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Spatial Heterogeneity in Modeling Environmental and Human Health Impacts of Chemicals

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Spatial Heterogeneity in Modeling Environmental and Human Health Impacts of Chemicals

A dissertation submitted in partial satisfaction of the
requirements for the degree Doctor of Philosophy
in Environmental Science and Management

by

Mengya Tao

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December 2019

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October 2019

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Lastly, I would like to thank my three family communities here in the U.S. that always make me feel like home and supported all the time, my family at Bren, my family at Bellydance Land, and my family at Arthur Murray Santa Barbara Dance Studio.

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EDUCATION

Ph.D. in Environmental Science and Management **2019**

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Master of Art in Applied Statistics **2017**

Department of Statistics and Applied Probability

University of California, Santa Barbara

Master of Environmental Science and Management **2014**

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Bachelor of Science in Environmental Science **2012**

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PUBLICATIONS

1. **Tao, M.**; Li, D.; Song, R.; Suh, S.; Keller, AA (2018). "OrganoRelease–A framework for modeling the release of organic chemicals from the use and post-use of consumer products" *Environmental Pollution*, 234, 751-761.
2. Yang, Y.*; **Tao, M.***; Suh, S (2017). "Geographic variability of agriculture requires sector-specific uncertainty characterization" *The International Journal of Life Cycle Assessment*, 23 (8), 1581-1589. (* equal contribution)
3. Adeleye, AS., Oranu, EA., **Tao, M.**, Keller, AA. (2016). Release and detection of nanosized copper from a commercial antifouling paint. *Water research* 102, 374-382.
4. Aceves-Bueno, E., Adeleye, AS., Feraud, M., Huang Y., **Tao, M.**, Yang Y., Anderson, SE (2017). The accuracy of citizen science data: a quantitative review. *The Bulletin of the Ecological Society of America*, 98 (4), 278-290.

5. Feinberg L., Jylkka, Z., **Tao, M.**, White, B., Xu, Y., Kendall B (2014). "Evaluating Offshore Wind Energy Feasibility off the California Central Coast", Master Thesis.

AWARDS and Professional Activities

2018	UC-Wide 4th Grad Slam Tournament People's Choice, UC
2018	UCSB 6th Annual Grad Slam Champion, UCSB
2017	Mellichamp Sustainability Fellowship, UCSB
2015 – 2017	Decker's Outstanding Graduate Student Scholarship, UCSB
2018	Gordon Research Seminar on Industrial Ecology (Chair)
2017	Bren School PhD Student Symposium (Chair)

ACADEMIC RESEARCH

Graduate Student Researcher, Chemical Life Cycle Collaborative (CLiCC) Project – UCSB, Santa Barbara, CA (9/14–Present)

- Worked on an EPA-funded project of rapidly assessing chemical life cycle impacts, human health and ecological risks.
- Developed a comprehensive framework to estimate chemical releases to the environment.
- Designed and implemented hyperparameter optimization, a combination of grid search and random search methods, for an artificial neural network (ANN) model to classify chemical functional uses.
- Built a complex mathematical model to track and estimate organic chemical behavior, fate, and transport in the environment dynamically using ordinary differential equations.
- Developed a web-based tool and collaborated with 10+ software developers to design and implement 30+ RESTful APIs in Python.
- Trained and managed 14 graduate and undergraduate students on data collection and back-end APIs development.
- Collected and processed data from 15+ data sources and built a relational database using Python and MySQL.

- Designed and automated downloadable 3-page summary reports and 20-page detailed reports in Python.

Graduate Student Researcher, Dr. Sangwon Suh – UCSB, Santa Barbara, CA (9/15–Present)

Climate Change Impacts on Agricultural Pesticide Use (8/18–Present)

- Mapped the spatial disparity of U.S. farmers' pesticide use for corn production and its associated environmental and human health impacts at county-level.
- Building regression models to show the relationship between climate change-induced extreme weather and pesticide toxicity impacts to understand climate change impacts on pesticide use.

Life Cycle Assessment of Agriculture Production System (9/16–8/17)

- Compiled state-specific life cycle inventory data (e.g., pesticide, fertilizer, energy) for 4 major crops (corn, soybean, cotton, and wheat) in the U.S. and evaluated geographic variability in the characterized results.
- Evaluated the consequence of choosing average or region-specific LCIs in understanding the life cycle environmental implications of 9 impact categories (e.g., global warming, acidification).
- Analyzed the implications in characterizing the uncertainties associated with geographic variability.

Master's Wind Energy Group Project – UCSB, Santa Barbara, CA (3/13–6/14)
Evaluating Offshore Wind Energy Feasibility off the California Central Coast
Client: Infinity Wind Power (now ENGIE North America)

- Interviewed 10+ stakeholder groups and conducted an online general public survey with 450+ participants to evaluate stakeholder perceptions of offshore wind.
- Performed spatial analysis of offshore wind potential and mapped conflicting uses of marine environment.
- Identified federal, state, and local permitting pathways to the approval of offshore wind in California.

Lab Research Assistant, Keller's Lab on Nanomaterials – UCSB, Santa Barbara, CA (6/13–3/14)

- Assisted with lab research on the release and detection of nanosized copper from a commercial antifouling paint.
- Monitored release of different fractions of copper (dissolved, nano, and bulk).
- Characterized the copper releases into water for environmental fate and effects analysis.

TEACHING and MENTORING EXPERIENCE

Teaching Assistant – UCSB, Santa Barbara, CA (9/18–12/18)

- Taught biostatistics to 75 students in 3 sessions and remembered every student's name and performance.

Teaching Assistant – California Department of Toxic Substance Control (DTSC), Sacramento, CA (8/18)

- Alternative Analysis Workshop on Life Cycle Impacts and Exposure Assessment (2-day workshop)
- Designed and constructed teaching materials and hands-on exercises, and coordinated with DTSC staff for the workshop planning with 100+ participants.

Workshop Instructor – American Center for Life Cycle Assessment, Portsmouth, NH (10/17)

- Introduction to Chemical Life Cycle Collaborative (CLiCC Tool) (1-day workshop)
- Taught LCA and CLiCC tool to 20+ professionals attending ACLCA conference.

WORK EXPERIENCE

Life Cycle Assessment Researcher – VitalMetrics (IERS, LLC), Goleta, CA (3/13–9/14)

- Analyzed the life cycle environmental implications for the goods purchased by the U.S. General Services Administration (GSA) using input-output LCA.

- Identified hotspots to reduce environmental impacts through sustainable procurement strategies.
- Developed an Excel automation tool to implement LCA data analysis and visualization.

Research Intern – Oregon Department of Environmental Quality, Portland, OR (6/11–8/11)

- Revised mercury TMDL criteria in Willamette Basin, based on present criteria and historical data.
- Conducted data analysis and visualized bacteria load curves and demonstrated its causal relationship.

TALKS

1. **Tao, M.**, Li, D., Garner, K., Song R., Keller, A.A., Suh, S (2018). Rapid risk assessment of chemicals in consumer products. *Gordon Research Conference on Industrial Ecology*. Les Diablerets, Switzerland.
2. **Tao, M.**, Keller, A. A., & Suh, S (2017). Rapid spatially-explicit life cycle risk assessment of chemicals in consumer products: Case study of methylene chloride in paint strippers. *38th Annual Meeting: Society of Environmental Toxicology and Chemistry*, North America. Minneapolis, Minnesota, USA.
3. **Tao, M.**, Keller, A. A., & Suh, S (2017). Rapid, region-specific risk assessment of chemicals in consumer products - Case study of paint strippers. *American Center for Life Cycle Assessment XVII Conference*. Portsmouth New Hampshire, USA.
4. **Tao, M.**, Qin, Y., Garner, K., Song, R., Suh, S (2017). Chemical Life Cycle Collaborative: A New Web-Based Tool for Assessing Chemicals' Life Cycle Impacts. *American Center for Life Cycle Assessment XVII Pre-conference Workshop.* Portsmouth, New Hampshire, USA.
5. **Tao, M.**; Li, D.; Keller, A.A.; Suh, S (2016). Applying QSAR models in a holistic rapid assessment of chemical life cycle impact. *17th International Conference on QSAR in Environmental and Health Sciences*, Miami, Florida, USA.
6. **Tao, M.**, Li, D; Song, R; Suh, S; Keller, A.A (2016). Framework for modeling the release of chemicals during the use phase of consumer products. *37th Annual Meeting: Society of Environmental Toxicology and Chemistry*. Orlando, Florida, USA.
7. **Tao, M.**, Song, R., Li, D., Suh, S., Keller, A.A (2016). Framework for modeling the release of chemicals during the use phase of consumer products. *Gordon Research Seminar on Industrial Ecology*. Stowe, Vermont, USA.

8. **Tao, M.**, Song, R., Li, D., Suh, S., Keller, A.A (2016). Geographic variability of agriculture requires sector-specific uncertainty characterization. *Gordon Research Conference on Industrial Ecology*. Stowe, Vermont, USA.

ABSTRACT

Spatial Heterogeneity in Modeling Environmental and Human Health Impacts of Chemicals

by

Mengya Tao

The number of chemicals that humanity is using for producing goods and services is rapidly increasing, while our understanding of their environmental and human health impacts improves slowly. Life cycle assessment (LCA) is one of the tools that evaluate environmental and human health impacts of chemicals. Traditional LCAs often rely on the models that represent broad spatial boundaries at regional, national, or global scales. However, the use, release, fate, and transport of chemicals, which are collectively referred to as biophysical processes, may vary substantially within those boundaries. This misalignment in spatial attributes between LCA models and the biophysical processes that determine environmental and human health impacts is one of the major sources of uncertainties in LCA. This dissertation is an attempt to close the gap between the spatial resolutions of the models used in LCA and the biophysical processes relevant for understanding the environmental and human health impacts of chemicals.

This dissertation covers three topics that collectively addresses the aforementioned objectives: (1) measuring spatial variabilities in LCI, (2) modeling the fate of chemicals in the environment at the spatial resolution that matches with the underlying biophysical processes, (3) building a systematic release framework to estimate chemical releases that supports the fate modeling.

First, I demonstrated that spatial disparities in state-specific LCI for four major crops in the USA can lead to two to fourfold differences in characterized results for most impact categories. The differences, however, increase to over an order of magnitude for freshwater ecotoxicity and human health non-cancer. Among the crops analyzed, winter wheat shows higher variability partly due to a larger difference in yield. As a result, the use of national average data derived from top corn and soybean producing states significantly underestimates the characterized impacts of corn and soybean in the states where land conversion from cotton to corn or soybean actually took place.

Secondly, I developed a spatially explicit and time-dependent multimedia fate modeling framework, ChemFate, that can be incorporated into regional LCIA. ChemFate consists of four multimedia fate models: (1) organoFate, a model for non-ionizable organic chemicals, (2) ionOFate, a model for ionizable organic chemicals, (3) metalFate, a model for metals, and (4) nanoFate, a model for nanomaterials. ChemFate is able to not only provide predictions for four different classes of chemicals, but also incorporate dynamic emissions and dynamic environmental conditions. The dynamic capability of ChemFate supports the model to simulate with real regional climatic data and produce better model performance.

Thirdly, I built a comprehensive release framework, OrganoRelease, to estimate the release of organic chemicals from the use and post-use of consumer products with limited information. OrganoRelease connects 19 unique functional uses and 14 product categories across 4 data sources and provides multiple pathways for chemical release estimation. The results can be used as input for methods estimating environmental fate and exposure.

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I. Introduction

A. Background

Chemicals are significant to humans and they are everywhere in our everyday items: food, clothing, personal care products, cleaning products, plastics, furniture, and paints. While chemicals bring benefits to our modern society, they can also pose harmful impact to humans and the environment. As of February 2019, over 86,200 chemicals are listed on the U.S. Toxic Substance Control Act (TSCA) Inventory, with over 40,600 active substances in U.S. commerce today (US EPA, 2015). However, only over 1,000 of them are regulated by major U.S. environmental statutes (Schwarzman and Wilson, 2009). As a result, the human health and ecological risks associated with those chemicals are poorly understood. Furthermore, over 140 million chemicals are registered under the Chemical Abstract Service, which includes every chemical substance described in the open scientific literature (CAS, 2018). With this number grows rapidly, our understanding of these chemicals environmental and human health impacts is facing a huge challenge (Li and Suh, 2019). Spatial patterns are important when considering ecosystem dynamics (Reap et al., 2003). The impact of a chemical to ecosystem and human health depends on spatial variables that are not homogenous across geographies due to the differences in 1) the amount of chemicals usage, 2) the amount of chemicals release, and 3) the behavior of chemicals in the environment.

Life cycle assessment (LCA) is a tool that is widely used to evaluate environmental and human health impacts of chemicals as well as products and processes where chemicals play a central role (Fantke and Ernstoff, 2018). LCA is the process of compiling inputs and outputs, and evaluating their potential environmental impacts of a product system throughout its life cycle, including raw material extraction, manufacturing, production, use and disposal

(Heijungs and Guineév, 2012). LCA calculation primarily consists of two parts, life cycle inventory (LCI) analysis and life cycle impact assessment (LCIA). LCI involves the compilation and quantification of input and output data, while LCIA translates the inventory of emissions (chemicals) into an environmental profile of a wide range of environmental impacts, such as freshwater ecotoxicity, human health cancer, global warming potential, acidification, ozone depletion, etc. This translation is through a substance-specific Characterization Factor (CF), which is independent of spatial and temporal information. While generic CFs work for global impacts, such as global warming potential and ozone layer depletion, they can misrepresent the environmental burdens of site-specific impact categories, such as ecotoxicity and acidification, due to huge variations in receiving environments (Potting and Hauschild, 2006).

Traditionally, LCA has relied on global or national inventory data and CFs, and spatial heterogeneity has not been thoroughly studied due to: (1) the lack of modeling frameworks with explicit consideration of spatial information, and (2) the lack of data on emission locations that associated with products production, use, and disposal (Chaplin-Kramer et al., 2017; Reap et al., 2003). Emission locations, in most cases, are not known in most LCA studies (Fantke and Ernstoff, 2018). LCIA methods are generally based on regional or global averages for chemicals (Kounina et al., 2014). However, the exclusion of geographical differences in LCI and the lack of spatial considerations in LCIA could introduce huge uncertainties into LCA results that may mislead decision-making (Ciroth et al., 2002; Reap et al., 2003; Ross and Evans, 2002).

While research on regional LCA, with region-specific inventories and CFs, has been largely focused on LCIA methods, particularly on acidification (Bare et al., 2003; Civit et al., 2014; Fréchette-Marleau et al., 2008; Roy et al., 2014) and eutrophication (Gallego et al., 2010; Helmes et al., 2012), less attention has been paid to how LCIs may vary across regions. Furthermore, the fate models, which is used to track chemical behaviors in the environment in LCIA methods for ecotoxicity and human health impact categories, still ignore spatial differentiation in the modeling framework (Rosenbaum et al., 2011). In addition, although understanding the amount of chemicals released to the environment is a key in all these models, there is no established method to estimate this quantity based on the key characteristics of chemicals in products.

B. Objectives

The objective of this dissertation is to close the gap between the spatial resolutions of LCA models and the relevant biophysical processes for better understanding the environmental and human health impacts of chemicals. This dissertation addresses the objective with three topics: (1) measuring spatial variabilities in LCI, (2) modeling the fate of chemicals in the environment at the spatial resolution that matches with the underlying biophysical processes, (3) building a model that estimates the release of chemicals during the use and post-use phase to allow location-specific fate modeling.

In Chapter 2, I demonstrated that spatial disparities in state-specific LCI for four major crops in the USA can lead to two to fourfold differences in characterized results for most impact categories. The differences, however, increase to over an order of magnitude for freshwater ecotoxicity and human health non-cancer. Among the crops analyzed, winter wheat shows higher variability partly due to a larger difference in yield. As a result, the use of national average data derived from top corn and soybean producing states significantly underestimates the characterized impacts of corn and soybean in the states where land conversion from cotton to corn or soybean actually took place. This chapter highlights the sheer influence of spatial heterogeneity across states to the overall uncertainties of agricultural crop LCAs and the need to minimize it to improve reliability of LCAs.

In Chapter 3, I developed a spatially explicit and time-dependent multimedia fate modeling framework, ChemFate, that can be incorporated into regional LCIA. ChemFate consists of four multimedia fate models: (1) organoFate, a model for non-ionizable organic chemicals, (2) ionOFate, a model for ionizable organic chemicals, (3) metalFate, a model for metals, and (4) nanoFate, a model for nanomaterials. ChemFate is able to not only provide predictions for four different classes of chemicals, but also incorporate dynamic emissions and dynamic environmental conditions. The dynamic capability of ChemFate supports the model to simulate with real regional climatic data and produce better model performance. This chapter enables the fate modeling of chemicals in life cycle impact assessment (LCIA) that can match the spatial resolution of the chemical use and release.

In Chapter 4, I present a comprehensive release model, OrganoRelease, for estimating the release of organic chemicals from the use and post-use of consumer products with limited information. First, a novel Chemical Functional Use Classifier was constructed to estimate chemical functional uses based on chemical structure. Then, the quantity of chemicals entering different product streams is estimated based on market share data of the chemical functional uses. Finally, chemical releases are estimated based on either chemical product categories or functional uses. OrganoRelease connects 19 unique functional uses and 14 product categories across 4 data sources and provides multiple pathways for chemical release estimation. The results of this chapter can be used as input for location-specific fate modeling by considering local population density and product use behaviors.

Chapter 2 helps advance the understanding of spatial heterogeneity of environmental and human health impacts of agricultural production. Chapter 3 and Chapter 4 contribute to the modeling of chemical release, fate and transport to close the gap of spatial differentiation in regional LCA.

