

Chapter 32

LCA of Nanomaterials

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Abstract Application of nanomaterials in products has led to an increase in number of nanoproducts introduced to the consumer market. However, along with new and improved products, there is a concern about the potential life cycle environmental impacts. Life cycle assessment is able to include a wide range of environmental impacts but, due to data limitations, it is commonly applied with focus on the cradle-to-gate part of the nanoproducts life cycle, neglecting use and disposal of the products. These studies conclude that nanomaterials are more energy demanding and have an inferior environmental profile than conventionally used materials, but functional units of these comparisons need to consider the use stage benefits attained through nanomaterials. A particular assessment challenge is the lack of understanding of the toxicological mechanisms related to potential release, fate and effects of nanomaterials when penetrating into living organisms. This is especially relevant for the freshwater compartment, as it is a common recipient.

32.1 Introduction

The basis of the nano-technology terminology is the nanometre, which is one billionth of a metre (10^{-9} m). Nano-scale is defined as the range from 0.1 to 1000 nm, nanomaterial as a material with at least one dimension within 1–100 nm, and nanoparticle as a particle with all three dimensions within the 1–100 nm range (ISO 2008; SCENIHR 2007). Nanomaterials and nanoparticles can be naturally or unintentionally produced, and they have always been present in the human surroundings. Examples of these are, e.g., soil (dust) and salt particles in air, to which humans are commonly exposed. These naturally occurring nanomaterials are in general known to cause little harm to humans (Buzea et al. 2007). Other sources of naturally occurring nanoparticles are, e.g. forest fires or volcanos, the particles from

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which may have the same impacts as the anthropogenic, unintentionally produced particles arising from combustion activities. The other path to production of nanomaterials is by manufacturing activities through either downscaling (from bulk) or upscaling (from atomic or molecular) materials. These activities may also cause unintentional emissions of nanoparticles. The manufacturing of engineered nanomaterials (ENMs) is linked to application of nanomaterials in/on consumer products, as alteration of materials from bulk to nano-size leads to an increase in surface area and improved functionality. As an example, gold becomes reactive at nano-size and may be used to increase oxidation in car catalytic converters so emission of pollutants is reduced. Further, ENMs may also provide products with, e.g. improved material properties like hydrophobicity (lotus effect), strength and/or electrical conductivity.

For quite some time particles from combustion have been known to cause harmful human impacts, even though the exact mechanisms are still being researched. ENMs and nanoparticles (ENPs) also cause concerns, but their behaviour in the environment and potential impacts to environment and humans are to a large extent still unknown (Miseljic and Olsen 2014; Jolliet et al. 2013). In order to embrace the entire life cycle of ENMs, and to avoid burden shifting, the LCA approach is favourable in order to quantify potential environmental impacts—not only as a single approach but also as a framework to be applied along with other methods (Som et al. 2010).

32.1.1 Nanoproducts and Environmental Assessment

Nano-technology and the application of ENMs in products have developed much in recent years. The reason for this is that ENMs are able to improve properties and functionalities of different materials, and thereby the consumer products. This has meant that companies have developed new and smart products, resulting in more nanoproducts being introduced to the consumer market (see Figs. 32.1 and 32.2). The more common nanoproducts are within the product category of health and fitness, with TiO₂-enhanced sunblock and Ag-enhanced clothing as prominent examples. The home and garden product category contains the second most widely used nanoproducts (mainly as sealants and coatings), as seen in Fig. 32.1. The most commonly used nanomaterials are based on Ag, TiO₂, and carbon as shown in Fig. 32.2.

In line with the rapid introduction of nanoproducts, and thereby ENMs, concerns are raised in terms of the potential environmental impacts these may have along their life cycle. Currently, chemical risk assessment (RA) is commonly performed on ENMs, but this approach has a different scope compared to LCA. RA is a procedure applied in order to estimate if a toxicological risk occurs from a substance, thus calculating, measuring or modelling the existence of risk derived from chemical exposure (European commission 2007; ECHA 2010, see also Sect. 31.1.3). LCA on

Fig. 32.1 Number of ENM products per product category (PoEN 2014; Hansen 2009; Miseljic 2014)

