

*Chapter 1*

## **VERY SMALL FLOWS? REVIEW OF THE SOCIETAL METABOLISM OF NANOMATERIALS**

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### **ABSTRACT**

Nanotechnology, creating the basis for the production of various nanomaterials, has been denoted the next industrial revolution. Nanomaterials are commonly defined as having at least one size dimension within the range of 1 to 100 nm, whereas nanoparticles have all their size dimensions within this range. A large number of applications for nanomaterials, ranging from medicine to energy, are expected to emerge within this field and societal flows of these materials are projected to increase in the future. In this chapter, existing studies on the societal metabolism in terms of flows and stocks of nanomaterials are reviewed with the aim to investigate current knowledge and methodological practices. The review particularly focuses on which methods that were used, which nanomaterials were studied, the magnitude of quantitative results, and knowledge gaps. Suggestions for future research are then provided based on the identified knowledge gaps.

The methods material, substance and particle flow analysis (MFA, SFA and PFA) have been used to assess flows of nanomaterials with a focus on estimating emissions. A number of studies were identified from the review that applied these methods to varying extents. Fourteen different nanomaterials were covered in the reviewed studies. Titanium dioxide, silver, carbon nanotubes, and zinc oxide are most frequently occurring in the studies. The maximum current global production of nanomaterials according to the reviewed studies is about 300 thousand metric tons per year. In addition, about 10 million tons of the nanomaterial carbon black is produced each year. In general, the reviewed studies of the societal metabolism of nanomaterials are characterized by limited information about nanomaterial production data and environmental emissions of nanomaterials. Furthermore, knowledge related to the societal transformation and waste handling of these materials is scarce.

Suggestions for future research include conducting additional case studies, both for MFA, SFA and PFA, as well as developing approaches for considering future flows and stocks of nanomaterials. Such approaches could potentially facilitate informed decision-making towards a resource-efficient and environmentally benign societal metabolism of nanomaterials. Nanomaterial flows do consist of very small entities, and the current flows of nanomaterials are very small compared to those of many conventional materials used in society. Nevertheless, the future flows of nanomaterials might not remain as small.

**Keywords:** nanomaterials, nanoparticles, material flow analysis (MFA), substance flow analysis (SFA), particle flow analysis (PFA)

## 1. INTRODUCTION

The societal metabolism of materials has varied throughout the history of mankind. First, materials such as stone and wood were used in forms that were relatively unaltered from how they are found in nature. Later, metals such as copper, bronze, iron and steel emerged as important parts of the societal metabolism. Wood and metals continue to play major roles in society, and so do different types of ‘stone,’ or ceramic materials, for example in the form of cement. In the 1900s, plastic made from fossil resources began to replace some conventional materials, and also enabled completely new material applications. In the 2000s, a new type of materials has begun to be produced in society – nanomaterials. In previous changes of the societal metabolism, new material types have typically had other chemical compositions than existing materials. For example, wood consists mainly of carbon hydrates such as cellulose, whereas plastics mainly consist of different hydrocarbons. For nanomaterials, however, it is not a new chemical composition that makes these materials different from existing materials. Rather, it is their physical forms that set them apart. Although nanomaterials have been used historically, for example to dye ceramic materials in medieval times (Erhardt, 2003), these uses were comparatively small and without detailed knowledge about the physical form of the material.

A number of different attempts have been made to define nanomaterials in order to unambiguously differentiate them from other chemical substances and materials. Commonly, nanomaterials are defined as having at least one size dimension within the range of 1 to 100 nm, while nanoparticles have all their dimensions within this range (Foss Hansen et al., 2007). In addition, it is sometimes suggested that to be counted as a nanomaterial, the material must have properties different from those of the bulk form of the same chemical substance (Foss Hansen et al., 2007). A detailed review of different definitions of nanomaterials, with a focus on regulation, has been conducted by Lövestam et al. (2010).

One way of categorizing nanomaterials is on the basis of physical shape (Foss Hansen et al., 2007). This includes three main categories: (1) Bulk nanomaterials, (2) nanomaterials that constitute surfaces, and (3) nanoparticles (Figure 1). The nanoparticles in turn can be in several forms: Airborne, surface-bound, or suspended in a liquid or a solid. In addition to this, Jiang et al. (2009) presented a more detailed typology for nanoparticles, which includes free particles, agglomerates held together by van der Waals bonds, and aggregates (or sintered particles) held together by covalent bonds. Another way to categorize nanomaterials is on the basis of their chemical composition. An often-used typology includes the following

categories: (1) Carbon nanomaterials, such as fullerenes and carbon nanotubes (CNT), (2) metal oxide nanomaterials, such as titanium dioxide (TiO<sub>2</sub>) and zinc oxide (ZnO), (3) metal nanomaterials, such as silver (Ag) and iron (Fe), and (4) others, such as quantum dots (QD) and nanoclays (Ju-Nam and Lead, 2008; Ma et al., 2010). Furthermore, nanomaterials can be either natural or man-made. This later type of nanomaterials is often said to be manufactured (Klaine et al., 2008) or engineered (Colvin, 2003).

The societal flows of nanomaterials have increased considerably in recent years, and are expected to increase even more in the future (Keller and Lazareva, 2014). Furthermore, a large number of applications for nanomaterials, ranging from medicine to energy, are expected to emerge (Keller and Lazareva, 2014). According to the most comprehensive database on consumer products containing nanomaterials, there are currently more than 1600 products containing nanomaterials on the market, and the number has increased notably between 2006 and 2013 (Project on emerging nanotechnologies, 2013; Vance et al., 2015). To the extent that the number of products provides an indication, the societal metabolism of nanomaterials thus seems to be increasing.

The aim of this study is to investigate the current knowledge and methodological practices related to the societal metabolism of nanomaterials. This is done by reviewing existing studies on flows and stocks of these materials with a focus on which methods were used, which nanomaterials were studied, the magnitude of quantitative results, and identified knowledge gaps. Based on the identified knowledge gaps, suggestions for further research are provided. First, however, a description of three methods for analysing the societal metabolism is provided: Material, substance and particle flow analysis (MFA, SFA and PFA).

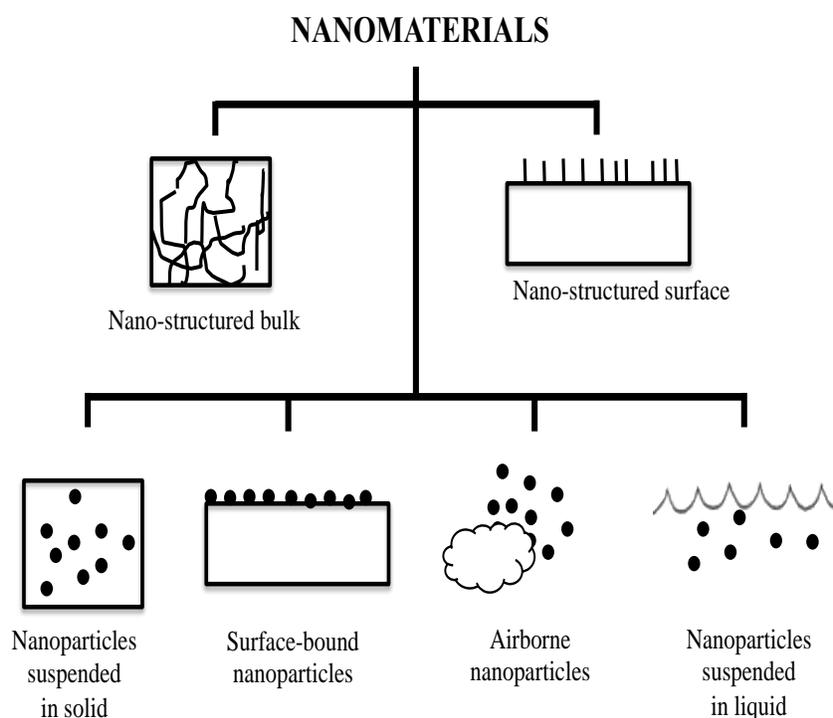


Figure 1. Typology for nanomaterials based on physical shape. Adopted by permission from Arvidsson (2012) and originally modified from Foss Hansen et al. (2007).