C. References

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Kounina, A., Margni, M., Shaked, S., Bulle, C., Jolliet, O., 2014. Spatial analysis of toxic emissions in LCA: A sub-continental nested USEtox model with freshwater archetypes. Environ. Int. 69, 67–89. https://doi.org/10.1016/j.envint.2014.04.004

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Reap, J., Bras, B., Newcomb, P.J., Carmichael, C., 2003. Improving Life Cycle Assessment by Including Spatial, Dynamic and Place-Based Modeling 77–83. https://doi.org/10.1115/DETC2003/DFM-48140

Rosenbaum, RalphK., Huijbregts, MarkA.J., Henderson, AndrewD., Margni, M., McKone, ThomasE., van de Meent, D., Hauschild, MichaelZ., Shaked, S., Li, D., Gold, LoisS., Jolliet, O., 2011. USEtox human exposure and toxicity factors for comparative assessment of toxic emissions in life cycle analysis: sensitivity to key chemical properties. Int. J. Life Cycle Assess. 16, 710–727. https://doi.org/10.1007/s11367-011-0316-4

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II. Geographic variability of agriculture in LCA and the need for sector-specific uncertainty characterization

A. Introduction

When conducting a Life cycle assessment (LCA), primary data supplied directly from the upstream or downstream processes are the gold standard. However, due to the limitations in time and resources, LCA practitioners are often relying on Life Cycle Inventory (LCI) databases especially for background processes (Guinée 2001). LCA has traditionally relied on global or national inventory data and characterization factors, while the risk of misrepresenting the environmental impacts of a product at regional or local levels has been well pointed out in the literature (Potting and Hauschild 2006). This is especially true when there is large variation in technology or environmental factors within the geographic boundary studied. By contrast, regional LCA, with region-specific inventories and characterization factors, improves the accuracy and relevance of LCA results (Yang and Suh 2015a; Yang 2016).

Over the past two decades, research in regional LCA has largely focused on impact assessment methods. A large number of regional characterization models have been developed for acidification (Bare et al. 2003; Fréchette-Marleau et al. 2008; Civit et al. 2014), eutrophication (Gallego et al. 2010; Helmes et al. 2012; Azevedo et al. 2013), and water (Pfister et al. 2009; Boulay et al. 2011; Hybel et al. 2015; Núñez et al. 2015). In comparison, relatively less attention has been paid to how Life Cycle Inventories (LCIs) may vary across regions at subnational levels. The practice is still pervasive of using national average data to represent products that are highly geographically variable (Weber and Matthews 2008; Hill et al. 2009; Rehr et al. 2010; Tessum et al. 2014).

Building on our previous research (Yang et al. 2012; Yang 2013; Yang and Suh 2015a, b), here we compiled region-specific life cycle inventories (i.e., inputs and emissions) for major crops in the U.S. and evaluated how they may contribute to geographic variability in environmental impacts. We also compared the implications of choosing average data *versus* region-specific data in assessing the environmental impacts of land use change from one crop to another. Last, we analyzed the implications of our findings in characterizing the uncertainties associated with geographic variability under the conventional pedigree approach. This paper aims to measure the spatial variability of intermediate inputs to agriculture in the U.S., and to understand its implications in interpreting LCA results.

B. Materials and Methods

Goal and scope. The functional unit of our study is defined as 1 metric ton crop harvested in a state, and the system boundary covers both direct emissions on site and indirect emissions from the supply chain (i.e., cradle to gate). Direct emissions including CO_2, N_2O, NH_3, and NO_x result from the use of agricultural inputs such as fertilizers, pesticides, and fuels. Indirect emissions include those from transportation and production of agricultural inputs. We begin by compiling inputs data and then estimate direct emissions based on emission factors from various models and studies (section 2.2.). Indirect emissions are derived from the Ecoinvent database (v2.2) (Ecoinvent 2014). Two recent production years are selected for each crop to mitigate potential temporal variation (Yang and Suh 2015b).

Next, characterization is performed for each crop using TRACI (Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts, v2.0), a tool developed by US Environmental Protection Agency (Bare 2011). Impact categories included are:

acidification (air), eutrophication (water), global warming, ozone layer depletion, smog air formation, freshwater ecotoxicity, human health respiratory, cancer, and non-cancer (e.g., developmental and neurotoxic effects). Water consumption and its impacts are not considered in our study given that their geographic variability has been already well documented by previous research (Mekonnen and Hoekstra 2010; Fulton et al. 2013; Wu et al. 2014; Gobin et al. 2017).

Agricultural inputs and direct emissions. Data on fertilizers and pesticides – including herbicides and insecticides – at the state level are from the agricultural chemical use program run by the National Agricultural Statistics Service (NASS) at the US Department of Agriculture (USDA 2016). NASS surveys farmers in top-producing states on a selective and rotational basis. Surveys for the following years are selected for our analysis: (1) corn: 2005 and 2010; (2) cotton: 2003 and 2007; (3) soybean: 2006 and 2012; (4) wheat: 2004 and 2009. On-farm energy use data for each crop at the state level are from surveys conducted by the Economic Research Service at USDA (USDA 2004).

We estimate the following emissions from the inputs used: 1) soil N_2O, NO_x and NH_3 emissions to air due to the use of nitrogen fertilizers (Yienger and Levy 1995; Krauter et al. 2002; Goebes et al. 2003; Ogle et al. 2008); 2) N and P runoff and leaching (USDA 2006); 3) pesticide emissions to various compartments after application (Berthoud et al. 2011); 4) CO_2, CH_4, N_2O, NO_X, SO_X, PM_{10}, $PM_{2.5}$, CO, and VOCs emissions from fuel combustion (Wang 2013); and 5) heavy metals contained in phosphate fertilizers (Mortvedt 1995). Our inventory covers dozens of chemicals, mostly pesticides and volatile organic compounds (VOCs). In estimating the fate of pesticides, we adopt the approach by Berthoud et al (2011). They used the vapor pressure of a pesticide to estimate the fraction emitted to air, assume 0.5% runoff to water, and assume the remaining fraction, capped at 85% in soil.

Exclusion of insecticides. In processing pesticides data from NASS, we exclude insecticides and include only herbicides. Pesticides data from the USDA are often partially available as the agency withholds usage data on certain pesticides to avoid disclosing information for individual farms (Yang 2015). This is especially true for insecticides: over 80% by mass are withheld for over a third of the states surveyed, as opposed to a large proportion of herbicides being released for most of the states (Figure 1).

Because insecticides are a major contributor to a crop's life cycle ecotoxicity impact (Yang 2013; Yang and Suh 2015b), including them would run of the risk of significantly misrepresent the geographic variability of the impact. That one state showed a much higher ecotoxicity impact than the other could be merely a result of data gap, namely, insecticides data were available for the former and not for the latter. Given the issues of data gap and the focus of our study on geographic variability instead of total impact, we exclude insecticides in our analysis.

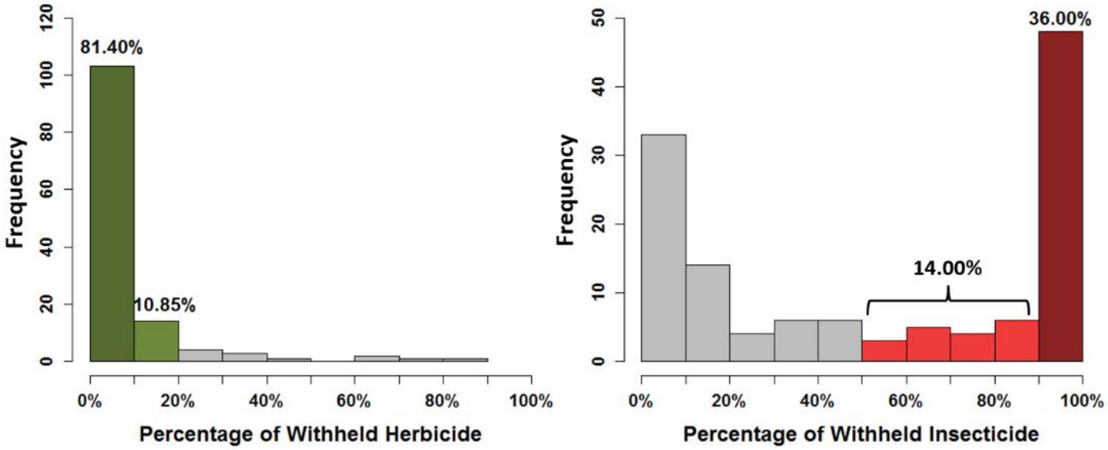

Fig. 1 Number of states for which herbicide (left) and insecticide (right) data are withheld at various percentages (by mass)

C. Results

Geographic variability of major inputs. Table 1 summarizes the geographic variability across the states with regard to inputs and yields. Due to limited space, only the most recent data are presented for each crop. However, similar patterns are observed for other, earlier years. Overall, the inputs and yields all show wide variations, probably due to the spatial variabilities in the weather, soil characteristics, geography, and management practices. Yields vary by a factor of 2 to 4 (maximum/minimum), and inputs in most cases vary by a factor of 2 to 7, while in some cases they vary by over a factor of 10. This is consistent with coefficient of variation, which is generally higher for inputs than for yields (except for the case of cotton). Electricity use (kWh ha^{-1}) seems to be the most variable input, with the largest coefficient of variation ranging from 1.5 to 3.3. Electricity is used primarily for irrigation; the high variation thus reflects the wide difference in the factors such as precipitation, water source (e.g., surface water or groundwater), groundwater depth, and irrigation methods (e.g., gravity or sprinkler). In general, regions with less precipitation and high reliance on groundwater are expected to use more electricity for irrigation per hectare.

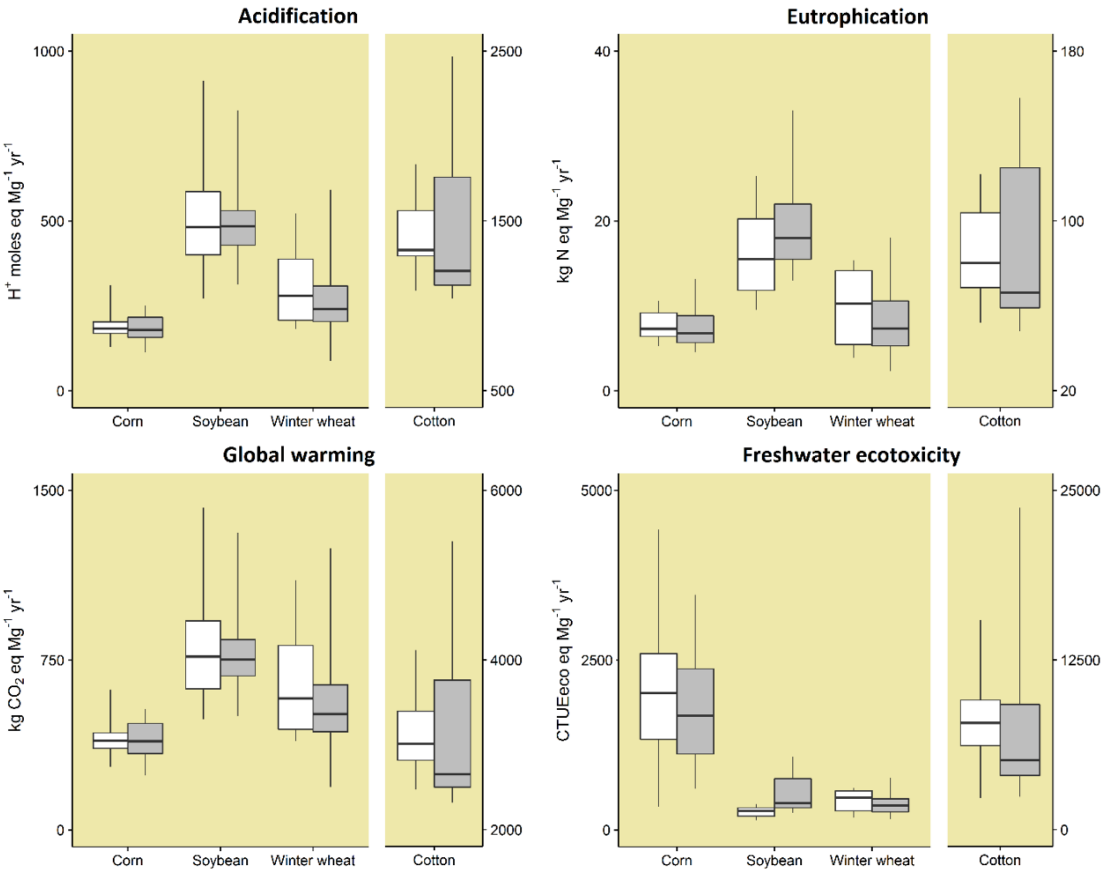

Fig. 2 Geographic variability in select impact categories: acidification, eutrophication, global warming, and freshwater ecotoxicity. Whiskers indicate states with maximal and minimal impacts, boxes indicate upper and lower quartile, and lines across boxes indicate median impacts. Blank boxes are based on results for the older years (2005 for corn, 2003 for cotton, 2006 for soybean, and 2004 for wheat) and grey boxes based on results for the recent years (2010 for corn, 2007 for cotton, 2012 for soybean, and 2009 for wheat) compiled for each crop. The vertical axes on the left are for corn, soybean, and winter wheat, and those on the right are for cotton.

Geographic variability in environmental impacts. As a result of the variation in inputs and yields (Table 1), environmental impacts per ton of crop produced show high variability across the states assessed. Table 2 shows the minimum, mean, maximum, standard deviation, and coefficient of variation of environmental impacts. Again, given the limited spaces, only the most recent results are shown for each crop. For most crops and impact categories, impacts vary by a factor of 2 to 4 across the states. Among the impact categories, freshwater ecotoxicity and human health non-cancer show relatively higher variability compared with others, with coefficient of variation ranging from 0.4 to 0.8. Freshwater ecotoxicity impact is associated primarily with pesticide use. The variation in the freshwater ecotoxicity impacts can be explained by the spatial variabilities in the type and the amount of pesticides used, as well as those in the yields. The variations in human health non-cancer impacts are due to the differences in both yield and the amount of phosphate input, which contains heavy metal impurities that are major contributors to the non-cancer human health impacts. The variation in other impact categories such as smog formation, acidification, eutrophication, and global warming largely reflects the variations in nitrogen input. In the case of cotton, fuel combustion also contributes significantly to these impact categories as cotton farming relies heavily on farm machinery.

Among the crops analyzed, wheat in general shows higher variability (see coefficient of variation in Table 2). This is due in large part to the variation in both yield and nutrient use. Wheat yield varies up to a factor of 3.7 in 2009, as opposed to 1.5-2 for corn and 2 for soybean. Nutrient inputs for wheat also show relatively larger variability. N application rate varies by a factor of 4-6 for wheat, but only 1-2 for corn and 2-3 for cotton. Phosphate application rate varies by a factor of 12 for wheat, but only 3-6 for corn and 3-4 for cotton. Figure 2 shows the geographic variability in selected impact categories.

Table 1. Geographic variability of inputs and yield.

Crop	**Year**	**Input & Yield**	**Unit ($ha^{-1)}$**	**Mean (min, max)**	**Standard Deviation**	**Coefficient of Variation**
Corn	2010	Nitrogen	kg	159 (99, 214)	29	0.2
		Phosphate	kg	52 (18, 106)	22	0.4
		Herbicide	kg	3.0 (1.8, 5.7)	1.0	0.3
		Fuel	Mj	4.6 (1.4, 19)	4.0	0.9
		Electricity	kWh	93 (0, 916)	204	2.2
		Yield	Mg^{-1}	9.1 (5.7, 11)	1.3	0.1
Cotton	2007	Nitrogen	kg	107 (72, 133)	19	0.2
		Phosphate	kg	38 (21, 63)	14	0.4
		Herbicide	kg	3.1 (1.4, 3.8)	0.7	0.2
		Fuel	Mj	9.0 (5.1, 17)	3.1	0.4

		Electricity	kWh	44 (0, 163)	65	1.5
		Yield	Mg^{-1}	1.0 (0.5, 1.7)	0.3	0.4
Soybean	2012	Nitrogen	kg	193 (97, 257)	49	0.3
		Phosphate	kg	21 (6.9, 41)	8.2	0.4
		Herbicide	kg	2.2 (1.6, 3.2)	0.4	0.2
		Fuel	Mj	2.0 (1.0, 7.2)	1.4	0.7
		Electricity	kWh	9 (0, 97)	23	2.4
		Yield	Mg^{-1}	2.7 (1.5, 3.1)	0.4	0.2
Winter Wheat	2009	Nitrogen	kg	82 (23, 137)	30	0.4
		Phosphate	kg	27 (5.4, 65)	17	0.6
		Herbicide	kg	0.8 (0.0, 1.7)	0.5	0.7
		Fuel	Mj	0.9 (0.6, 1.4)	0.2	0.2
		Electricity	kWh	79 (0, 1008)	258	3.3
		Yield	Mg^{-1}	3.2 (1.5, 5.4)	1.0	0.3

Table 2. Geographic variability of environmental impacts.

