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ORIGINAL PAPER

Comparing ecotoxicity risks for nanomaterial production and release under uncertainty

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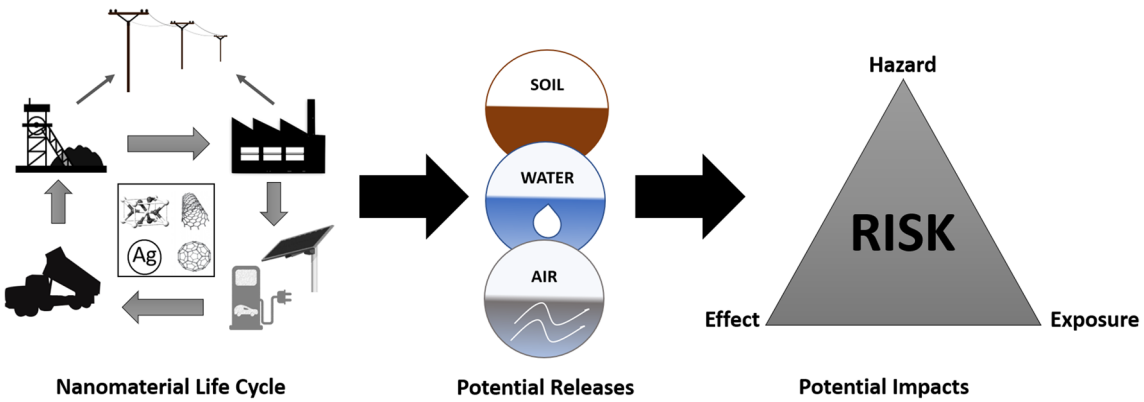
Abstract

Innovations in clean energy technology are expected to reduce fossil fuel dependence and mitigate greenhouse gas emissions, but these benefits are contingent on concurrent innovations in energy materials that can improve performance and reduce cost. Engineered nanomaterials have been promoted as a transformative advance in renewable energy generation and storage, but their adoption has also raised concerns that potential environmental impacts of nanomaterial production and use may outweigh their potential benefits. Life cycle assessment (LCA) has the potential to quantify nanomaterial environmental impacts and compare trade-offs between potential benefits and adverse impacts. However, LCA for nanomaterials is sparse due to lack of data and models that link physicochemical parameters with the overall toxicity and chemical fate of nanomaterial emissions. This study develops preliminary life cycle impact characterization factors for representative case study nanomaterials used in clean energy applications and then compares the environmental impact of direct nanomaterial release to the indirect impacts across the nanomaterial supply chain. Scenario analysis is used to model the uncertainty and variability of nanomaterial parameters that contribute to estimated freshwater aquatic ecotoxicity for carbon nanotubes, C60 fullerenes, nano-TiO_2, and nano-Ag. Results suggest that supply chain energy consumption, largely due to complex nanomaterial synthesis processes, may result in greater ecotoxicity than the direct nanomaterial release in many realistic cases. The exception to this trend was nano-Ag, which was intrinsically more toxic, primarily due to upstream processes leading to metal releases (silver mining), but also due to the potential for downstream silver releases in the worst-case scenario. This systematic approach can aid decision-makers in mitigating unintended consequences from nanomaterial use in clean energy technologies through informed life cycle and uncertainty modeling.

Graphical abstract



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Extended author information available on the last page of the article

Keywords Nanomaterials · Characterization factors · LCA · Ecotoxicity · Clean energy technology

Introduction

Engineered nanomaterials are characterized by having at least one dimension in the nanometer range, 10^{-9} m (European Commission 2011). Their size confers many unique properties that may provide enhanced performance in clean technologies and consumer products (Chen et al. 2012; Gilbertson et al. 2015; Hussein 2015). Nanomaterials are increasingly used to meet the growing demand for electricity grids powered by renewable energy technologies (United Nations 2016), where their integration can contribute to greater energy storage and conversion efficiency (Chen et al. 2015). For example, nano-TiO_2 has been found to increase the efficiency of lithium-ion batteries (Vázquez-Santos et al. 2017), and research on nano-Ag for polymer solar cells shows increased solar transmittance (Yu et al. 2011). Carbon nanotubes (CNTs) are utilized in fuel cell technologies for improved membrane strength and conductivity (Zhang et al. 2017), and fullerenes exhibit increased device efficiency in small molecule and polymer solar cells (Anctil et al. 2013). Despite the energy performance benefits of nanomaterials, key questions remain regarding the trade-off between their potential technological gains and their risks to human and environmental health (Colvin 2003; Moore et al. 2018). Quantifying this trade-off requires an understanding of the holistic life cycle environmental impacts of these materials and an ability to disaggregate this impact to identify processes with the greatest potential for improvement, whether they be upstream mining and processing of precursors that are synthesized into nanomaterials or the direct release of the nanomaterials into a natural ecosystem.

Life cycle assessment (LCA) is a decision-directed tool that holistically quantifies environmental performance of a technology or material from raw material extraction to disposal at end-of-life. While applying LCA to emerging nanomaterials has shown potential for informing cleaner design, manufacturing, and technology integration, this method also faces several challenges (Eckelman et al. 2012; Upadhyayula et al. 2012). A critical barrier is the lack of nanospecific life cycle impact assessment (LCIA) methods, which are required to model the ultimate effect of nanomaterial release on human and environmental health. LCIA models are typically built using comprehensive datasets of empirically derived and modeled parameters that describe a material's transport and fate in environmental compartments, potential for exposure to humans and other aquatic and terrestrial organisms, and ultimate impact to mortality, health, and other ecologically relevant end points. Because nanoscale physical and chemical properties differ dramatically from their larger scale, i.e., bulk, counterparts, fundamental characterization of fate, transport, exposure, and toxicity must be carried out anew, a challenge given the accelerating rate at which new nanomaterials are introduced to the market each year (Salieri et al. 2015), the difficulty of accurately modeling environmental fate and toxicity (Gavankar et al. 2012; Keller et al. 2013), and the lack of LCIA modeling techniques to integrate nanospecific impacts with other non-nanoemissions across the life cycle (Theis et al. 2011).

Due to the scarcity of needed data and models, many LCAs on nanoenabled technologies have limited their scope to impacts of bulk material emissions or upstream processes (e.g., toxicity impacts associated with energy consumption during nanomaterial synthesis), but omitted the impacts caused directly by nanomaterial emissions released during production or use (Gavankar et al. 2012; Hischier and Walser 2012). However, in recent years, nanospecific ecotoxicity characterization factors have been modeled in a handful of cases (Eckelman et al. 2012; Rodriguez-Garcia et al. 2014; Salieri et al. 2015; Deng et al. 2017). One such study finds that for the case of carbon nanotubes in a realistic scenario, direct emissions of CNTs to freshwater would contribute a negligible portion of the total ecotoxicity when compared to the contributions of non-nanoemissions from upstream processes, such as electricity production required for CNT synthesis (Eckelman et al. 2012). Whether such a finding is generalizable to other nanomaterials is still unknown, particularly because CNT production is extremely energy intense (Upadhyayula et al. 2012). There is a clear need to determine contributions to ecotoxicity risks for other nanomaterials used in clean energy technologies and to understand the extent to which these findings may hinge on resolving uncertainty surrounding LCIA modeling for nanomaterials.