## 2. METHODS FOR ANALYSING SOCIETAL METABOLISM

### 2.1. Historical Background

Industrial ecology strives to develop methods to support the restructuring of the economy into a sustainable system (Brunner and Rechberger, 2004). Therefore, understanding the structure and performance of the societal metabolism, that is exploring the flows of energy and matter through the economy (Brunner and Rechberger, 2004; Fischer-Kowalski and Hüttler, 1998), is at the core of industrial ecology (Bringezu and Moriguchi, 2002).

Analogues have been drawn between the societal metabolism and the ecosystem (Husar, 1994). The ecosystem constitutes a desirable metaphor for the description of the anthropogenic system and human-induced material flows due to the demonstrated capability of the natural system to have a sustained development compared with many anthropogenic systems. The ecosystem consists of flows of materials and energy in the environment from producers, via consumers, to recyclers (Husar, 1994). In an ecosystem, plants have the function of producers. Animals then consume a fraction of what is produced and microorganisms are responsible for the recycling. Comparing the ecosystem with an anthropogenic system, the production of materials is instead conducted through different manufacturing processes. Then humans consume the main part of what is produced and finally recycling processes, such as wastewater treatment and metal recycling, take place in some cases.

According to Husar (1994), the main difference between the ecosystem and the anthropogenic system is that the latter is lacking efficient recyclers. This implies that human-induced flows might lead to the build-up of concentrations of unwanted materials in the environment, which leads to pollution. Furthermore, there might be a very large physical separation between the producers, consumers and recyclers in an anthropogenic system. Global trade flows of oil provide an example of an anthropogenic system with this potentially very large physical separation (Husar, 1994). Natural cycles, such as the cycles of water and carbon, typically show cyclical behaviours, while this is scarcely the case for anthropogenic systems (Brunner and Rechberger, 2004).

The analysis of the societal metabolism in the fields of resource conservation and environmental management using variants of MFA goes back to the 1970s (Brunner and Rechberger, 2004). In the beginning, MFA-type methods were mainly used to analyse the metabolism of cities. Today, the application of MFA and similar methods is widespread and they are used in many different fields, including waste handling, wastewater treatment, product design, and life cycle assessment (Brunner and Rechberger, 2004). In the 1970s, it also became evident that quantifying bulk flows by MFA was not enough for a successful waste management, since such effort tended to implicate common waste handling processes such as incineration and landfilling (Brunner and Ma, 2009). This was problematic, since incineration and landfilling only relocated the problematic substances to another place or phase. For example, incineration of toxic wastes caused damage on vegetation through emissions of toxic combustion emissions (Brunner and Ma, 2009). The inability of such processes to solve the problem of increasing amount of toxic wastes highlighted the need for the consideration, control and direction of specific substances into proper treatments and sinks. This insight fostered the development of SFA, which is a special type of MFA were

specific substances are in focus. The SFA method has, for example, been used to investigate flows of heavy metals connected to incineration and contributed to the design of air pollution systems (Brunner and Ma, 2009).

The MFA and SFA methods, which are described in the two following sections, are similar in many ways. They are both based on the same principle of mass conservation (Lavoisier, 1789), and both methods are commonly used in the analysis of societal metabolism. However, their specific applications differ (van der Voet, 2002). MFA typically studies bulk flows of, for example, plastics and glass, and provide macroeconomic indicators. SFA usually provides information about environmental problems related to emissions of chemical substances, such as heavy metals and nutrients.

## 2.2. Material Flow Analysis (MFA)

When flows and stocks of materials are analysed from a systems perspective, the assessment is referred to as MFA (Brunner and Rechberger, 2004). The principle of mass balance implies that mass cannot be created or destroyed, but only transformed into other forms. This feature is used to verify MFA results by balancing inputs, outputs and stocks in the different system processes. Thus, no stock or flow can unintentionally be omitted from the analysis. This facilitates a comprehensive inclusion of all flows and stocks related to a process. According to Brunner and Rechberger (2004), it is this possibility to verify the results through the principle of mass balance that makes MFA so attractive as decision-support in environmental management. There exist a number of specific terms that are commonly used in MFA and some of these are described in Table 1.

**Table 1. Description of some important terms in MFA, based on Brunner and Rechberger (2004)**

<i>Flow</i>	A transfer of mass or energy that can be measured in ton or MJ per year.
<i>Process</i>	Activity related to transformation, such as extraction of metals from mineral ores, transport, such as relocation of products from one geographical point to another, or storage of materials, such as storage in a landfill.
<i>Products</i>	One or several substances that have a positive or negative economic value. Products with positive economic value can for example be cars and wood, while products with negative economic value can be municipal solid waste. Energy can also be a product.
<i>Material</i>	A term that includes both substances and products, but not energy.
<i>Metabolism</i>	Transfer, storage and transformation of materials and energy within a system. Also include exchanges of materials and energy with the system's environment, which is located outside the system's defined boundary.
<i>Stock</i>	A reservoir of materials or energy stored within a system or process, which can be expressed in tons or MJ. If there is accumulation or depletion of materials and energy within the system, the size of the stock increases or decreases, respectively.
<i>Substance</i>	Identical units of materials that are chemical elements, such as cadmium and nitrogen, or chemical compounds, such as ethylene and ammonium.
<i>System</i>	The object of investigation in an MFA. Defined by a group of entities that could include flows, stocks and processes, the interactions among the entities, and the system boundary that distinguishes the system of entities from other systems.
<i>System boundary</i>	Defines the system through a geographical boundary, which might be a region or a country, and a temporal boundary, which might cover one or several years.

Both Bringezu and Moriguchi (2002) and Brunner and Rechberger (2004) provided descriptions of a general MFA procedure. In their descriptions, three steps are identified: Problem and scope definition, quantification of flows and stocks, and presentation and evaluation of results (Figure 2). The steps are briefly explained in this section and further described in relation to SFA in Section 2.3. Although the general procedure of MFA can be described through discrete steps, Brunner and Rechberger (2004) highlighted the need to perform these steps in an iterative manner to assure that the objectives of the study are repeatedly verified and permeate the whole study.