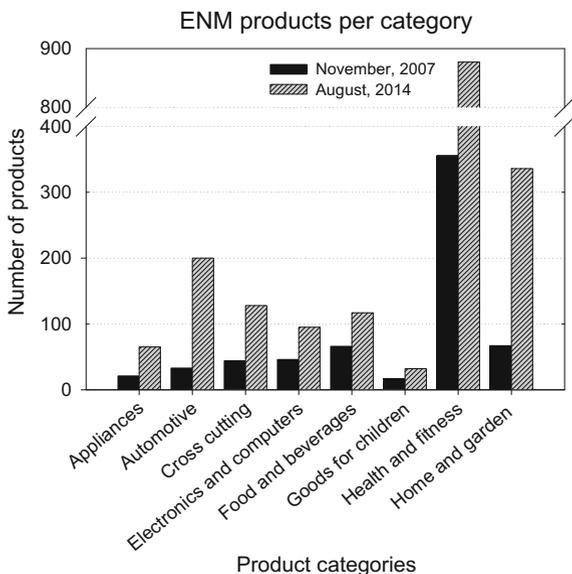
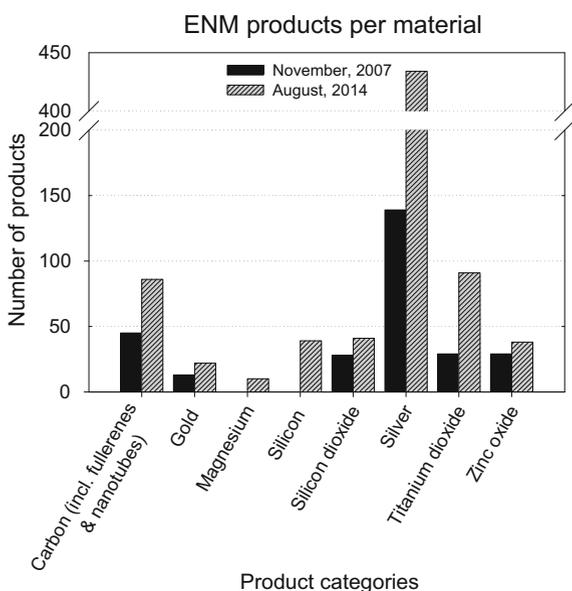


Fig. 32.2 ENM products per most frequently used material, in year 2007 and 2014 (PoEN 2014; Hansen 2009; Miseljic 2014)



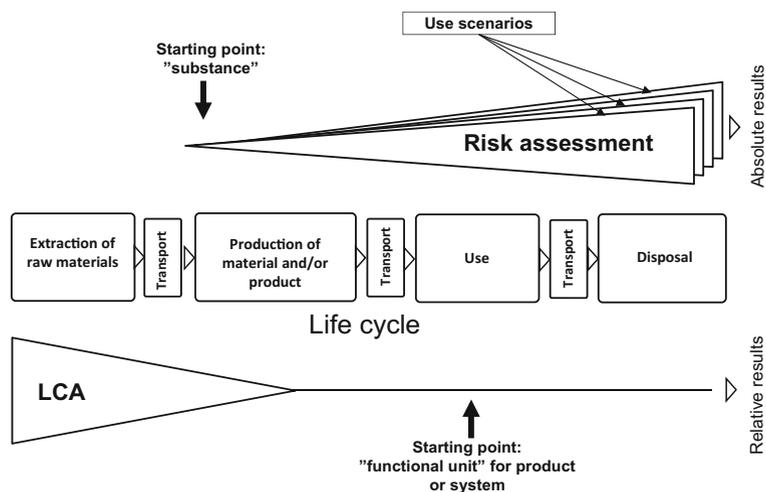


Fig. 32.3 RA and LCA coverage and starting point when addressing a product or system (Grieger et al. 2012)

the other hand is a relative environmental impact assessment method that also considers a wide range of environmental impact categories (ISO 2006). In LCA, the starting point of the assessment is the functional unit, where products or systems are studied, whereas RA is commonly substance oriented, see also Fig. 32.3.

There are benefits and challenges to applying both RA and LCA, due to these two approaches being developed with different initial purposes. The benefit of applying RA for ENMs is that it targets specific emission, exposure and dose-response conditions, but the downside is that the uncertainty is still high due to lack of data and lack of proper regulation. A refinement of regulation is needed, e.g. for the characterisation of ENMs applied in laboratory testing and thereby the use of appropriate metrics for expressing hazard and exposure. In addition to ENM mass, Aitken et al. (2011) and Hankin et al. (2011) proposed particle number and surface area as additional characteristic metrics.

