daous (2009)	Ecoinvent from 2007	No	Energy requirements & global warming potential	55 % of the CED is due to electricity use (IT) during the synthesis of molybdenum sulphide nanopowder and to that 63 % of global warming impact during production is associated to the electricity consumption. The impacts during production are dominated by electricity use and the rest of the energy/ impacts are due to use of reactants as ammonium molybdate, citric acid and ammonium sulphide
LeCorre et al. (2012)/ Starch nanocrystals and organically modified clay	Manufacturing	TRACI 2 and Eco-Indicator 99(H)	Simapro 7	Manufacturing: Joshi (2008), Angellier et al. (2004), Franklin Associates (2007)	Ecobilan database 2006	No	Global warming potential, energy requirements, radiation, ozone layer depletion, ecotoxicity, acidification, ozone depletion, respiratory effects (organic and inorganic), land use, mineral use (non-renewable resources), fossil fuel use, smog generation	The production of starch nanocrystals requires less energy to produce than organically modified nanoclay. Nonetheless, starch nanocrytals have a higher impact in global warming and acidification impact categories. The other benefit of starch nanocrystals is that they have a better score in non-renewable energy and mineral depletion, as starch nanocrystals are renewable and biodegradable

Table 2 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Weil et al. (2012)/ Comparison of black carbon and activated carbon with single- and multi-wall carbon nanotubes (SWCNT & MWCNT through CVD or laser ablation) for supercapacitor based electric energy storage in hybrid and full electric cars	Cradle-to-gate	CED (Cumulative energy demand)	–	Manufacturing: Healy et al. (2008), Steinfeld and Arrnin (2008), Kushnir and Sandén (2008)	Noijuntura and Kittisupakorn (2009), Ecoinvent database (unspecified version), some own data	No	Energy requirements	The production of an SWNT electrode for supercapacitors is by far worst through a laser ablation compared to CVD, as the energy required is approximately twice as high. Far less energy is needed to produce MWCNT (through CVD, fluidized bed or floating catalyst approach) and activated and black carbon. Hereby it is underlined that the production of nano-scale carbon is highly energy demanding, even though the technology is promising in the use for supercapacitor based electric energy storage in vehicles
Wender et al. 2011/Quantification of energy use during manufacturing of SWCNT anode for a lithium-ion battery. Further the material inputs and outputs are also quantified for the entire battery	Cradle-to-gate	Energy demand (manufacturing)	–	Manufacturing: Ganter et al. (2010)	–	No	Energy requirements, material inputs/ outputs	To produce SWCNT anodes in Li ion batteries 45-130 MWh of electricity is needed per kWh of battery storage capacity. The material need, for the entire lithium-ion battery, is mainly represented by DI water, graphite CaCO3 and the material inputs become waste in almost equal quantities
Merugula et al. (2010)/ Glass fibre-reinforced plastics (GFRP) and the vapour-grown carbon nanofibres (CNF) to reinforce the interface of a glass fibre/epoxy matrix aimed for reinforcing large wind turbine blades	Cradle-to-grave (use and disposal assumed to be the same for the CNF based product as for the commonly used)	CED (Cumulative energy demand)	Simapro 7	Manufacturing: Unspecified, but likely Khanna et al. (2007, 2008)	Vestas (2006), Tremeac and Meunier (2008), Weinzettel et al. (2009), Vestas (2004) and not specified databases from Simapro 7	No	Energy requirements	Cradle-to-gate processing energy of the new CNF added material is 1.4-1.7 times greater than for the original GFRP material on a MJ/kg basis and implicit assumption of weight savings of 20 %. Effects on energetic return on investment (EROI) vary from insignificant to substantial based on the upstream production processes of CNF manufacturing and solvent handling. The choices of solvent application and CNF production are dominant in the potential for energy benefits; ergo it is not substantiated whether CNF introduction is advantageous mechanically and energetically

Table 2 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Steinfeldt et al. (2010a)/Carbon nanotube composite materials (carrier tray e.g. for toner cartridge) and films (for wind power plant)	Cradle-to-grave	CML & CED (Cumulative energy demand)	Umberto	Manufacturing: Steinfeldt et al. (2010b)	Ecoinvent (unspecified version)	No	Global warming potential and energy requirements	The first case study (CNT composite material as carrier tray) shows that an increase in the production efficiency plays an important role in the environmental impact. This also resembles in the second case study (CNT films for wind power applications), where a small increase in energy production efficiency of 0.25 % can result in an environmental impact improvement between 3,7–11 %
Dahlben and Isaacs (2009)/ Manufacturing of two carbon nanotubes (CNT) containing applications of a switch and a polymer mesh	Cradle-to-gate	Eco-Indicator 1999 (EI99)	Simapro 7	Manufacturing: Observation of laboratory practices at Center for High-rate Nanomanufacturing (CHN), Healy et al. (2008)	Not specified (databases from Simapro 7)	No	Climate change, acidification, eutrophication, land use, mineral depletion, ecotoxicity, ozone layer depletion, airborne inorganics, fossil fuels and carcinogens	Results indicate significant environmental impact contribution to airborne inorganics, climate change and fossil fuels due to materials used in cleansing processes and electricity consumption. In every impact category the CNT-polymer mesh manufacture has a slightly greater environmental burden than the CNT switch. This is most likely because the CNT-polymer mesh manufacture process produces fewer devices per 3-inch wafer than the CNT switch (72 CNT-polymer meshes versus 84 CNT switches)
Ganter et al. (2010)/ Manufacturing of single wall carbon nanotubes (SWNCT) through laser vaporisation	Manufacturing	CED (Cumulative energy demand)	Simapro (unspecified version), EIO model from Carnegie Mellon (2008)	Laboratory testing, unknown database through Simapro and EIO model from Carnegie Mellon (2008)	–	No	Energy requirements	To produce 1 kg SWCNTs at the laboratory scale, through laser vaporisation, 0.13-0.19 GWh energy is needed. From that, 0.114 GWh electrical energy is used. The energy consumed is mainly related to thermal and resistive losses of the laser and single zone use in the production. The total energy measured in the laboratory was in line with energy reported databases, such as data from the LCA software Simapro and economic input–output LCA (the exact database source was not specified). This is in contrast to many conclusions that laboratory production usually requires a lot more energy

