Developing Anticipatory Life Cycle Assessment

Tools to Support Responsible Innovation

by

Benjamin Wender

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Graduate Supervisory Committee:

Thomas Seager, Chair
David Guston
Paul Westerhoff

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ABSTRACT

Several prominent research strategy organizations recommend applying life cycle assessment (LCA) early in the development of emerging technologies. For example, the US Environmental Protection Agency, the National Research Council, the Department of Energy, and the National Nanotechnology Initiative identify the potential for LCA to inform research and development (R&D) of photovoltaics and products containing engineered nanomaterials (ENMs). In this capacity, application of LCA to emerging technologies may contribute to the growing movement for responsible research and innovation (RRI). However, existing LCA practices are largely retrospective and ill-suited to support the objectives of RRI. For example, barriers related to data availability, rapid technology change, and isolation of environmental from technical research inhibit application of LCA to developing technologies. This dissertation focuses on development of anticipatory LCA tools that incorporate elements of technology forecasting, provide robust explorations of uncertainty, and engage diverse innovation actors in overcoming retrospective approaches to environmental assessment and improvement of emerging technologies. Chapter one contextualizes current LCA practices within the growing literature articulating RRI and identifies the optimal place in the stage gate innovation model to apply LCA. Chapter one concludes with a call to develop anticipatory LCA – building on the theory of anticipatory governance – as a series of methodological improvements that seek to align LCA practices with the objectives of RRI.

Chapter two provides a framework for anticipatory LCA, identifies where research from multiple disciplines informs LCA practice, and builds off the

recommendations presented in the preceding chapter. Chapter two focuses on crystalline and thin film photovoltaics (PV) to illustrate the novel framework, in part because PV is an environmentally motivated technology undergoing extensive R&D efforts and rapid increases in scale of deployment. The chapter concludes with a series of research recommendations that seek to direct PV research agenda towards pathways with the greatest potential for environmental improvement.

Similar to PV, engineered nanomaterials (ENMs) are an emerging technology with numerous potential applications, are the subject of active R&D efforts, and are characterized by high uncertainty regarding potential environmental implications. Chapter three introduces a Monte Carlo impact assessment tool based on the toxicity impact assessment model USEtox and demonstrates stochastic characterization factor (CF) development to prioritize risk research with the greatest potential to improve certainty in CFs. The case study explores a hypothetical decision in which personal care product developers are interested in replacing the conventional antioxidant niacinamide with the novel ENM C_{60}, but face high data uncertainty, are unsure regarding potential ecotoxicity impacts associated with this substitution, and do not know what future risk-relevant experiments to invest in that most efficiently improve certainty in the comparison. Results suggest experiments that elucidate C_{60} partitioning to suspended solids should be prioritized over parameters with little influence on results. This dissertation demonstrates a novel anticipatory approach to exploration of uncertainty in environmental models that can create new, actionable knowledge with potential to guide future research and development decisions.

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CHAPTER 1

BACKGROUND, MOTIVATION, AND SUMMARY OF APPROACH

Several prominent research strategy organizations recommend applying life cycle assessment (LCA) early in the development of emerging technologies. For example, the US Environmental Protection Agency, (EPA 2012) the National Research Council, (NRC 2012) the Department of Energy, (DOE 2012) and the National Nanotechnology Initiative (NNI 2011) identify the potential for LCA to inform research and development (R&D) of photovoltaics and nanomaterial (NM)-enabled products. LCA is increasingly recognized as the proper framework for environmental assessment of products and technologies, because the broad boundaries called for prevent shifting of environmental burdens from one life cycle phase or impact category to another (Curran 2004; Klopffer 2007; Eason 2011). Applying LCA early in the development of emerging technologies may identify environmentally problematic processes before significant investments are made in R&D and commercialization (Liebowitz and Margolis 1995; Theis, Bakshi et al. 2011). In this capacity, LCA offers a potential tool through which future environmental impacts of emerging technologies may be anticipated and environmental criteria integrated into R&D decisions.

I. The Growing Case for Responsible Innovation

Historically, the potential environmental impacts of emerging technologies have not been anticipated, but rather identified, regulated, and mitigated only *after* large-scale production and dissemination (Davies 2009). R&D advancement without consideration of potential future environmental impacts is problematic for at least three reasons:

1. Much of the environmental impact of an emerging technology becomes locked in by early R&D decisions and subsequent investments (Bhander, Hauschild et al. 2003),
2. In the later phases of technology development there is little flexibility for environmental considerations to redirect the innovation process (Stilgoe, Owen et al. 2013), and
3. Separation of environmental concerns from technology development can result in hidden liabilities and costs only identified by retrospective assessment and regulation (Owen and Goldberg 2010).

An alternative model is to promote innovation practices that integrate and are responsive to broader environmental concerns identified by applying environmental assessment tools like LCA early technology R&D. For example, LCA may be applied during prototyping activities early in the stage-gate model of innovation – in which technologies are advanced to increasing stages of readiness through 'gates' only when specific criteria are met. It is increasingly recognized that design criteria drawn from environmental objectives may structure interventions in the nascent stages of development that are more effective than retrospective approaches (Owen, Baxter et al. 2009). This alternative model is consistent with the growing literature describing Responsible Research and Innovation as the process through which "…innovators become mutually responsive…with a view on the (ethical) acceptability, sustainability, and societal desirability of the innovation process and its marketable products…" (Schomberg 2012)" – a vision that is gaining recognition in European and US research institutions (EC 2013; Guston, Fisher et al. 2014). While responsible innovation is an intuitively worthwhile goal, there is a lack of practical tools that make the abstract concept tangible to government, academic, and industry actors involved in innovation processes.

II. Life Cycle Assessment as a Practical Tool for Responsible Innovation

LCA offers one possible tool to support environmentally responsible innovation, yet at present there is little formal discussion of LCA in the context of RRI. To promote responsible innovation, LCA must support the pillars of responsible innovation: 1) anticipation as the process of imagining potential future environmental impacts and building capacity to address them today, 2) engagement with a broad set of actors, stakeholders, and disciplinary perspectives to broaden the range of values and perspectives considered, 3) integration as the process of incorporating environmental criteria into R&D decisions to allow 4) reflexivity of the innovation process by which the technology trajectory can be redirected (Stilgoe, Owen et al. 2013). However, current practices in LCA do not promote these components of responsible innovation, and LCA of emerging technologies faces numerous methodological barriers that diminish the efficacy of LCA for responsible innovation. LCA historically has been applied to established industries (Hunt, Franklin et al. 1996), is data intensive, and portrays near-certain knowledge of fate, transport, and toxicity data for emitted substances. In the context of emerging technologies, data is scarce, proprietary, uncertain, and not representative of eventual commercial processes. As a result LCA has been largely ineffective at redirecting emerging technology development (Seager and Linkov 2009).

III. Making LCA Anticipatory for Responsible Innovation

LCA of emerging technologies proceeds in the context of high uncertainty, which pervades all steps defined in international standards (ISO 2006) and renders codified approaches unreliable. Critical uncertainties include:

1. Predicting a use-phase relevant functional unit that captures the benefits of the emerging technology given uncertain performance, market adoption, and consumer behavior (Wender and Seager 2011; Miller and Keoleian 2015),
2. Uncertainty in extrapolating laboratory scale manufacturing inventory data to commercial scale production volumes (Gutowski, Liow et al. 2010; Gavankar, Suh et al. 2014),
3. Uncertainty regarding potential releases and impacts associated with direct exposure to emerging materials in the environment or workplace (Oberdörster, Oberdörster et al. 2005; Wiesner, Lowry et al. 2006; Oberdörster, Stone et al. 2007; Benn and Westerhoff 2008; Kiser, Westerhoff et al. 2009; Zalk, Paik et al. 2009).

To date LCA researchers have made piecemeal advances that address specific barriers, but these advances have yet to be integrated into a comprehensive framework. For example, Wender, Foley et al. (2012) demonstrate how thermodynamic modeling can be used to explore potential improvement in manufacturing process efficiency that may accrue with increased scale and experience, yet these analyses focusing on life cycle inventory modeling do not inform impact assessment of emerging contaminants. Similarly, Eckelman, Mauter et al. (2012) demonstrate a scenario-based approach to developing aquatic ecotoxicity characterization factors – which convert the mass of material released to damages caused in the environment through coupled transport and toxicology models (Hauschild, Huijbregts et al. 2008) – using the impact assessment tool USETox (Rosenbaum, Bachmann et al. 2008) for nanomaterials, yet focus on impact assessment alone excludes use-phase performance modeling or release scenarios. To address these barriers simultaneously, there is a critical need for these isolated advances to be integrated into a comprehensive framework capable of prospectively relating functional benefits afforded by an emerging technology with potential life cycle damages including release and direct exposure to emerging contaminants. This approach may promote environmentally responsible innovation by embracing uncertainty, anticipating potential future environmental tradeoffs, and engaging diverse actors including R&D decision makers, environmental risk researchers, and social scientists studying broader behavioral, market, and political drivers of technology development.

This dissertation details development of an *anticipatory* LCA framework that incorporates elements of technology forecasting, provides robust explorations of uncertainty, and engages innovation actors in overcoming retrospective approaches to environmental assessment and regulation. Anticipatory LCA seeks to provide environmental criteria to R&D decision makers in order to broaden the range of values used in formulating hypothesis and experimental research agenda, and thereby support responsible innovation of emerging technologies. In this capacity, anticipatory LCA is a tool used to advance innovation, as opposed to retrospective LCA which emphasizes optimization within existing regulations (see Figure 1, Paper 1 below). The framework builds on previous advances in LCA and structures interdisciplinary interactions and knowledge creation to address the high uncertainty associated with emerging technologies. Specifically, anticipatory LCA identifies contributions from technology developers, environmental risk researchers, and social scientists, maps these inputs to critical LCA modeling decisions, and identifies three intervention points where knowledge feedback may inform innovation actors (see Figure 2, Paper 2 below). For example, the anticipatory LCA framework calls for engagement with environmental risk researchers to conduct probabilistic impact assessment for emerging contaminants such as NMs, the results of which can identify those NM parameters most influential to LCA results (see Figure 3, Paper 3 below). Communicating these parameters to research funders and environmental researchers can prioritize experiments with the greatest potential to reduce uncertainty in the life cycle environmental impacts of an emerging material, thereby conserving research resources.

IV. Summary and Synthesis of Research Papers

This dissertation consists of three related research papers, each focused on developing anticipatory LCA methods that promote responsible innovation but exploring the topic with different boundaries and scales. Research paper one (RP1) is the broadest, contextualizes LCA within the growing field of Responsible Research and Innovation, and draws a distinction between retrospective and anticipatory LCA with an emphasis on the innovation actors engaged by each approach. RP1 surveys conceptual barriers that cause misalignment between existing LCA methods and the goals of responsible innovation. Research paper two (RP2) draws narrower boundaries, and focuses on the specific emerging technology of photovoltaics by extending the conceptual discussion of barriers from RP1 to practical barriers faced in LCA of emerging PV devices. RP2 introduces a generalizable framework for anticipatory LCA that incorporates piecemeal methods advancements and structured interdisciplinary collaboration to address critical uncertainties. RP2 identifies three intervention points through which anticipatory LCA may promote responsible innovation practices, one of which is explored in detail in research paper 3 (RP3). RP3 draws the narrowest boundaries, focusing on development of novel approaches for probabilistic characterization factor development for two commercially-relevant nanomaterials. RP3 modifies the consensus impact assessment method USETox, which generates human and ecotoxicity characterization factors based on material parameters and toxicology data, to include mechanisms influential to NM fate and transport, operate with probabilistic ranges rather than point-value estimates, and identify the material parameters most influential to impact assessment results. Each research paper is described in greater detail below, and taken together the dissertation demonstrates the necessity, approach, and potential for anticipatory LCA to guide responsible innovation of emerging technologies.

Table 1: Research questions, tasks, and deliverables for chapter 2

Research question	How do current practices in LCA support, and/or fail to support, the objectives of RRI?
Research question	To support RRI, what is the appropriate position in stage gate innovation to apply LCA, and what actors should LCA target?
Task	Literature review of LCA applied to emerging technologies with emphasis on advances and barriers, and review of stated principals of RRI related to LCA
Task	Develop simplified model of stage-gate innovation process and relevant actors engaged by LCA.
Deliverable	Peer reviewed journal article in *Journal of Responsible Innovation*
Intellectual Merit	Identification of prototyping stage as appropriate for application of LCA for RRI and description of how actors involved in prototyping can use LCA.

Abstract: The goal of guiding innovation toward beneficial social and environmental outcomes—referred to in the growing literature as responsible research and innovation (RRI)—is intuitively worthwhile but lacks practicable tools for implementation. One potentially useful tool is life cycle assessment (LCA), which is a comprehensive framework used to evaluate the environmental impacts of products, processes, and technologies. However, LCA ineffectively promotes RRI for at least two reasons: 1) Codified approaches to LCA are largely retrospective, relying heavily on data collected from mature industries with existing supply chains, and 2) LCA underemphasizes the importance of stakeholder engagement to inform critical modeling decisions which diminishes the social credibility and relevance of results. LCA researchers have made piecemeal advances that address these shortcomings, yet there is no consensus regarding how to advance LCA to support RRI of emerging technologies. This paper advocates for development of *anticipatory* LCA as non-predictive and inclusive of uncertainty, which can be used to explore both reasonable and extreme-case scenarios of future environmental burdens associated with an emerging technology. By identifying the most relevant uncertainties and engaging research and development (R&D) decision-makers, such anticipatory methods can generate alternative research agenda and provide a practicable tool to promote environmental RRI.

Figure 1: Intervention Points for LCA and Relevant Actors as Technology Readiness Increases

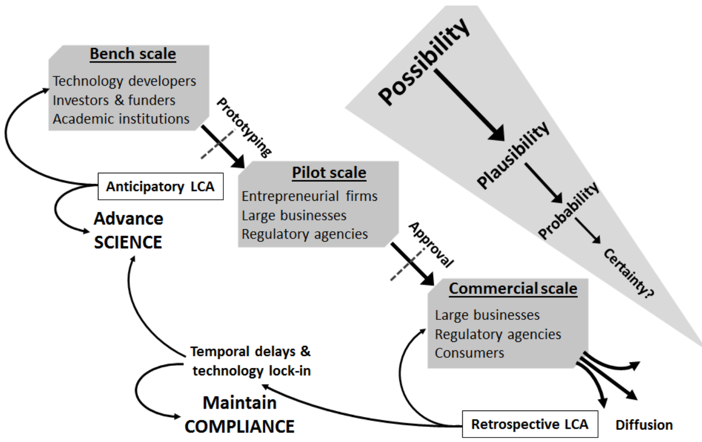

Figure 1. Applying LCA earlier in stage-gate innovation overcomes temporal delays and technology lock-in limiting retrospective LCA, and thereby has greater potential to reorient technology development

Table 2: Research question, task, and deliverable for chapter 3

Research question	Where do physical, environmental, and social science disciplines inform LCA practice and how do these disciplinary perspectives contribute to or reduce uncertainty in LCA models?
Task	Influence diagraming to create a "knowledge map" positioning existing tools and contributions from physical, environmental, and social sciences into LCA framework
Deliverable	Peer reviewed journal article in *Environmental Science & Technology*
Intellectual Merit	A generalizable framework for anticipatory LCA

Abstract: Current research policy and strategy documents recommend applying life cycle assessment (LCA) early in research and development (R&D) to guide emerging technologies toward decreased environmental burden. However, existing LCA practices are ill-suited to support these recommendations. Barriers related to data availability, rapid technology change, and isolation of environmental from technical research inhibit application of LCA to developing technologies. Overcoming these challenges requires methodological advances that help identify environmental opportunities prior to large

R&D investments. Such an *anticipatory* approach to LCA requires synthesis of social, environmental, and technical knowledge beyond the capabilities of current practices. This paper introduces a novel framework for anticipatory LCA that incorporates technology forecasting, risk research, social engagement, and comparative impact assessment, then applies this framework to photovoltaic (PV) technologies. These examples illustrate the potential for anticipatory LCA to prioritize research questions and help guide environmentally responsible innovation of emerging technologies.

Figure 2: A Framework for Anticipatory LCA of Emerging Technologies

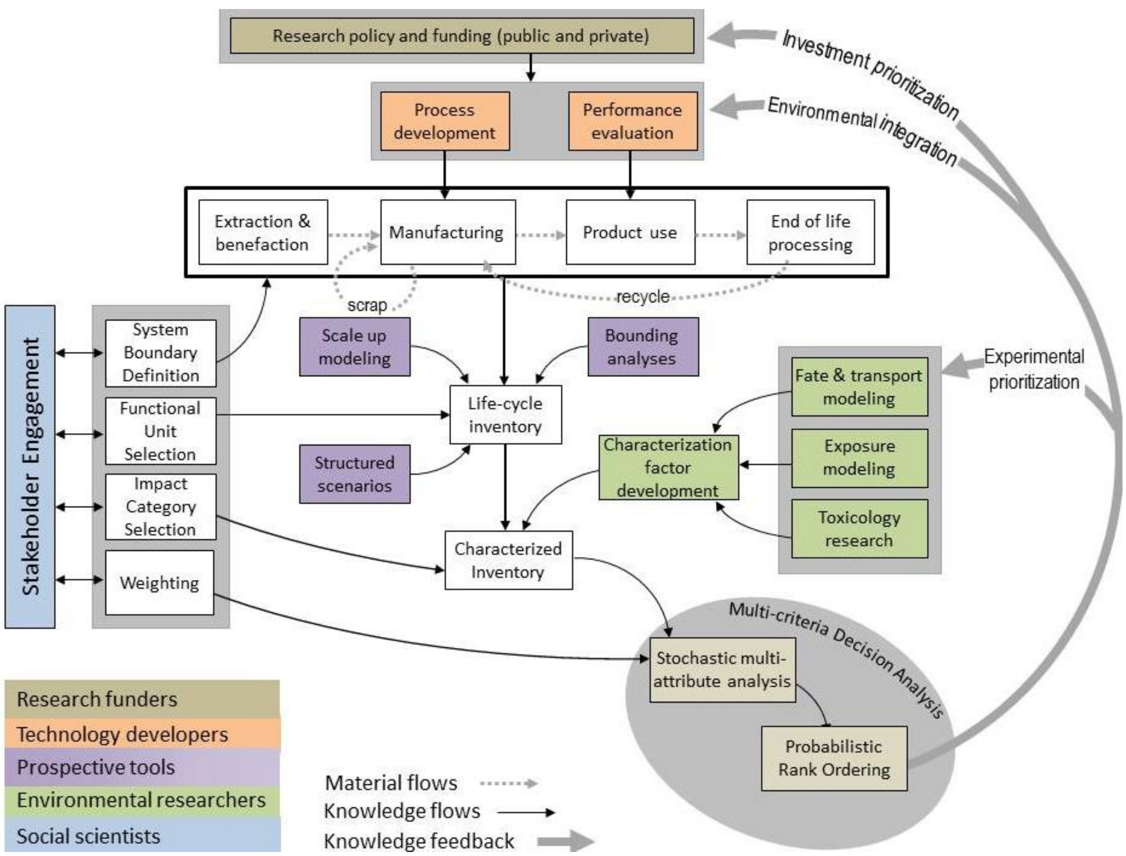

Figure 2. Anticipatory LCA structures interdisciplinary interactions and environmental interventions early in R&D. White boxes present current practices in LCA, which emphasize materials flows and feedback, whereas anticipatory LCA emphasizes knowledge flows and feedback.

Table 3: Components of Chapter 4 – Sensitivity-based research prioritization through stochastic characterization modeling

Research question	Can stochastic characterization factor (CF) development prioritize laboratory research of risk-relevant parameters?
Sub question 1	What material parameters are most influential to USETox CF results for each NM case study?
Task	Expert solicitation and meta-analysis of review articles to build parameter distributions and summarize disconnect between specific parameters used in USETox and actual environmental behavior of NMs
Task	Develop a Monte Carlo program within USETox and conduct global sensitivity analysis to identify those material parameters most influential for aquatic ecotoxicity CFs for case study NMs
Deliverable	Peer reviewed publication in *International Journal of Life Cycle Assessment* Modified stochastic USETox tool made freely available on Nanohub.org
Intellectual Merit	New probabilistic CFs for two commercially-relevant NMs Identification of material parameters most influential to CF results

Abstract Large data requirements, high uncertainty and complexity, and regulatory relevance of toxicity impact assessment motivates greater focus on model sensitivity toward input parameter variability. This is particularly useful for emerging contaminants like engineered nanomaterials (ENMs) to guide future efforts in data refinement and design of experiments. This study presents a Monte Carlo tool based on USEtox 1.0 that allows researchers to rapidly prioritize data needs according to influence on characterization factors (CFs). Using Monte Carlo analysis we demonstrate a sensitivity-based approach to prioritize research through a case study comparing aquatic ecotoxicity CFs calculated with USEtox 1.01 for the ENM C_{60} and the vitamin B derivative niacinamide, two antioxidants used in personal care products. We calculate CFs via 10,000 iterations assuming plus-or-minus one order of magnitude variance for fate and exposure-relevant inputs. Spearman Rank Correlation Indices are used for all variable

inputs to identify parameters with the largest influence on CFs, which we prioritize for data refinement and future experimental investigation. Based on the importance of aggregate multi-species toxicity (average log EC_{50}) and studies suggesting solvent residues may yield erroneous toxicity estimates, we recalculate C_{60} CFs omitting all studies using solvents in sample preparation.

For emissions to freshwater, the C_{60} CF is log-normally distributed with a geometric mean of 280 (geometric standard deviation, GSD: 2.1) PAF m^3 day/kg compared to 2.6 (GSD: 1.8) PAF m^3 day/kg for niacinamide. C_{60} CFs are most sensitive to varied suspended solids partitioning coefficients (Kpss) and average log EC_{50}, whereas variation of other substance parameters has comparatively little effect on model results. Insufficient experimental evidence hampers to revise assumptions for Kpss, and we suggest prioritizing future experiments that elucidate C_{60} interactions with suspended solids. Recalculating C_{60} CFs without toxicity studies that use solvents reduces the geometric mean by more than a factor of ten. This reinforces the importance of thorough source term characterization, in this case regarding the presence of solvent residues. Calculating stochastic CFs allows sensitivity-based prioritization of data needs and future experiments, which is particularly helpful in the context of emerging contaminants like C_{60}. Researchers can conserve resources and address parameter uncertainty by applying our approach when developing new or refining existing CFs for the inventory items that contribute most to toxicity impacts. The Monte Carlo tool can be applied to current toxicity characterization models like USEtox and is freely available

Figure 3: Relative Influence of Material Parameters input to USETox on CF Uncertainty

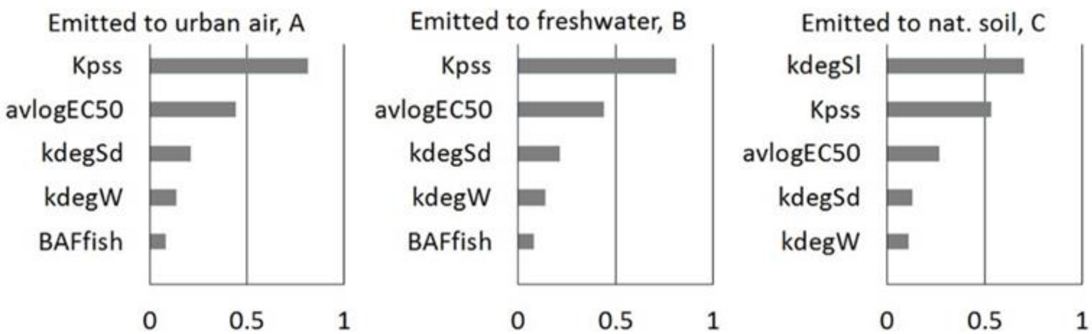

Fig 3 The five Spearman rank correlation indices with the greatest magnitude out of all variable inputs for three C_{60} aquatic ecotoxicity CFs. Greater magnitude indicates which input parameters have the greatest influence on CFs variability for each emission compartment.

CHAPTER 2

ANTICIPATORY LIFE CYCLE ASSESSMENT FOR RESPONSIBLE RESEARCH AND INNOVATION

Anticipatory Life Cycle Assessment for Responsible Research and Innovation

Ben A. Wender[1,2]*, Rider W. Foley[2], Troy A. Hottle[1], Jathan Sadowski[3], Valentina Prado-Lopez[1], Dan A. Eisneberg[1], Lise Laurin[4], and Thomas P. Seager[1]

[1]School of Sustainable Engineering and the Built Environment, Arizona State University, Tempe, AZ

[2]Center for Nanotechnology in Society, Arizona State University, Tempe, AZ

[3]Consortium for Science Policy & Outcomes, Arizona State University, Tempe, AZ

[4]Earthshift LLC, 31 Leach Road, Kittery ME 03904

*Corresponding author: bwender@asu.edu

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Anticipatory Life Cycle Assessment for Responsible Research and Innovation

Abstract

The goal of guiding innovation toward beneficial social and environmental outcomes—referred to in the growing literature as responsible research and innovation (RRI)—is intuitively worthwhile but lacks practicable tools for implementation. One potentially useful tool is life cycle assessment (LCA), which is a comprehensive framework used to evaluate the environmental impacts of products, processes, and technologies. However, LCA ineffectively promotes RRI for at least two reasons: 1) Codified approaches to LCA are largely retrospective, relying heavily on data collected from mature industries with existing supply chains, and 2) LCA underemphasizes the importance of stakeholder engagement to inform critical modeling decisions which diminishes the social credibility and relevance of results. LCA researchers have made piecemeal advances that address these shortcomings, yet there is no consensus regarding how to advance LCA to support RRI of emerging technologies. This paper advocates for development of *anticipatory* LCA as non-predictive and inclusive of uncertainty, which can be used to explore both reasonable and extreme-case scenarios of future environmental burdens associated with an emerging technology. By identifying the most relevant uncertainties and engaging research and development (R&D) decision-makers, such anticipatory methods can generate alternative research agenda and provide a practicable tool to promote environmental RRI.

Keywords: Anticipation, Technology Assessment, Foresight, Knowledge Integration

Introduction

Potential environmental impacts of emerging technologies are often only identified, regulated, and mitigated *after* large-scale production and dissemination (Davies 2009). Early research and development (R&D) suffers from a lack of integration of environmental research. This is problematic for at least three reasons: 1) many of the environmental impacts caused by a technology become locked-in by R&D decisions (Bhander, Hauschild et al. 2003); 2) in the early phases of technology development there exists greater flexibility for environmental considerations to guide the innovation process (Stilgoe, Owen et al. 2013); and 3) the separation of environmental research from technology development positions assessment and regulation as retrospective and reactive (Owen and Goldberg 2010). An alternative model is to integrate broader criteria into technology development (Fisher and Rip 2013). Rather than rely on retrospective approaches, design criteria explicitly drawn from social and environmental values can structure more effective interventions in the nascent stages of technology development, and thereby promote responsible research and innovation (RRI) practices (Owen, Baxter et al. 2009). However, there is a paucity of practicable design tools that effectively integrate broader values into technology R&D. This paper argues for the development of *anticipatory* life cycle assessment (LCA) methods as one tool to promote integration of environmental criteria early in the stage-gate innovation model and support the broader goals of RRI. Anticipatory LCA will be a collection of best practices from existing prospective studies as well as new methods, codified into a single, cohesive, easy-to-follow methodology.

1. *Life Cycle Assessment and its Discontents*

LCA—a comprehensive framework for evaluating environmental impacts of processes, products, or technologies—is the preferred analytic framework for environmental assessment because the broad boundaries used prevent the shifting of environmental burdens from one life cycle phase or environmental compartment to another. For example, the rapid growth in production of corn-derived ethanol was partially justified by amelioration of greenhouse gas emissions. However, the narrow policy focus on mitigation of climate change came at the expense of increased eutrophication impacts—a tradeoff easily identified by LCA (Miller, Landis et al. 2006).

To reduce the likelihood of unintended environmental consequences, research policy organizations increasingly recommend application of LCA to emerging technologies (NRC 2012). Implicit in such calls is a desire to foster environmental RRI by identifying potential impacts before commercial scale production and technology diffusion. However, traditional approaches to environmental LCA ineffectively promote RRI of emerging technologies for at least two reasons: 1) Codified practices rely extensively on data collected from mature industries with existing supply chains and are thereby largely retrospective, and 2) Established practices underemphasize the importance (and oversimplify the process) of stakeholder engagement in shaping LCA models and results, and thereby suffer diminished social credibility and relevance. Regarding the first point, there has been isolated progress in advancing LCA methods towards prospective identification and mitigation of environmental impacts, yet these tools have not been integrated into a comprehensive framework that supports RRI of emerging technologies. Regarding the second point, this manuscript emphasizes the importance of inclusion of diverse stakeholder values in critical environmental LCA modeling decisions, which may identify a need to generate multiple LCA models based on what values are included. Overcoming these barriers builds capacity for LCA to engage R&D decision makers with broader environmental values and provides a tool that contributes to environmental RRI of emerging technologies.