The present study aims to characterize this uncertainty by modeling scenarios that demonstrate the potential implications of omitting direct nanomaterial emissions from LCIA and by assessing nanospecific impact contributions to the total nanomaterial life cycle. These scenarios are built on bounded ranges that describe realistic and worst-case emissions of nanomaterial emissions using best available current knowledge. Using four case study material systems, the cradle-to-gate impacts of producing nanomaterials (including raw material extraction, energy production, and synthesis and purification steps) are compared to the potential impacts if the nanomaterials produced are then released, based on realistic scenarios of emissions and exposure. This approach fills a critical gap between past studies, which either examine only nanospecific impacts or which omit nanomaterial releases altogether. A key goal is understanding how life

cycle comparisons may vary with parameter uncertainty, including the uncertainty associated with nanomaterial release and ecotoxicity estimates. Ultimately, this approach can be integrated into broader LCA studies, once more information is available about how these materials are adopted and how their use changes or compares to the incumbent materials used in clean energy technologies.

Methods

Life cycle assessment overview

LCA is a model commonly used to examine "cradle-to-grave" environmental impacts of a material, product, process, or system. LCA is carried out in four steps: (1) goal and scope definition, (2) life cycle inventory (LCI), (3) life cycle impact assessment (LCIA), and (4) interpretation, which can include uncertainty analysis or recommendations for improvement. In the case of modeling nanomaterials, accounting for emissions after their production (i.e., during their use and management of end-of-life) is particularly challenging because they are used in a wide array of different applications, many of which are still unknown since these materials are early in their commercialization pathways. For that reason, this study focused on the more well understood processes taking place from "cradle-to-gate," or in other words, all upstream material extraction, refinement, and manufacturing that would bring the nanomaterial to the factory "gate" to be used in any application. This focus enables estimation and uncertainty characterization for all upstream or "embodied" impacts due to nanomaterial production. In addition, we also modeled the potential impacts associated with nanomaterial release, both during these upstream processes and under scenarios of accidental release after production. The upstream flows are modeled with existing LCI data, and impacts are calculated using established LCIA models. Because these LCIA models do not yet exist for nanomaterials, adaptations were made to calculate ecotoxicity impact characterization factors that account for the unique physical and chemical attributes of nanomaterials according to best available knowledge. These characterization factors were then parameterized using scenario-driven estimates collected from a comprehensive literature review, which also served to compile for the first time a wide array of nanomaterial fate, transport, and ecotoxicity data. Further details about all methodological aspects are provided in the following sections.

Goal and scope definition

The goal of this study was to characterize uncertainty in the life cycle ecotoxicity impacts of nanomaterial production and release and to identify processes with the highest potential for improvement. This goal was carried out using four case study nanomaterials: silver nanoparticles (nano-Ag), nano-titanium dioxide (nano-TiO_2), single-wall carbon nanotubes (SWNT), and 60-carbon spherical fullerenes (C60). These materials were selected based on their increasing use in clean energy applications (Hussein 2015), prevalence in risk assessment literature, popularity in consumer products (Vance et al. 2015), and importance for regional policy decisions (OECD 2012). Additionally, they also represent a wide cross section of material types (metals, metal oxides, and carbon-based), and their life cycle inventories are available in the open literature, which allows us to understand how calculated impacts, and uncertainty therein, vary across different material sets while ensuring consistency and transparency. However, because previous LCAs often do not contain information on nanomaterial releases, we also generated emissions estimates using published modeling methods (see section "Life cycle inventory development").

Figure 1 depicts the scope of the present study. For each case study material, impacts are related to the functional unit of one kilogram of pristine nanomaterial (not modified or functionalized). This basis was selected to simplify modeling and avoid major data gaps, since many of these nanomaterials are not yet widely commercialized and the clean energy applications in which they might ultimately find use are still unclear. While this approach does not account for the potential changes in functionality that a product or technology might have by use of a nanomaterial compared to an incumbent material, it does provide a uniform basis on which flows, emissions, and impacts can be compared, which aligns with our goal to investigate uncertainty and trends surrounding ecotoxicity impact modeling and comparisons.

Nano-Ag Listed most frequently in the Consumer Products Inventory, silver nanoparticles have antibacterial and optical properties that make them useful in dye-sensitized solar cells, textiles, consumer products, hand sanitizers and skin products, optical applications, and conductive inks (Hwang et al. 2013; HSDB 2013a; Oldenburg 2015). They can be produced using grinding, a pulsed plasma process, reduction of silver ions, laser ablation, or vapor deposition (Meyer et al. 2011, Pourzahedi and Eckelman 2015). Here, an LCI based on the flame spray pyrolysis method is adapted from Walser et al. (2011).

Nano-TiO_2 The second most common type of nanomaterial listed in the Project on Emerging Nanotechnologies' Consumer Product Inventory (Vance et al. 2015) is titanium dioxide. Nanoscale titanium dioxide can be found in the production of hydrogen fuel, as a photocatalyst, dye-sensitized solar cells, sunscreens, self-cleaning devices, UV-resistant materials, cosmetics, printing ink, chemicals, plastics, rubbers, wastewater treatment, and

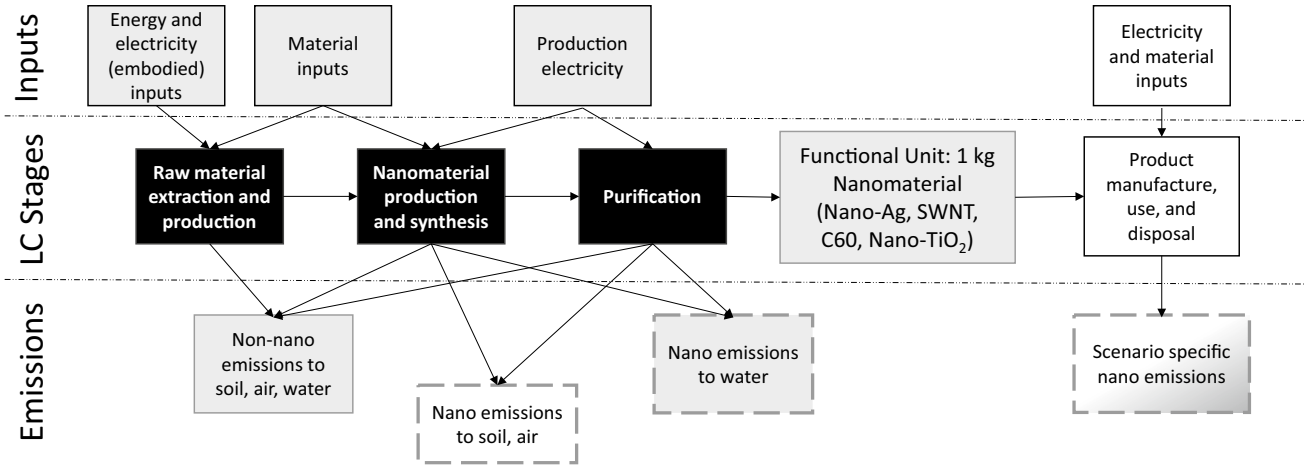

Fig. 1 Generalized scope. Black boxes represent processes within major life cycle stages. Gray boxes are materials, energy, and emissions that are included within the study scope. The boxes with a dotted border represent release estimates not previously studied in past LCAs. White boxes are life cycle stages typically represented in a full cradle-to-grave LCA, but not included here because of lack of information about end-uses of case study materials. The scenario-specific nano emissions box is both white and gray to indicate use-phase nano emissions are not modeled in the baseline analyses, but are included in two specific use-phase scenarios (see Section on "Other factors contributing to ecotoxicity variability")

