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Modeling the fate and end-of-life phase of engineered nanomaterials in the Japanese construction sector

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ABSTRACT

To date construction materials that contain engineered nanomaterials (ENMs) are available at the markets, but at the same time very little is known about their environmental fate. Therefore, this study aimed at modeling the potential fate of ENMs by using the example of the Japanese construction sector and by conducting a dynamic material flow analysis. Expert interviews and national reports revealed that about 3920–4660 tons of ENMs are annually used for construction materials in Japan. Nanoscale TiO_2, SiO_2, Al_2O_3 and carbon black have already been applied for decades to wall paints, road markings or concrete. The dynamic material flow model indicates that in 2016 about 95% of ENMs, which have been used since their year of market penetration, remained in buildings, whereas only 5% ended up in the Japanese waste management system or were diffusely released into the environment. Considering the current Japanese waste management system, ENMs were predicted to end up in recycled materials (40–47%) or in landfills (36–41%). It was estimated that only a small proportion was used in agriculture (5–7%, as ENM-containing sewage sludges) or was diffusely released into soils, surface waters or the atmosphere (5–19%). The results indicate that ENM release predominantly depend on their specific applications and characteristics. The model also highlights the importance of adequate collection and treatment of ENM-containing wastes. In future, similar dynamic flow models for other countries should consider, inasmuch as available, historical data on ENM production (e.g. like declaration reports that are annually published by relevant public authorities or associations), as such input data is very important regarding data reliability in order to decrease uncertainties and to continuously improve model accuracy. In addition, more environmental monitoring studies that aim at the quantification of ENM release and inadvertent transfer, particularly triggered by waste treatment processes, would be needed in order to validate such models.

1. Introduction

Due to the high market potential of nanotechnology there is a worldwide increase in consumer products that contain engineered nanomaterials (ENMs) (Hansen et al., 2016). Thus, ENMs are already used in many sectors, such as the construction, electronic, automobile, cosmetic, biomedical, pharmaceutical, or food industry. Particularly in the construction sector diverse ENMs have sparked increasing interest in application. For example, novel ENM-containing construction materials show beneficial effects regarding durability and mechanical properties. Carbon nanotubes (CNTs), nanoscale SiO_2 or TiO_2 are used in concrete to enhance concrete strength or to obtain concrete surfaces with self-cleaning or photo-catalytic properties (Lee et al., 2010; van Broekhuizen et al., 2011). A case study on the Swiss construction sector showed that nanoscale Ag, CeO_2, Fe_2O_3, SiO_2, TiO_2, ZnO are already used in construction materials in order to provide surface coatings and paints with, for example, antibacterial properties, improved scratch or fire resistance (Hincapié et al., 2015). However, ENM-containing construction materials are still niche products in Europe and very little information is currently available regarding their market share (van Broekhuizen et al., 2011).

Once ENMs are applied to products, environmental release generally depends on how ENMs are incorporated into a matrix (dispersed or bound in bulk materials or used in surface coatings)

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(Yang and Westerhoff, 2014). In addition, ENM release depends on material properties and on environmental conditions (material composition, temperature, prevailing pH, etc.) (Duncan and Pillai, 2015; Froggett et al., 2014). To date, the actual amounts of released ENMs into the environment and their release mechanisms into liquid media are not well known. Regarding the usage phase of construction materials, some experimental studies showed that very small amounts of nanoscale SiO_2 or TiO_2 in paints were detected in leachates after weathering processes (Al-Kattan et al., 2014, 2013). These studies indicate that hardening processes lead to a sufficient, solid incorporation of ENMs and, subsequently, reduces unintentional particle release. On the contrary, Kaegi et al. (2010) showed that more than 30% w/w of nano-Ag was released from façades after one year of exposure. However, during handling, processing, or by other mechanical impacts on construction materials, such as drilling, grinding, or demolishing, ENMs can be released and may lead to worker or environmental exposure (Lee et al., 2010). Furthermore, in the end-of-life phase of ENM-containing construction materials mechanical, thermal or chemical stress during waste treatment and landfilling may cause adverse environmental effects, triggered by air- or waterborne ENM release (Part et al., 2015). In this context, Mueller and Nowack (2008) have presented the first investigations aimed at modeling quantities of ENMs released into the environment from a complete life-cycle perspective. Other (static) fate models indicated that the majority of ENMs are likely to accumulate in waste streams and landfills, highlighting the importance of waste management systems (Caballero-Guzman et al., 2015; Hincapié et al., 2015; Keller and Lazareva, 2014). Dynamic flow models, which consider market dynamics and the lifetime of ENM-containing products, showed that the amounts of released ENMs increase year by year, considering a certain time lag between ENM production and release during their use as well as end-of-life phase (Bornhöft et al., 2016; Song et al., 2017; Sun et al., 2017).