In the first step in Figure 2, the objectives of the study are formulated. The scope of the study is defined, including the geographical, temporal and sometimes functional boundaries. A decision is also made regarding which flows and stocks that should be included and quantified, and for which materials this should be done. The choice of which flows to include is critical for a comprehensive analysis. If important flows are omitted, conclusions regarding which flows that are most important might become misleading. Information about flows and stocks can be provided from the scientific literature, from company or national reports, statistical databases, environmental protection agencies, and other sources. The geographical boundary in an MFA study commonly corresponds to a politically defined region, such as a country or city. The reason for this is that systematically collected data tend to be related to such regions. In addition, political and administrative stakeholders can often be more easily identified within these regions. Their interest in implementing recommendations from an MFA might be larger if those recommendations are related to the stakeholders' geographical area of jurisdiction. The temporal boundary in many MFA studies is one year, but if changes in stocks are considered, a longer time period is often employed.

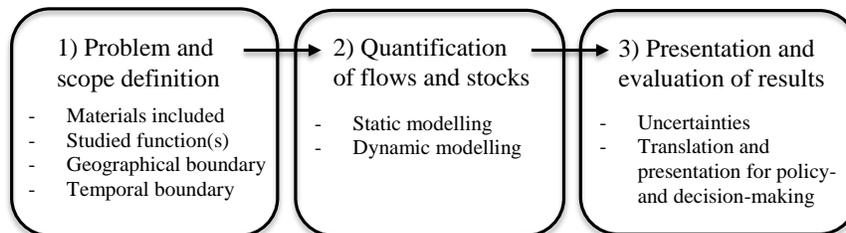


Figure 2. Three general steps in material flow analysis (MFA) based on Bringezu and Moriguchi (2002) and Brunner and Rechberger (2004).

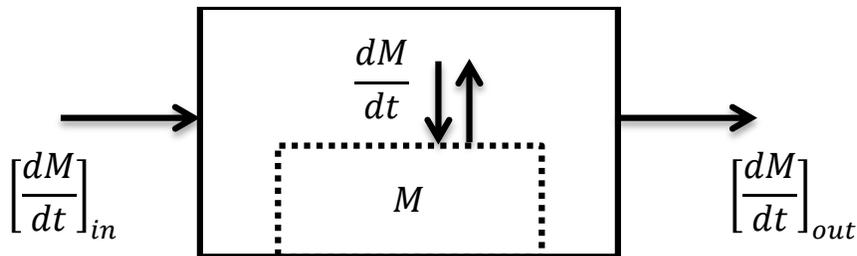


Figure 3. Illustration of flows of material  $M$  in and out from a system and the relation between the material stock and the change in the stock of  $M$ , either through accumulation or depletion. The figure is based on Brunner and Rechberger (2004).

In the second step in Figure 2, flows and stocks related to the defined processes in the system are quantified. The change in the stock of a material, which occurs either by accumulation or depletion, can be calculated as:

$$\frac{dM}{dt} = \left[ \frac{dM}{dt} \right]_{in} - \left[ \frac{dM}{dt} \right]_{out}$$

where  $M$  represents a material stock and  $t$  is the time.  $dM/dt$  thus represents a material flow or a change in a material stock. Figure 3 provides a graphical description of these terms.

The size of the stock of material  $M$  at the time  $t_1$  can be calculated based on a reference time  $t_0$  according to:

$$M(t_1) = \int_{t_0}^{t_1} \left[ \frac{dM}{dt} \right]_{in} dt - \int_{t_0}^{t_1} \left[ \frac{dM}{dt} \right]_{out} dt + M(t_0)$$

The quantification of flows and stocks can be conducted by using static or dynamic modelling (see further Section 2.3).

In the third step in Figure 2, the quantified flows and stocks from the MFA study are evaluated and presented. Uncertainties in information about flows and stocks can be handled and evaluated by using mathematical and statistical tools. There are two crucial audiences for an MFA study. Firstly, there are the technical experts on MFA methods. Secondly, there are the stakeholders governing policy- and decision-making processes, who probably are inexperienced with the MFA terminology and procedure.

### 2.3. Substance Flow Analysis (SFA)

In SFA, the flows and stocks of one or many substances in and out of a defined system are quantified with the aim to provide information that can support environmental management strategies for these substances (van der Voet, 2002). According to van der Voet (2002), there is no standardisation of the SFA method either, but there are some steps that such an analysis generally contains: Definition of the system, quantification of flows and stocks, and interpretation of results. These steps are very similar to the ones shown in Figure 2 for MFA. The framework provided by van der Voet (2002) is further described in this section.

In the first step, the system is defined with regard to space, time, functions, and substances. The system definition determines which processes, stocks and flows that are included. A flowchart can be drawn based on this information to provide a conceptual model of the studied system. Flows and changes in stocks are often in focus in SFA studies, while the total sizes of stocks are commonly excluded due to limited information. Commonly a region, such as a country, is analysed and all flows of the studied substance within this geographical boundary is analysed regardless of the substance's function. SFA can provide information about substances' contribution to specific pollution issues. Heavy metals, such as

lead and mercury in the study by Månsson et al. (2009), and nutrients, such as phosphorous in the study by Brunner (2010), belong to the most commonly studied substances in SFA.

In the second step, flows and stocks are quantified. The required information is identified, data is collected, and the system is modelled. The change in the stock of a substance, which is either by accumulation or depletion, can be calculated as:

$$\frac{dS}{dt} = \left[ \frac{dS}{dt} \right]_{in} - \left[ \frac{dS}{dt} \right]_{out}$$

where  $S$  represents a substance stock and  $t$  is time.  $dS/dt$  thus represents a substance flow or changes in a substance stock. The size of the stock of substance  $S$  at the time  $t_1$  can be calculated based on a reference time  $t_0$  according to:

$$S(t_1) = \int_{t_0}^{t_1} \left[ \frac{dS}{dt} \right]_{in} dt - \int_{t_0}^{t_1} \left[ \frac{dS}{dt} \right]_{out} dt + S(t_0)$$

van der Voet (2002) differentiated between static and dynamic modelling. Static modelling provides steady-state relations between flows and stocks, and thus describes the current societal metabolism. A set of linear equations describes the inter-dependence of different flows and stocks. In dynamic modelling, the variation in stocks with time is included and not only changes in stocks at a steady state. Since time is included as a modelling parameter, dynamic modelling is suitable for scenario analysis and predictions of future stocks. Information about the magnitude of stocks can serve as early warning signals for future emissions. Within dynamic modelling, there are mainly two approaches that are used to estimate emissions from stocks: The leaching model and the delay model. A specified fraction of the current stock contributes to the generation of emissions in the leaching model. In the delay model, the emissions from stocks are estimated based on information about past input into the economy and its residence time.

Finally, in the third step, the results in terms of quantified flows and stocks are interpreted. This can be done both by evaluating the robustness of the results and by translating them into policy-relevant terms. Interpreting results and making them available for policy-makers are important for the provision of knowledge support in environmental management.

## 2.4. Utility and Limitations of MFA and SFA

The main utility of MFA and SFA studies is the provision of support for effective resource and emission management (Brunner, 2012). This includes support for strategic and operational decisions linked to waste management (Brunner and Ma, 2009), and the identification of key processes and important sources of emissions (Brunner, 2010; Månsson et al., 2009). The knowledge and acknowledgement among policy-makers of the role of societal flows and stocks as initiators of environmental problems are gradually increasing (van der Voet, 2002). In addition, goal-oriented and effective waste management is hardly

possible without knowledge about the flows through waste management (Brunner and Ma, 2009).