for the degradation of pesticides (Hessen Ministry of Economy, Transport, Urban and Regional Development 2008; HSDB 2013b). Studies on nano-TiO_2 model either anatase nano-TiO_2, rutile nano-TiO_2, or a mixture of the two, which differ based on the crystal structure of TiO_2 (Shi et al. 2013). The production method modeled here is the Altairnano hydrochloride process, adapted from Grubb and Bakshi (2011).

Carbonaceous nanomaterials Single-wall carbon nanotubes (SWNT) are tubes comprised of graphene one wall thick. Specific advantages of SWNT are their tensile strength and Young's modulus (Healy et al. 2008). Potential uses are in semiconductors, batteries, hydrogen energy storage, and others. Another carbonaceous nanomaterial of interest is C60 fullerenes. Spherical fullerenes are molecules in the shape of hollow spheres commonly made of 60, 70, or more carbon atoms. They are currently used in cosmetics, skin creams, polymers, fuel cells, in lubricants, and organic photovoltaics (Ullmann 2003). SWNT and fullerenes can be produced using HiPco (high-pressure carbon monoxide), CVD (chemical vapor deposition), arc ablation (carbon arc discharge), laser ablation, and other methods. Fullerenes can additionally be produced using pyrolysis of naphthalene or toluene and the arc vaporization of graphite (Ullmann 2003). Here, the methods of production analyzed are carbon vapor deposition and arc ablation for SWNT (Healy et al. 2008), and pyrolysis using toluene and arc ablation for fullerenes (Anctil et al. 2011).

Life cycle inventory development

LCI data are taken from previous LCAs of the case study materials, specifically SWNT (Healy et al. 2008), nano-Ag (Walser et al. 2011), C60 fullerene (Anctil et al. 2011), and nano-TiO_2 (Grubb and Bakshi 2011). All nanospecific emissions were modeled from the literature, and all non-nano data were taken from ecoinvent 3.1, implemented in SimaPro 8. The electricity input to manufacturing processes was modeled as medium voltage average production mix for the USA, to replicate what is used primarily by industrial sources. Nanomaterial emissions during the production process are modeled using scenario analysis to bound the range of potential uncertainty around both release of these materials and their ultimate impact to the environment. The scenarios are (1) "no nano," which only considers upstream processes and non-nanoscale emissions associated with production processes (this scenario is included as a basis of comparison with past studies that omit nanospecific releases), (2) a "realistic" scenario, in which a broad range of scenarios are used to account for normal processing and environmental controls, and the median values are used, and (3) a "worst-case" scenario, in which the extreme values are used to demonstrate the range of impacts given unlikely circumstances (e.g., non-ideal operating conditions). Values for Scenarios 2 and 3 are provided in Table 1.

Previous studies have modeled realistic mass fraction lost during production as 0–2% and an additional 0–2%

Table 1 Life cycle inventory parameters applied to model direct nanomaterial release, by life cycle stage and scenario

Life cycle stage	Mass percent of NM *released* by stage
Upstream (nanomaterial production, purification)	All materials: 0–4% (Realistic); 2–6% (Worst case)
Downstream (scenario-specific use, end-of life)	Nano-TiO_2: 63%
	SWNT: 0.0094%
	C60 and Nano-Ag: not modeled
Waste management	Mass percent of NM *removed* by WWTP
Removal by WWTP (from Sun et al. 2014)	Nano-TiO_2: 80–100%
	SWNT and C60: 88–95%
	Nano-Ag: 39–99% (Realistic); 39–62% (Worst case)

during manufacture of nanomaterial-containing product and worst-case mass fraction released of 6% during production and 2% during manufacture (Gottschalk and Nowack 2011). While we did not consider any loss of material during the use of nanoenabled energy technologies, we did include the potential for these materials to ultimately be released at a wastewater treatment plant (WWTP), where between 20 and 98% of nanomaterials by mass are removed from influent to effluent, depending on the type of nanomaterial (Sun et al. 2014). While removal estimates are based on a European case study, estimates for the USA are similar, such that geographical differences are not expected to change the ultimate results (Lazareva and Keller 2014). The assumptions that are used for mass fraction released during production and manufacture are summarized in Table 1. These assumptions account for the fraction of nanomaterials that may ultimately enter the aquatic environment, taking into consideration the potential releases from processing or use as well as the potential for material removal when the effluent containing the nanomaterial is treated by conventional emissions control processes (WWTP). Further explanation of these additional emissions is provided in Supplemental Tables S1 and S2.

In addition to nanoemissions modeled as described above, the remaining (non-nano) emissions and inputs associated with upstream processes were grouped into three categories: (1) embodied electricity and energy (all energy inputs consumed by background processes occurring ahead of nanomaterial synthesis); (2) production electricity (consumed during the nanomaterial synthesis process); (3) other life cycle inputs, which includes raw material extraction or mining, transportation, and waste treatment. Energy is disaggregated this way to reflect past work that suggest that for ecotoxicity impacts, energy processes are key to understanding life cycle results.

Impact assessment models

A critical decision is the choice of the impact category, because environmental issues can be represented by many different midpoint-level impacts such as global climate change, ocean acidification, or mineral or resource depletion. For this study, we limited the analysis to the two issues that were quantifiable using available inventory data and impact assessment models and that connect with expected impacts of these materials: freshwater aquatic ecotoxicity (referred to simply as ecotoxicity throughout this paper) and cumulative energy demand (CED). Previous LCAs often used Ecoindicator 99 (Goedkoop 1999), ReCiPe (Goedkoop et al. 2009), and CED as impact categories, further confounding the ability to draw generalizations on results of LCA for different nanomaterials. Replicating the LCIs used previously and using a consistent impact assessment method allow for more effective discussion and comparison of the results between nanomaterials.