Crop	**Year**	**Impact**	**Mean (min, max)**	**Standard Deviation**	**Coefficient of Variation**
Corn	2010	ACD	183 (113, 250)	45	0.2
		EUT	7.3 (4.6, 13.2)	2.2	0.3
		GW	390 (240, 532)	91	0.2
		OLD	2.0E-05 (1.3E-05, 3.6E-05)	6.2E-06	0.3
		SF	48 (29, 92)	15	0.3
		HHR	0.31 (0.19, 0.46)	0.07	0.2
		FET	1812 (606, 3459)	889	0.5
		HHC	5.9E-06 (3.8E-06, 9.3E-06)	1.5E-06	0.3
		HHNC	4.7E-05 (2.1E-05, 1.0E-04)	1.9E-05	0.4
Cotton	2007	ACD	1472 (1044, 2469)	500	0.3
		EUT	89 (48, 158)	42	0.5
		GW	3235 (2319, 5400)	1054	0.3
		OLD	2.4E-04 (1.9E-04, 3.8E-04)	6.8E-05	0.3
		SF	434 (315, 710)	134	0.3
		HHR	3.6 (2.6, 5.7)	1.1	0.3
		FET	7815 (2405, 23710)	6094	0.8
		HHC	4.7E-05 (2.8E-05, 8.4E-05)	1.9E-05	0.4
		HHNC	3.7E-04 (1.7E-04, 6.7E-04)	2.1E-04	0.6
Soybean	2012	ACD	506 (313, 826)	135	0.3
		EUT	19 (13, 33)	6	0.3
		GW	799 (501, 1311)	207	0.3
		OLD	1.7E-05 (10.0E-06, 4.1E-05)	7.7E-06	0.5
		SF	115 (71, 195)	30	0.3
		HHR	0.38 (0.26, 0.62)	0.11	0.3
		FET	517 (244, 1080)	264	0.5

		HHC	3.3E-06 (1.7E-06, 6.0E-06)	1.1E-06	0.3
		HHNC	5.7E-05 (1.7E-05, 1.1E-04)	2.4E-05	0.4
Winter wheat	2009	ACD	263 (87, 592)	117	0.4
		EUT	7.9 (2.3, 18.1)	4.1	0.5
		GW	555 (190, 1245)	244	0.4
		OLD	2.3E-05 (1.1E-05, 4.9E-05)	9.2E-06	0.4
		SF	64 (23, 145)	28	0.4
		HHR	0.42 (0.16, 0.89)	0.18	0.4
		FET	376 (155, 763)	162	0.4
		HHC	8.3E-06 (2.6E-06, 1.8E-05)	3.8E-06	0.5
		HHNC	7.2E-05 (1.6E-05, 1.6E-04)	4.1E-05	0.6

Note: ACD – acidification, EUT – eutrophication, GW – global warming, OLD – ozone layer depletion, SF – smog formation, HHR – human health respiratory, FET – freshwater ecotoxicity, HHC – human health cancer, and HHNC – human health noncancer.

Differences in marginal impacts due to geographic variability. The geographic variability discussed in the previous section suggests that regional and local specifics of agricultural products may not be adequately captured in the national average LCI data. For example, 1 ton of cotton in the US in 2007 is estimated to generate 2.8 ton CO_2 eq on average, as opposed to 5.4 ton CO_2 eq for cotton in South Carolina. Had we used the national average to represent South Carolina cotton, we would have underestimated its climate change impact by 50%.

Regional inventories are also instrumental in understanding the environmental consequences of marginal changes, which may differ from that estimated using average data (Weidema et al. 1999; Varvel et al. 2008; Yang and Suh 2015c). In the past decade, biofuel policies in the US aimed at boosting ethanol production have contributed to rising corn/soybean prices and led the two crops to expand (Wallander et al. 2011). One of the crops displaced by them was cotton. It was estimated that between 2005 and 2009, hundreds of thousands of hectares of cotton were displaced by corn and soybean expansion (Wallander et al. 2011). As cotton is generally grown in the South, determining the environmental consequences of the expansion requires understanding how corn and soybean are cultivated in the South, instead of relying on national average data, which are largely a portrayal of corn and soybean in the Midwest (Yang and Suh 2015a) (Yang and Suh 2015a).

As Figure 3 indicates, there is indeed a significant difference, across all impact categories, between Southern corn/soybean and Midwest corn/soybean. Using average data derived from Midwest states in this case would significantly underestimate the environmental impacts of corn and soybean expansion.

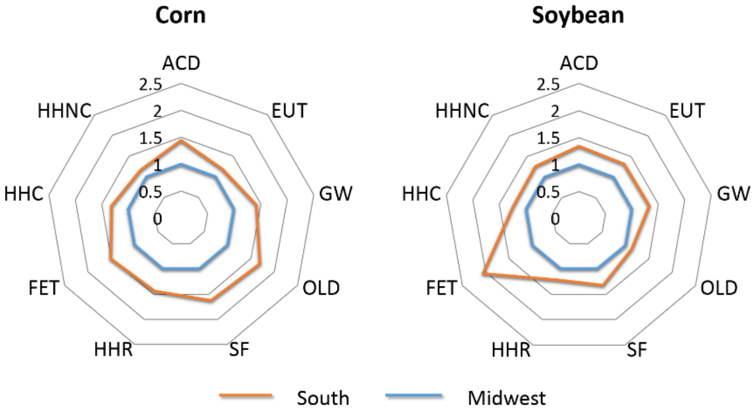

Fig. 3 Comparison between environmental impacts of corn/soybean in the Midwest, based on 9 major states (Shapouri et al. 2010), and that of corn/soybean in the South, where cotton is grown. Results are based on per Mg harvested and normalized to that of Midwest corn/soybean. In general, corn and soybean produced in the South generate higher environmental impacts than in the Midwest. This is partly because soil in the South is not as suited as soil in the Midwest to growing corn and soybean thus yield in the South is generally lower.

Uncertainty characterization using pedigree method. One of the dominant approaches to uncertainty characterization in LCA is the pedigree method (Weidema and Wesnæs 1996; Weidema 1998). The pedigree method assigns Geometric Standard Deviation (GSD) to unit process-level elementary flows based on qualitative characteristics of the unit process data. The original approach has been recently updated by Ciroth et al. (2013) partially based on empirical data including the European Pollutant Release and Transfer Register (E-PRTR).

The updated pedigree method uses five indicators, namely reliability, completeness, temporal correlation, geographical correlation, and further technological correlation, and assigns GSD accordingly. Table 3 compares the previous and updated GSD assignments to geographical correlation. It shows that the updated method generally assigns higher GSDs to geographical correlation, indicating potential underestimation of variability due to spatial differences.

Table 3. A Comparison of GSD (Geometric Standard Deviation) assignments to geographical correlation under the original and updated Pedigree method.

	1	**2**	**3**	**4**	**5**
Original method[*]	1	1.01	1.02		1.10
Updated method[†]	1	1.04	1.08	1.11	(n.a.)

References: [*](Weidema and Wesnæs 1996; Weidema 1998); [†](Ciroth et al. 2013)

In the updated method, five levels for geographical correlation are defined, namely, (1) data from the area under study, (2) average data from a larger area in which the area under study is included, (3) data from the area with similar production conditions, (4) data from the area with slightly similar production conditions, and (5) data from unknown or distinctly different area (Weidema and Wesnæs 1996; Ciroth et al. 2013). For each of these levels, different geometric standard deviation (GSD) values ranging from 1 to 1.11 are assigned (Table 3). Regional differences in intermediate inputs and outputs for iron and steel manufacturing or electronics, for example, may not be as large as those for agricultural crops, but the current pedigree approach assigns the same level of uncertainty to geographic correlation regardless of the sectoral identity. In other words, pedigree method is sector-generic; the same magnitude of uncertainty is assigned to LCIs regardless of the sector in question.

However, our results show that the current assignment of GSD values to geographical correlation is a significant underestimation for the crops analyzed in our study. Based on the data compiled, GSDs of agricultural inputs due to spatial differences can be as high as 7.1, with the average value of 2.2 and the median value of 1.9. The highest GSD under the current approach is 1.11 (Table 3). Figure 4 shows frequency distribution of GSDs for the overall and three individual types of agricultural inputs – energy, fertilizer, and herbicide. As can be seen from the lines, the computed average GSD values are all higher than the default highest GSD used in the pedigree approach. The number of states involved in generating those GSD values is shown in Electronic Supporting Material Figure S1. The two highest GSDs are both greater than 6.0, which are calculated based on 11 states' data points under the energy sector. This corresponds to the largest coefficient of variation value for the electricity input (Table 1). Around 65% of the GSDs are computed from more than 5 states. Table 4 indicates the average GSDs for four crops across three types of agricultural inputs. All of the average GSDs are above 1.7, much greater than the default highest GSD assigned to the geographical correlation in the pedigree method. Our findings reinforce those of Ciroth et al (Ciroth et al.

2013), who showed that the conventional pedigree approach yields an underestimation of GSDs.

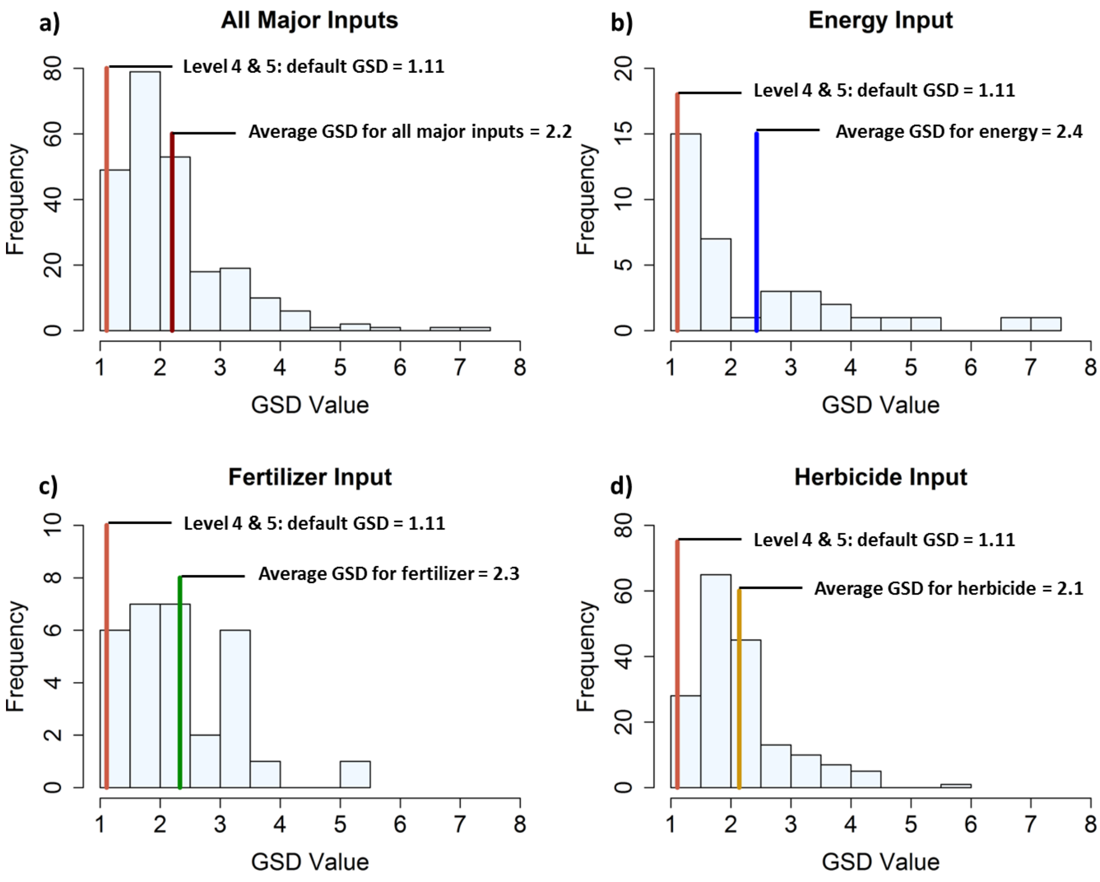

Fig. 4 Geometric standard deviation (GSD) of agricultural inputs. Panel a) displays the frequency distribution of the GSDs of all of the major agricultural inputs across the states for the four studied crops. Panel b), c), and d) show the individual frequency distribution of GSDs for energy, fertilizer, and herbicide. The red lines in a), b), c) and d) indicate the highest GSD corresponding to the geographical correlation levels 4 and 5 following the conventional pedigree approach.

Ideally, GSD values in LCI associated with pedigree criteria should be determined at a sectoral level, depending on the relevance of the geographical variability to the sector and corresponding technologies. Sectors and technologies that are heavily affected by the geographical variabilities such as temperature and its variation, precipitation (both frequency and intensity), soil characteristics, insulation, and abundance or scarcity of certain resources should be the priorities for developing sector-specific GSDs. Such sectors and technologies may go beyond those of agriculture: geographical variability plays an important role in determining the inputs, outputs, and the performance of heating and cooling equipment, insulation materials, and renewable energy.

D. Conclusion

In this study, we evaluated the geographic variability in environmental impacts of 4 major crops in the US across the states. Maximum characterized results can be 2 to 4 times larger than the minimum across the states depending on the impact categories. Exceptions include herbicide-related freshwater ecotoxicity and human health toxicity, which could vary by more than a factor of 10. Across the crops, wheat in general shows higher variability, because of a larger difference in yield and nutrient inputs used across the states. Note that the variability evaluated in our study is only due to the differences in input use and the emissions modelled, and region-specific fate, transport, exposure, and impacts were not included. The inventories compiled in this study can be combined with regional impact assessment models reflecting relevant regional conditions including population density and region-specific environmental variables.

We also showed that regional inventories can better serve the need of understanding the environmental impacts of marginal land use change in the case of cotton to corn and soybean conversion, which cannot be adequately captured by using national average data. This is especially important when marginal changes are substantially different from the average.

Lastly, we showed that the default GSDs assigned to geographical correlations in the pedigree approach substantially underestimate the underlying uncertainties in the LCIs of the crops analyzed. We recommend using a sector-specific instead of sector-generic approach to uncertainty characterization in LCIs.

E. Acknowledgement

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III. ChemFate: integrating dynamic fate modeling across multiple classes of chemicals

A. Introduction

A major challenge in comparing the risk of chemicals from radically different classes is that their behavior in the environment requires taking into consideration their particular properties and processes. For example, how should we compare a novel nanopesticide to existing ionizable or non-ionizable organic chemicals, or a pesticide based on metal salts, all used as fungicides? Or a sunscreen that may contain nano titanium dioxide or organic UV blockers/quenchers? While the characteristics of the geographical region of interest are the same, a correct assessment needs to consider the radically different properties of each one of these classes of chemicals. Multimedia fate and transport models are widely used in chemical risk assessment, chemical ranking and management support (Su et al., 2019; Zhu et al., 2016). They are designed to understand chemical transport behavior and fate in the environment and predict their environmental concentrations in environmental compartments.

However, most fate and transport models are specific to a particular class of chemicals. This makes a proper comparison of different classes of chemicals challenging, because different models are built under different modeling formulations and covers different sets of environmental compartments. The most widely adopted multimedia fate and transport model is Equilibrium Criterion (EQC), a fugacity-based model developed by Mackey in 1996 and recently updated in 2012 (Hughes et al., 2012). However, it only applies to non-ionizable organic chemicals and considers four environmental compartments (air, water, sediment, and soil). Another prevalent model, called Multimedia Activity Model for Ionics (MAMI), is an activity-based model that considers chemical speciation in the environment and applies to both non-ionizable and ionizable organic (acids and bases) chemicals (Franco and Trapp, 2010a; Trapp et al., 2010a). MAMI is able to model seven environmental compartments, including air, freshwater, freshwater sediment, marine, marine sediment, and three soil types (urban, agriculture, and other), but it is not applicable to metals or nanomaterials. TRANSPECII is a coupled metal speciation-fate model that is specific for metals, and it considers water, sediment (surficial and deep), and soil (surficial and deep) (Bhavsar et al., 2008). TRANSPECII keeps track of metal distribution among three phases: dissolved, colloidal, and particulate. Simplebox is a concentration-based nested multimedia fate model that is able to model non-ionizable organics, ionizable organics, and metals, and it contains the same seven environmental compartments as MAMI (Hollander et al., 2016a). However, Simplebox only models two phases for metals (particulate and dissolved), and ambient chemistry parameters are not considered in metal distributions. Table S1 shows a more detailed model comparisons regarding to model capability and environmental compartment specificity.

EQC and TRANSPECII are steady-state (level III) models with the assumption of constant environmental conditions and emissions (Bhavsar et al., 2008; Mackay et al., 1996). MAMI and Simbplebox has the qusai-dynamic feature that incorporates the variability in emissions (Franco and Trapp, 2010a; Hollander et al., 2016b). The dynamic pattern of chemical emissions is able to capture the temporal concentration changes in the past and providing realistic predictions in the future (Ao et al., 2009; Di Guardo et al., 2006; Luo et al., 2007; Su et al., 2018; Sweetman et al., 2002). However, without considering the dynamic nature of important environmental conditions (e.g., temperature, precipitation, hydrologic flow), the models may underestimate or overestimate chemical concentrations and exposures (Ghirardello et al., 2010; Morselli et al., 2018; Parker and A. Keller, 2019; Su et al., 2019).

Here we present ChemFate, a uniform framework that predicts chemical environmental concentrations for four classes of chemicals and incorporates both dynamic emissions and environmental conditions. ChemFate comprises four different models: (1) organoFate, a model specific for modeling the fate and transport of non-ionizable organic chemicals; (2) ionOFate, a model for ionizable organic chemicals; (3) metalFate, a model for metals; and (4) nanoFate, a model for nanomaterials. These models can be run with the same environmental characteristics for particular geographical regions (e.g. San Francisco, New York City, Des Moines), including the local temporal meteorological data. This allows the direct comparison of chemicals of radically different classes under comparable environmental conditions.

