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Influences of use activities and waste management on environmental releases of engineered nanomaterials

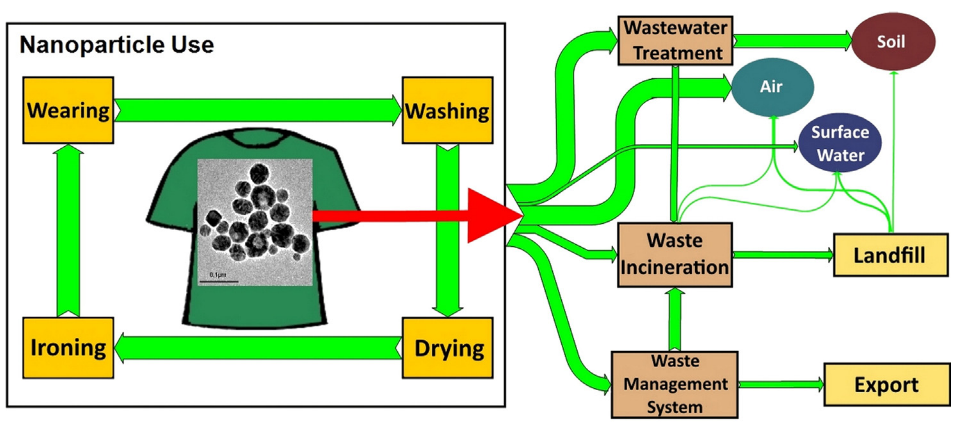
Henning Wigger [a,*], Stephan Hackmann [b], Till Zimmermann [a,c], Jan Köser [d], Jorg Thöming [e], Arnim von Gleich [a,c]

[a] Faculty of Production Engineering, Department of Technological Design and Development, University of Bremen, Badgasteiner Str. 1, 28359 Bremen, Germany
[b] UFT Center for Environmental Research and Sustainable Technology, Department of General and Theoretical Ecology, University of Bremen, Leobener Str., 28359 Bremen, Germany
[c] ARTEC — Research Center for Sustainability Studies, Enrique-Schmidt-Str. 7, 28359 Bremen, Germany
[d] UFT Center for Environmental Research and Sustainable Technology, Department of Sustainable Chemistry, University of Bremen, Leobener Str., 28359 Bremen, Germany
[e] UFT Center for Environmental Research and Sustainable Technology, Department of Sustainable Chemical Engineering, University of Bremen, Leobener Str., 28359 Bremen, Germany

HIGHLIGHTS

- Textile use activities and two waste management systems (WMSs) are investigated.
- Matrix material and use activities determine the ENM release.
- Counter-intuitive shifts of releases to air can happen during usage.
- WMS export can increase by 350% in case of short service life and minimal releases.
- Use and matrix material, ENM, and WMS should be considered in exposure assessment.

GRAPHICAL ABSTRACT



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ABSTRACT

Engineered nanomaterials (ENM) offer enhanced or new functionalities and properties that are used in various products. This also entails potential environmental risks in terms of hazard and exposure. However, hazard and exposure assessment for ENM still suffer from insufficient knowledge particularly for product-related releases and environmental fate and behavior. This study therefore analyzes the multiple impacts of the product use, the properties of the matrix material, and the related waste management system (WMS) on the predicted environmental concentration (PEC) by applying nine prospective life cycle release scenarios based on reasonable assumptions. The products studied here are clothing textiles treated with silver nanoparticles (AgNPs), since they constitute a controversial application. Surprisingly, the results show counter-intuitive increases by a factor of 2.6 in PEC values for the air compartment in minimal AgNP release scenarios. Also, air releases can shift from washing to wearing activity; their associated release points may shift accordingly, potentially altering release hot spots. Additionally, at end-of-life, the fraction of AgNP-residues contained on exported textiles can be increased by 350% when assuming short product lifespans and globalized WMS. It becomes evident that certain combinations

Abbreviations: AgNPs, silver nanoparticles; $COT_{max/min}$, cotton maximal/minimal scenario; DM, dry matter; ENM, engineered nanomaterial; EOL, end-of-life; LCA, life cycle assessment; OS_{Ger}, other silver sources scenario Germany; PEC, predicted environmental concentration; $PES_{max/min}$, polyester maximal/minimal scenario; SL, service life; WMS, waste management system; WIP, waste incineration plant; WWTP, wastewater treatment plant.

* Corresponding author.
E-mail address: hwigger@uni-bremen.de (H. Wigger).

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of use activities, matrix material characteristics, and WMS can influence the regional PEC by several orders of magnitude. Thus, in the light of the findings and expected ENM market potential, future assessments should consider these aspects to derive precautionary design alternatives and to enable prospective global and regional risk assessments.

