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**Advancing emission and impact modeling for
agricultural pesticides under tropical conditions, to
improve scientific foundation of the environmental
evaluation of tropical agri-food systems**

Céline Gentil-Sergent

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THÈSE POUR OBTENIR LE GRADE DE DOCTEUR DE MONTPELLIER SUPAGRO

En Sciences Agronomiques

École doctorale GAIA

Portée par

Unité de recherche HortSys du CIRAD

ADVANCING EMISSION AND IMPACT MODELING FOR AGRICULTURAL
PESTICIDES UNDER TROPICAL CONDITIONS, TO IMPROVE SCIENTIFIC
FOUNDATION OF THE ENVIRONMENTAL EVALUATION OF TROPICAL
AGRI-FOOD SYSTEMS

Présentée par Céline Gentil

Le 14 décembre 2020

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UBUNTU

PREFACE

This thesis work was co-supervised by Claudine Basset-Mens from CIRAD (The French Agricultural Research Centre for International Development) and by Peter Fantke from DTU (Danish Technical University).

The thesis was financially supported by CIRAD and ADEME of Martinique (the French Environment and Energy Management Agency) through the InnovACV project (Life Cycle Assessment (LCA) for innovation in cropping systems in Martinique, towards a better consideration of the impacts of pesticides). It benefited from two internship grants through the Rivage project funded by Martinique's European Regional Development Fund (MQ0003772-CIRAD). This thesis was also part of the OLCA-Pest project (Operationalizing Life Cycle Assessment of pesticides), an international project to build on current advances in life cycle inventory and life cycle impact assessment to operationalize and harmonize the emission quantification and impact characterization of pesticides in Life Cycle Assessment and product environmental footprinting.

This thesis was carried out at CIRAD on the CAEC (Campus Agro-Environnemental Caraïbes) site in Martinique.

ABSTRACT

The use of pesticides in agriculture leads to environmental and human health impacts, particularly in tropical regions where pedoclimatic conditions favorable to pests and diseases, encourage their use all year round. Life Cycle Assessment (LCA), an ISO-standardized methodology is widely applied to quantify the environmental performance of agri-food systems but generally do not account properly for impacts due to pesticide applications. A bibliographic review identified the characteristics that determine pesticide emissions and related impacts under tropical conditions, and assessed to what extent Life Cycle Inventory (LCI) and Life Cycle Impact Assessment (LCIA) models need to be adapted to better account for these conditions. The main goal of the thesis was to advance emission and impact modeling for agricultural pesticides under tropical conditions for improving scientific foundation of the environmental evaluation of tropical agri-food systems. First, we identified with a sensitivity analysis on the pesticide emission model PestLCI Consensus, aspects that needed be developed in priority. Then, the initial emission distribution was refined by developing a consistent set of foliar interception fractions as function of crop characteristics and spraying techniques for crops grown under tropical conditions. In addition, based on a literature review, measures of drift from application methods were extracted from experimentations specifically conducted in tropical conditions. Thanks to these developments, a consistent set of pesticide emission fractions were provided for direct use by LCA practitioners. A consistent coupling of pesticide LCI-LCIA models was proposed for all agricultural contexts including human toxicity due to exposure to pesticide residues in crops. The proposition was tested successfully on an LCA of an open-field tomato produced in Martinique (French West Indies) by parametrizing the model to the local conditions. Moreover, to evaluate the environmental sustainability of agricultural practices a framework was proposed to account for ground cover management in the modeling of pesticide emissions. These methodological, scientific and operational advances constitute an important step toward a more reliable modeling of field pesticide emissions, including emission to the consumed part of the crop, and their consistent conversion into (eco)-toxicity impacts, as part of LCA for agri-food systems in tropical regions. To improve further the assessment of agricultural pesticides in LCA, especially for tropical conditions, a better consideration of rain-related processes in pesticide emission model is needed.

Keywords: Active ingredient, models, tropical crops, farming practices, (eco)-toxicity, LCA

RESUME

L'utilisation des pesticides en agriculture a des impacts sur l'environnement et la santé humaine, en particulier dans les régions tropicales où les conditions pédoclimatiques favorables aux maladies et aux bio-agresseurs, motivent leur utilisation tout au long de l'année. L'Analyse du Cycle de Vie (ACV), une méthodologie normalisée par l'ISO, est largement utilisée pour quantifier la performance environnementale des systèmes agroalimentaires mais la prise en compte des impacts liés aux pesticides y représente un défi majeur. Une analyse bibliographique a permis d'identifier les caractéristiques qui déterminent les émissions de pesticides et leurs impacts associés dans les conditions tropicales, et d'évaluer dans quelle mesure les modèles d'inventaire et d'évaluation de l'impact doivent être adaptés pour mieux tenir compte de ces conditions. Le principal objectif de cette thèse était d'aller vers une meilleure modélisation des émissions et impacts des pesticides agricoles dans les conditions tropicales afin d'améliorer les bases scientifiques de l'évaluation environnementale des systèmes agroalimentaires tropicaux. Tout d'abord, grâce à une analyse de sensibilité du modèle d'émission des pesticides PestLCI Consensus, nous avons identifié les aspects qui devraient être développés en priorité. Ensuite, la distribution initiale des émissions de pesticides a été affinée en développant un ensemble cohérent de fractions d'interception foliaire en fonction des caractéristiques de la culture et des techniques de pulvérisation pour les cultures tropicales. De plus, des mesures de la dérive due aux méthodes d'application utilisées ont été extraites d'expérimentation spécifiquement menées dans des conditions tropicales. Ainsi, un jeu de fractions d'émission de pesticides a été produit pour une utilisation directe par les praticiens de l'ACV. Un couplage cohérent des modèles d'émission et d'impact des pesticides a été proposé pour tous les contextes agricoles, incluant la toxicité humaine due à l'exposition aux résidus de pesticides dans les produits récoltés. La proposition a été testée de façon satisfaisante sur l'ACV de la production de tomate plein champ en Martinique (Antilles françaises) en paramétrant le modèle aux conditions locales. De plus, pour évaluer la durabilité environnementale des pratiques agricoles, la modélisation de la gestion de la couverture du sol dans le modèle d'émissions des pesticides a également été proposée. Ces progrès méthodologiques, scientifiques et opérationnels constituent une étape importante vers une modélisation plus fiable des émissions au champ de pesticides, y compris l'émission dans la partie consommée de la culture, et leur conversion cohérente en impacts d'(éco)toxicité, dans le cadre de l'ACV des systèmes agroalimentaires des régions tropicales. Pour améliorer encore l'évaluation des pesticides agricoles dans le cadre de l'ACV, en particulier pour les conditions tropicales, il est nécessaire de mieux prendre en compte les processus liés à la pluie dans le modèle d'émission des pesticides.

Mot clés : substance active, modèles, cultures tropicales, pratiques agricoles, (éco-)toxicité, ACV

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INTRODUCTION

Why and how to assess pesticides for agri-LCA in tropical regions?

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State-of-the-art of pesticide LCI-LCIA models

Challenges and ways forward in pesticide emission and toxicity characterization modeling for tropical conditions

Thesis research questions & objectives

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Sensitivity of LCI model

New insights for PestLCI Consensus model in tropical conditions with a sensitivity analysis

CHAPTER 3
LCI-LCIA chain
Case study

Coupling pesticide emission and toxicity characterization models for LCA:
Application to open-field tomato production in Martinique

CHAPTER 4
LCI initial distribution

Quantifying pesticide emission fractions for tropical conditions

CHAPTER 5
Farming practices

Introducing ground cover management in pesticide emission modeling

CHAPTER 6

Discussion & Outlook

CONCLUSION

General conclusion



STRUCTURE OF THE THESIS

A brief **introduction** presents the thesis scientific context and the importance of modeling pesticide emissions and associated impacts on crops grown in tropical conditions through the Life Cycle Assessment (LCA) approach.

First, a state-of-the art of pesticide Life Cycle Inventory (LCI) and Life Cycle Impact Assessment (LCIA) models regarding their consideration of tropical conditions introduces the thesis work, and first recommendations of possible model refinements are proposed (**Chapter 1** – article published in Int J Life Cycle Assess).

This first chapter allows detailing the **research questions** and related **objectives** of the thesis and provide a summary of challenges and ways forward to adapt LCI and LCIA models to tropical conditions.

Second, the sensitivity of the pesticide emission model's outputs (i.e. PestLCI Consensus) and the related (eco-)toxicity impacts to the pesticide emission model's inputs is analyzed and the most important inputs and processes on which focusing adaptations to tropical conditions are identified (**Chapter 2** – presented as an article).

Third, the coupling of the pesticide emission and impact models is proposed and illustrated with a case study on tomato production in Martinique (**Chapter 3** – article published in J Clean Prod).

Four, the development of a set of crop growth stages and associated foliar interception fractions for crops grown under tropical conditions and the implementation of deposition curves defined in tropical conditions are presented (**Chapter 4** – article to be submitted).

Five, the inclusion of an important farming practice influencing pesticide transfers: the ground cover management is proposed in the PestLCI Consensus model (**Chapter 5** – article to be submitted).

Finally, a **discussion and outlook** part is presented and followed by a general **conclusion**.

The thesis is presented in English, except for the general conclusion which is in French.

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INTRODUCTION

WHY AND HOW TO ASSESS PESTICIDES FOR AGRI-LCA IN TROPICAL REGIONS?

1. Pesticides use on crops grown in tropical conditions leads to environmental and human health impacts

The tropical agricultural systems show a wide diversity of production situations, including export value chains to European markets, periurban cropping systems around cities in developing countries or cropping systems in French overseas departments, with contrasted challenges in terms of environmental impacts and health risks. In tropical regions¹, the growing demand for crop products on both the international and the domestic markets offers an opportunity for farmers to increase their activities and revenues (Weinberger and Lumpkin 2007). Moreover, a quick growing of cities in developing countries is leading to largely uncontrolled periurban productions of fruits and vegetables with high applications of pesticides² (Perrin et al. 2015). It is important to notice that in Latin America, sub-Saharan Africa, and South and East Asia, farm households have on average less than five hectares of agricultural land and ensure food security by producing more than 70% of the food calories in these regions (Samberg et al. 2016). To ensure the crop yield and to fight the pests, diseases and weeds, a wide diversity of pesticides can be used at a high frequency (Lewis et al. 2016), in particular in horticultural production systems (Racke et al. 1997), and almost all year round (Daam and van den Brink 2010) (Figure 1).

¹ Tropical regions or conditions are located mainly between the Tropic of Cancer and the Tropic of Capricorn. They are characterized according to Köppen-Geiger climate classification by warm temperatures with a small variation in daily and annual temperatures and with an alternating rainy and dry season, or by an equatorial climate with humid conditions prevailing all year round (Kottek et al. 2006; Beck et al. 2018).

² Pesticides or active ingredients in plant protection formulations are defined as “any substance, or mixture of substances of chemical or biological ingredients intended for repelling, destroying or controlling pests, or regulate plant growth” (FAO).

(a) Hand-operated sprayer

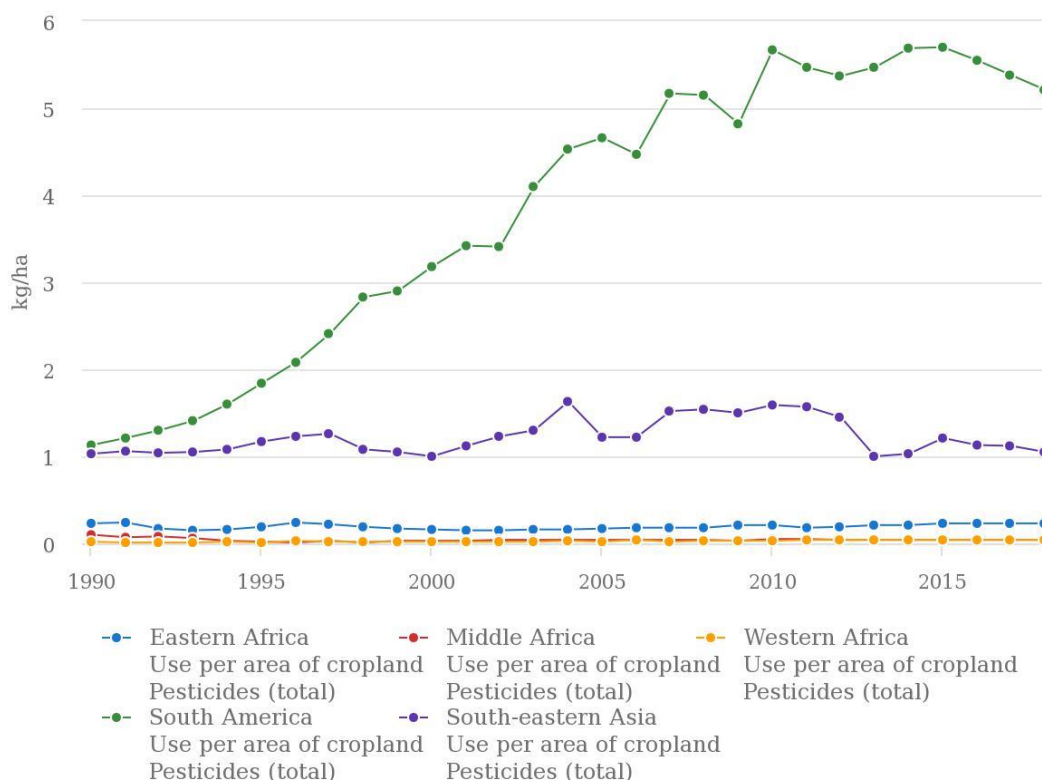


(b) Boom sprayer



Figure 1: Examples of pesticide application methods in tropical regions with (a) hand-operated spraying on vegetables in Martinique and (b) boom sprayer commonly used on pineapple fields in Costa Rica (Credits: Céline Gentil)

Figure 2 highlights the evolution of pesticide use per area of cropland since 1990. In South America, mainly showing tropical conditions as defined in Köppen-Geiger, the use of pesticides has increased by almost a factor of 5 over the last 30 years, with on average 5 kg of pesticide (i.e. active ingredient) applied per hectare and per year. For other tropical areas, pesticide uses have not changed substantially.



Source: FAOSTAT (Oct 10, 2020)

Figure 2: Pesticide use per area of cropland (kg/ha) and evolution since 1990, with details for regions of the world that are predominantly composed of tropical conditions (FAOSTAT)

In these tropical contexts, persistent pesticides are still used. In particular, in tropical developing countries, farmers use old chemicals with a high risk for the environment and human health (Sanchez-Bayo and Hyne 2011). Pesticides as all chemicals are widely disseminated in the environment and are responsible for human and ecosystem exposure and contamination (Landrigan et al. 2017), notably in tropical conditions (Arias-Estévez et al. 2008; Aktar et al. 2009; Lesueur Jannoyer et al. 2016). Pesticides can be detected in all environmental compartments in tropical regions, i.e. in the air (Fuhrmann et al. 2020), surface water (Bocquené and Franco 2005; Mottes et al. 2017a), groundwater (Sorensen et al. 2015), soil (Dickinson and Lepp 1984; Pillai 1986) and even in food crops (Hossain et al. 2015; Kariathi et al. 2016). Pesticide applications lead to non-target exposure of terrestrial ecosystems, on birds and mammals, on pollinating insects, and on marine and freshwater ecosystems (Daam and van den Brink 2010; Alves et al. 2013; Woodcock et al. 2017). Several routes of human pesticide exposure exist: directly by contact and inhalation at the pesticide manufacture level and for professional or domestic uses, and indirectly by inhalation of air, through the contact with contaminated surfaces and through water and food consumption (INSERM 2013).

Fantke and Jolliet (2016) identified the ingestion of pesticide residues in harvested crop components as the dominating exposure pathway for the general population. The risk of pesticide residues in food crop is often attributable to the non-compliance with the authorized pre-harvest period after the last application, the use of prohibited pesticides (Weinberger and Lumpkin 2007) and also to the non-compliance with the approved dose and frequency of application. The risk of ingestion of pesticide residues is increased for fruits and vegetables commonly consumed in fresh form (Weinberger and Lumpkin 2007). Some active ingredients used in agriculture and present in food are neurotoxic (as organophosphate pesticides), endocrine disruptors, carcinogenic etc. and there is strong evidence that chemicals and pesticides are responsible for ‘a global pandemic of neurodevelopmental toxicity’ (Landrigan et al. 2017).

To reduce the effects of pesticides on the environment and human health, it is critical to understand the relationship between farmers’ pest management and associated impacts.

2. Life Cycle Assessment for food and agriculture evaluation

Life Cycle Assessment (LCA) is an ISO-standardized methodology (ISO 14040 2006; ISO 14044 2006) to evaluate the environmental performance of product systems or services. LCA is aiming to identify the most environmentally friendly way(s) of fulfilling a function between different options, and constitutes a relevant tool for decision-makers. An LCA is divided in 4 steps (Figure 3). First, (i) “the goal and scope definition” phase, where the objectives of the study are defined, the system boundary is described and a functional unit is chosen with a definition of the product and its life cycle. Then (ii) the “Life Cycle Inventory (LCI)” phase in which physical fluxes of matter and energy are estimated over the whole life of the studied function, generally from “cradle-to-grave” or from “cradle-to-farm-gate” depending on the goal and scope of the study. These inventory fluxes are then aggregated and converted into indicators of potential environmental impacts by applying characterization factors and finally expressed per functional unit, at the “Life Cycle Impact Assessment (LCIA)” phase (iii). Results can be expressed at two levels: as midpoint and endpoint indicators. At midpoint level, there are 15 recommended impact categories (European Commission and Joint Research Centre 2010) (e.g. freshwater ecotoxicity and human toxicity regarding pesticide emissions). At the endpoint level, representing the ultimate objects to protect, the so-called areas of protection, they are three categories: abiotic resources, human health and ecosystem quality. The fourth step (iv) corresponds to the interpretation of the results in which the robustness of the results is discussed in relation to the objectives of the study and methods, and data used.

To evaluate the potential environmental and health impacts of crop production systems in relation to their function as part of a global environmental assessment, LCA is widely used (Dijkman et al. 2018; Poore and Nemecek 2018). A review on LCA study at the farm stage on perennial crops presented more than 70

LCA studies with some tropical crops such as sugarcane, coconut, banana, coffee and cocoa (Bessou et al. 2013) and other reviews presented LCA of vegetable products (Perrin et al. 2014). **LCAs of food products or agricultural production** systems generally aim at comparing the environmental performance of different production modes or farming practices fulfilling the same function and to identify for each production mode the margins of progress. For example, LCA is applied for comparisons between locally-grown and imported products (Basset-Mens et al. 2016b), or for eco-labelling and eco-design of new agricultural practices (Rouault et al. 2020). However, in the transition towards more agro-ecological practices, the LCA methodology is questioned for its completeness and relevance in comparing conventional, organic and agro-ecological production systems (Meier et al. 2015; van der Werf et al. 2020). One of the main challenges remains the proper accounting of the impacts due to the pesticide applications in conventional and organic (e.g. metal-based substances) cropping systems.

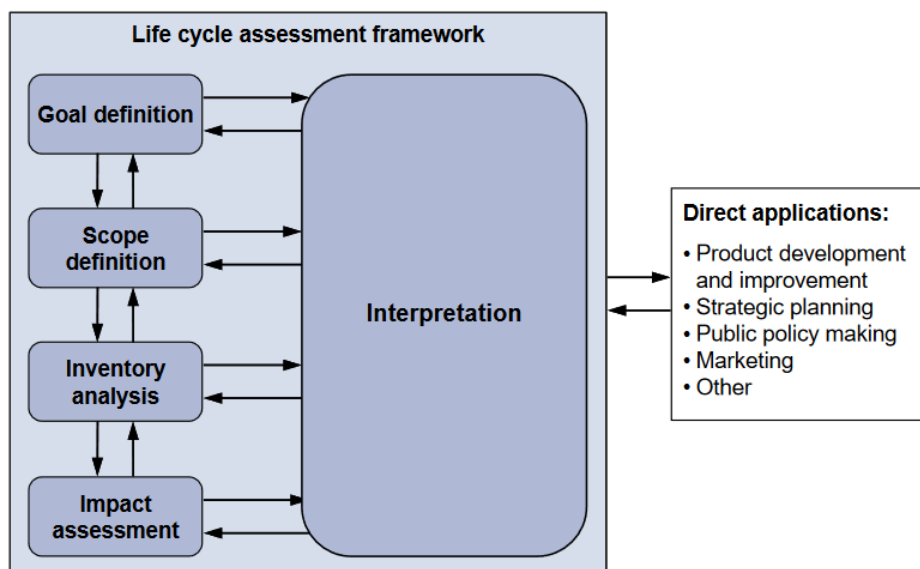


Figure 3: Framework for Life Cycle Assessment (European Commission and Joint Research Centre 2010)

3. A generally simplified assessment of pesticides in agri-LCA studies

Along the life cycle of pesticides (Figure 4), impacts come from the chemical manufacturing, the application at the farm stage, the ingestion of pesticide residues by consumers and the end of life treatment (Fantke 2019). In the LCA of agricultural products, pesticide applications are generally one of the main contributors to human and ecosystem toxicity impacts in cradle-to-farm-gate LCA studies (Bessou et al. 2013). Nevertheless, in many current LCA studies, pesticide emissions and related-impacts are not assessed

(Perrin et al. 2014). When pesticides are considered, 100% of the pesticides applied are assumed to be emitted to the agricultural soil (e.g. Nemecek and Schnetzer 2011) or a generic distribution between air and soil is assumed (e.g. Oliquino-Abasolo 2015) without considering any crop characteristics, application method, farming practices, soil or climate conditions. These field conditions and practices are highly influencing pesticides transfer to the environment, but differently in tropical conditions than in temperate ones (Sanchez-Bayo and Hyne 2011). Indeed, in tropical conditions, higher temperatures enhance degradation and volatilization of pesticides (Racke et al. 1997; Daam and van den Brink 2010). Intense rainfalls cause more runoff and leaching, leading to emissions to surface water and groundwater compartments (Sanchez-Bayo and Hyne 2011). Due to this pedoclimatic conditions, crop canopy and associated application methods differ from those under temperate conditions. Furthermore, some exposure pathways are frequently omitted, such as exposition by ingestion of pesticide residues in harvested crop components (Fantke et al. 2011b).

Overall, there is a need to evaluate the environmental performance of crop production including the (eco-)toxicity impacts due to pesticide applications (Perrin et al. 2014; Meier et al. 2015; Knudsen et al. 2019; van der Werf et al. 2020), notably due to pesticide residues (Fantke et al. 2011b). A set of the most updated models allows assessing more consistently pesticide emissions and related-impacts, considering notably the crop characteristics, application methods, soil and climate conditions.

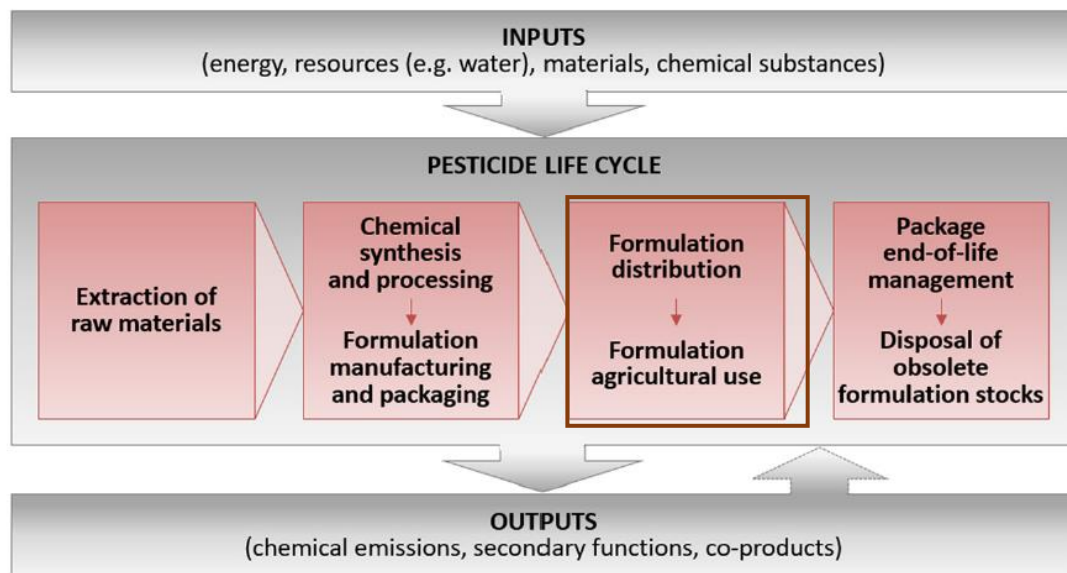


Figure 4: Pesticide application stage in the complete pesticide Life Cycle stages from Fantke (2019)

4. Most-up-to-date pesticide LCI-LCIA models and challenges

A brief overview of the available state-of-the-art pesticide emission and toxicity-related characterization models applicable for LCA, namely PestLCI Consensus, USEtox, and dynamiCROP, is given in the following (Figure 5).

Based on the framework proposed by Hauschild (2000), the LCI model **PestLCI** (Birkved and Hauschild 2006; Dijkman et al. 2012) estimates emission fractions to air, field soil, field crop, groundwater and off-field surfaces from pesticide application in open-field. It has been further advanced into PestLCI Consensus (Fantke et al. 2017a) and into a web-tool accessible from the web address <https://pestlciweb.man.dtu.dk/>. PestLCI uses two levels of emission distributions: initial (or primary) mass distribution covering initial processes within minutes after pesticide application and secondary emission distribution covering more continuous processes on crop leaves (degradation, volatilization, plant uptake) and soil (e.g. volatilization, leaching) within a longer period (e.g. 1 day) after application.

As a scientific consensus model, **USEtox** is a global and generic continental-scale model, to predict fate and exposure to chemicals. USEtox is widely applied in LCA for (eco-)toxicity characterization of chemical emissions in LCIA (Rosenbaum et al. 2008; Westh et al. 2015; Fantke et al. 2018a, b). In USEtox, human exposure factors matrix considers air inhalation, drinking water, exposed produce (= leaf, fruit and cereals), unexposed produce (= root crops), meat, dairy products and fish and recently pesticide residues in crop consumption (with the integration of dynamiCROP model).

The dynamic plant uptake model **dynamiCROP** was developed to assess pesticide residues exposure in crops through food consumption for LCA (Fantke et al. 2012b; Fantke and Jolliet 2016). The model is based on a “crop-specific multi-compartment system”, describing pesticide transfers across compartments (e.g. fruit surface deposit, stem, root-zone soil) over time. To calculate human impacts of pesticide through food ingestion, dynamiCROP follows the general LCIA cause-effect chain by combining factors representing environmental fate, human exposure and health effects into characterization factors (Fantke et al. 2011).

These three models were developed in temperate conditions and have been further completed, implementing worldwide climate and soil database. Nevertheless, few studies have used these LCI models (PestLCI and dynamiCROP) in tropical contexts (Ingwersen 2012; Fantke et al. 2012b). Furthermore, LCA studies usually do not couple state-of-the-art pesticide emission with (eco-)toxicity impact and pesticide crop residue models to ensure a consistent modeling from application to impacts (Martínez-Blanco et al. 2011; van Zelm et al. 2014; Perrin et al. 2014; Fantke 2019).

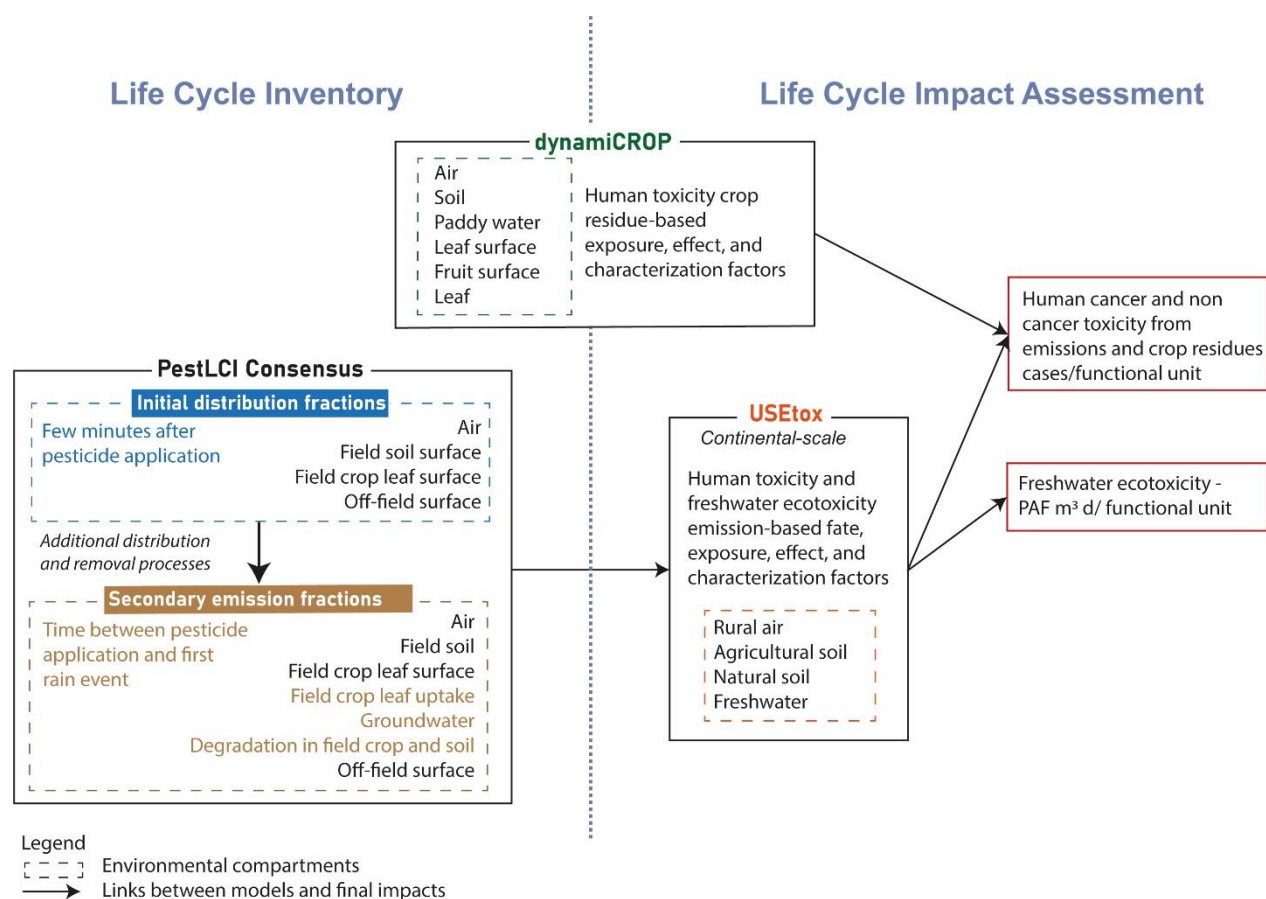


Figure 5: Life Cycle Inventory and Impact Assessment models for evaluating pesticides prior to this thesis work

In **conclusion**, evaluating emissions and associated impacts due to pesticide applications is challenging especially in a tropical context. For the LCA of crop products in tropical regions, operational and reliable models to estimate all fractions of pesticides applied into the environment including the harvested part of the crop are needed.

In order to define more specific research questions for the thesis and related objectives, we undertook a bibliographic review (chapter 1) to identify the characteristics and processes that determine pesticide emissions and related impacts under tropical conditions, and to assess to what extent LCI and LCIA models need to be adapted to better account for these conditions.

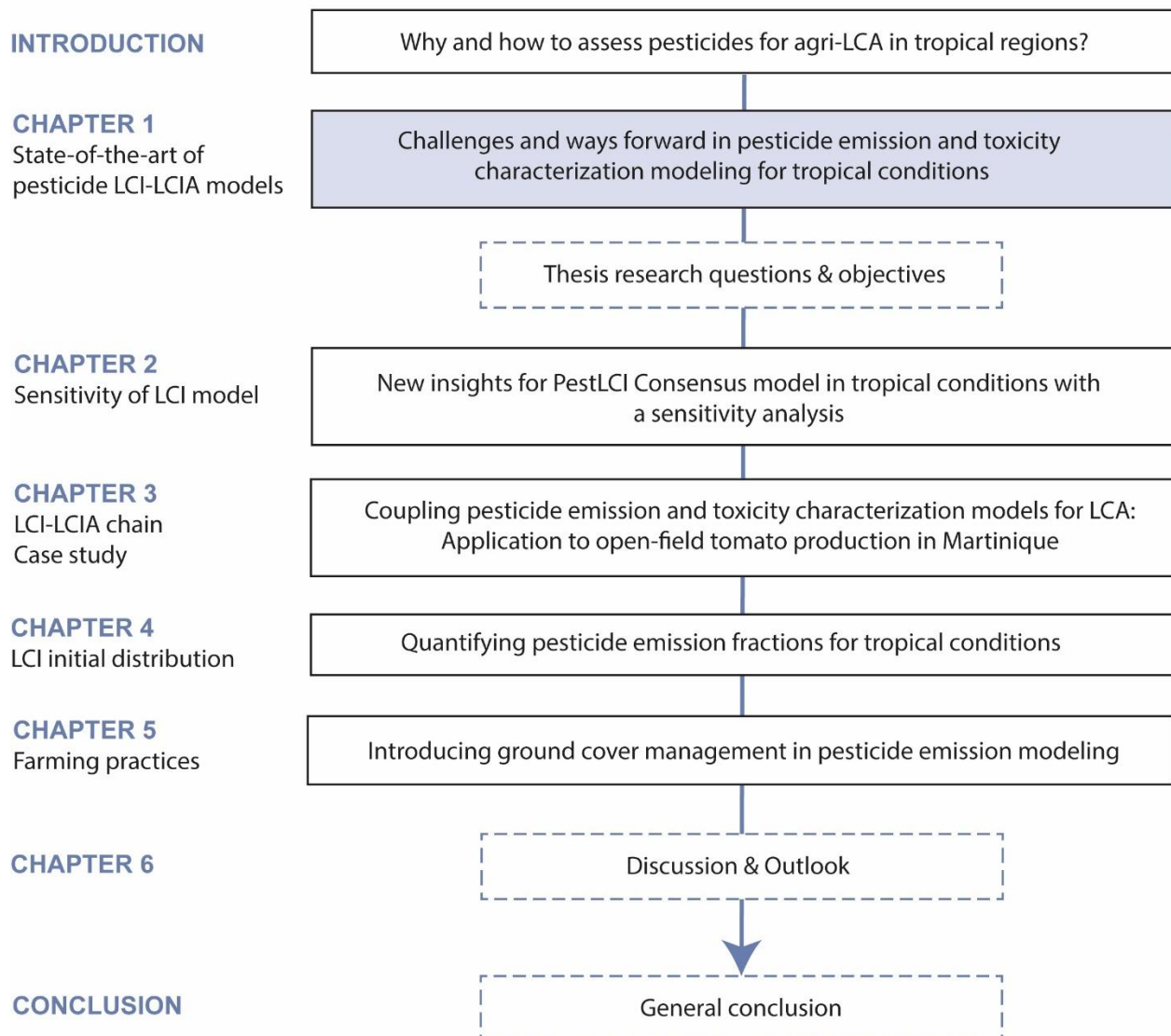
References Introduction

- Aktar MdW, Sengupta D, Chowdhury A (2009) Impact of pesticides use in agriculture: their benefits and hazards. *Interdisc Toxicol* 2:1–12. doi.org/10.2478/v10102-009-0001-7
- Alves PRL, Cardoso EJBN, Martines AM, et al (2013) Earthworm ecotoxicological assessments of pesticides used to treat seeds under tropical conditions. *Chemosphere* 90:2674–2682. doi.org/10.1016/j.chemosphere.2012.11.046
- Arias-Estévez M, López-Periago E, Martínez-Carballo E, et al (2008) The mobility and degradation of pesticides in soils and the pollution of groundwater resources. *Agri Ecosyst Environ* 123:247–260. doi.org/10.1016/j.agee.2007.07.011
- Basset-Mens C, Vanniere H, Grasselly D, et al (2016) Environmental impacts of imported and locally grown fruits for the French market: a cradle-to-farm-gate LCA study. *Fruits* 71:93–104. doi.org/10.1051/fruits/2015050
- Beck HE, Zimmermann NE, McVicar TR, et al (2018) Present and future Köppen-Geiger climate classification maps at 1-km resolution. *Scientific Data* 5:180214. doi.org/10.1038/sdata.2018.214
- Bessou C, Basset-Mens C, Tran T, Benoist A (2013) LCA applied to perennial cropping systems: a review focused on the farm stage. *Int J Life Cycle Assess* 18:340–361. doi.org/10.1007/s11367-012-0502-z
- Birkved M, Hauschild MZ (2006) PestLCI—A model for estimating field emissions of pesticides in agricultural LCA. *Ecol Model* 198:433–451. doi.org/10.1016/j.ecolmodel.2006.05.035
- Bocquené G, Franco A (2005) Pesticide contamination of the coastline of Martinique. *Mar Pollut Bull* 51:612–619. doi.org/10.1016/j.marpolbul.2005.06.026
- Daam MA, van den Brink PJ (2010) Implications of differences between temperate and tropical freshwater ecosystems for the ecological risk assessment of pesticides. *Ecotoxicology* 19:24–37. doi.org/10.1007/s10646-009-0402-6
- Dickinson NM, Lepp NW (1984) Pollution of tropical plantation crops by copper fungicides: a copper budget for Kenyan coffee plantation. *Stud Environ Sci* 341–346
- Dijkman TJ, Basset-Mens C, Antón A, Núñez M (2018) LCA of Food and Agriculture. In: Hauschild MZ (ed) *Life Cycle Assessment - Theory and Practice*. Springer, Cham, Switzerland, pp 723–754
- Dijkman TJ, Birkved M, Hauschild MZ (2012) PestLCI 2.0: a second generation model for estimating emissions of pesticides from arable land in LCA. *Int J Life Cycle Assess* 17:973–986. doi.org/10.1007/s11367-012-0439-2
- European Commission, Joint Research Centre (2010) *ILCD handbook: general guide for life cycle assessment: detailed guidance*. Publications Office of the European Union, Luxembourg
- Fantke P (2019) Modeling the environmental impacts of pesticides in agriculture. In: Weidema, B.P. (Ed) *Assessing the environmental impact of agriculture*, Burleigh Dodds Science Publishing, Cambridge, United Kingdom. doi.org/10.19103/AS.2018.0044.08