ChemFate predicts chemical concentrations in nine compartments: atmosphere (including air and aerosols), four types of soils (including urban, natural, agricultural with and without applied biosolids) divided into surface soil solids, soil air, soil pore water, and deep soil compartments, two types of water (including freshwater, costal water, and suspended sediment in both), and freshwater and coastal marine sediments. ChemFate uses well developed equations that take into account of chemical specificity and climatic variability. For example, ChemFate uses Stoke's law to calculate the deposition rates by considering particle density, fluid (air and water) density, and dynamic viscosity, which was well adopted in modeling particle transport and deposition (Nho-Kim et al., 2004; Tsuda et al., 2013). Moreover, soil runoff, soil erosion, and infiltration processes are modeled dynamically by considering daily precipitation intensity, regional slope and soil erodability. ChemFate considers a wider range of processes that are not modeled in the above mentioned models, including aerosol resuspension from coastal splash and soils by wind that results in transfer back to the aerosol compartment.(Gillette and Passi, 1988; Qureshi et al., 2009)

ChemFate is unique because of (i) the consistent modeling framework of dynamic inputs and time-dependent outputs to predict a comprehensive list of four different classes of chemicals under comparable environmental conditions; (ii) the type and structural detail of compartments included; (iii) the key transport processes that considers chemical and regional specificity instead of default rate constants. ChemFate is also developed in a way that is extensible to include other environmental compartments and additional transport processes. In the following sections, we present the general conceptual framework of ChemFate and the key processes considered in all models, then we evaluate each model in ChemFate against other models and observational data available.

B. Materials and Methods

ChemFate Framework. ChemFate is framework that comprises four dynamic (level IV) multimedia models that predicts the fate of four classes of chemicals: (1) organoFate, a model for non-ionizable organic chemicals; (2) ionOFate, a model for ionizable organic chemicals; (3) metalFate, a model for metals; (4) nanoFate, a model for engineered nanomaterials. ChemFate is designed to predict daily time-dependent concentrations in a specific region and non-steady state. Figure 1 show the conceptual diagram of ChemFate

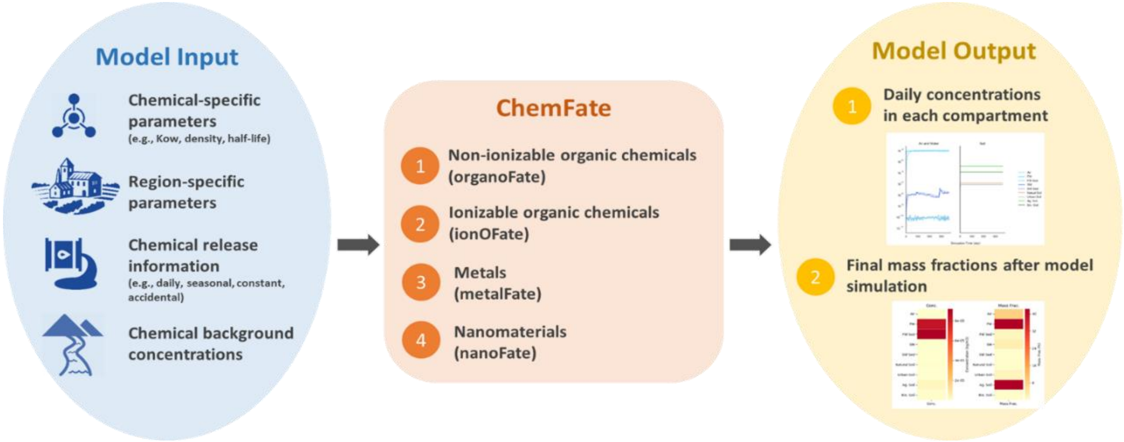


Fig 2. Conceptual diagram of ChemFate with model inputs and outputs

with model input and output information. Four major model inputs are chemical-specific parameters, region-specific parameters, chemical release information, and chemical background concentrations. ChemFate considers different chemical parameters for different types of chemicals, for example, chemical half-life in various environmental compartments are needed for non-ionizable and ionizable organic chemicals, but not for metals due to their negligible degradation process. Region specificity is reflected in the consideration of compartment dimensions and characteristics (e.g., soil properties, water chemistry) and in the incorporation of local observed daily temperature, precipitation, wind speed, and river flow. ChemFate can also model various chemical release scenarios, including daily constant release, daily various release, seasonal release and accidental spill, as chemical release information can be input at a daily level. For chemicals with known background concentrations in environmental compartments, ChemFate incorporates them into the model simulations. To provide a rapid evaluation of chemical fate in various regions, we have collected a selection of 12 default environments to represent a range of distinct climate conditions and land use types and a global environment. Table 1 shows the list of default regions with eight regions in the U.S. and four regions in the Europe. We also documented the detailed procedures to guide the user to develop a customized region (Supporting Information User Guide (SIUG), Section 6). Since the modeling part for engineered nanomaterials has been published (Garner et al., 2017), this paper focuses on the other three classes of chemicals.

Table 1. The list of default regions available in ChemFate

Region	State	Country
Austin	Texas	United States
Des Moines	Iowa	United States
Los Angeles	California	United States
San Francisco	California	United States
Visalia	California	United States
Miami	Florida	United States
Salem	Oregon	United States
New York City	New York	United States
London		United Kingdom
Rome		Italy
Stockholm		Sweden
Zurich		Switzerland
Global Environment		

ChemFate Methodology. ChemFate uses fugacity concept for non-ionizable organic chemicals and aquivalence concept for ionizable organic chemicals and metals. The concept of fugacity was firstly defined by Mackay (Mackay and Paterson, 1981) based on the behavior of ideal gases to model the chemical fate in the environment. However, the use of fugacity is not suitable for involatile chemicals, such as ionic species or metals (Mackay et al., 2003; Mackay and Diamond, 1989). Thus, the approach of aquivalence was introduced as an equilibrium criteria for ionizable chemicals, along with a pseudo single species mass balance modeling technique to multi-species chemicals (Diamond et al., 1992). Both fugacity and aquivalence are linearly related to concentration through chemical capacity Z values, and also D values are used to quantify transport and transformation rates for all chemicals (Diamond et al., 1992; Csiszar et al., 2011). The concentration C (mol/m3) of a chemical species is calculated by fugacity (Pa) and fugacity capacity Z_f (mol/m3-Pa) for non-ionizable organic chemicals, and aquivalence Q (mol/m3) and aquivalence capacity Z_Q (dimensionless) for ionizable organic chemicals and metals, $C = fZ_f = QZ_Q$. Z values are generated by phase partition coefficients and Z_Q values are species-specific for each phase and compartment. The detailed calculations of Z values and partition coefficients are described in SIUG, Section 3.1.

ChemFate predicts chemical concentrations in nine compartments: atmosphere (including air and aerosols), four kinds of soils (including urban, natural, agricultural with and without applied biosolids) divided into surface soil solids, soil air, soil pore water, and deep soil compartments, two types of water (including freshwater, costal water, and suspended sediment in both), and freshwater and coastal marine sediments (Figure 2). ChemFate simulates chemical transformations and transfers between compartments, tracking neutral form for non-ionizable organic chemicals and both neutral and primary ionic forms for ionizable organic chemicals. For metals, ChemFate tracks chemical concentrations in three phases, which are particulate, colloidal, and dissolved phases. Metals in particulate phase is defined as the metal sorbed to particulate matters, metals in colloidal phase is the metal bound to dissolved organic matters, and metals in dissolved phase include free ions and inorganic metal complexes (Bhavsar et al., 2004a).

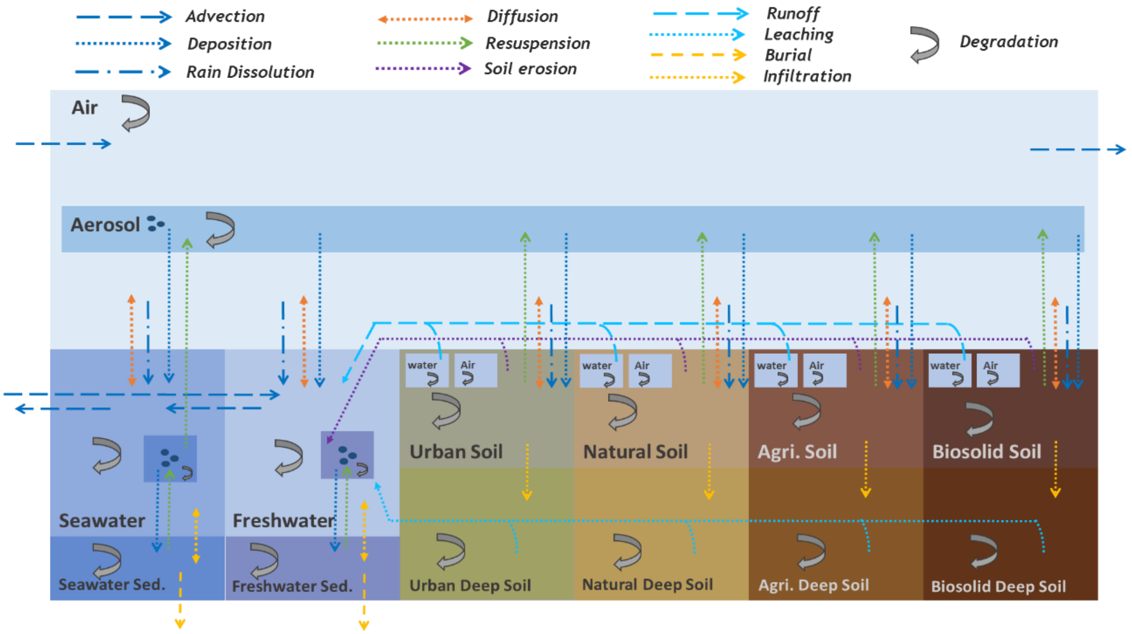


Fig 3. Environmental compartments and major transport and transformation processes considered in ChemFate.

Following the simplicity tradition of multimediate fate models, we assume an instantaneous and homogenous distribution of species within an environmental compartment (Mackay, 2001; Bhavsar et al., 2004a, 2008; Franco and Trapp, 2008; Trapp et al., 2010a; Csiszar et al., 2011). Furthermore, we also considers the overall reaction process following a first order reaction process and converts chemical half-lives into reaction rate constants (Mackay, 2001). ChemFate accounts for intercompartmental transport through intermedia diffusion and advective processes. The diffusion exchange is considered between air-water, air-soil, and sediment-water for non-ionizable organic and ionizable organic chemicals. For metals, only diffusion between sediment-water is included as other diffusion exchanges are not applied to metals (Bhavsar et al., 2004a, 2008). A suite of advective transports are considered in ChemFate, including advection in and out of the system by wind in air and by water flow in freshwater and coastal seawater and their sediments, dry and wet deposition (from air and aerosols), soil water runoff and soil erosion during precipitation events, suspended sediment sedimentation in water, resuspension and burial in sediment, aerosol resuspension from surface soil and coastal seawater, infiltration from surface soil to deep soil, and horizontal leaching from deep soil to freshwater. Those transport processes are described in detail in the SIUG, Section 3.4 and the mass balance equations for organoFate, ionOFate, and metalFate are described in the SIUG, Section 3. Table 2 shows the chemical forms and phases that are tracked in the modeled transformation and transport processes for organoFate, ionOFate, and metalFate.

Table 2. Key transport and transformation processes in the environmental compartments with tracked chemical forms (i.e., neutral-n and ionic-i) and phases (i.e., dissolved-d, particulate-p, and colloidal-c).

Compartment	Process	organoFate	ionOFate	metalFate
Air	Advection In/Out	n (d, p)	n (d, p), i (d, p)	i (d, p, c)
	Rain Dissolution	n (d)	n (d), i (d)	--
	Aerosol Dry Deposition	n (p)	n (p), i (p)	i (d, p)
	Aerosol Wet Deposition	n (p)	n (p), i (p)	i (d, p)
	Air-Water Diffusion	n (d)	n (d), i (d)	--
	Air-Soil Diffusion	n (d)	n (d), i (d)	--
	Degradation	n (d, p)	n (d, p), i (d, p)	i (d, p, c)
Water	Advection In/Out	n (d, p)	n (d, p), i (d, p)	i (d, p, c)
	Water-Air Diffusion	n (d)	n (d)	--
	Suspended Particle Deposition	n (p)	n (p), i (p)	i (p)
	Water-Sediment Diffusion	n (d)	n (d), i (d)	i (d, c)
	Aerosol Resuspension	n (p)	n (p), i (p)	i (p)
Sediment	Advection In/Out	n (d, p)	n (d, p), i (d, p)	i (d, p, c)

	Sediment Resuspension	n (p)	n (p), i (p)	i (p)
	Sediment-Water Diffusion	n (d)	n (d), i (d)	i (d, c)
	Burial	n (p)	n (p), i (p)	i (p)
Soil	Soil-Air Diffusion	n (d)	n (d), i (d)	--
	Runoff (water)	n (d)	n (d), i (d)	i (d, c)
	Erosion (solid)	n (p)	n (p), i (p)	i (p)
	Wind Erosion	n (p)	n (p), i (p)	i (p)
	Leaching (horizontal)	n (d)	n (d), i (d)	i (d, c)
	Infiltration (vertical)	n (d)	n (d), i (d)	i (d, c)

Model Evaluation. We selected three chemicals, benzene, 2,4-D, and Zn, to evaluate the three models in ChemFate. Benzene was chosen as a non-ionizable organic chemicals to validate organoFate, 2,4-D was chosen as an ionizable organic acid to validate ionOFate, and Ni was chosen as a metal to validate metalFate. In this study, we primarily focus on the comparison of model results with other widely used models, specifically, EQC (Hughes et al., 2012), MAMI (Franco and Trapp, 2010a; Trapp et al., 2010b), SimpleBox (Hollander et al., 2016), and TRANSPEC (Bhavsar et al., 2008, 2004b). Even though ChemFate is a dynamic level IV model and other models are level III, ChemFate model results are comparable to theirs with constant climate data, emission scenarios and a long 10-year simulation time. To demonstrate ChemFate's dynamic capability as a level IV model, we used 2,4-D as a case study to compare the results with field studies compiled in Franco's paper (Franco and Trapp, 2010b). In a separate study, we compare the outcome of ChemFate with field monitoring data for further evaluations [cited Nicol's].

Model Availability. ChemFate is publically available on Github and a simple graphical user interface (GUI) is downloadable to run the model. The source code would be available upon request.

C. Results

Predicted Exposure Concentrations for organoFate. Benzene was selected as an non-ionizable organic chemical for the evaluation of organoFate to compare the model results with EQC, MAMI, and SimpleBox. The chemical input data for benzene were gathered from EQC substance database (Table 3), and the environmental parameters (Table S1) were chosen from a Canada region described in MAMI(Franco and Trapp, 2010b). EQC only considers one general water compartment and one general soil compartment. For model comparison purpose, we parametrized the water compartment as a freshwater compartment and the volume to be the sum of freshwater and seawater volumes. The similar approach was also applied to EQC's soil compartment as we assumed the soil compartment as a natural soil with a total volume as the sum of natural soil, agricultural soil, and other soil described in MAMI. Even though organoFate considers four soil types, to make the results comparable to MAMI and Simplebox, we turned off the fourth soil type – agricultural soil with biosolids in organoFate. To make the models comparable, the rate constants in major transport processes were also parametrized to be the same in those models (Table S2). The emission scenario for benzene was set to be 24000 kg/day released to air only.

Table 3. Chemical input properties for benzene.

Chemical Property	Value	Unit
Molecular Weight	78	g/mol
Molecular Density	0.88	g/cm3
Octanol/Water Partition Coefficient	134.90	--
Organic Carbon/Water Partition Coefficient	67	L/kg

Air/Water Partition Coefficient	1.48	--
Aerosol/Air Partition Coefficient	2.37E-10	m3/ug
Half Life in Air	17	hr
Half Life in Freshwater	170	hr
Half Life in Freshwater Sediment	1700	hr
Half Life in Seawater	170	hr
Half Life in Seawater Sediment	1700	hr
Half Life in Natural Soil	17	hr
Half Life in Agricultural Soil	17	hr
Half Life in Other Soil	17	hr
Note: EQC provides half-lives in water, sediment, and soil compartments, and we assume the half-lives are the same in freshwater and seawater and among different types of soil.		

In this example, the first dominant transport processes were aerosol dry/wet deposition and rain dissolution that brought benzene from air to water and soils, and then the other diffusion and advective processes (e.g., sedimentation, runoff, and erosion) followed for compartmental transfers. Figure 3 displays the predicted chemical concentrations in eight environmental compartments calculated by organoFate at the end of 10-year simulation time, compared with the results obtained from MAMI, Simplebox, and EQC. organoFate results for all compartments almost fall into the same order of magnitude as the results from the other three models. The concentrations in air compartment predicted by organoFate and by other three models are very similar. The concentrations in other compartments predicted by organoFate are in close agreement with MAMI and EQC.