Table 2 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Bauer et al. (2008)/FED screen (incl. carbon nanotubes)	Cradle-to-grave (excl. disposal)	CED (Cumulative energy demand) & CML 2001	Umberto	Manufacturing: Hwang et al. (2005), Dean et al. (2005), Kim et al. (2000), Gröning et al. (2003)	Empa communication and Ecoinvent v1.2	No	Energy demand in percent according to life stages and parts, ecotoxicity aquatic, ionising radiation, photo-oxidant formation, ozone depletion, human toxicity, climate change, eutrophication, acidification, depletion of resources	The use phase is dominating the overall impacts due to the electricity consumption during operation and these impacts account aprox. from 60-75 %. Secondly, 15-30 % of the impacts can be associated to the back glass coating and more precisely to the cathode and thus also to the CNT layer coating (applied by a CVD process)
Healy et al. (2008)/ Manufacturing of SWNT by arc, CVD HiPco processes	Manufacturing	Materials and energy use (cost model by Tanwani (2005))	Simapro (unspecified version)	Manufacturing: Healy (2006), Isaacs et al. (2006), Tanwani (2005)	Not specified (databases from Simapro)	No	Climate change, acidification, land use, mineral depletion, ecotoxicity, carcinogens, respiratory inorganics (airborne inorganics)	Nearly all emissions resulted from the generation of electricity for all three production methods. The CVD production process contributed with the largest quantity of other emissions (beside the ones from electricity), which were an order of magnitude larger than those of the arc and HiPco production processes. In order to perform a more representative LCA, e.g. SWNT worker exposure will also have to be considered
Joshi (2008)/Production of fibre-biopolymers and organically modified montmorillonite (OMMT) nanoclay-biopolymer composites	Manufacturing	Energy demand and GHG emission	–	Manufacturing: Franklin Associates (2006)	Ecobilan DEAM^{TM} LCA database and biopolymer data mainly from Dornburg et al. (2004)	No	Energy use and GHG emissions	The comparison of the nanoclay-biopolymer composites with the fibre-biopolymer composites shows that the environmental burdens from nanoclays are worse than those from the fibre-biopolymers. Except from the phosphate and nitrate emissions. Though in term of GHG emissions and the energy use the nanoclays are performing environmentally better than the glass fibres. These conclusions are based on a kg basis comparison, and the conclusion is that a product specific LCA is recommended in order to better describe the potential environmental impacts

Table 2 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Khanna et al. (2008)/ Production of polymer nanocomposites (PNC): Carbon nanofibre (CNF) and carbon nanofibre-glass fibre (CNF-GF) hybrid nanocomposites - also compared with steel	Cradle-to-gate	Not specified	Simapro 7	Manufacturing: Khanna et al. (2007), Finegan et al. (2003), Howe et al. (2006), van Hattum et al. (2006), Wu et al. (2007), Gordeyev et al. (2000), Gang (2007)	Not specified databases from Simapro 7, Corbiere-Nicollier et al. (2001)	No	Non-renewable based energy use	Cradle-to-gate PNC is 1.3-10 times more energy intensive than steel (per component produced), but in the case of automotive body panels the PNC life energy savings are realised through the automobile use phase. In cases where PNC is applied in a structural function the energy intensive production overshadows the environmental profile compared to conventional materials as steel. The release and exposure of nanoparticles is unknown, but is more likely to occur in the disposal stage and the knowledge of this impact is currently severely lacking
Kushnir and Sandén (2008)/Energy requirements of carbon nanoparticle production	Cradle-to-gate	CED (Cumulative energy demand)	–		Various literature data (specified in article)	No	Energy requirements for production methods, given in MJ/kg material produced	Carbon nano particle production is found to be potentially 2-100 times more energy intensive than aluminium production. The higher energy requirement in the cradle-to-gate system can be counterbalanced in the use phase, with savings in energy
Khanna et al. (2007)/ Carbon nanofibres production	Cradle-to-gate	CML (midpoint) & Eco-Indicator 99 (end-point)	Simapro 6	Manufacturing: Laboratory experiments and Tibbetts and Gorkiewicz (1993), Tibbetts et al. (1994), Fan et al. (2000)	Not specified (databases from Simapro)	No	Energy requirements and global warming potential, marine ecotoxicity potential, damage to human health, damage to ecosystems	The preliminary energy analysis indicates a 6-60 times higher life cycle energy requirement for carbon nanofibres than aluminium, steel and polypropylene. Carbon nanofibres may have a higher environmental burden than conventional materials, when compared on an equal mass basis. The human and ecotoxicological impacts are not fully accounted as this lacks knowledge and information on potential release and related impacts

Table 2 continued

Study/product	Assessment boundaries	LCA methodology	LCA software	LCI-data sources		Assessed impacts		Assessment conclusion
				ENM	Other	ENM release	Other	
Roes et al. (2007)/PP-layered silicate nanocomposite and conventional packaging film, agricultural film and automotive panels	Cradle-to-grave	CML 2000 & LCC (non-standardised approach)	Simapro 7	Manufacturing: Institute for Polymer Research (IPF, Dresden, Germany)	Ecoinvent (unspecified version), APME (2000), Energy Efficiency Office (1993), Qiu et al. (1998), Spielmann et al. (2004), Phylipsen et al. (2002), Doka (2003)	No	Non-renewable energy use (NREU), climate change (GWP 100 years), abiotic depletion, ozone layer depletion, photochemical oxidant formation, acidification and eutrophication	The production and incorporation of nanoclays may be compensated by the weight reduction provided to the end-product. The polypropylene nanocomposite use for agricultural film has clear environmental benefits, but on the other side the use of these nanocomposites in packaging film and automotive panels has no obvious environmental benefits In terms of economy the use of nanocomposites is advantageous if the nanoclay is not higher than € 5,000/ton. Depending on which material and energy prices are assumed the life cycle costs for agricultural film with a polypropylene nanocomposite can be reduced by 26–39 %. The economic advantage for using nanocomposites in automotive applications can be reduced with 3–6 %
Isaacs et al. (2006)/ Single wall carbon nanotubes (SWNT) produced through: arc ablation, chemical vapour deposition (CVD) and HiPco processes	Manufacturing	Environmental Priority Strategies (EPS) 2000 (through SimaPro)	Simapro (unspecified version)	Manufacturing: Not specified (inventory simply presented in article)	Not specified	No	Economy, life expectancy, severe morbidity, morbidity, severe nuisance, nuisance, crop and wood growth capacity, fish and meat production, soil acidification, prod. cap. irrigation and drinking water, depletion of reserves, species extinction	HiPco is economy-wise the cheapest method, approximately 4 times cheaper than the other two production methods. In terms of environmental potential attributes the data background is not proper, but the CDV has the greatest and arc ablation the least environmental burden. Based on the process comparison of resources used and emissions generated the electricity consumption during synthesis is the major contributor to the environmental burden
Lloyd & Lave (2003)/ Clay-polypropylene nanocomposite instead of steel or aluminium in light-duty vehicle body panels	Cradle-to-grave	Economic input–output LCA (EIO-LCA)	EIO model from Carnegie Mellon (2008)	Manufacturing and purchase data from 4 U.S. commodity sectors (unspecified source)	Production and purchase data from 4 U.S. commodity sectors (unspecified source)	No	Non-renewable based energy use, global warming, toxic release and costs	Despite being more expensive and having a higher environmental production impact than steel and aluminium, clay-polypropylene nanocomposite has a reducing energy and environmental impact advantage by improving car fuel-economy. This, along with the design possibilities of this material, could entice consumers to purchase lower weight vehicles and thereby reduce the environmental impact in the use-phase of the car

nanocomposite obtain the needed properties at a lower weight. In many LCAs of ENMs it may be difficult to identify the property that is the most important for defining the functional unit. To perform a fair LCA comparison, the functional properties must be central and it should be evaluated whether there is a need to scale the functional unit as Roes et al. (2007) does.