LCA conveys an assessment of a wider range of environmental impacts, but as for RA there are also challenges to this approach. One of these is, as for RA, the lack of needed data and another is the lack of consideration of impact from ENM/ENP released to the environment.

Sweet and Strohm (2006) and Som et al. (2010) outlined that RA should consider more life cycle concepts and LCA should be more risk-based, when dealing with future environmental impact assessment of ENMs. This underlines the general approach that LCA should be applied as a complimentary framework along with other environmental impact assessment approaches (Grieger et al. 2012).

32.2 Literature Review

In order to evaluate the current assessment status of ENMs, and the application of LCA in this context, published scientific articles were reviewed. Only 29 LCA case studies were found and reviewed according to strengths and weaknesses, and the challenges the assessments represent (Miseljic and Olsen 2014). Jolliet et al. (2013) identified 43 studies but use a broader definition of LCA. Some studies did not perform LCA according to usual guidelines (e.g. ISO 2006), but claimed to include life cycle thinking and commonly focused on energy consumption of ENM manufacturing.

32.2.1 *Impact Hotspots*

Commonly, among the reviewed studies in Miseljic and Olsen (2014) and Jolliet et al. (2013), it can be concluded that, on a same produced mass basis, the manufacture of nanomaterials is found to have significantly higher energy requirements compared to the production of conventional materials such as aluminum or steel (shown e.g. in Khanna et al. 2007, 2008). Even though there are discrepancies of up to several orders of magnitude between the energy requirements for the same nanomaterials reported by different studies this often results in a less favourable cradle-to-gate environmental impact profile of ENM products compared to comparable conventional products. The high energy demand for production emphasise that one should be careful with normally accepted cut-off criteria when making inventories of nanoproducts since the product generally contains only few percent of ENMs. In terms of impact categories, those that are dominating for ENM production are nonrenewable resource depletion, global warming, acidification and impacts caused by airborne inorganics. However, due to the improved product functionality, the use stage of ENM products is often more environmentally friendly than for conventional products. This is exemplified in Lloyd and Lave (2003) and Lloyd et al. (2005), where ENM enhanced clay-polypropylene ENMs in car body panels and platinum-group metal particles in car catalysts have an environmentally friendlier use stage. However, potential release of ENMs/ENPs to the environment is not considered in these studies.

Most of the reviewed LCA studies focused on the manufacturing of nanomaterials, thus limiting the assessment scope to the energy consumption of the cradle-to-gate part. The use and disposal stages are commonly not considered, with few exceptions, e.g. Walser et al. (2011) who also consider toxicity from ENMs/ENPs from nanoproducts. However, the tendency is also that these studies rely on generic data, as seen in e.g. Grubb and Bakshi (2010), Osterwalder et al. (2006), Tibbetts et al. (1994), Hwang et al. (2005), Healy (2006), and Healy et al. (2008).

32.2.2 Overall Findings

The tendencies across the identified LCAs on ENMs can be summed up and also reflect the LCA state of knowledge regarding ENMs (Miseljic and Olsen 2014):

- Usually the studies consider cradle-to-gate or manufacturing system boundary.
- Use and disposal stages of the life cycle are poorly covered.
- Functional unit does not always consider functional benefits of ENMs.
- Common use of generic LCI data and assumptions, e.g. for upscaling of laboratory data.
- Almost no consideration of ENM release (e.g. in use or disposal stages) and the potential toxic impacts from these (fate, exposure and effect consideration). Walser et al. (2011) and Meyer et al. (2010) are exceptions.
- Cradle-to-gate LCA comparison of counterpart products (with ENMs and without) show that ENM products are more energy demanding and therefore have a worse cradle-to-gate environmental profile, e.g. in polymer nanocomposites versus 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) show that the use stage 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). End-of-life performance of ENM products is rarely considered.