2. *From Retrospective to Prospective LCA*

Most LCA applications are retrospective in that they occur after commercial scale production by large businesses and distribution to consumers according to laws set by regulatory agencies. Such analyses are useful for informing consumers and regulators about the environmental impacts of a product (e.g., carbon footprints, eco-labeling), yet have limited ability to reorient technology trajectory because temporal delays and large capital investments contribute to technology lock-in (Collingridge 1980). Qualitative approaches such as life cycle thinking (Thabrew, Wiek et al. 2009) can provide useful heuristics early in R&D but lack the quantitative rigor of LCA. To address this shortcoming, a growing literature of *prospective* LCA employs modeling tools that require less accurate datasets and focus analyses on potential environmental impacts arising from R&D decisions. Drawing from diverse fields ranging from future studies to thermodynamics, published advances include incorporation of backcasting (Herwich 2005), foresight tools, and scenario development into LCA and material flow analysis (Pesonen, Ekvall et al. 2000; Spielmann, Scholz et al. 2004; Wender and Seager 2011; Eckelman, Mauter et al. 2012; Dale, Pereira de Lucena et al. 2013; Simon and Weil 2013; Zimmerman, Dura et al. 2013), dynamic LCA process modeling (Collinge, Landis et al. 2013), thermodynamic modeling of manufacturing processes (Gutowski, Branham et al.

2009; Gutowski, Liow et al. 2010), and stochastic decision analysis (Canis, Linkov et al. 2010; Linkov, Bates et al. 2011; Prado-Lopez, Seager et al. 2014). These tools advance LCA methods and call attention to potential future environmental impacts of emerging technologies while early in R&D.

3. *Integrating Societal Values*

Application of LCA early in R&D is insufficient to promote environmental RRI if societal values are not integrated and alternative perspectives explored. Critiques of LCA identify long standing challenges in recognizing where and how to incorporate stakeholder value preferences into environmentally-focused analysis (Berube 2013), which increases the social credibility and relevance of LCA results. Inclusion of stakeholder values in environmental LCA is distinct from the rapidly expanding field of social life cycle assessment (S-LCA), which quantifies burdens in defined social impact categories such as child labor and indigenous rights (UNEP 2013) or life cycle sustainability assessment (LCSA) (Guinee, Heijungs et al. 2011), which entails concurrent application of LCA, S-LCA, and life cycle costing to identify environmental, social, and economic impacts respectively. While S-LCA and LCSA have a broader scope than environmental LCA and are designed to explicitly represent social impacts, these tools may suffer from a similar lack of stakeholder engagement to guide model construction.

While stakeholder engagement is discussed in ISO standards for environmental LCA, practitioners typically do not have the requisite training to identify affected parties and elicit the relevant value preferences. There are numerous decisions in environmental LCA that are normative, including: 1) system boundary definition (what activities are included), 2) functional unit selection (what service the technology provides), 3) impact category selection (what environmental impacts are considered), and 4) weighting (how much impacts in one category matter relative to another). As opposed to a practitioner making these decisions in isolation, environmental LCA should employ social science engagement methods to identify impacted stakeholders, elicit their value preferences, and use these numerous – often conflicting – perspectives to inform modeling decisions.

Explicit statement and inclusion of these values may result in several model configurations (e.g., multiple system boundaries or functional units based on what stakeholder values are represented). The process should be iterative and reflexive – for example, system boundary definition influences initial stakeholder identification, what activities are included, and how benefits and impacts are distributed. Conversely, a detailed secondary stakeholder analysis may reveal the need to redefine system boundaries. Rather than ignoring stakeholder differences in an attempt to be unbiased, LCA should explicitly account for these values and biases and provide a tool to quantitatively explore alternative perspectives to complement value sensitive design (Taebi et al. 2014).

4. *Toward Anticipatory LCA for Responsible Research and Innovation*

There is an opportunity to remake LCA as a tool to guide environmentally responsible product innovation by building on prospective modeling advances and exploring multiple configurations of system boundaries, functional units, impact categories, and weights based on modeled stakeholder values. The goal is to create a tool that integrates environmental concerns into the technology development process in a way that anticipates foreseeable negative consequences, identifies opportunities for improving the environmental profile of emerging technologies, and communicates findings to R&D decision-makers in time to reorient research. With this objective, we build upon advances in the domain of anticipatory governance (Guston 2013) – borrowing the terminology to define *anticipatory* LCA as a forward looking, non-predictive tool that increases model uncertainty through inclusion of prospective modeling tools and multiple social perspectives. As opposed to prospective LCA, which treats uncertainty largely as a measure of model reliability, anticipatory LCA should not seek to create a realistic model but rather to expand uncertainty and perform global sensitivity analysis to identify the most environmentally promising research agendas. In this capacity, anticipatory LCA may generate many models all with a high degree of uncertainty in order to explore a broad spectrum of possible futures (as opposed to a select few, most likely) to build capacity to prepare for many potential outcomes. Using anticipatory LCA as a tool not to predict the future, but to prepare for it, provides one approach to contribute to the broader goals of RRI.

Figure 4 illustrates a sequential stage-gate model of increasing market readiness that product innovations typically progress through (Robinson 2009), compares intervention points for retrospective and anticipatory LCA, and lists relevant actors associated with each stage. In early R&D activities (bench scale and prototyping phase), technology developers and research funders from both industry and academia begin to assess the technical performance and financial returns on investment characteristics of the technology (Foley and Wiek 2013). Gates (dotted lines on Figure 1) open and product development proceeds only when specific objectives – typically technical, financial, and legal – are met.

Intervention Points for LCA and Relevant Actors as Technology Readiness Increases

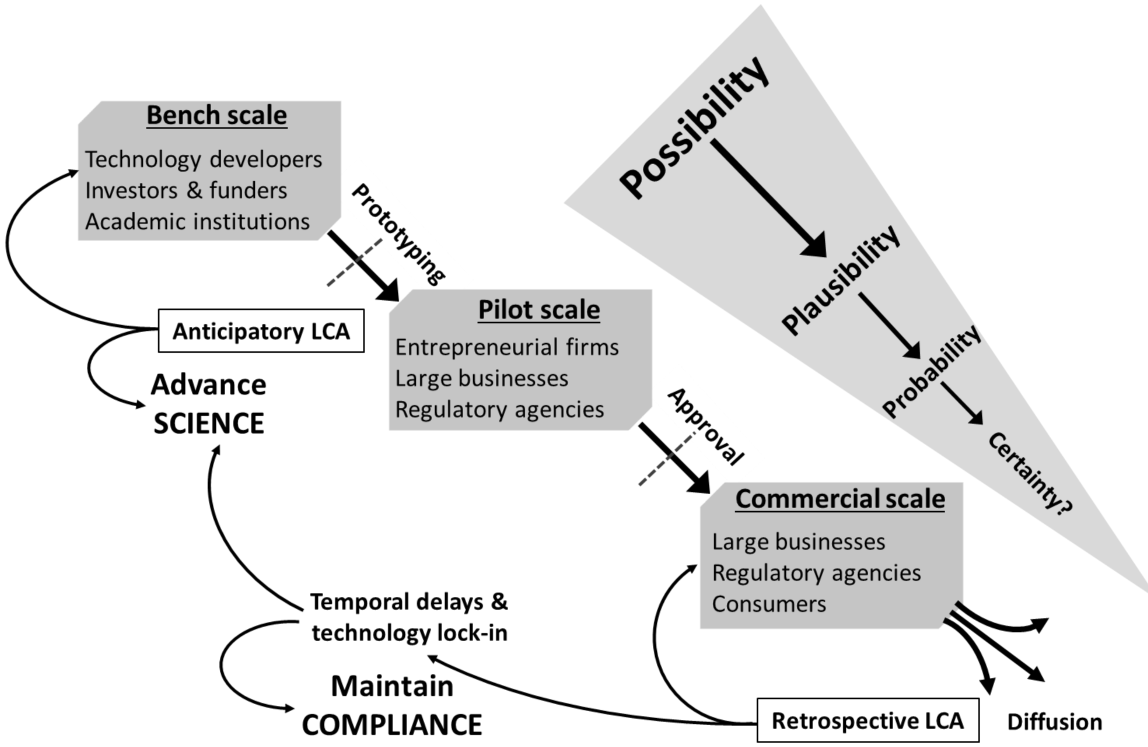

Figure 4. Applying LCA earlier in stage-gate innovation overcomes temporal delays and technology lock-in limiting retrospective LCA, and thereby has greater potential to reorient technology development through integration of broader criteria into bench scale research.

The stage-gate model of product innovation is criticized for considering only technical and economic criteria during laboratory scale research and prototyping activities, whereas broader socio-environmental impacts (albeit highly uncertain) occurs in later stages, if at all (Stilgoe, Owen et al. 2013). Applying LCA after commercial production and diffusion – termed retrospective LCA – filters out unacceptable technologies and serves as a tool to maintain compliance. Alternatively, anticipatory LCA should seek to provide broader environmental criteria early in R&D to promote formulation of new research agenda, and in doing so become a tool that advances science.

The proposed design and assessment tool is not the singular solution to achieve RRI, and significant work remains to develop generalizable methods for anticipatory LCA. Nonetheless, as discussed here, it adds reflexivity earlier into the product innovation process, integrates knowledge from disparate disciplines, is inclusive of broader societal values, and anticipates foreseeable future implications. While not all impacts can be identified or avoided, when implemented in an adaptive approach that leverages continuous learning this tool can aid in reducing negative environmental impacts. In this way anticipatory LCA embodies the core principals of RRI outlined by Stilgoe et al. (2013) and aligns normative goals regarding socio-ecological impacts with von Schomberg's notion of 'acceptability, sustainability and societal desirability' (2013, 64). A diversity of researchers, government agencies, and private organizations can participate in moving this research agenda forward.

5. *Who Can Use Anticipatory LCA*

Anticipatory LCA requires further attention and development as a practicable design tool used to implement environmental RRI into R&D processes. It provides a conceptual model to structure knowledge communication and collaboration between numerous stakeholders and a wide range of actors involved in innovation. *Research funders* could apply anticipatory LCA to systematically and quantitatively generate scenarios of potential impacts arising from alternative investment strategies. As the technology remains in a formative stage of development scenarios can overcome temporal delays by assessing future, broader impacts. This information complements economic and technical metrics to prioritize investment strategies that maximize positive social and environmental outcomes. Physical scientists, engineers, and other technology developers could apply anticipatory LCA to explore potential broader impacts associated with their laboratory research decisions, and could be engaged in structuring R&D activities that are responsive to social and environmental concerns. As a design tool, anticipatory LCA could provide timely feedback to technology developers and inform initial material selection, energy targets, end of life management strategies, maintenance options, and user demands. Social scientists that engage diverse stakeholders and explore the societal implications of emerging technologies could employ anticipatory LCA as a tool with increased technical detail than other foresight methods. Furthermore, this tool could provide an opportunity to integrate social scientists with environmental and technical researchers while yielding holistic metrics of technology trajectories and communicating findings to research funders. Environmental researchers can use anticipatory LCA to prioritize experimental research that will lead to the greatest reductions in uncertainty and most environmental improvement across the life-cycle of emerging technologies. Together, these activities engage a broad spectrum of actors in innovation processes and can contribute to environmental RRI.

References

Berube, David M. 2013. "Socialis Commodis and Life Cycle Analysis: A Critical Examination Of." In *Emerging Technologies: Socio-Behavioral Life Cycle Approaches*, edited by Nora Savage, Michael Gorman, and Anita Street, 139-164. Singapore: Pan Stanford Publishing.

Bhander, Gurbakhash Singh, Michael Hauschild, and Tim McAloone. 2003. "Implementing Life Cycle Assessment in Product Development." *Environmental Progress* 22 (4): 255-67.

Canis, Laure, Igor Linkov, and Thomas P. Seager. 2010. "Application of Stochastic Multiattribute Analysis to Assessment of Single Walled Carbon Nanotube Synthesis Processes." *Environmental Science & Technology* 45 (12): 5068-5074.

Collinge, William O., Amy E. Landis, Alex K. Jones, Laura A. Schaefer, and Melissa Bilec. 2013. "A Dynamic Life Cycle Assessment: Framework and Application to an Institutional Building." *International Journal of Life Cycle Assessment* 18 (3): 538-552.

Collingridge, David. 1980. *The social control of technology*. London: Pinter.

Dale, Alexander T., Andre F. de Lucena, Joe Marriott, Bruno S.M.C. Borba, Roberto Schaeffer, and Melissa Bilec. 2013. "Modeling Future Life-Cycle Environmental Impacts of Electricity Supplies in Brazil." *Energies* 6: 3182-3208.

Davies, J. Clarence. 2009. "Oversight of Next Generation Nanotechnology." Washington, DC: Woodrow Wilson International Center for Scholars.

Eckelman, Matthew J., Meagan S. Mauter, Jacqueline A. Isaacs, and Menachem Elimelech. 2012. "New perspectives on nanomaterial aquatic ecotoxicity: production impacts exceed direct exposure impacts for carbon nanotoubes."*Environmental science & technology* 46(5): 2902-2910.

Fisher, Erik, and Arie Rip. 2013. "Responsible Innovation: Multi-Level Dynamics and Soft Intervention Practices." In *Responsible Innovation: Managing the Responsible Emergence of Science and Innovation in Society,* edited by Richard Owen, John Bessant, and Maggy Heintz, 165-83. West Sussex, UK: John Wiley & Sons, Ltd.

Foley, Rider W., and Arnim Wiek. 2013. "Patterns of Nanotechnology Innovation and Governance within a Metropolitan Area." *Technology in Society* 35 (4): 233-47.

Guinee, Jeroen B., Reinout Heijungs, Gjalt Huppes, Alessandra Zamagni, Paolo Masoni, Roberto Buonamici, Tomas Ekvall, and Tomas Rydberg. 2011. “Life Cycle Assessment: Past, Present, and Future.” *Environmental Science & Technology* 45 (1): 90-96.

Guston, D. 2013. “Understanding Anticipatory Governance”. *Social Studies of Science*, DOI: 0306312713508669.

Gutowski, Timothy G., John Y.H. Liow, and Dusan P. Sekulic. "Minimum Exergy Requirements for the Manufacturing of Carbon Nanotubes." Paper presented at the Sustainable Systems and Technology (ISSST), IEEE International Symposium on, 17-19 May 2010.

Gutowski, Timothy G., Matthew S. Branham, Jeffrey B. Dahmus, Alissa J. Jones, Alexandre Thiriez, and Dusan P. Sekulic. 2009. "Thermodynamic Analysis of Resources Used in Manufacturing Processes." *Environmental Science & Technology* 43(5): 1584-90.

Herwich, Edgar. 2005. “Life Cycle Approaches to Sustainable Consumption: A Critical Review” *Environmental Science & Technology* 39(13): 4673-4684.

Linkov, Igor, Matthew E. Bates, Laure J. Canis, Thomas P. Seager, and Jeffrey M. Keisler. 2011. "A Decision-Directed Approach for Prioritizing Research into the Impact of Nanomaterials on the Environment and Human Health." *Nature Nanotechnology* 6 (12): 784-87.

Miller, Shelie A., Amy E. Landis, and Thomas L. Theis. 2006. "Use of Monte Carlo Analysis to Characterize Nitrogen Fluxes in Agroecosystems." *Environmental Science & Technology* 40 (7): 2324-32.

NRC, National Research Council. 2012. A Research Strategy for Environmental, Health, and Safety Aspects of Engineered Nanomaterials. Edited by Health Committee to Develop a Research Strategy for Environmental and Safety Aspects of Engineered Nanomaterials: The National Academies Press.

Owen, Richard, David Baxter, Trevor Maynard, and Michael Depledge. 2009. "Beyond Regulation: Risk Pricing and Responsible Innovation." *Environmental Science & Technology* 43(18): 6902-06.

Owen, Richard, and Nicola Goldberg. 2010. "Responsible Innovation: A Pilot Study with the U.K. Engineering and Physical Sciences Research Council." *Risk Analysis* 30 (11): 1699-707.

Pesonen, Hanna-Leena, Tomas Ekvall, Günter Fleischer, Gjalt Huppes, Christina Jahn, Zbigniews Klos, Gerald Rebitzer, et al. 2000. "Framework for Scenario Development in LCA." *The International Journal of Life Cycle Assessment* 5(1): 21-30.

Prado-Lopez, Valentina, Thomas P. Seager, Mikhail Chester, Lise Laurin, Melissa Bernardo, and Steven Tylock. 2014. “Stochastic multi-attribute analysis (SMAA) as an interpretation method for comparative life-cycle assessment (LCA)” *The International Journal of Life Cycle Assessment* 19: 405-416.

Robinson, Douglas K. R. 2009. "Co-Evolutionary Scenarios: An Application to Prospecting Futures of the Responsible Development of Nanotechnology." *Technological Forecasting and Social Change* 76(9): 1222-39.

Simon, Balint and Marcel Weil. 2013. “Analysis of Materials and Energy Flows of Different Lithium Ion Traction Batteries.” *Revue de Métallurgie* 110(S): 65-76

Spielmann, Michael, Roland Scholz, Olaf Tietje, and Peter de Haan. 2004. "Scenario Modeling in Prospective Lca of Transport Systems. Application of Formative Scenario Analysis." *The International Journal of Life Cycle Assessment* 10(5): 325-35.

Stilgoe, Jack, Richard Owen, and Phil Macnaghten. 2013. "Developing a Framework for Responsible Innovation." *Research Policy* 42 (9): 1568-80.

Taebi, Benham, Aad Correlje, Edwin Cuppen, Marloes Dignum, and Udo Pesch. 2014. “Responsible Innovation as an Endorsement of Public Values: The Need for Interdisciplinary Research.” *Journal of Responsible Innovation* DOI: 10.1080/23299460.2014.882072

Thabrew, Lanka, Arnim Wiek, and Robert Ries. 2009. “Environmental Decision Making in Multi-stakeholder Contexts: Applicability of Life Cycle Thinking in Development Planning and Implementation.” *Journal of Cleaner Production* 17(1): 67-76.

UNEP, United Nations Environmental Programme. 2013. “The Methodological Sheets for Subcategories in Social Life Cycle Assessment (S-LCA).”

von Schomberg, Rene. 2013. “A Vision of Responsible Research and Innovation.” In *Responsible Innovation: Managing the Responsible Emergence of Science and Innovation in Society*, edited by Richard Owen, John Bessant, and Maggy Heintz, 165-83. West Sussex, UK: John Wiley & Sons, Ltd.

Wender, Ben, Rider W. Foley, Thomas P. Seager, David H. Guston, and Arnim Wiek. (2012-2013). “Anticipatory Governance and Anticipatory Life Cycle Assessment of Single Wall Carbon Nanotube Anode Lithium Ion Batteries.” *Journal of Nanotechnology Law and Business* 9(3): 201-216.

Wender, Ben and Thomas P. Seager. 2011. “Towards Prospective Life Cycle Assessment: Single Wall Carbon Nanotubes for Lithium-ion Batteries.” International Symposium on Sustainable Systems and Technology, Chicago, IL 16-18 May.

Zimmermann, Benedikt, Hanna Dura, Manuel Baumann, and Marcel Weil. 2013. "Towards prospective time-resolved LCA of fully electric supercap-vehicles in Germany" *19th SETAC LCA Case Study Symposium.* Rome, Italy, December 11.

CHAPTER 3

ILLUSTRATING ANTICIPATORY LIFE CYCLE ASSESSMENT FOR EMERGING PHOTOVOLTAIC TECHNOLOGIES

Ben A. Wender,[1,2,3]* Rider W. Foley,[2] Valentina Prado-Lopez,[1] Dwarakanath Ravikumar,[1] Daniel A. Eisenberg,[1] Troy A. Hottle,[1] Jathan Sadowski,[4] William P. Flanagan,[5] Angela Fisher,[5] Lise Laurin,[6] Matthew E. Bates,[7] Igor Linkov,[7] Thomas P. Seager,[1,3] Matthew P. Fraser[1,3] and David H. Guston[2]

[1]School of Sustainable Engineering and the Built Environment, Arizona State University (ASU)

[2]Center for Nanotechnology in Society, ASU

[3]Quantum Energy and Sustainable Solar Technologies, NSF-DOE Engineering Research Center, ASU

[4]Consortium for Science, Policy & Outcomes, ASU

[5]Ecoassessment Center of Excellence, General Electric Company, Niskayuna NY

[6]EarthShift LLC, 31 Leach Road, Kittery, ME 03904

[7]US Army Engineer Research and Development Center, US Army Corps of Engineers

*Corresponding author: bwender@asu.edu

Abstract

Current research policy and strategy documents recommend applying life cycle assessment (LCA) early in research and development (R&D) to guide emerging technologies toward decreased environmental burden. However, existing LCA practices are ill-suited to support these recommendations. Barriers related to data availability, rapid technology change, and isolation of environmental from technical research inhibit application of LCA to developing technologies. Overcoming these challenges requires methodological advances that help identify environmental opportunities prior to large R&D investments. Such an *anticipatory* approach to LCA requires synthesis of social, environmental, and technical knowledge beyond the capabilities of current practices. This paper introduces a novel framework for anticipatory LCA that incorporates technology forecasting, risk research, social engagement, and comparative impact assessment, then applies this framework to photovoltaic (PV) technologies. These examples illustrate the potential for anticipatory LCA to prioritize research questions and help guide environmentally responsible innovation of emerging technologies.

Introduction

Research strategy and policy documents published by multiple organizations[1,2,3,4] recommend applying life cycle assessment (LCA) early in the development of emerging technologies such as photovoltaics and nanotechnology. These calls envision LCA as a tool to provide research and development (R&D) decision-makers with environmental guidance for consideration alongside technical and economic measures of technology readiness. In this capacity, LCA could proactively identify environmental opportunities and reorient research trajectories prior to significant investments in product scale-up and commercial dissemination. However, there are at least four critical challenges that make LCA ineffective in the context of emerging technologies: 1) Manufacturing and emissions databases rely on historical data collected predominantly from mature industries, 2) Current practices underemphasize the importance of engaging stakeholders to inform critical modeling decisions, 3) Impact assessment tools lack quantitative data describing the fate, transport, and toxicity of novel substances, and 4) Existing approaches to interpretation of comparative LCA results with high uncertainty present unresolved multi-criteria problems. Fulfilling the aforementioned expectations for application of LCA to guide R&D of emerging technologies requires methodological advances and interdisciplinary collaboration beyond the scope of existing practices. Amongst other challenges faced by LCA practitioners, this paper explores these four as they arose from efforts to use LCA proactively in a large multi-disciplinary photovoltaic research center, introduces a framework designed to help LCA practitioners overcome them, and applies the proposed framework to examine photovoltaic technologies.

1.1 Making LCA Prospective

The inherent lack of data across the life cycle of emerging technologies contributes to high uncertainty[5] that renders automated approaches impracticable and potentially misleading. For example, current LCA practices often rely on point-value estimates for data including manufacturing and emissions inventories, characterization factors that convert masses emitted into the potential impacts they cause, weights used to aggregate impacts across diverse impact categories into a single-score indicator, and normalization references used to contextualize the magnitude of reported impacts. These practices are inappropriate for prospective assessment of the environmental implications of emerging technologies, where parameter uncertainties are compounded by scenario and model uncertainty.[6,7] For emerging technologies, such as cutting-edge photovoltaics (PV) that are experiencing rapid rates of innovation even as they mature, critical data are unknown or highly uncertain, including: technology-specific commercial-scale manufacturing inventories, use-phase product performance, end-of-life disposal pathways, life cycle material releases, and risk-relevant properties are uncertain or entirely unknown. This challenge is distinct from traditional data quality issues that beguile LCA of emerging and established technologies alike in that no amount of increased effort in inventory data collection will yield representative data sets. In emerging technology cases,[8] LCA practitioners have responded with a number of strategies including: developing structured scenarios within LCA models,[9,10] thermodynamic process modeling,[11] consideration of experience curves from analogous industries to identify potential future improvements in efficiency,[12] dimensional analysis to explore scaling effects,[13] exploring market-driven impacts through consequential

LCA,[14] and uncertainty bounding analyses to provide upper and lower limits to environmental impact.[15] These advances – often grouped under the term prospective LCA[16,17] – allow the development of life cycle inventories descriptive of future technological developments.

1.2 Supporting Social Engagement

Accurate and meaningful inclusion of stakeholder values in environmental LCA is distinct from social LCA – which quantifies social impacts in defined categories such as human rights[18] – and little guidance exists with regard to emerging technologies. Environmental LCA is typically conducted without efforts to engage stakeholders on broader issues including public perception, behavioral responses to new technologies, and stakeholder priorities that inform modeling and interpretation of results. As a result, LCA practitioners make normative modeling decisions[19,20] that may overlook impacted parties or privilege one stakeholder perspective. These assumptions are sources of scenario uncertainty that often go unexplored, which is particularly important for emerging technologies because assumptions made about market adoption and user behavior may determine results. Critical modeling decisions in LCA including system boundary definition, functional unit selection, impact category selection, and weight determination all have social consequences that should be made explicit to systematically explore scenario uncertainty. To this end, application of LCA to emerging technologies must include stakeholder engagement activities[21] that inform these modeling decisions. Workshops facilitated by social scientists should bring a diversity of actors across the technology life cycle and elicit position statements identifying parameters, processes, and uncertainties most relevant to their position along the value chain. These workshops provide LCA practitioners an opportunity to communicate significant data gaps and assumptions for stakeholder validation, as well as solicit additional data. Targeted surveys designed to probe user behavior may inform multiple scenarios of technology adoption and usage behavior. These social dimensions data allow creation of complementary analyses based on different modeling assumptions and decision contexts, the results of which identify tradeoffs and opportunities unique to each stakeholder group.

1.3 Integrating Risk-relevant Research

Risk characterization requires quantification of the hazard and exposure potential associated with emerging technologies, and combination of these factors into models that estimate overall risk. Ideally risk-based data will constitute the basis for life cycle impact assessment models and characterization factor databases used in LCA.[22] However, risk assessment for novel chemicals can take decades or more to complete,[23] and the delay between technology introduction and risk quantification presents a serious challenge to assessment of the potential environmental hazards posed by emerging technologies.[24] As with LCA in general, the need for extensive data promotes retrospective risk assessment, while prospective assessments are more rare and controversial.[25] Nonetheless, a lack of validated data does not justify omission of these risks from analysis.[26] LCA practitioners need new methods to incorporate risk research that is characterized by high parameter uncertainty and data gaps to have transparent representation of possible risks. Tools such as weight of evidence[27] and Monte Carlo exploration of impact assessment models[15] help integrate uncertainty data into existing impact assessment models to produce timely results. Where objective data is lacking, comparative risk assessments that integrate expert judgments can model relative risks and inform decision-makers despite high uncertainty.

1.4 Supporting Complex Decisions

Interpretation of LCA results must support decision-makers presented with inconclusive findings, a challenge that is exacerbated for emerging technologies with parameter, scenario, and model uncertainties that cumulatively may span orders of magnitude. One approach to truncate data needs is to evaluate emerging technologies in a comparative manner, for example relative to existing products or alternative process configurations. Comparative LCAs of emerging technologies can benefit from new interpretation methods capable of reconciling tradeoffs between impact categories or technology alternatives, and present results in a manner that both portrays uncertainty and is easy to interpret. To this end, researchers have incorporated multi-criteria decision analysis tools to inform decision making.[22] Examples include use of stochastic multi-attribute analysis after characterization to generate a probabilistic rank ordering of alternatives according to their overall life cycle impacts.[28] Such tools can support R&D decision-makers in reducing environmental burdens by systematically identifying the uncertainties or data gaps that are most influential to changing a decision outcome. Uncertainties that have little impact on decision-maker choice may be revealed as a low priority thereby conserving research resources.

1.5 A Model of Anticipatory LCA

Current solutions to the four barriers described above are practiced in isolation, and their coordinated implementation requires interdisciplinary collaboration and knowledge transfer beyond the scope of existing practice. To organize interdisciplinary knowledge sharing around the life cycle of emerging technologies, we introduce a generalizable framework for anticipatory LCA, shown in Figure 1. The framework – which is neither exclusive nor exhaustive – provides examples of interactions between multiple actors, builds on the piecemeal modeling advances described above, and engages R&D decision-makers in guiding emerging technologies away from anticipated environmental impacts. Anticipatory LCA is not meant to be predictive. Informed by anticipatory governance strategies,[29] anticipatory LCA complements alternatives-based approaches such as green chemistry to stimulate the imagination of relevant actors, and generate research hypotheses and other governance strategies that reorient the technology trajectory towards environmentally advantageous outcomes.