Inventory data

Production data of ENMs are scarce and the 29 studies in Tables 1 and 2 rely to a high extent on generic data e.g. those presented in Grubb and Bakshi (2010), Osterwalder et al. (2006), Tibbetts et al. (1994), Hwang et al. (2005) and Healy (2006), Healy et al. (2008). Primary process data of ENMs, coming from industry, are often not openly disclosed in this technology domain due to the relative novelty of the scientific field. This leads to a higher level of uncertainty as it needs to be covered by estimations and secondary and generic data, like the approach in Bauer et al. (2008), Joshi (2008), Merugula et al. (2010) and Isaacs et al. (2006). The lack of primary data limits the scope of the studies. Very often, the use and disposal stages of ENMs are unknown resulting in cradle-to-gate LCA study approaches, as seen in Tables 1 and 2.

Life-Cycle Inventory (LCI) date are crucial for making an LCA, and the relatively few case studies of ENMs underline the limited inventory data availability. In addition to the case studies presented in Tables 1 and 2, a number of scientific articles contribute to the slowly growing LCI. Some of the studies, contributing to the inventory, are Geranio et al. (2009), Köhler et al. (2008), Künniger et al. (2010), Som et al. (2011), Suppen et al. (2005), Durucan et al. (2006) and Gutowski et al. (2010). For example, Geranio et al. (2009) describes the release of Ag ENPs during textile washing, while Künninger et al. (2010) describes the nano-Ag release from facades due to weathering. There are several such studies that do not perform LCAs per se, but anyway present valuable LCI data that are needed when pursuing to do LCAs of ENMs and the products they are included in. These studies contribute in developing the needed inventory.

Impact hotspots

Currently, the conclusion, based on Tables 1 and 2, is that compared to their conventional counterpart products, the ENM products are more energy demanding and have an inferior cradle-to-gate environmental impact profile, e.g. in Khanna et al. (2007, 2008), where the production of nanofibres and polymer nanocomposites is more energy demanding than a steel product. On the other hand, Lloyd and Lave (2003) and Lloyd et al. (2005) show that the use stage for ENM products is more environmentally friendly than for their conventional counterpart products by assessing clay-polypropylene ENMs in car body panels and platinum-group metal particles in car catalysts, respectively. However, impacts such as potential ENP release are not considered. Gavankar et al. (2012) and Hischier and Wasler (2012) conclude that the assessment of nano-specific impacts must be developed but in order to do so the level of understanding of ENM behaviour, exposure and effects must be raised substantially.

Overall findings

On the basis of the 29 found studies, certain tendencies can be identified reflecting the current state of knowledge regarding LCA of ENMs:

- Usual cradle-to-gate or manufacturing system boundary consideration.
- Use and disposal life-cycle stages are poorly covered.
- Common use of generic life-cycle inventory (LCI) data and assumptions.
- Almost no consideration of release of ENMs (e.g. in the use or disposal stages) and the potential toxic impacts of these (fate, exposure and effect consideration). Walser et al. (2011) is an exception.
- Cradle-to-gate LCA comparison of counterpart products (with ENMs and without) shows that ENM products are more energy demanding and, therefore, have a worse cradle-to-gate environmental profile, e.g. in polymer nanocomposites vs. steel and socks with and without nano (Moign et al. (2010); Meyer et al. 2010).
- Cradle-to-grave LCA comparison of counterpart products (with ENMs and without) shows that the use phase is better for ENM products as usually an improved functionality is achieved, e.g. comparing clay-propylene nanocomposites with steel or aluminium in light-duty vehicles (Osterwalder et al. 2006).

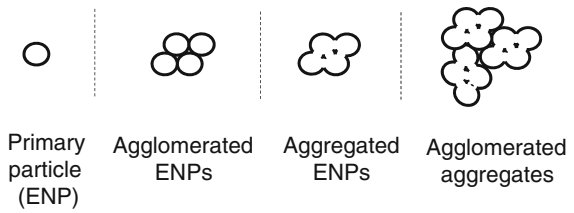

Fig. 2 Primary particle (ENP) appearance. Reproduced from Oberdörster et al. (2007)

Release of ENMs and assessment of their impacts contain a degree of complexity that makes them difficult to grasp scientifically and to include in a LCA approach. In the following, focus will be on the challenges related to assessment of the toxicological impacts, and afterwards a sensitivity analysis is performed on the potential toxicological impacts from release of ENMs/ENPs during use of products.

Difficulties in toxicological impact characterisation of ENPs

The potential toxicological impact of ENMs depends on the possible release of ENPs during an ENM products life, on their environmental fate and their potential effect when penetrating into living organisms. There is currently a lack in understanding of fate, exposure and effect of released ENPs in the environment—which is problematic especially in the freshwater compartment, as it is a common recipient (Quik et al. 2011; Lowry et al. 2012; Som et al. 2010). Some important parameters are particle appearance, transport, transformation and physico-chemical characteristics of ENPs. Current knowledge on these will be summarised below.

Particle appearance

Nanoparticles tend to agglomerate (coagulate), aggregate (fuse) or a combination thereof (e.g. like carbon black and TiO_2), see Fig. 2. The bonding and interaction happen in order to reduce the high-surface energy. The interaction between two particles, in liquid and air, can in general be described by forces of van der Waals attractions and electrostatic repulsions (Rupasinghe R-A-TP 2011). The particle appearance influences the toxicity of ENMs in water (Oberdörster et al. 2007).

Transformation

The transformation upon release into freshwater environment can be performed biotically (interaction with plants, water flea, fish etc.) or abiotically (interaction with water, sand, light, etc.) and can alter shape, size, surface chemistry and ultimately the fate of ENMs. Physico-chemical properties of ENMs define their differentiated behaviour, and the processes considered important for ENMs after release to water are (Vonk et al. 2009; Lowry et al. 2012):

- Dissolution
- Change in surface structure of ENMs/ENPs
- Aggregation/agglomeration
- Sedimentation

ENMs commonly tend to sorb to high-surface-area colloids and then aggregate/agglomerate and sediment (Klaine et al. 2008).

Transport

Transport is partially controlled by the aggregation/agglomeration and subsequent deposition/sedimentation of ENMs. Aggregation of ENMs in the environment will depend on these parameters (Lowry and Casman 2009; Lowry et al. 2012):

- Hydrophobicity
- Chemical bonding between nanoparticles
- Ionic strength
- Ionic composition

The ionic strength, which is higher in marine than freshwaters, and pH and the presence of divalent cations such as Ca^{2+} and Mg^{2+} will influence the rate and extent of aggregation/agglomeration (Lowry and Casman 2009). Brant et al. (2005) suggest that if C_{60} fullerenes are released into natural waters with an ionic strength higher than 0.001 M, these may form large aggregates/agglomerates that will sorb to other particles or media and eventually become immobilised.

Important physico-chemical characteristics of ENPs

Based on the developing understanding of ENM fate in e.g. freshwater it seems that the following characteristics are important to consider (Batley and McLaughlin 2010) (Klaine et al. 2008):

- Chemical composition
- Mass
- Particle number and concentration
- Surface area concentration
- Size distribution
- Specific surface area
- Surface charge/zeta potential
- Surface contamination and the nature of any shell and capping
- Solubility
- Crystal structure

The natural conditions of the environment are also important along with the common fate consideration of aggregation/agglomeration and dissolution. However, these and other co-related mechanisms especially in water are neither fully understood nor well represented with characterisation data (Farré et al. 2011). An overview is shown in Fig. 3.