32.3 General Methodological Issues

32.3.1 Goal and Scope Definition

Generally, it is found that the *system boundaries* for a fair comparison to products containing ENMs should include both the use stage where the beneficial functionalities of the ENMs are expressed, and the disposal stage. Certain reviewed studies consider the use and disposal stages, but their coverage is rather incomplete (Lloyd and Lave 2003; Lloyd et al. 2005; Babaizadeh and Hassan 2012; Manda et al. 2012; Roes et al. 2007; Steinfeldt et al. 2010). In the quoted example by Lloyd and Lave (2003) on clay-polypropylene ENMs in car body panels, the use stage is assessed by solely including the resource savings (fuel consumption) when driving with the lighter ENM enhanced panels. Other supplementary materials are not considered, meaning that release of other agents is not considered in the use stage (Som et al. 2010). In addition, the disposal stage is commonly not dealt with due to lack of knowledge in end-of-life treatment of ENMs and also which disposal processes they will be subject to (landfilling, incineration, or recycling).

In order to perform a comparable LCA the *functional unit* is central, as a comparable functionality of products or systems needs to be applied. Certain ENM studies tend to apply a simplified mass based functional unit, e.g. relating to 1 kg of an ENM product (Joshi 2008; Kushnir and Sandén 2008; Grubb and Bakshi 2010). However, a mass-based functional unit does not make sense when comparing ENMs with conventional products, as functionality is not proportional with weight (Hischier and Walser 2012). The improved material functionality, when using ENMs in products, needs to be considered in the functional unit. This means that higher resource and energy use in ENM production, compared to conventional additives, may be justified in the use stage by leading to less environmental impacts.

As an example, Roes et al. (2007) include elasticity (Young modulus) and tensile strength in the functional unit when comparing polypropylene (PP)/layered silicate nanocomposites with conventional PP, reason being that the nanocomposite and PP mix obtains the needed material properties at a lower weight. In this approach Roes et al. (2007) scale the functional unit, but in general it may be difficult to identify the most important properties that are to be applied in a functional unit of a fair LCA comparison.

32.3.2 *Inventory Data*

The inventory modelling of a nanoparticle typically involves background processes that are not specific to nano and thus not of specific interest here, while foreground process (e.g. data for production of ENM; and direct nanoparticle emissions) should be specific of the considered process (Jolliet et al. 2013). Commonly, as identified in the 29 LCA on ENMs studies from Miseljic and Olsen (2014), the studies rely on generic data for production of ENMs. Primary process data, on the other hand, is often difficult to acquire from the ENM industry. This can be due to several factors, but mainly to the relative novelty of the scientific field and the competition within the technology domain. This ongoing tendency leads to a higher level of uncertainty, as generic data and estimations need to be applied (e.g. in Bauer et al. 2008; Joshi 2008; Merugula et al. 2010; Isaacs et al. 2006). The immaturity of the field also lies behind the incomplete life cycle coverage in the performed LCAs with the very frequent omission of use and disposal stages from the product system modelling.

Life cycle inventory (LCI) modelling is gradually improving for ENMs, studies such as Geranio et al. (2009), Köhler et al. (2008), Künninger et al. (2010), Som et al. (2011), Suppen et al. (2005), Durucan et al. (2006) and Gutowski et al. (2010) do not perform LCAs per se, but are providing valuable LCI data on specific processes as well as some estimates of the direct release of nanoparticles. Geranio et al. (2009) quantifies the release of Ag ENPs during textile washing and Künninger et al. (2010) the release of nano-Ag from facades due to weathering.

Concerning the direct emissions of nanoparticles, the physical and chemical characteristics of the emitted nanoparticles are also of interest since these characteristics are needed to link up with the impact assessment.

32.3.3 *Impact Assessment*

Generally, the impact from background and foreground process is no different from other LCAs. Therefore, the main aspect to consider is the potential impacts from direct emissions of nanoparticles. Here the potential toxic effects have caused highest concern and the next chapter will deal exclusively with this.

32.3.4 *Interpretation*

It is evident from the previous text that LCAs of nanoproducts are rather uncertain and that an understanding of the main uncertainties is important (Jolliet et al. 2013). It is suggested to use tools for uncertainty assessment and sensitivity analysis as explained in Chap. 11.