Predicted Exposure Concentrations for ionOFate. 2,4-D was selected as an ionizable organic chemical for the evaluation of ionOFate to compare the model results among ChemFate, MAMI, and Simplebox. Furthermore, since the field studies of 2,4-D for air and freshwater compartments in the interested region were compiled in Franco's study, those model outputs were also compared to those observational data. The chemical input data (Table 3), environmental parameters (Table S2), and rate constants for major transport processes (Table S3) for 2,4-D were gathered from Franco's study.(Franco and Trapp, 2010b) A realistic emission scenario was implemented for 2,4-D with 21.22 kg/day released to air and 509.28 kg/day released to agricultural soil.(Franco and Trapp, 2010b)

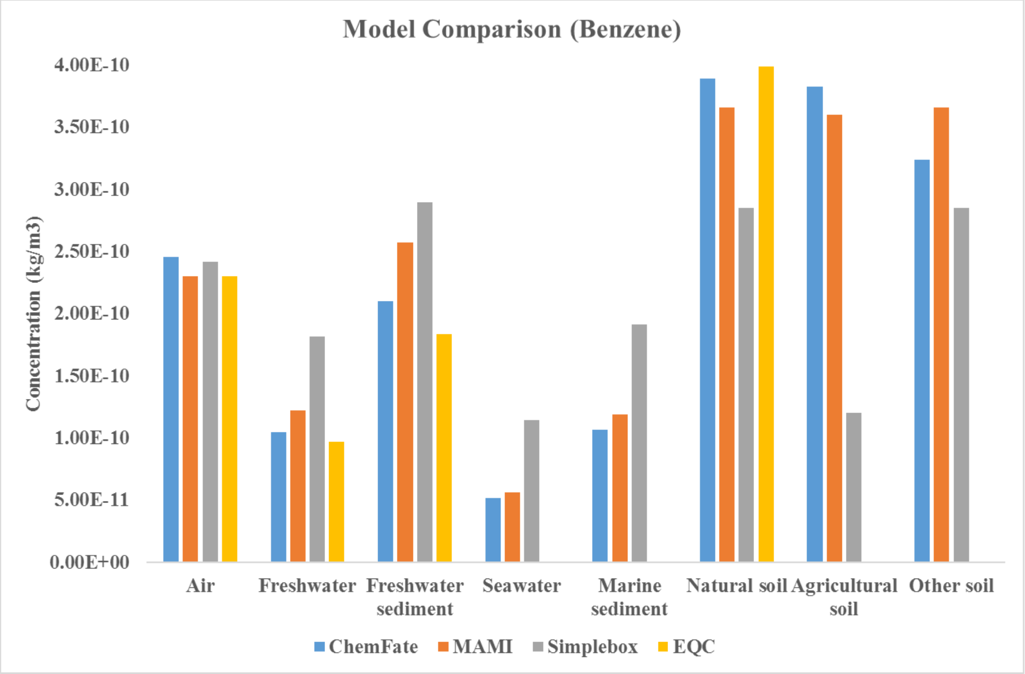


Fig 4. Model results comparison for benzene. The simulation results from organoFate are at the end of 10-year simulation time.

Model with Default Rate Constants. Even though ionOFate is able to model the dynamic deposition rates of aerosol dry deposition and suspended sediment deposition, we used the same deposition rate constants for direct comparisons here. The same approach was also applied to soil solid erosion and soil water runoff processes. The functions of aerosol resuspension from coastal splash and soils by wind were turned off, as they were not modeled in neither MAMI nor Simplebox. The major loss processes are advection from air and biodegradation in compartments. After the release of 2,4-D to air, it is rapidly removed from air to other compartments by rain dissolution and wet deposition. The concentration in agricultural soil drops gradually by soil runoff and erosion processes, transporting 2,4-D to freshwater environment. The concentration of 2,4-D in marine is transported from freshwater through advection flows. 2,4-D is removed at a slower rate in freshwater, marine, and their sediments.

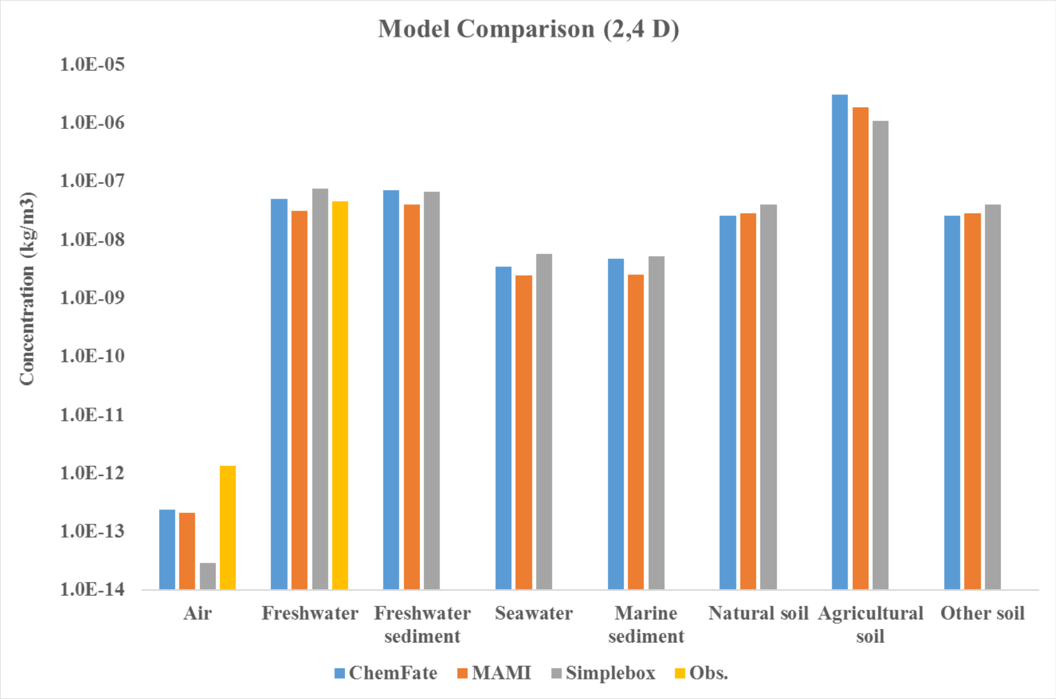


Fig 5. Model results comparison for 2,4-D. The simulation results from ionOFate are at the end of the 10-year simulation time and the median values are selected as observation data for air and freshwater.

Figure 4 shows the predicted chemical concentrations calculated by ionOFate at the end of 10-year's simulation time, compared with the results obtained from MAMI, Simplebox and observation data. The concentrations predicted by ionOFate and by MAMI are almost identical for air, natural soil, and other soil, and very similar to other compartments. The predictions from Simplebox are also similar to those from ionOFate and MAMI, except for the concentration in air compartment. Simplebox underestimates the concentration in air by two orders of magnitude, compared with the median measured concentration.

Model with Dynamic Kinetics. After confirming ionOFate's validity from the previous comparisons, we turned on ionOFate's dynamic capabilities to evaluate ionOFate by comparing the model's outputs with the range of observational data. Since we only have observational data for air and freshwater compartments, the comparisons are limited to those two compartments. Furthermore, because the observational data were collected from the Canadian Prairie Provinces that covered many large regional areas in Saskatchewan, Manitoba, British Columbia, Quebec, and Ontario over the time period 1989-2005 (Franco and Trapp, 2010b), building a climatic dataset that is specific to this region would be impossible. Therefore, we selected one of our default region New York and used its daily climatic data during 2005-2014 as a proxy to run ionOFate. The goal of the comparison is to demonstrate ionOFate's dynamic simulation capabilities with a more realistic climatic data.

The daily distributions of climatic data (i.e., temperature, wind speed, precipitation, and water flow) over the 10-years simulation time 2005-2014 are displayed in Figure S1.

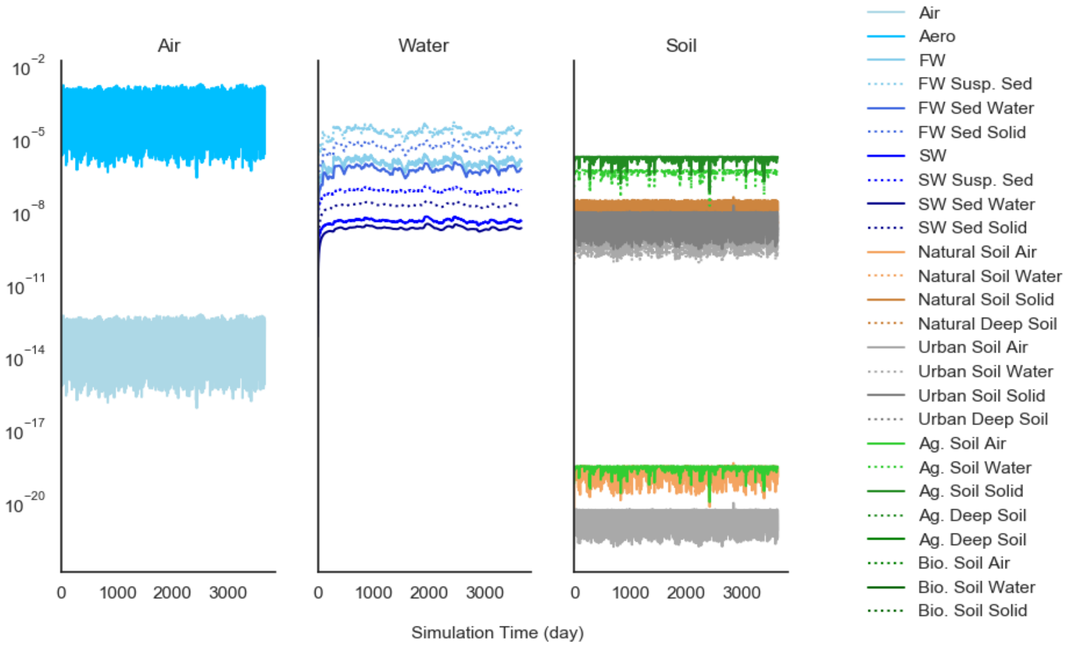
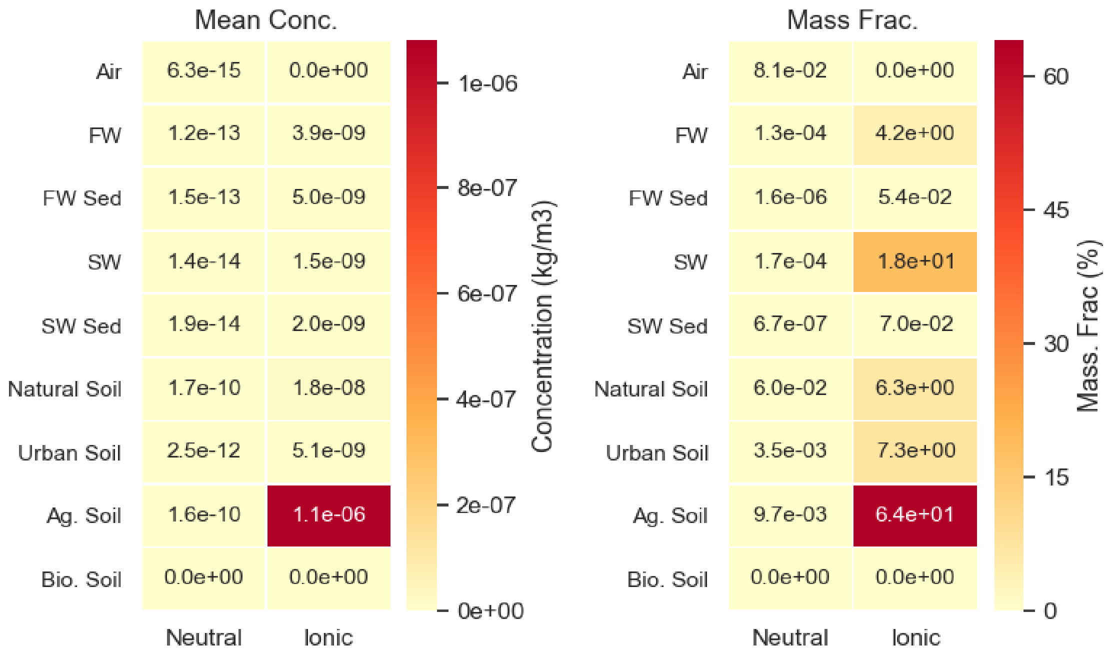


Fig 6. 2,4-D accumulation across environmental media, including air, water (freshwater and seawater), and soil (natural soil, urban soil, agricultural soil, and agriculture with biosolids application), under the release scenario of 21.22 kg/day to air and 509.28 kg/day to agricultural soil over ten years of model simulation with New York region climatic data.

Figure 5 shows the 2,4-D daily concentrations in environmental media of sub-compartments over ten years of model simulation. The highest concentrations can be found in aerosols and agricultural soil solids. The concentrations in freshwater and marine subcompartments are accumulated over the time. The substantial variability of 2,4-D concentrations in air and soil is mainly caused by the large fluctuations of environmental processes that involved wind speed and precipitation parameters, such as air/aerosol advection, aerosol wet deposition, soil runoff and soil erosion. The dominant species of 2,4-D is the anionic species, with over 99% in all compartments, except for air (Figure 6). The mean predicted concentrations of 2,4-D are the highest in agricultural soil, followed by natural soil, urban soil, and freshwater sediment. In terms of mass fraction by the end of model simulation, the highest mass are also associated with soils (Figure 7).

By combining the subcompartment concentrations in air and freshwater into bulk environmental concentrations, the boxplots in Figure 7 display the overall daily concentration distributions over ten years of model simulation in ionOFate, along with the range of concentrations from various field studies over years. The orange and purple dots show the outputs from MAMI and Simplebox. From Figure 7, we can see that ionOFate results capture most of the high variabilities of 2,4-D concentrations in both air and freshwater compartments, indicating a more realistic comparison between model outputs and observational data.



Mean Conc.

	Neutral	Ionic
Air	6.3e-15	0.0e+00
FW	1.2e-13	3.9e-09
FW Sed	1.5e-13	5.0e-09
SW	1.4e-14	1.5e-09
SW Sed	1.9e-14	2.0e-09
Natural Soil	1.7e-10	1.8e-08
Urban Soil	2.5e-12	5.1e-09
Ag. Soil	1.6e-10	1.1e-06
Bio. Soil	0.0e+00	0.0e+00

Mass Frac.

	Neutral	Ionic
Air	8.1e-02	0.0e+00
FW	1.3e-04	4.2e+00
FW Sed	1.6e-06	5.4e-02
SW	1.7e-04	1.8e+01
SW Sed	6.7e-07	7.0e-02
Natural Soil	6.0e-02	6.3e+00
Urban Soil	3.5e-03	7.3e+00
Ag. Soil	9.7e-03	6.4e+01
Bio. Soil	0.0e+00	0.0e+00

Fig 6. 2,4-D mean concentration over the 10-year simulation time in environmental bulk compartment for neutral and ionic forms (left) and mass fraction distribution at the end of 10-year simulation time.

Predicted Exposure Concentrations for metalFate. Ni was selected as a metal for the evaluation of metalFate to compare the model results with TRANSPECII and Simplebox. The models were applied to the Kelly Lake watershed described by Pearson et al. (1999) and Bhavsar et al. (2008) (Bhavsar et al., 2008; Pearson et al., 1999). Kelly Lake watershed is located in the Greater City of Sudbury, Ontario, Canada, and the soils surrounding Sudbury contain Ni due to local geology and atmospheric releases from mining operations (Bhavsar et al., 2008). Therefore, Ni has been transported to nearby water bodies through direct atmospheric deposition and soil runoff. The chemical input data and the environmental parameters used in the models are gathered from Bhavsar et al. (2008) and displayed in Table S4-S5 (Bhavsar et al., 2008). TRANSPECII considers two three compartments, water, sediment and soil. The sediment part comprises surficial (0-5 cm) and deeper (5-20 cm) and the soil part comprises surficial (0-5 cm) and lower (5-20 cm) soil layers. To facilitate the direct model comparison, metalFate compartments' existences and depths were adjusted accordingly. The emission scenario selected from Bhavsar et al. (2008) is 0.01 kg/day release to water, 17.5 kg/day release to water sediment, 1.9 kg/day release to surface soil, and 0.5 kg/day release to deep soil. Simplebox was not modeled in this case because the releases to water sediment and deep soil cannot be incorporated into the model. The water compartment concentration is the target media for model results evaluation and comparison with the observation data. Figure 7 shows the model outputs from metalFate and TRANSPECII and their results are very close. Both of the model outputs are within the observation data range of 0.4-0.8 kg/m^3.

D. Discussion

Model Application. The results from organoFate, ionOFate, and metalFate are comparable with other widely used models (i.e. EQC, MAMI, Simplebox, and TRANSPECII) under the same model parametrizations. The advantages of ChemFate are in the following ways. First, ChemFate provides a consistent modeling framework that allows the direct comparison of chemicals of radically different classes under the same environmental conditions. Second, the models in ChemFate are able to simulate time-dependent daily chemical concentrations with real regional climatic data. The fundamental determinant of concentrations is the emission rate (Sweetman et al., 2002), and ChemFate allows for more realistic emission inputs and variabilities. This is particularly important for chemicals that are used in seasonal trends or are released due to an accident spill. Environmental fluctuations and release fluctuations have the potential to cause short-term

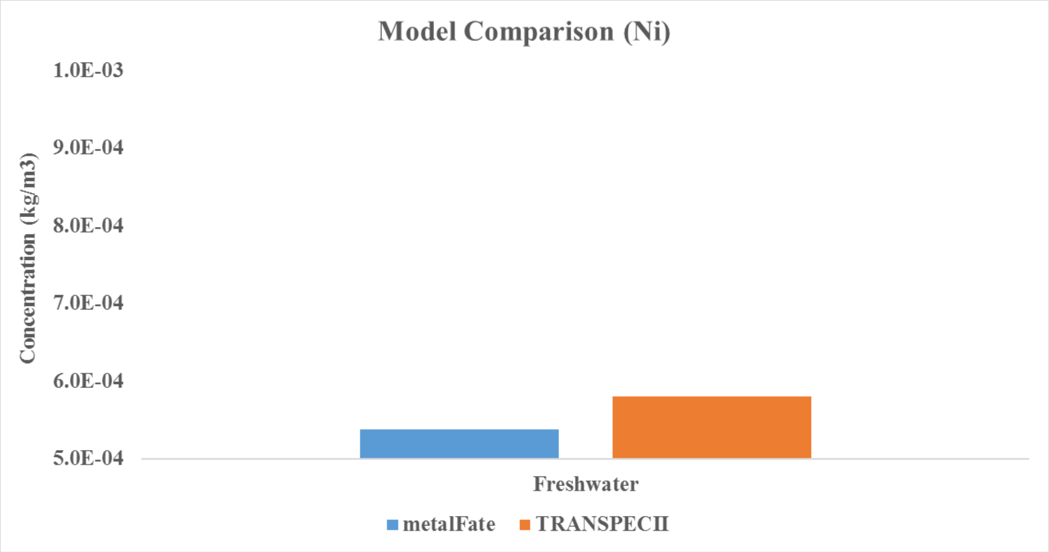


Fig 7. Model results comparison between metalFate and TRANSPECII in water compartment.

toxic effects (Garner et al., 2017), and ChemFate enables to capture the temporal risks of high contaminant concentrations, particularly to sensitive species in the environment (Parker and A. Keller, 2019). And the time-dependent concentrations also make it more useful to compare with monitoring data for validation purposes. Third, for ionizable organic chemicals and metals, ionOFate and metalFate not only give outputs for total chemical concentrations, they can also differentiate concentrations in different forms and phases. Fourth, ChemFate considers a wider range of processes, such as aerosol resuspension and transformation for ionic species, and use transport processes that consider the variations of climatic data instead of assuming constant rates, such as soil runoff and soil erosion. Lastly, ChemFate are coded in Python and it would be easily extensible to incorporate more environmental compartments and transport processes for future improvement. For general users, ChemFate has a user-friendly graphical user interface (GUI) to guide users to run the tool.