Figure 5 depicts how knowledge generated by researchers from social, environmental, and physical sciences informs anticipatory LCA model formulation. Public and private funding organizations (gold) provide resources for physical scientists and engineers (orange) to advance technologies through R&D towards commercial applications. Data collected by metering energy consumption, logging chemical inventories, and characterizing emission streams from laboratory-scale research is used in life cycle modeling software to capture up- and down-stream impacts. Performance characterization and measurements are used to inform functional unit modeling. Prospective modeling tools (purple) such as structured scenario analysis, scale-up modeling, and uncertainty bounding analyses (among other tools introduced in section 1.1) are used to account for parameter and scenario uncertainty in exploring how the life cycle inventory may change with future developments and alternative process configurations. Social scientists (blue) facilitate stakeholder engagement to inform practitioner modeling of multiple system boundaries, functional units, impact categories, and weights as modeling variables to systematically explore scenario uncertainty. Stochastic development of characterization factors incorporates variable risk data collected by environmental researchers (green) for emerging contaminants. The characterized inventory is explored with decision analysis tools (tan) to identify the most significant tradeoffs relative to data uncertainty and present results as a probabilistic rank ordering of alternatives.[28] Knowledge feedback (grey arrows) enables interventions in research funding, technology development, and risk research by identifying the uncertainties that undermine confidence in the analysis and prioritizing these for further research.

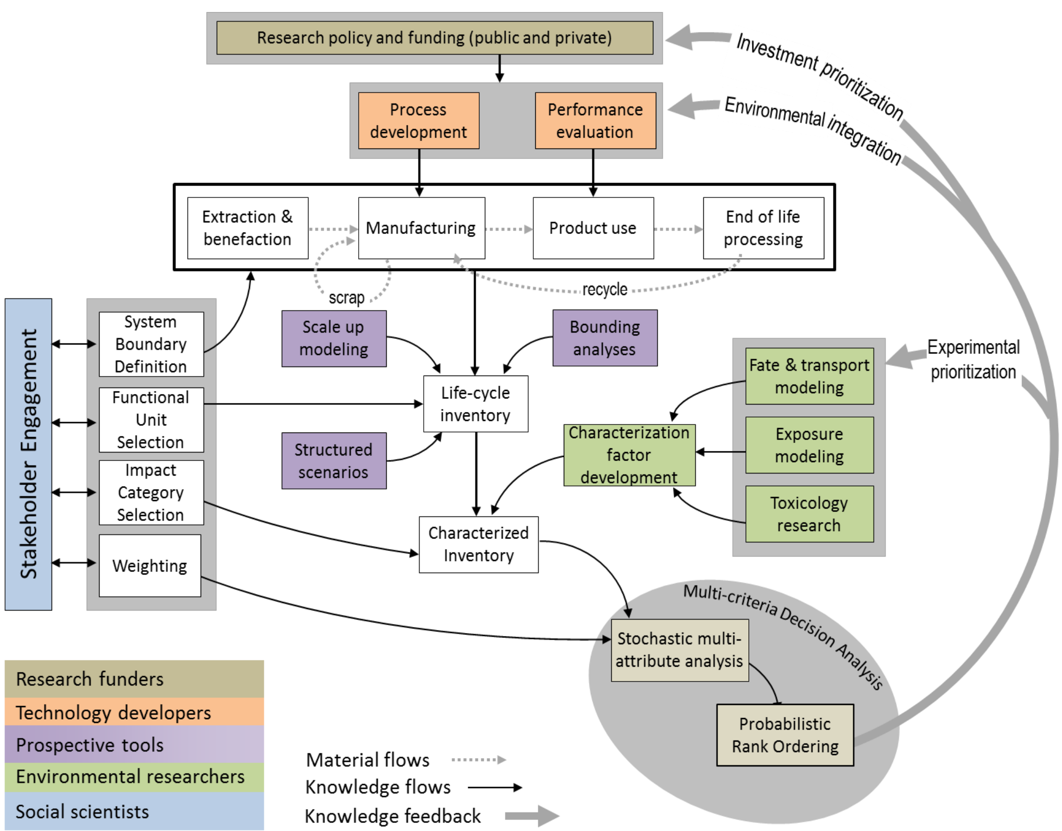

Figure 5: Anticipatory LCA structures interdisciplinary interactions and environmental interventions early in R&D. White boxes present current practices in LCA, which

emphasize material flows and feedback, whereas anticipatory LCA emphasizes knowledge flows and feedback.

Illustrating Anticipatory LCA

To demonstrate the modeling components and knowledge feedbacks contained in the anticipatory LCA framework, we present illustrative examples germane to rapidly expanding commercial PV technologies and emerging PV devices containing carbon nanotubes (CNTs). Production and adoption of PV technologies is in part driven by the goal of reducing greenhouse gas emissions and improving the environmental profile of electricity production, thus use of anticipatory LCA to guide PV innovation may result in development of products with greater potential for environmental benefit. The following illustrative examples span the entire anticipatory LCA framework and demonstrate: 1) Inclusion of technology foresight and treatment of scenario uncertainty through the creation of structured scenarios relative to thermodynamic limits, 2) Incorporation of multiple stakeholder perspectives through modeling of multiple system boundaries and functional units, 3) Integration of variable CNT risk-data through Monte Carlo simulation within existing impact assessment tools, and 4) Improved treatment of uncertainty and presentation of results using novel interpretation practices tailored to a specific decision context.

2.1 Structured Scenarios of Future Advances in mono-Crystalline Silicon Photovoltaic Devices

The life cycle greenhouse gas benefits of PV are proportional to the energy generated by the panel over its lifetime and inversely proportional to the energy consumed in manufacturing the panel.[30] These parameters are dynamic, sensitive to manufacturing and deployment locations, and responsive to alternative PV research agendas (e.g., research emphasis on increasing efficiency versus reducing manufacturing burdens). The historical trends (solid lines, 1998-2008) in manufacturing energy consumption (left axis) and cell efficiency (right axis) are used to formulate quantitative scenarios of future changes (dashed lines, 2008-2018), shown in Figure 6.

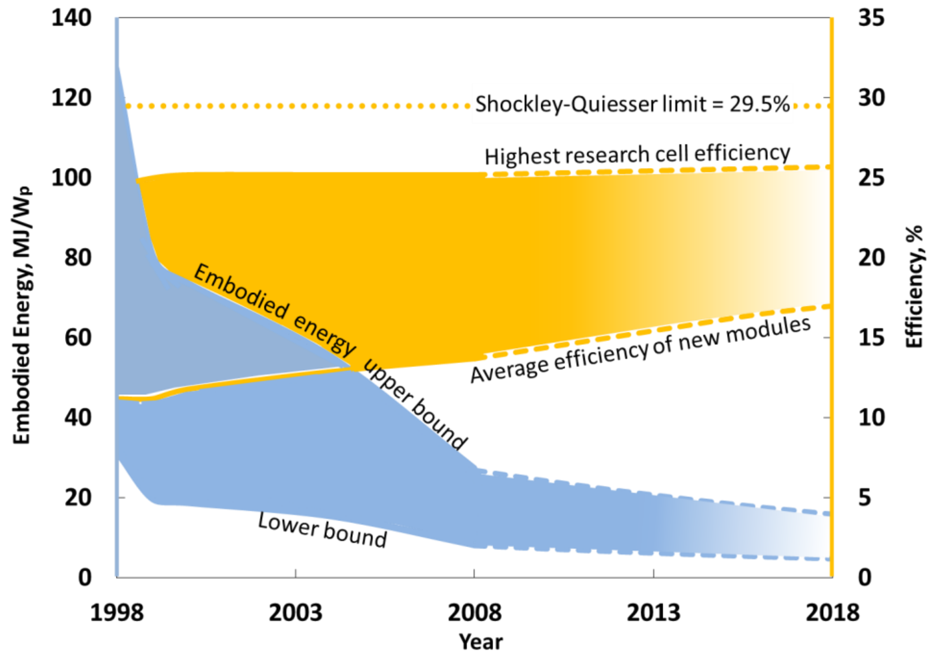

Figure 6: Historical trends (solid) and future scenarios (dashed) of manufacturing energy consumption (blue region, left axis) and cell efficiency (yellow region, right axis) over time for single-junction mono-crystalline silicon PV devices. Comparison to the Shockley-Quiesser limit (dotted) indicates that laboratory research directed at increasing cell efficiency has limited potential for improvement, whereas increasing the efficiency of commercially available cells and continuing to reduce the embodied energy of single-junction mono-crystalline PV devices has greater potential to result in environmental improvements.

Published estimates of the cradle-to-gate energy consumption of single-junction mono-crystalline PV production between the years 1998-2008[31,32,33,34] are varied by +/- 30% and normalized to high and low estimates of the Watt-peak (Wp) capacity. Under standard conditions the Wp capacity is a function of cell efficiency, which was bounded by an upper limit of the highest reported research cell efficiency[35,36,37,38,39] and a lower limit of the average efficiency of new panels entering the market.[40] Solar cell efficiencies are presented with respect to the Shockley-Quiesser limit[41] (dotted line) – the maximum possible efficiency of a single-junction cell based on the electronic properties of the semiconductor material and the characteristics of the solar spectrum – which is 29.5% in the case of mono-crystalline silicon. Current research cells are within 5% of this limit but have hardly improved over the last decade, whereas manufactured cells remain significantly less efficient but have shown steady increases. The embodied energy per unit area of panel has decreased historically and will likely continue this trend, although with a slower rate of improvement. These scenarios suggest that R&D resources allocated to furthering reductions in manufacturing energy consumption and improving the efficiency of manufactured cells have greater potential to improve life cycle greenhouse gas savings than investments in increasing laboratory cell efficiency marginally closer to their theoretical limit.[30] For example, feedback of this environmental knowledge to technology developers and research funders at the Quantum Energy and Sustainable Solar Technologies (QESST) Engineering Research Center contributed to formulation of research agenda focused on reducing silicon material use in PV devices through development of thin film silicon devices and novel laser processing approaches that reduce material losses in wafering.

2.2 Stakeholder Engagement Informs Modeling

In addition to the technical characteristics of installed PV panels and emissions associated with manufacturing, the net greenhouse gas savings associated with PV adoption is influenced by consumer behavior in the PV use-phase. Research on some renewable energy and efficiency-increasing technologies – for example light emitting diodes for domestic lighting[42] – suggests that environmental benefits do not accrue *de facto* because gains in efficiency are surpassed by increased consumption, a phenomenon termed the direct rebound effect.[43,44] Conversely, an energy consumer with newly installed PV panels may monitor their usage with greater scrutiny, leading to environmental improvements derived from both increased efficiency and reduced consumption (termed a negative rebound effect). This type of epistemic uncertainty influences LCA results and illustrates the importance of meaningful inclusion of user behavior through engagement activities that directly inform development of structured scenarios and alternative model configurations. Figure 7 contrasts a PV manufacturer perspective (green line, left axis) – which emphasizes cradle-to-gate CO_2 emissions associated with their product – and consumer perspective (blue region, right axis), which emphasizes CO_2 emissions produced by an average US household.

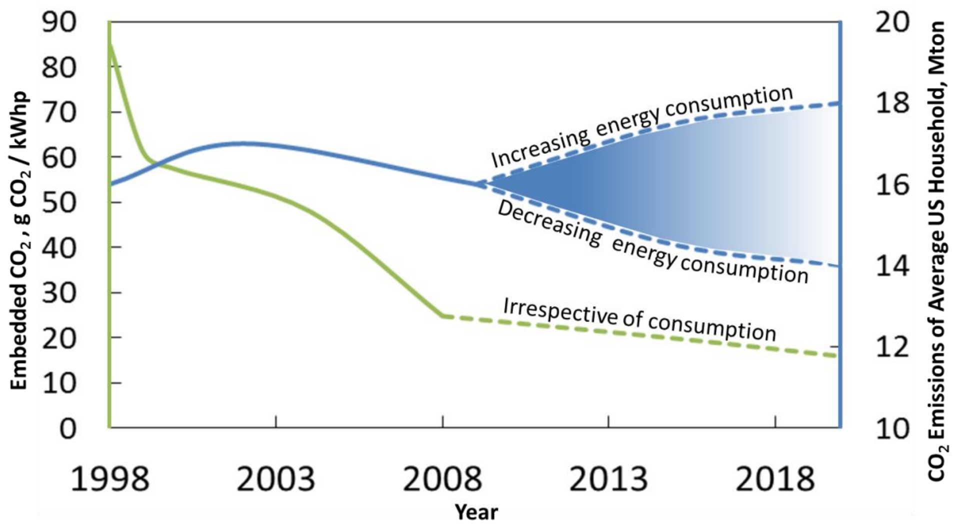

Figure 7: Historical data (solid) and future scenarios (dashed) of cradle-to-gate CO_2 emissions associated with production of 1 kWhp capacity of mono-crystalline PV (green line, left axis) and CO_2 emissions of the average US household (blue line, right axis).

Producing two analyses using different boundaries and functional units tailors results to a specific stakeholder and quantitatively incorporates scenario uncertainty arising from normative modeling decisions.

Historical data (solid) for cradle-to-gate emissions and future scenarios (dashed) correspond to the upper bound from Figure 2, converted to CO_2 emissions using a global average carbon intensity of 61 gCO_2 per MJ of primary energy.[45] Historically the CO_2 emissions of the average US household oscillated between 15 and 17 Mmt,[46] with future scenarios corresponding to an increase or decrease to 18 Mmt or 14 Mmt respectively. Producing two complementary analyses informed by engagement activities tailors results and identifies opportunities unique to individual stakeholder perspectives, in this case the manufacturer's perspective illustrates the potential for further reductions in embedded CO_2 whereas the consumer perspective illustrates scenarios of positive and negative rebound effects driving household emissions. Comparing these analyses provides insights into the relative magnitude of uncertainties associated with each perspective, in this case that continued reductions in the embodied energy of PV may be inconsequential if end-user consumption increases. Through targeted stakeholder engagement, decision making power does not lie solely with the LCA practitioner, but is explored and negotiated with diverse actors.

2.3 Stochastic Development of Characterization Factors for Novel Materials

When emerging PV technologies incorporate novel materials that lack risk data entirely or demonstrate high parameter uncertainty LCA practitioners have been unable to include impacts associated with their release.[26] For example, researchers are exploring incorporation of carbon nanotubes (CNTs) into dye-sensitized solar cells because the high electron mobility an tunable electronic properties may improve device performance,[47,48] but have no guidance regarding potential life cycle environmental implications. The heterogeneity of CNTs, diverse synthesis and post-synthesis treatment pathways, and experimental challenges encountered while measuring nanomaterial risk-relevant parameters[49] further contributes to uncertainty in data required to calculate ecosystem quality and human health characterization factors (CFs).[50] Applying a stochastic approach within existing impact assessment tools allows probabilistic development of CFs, shown in Figure 8, in place of single value estimates used for chemicals with less uncertain data.

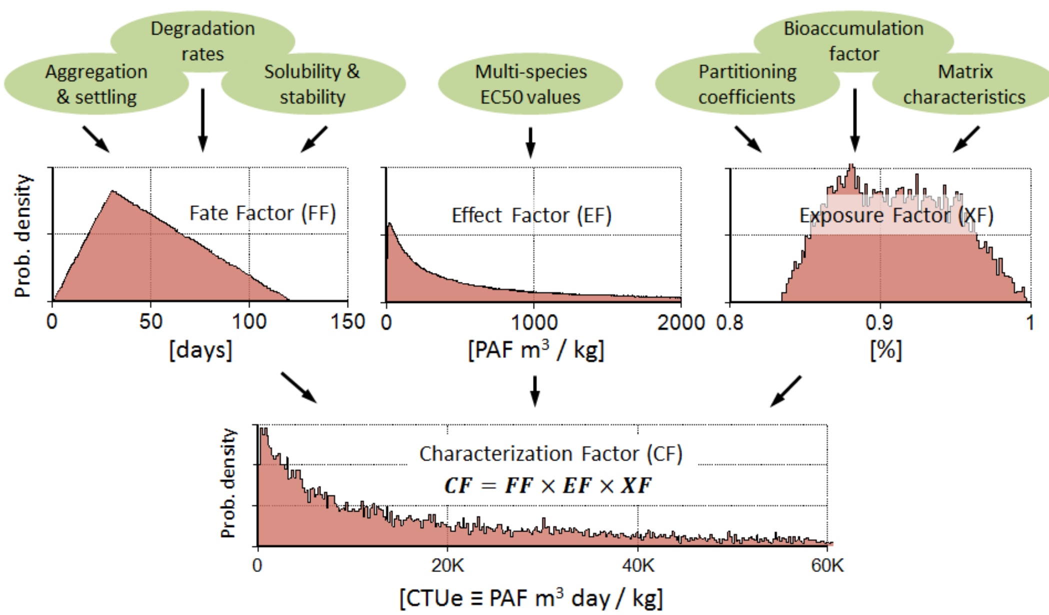

Figure 8: Stochastic development of a freshwater ecotoxicity CF for CNTs following the approach used in the consensus impact assessment tool USETox. Results are presented as a probability distribution with uncertainty derived from conflicting estimates of material properties (e.g., solubility, EC_{50}) and behavior (e.g., partitioning, bioaccumulation) in freshwater. Rank correlation identifies the uncertainties that drive CF results.

Extension of prior[15] probabilistic explorations of the consensus impact assessment tool USETox[51,50] to estimate a CNT aquatic ecotoxicity CF allows development of full distributions in place of best- and worst-case scenarios. USETox calculates CFs as the product of a fate factor (FF), an effect factor (EF), and an exposure factor (XF). Using the same data and modeling assumptions described in Eckelman et al 2012, we produce a full distribution for XF, which represents the fraction of CNTs that are bioavailable to aquatic organisms in the water column. Similarly, we reproduce EF, which represents the toxic effects leading to reductions in species populations in a unit volume of freshwater per kg CNT emitted [PAF m^3/kg], but model EF as a continuous lognormal distribution as is common in hazard assessment.[52] Figure 4 shows EF lognormally distributed a mean of 200 PAF m^3/kg and truncated with minimum of 20 and maximum of 2000 PAF m^3/kg corresponding to HC_{50} values of 25 mg/L and .25 mg/L respectively.[15] However, we deviate from prior studies and USETox to estimate FF, which represents the residence time [days] over which CNTs are bioavailable in the freshwater column, by using uncertain data taken directly from literature. Based on available data, FF is modeled as a triangular distribution with a lower limit of 2 hours, mode of 30 days,[53] and upper limit of 122 days.[15] The low end of this distribution corresponds to non-functionalized CNTs that rapidly settle out of water with low concentrations of natural organic matter (NOM), which has been shown to stabilize CNTs. The mode value corresponds to NOM-stabilized CNTs in freshwater with realistic NOM concentrations, and was informed (along with the distribution shape) by expert elicitation. The product of FF, EF, and XF yields the CNT CF [PAF m^3 day/kg], which represents the time and space integrated freshwater ecotoxicity impacts associated with direct emission of one kg of CNTs. This analysis informs PV researchers about the potential risks associated with inclusion of CNTs into PV devices. Rank correlation statistical analysis of the characterization factor inputs can be used to determine which material parameters most influence CF results – in this case EF is most uncertain and further research efforts would be best allocated to improving certainty in measured EC_{50} values for aquatic species.

2.4 Decision Analysis Simplifies Uncertain Environmental Results

Existing interpretation practices struggle to present actionable results for seemingly simple decisions, such as choosing the PV technology with the lowest overall environmental burden for a given installation, where data uncertainties are large relative to differences in environmental impacts associated with each alternative. In place of bar charts produced by commercially available software, anticipatory LCA follows an alternative interpretation method[54] to compare the impacts of 1 kWh of electricity produced by a 3kWp installation of either mono-crystalline silicon, multi-crystalline, or cadmium-tellurium (CdTe) panels (all inventory data taken from ecoinvent 2.2 for 3kWp slanted roof installation in Switzerland). Using the impact assessment tool ReCiPe[55] and pedigree matrix uncertainty[56] we overlay probability distributions for each technology and compare these in each impact category, shown in Figure 9.

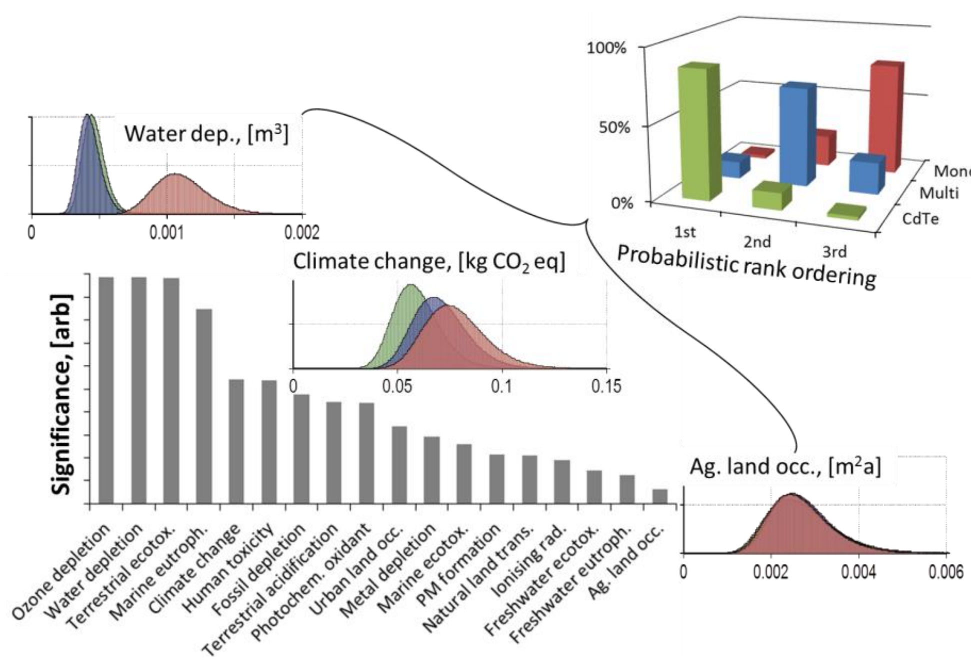

Figure 9: Decision-driven comparative LCA results for 1 kWh of electricity produced by a 3 kWp mono-crystalline silicon (red), multi-crystalline silicon (blue), or cadmium tellurium (green) PV system. Significance is estimated based on the overlapping area of each distribution, with a smaller overlaps corresponding to greater significance. Aggregating these impact categories together as a probabilistic rank ordering of alternatives incorporates uncertainty and presents results in a manner that is easy to interpret.

Presenting results this way identifies those categories in which there is significant difference in impact relative to data uncertainty and those categories in which high uncertainty overwhelms confidence in the comparison. Significance is estimated by comparing overlap area of each distribution – with greater overlap area corresponding to similar performance and a less significant tradeoff – which systematically identifies impact categories for which greater certainty is necessary to support the comparison. Using equal weights and published outranking algorithms[54] aggregating impact categories together into a probabilistic rank ordering of technology alternatives presents decision makers with an easy-to-interpret output that shows the likelihood of a given alternative outperforming the others. Figure 5 indicates that CdTe PV panels have an 80% likelihood of being ranked first (lowest overall environmental burden) whereas mono-crystalline silicon PV is almost always the most burdensome technology. Decision-driven interpretation of comparative LCA results accommodates inclusion of large uncertainties throughout modeling, systematically identifies impact categories in which greater certainty is necessary to inform decision-makers, and can promote uptake of LCA results by simplifying presentation.

Enacting Anticipatory LCA for Environmentally Responsible Innovation

This paper identifies four limitations – among numerous other pitfalls identified in the literature – that diminish the efficacy of current LCA practices in the context of emerging technologies, and introduces an interdisciplinary framework for anticipatory LCA that represents an early attempt to structure LCA as a process not a product in itself.[8] Anticipatory LCA is not predicative, but rather systematically and iteratively explores uncertainties across the life cycle of an emerging technology to prioritizing research with the greatest potential for environmental improvement and potentially contribute to responsible innovation[57,58,59] by redirecting a technology's development pathway. While this paper focuses on applying anticipatory LCA to PV, it is generalizable to other emerging technologies and customizable to fit specific decision contexts. Nonetheless a framework alone is inadequate, and enacting anticipatory LCA (and other large, transdisciplinary research efforts) requires advances in data sharing and institutional organization that parallel methods advancements. One practicable first step is the creation of formalized working groups within international organizations such as the Life Cycle Initiative or International Society for Industrial Ecology, which can galvanize support within the LCA community as well as direct contributions from relevant disciplines. With institutionalized support and the continued efforts of researchers from numerous backgrounds, it is possible for LCA to begin guiding innovation rather than retrospectively assessing its outcomes.

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References

(1) *Lithium-ion Batteries and Nanotechnology for Electric Vehicles: A Life Cycle Assessment*; EPA 744-R-12-001; United States Environmental Protection Agency: Washington, DC, **2012**; http://www.epa.gov/oppt/dfe/pubs/projects/lbnp/final-li-ion-battery-lca-report.pdf

(2) *A Research Strategy for Environmental, Health, and Safety Aspects of Engineered Nanomaterials*; National Research Council: Washington, DC; **2012**; http://www.nap.edu/catalog.php?record_id=13347

(3) *SunShot Vision Study*; Department of Energy; Washington, DC, **2012**; http://www1.eere.energy.gov/solar/pdfs/47927.pdf.

(4) *Environmental, Health, and Safety Research Strategy*; National Nanotechnology Initiative; Washington, DC, **2011**; http://www.nano.gov/sites/default/files/pub_resource/nni_2011_ehs_research_strategy.pdf

(5) National Research Council. *Models in Environmental Regulatory Decision Making.* Washington, DC: The National Academies Press, **2007**.

(6) Lloyd, S. M.; Ries, R., Characterizing, Propagating, and Analyzing Uncertainty in Life-Cycle Assessment: A Survey of Quantitative Approaches. *Journal of Industrial Ecology* **2007,** *11* (1), 161-179.

(7) Heijungs, R.; Huijbregts, M. A. In *A review of approaches to treat uncertainty in LCA*, iEMSs 2004 International Congress:" Complexity and Integrated Resources Management". International Environmental Modelling and Software Society, Osnabrueck, Germany, **2004**.

(8) McKone, T.; Nazaroff, W.; Berck, P.; Auffhammer, M.; Lipman, T.; Torn, M.; Masanet, E.; Lobscheid, A.; Santero, N.; Mishra, U., Grand challenges for life-cycle assessment of biofuels. *Environmental Science & Technology* **2011,** *45* (5), 1751-1756.

(9) Pesonen, H.-L.; Ekvall, T.; Fleischer, G.; Huppes, G.; Jahn, C.; Klos, Z.; Rebitzer, G.; Sonnemann, G.; Tintinelli, A.; Weidema, B.; Wenzel, H. Framework for scenario development in LCA. *Int. J. Life Cycle Assess.* **2000,** *5* (1), 21-30.

(10) Scown, C. D.; Nazaroff, W. W.; Mishra, U.; Strogen, B.; Lobscheid, A. B.; Masanet, E.; Santero, N. J.; Horvath, A.; McKone, T. E., Lifecycle greenhouse gas implications of US national scenarios for cellulosic ethanol production. *Environmental Research Letters* **2012,** *7* (1), 014011.

(11) Grubb, G. F.; Bakshi, B. R. Appreciating the Role of Thermodynamics in LCA Improvement Analysis via an Application to Titanium Dioxide Nanoparticles. *Environ. Sci.Technol.* **2011,** *45* (7), 3054-3061.

(12) Wender, B. A.; Seager, T.P. Anticipatory life cycle assessment of SWCNT-enabled lithium ion batteries. In *Nanotechnology for Sustainable Manufacturing*, Rickerby, D., Ed. Maralte BV: Lieden, NL., **2014.** ISBN: ISBN 9781482214826.

(13) Caduff, M.; Huijbregts, M. A. J.; Althaus, H.-J.; Koehler, A.; Hellweg, S. Wind Power Electricity: The Bigger the Turbine, The Greener the Electricity? *Environ. Sci. Technol.***2012,** *46* (9), 4725-4733.

(14) Weidema, B. P. *Market information in life cycle assessment*. Miljøstyrelsen: **2003**; Vol. 863.

(15) Eckelman, M. J.; Mauter, M. S.; Isaacs, J. A.; Elimelech, M. New Perspectives on Nanomaterial Aquatic Ecotoxicity: Production Impacts Exceed Direct Exposure Impacts for Carbon Nanotoubes. *Environ. Sci.Technol.* **2012,** *46* (5), 2902-2910.

(16) Walser, T.; Demou, E.; Lang, D. J.; Hellweg, S. Prospective environmental life cycle assessment of nanosilver T-shirts. *Environ. Sci. Technol.* **2011**, 45 (10), 4570-4578.

(17) Arvidsson, R.; Kushnir, D.; Sandén, B. A.; Molander, S. Prospective Life Cycle Assessment of Graphene Production by Ultrasonication and Chemical Reduction. *Environ. Sci. Technol.* **2014, DOI:** 10.1021/es405338k.

(18) *Guidelines for Social Life Cycle Assessment of Products*; UNEP DTI/1164/PA; United Nations Enviornmental Program; **2012**.

(19) Hertwich, E. G.; Hammitt, J. K.; Pease, W. S., A Theoretical Foundation for Life-Cycle Assessment. *Journal of Industrial Ecology* **2000,** *4* (1), 13-28.

(20) Berube, D. M., Socialis Commodis and Life Cycle Analysis: A Critical Examination of Uncertainty. In *Emerging Technologies: Socio-Behavioral Life Cycle Approaches* **2013**, 139.