Toxicity

The toxic effect of ENP depends on a number of parameters, e.g. size, dissolution, surface structure and aggregation/agglomeration. Variation in ENP size does show difference in toxicity, e.g. Zhu et al. (2009) stated that a 48 h test showed a 143 mg/L LC50 on Daphnia magna from <20 nm TiO_2 ENMs, while Heinlaan et al. (2008) observed a 48 h LC50 of 20,000 mg/L on Daphnia magna from 25 to 75 nm TiO_2 in water. These indicate the variation in toxicity between different sizes of tested ENMs, but still different tests are difficult to compare due to variations in test conditions. Kashiwada (2006) also showed that particle size has an effect on accumulation of fluorescent nanoparticles in the Japanese medeka (Oryzias latipes), as smaller particles accumulate rapidly. On cell level, Hussain et al. (2009) showed that there is a size-dependent cellular interaction of silver nanoparticles in alveolar macrophages, and the cytotoxicity was 10 times higher for ENM particles of 15 nm than for those of 30 nm. The crystal structure of ENM materials also plays a role, e.g. TiO_2 in anatase crystal structure form is more toxic to organisms than the rutile form (Hall et al. 2009; Wang et al. 2008).

During dissolution, ENMs become smaller and may completely dissolve, depending on material and environmental conditions, which may influence the

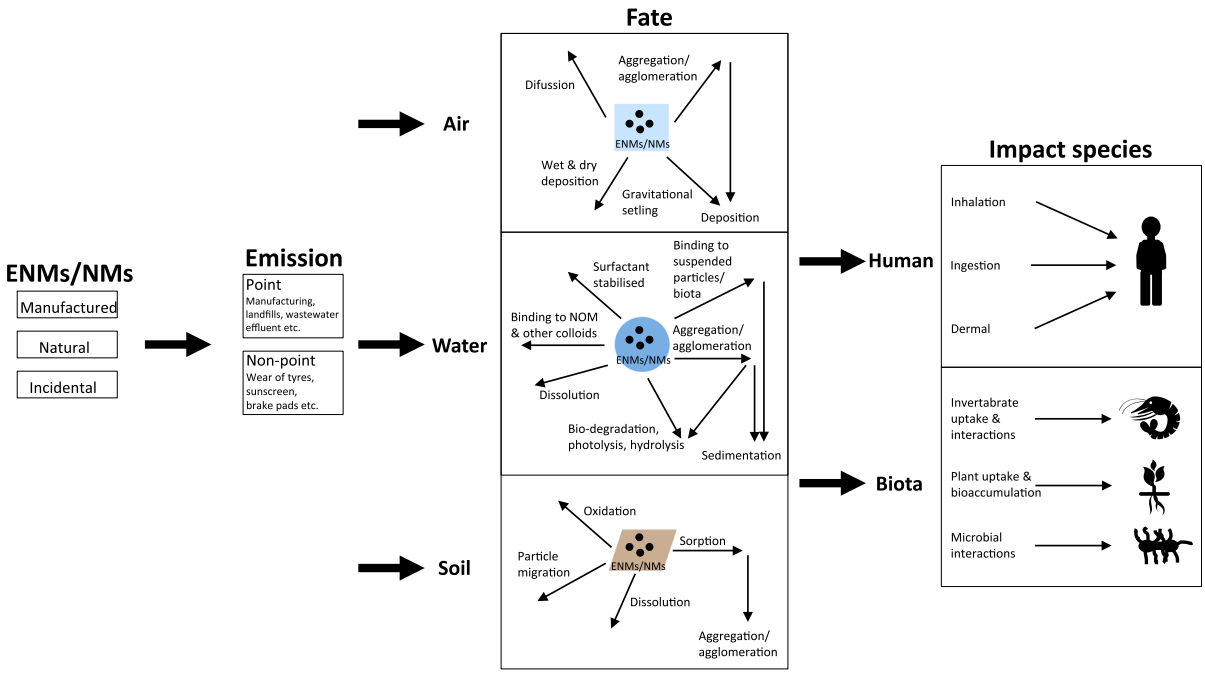

Fig. 3 Potential environmental pathways of released engineered and naturally occurring nanomaterials (ENMs and NMs) in air, water and soil and related to common impact organisms (based on Klaine et al. 2008; Batley and McLaughlin 2010; Wiesner et al. 2006)

toxicity significantly. The toxicity, in the case of Ag ENMs released to freshwater, will depend on the intrinsic toxicity potential of ENMs and the ions formed through oxidative dissolution (Scheringer et al. 2010). In continuation, Scheringer et al. (2010) mentions that the high toxicity potential of free Ag ions in natural waters may be disrupted by the presence of complexing ligands, as they will reduce the silver ion concentration and the related bioavailability of those. The toxicological effects are also related to change in surface structure of ENPs which may be caused by removal/alteration of the coating. Change in surface structure, e.g. by natural and anthropogenic chemicals in the environment, may result in enhanced mobility, bioavailability, aggregation (mainly hydrophobic surfaces), sedimentation, dissolution and dispersion (mainly hydrophilic surfaces), and consequently the actual exposure and toxicity may increase (Vonk et al. 2009; Lowry and Casman 2009). Also, solution pH and the presence of adsorbing molecules and ions have an influence. The closer the pH is to the isoelectric point, the particle charge becomes lower and a change in the repulsive forces is able to promote aggregation/agglomeration (Franklin et al. 2007; Illés and Tombácz 2006). The correlation of these processes, in contrast to single-chemical behaviour, means that single-chemical impact models are usually not suitable to use for ENMs (Lowry and Casman 2009).

After aggregation, the gravitational forces cause sedimentation and ENMs are removed from the water layer and the material is less available to certain water organisms, but more to the benthic organisms in the water environment (Klaine et al. 2008; Lowry et al. 2012). Turbulent motion in benthos and bio-turbulation of sediments can cause re-suspension and make the material more available to organisms in the water layer (Klaine et al. (2008).

Sensitivity analysis—potential ecotoxicological impacts from Ag and TiO_2 ENM products

The few LCA studies that mention the release of ENPs, mainly in the use phase, have not taken the step further towards assessment of the effects, e.g. Meyer et al. (2010). Only Walser et al. (2011) takes this step even though a nanomaterials specific impact characterisation factor was not calculated. Meyer et al. (2010) concludes that ENPs are released from Ag ENM-fabrics during washing, and Geranio et al. (2009) quantifies the potential release of silver during washing. However, the potential impact of the release is not assessed and that is the aim of the sensitivity analysis in this study. In a LCA perspective, the effect factor (EF) should be applied along with the fate factor and exposure factor in order to derive the characterisation factor (see Eq. 2). Due to the challenges in modelling the fate and exposure, described in "Difficulties in toxicological impact characterisation of ENPs" Section, a scenario of direct emission to freshwater with subsequent exposure has been made in this sensitivity analysis. Direct emission to freshwater is a worst-case scenario, while the fate factor (FF) set to 1 day is a best-case scenario due to the short substance residence time in freshwater. The EF for Ag and TiO_2 ENM was calculated from the potentially affected fraction (PAF) of species (Larsen and Hauschild 2007a, b). In comparative assessments like LCA, the effect factors should be based on HC50 (hazardous concentration for 50 % of the species) and PAF (Rosenbaum et al. 2008; Larsen and Hauschild 2007a, b):

$$\mathrm{EF} = \frac{0.5}{\mathrm{HC50}} = \frac{0.5\,\mathrm{PAF}}{\mathrm{HC50_{EC50}}} \tag{3}$$

HC50 values

HC50 is defined as the hazardous concentration in kg/m^3 at which 50 % of species in an aquatic environment are exposed to a concentration above their EC50. As normally in standardised testing, three trophic levels are included and it is recommended that at least three toxicity values are available at each trophic level (Rosenbaum et al. 2008; Larsen and Hauschild 2007a, b). In this study, the $HC50_{chronic}$ values are based on chosen $EC50_{acute}$ values according to standards from Rosenbaum et al. (2008) and Larsen and Hauschild (2007a, 2007b). In the supporting information, in Tables 3 and 4, the calculation of $HC50_{EC50}$ for Ag and TiO_2 ENMs (mainly <200 nm diameter size, see Tables 3 and 4 in supporting information) from freshwater species data for three trophic levels is shown, according to Larsen and Hauschild (2007a, 2007b) and Rosenbaum et al. (2008).