The utility of MFA and SFA results depends on their effective translation into policy-relevant terms, which can be done by defining indicators (van der Voet et al., 1999; van der Voet, 2002). Such indicators reduce the complexity of MFA and SFA results by aggregating large sets of information into a limited set of measures relevant for environmental policy. Examples of indicators include domestic extraction of fossil fuels and minerals, domestic waste production, total material requirement (TMR), material input per service unit (MIPS), resource efficiency, and emissions to the environment. According to van der Voet (2002), it is important to be aware of the limitations of SFA, and interpretations of the results must not go beyond the system definition. For example, problem-shifting to other substances or changes in energy use are not considered in SFA studies where only one substance is examined (van der Voet et al., 1999).

The utility of some SFA studies conducted in the Stockholm region in Sweden are discussed by Månsson et al. (2009). The authors evaluated the impact that these studies had on environmental policy for the urban management of hazardous substances. The SFA studies mainly considered metals such as lead (Pb) and mercury (Hg) but also some organic substances. The utility was evaluated by examining the references to the SFA studies in environmental policy documents. Månsson et al. (2009) concluded that the availability of empirical data, the focus on emissions, and mapping of their sources were some aspects that explained the high utility of many of the SFA studies in the Stockholm region. Furthermore, the studies were more utilized if information about diffuse emissions was provided and if emissions sources were related to control measures. The involvement of the users of the information from the SFA studies also affected the utility.

## 2.5. Particle Flow Analysis (PFA)

It has been suggested that mass does not describe the flows of nanoparticles in an adequate way (Handy et al., 2008). The reason for this is that the number – and not just the mass – of nanoparticles that is emitted might have implications for the environmental fate and exposure (Praetorius et al., 2012). Depending on their size, there might be very different numbers of nanoparticles in mass flows of similar magnitude. Recognizing the need for an assessment method adapted to nanoparticles, the method of PFA was developed by Arvidsson et al. (2012). PFA estimates emissions of nanoparticles from society to the environment and is a modified form of SFA where particle number is used as the flow and stock metric instead of mass.

It is not only the utilisation of particle number as flow metric that differentiates PFA from MFA and SFA. Although the mass of nanoparticles is conserved in PFA, same as in MFA and SFA, the particle number might not always be conserved. Processes such as agglomeration, dissolution, melting, grinding, and weathering might increase or decrease the number of particles. This has been included in the PFA method in the form of a sink/source term. The change in the particle stock of a nanoparticulate material can thus be calculated as:

$$\frac{dP}{dt} = \left[ \frac{dP}{dt} \right]_{in} - \left[ \frac{dP}{dt} \right]_{out} + P_s$$

where  $P$  represents particle number stock,  $t$  is the time,  $dP/dt$  is a particle flow or change in particle stock, and  $P_s$  is the source/sink term. The parameter  $P_s$  can be a source or a sink depending on its sign (positive or negative). A sink is thus seen as a negative source, and is analogous to loss functions used in environmental fate modelling of chemicals, such as biodegradation (van der Meent and de Bruijn, 2007). Such sinks and sources could be present in any type of process throughout the particle flow, including raw material extraction, production, use, and waste handling.

It is possible to go between mass and particle number. This can be done in a proxy fashion by using the average diameter  $d$  of the particles:

$$P = \frac{6m}{\rho\pi d^3}$$

where  $P$  is again a particle number stock,  $m$  is the mass,  $\rho$  is the particle density, and  $d$  is the average particle diameter. Note that this equation is only valid for spherical particles, and for cases where the particle size distribution is relatively narrow. If the distribution is wider, more detailed knowledge about the size distribution is required, together with a model that integrates over the particle size range. For other shapes than spherical, more complex models are required. PFA is a much more recent method than MFA and SFA, and has until now only been applied in two case studies. The future utility and limitations of this method is therefore an issue for future investigation.

### 3. MFA, SFA AND PFA STUDIES OF NANOMATERIALS

In this section, existing MFA, SFA and PFA studies of the societal metabolism of nanomaterials are reviewed. Twelve studies that explore flows of nanomaterials using MFA, SFA and PFA were identified. Three different approaches for analysing the societal metabolism of nanomaterials can be distinguished in these studies. The approaches are presented in this section together with information about the included nanomaterials and the results of the studies. In total, fourteen different nanomaterials were identified: Silver (Ag), aluminium oxide ( $Al_2O_3$ ), carbon black (CB), carbon nanotubes (CNT), cerium oxide ( $CeO_2$ ), copper and copper oxides (Cu and Cu oxides), copper carbonate ( $CuCO_3$ ), fullerenes, iron and iron oxide (Fe and Fe oxides), nanoclays, quantum dots (QD), silicon dioxide ( $SiO_2$ ), titanium dioxide ( $TiO_2$ ) and zinc oxide (ZnO). Knowledge gaps identified in the reviewed studies are also described in this section. Note that some of the reviewed studies are environmental risk assessment (ERA) studies in which MFA or SFA is conducted as part of the study in order to estimate emissions of nanomaterials. Only ERA studies of nanomaterials that include a detailed modelling of the societal metabolism have been considered in this review.

### 3.1. Approaches for Analysing the Societal Metabolism of Nanomaterials

In the reviewed literature about the societal metabolism of nanomaterials, three different approaches could be identified. The approaches are described in the following sections, with references to the respective studies (Table 2). Although some studies have used the term SFA (Mueller and Nowack, 2008) and MFA explicitly (Gottschalk et al., 2015), the collapsed term MFA/SFA is used in this section. This is because most of the studies do not differentiate between MFA and SFA, and sometimes it is not obvious if the studied nanomaterial should be regarded as a material or a substance. Indeed, as described in Section 2, there are many similarities between these two methods, to the extent that they can be seen as two variants of the same method.

**Table 2. Summarized description of the identified studies of the societal metabolism of nanomaterials. The studies are divided into three approaches: The MFA/SFA approach, the limited MFA/SFA approach, and the PFA approach. In relation to temporal boundaries, ‘current’ refers to the approximate year at which the study was conducted, whereas ‘future’ refers to some prospective scenario. The fourteen nanomaterials included in the reviewed studies are: Silver (Ag), aluminium oxide (Al<sub>2</sub>O<sub>3</sub>), carbon black (CB), carbon nanotube (CNT), cerium oxide (CeO<sub>2</sub>), copper and copper oxides (Cu and Cu oxides), copper carbonate (CuCO<sub>3</sub>), fullerenes, iron and iron oxide (Fe and Fe oxides), nanoclays, quantum dots (QD), silicon dioxide (SiO<sub>2</sub>), titanium dioxide (TiO<sub>2</sub>) and zinc oxide (ZnO)**