(21) Thabrew, L.; Wiek, A.; Ries, R. Environmental decision making in multi-stakeholder contexts: applicability of life cycle thinking in development planning and implementation. *J. Cleaner Prod.* **2009,** *17* (1), 67-76.

(22) Linkov, I.; Seager, T. P., Coupling Multi-Criteria Decision Analysis, Life-Cycle Assessment, and Risk Assessment for Emerging Threats. *Environ. Sci. & Technol* **2011,** *45* (12), 5068-5074.

(23) National Research Council. *Science and Decisions.* Washington, DC: The National Academies Press, **2009**.

(24) Lambert, J. H.; Farrington, M. W., Risk-Based Objectives for the Allocation of Chemical, Biological, and Radiological Air Emissions Sensors. *Risk Analysis* **2006,** *26* (6), 1659-1674.

(25) Potts, H.; Anderson, J.; Colligan, L.; Leach, P.; Davis, S.; Berman, J., Assessing the validity of prospective hazard analysis methods: a comparison of two techniques. *BMC Health Services Research* **2014,** *14* (1), 41.

(26) Gavankar, S.; Suh, S.; Keller, A. Life cycle assessment at nanoscale: review and recommendations. *Int. J.Life Cycle Assess.* **2012,** *17* (3), 295-303.

(27) Zuin, S.; Micheletti, C.; Critto, A.; Pojana, G.; Johnston, H.; Stone, V.; Tran, L.; Marcomini, A., Weight of Evidence approach for the relative hazard ranking of nanomaterials. *Nanotoxicology* **2011,** *5* (3), 445-458.

(28) Canis, L.; Linkov, I.; Seager, T. P., Application of stochastic multiattribute analysis to assessment of single walled carbon nanotube synthesis processes. *Environ. Sci. & Technol.* **2010,** 44 (22), 8704-8711.

(29) Guston, D. Understanding 'Anticipatory Governance'. **2013,** *Soc. Stud. Sci.* 44 (2), 218-242.

(30) T.R. Dwarakanath; Wender, B.A.; Seager, T.P.; Fraser, M.P. Towards anticipatory lifie cycle assessment of photovoltaics, *Proceedings of the 39th IEEE Photovoltaics Specialist Conference*, Tampa Fl, Tampa Fl, **2013**.

(31) Alsema, E.; Frankl, P.; Kato, K. Energy pay-back time of photovoltaic energy systems: present status and prospects. Presented at the *2nd World Conference on Photovoltaic Solar Energy Conversion,* Vienna, AU; **1998**.

(32) Alsema, E. A. Energy pay-back time and CO2 emissions of PV systems. *Progress in Photovoltaics: Research and Applications* **2000,** *8* (1), 17-25.

(33) Alsema, E.; de Wild-Scholten, M. The real environmental impacts of crystalline silicon PV modules: an analysis based on up-to-date manufacturers data, Presented at the *20th European Photovoltaic Solar Energy Conference*, Barcelona, ES; **2005**.

(34) Fthenakis, V.; Held, M.; Kim, H.; Raugei, M. Update of energy payback times and environmental impacts of photovoltaics. Presented at the *24th European Photovoltaic Solar Energy Conference and Exhibition*, Hamburg, DE; **2009**.

(35) Green, M. A.; Emery, K.; Bücher, K.; King, D. L.; Igari, S. Solar cell efficiency tables (version 11). *Prog. Photovoltaics* **1998,** *6* (1), 35-42.

(36) Green, M. A.; Emery, K.; Bücher, K.; King, D. L.; Igari, S. Solar cell efficiency tables (version 13). *Prog. Photovoltaics* **1999,** *7* (1), 31-37.

(37) Green, M. A.; Emery, K.; Hishikawa, Y.; Warta, W. Solar cell efficiency tables (Version 31). *Prog. Photovoltaics* **2008,** *16* (1), 61-67.

(38) Green, M. A.; Emery, K.; King, D. L.; Igari, S. Solar cell efficiency tables (version 15). *Prog. Photovoltaics* **2000,** *8* (1), 187-195.

(39) Green, M. A.; Emery, K.; King, D. L.; Igari, S.; Warta, W. Solar cell efficiency tables (version 24). *Prog. Photovoltaics* **2004,** *12* (5), 365-372.

(40) Siemer, J.; Knoll, B. Still more than enough. *Photon International* **2013,** February 2013, 73.

(41) Shockley, W.; Queisser, H. J. Detailed balance limit of efficiency of p-n junction solar cells. *J. of applied physics* **1961,** *32* (3), 510-519.

(42) Hicks, A.; Theis, T. An agent based approach to the potential for rebound resulting from evolution of residential lighting technologies. *Int. J Life Cycle Assessment* **2014,** *19* (2), 370-376.

(43) Maxwell, D.; Owen, P.; McAndrew, L.; Muehmel, K.; Neubauer, A. *Addressing the Rebound Effect, a report for the European Commission DG Environment*; 2011

(44) Andersen, O. *Unintended Consequences of Renewable Energy*. Springer London, London: **2013**.

(45) Sims, R. E. H.; Schock, R.N.; Adegbululgbe, A.; Fenhann, J.; Konstantinaviciute, I.; Moomaw, W.; Nimir, H.B.; Schlamadinger, B.; Torres-Martínez, J.; Uchiyama, C. T., Y.; Vuori, S.J.V.; Wamukonya, N.; Zhang, X. *Energy supply.* In Intergovernmental Panel on Climate Change: Cambridge, UK and New York, USA, **2007**.

(46) *US Carbon Emissions and Intensities Over Time: A Detailed Accounting of Industries, Government and Households*; US Department of Commerace: Economics and Statistics Administration: **2010**.

(47) Bhattacharyya, S.; Kymakis, E.; Amaratunga, G. A. J. Photovoltaic Properties of Dye Functionalized Single-Wall Carbon Nanotube/Conjugated Polymer Devices. *Chemistry of Materials* **2004,** *16* (23), 4819-4823.

(48) Dang, X.; Yi, H.; Ham, M.-H.; Qi, J.; Yun, D. S.; Ladewski, R.; Strano, M. S.; Hammond, P. T.; Belcher, A. M. Virus-templated self-assembled single-walled carbon nanotubes for highly efficient electron collection in photovoltaic devices. *Nat Nano* **2011,** *6* (6), 377-384.

(49) Wiesner, M. R.; Lowry, G. V.; Jones, K. L.; Hochella, J. M. F.; Di Giulio, R. T.; Casman, E.; Bernhardt, E. S. Decreasing Uncertainties in Assessing Environmental Exposure, Risk, and Ecological Implications of Nanomaterials. *Environ. Sci. Technol.* **2009,** *43* (17), 6458-6462.

(50) Henderson, A.; Hauschild, M.; Meent, D.; Huijbregts, M. J.; Larsen, H.; Margni, M.; McKone, T.; Payet, J.; Rosenbaum, R.; Jolliet, O. USEtox fate and ecotoxicity factors for comparative assessment of toxic emissions in life cycle analysis: sensitivity to key chemical properties. *Int. J. Life Cycle Assess.* **2011,** *16* (8), 701-709.

(51) Huijbregts, M. A.; Hauschild, M.; Jolliet, O.; Margni, M.; McKone, T.; Rosenbaum, R.K.; van de Meent, D. USEtox User Manual. **2010**; http://www.usetox.org/sites/default/files/support-tutorials/user_manual_usetox.pdf

(52) Golsteijn, L.; Hendriks, H. W. M.; van Zelm, R.; Ragas, A. M. J.; Huijbregts, M. A. J. Do interspecies correlation estimations increase the reliability of toxicity estimates for wildlife? *Ecotoxicology and Environmental Safety* **2012,** *80* (0), 238-243.

(53) Hyung, H.; Fortner, J. D.; Hughes, J. B.; Kim, J.-H. Natural Organic Matter Stabilizes Carbon Nanotubes in the Aqueous Phase. *Environ Sci & Technol* **2006,** *41* (1), 179-184.

(54) Prado-Lopez, V.; Seager, T.; Chester, M.; Laurin, L.; Bernardo, M.; Tylock, S., Stochastic multi-attribute analysis (SMAA) as an interpretation method for comparative life-cycle assessment (LCA). *The International Journal of Life Cycle Assessment* **2013**, 1-12.

(55) Goedkoop, M.; Heijungs, R.; Huijbregts, M.; De Schryver, A.; Struijs, J.; van Zelm, R., ReCiPe 2008. *A life cycle impact assessment method which comprises harmonised category indicators at the midpoint and the endpoint level* **2009,** *1*.

(56) Ciroth, A.; Muller, S.; Weidema, B.; Lesage, P. Empirically based uncertainty factors for the pedigree matrix in ecoinvent. *Int. J. Life Cycle Assess.* **2013**, 1-11; DOI 10.1007/s11367-013-0670-5

(57) Stilgoe, J.; Owen, R.; Macnaghten, P. Developing a framework for responsible innovation. *Research Policy* **2013**, 42 (9), 1568-1580.

(58) von Schomberg, R. A Vision of Responsible Research and Innovation. In R. Owen, J. Bessant, M. Heints (Eds) *Responsible Innovation: Managing the Responsible Emerginged of Science and Innovation in Sociery,* Wiley, London **2013**, 51-74.

(59) *Regulation of the Euproean Parliament and of the council establishing Horizon 2020 - The framework programme for research and innovation (2014-2020)*; EC COM(2011) 809; European Commission: Brussels, BE, **2011;** http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:52011PC0809&from=EN

CHAPTER 4

SENSITIVITY-BASED RESEARCH PRIORITIZATION THROUGH STOCHASTIC CHARACTERIZATION MODELING

BA Wender[1*], V Prado[2], P Fantke[3], A Cano[1] and TP Seager[1]

[1]School of Sustainable Engineering and the Built Environment, Arizona State University, 660 S. College Ave. Rm 507. Tempe, AZ 85287

[2]Institute of Environmental Sciences CML, Leiden University, Einsteinweg 2, 2333 CC, Leiden

[3]Quantitative Sustainability Assessment Division, Department of Management Engineering, Technical University of Denmark, Produktionstorvet 424, 2800 Kgs. Lyngby, Denmark

[*]Corresponding author: bwender@asu.edu

Abstract

Large data requirements, high uncertainty and complexity, and regulatory relevance of toxicity impact assessment motivates greater focus on model sensitivity toward input parameter variability. This is particularly useful for emerging contaminants like engineered nanomaterials (ENMs) to guide future efforts in data refinement and design of experiments. This study presents a Monte Carlo tool based on USEtox 1.0 that allows researchers to rapidly prioritize data needs according to influence on characterization factors (CFs). Using Monte Carlo analysis we demonstrate a sensitivity-based approach to prioritize research through a case study comparing aquatic ecotoxicity CFs calculated with USEtox 1.01 for the ENM C_{60} and the vitamin B derivative niacinamide, two antioxidants used in personal care products. We calculate CFs via 10,000 iterations assuming plus-or-minus one order of magnitude variance for fate and exposure-relevant inputs. Spearman Rank Correlation Indices are used for all variable inputs to identify parameters with the largest influence on CFs, which we prioritize for data refinement and future experimental investigation. Based on the importance of aggregate multi-species toxicity (average log EC_{50}) and studies suggesting solvent residues may yield erroneous toxicity estimates, we recalculate C_{60} CFs omitting all studies using solvents in sample preparation.

For emissions to freshwater, the C_{60} CF is log-normally distributed with a geometric mean of 280 (geometric standard deviation, GSD: 2.1) PAF m^3 day/kg compared to 2.6 (GSD: 1.8) PAF m^3 day/kg for niacinamide. C_{60} CFs are most sensitive to varied suspended solids partitioning coefficients (Kpss) and average log EC_{50}, whereas variation of other substance parameters has comparatively little effect on model results.

Insufficient experimental evidence hampers to revise assumptions for Kpss, and we suggest prioritizing future experiments that elucidate C_{60} interactions with suspended solids. Recalculating C_{60} CFs without toxicity studies that use solvents reduces the geometric mean by more than a factor of ten. This reinforces the importance of thorough source term characterization, in this case regarding the presence of solvent residues. Calculating stochastic CFs allows sensitivity-based prioritization of data needs and future experiments, which is particularly helpful in the context of emerging contaminants like C_{60}. Researchers can conserve resources and address parameter uncertainty by applying our approach when developing new or refining existing CFs for the inventory items that contribute most to toxicity impacts. The Monte Carlo tool can be applied to current toxicity characterization models like USEtox and is freely available.

Introduction

Coupled fate-exposure-effect models like USEtox (Rosenbaum et al. 2008), Impact2002 (Pennington et al. 2005), and USES-LCA (van Zelm et al. 2009) are widely used to calculate characterization factors (CFs) for human toxicity and ecotoxicity impacts in life cycle assessment (LCA). CFs allow practitioners and decision makers to quantify potential toxic impacts associated with emissions quantified in the life cycle inventory. These models are complicated, require various substance-specific input parameters, and their results are typically characterized by an overall uncertainty of two to three orders of magnitude depending on emission compartment, exposure scenario, and data availability (Jolliet and Fantke 2015; Rosenbaum 2015). Thus, life cycle impact assessment (LCIA) models for characterizing human toxicity and ecotoxicity require further improvement, although significant achievements have been made over the last decade. For example, sustained harmonization efforts between divergent ecotoxicity LCIA models resulted in the consensus model USEtox (Rosenbaum et al. 2008; Westh et al. 2015) and the recently released USEtox 2.0 (http://usetox.org), which are considered best practice (Hauschild et al. 2013), recommended by the ILCD handbook (EC 2011), and implemented in TRACI (Bare et al. 2012). The extensive inter-model comparisons and streamlining activities addressed model uncertainty and improved transparency and credibility (Hauschild et al. 2008).

However, further development and adoption of current human toxicity and ecotoxicity LCIA models faces challenges related to the large number and diverse properties of relevant emitted substances, limited availability of high quality data, and sparse treatment of parameter uncertainty or variability (Alfonsín et al. 2014; Gust et al.

2015; Rosenbaum 2015). For example, there is a large discrepancy between the ≈ 10,000 substances included in the latest Ecoinvent inventory library (Weidema 2013) and the ≈ 3,500 human and ecotoxicity CFs available from the largest toxicity characterization models USEtox and USES LCA (Henderson et al. 2011; van Zelm et al. 2009). In the parsimonious model USEtox, each individual CF requires approximately ten substance-specific input parameters, thereby challenging the experimental and data curation efforts required for database validation and expansion. As a result, a large share of CFs in USEtox relies on substance data estimated using outputs from quantitative structure activity relationships (QSARs) such as EPI Suite (USEPA 2015b), which are essential for filling data gaps but lack experimental evidence and therefore are considered of lower quality than measured values (Huijbregts 2010a). Thus, there is a critical need to explore the sensitivity of human toxicity and ecotoxicity LCIA results – and those used in other impact categories – to variability and uncertainty in required substance input data, which may help expedite database expansion, refinement, and development of future research agenda (Cellura et al. 2011; Cucurachi and Heijungs 2014).

The best available method to evaluate LCIA model sensitivity to variability in substance data is to use Monte Carlo analysis to sample from specified distributions (Sonnemann et al. 2003) and calculate CFs as frequency distributions as opposed to point values (Lloyd and Ries 2007; van Zelm and Huijbregts 2013). Calculating stochastic CFs enables sensitivity analyses that can help expedite data collection by identifying the substance-specific parameters with the greatest influence on model results (Saltelli et al. 2008). This can help define research agenda and conserve resources by focusing attention on experiments with the greatest potential to reduce uncertainty of model results, while substance data with little impact on results may be revealed as a low investigative priority.

The benefits of applying sensitivity-based research prioritization may be greatest in the context of emerging contaminants such as engineered nanomaterials (ENMs). Widespread concern regarding potential toxicity-related impacts associated with emissions of ENMs galvanized an active research community and produced volumes of published data that demonstrates high variability between published parameter estimates (NSTCCT 2014). The suitability of human and ecotoxicity LCIA models for ENMs is a known issue (Klopffer 2007) and relatively well covered in recent literature (Gilbertson et al. 2015; Miseljic and Olsen 2014b; Salieri et al. 2015). Less emphasized are critical data-related challenges include:

1) The large number of commercially-relevant ENMs and possible permutations made through alternative surface coatings leaves comprehensive characterization and collection of sufficient data for all ENM emissions impracticable (Alvarez et al. 2009; Cohen et al. 2013; Grieger et al. 2010),
2) Material heterogeneity within even narrow classes of ENMs – for example carbon nanotubes with differing lengths, number of walls, chirality – results in high variability in risk-relevant parameters reported in the literature (Hendren et al. 2015; Saleh et al. 2015; Seager and Linkov 2008), and
3) Computational approaches to estimating substance properties for ENMs are nascent (Alvarez et al. 2009; Cohen et al. 2013; Eisenberg 2015) and QSARs designed for conventional chemical pollutants may be inapplicable. For example,

EPI Suite does not apply to the ENM C_{60} because the closed-cage structure is incomparable to other carbonaceous materials.

Together these challenges limit the applicability of existing human and ecotoxicity LCIA models to ENMs, and to date there are no CFs specific for ENMs included in any commercial LCA software package or database. Nanomaterial LCA review articles identified the lack of ENM-specific CFs as preventing quantification of toxicity impacts associated with ENM emissions (Gavankar et al. 2012; Hischier and Walser 2012; Miseljic and Olsen 2014a). In the literature fewer than five studies have developed aquatic ecotoxicity CFs for ENMs, predominantly through innovative modifications of USEtox including: development of realistic and worst-case scenarios for the ENM's CF (Eckelman et al. 2012), precautionary assumptions (Miseljic and Olsen 2014a), qualitative discussion of uncertainty (Rodriguez-Garcia et al. 2014), and development of simplified colloidal transport models within USEtox (Salieri et al. 2015). Only Eckelman et al (2012) conducts a thorough Monte Carlo sensitivity analysis on substance properties, but the emphasis was on comparing the magnitude of cumulative upstream ecotoxicity impacts with those directly from ENM releases, and therefor did not include the relative influence of variable substance data on characterization results.

The present paper introduces a Monte Carlo tool that can be combined with USEtox 1.01 that allows users to specify substance data as variable distributions, as opposed to point value estimates, and presents resulting CFs as frequency distributions. We apply the tool to compare aquatic ecotoxicity CFs of the ENM C_{60} (CAS 99685-96-8) and the vitamin B derivative niacinamide (CAS 98-92-0), both of which are used at low concentrations in commercial personal care products because of their antioxidant properties (Benn et al. 2011; Lens 2009; PEN 2013). The comparison represents a hypothetical decision context in which personal care product developers are considering substitution of the emerging material C_{60} for a conventional alternative performing the same function. Given high environmental and regulatory uncertainty regarding ENMs, product developers are unsure of potential toxicity impacts and what further research is necessary to improve confidence in the material comparison. Differences in performance, which are often the motivation for adoption of new materials, would be reflected in functional unit definition and differences in emitted mass are tracked in the life cycle inventory, both of which are beyond the scope of this manuscript. More importantly, the comparison illustrates one component of an *anticipatory* approach to LCA that compares an emerging technology to conventional alternatives in order to guide research and development decisions towards reduced environmental impacts (Wender et al. 2014b).

2.0 Methods

USEtox calculates freshwater ecotoxicity CFs per unit mass of emitted substance, measured in comparative toxicity units CTUe (PAF m^3 d/kg), as the product of a fate factor (FF, d), an exposure factor (XF, dimensionless), and an effect factor (EF, PAF m^3/kg) (Equation 1). FF, XF, and EF represent the residence time in freshwater, dissolved fraction in freshwater, and aggregated multi-species toxicological response, respectively (Henderson et al. 2011; Huijbregts 2010a):

$$CF = FF * XF * EF \qquad \text{Eq. 1}$$

Model structure, assumptions, and landscape data of USEtox 1.01 were not targeted in our Monte Carlo tool and thus model uncertainty is not addressed in this study as the focus is exclusively on substance data research prioritization.

2.1 Description of the Monte Carlo Tool

To facilitate Monte Carlo operation, we developed a user-friendly interface where USEtox-required substance data can be described as any combination of uniform, normal, log-normal and triangular distributions, or remain point values as applied in USEtox. These distributions are sampled independently n-specified times, the values were used as input to USEtox, and resulting CFs plotted as frequency distributions along with descriptive statistics. Additionally, the Monte Carlo tool calculates Spearman Rank Correlation Indices for all inputs that are not point values (SI 2.1). Results for each material presented are based on 10,000 Monte Carlo runs, taking approximately one hour to complete (2.0 GHz intel i7). The JAVA-based tool has been made open source and a beta version made available for download.

2.2 Fate and Exposure Data and Modeling Assumptions

C_{60} partitions strongly to dissolved organic carbon, suspended solids, and natural organic matter (Yang et al. 2015). Thus, we implement values from available literature according to USEtox requirements for metals as shown in Table 4. The large quantity of publications detailing fate-relevant studies for C_{60} and its aggregates, combined with inconsistent reporting of nanomaterial and matrix characteristics, prohibits a comprehensive review. To emphasize the method of sensitivity-based research prioritization we have selected only studies which report USEtox-required parameters by name, for example as opposed to studies reporting removal percentages by biomass.

Table 4 Fate and exposure relevant data and modeled variance for C_{60}

Parameter	Description	Units	Midpoint value(s)	Baseline variance	Reference
MW	Molecular weight	g/mol	721	721	Chemical formula
Kow	Octanol-water partitioning coefficient	L/L	4.6×10^{6}	4.6×10^{5} - 4.6×10^{7}	Jafvert & Kulkarni, 2008
Koc	Organic carbon partitioning coefficient	L/kg	1.2×10^{7} 5×10^{3}	5×10^{3} - 1.2×10^{7}	Chen & Jafvert, 2009 Avanasi et al, 2014
Kh	Henry's law constant	Pa m^3/mol	1×10^{-20}	1×10^{-20}	USEtox manual
Pvap	Vapor pressure	Pa	6×10^{-4} 1×10^{-20}	1×10^{-20} - 6.6×10^{-4}	SES Research, 2010 USEtox manual
Sol	Solubility in water	mg/L	2-8 $\times 10^{-6}$ <100 nC_{60}	5×10^{-6} - 1×10^{2}	Jafvert & Kulkarni, 2008 Fortner et al, 2005
Kdoc Kpss Kpsl Kpsd	Partitioning coefficient between: dissolved organic carbon; Suspended solids; Soil particles; Sediment particles	L/kg	3.2×10^{4}	3.2×10^{3} - 3.2×10^{5}	USEtox regression: Kdoc=0.08*Kow Assume Kdoc = Kpss = Kpsl = Kpsd
Kdeg, air	Degradation rate in air		1×10^{-20} 2×10^{-5}	1×10^{-20} - 2×10^{-5}	USEtox manual, Tiwari et al, 2014
Kdeg, water	Degradation rate in water	1/s	4.5×10^{-8}	4.5×10^{-9} - 4.5×10^{-7}	Avanasi et al, 2014 USEtox manual
Kdeg, soil	Degradation rate in soil		2.25×10^{-8}		

				2.2×10^{-9} - 2.2×10^{-7}	
Kdeg, sed	Degradation rate in sediment		5 x 10^{-9}	5×10^{-10} - 5×10^{-8}	
BAF fish	Bioaccumulation factor in fish	L/kg	3.2 x 10^{4} 5.12 x 10^{5}	5×10^{4} - 5×10^{6}	Li et al, 2010 Jafvert & Kulkarni, 2008

A growing weight of evidence suggests that C_{60} released to water partitions to natural organic matter, biological membranes, and settles to sediment rapidly (PubChem 2015a; Pycke et al. 2012; USEPA 2010). Nonetheless some fate-relevant parameters published data show high variability, for example Chen and Jafvert (2009) reported the first estimate of an organic carbon-water partitioning coefficient (Koc) of $\approx 1.2 \times 10^{7}$ mL/g, whereas five years later Avanasi et al. (2014) report Koc values as low as 5×10^{3} mL/g based on soil type. We model Koc as a uniform distribution across this range. C_{60} solubility ranges from virtually insoluble ($<10^{-9}$ mg/L) as isolated particles to nearly 100 mg/L as water-stable aggregates (Avanasi et al. 2014), which we model as uniform between 5×10^{-6} and 100 mg/L. Similarly, atmospheric degradation rates (Kdeg, air) of 2×10^{-5} 1/s by environmentally-relevant ozone concentrations was shown in Tiwari et al. (2014), although other carbon nanomaterials have been modeled as resistant to degradation (e.g., 1×10^{-20} 1/s) (Rodriguez-Garcia et al. 2014). Thus we model Kdeg, air as uniform between these two values. In part the variability in fate and exposure relevant substance data for C_{60} is related to the large number of publications on the ENM, as compared to the less-studies niacinamide. Thus, future efforts can incorporate the number of studies into estimates of parameter uncertainty or variability as has recently been demonstrated for pesticide dissipation half lives in plants (Fantke et al. 2014).

Fate and exposure relevant parameters for which only point values are reported in literature or available from QSAR programs, we assume an arbitrary baseline scenario of uniform variable distributions of plus-or-minus one order of magnitude from the midpoint value. The USEtox 1.01 manual describes a simple regression to estimate the dissolved organic carbon partitioning coefficient (Kdoc) as 0.08 x Kow, giving the midpoint value of 3.2 x 10^4 L/kg. In the absence of experimental data, we assume Kdoc is equal to the suspended solids partitioning (Kpss), sediment particle partitioning (Kpsd), and soil particle partitioning (Kpsl) coefficients (Eckelman et al. 2012). Based on the classification of C_{60} as recalcitrant (Avanasi et al. 2014; Kümmerer et al. 2011) and the USEtox manual (Huijbregts 2010b), we model the aquatic degradation rate (Kdeg, water) as 4.5 x 10^{-8} 1/s, and the soil and sediment degradation rates as 1/2 and 1/9 of Kdeg, water respectively. Bioaccumulation factors for fish (BAF fish) have been reported as ≈ 3 x 10^4 L/kg (Li et al. 2010) and 5 x 10^5 L/kg (Jafvert and Kulkarni 2008), which is less than the assumed baseline variability, thus we model BAF fish as uniform between 5 x 10^4 and 5 x 10^6 L/kg.

The conventional antioxidant niacinamide that C_{60} may replace is the subject of relatively fewer studies, which is why we rely primarily on EPISuite (USEPA 2015b) and supplement with available literature as summarized in Table 5.

Table 5 Fate and exposure relevant data and modeled variance for niacinamide

Parameter	**Description**	**Units**	**Midpoint value(s)**	**Baseline variance**	**Reference**
MW	Molecular weight	g/mol	122	 122	Chemical formula
Kow	Octanol-water partitioning coefficient	L/L	0.42	 4.2x10^{-2} - 4.2	OECD SIDS

Koc	Organic carbon partitioning coefficient	L/kg	8.5	0.85 - 85	EPISuite, Kocwin
Kh	Henry's law constant	Pa m^3/mol	2.9 x 10^{-7} 6.45 x10^{-6}	2.9x10^{-8} - 2.9x10^{-6}	PubChem database USEtox Guidance
Pvap	Vapor pressure	Pa	0.026 0.05	5x10^{-3} - 0.5	EPISuite, MPBPVP PubChem database
Solubility	Solubility in water	mg/L	5e5 6.9-10 x 10^5	5x10^4 - 5x10^6	EPISuite, exper. OECD SIDS
Kdeg, air	Degradation rate in air	1/s	1.8 x 10^{-6}	1.8x10^{-7} - 1.8x10^{-5}	EPISuite, AOPWin USEtox manual
Kdeg, water	Degradation rate in water		2.1 x 10^{-7}	2.1x10^{-8} - 2.1x10^{-6}	EPISuite, Biowin USEtox manual
Kdeg, soil	Degradation rate in soil		1 x 10^{-7}	1x10^{-8} - 1x10^{-6}	
Kdeg, sed	Degradation rate in sediment		2.3 x 10^{-8}	2.3x10^{-9} - 2.3x10^{-7}	
BAF fish	Bioaccumulation factor in fish	L/kg	0.9	0.09 to 9.0	EPISuite, BCFBAF

Niacinamide was not included in USEtox 1.01, but was covered in the recently released USEtox 2.0 (http://usetox.org) with fate and exposure-relevant parameter values nearly identical to those presented in Table 2 (SI, 2.2.2). We collect parameter estimates from an OECD Screening Information Dataset, which reports experimentally-determined estimates for Kow of 0.42 and solubility of 6.9-10 x 10^5 mg/L (UNEP 2002), which correspond closely with values reported in EPISuite (USEPA 2015b). The National Center for Biotechnology Information database reports Henry's Constant (Kh) as 2.9 x

10^{-7} Pa m^3/mol and a vapor pressure of 0.05 Pa (PubChem 2015b). We combine EPISuite outputs and the USEtox organics manual (Huijbregts 2010c) to model uniform distributions for all degradation rates and BAF fish following the baseline scenario of plus-or-minus one order of magnitude from these midpoint values.