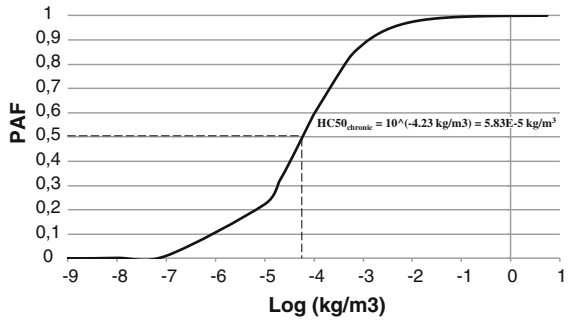

Fig. 4 Aquatic species (algae, daphnia, fish) sensitivity distribution for Ag ENM with PAF curve derived from $HC50_{EC50}$ values from Table 3 (supporting information)

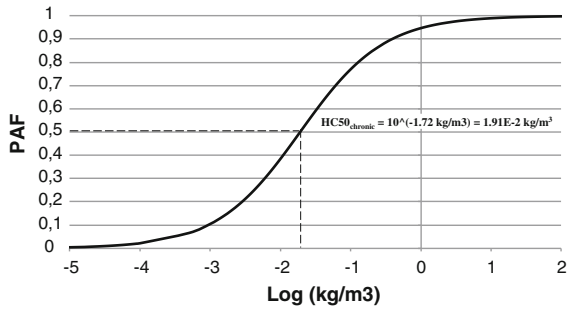

Fig. 5 Aquatic species (algae, daphnia, fish) sensitivity distribution for TiO_2 ENM with PAF curve derived from $HC50_{EC50}$ values from Table 4 (supporting information)

Freshwater species sensitivity distribution

Following the derived $HC50_{EC50}$ geometric mean results for Ag and TiO_2 ENM, the aquatic species sensitivity distributions can be derived, where C is the concentration of Ag or TiO_2 ENM, α is the average chronic logHC50 for Ag or TiO_2 ENM and calculated on the basis of Tables 3 or 4 (supporting information), β is the scale parameter and σ is the standard deviation of $HC50_{EC50}$ values in Tables 3 or 4 (supporting information) (Larsen and Hauschild 2007a, b):

$$\mathrm{PAF} = \frac{1}{1 + e^{\frac{-(\log C - \alpha)}{\beta}}} \tag{4}$$

$$\beta = \frac{\sqrt{3}}{\pi} \cdot \sigma \tag{5}$$

The PAF curve is thus derived and shows the species sensitivity, on the basis of Tables 3 and 4 from supporting information, in freshwater in relation to ENM Ag and TiO_2 concentrations (Figs. 4, 5).

Freshwater characterisation factor

The PAF curves show that in order to affect 50 % of species in freshwater, the concentration must be $5.83\mathrm{E}-5$ kg Ag ENM/m^3 and $1.91\mathrm{E}-2$ kg TiO_2 ENM/m^3 ($HC50_{chronic}$). Hereby, Ag ENMs are considered more toxic towards the freshwater species than TiO_2 ENMs, in accordance to e.g. Klaine et al. (2008). From the $HC50_{chronic}$, the effect factor (EF) can be derived with the aim of generating characterisation factors (CF) expressing the PAF resulting from release of 1 kg of Ag or TiO_2 ENM. As mentioned in "Life-cycle impact assessment and life-cycle assessment" Section, the exposure factor (XF) is neglected for ecotoxicity and the fate factor (FF) is due to severe challenges (mainly many unknowns) simplified to 1 day assuming rapid transformation, aggregation and sedimentation of ENMs:

$$\mathrm{CF} = \mathrm{EF} \cdot \mathrm{XF} \cdot \mathrm{FF} \approx \mathrm{EF} \cdot \mathrm{FF}_{\mathrm{simplified}} \tag{6}$$

$$\mathrm{CF}_{\mathrm{Ag\,ENM}} = \mathrm{EF} \cdot \mathrm{FF}_{\mathrm{simplified}} = \frac{0.5\,\mathrm{PAF}}{\mathrm{HC50}_{\mathrm{chronic}}} \cdot 1\ \mathrm{day} = \frac{0.5}{5.83\mathrm{E}-5\,\frac{\mathrm{kg}}{\mathrm{m}^3}} \cdot 1\ \mathrm{day} = 8.57\mathrm{E}3\,\frac{\mathrm{PAF} \cdot \mathrm{m}^3 \cdot \mathrm{day}}{\mathrm{kg}} \tag{7}$$

$$\mathrm{CF}_{\mathrm{TiO_2\,ENM}} = \mathrm{EF} \cdot \mathrm{FF}_{\mathrm{simplified}} = \frac{0.5\,\mathrm{PAF}}{\mathrm{HC50}_{\mathrm{chronic}}} \cdot 1\ \mathrm{day} = \frac{0.5}{1.91\mathrm{E}-2\,\frac{\mathrm{kg}}{\mathrm{m}^3}} \cdot 1\ \mathrm{day} = 2.62\mathrm{E}1\,\frac{\mathrm{PAF} \cdot \mathrm{m}^3 \cdot \mathrm{day}}{\mathrm{kg}} \tag{8}$$

Characterisation factors applied on Ag and TiO_2 ENM case studies

The derived characterisation factors, in Freshwater characterisation factor Section, for Ag and TiO_2 ENM were applied to the release in the case studies (case studies details can be seen in supporting information). These studies represent the actual release, achieved through laboratory testing. The release is based on the total silver or titanium(-dioxide) content, and thus may not be in ENM or ENP size definition (ISO 2008; SCENIHR 2007). The release is assumed to be in the ENM size, even if not all may be within that range, and the release is assumed to directly being led to freshwater environment. Thus, this scenario represents the worst case as it is based on total Ag and TiO_2 content released, and assumes that washing water will be led directly to a freshwater recipient without passing a wastewater treatment facility. Tables 5 and 6, in the supporting information, summarise the results, comprising potential release and characterised ecotoxicity impact results from ENM products.

Figures 6 and 8 show the characterised impact results, also seen in the last column in Tables 5 and 6 in the supporting information. The ecotoxicity impact from 1 g of Ag ENM products varies within the range of 1E − 7 to 1E4 PAF*m^3*day, dependent on the amount released from the products. For Ag ENM, according to the linear regression, the ecotoxicity impact is 1E − 6 PAF*m^3*day per µg Ag ENM content in the products (Fig. 7).

