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Probabilistic modeling of the flows and environmental risks of nano-silica

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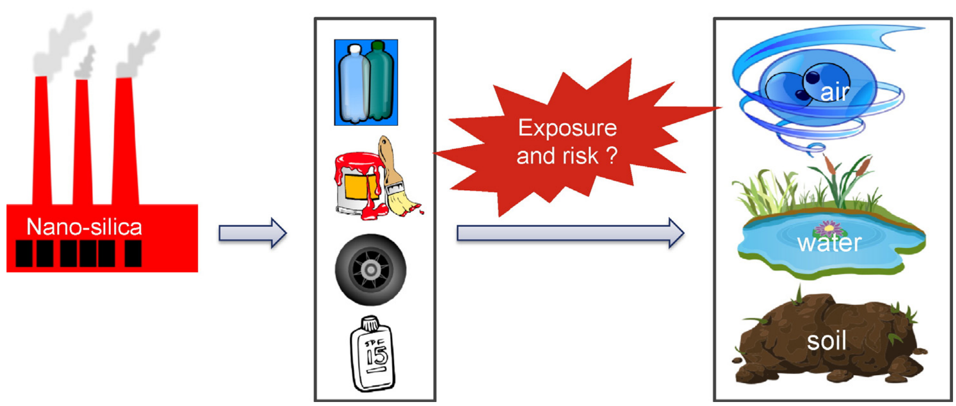
HIGHLIGHTS

- We quantify the exposure of nano-silica to technical systems and the environment.
- The median concentration in surface waters is predicted to be 0.12 μg/L in the EU.
- Probabilistic species sensitivity distributions were computed for surface waters.
- The risk assessment suggests that nano-silica poses no risk to aquatic organisms.

GRAPHICAL ABSTRACT



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ABSTRACT

Nano-silica, the engineered nanomaterial with one of the largest production volumes, has a wide range of applications in consumer products and industry. This study aimed to quantify the exposure of nano-silica to the environment and to assess its risk to surface waters. Concentrations were calculated for four environmental (air, soil, surface water, sediments) and two technical compartments (wastewater, solid waste) for the EU and Switzerland using probabilistic material flow modeling. The corresponding median concentration in surface water is predicted to be 0.12 μg/l in the EU (0.053–3.3 μg/l, 15/85% quantiles). The concentrations in sediments in the complete sedimentation scenario were found to be the largest among all environmental compartments, with a median annual increase of 0.43 mg/kg·y in the EU (0.19–12 mg/kg·y, 15/85% quantiles). Moreover, probabilistic species sensitivity distributions (PSSD) were computed and the risk of nano-silica in surface waters was quantified by comparing the predicted environmental concentration (PEC) with the predicted no-effect concentration (PNEC) distribution, which was derived from the cumulative PSSD. This assessment suggests that nano-silica currently poses no risk to aquatic organisms in surface waters. Further investigations are needed to assess the risk of nano-silica in other environmental compartments, which is currently not possible due to a lack of ecotoxicological data.

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1. Introduction

As a result of the development of nanotechnology, the production of engineered nanomaterials (ENMs) has increased significantly over the

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last decades and numerous applications penetrate almost every aspect of our life. Although nanomaterials benefit our everyday life, they can also have adverse impacts on humans and the environment (Handy et al., 2008). Due to their small size and large specific surface area, nanoparticles are more likely than large-scale materials to penetrate cells, pass through biological barriers and generate free radicals and reactive oxygen species (Krug and Wick, 2011; Nel et al., 2006). Thus it is critical to evaluate human and environmental risks posed by ENMs, especially those with high production volume. This field has received a lot of attention from researchers and regulators (Klaine et al., 2012).

Nano-silica represents silica particles and agglomerates with a size of the primary particles between 1 and 100 nm (Dekkers et al., 2013). Synthetic amorphous silica (SAS) has been produced since the 1950s and has a worldwide production of several million tons (SASSI, 2008). Nano-silica is used in various pharmaceutical products, cosmetics, printer toners and food products because it can provide materials with a desired consistency and prevents separation of various ingredients (Napierska et al., 2010). In addition, it is also widely used in paints, surgical tools, medical equipment, textiles and all sorts of surfaces and substrates due to its high water repellence (Kaiser et al., 2013). With desired surface and internal functionality, nano-silica also has a high potential for application in different biorecognition agents, e.g. antibodies, protein complexes, etc. (Salata, 2004). Furthermore, the silica matrix has a negative charge and can provide a high number of electrostatic binding sites. This property is widely used for drug delivery purposes (Jin et al., 2009).

The production and use of nano-silica unavoidably cause environmental release of it. However, the knowledge about environmental exposure to nano-silica still remains scarce. Currently, only a few release studies are available that have investigated the release of nano-silica from products (Froggett et al., 2014). For example, the release of nano-silica from nanocomposites by three different release mechanisms has been studied (Wohlleben et al., 2011). Nguyen et al. (2012) have reported the release of nano-silica during the irradiation of polymer composites and Al-Kattan et al. (2015) and Zuin et al. (2014) have quantified the release of silica from paint containing nano-silica into water.