- Fantke P, Antón A, Grant T, Hayashi K (2017) Pesticide emission quantification for life cycle assessment: A global consensus building process. *J Life Cycle Assess* 13:245–251
- Fantke P, Aurisano N, Bare J, et al (2018a) Toward harmonizing ecotoxicity characterization in life cycle impact assessment. *Environ Toxicol Chem* 37:2955–2971. doi.org/10.1002/etc.4261
- Fantke P, Aylward Lesa, Bare Jane, et al (2018b) Advancements in Life Cycle Human Exposure and Toxicity Characterization. *Environmental Health Perspectives* 126:125001. doi.org/10.1289/EHP3871
- Fantke P, Jolliet O (2016) Life cycle human health impacts of 875 pesticides. *Int J Life Cycle Assess* 21:722–733. doi.org/10.1007/s11367-015-0910-y
- Fantke P, Juraske R, Antón A, et al (2011) Dynamic multicrop model to characterize impacts of pesticides in food. *Environ Sci Technol* 45:8842–8849. doi.org/10.1021/es201989d
- Fantke P, Wieland P, Juraske R, et al (2012) Parameterization models for pesticide exposure via crop consumption. *Environ Sci Technol* 46:12864–12872. doi.org/10.1021/es301509u
- Fuhrmann S, Klánová J, Příbylová P, et al (2020) Qualitative assessment of 27 current-use pesticides in air at 20 sampling sites across Africa. *Chemosphere* 258:127333. doi.org/10.1016/j.chemosphere.2020.127333
- Hauschild MZ (2000) Estimating pesticide emissions for LCA of agricultural products. In: Weidema BP (ed) *Agricultural data for life cycle assessments*. Agricultural Economics Research Institute (LEI), The Hague, pp 64–79
- Hossain MS, Fakhrudin ANM, Chowdhury MAZ, et al (2015) Health risk assessment of selected pesticide residues in locally produced vegetables of Bangladesh. *Int Food Res J* 22:110–115
- Ingwersen WW (2012) Life cycle assessment of fresh pineapple from Costa Rica. *J Clean Prod* 35:152–163. doi.org/10.1016/j.jclepro.2012.05.035
- INSERM (2013) *Pesticides : Effets sur la santé*. INSERM, Paris
- ISO 14040 (2006) *Management environnemental — Analyse du cycle de vie — Principes et cadre*. ISO
- ISO 14044 (2006) *Environmental management — Life cycle assessment — Requirements and guidelines*. International Standards Organization, 14044:2006
- Kariathi V, Kassim N, Kimanya M (2016) Pesticide exposure from fresh tomatoes and its relationship with pesticide application practices in Meru district. *Cogent Food & Agriculture* 2:1196808. doi.org/10.1080/23311932.2016.1196808
- Knudsen MT, Dorca-Preda T, Djomo SN, et al (2019) The importance of including soil carbon changes, ecotoxicity and biodiversity impacts in environmental life cycle assessments of organic and conventional milk in Western Europe. *J Clean Prod* 215:433–443. doi.org/10.1016/j.jclepro.2018.12.273
- Kottek M, Grieser J, Beck C, et al (2006) World Map of the Köppen-Geiger climate classification updated. *Meteorol Z* 15:259–263. doi.org/10.1127/0941-2948/2006/0130

- Landrigan PJ, Fuller R, Acosta NJR, et al (2017) The Lancet Commission on pollution and health. *The Lancet*. doi.org/10.1016/S0140-6736(17)32345-0
- Lesueur-Jannoyer M, Cattan P, Woignier T, Clostre F (2016) *Crisis management of chronic pollution: contaminated soil and human health*, 1st Edition. CRC Press
- Lewis SE, Silburn DM, Kookana RS, Shaw M (2016) Pesticide behavior, fate, and effects in the tropics: an overview of the current state of knowledge. *J Agric Food Chem* 64:3917–3924. doi.org/10.1021/acs.jafc.6b01320
- Martínez-Blanco J, Muñoz P, Antón A, Rieradevall J (2011) Assessment of tomato Mediterranean production in open-field and standard multi-tunnel greenhouse, with compost or mineral fertilizers, from an agricultural and environmental standpoint. *J Clean Prod* 19:985–997. doi.org/10.1016/j.jclepro.2010.11.018
- Meier MS, Stoessel F, Jungbluth N, et al (2015) Environmental impacts of organic and conventional agricultural products – Are the differences captured by life cycle assessment? *J Environ Manag* 149:193–208. doi.org/10.1016/j.jenvman.2014.10.006
- Mottes C, Lesueur Jannoyer M, Le Bail M, et al (2017) Relationships between past and present pesticide applications and pollution at a watershed outlet: The case of a horticultural catchment in Martinique, French West Indies. *Chemosphere* 184:762–773. doi.org/10.1016/j.chemosphere.2017.06.061
- Nemecek T, Schnetzer J (2011) *Methods of assessment of direct field emissions for LCIs of agricultural production systems. Data v3.0*. Swiss Center for Life Cycle Inventories, Duebendorf, Switzerland
- Oliquino-Abasolo A (2015) *Agro-environmental sustainability of conventional and organic vegetable production systems in Tayabas, Quezon, Philippines*. FAO University Library, University of the Philippines at Los Baños
- Perrin A, Basset-Mens C, Gabrielle B (2014) Life cycle assessment of vegetable products: a review focusing on cropping systems diversity and the estimation of field emissions. *Int J Life Cycle Assess* 19:1247–1263. doi.org/10.1007/s11367-014-0724-3
- Perrin A, Basset-Mens C, Huat J, Yehouessi W (2015) High environmental risk and low yield of urban tomato gardens in Benin. *Agron Sustain Dev* 35:305–315. doi.org/10.1007/s13593-014-0241-6
- Pillai MKK (1986) Pesticide pollution of soil, water and air in Delhi area, India. *Sci Total Environ* 55:321–327. doi.org/10.1016/0048-9697(86)90189-0
- Poore J, Nemecek T (2018) Reducing food's environmental impacts through producers and consumers. *Science* 360:987–992. doi.org/10.1126/science.aaq0216
- Racke KD, Skidmore MW, Hamilton DJ, et al (1997) Pesticides report 38. Pesticide fate in tropical soils - (Technical report). *Pure Appl Chem* 69:1349–1371. doi.org/10.1351/pac199769061349
- Rosenbaum RK, Bachmann TM, Gold LS, et al (2008) USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment. *Int J Life Cycle Assess* 13:532. doi.org/10.1007/s11367-008-0038-4

- Rouault A, Perrin A, Renaud-Gentié C, et al (2020) Using LCA in a participatory eco-design approach in agriculture: the example of vineyard management. *Int J Life Cycle Assess* 25:1368–1383. doi.org/10.1007/s11367-019-01684-w
- Samberg LH, Gerber JS, Ramankutty N, et al (2016) Subnational distribution of average farm size and smallholder contributions to global food production. *Environ Res Lett* 11:124010. doi.org/10.1088/1748-9326/11/12/124010
- Sanchez-Bayo F, Hyne RV (2011) Comparison of environmental risks of pesticides between tropical and nontropical regions. *Integr Environ Assess Manag* 7:577–586. doi.org/10.1002/ieam.189
- Sorensen JPR, Lapworth DJ, Nkhuwa DCW, et al (2015) Emerging contaminants in urban groundwater sources in Africa. *Water Research* 72:51–63. doi.org/10.1016/j.watres.2014.08.002
- van der Werf HMG, Knudsen MT, Cederberg C (2020) Towards better representation of organic agriculture in life cycle assessment. *Nature Sustainability* 3:419–425. doi.org/10.1038/s41893-020-0489-6
- van Zelm R, Larrey-Lassalle P, Roux P (2014) Bridging the gap between life cycle inventory and impact assessment for toxicological assessments of pesticides used in crop production. *Chemosphere* 100:175–181. doi.org/10.1016/j.chemosphere.2013.11.037
- Weinberger K, Lumpkin TA (2007) Diversification into horticulture and poverty reduction: a research agenda. *World Development* 35:1464–1480. doi.org/10.1016/j.worlddev.2007.05.002
- Westh TB, Hauschild MZ, Birkved M, et al (2015) The USEtox story: a survey of model developer visions and user requirements. *Int J Life Cycle Assess* 20:299–310. doi.org/10.1007/s11367-014-0829-8
- Woodcock BA, Bullock JM, Shore RF, et al (2017) Country-specific effects of neonicotinoid pesticides on honey bees and wild bees. *Science* 356:1393–1395. doi.org/10.1126/science.aaa1190



CHAPTER 1

CHALLENGES AND WAYS FORWARD IN PESTICIDE EMISSION AND TOXICITY CHARACTERIZATION MODELING FOR TROPICAL CONDITIONS

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Abstract

In tropical cropping systems, pesticides are extensively used to fight pests and ensure high crop yields. However, pesticide use also leads to environmental and health impacts. While pesticide emissions and impacts are influenced by farm management practices and environmental conditions, available Life Cycle Inventory (LCI) emission models and Life Cycle Impact Assessment (LCIA) toxicity characterization models are generally designed based on temperate conditions. There is, hence, a need for adapting LCI and LCIA models for evaluating pesticides under tropical conditions. To address this need, we aim to identify the characteristics that determine pesticide emissions and related impacts under tropical conditions, and to assess to what extent LCI and LCIA models need to be adapted to better account for these conditions.

We investigated the state-of-knowledge with respect to characteristics that drive pesticide emission patterns, environmental fate, human and ecological exposures, and toxicological effects under tropical conditions. We then discuss the applicability of existing LCI and LCIA models to tropical regions as input for deriving specific recommendations for future modelling refinements.

Our results indicate that many pesticide-related environmental processes, such as degradation and volatilization, show higher kinetic rates under tropical conditions mainly due to higher temperatures, sunlight radiation and microbial activity. Heavy and frequent rainfalls enhance leaching and runoff. Specific soil characteristics (e.g. low pH), crops and cropping systems (e.g. mulching) are important drivers of distinct pesticide emission patterns under tropical conditions. Adapting LCI models to tropical conditions implies incorporating specific features of tropical cropping systems (e.g. intercropping, ground cover management), specific drift curves for tropical pesticide application techniques and better addressing leaching processes. The validity domain of the discussed LCI and LCIA models could be systematically extended to tropical regions by considering tropical soil types, climate conditions, and crops, and adding active substances applied specifically under tropical conditions, including the consideration of late applications of pesticides before harvest and their effect on crop residues and subsequent human intake.

Current LCI and LCIA models are not fully suitable for evaluating pesticide emissions and impacts for crops cultivated in tropical regions. Models should be adapted and parameterized to better account for various characteristics influencing emission and impact patterns under tropical conditions using best available data and knowledge. Further research is urgently required to improve our knowledge and data with respect to understanding and evaluating pesticide emission and impact processes under tropical conditions.

Keywords: Life cycle assessment; pesticides; emission models; toxicity characterization models; tropical regions

1.1 Introduction

Tropical conditions are located mainly between the Tropic of Cancer and the Tropic of Capricorn. They are characterized according to Köppen-Geiger climate classification by warm temperatures with an alternating rainy and dry season, or by an equatorial climate with humid conditions prevailing all year round (Kottek et al. 2006; Beck et al. 2018). These conditions are very suitable for diversified agricultural production. A common characteristic of tropical crop farming is the possibility to grow crops all year round without interruption by a cold season, as is the case in temperate climates. However, such environmental conditions are also favorable for the occurrence of pests (insects, weeds, fungi, etc.). To fight pests and to preserve high crop yields and quality, most farmers use a wide diversity of pesticides at high frequency (Racke et al. 1997; Lewis et al. 2016) and sometimes all year round (Daam and van den Brink 2010; Mottes et al. 2017b). In many tropical contexts, farmers have received no or little training about proper pesticide application and have a limited awareness of pesticide risk (Williamson et al. 2008; Raksanam et al. 2012). Therefore, farmers often do not respect good application

practices (Settle et al. 2014; Houbraken et al. 2017; Elibariki and Maguta 2017), practice excessive use and misuse of pesticides (Montes et al. 2012; Pouokam et al. 2017). Furthermore, structural adjustment policies e.g. in African countries, led to a decline in compliance control services and an increase in reported misuses, including the use of unauthorized pesticides (de Bon et al. 2014). Under such circumstances, pesticide uses lead to increased environmental and health pressure (Arias-Estévez et al. 2008; Aktar et al. 2009). As in all cultivated areas in the world, pesticides can be detected in all environmental compartments in tropical regions, including groundwater (Sorensen et al. 2015). For example, Bocquené and Franco (2005) found pesticides in water and sediment from rivers in Martinique, Pillai (1986) in soil and air in India, and Dickinson and Lepp (1984) in non-agricultural plants and soils in Kenya. Moreover, Arias-Andrés et al. (2018) observed ecotoxicity effects of pesticides on freshwater organisms in Costa-Rica, and Peters et al. (1997) on marine organisms in tropical conditions. Pesticides have also been detected in food crops grown in tropical regions, which might exceed the recommended maximum residue limits (e.g. tomato (*Solanum lycopersicum*) in Tanzania: Kariathi et al. 2016, and vegetables in Bangladesh: Hossain et al. 2015). Finally, food crop consumption was identified as main pesticide exposure pathway for the general human population (Fantke and Jolliet 2016). This is particularly relevant for fruits and vegetables, mainly consumed in fresh form, which increases the risk of ingestion of pesticide residues (Weinberger and Lumpkin 2007).

To evaluate the potential environmental and health impacts of crop production systems in relation to their function as part of a global environmental assessment, Life Cycle Assessment (LCA) is widely used, and pesticides are generally one of the main contributors to human and ecosystem toxicity impacts in cradle-to-farm-gate LCA studies (Bessou et al. 2013). However, current models for applied to evaluate emissions and toxicity-related impacts of pesticides in LCA were developed and parameterized typically reflecting temperate conditions for climate, soil, application techniques and crops (Rosenbaum et al. 2008; Fantke et al. 2011b; Dijkman et al. 2012; Fantke et al. 2017a), questioning their relevance for tropical conditions. Under tropical conditions, pesticide emissions and impacts are not as well understood or supported by measurements as in temperate regions (Racke et al. 1997; Sanchez-Bayo and Hyne 2011). For example, ecotoxicological tests on aquatic ecosystems are rare in tropical countries (Castillo et al. 1996).

Fantke et al. (2017a) highlighted the lack of data and model parameters for characterizing emission patterns under tropical conditions, suggesting to develop specific pesticide application scenarios for tropical regions. However, the development of such scenarios requires pesticide emission and impact characterization models that are able to compare agricultural practices and conditions in tropical regions. There is an urgent need for adapting and parameterizing existing Life Cycle Inventory (LCI) emission and Life Cycle Impact Assessment (LCIA) toxicity characterization models to better account for pesticide impacts under tropical conditions.

This paper, hence, aims at providing an overview of how to adapt currently available LCI and LCIA models to evaluate the use of pesticides in agriculture under tropical conditions, based on addressing three specific objectives. First, we explore factors influencing emission and toxicity impact patterns under tropical conditions. Second, we analyze current LCI and LCIA models with focus on how they currently reflect tropical conditions. Third, we provide insights of model improvements to better account for tropical conditions, and derive specific recommendations for future research to adapt LCI and LCIA models to address emissions and impacts of pesticides applied under tropical conditions.

1.2 Methods

1.2.1 Study area – tropical conditions and crops

The tropical conditions defined by the Köppen-Geiger climate classification (Kottek et al. 2006; Beck et al. 2018) are divided into three categories: tropical rainforest climate, tropical monsoon climate, and tropical savannah climate with dry summer or winter. These climates, the regions and countries they cover, and the main related crops are summarized in Table 1. In this paper, we always refer to this Köppen-Geiger climate classification. We note that a large diversity of climates might be present even in a small territory, for example in the Martinique Island, where all three above-described tropical climates are present on an area of only 1128 km². Other climatic conditions between the Tropics of Cancer and Capricorn, as arid desert conditions or temperate conditions in highlands (e.g. mountain area in South America), are not considered as tropical conditions, and are hence not addressed in our study. In some regions, tropical conditions are found only part of the year. These climates are called humid subtropical climate, and are characterized by hot and humid summer (e.g. east of Australia, east of China), and are not considered in this study.

Even if climate conditions are widely different between tropical and temperate regions, certain crops can be grown in both, e.g. maize (*Zea mays*) and tomato. However, some crops can usually only be grown under tropical conditions, e.g. palm oil (*Elaeis guineensis*), cassava (*Manihot esculenta*) or sugarcane (*Saccharum officinarum*), because the sum of temperatures in temperate climates is too low to finish the crop cycle during the warm season. Some crops are present in the three different equatorial climates (e.g. banana *Musa spp.*). According to FAOSTAT (2019), the main crops in terms of area harvested in the countries with tropical climates are in decreasing order of importance: rice (*Oriza spp.*), maize, soybean (*Glycine max*), sorghum (*Sorghum bicolor*), bean (*Phaseolus vulgaris*), cassava, sugar cane and palm oil. As synthesized in Racke et al. (1997), in tropical conditions depending on rain patterns, two types of crop production exist. In equatorial humid climates, the main staple crops are roots and tubers (e.g. sweet potato *Ipomoea batatas* and cassava), and fruits (e.g. banana), whereas in tropical climates with a dry season, the staple foods are cereals (e.g. rice, maize and sorghum). Beyond the

mentioned crops, the production of fruits and vegetables is generally important in these regions for the nutritional balance of people's diet (Weinberger and Lumpkin 2007; Williamson et al. 2008). Fruits and vegetables thereby constitute preferential targets for pests, and as nutritional high value crops they usually receive intensive pesticide applications.

Table 1: Presentation of tropical climates, their main characteristics, regions/countries and main crops grown

Tropical climates	Main characteristics		Regions/countries	Main crops grown in these climates
	Temperature	Humidity		
Equatorial rainforest, fully humid	$T_{\min} \geq +18\text{ }^{\circ}\text{C}$	$P_{\min} \geq 60\text{ mm}$	Generally found within 15° North and South of the equator: in Central Africa (e.g. Uganda), in Southeast Asia (e.g. Malaysia, Indonesia), parts of Central and South America (e.g. Colombia, Costa Rica)	Rice, palm oil, roots and tubers
Equatorial monsoon		$P_{\text{ann}} \geq 25 \times (100 - P_{\min})$	Caribbean islands (e.g. Dominican Republic), west and central Africa (e.g. Guinea, Cameroon), South Asia (e.g. Philippines)	Rice, pulses, sorghum, sugar cane
Equatorial savannah with dry summer or dry winter		$P_{\min} < 60\text{ mm}$ in summer or $P_{\min} < 60\text{ mm}$ in winter	Central and Northern parts of South America (e.g. Brazil), Central Africa (e.g. Tanzania, Madagascar), Southeast Asia (e.g. Thailand, India), Northern Australia	Rice, maize, sorghum, pulses, groundnuts (<i>Arachis hypogaea</i>), sugar cane

T_{\min} : monthly mean temperature of the coldest month

P_{\min} : mean precipitation of the driest month

P_{ann} : mean cumulative annual precipitation

1.2.2 Methodological approach

As first step, available literature was reviewed to analyze the state-of-the-art in data describing pesticide emissions and impacts under tropical conditions. According to the tropical climates from Köppen-Geiger classification, we identified all countries with at least one of the three tropical climates. Using these countries as spatial scope, we did a bibliographic search (on Agricola, Agris and CAB Abstract) using an “AND” combination of two main search criteria. One criterion was related to pesticides (using search keywords, such as “fungicide”, “insecticide”, etc.) and one criterion related to Life Cycle Assessment, (eco)-toxicity and environmental dissipation of pesticides (using search keywords, such as “degradation”, “leaching”, “fate”, etc.). More than 600 articles were identified and

576 were selected and organized in three categories by reading the abstracts: environmental processes ($n=288$), (eco)-toxicity ($n=331$) and farmers' behavior with pesticide application ($n=41$). Some articles could appear in more than one category. These articles were analyzed and classified considering different aspects: country or region, crop, pesticide target class, environmental compartments and processes, etc. This bibliographic work enabled us to realize that there were many articles available on the subject. Some key review papers allowed us to identify the major and specific processes of pesticide dissipation in the environment under tropical conditions (e.g. Racke et al. 1997; Sanchez-Bayo and Hyne 2011). Consequently, another, more focused bibliographic search has been conducted, using Web of Science. Subsequently, those publications that were most relevant with respect to the focus on tropical conditions were analyzed in more detail, with 17 articles studying water flow processes (e.g. leaching, runoff), 17 articles focusing on pesticide drift when applying pesticides, and 20 articles analyzing effects of farm management practices on pesticide dissipation. Overall, there is a rich body of risk assessment literature, which could additionally be used to improve and refine existing models applied in LCI and LCIA for pesticides (e.g. Brock et al. 2009; Thorbek et al. 2009).

As a final step, the validity and completeness of state-of-the-art models for characterizing pesticide emissions and toxicity-related impacts in LCA, namely PestLCI, USEtox and dynamiCROP, were assessed based on the references presented in Table 2. Furthermore, the equations, assumptions and database of the PestLCI model were analyzed in detail. Finally, improvement recommendations were derived according to the relevant processes and characteristics identified for tropical conditions and with specific relation to the studied models.

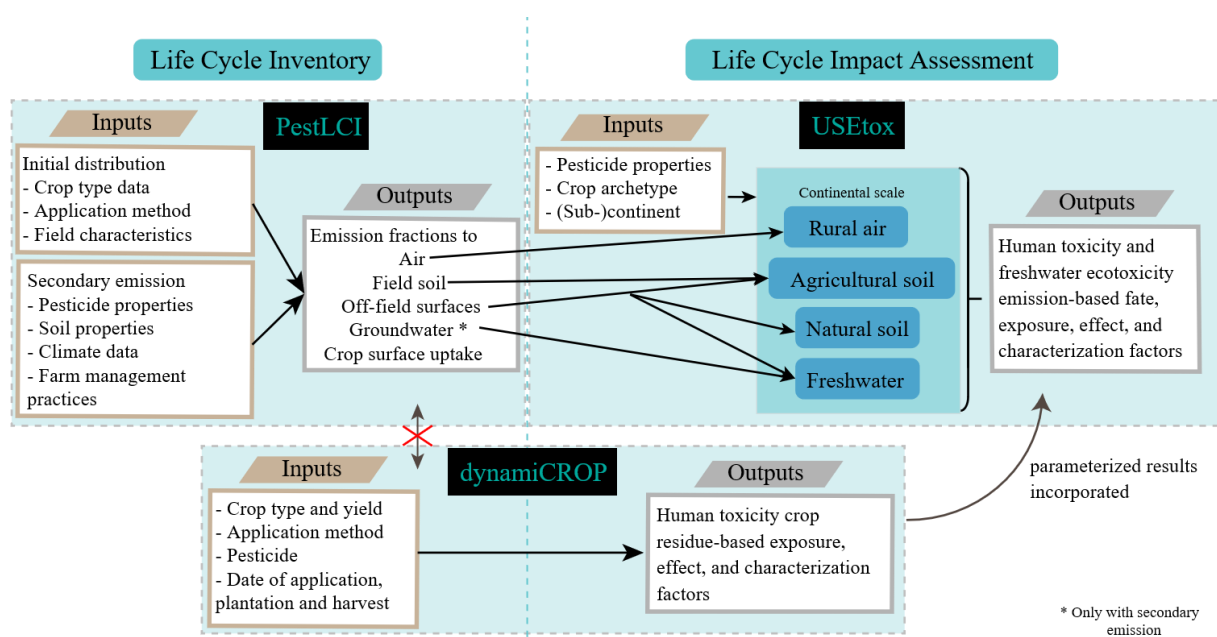
1.2.3 Pesticide modelling in LCA

In order to estimate pesticide impacts in LCA, models are required as well as an exhaustive and reliable inventory of pesticide emissions under various relevant production systems and conditions. We selected the three most up-to-date pesticide emission and toxicity-related characterization models, namely the adapted PestLCI model as pesticide emission inventory model, USEtox 2.1 as general (eco-)toxicity characterization model, and dynamiCROP 3.1 as model characterizing human exposure to pesticide residues in food crops. The models are presented in detail further below, and related references summarized in Table 2. The inter-connections between the considered models along with their inputs and outputs are presented in Figure 6. As part of a global pesticide consensus building effort for LCA (Rosenbaum et al. 2015; Fantke et al. 2017a), some modifications to the PestLCI 2.0 model were proposed, mainly focusing on including additional drift functions and adapting certain model parameters. We refer to this version in the following text as 'adapted PestLCI model'.

Table 2: References used for the analysis of the models PestLCI, USEtox and dynamiCROP

Models	Original publications	Consensus publications / model update / model analyses	Publications on model case studies
PestLCI	Birkved and Hauschild 2006	Dijkman et al. 2012; Dijkman 2013; Fantke et al. 2017a; Fantin et al. 2019; Rosenbaum et al. 2015	<i>PestLCI 1.0:</i> Vázquez-Rowe et al. 2012; Salomone and Ioppolo 2012; Bojacá et al. 2012; Ingwersen 2012* <i>PestLCI 2.0:</i> Dijkman 2013; Nordborg et al. 2014, 2017; Xue et al. 2015; Renaud-Gentié et al. 2015; Vázquez-Rowe et al. 2017
USEtox	Rosenbaum et al. 2008; Fantke et al. 2017b	Henderson et al. 2011; Rosenbaum et al. 2011; Westh et al. 2015	Juraske et al. 2009; Berthoud et al. 2011; Vázquez-Rowe et al. 2012; Dijkman et al. 2017; Hunt et al. 2017 <i>Used in combination with PestLCI or dynamiCROP:</i> Fantke et al. 2011b; Vázquez-Rowe et al. 2012, 2017; Ingwersen 2012; Dijkman 2013; Antón et al. 2014; Xue et al. 2015; Renaud-Gentié et al. 2015; Fantke and Jolliet 2016
dynamiCROP	Fantke et al. 2011a,b	Fantke et al. 2012b, 2013; Fantke and Jolliet 2016	Juraske et al. 2011; Fantke et al. 2011a; Juraske et al. 2012; Itoiz et al. 2012; Fantke et al. 2012a; Jacobsen et al. 2015; Fantke and Jolliet 2016; Feng et al. 2018

*only one study for tropical conditions

**Figure 6:** Life cycle inventory and impact assessment models for evaluating pesticides, their inputs, outputs and inter-connections

1.2.3.1 PestLCI to quantify pesticide emissions

The adapted PestLCI model is a model to estimate pesticide emissions for LCA of agricultural products (Dijkman et al. 2012; Fantke et al. 2017a). Based on the framework proposed by Hauschild (2000), this model estimates emissions to air, surface water, groundwater and soil from pesticide application in open fields, through two sets of distributions. Primary distribution covers initial processes within few minutes after pesticide application. When the pesticides have been deposited on crop, soil and off-field surfaces, and emitted to air via wind drift, secondary distribution estimates emissions covering more continuous processes on crop leaves (degradation, volatilization, plant uptake) and soil (volatilization, degradation, leaching, runoff). As result of the secondary distribution, pesticides are emitted to surface water, to groundwater, to soil, to air and to plant compartments (Dijkman et al. 2012). These processes are captured until the first rainfall event occurs (according to the frequency of rainfall events by month). The model was not developed for pesticide emission quantification for greenhouse production. However, from the same framework by Hauschild (2000), Antón et al. (2004) developed a proposition to evaluate pesticide emissions in greenhouses for LCA studies. Depending on the goal and scope of an LCA study, the agricultural soil and the buffer zone may be considered part of the ecosphere (i.e. environment) or the technosphere (i.e. agricultural production system). This will influence the results, as in LCA, an emission is a chemical flow crossing the boundary between technosphere and ecosphere. The interest of using PestLCI has been demonstrated by Vázquez-Rowe et al. (2012, 2017) ; Renaud-Gentié et al. (2015) ; Fantke et al. (2017a); Fantke (2019), in comparison to other methods as e.g. ecoinvent where 100% of the applied dose of pesticides is assumed to be emitted directly to the agricultural soil (Nemecek and Schnetzer 2011). In contrast, PestLCI allows to integrate much more specificity than such generic assumptions, and estimates are derived as function of crop, location, growing season, pesticide, farming practice, and application method (Vázquez-Rowe et al. 2017). The adapted PestLCI model reflects the state-of-the-art in estimating pesticide emissions in LCA. This model is hence used to analyze its suitability for quantifying pesticide emissions under tropical conditions. The LCA study from Ingwersen (2012) of fresh pineapple (*Ananas comosus*) in Costa Rica constitutes a first application of the PestLCI model to estimate emissions under tropical conditions, which we therefore include in the discussion of model suitability.

1.2.3.2 USEtox to characterize human toxicity and ecotoxicity impacts

For characterizing human toxicity and ecotoxicity impacts in LCA, the most consensual model is USEtox (Hauschild et al. 2013). USEtox was developed as outcome of a global scientific consensus building process aiming to harmonize existing LCIA models for assessing environmental and health exposure to toxic substances (Rosenbaum et al. 2008; Westh et al. 2015). USEtox is a continental-scale

model with six environmental compartments at continental level: urban air, rural air, agricultural soil, natural soil, freshwater and coastal marine waters. This steady-state model allows calculating two impact categories with three indicators, two for human toxicity, namely human cancer toxicity and human non-cancer toxicity, and one for freshwater aquatic ecotoxicity of chemical emissions (including pesticides). Depending on the goal and scope of each LCA, USEtox allows calculating potential impact results at the midpoint level or damage results at the endpoint level. Ecotoxicity impact results are expressed as potentially affected fraction of species (midpoint) and potentially disappeared fraction of species (damage), integrated over exposure time and volume per unit mass of a substance emitted. Human toxicity impact results are expressed as number of disease cases (midpoint) and disability-adjusted life years (damage) per unit mass of a substance emitted. Human exposure factors account for air inhalation, and ingestion of drinking water, exposed produce (= leaf, fruit and cereals), unexposed produce (= root crops), meat, dairy products and fish. The consideration of pesticide residues in food crops was recently included based on the parameterization of the dynamiCROP model for six major food crops (Fantke et al. 2017b).

1.2.3.3 dynamiCROP to characterize exposure to crop residues

The dynamic plant uptake model dynamiCROP was developed, to include in LCA human exposure to pesticide residues in food crops as a predominant exposure pathway for the general human population (Juraske et al. 2009; Fantke et al. 2012b). This model estimates pesticide plant uptake and residue exposure in open field contexts, but also allows for pesticide greenhouse applications. To calculate human impacts of pesticides residues through food crop consumption, dynamiCROP follows the general LCIA cause-effect chain combining factors representing environmental fate, human exposure and health effects (Fantke et al. 2011b). The dynamiCROP model relates the mass that is ultimately taken in by humans via crop residues to the mass of applied pesticide (Fantke and Jolliet 2016). Food processes, such as peeling of fruits, are taken into account using generic food processing factors. Pesticide fate in plants is mainly influenced by degradation in and on crops, time between pesticide application and crop harvest, overall residence times in soil, and substance molecular weight, based on a detailed analysis of influencing factors (Fantke et al. 2013; Fantke et al. 2014). These five key parameters are responsible for between 80% and 93% of the variation in pesticides residues and allowed to create parameterized exposure models for six important food crops (Fantke et al. 2012b). When combining human exposure estimates with human toxicity effect information, human toxicity results in dynamiCROP are compatible with results from USEtox and are expressed in disease cases (midpoint) or DALY (endpoint) per kg pesticide applied (or per kg emitted based on linking mass applied to mass emitted). Since 2016, parameterized dynamiCROP results are incorporated in USEtox (direct input in intake fraction matrix), but to use the model in its full version, LCA practitioners will need to couple dynamiCROP results with

the applied pesticide mass on crops, which is currently not included in LCI databases (Rosenbaum et al. 2015).

1.3 Results and discussion

1.3.1 Characteristics of pesticide emission and impact patterns under tropical conditions

Tropical crop production systems and their use of pesticides are very different compared to cropping systems under temperate conditions. To highlight the main differences, we first present the specificities of tropical abiotic and biotic features. Second, agricultural practices and farmers' behaviors are detailed. Figure 7 summarizes the identified key processes and characteristics influencing pesticide emission distribution under tropical conditions.

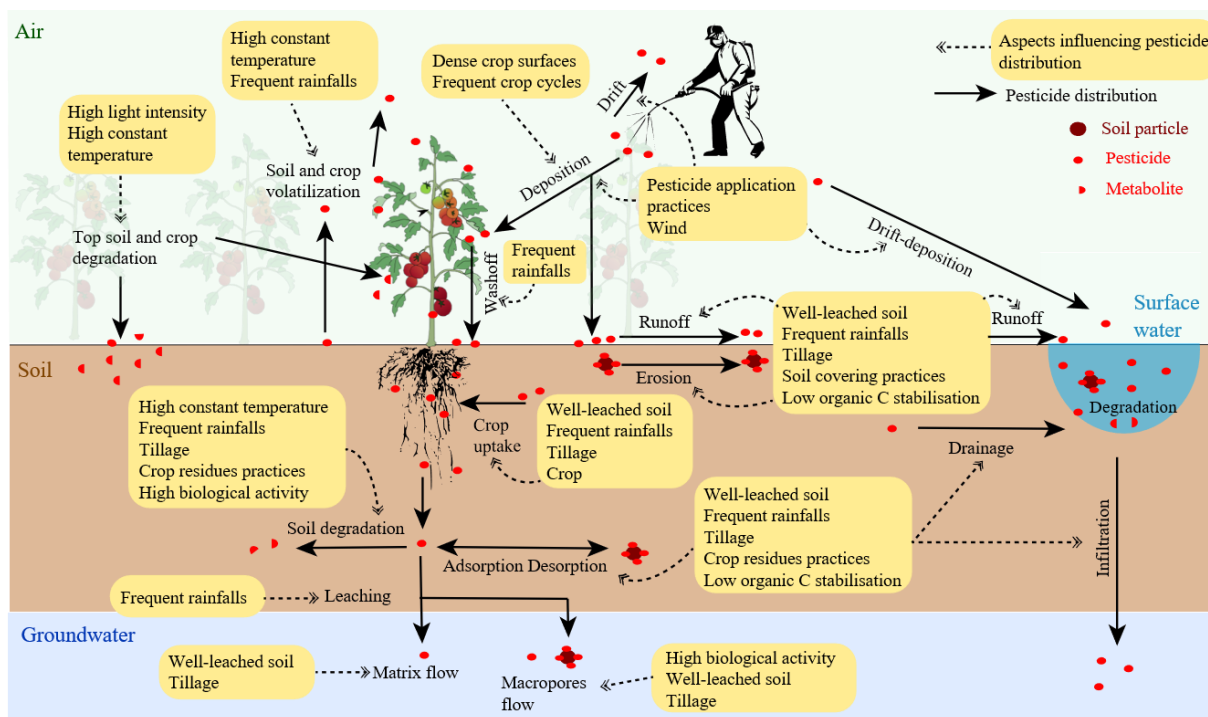


Figure 7: Key processes and characteristics that influence pesticide emission distribution in the tropics, normal arrows represent the pesticide distribution and dotted arrows represent aspects influencing pesticide distribution under tropical conditions

1.3.1.1 Environmental characteristics of tropical conditions

Abiotic conditions. Temperature dependent processes, such as degradation, have higher kinetic rates under tropical conditions due to higher average temperatures (Daam and van den Brink 2010), which facilitates higher biological activity (Racke et al. 1997). Increasing temperature also enhances volatilization (Sanchez-Bayo and Hyne 2011) because of higher vapor pressure (Figure 7). Even if vapor pressure is an intrinsic pesticides' characteristic, higher temperatures enhance the ability of a pesticide to turn into vapor and volatilize into the air. Just after pesticide application, a higher sunlight radiation allows for increased photodegradation on plant leaf surfaces compared to that under more average latitudes. Likewise, the photolysis of pesticides on soil surfaces is more important in tropical regions especially for pesticides applied on soil, without shadow from the crop, notably at the beginning of crop growth (Daam and van den Brink 2010).

Pesticide emission patterns are also influenced by rainfall distribution patterns and intensity, especially in tropical conditions, where extreme rainfall events and/or very dry seasons occur frequently (Figure 7). Extreme rainfall causes high runoff and leaching, resulting in more pesticide residues found in surface waters and higher mobility toward groundwater (Sanchez-Bayo and Hyne 2011). In Martinique (French West Indies), where the climate is humid tropical, Mottes et al. (2017) showed high rates of river contamination by pesticides, especially herbicides. In these circumstances, rainfall events directly after pesticide application generated the pollution peaks. Likewise, in Guadeloupe, Charlier et al. (2009) highlighted for cadusaphos that the main contributor to stream contamination is shallow groundwater, due to the permeability of the soil and abundant rainfall.

In such tropical climatic conditions, a huge diversity of soils is present with various characteristics. One particularity of tropical soils is their substantial anion exchange capacity (Racke et al. 1997; Sansoulet et al. 2007). Soil characteristics (organic carbon content and pH) are also important influencing factors of pesticide distribution under tropical conditions, as demonstrated by e.g. Sanchez-Bayo and Hyne (2011) (Figure 7). In tropical contexts, the decomposition of organic matter is five times higher as compared to temperate environments (Racke et al. 1997). The organic carbon content might be low due to high rainfall and high microbial activity, which results in less adsorption and consequently more availability to be transferred to water and air (Sanchez-Bayo and Hyne 2011). Nevertheless, organic carbon content deficit can be compensated by organic matter inputs from crop residues, leading to soil stabilization; especially where microbial degradation and biomass production occurs all year round. Furthermore, when soil pH is low, which is the case for most soils in Brazil and Southeast Asia, the desorption of acidic herbicides leaves more residues available for leaching (Sanchez-Bayo and Hyne 2011).

Biotic conditions. In tropical regions, biodiversity in terrestrial and freshwater aquatic ecosystems is generally higher than in temperate regions, mainly due to less limiting aspects related to rain and temperature (Brown 2014). Few studies exist, however, on the specificity of environmental impact of pesticide use under these conditions. In their study, Kwok et al. (2007) compared the sensitivity of tropical and temperate freshwater organisms for 18 chemical substances, and demonstrated that tropical aquatic organisms seem to be more sensitive to some organic chemicals and less sensitive to metals. Furthermore, the potential effect of pesticides on aquatic organisms might be reinforced by potentially high pesticide concentrations in water due to pesticide distribution processes in tropical environments (e.g. runoff) (Sanchez-Bayo and Hyne 2011). For pesticide impacts on terrestrial ecosystems, it has been reported that different pesticides show negative effects on non-target tropical species (e.g. Alves et al. 2013). Arias-Andrés et al. (2018) presented specific acute toxicity data with tropical native species (earthworm). A recent study also synthesized available toxicological data of freshwater shrimps for insecticides and fungicides under tropical conditions (Daam and Rico 2018). These data were compared to data for temperate species (*Daphnia magna* and aquatic invertebrates), showing that the shrimps were less sensitive to sodium channel modulator insecticide, e.g. lindane, cypermethrin and to acetylcholinesterase inhibiting fungicides and insecticides e.g. diazinon. In their review, Aktar et al. (2009) also warned against the risks for soil microorganisms, insects, plants, aquatic organisms and birds, associated to the use of organochlorine pesticides in India, which are usually phased out and not used anymore in temperate regions dominated by developed countries.