The TiO_2 ENM ecotoxicity impact, per gram product, is within the range of 2E − 6 to 4E − 5 PAF*m^3*day, while the linear regression based on these products indicates an increase of 2E − 9 PAF*m^3*day per µg TiO_2 ENM content in product.

The conclusion based on the Ag and TiO_2 ENM case studies and the ENM characterisation factors is that the ecotoxicity impact is higher for Ag than for TiO_2 cases. This can be seen from the derived characterisation factors in "Freshwater characterisation factor" Section and the impacts displayed in Figs. 6 and 8, as these show a higher potential ecotoxicity impact to freshwater organisms (the ones included in Tables 3 and 4 that can be seen in the supporting information). Also, as seen in Fig. 6, there is a linear regression for Ag cases, while for TiO_2 in Fig. 8 this cannot be concluded (based on only 11 release cases found). This means, for Ag cases, that the content of Ag ENMs in products is determining for amount of Ag released (mainly due to washing), and consequently to the ecotoxicity impacts to the freshwater species.

In Fig. 7, it is shown for Ag ENM products that the most common products (textiles, socks and shirts) represent the highest impacts, except for the medical mask and cloth that represents only two datasets. These impacts, only originating from the Ag release, are ca. 1–3.5 orders of magnitude higher according to median (excl. baby plush toys & baby blanket) than

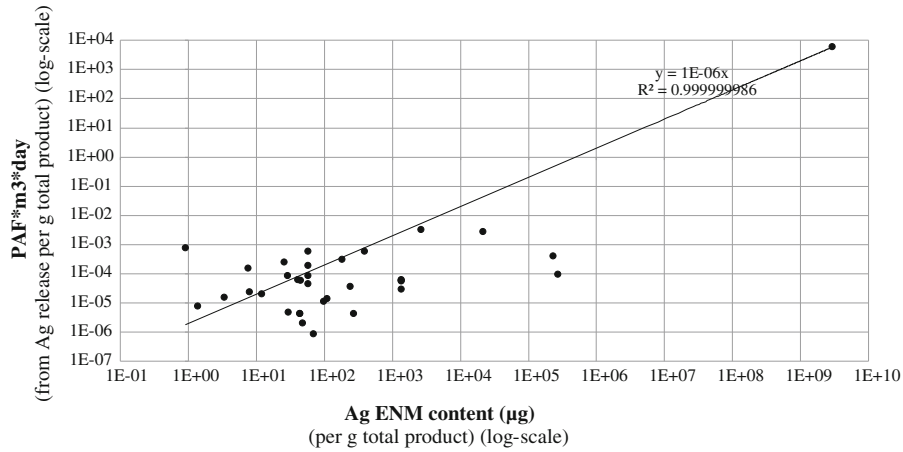

Fig. 6 Ecotoxicity impact from release of Ag (total metal) from products containing Ag ENM, data displayed according release related ecotoxicity per g total product. Based on data from Table 3 (Fig. 4) and 5 (supporting info.). A detailed chart can be seen in the supporting information (Fig. 11)

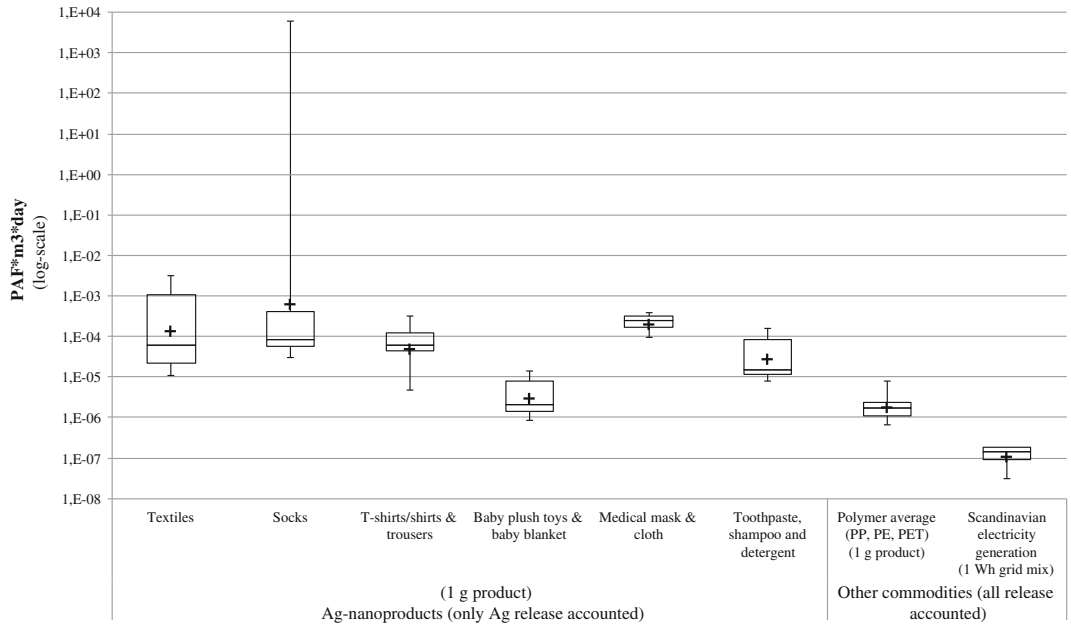

Fig. 7 Ecotoxicity impact per gram of Ag ENM product and also compared to non-ENM commodities as ecotoxicity impact per gram of polymer (PP, PE, PET European (RER) average, Ecoinvent v2.2 and Buwal database) and per Wh electricity generated (Scandinavian countries grid mix average, Ecoinvent v2.2 database). Based on Table 3 and 5 from supporting information (see also Figs. 4 and 6). Cross (+) indicates the geometric mean

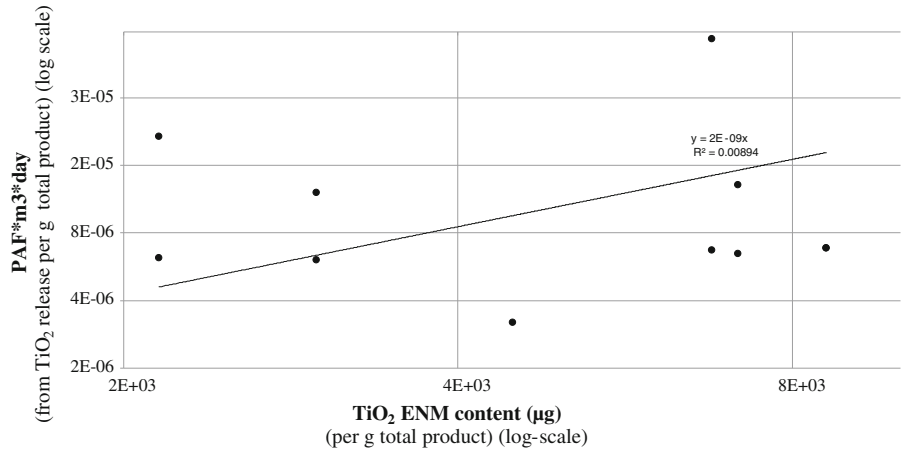

Fig. 8 Ecotoxicity impact from release of TiO2 (total metal) from products containing TiO2 ENM, data displayed according release related ecotoxicity per g total product. Based on data from Table 4 (Fig. 5) and 6 (supporting info.). A detailed chart can be seen in the supporting information (Fig. 12)