To date, there are no analytical methods available to detect trace concentrations of inorganic ENMs such as nano-silica in environmental samples (von der Kammer et al., 2012). Environmental exposure assessment therefore has to depend on modeling approaches to predict the environmental concentrations (Gottschalk et al., 2013b). In the past few years, several models have been developed to assess release and environmental concentrations of ENM (Arvidsson et al., 2012; Boxall et al., 2007; Gottschalk et al., 2009; Keller and Lazareva, 2014; O'Brien and Cummins, 2010; Sun et al., 2014). Some modeling studies have also included nano-silica. Boxall et al. (2007) have modeled the environmental exposure to more than ten ENMs in the UK environment including nano-silica and the concentrations of nano-silica were predicted to be 0.0007 μg/l in water and 0.03 μg/kg in soil, assuming a 10% market penetration of the nano-products. Keller et al. (2013) have predicted a global release of 10,600 metric tons of nano-silica to soil and 2100 metric tons to water in 2010. Furthermore, they have performed a local modeling study to calculate the release of nano-silica in effluents from sewage treatment plants (STP) in California (0.63–53 metric tons/year) and San Francisco bay (120–10,000 kg/year) in 2010 (Keller and Lazareva, 2014). However, the methods used in these models did not consider the high uncertainties of almost all model parameters. A simplistic algorithm approach was applied in Boxall's model considering only one use and complete release. In Keller's model, linear equations to calculate the concentrations of ENMs in the environment were used without considering the uncertainty of the input data. The probabilistic material flow model developed by Gottschalk et al. (2010) is able to consider the uncertainty of all model parameters. The release of a variety of different nanomaterials has been studied using this model (Gottschalk et al., 2015; Gottschalk et al., 2009; Sun et al., 2014).

The hazards of nano-silica have been investigated by several toxicological and ecotoxicological studies (Chang et al., 2007; Napierska et al., 2010; Van Hoecke et al., 2011; Yang et al., 2014). Napierska et al. (2010) have evaluated both nano-silica and synthetic amorphous silica in *in vivo* and *in vitro* studies and apoptosis was only detected after exposure to nanoparticles (14 nm) and no effect was observed after treatment with micro particles (1–5 μm). Dekkers et al. (2013) described a higher toxic potential of nano-silica in food after intravenous injection than after oral administration and adverse effects were detected after exposure to 3 mg/kg body weight per day for intravenous injection in comparison with 1,000–2,000 mg/kg body weight per day/day for oral administration. The effects of nano-silica on aquatic organisms have also been investigated to some extent. Yang et al. (2014) for example have analyzed the ecotoxicological effect of nano-silica and bulk silica to *Daphnia magna*. The study indicated that nano-silica has a dose-dependent effect on *D. magna* but bulk silica does not.

Risk assessments for nano-silica have been performed so far only for specific applications. One example is the risk assessment of a glass cleaner formulation (spray application) containing nano-silica (Michel et al., 2013). In addition, Dekkers et al. (2013) have conducted a risk assessment of nano-silica in food. However, no environmental risk assessment covering a broad range of products containing nano-silica has been performed so far.

Considering the lack of exposure concentrations and the absence of generalized environmental risk assessments for nano-silica, the aim of this study is to evaluate the environmental release and risks of nano-silica using probabilistic material flow and environmental risk assessment models.

2. Methods

2.1. General layout of the model

Material flow analysis (MFA) was used to determine the flows and stocks of nano-silica. The principle of MFA is to express all available information concerning a material presenting in processes, flows and stocks. Fig. 1 presents the general structure of the MFA model which is based on Gottschalk et al. (2009) and Sun et al. (2014). The idea behind the model is to trace nano-silica during all phases of the product life cycle (e.g. production, manufacturing, use, and disposal) and to quantify the transport to technical compartments and transfer from technical compartments to the environment. The system boundaries considered in this work are the EU and Switzerland.

The model contains four environmental compartments: soil (split into general soils and sludge treated soils); surface water; air; sediments; and seven technical compartments: production, manufacturing, consumption (PMC); sewage treatment plants (STP); waste incineration plants (WIP); landfills; recycling; cement; export.

2.2. Model input parameters

2.2.1. Input data treatment

Due to limited knowledge about input parameters (production, allocation of nano-silica to product categories and transfer coefficients), the output of the model is inherently uncertain. To deal with this high uncertainty, all input values were introduced into the model as probability distributions. Two types of distributions – triangular and uniform distributions – were used to process the input parameters. The principle of applying these distributions to different data sources was shown in Sun et al. (2014).

The Degree of Belief (DoB) parameter was assigned to production volumes and STP removal efficiency from different sources as a measure of their reliability. Two levels of DoB were used: 20% and 80% DoB, which were assigned to the sources of low and high reliability, respectively. This method also was used in a previous study (Sun et al., 2014). High reliability was assigned to data from reviewed studies

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