Study	Nanomaterials	Specific applications (functions) studied	Geographical boundaries	Temporal boundaries
<b>MFA/SFA approach</b>				
Mueller and Nowack (2008)	Ag, CNT, TiO <sub>2</sub>	Eleven applications including textiles, cosmetics, plastics, electronics, etc.	Switzerland	Current
Gottschalk et al. (2009)	Ag, CNT, fullerenes, TiO <sub>2</sub> , ZnO	Fifteen applications including cosmetics, plastics, consumer electronics, etc.	United States, Europe, Switzerland	Current and future
Keller et al. (2013)	Ag, Al <sub>2</sub> O <sub>3</sub> , CeO <sub>2</sub> , CNT, Cu+Cu oxides, Fe+Fe oxides, nanoclays, SiO <sub>2</sub> , TiO <sub>2</sub> , ZnO,	Sixteen applications including catalysts, coating, paints, pigments, cosmetics, electronics and optics, textiles, etc.	Global	Current
Keller and Lazareva (2014)	Ag, Al <sub>2</sub> O <sub>3</sub> , CeO <sub>2</sub> , CNT, Cu+Cu oxides, Fe+Fe oxides, nanoclays, SiO <sub>2</sub> , TiO <sub>2</sub> , ZnO,	Twenty applications including cosmetics, pigments, coatings, paints, etc.	Global, eight world regions, United States, San Francisco Bay Area	Current
Sun et al. (2014)	Ag, CNT, fullerenes, TiO <sub>2</sub> , ZnO	Thirty two applications including cosmetics, paints, textiles, catalysts, etc.	Europe, Switzerland	Current

**Table 3. (Continued)**

Study	Nanomaterials	Specific applications (functions) studied	Geographical boundaries	Temporal boundaries
<b>MFA/SFA approach</b>				
Caballero-Guzman et al. (2015)	Ag, CNT, TiO <sub>2</sub> , ZnO	Thirty applications including cosmetics, paints, textiles, catalysts, etc.	Switzerland	Current
Gottschalk et al. (2015)	Ag, CB, CeO <sub>2</sub> , CNT, CuCO <sub>3</sub> , TiO <sub>2</sub> , QD, ZnO	Thirty five applications including catalysts, paints, cosmetics, wood preservation, tires, polymer composites, imaging, etc.	Denmark	Current (except for CuCO <sub>3</sub> for which a future scenario was created)
Sun et al. (2015)	Ag, CNT, fullerenes, TiO <sub>2</sub> , ZnO	Thirty six applications including cosmetics, paints, composites, etc.	South Australia	Current
<b>Limited MFA/SFA approach</b>				
Robichaud et al. (2009)	TiO <sub>2</sub>	Not specified	United States	Current and future
Hendren et al. (2011)	Ag, CeO <sub>2</sub> , CNT, fullerenes, TiO <sub>2</sub>	Not specified	United States	Current
<b>PFA approach</b>				
Arvidsson et al. (2011)	Ag	Wound dressings, textiles, electronic circuitry	Global	Current and future
Arvidsson et al. (2012)	TiO <sub>2</sub>	Sunscreen, paint, self-cleaning cement	Global	Current and future

### 3.1.1. The MFA/SFA Approach

The first approach identified is the MFA/SFA approach. In the studies using this approach, flows of nanomaterials and in particular flows of nanomaterials to the environment (emissions) are estimated from a material or substance flow perspective.

In the study by Keller et al. (2013), the releases of engineered nanomaterials from manufacturing, use and disposal into landfill, water, air and soil were estimated. The intermediate steps of wastewater treatment plants and waste incineration plants were considered. The engineered nanomaterial flows from these plants into sludge, air and effluent were also studied. Lower and higher estimates for the global annual production of nanomaterials were provided based on interviews and surveys from a market study involving manufacturers. Keller et al. (2013) assumed that all engineered nanomaterials will eventually become dissipated and either reach the environment or end up in landfills. Recycling was thus not considered. Stocks of engineered nanomaterials in society are assumed to be negligible. Lower and higher coefficients quantifying the release of engineered nanomaterials from manufacturing and the use phase of different applications were generic for all engineered nanomaterials. Release factors of engineered nanomaterials from waste incineration and wastewater treatment were also generic for all the nanomaterials.

In the study by Keller and Lazareva (2014), releases of engineered nanomaterials were estimated for a number of geographical regions: Global, regional (eight world regions), the

United States, all states in the United States, and the San Francisco Bay area in California. The study was conducted by the same group as also conducted the study by Keller et al. (2013) and was in part based on the same data. The lower and higher global production estimates used by Keller et al. (2013) were converted into regional, national and local estimates by Keller and Lazareva (2014). The conversion was conducted by population-weighted national development proxies, which took into consideration the varying capability to purchase engineered nanomaterial products. Then, the release of engineered nanomaterials for different geographical boundaries was estimated by considering regional differences in waste handling.

Material flows of some nanomaterials have also been modelled in a number of studies conducted by a research group in Zurich, Switzerland. In the study by Mueller and Nowack (2008), nanomaterial emissions to the environment were estimated based on an SFA study for Switzerland. The same group has later conducted numerous additional studies, containing various methodological improvements, such as the application of Monte Carlo simulations as a strategy to consider uncertainty (Gottschalk et al., 2009; Sun et al., 2014; Gottschalk et al., 2015). The study by Gottschalk et al. (2009) furthermore conducted a prospective assessment of flows of engineered nanomaterials based on the presumed market evolution of products including these materials. Gottschalk et al. (2015) also created a future scenario for one nanomaterial. The future scenario was based on the assumption of a widespread use of micronized  $\text{CuCO}_3$  in wood protection, substituting the current use of non-micronized  $\text{CuCO}_3$  in Denmark. In the study by Caballero-Guzman et al. (2015), flows of nanomaterials are modelled specifically for waste handling systems, such as recycling, landfill, and combustion. Sun et al. (2015) estimated emissions of nanomaterials to soils and water based on substance or material flow modelling and considered spatial and temporal variability.

### ***3.1.2. The Limited MFA/SFA Approach***

The second approach that has been identified is the limited MFA/SFA approach. This approach is called limited because it only focuses on estimating production rates and does not consider the entire societal metabolism of the nanomaterials. Although the studies using this approach only provide quantitative results for the production flow, the results are discussed and put into an MFA/SFA context. Using this approach, nanomaterial production is estimated as an upper limit for emissions to the environment, without considering specific applications but rather providing numbers for total production (Hendren et al., 2011; Robichaud et al., 2009). This information constitutes a basis for further emission assessment.

Robichaud et al. (2009) estimated the United States' production of  $\text{TiO}_2$  nanomaterials based on information from patents, academic publications, and company interviews. The current bulk material production was then assumed as an upper limit for future nanomaterial production. A description of how the share of the total production of  $\text{TiO}_2$  that is in the nano-form could increase in the future until 2025 was provided. By 2025, they assumed that all  $\text{TiO}_2$  produced would be in the nano-form. The study by Hendren et al. (2011) estimated the United States' production of nanomaterials based on information about the production volume of companies and their production capacities. Proxy indicators, such as the number of employees and annual revenue, were used to describe the size of companies. This information was used to conduct extrapolations of the known production of one company to other companies for which information about nanomaterial production was not available.