the impacts from production of 1 g PP, PE and PET polymer average or 1 Wh of grid mix electricity from Scandinavia. The polymer and electricity production data were attained from Ecoinvent v2.2 and Buwal database, and the ecotoxicity impacts were derived through Impact 2002+ LCA method (Jolliet et al. 2003). The ecotoxicity impact potential from the polymer and energy production originates from the entire production chain, in contrast to the ENM products where the accounted impacts are based solely on the release of Ag in the use-stage (see Fig. 7). These are provided in order to place the potential impacts of released Ag ENM in perspective. Even though assumptions are made in order to quantify the

impact potential from e.g. washing of t-shirts containing different Ag ENM textiles, it should still be clear that the release will occur (Geranio et al. 2009) and the related freshwater impacts will be created if the waste-water treatment is not suited for the complete removal of Ag ENM. Also, impacts of Ag ENM in freshwater could be related to dissolution into silver ions as according to Ratte (1999) the silver ion (Ag+) is defined as the most toxic form of silver in water. Silver speciation commonly occurs in freshwater with sulphide and chloride and this reduces the silver bioavailability, but even though this is the most realistic fate, the toxicity studies are quite simple and may to a lesser extent include speciation in the laboratory setup for toxicity measurements to freshwater organisms (Scheringer et al. 2010; Fabrega et al. 2011; Jones and Grainger 2009). Due to difficulties in empirically testing different scenarios, biological models have been introduced and one of those is the Free Ion Activity Model (FIAM) and the Biotic Ligand Model (BLM) used to predict acute metal toxicity to aquatic vertebrates and invertebrates (Fabrega et al. 2011). The main issue with such a model is that it is equilibrium based and not always suitably models what happens in real life (Slaveykova and Wilkinson 2005; Fabrega et al. 2011). Further, it is unknown how suitable some of the assumptions are for ENMs, e.g. dominance of free ion activities in determining bioavailability, aqueous phase chemistry is at equilibrium and that uptake and flux across the membrane is rate limiting (Fabrega et al. 2011).

Regarding TiO_2 ENM products, as seen from the boxplot in Fig. 9, the t-shirts washed 10-times have higher ecotoxicity impact due to higher release, than e.g. after one wash. The impacts, only originating from the TiO_2 release, are ca. 1–2 orders of magnitude higher (median) than the impacts from production of 1 g PP, PE and PET polymer average or 1 Wh of grid mix electricity from Scandinavia. The release of Ag or TiO_2 in the use stage has in almost all cases higher impacts than the total ecotoxicity impacts from production of 1 g polymer (PP, PE and PET average) and 1 Wh of Scandinavian electricity (grid mix). This sensitivity underlines the need to consider the release of ENPs and/or ENMs during use, solely due to potential ecotoxicity impacts.

According to Jones and Grainger (2009), the main hurdle is still to model and predict the actual fate of ENPs

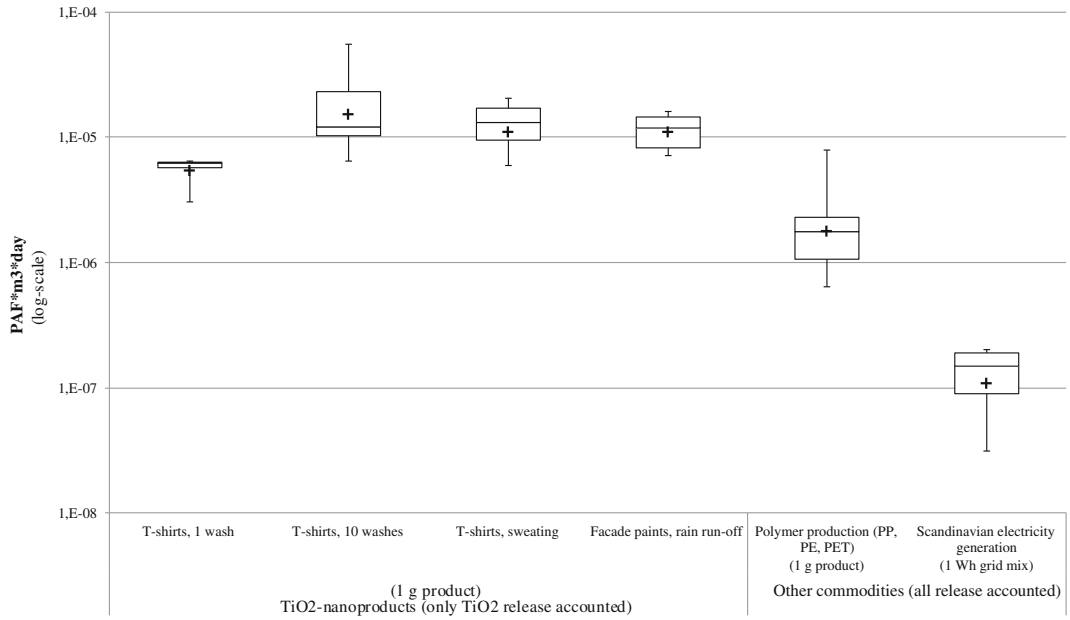

Fig. 9 Ecotoxicity impact per gram of TiO_2 ENM product and also compared to non-ENM commodities as ecotoxicity impact per gram of polymer (PP, PE, PET European (RER) average, Ecoinvent v2.2 and Buwal database) and per Wh electricity generated (Scandinavian countries grid mix average, Ecoinvent v2.2 database). Based on Tables 4 and 6 from supporting information (see also Figs. 5 and 8). Cross (+) indicates the geometric mean