32.4 Specific Methodological Issues for Ecotoxicological Impact Characterisation of ENMs/ENPs

As concluded in the previous section, the potential release of ENMs throughout the life cycle of ENM products is generally not considered. A sensitivity analysis of the importance of potential freshwater ecotoxicity impacts from Ag and TiO₂ ENM release from products underlined the need to consider potential impacts of such releases throughout a product's entire life cycle (Miseljic and Olsen 2014). The analysis also illustrates the differences between the impacts of different ENPs (i.e. higher freshwater ecotoxicity from Ag ENMs, compared to TiO₂ ENMs). The assessment of impacts of ENM release from cradle to grave improve the LCA of ENMs, and address an increasing environmental concern (Buzea et al. 2007; Bauer et al. 2008; Oberdörster et al. 2007; Jolliet et al. 2013). However, current lack of understanding of the mechanisms leading to toxicity, more precisely the ENM release, fate and potential effects when penetrating into living organisms, pose challenges for the assessment while being highly relevant; especially for the freshwater compartment, as it is a common recipient (Quik et al. 2011; Lowry et al. 2012; Som et al. 2010). According to Jones and Grainger (2009), the main hurdle is to predict the actual fate of the released ENMs. In the following, the most important aspects related to such an assessment are discussed as an example of the challenges specific for LCIA of nano-technology.

32.4.1 *Particle Appearance*

The primary ENP appearance differs over time and according to environmental conditions, as ENPs tend to agglomerate (coagulate), aggregate (fuse) or a combination thereof. The interaction and bonding happens in order to reduce the high-surface energy. Interaction between two ENPs, e.g. in liquid and air, can in theory be described by forces of van der Waals attractions and electrostatic repulsions (Rupasinghe R-A-TP 2011). The appearance of ENPs, along with the forces causing this, influences the toxicity of ENMs in e.g. water (Oberdörster et al. 2007).

32.4.2 *Transformation*

After release to freshwater, which is considered a common recipient, the ENMs are either subject to biotic (interaction with plants, water flea, fish etc.) or abiotic (interaction with water, sand, light, etc.) transformation. These can alter the shape, size, surface chemistry, and ultimately the fate of the released ENMs. Thus, physico-chemical properties of ENMs are important for the differentiated behaviour of ENMs in water and the water-phase processes considered important are:

- Aggregation/agglomeration
- Dissolution
- Sedimentation (and resuspension)
- Change in surface structure of ENMs/ENPs.

Commonly, freshwater fate of ENMs tends to be dominated by sorption to high-surface-area colloids with subsequent sedimentation (Klaine et al. 2008). Within the sediment the ENMs can be transported, and also re-suspended to the water phase, see Fig. 32.4.

32.4.3 *Transport*

Transport of ENMs is partially controlled by aggregation/agglomeration, which subsequently is followed by sedimentation. The aggregation will depend on parameters such as (Lowry and Casman 2009; Lowry et al. 2012):

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

Ionic strength, pH and the presence of divalent cations such as Ca^{2+} and Mg^{2+} will influence the rate and extent of aggregation/agglomeration (e.g. a higher ionic