1. Introduction

Engineered nanomaterials (ENMs) offer new opportunities for several applications due to their improved or novel properties. Consequently, various products have already entered the market. Among these, textiles with silver nanoparticles (AgNPs) (Wijnhoven et al., 2010) represent a particularly controversial ENM application. Besides their promising possibilities, ENMs—as nearly each technological innovation—come with potential risks. To assess such risks, several research studies have been conducted and first indications of potential mechanisms causing adverse effects have been found. Nevertheless, major challenges for determining potential hazards still remain. Moreover, the diversity in ENM characterization protocols applied in experiments and the lack of reference materials complicate predictive hazard estimations. These challenges still hinder the determination of specific physico-chemical properties responsible for the mode of action (Schafer et al., 2013). Even more so, as compared to hazard assessment, only very few environmental exposure assessment studies have been conducted so far (Gottschalk et al., 2013), despite the fact that these are as important in risk assessment as studies on potential hazards. Typically a mass-balance approach with a life cycle thinking approach is used in environmental exposure modeling. The predicted environmental concentration (PEC) in the respective compartment is determined by balancing input and output flows for each process in every life cycle stage. For this purpose, exposure models are primarily based on the allocation of the global ENM production volume and are weighted based on the population or economic power of the geographical area of interest (Hendren et al., 2013). The production volume estimations are mostly based on the first publications in this field: Blaser et al. (2008), Müller and Nowack (2008), Gottschalk et al. (2009), and Piccinno et al. (2012). Furthermore, existing exposure models (e.g., Keller and Lazareva, 2013; Sun et al., 2014) predominantly refer to large geographical areas like the United States, Europe, and less to regional extent. The focal life cycle stage depends also on the goal and scope of the study. Particularly, the end-of-life (EOL) stage gets increased attention, because ENM will likely end up in landfills, wastewater treatment plants (WWTPs), or waste incineration plants (WIPs), as shown by Gottschalk et al. (2009), Keller and Lazareva (2013), and Sun et al. (2014).

In addition to the described challenges in exposure assessment, there is a lack of knowledge regarding production volumes as well as products and fate and behavior of released ENMs. In particular, the latter aspect is rather complex, because ENMs can transform and thus change their properties during their life cycle. Therefore, currently existing fate and behavior models that are widely used for organic chemicals, need to be adapted (Westerhoff and Nowack, 2013). Such a modified approach using partition coefficients (e.g., octanol–water coefficient) can identify the distribution of ENMs and major exposure pathways in risk assessment (Westerhoff and Nowack, 2013). Consequently, several studies focus on the applicability of partition coefficients for ENM to predict their environmental behavior. However, recent findings have highlighted that partition coefficients seem not applicable for ENM and thus alternative methods are needed. This is mainly because the transport and behavior of ENMs in environmental compartments represent a major determinant of exposure (Praetorius et al., 2012; Praetorius et al., 2014; Meesters et al., 2014).

Irrespectively of the kind of finally transformed ENMs, which may be the origin of adverse effects, the strength of the emission source defines the exposure and thus determines the potentially necessary precautionary measures (IPCS, 2004; Kümmerer, 2010). Besides environmental emissions caused through industrial processes, products can be regarded as the main output source (Held, 1991) and are thus related to their environmental release and exposure. Up to now, ENM release studies for products are rare. However, they are urgently needed to determine reliable PECs (Markus et al., 2013). Products that have been considered in field studies published so far include surface coatings, food packaging, sprays, and textiles (Lorenz et al., 2012; von Goetz et al., 2013; Mitrano et al., 2014). Their most substantial findings were that ENM release depends on the incorporation into the product, and that most of the ENMs are likely to end up in wastewater (Lorenz et al., 2012; von Goetz et al., 2013; Mitrano et al., 2014). Due to the scarce data on releases, the majority of the exposure assessment studies have limited the usage to particular activities (e.g., washing) and ignored the product related EOL. Published studies on textiles generally assume *a-priori* total relative releases of ENM to water of 90–95% and 5% to air (Blaser et al., 2008; Müller and Nowack, 2008; Keller and Lazareva, 2013). Only two studies differentiated between relative releases from several products to air, wastewater, surface water, and export (Gottschalk et al., 2009; Sun et al., 2014). Finally, the usage and its influence on the PEC remain an open question and thus need more detailed investigations.

Apart from exposure assessment and the life cycle perspective, most ENMs are embedded in products and the potential environmental release depends on the usage of these products. Therefore, similarities to product life cycle assessment (LCA) are apparent and existing experiences should be considered in environmental exposure assessment as both apply a life cycle thinking perspective using the mass-balance approach (Grieger et al., 2012). For instance, even if the influence of textile use activities on LCA results remains to be validated, existing LCA studies have shown the influence of changing use patterns and the kind of ENM on LCA results (Walser et al., 2011; Laitala and Boks, 2012). User behavior furthermore determines product lifespan and disposal, which influences the subsequent waste treatment and release of ENM during EOL (Laitala and Boks, 2012; Velden et al., 2013). Generally waste management systems (WMSs) depend on national regulations, product waste trading, and product type (Pires et al., 2011). When focusing on regional or local scales (Keller and Lazareva, 2013), the impact of trans-border relations for disposed products becomes more important, especially for low ENM releases during use and short product lifespan. Hence, product type as well as national WMS and related ENM release points are strongly related. However, the actual influence of regional differences regarding use activities, product lifespan, and product-related waste treatment on the PEC remains to be clarified.

Thus far, only little knowledge on potential releases and related risks has been gained, primarily due to the early stage of research and development of nanotechnologies. In this context, our study applies nine different life cycle release scenarios in a preliminary assessment of ENM releases from clothing textiles. In particular, previous studies on clothing textiles have shown controversial results, since very high as well as very low release rates of AgNPs have been reported (Benn and Westerhoff, 2008; Geranio et al., 2009; KEMI, 2012). Thereby our study addresses three relevant aspects. First, it is hypothesized that both product materials and use activities may have a relevant influence on the release points and quantities. To demonstrate this, environmental releases were derived based on ENM literature and reasonable assumptions. Second, with the growing focus on the EOL and the differences of regional

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