1.3.1.2 Agricultural practices and farmers' behavior

Thanks to the particular environmental conditions of tropical cropping systems and according to the crop production target for export (e.g. palm oil, cocoa (*Theobroma cacao*), rice, banana), local market (e.g. fruits and vegetables, rice) or self-sufficiency (e.g. roots and tubers, fruits and vegetables), farming systems and associated crops cultivated are highly diversified (Biénabe et al. 2016). While in certain cropping systems one single crop is grown intensively and years after years over a vast and flat area (e.g. sugarcane), in small-scale farming systems, agro-forestry systems cash crops (e.g. coffee (*Coffea spp.*), cocoa can be combined with fruit trees (e.g. avocados (*Persea americana*), guava tree (*Psidium guajava*) and/or vegetable crops) in intercropping in mountain areas. Thus, application methods and farm management practices can vary a lot depending on cropping systems and environmental characteristics. Due to these different types of cropping systems and practices, pesticide emissions to the environment also greatly differ from temperate conditions. Tropical crops also have specific characteristics that can influence pesticide distribution. Their canopy might have a greater volume compared to temperate crops and due to faster crop growth in the tropics, several production cycles can be run one after the other and generally all year round.

Pesticide use and application methods. In tropical agriculture, one of the main application methods used is the hand operated sprayer, such as the backpack sprayer or knapsack sprayer as for horticulture production (Charlier et al. 2009) and for cash crops such as coffee and cocoa as shown by Matthews et al. (2003) in Cameroon. Hand operated sprayers constitute for instance over 90% of the spraying equipment in Kenya (Mitoko 1997). More conventional boom sprayers and air blast sprayers are also used, respectively, in monocultures such as soybean (Bueno et al. 2017) and in fruit production (Alves and da Cunha 2014). Aerial application is banned in certain countries but is still in use in others, in particular in huge tropical banana or sugar cane cropping systems. Therefore, since pesticide applications are mainly done directly by hand or with manual applicators, occupational exposure might be higher in the tropics especially in developing countries where farmers often have a reduced education on pesticides' risks for their health. In addition to that, the warm and humid conditions make the wearing of personal protective equipment (PPE) difficult (Raksanam et al. 2012). Furthermore, due to generally high diversity of crop productions and pests under tropical conditions, there are certain pests for which no authorized pesticides are currently available (Laplace 2018). These orphan uses represent a major risk for human health and the environment, since farmers might be tempted to use unauthorized plant protection products for their crops.

Due to a low level of knowledge about pesticide use (no or little training in pesticide application) and weak awareness of pesticide risk (Settle et al. 2014; Houbraken et al. 2017; Elibariki and Maguta 2017), non-compliance with the approved dose and/or with the frequency of application is also common in tropical regions. The non-compliance with the minimum required pre-harvest period after the last application and the use of prohibited pesticides (Weinberger and Lumpkin 2007; Montes et al. 2012; Pouokam et al. 2017) further increase human and ecological risks under tropical conditions.

Agricultural practices. Specific agricultural practices in the tropics affect pesticide emissions at the field and catchment scales (Mottes et al. 2014), in particular practices influencing the hydraulic processes in and on the soil, and concern mostly tillage and ground cover management (Figure 7). For example, the implementation of cover plants or crops between crop rows allows to reduce runoff, as shown for banana plantations in intercropping with pineapple (Abbasi and Jamal 1999). Likewise, the use of crop residues as straw in sugar cane production (Pereira-Junior et al. 2015) or biochars (Kookana 2010; Mendes et al. 2018) increases herbicide sorption and reduces leaching. In conclusion, these specificities of tropical conditions and farm management practices are expected to strongly influence emission patterns and should therefore be accounted for in modelling pesticide emissions.

1.3.2 Improvements to better model pesticides under tropical conditions in LCA

The results of our critical analysis of the models' validity for tropical conditions are summarized in Table 3, providing specific recommendations for model improvement, which are either specific to

tropical conditions or considered generally applicable for all contexts. Subsequently, margins of improvements and proposals for further research for the adapted PestLCI model, the impact model USEtox and the dynamiCROP model are presented.

Table 3: Recommendations for improving the adapted PestLCI model, USEtox and dynamiCROP to be suitable for assessing pesticides under tropical conditions, and more generally for agricultural LCA.

	Adapted PestLCI model	USEtox	dynamicroP
General context	<ul style="list-style-type: none"> - Adding ground cover management and other agricultural practices or techniques (such as greenhouse production) ○ 	<ul style="list-style-type: none"> - Adding missing characterization factors for active substances (for organic, metal, inorganic and biological/natural pesticides) ○ - Including marine, terrestrial, pollinators and birds' toxicity ○ - Adding exposure pathways for human toxicity (bystanders, workers) ○ - Adding groundwater, sediment and plant compartments ○ 	<ul style="list-style-type: none"> - Adding missing exposure factors for active substances (for organic, metal, inorganic and biological/natural pesticides) ● - Improving the estimation of human intake due to late application of pesticides before harvest ○
	<ul style="list-style-type: none"> - Adding active substances, notably biological/natural substances, metals and inorganics; and methods for characterizing additional substances (e.g. inorganic salts and plant-based pesticides) ○ - Offering the possibility to parameterize the models with users' set of data (crop, soil and climate) ● - Including metabolites emission and impact modelling, notably with guidance ○ - Incorporating seedlings and seeds, and post-harvest pesticide applications ○ 		
Tropical context	<ul style="list-style-type: none"> - Adding tropical crop data (plant development and growth, archetypes) ● - Adding drift curves for tropical pesticide application techniques ● - Correction of empirical equations defined based on temperate conditions ○ - Allowing modelling of small plots (less than 50 m) ○ - Integrating processes occurring after the first rainfall event (such as leaching) ○ - Including drainage ditches ○ 	<ul style="list-style-type: none"> - Adding ecotoxicological data specific to tropical biota ○ 	<ul style="list-style-type: none"> - Adding crop data (plant development and growth, archetypes) for tropical crops ○
	<ul style="list-style-type: none"> - Adding tropical climate and soil types ● - Adding specific pesticides characteristics depending on tropical conditions and crops ● - Complementing substance data (e.g. half-lives) under tropical conditions ○ 		

“●” for aspects that require limited effort

“○” for aspects that require substantial model adaptations and additional research.

1.3.2.1 General limitations of the use of models

There are some general limitations of the discussed models, which reduce the possible extension of their validity to assess pesticides under tropical conditions. The uncertainty of toxicity impact results increases dramatically when generic or average values across pesticides belonging to a certain chemical family are used or when applying an averaged characterization factor across pesticides as proxy for certain substances (Basset-Mens et al. 2019). In emission and impact modelling, biological/natural substances, metal-based pesticides or inorganic substances are currently not included (Meier et al. 2015), such as copper pesticide (Peña et al. 2018). Likewise, active substances' metabolites, which may have even higher toxic effects than their parent compounds (e.g. diuron) are not accounted for in practice (Oturán et al. 2008).

Beyond the improvements relevant for each model, the interface between emission and impact characterization models is not perfect; as presented in Figure 6. More research is needed on the best way to deal with the soil, which could in fact belong to both the ecosphere and the technosphere, which affects both the modeling of emissions and related impacts on humans and ecosystems (van Zelm et al. 2014). Despite remaining research needs, preliminary recommendations have been proposed to achieve a coherent use of LCI and LCIA models for pesticides (Rosenbaum et al. 2015; Fantke et al. 2016, 2017a, 2018a). The recommendations mainly refer to the delineation between technosphere and ecosphere and what to include in LCI or LCIA in order to model pesticides in a consistent way. However, the adapted PestLCI model and the impact model USEtox have different spatiotemporal boundary systems (Dijkman et al. 2018). To avoid omissions or double counting of mass transport processes, only the primary pesticide distribution from the adapted PestLCI model was recommended to be used as direct input for the LCIA model USEtox and the recommended timeframe of the adapted PestLCI model to consider environmental processes was one day. This, however, means that processes beyond the considered timeframe are currently not considered in LCI emission models for pesticides. This affects for example leaching and run-off, two processes that are particularly relevant in tropical contexts (Racke et al. 1997). Instead, these rather long-term processes are currently considered in USEtox and dynamiCROP, which do not have the spatial level of disaggregation of the emission model to account for variations in soil and climate characteristics. Consequently, soil and climate characteristics influencing these processes are not taken into account properly, while the calculation of emission fractions is exclusively depending on drift curves and crop stage. One possible solution is to extend the temporal coverage of environmental processes in the emission modeling. Another possible solution is to spatialize the impact assessment modeling as recently proposed by Wannaz et al. (2018). Moreover, the use of the primary distribution only in the adapted PestLCI model to estimate pesticide emission fractions in a tropical context is even less relevant, since the drift curves are defined for temperate crops, climates and application methods. In conclusion, some of these recommendations do not allow a good consideration of pesticides in LCA, by excluding key factors influencing pesticide emissions.

1.3.2.2 Emission modeling for tropical conditions

The proposed improvements that were summarized in Table 3 are detailed in Table 4 according to the relevance of the processes for modelling pesticide emissions under tropical conditions. Adaptations and recommendations from previous case studies using PestLCI 1.0 and PestLCI 2.0 are also presented in Table 4. In open fields, PestLCI 2.0 has been tested only under temperate conditions by Dijkman (2013) on barley (*Hordeum vulgare*) and kiwi (*Actinidia deliciosa*), by Xue et al. (2015) on maize, by Renaud-Gentié et al. (2015) and Vázquez-Rowe et al. (2017) on vineyard. In tropical conditions, only PestLCI 1.0 has been tested by Ingwersen (2012) on pineapple. More generally, we recommend that PestLCI is made more user-friendly by e.g. running multiple scenarios in a single run and by offering an import/export function to common LCA software formats.

Extension of the validity domain of the model. First, the validity domain of the model needs to be extended to account for tropical crop type and cropping systems (e.g. multi-annual banana and sugar cane production and their associated farm management practices, crop morphology), and climate and soil characteristics. When tested in case studies, the model was generally adapted to local conditions (Table 4). A first analysis of the equations and parameters used in the emission model highlights that some parameters rely on fixed default or average values. As highlighted by Renaud-Gentié et al. (2015) based on a sensitivity analysis, some of these parameters have a strong effect on final results. In certain contexts, results might be sensitive to the fraction of continuous macropores in the soil (e.g. in vineyard soils Renaud-Gentié et al. 2015), to the volume fraction of water in soil, and to the fraction intercepted by leaves. Consequently, the Leaf Area Index and associated fraction intercepted by leaves should be adapted for all crops and growth stages, with a correspondence table as done by Linders et al. (2000) but including the main tropical crops. Furthermore, Renaud-Gentié et al. (2015), argue for the possibility to add the user's own dataset on climate and soil, because soil and climate characteristics have strong influences on results. Furthermore, some equations for important processes of pesticide distribution in tropical conditions are based on Danish circumstances as the calculation of the length of a rainfall event in case of macropores flow, which is used to estimate the leached fraction. A deeper understanding is necessary of how and to what degree certain aspects like temperature influence emission results, in order to extend the applicability of the model to tropical conditions. Further research is also required on modelling the emissions of metals and other inorganic substances. These substances can largely increase impact on freshwater ecosystems (Vázquez-Rowe et al. 2012), but another approach is required to integrate their specific behavior in the environment into the adapted PestLCI model (Renaud-Gentié et al. 2015a; Peña et al. 2018). Adapting the PestLCI model for special cropping systems is finally relevant, for example for flooded crops as rice, with specific pesticide field emissions, getting inspiration from models specifically parameterized for such cropping systems (Capri and Miao 2002; Inao et al. 2018).

Table 4: Factors influencing pesticide emissions under tropical conditions, and how they are currently considered in the adapted PestLCI model (as part of primary pesticide distribution or as part of secondary emissions) based on conclusions and improvements proposed by previous studies applying this model

Type	Aspect	Relevance for modelling pesticides in tropical regions (processes)	References for relevance of process	Integration in the adapted PestLCI model	Distribution in the adapted PestLCI model	References with case studies	
Agricultural practices	Application technique and target	Airborne emissions, volatilization and plant uptake	(García-Santos et al. 2016; Lewis et al. 2016)	Missing plant growth/stage No drift curve from tropical conditions	Primary (drift) - secondary	Adapted in (Renaud-Gentié et al. 2015a)	
	Ground cover (mulching, crops residues, cover crops etc.)	Sorption, dissipation at field scale and washoff from ground cover	(Mottes et al. 2014; Lewis et al. 2016)	Practices not integrated	Secondary	Developed and added in (Renaud-Gentié et al. 2015a)	
	Other farm management practices (e.g. tillage, buffer zone, biochar, drainage ditches)	Pesticide transport to water compartments	(Pereira-Junior et al. 2015; Mutua et al. 2016; Mendes et al. 2018)	Practices integrated: tillage, buffer zone and pipe drainage Practices not integrated: biochar, drainage ditches	Secondary		
Environmental characteristics	Climate	Temperature	Chemical and physical processes that change with temperature to different degrees (e.g. volatilization, degradation on leaf)	(Daam and van den Brink 2010)	Most of the processes are temperature-dependent (average temperature in the month of application)	Secondary	Adapted in (Ingwersen 2012; Xue et al. 2015; Renaud-Gentié et al. 2015a; Vázquez-Rowe et al. 2017)
		Rainfall	Pesticide transport outside the field by runoff, leaching, drainage, washoff	(Daam and van den Brink 2010)	No modelling of processes occurring after the first rainfall, Definition of rainfall event questionable	Secondary	
		Sunlight	Photodecomposition and volatilization	(Daam and van den Brink 2010)	Parameter not integrated in the processes	Secondary	
Soil properties (e.g. OC content, pH)	Soil adsorption and desorption, plant uptake, macropores flow	(Charlier et al. 2009; Lewis et al. 2016)	Parameters integrated in the model	Secondary			
Active substances	Chemical properties (e.g. DT50 soil, DT50 plant, solubility, Koc)	Plant uptake and transfer to water bodies	(Racke et al. 1997; Arias-Estévez et al. 2008; Sanchez-Bayo and Hyne 2011; Fantke and Juraske 2013; Fantke et al. 2016)	Variability of some chemical properties due to plant and environmental conditions not integrated (e.g. biological degradation level, temperature dependency, pesticide-plant species combinations)	Secondary	Completed for missing substances in (Xue et al. 2015; Renaud-Gentié et al. 2015a; Vázquez-Rowe et al. 2017)	
	Inorganic substances / metals	Integrate toxicity impact of these substances, need to evaluate distribution into the environment	(Peña et al. 2018)	No modelling of inorganic active substance	Primary – secondary	Issue highlighted in (Renaud-Gentié et al. 2015a)	

Primary drift distribution. In the primary distribution, the spray drift allows to estimate the fraction of pesticides dropped out of the field within few minutes after field application. This corresponds to the transfer by air of spray droplets out of the treated field, which could be on water surfaces, vegetation, soil or urbanized area. Current drift curves available in the adapted PestLCI model mainly consider German (Ganzelmeier et al. 1995) and Dutch (Holterman and van de Zande 2003) conditions. These drift curves were calibrated for crops present in these two countries and for the application methods prevalent in these countries, and more generally in Europe. While drift curves are mainly categorized according to application methods, several factors that vary widely between tropical and temperate conditions influence wind drift, such as crop density, soil conditions, and rain patterns. Hence, currently implemented drift curves focus on a temperate climate, defined as oceanic climate (Kottek et al. 2006). As one of the main application methods under tropical conditions, the knapsack sprayer has a drift curve available. It was calculated on potato (*Solanum tuberosum*) production with the IMAG calculator (from the Dutch model) in Boyacá, a highland region in Colombia (García-Santos et al. 2016), which corresponds to a temperate oceanic climate. Given that the main factors of the variation of spray drift are the application method and the nozzle's type, the climatic conditions (temperature, hygrometry, wind), the formulation of pesticide (e.g. powder, liquid) and his adjuvant, the composition and state of vegetation, and the farmers' use of equipment (Franke et al. 2010), the use of this drift curve in tropical conditions is questionable and should be done with caution. A literature review on drift in tropical conditions highlighted the low number of studies on pesticide drift under these conditions. Gouda et al. (2018) presented a drift experiment and calculated a drift curve for a knapsack sprayer on cotton (*Gossypium spp.*) production in Benin. Awadhwal et al. (1991) and Snelder et al. (2008) studied pesticide drift for a knapsack sprayer on rice production in the Philippines, and da Cunha et al. (2003) studied drift curves for an air blast sprayer on bean production in Brazil. Due to differences on the methodology of drift measurements, a more in-depth analysis of these studies is required to validate their potential integration into the adapted PestLCI model as additional drift curves for tropical conditions. Drift models should also be explored, since several could be relevant for tropical conditions. In conclusion, research is still needed on the mechanisms of pesticide drift during field application under tropical conditions. New drift curve estimates are required to best estimate pesticide field emission in these contexts.

Secondary emission distribution. In the adapted PestLCI model, water flows are not adequately modelled in secondary distribution estimates. After the first rainfall event, no more biophysical processes occur whereas, in the reality, runoff and leaching continue to occur several days or months after the pesticide has been applied depending on its persistence in the environment. This is particularly relevant under tropical conditions and must be considered, especially where farm management practices have an effect on pesticide transfers to water. Ground cover management is particularly critical in

tropical conditions. Weeds are present almost all year, soil moisture should be maintained in the dry season and soil erosion due to heavy rainfall events should be reduced as much as possible. As presented in Table 4, some farm management practices are already modelled in the adapted PestLCI model, as tillage, buffer zone and pipe drainage. However, other farm management practices, which also influence the mobility of pesticides to the environment, are not yet considered (see Table 4). PestLCI 2.0 was customized for viticulture to take into account the effect of cover plants (grass) between the rows of vines (*Vitis vinifera*) on pesticide distribution (Renaud-Gentié et al. 2015a). This adaptation is not yet included in the available adapted PestLCI model. This specific module could be extended to tropical productions, such as banana (Abbasi and Jamal 1999), with the possibility to choose the type of ground cover between rows by either a cover crop, a cover plant or a mulch. Other farm management practices have a strong influence on pesticide drift, such as the control of the tractor speed or the compliance of recommended climatic conditions to apply pesticides (Arvidsson et al. 2011). However, in general, this level of detail of information cannot be accounted for in LCA, and some uncertainty from the farmers practice remains. Furthermore, some practices already integrated in the model could be improved by adding more possibilities, such as drainage ditches. Finally, the diversity of cropping systems should be integrated in the model, such as intercropping.

1.3.2.3 (Eco-)toxicity characterization for tropical conditions

Built as a mechanistic model, USEtox is based on averaging conditions across all continents, yielding recommended factors for a generic average continent. However, while continental and sub-continental parameterizations are available, they are only recommended for sensitivity analysis and do not reflect the specific conditions of tropical regions, where e.g. the ecotoxicity effects on tropical species or temperature dependent processes are considered. More specifically, information on effects on tropical ecosystem species is required to assess pesticides in tropical conditions. When such information is missing, pesticides cannot be characterized, leading to a possible underestimation in results whenever such pesticides are used, but not considered in related impact scores (Vázquez-Rowe et al. 2017)

In Pennington et al. (2005), the authors highlighted that the uncertainty of toxicity characterization factors evaluated with a non-spatial model (IMPACT 2002) is at least of 2 to 3 orders of magnitude for some chemicals. As a consequence, Ingwersen (2012) proposed to customize USEtox to the Costa Rican environment in his LCA study of fresh pineapple. USEtox could use spatial differentiation considering tropical conditions and ecosystems (e.g. mangrove) on the one hand and temperate conditions on the other, as already proposed in other spatial differentiation models (e.g. Wannaz et al. 2018). However, the benefit of applying spatial differentiation impact assessment models relies on the availability of spatial data for underlying input parameters, such as species distribution, life cycle emissions and background exposure levels, which is often not available in LCA studies with a tropical context. Given

that in many LCA studies, spatially explicit data are often not available, current models are not primarily designed for regionalized toxicity characterization. In cases, where spatial data are becoming available, the models should be adapted to account for spatial aspects, which is discussed e.g. in Peña et al. (2018, 2019). More generally, for all pedo-climatic contexts, some impacts are not adequately taken into account or not included at all in some models, such as terrestrial and marine ecotoxicity (Notarnicola et al. 2017). As recommended by Fantke et al. (2018), efforts are required on on-field impacts and missing impact categories including terrestrial/pollinator ecotoxicity; especially so in tropical conditions (Brown 2014). USEtox could have a groundwater compartment to better distribute emission fractions from the adapted PestLCI model to surface water and groundwater, currently fully allocated to the freshwater compartment by default (Fantke et al. 2018a) (see Figure 6), but also to take into account ecotoxicity in groundwater where biodiversity is specific (Danielopol et al. 2000). Whereas groundwater is relevant in most climate conditions, it plays a particular role in tropical contexts with plenty of rain and high water flow rates. Although a different sensitivity of tropical species has been demonstrated for some active substances compared to that of temperate species studies (Kwok et al. 2007; Daam and van den Brink 2010), ecotoxicological data for tropical species is scarce and current knowledge in ecotoxicology mainly comes from Europe and North America with temperate conditions (Kwok et al. 2007). The few data available on tropical species sensitivity to pesticides (Alves et al. 2013; Arias-Andrés et al. 2018; Daam and Rico 2018) should be integrated into the model. However, Kwok et al. (2007), Daam and van den Brink (2010), and Leboulanger et al. (2011) highlighted the need for further development of toxicity tests with indigenous species in tropical conditions. In priority, for a better estimation of pesticide environmental impacts in tropical conditions, further research on indigenous species in tropical conditions and organisms' sensitivity to pesticides from tropical origins is required. Some pesticides exposure pathways in agriculture are missing as occupational exposure (when preparing and applying pesticides) (Ingwersen 2012), residential bystanders and family of exposed workers (Ryberg et al. 2018). This is of particular concern in tropical conditions, where human exposure to pesticides might be higher because of the proximity of dwellings to treated plots, the mainly manual use of pesticides without, most of the time, personal protective equipment and skills to apply them, and the frequent storage of pesticides in households (Williamson et al. 2008). A framework for assessing residential bystander exposure to field pesticide applications (potatoes) in LCIA has been recently presented by Ryberg et al. (2018). However, this work must be further adapted and extended to additional tropical crops and application methods, in particular to hand-operated spraying, which is important under tropical conditions. Other sources of pesticide exposure and contamination could be added as post-harvested treatment and seed treatment, that can have environmental impact (e.g. seed treatment on terrestrial ecosystem - worms Alves et al. 2013) and impacts on human health (e.g. pesticide residues from post-harvest treatments Bajwa and Sandhu 2014).

1.3.2.4 Crop residue modeling for tropical conditions

In dynamiCROP, the development of a user interface to change easily default values on climate data (e.g. average relative humidity, mean temperature in air, precipitation rate during wet period), soil data (e.g. pH, soil organic carbon content), and plant characteristics (e.g. leaf area index, density of fruit) would be useful to optimize the use of dynamiCROP for tropical (and other) contexts. Furthermore, important crop archetypes relevant for tropical contexts are missing as for example banana or soybean production. As part of a study on passion fruits cultivated in Colombia (Juraske et al. 2012), the passion fruit crop model has been parameterized into dynamiCROP, and constitutes the unique tropical crop. Archetypes already modelled should be used carefully for assessments focusing on tropical conditions, because crop growth and varieties are different and could imply differences in pesticide distribution in the modeled plant-environment system. Hence, plant characteristics should always be checked prior to their use in related models. For example, to model the uptake of pesticides by taro (*Colocasia esculenta*) crops (a tuber), currently the user must choose the potato archetype, whereas the crops' family and farm management practices are widely different from taro. Currently, data for pesticide dissipation half-lives in plants can be estimated based on Fantke et al. (2014), where dissipation from plant is influenced by active substance properties, plant characteristics and environmental conditions (mainly temperature). However, differences in rain, sunlight and other conditions relevant for tropical regions are currently not considered in such estimates and, hence, further research is required to measure half-lives on plants for tropical crops and conditions. Furthermore, the estimation of human intake due to the late application of pesticides before harvest should be improved. This is particularly important in many tropical agricultural productions, where harvesting takes place throughout the year with plants and fruits/legumes at different stages of maturity in the same plot (e.g. vegetable production). Finally, dynamiCROP has never been used in combination with the adapted PestLCI model. The calculation chain between both models could be achieved by using the fraction of drift and volatilization in the adapted PestLCI model to refine the respective fixed fractions lost to air per crop in dynamiCROP. With this step, we could have a consistently estimated fraction of pesticide reaching the plant. The model is currently tested in combination with the adapted PestLCI model and USEtox on open-field tomato production in tropical soils and climates, in the Martinique Island. When related results become available, recommendations are expected how to best align the combined use of the three discussed models.

1.4 Conclusions

The present study showed that processes driving pesticide emission and impact patterns under tropical conditions are specific in relation to soil, climate, cropping practices and crops. The three most up-to-date and consensual LCI and LCIA models commonly reflect temperate conditions for climate, soil, application techniques and crops and are not yet suited for the evaluation of the impacts of

pesticides in LCA of crops cultivated in tropical regions. Under tropical conditions, higher temperatures, sunlight radiation, and microbial activity enhance degradation and volatilization of pesticides. Heavy and frequent rainfall events lead to higher leaching and runoff. Tropical crops, cropping systems and practices also widely differ as compared to temperate regions in relation to natural and human drivers, and we demonstrated how these aspects can alter the transfers and impacts of pesticides. In developing countries with a tropical climate, pesticides are most often applied manually or with a hand operated sprayer. Farmers generally have less awareness of pesticides' danger and less training on good practices of application leading to higher risks of transfers and impacts on the environment and human health. Under tropical conditions, certain practices, such as ground cover management, can play a major role in the transfer of pesticides to the water compartment by affecting the soil hydraulic processes. Furthermore, biodiversity is naturally higher in tropical regions and preliminary research revealed a different sensitivity of tropical species to pesticides compared to species in temperate regions, while in most ecotoxicity experiments they are currently not represented.

We provided a set of recommendations to better account for the specificities of pesticide emissions and impacts in tropical conditions in the three discussed models. Databases need to be extended to integrate tropical crops characteristics, soil and climate specificities for tropical conditions, and active substances' chemical characteristics. In the primary distribution of the adapted PestLCI model, the addition of specific drift curves for tropical conditions has the potential to make a difference on the results and to reduce uncertainty. In the secondary emission distribution, specific features of tropical cropping systems and farm management practices should be included, especially ground cover management (e.g. mulching) in combination with a better accounting of the leaching process over time. In dynamiCROP, the development of a user interface to change easily default values on climate, soil and plant characteristics would be useful and important crop archetypes for tropical conditions should be added. The estimation of human intake due to the late application of pesticides before harvest should also be improved to account for characteristics of cropping systems in the tropics.

Additional perspectives for the future include the need to test in combination PestLCI, USEtox and dynamiCROP in real LCA case studies under tropical conditions, in order to contribute to their better understanding and parametrization.

In summary, emission and impact evaluation processes are not as well understood for tropical conditions as they are for temperate conditions, with fewer measurements available for the former. Further experimental research in these contexts is therefore urgently needed. In the adapted PestLCI model, mechanisms of pesticide drift during field application, according to tropical conditions and applications methods and mechanisms to account for metals and other inorganic substances should be explored and included. Research on organisms' sensitivity to pesticides from tropical origins is also essential. In current impact models, different exposure populations (consumers, workers and bystanders)

and effects on relevant organisms (freshwater, marine, terrestrial, pollinators and birds) should progressively be modelled. Reflection and consensus on the conceptual framework of the LCI-LCIA calculation chain and the soil's belonging to ecosphere or technosphere are needed for a better consistency of the modelling of pesticide emissions and impacts in LCA studies focusing on tropical conditions. Current model limitations highlighted in our study are a useful starting point for focusing future research and model refinement efforts, with the aim to help reducing uncertainty in LCA results representing tropical conditions.

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References Chapter 1

- Abbasi MA, Jamal T (1999) Soil loss and runoff measurement from banana-pineapple intercropping system. *Pakistan Journal of Biological Sciences* 2:689–692. doi.org/10.3923/pjbs.1999.689.692
- Aktar MW, Sengupta D, Chowdhury A (2009) Impact of pesticides use in agriculture: their benefits and hazards. *Interdiscip Toxicol* 2:1–12. doi.org/10.2478/v10102-009-0001-7
- Alves GS, da Cunha JPAR (2014) Field data and prediction models of pesticide spray drift on coffee crop. *Pesquisa Agropecuária Brasileira* 49:622–629. doi.org/10.1590/S0100-204X2014000800006
- Alves PRL, Cardoso EJBN, Martines AM, et al (2013) Earthworm ecotoxicological assessments of pesticides used to treat seeds under tropical conditions. *Chemosphere* 90:2674–2682. doi.org/10.1016/j.chemosphere.2012.11.046
- Antón A, Castells F, Montero JI, Huijbregts M (2004) Comparison of toxicological impacts of integrated and chemical pest management in Mediterranean greenhouses. *Chemosphere* 54:1225–1235. doi.org/10.1016/j.chemosphere.2003.10.018
- Antón A, Torrellas M, Núñez M, et al (2014) Improvement of Agricultural Life Cycle Assessment Studies through Spatial Differentiation and New Impact Categories: Case Study on Greenhouse Tomato Production. *Environ Sci Technol* 48:9454–9462. doi.org/10.1021/es501474y
- Arias-Andrés M, Rämö R, Torres FM, et al (2018) Lower tier toxicity risk assessment of agriculture pesticides detected on the Río Madre de Dios watershed, Costa Rica. *Environ Sci Pollut Res* 25:13312–13321. doi.org/10.1007/s11356-016-7875-7

- Arias-Estévez M, López-Periago E, Martínez-Carballo E, et al (2008) The mobility and degradation of pesticides in soils and the pollution of groundwater resources. *Agri Ecosyst Environ* 123:247–260. doi.org/10.1016/j.agee.2007.07.011
- Arvidsson T, Bergström L, Kreuger J (2011) Spray drift as influenced by meteorological and technical factors. *Pest Manag Sci* 67:586–598. doi.org/10.1002/ps.2114
- Awadhwal NK, Cabrido EF, Quick GR (1991) Minimizing drift and exposure from knapsack sprayers. *J Environ Sci Health B* 26:589–600. doi.org/10.1080/03601239109372758
- Bajwa U, Sandhu KS (2014) Effect of handling and processing on pesticide residues in food- a review. *J Food Sci Technol* 51:201–220. doi.org/10.1007/s13197-011-0499-5
- Basset-Mens C, Edewa A, Gentil C (2019) An LCA of French beans from Kenya for decision-makers. *Indonesien Journal of Life Cycle Assessment and Sustainability* (in press)
- Beck HE, Zimmermann NE, McVicar TR, et al (2018) Present and future Köppen-Geiger climate classification maps at 1-km resolution. *Scientific Data* 5:180214. doi.org/10.1038/sdata.2018.214
- Berthoud A, Maupu P, Huet C, Poupart A (2011) Assessing freshwater ecotoxicity of agricultural products in life cycle assessment (LCA): a case study of wheat using French agricultural practices databases and USEtox model. *Int J Life Cycle Assess* 16:841. doi.org/10.1007/s11367-011-0321-7
- Bessou C, Basset-Mens C, Tran T, Benoist A (2013) LCA applied to perennial cropping systems: a review focused on the farm stage. *Int J Life Cycle Assess* 18:340–361. doi.org/10.1007/s11367-012-0502-z
- Biénabe E, Rival A, Loeillet D (2016) *Développement durable et filières tropicales*, Quae. Versailles, France
- Birkved M, Hauschild MZ (2006) PestLCI—A model for estimating field emissions of pesticides in agricultural LCA. *Ecol Model* 198:433–451. doi.org/10.1016/j.ecolmodel.2006.05.035
- Bocquené G, Franco A (2005) Pesticide contamination of the coastline of Martinique. *Mar Pollut Bull* 51:612–619. doi.org/10.1016/j.marpolbul.2005.06.026
- Bojacá CR, Gil R, Casilimas H, et al (2012) Modelling the environmental impact of pesticides sprayed on greenhouse tomatoes: a regional case study in Colombia. *Acta Hort* 61–68. doi.org/10.17660/ActaHortic.2012.957.6
- Brown JH (2014) Why are there so many species in the tropics? *J Biogeogr* 41:8–22. doi.org/10.1111/jbi.12228
- Bueno MR, da Cunha JPAR, de Santana DG (2017) Assessment of spray drift from pesticide applications in soybean crops. *Biosyst Eng* 154:35–45. doi.org/10.1016/j.biosystemseng.2016.10.017
- Capri E, Miao Z (2002) Modelling pesticide fate in rice paddy. *Agronomie* 22:363–371. doi.org/10.1051/agro:2002020
- Castillo LE, de la Cruz E, Ruepert C (1996) Ecotoxicology and pesticides in tropical aquatic ecosystems of Central America. *Environ Toxicol Chem* 16:41–51. doi.org/10.1002/etc.5620160104

- Charlier J-B, Cattan P, Voltz M, Moussa R (2009) Transport of a nematicide in surface and groundwaters in a tropical volcanic catchment. *J Environ Qual* 38:1031–1041. doi.org/10.2134/jeq2008.0355
- da Cunha JPAR, Teixeira MM, Coury JR, Ferreira LR (2003) Evaluation of strategies to reduce pesticide spray drift. *Planta Daninha* 21:325–332. doi.org/10.1590/S0100-83582003000200019
- Daam MA, Rico A (2018) Freshwater shrimps as sensitive test species for the risk assessment of pesticides in the tropics. *Environ Sci Pollut Res* 25:13235–13243. doi.org/10.1007/s11356-016-7451-1
- Daam MA, van den Brink PJ (2010) Implications of differences between temperate and tropical freshwater ecosystems for the ecological risk assessment of pesticides. *Ecotoxicology* 19:24–37. doi.org/10.1007/s10646-009-0402-6
- Danielopol DL, Pospisil P, Rouch R (2000) Biodiversity in groundwater: a large-scale view. *Trends Ecol Evol* 15:223–224. doi.org/10.1016/S0169-5347(00)01868-1
- Dickinson NM, Lepp NW (1984) Pollution of tropical plantation crops by copper fungicides: a copper budget for Kenyan coffee plantation. *Stud Environ Sci* 341–346
- Dijkman TJ (2013) Modelling of pesticide emissions for Life Cycle Inventory analysis: Model development, applications and implications. Dissertation, Department of Management Engineering, Technical University of Denmark
- Dijkman TJ, Basset-Mens C, Antón A, Núñez M (2018) LCA of Food and Agriculture. In: Hauschild MZ (ed) *Life Cycle Assessment - Theory and Practice*. Springer, Cham, Switzerland, pp 723–754
- Dijkman TJ, Birkved M, Hauschild MZ (2012) PestLCI 2.0: a second generation model for estimating emissions of pesticides from arable land in LCA. *Int J Life Cycle Assess* 17:973–986. doi.org/10.1007/s11367-012-0439-2
- Dijkman TJ, Birkved M, Saxe H, et al (2017) Environmental impacts of barley cultivation under current and future climatic conditions. *J Clean Prod* 140:644–653. doi.org/10.1016/j.jclepro.2016.05.154
- Elibariki R, Maguta MM (2017) Status of pesticides pollution in Tanzania – A review. *Chemosphere* 178:154–164. doi.org/10.1016/j.chemosphere.2017.03.036
- Fantin V, Buscaroli A, Dijkman T, et al (2019) PestLCI 2.0 sensitivity to soil variations for the evaluation of pesticide distribution in Life Cycle Assessment studies. *Sci Total Environ* 656:1021–1031. doi.org/10.1016/j.scitotenv.2018.11.204
- Fantke P (2019) Modeling the environmental impacts of pesticides in agriculture. In: Weidema, B.P. (Ed) *Assessing the environmental impact of agriculture*, Burleigh Dodds Science Publishing, Cambridge, United Kingdom. doi.org/10.19103/AS.2018.0044.08
- Fantke P, Antón A, Grant T, Hayashi K (2017a) Pesticide emission quantification for life cycle assessment: A global consensus building process. *Journal of Life Cycle Assessment, Japan* 13:245–251
- Fantke P, Arnot JA, Doucette WJ (2016) Improving plant bioaccumulation science through consistent reporting of experimental data. *J Environ Manage* 181:374–384. doi.org/10.1016/j.jenvman.2016.06.065