### **3.1.3. The PFA Approach**

Arvidsson et al. (2011) and Arvidsson et al. (2012) analysed the societal metabolism of nanomaterials using PFA. This approach is the only one of the four identified from the literature review that used particle number as flow metric, rather than mass. The PFA method, together with the reasons behind the development of this method and the motives behind the usage of particle number as flow metric instead of mass, is described in Section 2.5. In the PFA method, flows and stocks of nanoparticles are analysed and quantified in the unit particle number. However, the studies by Arvidsson et al. (2011) and Arvidsson et al. (2012) focused only on the use phase and emissions from this phase. The reason for this was the lack of information related to the production and waste handling phases. Furthermore, both a current situation and a future, explorative scenario, were considered in order to account for potential future technology diffusion.

## **3.2. Nanomaterials Included**

In total fourteen nanomaterials are included in the reviewed studies of the societal metabolism of nanomaterials (Table 2). Estimations of annual global production rates are provided in Table 4, since they are indicative of the magnitude of the societal metabolism of the nanomaterials. A summary of qualitative assessment results from the studies for specific nanomaterial groups is provided in the following sections. The nanomaterials are divided into groups based on their chemical composition: Metal oxide nanomaterials, metal nanomaterials, carbon nanomaterials and other nanomaterials. This is in accordance with the commonly used typology presented by, for example, Ju-Nam and Lead (2008) and Ma et al. (2010). Note that few of the reviewed studies differentiate between different physical forms of nanomaterials (see Figure 1).

### **3.2.1. Metal Oxide Nanomaterials**

This type of nanomaterials includes the following ones identified in the reviewed literature:  $\text{Al}_2\text{O}_3$ ,  $\text{CeO}_2$ ,  $\text{CuCO}_3$ , Cu oxides, Fe oxides,  $\text{SiO}_2$ ,  $\text{TiO}_2$  and ZnO. Overall, with the exception of the carbon nanomaterial CB, metal oxide nanomaterials seem to currently dominate the global flows of nanomaterials.

The global releases of  $\text{SiO}_2$ , under the assumption of maximum production and emission rates, were estimated to mainly end up in landfill sinks (Keller et al., 2013).  $\text{SiO}_2$  was found to dominate disposal flows of nanomaterials to landfill globally. Despite the large production and material flows of  $\text{SiO}_2$ , this nanomaterial was only included in two of the reviewed studies (Keller et al., 2013; Keller and Lazareva, 2014).  $\text{TiO}_2$ , on the other hand, is the most frequently studied nanomaterial in the reviewed literature. It is included in all reviewed studies except for one. In the study by Arvidsson et al. (2012),  $\text{TiO}_2$  in the specific applications of paint, sunscreen and self-cleaning cement was analysed using a global boundary. The results suggested that sunscreen was the application with the currently largest emissions of  $\text{TiO}_2$  nanoparticles. This was despite that the quantity of these nanoparticles measured in mass was not the largest. However, when particle number was used as flow metric instead, the flow became larger since the  $\text{TiO}_2$  nanoparticles used in sunscreens were much smaller. The explorative scenario, assuming a high consumption together with a larger population and technology diffusion, suggested that the largest flows and stocks of  $\text{TiO}_2$

nanoparticles in the future could instead be related to the application of TiO<sub>2</sub> in self-cleaning cement.

Keller et al. (2013) reported that TiO<sub>2</sub> was one of the engineered nanomaterials with highest release. Most of the global TiO<sub>2</sub> flows were estimated to end up in soil and landfill sinks. Furthermore, a large part of the flow was estimated to pass wastewater treatment plants on their way to these sinks. This situation for the major flows and environmental release of TiO<sub>2</sub> is recognized in many of the other reviewed studies despite different geographical boundaries. The studies of TiO<sub>2</sub> flows in Switzerland (Mueller and Nowack, 2008), in Europe, United States, and Switzerland (Gottschalk et al., 2009), in Europe and Switzerland (Sun et al., 2014), in Denmark (Gottschalk et al., 2015), and in South Australia (Sun et al., 2015) all estimated that these flows of TiO<sub>2</sub> were the most important. Exceptions are found in the studies by Mueller and Nowack (2008), Gottschalk et al. (2009) and Sun et al. (2014), where more minor releases to soils were estimated in the cases for Switzerland. The comparatively smaller share of TiO<sub>2</sub> release to soil was described in these studies as a consequence of the prohibition to apply sludge from wastewater treatment plants to soil (or directly to landfills) in Switzerland. The main part of the TiO<sub>2</sub> flow in Switzerland was estimated to first pass through wastewater treatment plants and then waste incineration plants before reaching its sink in landfills in the studies by Mueller and Nowack (2008), Gottschalk et al. (2009) and Sun et al. (2014).

Notably, two forms of TiO<sub>2</sub> flows were distinguished in this study by Gottschalk et al. (2015): Photostable and photocatalytic TiO<sub>2</sub>. These were analysed separately since the latter is considered to be more toxic. The most important flows of these forms of TiO<sub>2</sub> were to wastewater (for photostable TiO<sub>2</sub> due to its main application in cosmetics), and recycling and landfill sinks (for photocatalytic TiO<sub>2</sub>). Gottschalk et al. (2015) suggested that it is crucial to make the differentiation between photostable and photocatalytic TiO<sub>2</sub> in order to provide an improved foundation for consecutive ERA studies. Furthermore, the study by Sun et al. (2015), considered the changes in stocks of engineered nanomaterials (such as TiO<sub>2</sub>) in South Australian landfills, soils and sediments over a seven year time period. Soil was found to contain the largest stock. Caballero-Guzman et al. (2015) identified waste incineration plants (incineration of plastics, treated wood, and ceramics) and landfills (disposal of construction waste, including paint) as the most important sinks for TiO<sub>2</sub> in the recycling system of Switzerland.

Global releases of Al<sub>2</sub>O<sub>3</sub> have been estimated to mainly end up in landfill sinks (Keller et al., 2013). Fe oxides are assessed together with their non-oxide forms in two of the reviewed studies (Keller et al., 2013; Keller and Lazareva, 2014). According to Keller et al. (2013), global flows of Fe and Fe oxides originate mostly from the applications of electronics and optics, and catalysts, and they will mainly end up in landfills. In addition, Fe oxides were mentioned by Keller et al. (2013) as one of the main nanomaterials used in cosmetics.

ZnO is frequently included in the reviewed studies. Keller et al. (2013) estimated that the largest fraction of ZnO globally is used in cosmetics, medical applications, electronics, optics, coatings, paints, and pigments. The global release of ZnO was estimated to end up in landfills partly via wastewater treatment plants and waste incineration plants in that study. The most important flows for ZnO in the United States were from wastewater treatment to soils due to the application of sludge as fertilizer (Gottschalk et al., 2009). In other studies, considering flows of ZnO in Europe and Switzerland (Sun et al., 2014), in Denmark (Gottschalk et al., 2015), and in South Australia (Sun et al., 2015), the same flows were identified as important.

Generally, main flows entering wastewater was estimated to occur as a consequence of the dominating application of ZnO in cosmetics. However, releases to the environment of ZnO were estimated to be restricted in these other studies since it was assumed that the majority of the ZnO was transformed to non-nanomaterial forms during wastewater treatment. Another study identified waste incineration plants (incineration of painted wood) and landfills (disposal of construction waste) as the most important sinks for ZnO in the recycling system of Switzerland (Caballero-Guzman et al., 2015).