Model Limitations. For all multi-media fate and transport models, the general assumption of homogenous mixing is applied. And ChemFate assumes an instantaneously homogenous distribution of various chemical forms and phases within the same environmental compartment. For example, in a confined freshwater, metalFate assumes metals to be distributed among the dissolved, colloidal, and particulate phases right away based on their partition coefficients. This may not be true for all chemicals, but we have this assumption due to the lack of kinetics data to generate the rate of distribution (Bhavsar et al., 2008). Therefore, ChemFate does not consider the interactions of ionic species with other water solutes in ionOFate and metalFate.

ChemFate is designed to model a region with a combination of different environmental compartments, so it is not applied for a river network that connects several sub-compartments to simulate river reaches. Some other models and tools are available to simulate soil-water interactions with more in-depth hydrological modeling of the connected watershed network, such as SWAT (Neitsch et al., 2011) and WARMF (Chen et al., 2001). Furthermore, ChemFate only models one region at a time, and the advection transport out of air and costal marine compartments are considered as loss processes. This is different from global or large regional scale fate models that simulate chemicals transporting from one grid to another (MacLeod et al., 2001; Su et al., 2018; Toose et al., 2004).

Although we should always try to compare the model results with observational data in model evaluations, the observational data are rarely at the scale that can perfectly verify the models. The release scenarios we usually explore are at large scale, which makes it unfeasible to compare against the field observations directly. Despite this challenge, models still need to be continuously improved and validated with monitoring data as close as possible to avoid systematic errors because of omitted or poorly described processes (Mackay et al., 2009).

Since models are simplifications of a real world, they always embed uncertainties in themselves. Two primary types of uncertainty exist in ChemFate, which are model uncertainty and parameter uncertainty (Dubus et al., 2003; Hertwich et al., 2000; MacLeod et al., 2002). In ChemFate, the model uncertainty includes the transport and transformation processes involved in the model and the mathematical equations that are used to model each process. Even though the state-of-art or widely used equations for major processes are incorporated in ChemFate, the model uncertainty still exists and may be improved iteratively through constant model evaluation and calibration. The major source of parameter uncertainty in ChemFate come from the input parameters of physical-chemical properties and environmental parameters, which is usually addressed through Monte Carlo method (Hertwich et al., 2000; Luo and Yang, 2007).

E. Conclusion

Four multi-media fate and transport models are developed under the ChemFate framework to predict chemical time-dependent daily concentrations in environmental media: (1) organoFate, a model for non-ionizable organic chemicals, (2) ionOFate, a model for ionizable organic chemicals, (3) metalFate, a model for metals, and (4) nanoFate, a model for nanomaterials. ChemFate presents the most comprehensive capabilities that is able to not only provide predictions for four different classes of chemicals, but also incorporate dynamic emissions and dynamic environmental conditions. Model results from organoFate, ionOFate and metalFate are comparable with the widely adopted models (i.e. EQC, MAMI, Simplebox, and TRANSPECII) under the same model parametrizations. While the dynamic capability of ChemFate supports the model to simulate with real regional climatic data and produce better model performance that are more comparable to observational data. ChemFate aims to provide a consistent modeling framework that enables the direct comparisons of different classes of chemicals under the same environmental conditions. In short, ChemFate enables the use of more realistic release and climatic data inputs, and generate more realistic time-dependent chemical concentration outputs, for both chemical hazard assessment and risk assessment.

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IV. OrganoRelease – a framework for modeling the release of organic chemicals from the use and post-use of consumer products

A. Introduction

Concern over health risks of chemicals in consumer products has been increasing due to human exposure to chemicals released both indoors and outdoors (Trudel et al., 2008, 2011; Zota et al., 2014). Release of chemicals to the general environment from consumer products is also a concern for the health of the ecosystem especially for chemicals that are persistent and induce ecotoxicity, such as perfluorinated compounds, flame retardants, and certain antibiotics (Clarke and Smith, 2011; Ezechias et al., 2014; Janecko et al., 2016; Kunhikrishnan et al., 2015; Ortiz de Garcia et al., 2014; Rosal et al., 2010). As a result, chemicals contained in consumer products have become the focus of recent regulatory developments including the Safer Consumer Products Act of California, the Safe Chemical Act of the U.S., and the Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) program of the European Union.

A major challenge to minimizing the human and ecological health risk of chemicals in consumer products is the lack of available release information on the vast majority of chemicals and the economical infeasibility of obtaining relevant information for all chemicals through traditional experimental testing. To determine release estimates of chemicals from consumer products, one must first determine the plausible uses of a given chemical. Recently, Phillips et al., (2017) proposed a methodology to screen out candidate chemical alternatives based on functional use similarities and hazard information by combining quantitative structure-use relationship models and high-throughput toxicity screening. However, to assess the human and ecological health risk chemicals may pose, exposure as well as the fate and transport of the chemicals must be characterized, which requires estimates of the amount of chemical released from different products and applications. Traditional exposure assessments have been done for chemicals in consumer products (Aronson et al., 2007; Dann and Hontela, 2011; Goebel et al., 2012; Kienhuis et al., 2015). But they rely on measuring the concentrations of chemicals in different environmental media, which would be cost prohibitive for evaluating thousands of chemicals in consumer products. Fate and transport models for both indoor and outdoor environments can be used to estimate the concentrations of the chemicals of interest (Garner et al., 2017; Hollander et al., 2016; Liagkouridis et al., 2015; Rosenbaum et al., 2011). However, a vital input for these models is the release information of the chemical(s) to different environmental compartments during and after the use of products that contain the chemical(s).

Holmgren et al., (2012) presented a generic emission model for organic chemicals embedded in solid materials but this model is rather mathematically complex with dozens of parameters for each chemical, the shape of the consumer product, and the room characteristics. A less complicated and parsimonious model was recently developed for the release of volatile organic compounds encapsulated in products (Huang and Jolliet, 2016a) but is limited to volatile organic compounds. Furthermore, both models (Holmgren et al., 2012; Huang and Jolliet, 2016a) provide release estimates only for indoor air. Various models to estimate the release of different types of nanoparticles to multiple environmental compartments during the life cycle of various products have been published (Gottschalk et al., 2010; Gottschalk and Nowack, 2011; Keller et al., 2013, 2014; Keller and Lazareva, 2013; Sun et al., 2016). However, such studies do not address the release of organic chemicals, which make up the majority of both existing and new chemicals.

Given the current status of research, a systematic methodology for quantitatively estimating the release of organic chemicals contained in a wide range of consumer products is needed as input to models used to characterize potential human and ecosystem exposure to chemicals. Here we present the OrganoRelease framework, a methodology to estimate the distribution (as a fraction of the total amount used) of the release of a chemical to different environmental compartments during the use and post-use phases of consumer products, when product-specific chemical release measurements are lacking. Post-use phase is when chemicals are being transported to wastewater treatment plants or waste treatment plants after the direct use of consumer products. OrganoRelease was designed to accommodate different levels of data availability for any given organic chemical, with the minimum required input being the chemical structure alone. To achieve this goal, in OrganoRelease we connect the chemical's structure (e.g., topological, physicochemical properties, etc.), possible functional uses which are grouped based on the chemical's primary function (e.g., surfactant, solvent, etc.), potential product categories (e.g., cosmetics, paints, etc.), and release factors (as fractions of the chemical of interest released to indoor or outdoor air, wastewater, soil, and waste during use and post-use phases). OrganoRelease can provide rapid screening-level estimates of the release of chemicals contained in a range of consumer products during the use and post-use phases.

B. Materials and Methods

OrganoRelease estimates the distribution (in percent) of the release of a chemical directly released to indoor air, outdoor air, wastewater, soil, and waste, without considering any post-release fate and transport processes. OrganoRelease consists of three components that connect the functional use (defined as the chemical categories grouped based on their primary function), product category, and release factors together: 1) a Chemical Functional Use Classifier that estimates the chemical's functional use if not known; 2) market share data that links the functional uses with different product categories to quantify the mass fraction of chemical(s) entering corresponding product streams; and 3) release factors, based on functional uses and product categories, to estimate the initial release of the chemical(s) in products to the environment. The conceptual framework of OrganoRelease is shown in Figure 1.

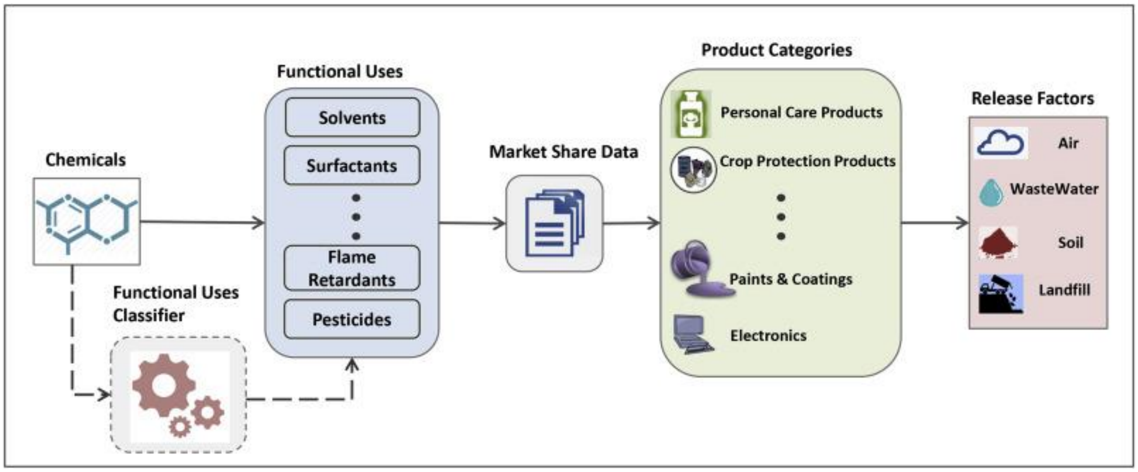

Fig. 1. Conceptual framework of OrganoRelease

Chemical Functional Use Classifier. An artificial neural network (ANN) was employed to develop a Chemical Functional Use Classifier ("Classifier" for short) that estimates chemical functional uses such as solvents and surfactants based on the molecular structure of the chemical. The molecular structure information is represented by the molecular descriptors, including constitutional, topological, chemical properties, and many other descriptor blocks (Todeschini and Consonni, 2009). ANNs serve as a nonlinear, universal approximation model to extract the intrinsic knowledge within a large amount of data (Hornik et al., 1989).

Pairs of chemical-functional use data points were collected from Chemical Book (ChemicalBook, 2016) based on chemical functional uses with available market share data.

This included nine functional uses: aerosol propellants, antibacterial agents, flame retardants, flavors and fragrances, solvents, surfactants, fungicides, herbicides, and insecticides, which are the major functional uses in consumer products. This is to ensure a seamless connection between estimating product use categories and functional uses. Since the molecular descriptors can only be computed for organic compounds, we removed mixtures, inorganics, salts, and organometallics. After the data curation process, we had a total of 2,900 pairs of chemical-functional use to build the Classifier.

Dragon 7 (Dragon 7.0, 2016) was used to generate over 4,000 molecular descriptors for each chemical, including constitutional, topological, chemical properties, and many other descriptor blocks. In order to reduce the number of molecular descriptors to the recommended ratio of number of entries to number of variables for quantitative structure-activity relationship (QSAR) models (Dearden et al., 2009), a filter-based feature selection algorithm was performed to remove the molecular descriptors that have a variance lower than 15 or a correlation coefficient higher than 0.65 with other descriptors (Dutta et al., 2007; Gramatica, 2007).

The collected dataset was randomly split into training, validation and test datasets. For each functional use category, 20 chemicals were first randomly selected as test dataset. For the rest of the data points within each functional use, 85% were randomly chosen as training set and 15% as validation set. The ANN models were built based on the training set, and the validation set was used to select the best fit model. The test set was applied to evaluate the performance of the final model. The Classifier was built in Python, using the Scikit-learn package (Pedregosa et al., 2011) for dataset random selection and final model evaluation. TensorFlow (Abadi et al., 2016) was implemented to build neural network models. A combination of grid search and random search methods were performed for hyperparameter optimization to optimize the set of parameters in the learning algorithm as well as avoid overfitting (Bergstra et al., 2013). Figure 2 illustrates the conceptual diagram of the Classifier building process.

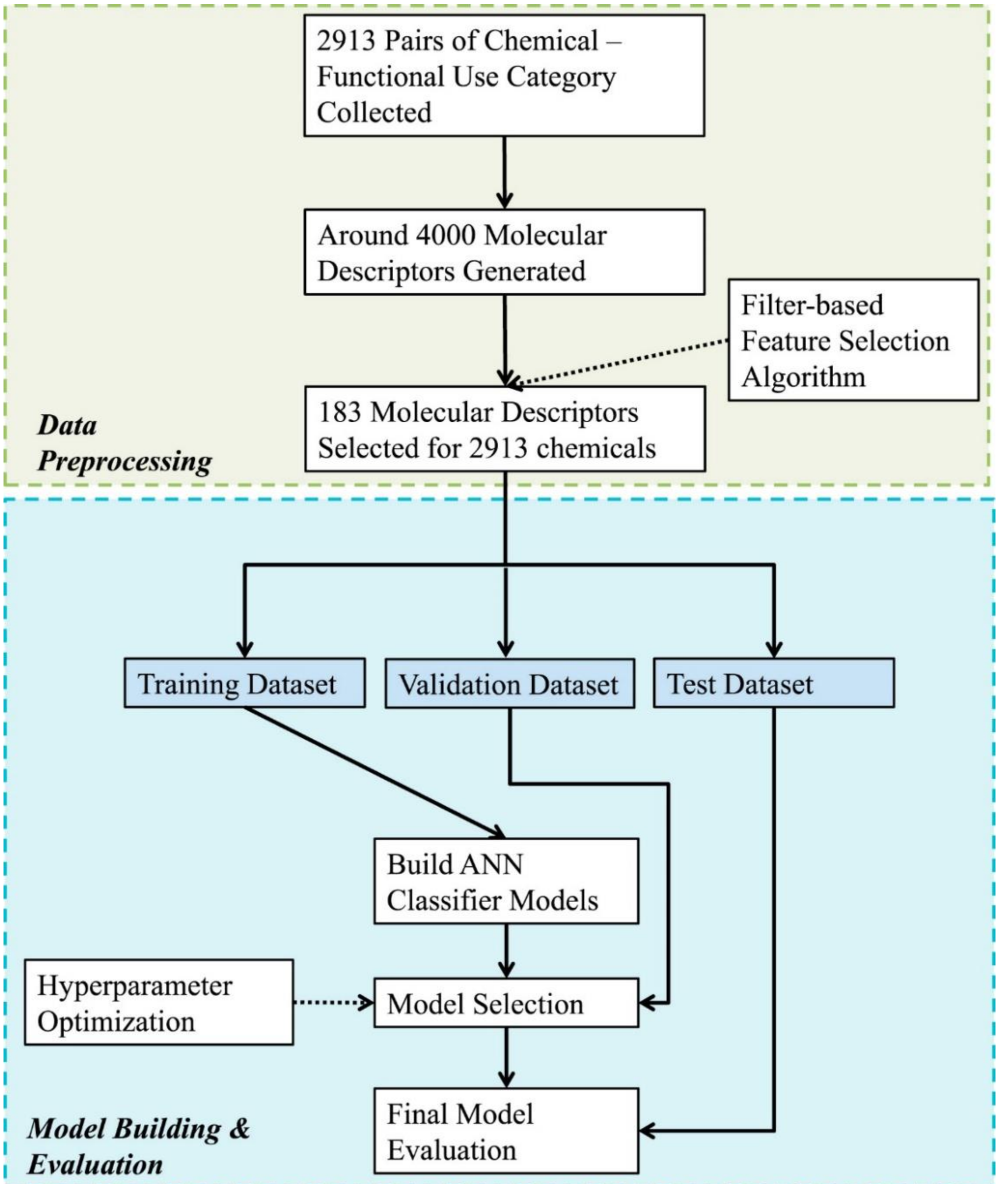

Fig. 2. Conceptual diagram of Chemical Functional Use Classifier module building process.