- Fantke P, Aurisano N, Bare J, et al (2018) Toward harmonizing ecotoxicity characterization in life cycle impact assessment. *Environ Toxicol Chem* 37:2955–2971. doi.org/10.1002/etc.4261
- Fantke P, Bijster M, Guignard C, et al (2017b) USEtox® 2.0 Documentation (Version1)
- Fantke P, Charles R, Alencastro LF de, et al (2011a) Plant uptake of pesticides and human health: Dynamic modeling of residues in wheat and ingestion intake. *Chemosphere* 85:1639–1647. doi.org/10.1016/j.chemosphere.2011.08.030
- Fantke P, Friedrich R, Jolliet O (2012a) Health impact and damage cost assessment of pesticides in Europe. *Environ Int* 49:9–17. doi.org/10.1016/j.envint.2012.08.001
- Fantke P, Gillespie BW, Juraske R, Jolliet O (2014) Estimating half-lives for pesticide dissipation from plants. *Environ Sci Technol* 48:8588–8602. doi.org/10.1021/es500434p
- Fantke P, Jolliet O (2016) Life cycle human health impacts of 875 pesticides. *Int J Life Cycle Assess* 21:722–733. doi.org/10.1007/s11367-015-0910-y
- Fantke P, Juraske R (2013) Variability of pesticide dissipation half-lives in plants. *Environ Sci Technol* 47:3548–3562. doi.org/10.1021/es303525x
- Fantke P, Juraske R, Antón A, et al (2011b) Dynamic multicrop model to characterize impacts of pesticides in food. *Environ Sci Technol* 45:8842–8849. doi.org/10.1021/es201989d
- Fantke P, Wieland P, Juraske R, et al (2012b) Parameterization models for pesticide exposure via crop consumption. *Environ Sci Technol* 46:12864–12872. doi.org/10.1021/es301509u
- Fantke P, Wieland P, Wannaz C, et al (2013) Dynamics of pesticide uptake into plants: From system functioning to parsimonious modeling. *Environ Modell Softw* 40:316–324. doi.org/10.1016/j.envsoft.2012.09.016
- FAOSTAT (2019) FAOSTAT. In: FAOSTAT. <http://www.fao.org/faostat/en/#home>. Accessed 15 Jan 2019
- Feng X, Wang K, Pan L, et al (2018) Measured and modeled residue dynamics of famoxadone and oxathiapiprolin in tomato fields. *J Agric Food Chem* 66:8489–8495. doi.org/10.1021/acs.jafc.8b02056
- Franke AC, Kempenaar C, Holterman HJ, van de Zande JC (2010) Spray drift from knapsack sprayers, A study conducted within the framework of the Sino-Dutch Pesticide Environmental Risk Assessment Project PERAP. Plant Research International B.V., Wageningen
- Ganzelmeier H, Rautmann D, Spangenberg R, et al (1995) Studies on the spray drift of plant protection products. Federal Biological Research Centre for Agriculture and Forestry, Berlin
- García-Santos G, Feola G, Nuyttens D, Diaz J (2016) Drift from the Use of Hand-Held Knapsack Pesticide Sprayers in Boyacá (Colombian Andes). *J Agric Food Chem* 64:3990–3998. doi.org/10.1021/acs.jafc.5b03772
- Gouda AI, Mehoba MHL, Toko II, et al (2018) Comparison of drift of two types of sprayers used in cotton production in Benin. *Biotechnol Agron Soc* 22:94–105
- Hauschild MZ (2000) Estimating pesticide emissions for LCA of agricultural products. In: Weidema BP (ed) *Agricultural data for life cycle assessments*. Agricultural Economics Research Institute (LEI), The Hague, pp 64–79

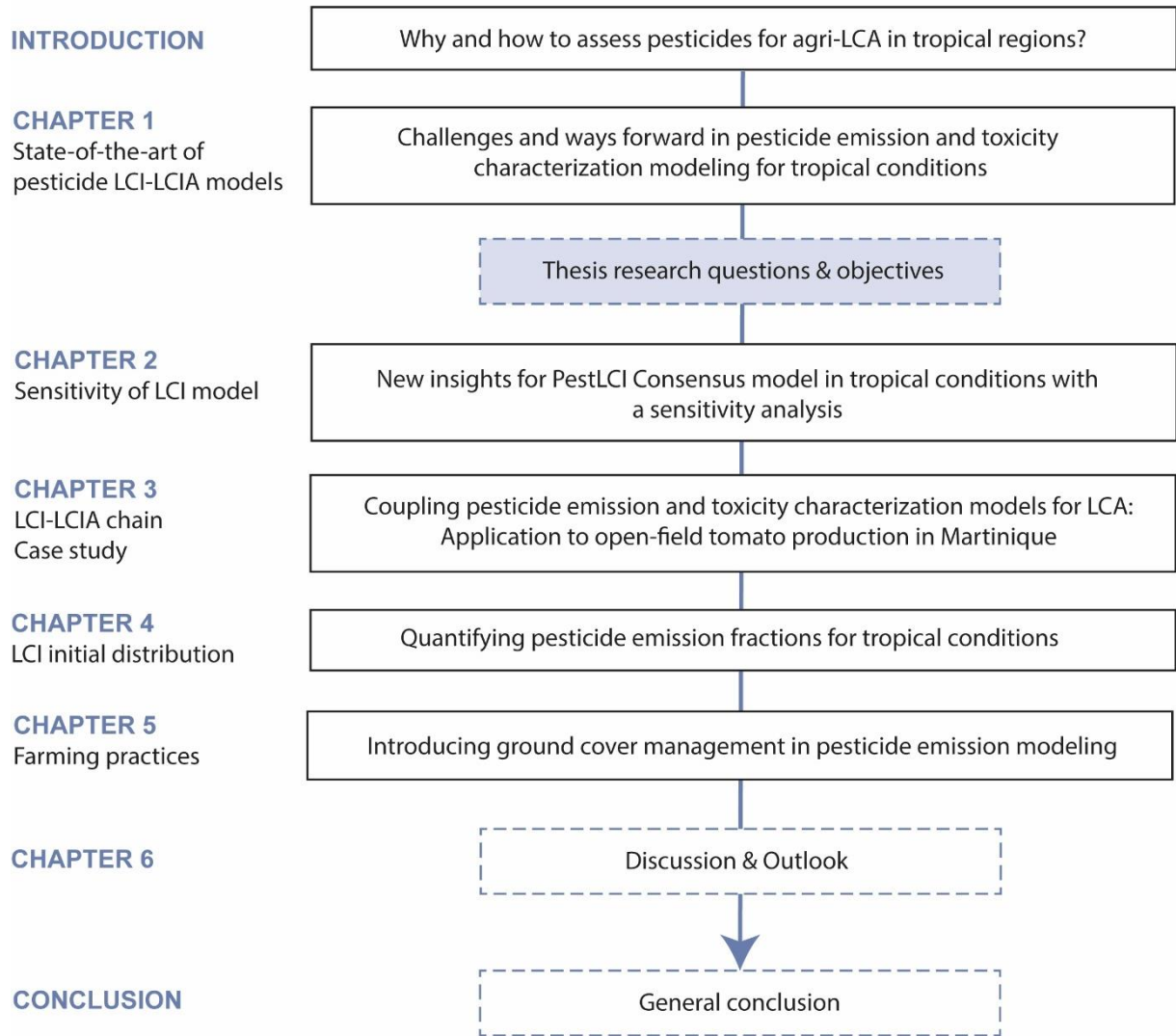
- Hauschild MZ, Goedkoop M, Guinée J, et al (2013) Identifying best existing practice for characterization modeling in life cycle impact assessment. *Int J Life Cycle Assess* 18:683–697. doi.org/10.1007/s11367-012-0489-5
- Henderson AD, Hauschild MZ, van de Meent D, et al (2011) USEtox fate and ecotoxicity factors for comparative assessment of toxic emissions in life cycle analysis: sensitivity to key chemical properties. *Int J Life Cycle Assess* 16:701. doi.org/10.1007/s11367-011-0294-6
- Holterman HJ, van de Zande J. (2003) IMAG Drift Calculator v1.1: user manual. http://www.pesticidemodels.eu/sites/default/files/documents/IDCmanual_0.pdf. Accessed 28 Dec 2018
- Hossain MS, Fakhruddin ANM, Chowdhury MAZ, et al (2015) Health risk assessment of selected pesticide residues in locally produced vegetables of Bangladesh. *International Food Research Journal* 22:110–115
- Houbraken M, Habimana V, Senaeve D, et al (2017) Multi-residue determination and ecological risk assessment of pesticides in the lakes of Rwanda. *Sci Total Environ* 576:888–894. doi.org/10.1016/j.scitotenv.2016.10.127
- Hunt ND, Hill JD, Liebman M (2017) Reducing freshwater toxicity while maintaining weed control, profits, and productivity: effects of increased crop rotation diversity and reduced herbicide usage. *Environ Sci Technol* 51:1707–1717. doi.org/10.1021/acs.est.6b04086
- Inao K, Iwafune T, Horio T (2018) An improved PADDY model including uptake by rice roots to predict pesticide behavior in paddy fields under nursery-box and submerged applications. *J Pestic Sci* 43:142–152. doi.org/10.1584/jpestics.D17-084
- Ingwersen WW (2012) Life cycle assessment of fresh pineapple from Costa Rica. *J Clean Prod* 35:152–163. doi.org/10.1016/j.jclepro.2012.05.035
- Itoiz ES, Fantke P, Juraske R, et al (2012) Deposition and residues of azoxystrobin and imidacloprid on greenhouse lettuce with implications for human consumption. *Chemosphere* 89:1034–1041. doi.org/10.1016/j.chemosphere.2012.05.066
- Jacobsen RE, Fantke P, Trapp S (2015) Analysing half-lives for pesticide dissipation in plants. *SAR QSAR Environ Res* 26:325–342. doi.org/10.1080/1062936X.2015.1034772
- Juraske R, Fantke P, Ramírez ACR, González A (2012) Pesticide residue dynamics in passion fruits: Comparing field trial and modelling results. *Chemosphere* 89:850–855. doi.org/10.1016/j.chemosphere.2012.05.007
- Juraske R, Mutel CL, Stoessel F, Hellweg S (2009) Life cycle human toxicity assessment of pesticides: Comparing fruit and vegetable diets in Switzerland and the United States. *Chemosphere* 77:939–945. doi.org/10.1016/j.chemosphere.2009.08.006
- Juraske R, Vivas CSM, Velásquez AE, et al (2011) Pesticide uptake in potatoes: model and field experiments. *Environ Sci Technol* 45:651–657. doi.org/10.1021/es102907v
- Kariathi V, Kassim N, Kimanya M (2016) Pesticide exposure from fresh tomatoes and its relationship with pesticide application practices in Meru district. *Cogent Food & Agriculture* 2:1196808. doi.org/10.1080/23311932.2016.1196808
- Kookana RS (2010) The role of biochar in modifying the environmental fate, bioavailability, and efficacy of pesticides in soils: a review. *Soil Res* 48:627–637. doi.org/10.1071/SR10007

- Kottek M, Grieser J, Beck C, et al (2006) World Map of the Köppen-Geiger climate classification updated. *Meteorol Z* 15:259–263. doi.org/10.1127/0941-2948/2006/0130
- Kwok KW, Leung KM, Lui GS, et al (2007) Comparison of tropical and temperate freshwater animal species' acute sensitivities to chemicals: Implications for deriving safe extrapolation factors. *Integr Environ Asses* 3:49–67. doi.org/10.1002/ieam.5630030105
- Laplace D (2018) L'usage des phytosanitaires dans le bassin amazonien : entre problèmes spécifiques à l'agriculture en zone tropicale et fragilité de l'écosystème. *Innovations Agronomiques* 1–10. doi.org/dx.doi.org/10.15454/1.5407989444908796E12
- Leboulanger C, Schwartz C, Somville P, et al (2011) Sensitivity of two mesocyclops (Crustacea, Copepoda, Cyclopidae), from tropical and temperate origins, to the herbicides, Diuron and Paraquat, and the insecticides, Temephos and Fenitrothion. *B Environ Contam Tox* 87:487–493. doi.org/10.1007/s00128-011-0406-9
- Lewis SE, Silburn DM, Kookana RS, Shaw M (2016) Pesticide Behavior, Fate, and Effects in the Tropics: An Overview of the Current State of Knowledge. *J Agric Food Chem* 64:3917–3924. doi.org/10.1021/acs.jafc.6b01320
- Linders J, Mensink H, Stephenson G, et al (2000) Foliar interception and retention values after pesticide application: a proposal for standardised values for environmental risk assessment. *Pest Manag Sci* 58:315–315. doi.org/10.1002/ps.448
- Matthews G, Wiles T, Baleguel P (2003) A survey of pesticide application in Cameroon. *Crop Prot* 22:707–714. doi.org/10.1016/S0261-2194(03)00008-5
- Meier MS, Stoessel F, Jungbluth N, et al (2015) Environmental impacts of organic and conventional agricultural products – Are the differences captured by life cycle assessment? *J Environ Manag* 149:193–208. doi.org/10.1016/j.jenvman.2014.10.006
- Mendes KF, Hall KE, Takeshita V, et al (2018) Animal bonechar increases sorption and decreases leaching potential of aminocyclopyrachlor and mesotrione in a tropical soil. *Geoderma* 316:11–18. doi.org/10.1016/j.geoderma.2017.12.017
- Mitoko GJAO (1997) Occupational pesticide exposure among Kenyan agricultural workers: an epidemiological and public health perspective. Ohayo-Mitoko
- Montes AM, González-Farías FA, Botello AV (2012) Pollution by organochlorine pesticides in Navachiste-Macapule, Sinaloa, Mexico. *Environ Monit Assess* 184:1359–1369. doi.org/10.1007/s10661-011-2046-2
- Mottes C, Lesueur Jannoyer M, Le Bail M, et al (2017) Relationships between past and present pesticide applications and pollution at a watershed outlet: The case of a horticultural catchment in Martinique, French West Indies. *Chemosphere* 184:762–773. doi.org/10.1016/j.chemosphere.2017.06.061
- Mottes C, Lesueur-Jannoyer M, Bail ML, Malézieux E (2014) Pesticide transfer models in crop and watershed systems: a review. *Agron Sustain Dev* 34:229–250. doi.org/10.1007/s13593-013-0176-3
- Mutua GK, Ngigi AN, Getenga ZM (2016) Degradation characteristics of metribuzin in soils within the Nzoia River Drainage Basin, Kenya. *Toxicol Environ Chem* 98:800–813. doi.org/10.1080/02772248.2015.1128938

- Nemecek T, Schnetzer J (2011) Methods of assessment of direct field emissions for LCIs of agricultural production systems. Data v3.0. Agroscope, Zurich
- Nordborg M, Cederberg C, Berndes G (2014) Modeling potential freshwater ecotoxicity impacts due to pesticide use in biofuel feedstock production: the cases of maize, rapeseed, salix, soybean, sugar cane, and wheat. *Environ Sci Technol* 48:11379–11388. doi.org/10.1021/es502497p
- Nordborg M, Davis J, Cederberg C, Woodhouse A (2017) Freshwater ecotoxicity impacts from pesticide use in animal and vegetable foods produced in Sweden. *Sci Total Environ* 581–582:448–459. doi.org/10.1016/j.scitotenv.2016.12.153
- Notarnicola B, Sala S, Antón A, et al (2017) The role of life cycle assessment in supporting sustainable agri-food systems: A review of the challenges. *J Clean Prod* 140:399–409. doi.org/10.1016/j.jclepro.2016.06.071
- Oturan N, Trajkovska S, Oturan MA, et al (2008) Study of the toxicity of diuron and its metabolites formed in aqueous medium during application of the electrochemical advanced oxidation process “electro-Fenton.” *Chemosphere* 73:1550–1556. doi.org/10.1016/j.chemosphere.2008.07.082
- Peña N, Antón A, Kamilaris A, Fantke P (2018) Modeling ecotoxicity impacts in vineyard production: Addressing spatial differentiation for copper fungicides. *Sci Total Environ* 616–617:796–804. doi.org/10.1016/j.scitotenv.2017.10.243
- Pennington DW, Margni M, Ammann C, Jolliet O (2005) Multimedia fate and human intake modeling: spatial versus nonspatial insights for chemical emissions in Western Europe. *Environ Sci Technol* 39:1119–1128. doi.org/10.1021/es034598x
- Pereira-Junior EV, Giori FG, Nascimento AL, et al (2015) Effects of soil attributes and straw accumulation on the sorption of hexazinone and tebuthiuron in tropical soils cultivated with sugarcane. *J Environ Sci Health B* 50:238–246. doi.org/10.1080/03601234.2015.999588
- Peters EC, Gassman NJ, Firman JC, et al (1997) Ecotoxicology of tropical marine ecosystems. *Environ Toxicol Chem* 16:12–40. doi.org/10.1002/etc.5620160103
- Pillai MKK (1986) Pesticide pollution of soil, water and air in Delhi area, India. *Sci Total Environ* 55:321–327. doi.org/10.1016/0048-9697(86)90189-0
- Pouokam GB, Lemnyuy Album W, Ndikontar AS, Sidatt MEH (2017) A pilot study in Cameroon to understand safe uses of pesticides in agriculture, risk factors for farmers’ exposure and Management of Accidental Cases. *Toxics* 5:. doi.org/10.3390/toxics5040030
- Racke KD, Skidmore MW, Hamilton DJ, et al (1997) Pesticides report .38. Pesticide fate in tropical soils - (Technical report). *Pure Appl Chem* 69:1349–1371. doi.org/10.1351/pac199769061349
- Raksanam B, Taneepanichskul S, Siriwong W, Robson MG (2012) Factors associated with pesticide risk behaviors among rice farmers in rural community, Thailand. *Journal of Environment and Earth Science* 2:32–39. doi.org/doi:10.7282/T30Z71NB
- Renaud-Gentié C, Dijkman TJ, Bjørn A, Birkved M (2015) Pesticide emission modelling and freshwater ecotoxicity assessment for Grapevine LCA: adaptation of PestLCI 2.0 to viticulture. *Int J Life Cycle Assess* 20:1528–1543. doi.org/10.1007/s11367-015-0949-9
- Rosenbaum RK, Antón A, Bengoa X, et al (2015) The Glasgow consensus on the delineation between pesticide emission inventory and impact assessment for LCA. *Int J Life Cycle Assess* 20:765–776. doi.org/10.1007/s11367-015-0871-1

- Rosenbaum RK, Bachmann TM, Gold LS, et al (2008) USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment. *Int J Life Cycle Assess* 13:532. doi.org/10.1007/s11367-008-0038-4
- Rosenbaum RK, Huijbregts MAJ, Henderson AD, et al (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. doi.org/10.1007/s11367-011-0316-4
- Ryberg MW, Rosenbaum RK, Mosqueron L, Fantke P (2018) Addressing bystander exposure to agricultural pesticides in life cycle impact assessment. *Chemosphere* 197:541–549. doi.org/10.1016/j.chemosphere.2018.01.088
- Salomone R, Ioppolo G (2012) Environmental impacts of olive oil production: a Life Cycle Assessment case study in the province of Messina (Sicily). *J Clean Prod* 28:88–100. doi.org/10.1016/j.jclepro.2011.10.004
- Sanchez-Bayo F, Hyne RV (2011) Comparison of environmental risks of pesticides between tropical and nontropical regions. *Integr Environ Assess Manag* 7:577–586. doi.org/10.1002/ieam.189
- Sansoulet J, Cabidoche YM, Cattani P (2007) Adsorption and transport of nitrate and potassium in an Andosol under banana (Guadeloupe, French West Indies). *Eur J Soil Biol* 58:478–489. doi.org/10.1111/j.1365-2389.2007.00904.x
- Settle W, Soumare M, Sarr M, et al (2014) Reducing pesticide risks to farming communities: cotton farmer field schools in Mali. *Philosophical Transactions of the Royal Society B Biological Sciences* 369:20120277. doi.org/10.1098/rstb.2012.0277
- Snelder DJ, Masipiqueña MD, de Snoo GR (2008) Risk assessment of pesticide usage by smallholder farmers in the Cagayan Valley (Philippines). *Crop Prot* 27:747–762. doi.org/10.1016/j.cropro.2007.10.011
- van Zelm R, Larrey-Lassalle P, Roux P (2014) Bridging the gap between life cycle inventory and impact assessment for toxicological assessments of pesticides used in crop production. *Chemosphere* 100:175–181. doi.org/10.1016/j.chemosphere.2013.11.037
- Vázquez-Rowe I, Torres-García JR, Cáceres AL, et al (2017) Assessing the magnitude of potential environmental impacts related to water and toxicity in the Peruvian hyper-arid coast: A case study for the cultivation of grapes for pisco production. *Sci Total Environ* 601–602:532–542. doi.org/10.1016/j.scitotenv.2017.05.221
- Vázquez-Rowe I, Villanueva-Rey P, Moreira MT, Feijoo G (2012) Environmental analysis of Ribeiro wine from a timeline perspective: Harvest year matters when reporting environmental impacts. *J Environ Manag* 98:73–83. doi.org/10.1016/j.jenvman.2011.12.009
- Wannaz C, Fantke P, Lane J, Jolliet O (2018) Source-to-exposure assessment with the Pangea multi-scale framework – case study in Australia. *Environ Sci: Processes Impacts* 20:133–144. doi.org/10.1039/C7EM00523G
- Weinberger K, Lumpkin TA (2007) Diversification into horticulture and poverty reduction: a research agenda. *World Development* 35:1464–1480. doi.org/10.1016/j.worlddev.2007.05.002
- Westh TB, Hauschild MZ, Birkved M, et al (2015) The USEtox story: a survey of model developer visions and user requirements. *Int J Life Cycle Assess* 20:299–310. doi.org/10.1007/s11367-014-0829-8

- Williamson S, Ball A, Pretty J (2008) Trends in pesticide use and drivers for safer pest management in four African countries. *Crop Prot* 27:1327–1334. doi.org/10.1016/j.cropro.2008.04.006
- Xue X, Hawkins TR, Ingwersen WW, Smith RL (2015) Demonstrating an approach for including pesticide use in life-cycle assessment: Estimating human and ecosystem toxicity of pesticide use in Midwest corn farming. *Int J Life Cycle Assess* 20:1117–1126. doi.org/10.1007/s11367-015-0902-y



THESIS RESEARCH QUESTIONS AND OBJECTIVES

The initial research question asked in this thesis was “Why and how to assess pesticides for agricultural LCA in tropical regions?” So, the first chapter presented a bibliographic review highlighting the most important issues for currently assessing pesticides of crops grown in tropical conditions, at both emission (LCI) and impact (LCIA) stages. The most up-to-date and consensual LCI and LCIA models commonly reflect temperate conditions for climate, soil, application methods and crops, and not those reflecting crops cultivated in tropical regions. The LCI parameters driving variability in pesticide emission and impact score results need to be defined and the model’s parametrization have to be tested for crops grown in tropical conditions. To assess pesticide emissions, tropical crops and associated growth stages are not represented, therefore, foliar interception fractions are unknown when applying pesticides. There is no application method and its drift defined for crops grown in tropical conditions. In the secondary emission distribution, specific features of tropical cropping systems and farm management practices should be included, especially ground cover management. In order to fully assess pesticide emissions and related impacts in tropical conditions and more generally for all agricultural contexts, a consistent combination of LCI and LCIA models is required, with the inclusion of plant uptake and residue exposure in crop.

Hence, the **overarching goal** of the thesis is:

Advancing emission and impact modeling for agricultural pesticides under tropical conditions, to improve scientific foundation of the environmental evaluation of tropical agricultural food systems.

The thesis aims to answer the following **research questions and specific objectives**:

✓ **Which LCI parameters are driving variability in pesticide emission and impact score results?**

To answer to the research question, specific objectives are defined with the following questions: How do changes in emission model inputs affect outputs at emission and impact results level? Which of the tested input variables are most relevant to assess pesticide emissions of crops grown specifically under tropical conditions? Is the sensitivity of emission results to model input data representative of processes occurring in tropical conditions? (Chapter 2)

✓ **Is it possible to consistently assess emissions and (eco-)toxicity impacts from pesticide applications under tropical conditions in LCA considering pesticide residues in consumed crop?**

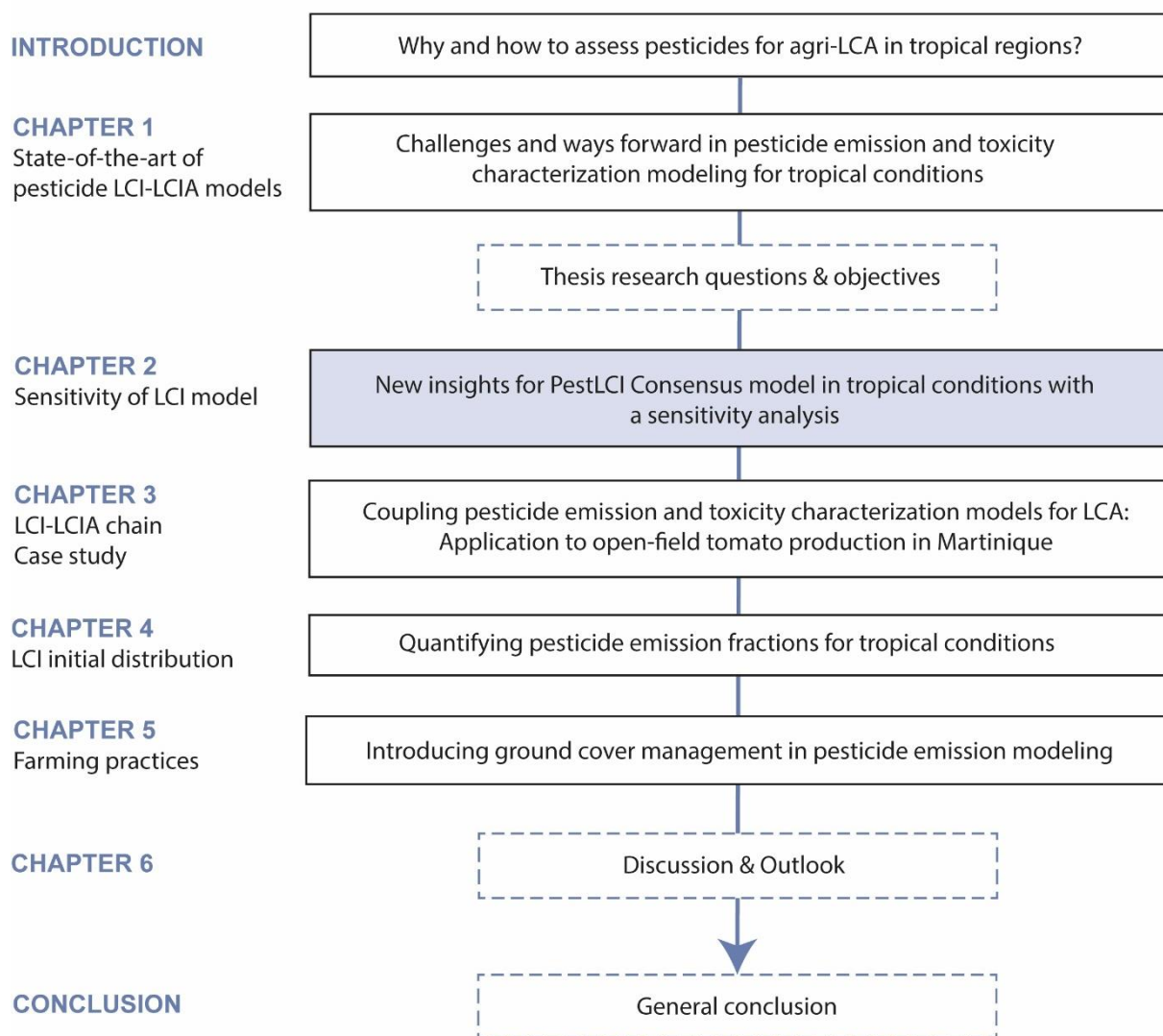
In chapter 3, we will propose a consistent combination of state-of-the-art emission and impact models for characterizing pesticides in LCA, including exposure to pesticide residues in crops. The model coupling will be illustrated with real-life case study on open-field tomato production in Martinique, testing different methodological choices.

✓ **How to quantify initial pesticide emission fractions for crops grown under tropical conditions?**

To provide LCA practitioners with a set of emission fractions for different crops grown in tropical conditions, we will develop a consistent set of crop growth stages and associated foliar interception fractions according to crop growth and spraying technique under tropical conditions. In addition, drift deposition curves defined for tropical crop conditions and application methods will be proposed in the PestLCI Consensus model (Chapter 4).

✓ **How to account for agroecological practices in pesticide emission modeling?**

To assess and compare different production systems, such as conventional agriculture and more agro-ecological agriculture, we will propose a framework for considering ground cover management (GCM) in a state-of-the-art pesticide emission model (i.e. PestLCI Consensus model). Our approach will be tested in a case study using different GCM types under temperate and tropical conditions and the effect of GCM on distributions of emissions and related freshwater ecotoxicity impacts will be analyzed (Chapter 5).



CHAPTER 2

NEW INSIGHTS FOR PESTICIDE EMISSION MODELING UNDER TROPICAL CONDITIONS WITH A SENSITIVITY ANALYSIS

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Abstract

Life Cycle Assessment of agricultural products requires the consideration of pesticide emissions to quantify their toxicity impacts on humans and ecosystems. The PestLCI model estimates pesticide emissions in various environmental compartments. Recently updated and implemented as a web-tool, the Consensus version of PestLCI was initially developed to assess pesticide emissions under temperate conditions. To identify the related key input variables that would have to be adapted, we tested the sensitivity of model output using a scenario analysis, where numerical and categorical input variables were varied. Results were combined with impact characterization factors to understand the influence of emission input data at impact level.

The ‘mean absolute deviation’ method was applied to evaluate categorical and continuous emission model input variables. Following this method, we used an index of the change induced by the variation of inputs as compared to a reference scenario of an insecticide application on tomato under tropical conditions. Overall, 282 scenarios were simulated, changing the value of one input variable at a time. The emission fractions (emission model output) were characterized in terms of toxicity-related impacts,

by multiplying them with corresponding characterization factors according to the relevant environmental compartments, using freshwater ecotoxicity and human non-cancer toxicity as indicators.

As most influential input variables, we identified the foliar interception fraction and application method for initial pesticide distribution fractions within minutes after application, and pesticide, soil and climate type for secondary emission distribution fractions that also consider processes within 24 hours after application. These variables should hence be the focus for adaptation of the emission model for tropical conditions. The main compartments contributing to overall impact scores per pesticide are emissions to off-field surfaces, groundwater and field soil that are allocated to agricultural soil and freshwater in the impact assessment. For freshwater ecotoxicity, impacts based on secondary emission distributions are substantially higher than those based on initial distributions. For human non-cancer toxicity, impact results are similar for both emission distribution levels.

In order to estimate pesticide emissions for agri-food systems under tropical conditions in LCA, we identified aspects that should be in priority developed in future research efforts. These include application method, crop interception fractions and processes occurring in soil influencing emissions to water bodies, in particular for foreground LCA studies. Our results furthermore emphasize that impacts from ingestion via crop residues should be included to cover an additionally relevant pathway for human toxicity impacts, aligned with current pesticide emission outputs. Overall, our sensitivity analysis constitutes a viable starting point for further improving and refining the assessment of pesticides in a life cycle assessment context, with special emphasis on pesticides applied under tropical conditions.

Keywords: Active ingredient, pesticide modeling, Life Cycle Assessment, tropical crops, scenario analysis

2.1 Introduction

Life Cycle Assessment of agricultural products requires the estimation of pesticide emissions to quantify their toxicity-related impacts on humans and ecosystems. Based on the framework proposed by Hauschild (2000), the pesticide life cycle inventory (LCI) model PestLCI estimates pesticide emissions in various compartments: air, soil, crop, groundwater and off-field surfaces. Several versions followed, starting with PestLCI 1.0 (Birkved and Hauschild 2006), then PestLCI 2.0 (Dijkman et al. 2012), which was further adapted for grapevine production (Renaud-Gentié et al. 2015b) considering cover crops in a specific version, and finally the PestLCI Consensus version building on global recommendations derived in the Glasgow Consensus process (Rosenbaum et al. 2015). PestLCI Consensus was recently implemented as a web-tool (Fantke et al. 2017a).

Along the updates done on PestLCI, several sensitivity analyses have been conducted on the different versions of the model. A sensitivity analysis consists of varying the input variables and

observing how this influences the variability of model outputs (Faivre et al. 2013; Pianosi et al. 2016), allowing to test the robustness of a model toward its input parameters. Birkved and Hauschild (2006) did a first sensitivity analysis on PestLCI 1.0 for emission fractions to air, surface water and groundwater, and the largest influence on all emission fractions were due to variables related to solar radiation of the month of application, pesticide dissipation in soil and soil characteristics. Next, Dijkman et al. (2012) and Fantin et al (2018) provided a sensitivity analysis of PestLCI 2.0. They concluded that combining climate and soil variations had an important effect on model output variations. The most influential variables were again the soil characteristics and climate conditions, notably the solar radiation. PestLCI 2.0 was composed of more inputs than PestLCI 1.0. Next, Renaud-Gentié et al. (2015) provided a sensitivity analysis of the version of PestLCI 2.0 for viticulture. The main conclusions revealed that soil and climate characteristics were again the most influential variables, along with canopies development (of vine and cover crop) and the application method characteristics.

Several modifications were introduced in the PestLCI Consensus model, mainly related to the overall mass balance of applied pesticide, on the drift deposition estimation and on processes occurring few days after pesticide application (secondary emissions) such as volatilization, leaf uptake or even degradation on crops leaves (Fantke et al. 2017a). A sensitivity analysis of this new version has not yet been performed, and due to modifications from the previous versions, conclusions of latest sensitivity analyses need to be updated. Furthermore, initially developed for application to temperate regions, the model was extended with worldwide databases on soil and climate conditions. Processes driving pesticide transfers to the environment are specific to soil, climate, crop and farming practices (Racke et al. 1997; Daam and van den Brink 2010; Sanchez-Bayo and Hyne 2011; Mottes et al. 2014). In a bibliographic review, the use of PestLCI Consensus model for crops grown in tropical conditions was recently evaluated (Gentil et al. 2020b), and first recommendations were derived to better account for the specificities of pesticide emissions under tropical conditions. To refine those recommendations and to evaluate how inputs used in PestLCI Consensus affect both emission results as well as impact scores derived from combining those emission results with state-of-the-art toxicity characterization results, a sensitivity analysis of emission model inputs at the level of emission and impact results is required, with special emphasis on aspects relevant for tropical conditions. This will help to identify the most important variables for crops grown under tropical conditions in pesticide emission modeling, and better assess the environmental performance of agri-food systems in tropical regions for LCA.

In response to these needs, we aim in the present study at answering the following three questions: (1) How do changes in emission model inputs affect outputs at emission and impact results level? (2) Which of the tested input variables are most relevant to assess pesticide emissions of crops grown specifically under tropical conditions? (3) Is the sensitivity of emission results to model input data representative of processes occurring in tropical conditions?

2.2 Materials and methods

To test the robustness of the PestLCI Consensus model for estimating emissions from application of pesticides in tropical conditions and to identify which input variables are most relevant under tropical conditions, a sensitivity analysis using different scenarios was carried out, evaluating how varying emission model inputs affect variability of emission and related impact results. For assessing impact results-level sensitivity, we combined emission results from PestLCI Consensus with characterization factors derived for human toxicity and freshwater ecotoxicity with the scientific consensus model USEtox (Rosenbaum et al. 2008) into impact scores per emission compartment. Combining impact scores across emission compartments allows us to assess the relative contribution of the different environmental emission compartments to the total impact score and the effect of the choice of emission distribution used.

2.2.1 PestLCI model characteristics

PestLCI Consensus (<https://pestlciweb.man.dtu.dk>) is a life cycle inventory (LCI) mass balance model, which proposes two types of emission distributions for pesticides applied to agricultural fields, namely the initial (primary) distribution considering the first minutes after the pesticide application and the secondary emissions considering additional processes within 24 hours after application. Few minutes after pesticide application, pesticides are distributed to air, field soil surface, field crop leaf surface and off-field surfaces (Dijkman et al. 2012; Fantke et al. 2017a). After pesticide deposition, additional distribution and removal processes lead to secondary emission fractions to air, field soil, field crop leaf surface, groundwater, off-field surfaces, as well as to field crop leaf uptake, and degradation in field crop and soil (Figure 8)³. To define these emission fractions, the model is composed of three categories of input variables, (i) mandatory user inputs, (ii) optional user inputs (default values given where no user value is provided) and (iii) default model parameters (user adjustable, where advanced process refinement is needed), being either continuous or categorical variables (Table 5). The current model version is applicable to organic, xenobiotic pesticides. Developments to include metal-based substances are being currently evaluated in the OLCA-Pest project (<http://qsa.man.dtu.dk/research/research-projects/olca-pest>), particularly for secondary emission processes. The model is composed of several databases for pesticides, which is a compilation of several pesticide databases (e.g. REACH, EFSA, EPISUITE), application methods, soils from the FAO (soil profiles available on <http://www.fao.org/soils-portal/en/>), and climates from the Köppen-Geiger classification (Kottek et al. 2006; Beck et al. 2018). The PestLCI Consensus model also allows users to feed the model with their

³ Since this study was realized, the field crop leaf uptake was separated from the fraction degraded in field crop leaf and soil.

own climate and soil characteristics, as well as adding new pesticides to the existing databases or modifying properties of already included pesticides.

Table 5: Input variables for PestLCI Consensus initial distribution and secondary emissions indicating whether the variable is mandatory (*) or optional (**). Details on PestLCI variables, units and definitions are presented in the Supplementary Material (SM), Table S2.1.

	Type of variables	
	Continuous variables	Categorical variables
Initial distribution	Foliar interception fraction* Buffer zone presence inside or outside the field* Buffer zone width (default:0)* Buffer zone foliar interception fraction (default: 0.4)** Field width* Field length*	Pesticide – active ingredient* Application method* Drift reduction type (default: 0)**
Secondary emissions	Slope (default: 0)** Drainage depth and fraction drained (default: 0)** Annual irrigation (default: 0)**	Climate* Soil* Tillage type (default: conventional tillage)**
Default parameters (user adjustable)		Soil material density Reference soil moisture content Fraction macropores Response factor for biodegradation rate Q-value Air boundary layer D(lam) A(p,ref)

2.2.2 Sensitivity analysis approach

We tested the sensitivity of emission results (derived using PestLCI Consensus) and impact score results (derived combining emission results with toxicity characterization results using USEtox) to both continuous and categorical PestLCI Consensus input variables. A scenario analysis was performed using the Mean Absolute Deviation (MAD) method, which is an index of the change of model outputs (emission results and combined emission and impact characterization results) between a given pesticide “application scenario” and a pre-defined “reference scenario”. The MAD method is defined as the arithmetic mean of the absolute deviations of a set of application scenarios from a reference scenario (i.e. from a reference scenario of pesticide application with defined values for all input variables), which is used as reference point across application scenarios for each set of tested variables. Mathematically, it is described as:

$$MAD = \frac{\sum_{i=1}^n |x_i - x_{\text{ref}}|}{n} \quad (2.1)$$

for a set of $x_i \in \{x_1, x_1, \dots, x_n\}$ emission scenarios, with reference scenario x_{ref} as chosen ‘measure of central tendency’ of application scenario set X , i.e. $x_{\text{ref}} \triangleq m(X)$.

MAD has the same unit as the application scenario results. To get the percent deviation from the reference scenario, we can calculate the mean relative difference rD_{ref} from the reference scenario as:

$$rD_{\text{ref}} = 100 \times \frac{MAD}{x_{\text{ref}}} \quad (2.2)$$

rD_{ref} has the unit of % of the reference scenario unit.

The reference scenario corresponded to a typical situation of pesticide application on a tomato open-field production in Martinique and set of tested input variables are summarized in Table 6. Overall, 282 scenarios were simulated, changing the value of one input variable at a time. For the continuous input variables (e.g. field slope), ten contrasted values within a realistic range were selected. For the categorical input variables (e.g. soil type), a set of different categorical variables were tested. To assess secondary emissions, a running time of the model was set at 7 days, in order to consider, for all climates and months tested, the effect of one rain event in the modeling of pesticide emissions.