In general, material flow analysis (MFA) is a common method for modeling the environmental fate of materials or substances along their entire life cycle (Nowack et al., 2015). For example, static material flow models were used to describe potential flows of ENMs and their distribution in the anthroposphere (Caballero-Guzman and Nowack, 2016). Moreover, so-called dynamic MFA methods can be used – based upon historical data and lifetime distribution – to estimate and extrapolate future production amounts and market dynamics of a material (Bornhöft et al., 2016; Müller et al., 2014; Song et al., 2017; Sun et al., 2017). But for lack of information on yearly ENM production amounts it is currently very challenging to predict future market dynamics and distribution of ENM-containing products (Vance et al., 2015). However, simplifications as well as a lack of information on production, trade or consumption statistics obviously leads to unavoidable data uncertainties that need to be documented (Brunner and Rechberger, 2004).

This study aimed at applying a dynamic MFA focusing on the use of ENM-containing products that are solely used in the Japanese construction sector. Relevant Japanese Associations have been interviewed to obtain information about production amounts as well as their market shares and dynamics (as input data for the MFA). So-called survival-debris-functions, which are based on historical statistical data on construction activities and which consider the very long life spans for buildings and roads, were used to calculate the outflows of the in-use stock. ENM-specific transfer coefficients, derived from the literature, were used to calculate ENM outflows of waste management processes. The results from dynamic flow modeling show that, in 2016, the majority of ENMs remained in buildings (~95%), whereas small quantities (~5%) ended up in the Japanese waste management system or were diffusely released into the environment.

2. Material and methods

2.1. Modeling approach

In general, a dynamic flow model enables extrapolating and determining temporal changes of material flows and (in-use/end-of-life) stocks within a defined system (Müller et al., 2014). The system boundaries of this dynamic flow model cover the life cycle from use to disposal of ENM-containing construction materials, available in the Japanese market since their year of market penetration. All required modeling steps are explained in more detail in the following sections:

1. Definition of system boundaries and material-specific processes (see Section 2.2)
2. Determination of data base (model input data) (see Section 2.3)
3. Estimation of ENM flows and stocks (see Section 2.4)
4. Determination of ENM distribution and transfer coefficients (see Section 2.5).

Due to the lack of information on ENM market dynamics it was assumed that the production amounts of ENMs remained constant for the whole period of interest. Such linear extrapolation model enabled to calculate accumulating ENM flows and stock data since their year of market penetration. In addition, based upon previous waste management plans it was expected that the status quo of the Japanese construction and demolition (C&D) waste management system did not change significantly during the whole period of interest. So-called survival-debris-functions, which are based on historical statistical data and which consider the very long life spans for buildings and roads, allowed for extrapolation of ENM flows that are generated through the use as well as disposal of construction materials (see also Section 2.4).

2.2. System boundaries and ENM-specific processes

This dynamic MFA focuses on ENMs that are explicitly produced in Japan and solely applied to construction materials at the Japanese market. The system boundaries for the dynamic flow model include the use and disposal phase of ENM-containing construction materials. All ENM flows within the system – i.e. TiO_2, SiO_2, Al_2O_3 and carbon black – are separately calculated and presented for the year 2016. In Japan, ENMs are used as additives in concrete or paints (e.g. for facades or road markings) that are consequently applied for *roads*, *infrastructures*, *wooden* or *non-wooden buildings*. We note that these classifications were based upon statistical data on construction activities in Japan (see Table S1 in the Supplementary material). ENM applications illustrate processes during the use phase of an ENM-containing construction material, by which in-use stocks are generated (from the year of market penetration of an ENM until its end of useful life). Therefore, these generated in-use ENM stocks exist for a certain time depending on the very long life span of an ENM-containing application and, consequently, are temporal sinks.

At the end of its useful life, ENM-containing buildings or roads will be demolished and collected as construction and demolition (C&D) wastes. According to Japanese waste statistics, C&D waste is collected as *cement concrete*, *mixed demolition waste*, *wooden materials* and *asphalt concrete* (MLIT, 2014a). In this dynamic MFA, C&D waste collection and waste treatment (recycling) plants illustrate processes of the end-of-life phase. The output flows of waste treatment plants are either used for recycled materials or are dumped in landfills, whereas wooden and other high calorific ENM-containing residues are further treated in waste incineration plants (MLIT, 2014a). The basic concept for modeling and

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