The performance of the Classifier is measured by its precision, recall, and F1 score, which are defined in Equations (1) – (3). Precision indicates the ability of the Classifier to not label a negative sample as positive, while recall implies the ability of the Classifier to find all positive samples (Powers, 2011). F1 score represents a weighted harmonic mean of precision and recall. All three metrics range from 0 to 1; values closer to 1 indicate better model performance.

$$\text{Precision} = \frac{\text{True Positive}}{\text{True Positive+False Positive}} \qquad (1)$$

$$\text{Recall} = \frac{\text{True Positive}}{\text{True Positive+False Negative}} \qquad (2)$$

$$\text{F1} = 2 \times \frac{\text{Precision}\times\text{Recall}}{\text{Precision+Recall}} \qquad (3)$$

Market share data. The use of market share data offers an approach to quantify the percentages of the functional use(s) of the chemical of interest entering the corresponding product categories. Grand View Research (Grand View Research Inc, 2016) (GVR) and The Freedonia Group (Freedonia Group, 2016) provided market research information on global market volumes (in tons) by application, including aerosol propellants, antibacterials, flame retardants, flavors and fragrances, pesticides, solvents, and surfactants. The three types of pesticides (fungicides, insecticides, herbicides) were combined into agricultural chemicals, since the release factors, even for consumer use, are not differentiated in the database developed by the European Crop Protection Agency (ECPA, 2013). The release category of crop protection products (Table S-1) also includes the use of the three types of pesticides by consumers in residences and commercial operations. The market volumes were transformed into percentages to capture the distribution of product applications for each chemical functional use. The original market volume data was developed by the consulting companies using interviews with suppliers, distributors as well as buyers. A certain level of uncertainty was associated with the market share estimates. To capture the uncertainty, we assumed a variation of ±10% in the market share percentage estimates based on our communications with the market research companies. We transformed the market volume data into market fractions with an uncertainty range.

Release factor data sources. Release factors to environmental compartments (indoor air, outdoor air, wastewater and soil) for chemicals in different products were gathered from the European Union (EU) specific environmental release categories (SPERCs) (Sättler et al., 2012). The scenario specific SPERCs were developed by a variety of European trade groups and sector organizations, intending to provide realistic yet conservative default release estimates, considering narrower use scenarios and good operational practices. SPERC-based emission estimates have been incorporated into several environmental exposure assessment tools (CEFIC, 2012), such as CHESAR (CHEmical Safety Assessment and Reporting Tool) (ECHA, 2016), ECETOC TRA (European Center for Ecotoxicology and Toxicology of Chemicals Targeted Risk Assessment) (ECETOC, 2014), and EasyTRA (Jansen-Systems, 2014). Since we are only interested in consumer products used in wide dispersive use (also called consumer use) or professional use, 64 out of 190 SPERCs files were reviewed and extracted to use as release factors, which are shown in Table S-1 in the Supplemental Information (SI). The industry sector groups and their corresponding websites are shown in Table S-2 in the SI. The default release factors in SPERCs consider the application type (e.g., down the drain, spray), use scenarios (e.g., indoor or outdoor use), and physicochemical properties, specifically vapor pressures for nine product sub-categories and water solubility for one product category (SI Table S-1). Among the nine product sub-categories that consider vapor pressures, four have varying release factors based on tiered vapor pressure values while the other broadly consider whether the chemical is solid or volatile. Most of the products in these categories are used indoors, and would thus result in an indoor air release. Some categories may be used both indoors and outdoors, such as paints and cleaning products. Another small number of categories, such as pesticides and fertilizers, are mainly used outdoor, leading to an outdoor air release. Therefore, the release factors to air could be assigned to either indoor or outdoor air depending on the use scenarios.

Another major data source for release factor estimates was the A-table of the European Union Technical Guidance Documents (EU TGD), specifically, under industrial category number 5 for “personal/domestic use”, which considers the private use phase in a chemical life cycle (European Commission, 2003; Vermeire et al., 2005). There are 55 use categories, and category 55 is designated as “Other” uses (SI Table S-3). Three categories in EU TGD have varying release factors based on the chemical’s water solubility and six categories based on vapor pressure. Most of these functional use categories result in release to indoor air since these consumer products are used indoors, with the exception of fertilizers and pesticides.

Since most of the release factors in SPERCs take into account the values from EU TGD, measured release data, market research data, and expert judgement via questionnaires (CEFIC, 2012; Sättler et al., 2012; Umweltbundesamt, 2011), OrganoRelease gives top priority for the default values in SPERCs. When release factors for certain product categories are not available, the default values in EU TGDs are used. For the fraction of chemical that is not released into air, wastewater, or soil, we assume it would be disposed as waste at the post-use phase. OrganoRelease does not consider additional potential releases from waste management (e.g. landfill leachate).

Framework workflow. The workflow of OrganoRelease is shown in Figure 3. Based on different levels of knowledge available for a given the chemical, the framework provides different paths to assess the potential environmental release of a chemical during the use and post-use phase of various consumer products. These release factors can be directly applied to chemicals for which the consumer product categories and use amounts (mass) are known (Path 1). When a chemical's product categories are uncertain but the likely functional uses are known, which is often the case for upstream chemical producers, two paths can be applied depending on the availability of market share data for those functional uses. If the market share data is available, it will be employed to match the functional use with the product use categories (Path 2a). If the market share data for a certain functional use is not available, the release factors will be generated solely based on functional use (Path 2b). For a chemical with both product categories and functional uses unknown, the Chemical Functional Use Classifier will be used to predict the most probable functional use. The release factors will then be assigned based on the predicted functional use and the availability of market share data (Path 3a if market share data available and Path 3b if not).

The framework workflow can be mathematically represented in Equations (4) and (5) as shown below:

Path 1:

$$M_k = M_{use}(\sum_j F_{j,k} \times \varphi_j) \tag{4}$$

Path 2a/b and Path 3a/b:

$$M_k = M_{use}(\sum_i \sum_j F_{i,j,k} \times \varphi_{i,j} \times \Phi_i) \tag{5}$$

where M_k is the amount of chemical release into a compartment k (indoor air, outdoor air, wastewater, soil, or waste), M_{use} is the amount of chemical consumed in the use phase, $F_{i,j,k}$ is the release factor to compartment *k* of the chemical used in product category *j* of functional use *i*, Φ_i is the fraction of chemical used in functional use *i*, and $\varphi_{i,j}$ is the fraction of chemical used in product category *j* of functional use *i*. OrganoRelease can only provide the release fractions of a chemical to various environmental compartments. To calculate the total amount of a chemical used (M_{use} in kg) in a particular location, country or globally, the user needs external data. For certain chemicals, production amount may serve as a surrogate of use as shown in a recent study (Shin et al., 2015).

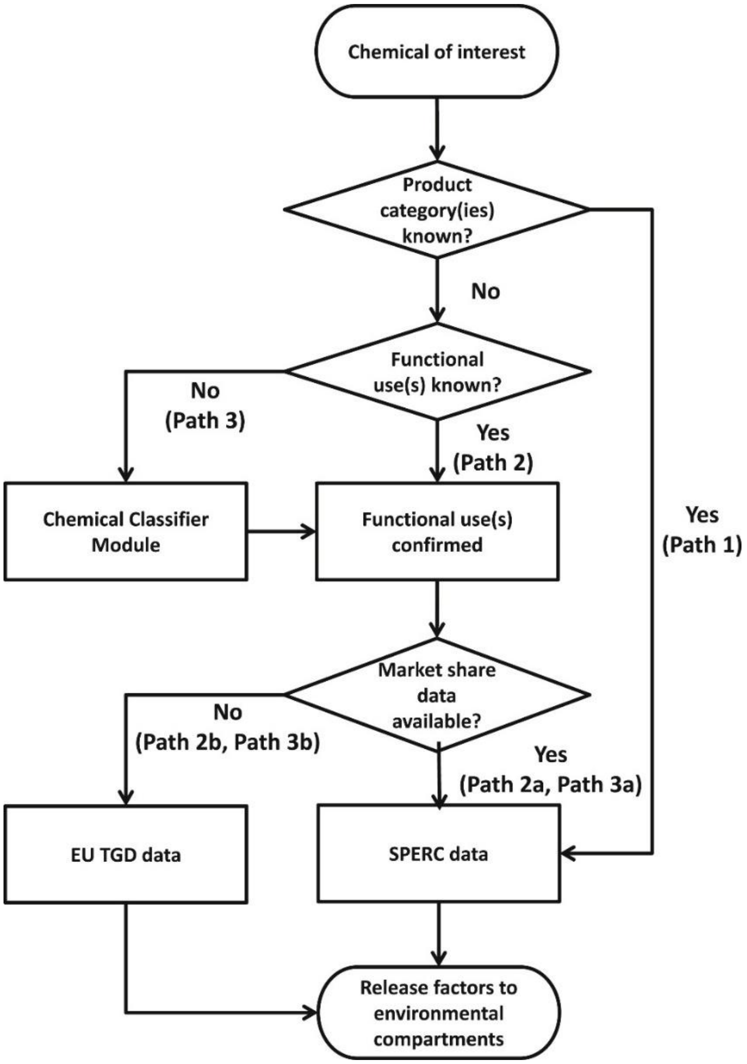

Fig. 3. Workflow of OrganoRelease, depicting the various paths depending on the level of known information available for the chemical(s) of interest. The diamond symbol represents a decision that needs to be made and the rectangle indicates a process to identify the release factors.

We developed a case study using tertiary-butyl acetate (TBAC) to demonstrate the workflow of the OrganoRelease framework. TBAC is a commonly used solvent with wide applications such as paints, adhesives and cleaners (Geiser, 2015), listed by US EPA as a high production volume chemical with 5000-25000 metric tons produced in 2015 (EPA, 2017). Since TBAC is not within our Classifier's training dataset, we used the Classifier to predict its functional use to demonstrate Paths 3a/b. Paths 1 and 2a/b were also considered in the demonstration.

C. Results

3.1. Functional uses and product categories considered

Table 1 presents the functional uses included in the Classifier, with market share data, or with default release factors in EU TGD. Two lists of product categories with default release factors in EU TGD and SPERC are also shown in Table 1. The release factors for the "Others" category in EU TGD depend solely on the chemical's water solubility and vapor pressure. Currently, the Classifier can estimate nine functional uses and market share data is available for seven. Note that the Classifier was developed based on the highest quality of data we obtained and therefore it differentiated pesticides into fungicides, herbicides, and insecticides. However, these three functional uses in the Classifier are indifferently matched to pesticides in the market share data since such differentiation was not available for the market share data.

All currently published SPERCs that are relevant to consumer products were collected from eight industrial sectors (SI Table S-1). Even though the EU TGD contains 55 categories, only 15 functional uses and 5 product categories with quantitative release information were available, including the category of "Others", which covers anything outside the categories documented in EU TGD.

Table 4. Functional uses and product categories in different components of OrganoRelease.

Data Source	Type	Number of Types	Description
Classifier	Functional Use	9	Aerosol Propellants, Antibacterials, Flame Retardants, Flavors and Fragrances, Fungicides, Herbicides, Insecticides, Solvents, Surfactants
Market Share Data	Functional Use	7	Aerosol Propellants, Antibacterials, Flame Retardants, Flavors and Fragrances, Pesticides (includes Fungicides, Herbicides, Insecticides), Solvents, Surfactants
EU TGD	Functional Use	15	Aerosol Propellants, Anti-freezing Agents, Anti-static Agents, Bleaching Agents, Cleaning Agents and Additives, Colorants, Complexing Agents, Corrosion Inhibitors, Fertilizers, Odor Agents, Pharmaceuticals, Softeners, Solvents, Surfactants, Others
	Product Category	5	Adhesives, Cosmetics, Food and Feedstuff Additives, Lubricants and Additives, Plant Protection Products (Agricultural)
SPERC	Product Category	13	Adhesives and Sealants, Coatings (Automotive), Coatings (Coal), Coatings and Inks and Artist Colors, Construction Chemicals, Crop Protection Products, Detergents and Cleaners, Metal Treatment Products, Hair and Skin Care Products, Lubricants and Greases, Paints and Coatings in Metal Packaging, Solvent in different products

3.2. Chemical Functional Use Classifier performance

The performance of the Chemical Functional Use Classifier on nine functional uses is presented in Table 2. The Classifier achieved both high precision and recall rate for aerosol propellants, flame retardants, flavors and fragrances, surfactants, and herbicides, with F1 scores over 0.89. For other functional uses the F1 scores were over 0.70 except for fungicides, which had an F1 score of 0.60. A detailed misclassification table of the Classifier on 180 test chemicals (20 test chemicals for each functional use) can be found in SI Table S-4. The lower precision and recall rates of fungicides and insecticides is partially due to the high structure similarity between fungicides and insecticides, which leads to 20% of fungicides being misclassified as insecticides and 20% of insecticides being misclassified as fungicides (SI Table S-4). An alternative model was also constructed by aggregating fungicides, herbicides, and insecticides into one category of pesticides while keeping other functional uses. The overall F1 score for the alternative model is 0.88. SI Table S-5 shows the detailed value of precision, recall, F1 score, and SI Table S-6 displays the misclassification information for each functional use in the alternative model.

Table 5. Performance of the Chemical Functional Use Classifier on nine functional uses

Functional Use	Number of Chemicals*	Precision	Recall	F1 Score	Number of Test Chemicals
Aerosol Propellants	68	1	0.9	0.95	20
Antibacterials	582	0.77	0.85	0.81	20
Flame Retardants	146	1	1	1	20
Flavors and Fragrances	488	0.86	0.95	0.9	20
Solvents	188	0.79	0.75	0.77	20
Surfactants	195	1	0.9	0.95	20
Fungicides	290	0.6	0.6	0.6	20
Herbicides	429	0.94	0.85	0.89	20
Insecticides	527	0.65	0.75	0.7	20
Average/Total		0.85	0.84	0.84	180

*Total number of chemicals considered in a given functional use for Classifier

Training data is crucial to develop the Classifier. The Classifier can incorporate additional functional uses and expand its coverage as more data becomes available. This will increase the overlap between functional uses that can be estimated by the Classifier and included in the EU TGD to improve the connections between these two components in OrganoRelease.

3.3. Market share data results

Market share data was compiled for nine major functional uses that are relevant to consumer products, including antibacterial agents, aerosol propellants, flavors and fragrances, flame retardants, insecticides, herbicides, fungicides, solvents, and surfactants (Freedonia Group, 2016; Grand View Research Inc, 2016). Insecticides, herbicides and fungicides were combined into pesticides, since their product category is the same, in terms of the release factors (i.e. release to air and soils, since it is assumed they are not released to wastewater). Table 3 shows the distribution structure of each functional use across the product categories along the year of the data was collected. In some cases, it is possible that the functional use is assigned to a primary product category and is not reflected in the final product category (e.g. solvents are used in paints, which may end up in automotive uses, but are not captured explicitly). As mentioned in the previous section of this paper, a factor of ±10% was considered to reflect the uncertainty in market shares. Under the scenario of unknown chemical product uses, the market share data provides valuable information on product flows for every available functional use, which can further improve the estimation accuracy of chemical releases. The coverage of functional uses and market share data can also be expanded and updated as additional information becomes available. At this stage our objective was to demonstrate proof-of-concept.

Table 6. Market fraction of functional uses across product categories[1].

Functional Use / Product Category	Aerosol Propellants[2]	Antibacterial Agents[2]	Flame Retardants[3]	Flavors and Fragrances [3]	Pesticides[2]	Solvents[2]	Surfactants[2]
Year of data	2014	2014	2013	2011	2014	2013	2014
Adhesives and Sealants						2.5% - 3.0%	
Agricultural Chemicals					100.0%		3.2% - 4.0%
Automotive			12.8% - 15.6%				
Construction Chemicals			24.2% - 29.5%				1.7% - 2.1%
Personal Care Products	33.8% - 41.4%	16.6% - 20.3%				2.5% - 3.0%	20.8% - 25.4%
Electronics			20.2% - 24.7%				
Emulsion Polymerization (plastics)							2.8% - 3.4%
Foods and Beverages	5.8% - 7.0%	21.4% - 26.2%		41.7% - 51.0%			3.7% - 4.5%
Home Care Products	30.5% - 37.3%						36.8% - 45.0%
Industrial and Institutional Cleaner				37.4% - 45.7%			4.4% - 5.4%
Oilfield Chemicals							3.3% - 4.1%
Others	3.1% - 3.7%		9.4% - 11.5%	10.9% - 13.3%		14.8% - 18.1%	9.3% - 11.3%
Paints and Coatings	14.0% - 17.1%	12.1% - 14.8%				54.8% - 67.0%	1.9% - 2.3%
Pharmaceuticals	2.9% - 3.5%					6.8% - 8.3%	
Printing Inks						8.6% - 10.6%	
Textiles			3.2% - 4.0%				2.0% - 2.4%
Water Treatment Chemicals		27.0% - 33.0%					
Wires and Cables			20.2% - 24.7%				
Wood Preservatives		12.9% - 15.8%					

1. Each column shows the fraction of each functional use distributed across the product categories based on the market volumes. Pesticides includes fungicides, insecticides and herbicides.

2. Compiled from Grand View Research Inc, 2016

3. Compiled from Freedonia Group, 2016

Release fraction selection. The release factor selection is based on varying degrees of knowledge for a given functional use and/or product category as described in the methods section. For the functional use and product category pairs with matching SPERC document available, release factors from SPERC documents are selected. For example, ESVOC SPERC 8.3c.v1 (uses in coatings: consumer) was chosen for solvents used in paints and coatings; ECPA SPERC 8d (crop protection products) was chosen for pesticides used as agricultural chemicals. If the product category (e.g., pharmaceuticals) for a chemical does not have a SPERC, release factors from EU TGD are selected to match the functional use (either known or estimated by the Classifier). Due to the spread in physicochemical properties (e.g. vapor pressure and water solubility), the release factors can vary across a wide range since these properties are considered in many of the SPERCs and EU TGDs. A comprehensive release factor selection table can be found in SI Table S-7. The release estimates may be applied to all regions, but wastewater collection rate and treatment efficiency in different regions (summarized in Keller and Lazareva, 2013) need to be considered to improve the accuracy of estimates for the fraction released to water. A wastewater treatment plant model, such as the Sewage Treatment Plant (STP) module in EPI Suite from US EPA, can be used to estimate the fractions of the chemical removed by various processes (e.g., biodegradation, and air stripping), released to the environment as effluent, or accumulated in biosolids (US EPA, 2012). The data from SPERC and EU TGD provides a starting point for estimating chemical releases, but product- or use-specific information should be used in OrganoRelease whenever available, to improve the accuracy of the estimates.