2.2.3 Pesticide impacts calculation

The emissions were converted into impacts with the corresponding USEtox continental-scale emission compartments, namely agricultural soil, natural soil, freshwater and rural air (Rosenbaum et al. 2008; Fantke et al. 2018b). The environmental compartments from PestLCI Consensus and USEtox differ. To align the compartments, the emissions to groundwater from PestLCI Consensus were linked to the freshwater emission compartment of USEtox, and the off-field surfaces compartment from PestLCI Consensus was linked to 3 different compartments in USEtox, namely agricultural soil, natural soil and freshwater, according to the statistical soil occupation in our reference scenario in Martinique (Figure 8). Due to the absence of a plant that is modeled in USEtox as environmental emission compartment, emissions to field crop have not been assigned to any compartment, and related impacts due to direct pesticide ingestion have not be assessed in the present study. The impact score (IS) of each scenario simulation was calculated, multiplying the emission fractions of each USEtox compartment by the corresponding characterization factors (CF), for the freshwater ecotoxicity and the human non-cancer toxicity.

Table 6: Reference pesticide application scenario and set of categorical and continuous variables tested in the sensitivity analysis of PestLCI Consensus model inputs

Input variable	Reference scenario	Categorical and continuous variables tested
Name of pesticide	Abamectin	acetamiprid – azoxystrobin – cyazofamid – deltamethrin – emamectin benzoate – glufosinate ammonium – glyphosate – indoxacarb – lambda-cyhalothrin – mancozeb – metaldehyde – pymetrozine – pyriproxyfen – spinosad – tricosane – endosulfan – kasugamycin hydrochloride hydrate – cyhalofop-butyl – 1,2-dibromoethane – fuberidazole
Crop foliar interception fraction	0.5	10 values between 0 and 1
Climate*	Af	Am – As – Aw – Cfb – Dfc – SVn – SSA – NPr
Month of application	September	All months
Soil type*	Nd	Bv – Je – Th – Tv – Vp – Bk – Q – Fh
Application method	Knapsack sprayer	All available application methods (27)
Drift reduction	No	Yes
Buffer zone	No	Yes, inside the field Yes, outside the field
Buffer zone width (m)	0	10 values between 0 and 10
Buffer zone interception fraction	0	10 values between 0 and 1 (default 0.4)
Field width (m)	100	10 values between 10 and 200
Field length (m)	100	10 values between 10 and 200
Field slope (%)	0	10 values between 0 and 25
Drainage depth (m)	0	10 values between 0 and 1
Drainage fraction effectiveness	0	10 values between 0 and 1
Annual irrigation (mm)	700	10 values between 0 and 1500
Tillage type	Conventional tillage	No tillage; Reduced tillage

*Details on climate and soil types tested are presented in SM, Table S2.2.

The total IS for all scenarios was then calculated as sum over emission compartment-specific impact scores:

$$IS = \sum_{p,c} (m_{\text{emi},p,c} \times CF_{p,c}) \quad (2.3)$$

where $m_{\text{emi},p,c}$ ($\text{kg}_{\text{emitted}}/\text{kg}_{\text{applied}}$) is the total emitted mass of pesticide p from the tomato production (reference crop for all scenarios) into a given environmental compartment c , and $CF_{p,c}$ ($\text{impact}/\text{kg}_{\text{emitted}}$) is the corresponding characterization factor for freshwater ecotoxicity (in PAF $\text{m}^3 \text{d}/\text{kg}_{\text{emitted}}$) and for human population-level toxicity (incidence cases/ $\text{kg}_{\text{emitted}}$). The characterization factors for the following

active ingredients were missing for these two impact categories and consequently, these active ingredients were removed from the analysis (for the freshwater ecotoxicity: tricosane, endothion, kasugamycin hydrochloride hydrate, cyhalofop-butyl and 1,2-dibromoethane; for the human non-cancer toxicity: tricosane, endothion, kasugamycin hydrochloride hydrate, cyhalofop-butyl and 1,2-dibromoethane, fuberidazole). Details of CF are presented in SM, Table S2.3.

The total (eco)-toxicity impact scores and the relative contribution of the different environmental compartments for the freshwater ecotoxicity and human non-cancer toxicity were analyzed summing the median of all scenarios for each emission compartment.

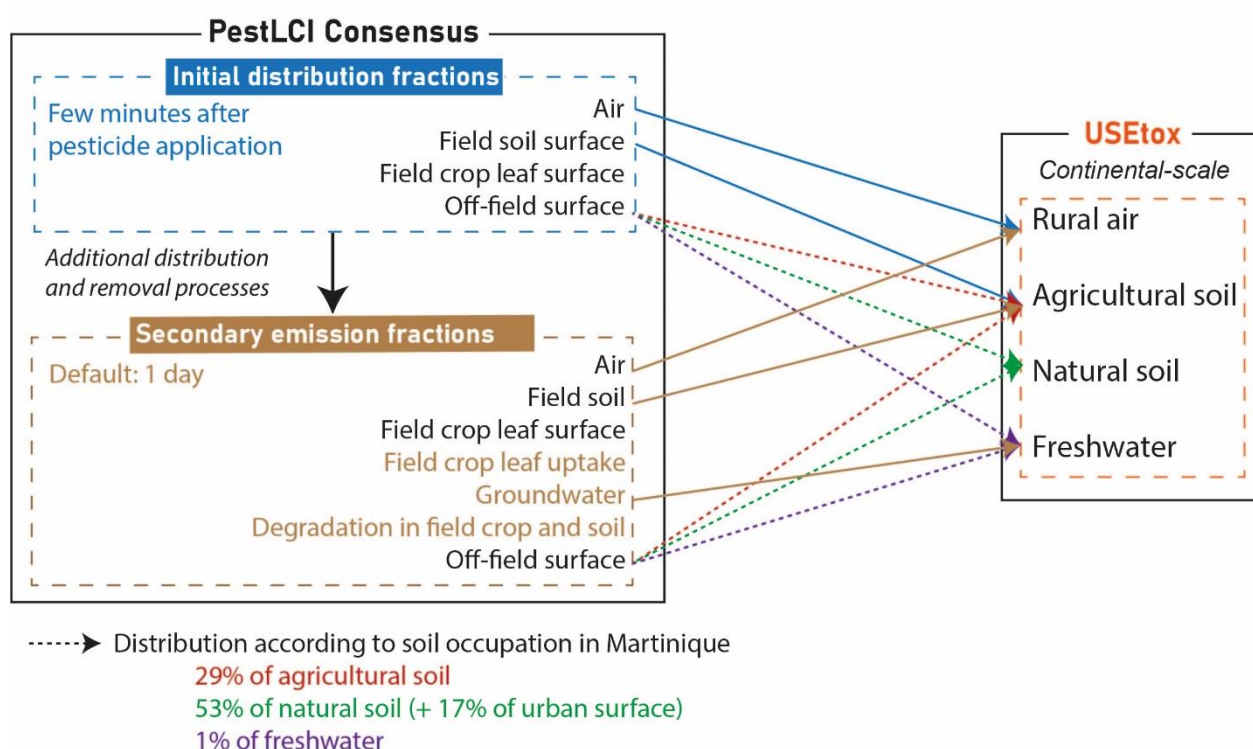


Figure 8: PestLCI emissions fractions and coupling with USEtox environmental compartments

2.3 Results and discussion

2.3.1 Scenario analysis at pesticide emission results level

We first analyzed the sensitivity of the diverse initial and secondary emission fractions to each emission model input variation. The sensitivity of output emission fractions to input variations is detailed in SM, Table S2.4.

The sensitivity analysis results using the MAD approach on initial distribution fractions are presented in Table 7. According to the tomato case study reference scenario, the most influential

variables for the initial distribution were the crop foliar interception fraction, the application method and its drift reduction, and the field width; mostly affecting field crop and soil fractions in PestLCI Consensus. Application method and its drift reduction were the only variables influencing pesticide distribution to air. For off-field surfaces, the most influential inputs were the field width, followed by the presence of a buffer inside the field and then by the application method. Finally, for initial fractions reaching field crop and field soil surfaces, the most influential variable was the foliar interception fraction, followed by the application method.

Table 7: Influence of emission model input variables on initial pesticide distribution fractions (rD_{ref} : the mean relative difference from the reference scenario). Example: the foliar interception fraction input deviates the emission to field crop from the reference scenario by almost 30%.

Input variable \ Emission compartments	Air	Off-field surfaces	Field crop	Field soil
Foliar interception fraction	0	0	27.97%	27.97%
Application method	2.00%	1.25%	1.63%	2.19%
Application drift reduction	3.07%	0.93%	1.75%	0.18%
Field width	0	1.88%	0.94%	0.94%
Buffer zone width, inside the field	0	1.23%	0.62%	0.62%
Buffer zone interception fraction, inside the field	0	1.34%	1.09%	0.35%
Buffer zone width, outside the field	0	0.20%	0.10%	0.10%
Buffer zone interception fraction, outside the field	0	0.20%	0.10%	0.10%

The sensitivity analysis results using the MAD approach on secondary emissions fractions are presented in Table 8. According to the tomato case study reference scenario, the most influential variables were the foliar interception fraction, the pesticide, the climate and soil types, and the application method and its drift reduction; mostly affecting emissions to field soil and the degraded fraction (i.e. in field crop and soil), and the fraction taken up into the crop leaves. Emissions to air were mostly influenced by the pesticide, mostly influencing volatilization occurring after initial distribution. For off-field surfaces, the most influential inputs were similar to those for initial distributions, i.e. field width, presence of a buffer inside the field and, application method and its drift reduction. Most input variables had limited influence on groundwater emissions, which were mainly influenced by the tillage type. For field soil and degraded fraction (i.e. in field crop and soil) and crop leaf uptake, the most influential variable was the foliar interception fraction, followed by the pesticide, the climate and the soil type.

Table 8: Influence of emission model input variables on secondary pesticide emission fractions (rD_{ref})

Emission compartments	Air	Off-field surfaces	Groundwater	Field soil	Degradation and uptake
Input variable					
Foliar interception fraction	0	0.01%	0.11%	22.91%	23.02%
Pesticide	4.20%	0.01%	0.30%	16.90%	16.71%
Climate type	0	0.01%	0.08%	5.81%	5.82%
Soil type	0	0.01%	0.24%	4.53%	4.30%
Application drift reduction	3.07%	0.93%	0.01%	1.62%	2.50%
Application method	2.00%	1.25%	0.01%	1.51%	1.73%
Field width	0	1.88%	<0.01%	0.87%	1.00%
Buffer zone interception fraction, inside the field	0	1.35%	<0.01%	0.97%	0.41%
Buffer zone width, inside the field	0	1.24%	<0.01%	0.58%	0.66%
Tillage type	0	0	0.92%	0.92%	0
Month of application	0	<0.01%	0.05%	0.88%	0.90%
Soil material density	0	<0.01%	0.08%	0.43%	0.36%
Q-value	0	<0.01%	<0.01%	0.27%	0.28%
Buffer zone width, outside the field	0	0.20%	<0.01%	0.09%	0.11%
Buffer zone interception fraction, outside the field	0	0.20%	<0.01%	0.09%	0.11%
Field slope	0	0.16%	<0.01%	0.16%	0
Fraction macropores	0	0	<0.01%	<0.01%	0

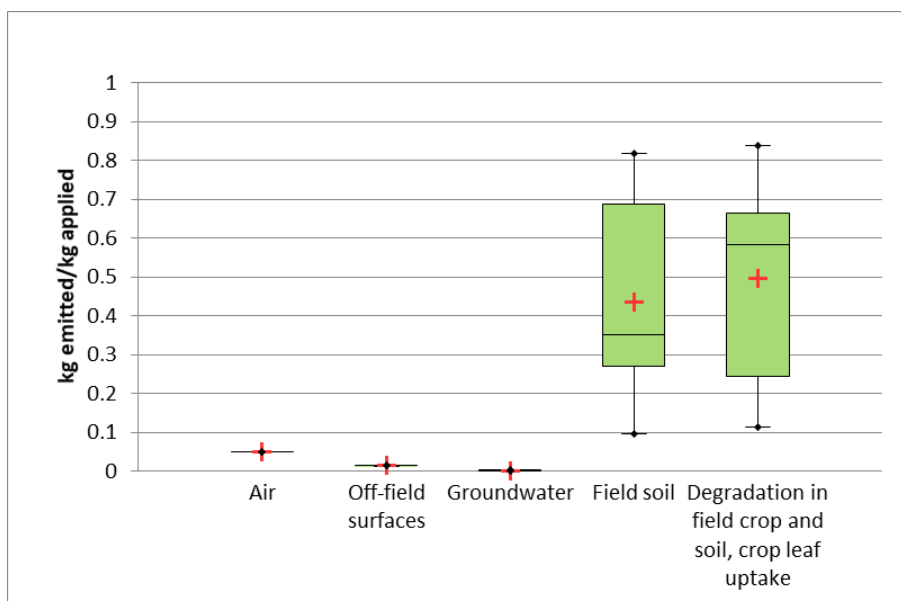
Default variables are only used for secondary emission processes estimation. Among the 8 default variables, 5 did not influence any emission fractions, which was of particular concern.⁴ The three others (soil material density, fraction macropores and Q-value) had low influence, mainly on field soil. The lack of sensitivity of emission fractions to drainage, irrigation and field length variables was likewise of concern and revealed several limitations of the current PestLCI model, mainly regarding water fluxes modeling in soil during the secondary emissions calculation.

Overall, the most influential input variables on emission fraction results were the foliar interception fraction and the pesticide. Figure 9 illustrates the variation of emission fractions (kg emitted/kg applied) for a set of scenarios simulated by varying only one input variable at a time compared to the reference scenario, for (a) the foliar interception fraction and (b) the pesticide. The foliar interception fraction is the fraction of pesticide that the crop leaves can intercept from an applied pesticide amount, which

⁴ After this sensitivity analysis, the issue with those non-influential variables was identified and the model was updated accordingly. However, the updated process descriptions were not considered in our sensitivity analysis.

depends on the crop type and its growth stage, as defined by Linders et al. (2000) using the growth stages with the BBCH-scale (Meier 2018). According to the foliar interception fraction, the fraction emitted to field soil varied from 0.1 to 0.82 and the fraction degraded (i.e. in field crop and soil) and taken up by the crop leaves varied from 0.11 to 0.84. According to pesticide characteristics (e.g. soil DT50), the fraction emitted to field soil varied from 0.01 to 0.78 and the fraction degraded (i.e. in field crop and soil) and taken up by the crop leaves varied from 0.06 to 0.93, which means that defining these input variables properly is absolutely crucial as these results range from almost zero to almost 100%. Moreover, the soil and climate did not have a strong influence on secondary emission fractions, contrary to what we might expect under tropical conditions.

(a) Foliar interception fraction (10 scenarios)



(b) Pesticides (21 scenarios)

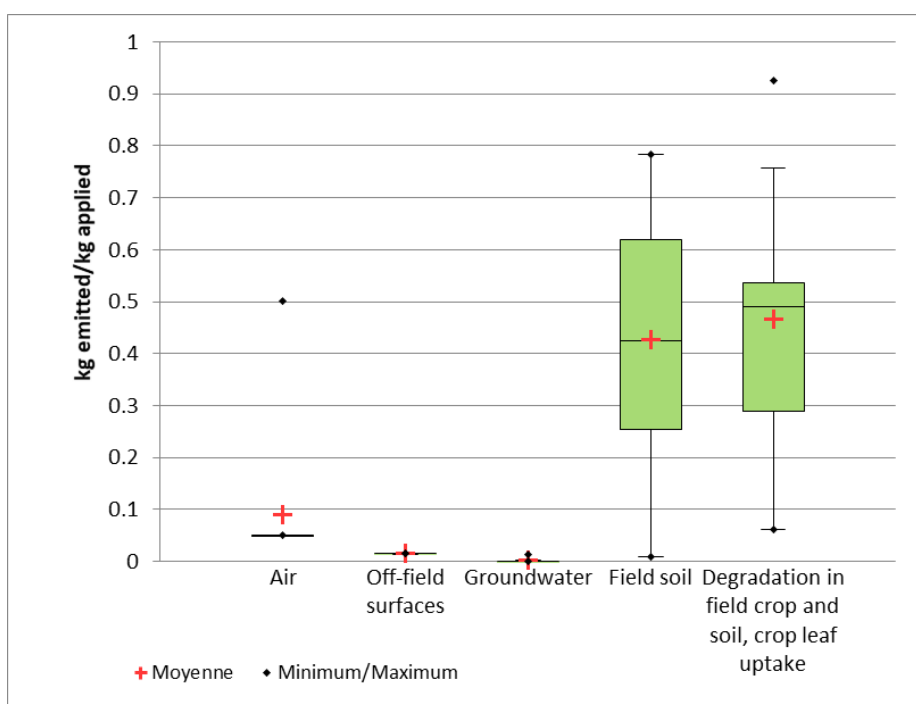


Figure 9: Box plots representing the variation of secondary emission fractions for a set of scenarios simulated by varying only one input at a time compared to the reference scenario, for (a) the foliar interception fraction and (b) the pesticide.

2.3.2 Propagation of the scenario analysis to impact score level

The emission results were combined with impact results into impact scores based on linking emissions into above-defined compartments with the respective characterization factors for the

corresponding USEtox continental compartments, namely agricultural soil, natural soil, freshwater and rural air. The total impact scores from the different environmental compartments for (a) the freshwater ecotoxicity and (b) human non-cancer toxicity are presented in Figure 10.

For freshwater ecotoxicity, using either initial or secondary distributions, impacts came from two main compartments, namely agricultural soil and freshwater. With the initial distribution, the agricultural soil compartment was the main contributor to overall impact scores (72%). Whereas, when using the secondary emissions, the freshwater compartment was the most important contributor with 75% of the total impact, while the agricultural soil was the second most important compartment with 21% contribution to overall impact scores. These impacts to agricultural soil and freshwater compartments came from pesticide distribution on field soil, off-field surfaces compartments for initial distribution, and emissions to groundwater in secondary emissions. For the human non-cancer toxicity, the major contributor to impact scores was the agricultural soil compartment, using either initial or secondary emissions, representing for both around 90% of the total impact scores.

With our tomato case study as reference scenario, the freshwater ecotoxicity impact was three times higher when using the secondary emissions than when using the initial distribution. For human non-cancer toxicity, the most influential compartments in USEtox being the same regardless of the distribution from PestLCI, the difference of impact when using the initial or secondary emissions was almost zero. These results must be relativized compared to the high uncertainty of the pesticide characterization factors for which uncertainties of one to three orders of magnitude apply, depending on emission compartment and impact pathways considered (Rosenbaum et al. 2008).

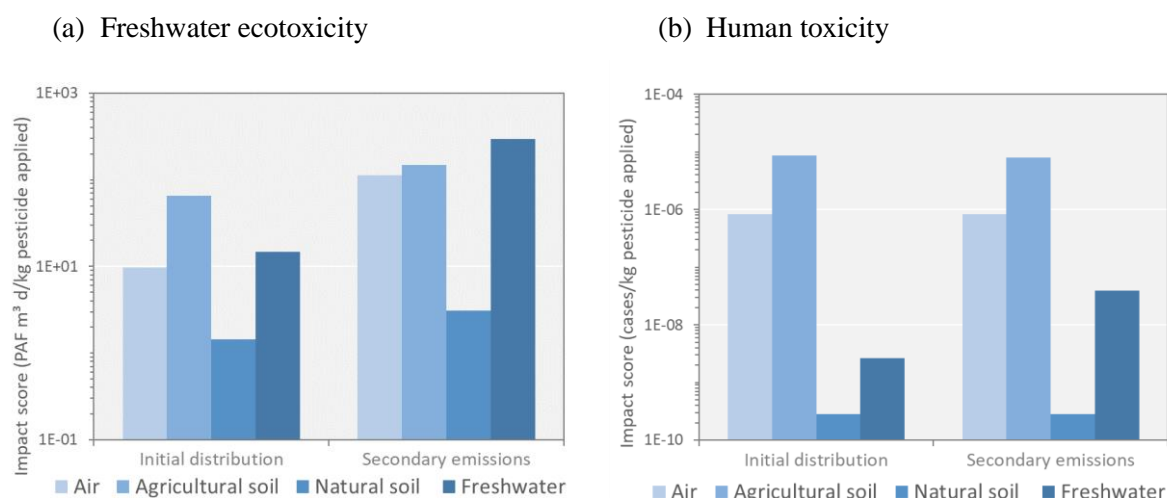


Figure 10: Illustration of the total impact scores from the different environmental compartments for (a) the freshwater ecotoxicity and (b) human non-cancer toxicity (sum of the medians in each environmental compartment of emissions).

The emission fractions to freshwater from off-field surfaces and to groundwater were very small, being respectively 1.5% and 0.2% for the secondary emissions (median of the 282 scenarios). However, the freshwater compartment contributed to 75% of freshwater ecotoxicity. These results are mainly explained by the high freshwater ecotoxicity characterization factors for freshwater compartments, and because the ecotoxicity impact category targets freshwater organisms.

As already mentioned, pesticide emissions on field crop were not assigned to any environmental impact compartment. Consequently, impacts from pesticide residues in edible field crops and on remaining pesticide fractions in unharvested cover crops from pesticide on crop were not considered in the present study. This is a limitation of our sensitivity analysis, since across the 282 scenarios with the tomato case study as reference scenario, on average 46.4% of the pesticides were distributed on crop leaves. With that, neglecting emissions and subsequent impacts from residues in food crops may lead to underestimations of overall impacts from any product system in LCA.

2.3.3 Proposition to better consider tropical conditions for pesticide assessment

According to the identified influential variables in pesticide emission modeling, **the foliar interception fraction** is the most sensitive variable for both initial and secondary emission processes. This fraction depending on crop growth stages is defined according to Linders et al. (2000) for a wide diversity of crops including some that can grow both in temperate and tropical conditions (e.g. tomato, bean, tobacco). However, various crops growing in tropical conditions are missing, such as banana or pineapple. A set of crop growth stages and associated foliar interception fractions for crops grown under tropical conditions should therefore be developed to accurately consider the most relevant model input variable in pesticide emission modeling.

For initial distribution fractions, the second most influential variable was the **application method and its drift reduction**. Currently in PestLCI Consensus model, among the application methods and its associated drift deposition curve, none have been developed under tropical conditions. Moreover, the aerial application method, common in tropical regions is not available, regardless of the region of application. A bibliographic review on drift deposition under tropical conditions should be done to explore whether the corresponding curves could be integrated into the model.

Furthermore, some **farming practices** might strongly influence pesticide emissions to freshwater (Mottes et al. 2014) such as tillage and ground cover management (Reichenberger et al. 2007; Alletto et al. 2010). Currently, among the farming practices (e.g. tillage) considered in PestLCI Consensus model, none had a substantial influence on pesticide emissions, which was not expected, and might be mostly attributable to remaining refinement needs in the model related to secondary environmental processes. Moreover, some rain-related processes are defined with temperate climate and soil data. A refinement of these processes influencing pesticides transfer to water bodies would be relevant, in particular under tropical conditions where intense rainfall are causing more runoff and leaching than in temperate

conditions (Sanchez-Bayo and Hyne 2011). To this end, processes related to soil and climate data would have to be analyzed in a dedicated effort to assess the validity of those processes for tropical conditions. Overall, aspects predominantly relevant for crops grown in tropical conditions are currently not well reflected in the emission model under investigation, and should be carefully refined in order to render the model applicable also to tropical cropping practices.

Current pesticide impact assessment should be complemented with a terrestrial ecotoxicity impact category to better assess impacts from emissions to field soil, building on actual soil organism effect data and considering various soil characteristics that might influence field emissions as well as environmental fate (such as degradation). Moreover, impacts from pesticide residues in edible field crops and on remaining pesticide fractions in unharvested cover crops from pesticide on crop should be assessed to avoid underestimations and bias when assessing overall environmental performance of agri-food systems. The impacts from ingestion via crop residues could be considered by aligning the pesticide emission model PestLCI, and the plant uptake and crop residue exposure model, dynamiCROP (Fantke et al. 2011b), recently adjusted for LCA (Fantke and Jolliet 2016).

2.3.4 Limitations of the present study

We used the MAD approach to test the sensitivity of emission outputs and related impact scores to PestLCI Consensus inputs variations. This approach allowed accounting for the sensitivity to both continuous and categorical input variables. However, continuous data are also aggregated into categorical variables, such as pesticide (e.g. solubility, soil DT50), climate (e.g. average temperature, rain frequency) and soil input (e.g. soil composition of sand, silt and clay) variables. Sensitivity to these continuous variables should also be evaluated, notably to better assess the validity of the model for tropical conditions, especially climate conditions. Contrary to previous PestLCI model versions, the development of the model as a web-tool does not allow yet to do a sensitivity analysis on all variables and detailed parameters. Moreover, to complete the sensitivity analysis, the possible covariance between variables should be assessed.

For the reference scenario, one active ingredient and one crop type have been selected, i.e. the insecticide abamectin on a tomato crop. However, abamectin is an organism-derived pesticide with unclear half-lives and properties, consequently for the reference scenario a substance whose properties are better known should be chosen. Furthermore, knowing the high sensitivity of impact results to the choice of pesticide, the scenario analysis should be repeated with reference scenarios using contrasted pesticides in terms of characterization factors per compartment and impact category and contrasted crops, to confirm the most influential variables regardless of the intrinsic characteristics of the pesticide, at least at the level of emission results. This is also to allow for better considering agricultural management practices (e.g. emission reduction technologies), which have a greater influence on emission results than

chemical properties, given that pesticides are applied in aqueous or oily formulations, with what the properties of the actual active ingredients have limited effect on at least initial emission distributions.

When analyzing the results at impact level, certain characterization factors were missing and it was not possible to assess the sensitivity at impact level to all active ingredients selected. Despite these limitations, our analysis already provides important insights into the most influential parameters influencing emission and impact results for the most recent version of PestLCI.

2.4 Conclusions

To improve the assessment of agri-food systems with LCA in tropical regions, our study aimed to test the sensitivity of emission results and impact scores to PestLCI Consensus input variables in relation to tropical conditions.

The sensitivity analysis using different scenarios highlighted the most influential variables for each emission distribution level used. By order of importance, for the initial distribution, it was the fraction intercepted by crop leaves and the application method and its drift reduction; for the secondary emissions, it was the foliar interception fraction, the pesticide, climate and soil type and then the application method and its drift reduction. Multiplying the emission fractions to each compartment by the respective characterization factor for freshwater ecotoxicity and human non-cancer toxicity, revealed the relative contribution of the different environmental compartments to the total impact score. The main influential compartments were agricultural soil and freshwater compartments in USEtox, corresponding to emission results for the off-field surfaces, groundwater and field soil compartments in PestLCI. For the freshwater ecotoxicity, the impacts obtained with the secondary emissions were higher than those obtained with the initial distribution. For the human non-cancer toxicity, results were similar with both emission distribution levels.

In order to estimate pesticide emissions for agri-food systems in LCA, aspects that should be developed in priority for tropical conditions are application method, crop interception fractions and soil processes influencing emissions to water bodies, in particular for foreground LCA studies, including farming practices such as ground cover management.

Overall, our sensitivity analysis constitutes a viable starting point for further improving and refining the assessment of pesticides in a life cycle assessment context, with special emphasis on pesticides applied under tropical conditions.

Acknowledgments

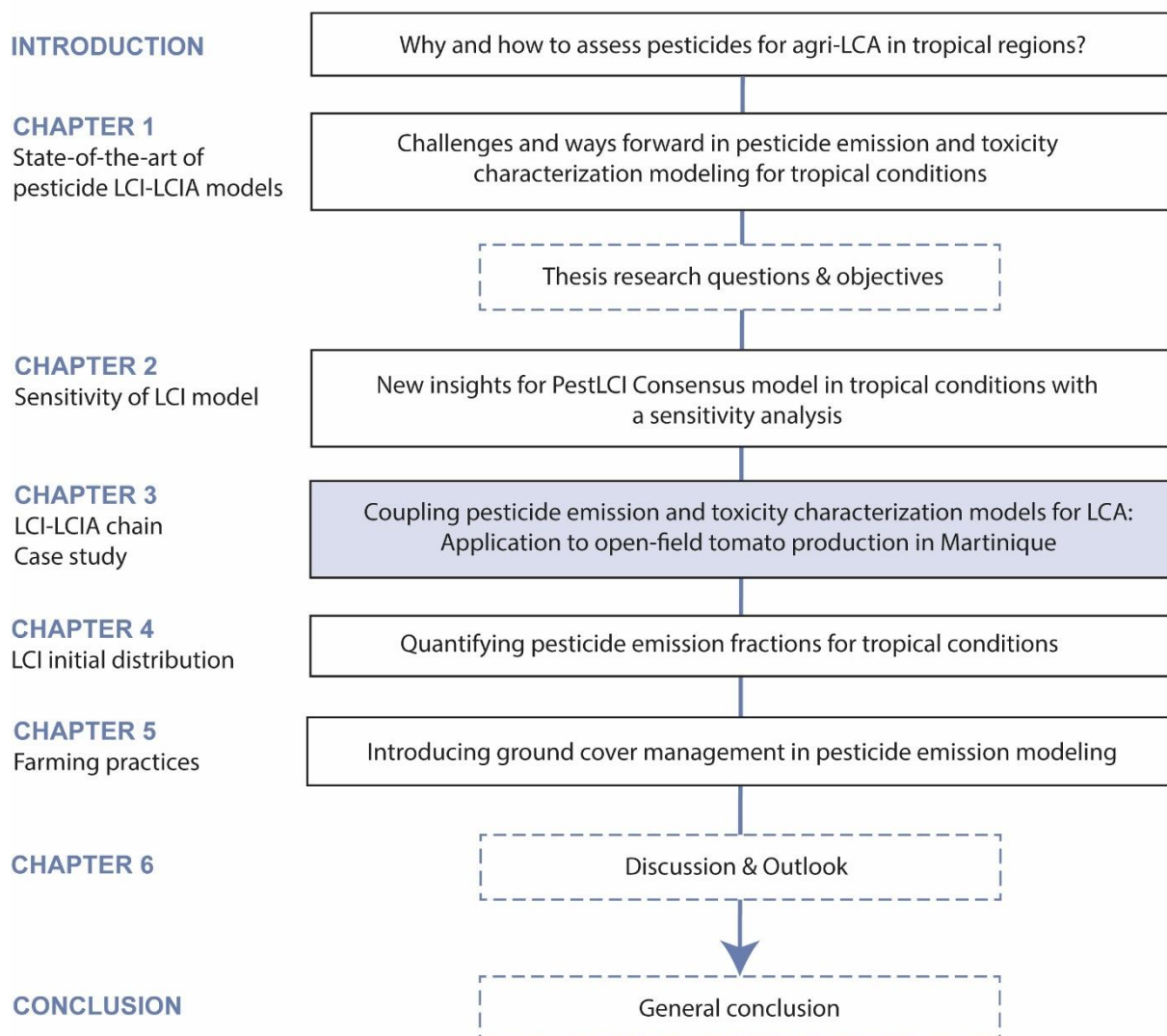
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References Chapter 2

- Alletto L, Coquet Y, Benoit P, et al (2010) Tillage management effects on pesticide fate in soils. A review. *Agron Sustain Dev* 30:367–400. doi.org/10.1051/agro/2009018
- Beck HE, Zimmermann NE, McVicar TR, et al (2018) Present and future Köppen-Geiger climate classification maps at 1-km resolution. *Scientific Data* 5:180214. doi.org/10.1038/sdata.2018.214
- Birkved M, Hauschild MZ (2006) PestLCI—A model for estimating field emissions of pesticides in agricultural LCA. *Ecol Model* 198:433–451. doi.org/10.1016/j.ecolmodel.2006.05.035
- Daam MA, van den Brink PJ (2010) Implications of differences between temperate and tropical freshwater ecosystems for the ecological risk assessment of pesticides. *Ecotoxicology* 19:24–37. doi.org/10.1007/s10646-009-0402-6
- Dijkman TJ, Birkved M, Hauschild MZ (2012) PestLCI 2.0: a second generation model for estimating emissions of pesticides from arable land in LCA. *Int J Life Cycle Assess* 17:973–986. doi.org/10.1007/s11367-012-0439-2
- Faivre R, Iooss B, Mahévas S, et al (2013) Analyse de sensibilité et exploration de modèles. Editions Quae
- Fantin et al (2018) PetLCI 2.0 sensitivity to soil variations for the evaluation of pesticide distribution in Life Cycle Assessment studies
- Fantke P, Antón A, Grant T, Hayashi K (2017) Pesticide emission quantification for life cycle assessment: A global consensus building process. *J Life Cycle Assess* 13:245–251
- Fantke P, Aylward Lesa, Bare Jane, et al (2018) Advancements in Life Cycle Human Exposure and Toxicity Characterization. *Environmental Health Perspectives* 126:125001. doi.org/10.1289/EHP3871
- Fantke P, Jolliet O (2016) Life cycle human health impacts of 875 pesticides. *Int J Life Cycle Assess* 21:722–733. doi.org/10.1007/s11367-015-0910-y
- Fantke P, Juraske R, Antón A, et al (2011) Dynamic multicrop model to characterize impacts of pesticides in food. *Environ Sci Technol* 45:8842–8849. doi.org/10.1021/es201989d
- Gentil C, Fantke P, Mottes C, Basset-Mens C (2020) Challenges and ways forward in pesticide emission and toxicity characterization modeling for tropical conditions. *Int J Life Cycle Assess* 25:1290–1306. doi.org/10.1007/s11367-019-01685-9
- Hauschild MZ (2000) Estimating pesticide emissions for LCA of agricultural products. In: Weidema BP (ed) *Agricultural data for life cycle assessments*. Agricultural Economics Research Institute (LEI), The Hague, pp 64–79
- Kottek M, Grieser J, Beck C, et al (2006) World Map of the Köppen-Geiger climate classification updated. *Meteorol Z* 15:259–263. doi.org/10.1127/0941-2948/2006/0130

- Linders J, Mensink H, Stephenson G, et al (2000) Foliar interception and retention values after pesticide application: a proposal for standardised values for environmental risk assessment. *Pest Manag Sci* 58:315–315. doi.org/10.1002/ps.448
- Meier U (2018) Growth stages of mono- and dicotyledonous plants: BBCH Monograph. Open Agrar Repository
- Mottes C, Lesueur-Jannoyer M, Bail ML, Malézieux E (2014) Pesticide transfer models in crop and watershed systems: a review. *Agron Sustain Dev* 34:229–250. doi.org/10.1007/s13593-013-0176-3
- Pianosi F, Beven K, Freer J, et al (2016) Sensitivity analysis of environmental models: A systematic review with practical workflow. *Environmental Modelling & Software* 79:214–232. doi.org/10.1016/j.envsoft.2016.02.008
- Racke KD, Skidmore MW, Hamilton DJ, et al (1997) Pesticides report 38. Pesticide fate in tropical soils - (Technical report). *Pure Appl Chem* 69:1349–1371. doi.org/10.1351/pac199769061349
- Reichenberger S, Bach M, Skitschak A, Frede H-G (2007) Mitigation strategies to reduce pesticide inputs into ground- and surface water and their effectiveness; a review. *Sci Total Environ* 384:1–35. doi.org/10.1016/j.scitotenv.2007.04.046
- Renaud-Gentié C, Dijkman TJ, Bjørn A, Birkved M (2015) Pesticide emission modelling and freshwater ecotoxicity assessment for Grapevine LCA: adaptation of PestLCI 2.0 to viticulture. *Int J Life Cycle Assess* 20:1528–1543. doi.org/10.1007/s11367-015-0949-9
- Rosenbaum RK, Antón A, Bengoa X, et al (2015) The Glasgow consensus on the delineation between pesticide emission inventory and impact assessment for LCA. *Int J Life Cycle Assess* 20:765–776. doi.org/10.1007/s11367-015-0871-1
- Rosenbaum RK, Bachmann TM, Gold LS, et al (2008) USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment. *Int J Life Cycle Assess* 13:532. doi.org/10.1007/s11367-008-0038-4
- Sanchez-Bayo F, Hyne RV (2011) Comparison of environmental risks of pesticides between tropical and nontropical regions. *Integr Environ Assess Manag* 7:577–586. doi.org/10.1002/ieam.189



CHAPTER 3

COUPLING PESTICIDE EMISSION AND TOXICITY CHARACTERIZATION MODELS FOR LCA: APPLICATION TO OPEN-FIELD TOMATO PRODUCTION IN MARTINIQUE

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Abstract

The environmental evaluation of fruits and vegetables in life cycle assessment (LCA) requires a proper estimation of pesticide emissions and associated (eco-)toxicity impacts. In response, we developed an approach to consistently combine state-of-the-art emission inventory and impact assessment models for assessing human toxicity and freshwater ecotoxicity impacts from pesticide applications, and tested our approach in an LCA case study on pesticides applied to an open-field tomato produced in Martinique (French West Indies). Our results show that impact scores vary over several orders of magnitude, mainly as function of differences in pesticide properties and application time in

relation to crop growth stage. Overall, impacts related to pesticide field emissions leading to exposure to pesticide residues in crop harvest are a main contributor to LCA performance results for tomato produced in Martinique, with fertilizer and packaging manufacturing as other dominating aspects. While the proposed approach is applicable to refine currently LCA methods for assessing pesticides, large uncertainties remain. These are mostly related to the parametrization of impact factors for tropical species. Based on our findings, we recommend using initial emission distribution fractions in combination with steady-state characterization factors for environmental emissions and with time-dependent characterization factors for pesticide residues in crop harvest in LCA, while further improving the use of secondary emission fractions to allow for better consideration of local field, soil and climate characteristics.

Keywords: Active ingredient, modeling, human health, Life Cycle Assessment, tropical conditions

3.1 Introduction

In tropical regions, fruit and vegetable production for local markets has become a key challenge. This is especially relevant for remote islands, such as Martinique (French West Indies), which receives its main supply from other Caribbean islands, main France or other countries, such as Spain and Morocco. Moreover, current production systems facing high pest pressure in the tropics mainly rely on the use of pesticides all year round (Racke et al. 1997; Daam and van den Brink 2010; Lewis et al. 2016; Mottes et al. 2017b), potentially harming environmental and human health (Arias-Estévez et al. 2008; Aktar et al. 2009; Lesueur Jannoyer et al. 2016). In Martinique and other remote islands, there is hence a demand for fruits and vegetables that are grown locally and in an environmentally sustainable way. This includes the production of tomato (*Solanum lycopersicum*), a highly demanded vegetable in Martinique with an average annual local production of 1800 tonnes, but difficult to produce locally due to various diseases, weeds and pests that affect plant health and production. In support of improving local crop production, there is a need to evaluate the environmental performance of locally grown produce including the characterization of pesticide emissions and associated (eco-)toxicity impacts (Perrin et al. 2014; Meier et al. 2015; Knudsen et al. 2019; van der Werf et al. 2020).