To model freshwater aquatic ecotoxicity impacts, we used an adapted version of USEtox, a consensus-based method developed by the Task Force on Toxic Impacts under the United Nations Environment Programme/Society of Environmental Toxicologists and Chemists (UNEP/SETAC) Life Cycle Initiative (Rosenbaum et al. 2008). In the past, criticism has been cast on attempts to tailor USEtox for nanomaterial toxicity because the method was originally intended for assessing toxicity midpoints for organic chemicals, and the ability to adapt the exposure equation and fugacity model for fate calculations was uncertain. However, recent work has begun exploring use of USEtox for nanomaterials, as outlined in review (Gilbertson et al. 2015) and modeling (Salieri et al. 2015) sources; the conclusions of which are that in the absence of an adequate modeling framework, the basic USEtox impact assessment framework and some of the equations can be adapted for preliminary nanomaterial toxicity assessment. In fact, the method has been applied

successfully to nanomaterials in recent cases (Eckelman et al. 2012; Deng et al. 2017).

Generally, impact assessment is performed by multiplying substance-specific characterization factors (CFs), by the amount of chemical emitted, M, to quantify an Impact Score (IS):

$$IS_t = \sum_i \left(CF_{t,i} \times M_i\right),$$

where t is the impact category, in this case, freshwater aquatic ecotoxicity measured in comparative toxic units (CTU_e). M_i is the mass of chemical emitted to an environmental compartment, in this case, freshwater ecosystems (as determined by LCI as described above). The basic framework used by USEtox quantifies the characterization factor (CF) as:

$$CF = FF \times XF \times EF,$$

where EF is the effect factor, XF is the exposure probability, and FF is the fate factor. To determine the most representative data to parameterize this framework, the scenario analysis (realistic and worst case) incorporated a wide range of physicochemical properties that might impact EF, XF, and FF. The necessary data are taken from literature sources or from experimental data for fate and exposure from the literature as described below and summarized in Tables S3–S7 of the Supplemental Information.

Fate The fate factor describes the duration that the nanomaterial will reside in the environmental compartment (Rosenbaum et al. 2008), i.e., freshwater in this case. Debate still exists as to whether an equilibrium partitioning approach as used in USEtox and ReCiPe (Goedkoop et al. 2009) or a colloidal model is more appropriate for modeling the fate of nanomaterials in environmental media (Gilbertson et al. 2015). The recommendations from Gilbertson et al. (2015) are that given the lack of an adequate colloidal model, the use of a partitioning model for fate calculations is still the best available interim approach. Here, the SimpleBox4Nano and USEtox models are compared for fate using the recent literature data on physicochemical properties for the three insoluble nanomaterials. The freshwater fate factor, measured in days, is calculated for each substance using the best available substance-specific data or generic data when not specifically required by the fate model (See Table S8). Assumptions about fate are based on both the predicted values using substance data and measured values in a laboratory or actual freshwater, as reported in the literature. Nano-TiO_2 fate factors are explicitly based upon previous findings which use SimpleBox4Nano (Salieri et al. 2015).

Exposure The environmental exposure factor in traditional models is the fraction of chemical dissolved in freshwater, thereby determining the probability that an aquatic organism will be exposed to the chemical. However, most of the case study materials, with the exception of nano-Ag, are not likely to solubilize. We have adopted the approach published in previous studies (Salieri et al. 2015) to assume that EF = 100%, or in other words, that all insoluble nanomaterials are bioavailable.

Effect To align with previous toxicity assessment methods, a PAF (Potentially Affected Fraction of Species) approach is taken, where the effect factor is calculated as $EF_{eco} = 0.5/HC_{50}$, the HC_{50} being the calculated geometric mean of all the species-specific LC_{50} or EC_{50} values (Eckelman et al. 2012; Salieri et al. 2015; Deng et al. 2017; Rosenbaum et al. 2008), where the LC_{50} and EC_{50} are the exposure concentrations at which 50% of the test organisms exhibit the effect relative to the control. The LC_{50} describes a concentration at which mortality is experienced, whereas EC_{50} generally refers to an effect other than mortality. Ecotoxicity from chemical emissions to freshwater is generally estimated by calculating the effect to at least three phyla: aquatic invertebrates, aquatic vertebrates, and plants. The LC_{50} and EC_{50} values used herein are taken from the literature (see Tables S4–S7 in the SI). Uncertainty around these values is based on variability in particle size, surface charge, surface coating, purity, and other properties reported in the literature.

The realistic EF_{eco} is derived from LC_{50} values (mortality only) calculated from acute and chronic toxicity tests for all available phyla and for any permutation of a given nanomaterial (size, crystalline structure, etc.). All these types of nanomaterials are included to represent the diversity of nanomaterials available in commerce and the influence of a wide range of toxicity results on the overall ecotoxicity. The worst-case scenario (WCS) EF_{eco} is derived from the most sensitive end points for three phyla. An acute-to-chronic (ACR) factor of 10 was applied where appropriate (Rosenbaum et al. 2008). Studies where all tested concentrations did not result in significant ecotoxicity were not included in any calculations.

Nano-Ag modeling Nano-Ag presents a somewhat different issue than the three other nanomaterials, in that the core material, silver, has significantly different chemical properties as a partially soluble metal. Much debate exists in the nano-LCA community as to the proper way to model the contribution of ionic silver to nano-Ag toxicity (Kennedy et al. 2010). This is an important distinction given that in over 90% of past studies, ionic silver was found to be more toxic than nano-Ag (Notter et al. 2014). Furthermore, USEtox provides an interim CF for ionic silver which itself has a high degree of uncertainty when compared to the recommended set of USEtox characterization factors. As such, considerations are taken for modeling the CF for nano-Ag and comparing it to the ionic silver CF.

The scenario approach here assumes no dissolution to avoid the issues associated with disaggregating the toxic

impacts of nano-Ag from ionic silver. A very simple scenario tests the sensitivity of this choice and is presented with the CF results in Table S8 in the SI file. In this sensitivity analysis, nano-Ag dissolution is included at 5% and 25%, for RS and WCS, respectively, based upon findings from nanotoxicity studies, to demonstrate the potential toxicity of nano-Ag when the dissolved portion of the emission can increase the overall toxicity of emissions. No further speciation of nano-Ag is assumed, despite the evidence that this could be revised in future studies (Mitrano et al. 2014). In this scenario, the CF equation is as follows:

$$CF = EF_{NanoAg} * XF_{NanoAg} * FF_{NanoAg} + \left(1 - XF_{NanoAg}\right) * CF_{Ag(I)},$$

where $XF_{NanoAg} = 5$ and 25% in RS and WCS, respectively. However, improved assumptions about dissolution and allocation of ecotoxicity are discussed in section "Other factors contributing to ecotoxicity variability".