2.3 Effect Factor Data and Modeling Assumptions

We calculate EF for both materials using variable toxicology data from acute and chronic toxicity tests on producers (algae), primary consumers (invertebrates), and secondary consumers (fish) (Hauschild and Huijbregts 2015; Huijbregts 2010a). Toxicity data for C_{60} and niacinamide – typically reported as the concentration at which 50 percent of the exposed organisms over background exhibit the studied effect (EC_{50}), inhibited growth (IC_{50}), or lethality (LC_{50}) – was taken from available literature and is summarized in Table 6 and Table 7, respectively.

Table 6 Data from individual ecotoxicity studies of C_{60}

Reference	Species (n=10)	Test type and endpoint	Reported value(s)	Stabilization method	EC_{50} value
Producers					
Tao et al, 2015	*S. obliquus*	72h Chronic IC_{50}	1.94 mg/L	THF then membrane filtered	1.9 mg/L
Gelca et al, 2012	*S. capricornutum*	5d Chronic IC_{50} dark	0.04 mg/L	Stirred then filtered, average of size ranges taken	0.04 mg/L
		5d Chronic IC_{50} light	0.02 mg/L		0.02 mg/L
Baun et al, 2008*	*P. subcapitata*	48h Chronic IC_{30}	90 mg/L	Stirring	90 mg/L
Blaise et al, 2008*	*P. subcapitata*	72h Chronic IC_{25}	100 mg/L	Mixing	100 mg/L
Seki et al, 2008**	*P. subcapitata*	72h Chronic IC_{50}	14.8 mg/L extrapolated	Grinding with sugar and oil	15 mg/L
Primary Consumers					

Seki et al, 2008	*D. magna*	48h Acute EC_{50} immobilization	>2.25 mg/L (LOEC)	Grinding with sugar and oil	5 mg/L
Blaise et al, 2008	*T. platyurus*	24h Acute LC_{50}	>10 mg/L	Mixing	5 mg/L
	H. attenuata	96h Acute EC_{50} morphological	>10 mg/L		5 mg/L
Lovern & Klaper, 2006	*D. magna*	48h Acute LC_{50}	7.9 mg/L	Sonication	3.9 mg/L
			0.46 mg/L	THF, filtered then evaporated	0.2 mg/L
Zhu et al, 2009	*D. magna*	48h Acute LC_{50}	10.5 mg/L	Shaken	5.3 mg/L
		48h Immobility EC_{50}	9.34 mg/L		4.6 mg/L
Ji et al, 2014	*D. magna*	96h Acute LC_{50} dark	1.85 mg/L (NOEC)	Mixing then filtered through .2 micron	17 mg/L
		96h Acute LC_{50} light	0.46 mg/L (NOEC)		4.1 mg/L
	M. macrocopa	96h Acute LC_{50} dark			4.1 mg/L
		96h Acute LC_{50} light			4.1 mg/L
Tao et al, 2009	*D. magna*	48h Acute LC_{50} neonatal	0.44 mg/L	THF then evaporated	0.2 mg/L
Zhu et al, 2006	*D. magna*	48h Acute LC_{50}	0.8 mg/L	THF then evaporated	0.4 mg/L
Oberdorster et al, 2006	*D. magna*	96h Acute LC_{50}	>35 mg/L (LOEC)	Stirring	78 mg/L
		21d Chronic Molting delay, number of offspring	2.5 mg/L (LOEC)		5.6 mg/L
Baun et al, 2008	*D. magna*	48h Chronic Mobility	<50 mg/L (NOEC)	Stirring	450 mg/L
Secondary consumers					
Seki et al, 2008	*O. latipes*	96h Acute LC_{50}	>2.15 (NOEC)	Grinding with sugar and oil	19 mg/L
Oberdorster et	*O. latipes*	96h Acute LC_{50}	0.5 mg/L (NOEC)	Stirring	4.5 mg/L

al, 2006	*P. promelas*		1 mg/L (NOEC)		9 mg/L
Usenco et al, 2007	*D. rerio*	96h Acute LC_{50} embryonic	0.2 mg/L	C_{60} or C_{70} sonicated in DMSO	0.1 mg/L
			4 mg/L	$C_{60}(OH)_{24}$	2 mg/L
Usenco et al, 2008	*D. rerio*	5d Acute LC_{50} dark	0.3 mg/L	C_{60} sonicated in DMSO	0.15 mg/L
		5d Acute LC_{50} light	0.2 mg/L		0.1 mg/L
		5d Chronic EC_{50} Fin malformation	0.15 mg/L		0.15 mg/L
Zhu et al, 2007	*D. rerio*	96h Chronic EC_{50} developmental	1.5 mg/L	C_{60} in THF then evaporated	1.5 mg/L
			50 mg/L (NOEC)	$C_{60}(OH)_{24}$	450 mg/L

*Although USEtox manual specifies EC_{50} values, we retain data from studies reporting 25 and 30 percent effected concentrations as additional uncertainty is included in EF modeling.
**Seki et al (2008) do not reach 50 percent inhibitory concentrations but report an extrapolated EC_{50} value based on lower effect-level concentrations.

This curated data set demonstrates high variability between reported values, with at least two orders of magnitude difference in every trophic level and five orders of magnitude difference across all species. In spite of ongoing improvements to toxicity testing for ENMs (Petersen et al. 2015) there is general consensus that C_{60} presents relatively low hazard to aquatic species (Andrievsky et al. 2005). As noted in Table 3, many of the studies compare fullerene toxicity between:

1) Alternative sample preparation methods (Lovern and Klaper 2006; Seki 2008; Usenko et al. 2007; Zhu et al. 2006; Zhu et al. 2007) to elucidate the extent to which solvents or other contaminants may cause erroneously high toxicity estimates (Henry et al. 2011; Kovochich et al. 2009), and

2) Testing conditions exposed to light or kept in darkness (Gelca et al. 2012; Ji et al. 2014; Usenko et al. 2008) to understand the importance of photoexcitation and degradation in driving toxicity (Kolosnjaj et al. 2007).

A noteworthy source of uncertainty is converting acute, no observed effect concentration (NOEC), and lowest observed effect concentration (LOEC) endpoints reported in the majority of studies into equivalent chronic EC_{50} values by dividing by an acute to chronic ratio of 2 (Huijbregts 2010a), 1/9, and 4/9 respectively, following studies for non-cancer endpoints (Eckelman et al. 2012; Huijbregts et al. 2005). We apply these factors consistently across both materials, and do not test the sensitivity of CFs to these assumptions.

The conventional alternative niacinamide again is the subject of relatively fewer studies than the emerging material C_{60}. Reported toxicity data for niacinamide are consistently greater than C_{60} by at least two orders of magnitude, and all exceed 1 g/L as shown in Table 7.

Table 7 Data from individual ecotoxicity studies of niacinamide

Reference	Species n=3	Test type and endpoint	Reported value(s)	EC_{50} value
Producers				
OECD SIDS, 2002	*S. subspicatus*	72h Acute EC_{50}	>1000 mg/L	500 mg/L
	Algae - generic	QSAR, 96h Accute EC_{50}	8,934 mg/L	4,500 mg/L
Primary consumers				
OECD SIDS, 2002	*D. magna*	24h Acute EC_{50}	>1000 mg/L	500 mg/L
	Daphnid - generic	48h Acute EC_{50}, QSAR	16,456 mg/L	8,000 mg/L
Secondary consumers				
OECD SIDS, 2002	*P. reticulata*	96h Acute LC_{50}	>1000 mg/L	500 mg/L

	Fish - generic	96h Acute LC_{50}, QSAR	18,189 mg/L	9,000 mg/L
ECOTox database*	*X. laevis*	96h Acute EC_{50}, embryonic	0.34 mg/L	0.17 mg/L

*Misclassified data point contained in ECOTox database.

Consistent with our treatment of C_{60} ecotoxicity studies we multiply the acute toxicity data reported in Table 4 by 1/2. The dataset contains a misclassified acute EC_{50} value of 0.34 mg/L reported in the ECOTox and RIVM ETox databases (RIVM 2015; USEPA 2015a), which references a study that considers nicotine and 6-aminonicotinamide (Dawson and Wilke 1991) not nicotinamide, and has been brought to the attention of the respective database managers. Unfortunately, this is the only value implemented in the recently released USEtox 2.0, which results in a niacinamide ecotoxicity CF for emission to freshwater on the order of 10^5 PAF m^3 d/kg – surprisingly large for a vitamin B derivative widely considered to be innocuous at relevant commercial and environmental concentrations (CIREP 2005). Thus we exclude this value in calculating EFs for niacinamide, although the influence of the data point on aggregate multi-species toxic concentration (aveLog EC_{50}) estimation and standard error on the mean (SEM) calculation is significant (SI 2.3.1).

To calculate aveLog EC_{50} from the individual studies reported in Tables 3 and 4, we take the log of the geometric mean of each trophic class, and then calculate the arithmetic mean of these values (Huijbregts 2010a) (SI 2.3.2). This represents the concentration at which half of aquatic species are exposed above their median EC_{50} values, and is 0.43 and 3.2 log mg/L for C_{60} and niacinamide respectively. We calculate the SEM from the log EC_{50} data, which is 0.12 for C_{60} and 0.04 for niacinamide (SI 2.3.2). Uncertainty in the average toxicity ($\overline{ave\ Log}$) follows a Student's t distribution

(Golsteijn et al. 2012; Van Zelm et al. 2007) centered around aveLog EC_{50} and scaled by the SEM, shown in Eq. 2:

$$\overline{ave\ Log} = ave\ Log\ EC_{50} + SEM * t \qquad \text{Eq. 2}$$

Where t represents a two-tailed t-distribution with n-1 degrees of freedom from n different species with experimental toxicity data (SI 2.3.2).

3 Results and Discussion

Freshwater aquatic ecotoxicity CFs for C_{60} and niacinamide emitted directly to urban air, continental freshwater, and natural soil (Figure 10 A-C) show approximately two orders of magnitude variability resulting from the assumed plus-or-minus one order of magnitude in the baseline scenario. These results are generated through the full sampling of distributions specified in Tables 4 and 5 as well as $\overline{ave\ Log}$ for each material, and thus represent the global sensitivity of freshwater aquatic ecotoxicity CFs to simultaneous changes in all substance properties. Emissions to rural air and agricultural soil show similar variability and order of preference, and niacinamide emissions to marine water are more than 15 orders of magnitude greater than C_{60} due to its resistance to removal via sedimentation (SI 3.1).

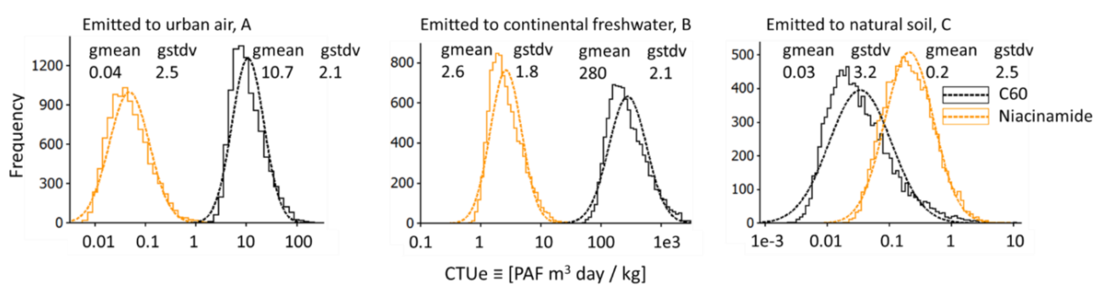

Fig 10 Stochastic aquatic ecotoxicity CFs for C_{60} (black) and niacinamide (orange) antioxidants emitted to urban air (A), freshwater (B), and natural soil (C) compartments. Solid lines are frequency distributions from 10,000 Monte Carlo runs and dashed lines

are normal distributions fit to the log-transformed data (i.e., CFs are log normal distributions).

For emissions to air and freshwater, niacinamide is characterized by a lower toxicity potential per unit mass than C_{60}, as opposed to emissions to soil in which case C_{60} has a lower average CF due to its strong partitioning to soil over water. For emission to freshwater, stochastic CFs for C_{60} and niacinamide are log normally distributed with a geometric mean of 280 and 2.6 and geometric standard deviation of 2.1 and 1.8, respectively. All of these differences are significant (Welch's t-test $p < 0.001$), with the closest scenario (i.e., emission to soil) yielding a Welch's t-test statistic < 0.05 (SI 3.2) (Fagerland and Sandvik 2009). Although model uncertainty is relatively well studied and beyond the scope of this study, these differences are significant with respect to model uncertainty, and variability in CFs in the baseline scenario is smaller in magnitude than estimated model uncertainty (Rosenbaum et al. 2008) (SI 3.3). Given baseline scenario assumptions, the hypothetical product developers can conclude that C_{60} has greater potential for ecotoxicity impacts per unit mass than niacinamide,

3.1 Identifying the Most Influential Substance Parameters

To estimate the relative influence of varied input parameters used to calculate C_{60} CFs we take the absolute value of the Spearman Rank Correlation Index for emissions to urban air, continental freshwater, and natural soil (Figure 11A-C). Spearman rank correlation assumes independence of observations within each parameter and makes no assumptions about the distribution type (Gauthier 2001). Many of the substance parameters in USEtox are themselves calculated as function of other substance input parameters using simple regressions, for example estimating Kdoc based on Kow, and are thereby not independent. We do not account for the interdependence of parameters as the focus is on identifying only the few most influential substance properties, although Fantke et al. (2012) demonstrate how to decouple true parameter uncertainty (e.g., in Kdoc) from regression-related uncertainty.

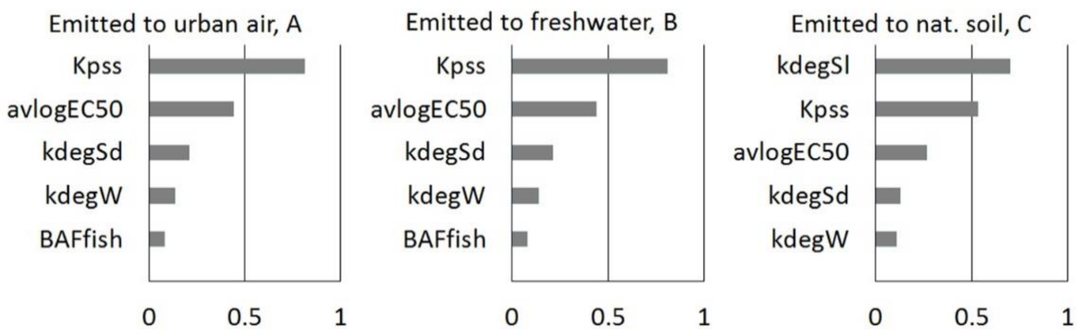

Fig 11 The five Spearman rank correlation indices with the greatest magnitude out of all variable inputs for three C_{60} aquatic ecotoxicity CFs. Greater magnitude indicates which input parameters have the greatest influence on CFs variability for each emission compartment.

Figure 11 calls attention to the importance of variability in the suspended solids partitioning coefficient (Kpss), $\overline{ave\ Log}$ aggregate ecotoxicity, and to a lesser extent sediment, aquatic, and soil degradation rates (KdegSd, KdegW, KdegSl) as driving variance in C_{60} CF results. Despite the large variability modeled for C_{60} solubility, this parameter has negligible effect on CFs (SI 2.2.1). The importance of removal through aggregation and sedimentation is consistent with recent reports for other ENMs (Dale et al. 2015). Thus we prioritize these parameters for C_{60} for further data refinement and future experimental research. In the case of niacinamide, uncertainty in degradation rates in air, water, and soil have the greatest influence for all emission scenarios, followed by Henry's constant, the organic-carbon partitioning coefficient, and average ave Log EC_{50} (SI 3.4).

3.2 Decomposing CFs into Fate, Exposure, and Effect Components

The two antioxidant compounds display significant differences in terms of their freshwater residence time (fate factor FF), dissolved fraction (exposure factor XF), and aggregate multi-species toxicity (effect factor EF) as shown in Figure 12A-C, and the product of these three yields the CF following equation 1.

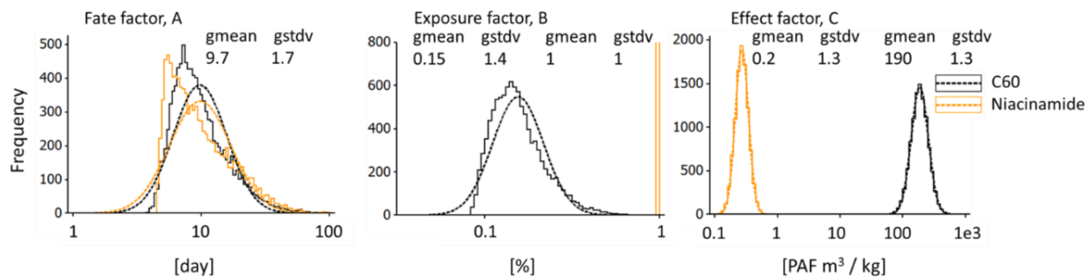

Fig 12 Component fate (A), exposure (B), and effect factors (C) for niacinamide (orange) and C_{60} (black) identify significant differences between the two antioxidants, specifically the high exposure and low toxicity of niacinamide compared to C_{60}. Solid lines are frequency distributions of 10,000 Monte Carlo runs and dashed lines are normal distributions fit to the log-transformed data.

FF for each material is equivalent, with partitioning and sedimentation the dominant removal route for C_{60} and biodegradation dominant for niacinamide. XF for niacinamide is effectively 1 – representing 100 percent of the emission being bioavailable – whereas the C_{60} XF has a geometric mean of 0.1 (corresponding 10 percent dissolved and bioavailable) because of strong partitioning to suspended solids, dissolved organic carbon, and biomass. The greatest difference between the two antioxidants is in EF, where C_{60} exceeds niacinamide by three orders of magnitude (geometric mean 190 vs 0.2), which is not surprising given the low ecotoxicity values reported for niacinamide in Table 4.

3.3 Refining Estimates of Variability for C_{60} Substance Data

Figure 2 indicates that, for the majority of input parameters in Tables 1 & 2, the assumed variability of plus-or-minus one order of magnitude has little influence on C_{60} aquatic ecotoxicity CFs. In the case of direct emission to freshwater, the suspended solids partitioning coefficient (Kpss) and average toxicity (aveLog EC_{50}) are prioritized for data refinement and promising candidates for further experimental investigation. The assumed Kpss with uniform variability between 3×10^3 and 3×10^5 L/kg is based on the USEtox 1.01 regression for estimating Kdoc from Kow, which does not warrant reduction from our high-uncertainty baseline scenario even though experimental values for Kow are available. C_{60} is expected to exhibit strong partitioning to suspended solids based on reported Koc values (PubChem 2015a), although there are reports of variable removal between 10 and 90 percent by high concentrations of heterogeneous biomass (which likely has a higher organic content than suspended solids) between alternative C_{60} preparation methods (Kiser et al. 2010). Thus, further reduction of variability in Kpss requires identification of dominant preparation methods and experimental investigation of C_{60} partitioning to suspended solids with realistic compositions and concentrations.

Uncertainty in aveLog EC_{50} for C_{60} is similarly influential to CFs and complicated by differences between C_{60} preparation methods, particularly regarding the presence of solvent residues and their potential contribution to erroneously high toxicity estimates. C_{60} used in cosmetics is commonly stabilized in castor oil or polymer coatings such as polyvinylpyrrolidone (Benn et al. 2011; Lens 2009), and likely will not be prepared using solvents. To explore the sensitivity of C_{60} EFs and CFs to preparation method, we exclude all studies in Table 3 that used solvents to stabilize C_{60} and calculate a revised EF with a geometric mean of 72 and revised CF of 31, as opposed to 187 and 280 in the baseline scenario including all preparation methods, (Figure 13A&B).

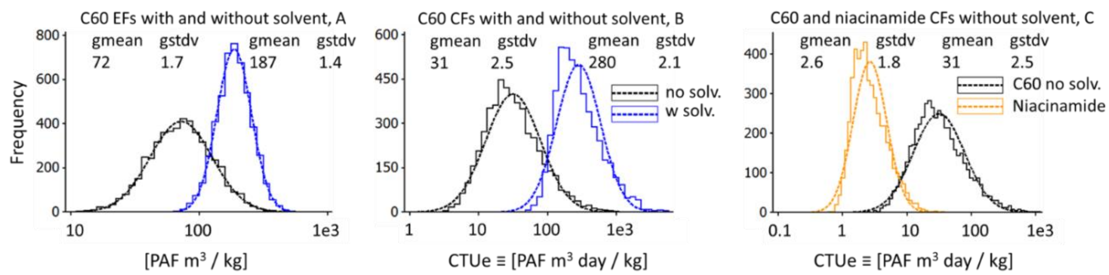

Fig 13 Removal of all ecotoxicity studies relying on solvents (black without, blue with) reduces the C_{60} effect factor (A) and characterization factor (B) by more than one order of magnitude. With no solvents the toxicity potential of C_{60} is closer to niacinamide (orange) but still significantly different for emissions to freshwater (C).

The revised CF for C_{60} emissions to freshwater still exceeds niacinamide by an order of magnitude (4C) and is significantly different (Welch's t test $p < 0.001$). This suggests that, if solvent residues are not present in C_{60} emissions, the aquatic ecotoxicity potential is marginally greater than niacinamide for direct emission to freshwater. For emissions to rural and continental air, the geometric mean of the C_{60} CF is at least two orders of magnitude greater than niacinamide, whereas for emissions to natural soil, agricultural, and marine water niacinamide significantly exceeds C_{60} (SI 3.4). Thus, the order of preference for the materials depends on the emission compartment. Furthermore, there is a critical need to: 1) characterize the form of C_{60} released regarding the presence of solvent residues, and 2) to design new experiments to elucidate suspended solids partitioning behavior.

4.0 Conclusion

LCIA method developers can apply the Monte Carlo tool to expedite expansion and review of toxicity databases by identifying the most influential substance data for distinct chemical classes, and then focusing their efforts on reducing parameter uncertainty on these estimates by finding or providing experimental references. Analogous to the case shown above, it is likely that only a few model input parameters are significant for each chemical class, and building consensus about uncertainty estimates for these parameters may allow future quantification of parameter uncertainty for all chemicals currently included in LCIA models (similar to what has been done for global estimates of model uncertainty). Furthermore, we encourage LCA practitioners to apply the Monte Carlo tool to the life cycle inventory items that contribute most to ecotoxicity impacts to increase confidence in interpretation of LCIA results.

In the context of emerging contaminants, calculating CFs stochastically allows practitioners to identify which input parameters are most influential to characterization results, and use this information to help prioritize experimental research agenda. Our results suggest that focusing experimental resources on improving data for suspended solids partitioning behavior and multi-species toxicity indicators has the greatest potential to reduce uncertainty of current C_{60} CF estimates. In this capacity, stochastic evaluation of impact assessment models to identify the most influential parameter uncertainties and inform future research agenda constitutes an example of *anticipatory* LCA (Wender et al. 2014a; Wender et al. 2014b).

The approach outlined in the present paper has potential for broader application to different LCIA models and other impact categories that use simplified fate and effect modeling based on variable substance properties. The controversy, parameter, and mechanistic uncertainty surrounding the environmental impacts of ENMs represents an opportunity to reevaluate LCIA estimates for commercially-available, well-studied chemicals. No midpoint impact assessment methods include formal uncertainty analysis, thus this approach could improve treatment and presentation of uncertainty for LCA of emerging and established technologies alike.

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References

Alfonsín C, Hospido A, Omil F, Moreira MT, Feijoo G (2014) PPCPs in wastewater – Update and calculation of characterization factors for their inclusion in LCA studies Journal of Cleaner Production 83:245-255 doi:http://dx.doi.org/10.1016/j.jclepro.2014.07.024

Alvarez PJ, Colvin V, Lead J, Stone V (2009) Research priorities to advance eco-responsible nanotechnology ACS nano 3:1616-1619

Andrievsky G, Klochkov V, Derevyanchenko L (2005) Is the C60 fullerene molecule toxic?! Fullerenes, Nanotubes, and Carbon Nanostructures 13:363-376

Avanasi R, Jackson WA, Sherwin B, Mudge JF, Anderson TA (2014) C60 Fullerene Soil Sorption, Biodegradation, and Plant Uptake Environmental Science & Technology 48:2792-2797 doi:10.1021/es405306w

Bare J, Young D, QAM S, Hopton M, Chief SAB (2012) Tool for the Reduction and Assessment of Chemical and other Environmental Impacts (TRACI)

Benn TM, Westerhoff P, Herckes P (2011) Detection of fullerenes (C 60 and C 70) in commercial cosmetics Environmental Pollution 159:1334-1342

Cellura M, Longo S, Mistretta M (2011) Sensitivity analysis to quantify uncertainty in Life Cycle Assessment: The case study of an Italian tile Renewable and Sustainable Energy Reviews 15:4697-4705 doi:http://dx.doi.org/10.1016/j.rser.2011.07.082

Chen C-Y, Jafvert CT (2009) Sorption of Buckminsterfullerene (C60) to Saturated Soils Environmental Science & Technology 43:7370-7375 doi:10.1021/es900989m

CIREP (2005) Cosmetic Ingredient Review Expert Panel Final Report of the Safety Assessment of Niacinamide and Niacin International Journal of Toxicology 24:1-31 doi:10.1080/10915810500434183

Cohen Y, Rallo R, Liu R, Liu HH (2013) In Silico Analysis of Nanomaterials Hazard and Risk Accounts of Chemical Research 46:802-812 doi:10.1021/ar300049e

Cucurachi S, Heijungs R (2014) Characterisation factors for life cycle impact assessment of sound emissions Science of The Total Environment 468–469:280-291 doi:http://dx.doi.org/10.1016/j.scitotenv.2013.07.080

Dale AL, Lowry GV, Casman EA (2015) Stream Dynamics and Chemical Transformations Control the Environmental Fate of Silver and Zinc Oxide Nanoparticles in a Watershed-Scale Model Environmental Science & Technology 49:7285-7293 doi:10.1021/acs.est.5b01205

Dawson DA, Wilke TS (1991) Evaluation of the frog embryo teratogenesis assay: Xenopus (FETAX) as a model system for mixture toxicity hazard assessment Environmental Toxicology and Chemistry 10:941-948 doi:10.1002/etc.5620100710

EC (2011) International reference life cycle data system (ILCD) handbook - Recomendations for life cycle impact assessment in the European context. First edition edn. Office of the European Union, Luxemburg

Eckelman MJ, Mauter MS, Isaacs JA, Elimelech M (2012) New Perspectives on Nanomaterial Aquatic Ecotoxicity: Production Impacts Exceed Direct Exposure Impacts for Carbon Nanotoubes Environmental Science & Technology 46:2902-2910 doi:10.1021/es203409a

Eisenberg DG, K; Hristozov, D; Bates, M and Linkov, I (2015) Risk Assessment, Life Cycle Assessment, and Decision Methods for Nanomaterials. In: Satinder Kaur Brar TCZ, Mausam Verma, Rao Y. Surampalli, and Rajeshwar D. Tyagi (ed) Nanomaterials in the Environment. American Society of Civil Engineers, Reston, Virgina, pp 383-419. doi:doi:10.1061/9780784414088.ch15

Fagerland MW, Sandvik L (2009) Performance of five two-sample location tests for skewed distributions with unequal variances Contemporary Clinical Trials 30:490-496 doi:http://dx.doi.org/10.1016/j.cct.2009.06.007

Fantke P, Gillespie BW, Juraske R, Jolliet O (2014) Estimating Half-Lives for Pesticide Dissipation from Plants Environmental Science & Technology 48:8588-8602 doi:10.1021/es500434p

Fantke P, Wieland P, Juraske R, Shaddick G, Itoiz ES, Friedrich R, Jolliet O (2012) Parameterization Models for Pesticide Exposure via Crop Consumption Environmental Science & Technology 46:12864-12872 doi:10.1021/es301509u

Gauthier TD (2001) Detecting Trends Using Spearman's Rank Correlation Coefficient Environmental Forensics 2:359-362

Gavankar S, Suh S, Keller AF (2012) Life cycle assessment at nanoscale: review and recommendations The International Journal of Life Cycle Assessment 17:295-303 doi:10.1007/s11367-011-0368-5

Gelca R, Anderson TA, Cox SB (2012) The Effect of Cluster Size on the Breakdown of C₆₀ Water Suspensions Into Toxic Compounds Advanced Science, Engineering and Medicine 4:205-210 doi:10.1166/asem.2012.1181

Gilbertson L, Wender B, Zimmerman J, Eckelman MJ (2015) Coordinating Modeling and Experimental Research of Engineered Nanomaterials to Improve Life Cycle Assessment Studies Environmental Science: Nano doi:10.1039/c5en00097a

Golsteijn L, Hendriks HWM, van Zelm R, Ragas AMJ, Huijbregts MAJ (2012) Do interspecies correlation estimations increase the reliability of toxicity estimates for wildlife? Ecotoxicology and Environmental Safety 80:238-243 doi:http://dx.doi.org/10.1016/j.ecoenv.2012.03.005

Grieger K, Baun A, Owen R (2010) Redefining risk research priorities for nanomaterials Journal of Nanoparticle Research 12:383-392 doi:10.1007/s11051-009-9829-1

Gust KA, Collier ZA, Mayo M, Stanley JK, Gong P, Chappell M (2015) Limitations of toxicity characterization in life cycle assessment–can adverse outcome pathways provide a new foundation? Integrated Environmental Assessment and Management

Hauschild M et al. (2013) Identifying best existing practice for characterization modeling in life cycle impact assessment The International Journal of Life Cycle Assessment 18:683-697 doi:10.1007/s11367-012-0489-5

Hauschild MZ et al. (2008) Building a Model Based on Scientific Consensus for Life Cycle Impact Assessment of Chemicals: The Search for Harmony and Parsimony Environmental Science & Technology 42:7032-7037 doi:10.1021/es703145t

Hauschild MZ, Huijbregts MA (2015) Life Cycle Impact Assessment LCA Compendium–The Complete World of Life Cycle Assessment (

Henderson A et al. (2011) USEtox fate and ecotoxicity factors for comparative assessment of toxic emissions in life cycle analysis: sensitivity to key chemical properties The International Journal of Life Cycle Assessment 16:701-709 doi:10.1007/s11367-011-0294-6

Hendren CO, Lowry GV, Unrine JM, Wiesner MR (2015) A functional assay-based strategy for nanomaterial risk forecasting Science of The Total Environment 536:1029-1037 doi:http://dx.doi.org/10.1016/j.scitotenv.2015.06.100

Henry TB, Petersen EJ, Compton RN (2011) Aqueous fullerene aggregates (nC60) generate minimal reactive oxygen species and are of low toxicity in fish: a revision of previous reports Current Opinion in Biotechnology 22:533-537 doi:http://dx.doi.org/10.1016/j.copbio.2011.05.511

Hischier R, Walser T (2012) Life cycle assessment of engineered nanomaterials: State of the art and strategies to overcome existing gaps Science of The Total Environment 425:271-282 doi:http://dx.doi.org/10.1016/j.scitotenv.2012.03.001

Huijbregts MA, Hauschild, M. Jolliet, O., Margni, M., McKone, T., Rosenbaum, R.K., and van de Meent, D. (2010a) USEtox User Manual.