Case Study. Tertiary-butyl acetate (TBAC, CAS 540-88-5), a chemical not in the training dataset of the Chemical Functional Use Classifier, was selected to showcase the workflow of OrganoRelease. TBAC is generally used as a solvent in paints (Cooper et al., 2001). It has a solubility of 8.3 g/L in water (US EPA, 2012) and a vapor pressure of 47 mm Hg at 25 °C (Pubchem, 2017). For demonstration purposes of the various paths of the framework, different levels of knowledge of the chemical were assumed: 1) TBAC used as solvent (functional use) only in paints (product category) as known information (Path 1); 2) TBAC used as solvent (functional use) as the only known information (Path 2a); and 3) no known use information for TBAC available (Path 3a). Corresponding results are shown in Table 4. The diagram in Fig. 4 displays the mass flow of TBAC from solvent to product categories and releases to air, wastewater, soil, and waste in Path 2a.

The market share data of solvents was utilized to quantify the mass flow from solvent to various product categories. For each product category, the default release factors in the corresponding SPERC and EU TGD were selected to quantify the releases to various environmental compartments. In Table 4, the release factors for Path 2a are ranges instead of point values due to the assumed ±10% uncertainty in the market share data. In Path 3a, the Classifier correctly estimated TBAC to be a solvent, so the release factors are the same as in Path 2a. Path 2b and Path 3b are not applicable as the market share data of solvent is available. Those two paths would only be applied when the functional use does not find a match in the market share data.

Table 7. Release factors to various environmental compartments for TBAC assuming different levels of knowledge. Path 2a and Path 3a have taken into account of ±10% uncertainty in market share data.

Framework Path	Release Data Description	Release Factor to Environmental Compartment				
		Indoor Air	Outdoor Air	Wastewater	Soil	Waste
Path 1	ESVOC SPERC 8.3c.v1 (solvent uses in coatings: consumer)	98.50%	0%	1.00%	0.50%	0%

Path 2a	Solvent market share data; release factors from SPERC and EU TGD	79.0% - 96.5%	0%	5.3% - 6.4%	0.68% - 0.84%	5.1% - 6.2%
Path 2b	Not applicable (N/A)	N/A	N/A	N/A	N/A	N/A
Path 3a	The Classifier predicts the chemical to be solvent	79.0% - 96.5%	0%	5.3% - 6.4%	0.68% - 0.84%	5.1% - 6.2%
Path 3b	Not applicable (N/A)	N/A	N/A	N/A	N/A	N/A

Path 1 has the most accurate estimates, since it uses known information. There is uncertainty in the release factors for Path 1, given that these are generic values for all chemicals used as solvents in paints, but Path 1 serves as a basis for the comparison. In Path 2, since the only known information is the functional use, OrganoRelease considers possible product categories that use solvents and the corresponding SPERCs or TGDs, which results in a wider range of estimates and also a decrease in the estimated release to air, and a corresponding increase in release to wastewater (from treatment) or waste. For a given calculation, the sum of all releases must add up to 100% (as shown in Figure 4), but we present the range of predicted values. The increase in predicted release to wastewater and waste is mostly due to the consideration of the use of TBAC in pharmaceuticals, printing inks and others. If the user has additional information indicating that a chemical of interest cannot be used in those product categories, OrganoRelease can be re-run to obtain more accurate estimates.

In this case study, we explored the use of OrganoRelease for estimating chemical releases to various environmental compartments. Although more knowledge for the chemical of interest can reduce the uncertainty of the results, OrganoRelease is able to provide a starting point for the release estimations based on only the molecular structure of the chemical with the built-in Classifier and various databases that work in coordination.

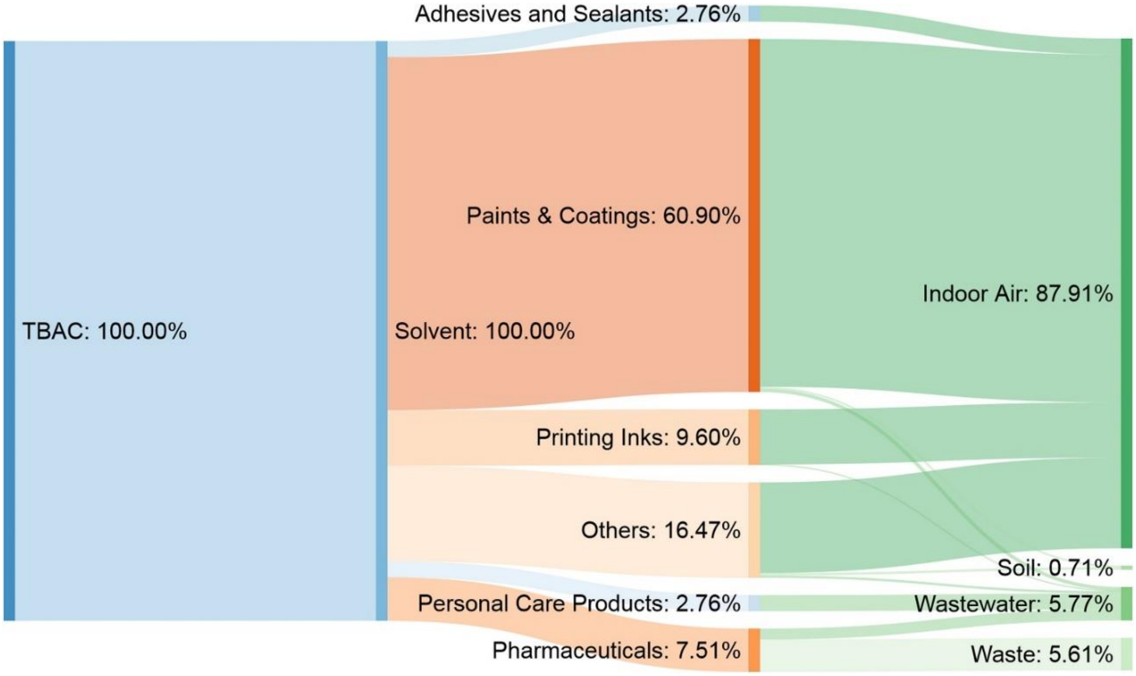

Fig. 4. Mass flow (in percent) of TBAC in Path 2a based on solvent market share data, using the midpoint of the range of values. The release factors for each product category are selected from SPERC and EU TGD.

D. Discussion

Applications of OrganoRelease. The quantitative range of release estimates generated by OrganoRelease can be used as input for other environmental models answering various questions. The first-tier release factors from OrganoRelease can be used with second tier models such as sewage treatment plant (STP) (US EPA, 2012), to obtain aggregated release information of chemicals to different environmental compartments and provide the necessary input data for fate and transport models (e.g., Brandes et al., 1996; Garner et al., 2017; Mackay and Paterson, 1991; Nijhof et al., 2016), to predict the environmental concentrations of the chemicals, which can then be used as input to evaluate potential damages to humans and ecosystems. If the product application is indoors (e.g. paints, application of cosmetics), the estimated release to the indoor air environment from OrganoRelease can be used as an input for indoor air models (Chaudhary and Hellweg, 2014; Wenger et al., 2012) to calculate human exposure and assess risks.

OrganoRelease can also make contributions to Life Cycle Assessment (LCA). LCA is a tool to assess the environmental and human health impacts of a product throughout its entire life cycle, from raw material extraction, manufacturing, use, to end of life (Azapagic, 1999; Gavankar et al., 2012; Hellweg and Canals, 2014). Although emissions of chemicals during the cradle-to-gate phases (i.e., from raw materials to completion of manufacturing) is systematically covered by existing life cycle inventory (LCI) databases (National Renewable Energy Laboratory, 2012; Wernet et al., 2016), there is great need for similar data on the release information for the use and post-use phases of chemicals in consumer products to complete LCA's holistic goal. By integrating OrganoRelease as input to current life cycle impact assessment (LCIA) methodologies, the challenge of data gaps in use and post-use releases of chemicals in consumer products can be addressed with information on the use amount of chemicals in consumer products. This can potentially help decision makers understand the relative contributions of impacts to human health and ecosystem quality from the use and post-use phases of consumer products in the context of their entire life cycle impacts. Coupled with regionalized socioeconomic data or detailed regional market share data for different product categories, this framework may provide insights on the different release factors to the environment based on the location where the products are consumed (Hodges et al., 2014; Keller and Lazareva, 2013). However, it should be noted that OrganoRelease cannot address near-field exposure related questions such as dermal contact and ingestion of leachate from packaging materials. Specialized exposure models such as PiF

(Jolliet et al., 2015) and SHEDS-HT (Isaacs et al., 2014) should be considered to for such exposure studies. In addition, the release factors estimated by OrganoRelease may be more conservative due to the methodologies of SPERC and EU TGD. This is in contrast with the average estimates of chemical emissions in current LCI databases. LCA practitioners should be mindful of this difference when applying OrganoRelease with other LCI databases.

Coverage of OrganoRelease and comparison with other studies. Although detailed information of the chemical/product combination is always preferred, OrganoRelease is applicable even when minimal information other than the chemical structure is available. The Chemical Functional Use Classifier is built using machine learning to increase the applicability of OrganoRelease as more information becomes available. Machine learning techniques have been employed recently to solve challenging problems regarding sustainability of chemicals and consumer products. Isaacs et al., (2016) developed classifier models to predict chemical functions and their weight fraction in personal care products using random forest. Using the same random forest technique, Phillips et al., (2017) built binary models to predict the functional uses of chemicals and combined this information with high-throughput toxicity screening results to identify alternatives to existing ingredients of concern within consumer products. The F1 score calculated for the Classifier measures the model’s accuracy of classification and hence is comparable to the complement of misclassification error reported in Phillips et al., (2017) , even when our Classifier is multiclass while the models developed by Phillips are binary. For the same functional uses covered in both studies (antibacterials, flame retardants, flavors and fragrances, solvents, and surfactants), the F1 scores of our Classifier are similar to the complements of misclassification errors from Phillips et al., (2017) indicating similar level of performance for these functional uses. Another distinction between the OrganoRelease classifier and that of Philips et al. is with regards to the functional uses covered. The Classifier in this model links with the market fraction data available in this study so that release information for a range of chemicals can be estimated. The binary models built by Phillips et al. would require market fraction data on several additional functional uses to be compatible with OrganoRelease. In addition, since the binary classifiers were built and trained with different data and features, the output from each classifier may not be directly comparable. OrganoRelease's Classifier uses the same features for estimating the probability of all functional uses, making each probability comparable before selecting the functional use with the highest probability as output.

The versatility of OrganoRelease, in terms of the amount of information needed to generate an estimate, can be compared with other studies on consumer products. Case studies on release of certain chemicals in specific products exist but cannot easily offer estimates for other chemical and products combinations (Quadros et al., 2013; van Wezel et al., 2016). Mechanistic diffusion models that estimate the release of chemicals from a product to indoor air are available but generally have high complexity and the limited availability of parameter values could hinder their application in screening risk or life-cycle assessments (Huang et al., 2017). The minimal requirements for OrganoRelease could be useful when information for a more complex model is not available.

The Consumer Exposure Model developed by US EPA requires information of the weight fractions of chemicals to complete estimates of release (US EPA, 2015). Obtaining this information can be challenging, as it is often proprietary knowledge with commercial value. The predictive model estimating the weight fractions of chemicals in consumer products developed by Isaacs et al., (2016) offers an opportunity to address this challenge. However, the Isaacs model is specialized in the relatively data-rich product category of personal care products and is not yet applicable to other products. The Chemical/Product Categories Database (CPCat) covers a much wider range of product categories, but the information it contains is rather general and includes all possible product applications where a known chemical may be found and all information is subject to the availability of chemicals already included in CPCat (Dionisio et al., 2015). In contrast, the Chemical Functional Use Classifier developed in this study allows OrganoRelease the ability to generate preliminary release estimates using mostly chemical structure (along with the available market fraction) thus being able to cover more chemicals.

The Chemical Functional Use Classifier developed in this study may also be useful to explore potential new functional uses of existing chemicals, using the “misclassification” results. This may also help in identifying unintended uses of chemicals and resulting consequences to human health and environment.

Limitations of OrganoRelease. The final results produced by OrganoRelease largely depend on the SPERC and EU TGD data. While offering a wide range of coverage over multiple product categories and functional uses with some considerations of physicochemical properties, SPERC and EU TGD data are not chemical specific but generic within each product category/functional use and should be interpreted as a preliminary estimate, useful when more specific information is absent. Isaacs et al., (2014) demonstrated product composition, amount of product used, and frequency of product used are among the most sensitive parameters in estimating human exposure in the use phase of consumer products. Thus, OrganoRelease should be used as a screening level tool for chemicals when there are major data limitations and not be used when relevant data can be obtained in more detailed or product oriented studies. It should be noted that although OrganoRelease seeks to provide accurate release estimates, some of the SPERCs or EU TGD could be conservative in nature, and may result in conservative release estimates.

Based on the EU TGD data, OrganoRelease considers dermal application of certain pharmaceuticals, in which case 25% is released to wastewater; if the pharmaceutical is ingested, the EU TGD consider that only 5% is released to wastewater. We assume that the balance of the chemical in a pharmaceutical is absorbed and metabolized. As indicated before, OrganoRelease is intended for a generic release estimate; if a manufacturer or user has better information on the application, it should be used instead. OrganoRelease does not account for the possible transformation (e.g. photodegradation) of a chemical during the use phase, which may be significant for some chemicals or applications. More specific models would be needed for those cases.

The market data gathered so far covers nine functional uses. Although these categories represent a wide range of applications in different consumer products, there are gaps in linking other functional uses with product categories. Thus, expanding the market data is among the top priorities for future development of OrganoRelease. In addition, several product categories (wood preservative, wires and cable, electronic, automotive, textile, etc.) in the collected market share data cannot be matched to the SPERC database. The OrganoRelease framework thus estimates the release factors based on the functional use, which is less specific. Depending on the information available, the user may opt to not consider certain product categories if additional (external) information (e.g. toxicity) can be used to determine that a chemical is not suitable for use in a given product category.

The Chemical Functional Use Classifier predicts the potential functional uses among the nine developed categories when no detail information is available. The functional uses that can be estimated are limited by the amount of training data we were able to collect. To improve the coverage of the Classifier more training data with more diverse functional uses are needed. However, the Classifier estimates only the functional use with highest probability. If the user considers only the "most probable" functional use, then that limits the release to the product categories associated with that functional use. If the user has additional information on functional uses, those functional uses can be selected to consider a broader range of product categories.

Although the uncertainty in classification and market fractions is quantifiable in OrganoRelease, we were not able to characterize uncertainty for the SPERC and EU TGD data, since that information is not available. The performance metrics of the Chemical Functional Use Classifier indicate the accuracy and uncertainty of its results. Without further insights, we refrain from assigning arbitrary uncertainty values for the SPERC and EU TGD data.

Finally, the nomenclatures for functional uses/product categories across the Chemical Functional Use Classifier, market share data, EU TGD data, and SPERC data are not always the same. Despite our best effort to match relevant nomenclatures in this framework, there may be discrepancies in connecting different data sources, which is a common challenge in studies on consumer products (Dionisio et al., 2015; Goldsmith et al., 2014).

E. Conclusion

This study presents the framework OrganoRelease to addresses the challenge of data availability of the release information for chemicals in consumer products during the use and post-use phases. We have developed a Chemical Functional Use Classifier and connected it with existing market share data and release factor estimates to cover 19 unique functional uses and 14 product categories. The versatility of OrganoRelease enables it to provide quantitative release estimates based mostly on chemical structure information, although better results can be obtained if market data is considered. The release estimates can be used as input for other models to assess the human exposure and ecological health risks of chemicals under different scenarios. OrganoRelease can be further expanded to cover more functional uses and product categories of chemicals by incorporating additional data as it becomes available.

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V. Conclusions

This dissertation aims to close the gap between the spatial resolutions of the models used in LCA and the biophysical processes relevant for understanding the environmental and human health impacts of chemicals. It covers three topics that collectively addresses the objectives: (1) measuring spatial variabilities in LCI, (2) modeling the fate of chemicals in the environment at the spatial resolution that matches with the underlying biophysical processes, (3) building a systematic release framework to estimate chemical releases that supports the fate modeling.

In Chapter 2, I demonstrated that spatial disparities in state-specific LCI for four major crops in the USA can lead to two to fourfold differences in characterized results for most impact categories. The differences, however, increase to over an order of magnitude for freshwater ecotoxicity and human health non-cancer. As a result, the use of national average data derived from top corn and soybean producing states significantly underestimates the characterized impacts of corn and soybean in the states where land conversion from cotton to corn or soybean actually took place.

In Chapter 3, I developed a spatially explicit and time-dependent multimedia fate modeling framework, ChemFate, that can be incorporated into regional LCIA. ChemFate consists of four multimedia fate models: (1) organoFate, a model for non-ionizable organic chemicals, (2) ionOFate, a model for ionizable organic chemicals, (3) metalFate, a model for metals, and (4) nanoFate, a model for nanomaterials. ChemFate is able to not only provide predictions for four different classes of chemicals, but also incorporate dynamic emissions and dynamic environmental conditions. The dynamic capability of ChemFate supports the model to simulate with real regional climatic data and produce better model performance.

In Chapter 4, I built a comprehensive release framework, OrganoRelease, to estimate the release of organic chemicals from the use and post-use of consumer products with limited information. OrganoRelease connects 19 unique functional uses and 14 product categories across 4 data sources and provides multiple pathways for chemical release estimation. The results can be used as input for methods estimating environmental fate and exposure.