**Table 4. List of nanomaterials identified, information about their annual global production rates, and their main applications are here provided based on data from the reviewed studies (Table 2). The fourteen included nanomaterials in the reviewed studies are: Silver (Ag), aluminium oxide (Al<sub>2</sub>O<sub>3</sub>), carbon black (CB), carbon nanotube (CNT), cerium oxide (CeO<sub>2</sub>), copper and copper oxides (Cu and Cu oxides), copper carbonate (CuCO<sub>3</sub>), fullerenes, iron and iron oxides (Fe and Fe oxides), nanoclays, quantum dots (QD), silicon dioxide (SiO<sub>2</sub>), titanium dioxide (TiO<sub>2</sub>) and zinc oxide (ZnO)**

Nanomaterial	Type of nanomaterial	Annual global production rate [metric ton/year]	Main applications
Ag	Metal	0.1 – 800	Textiles, wound dressings, cosmetics, cleaning agents, paints
Al <sub>2</sub> O <sub>3</sub>	Metal oxide	20 000 – 40 000	Coatings and paints, catalysts
CB	Carbon	10 million	Tires, rubber, pigment
CNT	Carbon	100 – 3000	Composite and polymer additive, Conductive and sensory textiles and fibers
CeO <sub>2</sub>	Metal oxide	6 – 10 000	Catalysts, polishing, electronics and optics
Cu + Cu oxides	Metal + Metal oxide	20 – 200	Catalysts, anti-microbial agent in coatings of plastics and textiles
CuCO <sub>3</sub>	Metal oxide	-	Wood preservatives
Fullerenes	Carbon	0.2 – 200	Composites, cosmetics
Fe + Fe oxides	Metal + Metal oxide	30 000 – 40 000	Pigments in coatings, plastics and cosmetics, catalysts
Nanoclays	Other	9000 – 10 000	Rheological modifier in paints and cosmetics, polymer nano-composites, water treatment
QD	Other	0.6 – 6	Medical imaging devices, solar cells, light emitting devices,
SiO <sub>2</sub>	Metal oxide	50 – 100 000	Filler in rubber and polymer compounds, anti-corrosion and wear-resistant coatings
TiO <sub>2</sub>	Metal oxide	600 – 90 000	Sunscreen, paint and protective coatings, self-cleaning cement
ZnO	Metal oxide	20 – 30 000	UV filter in cosmetics and sunscreen, conductive thin-films, antimicrobial agent in medical and sanitary products

Environmental releases of CeO<sub>2</sub> in Denmark were estimated to be quite limited (below half a tonne per year) in the study by Gottschalk et al. (2015). Keller et al. (2013) estimated

that the global releases of this engineered nanomaterial, under the assumption of maximum production and emission rates, mainly ended up in landfill sinks. Cu oxides were (as for Fe oxides) assessed together with their non-oxide forms in two of the reviewed studies (Keller et al., 2013; Keller and Lazareva, 2014). According to Keller et al. (2013), global flows of Cu and Cu oxides originated mostly from electronics, optics, and catalysts, and they will mainly end up in landfills. In the study by Gottschalk et al. (2015), landfills were identified as important sinks for  $\text{CuCO}_3$  in Denmark. Direct emissions to soil were also considered important for this nanomaterial.

### 3.2.2. *Metal Nanomaterials*

Ag, Cu and Fe are the metal nanomaterials that were identified in the reviewed literature. Overall, metal nanomaterials seem to constitute a smaller share of the global flows of nanomaterials than do metal oxides. Ag, in turn, constitutes a smaller share of the global production of metal nanomaterials, but are more frequently studied in the reviewed studies than are Cu and Fe. Regarding the metal nanomaterials of Cu and Fe, these are assessed together with their oxide forms only in two of the reviewed studies (Keller et al., 2013; Keller and Lazareva, 2014). The results from these studies are presented in Section 3.2 and are therefore not repeated here.

Ag nanoparticles have been increasingly produced for antibacterial purposes in textile applications such as socks, sports apparel and wound dressings. In addition, possibilities to reduce resource demands have motivated the utilisation of Ag nanomaterial ink in electronics. The potential environmental impact from Ag nanoparticles has also been highlighted in literature on nanomaterials risks (Luoma, 2008). Arvidsson et al. (2011) investigated the use phase of Ag nanoparticles and focused on the applications of Ag in textiles, wound dressings, and electric circuitry. The results indicated that the highest emissions of Ag nanoparticles are from textiles, both for the current situation and for a future, explorative scenario. The study also highlighted the contribution from the dissipative use of Ag (for example in textiles) to resource scarcity. Caballero-Guzman et al. (2015) focused on the recycling system of engineered nanomaterials, including Ag in Switzerland. The largest flow of Ag in this system was identified as the incineration of Ag in plastics, filter components, and treated wood.

The study on global flows of engineered nanomaterials by Keller et al. (2013) reported that the majority of Ag flows will end up in landfills and soils. Flows to water and air were estimated to be considerably smaller. In the study by Mueller and Nowack (2008), the most important flows of Ag in Switzerland were estimated to be to landfills through wastewater treatment plants and waste incineration plants. Similarly, the most prominent flows of Ag were considered to be to landfills for Switzerland in the study by Gottschalk et al. (2009). However, when the geographical boundaries of Europe and the United States were studied in the latter study, flows of Ag to soil through the application of sewage sludge were also found to be important. This difference between the considered regions was reported to originate from the different sewage sludge handling in these regions.

The environmental release of Ag nanomaterials was estimated to be rather limited in other studies (Sun et al., 2014; Sun et al., 2015; Gottschalk et al., 2015). The reason for this was that considerable flows entered wastewater, and most Ag was there transformed into non-nano forms of Ag. According to Sun et al. (2014), flows of Ag in Europe and Switzerland instead mainly enter recycling and landfills. The dominating release of Ag to wastewater, where the Ag was largely transformed, originated from the application of Ag in textiles. Sun

et al. (2015) investigated flows of Ag in South Australia. The most important flow for Ag was identified to be to landfills. According to Sun et al. (2015), the reason for this was the dominating applications of Ag in plastics and electronics.

### **3.2.3. Carbon Nanomaterials**

The carbon nanomaterials identified include CB, CNT, and fullerenes. Overall, compared to metal and metal oxide nanomaterials, CNT and fullerenes seem to constitute a smaller share of the global nanomaterial production. CB, on the other hand, seems to be the most produced nanomaterial of all the reviewed ones.

Only one reviewed study included CB (Gottschalk et al., 2015). Among nine investigated engineered nanomaterials in Denmark, CB was the nanomaterial with the highest estimated environmental emissions in that study. This was a result originating from the high production rate and high release from the use phase of CB. The releases to the environment were expected from wear and tear processes of products such as tires containing CB. The estimates were based on the assumption that the total release of CB particles would be in the nano-size range. Gottschalk et al. (2015) admitted that this assumption implied a conservative assessment of the environmental releases of CB.