Life cycle assessment (LCA) is an ISO-standardized methodology to evaluate the environmental performance of product systems. However, in many current studies, pesticide emissions and related-impacts are not assessed as demonstrated in the review on LCA of vegetables of Perrin et al. (2014); while in cradle-to-farm gate LCA studies, one of the main contributors to human and ecosystem toxicity is generally pesticide (Bessou et al. 2013). Completing the reviews from Perrin et al. (2014) and Bessou et al. (2013), an analysis of most recent LCA studies (presented in the Supplementary Material (SM), Section S3.11) confirmed that, when pesticides are considered, emissions are instead derived from

generic emission fractions, assuming that all pesticides are either going 100% to agricultural soil (e.g. Nemecek and Schnetzer 2011) or follow a generic distribution between air and soil (e.g. Oliquino-Abasolo 2015). Indeed, generally, only the amount of pesticides applied on the field is known (Fantke et al. 2012a; Fantke and Jolliet 2016). This approach is simpler but according to our previous work is very likely to lack accuracy, especially in tropical conditions (Gentil et al. 2020). Such generic emission fractions ignore differences across application methods, crops and other important characteristics influencing pesticide emission distributions, such as in tropical conditions with higher temperature enhancing degradation and volatilization of pesticides (Racke et al. 1997; Daam and van den Brink 2010), with intense rainfall causing more runoff and leaching, leading to emissions to surface waters and groundwater compartments (Sanchez-Bayo and Hyne 2011) or crop canopy and associated application methods. Furthermore, some exposure pathways are frequently omitted, such as exposition by ingestion of pesticide residues in harvested crop components (Fantke et al. 2011b). LCA studies usually do not couple state-of-the-art pesticide emission with (eco-)toxicity impact and pesticide crop residue models to ensure a consistent modeling from application to impacts (Martínez-Blanco et al. 2011; Perrin et al. 2014; van Zelm et al. 2014).

A brief overview of the currently available state-of-the-art pesticide emission and toxicity-related characterization models applicable for LCA, namely PestLCI, USEtox, and dynamiCROP, is given in the following. The pesticide life cycle inventory (LCI) model PestLCI (Dijkman et al. 2012) estimates emission fractions to air, field soil, field crop, groundwater and off-field surfaces, and has been further advanced into PestLCI Consensus (Fantke et al. 2017a). PestLCI uses two levels of emission distributions. Initial (or primary) mass distributions cover initial processes within minutes after pesticide application, whereas secondary emission distributions also consider additional transport and degradation processes within a given period (e.g. 1 day) after application. The scientific consensus model USEtox is widely applied in LCA for (eco-)toxicity characterization of chemical emissions in life cycle impacts assessment (LCIA) (Rosenbaum et al. 2008; Westh et al. 2015; Fantke et al. 2018a, b). For characterizing human toxicity impacts from pesticides, Fantke and Jolliet (2016) identified ingestion of pesticide residues in harvested crop components as dominating exposure pathway for the general population, which is of particular concern for freshly consumed fruits and vegetables (Weinberger and Lumpkin 2007; Fantke et al. 2012a). Specific pesticide residues in crops scenarios can be assessed using a dedicated LCIA plant uptake and crop residue exposure model, such as dynamiCROP (Fantke et al. 2011a, b, van Zelm et al. 2014).

Those models have been primarily developed for temperate conditions, the suitability of these models to evaluate pesticide-related impacts for crops grown under tropical conditions has recently been questioned (Gentil et al. 2020b), with only two studies applying these models separately in a tropical context, namely PestLCI for pineapple production in Costa Rica (Ingwersen 2012) and dynamiCROP for passion fruit production in Colombia (Juraske et al. 2012). Furthermore, all these models were

designed separately with different system boundaries and considered compartments (Gentil et al. 2020), leading to potential overlaps and gaps between LCI and LCIA (van Zelm et al. 2014; Rosenbaum et al. 2015). Combining these models in a consistent way, hence, constitutes a challenge for practitioners on the most consistent way to assess pesticides.

Assessing pesticide-related emissions and impacts, therefore, requires a consistent coupling of these models and a parameterization to tropical conditions. The purpose of our study is to propose a consistent combination of emission and impact models with a case study on an open-field tomato production LCA in the tropical conditions of Martinique.

To address these challenges, we aim at answering the question “How can emissions and (eco-)toxicity impacts from pesticide applications under tropical conditions be consistently evaluated in LCA?” We defined three specific objectives: (i) To propose a consistent combination of state-of-the-art emission and impact models for characterizing pesticides in LCA, including exposure to pesticide residues in crops. (ii) To apply the coupled models in a real-life case study on open-field tomato production in Martinique, testing different methodological choices. (iii) To derive recommendations for LCA and define future research needs for an improved evaluation of pesticide emissions and impacts applied under tropical conditions.

3.2 Materials and methods

3.2.1 Overview of the followed approach

To assess pesticide emissions and impacts according to the relevant ISO norms (ISO 14040 2006; ISO 14044 2006), we developed a cradle-to-farm gate LCA on open-field tomato production in Martinique. We used SimaPro (version 9.0.0.35) as LCA software and USEtox 2.11 as (eco-)toxicity characterization method. In the following, we refer by the term “pesticide” to the active ingredient in a given plant protection product formulation. We transferred the life cycle inventory pesticide data from PestLCI outputs to SimaPro with the ELDAM software (Coste et al. 2018), to ensure data set quality and review.

We separately evaluated freshwater ecotoxicity and human toxicity impacts for organic and metal-based substances for best-possible transparency according to current recommendations, since the characterization of metals and organics follows different approaches for fate, exposure and effect modeling (Frischknecht and Jolliet 2019). While non-cancer effect information was available for all considered pesticides, cancer data were only available for deltamethrin showing no carcinogenic effects. According to USEtox guidelines, we calculated missing characterization factors for emission- and crop

residue-based human toxicity and freshwater ecotoxicity, with model input data for these pesticides given in the SM (Sections S3.1 and S3.2).

The considered life cycle stages include production, transport to the farm and use on the farm of all inputs (fertilizers, pesticides, field materials, pesticide spray equipment, irrigation system and packaging manufacturing). Due to the lack of consistent and valid model for tropical conditions, we didn't account for field emissions of heavy metals from fertilizers. Background processes for the manufacturing and transport of farm inputs, such as fertilizers, pesticides, field material, and packaging boxes, were obtained from ecoinvent 3.5 LCI database (cut-off version). We defined the functional unit (FU) as 1 kg of fresh tomatoes produced in Martinique (French West Indies). For illustrative purposes, we used a mass-based FU, whereas a more nutrition-oriented FU might be more appropriate in actual food-related LCAs (Weidema and Stylianou 2019).

3.2.2 Coupling emission and impact models for pesticides

As starting point, the impact score for (eco-)toxicity impacts of pesticide emissions related to our tomato production, IS (impact/functional unit), is calculated as:

$$IS = \sum_{p,c} (m_{\text{emi},p,c} \times CF_{p,c}) \quad (3.1)$$

where $m_{\text{emi},p,c}$ ($\text{kg}_{\text{emitted}}/\text{FU}$) is the total emitted mass of pesticide p from the tomato production into a given environmental compartment c , and $CF_{p,c}$ ($\text{impact}/\text{kg}_{\text{emitted}}$) is the corresponding characterization factor for a given impact category (i.e. human toxicity or ecotoxicity).

Emission mass is usually not known to LCA practitioners (Rosenbaum et al. 2015), but can be obtained from the pesticide mass applied to the tomato fields, $m_{\text{app},p}$ ($\text{kg}_{\text{applied}}/\text{FU}$), which we collected from farmers using semi-directive interviews (i.e. interviews using open-ended and targeted questions) and the related mass fraction that is emitted into different environmental compartments, $m_{f,p,c}$ ($\text{kg}_{\text{emitted}}/\text{kg}_{\text{applied}}$):

$$m_{\text{emi},p,c} = m_{\text{app},p} \times m_{f,p,c} \quad (3.2)$$

When pesticides contain metal ions, they cannot be characterized as organic substances, since characterizing metals requires to consider speciation and other metal-relevant characteristics (Dong et al. 2014). Emission fractions for pesticides, which need to be characterized as metal ions, hence require a correction factor that accounts for the mass contribution of the metal ion to the overall mass of the emitted pesticide molecule:

$$m_{\text{emi},p,c} = m_{\text{app},p} \times m_{f,p,c} \times \frac{MW_{i \in p} \times n_i}{MW_p} \quad (3.3)$$

where $MW_{i \in p}$ (g/mol) is the molecular weight of the metal ion i found in pesticide p , n_i is the number of metal ions apparent in the pesticide molecule, and MW_p (g/mol) is the molecular weight of the pesticide (an example is presented in SM, Section S3.3).

In most LCAs, applied mass is derived from reported doses applied to a certain crop area, summed over different treatments, and is assumed to reach only field soil, i.e. $m_{f,p,c} = 1$ for $c =$ field soil, and $m_{f,p,c} = 0$ for all other compartments across pesticides (Nemecek and Schnetzer 2011). This approach, however, is too simplistic and can be misleading, since relevant emission fractions might reach other compartments and field crop surfaces. Instead, we apply a mass-balance model that accounts for pesticide distribution processes after field application, considering crop and field characteristics (e.g. crop growth stage and field width) along with agricultural practices (e.g. application method). Such a model is PestLCI 2.0 (Dijkman et al. 2012), which was further adapted and implemented as a web-based tool (Fantke et al. 2017a); details are presented in SM, Section S3.4. Using this adapted PestLCI Consensus model Pesticide, we estimated initial distribution (first minutes after application) and secondary emission (until first rain event) fractions. Emission distribution fractions sum up to $\sum_c m_{f,p,c} = 1$ for any given pesticide. Emission input data are detailed in SM (Section S3.5).

Characterization factors, $CF_{p,c}$ (impact/kg_{emitted}), use the pesticide mass emitted into a given environmental compartment as starting point to evaluate related impacts (either on humans or on ecosystems) based on characterizing for each pesticide its environmental fate, exposure and (eco-)toxicity effects. We apply the scientific consensus model USEtox (Rosenbaum et al. 2008), version 2.11 (<https://usetox.org>), to obtain (eco-)toxicity-related characterization factors as:

$$CF_{p,c} = FF_{p,c} \times XF_{p,c} \times EF_p = iF_{p,c} \times EF_p \quad (3.4)$$

where $FF_{p,c}$ (kg_{in compartment} per kg_{emitted}/d) is the fate factor denoting the increase in pesticide mass in compartment c for an emission into any compartment, $XF_{p,c}$ (kg_{intake}/d per kg_{in compartment} or kg_{dissolved}/kg_{in compartment}) is the exposure factor relating population intake (for human exposure) or dissolved pesticide mass (for ecosystem exposure) to total mass in the given compartment, and EF_p (impact/kg_{intake} or impact/kg_{dissolved}) is the effect factor finally relating exposure to impacts. For human toxicity, fate and exposure factors can be summarized into human population intake fractions, $iF_{p,c}$ (kg_{intake}/d per kg_{emitted}/d).

For human toxicity, impacts are expressed as population-level disease incidence risk, which is denoted as incidence or disease ‘case’ when cumulatively exceeding 1, and for ecotoxicity, impacts are expressed as potentially affected fraction (PAF) of exposed species, integrated over compartment

volume and the pesticide's residence time in the environment. We applied the following mapping of PestLCI Consensus to USEtox compartments for consistently combining initial distribution and secondary emission fractions to respective characterization factors (see Figure 11). Air (PestLCI Consensus) is assigned to continental rural air (USEtox), field soil surface and field soil are assigned to continental agricultural soil, and groundwater is assigned to continental freshwater. Off-field surfaces are assigned to continental agricultural soil, natural soil (including urban areas) and freshwater according to the area share of each compartment in a given region (i.e. respectively 29%, 70% and 1% in Martinique). Other initial distribution and emission compartments (crop components and degradation) are not linked to USEtox.

Eq. 3.4 is valid when characterization factors relate to emitted pesticide mass. Impacts related to pesticide mass ending up in the harvested components of the treated field crops (tomato in our case study) consumed by humans are a major contributor to human disease burden, but are usually missing in LCA studies (Fantke et al. 2012b), and related emissions to field crop surface (output of PestLCI Consensus) are hence not characterized. To consider such impacts, we applied the dynamiCROP model (Fantke et al. 2011a, b), recently integrated for some parameterized scenarios into USEtox (Fantke and Jolliet 2016). We used dynamiCROP, version 3.12 (<https://dynamicrop.org>), to obtain residue-related characterization factors for crop x (i.e. impacts from intake of pesticide residues in consumed crop components) as:

$$CF_{p,c} = hF_{p,c}(t) \times PF_f \times EF_p = iF_{p,c} \times EF_p \quad (3.5)$$

where $hF_{p,c}(t)$ ($\text{kg}_{\text{in crop harvest}}/\text{kg}_{\text{emitted}}$) is the harvest fraction relating pesticide residues at harvest time t (d) in crop components that are harvested for human consumption to pesticide mass emitted into a given environmental compartment, PF_f ($\text{kg}_{\text{in processed food}}/\text{kg}_{\text{in crop harvest}}$) is a residue reduction factor due to food processing step f (e.g. washing, cooking), and EF_p ($\text{impact}/\text{kg}_{\text{intake of processed food}}$) is the human toxicity effect factor as defined in eq. 3.4. Harvest fraction and food processing factor can be combined into residue-related intake fractions, $iF_{p,c}$ ($\text{kg}_{\text{intake}}/\text{kg}_{\text{emitted}}$), consistent with intake fractions from USEtox (see eq. 3.4). Assuming that tomatoes are mainly consumed freshly, we applied a washing-related $PF_f = 0.56$ across pesticides (Kaushik et al. 2009). The harvest fraction as originally defined in dynamiCROP refers to total pesticide residues in crops (via all emission compartments) and relates to mass applied (see e.g. Fantke et al. 2013). Related characterization factors for pesticide residues in crops, however, would not be consistent with using initial distribution or secondary emissions into different environmental compartments as defined in eq. 3.1. We therefore adapted $hF_{p,c}(t)$ to relate to initial distributions and emissions, thereby consistently coupling dynamiCROP with PestLCI Consensus results:

$$hF_{p,c}(t) = \frac{\sum_h m_{res,p,h}(t)}{m_{app,p} \times mf_{p,c}} = \frac{\sum_h m_{res,p,h}(t)}{m_{emi,p,c}} \quad (3.6)$$

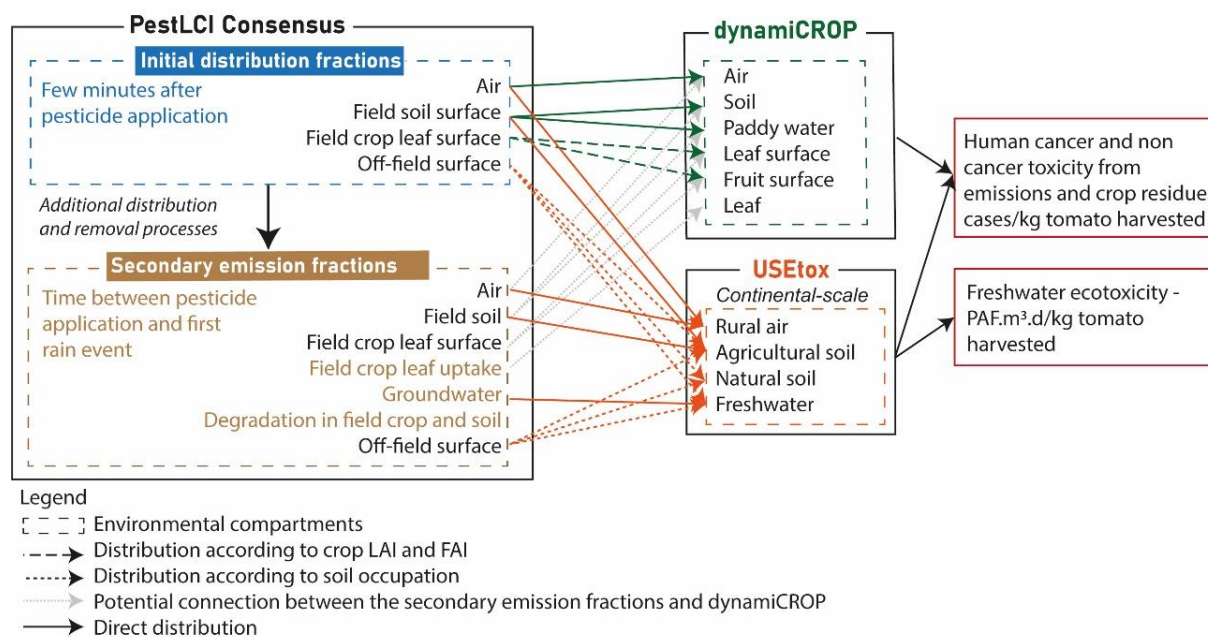
where $m_{res,p,h}(t)$ ($\text{kg}_{\text{in crop harvest}}/\text{FU}$) is the pesticide residual mass in crop components h harvested at time t (d) for human consumption. Since dynamiCROP uses matrix algebra to simultaneously solve a system of differential equations for $m_{res,p,h}(t)$, we realized the adaptation by transforming the input mass vector (i.e. $m_{emi,p,c}$ at time $t = 0$) into a diagonal matrix. Combining these initial conditions diagonal matrix with the fundamental matrix (i.e. mass fractions transferred between compartments at time t) yields emission compartment-specific $m_{res,p,h}(t)$. For details about the underlying matrix solution, see Fantke et al. (2013). Mass applied, $m_{app,p}$ ($\text{kg}_{\text{applied}}/\text{FU}$), distribution fractions, $mf_{p,c}$ ($\text{kg}_{\text{emitted}}/\text{kg}_{\text{applied}}$), emitted to a given environmental compartment matched between PestLCI Consensus (emission output) and dynamiCROP (input for residue calculations), and emitted mass, $m_{emi,p,c}$ ($\text{kg}_{\text{emitted}}/\text{FU}$) are defined in eq. 3.2. The air compartment (PestLCI Consensus) is assigned to air (dynamicalCROP), field soil surface is assigned to soil, and field crop surface is assigned to leaf surface and fruit surface (see Figure 11) according to their total crop surface area contributions:

$$\begin{aligned} mf_{p,c,\text{leaf}} &= \frac{LAI}{LAI + FAI} \text{ for leaf surfaces} \\ mf_{p,c,\text{fruit}} &= \frac{FAI}{LAI + FAI} \text{ for fruit surfaces} \end{aligned} \quad (3.7)$$

where $mf_{p,c,\text{leaf}}$ ($\text{kg}_{\text{emitted to leaf surface}}/\text{kg}_{\text{emitted to field crop surface}}$) and $mf_{p,c,\text{fruit}}$ ($\text{kg}_{\text{emitted to fruit surface}}/\text{kg}_{\text{emitted to field crop surface}}$) are the initial mass fractions emitted to compartment $c = \{\text{field crop surface}\}$ reaching respectively crop leaf and fruit surface areas, and LAI ($\text{m}^2_{\text{leaf surface}}/\text{m}^2_{\text{soil}}$) and FAI ($\text{m}^2_{\text{fruit surface}}/\text{m}^2_{\text{soil}}$) are respectively the crop-specific leaf and fruit area indices. The dynamicalCROP model is currently applicable for assessing organic substances. Further details describing the dynamicalCROP model version adapted for LCA are available in the SM (Section S3.6).

Combining residue-related characterization factors (dynamicalCROP) with characterization factors for environmentally mediated exposures of the general population (USEtox) finally ensures that all relevant initial distribution and emission fractions (PestLCI Consensus) are accounted for, building on a consistent combination of the three underlying models (see Figure 11). Connecting compartments between PestLCI Consensus and dynamicalCROP for secondary emission fractions requires further research as there are currently potential overlaps in modeled plant uptake processes (light gray arrows in Figure 11). Occupational exposure of farmers to pesticides are currently not considered within the existing models.

Figure 11: Connection of the emission compartments of PestLCI to those of dynamiCROP and USEtox according to the pesticide mass applied per kg of the harvested crop; adapted from Fantke (2019).



3.2.3 Assessing sensitivity of different choices

PestLCI and dynamiCROP were customized with site-specific climate and soil data, using local data from Météo France and FAO soil (for details, see SM, Section S3.6).

The sensitivity of different methodological choices has been tested. The sensitivity of (eco-)toxicity impacts has been tested against three emission estimation methods, namely applying the common hypothesis of 100% of pesticide being emitted to agricultural soil, using initial distribution fractions from PestLCI Consensus (only considering processes within first minutes after application), and using secondary emission fractions from PestLCI Consensus (considering processes over a longer timeframe after application). For secondary emissions, we defined the modeled time between pesticide application and emission output as 5 days based on the highest rainfall frequency for all months for the three considered climates in our case study. For copper sulfate, we only derived initial distributions, since secondary emission processes are currently not adapted for metal speciation. Further, we compared the sensitivity of residue-related impacts using initial distribution fractions parametrized to tropical conditions with generic distribution fractions that were available in dynamiCROP. With a test of Wilcoxon's signed ranks, initial and secondary emission fractions were compared for both human toxicity and freshwater ecotoxicity.

3.2.4 Pesticide life cycle inventory data

From a sample of six conventional farms in Martinique we collected primary data. To assess the variability of the secondary emission fractions, farms were distributed as follows: three farms in the North Caribbean (Municipality of Le Prêcheur) and three in the South Atlantic (Municipalities of Vauclin and Sainte-Anne), for the years 2017 and 2018. These two areas of production feature contrasted soil and climate conditions. In this sample, we considered, one field plot per farm, which is six field plots, with one production cycle each. The field plot farms are referred to as scenarios, indicated by letters A to F in our study. Table 9 summarizes the characteristics of the different scenarios, with details on the study system and applied pesticides in SM (Sections S3.7 and S3.8). Required variables for determining initial distribution fractions are the application method (i.e. a knapsack sprayer for all scenarios), the crop and its growth stage, the presence of a buffer zone, field dimensions (variables represented with a gray background in Table 9), and the active ingredient and its applied dose for the impact assessment, including crop characteristics for residue calculations.

Table 9: Characteristics of the considered scenarios of tomato production in Martinique. Required variables for determining initial pesticide distribution fractions are highlighted in gray background.

Scenario ID (farm)	A	B	C	D	E	F	
Climate*	Npr		Svn		Ssa		
Soil	Vitric andosol (TV)			Vertic cambisol (BV)			
Practices	Conventional tillage			yes			
	Irrigation***			no			
	Buffer zone width (m)***			2			
				no buffer zone			
Field plot characteristics	Area (ha)	0.15	0.9	0.05	0.67	0.04	1
	Length (m)	60	100	25	90	17	125
	Width (m)	25	90	20	75	25	80
	Slope (%)	5	25	9	7	20	20
Pesticide application count**	Herbicide	1	2	2	0	1	0
	Insecticide	11	19	10	7	8	8
	Fungicide	9	8	3	2	0	8
Crop yield	Yield (kg/m ²)	0.67	1	4	5.2	3.5	2

*Npr: North Prêcheur, 2300 mm rain/year, 25.4 °C average annual temperature; Svn: South Vauclin, 1200 mm rain/year, 27.2 °C average annual temperature; Ssa: South Sainte-Anne, 1600 mm rain/year, 27.2 °C average annual temperature

**Considered pesticides are detailed in SM (Section S3.8)

***Practices not assessed in the present study

3.3 Results

3.3.1 Emission and impact factors from coupled LCI and LCIA models

Consistently coupling pesticide initial distribution fractions from PestLCI Consensus with characterization factors for environmental emissions from USEtox and for exposure to pesticide residues in crops from dynamiCROP yields a set of aligned LCI and LCIA results for all six considered tomato production scenarios. The variability of initial emission distribution fractions across pesticides and scenarios is summarized in Figure 12. After initial distribution (i.e. some minutes after application), pesticides were mainly deposited on field soil (up to 89% for glufosinate-ammonium at pre-emergence) and field crop (up to 60% across several pesticides) surfaces, varying according to application time in relation to crop growth stage. According to the considered application method (knapsack sprayer without drift reduction system), we fixed an airborne fraction at 5% across scenarios. Fractions reaching off-field surfaces vary only slightly as function of field characteristics and are generally low (<10%).

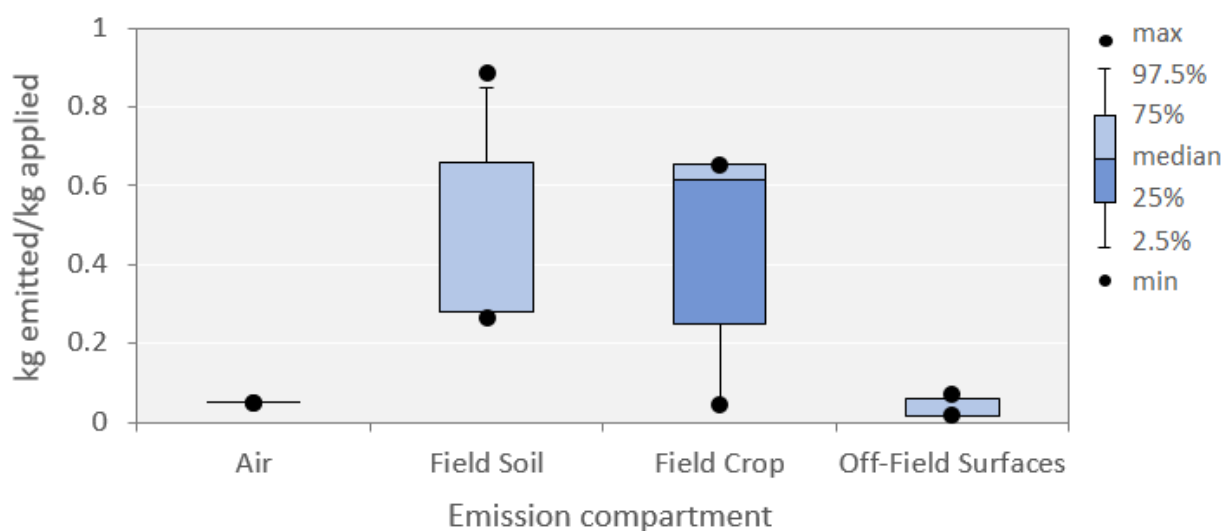


Figure 12: Box-and-Whisker plot indicating the variability of initial pesticide distribution fractions to the different environmental compartments across 80 considered pesticide-field combinations.

In Figure 13, initial distribution fractions are combined with pesticide-specific mass applied per kg tomato harvested, and plotted against impact characterization results (incidence risk per kg emitted) across our six scenarios. Combining these results yields impact scores in terms of impact per kg produced tomato, plotted along diagonal equi-impact lines in Figure 13, where data points on the same diagonal line indicate equal impact, either driven by emissions (x-axis), characterization results (y-axis) or a combination of both. Human toxicity impacts are mainly related to pesticide residues in crops and high emission fractions to agricultural soil, with highest impact scores for metaldehyde, glufosinate-ammonium, acetamiprid and spinosad. This demonstrates the importance of considering exposure to

pesticide residues in crops in LCA. However, impacts span a wide range across pesticides, showing the importance of physicochemical substance properties. Emissions to agricultural soil and for some pesticides to freshwater (via off-field surfaces deposition) and air drive freshwater ecotoxicity. Copper sulfate dominates ecotoxicity impacts, but might be overestimated and depend on the chosen water chemistry that influences metal speciation. Highest impact scores across organic substances are found for lambda-cyhalothrin, azoxystrobin and mancozeb. With exceptions, there is a trend that pesticides applied and emitted in high quantities (e.g. herbicides) are less toxic than pesticides with low application dose (e.g. some insecticides).

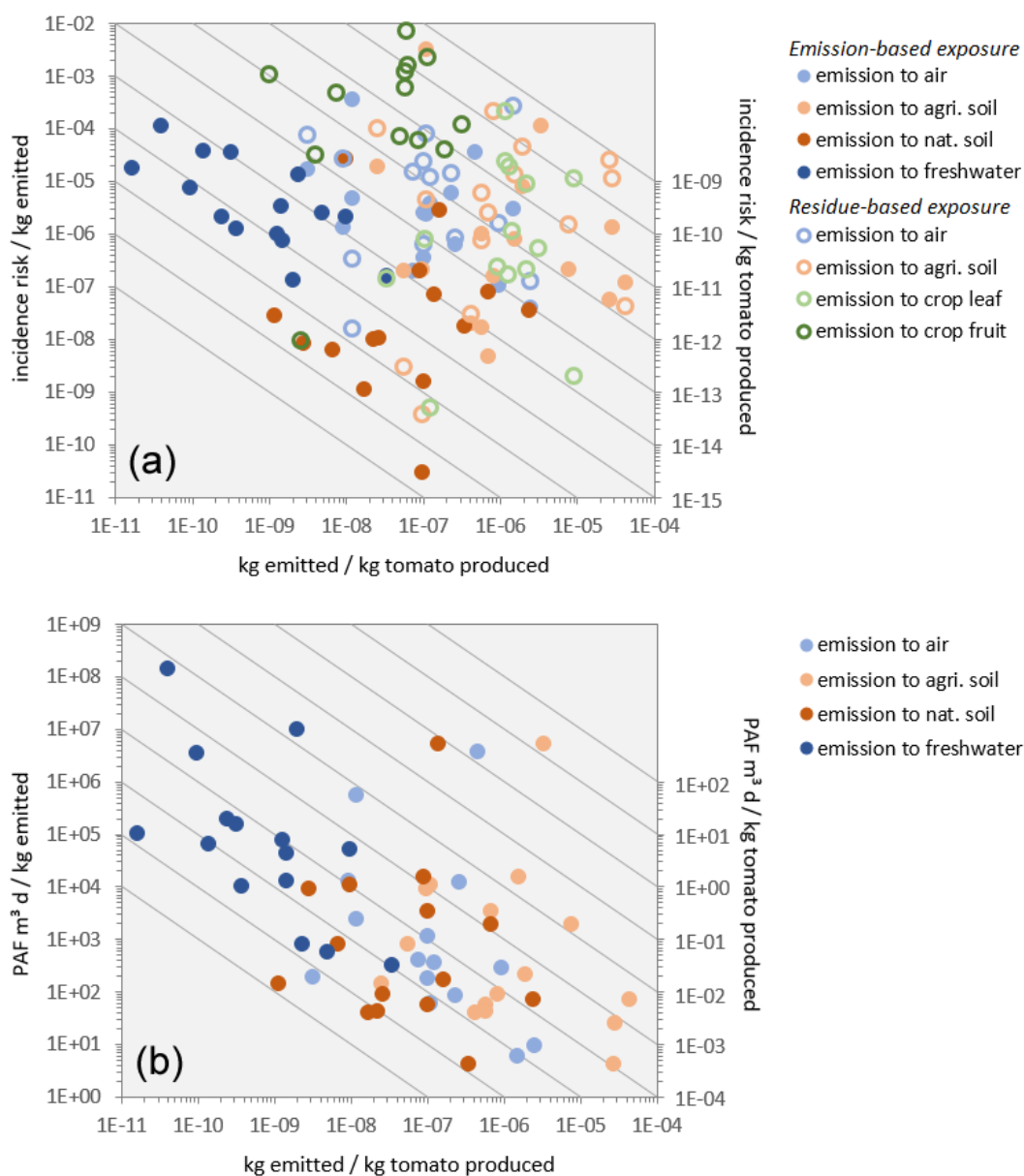


Figure 13: Characterization factors (left-side y-axes) for (a) human PAF toxicity and (b) freshwater ecotoxicity plotted as function of pesticide initial distribution fractions for six tomato fields in Martinique (x-axis). Diagonal equi-impact lines (right-side y-axes) show the respective impact scores.

3.3.2 Freshwater ecotoxicity impacts across tomato production scenarios

Based on initial distribution as underlying emission estimation approach and combining impact scores for pesticides applied to tomato fields in Martinique with life cycle impacts of other scenario inputs yields cradle-to-farm gate results per FU (i.e. 1 kg fresh tomatoes harvested). Figure 14 summarizes the contribution of cradle-to-farm gate stages to total freshwater ecotoxicity and human toxicity impacts across our six scenarios, separately plotted for organic substances and metal-based substances in line with current recommendations. Aggregated average freshwater ecotoxicity impacts were 0.13 PAF m³ d/kg tomato for organic substances and 92 PAF m³ d/kg tomato for metal-based substances across scenarios.

With >50%, pesticide field emissions constitute the main contributor to freshwater ecotoxicity from organic substances, varying by a factor 1800 between the lowest (F) and highest (B) scenario. Dominating pesticides are the fungicides azoxystrobin and mancozeb, and the insecticide lambda-cyhalothrin, with average application doses between 3 and 150 g/ha. The small doses of lambda-cyhalothrin were compensated by higher characterization factors (for individual characterization factors see Figure 13 and SM, Section S3.2). Fertilizer and packaging manufacturing represent the second most important contributors with an average contribution to total impacts of ~13% each, and reaching up to 36% for fertilizer manufacturing in scenario E and 45% for packaging manufacturing in scenario D.

Fertilizer and packaging manufacturing furthermore constitute the greatest contributors to freshwater ecotoxicity from metal-based substances with respectively 37% and 34%, dominated by aluminum-, iron- and copper-related emissions. Pesticide field emissions only represent ~4%, reaching up to 12% for scenario B due to application of copper sulfate. Overall, copper sulfate dominated freshwater ecotoxicity from pesticide field emissions, with in average impacts 2 orders of magnitude higher than impacts from organic substances.

We divided impacts from pesticide field applications into three stages: field emissions, manufacturing and spray equipment. For the latter two, variability across scenarios follows the same trend for organic and metal-based substances, with manufacturing only contributing on average < 2% to freshwater ecotoxicity across substances. Impact variations was mainly driven by active ingredient and applied quantity. Pesticide spray equipment (knapsack sprayer in all scenarios) contributes on average < 1% to freshwater ecotoxicity for organic substances and 4% for metal-based substances. Variation of knapsack sprayer impacts is mainly due to variation in the number of pesticide applications per scenario (5 to 23 pesticide applications per crop cycle) and area sprayed.

Freshwater ecotoxicity from processes belonging to field operations (tillage, field material and irrigation system) is on average 5.7 PAF m³ d/kg tomato for metal-based substances and three orders of magnitude lower for organic substances, representing respectively 6% and 3% of total impacts.

Freshwater ecotoxicity from processes belonging to other aspects (i.e. transport of inputs, fertilizer manufacture and packaging manufacturing) represents respectively 14% and 85% of the total impact for organic and metal-based substances.

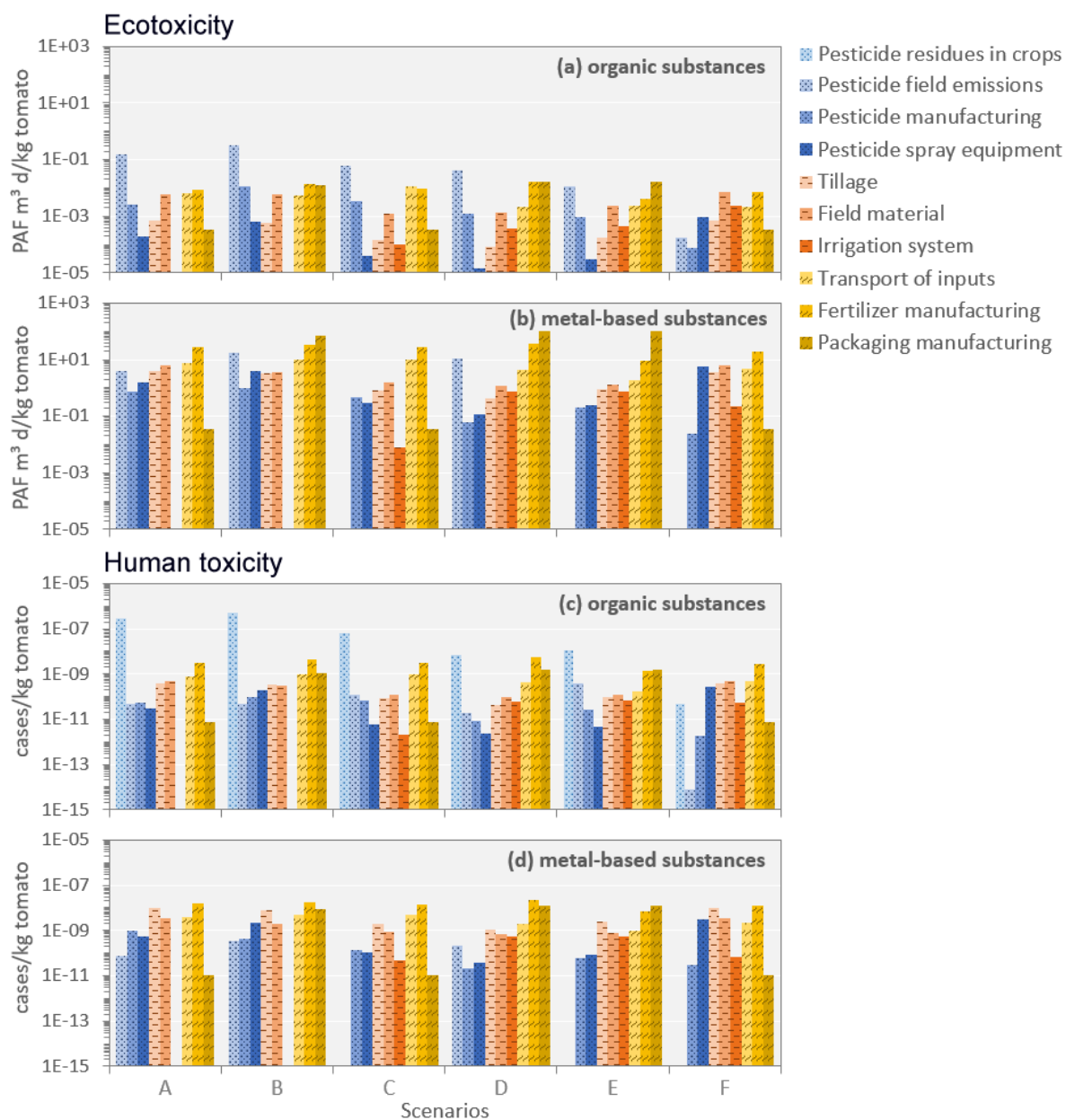


Figure 14: Contribution of cradle-to-farm gate stages to freshwater ecotoxicity and human toxicity impacts per kg tomato harvested for six scenarios, based on initial emission distributions for organic substances (a, c) and for metal-based substances (b, d). Dotted processes belong to pesticides, horizontally dashed processes belong to field operations, and diagonally dashed processes belong to other aspects. Scenarios A-F indicate different combinations of climate, soil and agricultural practice (see Table 9).