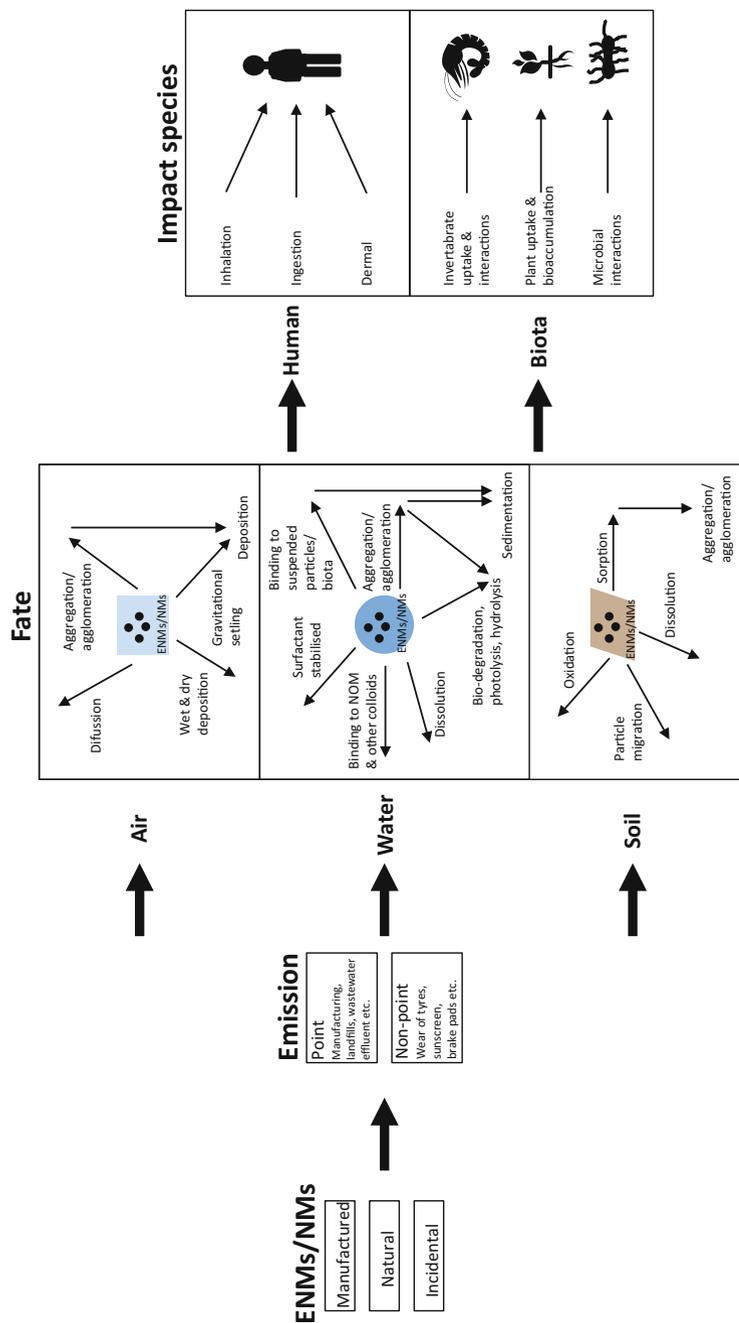


Fig. 32.4 Potential environmental pathways of released engineered and naturally occurring nanomaterials (ENMs and NMs) in air, water, and soil and related to common impact organisms (Miseljic and Olsen 2014)

strength will lead to more aggregation/agglomeration, as is the case for marine waters compared to freshwater). Brant et al. (2005) exemplify that if the ENM C₆₀ fullerene is released into water with an ionic strength higher than 0.001 M the formed aggregates/agglomerates will sorb to other particles and media and eventually become immobilised. This happens in particular when pH is close to the isoelectric point (i.e. pH at which a molecule has no net electrical charge), since the particle charge is then lower and a change in repulsive forces is able to promote aggregation/agglomeration (Franklin et al. 2007; Illés and Tombác 2006).

In water, the gravitational forces cause the aggregates/agglomerates to sediment, thus becoming less available to certain aquatic organisms, but more to the benthic organisms (Klaine et al. 2008; Lowry et al. 2012). Subsequently, turbulent motion in benthos and bio-turbation in the sediment can cause the ENM to be re-suspended and become more available again in the water phase.

32.4.4 Important Physico-Chemical Characteristics

Considering the developing understanding of the fate of ENMs in freshwater, the following physico-chemical ENM properties 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.

In addition to the ENM specific properties, the natural conditions of the surrounding environment are also important when dealing with ENM fate. However, aggregation/agglomeration and dissolution alongside other co-related mechanisms are neither fully understood nor well represented with characterisation data, in relation to mechanisms shown in Fig. 32.4 (Farré et al. 2011).

32.4.5 Toxicity

The toxic effects from ENPs and ENMs depend on several parameters, e.g. size, surface/crystal structure, dissolution and aggregation/agglomeration. Size of ENPs is proven to have an influence on the level of toxicity, e.g. 48 h testing on *Daphnia*

magna showed, according to Zhu et al. (2009), a 143 mg/L LC₅₀ for <25 nm TiO₂ (20% rutile and 80% anatase crystal structure), while Heinlaan et al. (2008) observed a 20,000 mg/L LC₅₀ for 25–75 nm TiO₂ (crystal structure not reported). These two studies along with several others (e.g. Kashiwada 2006; Hussain et al. 2009) indicate large variations between different nominal sizes of ENPs, but it needs to be underlined that tests are difficult to compare due to variations in test conditions. In addition to size, crystal structure also has an influence on toxicity, where, e.g. TiO₂ in anatase crystal structure is known to be more toxic to organisms than in rutile form.

Toxicity is dependent on the intrinsic toxicity potential of the ENMs, influenced by, e.g. size, and ions formed through oxidative dissolution (Scheringer et al. 2010). The high toxicity potential of free Ag ions in natural waters may be disrupted by the presence of complexing ligands, as they reduce the Ag ion concentration and thus the bioavailability (Scheringer et al. 2010). In addition, toxicological effects also depend on the surface structure of ENPs, where surface structure can be removed/altered, e.g. by natural and anthropogenic chemicals in the environment. Change in surface structure 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; Ratte 1999). Further, the pH and presence of adsorbing molecules and ions have an influence on ENM fate and eventually the toxicity.