According to Keller et al. (2013), most of the global release of CNT were expected to go directly into landfills, without passing any wastewater treatment plants or waste incineration plants. In another study of environmental releases of engineered nanomaterials in Switzerland, the most important flows of CNT were estimated to end up in landfills after passing through waste incineration plants (Mueller and Nowack, 2008). Similarly, the most important flows of CNT in the United States estimated in the study by Gottschalk et al. (2009) were found go to landfills through waste incineration plants. Material flows of CNT in Denmark were estimated to mainly end up in recycling processes, waste incineration and landfilling (Gottschalk et al., 2015). Furthermore, the release of CNT to the environment was predicted to be minor. Sun et al. (2014) identified that flows of CNT to recycling and waste incineration plants were the most prominent in Europe and Switzerland. Furthermore, CNT were assumed to be largely eliminated in these processes and thus the environmental releases of CNT were small. The study by Sun et al. (2015) reported that the majority of CNT flows will either end up in landfills or recycling in South Australia as well. This was explained as a consequence of the CNT main application, which was as reinforcement in polymer composites. Caballero-Guzman et al. (2015) focused on the recycling system of engineered nanomaterials, such as CNT, in Switzerland. The largest mass flows in the recycling system of CNT were identified as applications (such as batteries and chips in printed circuit boards) being exported.

The study by Gottschalk et al. (2009) reported that the most important flows of fullerenes go to landfills but also to surface waters in the United States. The flows of fullerenes in Europe and Switzerland to recycling and waste incineration plants were identified as the most important flows by Sun et al. (2014). Fullerenes were, however, considered to be eliminated in these processes, and therefore their environmental releases were estimated to be limited. The study by Sun et al. (2015) for South Australia also showed that the majority of fullerene flows will either end up in landfills or go to recycling. This was because they followed the fate of their main application, which were polymer composites.

#### **3.2.4. Other Nanomaterials**

There exist other types of nanomaterials that are neither metal-, metal oxide-, nor carbon-based. The two examples found in the reviewed literature are nanoclays and QD. Overall, the flows of these other nanomaterials seem to constitute a minor share of the global nanomaterial production.

Concerning nanoclays, most of this nanomaterial was found to end up in landfills in the study by Keller et al. (2013). The nanoclay flows to landfills were further reported to mainly originate from its applications in composites and the automotive industry. The only study among the reviewed ones that included QD was the study by Gottschalk et al. (2015). Almost no environmental release was estimated for QD in that study. This was due to the low production rate of QD together with its application in products with limited release during use.

### **3.3. Knowledge Gaps Identified**

A number of knowledge gaps have been identified in the reviewed literature related to the societal metabolism of nanomaterials, of which many are recurrent in most of the studies. One commonly mentioned knowledge gap is related to the limited information about production of nanomaterials, and particularly to the absence of application-specific production data. Almost all the reviewed studies emphasized high uncertainties in their production estimates. Keller et al. (2013) reported considerable uncertainties in their estimations of emissions related to the data on global production of engineered nanomaterials. Gottschalk et al. (2009) stated that nanomaterial production is a crucial input data, since modelled release of nanomaterials are dependent on production rates. The criticality of information about production for estimations of nanomaterials' exposure were also emphasized by Sun et al. (2014). Hendren et al. (2011) discussed both the scarcity of information related to nanomaterial production and the general inconsistency in existing data sources. According to Hendren et al. (2011), the general lack of information of nanomaterial production was "striking." For example, almost 90% of their data on TiO<sub>2</sub> production had to be extrapolated using proxy indicators. Robichaud et al. (2009), who also faced many difficulties in estimating United States' production of TiO<sub>2</sub>, stated that data were often confidential. Mueller and Nowack (2008) stated that the quality of estimations of environmental exposure of nanomaterials only reflects the quality of input data, which they considered to be very uncertain.

Another identified knowledge gap is related to the release of nanomaterials. Such release depends on parameters such as product life time, the type of incorporation of nanomaterials in products, how much nanomaterials the product contains, and how the product is used (Mueller and Nowack, 2008). Therefore, the release of nanomaterials typically depends on the nature of the products that contain nanomaterials. Notably, the studies by Hendren et al. (2011) and Robichaud et al. (2009) were not application-specific in their estimations of nanomaterial production rates. Information about the production and release from specific applications for nanomaterials is in general limited in the reviewed studies. For example, Arvidsson et al. (2012) and Arvidsson et al. (2011) found that release factors were difficult to obtain or estimate for TiO<sub>2</sub> and Ag for the considered applications.

Another factor that might affect the release of nanomaterials is transformation of these materials during their societal metabolism. This has been highlighted as an important aspect

for improving release estimates for nanomaterials (Keller et al., 2013; Keller and Lazareva, 2014). Studies of the environmental release of Ag and ZnO estimated quite limited emissions if the transformation of these nanomaterials in wastewater was considered (see Section 3.2.1 and 3.2.2). Some other studies did not take the transformation of these nanomaterials into their non-nano forms into consideration, which sometimes led to notably different results. This highlights the importance of transformations of nanomaterials throughout their societal metabolism.

Furthermore, the understanding of waste handling processes of nanomaterials has been highlighted as limited, to the extent that they were excluded in some of the reviewed studies (Arvidsson et al., 2011; Arvidsson et al., 2012). Other studies suffered from the scarcity of information related to nanomaterials during waste handling and recycling (Caballero-Guzman et al., 2015; Sun et al., 2014).

## CONCLUSION

The number of studies concerning the societal metabolism of nanomaterials is limited and so is the number of nanomaterials covered. The nanomaterials of TiO<sub>2</sub>, Ag, CNT and ZnO are the most frequently investigated. The reviewed studies generally suffer from limited information about nanomaterial production rates, the amount of nanomaterials in different products, and the use of these products. These factors have a clear influence on the potential for subsequent releases of nanomaterials. Furthermore, information about waste handling processes of nanomaterials was limited. These limitations manifested themselves in partly varying results in the reviewed studies.

The maximum current total global production of nanomaterials according to the reviewed studies is about 300 thousand metric tons per year (Table 4). In addition, about 10 million tons of CB is produced each year. In order to obtain an understanding of the magnitude of these numbers, they can be compared to the global production of other, conventional materials. For example, the total world production of crude steel was about 1700 million metric tons in 2014 (Worldsteel, 2015), and the world production of plastics were about 310 million tons in 2014 (PlasticsEurope, 2015). Thus, comparing the flows of nanomaterials with the flows of steel and plastics suggests that the societal metabolism of nanomaterials is still comparatively small. However, there might exist a number of hidden nanomaterials that have not yet been investigated from a societal metabolism perspective. Tungsten carbide (WC) might be an example of such a hidden nanomaterial (Bastian et al., 2009). In addition, considering their early state of technological development, the quantities of produced nanomaterials are likely to increase in the future. Which nanomaterials that constitute the top produced ones might also change with time. Several studies acknowledge the need to investigate future releases of nanomaterials due to the likely increase in use of these materials. Such prospective studies could provide useful information not only about the current societal metabolism of nanomaterials, but also about future, potential metabolisms.

Conducting additional case studies for nanomaterials, both for MFA, SFA and PFA, as well as developing approaches for considering future flows and stocks of nanomaterials, are the main recommendations for future research. The employment of these methods is important in order to facilitate informed decision-making towards a resource-efficient and

environmentally benign societal metabolism of nanomaterials. Considering that much of the current flows of nanomaterials seem to end up either in landfills or as emissions to the environment, such efforts are highly motivated. Although nanomaterial flows consist of very small entities, and the current flows of nanomaterials are very small compared to conventional materials used in society, the flows of nanomaterials might not remain as small in the future.

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