Huijbregts MAJ, Rombouts LJA, Ragas AMJ, van de Meent D (2005) Human-toxicological effect and damage factors of carcinogenic and noncarcinogenic chemicals for life cycle impact assessment Integrated Environmental Assessment and Management 1:181-244 doi:10.1897/2004-007R.1

Huijbregts MM, M; van de Meent, D; Jollier, O; Rosenbaum, RK; McKone, T and Hauschild, M. (2010b) USEtox chemcial-specific database: Organics.

Huijbregts MM, M; van de Meent, D; Jollier, O; Rosenbaum, RK; McKone, T and Hauschild, M. (2010c) USEtoxTM Chemical-specific database: organics

Jafvert CT, Kulkarni PP (2008) Buckminsterfullerene's (C60) Octanol−Water Partition Coefficient (Kow) and Aqueous Solubility Environmental Science & Technology 42:5945-5950 doi:10.1021/es702809a

Ji K-h, Kim J-k, Choi K-h (2014) Sunlight Enhances Toxicity of Fullerene (C 60) to Freshwater Invertebrates Daphnia magna and Moina macrocopa The Korean Journal of Public Health 51:35-45

Jolliet O, Fantke P (2015) Human Toxicity. In: Life Cycle Impact Assessment. Springer, pp 75-96

Kiser MA, Ryu H, Jang H, Hristovski K, Westerhoff P (2010) Biosorption of nanoparticles to heterotrophic wastewater biomass Water research 44:4105-4114 doi:http://dx.doi.org/10.1016/j.watres.2010.05.036

Klopffer W, Curran, M.A., Frankl, P., Heijung, R., Kohler, A., and Olsen, S.I. (2007) Nanotechnology and life cycle assessment: A systems approach to nanotechnology and the environment. Woodrow Wilson International Center for Scholars,

Kolosnjaj J, Szwarc H, Moussa F (2007) Toxicity Studies of Fullerenes and Derivatives. In: Chan WW (ed) Bio-Applications of Nanoparticles, vol 620. Advances in Experimental Medicine and Biology. Springer New York, pp 168-180. doi:10.1007/978-0-387-76713-0_13

Kovochich M et al. (2009) Comparative Toxicity of C60 Aggregates toward Mammalian Cells: Role of Tetrahydrofuran (THF) Decomposition Environmental Science & Technology 43:6378-6384 doi:10.1021/es900990d

Kümmerer K, Menz J, Schubert T, Thielemans W (2011) Biodegradability of organic nanoparticles in the aqueous environment Chemosphere 82:1387-1392 doi:http://dx.doi.org/10.1016/j.chemosphere.2010.11.069

Lens M (2009) Use of Fullerenes in Cosmetics Recent Patents on Biotechnology 3:118-123 doi:10.2174/187220809788700166

Li D, Fortner JD, Johnson DR, Chen C, Li Q, Alvarez PJJ (2010) Bioaccumulation of 14C60 by the Earthworm Eisenia fetida Environmental Science & Technology 44:9170-9175 doi:10.1021/es1024405

Lloyd SM, Ries R (2007) Characterizing, Propagating, and Analyzing Uncertainty in Life-Cycle Assessment: A Survey of Quantitative Approaches Journal of Industrial Ecology 11:161-179 doi:10.1162/jiec.2007.1136

Lovern SB, Klaper R (2006) Daphnia magna mortality when exposed to titanium dioxide and fullerene (C60) nanoparticles Environmental Toxicology and Chemistry 25:1132-1137

Miseljic M, Olsen S (2014a) Life-cycle assessment of engineered nanomaterials: a literature review of assessment status Journal of Nanoparticle Research 16:1-33 doi:10.1007/s11051-014-2427-x

Miseljic M, Olsen SI (2014b) Life-cycle assessment of engineered nanomaterials: a literature review of assessment status Journal of Nanoparticle Research 16:1-33

NSTCCT (2014) Progress review on the coordinated implementation of the national nanotechnology initiative 2011 environmental, health, and safety research strategy. Washington, DC

PEN (2013) Consumer Products Inventory. Project on Emerging Nanotechnologies. http://www.nanotechproject.org/cpi. Accessed Spetember 2015

Pennington DW, Margni M, Ammann C, Jolliet O (2005) Multimedia Fate and Human Intake Modeling: Spatial versus Nonspatial Insights for Chemical Emissions in Western Europe Environmental Science & Technology 39:1119-1128 doi:10.1021/es034598x

Petersen EJ et al. (2015) Adapting OECD Aquatic Toxicity Tests for Use with Manufactured Nanomaterials: Key Issues and Consensus Recommendations Environmental Science & Technology 49:9532-9547 doi:10.1021/acs.est.5b00997

PubChem (2015a) Compound Summary for Buckminsterfullerene. National Center for Biotechnology Information. https://pubchem.ncbi.nlm.nih.gov/compound/123591. Accessed October 27 2015

PubChem (2015b) Compound Summary for Nicotinamide. National Center for Biotechnology Information. https://pubchem.ncbi.nlm.nih.gov/compound/936. Accessed September 9 2015

Pycke BF, Chao T-C, Herckes P, Westerhoff P, Halden RU (2012) Beyond nC60: strategies for identification of transformation products of fullerene oxidation in aquatic and biological samples Analytical and bioanalytical chemistry 404:2583-2595

e-ToxBase (2015) National Institute for Public Health and the Environment. http://www.ru.nl/environmentalscience/research/themes-0/risk-assessment/e-toxbase/.

Rodriguez-Garcia G, Zimmermann B, Weil M (2014) Nanotoxicity and Life Cycle Assessment: First attempt towards the determination of characterization factors for carbon nanotubes IOP Conference Series: Materials Science and Engineering 64:012029

Rosenbaum R et al. (2008) USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment The International Journal of Life Cycle Assessment 13:532-546 doi:10.1007/s11367-008-0038-4

Rosenbaum RK (2015) Ecotoxicity. In: Life Cycle Impact Assessment. Springer, pp 139-162

Saleh NB, Aich N, Plazas-Tuttle J, Lead JR, Lowry GV (2015) Research strategy to determine when novel nanohybrids pose unique environmental risks Environmental Science: Nano 2:11-18 doi:10.1039/c4en00104d

Salieri B, Righi S, Pasteris A, Olsen SI (2015) Freshwater ecotoxicity characterisation factor for metal oxide nanoparticles: A case study on titanium dioxide nanoparticle Science of The Total Environment 505:494-502 doi:http://dx.doi.org/10.1016/j.scitotenv.2014.09.107

Saltelli A et al. (2008) Global sensitivity analysis: the primer. John Wiley & Sons,

Seager TP, Linkov I (2008) Coupling multicriteria decision analysis and life cycle assessment for nanomaterials Journal of Industrial Ecology 12:282-285

Seki MF, S; Gondo, Y; Inoue, Y; Nozaka, T and Takatsuki, M (2008) Acute toxicity of fullerene C60 in aquatic organisms Environmental Science (Japan) 21:53-62

Sonnemann GW, Schuhmacher M, Castells F (2003) Uncertainty assessment by a Monte Carlo simulation in a life cycle inventory of electricity produced by a waste incinerator Journal of Cleaner Production 11:279-292 doi:http://dx.doi.org/10.1016/S0959-6526(02)00028-8

Tiwari AJ, Morris JR, Vejerano EP, Hochella MF, Marr LC (2014) Oxidation of C60 Aerosols by Atmospherically Relevant Levels of O3 Environmental Science & Technology 48:2706-2714 doi:10.1021/es4045693

UNEP (2002) OECD SIDS Initial Assessment Report - nicotinamide.

Usenko CY, Harper SL, Tanguay RL (2007) In vivo evaluation of carbon fullerene toxicity using embryonic zebrafish Carbon 45:1891-1898 doi:http://dx.doi.org/10.1016/j.carbon.2007.04.021

Usenko CY, Harper SL, Tanguay RL (2008) Fullerene C60 exposure elicits an oxidative stress response in embryonic zebrafish Toxicology and Applied Pharmacology 229:44-55 doi:http://dx.doi.org/10.1016/j.taap.2007.12.030

USEPA (2010) Interim Technical Guidance for Assessing Screening Level Environmental Fate and Transport of, and General Population, Consumer, and Environmental Exposure to Nanomaterials. United States Environmental Protection Agency,

ECOTox User Guide: ECOTOX Database System. Version 4.0 (2015a) http:/www.epa.gov/ecotox.

USEPA (2015b) Estimation Programs Interface Suite™ for Microsoft® Windows, v 4.11. United State Enivronmental Protection Agency, Washington, DC, USA

Van Zelm R, Huijbregts MA, Harbers JV, Wintersen A, Struijs J, Posthuma L, Van de Meent D (2007) Uncertainty in msPAF-based ecotoxicological effect factors for freshwater ecosystems in life cycle impact assessment Integrated Environmental Assessment and Management 3:e6-e37

van Zelm R, Huijbregts MAJ (2013) Quantifying the Trade-off between Parameter and Model Structure Uncertainty in Life Cycle Impact Assessment Environmental Science & Technology 47:9274-9280 doi:10.1021/es305107s

van Zelm R, Huijbregts MJ, van de Meent D (2009) USES-LCA 2.0—a global nested multi-media fate, exposure, and effects model The International Journal of Life Cycle Assessment 14:282-284 doi:10.1007/s11367-009-0066-8

Weidema BPB, Ch.; Hischier, R.; Mutel, Ch.; Nemecek, T.; Reinhard, J.; Vadenbo, C.O.; Wernet, G (2013) The ecoinvent database: Overview and methodology, Data quality guideline for the ecoinvent database version 3.

Wender BA et al. (2014a) Anticipatory life-cycle assessment for responsible research and innovation Journal of Responsible Innovation 1:200-207 doi:10.1080/23299460.2014.920121

Wender BA et al. (2014b) Illustrating Anticipatory Life Cycle Assessment for Emerging Photovoltaic Technologies Environmental Science & Technology 48:10531-10538 doi:10.1021/es5016923

Westh T, Hauschild M, Birkved M, Jørgensen M, Rosenbaum R, Fantke P (2015) The USEtox story: a survey of model developer visions and user requirements The International Journal of Life Cycle Assessment 20:299-310 doi:10.1007/s11367-014-0829-8

Yang Y, Wang Y, Hristovski K, Westerhoff P (2015) Simultaneous removal of nanosilver and fullerene in sequencing batch reactors for biological wastewater treatment Chemosphere 125:115-121 doi:http://dx.doi.org/10.1016/j.chemosphere.2014.12.003

Zhu S, Oberdörster E, Haasch ML (2006) Toxicity of an engineered nanoparticle (fullerene, C60) in two aquatic species, Daphnia and fathead minnow Marine Environmental Research 62, Supplement 1:S5-S9 doi:http://dx.doi.org/10.1016/j.marenvres.2006.04.059

Zhu X, Zhu L, Li Y, Duan Z, Chen W, Alvarez PJ (2007) Developmental toxicity in zebrafish (Danio rerio) embryos after exposure to manufactured nanomaterials: buckminsterfullerene aggregates (nC60) and fullerol Environmental Toxicology and Chemistry 26:976-979

CHAPTER 5

SYNTHESIS

This synthesis chapter integrates findings from component studies to address the guiding research question of "**how life cycle assessment (LCA) can be improved to inform responsible research and innovation (RRI) of emerging technologies?"** In answering this question the dissertation: 1) Identifies several limitations in current LCA practice that impede application of LCA early in research and development based on capacities summarized in the growing literature describing RRI (Wender et al. 2014a), 2) Introduces a framework for *anticipatory* LCA that addresses these limitations by including elements of technology forecasting, social engagement, stochastic risk modeling, and multi criteria decision analysis (Wender et al. 2014b), and 3) Demonstrates a Monte Carlo human and ecotoxicity impact assessment tool based on the consensus model USEtox that explores uncertainty to inform development of future research agenda. The component chapters and contributions address specific questions, yet are interrelated and build off one another as summarized in the dissertation graphical abstract shown in Figure 14.

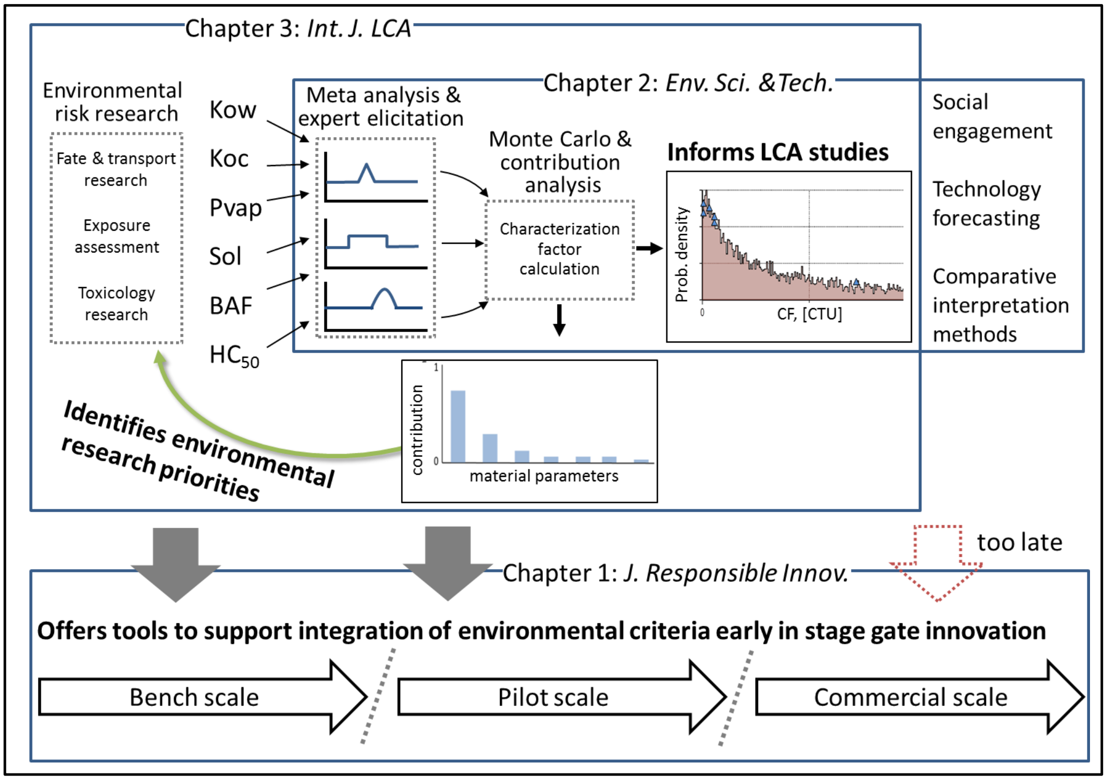


Figure 14 Graphical Abstract Development of anticipatory LCA tools facilitates exploration of uncertainty in life cycle inventory data and impact assessment models for emerging technologies, identifies those parameters with the greatest contribution to uncertainty in life cycle environmental impacts, and offers a pathway to integrate environmental criteria early in technology development**.**

Each chapter adopts increasingly narrow boundaries of analysis: Chapter One begins with the broader social context, Chapter Two identifies three specific interventions, and Chapter Three explores one of these interventions in a detailed case study. The dissertation describes the limitations of current LCA practices and proposes methodological advances using illustrative examples of nanotechnology and photovoltaics, nonetheless the work is focused the process and methods of environmental assessment. Thus, this dissertation is not about nanomaterials or photovoltaics being good or bad for the environment. Instead, the work contributes improved LCA methods that emphasize uncertainty and sensitivity analyses to inform contemporary decisions with an eye towards the research and decisions with the greatest potential to improve environmental attributes.

Chapter One contextualizes LCA within literature from the burgeoning field of RRI (Guston et al. 2014; Stilgoe et al. 2013; von Schomberg 2013), and suggests LCA as a holistic technology assessment method with potential to operationalize the otherwise conceptual discussions of RRI. The chapter reviews current practices in LCA and emphasizes the limitations and opportunities that are relevant to RRI. Specifically, the chapter concludes that the extensive reliance on commercial-scale data – data that is inherently not available for emerging technologies and their product applications – renders LCA as retrospective and ill-suited for enabling the objectives RRI. Another limitation identified in Chapter One is the lack of treatment of alternative stakeholder perspectives in LCA practice. To broaden the range of values and perspectives included in LCA, the chapter identifies several practitioner decisions that are value-based and suggest conducting social engagement activities to evaluate how various stakeholders may select differently. These limitations speak directly to the core RRI capacities of anticipation and inclusion, and the chapter concludes that advancements in LCA methods are necessary to overcome these limitations and align LCA with RRI.

Chapter Two builds on the objectives and critiques presented above by introducing an iterative framework for anticipatory LCA that is forward looking, decision directed, inclusive of alternative stakeholder perspectives, and focused on uncertainty and sensitivity analysis to inform future research agenda. The chapter demonstrates each component of the anticipatory LCA framework using the illustrative example of photovoltaic (PV) technologies to identify:

1) The greatest opportunity to improve life cycle greenhouse gas (GHG) savings of monocrystalline silicon PV panels is to reduce manufacturing energy consumption as opposed in marginally improving use phase efficiency, although the latter dominates current research efforts;
2) The environmental benefits of PV, and greatest opportunities for further improvements, quantified in an LCA depend strongly on the selection of functional unit and system boundaries, which will be specified differently by PV users and manufacturers;
3) Instances when PV panels contain novel materials – for example incorporation of carbon nanotubes or C_{60} fullerenes into organic PV modules – require impact assessment methods that account for relatively greater uncertainty, and the results of which have potential to prioritize future research to focus on the risk relevant parameters with the greatest influence on environmental impacts; and
4) Data demands and cognitive limitations faced in interpretation of comparative LCA results can be greatly reduced through inclusion of decision analysis tools that sort environmental impacts relative to data certainty.

The proposed anticipatory LCA framework identifies three intervention points (among many) at which communication of environmental findings to specific innovation actors can guide future research agenda. Identification and communication of the processes and material parameters along the product life cycle with the greatest potential for environmental improvement can support integration of environmental criteria into design, funding, and risk research decisions. The chapter concludes that applying this framework iteratively alongside development of emerging technologies can help operationalize the principals of RRI.

Chapter three focuses on one of the identified interventions in greater detail, specifically exploring how uncertainty and sensitivity analysis of life cycle impact assessment (LCIA) methods can inform risk research for emerging technologies by identifying the material parameters and associated uncertainties that are most influential to model results. The chapter introduces a Monte Carlo tool based on the consensus human and ecotoxicity impact assessment model USEtox (Westh et al. 2015) that allows users to specify all required substance data as probability distributions, presents CF results as frequency distributions, and compares the relative influence of variability in each material parameter. Applying this tool to a comparative case study of niacinamide and the engineered nanomaterial C_{60}, both of which are used in low concentrations in commercial personal care products, suggests that research to improve understanding in C_{60}-suspended solids partitioning behavior has the greatest potential to improve certainty in human and ecotoxicity estimates for this emerging contaminant. The sensitivity-based approach to research prioritization demonstrated in this chapter has potential for broader applicability to other emerging contaminants characterized by high uncertainty or to other impact categories beyond human or ecotoxicty. The completed software is freely available and continuing collaborations with USEtox model developers will promote broad dissemination.

Synthesizing the recommendations of each chapter into a brief answer to the dissertation's guiding question: to support the objectives of RRI LCA methods must become forward looking (e.g., by overcoming reliance on historical data), integrative of diverse viewpoints (e.g., by comparing alternative model assumptions informed by stakeholder engagement), and generate knowledge that is useful for contemporary decisions that influence the products eventual environmental impacts.

Dissertation boundaries and limitations

Although this dissertation draws motivation from the growing movement for RRI, the work stops short of claiming RRI as an explicit outcome. More subtly, this work aligns with the principals of RRI but focuses on the methods that make this otherwise conceptual work practicable. Thus, the principal outcome of this work is a framework and impact assessment tool that can be applied to help anticipate potential environmental impacts of emerging technologies, explore diverse viewpoints through environmental assessment, and integrate environmental criteria into technology development criteria. Although case studies are used to demonstrate these tools, the dissertation also does not claim improved photovoltaic or nanotechnologies as an outcome. Furthermore, although the dissertation focuses on improving technology assessment methods to align with RRI, the advances presented herein will not *de facto* result in RRI or product improvements, but will require sustained efforts. The remainder of this synthesis surveys noteworthy limitations and concludes with two examples of research organizations applying the Monte Carlo tool based on USEtox to support environmentally responsible innovation, although the associated projects and future publications are outside the scope of this dissertation.

LCA for the process not the product – The methods presented in this work must be applied iteratively, as opposed to being applied once to reach a conclusive result, which requires a shift in perspective by some decision makers and practitioners. For example, the Monte Carlo impact assessment tool based on USEtox can identify research strategies with the greatest potential to reduce uncertainty, which in turn will result in new parameter estimates for input to the model and new results generated. In this capacity, anticipatory LCA is not static, co-evolves with the technology being studied, and serves as a holistic framework to organize existing knowledge and prioritize future data needs (McKone et al. 2011).

Diverse disciplinary perspectives – The anticipatory LCA framework requires input from a breadth of disciplinary perspectives and cannot be applied by an analyst and database in isolation. For example, Chapter 2 describes the need for stakeholder engagement through social science methods such as interviews, focus groups, and structured workshops to iteratively explore alternative perspectives (e.g., technology user versus manufacturer) in modeling assumptions and results. This requires training and skills not typical for LCA practitioners, and may be prohibitive of broader application. Thus, the anticipatory LCA framework is best suited for application by large research organizations (e.g., Intel or GE), large funding agencies or government technology assessment organizations (e.g., GAO), or interdisciplinary research teams.

Reduced data needs at the expense of greater modeling efforts – A benefit of the anticipatory LCA framework and stochastic impact assessment tool is that their application addresses data shortages and high uncertainty in the context of emerging technologies. In the absence of low-uncertainty data, the anticipatory approach emphasizes sensitivity analyses and iterative model refinement to identify which data needs are most relevant in a specified decision context. In practice this shifts efforts data collection needs toward increased efforts in analysis and interpretation. Specifically, the framework and stochastic impact assessment tool are designed to be applied iteratively and with the understanding that there is not necessarily one correct answer. Analysts must be prepared to develop multiple life cycle models based on different assumptions (e.g., system boundaries, functional units), compare results and identify salient differences, communicate findings to decision makers and active stakeholders, and then reevaluate each model. Thus reductions in data needs may be offset by greater demand in analysis and interpretation.

Securing decision maker buy in and describing the decision context – The tools introduced in this dissertation require contributions and effort on the part of the decision maker, not just in interpretation of results but in defining the decision context. This must include what technologies are being compared, what essential functions these serve and what figures of merit are used to compare alternatives, and what influence the decision maker can have on the extended product system. All of this in turn will shape the boundaries of analysis and inform iterative modeling decisions such as definition of functional unit. In the two illustrative use scenarios presented below, the decision context is specified for use of the stochastic impact assessment tool by two different research organizations.

Example usage scenarios

USEtox model developers – The USEtox human and ecotoxicity impact assessment model was developed by an international consortium of researchers under sponsorship of the United Nations Environmental Program. The principal mission of USEtox was to help provide tools that address human and ecotoxicity impacts in comparative technology assessment. USEtox is considered best practice in LCA and widely viewed as successful in building consensus around best methods to include this important category of impacts in technology assessment. Unfortunately, the USEtox model has large substance-specific data requirements, high uncertainty, and greater complexity than models used in other impact categories. Thus, USEtox model developers face serious challenges in quantifying the magnitude and significance of parameter uncertainty associated with USEtox CFs, as to date there are only estimates of model uncertainty.

The Monte Carlo tool based on USEtox has been shared with model developers to help reduce data needs and expedite expansion and adoption of the model. Specifically, calculating stochastic CFs allows full sensitivity analysis of all variable inputs, which in turn identifies the material parameters that are most influential to results as high priority for further efforts in data collection. Likewise, parameters that have little influence on Cfs for a given chemical class do not require efforts to define estimates of parameter uncertainty. Thus, USEtox model developers only have to define parameter uncertainty estimates for 2-4 substance parameters per chemical class (often 6 used), which is small and achievable compared to more than 10 parameters for all substances. In this capacity, the USEtox team will apply the tool to rapidly estimate the parameter uncertainty associated with USEtox CFs. This will improve decision maker confidence in interpreting comparative toxicity results and allow direct comparison of parameter and model uncertainty.

The US Army Corps of Engineers – The USACE is tasked with environmental assessment and improvement of the Nation's military assets, and is in the process of evaluating the potential environmental implications of novel munitions compounds that will replace the explosive TNT. USACE researchers are applying LCA to develop a holistic understanding of the environmental impacts of these novel compounds, which requires significant experimental efforts to collect sufficient fate, exposure, and toxicological data. In personal communications USACE researchers have shared Gantt charts detailing years of planned experiments in addition to the numerous studies already published. Thus USACE researchers are faced to allocate fixed research resources across a portfolio of possible efforts without clear understanding of the significance of each experiment on life cycle impacts.

Development of the Monte Carlo tool based on USEtox was funded in part by the USACE because the tool may help inform future investments in risk research of novel munitions compounds by identifying material parameters with the greatest influence on life cycle toxicity potential as high priorities for further experimental investigation. Conversely, material parameters with relatively little influence on results can be made a lower priority, thereby conserving research resources. In this way, USACE researchers can utilize the Monte Carlo tool based on current understanding of material properties and associated uncertainty to understand: 1) the human and ecotoxic potential of novel munitions compounds as compared to conventional alternatives, 2) identify the components within novel munitions compounds that are most problematic, and 3) prioritize experimental agenda with the greatest potential to improve certainty in interpretation of model results.

References

Guston DH, Fisher E, Grunwald A, Owen R, Swierstra T, van der Burg S (2014) Responsible innovation: motivations for a new journal Journal of Responsible Innovation 1:1-8

McKone T et al. (2011) Grand challenges for life-cycle assessment of biofuels Environmental Science & Technology 45:1751-1756

Stilgoe J, Owen R, Macnaghten P (2013) Developing a framework for responsible innovation Research Policy 42:1568-1580 doi:http://dx.doi.org/10.1016/j.respol.2013.05.008

von Schomberg R (2013) A Vision of Responsible Research and Innovation. In: Responsible Innovation. John Wiley & Sons, Ltd, pp 51-74. doi:10.1002/9781118551424.ch3

Wender BA et al. (2014a) Anticipatory life-cycle assessment for responsible research and innovation Journal of Responsible Innovation 1:200-207 doi:10.1080/23299460.2014.920121

Wender BA et al. (2014b) Illustrating Anticipatory Life Cycle Assessment for Emerging Photovoltaic Technologies Environmental Science & Technology 48:10531-10538 doi:10.1021/es5016923

Westh T, Hauschild M, Birkved M, Jørgensen M, Rosenbaum R, Fantke P (2015) The USEtox story: a survey of model developer visions and user requirements The International Journal of Life Cycle Assessment 20:299-310 doi:10.1007/s11367-014-0829-8

REFERENCES

Alfonsín C, Hospido A, Omil F, Moreira MT, Feijoo G (2014) PPCPs in wastewater – Update and calculation of characterization factors for their inclusion in LCA studies Journal of Cleaner Production 83:245-255 doi:http://dx.doi.org/10.1016/j.jclepro.2014.07.024

Alsema, E. A. Energy pay-back time and CO2 emissions of PV systems. Progress in Photovoltaics: Research and Applications 2000, 8 (1), 17-25.