Uncertainty analysis

The software program @RISK was used to create Monte Carlo numerical simulations to explore uncertainties in the three key parameters (FF, EF, and XF) for the developed characterization factors. The fate factor was modeled with a uniform distribution because the underlying data were drawn from a limited number of sources such that normality could not be inferred. The uniform distribution reflects an equal probability of values being within the range between the realistic scenario (minimum value) and the worst-case scenario (maximum value). Because empirically determined exposure factors are not typically available, most impact modeling studies assume a value of 1; therefore, a Beta-PERT distribution was applied. This distribution prioritizes the likely value (as determined by expert judgment) over values around the edges of the distribution. However, for the case of nano-Ag, published values suggest a range between 0.6 and 0.8, and thus, a most likely value of 0.7 was selected for the exposure factor. For the effect factor, a normal Gaussian distribution was applied because sufficient data were available to calculate a mean and standard deviation. Both the nano-TiO_2 and nano-Ag cases had to be truncated on the left at zero to prevent negative value scenarios. Figure 2 summarizes the method used to calculate the ranges, Table S8 lists values used to develop the distributions, and Table S9 summarizes inputs to the @RISK models. Five thousand iterations were run to produce a CF distribution, the standard number of iterations for Monte Carlo sampling.

Results and discussion

Characterization factor development and variability

Relevant life cycle data and fate, effect, and exposure factor parameters were compiled to develop characterization factors (CFs) for each nanomaterial. The CF values for case study materials studied represent a large range in magnitude of values as a result of the variability modeled in the fate and/or effect factor calculations (Fig. 3).

For instance, nano-TiO_2 has a relatively low, yet wide range of possible values as a result of the uncertainty surrounding the calculation of the fate factor (Table S3), which can be explained by the lack of available data to characterize its partitioning behavior in environmental media. While the realistic nano-Ag CF value is similar to many of the USEtox inorganic CFs (Figure S1), the WCS CF for nano-Ag was 250% larger than the largest inorganic material (i.e., the USEtox Ag(I) CF). The difference

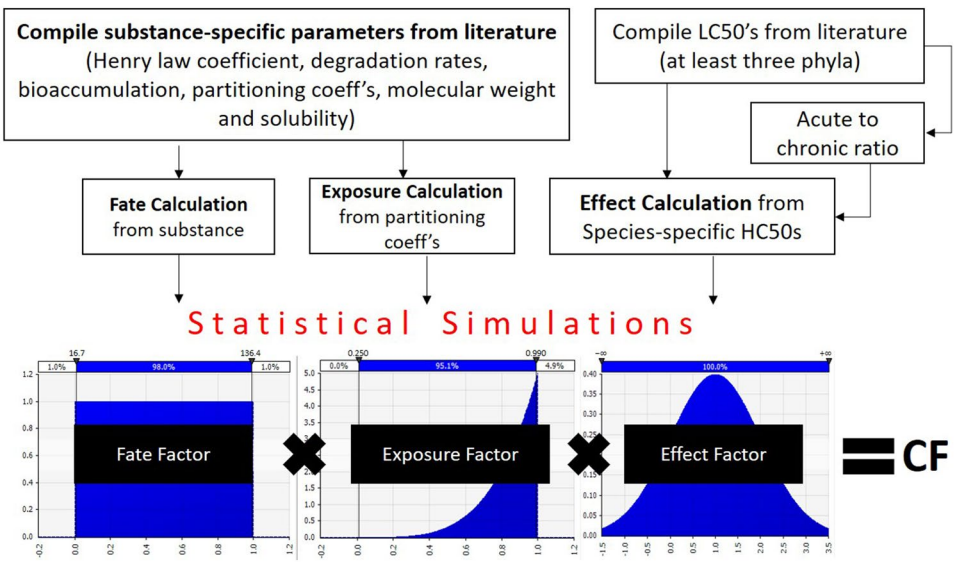

Fig. 2 Uncertainty analysis modeled ranges of physico-chemical properties and fate, exposure, and effect factors. Statistical simulations were used to generate the ranges for both realistic and worst-case scenarios based on assumptions about those scenarios described in this section

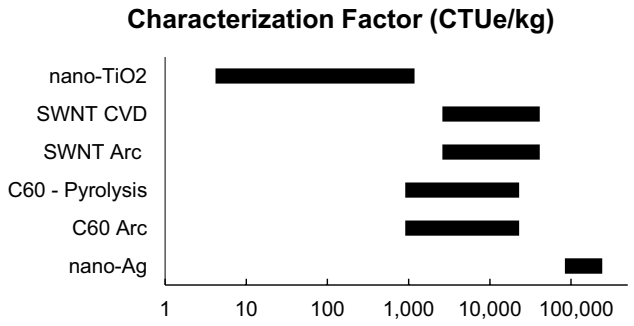

Fig. 3 Log-scale ecotoxicity characterization factor ranges (between realistic and worst-case estimates) by nanomaterial

between the realistic and WCS CFs is primarily due to the large range in toxicity results reported in the literature, which themselves have inherent variability due to experimental conditions (Table S7) and the inclusion or exclusion of ionic silver. Carbon-based nanomaterials, C60 and SWNT, due to the similarities in the modeling of fate and exposure, result in a similar spread between WCS and realistic CFs, but the SWNT realistic CF is almost three times larger than the C60 realistic CF.

Because the key input parameters (fate factor, exposure factor, and emission factor) have a high degree of uncertainty in their quantification, Monte Carlo numerical simulation results help explore the most likely range of potential characterization factors. Figure 4 displays the distribution of expected CF values for the four case study materials. The CFs for inorganic materials (Fig. 4a and b) show a tighter, more normal distribution skewed to lower values, mainly due to the lower effect factors. Notably, both carbonaceous cases (Fig. 4c and d) show a wider, more uniform distribution, indicating higher uncertainty overall in CF calculation. Values shown in the SWNT distribution are also consistent with previous studies (Table S1). As a whole, results indicate that for LCA involving any of these nanomaterials with a realistic, i.e., not overly conservative, modeling approach, the nanomaterial CFs could be treated similarly to those of USEtox inorganics, although emphasizing that caution is necessary to understand the potential range of CFs. Beyond showing the likelihood for certain characterization factor values, these results can also inform the direction of research to minimize uncertainty surrounding the most variable factors.

Ecotoxicity impact comparisons

The calculated CFs were then used to model contributions to ecotoxicity for the four nanomaterials by combining the CF values with the potential release estimates for all materials under the realistic and worst-case scenario (Fig. 5).