3.3.3 Human toxicity impacts across tomato production scenarios

Figure 14 further presents the contribution of cradle-to-farm gate stages to total human toxicity impacts across scenarios. Average human toxicity impacts range from a cumulative population incidence risk of 2×10^{-7} cases/kg tomato for organic substances to 3×10^{-8} cases/kg tomato for metal-based substances.

Ingestion of pesticide residues in crops is the main contributor (68.7%) for organic substances, ranging from < 1% (scenario F) to 99% (scenario B). The second most important contributor is fertilizer manufacturing, representing ~19% of human toxicity from organic substances. Packaging manufacturing and input transportation represent less than 5% of impacts each for organic substances.

Fertilizer manufacturing drives with 45% contribution human toxicity from metal-based substances, with substances containing zinc, mercury, lead, arsenic and cadmium as main contributors. We divided impacts from pesticide field applications into four stages: residues ingestion, field emissions, manufacturing and spray equipment, of which the latter three on average contributed with less than 3% each across substances.

Human toxicity from processes belonging to field operations (tillage, field material and irrigation system) were a factor 10 lower for organic than for metal-based substances, and represent respectively less than 1% and 23% of total impacts. Human toxicity from processes belonging to other aspects (i.e. transport of inputs, fertilizer manufacture and packaging manufacturing) represent respectively 3% and 73% of total impact for organic and metal-based substances.

With respect to impacts related to ingestion of pesticide residues in crops, we find a wide variation across scenarios (see Figure 14c, dominating contributor in left-most column per scenario), with degradation in crops, time between pesticide application and crop harvest, overall residence time in soil and substance molecular weight as main influencing factors, as detailed in Fantke et al. (2012b). In our case study, application dose, toxicity potency, and tomato yield were additional aspects driving impact variability. Low residue-related impact in scenario F are mainly explained by low application doses, up to 10 times less than recommended doses. Residue-related impacts are driven by different pesticides in each scenario, namely mancozeb (scenarios A and F), glufosinate-ammonium (B), pymetrozine (C), acetamiprid (D), and spinosad (E). Except for using metaldehyde in scenario C, the European maximum residue limits (MRLs) were respected according to our residue estimates. This indicates that our considered Martinique's tomato producers generally respected the recommended pre-harvest interval and homologated doses. It is important to note here that although MRLs are respected, there is still a potential impact on humans, even though related risks are considered "acceptable" as per regulatory definitions. Residues are assumed to be generically further reduced by 44% by applying a washing-related reduction factor across pesticides.

3.3.4 Sensitivity of impact results to methodological choices

We tested the sensitivity of (eco-)toxicity impacts comparing the emission inventory methodology, comparing initial distribution fractions, secondary emission fractions, and the common assumption of 100% of pesticide being emitted to agricultural soil (Nemecek and Schnetzer 2011). Figure 15 summarizes the sensitivity of our impact results, contrasting as underlying emission inventory approach initial distribution fractions (i.e. initial minutes after application) against secondary emission fractions (i.e. longer timeframe after application, with climate, soil and agricultural practices as additional influencing aspects) and for field pesticides, and against the common assumption of 100% of applied pesticide being emitted to agricultural soil. Using the secondary emission fractions, freshwater ecotoxicity was significantly higher in all scenarios (test of Wilcoxon's signed ranks, p -value < 0.05), except for scenario A. Human toxicity results were not significantly different using initial or secondary emission fractions (p -value > 0.05). However, assuming uncertainties of pesticide emission results of at least a factor 5 to 10 (more accurate estimates are currently missing) and using reported uncertainties in characterization results of two to three orders of magnitude (Rosenbaum et al. 2008), differences in impacts using primary versus secondary emissions are mostly not significant. When assuming that 100% of applied pesticides are emitted to agricultural soil, impact results decrease up to a factor 20 for the metal-based substance copper sulfate and up to a factor 10 for organic substances (e.g. freshwater ecotoxicity for the insecticide acetamiprid). Using a model providing emission distribution fractions into different compartments is, hence, relevant for improving the estimation of pesticide impacts in LCA (see detailed impact results per scenario and pesticide in SM, Section S3.9).

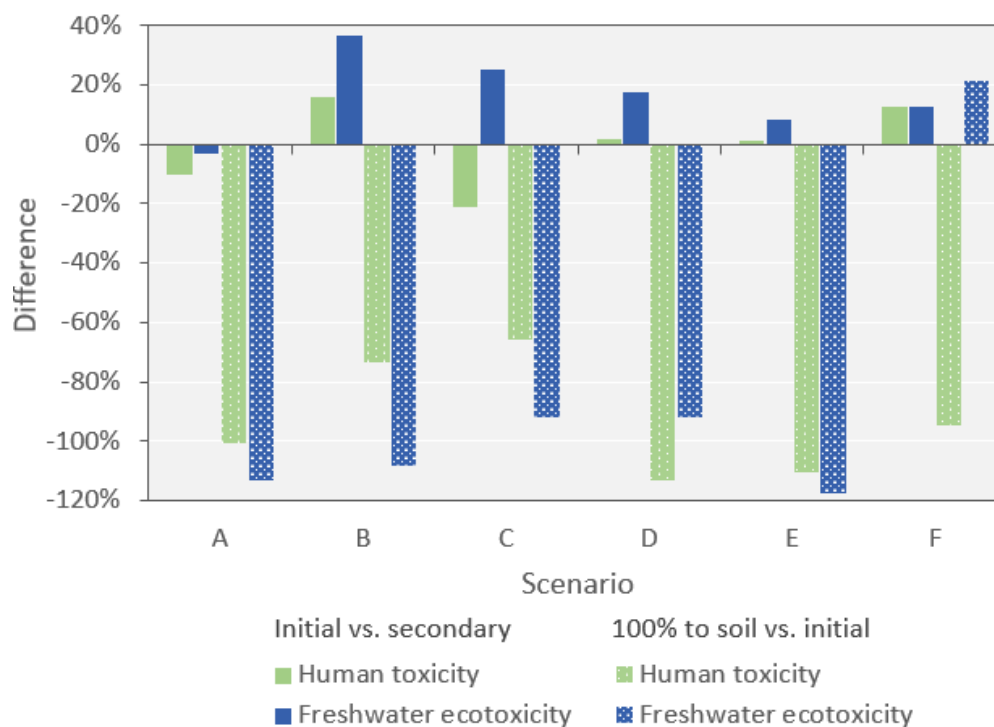


Figure 15: Difference for freshwater ecotoxicity and human toxicity impacts for organic substances contrasting initial distribution fractions, secondary emission fractions, and the common assumption of 100% of applied pesticide being emitted to agricultural soil as underlying approach. Scenarios A-F indicate different combinations of climate, soil and agricultural practice (see Table 9). For the comparison of initial and secondary, the difference was calculated as: $\text{variation}(\%) = (\text{sec} - \text{initial}) / \text{sec}$, where ‘sec’ is for (eco-)toxicity using secondary emission fractions and ‘initial’ for (eco-)toxicity using initial distribution fractions; for the comparison of 100% soil and initial, the difference was calculated as: $\text{variation}(\%) = (\text{initial} - 100\% \text{soil}) / \text{initial}$ where 100%soil is for (eco-)toxicity using an emission fractions of 100% to soil.”

We tested the sensitivity of impacts from pesticide residues in the harvested part of the crop comparing a parametrization of the emission model for tropical conditions against fixed emission fractions as described in the default plant uptake model. Across scenarios, residue impacts are on average a factor 25 higher when using initial distribution fractions parametrized to tropical conditions (up to a factor 147 for some substances; see details of residue impact per scenario and pesticide in SM, Section S3.10). With the new coupling of emission fractions and the parametrization of plant uptake processes to tropical conditions, we could, hence, significantly reduce the uncertainty associated with climate and soil characteristics in impact estimates due to residues, which in several scenarios dominate LCA human toxicity impacts at the product system level.

3.4 Discussion

3.4.1 Applicability and limitations of our approach

Our approach of coupled LCI and LCIA models, covering the different emission- and crop-residue related exposure pathways is applicable in LCA studies focusing on evaluating the environmental performance of crop production. Coupling the different models helps to overcome currently prevailing assumptions for pesticide emissions (leading to overestimation of freshwater ecotoxicity when considering field soil part of the ecosphere) and to consider pesticide residues in crops as contributor to human toxicity, which is currently mostly missing in LCA studies (leading to underestimation of human toxicity impacts). Coupling the different LCI and LCIA models required the adaptation of these models at different levels. We modified the dynamiCROP model to account for variable emission fractions from PestLCI Consensus as starting point instead of using generic estimates. This combination of models and the parametrization to local conditions (in particular tropical conditions) allows a consistent mass flow of pesticides from application to residues at crop harvest time. The coupling of secondary emission fractions to dynamiCROP requires further research related to removing overlaps in the modeled processes.

Although initial distribution fractions are less refined than secondary emission fractions, they can be consistently combined with existing steady-state LCIA models like USEtox. This approach is hence generally suitable for LCA studies, where the farm stage is both in the background and foreground system (consideration of application method and its drift). Using the secondary emission fractions is more demanding in primary data but seems more appropriate for LCA studies where the farm stage is part of the foreground system and where the purpose is to discriminate more specifically pest management practices and/or climate and soil characteristics. Nevertheless, current secondary emission modeling does not yet discriminate well enough farming practices and local conditions, such as soil and climate, in particular for tropical conditions. Especially water flux characteristics and related-processes should be better considered for tropical conditions and constitutes a current limitation (Gentil et al. 2020b). However, even if secondary emission fractions allow for better consideration of field properties and regional aspects associated with the pesticide application scenario not captured in generic LCIA models, the (eco-)toxicity impacts were not significantly different, mainly due to high uncertainty of characterization factors and missing characterization factors accounting for tropical conditions (i.e. tropical species' sensitivity to ecotoxicity effects). This, hence, constitutes another limitation of our approach, and requires further efforts to adapt characterization models to tropical conditions. Furthermore, coupling secondary emissions with LCIA models requires additional research to address current overlaps in modeling processes between application time and the time of secondary emissions, potentially leading to double counting of e.g. degradation and leaching (Rosenbaum et al. 2015).

3.4.2 LCA comparisons and limitations

Various LCA studies focusing on one or more aspects of tomato production exist (Bojacá et al. 2014; Khoshnevisan et al. 2014; Ingraio et al. 2019; Payen et al. 2015; Romero-Gómez et al. 2017; Zarei et al. 2019). Detail on existing LCA studies are presented in SM, Section S3.11). Out of these studies, all but one, namely Bojacá et al. (2014), applied generic emission factors instead of a mass balance-based emission model. In addition, none of these studies included pesticide residues in food crops as human exposure pathway, although it has been shown to be the predominant pathway contributing to human toxicity impacts in LCA (Juraske et al. 2009; Fantke and Jolliet 2016). Furthermore, almost all existing studies were conducted under conditions other than tropical climates. Indeed, LCA studies on open-field tomato in tropical conditions including (eco-)toxicity impacts due to pesticides are scarce (Perrin et al. 2014). The very few studies conducted under tropical conditions did not adapt their models for tropical conditions (Basset-Mens et al 2016, Payen et al 2015, Perrin et al 2015). These factors rendered it difficult to compare our findings with results from other LCA studies. However, for several of our own datasets for tomato LCA, namely for Rwanda (Basset-Mens et al. 2016a), Morocco (Payen et al. 2015) and Benin (Perrin et al. 2015), we updated (eco-)toxicity impacts from tomato production using USEtox 2.1 for organic substances (Figure 16) to facilitate a comparison with results from our present study. Across other studies, we applied the current assumption of 100% of applied pesticide mass is emitted to agricultural soil. Across studies, freshwater ecotoxicity was higher than in our open-field tomato study in Martinique. For tomato grown in cold greenhouses in Morocco (Payen et al. 2015) and in open-field in Martinique (this study) emissions of organic pesticides represented the major contributor to ecotoxicity impacts, dominated by insecticides and fungicides, in line with conclusions from other studies in Bojacá et al. (2013) and Kariathi et al. (2016). It was not possible to compare total human toxicity impacts for tomato in Martinique with results from Benin and Rwanda, since pesticide residues in tomato were not included in any of the other case studies. When only comparing results without residues, our study shows the lowest impacts across studies, mainly due to ingestion-related impacts from agricultural soil emissions in other studies. However, when including residue-related impacts, our toxicity impacts exceed impacts from all other studies.

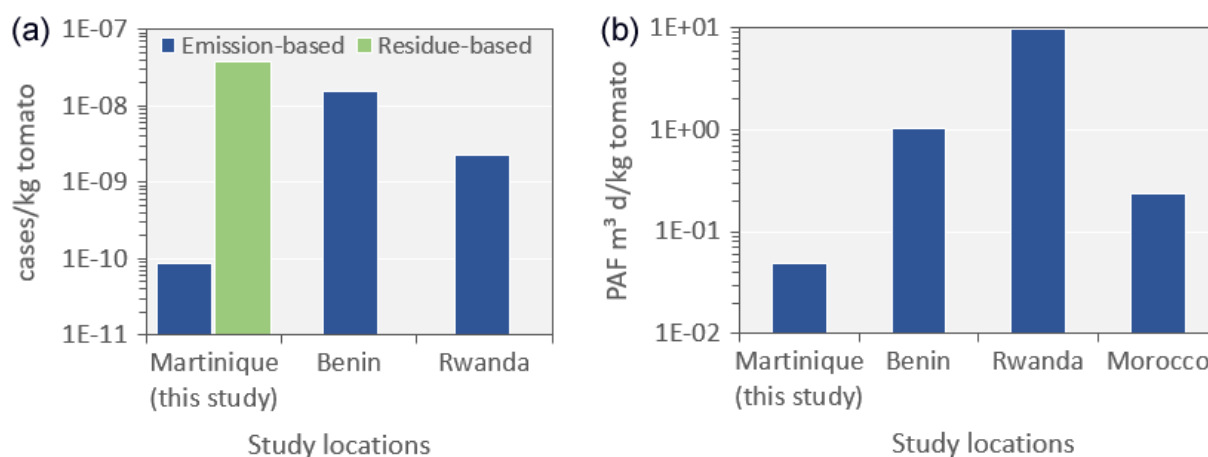


Figure 16: Comparison of (a) human non-cancer toxicity and (b) freshwater toxicity impacts for organic substances for cradle-to-farm gate tomato LCA studies in open-field production in Martinique (our study), Benin in open-field (Perrin et al. 2015), Rwanda in open-field (Basset-Mens et al. 2016a), and Morocco in cold greenhouse (Payen et al. 2015).

When ignoring residue-related impacts, the cradle-to-farm gate (eco-)toxic impacts per kg tomato for our sample of farms appeared relatively low compared to those from Rwanda, Benin and Morocco, despite high pests and weeds pressure on tomato crop in Martinique and relatively low yields (around 22 tonnes/ha on average). This can partly be explained by applying pesticides with higher toxicity potential in Rwanda and Benin as compared to Martinique, of which the latter only applied pesticides that are registered for agricultural use in the European Union. Furthermore, farmers in Martinique generally respect the European recommended application doses and pre-harvest intervals, which is often not respected in other tropical countries, leading to over- and misuse of pesticides ; such as in Tanzania, where 18% of farmers overdosed pesticides (Kariathi et al. 2016) or in urban gardens as in Benin (Perrin et al. 2017), potentially increasing the risk for pest resistance and high accumulation of residues in tomato and in the environment. Moreover, this detailed data on pesticide practices of Martinique tomato producers is a pioneering achievement and could not be compared with other quantitative data on pest management for tomato in Martinique. This highlights the drastic lack of quantitative data on pesticide practices in vegetable crops in Martinique in particular and under tropical conditions in general.

Pest management on tomato crop in Martinique still presents some margins of improvement. In our sample, two types of pesticide treatment strategies can be distinguished, curative and preventive. Some farmers apply a pesticide preventively every week alternating a fungicide and an insecticide. Better using crop rotations and associations and training farmers to identify pests early and then only apply a curative treatment, if necessary, could lead to a reduction in the number of applications per production cycle. Furthermore, the copper sulfate (Bordeaux mixture) is generally not considered as a chemical pesticide, being authorized in organic agriculture. However, we have shown that copper sulfate

has a significant impact on freshwater ecosystems. Therefore, we propose that this substance is systematically included in the assessment of ecotoxicity impacts in LCA studies, especially when comparing conventional with organic farming practices.

Despite a diversified sample of farms (in the North and South of the island with 2 distinct soil types and 3 climate types), only 6 farms could be surveyed. Identifying tomato production farms was difficult, as well as involving farmers in the semi-directive survey. Since we cannot evaluate the representativeness of our sample by lack of existing data on pest management on tomato in Martinique, an extrapolation of our results remains difficult. However, our estimates can be used along with other LCA studies to get an overview of the range of impacts related to pesticide use in tomato production systems. Results on specific pesticide practices thereby should be kept as detailed as possible whenever pest management practices are in focus in a given LCA study in order to highlight related hotspots and target processes for emission and impact reduction. Whenever pest management practice is just one out of many aspects that are evaluated, related processes can be aggregated for better comparison with other considered aspects or processes.

3.4.3 Future research needs

The main current limitations of the presented approach require additional research efforts as detailed in the following. The crop uptake and secondary emissions for metal-based substances cannot be evaluated with the current models. Adaptation from USEtox for environmental processes could be considered, but further research is required to consider metal speciation and equilibrium partitioning in emission distribution and plant uptake processes, which might additionally depend on the emission location (Peña et al. 2018). Coupling secondary emission fractions with LCIA models and addressing the potential overlap in modeled processes should furthermore be developed.

Pesticide emission and impact models are mainly parametrized for European conditions and have been extended to the whole world, using global databases of soils and climates. However, their validity for crops grown under tropical conditions remains questionable (Gentil et al. 2020b), which is confirmed in our present study. Drift curves developed under real tropical crop conditions are currently not implemented, and neither are growth phase-specific interception fractions for crops under tropical conditions. Furthermore, common farming practices under tropical conditions, such as ground cover management, could not be considered, while weeding between tomato rows is frequent in these conditions where the plots may feature a large slope (25%). Agro-ecological practices, such as ground cover management to reduce pesticide uses and emissions through soil protection (e.g. mulch), will have to be integrated in pesticide emission assessment. A dedicated module to include ground cover management in PestLCI Consensus is currently under development based on the work initiated by

Renaud-Gentié et al. (2015). Associated with intense rain events all year round in humid tropical conditions, these aspects will have consequences on water fluxes, which require further model adaptation (Sanchez-Bayo and Hyne 2011; Mottes et al. 2017b).

Tropical species' sensitivity to ecotoxicity effects and related characterization results should be further developed in order to reduce the main remaining uncertainty in impact results for pesticides. Finally, sources of uncertainty in pesticide emission and fate modeling are numerous, and their estimations are challenging (Dubus et al. 2003). Improving these aspects will ultimately help making LCA results for pesticides more reliable, and help linking product life cycles to targets for reducing chemical pollution (Fantke and Illner, 2019).

3.5 Conclusions

The main purpose of this study was to combine state-of-the-art LCI and LCIA models for assessing the emissions and (eco-)toxicity impacts due to pesticide applications, associated with the production of open-field tomato in Martinique as part of a complete cradle-to-farm gate LCA study. We developed an approach for consistently combining PestLCI Consensus, USEtox and dynamiCROP to allow an operational assessment of pesticide (eco-)toxicity impacts including the consideration of the main route of exposure for human health, namely pesticide residues in crops ingestion. Our formalization for properly connecting the models will help practitioners evaluate these impacts in their LCA studies. This will provide a more consistent estimation of pesticides' uptake into crops and an easier consideration of pesticide residues in crops in LCA studies of agricultural products. Regarding the use of PestLCI Consensus for LCA studies, where the agricultural stage is part of the foreground system, the secondary emission fractions allow to take into account farming practices, climate and soil conditions. However, the relatively small differences in impact results based on using initial distribution fractions versus secondary emission fractions indicate that using the former is suitable as a first proxy, while uncertainties in impact characterization should be addressed in complement of further refining emission estimates. With the presented model coupling, the initial distribution of PestLCI is fully consistent with USEtox and dynamiCROP, taking into account already application, active ingredient and the mass applied.

Applied to the best possible sample of open-field tomato plots in Martinique, the use of these three models together revealed that despite a high pest and disease pressure in tropical humid conditions, the freshwater ecotoxicity and human toxicity impacts were low compared to impacts for other tomato production systems. Our sample of tomato farmers in Martinique respected the homologated doses, pre-harvest interval and consequently did not exceed MRLs. However, pesticide management is a good starting-point to further improving the environmental performance of tomato production in Martinique.

Further pesticide use data should be collected to validate our results. As already demonstrated by Fantke and Jolliet (2016), the use of dynamiCROP allows highlighting the major impact of pesticide residues in crops on human toxicity. Analyzing separately organic and metal-based substances allows highlighting dominating contributors in both substance categories. Further adaptation of PestLCI Consensus remains necessary, especially on the inclusion of agro-ecological practices and a better accounting of water flows and on other specificities of tropical conditions and its crops. Yet, our proposed approach constitutes a valuable starting point for improving the assessment of pesticides in LCA. Next steps to advance and further refine our approach are to develop and implement additional drift deposition functions, account for ground cover management and address data gaps in the existing models, for example to consider inorganic substances.

Acknowledgments

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References Chapter 3

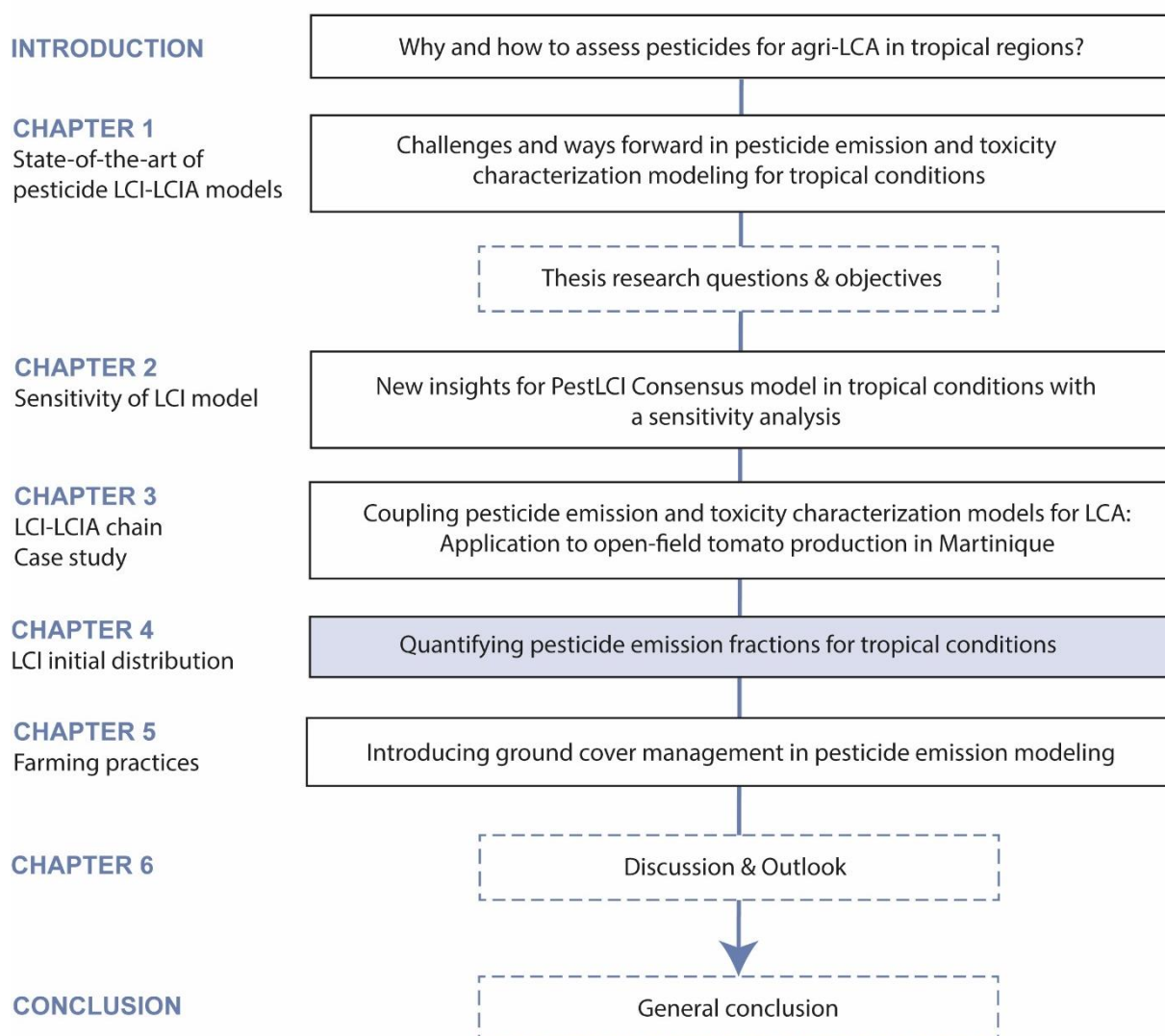
- Aktar MdW, Sengupta D, Chowdhury A (2009) Impact of pesticides use in agriculture: their benefits and hazards. *Interdiscip Toxicol* 2:1–12. doi.org/10.2478/v10102-009-0001-7
- Arias-Estévez M, López-Periago E, Martínez-Carballo E, et al (2008) The mobility and degradation of pesticides in soils and the pollution of groundwater resources. *Agri Ecosyst Environ* 123:247–260. doi.org/10.1016/j.agee.2007.07.011
- Basset-Mens C, Kleih U, Martin A (2016) Value chain analysis of the tomato value chain from Rwamagana, Rwanda. ISS-FANSSA-BX11 Project for the European commission – DEVCO 124
- Bessou C, Basset-Mens C, Tran T, Benoist A (2013) LCA applied to perennial cropping systems: a review focused on the farm stage. *Int J Life Cycle Assess* 18:340–361. doi.org/10.1007/s11367-012-0502-z
- Bojacá CR, Arias LA, Ahumada DA, et al (2013) Evaluation of pesticide residues in open field and greenhouse tomatoes from Colombia. *Food Control* 30:400–403. doi.org/10.1016/j.foodcont.2012.08.015

- Bojacá CR, Wyckhuys KAG, Schrevens E (2014) Life cycle assessment of Colombian greenhouse tomato production based on farmer-level survey data. *J Clean Prod* 69:26–33. doi.org/10.1016/j.jclepro.2014.01.078
- Coste G, Biard Y, Roux P, Helias A (2018) ELDAM, a new quality management system for LCI datasets exchange and review. In: Book of abstracts of the 11th International Conference on Life Cycle Assessment of Food 2018 (LCA Food) “Global food challenges towards sustainable consumption and production.” Life Cycle Assessment
- Daam MA, van den Brink PJ (2010) Implications of differences between temperate and tropical freshwater ecosystems for the ecological risk assessment of pesticides. *Ecotoxicology* 19:24–37. doi.org/10.1007/s10646-009-0402-6
- Dijkman TJ, Birkved M, Hauschild MZ (2012) PestLCI 2.0: a second generation model for estimating emissions of pesticides from arable land in LCA. *Int J Life Cycle Assess* 17:973–986. doi.org/10.1007/s11367-012-0439-2
- Dong Y, Gandhi N, Hauschild MZ (2014) Development of Comparative Toxicity Potentials of 14 cationic metals in freshwater. *Chemosphere* 112:26–33. doi.org/10.1016/j.chemosphere.2014.03.046
- Dubus IG, Brown CD, Beulke S (2003) Sources of uncertainty in pesticide fate modelling. *Science of The Total Environment* 317:53–72. doi.org/10.1016/S0048-9697(03)00362-0
- Fantke P (2019) Modeling the environmental impacts of pesticides in agriculture. In: Weidema, B.P. (Ed) *Assessing the environmental impact of agriculture*, Burleigh Dodds Science Publishing, Cambridge, United Kingdom. doi.org/10.19103/AS.2018.0044.08
- Fantke P, Antón A, Grant T, Hayashi K (2017) Pesticide emission quantification for life cycle assessment: A global consensus building process. *J Life Cycle Assess* 13:245–251
- Fantke P, Aurisano N, Bare J, et al (2018a) Toward harmonizing ecotoxicity characterization in life cycle impact assessment. *Environ Toxicol Chem* 37:2955–2971. doi.org/10.1002/etc.4261
- Fantke P, Aylward Lesa, Bare Jane, et al (2018b) Advancements in Life Cycle Human Exposure and Toxicity Characterization. *Environmental Health Perspectives* 126:125001. doi.org/10.1289/EHP3871
- Fantke P, Charles R, de Alencastro LF, et al (2011a) Plant uptake of pesticides and human health: Dynamic modeling of residues in wheat and ingestion intake. *Chemosphere* 85:1639–1647. doi.org/10.1016/j.chemosphere.2011.08.030
- Fantke P, Friedrich R, Jolliet O (2012a) Health impact and damage cost assessment of pesticides in Europe. *Enviro Int* 49:9–17. doi.org/10.1016/j.envint.2012.08.001
- Fantke, P, Illner, N, 2019. Goods that are good enough: Introducing an absolute sustainability perspective for managing chemicals in consumer products. *Curr. Opin. Green Sustain. Chem.* 15, 91e97. doi.org/10.1016/j.cogsc.2018.12.001.
- Fantke P, Jolliet O (2016) Life cycle human health impacts of 875 pesticides. *Int J Life Cycle Assess* 21:722–733. doi.org/10.1007/s11367-015-0910-y
- Fantke P, Juraske R, Antón A, et al (2011b) Dynamic multicrop model to characterize impacts of pesticides in food. *Environ Sci Technol* 45:8842–8849. doi.org/10.1021/es201989d

- Fantke P, Wieland P, Juraske R, et al (2012b) Parameterization models for pesticide exposure via crop consumption. *Environ Sci Technol* 46:12864–12872. doi.org/10.1021/es301509u
- Fantke P, Wieland P, Wannaz C, et al (2013) Dynamics of pesticide uptake into plants: From system functioning to parsimonious modeling. *Environ Modell Softw* 40:316–324. doi.org/10.1016/j.envsoft.2012.09.016
- Frischknecht R, Jolliet O (2019) *Global Guidance for Life Cycle Impact Assessment Indicators*. Volume 2. United Nations Environment Programme
- Gentil C, Fantke P, Mottes C, Basset-Mens C (2020) Challenges and ways forward in pesticide emission and toxicity characterization modeling for tropical conditions. *Int J Life Cycle Assess* 25:1290–1306. doi.org/10.1007/s11367-019-01685-9
- Ingwersen WW (2012) Life cycle assessment of fresh pineapple from Costa Rica. *J Clean Prod* 35:152–163. doi.org/10.1016/j.jclepro.2012.05.035
- ISO 14040 (2006) *Management environnemental — Analyse du cycle de vie — Principes et cadre*. ISO
- ISO 14044 (2006) *Environmental management — Life cycle assessment — Requirements and guidelines*. International Standards Organization, 14044:2006
- Juraske R, Fantke P, Ramírez ACR, González A (2012) Pesticide residue dynamics in passion fruits: Comparing field trial and modelling results. *Chemosphere* 89:850–855. doi.org/10.1016/j.chemosphere.2012.05.007
- Kariathi V, Kassim N, Kimanya M (2016) Pesticide exposure from fresh tomatoes and its relationship with pesticide application practices in Meru district. *Cogent Food & Agriculture* 2:1196808. doi.org/10.1080/23311932.2016.1196808
- Kaushik G, Satya S, Naik SN (2009) Food processing a tool to pesticide residue dissipation – A review. *Food Research International* 42:26–40. doi.org/10.1016/j.foodres.2008.09.009
- Khoshnevisan B, Rafiee S, Omid M, et al (2014) Environmental impact assessment of tomato and cucumber cultivation in greenhouses using life cycle assessment and adaptive neuro-fuzzy inference system. *J Clean Prod* 73:183–192. doi.org/10.1016/j.jclepro.2013.09.057
- Knudsen MT, Dorca-Preda T, Djomo SN, et al (2019) The importance of including soil carbon changes, ecotoxicity and biodiversity impacts in environmental life cycle assessments of organic and conventional milk in Western Europe. *J Clean Prod* 215:433–443. doi.org/10.1016/j.jclepro.2018.12.273
- Lesueur Jannoyer M, Cattan P, Woignier T, Clostre F (2016) *Crisis management of chronic pollution: contaminated soil and human health*, 1st Edition. CRC Press
- Lewis SE, Silburn DM, Kookana RS, Shaw M (2016) Pesticide behavior, fate, and effects in the tropics: an overview of the current state of knowledge. *J Agric Food Chem* 64:3917–3924. doi.org/10.1021/acs.jafc.6b01320
- Martínez-Blanco J, Muñoz P, Antón A, Rieradevall J (2011) Assessment of tomato Mediterranean production in open-field and standard multi-tunnel greenhouse, with compost or mineral fertilizers, from an agricultural and environmental standpoint. *J Clean Prod* 19:985–997. doi.org/10.1016/j.jclepro.2010.11.018

- Meier MS, Stoessel F, Jungbluth N, et al (2015) Environmental impacts of organic and conventional agricultural products – Are the differences captured by life cycle assessment? *J Environ Manag* 149:193–208. doi.org/10.1016/j.jenvman.2014.10.006
- Mottes C, Lesueur Jannoyer M, Le Bail M, et al (2017) Relationships between past and present pesticide applications and pollution at a watershed outlet: The case of a horticultural catchment in Martinique, French West Indies. *Chemosphere* 184:762–773. doi.org/10.1016/j.chemosphere.2017.06.061
- Nemecek T, Schnetzer J (2011) Methods of assessment of direct field emissions for LCIs of agricultural production systems. Data v3.0. Swiss Center for Life Cycle Inventories, Duebendorf, Switzerland
- Oliquino-Abasolo A (2015) Agro-environmental sustainability of conventional and organic vegetable production systems in Tayabas, Quezon, Philippines. FAO University Library, University of the Philippines at Los Baños:
- Payen S, Basset-Mens C, Perret S (2015) LCA of local and imported tomato: an energy and water trade-off. *J Clean Prod* 87:139–148. doi.org/10.1016/j.jclepro.2014.10.007
- Peña N, Antón A, Kamilaris A, Fantke P (2018) Modeling ecotoxicity impacts in vineyard production: Addressing spatial differentiation for copper fungicides. *Sci Total Environ* 616–617:796–804. doi.org/10.1016/j.scitotenv.2017.10.243
- Perrin A, Basset-Mens C, Gabrielle B (2014) Life cycle assessment of vegetable products: a review focusing on cropping systems diversity and the estimation of field emissions. *Int J Life Cycle Assess* 19:1247–1263. doi.org/10.1007/s11367-014-0724-3
- Perrin A, Basset-Mens C, Huat J, Gabrielle B (2017) The variability of field emissions is critical to assessing the environmental impacts of vegetables: A Benin case-study. *J Clean Prod* 153:104–113. doi.org/10.1016/j.jclepro.2017.03.159
- Perrin A, Basset-Mens C, Huat J, Yehouessi W (2015) High environmental risk and low yield of urban tomato gardens in Benin. *Agron Sustain Dev* 35:305–315. doi.org/10.1007/s13593-014-0241-6
- Racke KD, Skidmore MW, Hamilton DJ, et al (1997) Pesticides report 38. Pesticide fate in tropical soils - (Technical report). *Pure Appl Chem* 69:1349–1371. doi.org/10.1351/pac199769061349
- Renaud-Gentié C, Dijkman TJ, Bjørn A, Birkved M (2015) Pesticide emission modelling and freshwater ecotoxicity assessment for Grapevine LCA: adaptation of PestLCI 2.0 to viticulture. *Int J Life Cycle Assess* 20:1528–1543. doi.org/10.1007/s11367-015-0949-9
- Romero-Gámez M, Antón A, Leyva R, Suárez-Rey EM (2017) Inclusion of uncertainty in the LCA comparison of different cherry tomato production scenarios. *Int J Life Cycle Assess* 22:798–811. doi.org/10.1007/s11367-016-1225-3
- Rosenbaum RK, Antón A, Bengoa X, et al (2015) The Glasgow consensus on the delineation between pesticide emission inventory and impact assessment for LCA. *Int J Life Cycle Assess* 20:765–776. doi.org/10.1007/s11367-015-0871-1
- Rosenbaum RK, Bachmann TM, Gold LS, et al (2008) USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment. *Int J Life Cycle Assess* 13:532. doi.org/10.1007/s11367-008-0038-4
- Sanchez-Bayo F, Hyne RV (2011) Comparison of environmental risks of pesticides between tropical and nontropical regions. *Integr Environ Assess Manag* 7:577–586. doi.org/10.1002/ieam.189

- van der Werf HMG, Knudsen MT, Cederberg C (2020) Towards better representation of organic agriculture in life cycle assessment. *Nature Sustainability* 3:419–425. doi.org/10.1038/s41893-020-0489-6
- van Zelm R, Larrey-Lassalle P, Roux P (2014) Bridging the gap between life cycle inventory and impact assessment for toxicological assessments of pesticides used in crop production. *Chemosphere* 100:175–181. doi.org/10.1016/j.chemosphere.2013.11.037
- Weidema BP, Stylianou KS (2019) Nutrition in the life cycle assessment of foods—function or impact? *Int J Life Cycle Assess.* doi.org/10.1007/s11367-019-01658-y
- Weinberger K, Lumpkin TA (2007) Diversification into horticulture and poverty reduction: a research agenda. *World Development* 35:1464–1480. doi.org/10.1016/j.worlddev.2007.05.002
- Westh TB, Hauschild MZ, Birkved M, et al (2015) The USEtox story: a survey of model developer visions and user requirements. *Int J Life Cycle Assess* 20:299–310. doi.org/10.1007/s11367-014-0829-8
- Zarei MJ, Kazemi N, Marzban A (2019) Life cycle environmental impacts of cucumber and tomato production in open-field and greenhouse. *Journal of the Saudi Society of Agricultural Sciences* 18:249–255. doi.org/10.1016/j.jssas.2017.07.001



CHAPTER 4

QUANTIFYING PESTICIDE EMISSION FRACTIONS FOR TROPICAL CONDITIONS

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Abstract

The inventory model ‘PestLCI Consensus’, originally developed for temperate conditions, estimates initial pesticide emission fractions to air, to off-field surfaces by drift deposition, and to field crop and field soil surfaces according to crop foliar interception characteristics. Since crop characteristics and application techniques differ in tropical conditions, these aspects need to be included in the model in support of evaluating pesticide emissions under tropical conditions.