The correlation of various mechanisms in ENM behaviour and impacts, in contrast to single-chemical behaviour, means that single-chemical impact models are not suitable to be applied for ENMs (Lowry and Casman 2009).

32.5 Conclusion: What to Consider When Performing LCA on ENMs

Currently, LCA of ENMs is deficient in several areas. First, novelty of the nano-technology is limiting the availability of LCI data. Second, the potential release of ENMs/ENPs is difficult to include in LCA at this point. This is due to both lack of LCI data and an incomplete understanding of fate, exposure and effects (eco- and human toxicity characterisation factors). Figure 32.5 illustrates the current state of LCA for nano-technological products.

Based on already performed LCAs on ENMs there should be awareness of the following when aiming at performing an LCA on ENMs:

- Goal and scope: If possible consider the whole life cycle and be realistic when setting goals (see Fig. 32.5). The functional unit needs to take into account the potential differences in functionality of the product when using ENMs.
- LCI: Data is difficult to acquire, so either collaborate with the industry or base the data on already published studies and generic processes from databases as

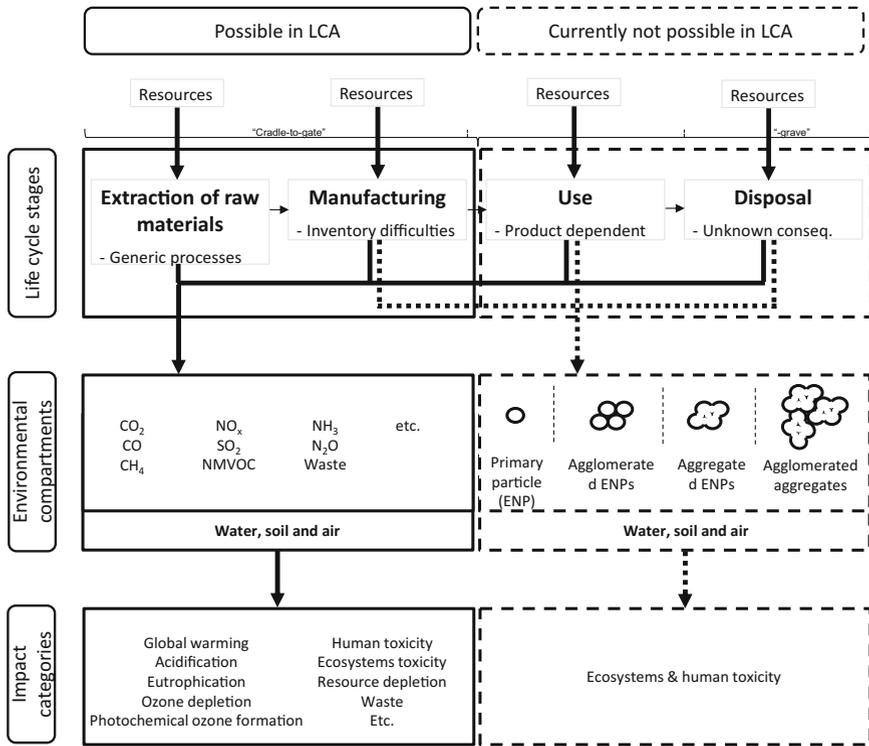


Fig. 32.5 Possibilities and limitations of LCA: what currently can be assessed in LCA (Miseljic and Olsen 2014). *Full lines* illustrate what is currently done (and is possible at the stage of current research) while *dotted lines* illustrate the challenges, especially related to the assessment of release of nanoparticles

e.g. ecoinvent. Be aware that lab-scale production data can be misleading and need to be scaled up.

- LCIA: A completely holistic impact assessment cannot yet be performed, mainly due to the challenges related to quantification 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 exposures to ENM/ENP 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?

So far, the LCAs performed on ENMs have been used to assess the accountable production-related emissions. Future LCAs should seek to develop the areas that are currently poorly covered, so that impact burden shifting is avoided. This may be done by including other approaches (e.g. RA) using LCA as a framework for gathering the best developed approaches in order to perform a holistic environmental impact assessment.

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