Whereas the CF values varied widely for each nanomaterial between the realistic and WCS, the ranges of total

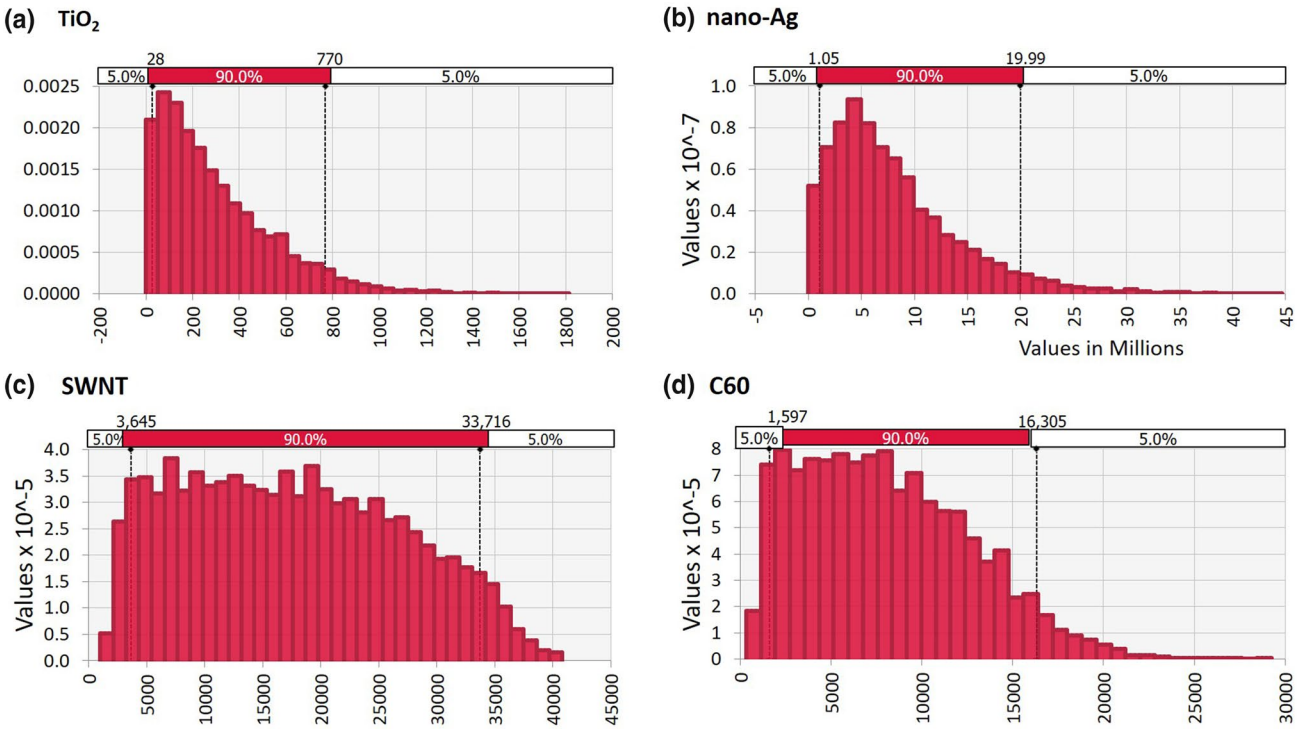

Fig. 4 Characterization factor distributions determined from Monte Carlo simulation for **a** nano-TiO_2, **b** nano-Ag, **c** SWNT, and **d** C60

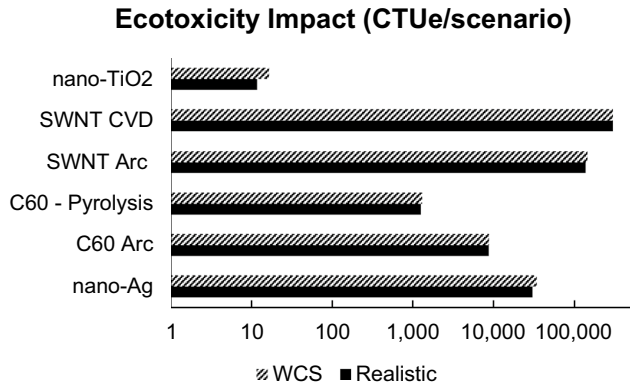

Fig. 5 Log-scale ecotoxicity impact (expressed as cumulative toxic unit equivalent) by nanomaterial for the worst-case and realistic scenarios of impact and release

ecotoxicity impact were much closer for all case studies. In other words, the differences in CF were less significant once the life cycle ecotoxicity accounted for non-nanoemissions and impacts. To further investigate what was causing this trend in results, contributions to ecotoxicity were disaggregated into four major categories: (1) direct nanomaterial release; (2) embodied electricity and energy; (3) production electricity; and (4) "other," which includes material mining and precursor preparation, transport, and waste treatment, as depicted in Fig. 6 (with more detailed results in SI Figures S2–S9). Note that production electricity refers to ecotoxicity associated with producing the electricity required during the nanomaterial synthesis or purification steps; while embodied electricity includes all other upstream energy inputs to material extraction and preparation. This distinction is included to help improve understanding about where to prioritize process improvements, e.g., reducing or substituting a high-impact precursor or changing nanomaterial synthesis conditions to minimize energy use.

A key finding from Fig. 6 is that differences in the direct nanomaterial impacts for each scenario were insignificant compared to the ecotoxicity impacts caused by non-nanoemissions, which echoes past findings (Eckelman et al. 2012). Production electricity was seen to be the primary contributor to ecotoxicity for SWNT in all scenarios and