Alsema, E.; de Wild-Scholten, M. The real environmental impacts of crystalline silicon PV modules: an analysis based on up-to-date manufacturers data, Presented at the 20th European Photovoltaic Solar Energy Conference, Barcelona, ES; 2005.

Alsema, E.; Frankl, P.; Kato, K. Energy pay-back time of photovoltaic energy systems: present status and prospects. Presented at the 2nd World Conference on Photovoltaic Solar Energy Conversion, Vienna, AU; 1998.

Alvarez PJ, Colvin V, Lead J, Stone V (2009) Research priorities to advance eco-responsible nanotechnology ACS nano 3:1616-1619

Andersen, O. Unintended Consequences of Renewable Energy. Springer London, London: 2013.

Andrievsky G, Klochkov V, Derevyanchenko L (2005) Is the C60 fullerene molecule toxic?! Fullerenes, Nanotubes, and Carbon Nanostructures 13:363-376

Arvidsson, R.; Kushnir, D.; Sandén, B. A.; Molander, S. Prospective Life Cycle Assessment of Graphene Production by Ultrasonication and Chemical Reduction. Environ. Sci. Technol. 2014, DOI: 10.1021/es405338k.

Avanasi R, Jackson WA, Sherwin B, Mudge JF, Anderson TA (2014) C60 Fullerene Soil Sorption, Biodegradation, and Plant Uptake Environmental Science & Technology 48:2792-2797 doi:10.1021/es405306w

Bare J, Young D, QAM S, Hopton M, Chief SAB (2012) Tool for the Reduction and Assessment of Chemical and other Environmental Impacts (TRACI)

Benn, T. M. and P. Westerhoff (2008). "Nanoparticle Silver Released into Water from Commercially Available Sock Fabrics." Environmental Science & Technology 42(11): 4133-4139.

Benn TM, Westerhoff P, Herckes P (2011) Detection of fullerenes (C 60 and C 70) in commercial cosmetics Environmental Pollution 159:1334-1342

Berube, David M. 2013. "Socialis Commodis and Life Cycle Analysis: A Critical Examination Of." In Emerging Technologies: Socio-Behavioral Life Cycle Approaches, edited by Nora Savage, Michael Gorman, and Anita Street, 139-164. Singapore: Pan Stanford Publishing.

Bhander, G. S., M. Hauschild, et al. (2003). "Implementing life cycle assessment in product development." Environmental Progress 22(4): 255-267.

Bhattacharyya, S.; Kymakis, E.; Amaratunga, G. A. J. Photovoltaic Properties of Dye Functionalized Single-Wall Carbon Nanotube/Conjugated Polymer Devices. Chemistry of Materials 2004, 16 (23), 4819-4823.

Caduff, M.; Huijbregts, M. A. J.; Althaus, H.-J.; Koehler, A.; Hellweg, S. Wind Power Electricity: The Bigger the Turbine, The Greener the Electricity? Environ. Sci. Technol.2012, 46 (9), 4725-4733.

Canis, L.; Linkov, I.; Seager, T. P., Application of stochastic multiattribute analysis to assessment of single walled carbon nanotube synthesis processes. Environ. Sci. & Technol. 2010, 44 (22), 8704-8711.

Cellura M, Longo S, Mistretta M (2011) Sensitivity analysis to quantify uncertainty in Life Cycle Assessment: The case study of an Italian tile Renewable and Sustainable Energy Reviews 15:4697-4705 doi:http://dx.doi.org/10.1016/j.rser.2011.07.082

Chen C-Y, Jafvert CT (2009) Sorption of Buckminsterfullerene (C60) to Saturated Soils Environmental Science & Technology 43:7370-7375 doi:10.1021/es900989m

CIREP (2005) Cosmetic Ingredient Review Expert Panel Final Report of the Safety Assessment of Niacinamide and Niacin International Journal of Toxicology 24:1-31 doi:10.1080/10915810500434183

Ciroth, A.; Muller, S.; Weidema, B.; Lesage, P. Empirically based uncertainty factors for the pedigree matrix in ecoinvent. Int. J. Life Cycle Assess. 2013, 1-11; DOI 10.1007/s11367-013-0670-5

Cohen Y, Rallo R, Liu R, Liu HH (2013) In Silico Analysis of Nanomaterials Hazard and Risk Accounts of Chemical Research 46:802-812 doi:10.1021/ar300049e

Collinge, William O., Amy E. Landis, Alex K. Jones, Laura A. Schaefer, and Melissa Bilec. 2013. "A Dynamic Life Cycle Assessment: Framework and Application to an Institutional Building." International Journal of Life Cycle Assessment 18 (3): 538-552.

Collingridge, David. 1980. The social control of technology. London: Pinter.

Cucurachi S, Heijungs R (2014) Characterisation factors for life cycle impact assessment of sound emissions Science of The Total Environment 468–469:280-291 doi:http://dx.doi.org/10.1016/j.scitotenv.2013.07.080

Curran, M. A. (2004). "The status of life-cycle assessment as an environmental management tool." Environmental Progress 23(4): 277-283.

Dale, Alexander T., Andre F. de Lucena, Joe Marriott, Bruno S.M.C. Borba, Roberto Schaeffer, and Melissa Bilec. 2013. "Modeling Future Life-Cycle Environmental Impacts of Electricity Supplies in Brazil." Energies 6: 3182-3208.

Dale AL, Lowry GV, Casman EA (2015) Stream Dynamics and Chemical Transformations Control the Environmental Fate of Silver and Zinc Oxide Nanoparticles in a Watershed-Scale Model Environmental Science & Technology 49:7285-7293 doi:10.1021/acs.est.5b01205

Dang, X.; Yi, H.; Ham, M.-H.; Qi, J.; Yun, D. S.; Ladewski, R.; Strano, M. S.; Hammond, P. T.; Belcher, A. M. Virus-templated self-assembled single-walled carbon nanotubes for highly efficient electron collection in photovoltaic devices. Nat Nano 2011, 6 (6), 377-384.

Davies, J. Clarence. 2009. "Oversight of Next Generation Nanotechnology." Washington, DC: Woodrow Wilson International Center for Scholars.

Dawson DA, Wilke TS (1991) Evaluation of the frog embryo teratogenesis assay: Xenopus (FETAX) as a model system for mixture toxicity hazard assessment Environmental Toxicology and Chemistry 10:941-948 doi:10.1002/etc.5620100710

DOE, Department of Energy (2012). SunShot Vision Study.

Eason, T., Meyer, D.E., Curran, M.A., and Upadhyayula, V.K.K., Guidance to Facilitate Decisions for Sustainable Nanotechnology, 2011, U.S. Environmental Protection Agency: National Risk Management Research Laboratory.

EC, European Commission. (2013). Horizon 2020: Science with and for Society. The EU Framework Programme for Research and Innovation; Available from: http://ec.europa.eu/programmes/horizon2020/en/h2020-section/science-and-society.

EC, European Comission. (2011) International reference life cycle data system (ILCD) handbook - Recomendations for life cycle impact assessment in the European context. First edition edn. Office of the European Union, Luxemburg

Eckelman, M. J.; Mauter, M. S.; Isaacs, J. A.; Elimelech, M. New Perspectives on Nanomaterial Aquatic Ecotoxicity: Production Impacts Exceed Direct Exposure Impacts for Carbon Nanotoubes. Environ. Sci.Technol. 2012, 46 (5), 2902-2910.

Eisenberg DG, K; Hristozov, D; Bates, M and Linkov, I (2015) Risk Assessment, Life Cycle Assessment, and Decision Methods for Nanomaterials. In: Satinder Kaur Brar TCZ, Mausam Verma, Rao Y. Surampalli, and Rajeshwar D. Tyagi (ed) Nanomaterials in the Environment. American Society of Civil Engineers, Reston, Virgina, pp 383-419. doi:doi:10.1061/9780784414088.ch15

EPA (2012). Lithium-ion Batteries and Nanotechnology for Electric Vehicles: A Life Cycle Assessment.

Fagerland MW, Sandvik L (2009) Performance of five two-sample location tests for skewed distributions with unequal variances Contemporary Clinical Trials 30:490-496 doi:http://dx.doi.org/10.1016/j.cct.2009.06.007

Fantke P, Gillespie BW, Juraske R, Jolliet O (2014) Estimating Half-Lives for Pesticide Dissipation from Plants Environmental Science & Technology 48:8588-8602 doi:10.1021/es500434p

Fantke P, Wieland P, Juraske R, Shaddick G, Itoiz ES, Friedrich R, Jolliet O (2012) Parameterization Models for Pesticide Exposure via Crop Consumption Environmental Science & Technology 46:12864-12872 doi:10.1021/es301509u

Fisher, Erik, and Arie Rip. 2013. "Responsible Innovation: Multi-Level Dynamics and Soft Intervention Practices." In Responsible Innovation: Managing the Responsible Emergence of Science and Innovation in Society, edited by Richard Owen, John Bessant, and Maggy Heintz, 165-83. West Sussex, UK: John Wiley & Sons, Ltd.

Foley, Rider W., and Arnim Wiek. 2013. "Patterns of Nanotechnology Innovation and Governance within a Metropolitan Area." Technology in Society 35 (4): 233-47.

Fthenakis, V.; Held, M.; Kim, H.; Raugei, M. Update of energy payback times and environmental impacts of photovoltaics. Presented at the 24th European Photovoltaic Solar Energy Conference and Exhibition, Hamburg, DE; 2009.

Garner, K. and A. Keller (2014). "Emerging patterns for engineered nanomaterials in the environment: a review of fate and toxicity studies." *Journal of Nanoparticle Research* 16(8): 1-28.

Gauthier TD (2001) Detecting Trends Using Spearman's Rank Correlation Coefficient Environmental Forensics 2:359-362

Gavankar, S., S. Suh, and A.A. Keller, The Role of Scale and Technology Maturity in Life Cycle Assessment of Emerging Technologies. Journal of Industrial Ecology, 2014: p. n/a-n/a.

Gavankar, S.; Suh, S.; Keller, A. Life cycle assessment at nanoscale: review and recommendations. Int. J.Life Cycle Assess. 2012, 17 (3), 295-303.

Gelca R, Anderson TA, Cox SB (2012) The Effect of Cluster Size on the Breakdown of C_{60} Water Suspensions Into Toxic Compounds Advanced Science, Engineering and Medicine 4:205-210 doi:10.1166/asem.2012.1181

Gilbertson L, Wender B, Zimmerman J, Eckelman MJ (2015) Coordinating Modeling and Experimental Research of Engineered Nanomaterials to Improve Life Cycle Assessment Studies Environmental Science: Nano doi:10.1039/c5en00097a

Goedkoop, M.; Heijungs, R.; Huijbregts, M.; De Schryver, A.; Struijs, J.; van Zelm, R., ReCiPe 2008. A life cycle impact assessment method which comprises harmonised category indicators at the midpoint and the endpoint level 2009, 1.

Golsteijn, L.; Hendriks, H. W. M.; van Zelm, R.; Ragas, A. M. J.; Huijbregts, M. A. J. Do interspecies correlation estimations increase the reliability of toxicity estimates for wildlife? Ecotoxicology and Environmental Safety 2012, 80 (0), 238-243.

Green, M. A.; Emery, K.; Bücher, K.; King, D. L.; Igari, S. Solar cell efficiency tables (version 11). Prog. Photovoltaics 1998, 6 (1), 35-42.

Green, M. A.; Emery, K.; Bücher, K.; King, D. L.; Igari, S. Solar cell efficiency tables (version 13). Prog. Photovoltaics 1999, 7 (1), 31-37.

Green, M. A.; Emery, K.; Hishikawa, Y.; Warta, W. (2008). "Solar cell efficiency tables (Version 31)." *Progress in Photovoltaics*, 16(1): 61-67.

Green, M. A.; Emery, K.; King, D. L.; Igari, S. (2000). "Solar cell efficiency tables (version 15)". *Progress in Photovoltaics*, 8(1): 187-195.

Green, M. A.; Emery, K.; King, D. L.; Igari, S.; Warta, W. (2004). "Solar cell efficiency tables (version 24)." *Progress in Photovoltaics*, 12(5): 365-372.

Grieger K, Baun A, Owen R (2010) Redefining risk research priorities for nanomaterials Journal of Nanoparticle Research 12:383-392 doi:10.1007/s11051-009-9829-1

Grubb, G. F.; Bakshi, B. R. (2011). "Appreciating the Role of Thermodynamics in LCA Improvement Analysis via an Application to Titanium Dioxide Nanoparticles." *Environmental Science & Technology*, 45 (7): 3054-3061.

Guinee, Jeroen B., Reinout Heijungs, Gjalt Huppes, Alessandra Zamagni, Paolo Masoni, Roberto Buonamici, Tomas Ekvall, and Tomas Rydberg. (2011). “Life Cycle Assessment: Past, Present, and Future.” *Environmental Science & Technology*, 45 (1): 90-96.

Gust KA, Collier ZA, Mayo M, Stanley JK, Gong P, Chappell M (2015) Limitations of toxicity characterization in life cycle assessment–can adverse outcome pathways provide a new foundation? Integrated Environmental Assessment and Management

Guston, D. (2013). “Understanding Anticipatory Governance”. *Social Studies of Science*, DOI: 0306312713508669.

Guston, D.H., et al. (2014). “Responsible innovation: motivations for a new journal.” *Journal of Responsible Innovation*, 1(1): p. 1-8.

Gutowski, T.G., J.Y.H. Liow, and D.P. Sekulic. Minimum exergy requirements for the manufacturing of carbon nanotubes. in Sustainable Systems and Technology (ISSST), 2010 IEEE International Symposium on. 2010.

Gutowski, Timothy G., Matthew S. Branham, Jeffrey B. Dahmus, Alissa J. Jones, Alexandre Thiriez, and Dusan P. Sekulic. (2009). "Thermodynamic Analysis of Resources Used in Manufacturing Processes." *Environmental Science & Technology* 43(5): 1584-90.

Hauschild, M. Z., M. Huijbregts, et al. (2008). "Building a Model Based on Scientific Consensus for Life Cycle Impact Assessment of Chemicals: The Search for Harmony and Parsimony." *Environmental Science & Technology*, 42(19): 7032-7037.

Hauschild M et al. (2013) Identifying best existing practice for characterization modeling in life cycle impact assessment The International Journal of Life Cycle Assessment 18:683-697 doi:10.1007/s11367-012-0489-5

Hauschild MZ, Huijbregts MA (2015) Life Cycle Impact Assessment LCA Compendium–The Complete World of Life Cycle Assessment

Heijungs, R.; Huijbregts, M. A. (2004). “A review of approaches to treat uncertainty in LCA,” iEMSs 2004 International Congress: Complexity and Integrated Resources Management". International Environmental Modelling and Software Society, Osnabrueck, Germany, 2004.

Henderson, A.; Hauschild, M.; Meent, D.; Huijbregts, M. J.; Larsen, H.; Margni, M.; McKone, T.; Payet, J.; Rosenbaum, R.; Jolliet, O. (2011). “USEtox fate and ecotoxicity factors for comparative assessment of toxic emissions in life cycle analysis: sensitivity to key chemical properties.” *International Journal of Life Cycle Assessment*, 16(8): 701-709.

Hendren CO, Lowry GV, Unrine JM, Wiesner MR (2015) A functional assay-based strategy for nanomaterial risk forecasting Science of The Total Environment 536:1029-1037 doi:http://dx.doi.org/10.1016/j.scitotenv.2015.06.100

Henry TB, Petersen EJ, Compton RN (2011) Aqueous fullerene aggregates (nC60) generate minimal reactive oxygen species and are of low toxicity in fish: a revision of previous reports Current Opinion in Biotechnology 22:533-537 doi:http://dx.doi.org/10.1016/j.copbio.2011.05.511

Hertwich, E. G.; Hammitt, J. K.; Pease, W. S. (2000). “A Theoretical Foundation for Life-Cycle Assessment.” *Journal of Industrial Ecology*, 4(1): 13-28.

Herwich, Edgar. (2005). “Life Cycle Approaches to Sustainable Consumption: A Critical Review” *Environmental Science & Technology*, 39(13): 4673-4684.

Hicks, A.; Theis, T. (2014). “An agent based approach to the potential for rebound resulting from evolution of residential lighting technologies.” *International Journal of Life Cycle Assessment*, 19(2): 370-376.

Hischier, R. and T. Walser (2012). "Life cycle assessment of engineered nanomaterials: State of the art and strategies to overcome existing gaps." *Science of The Total Environment*, 425(0): 271-282.

Huijbregts, M. A.; Hauschild, M.; Jolliet, O.; Margni, M.; McKone, T.; Rosenbaum, R.K.; van de Meent, D. USEtox User Manual. (2010a); http://www.usetox.org/sites/default/files/support-tutorials/user_manual_usetox.pdf

Huijbregts MAJ, Rombouts LJA, Ragas AMJ, van de Meent D (2005) Human-toxicological effect and damage factors of carcinogenic and noncarcinogenic chemicals for life cycle impact assessment Integrated Environmental Assessment and Management 1:181-244 doi:10.1897/2004-007R.1

Huijbregts MM, M; van de Meent, D; Jollier, O; Rosenbaum, RK; McKone, T and Hauschild, M. (2010b) USEtox chemcial-specific database: Organics.

Hunt, R., W. Franklin, and R. Hunt. (1996). “LCA — How it came about.” *The International Journal of Life Cycle Assessment*, 1(1): 4-7.

Hyung, H.; Fortner, J. D.; Hughes, J. B.; Kim, J.-H. (2006). “Natural Organic Matter Stabilizes Carbon Nanotubes in the Aqueous Phase.” *Environmental Science & Technol*ogy, 41 (1), 179-184.

ISO (2006). Environmental management - life cycle assessment - requirements and guidelines. Geneva, International Organization for Standards.

Jafvert CT, Kulkarni PP (2008) Buckminsterfullerene’s (C60) Octanol−Water Partition Coefficient (Kow) and Aqueous Solubility Environmental Science & Technology 42:5945-5950 doi:10.1021/es702809a

Ji K-h, Kim J-k, Choi K-h (2014) Sunlight Enhances Toxicity of Fullerene (C 60) to Freshwater Invertebrates Daphnia magna and Moina macrocopa The Korean Journal of Public Health 51:35-45

Jolliet O, Fantke P (2015) Human Toxicity. In: Life Cycle Impact Assessment. Springer, pp 75-96

Kiser, M. A., P. Westerhoff, et al. (2009). "Titanium Nanomaterial Removal and Release from Wastewater Treatment Plants." *Environmental Science & Technology*, 43(17): 6757-6763.

Klopffer, W., Curran, M.A., Frankl, P., Heijung, R., Kohler, A., and Olsen, S.I., (2007). “Nanotechnology and life cycle assessment: A systems approach to nanotechnology and the environment.” Woodrow Wilson International Center for Scholars.

Kolosnjaj J, Szwarc H, Moussa F (2007) Toxicity Studies of Fullerenes and Derivatives. In: Chan WW (ed) Bio-Applications of Nanoparticles, vol 620. Advances in Experimental Medicine and Biology. Springer New York, pp 168-180. doi:10.1007/978-0-387-76713-0_13

Kovochich M et al. (2009) Comparative Toxicity of C60 Aggregates toward Mammalian Cells: Role of Tetrahydrofuran (THF) Decomposition Environmental Science & Technology 43:6378-6384 doi:10.1021/es900990d

Kümmerer K, Menz J, Schubert T, Thielemans W (2011) Biodegradability of organic nanoparticles in the aqueous environment Chemosphere 82:1387-1392 doi:http://dx.doi.org/10.1016/j.chemosphere.2010.11.069

Lambert, J. H.; Farrington, M. W. (2006). “Risk-Based Objectives for the Allocation of Chemical, Biological, and Radiological Air Emissions Sensors.” *Risk Analysis*, 26 (6), 1659-1674.

Lens M (2009) Use of Fullerenes in Cosmetics Recent Patents on Biotechnology 3:118-123 doi:10.2174/187220809788700166

Li D, Fortner JD, Johnson DR, Chen C, Li Q, Alvarez PJJ (2010) Bioaccumulation of 14C60 by the Earthworm Eisenia fetida Environmental Science & Technology 44:9170-9175 doi:10.1021/es1024405

Liebowitz, S. J. and S. E. Margolis (1995). "Path dependence, lock-in, and history." *Journal of Law, Economics, & Organization*: 205-226.

Linkov, I.; Seager, T. P. (2011) Coupling Multi-Criteria Decision Analysis, Life-Cycle Assessment, and Risk Assessment for Emerging Threats. *Environmental Science & Technology*, 45 (12), 5068-5074.

Linkov, Igor, Matthew E. Bates, Laure J. Canis, Thomas P. Seager, and Jeffrey M. Keisler. (2011). "A Decision-Directed Approach for Prioritizing Research into the Impact of Nanomaterials on the Environment and Human Health." *Nature Nanotechnology*, 6 (12): 784-87.

Lloyd, S. M.; Ries, R. (2007). "Characterizing, Propagating, and Analyzing Uncertainty in Life-Cycle Assessment: A Survey of Quantitative Approaches." *Journal of Industrial Ecology*, 11 (1), 161-179.

Lovern SB, Klaper R (2006) Daphnia magna mortality when exposed to titanium dioxide and fullerene (C60) nanoparticles Environmental Toxicology and Chemistry 25:1132-1137

Maxwell, D.; Owen, P.; McAndrew, L.; Muehmel, K.; Neubauer, A. Addressing the Rebound Effect, a report for the European Commission DG Environment; 2011

McKone, T.; Nazaroff, W.; Berck, P.; Auffhammer, M.; Lipman, T.; Torn, M.; Masanet, E.; Lobscheid, A.; Santero, N.; Mishra, U. (2011). Grand challenges for life-cycle assessment of biofuels. *Environmental Science & Technology*, 45 (5), 1751-1756.

Miller, S. A. and G. A. Keoleian (2015). "A Framework for Analyzing Transformative Technologies in Life Cycle Assessment." *Environmental Science & Technology*, just accepted.

Miller, Shelie A., Amy E. Landis, and Thomas L. Theis. 2006. "Use of Monte Carlo Analysis to Characterize Nitrogen Fluxes in Agroecosystems." *Environmental Science & Technology*, 40 (7): 2324-32.

Miseljic, M. and S. Olsen (2014). "Life-cycle assessment of engineered nanomaterials: a literature review of assessment status." *Journal of Nanoparticle Research*, 16(6): 1-33.

NNI, Environmental, Health, and Safety Research Strategy, 2011: National Nanotechnology Initiative.

NRC, National Research Council. (2007). “Models in Environmental Regulatory Decision Making”. Washington, DC: The National Academies Press.

NRC, National Research Council. “Science and Decisions”. (2009). Washington, DC: The National Academies Press.

NRC, National Research Council. (2012). A Research Strategy for Environmental, Health, and Safety Aspects of Engineered Nanomaterials, The National Academies Press.

NSTCCT. (2014) Progress review on the coordinated implementation of the national nanotechnology initiative 2011 environmental, health, and safety research strategy. Washington, DC

Oberdörster, G., E. Oberdörster, et al. (2005). "Nanotoxicology: An Emerging Discipline Evolving from Studies of Ultrafine Particles." *Environmental Health Perspectives*, 113(7): 823-839.

Oberdörster, G., V. Stone, et al. (2007). "Toxicology of nanoparticles: A historical perspective." *Nanotoxicology*, 1(1): 2-25.

Owen, Richard, and Nicola Goldberg. (2010). "Responsible Innovation: A Pilot Study with the U.K. Engineering and Physical Sciences Research Council." *Risk Analysis*, 30 (11): 1699-707.

Owen, Richard, David Baxter, Trevor Maynard, and Michael Depledge. (2009). "Beyond Regulation: Risk Pricing and Responsible Innovation." *Environmental Science & Technology* 43(18): 6902-06.

PEN (2013) Consumer Products Inventory. Project on Emerging Nanotechnologies. http://www.nanotechproject.org/cpi. Accessed Spetember 2015

Pennington DW, Margni M, Ammann C, Jolliet O (2005) Multimedia Fate and Human Intake Modeling: Spatial versus Nonspatial Insights for Chemical Emissions in Western Europe Environmental Science & Technology 39:1119-1128 doi:10.1021/es034598x

Pesonen, Hanna-Leena, Tomas Ekvall, Günter Fleischer, Gjalt Huppes, Christina Jahn, Zbigniews Klos, Gerald Rebitzer, et al. 2000. "Framework for Scenario Development in LCA." *The International Journal of Life Cycle Assessment*, 5(1): 21-30.

Petersen EJ et al. (2015) Adapting OECD Aquatic Toxicity Tests for Use with Manufactured Nanomaterials: Key Issues and Consensus Recommendations Environmental Science & Technology 49:9532-9547 doi:10.1021/acs.est.5b00997

Potts, H.; Anderson, J.; Colligan, L.; Leach, P.; Davis, S.; Berman, J. (2014). "Assessing the validity of prospective hazard analysis methods: a comparison of two techniques. *BMC Health Services Research*, 14 (1), 41.

Pourzahedi, L. and M. J. Eckelman (2014). "Environmental Life Cycle Assessment of Nanosilver-Enabled Bandages." *Environmental Science & Technology* 49(1): 361-368.

Prado-Lopez, Valentina, Thomas P. Seager, Mikhail Chester, Lise Laurin, Melissa Bernardo, and Steven Tylock. 2014. "Stochastic multi-attribute analysis (SMAA) as an interpretation method for comparative life-cycle assessment (LCA)" *The International Journal of Life Cycle Assessment* 19: 405-416.

PubChem (2015a) Compound Summary for Buckminsterfullerene. National Center for Biotechnology Information. https://pubchem.ncbi.nlm.nih.gov/compound/123591. Accessed October 27 2015

PubChem (2015b) Compound Summary for Nicotinamide. National Center for Biotechnology Information. https://pubchem.ncbi.nlm.nih.gov/compound/936. Accessed September 9 2015

Pycke BF, Chao T-C, Herckes P, Westerhoff P, Halden RU (2012) Beyond nC60: strategies for identification of transformation products of fullerene oxidation in aquatic and biological samples Analytical and bioanalytical chemistry 404:2583-2595

RIVM (2015) e-ToxBase National Institute for Public Health and the Environment. http://www.ru.nl/environmentalscience/research/themes-0/risk-assessment/e-toxbase/.

Robinson, Douglas K. R. (2009). "Co-Evolutionary Scenarios: An Application to Prospecting Futures of the Responsible Development of Nanotechnology." *Technological Forecasting and Social Change*, 76(9): 1222-39.

Rodriguez-Garcia, G., B. Zimmermann, et al. (2014). "Nanotoxicity and Life Cycle Assessment: First attempt towards the determination of characterization factors for carbon nanotubes." *IOP Conference Series: Materials Science and Engineering* 64(1): 012029.

Rosenbaum, R., et al., (2008). “USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment.” *The International Journal of Life Cycle Assessment*, 2008. 13(7): p. 532-546.

Rosenbaum RK (2015) Ecotoxicity. In: Life Cycle Impact Assessment. Springer, pp 139-162

Saleh NB, Aich N, Plazas-Tuttle J, Lead JR, Lowry GV (2015) Research strategy to determine when novel nanohybrids pose unique environmental risks Environmental Science: Nano 2:11-18 doi:10.1039/c4en00104d

Salieri, B., S. Righi, et al. (2015). "Freshwater ecotoxicity characterisation factor for metal oxide nanoparticles: A case study on titanium dioxide nanoparticle." *Science of The Total Environment*, 505(0): 494-502.

Saltelli A et al. (2008) Global sensitivity analysis: the primer. John Wiley & Sons,

Schomberg, R. (2012). “Prospects for technology assessment in a framework of responsible research and innovation.” Technikfolgen abschätzen lehren. M. Dusseldorp and R. Beecroft, VS Verlag für Sozialwissenschaften: 39-61.

Scown, C. D.; Nazaroff, W. W.; Mishra, U.; Strogen, B.; Lobscheid, A. B.; Masanet, E.; Santero, N. J.; Horvath, A.; McKone, T. E. (2012). “Lifecycle greenhouse gas implications of US national scenarios for cellulosic ethanol production.” *Environmental Research Letters*, 7 (1), 014011.

Seager, T. and I. Linkov (2009). "Uncertainty in life cycle assessment of nanomaterials." Nanomaterials: Risks and Benefits: 423-436.

Seki MF, S; Gondo, Y; Inoue, Y; Nozaka, T and Takatsuki, M (2008) Acute toxicity of fullerene C60 in aquatic organisms Environmental Science (Japan) 21:53-62

Shockley, W.; Queisser, H. J. Detailed balance limit of efficiency of p-n junction solar cells. J. of applied physics 1961, 32 (3), 510-519.