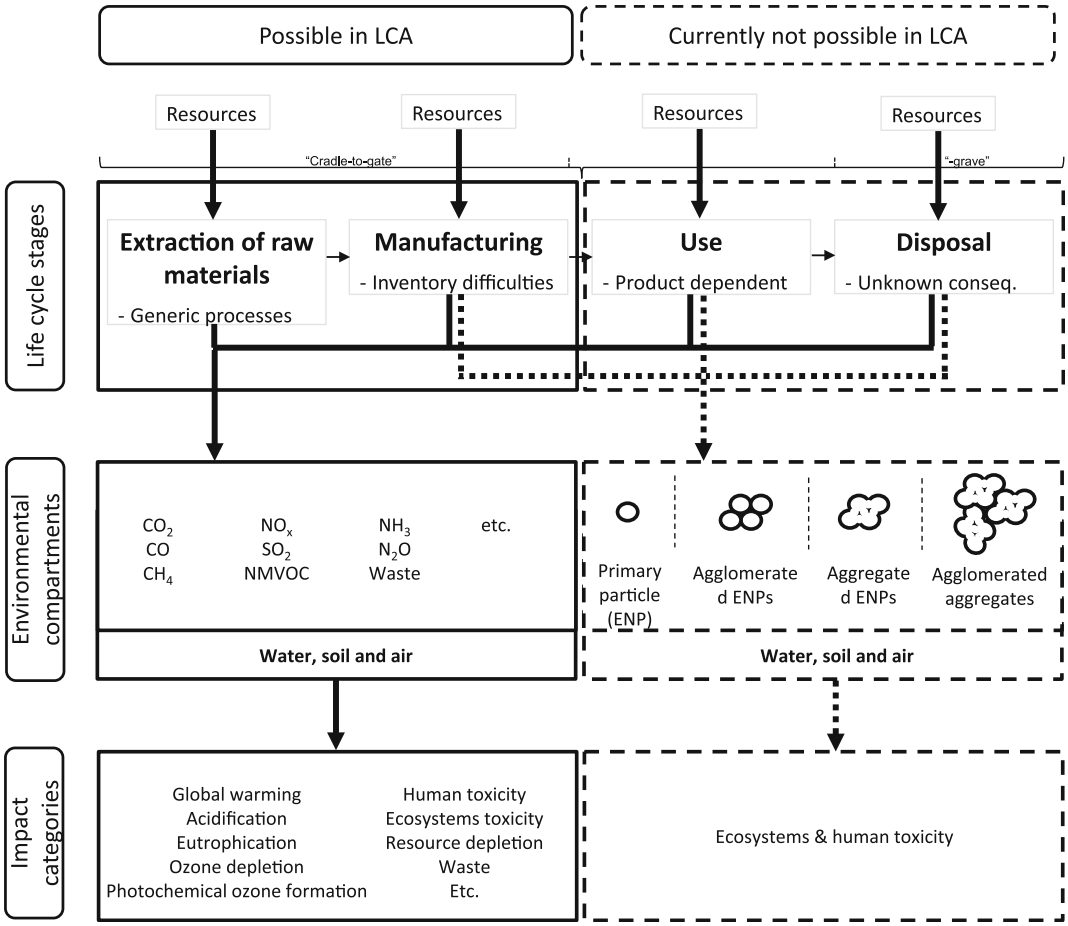

Fig. 10 Possibilities and limitations of LCA—what currently can be assessed in LCA. Figure is partially based on Som et al. (2010) and a reproduced figure from Oberdörster et al. (2007)

when they are released to the environment since more products with ENMs are introduced to the consumer market. For example, if more washable products contain Ag ENM, the actual hazard may be higher in terms of PAF, but this of course also depends on the actual fate of ENMs as described in "Difficulties in toxicological impact characterisation of ENPs" Section.

Limitations of LCA of ENMs—what needs to be considered

In order to perform an LCA of ENMs, certain aspects need to be considered and some are more important than others. As described in "Life Cycle Assessment case studies of ENMs" and "Difficulties in toxicological impact characterisation of ENPs" Section, the potential ENP/ENM release is commonly not included in LCAs. This is partly due lack of knowledge about the release, partly due to a missing understanding of how to quantify the ecotoxicity as a function of particle release from ENM products during its life cycle.

Figure 10 illustrates LCA limitations regarding the impact assessment of ENMs. The limitations are linked to the limited knowledge on fate and effects of released ENPs. Currently, the LCAs performed on ENMs disregard the potential (eco-)toxicity impacts of ENP release to environmental compartments. In order to include these, there is a need to develop characterisation factors in collaboration with the RA field. Currently, if aiming at performing an LCA one should be aware of the following:

- Goal and scope: If possible, consider the whole life cycle and set goals according to LCA possibilities (see Fig. 10). The functional unit needs to take into

account the functionality and property differences of the product when using ENMs.

- LCI: Data are difficult to acquire, so either collaborate with the industry or base the data on already published studies and generic processes from databases as e.g. Swiss Ecoinvent.
- LCIA: A completely holistic impacts assessment cannot be performed, mainly due to the unanswered questions regarding accounting of ENM/ENP release and the related impacts (Som et al. 2010; Bauer et al. 2008):
- How much ENM/ENP is released to environmental compartments (e.g. water) and technosphere (e.g. waste-water treatment)?
- Which exposure to ENMs/ENPs occur in the environment and what are the effects on biota and humans?
- At different times, what appearance (size, shape and composition) do the ENMs/ENPs take in the environment (primary particles (ENPs), agglomerated ENPs, aggregated ENPs, agglomerated aggregates)?
- What are the environmental consequences from different end-of-life treatment of ENM products?

These impact assessment-related problems need to be uncovered and solutions to these included in future LCA. The general approach, as mentioned previously, will be to include RA developments in LCA, so fate and end-point effects can be quantified.

Conclusion

Currently, the application of LCA of ENMs is not well developed and there are different reasons for this. Firstly, due to the relative novelty of ENMs, the inventory data available are limited making it difficult to establish the actual inventory data of ENM raw material extraction and production. Secondly, the potential release of ENP during the life cycle is not yet well investigated (partly due to measuring difficulties). Finally, after release of ENMs to the environment, there is still an incomprehensive understanding of processes controlling the fate and effect of released ENPs. In this context, only one study managed to include impacts related to ENM release in their LCA of a nanoproduct, although a nanomaterial specific impact characterisation factor was not calculated (Walser et al. 2011). To make LCA more robust when assessing ENMs the factors controlling transport, transformation, fate (e.g. aggregation and deposition) and effect of released ENPs must be uncovered and corresponding LCA environmental impact characterisation factors should be developed. Due to constraints of unknowns and time the prospect is to develop simplified models that may serve in preliminary fate and effects assessments of released ENMs. There is also a need to develop a better LCI database for ENMs, as Bauer et al. (2008) also underlines.

The conclusion of the sensitivity analysis underlines the need to consider this potential impact source during a products lifetime, as the quantified ecotoxicity impacts in this study for Ag and TiO_2 ENM products are higher compared to other frequently applied processes such as polymer production (European PP, PE and PET average) and electricity (Scandinavian grid mix). Further, Ag ENMs, compared to TiO_2 ENMs, contribute to a higher ecotoxicity to freshwater algae, daphnia and fish. The ecotoxicity impacts originating from the Ag and TiO_2 release from Ag and TiO_2 ENM product, pose up to ca. 3.5 orders of magnitude higher (median) ecotoxicity impact for Ag and ca. 1–2 orders of magnitude higher (median) ecotoxicity impact for TiO_2 than production of 1 g polymer (European PP, PE and PET average) or 1 Wh of grid mix electricity from Scandinavia. Additionally, for Ag ENM products, a linear regression was observed between Ag ENM content in the products and the release of total Ag during use (mainly washing), consequently leading to potential ecotoxicity impacts to algae, daphnia and fish freshwater species.

Some may question the need to apply LCA on ENM products, but LCA has been proven as one of the only holistic tools to understand the overall impact of products and systems—meaning that it covers broadly in terms of including the whole life cycle, all potential impacts and their range of categories. Thus, LCA has good potential to be developed into a robust tool for assessing a wide range of impacts deriving from ENM product life cycles. Based on the review, we observe that LCAs have been applied on ENM products, but so far mainly to assess the accountable production-related emissions. Thus, we find that the future aims should be to further map, develop and validate the application of LCA of ENMs and, importantly, collect inventory data and develop impact characterisation

factors to further the assessment of potential environmental and toxicological impacts from released ENMs/ENPs. To supplement this massive task that lies ahead, the sensitivity analysis performed in this study on Ag and TiO_2 identifies some of the aspects needed in development of impact characterisation factors, but the approach to calculate, i.e. the freshwater fate factor for each ENM especially needs more attention. In this sense, LCA should be used with caution as release, fate and effect of ENM/ENP is currently not properly accounted for and the actual environmental implications not enough investigated.

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