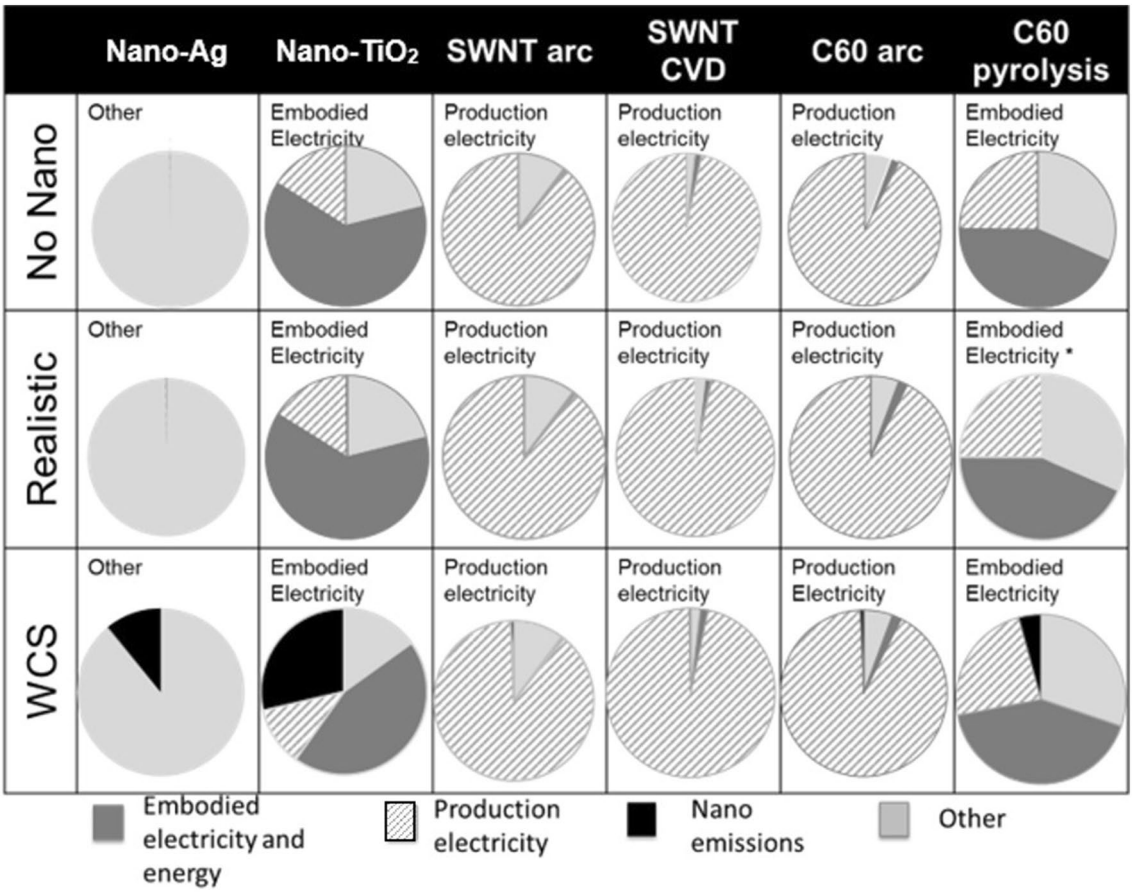

Fig. 6 Ecotoxicity contributions from four major categories across the three scenarios. The category with the highest impact among the four is named above the pie chart in each case. The asterisk (*) for realistic C60 pyrolysis reflects that embodied electricity is only slightly larger than the other categories

for the "no nano" and realistic scenarios for C60 produced through the arc plasma method. On the other hand, embodied electricity makes a larger contribution in the no nano and realistic cases for C60 produced by pyrolysis, primarily due to the significant energy demand associated with synthesis of the carbonaceous feedstock (Anctil et al. 2011). On the other hand, ecotoxicity impact for nano-Ag is associated primarily with silver release during mining and precursor preparation processes. This finding is consistent with Pourzahedi and Eckelman (2015), who demonstrated that upstream production of silver has the most significant impacts, even when comparing across multiple nanomaterial synthesis methods and for a wide array of life cycle impacts. Nanomaterial release contributes marginally to the overall impact in the worst-case scenario for all materials except SWNT. These results suggest that when the nanomaterial itself has a very energy-intense manufacturing process (as is the case for SWNT), and when most of that energy comes from electricity, which is still generated using a significant amount of coal in the US electric grid, then it is likely that these upstream processes will drive impact, even under worst-case scenarios of nanomaterial release. However, in cases where a greater fraction of nanomaterials are released and/or when their release ultimately leads to greater bioavailability and toxicity, considering direct ecotoxicity is important from a risk perspective.

Accidental release scenario

Although the upstream processes contribute most to impact in the two modeled scenarios, there is another case that should be considered from a proactive risk perspective. Specifically, we examined the unlikely scenario in which 100% of the nanomaterials produced are released to freshwater without going through any wastewater treatment. While this scenario is not likely, it could be conceived of as an accidental release (see SI Table S1) and reflects the modeling framework used by Eckelman et al. (2012) which found that SWNT emissions still contributed an insignificant portion of the impact when compared to the ecotoxicity of electricity production, even if all of the produced nanomaterials were released directly to a freshwater ecosystem. Here, however, the "accidental release" scenario depicts a case where nanomaterial emissions *could* contribute an overwhelmingly significant percent of the total ecotoxicity impact, for all nanomaterials *other* than SWNT (Fig. 7).

Other factors contributing to ecotoxicity variability

Nanoenabled application It is also important to note that a high CF does not necessarily indicate a high ecotoxicity value; the amount of material released and the ultimate application in which nanomaterials are adopted can influence the overall ecotoxicity throughout the life cycle. For instance, before C60 is integrated into clean energy applications such as organic photovoltaic cells (OPVs), it is first purified and often functionalized to create materials like [6,6]-phenyl-C_{61}-butyric acid methyl ester (PCBM) (Anctil et al. 2011). These processing steps require additional energy, increasing the potential ecotoxicity from production, but may also influence the fate, transport, and effect of the material itself. Because of the lack of ecotoxicity data for multiple phyla, a characterization factor cannot be calculated at this time for PCBM. To understand the life cycle impact of C60, future studies must generate data to model how ecotoxicity changes with processing and use. Further, as demonstrated with the use of nano-Ag in textiles, the greatest life cycle impacts ultimately depend on the impact category studied, the silver content, and consumer behavior (Hicks et al. 2015). Although nano-Ag use in textiles could reduce

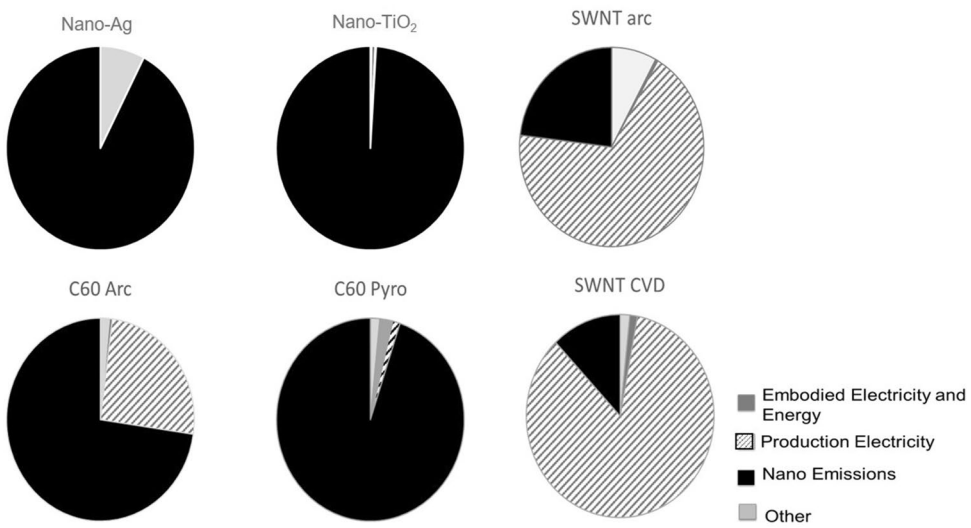

Fig. 7 Accidental release to freshwater is modeled by calculating the ecotoxicity relative to 100% of nanomaterial release into freshwater ecosystems to facilitate a comparison between the impacts of nanomaterial production and the maximum impact due to its release

the odor and laundering frequency, the ultimate consumer behavior strongly influences whether the potential benefit of reduced laundering outweighs the upstream energy and resources required to produce nano-Ag enabled textiles (Hicks and Theis 2017). In other instances, researchers have combined multiple nanomaterials into the same clean energy application. Yu et al. (2011) demonstrated the use of nano-Ag wires in a polymer solar cell electrode and PCBM in the active layer for device performance improvement. From a systems perspective, the ecotoxicity from integrating nanomaterials varies depending on application, consumer use, functionalization, and combination of materials and therefore should be considered when choosing nanomaterials to integrate into clean energy technologies.