Siemer, J.; Knoll, B. Still more than enough. Photon International 2013, February 2013, 73.

Simon, Balint and Marcel Weil. 2013. “Analysis of Materials and Energy Flows of Different Lithium Ion Traction Batteries.” Revue de Métallurgie 110(S): 65-76

Sims, R. E. H.; Schock, R.N.; Adegbululgbe, A.; Fenhann, J.; Konstantinaviciute, I.; Moomaw, W.; Nimir, H.B.; Schlamadinger, B.; Torres-Martínez, J.; Uchiyama, C. T., Y.; Vuori, S.J.V.; Wamukonya, N.; Zhang, X. Energy supply. In Intergovernmental Panel on Climate Change: Cambridge, UK and New York, USA, 2007.

Sonnemann GW, Schuhmacher M, Castells F (2003) Uncertainty assessment by a Monte Carlo simulation in a life cycle inventory of electricity produced by a waste incinerator Journal of Cleaner Production 11:279-292 doi:http://dx.doi.org/10.1016/S0959-6526(02)00028-8

Spielmann, Michael, Roland Scholz, Olaf Tietje, and Peter de Haan. 2004. "Scenario Modeling in Prospective Lca of Transport Systems. Application of Formative Scenario Analysis." The International Journal of Life Cycle Assessment 10(5): 325-35.

Stilgoe, Jack, Richard Owen, and Phil Macnaghten. 2013. "Developing a Framework for Responsible Innovation." Research Policy 42 (9): 1568-80.

T.R. Dwarakanath; Wender, B.A.; Seager, T.P.; Fraser, M.P. Towards anticipatory lifie cycle assessment of photovoltaics, Proceedings of the 39th IEEE Photovoltaics Specialist Conference, Tampa Fl, Tampa Fl, 2013.

Taebi, Benham, Aad Correlje, Edwin Cuppen, Marloes Dignum, and Udo Pesch. 2014. “Responsible Innovation as an Endorsement of Public Values: The Need for Interdisciplinary Research.” Journal of Responsible Innovation DOI: 10.1080/23299460.2014.882072

Thabrew, Lanka, Arnim Wiek, and Robert Ries. 2009. “Environmental Decision Making in Multi-stakeholder Contexts: Applicability of Life Cycle Thinking in Development Planning and Implementation.” Journal of Cleaner Production 17(1): 67-76.

Theis, T. L., B. R. Bakshi, et al. (2011). "A life cycle framework for the investigation of environmentally benign nanoparticles and products." physica status solidi (RRL) – Rapid Research Letters 5(9): 312-317.

Tiwari AJ, Morris JR, Vejerano EP, Hochella MF, Marr LC (2014) Oxidation of C60 Aerosols by Atmospherically Relevant Levels of O3 Environmental Science & Technology 48:2706-2714 doi:10.1021/es4045693

UNEP (2002) OECD SIDS Initial Assessment Report - nicotinamide.

UNEP, United Nations Environmental Programme. 2013. “The Methodological Sheets for Subcategories in Social Life Cycle Assessment (S-LCA).”

US Department of Commerce. *US Carbon Emissions and Intensities Over Time: A Detailed Accounting of Industries, Government and Households*; US Department of Commerce: Economics and Statistics Administration: 2010.

Usenko CY, Harper SL, Tanguay RL (2007) In vivo evaluation of carbon fullerene toxicity using embryonic zebrafish Carbon 45:1891-1898 doi:http://dx.doi.org/10.1016/j.carbon.2007.04.021

Usenko CY, Harper SL, Tanguay RL (2008) Fullerene C60 exposure elicits an oxidative stress response in embryonic zebrafish Toxicology and Applied Pharmacology 229:44-55 doi:http://dx.doi.org/10.1016/j.taap.2007.12.030

USEPA (2010) Interim Technical Guidance for Assessing Screening Level Environmental Fate and Transport of, and General Population, Consumer, and Environmental Exposure to Nanomaterials. United States Environmental Protection Agency.

USEPA (2015a) ECOTox User Guide: ECOTOX Database System. Version 4.0 http:/www.epa.gov/ecotox.

USEPA (2015b) Estimation Programs Interface Suite™ for Microsoft® Windows, v 4.11. United State Enivronmental Protection Agency, Washington, DC, USA

van Zelm R, Huijbregts MA, Harbers JV, Wintersen A, Struijs J, Posthuma L, Van de Meent D (2007) Uncertainty in msPAF-based ecotoxicological effect factors for freshwater ecosystems in life cycle impact assessment Integrated Environmental Assessment and Management 3:e6-e37

van Zelm R, Huijbregts MAJ (2013) Quantifying the Trade-off between Parameter and Model Structure Uncertainty in Life Cycle Impact Assessment Environmental Science & Technology 47:9274-9280 doi:10.1021/es305107s

van Zelm R, Huijbregts MJ, van de Meent D (2009) USES-LCA 2.0—a global nested multi-media fate, exposure, and effects model The International Journal of Life Cycle Assessment 14:282-284 doi:10.1007/s11367-009-0066-8

von Schomberg, R. A Vision of Responsible Research and Innovation. In R. Owen, J. Bessant, M. Heints (Eds) Responsible Innovation: Managing the Responsible Emerginged of Science and Innovation in Sociery, Wiley, London 2013, 51-74.

Walser, T.; Demou, E.; Lang, D. J.; Hellweg, S. Prospective environmental life cycle assessment of nanosilver T-shirts. Environ. Sci. Technol. 2011, 45 (10), 4570-4578.

Weidema, B. P. Market information in life cycle assessment. Miljøstyrelsen: 2003; Vol. 863.

Weidema BPB, Ch.; Hischier, R.; Mutel, Ch.; Nemecek, T.; Reinhard, J.; Vadenbo, C.O.; Wernet, G (2013) The ecoinvent database: Overview and methodology, Data quality guideline for the ecoinvent database version 3.

Wender, B. A.; Seager, T.P. Anticipatory life cycle assessment of SWCNT-enabled lithium ion batteries. In Nanotechnology for Sustainable Manufacturing, Rickerby, D., Ed. Maralte BV: Lieden, NL., 2014. ISBN: ISBN 9781482214826.

Wender, B., R. Foley, et al. (2012). "Anticipatory Governance and Anticipatory Life Cycle Assessment of Single Wall Carbon Nanotube Anode Lithium Ion Batteries." Nanotech. L. & Bus. 9: 201.

Wender, Ben and Thomas P. Seager. 2011. “Towards Prospective Life Cycle Assessment: Single Wall Carbon Nanotubes for Lithium-ion Batteries.” International Symposium on Sustainable Systems and Technology, Chicago, IL 16-18 May.

Westh T, Hauschild M, Birkved M, Jørgensen M, Rosenbaum R, Fantke P (2015) The USEtox story: a survey of model developer visions and user requirements The International Journal of Life Cycle Assessment 20:299-310 doi:10.1007/s11367-014-0829-8

Wiesner, M. R., G. V. Lowry, et al. (2006). "Assessing the Risks of Manufactured Nanomaterials." Environmental Science & Technology 40(14): 4336-4345.

Wiesner, M. R.; Lowry, G. V.; Jones, K. L.; Hochella, J. M. F.; Di Giulio, R. T.; Casman, E.; Bernhardt, E. S. Decreasing Uncertainties in Assessing Environmental Exposure, Risk, and Ecological Implications of Nanomaterials. Environ. Sci. Technol. 2009, 43 (17), 6458-6462.

Yang Y, Wang Y, Hristovski K, Westerhoff P (2015) Simultaneous removal of nanosilver and fullerene in sequencing batch reactors for biological wastewater treatment Chemosphere 125:115-121 doi:http://dx.doi.org/10.1016/j.chemosphere.2014.12.003

Zalk, D., S. Paik, et al. (2009). "Evaluating the Control Banding Nanotool: a qualitative risk assessment method for controlling nanoparticle exposures." Journal of Nanoparticle Research 11(7): 1685-1704.

Zhu S, Oberdörster E, Haasch ML (2006) Toxicity of an engineered nanoparticle (fullerene, C60) in two aquatic species, Daphnia and fathead minnow Marine Environmental Research 62, Supplement 1:S5-S9 doi:http://dx.doi.org/10.1016/j.marenvres.2006.04.059

Zhu X, Zhu L, Li Y, Duan Z, Chen W, Alvarez PJ (2007) Developmental toxicity in zebrafish (Danio rerio) embryos after exposure to manufactured nanomaterials: buckminsterfullerene aggregates (nC60) and fullerol Environmental Toxicology and Chemistry 26:976-979

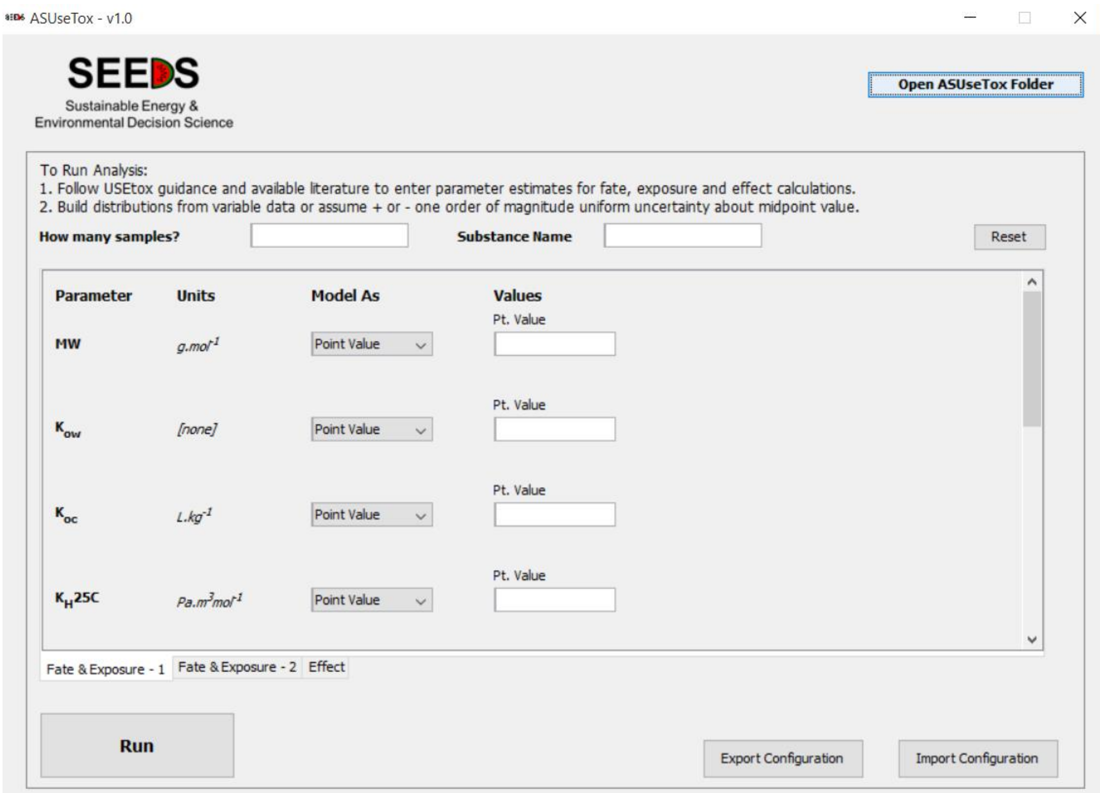
Zimmermann, Benedikt, Hanna Dura, Manuel Baumann, and Marcel Weil. 2013. "Towards prospective time-resolved LCA of fully electric supercap-vehicles in Germany" 19th SETAC LCA Case Study Symposium. Rome, Italy, December 11.

Zuin, S.; Micheletti, C.; Critto, A.; Pojana, G.; Johnston, H.; Stone, V.; Tran, L.; Marcomini, A., Weight of Evidence approach for the relative hazard ranking of nanomaterials. Nanotoxicology 2011, 5 (3), 445-458.

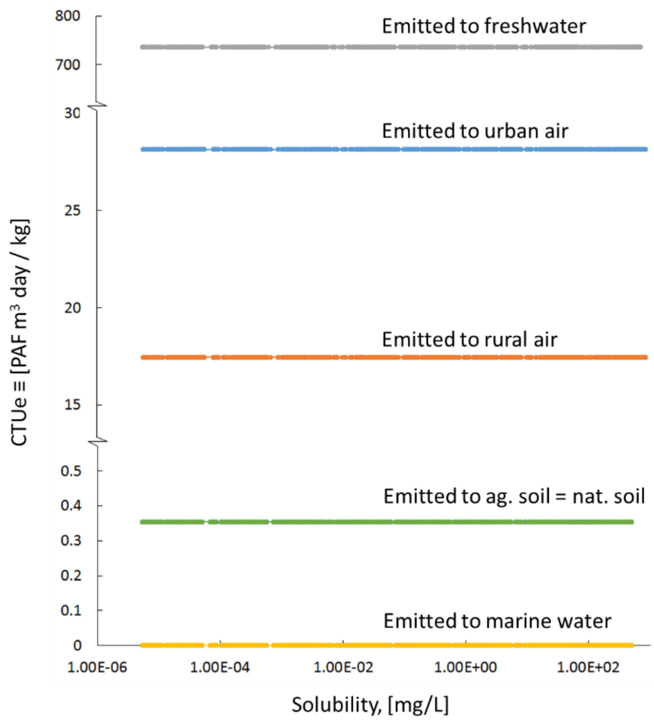
APPENDIX A

SUPPORTING INFORMATION FOR CHAPTER 4

Supporting Information 2.1 – Screenshots of the program interface for user specification of data as any combination of uniform, triangular, normal, or log normal distributions (A) and presentation of results as frequency distributions and column statistics (B). All randomly generated data and results are stored in an accessible spreadsheet allowing further statistical analysis.



Supporting information 2.2.1 – Local sensitivity of C60 aquatic ecotoxicity CFs to changes in solubility only shows that uncertainty in solubility – estimated as uniform between 10e-6 and 10e2 – has no effect on CF values for any emission scenario. Point values are assumed for all other parameters following Table 1 in the main text. Instances in which experimental and computational values are available in literature, we apply experimental values.

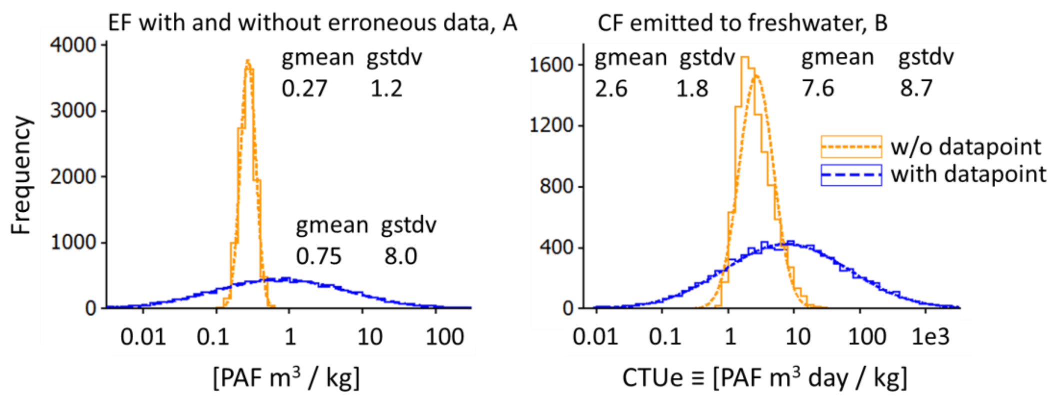


Supporting information 2.2.2 – Substance data for niacinamide (98-92-0) as implemented in new release of USEtox 2.0 are near identical to those used in this paper. The two notable exceptions are organic carbon water partitioning coefficient (Koc) which we apply from EPISuite's KOWIN sub routine and aveLog EC50 where we omit the erroneous data point from available ecotoxicity databases (discussed in main text at end of section 2.3 and supporting information 2.3.1).

Parameter	Units	Midpoint of value(s) used in this work	Value reported in USEtox 2.0
MW	g/mol	122	122.13
Kow	[none]	0.46	0.427
Koc	L/kg	8.5	none
Kh	Pa m^3/mol	2.9e-7 6.45e-6	6.45e-6
Pvap	Pa	0.026 0.05	0.0264
Solubility	mg/L	5e5 6.9-10e5	5e5
Kdeg, air	1/s	1.8e-6	1.75e-6
Kdeg, water		2.1e-7	2.14e-7
Kdeg, soil		1e-7	1.07e-7
Kdeg, sed		2.3e-8	2.38e-8
BAF fish	L/kg	0.9	0.901
aveLogEC50	log mg/L	3.2, SEM 0.04	-0.77, SEM N/R

Supporting information 2.3.1 – Including the misclassified ecotoxicity study reporting a 0.17 [mg/L] EC_{50} in the EPA ECOTox and RIVM ETox databases reduces aveLog EC50 from 3.27 to 2.8 log (mg/L) and increases the standard error on the mean (SEM) from 0.048 to 0.423. These differences correspond to EFs (A) with a geometric mean of 0.75 (with) and 0.27 (baseline) and aquatic ecotoxicity CFs (B) with a geometric mean of 7.6 (with) and 2.6 (baseline) and geometric standard deviations of 8.7 and 1.8 respectively.

Reported data	Acute to chronic converstion	Geometric mean of trophic level	Log of each geometric mean	Arithmetic mean of log values
Producers	*(Xi) n=2-3**		*(Ai) N=3*	(aveLog EC50)
1000	500	1494.48988	3.174492978	
8934	4467			
Consumers				
1000	500	2028.299781	3.307132144	2.814785902
16456	8228			
Secondary consumers				
1000	500	91.77673097	1.962732584	
18189	9094.5			
0.34***	0.17			
***Erroneous datapoint from ecotoxicity databases			Standard dev.	Strd. Error on mean
*n=3 for secondary consumers if erroneous data point excluded			0.740874098	0.42774386



Supporting information 2.3.2 – Following USEtox guidance aveLog EC50 is calculated by taking arithmetic mean of the geometric means (Ai) from variable EC_{50} data in chronic equivalents (Xi), as indicated in representative table in SI 2.3.1.

$$A_i = log \ \left(\prod_{i=1}^{n} X_i\right)^{1/n} \qquad \text{Equation 1}$$

And then calculate aveLog EC50 as the arithmetic mean of these values:

$$aveLog\ EC50 = \frac{1}{N}\sum_{i=0}^{N} A_i \qquad \text{Equation 2}$$

We calculate the standard error on the mean as:

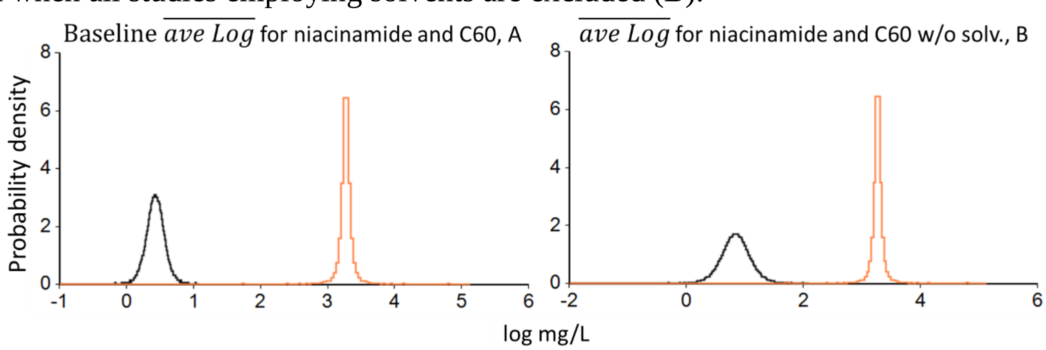
$$SEM = \frac{s}{\sqrt{n}} = \sqrt{\frac{\frac{1}{N-1}\sum_{i=0}^{N}(A_i-\bar{A})^2}{n}} \qquad \text{Equation 3}$$

Comparing niacinamide EFs (A) and CFs for emission to freshwater (B) with (blue) and without (orange) the erroneous data point show its inclusion increases the geometric mean (0.27 vs 0.75 and 2.6 vs 7.6 for EFs and CFs respectively) and significantly increases uncertainty (geometric standard deviations of 1.2 vs 8.0 and 1.8 vs 8.7 for EFs and CFs respectively).

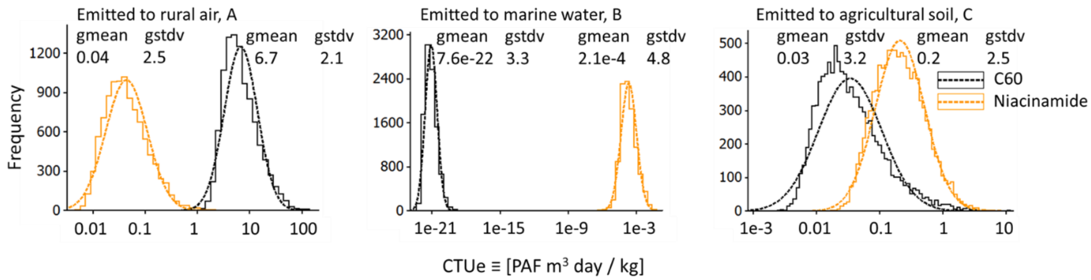
Uncertainty modeling in average toxicity indicator (aveLog EC50) is informed by Golsteijn, Hendriks et al. (2012) and Van Zelm, Huijbregts et al. (2007) with uncertainty in aveLog EC50 ($\overline{ave\ Log}$) modeled as:

$$\overline{ave\ Log} = ave\ Log\ EC50 + SEM * t$$

Where: ave Log EC50 is calculated according to USEtox guidance (Huijbregts 2010; Frantke 2015) from available literature and databases and t represents a two-tailed t-distribution with n-1 degrees of freedom from n different species with experimental toxicity data. As reported in Tables 3 and 4 in the main text, n=10 for C60 (n=9 for the no-solvent scenario, see Supporting information 3.4) and n=3 for niacinamide excluding *X. laevis*. The standard error on the mean (SEM) is calculated for each data set following Equation 2 in supporting information 2.3.1. Distributions of ($\overline{ave\ Log}$) for niacinamide (orange) and C60 (black) in the baseline scenario (A) show the relative lower toxicity and uncertainty of niacinamide despite the relatively few species for which data is available, even when all studies employing solvents are excluded (B).



Supporting information 3.1 – Comparison of freshwater ecotoxicity characterization factors for C_{60} and niacinamide emitted to rural air (A), marine water (B), and agricultural soil (C).



Supporting Information 3.2 – The statistical significance of the difference between stochastic aquatic ecotoxicity CFs of C_{60} and nicainamide are calculated following Welch's t test for distributions with unequal variance as:

$$t = \frac{\overline{X_1} - \overline{X_2}}{\sqrt{\frac{S_1^2}{n_1} + \frac{S_2^2}{n_2}}}$$

Where $\bar{X}$ is the distribution mean, $S^2$is the distribution variance, and n is the number of samples for distributions 1 and 2.

The degrees of freedom is given by:

$$v = \frac{\left(\frac{S_1^2}{n_1} + \frac{S_2^2}{n_2}\right)^2}{\frac{S_1^4}{n_1^2 * (N_1 - 1)} + \frac{S_2^4}{n_2^2 * (N_2 - 1)}}$$

All calculations are based on $n_1 = n_2 = 10{,}000$ simulations, with standard deviations reported in figure for parameter uncertainty and taken from Rosenbaum, Bachmann et al. (2008) for model uncertainty.

Supporting information 3.3 – Variability from uncertain parameters is smaller in magnitude than model uncertainty, which is quantified for emissions to rural air, freshwater, and agricultural soil with geometric standard deviations of 13.3, 4.2, and 10.2 respectively (Rosenbaum, Bachmann et al. 2008) (Figure 2A-C).

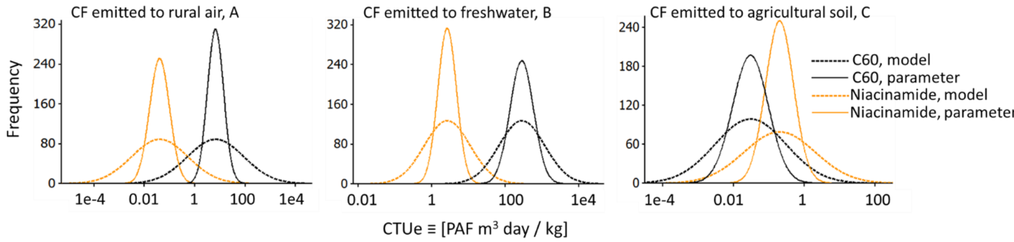
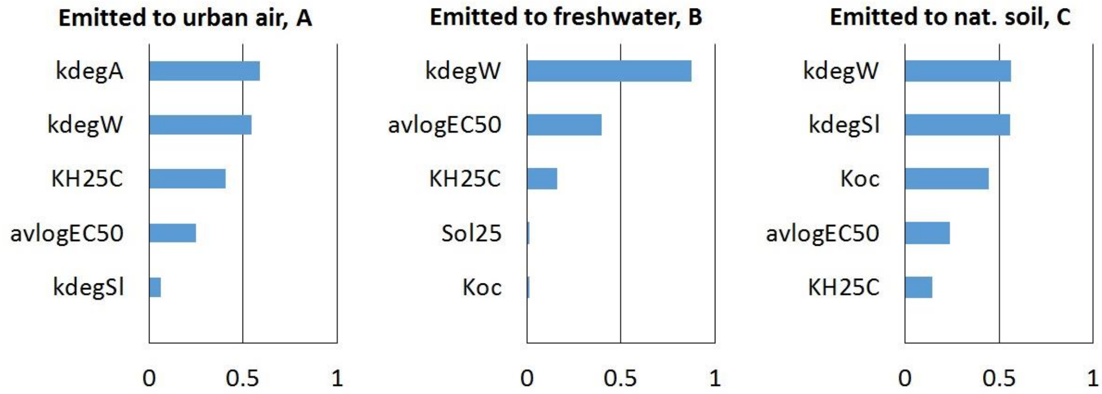


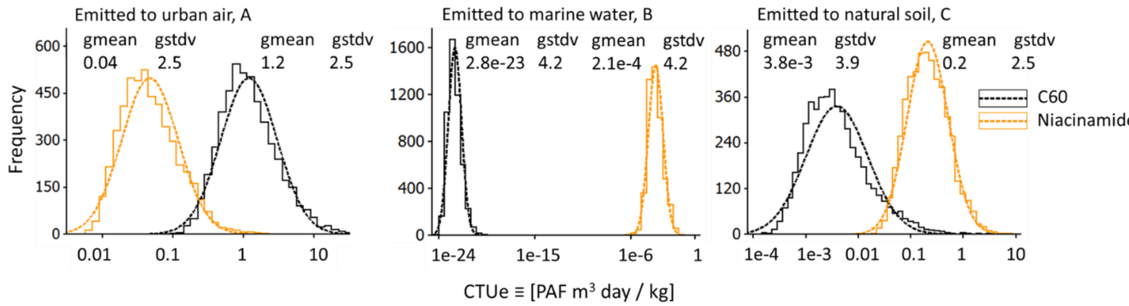
Figure 2. Comparison of parameter (solid) and model (dashed) uncertainty in freshwater aquatic ecotoxicity CFs for niacinamide (orange) and C_{60} (black) emissions to rural air (A), freshwater (B), and agricultural soil (C) compartments.

Nonetheless, the difference in CFs is significant with respect to model uncertainty (Welch's t test $p < 0.001$) for emissions to rural air and freshwater (2A&B), but not significant for emissions to soil compartments (2C) with a Welch's t test p-value > 0.2.

Supporting Information 3.3 – Spearman Rank Correlation Indices for all variable inputs used to calculate niacinamide aquatic ecotoxicity CFs for emissions to urban air (A), continental freshwater (B), and natural soil (C).



Supporting information 3.4 – Additional characterization factors for C60 prepared without solvent and niacinamide emitted to urban air (A), marine water (B), and natural soil (C). Emissions to rural air and agricultural soil are very similar to A and C respectively.



Frantke, P. (2015). USEtoc 2.0 User Manual (Version 2), UNEP-SETAC.
Golsteijn, L., H. W. M. Hendriks, et al. (2012). "Do interspecies correlation estimations increase the reliability of toxicity estimates for wildlife?" Ecotoxicology and Environmental Safety **80**(0): 238-243.
Huijbregts, M. A., Hauschild, M. Jolliet, O., Margni, M., McKone, T., Rosenbaum, R.K., and van de Meent, D. (2010). USEtox User Manual.
Rosenbaum, R., T. Bachmann, et al. (2008). "USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment." The International Journal of Life Cycle Assessment **13**(7): 532-546.
Van Zelm, R., M. A. Huijbregts, et al. (2007). "Uncertainty in msPAF-based ecotoxicological effect factors for freshwater ecosystems in life cycle impact assessment." Integrated Environmental Assessment and Management **3**(4): e6-e37.

APPENDIX B

STATEMENT OF COAUTHOR PERMISSIONS

All coauthors have given permission for each chapter to be published as chapters in Ben Wender's dissertation.