Type of nanomaterial production Past studies (Healy et al. 2008; Anctil et al. 2011; Pati et al. 2014) have shown that synthesis method can be a source of variation in overall nanomaterial life cycle impacts, but does not typically change the relative contribution of different stages to the overall impact. For example, in the case of nano-Ag, Pourzahedi and Eckelman (2015) found that even when the magnitude of life cycle ecotoxicity varied among different synthesis methods and material sources, the production of silver had the highest contribution to total life cycle impact in all cases. Although the number of *similar* production methods between the case study materials is limited, available results indicate that the type of material has a greater influence than synthesis method on the overall results. For example, pyrolysis methods are modeled for both C60 and nano-Ag, but the life cycle stage with greatest impact differs significantly between the two cases. The predominant contributor to ecotoxicity in the nano-Ag case is overwhelmingly the mining and refining of the silver precursor (Figure S3), due to the release of metallic or ionic silver in these upstream processes. On the other hand, C60 has lower upstream releases of emissions leading to ecotoxicity and far greater contribution from electricity required during precursor synthesis and nanomaterial manufacturing. These results underscore the importance of continuing to build nanospecific life cycle inventory libraries to further characterize different nanomaterial production pathways and impact potentials.

Limited life cycle data Results also varied depending on the volume and type of nanomaterial modeled as being released at some point in its life cycle. These findings are similar to Deng et al. (2017) where there was a large variation in the effect factor of graphene oxide because of the limited data available from the ecotoxicity literature. Using a realistic CF for nano-TiO_2, the impact of the use-phase emissions is 0.26 CTUe, but when using the WCS CF, the impact is 74 CTUe, over two orders of magnitude greater. On the other hand, the impact of the direct emissions of SWNT is at most 0.03% of the total impact even when a WCS CF is used, suggesting that use-phase emissions of SWNT may be considered negligible under the current knowledge of releases from manufacturing or WWTP. However, without more comprehensive information about nanomaterial use in products and long term releases, these estimates may underestimate emissions occurring during the use-phase or end-of-life. For example, SWNT could be released from lithium-ion batteries during recycling because of cross-product contamination (Köhler and Som 2008).

Interpretation and implications

Impact assessment tools were adapted to create a potential range of CFs using multiple modeling choices for four different nanomaterials, with results underscoring the effect on nano-CFs due to variability in both life cycle modeling parameters and fate, effect, and exposure parameter data. Future work is needed to resolve modeling uncertainty by advancing fate and exposure models specific to nanomaterials and their unique physical and chemical properties. On the other hand, the question could be raised about the extent to which these sources of uncertainty must be resolved, particularly because in many realistic cases explored in this study, electricity is the primary driver of ecotoxicity, not the release of nanomaterials themselves. Thus, the location of production and the associated electric grid fuel mix can greatly influence the ultimate ecotoxicity of a given nanomaterial. In many LCAs, the environmental impact of electricity consumption is well accounted for by the impact of cumulative energy demand (CED), which is a relatively straightforward metric to calculate given widespread availability of energy use data and consensus on characterization factor development. CED is a metric that describes the net life cycle energy used to produce a material or product (Louwen et al. 2016). Past LCA work has shown that the sensitivity of CED impacts to fossil sources in the electricity grid mix gives it high correlation to other life cycle environmental impacts, including ecotoxicity (Huijbregts et al. 2010; Laurent et al. 2012). The CED of the case study nanomaterials was calculated using data from the literature sources described in the "Methods" section to investigate whether CED can help predict impacts in the absence of more certain ecotoxicity parameters (Fig. 8).

Given the amount of detail that goes into nanoecotoxicity modeling and the similarities between Figs. 5 and 8, it may be possible to streamline early LCA studies by using CED as an approximation of net impacts prior to more detailed impact modeling, particularly when processes are known to consume significant amounts of electricity or other energy sources. These findings suggest that additional studies should be performed in order to determine the effectiveness of CED as a proxy for "hotspot" analysis until ecotoxicity uncertainties are more resolved. Only in the case of nano-Ag,

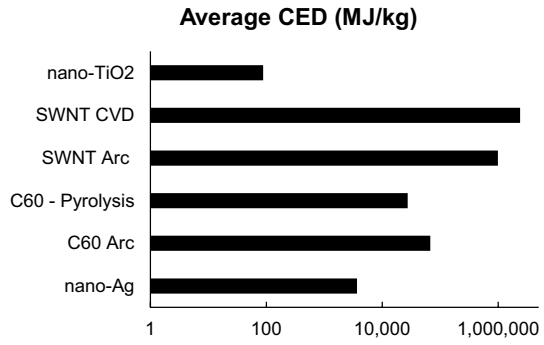

Fig. 8 Average cumulative energy demand per kg of nanomaterial produced, shown on a logarithmic scale

the only soluble nanomaterial included in this study, was the impact of nanomaterial emissions a significant contributor to the overall ecotoxicity. Thus, if a nanomaterial has physical or chemical properties expected to confer significant ecotoxicity, then it should be prioritized for developing detailed CFs that reflect variability associated with specific material properties and transformations through its life cycle and in the environment. On the other hand, ecotoxicity of materials characterized by lower uncertainty (e.g., the relatively uniform distributions of CFs for C60 and SWNT) may be adequately modeled using average or representative values until better data become available.

One of the significant challenges encountered here was choosing the correct parameters for modeling nanomaterial CFs, largely stemming from inconsistency in empirical studies from which these values were drawn. For example, LD50 values were obtained from studies applying widely varying methods for nanomaterial detection and measurement, toxicity assay controls, test environments, or from selection of relevant values when toxic concentrations exceed the maximum possible prepared concentrations. There are clear opportunities for fundamental ecotoxicity research to employ greater standardization using reference compounds, consistent assay approaches, reporting open data on physical and chemical properties of reference compounds and test materials, and including uncertainty analysis (Babbitt and Moore 2018). Some of these issues may be addressed via high-throughput screening which allows rapid toxicity testing in vitro, controlling for multiple variables. Making parameter assumptions was a particular challenge for nano-Ag given its partial solubility and the variability in approaches used to model dissolution and differentiate nano-Ag toxicity from ionic silver toxicity (Gaiser et al. 2012). Ongoing debate is seen in the literature regarding best practices for modeling and communicating nano-Ag toxicity. In general, across the field of nanomaterial toxicity model development, the documentation of physicochemical properties, test conditions, and limitations should be provided with the characterization factor, so the impact assessor can decide which toxicity values are most appropriate for the particular conditions of the assessment.

While these results focus on ecotoxicity produced on a per kilogram basis, they can be scaled to the appropriate amount used in a product and account for material-enabled benefits or expanded functionalities in products containing these case study nanomaterials. As many different nanomaterials could potentially be used for the same clean energy application (e.g., both SWNT and C60 can be used in the active layer of an organic photovoltaic cell), these results can be used to inform the trade-offs of material selection using life cycle assessment and uncertainty analysis. Future work should examine the broader context of the increasing demand for nanomaterials and the larger scale of risks associated with the adoption of nanomaterials in new technologies and products, particularly as it relates to potential for regulatory frameworks as a solution to minimize these risks.

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Compliance with ethical standards

Conflict of interest The authors declare no competing financial interest.

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