Based on published literature, a consistent set of crop foliar interception fractions was developed as function of crop characteristics and spraying techniques for tropical crops. In addition, we derived drift deposition fractions from published drift experiments specifically conducted under tropical conditions. Finally, we compiled a consistent set of pesticide emission fractions for application in life cycle assessment (LCA).

Foliar interception fractions are strongly influenced by the spraying technique, particularly for hand-operated applications. Drift deposition fractions to off-field surfaces were derived for air blast sprayer

on papaya and coffee, for boom sprayer on bean and soybean, for aerial application on soybean, sorghum, millet, corn and cotton, and for hand-operated application on cotton. Emission fractions vary for each combination of crop and application method. Drift deposition curves for missing crop-application method combinations can only partly be extrapolated from the set of considered combinations.

Overall, our proposed foliar interception fractions and drift deposition fractions for various crops grown under tropical conditions allow to estimate pesticide emissions in support of assessing the environmental performance of agrifood systems in LCA with focus on tropical regions.

Keywords: Active ingredient, pest management, drift deposition, Life Cycle Assessment, tropical crops, foliar interception

4.1 Introduction

In tropical contexts, environmental conditions are favorable to grow all types of agricultural products, but they are also favorable for the occurrence of pests (insects, weeds, fungi etc.), requiring adequate pest control measures all year round (Daam and van den Brink 2010; Mottes et al. 2017b). Indeed, under tropical conditions, most farmers use a wide diversity of pesticides often at high dose and frequency to control pests and to ensure high crop yields (Racke et al. 1997; Lewis et al. 2016), leading to potential pesticide-related environmental and human health impacts (Arias-Estévez et al. 2008; Lesueur Jannoyer et al. 2016). Hence, the crucial need to properly assess emissions and (eco-)toxicological impacts from pesticide applications in Life Cycle Assessment (LCA) of crop production in tropical conditions (Perrin et al. 2014; Meier et al. 2015; Knudsen et al. 2019; van der Werf et al. 2020).

Pesticide emissions in LCA are generally estimated using default factors that do not account for relevant crop or application characteristics (Oliquino-Abasolo 2015; Wang et al. 2018; Li et al. 2019), and in the most common approach,ecoinvent, 100% of the applied dose of pesticides is assumed to be emitted directly to agricultural field soil (Nemecek and Schnetzer 2011). This approach is simple but according to our previous work, is very likely to underestimate emissions reaching vulnerable non-target ecosystems, especially in tropical conditions (Gentil et al. 2020b). More sophisticated approaches exist, such as the pesticide life cycle inventory (LCI) model PestLCI (Birkved and Hauschild 2006; Dijkman et al. 2012), which allows to estimate emission fractions to air, field soil, field crop, groundwater and off-field surfaces, and has been further advanced into PestLCI Consensus (Fantke et al. 2017a). However, this model was parameterized primarily for temperate conditions, leading to a high uncertainty of results representing tropical conditions (Gentil et al. 2020b).

Initial pesticide mass distribution in PestLCI Consensus considers processes within the first minutes after application, including pesticide deposition on crop, soil and off-field surfaces by drift deposition, and to air by airborne drift, mainly modeled as a function of application method, field characteristics and foliar interception of field crops. The secondary emission distribution in PestLCI Consensus is additionally considering processes dependent on farming practices, climate and soil conditions, and pesticide properties. Drift deposition to off-field surfaces influences both initial and then secondary distributions; however, only drift deposition curves defined for crops grown in temperate conditions are available in the model, mainly from Germany and the Netherlands (Ganzelmeier et al. 1995; Rautmann et al. 2001; Holterman and van de Zande 2003; van de Zande et al. 2015; Gil et al. 2018). Drift during pesticide application is influenced by several parameters, which might differ under tropical conditions compared to temperate ones. This includes meteorological conditions, such as temperature (Nuyttens et al. 2006a), humidity, with a decrease of drift by less evaporation reducing the droplets size (Nuyttens et al. 2006a; Franke et al. 2010), and wind speed (not specific to tropical conditions) (Grella et al. 2017). In addition, application methods and nozzle types (Nuyttens et al. 2006b), pesticides and mixtures composition (de Schamphelleire et al. 2005; Hewitt et al. 2009; Langenbach and Caldas 2018) are other factors influencing drift, as well as the type of crop and its canopy shape (Garcerá et al. 2017; Gil et al. 2018). Thus, a refinement of drift deposition curves under tropical conditions is necessary (Gentil et al. 2020b), in particular to refine off-field surfaces emission estimates. Moreover, crop pesticide interception is another key variable of initial distribution of pesticides. It is influenced by the application method, the crop canopy and planting configuration, and also the crop growth stage (Larbi and Salyani 2012), which depend on climatic and agronomic conditions (Lammoglia et al. 2016). The foliar interception fraction is defined as the fraction that the crop leaves can intercept from an applied pesticide amount. In the current PestLCI Consensus model, these fractions were defined based on Linders et al. (2000) using the growth stages with BBCH-scale (Meier 2018). A wide diversity of crops including those that can grow both in temperate and tropical conditions (e.g. tomato, bean, tobacco) were determined, whereas various crops growing in tropical conditions are missing, such as banana or pineapple. Furthermore, the foliar interception fraction from Linders et al. (2000) are mostly calculated considering a default fraction of the dosage to the air of 0.1, whereas the foliar interception fraction in PestLCI Consensus is used to determine the pesticide fraction reaching the field area only, i.e. the field crop leaves according to the growth stage (and its related leaf area index: LAI) and then the soil. Few data exist on crop foliar interception for tropical crops. As considered by Mottes (2013), the spraying technique must be considered to define the crop foliar interception fraction, which determines the fraction that reaches crop leaf surfaces versus field soil surfaces. A first important step to better account for tropical conditions in LCI emission modeling for pesticides is to refine the initial distribution of PestLCI Consensus, considering drift deposition curves defined in tropical conditions and foliar interception fractions for tropical crops.

To address this aspect, we aim in the present study at answering the question “How can the calculation of initial pesticide emission fractions be adapted for crops grown under tropical conditions?” To answer this question, we defined three specific objectives: (i) To develop a consistent set of crop growth stages and associated foliar interception fractions according to crop growth and spraying technique under tropical conditions (ii) To propose drift deposition curves defined for tropical crop conditions and application methods (iii) To implement these data in the PestLCI Consensus model and to calculate related emission fractions for different crops grown in tropical conditions for LCA practitioners.

4.2 Materials and methods

4.2.1 Emission fraction mass balance and foliar interception fraction calculation

With a set of input variables, the initial distribution of PestLCI Consensus web-tool estimates pesticide depositions on crop and soil according to the foliar interception fraction, on off-field surfaces by drift according to application method, and airborne emissions via wind drift. The input variables influencing the output fractions are presented in Table 10, and related calculation steps are presented in the following. Figure 17 presents the initial emission fractions to air (f_{air}), off-field surfaces (f_{dep}), to field soil ($f_{\text{field} \rightarrow \text{soil}}$) and field crop ($f_{\text{field} \rightarrow \text{crop}}$) according to the spraying technique.

Table 10: Initial distribution input variables related to output fractions in PestLCI Consensus

Initial distribution input variables	Range of variables	Output fractions determined by initial distribution input variables
Foliar interception fraction	$0 \leq x \leq 1$	$f_{\text{field} \rightarrow \text{crop}} ; f_{\text{field} \rightarrow \text{soil}}$
Crop growth stage	Set of crop growth stages	$f_{\text{field} \rightarrow \text{crop}} ; f_{\text{field} \rightarrow \text{soil}}$
Application method	Set of application methods	$f_{\text{air}} ; f_{\text{dep}}$
Spraying technique	Application over – under the canopy, plant-by-plant application, soil application	$f_{\text{field} \rightarrow \text{crop}} ; f_{\text{field} \rightarrow \text{soil}}$

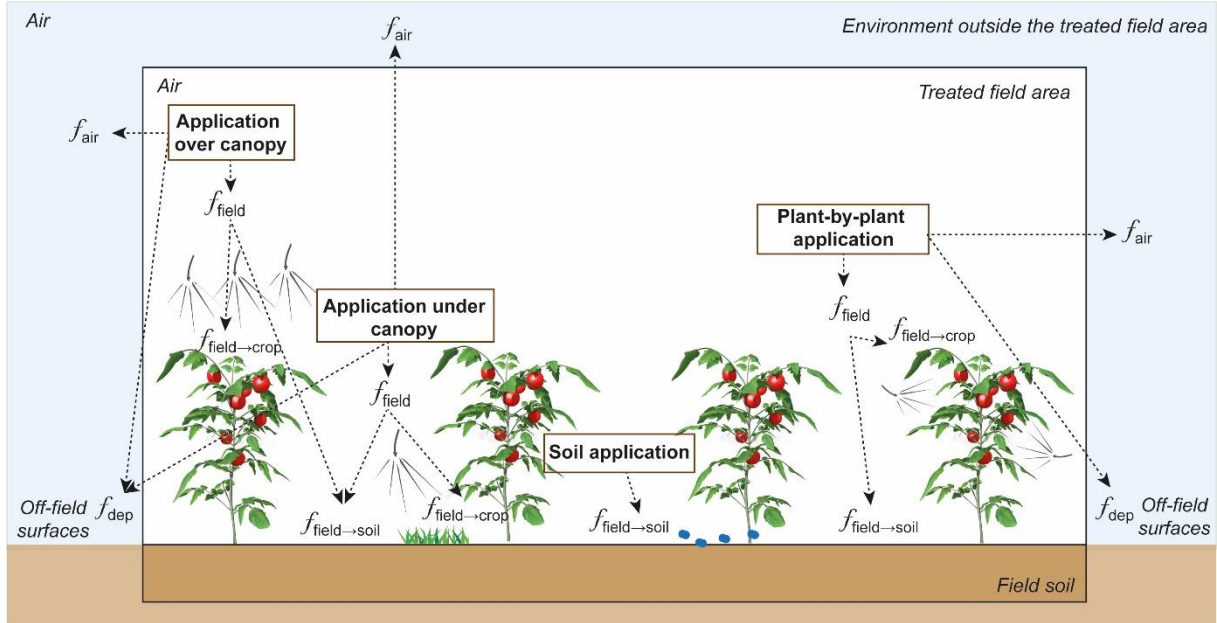


Figure 17: Conceptual overview of initial emission fractions to air (f_{air}), off-field surfaces (f_{dep}), field soil surfaces ($f_{\text{field} \rightarrow \text{soil}}$) and field crop surfaces ($f_{\text{field} \rightarrow \text{crop}}$) for different spraying techniques (application over canopy, under canopy, soil application and plant-by-plant application).

As a starting point, within minutes after pesticide application, a certain fraction of the pesticide mass is deposited to off-field surfaces (f_{dep}) derived from drift deposition functions specific to each application method and crop type (Eq. 4.2). Another fraction remains airborne and undergoes wind drift (f_{air}), derived as default fraction per application method and crop. The remaining fraction deposits on the field (f_{field} , Eq. 4.3), derived as follows (Eq. 4.1):

$$1 = f_{\text{field}} + f_{\text{air}} + f_{\text{dep}} \quad (4.1)$$

The drift deposition fraction obtained from the integration of any given drift deposition curve $f(x)$ as a function of distance x (m) to the field edge is calculated as (f_{dep}):

$$f_{\text{dep}} = \int_{x_1}^{x_2} f(x) dx \times \frac{1}{x_2} \times \frac{A_{\text{dep}}}{A_{\text{field}}}$$

with

$$A_{\text{dep}} = L_f \times Z_2$$

$$A_{\text{field}} = L_f \times W_f \quad (4.2)$$

where A_{dep} is the off-field deposition area in downwind direction from the treated field (m^2), and A_{field} is the agricultural field area relevant for the pesticide application (m^2), with L_f as field length (m) parallel to the edge between treated field and deposition area, W_f as field width (m) perpendicular to the edge between treated field and deposition area and Z_2 as upper validity limit of the drift deposition curve (m), which corresponds to the position of the last collector during the experiment. We realize that drift

functions are generic across field dimensions based on the defined setup of the underlying drift experiments, which derive drift deposition for a sufficiently large treated field area, so that any increase in treated area does not further increase drift deposition. A default square field with $L_f = W_f = 100$ m was hence assumed across our scenarios.

After re-arranging eq. 4.1, we obtain the fraction that deposits on the field surface area (f_{field}) as:

$$f_{\text{field}} = 1 - (f_{\text{air}} + f_{\text{dep}}) = f_{\text{field} \rightarrow \text{crop}} + f_{\text{field} \rightarrow \text{soil}} \quad (4.3)$$

The fraction reaching the field surface area (f_{field}) is partly depositing on crop leaves ($f_{\text{field} \rightarrow \text{crop}}$, Eq. 4.4) according to foliar interception fraction $f_{\text{intercept,crop}}$ derived from the crop growth stage, the application method and the pesticide spraying technique (details on Table 11), calculated as:

$$f_{\text{field} \rightarrow \text{crop}} = f_{\text{field}} \times f_{\text{intercept,crop}} \quad (4.4)$$

The remaining part of the fraction reaching the treated field is assumed to deposit on the field soil surface ($f_{\text{field} \rightarrow \text{soil}}$, Eq. 4.5), calculated as:

$$f_{\text{field} \rightarrow \text{soil}} = f_{\text{field}} \times (1 - f_{\text{intercept,crop}}) \quad (4.5)$$

The crop foliar interception fractions ($f_{\text{intercept,crop}}$) are defined as fractions that the crop can capture via its leaf surface area. The foliar interception fraction was derived from the fraction of field area that is covered by crop leaves, $fr_{\text{field-covered}}$, according to the main crop growth stages, using the leaf area index (LAI) and the pesticide spraying technique (Figure 17 and Table 11). The LAI is defined as the ratio of the leaf surface area (one side) per unit of ground surface area ($\text{m}^2 \text{m}^{-2}$).

The fraction of field covered by crop leaves (Eq. 4.6-4.7) was calculated from Mottes (2013) using the reported relation by Haverkort et al. (1991) and Cadarsa and Govinden (1999) with a default light extinction coefficient for cropland, $k_{\text{ext}} = 0.62$ (-) (Zhang et al. 2014), and a ‘maximum soil cover’ LAI of $a = 3 \text{ m}^2 \text{m}^{-2}$ (Allen and Pereira 2009 from the FAO-56 procedure); setting $fr_{\text{field-covered}}$ to 1 for $LAI \geq a$. For $LAI < a$, $fr_{\text{field-covered}}$ was calculated as:

$$fr_{\text{field-covered}} = 1.18437 \times (1 - e^{-0.62 \times LAI}) \quad (4.6)$$

where LAI is defined by a bibliographic review from (Mottes 2013), and completed for missing crops. All data are presented in the Supplementary Material (SM), Table S4.1. LAI could be refined according to specific LAI functions available as reported for six major crops in Fantke et al. (2011). Varying our default values for k_{ext} and a across crops, respectively, affected the resulting $fr_{\text{field-covered}}$ by less than 10%, which falls within the combined uncertainty of these parameters of at least a factor of 2 (Fantke et al. 2012b). However, if users find for a given crop very different k_{ext} and a , $fr_{\text{field-covered}}$ can be calculated as:

$$fr_{\text{field-covered}} = \frac{1 - e^{(-k_{\text{ext}} \times LAI)}}{1 - e^{(-k_{\text{ext}} \times a)}} \quad (4.7)$$

Each crop growth stage, namely (0) installation, (1) development, and (2) reproductive phase, corresponds to a range of growth stages according to the BBCH-scale of respectively 0-20, 30-50 and 60-80 for the first three (Meier 2018). A senescence phase (3) can be considered for BBCH-scale stage of 90. However, in tropical regions this phase is generally replaced by a physical destruction of the crop after few years of production (e.g. banana production) or after few months for vegetables, which is usually not relevant for pesticide application. Foliar interception fractions for a pesticide application at the senescence phase can also be estimated from the LAI, while the default LAI defined for the development stage (1) can be used where a more specific LAI is not available.

Then the spraying technique, i.e. the part of the field receiving the pesticide application related to the application method, is required to define the foliar interception fraction ($f_{\text{intercept,crop}}$). The different spraying techniques as defined in Mottes (2013) and in Mottes et al. (2014) are presented in Figure 17 and related calculation of the foliar interception fractions are detailed in Table 11.

Table 11: Pesticide spraying technique with corresponding application methods and calculation

Spraying technique	Corresponding application methods	Calculation
Application over the canopy	Aerial applicators, boom sprayers, air blast sprayers (e.g. on motorized field)	$f_{\text{intercept,crop}} = fr_{\text{field-covered}}$
Application under the canopy	Hand-operated sprayers (e.g. application of herbicide against weeds)	$f_{\text{intercept,crop}} = fr_{\text{field-covered}} \times 0.05$
Soil application	Granular or powder applicators, soil injectors (e.g. against soil insects)	$f_{\text{intercept,crop}} = 0$
Plant-by-plant application	Hand-operated sprayers* (e.g. on vegetable fruits)	$f_{\text{intercept,crop}} = LAI$ if $LAI < 0.95$, else $f_{\text{intercept,crop}} = 0.95$

*Hand-operated sprayers include several application methods, such as electric or manual air-pressure knapsack sprayer, backpack mist-blower (motorized) and low volume or ultra-low volume sprayer.

The spraying techniques are: (1) *application over the canopy*, where all sprays can reach the crop into the field according to $fr_{\text{field-covered}}$, (2) *application under the canopy*, where the pesticide is not applied on the main crop, although it is assumed that a small emission fraction reaches the crop, (3) *soil application*, where pesticide is applied on or in the soil, around the base of the crop (e.g. powder application), and (4) *plant-by-plant application*, done with a hand application method, where pesticides are applied plant-by-plant on all or part of the plant (e.g. only on fruits). Hand application is a common application method on crops grown in tropical conditions, generally on non-mechanized fields or crops such as horticulture production (Charlier et al. 2009) or on family farming systems. The spraying techniques are related to pesticide target classes. Application over the canopy is usually used for insecticides and fungicides, but can at times also be used for herbicides in specific crop production systems (e.g. on sugar cane at the beginning of the growth or on genetically modified glyphosate-tolerant

crop (Cuhra 2015)). Application under the canopy is usually used for herbicides in mono-cropping systems, but can also be used for insecticides or fungicides for double- or multi-cropping systems. Soil applications are composed of insecticides (generally in granular or powder). Plant-by-plant application is used for insecticides and fungicides to protect the whole crop or part of the crop (e.g. fruits).

For each pesticide spraying technique related to a set of application methods, foliar interception fractions were estimated for a range of crops or crop classes growing under tropical conditions as defined by climate A of the Köppen-Geiger classification (Kottek et al. 2006; Beck et al. 2018); for crops for which a drift deposition curve is available (i.e. bean, soybean, coffee) and for most important tropical crops according to area harvested in FAOSTAT (2020) (i.e. banana, pineapple, sugar cane, tubers, vegetable fruits, tropical fruit trees).

4.2.2 Drift deposition curves and application methods

According to a bibliographic research (detail in SM in Table S4.2) and a set of pre-defined criteria, drift deposition curves were selected. The criteria were defined as follows:

- (i) The drift deposition curves had to be developed under tropical weather conditions, mainly between the Tropic of Cancer and the Tropic of Capricorn. According to Köppen-Geiger climate classification, these weather conditions are characterized by warm temperatures with alternating rainy and dry seasons, or by an equatorial climate with all year round humid conditions (Kottek et al. 2006; Beck et al. 2018). Humid conditions correspond to $T_{\min} \geq +18\text{ }^{\circ}\text{C}$ (monthly mean temperature of the coldest month), $P_{\min} \geq 60\text{ mm}$ (mean precipitation of the driest month) for equatorial rainforest, $P_{\min} < 60\text{ mm}$ for equatorial savannah and $P_{\text{ann}} \geq 25 \times (100 - P_{\min})$ for equatorial monsoon (P_{ann} : mean cumulative annual precipitation).
- (ii) The experimentation to define drift deposition curves had to comply with ISO 22866:2005 (ISO 22866 2005) or with a similar standard (i.e. following a part but not all requirements of ISO 22866 when necessary). Indeed, it is important to note that ISO 22866 is well adapted for boom sprayer application methods, but for applications such as the aerial ones, or knapsack sprayer, or even air blast sprayer on tree production such as on papaya in Hernández-Hernández et al. 2007, ISO 22866 might not be fully adapted as notified in Gil et al. 2018, and procedure from ASABE (American Society of Agricultural and Biological Engineers) has to be followed, as in Baio et al. 2019 for the aerial application (ASABE 2009) and by Gouda et al. 2018 for knapsack application method. Details of experimental protocol of the drift deposition curves selected are presented in SM, Table S4.4.
- (iii) The drift deposition curve had to be applicable to open-field production (i.e. excluding greenhouse production systems, which have different drift deposition conditions and profiles).

Among the 8 identified studies, several application methods were tested to derive drift deposition functions, namely aerial application, hand-operated application, air blast sprayer and boom sprayer, with a diversity of operational techniques (e.g. atomizer, flat fan, nozzle types) and crop stages. Table 12 presents the identified studies, application methods, crops, experimental conditions and compliance to ISO norm. All drift deposition curves were developed under tropical savanna climate and mostly in South America. The identified drift deposition curves $f(x)$ allow to estimate deposited pesticide fractions to off-field surfaces (f_{dep}). The fractions emitted to air (f_{air}), corresponding to airborne emissions associated to a crop-application method pair, were not available in the corresponding studies. To derive fractions emitted to air for the considered scenarios, generic fractions available in PestLCI Consensus were adopted. These drift deposition curves $f(x)$ developed for crops grown in tropical conditions were compared to similar application methods and associated drift deposition curves defined in temperate conditions. These drift deposition curves are ultimately expressed as a drift deposition fraction, as a function of the x distance from the field edge and are usually calculated as $f(x) = \alpha x^\beta$ (Ganzelmeier and Rautmann 2000) or $f(x) = \alpha_0 e^{-x\alpha_1} + \beta_0 e^{-x\beta_1}$ (Holterman and van de Zande 2003; García-Santos et al. 2016)), with α_x and β_x as coefficients according to the application method and the crop (see details of the drift deposition curves in SM Table S4.3).

Table 12: Identified studies and main characteristics of their drift deposition experiments

Reference	Location	Climate	Application method	Crop	Wind speed	Temperature (°C)	Relative humidity	Compliance to ISO 22866	Comparisons
Gouda et al. 2018	Benin	Tropical savanna climate (Aw)	Ultra-low volume centrifugal cane and knapsack sprayer	Cotton (<i>Gossypium hirsutum</i>)	3.0±0.4 m.s ⁻¹	35±1 °C	64±4%	No	Knapsack-sprayer on potatoes (García-Santos et al. 2016)
Hernández-Hernández et al. 2007	Mexico	Tropical savanna climate (Aw)	Air blast sprayer	Papaya (<i>Carica papaya</i> L.)	1.6 m.s ⁻¹	35.7±0.8 °C	53.8±7.8%	No	Air blast sprayer on perennial crops
Alves and da Cunha 2014	Brazil	Tropical savanna climate (Aw)	Air blast sprayer	Coffee (<i>Coffea arabica</i> L.)	2.9±1.5 m.s ⁻¹	21.3±2.9 °C	69.5±14.1%	Yes	(Holterman and van de Zande 2003) and on olive trees (Gil et al. 2018)
Bueno et al. 2017	Brazil	Tropical savanna climate (Aw)	Boom sprayer	Soybean (<i>Glycine max</i>)	2.9±0.8 m.s ⁻¹	29.8±1.1 °C	46.5±5.2%	Yes	Boom sprayer (van de Zande et al. 2015)
Bueno et al. 2016	Brazil	Tropical savanna climate (Aw)	Boom sprayer	Bean (<i>Phaseolus vulgaris</i> L.)	3.3±1.4 m.s ⁻¹	26.8±3.4 °C	56.8±14.3%	Yes	
da Cunha et al. 2017a	Brazil	Tropical savanna climate (Aw)	Aerial	Soybean (<i>Glycine max</i>)	From 1.4 to 1.9 m.s ⁻¹	From 23.9 to 25.3 °C	From 74 to 81%	No	n/a
da Cunha et al. 2017b	Brazil	Tropical savanna climate (Aw)	Aerial	Soybean (<i>Glycine max</i>)	From 1.7 to 2.5 m.s ⁻¹	From 26.7 to 29.1 °C	From 65 to 71%	No	
Baio et al. 2019	Brazil	Tropical savanna climate (Aw)	Aerial	Sorghum (<i>Sorghum bicolor</i>), Millet (<i>Pennisetum glaucum</i>), Soybean (<i>Glycine max</i>), corn (<i>Zea mays</i>), Cotton (<i>Gossypium hirsutum</i>)	Mean 1.4 m.s ⁻¹	Mean 25.6 °C	Mean 67.4%	No	

4.3 Results

4.3.1 Pesticide foliar interception fractions for crops grown in tropical conditions

Table 13 presents the foliar interception fractions by crop or crop class according to the growth stage and related LAI, and the spraying technique. Since pesticides are applied as formulations, initial distribution fractions and leaf interception are not influenced by properties of the active ingredients, but only by formulation, type application method and crop growth stage.

For the soil application, the foliar interception fraction by the crop is zero for all crop types and growth stages. For application under the canopy, with a hand-operated sprayer, where the pesticide is not applied on the main crop, a small emission fractions reaching the crop was estimated to be always less than 5% of the total pesticide mass applied, with a median of 4% considering all crops and growth stages. Applications over the canopy by aerial applications, boom sprayers or air blast sprayers are fully dependent on the LAI. When the fraction of field area covered by crop canopy ($f r_{\text{field-covered}}$) is equal to 1, the crop leaves can intercept all the pesticide mass reaching the field, which is always the case during the reproductive phase (2) of the crop and sometimes also during the development phase (1), e.g. sugar cane and pineapple. Semi-perennial crops (i.e. banana, sugar cane, pineapple, cotton) and perennial crops (i.e. coffee) are grown for several years and therefore are most of the time at the crop growth stage (2), which can be defined as the default growth stage in case of lack of information on the growth stage of those crops.

Table 13: Foliar interception fraction according to pre-defined, classified crop growth stages (0: installation; 1: development; 2: reproductive phase) and spraying technique (application over the canopy, under the canopy application and plant-by-plant application)

Crop / crop class	Growth stage	LAI	Foliar interception fraction according to the spraying technique		
			Application over the canopy	Application under the canopy	Plant-by-plant application
Banana	0	0.2	0.14	0.01	0.20
	1	1.9	0.82	0.04	0.95
	2	3.8	1	0.05	0.95
Bean	0	0.3	0.20	0.01	0.30
	1	1.9	0.82	0.04	0.95
	2	4	1	0.05	0.95
Coffee	0	0.4	0.27	0.01	0.43
	1	1.6	0.73	0.04	0.95
	2	3.9	1	0.05	0.95
Cotton	1	1.6	0.75	0.04	0.95
	2	3.3	1	0.05	0.95
Pineapple	0	0.4	0.26	0.01	0.40
	1	4.3	1	0.05	0.95
	2	8.6	1	0.05	0.95
Soybean	0	0.3	0.20	0.01	0.30
	1	2.0	0.83	0.04	0.95
	2	4.1	1	0.05	0.95
Sugar cane	0	0.3	0.20	0.01	0.30
	1	3	1	0.05	0.95
	2	6	1	0.05	0.95
Tropical fruit trees	0	0.5	0.32	0.02	0.50
	1	5	1	0.05	0.95
	2	10	1	0.05	0.95
Tubers	0	0.1	0.07	0.004	0.10
	1	1.5	0.72	0.04	0.95
	2	3	1	0.05	0.95
Vegetable fruits	0	0.2	0.14	0.01	0.20
	1	2	0.84	0.04	0.95
	2	4	1	0.05	0.95

4.3.2 Drift deposition curves for pesticide applications on crops grown in tropical conditions

Figure 18 presents the drift deposition curve results for crop–application method pairs in tropical conditions, for (a) the aerial applications, (b) hand-operated applications, (c) air blast applications and (d) boom sprayer applications. The deposition dose fraction (kg deposited/ha deposition area per kg

applied/kg treated area) per distance from the field edge (m) is represented and compared to crop – application method pairs defined in temperate conditions (represented by black lines). The drift deposition curves using the 90th percentile of observed data are drawn with dotted lines and the drift deposition curves using means or medians of observed data are drawn with solid lines. This is to highlight that the latter are preferred for the comparative purpose of LCA. The drift deposition curves defined with the 90th percentile represent the upper limit of the drift emission and are generally not used for LCA purposes as they are difficult to compare and do not respect the overall pesticide emission mass balance. However, since drift deposition fractions usually are small compared to fractions reaching the field, a small overestimation of the former is acceptable in cases where no median deposition results are available. Details on each drift deposition curve equation, data used to define the curves (means, medians or 90th percentile of observed data), the coefficient of determination (R^2), and the lower and upper validity limits according to the experimentation and application method are presented in Table 14. Emission fractions to air (f_{air}) associated to an application method and a crop are details in SM table S4.5. The drift deposition curves were mostly defined according to two curve models, from the German (Ganzelmeier et al. 1995) and the Dutch (Holterman and van de Zande 2003) experiments (van de Zande et al. 2015) (see details of the drift deposition curves in SM table S4.3). The experimental protocol of the drift deposition curves selected is presented in SM table S4.4.

Aerial applications

The drift deposition curves for aerial application in tropical conditions were developed with means (i.e. central estimates) of observed data according to the German fitting model. For the drift deposition curves developed on soybean only (da Cunha et al. 2017a, b), the lower and upper fitting curve validity limits are respectively 20 m and 320 m from the field edge, whereas for the drift deposition curve developed based on experiments for diversity of crop (Baio et al. 2019), the curve is valid between 10 m and 2000 m from the field edge. Total drift deposition fractions reaching off-field surfaces range from 20% to 20.4% (da Cunha et al. 2017a, b) as function of nozzle type on soybean, and 10.3% (Baio et al. 2019) on a set of crops (cotton, millet, sorghum, soybean and corn) (Table 15). The difference between the two studies might be related to the height above crop canopy when spraying at, respectively, 3 m and 5 m. In the absence of drift deposition curves for aerial application in temperate conditions, those drift deposition curves developed for tropical conditions might be used carefully for crops grown in temperate conditions according to differences in crop and application method characteristics. Aerial applications with rotary atomizer, electrostatic system and atomizer at 55° angle were leading the lowest drift deposition in comparison of other aerial applications.

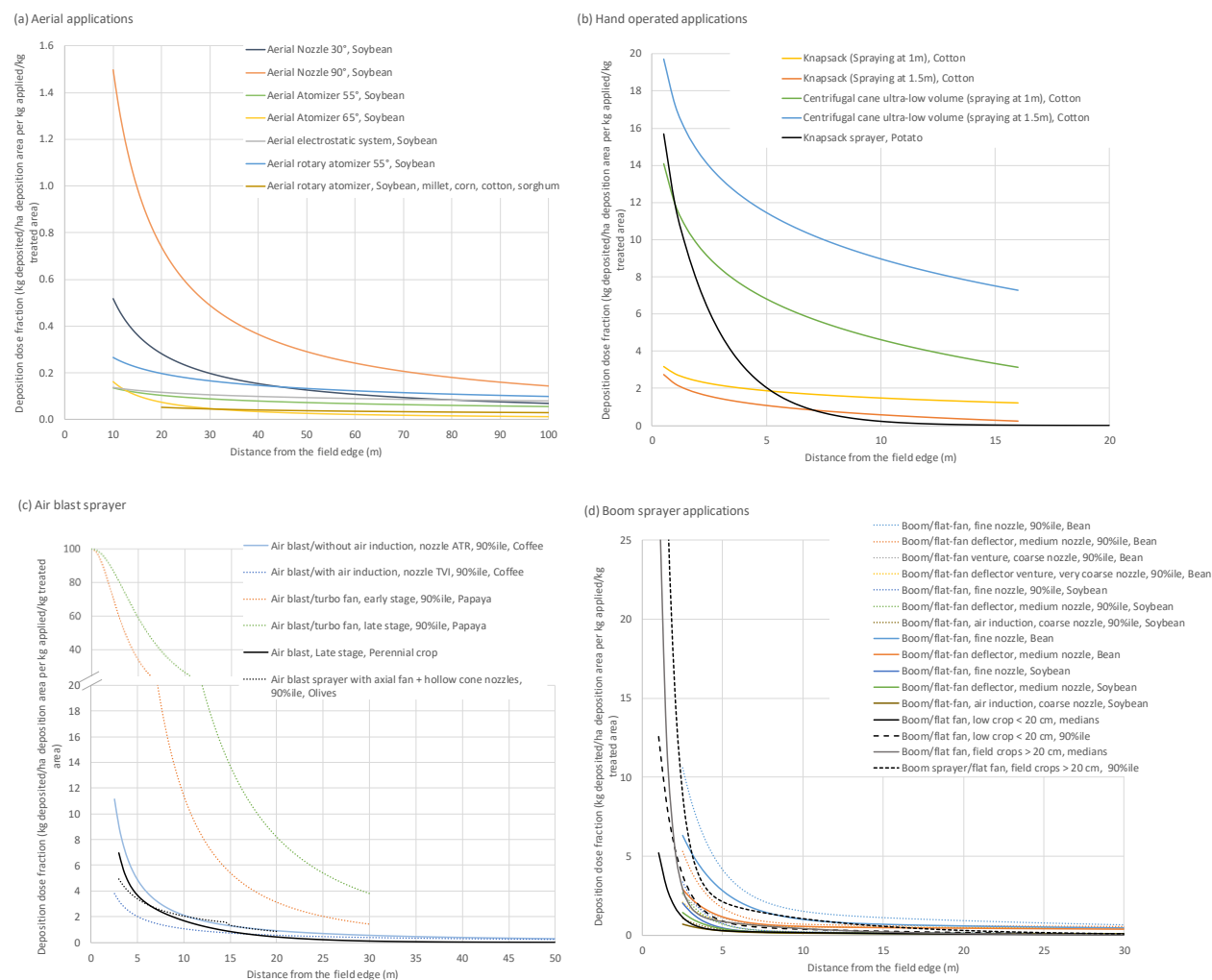


Figure 18: Drift deposition curves for aerial applications (a), hand-operated applications (b), air blast applications (c) and boom sprayer applications (d) defined in tropical regions representing the deposition dose fraction (kg deposited/ha deposition area per kg applied/kg treated area) per distance from the field edge (m), compared to crop – application method pairs defined in temperate conditions in black lines, with the drift deposition curves using the 90th percentile of observed data in dotted lines and the drift deposition curves using means or medians of observed data in solid lines.

Hand-operated applications

Drift deposition curves for hand-operated applications in tropical conditions were defined with means of observed data with an equation type $f(x) = \alpha \ln(x) + \beta$. The drift deposition fractions were twice as high with the knapsack sprayer compared to the ultra-low volume centrifugal cane sprayer. Until 5 m from the field edge, the drift deposition fractions were higher with the knapsack sprayer for potatoes in temperate conditions (García-Santos et al. 2016) (black curve), than those for cotton in tropical conditions (Gouda et al. 2018) for both spraying heights (1 m and 1.5 m). These results might be due to crop height variations between cotton (>1 m) and potatoes (~20 cm), and the corresponding distance between knapsack sprayer nozzles and crop canopy. In fact, a pesticide application with a knapsack sprayer on low height crops may lead to higher drift than on taller crops (Franke et al. 2010), due to a higher distance between nozzles and crop canopy (van de Zande et al. 2008). In the experiment on cotton in Benin, the less high the crop, the higher the drift (Gouda et al. 2018). Furthermore, Gouda et al. (2018) stated that beyond a certain height, reaching the crop canopy with a knapsack sprayer is more difficult, leading to more drift, in particular with the ultra-low volume application method. Applying pesticides with a knapsack sprayer results in a total drift deposition fraction to off-field surfaces of less than 1% on potatoes in temperate conditions (García-Santos et al. 2016), and even less on cotton in Benin (Gouda et al. 2018). The ultra-low volume application method could not be compared with those defined in temperate conditions, since this application method is exclusively used in tropical conditions for crops such as cotton and rice. Until 15 m from the field edge, the drift deposition fractions were between 3.3% and 7.5% for the ultra-low volume centrifugal cane drift deposition curves. In contrast to the knapsack sprayer, for the ultra-low volume centrifugal cane, the taller the crop, the higher the drift, with a drift deposition fraction to off-field surfaces of 1.4% at 1 m and 2.1% at 1.5 m. The ultra-low volume application method induces more drift than the knapsack sprayer, which can partly be explained by the smaller droplets generated with an ultra-low volume application (Gouda et al. 2018).

Air blast applications

Drift deposition for air blast sprayer application in tropical conditions was estimated with means and 90th percentile of observed data according to the German fitting model for coffee in Brazil (Alves and da Cunha 2014), and with 90th percentile observed data with equation type $f(x) = 1/(\alpha + \beta x^2)$ for papaya in Mexico (Hernández-Hernández et al. 2007). These drift deposition curves were compared with: “Air blast – perennial crop – late stage (in leaf)” (Holterman and van de Zande 2003) and “air blast sprayer with axial fan + hollow cone nozzles, 90thile, Olives” (Gil et al. 2018), with respectively a total drift deposition fraction to off-field surfaces of 3.4% and 3.3%. The drift deposition fractions from the two drift deposition curves defined for coffee production (using mean data) are lower than the selected drift deposition curves defined in temperate conditions. The total drift deposition fraction to off-field surfaces is slightly higher when using nozzles without air induction as compared to using air induction nozzles, with respectively 2.9% and 2.7%. Indeed, air induction nozzles produce larger droplets less

prone to drift, than nozzles without air inductions; consequently, the risk of drift is lower with air induction nozzle, as already discussed by Combella et al. (1996) and Grella et al. (2019).

In the papaya case study, no median deposition results were available; using the 90th percentile lead to a small overestimation of drift deposition fractions. From those drift deposition curves, the total drift deposition fractions to off-field surfaces were higher than results from other drift deposition curves developed in temperate and tropical conditions, with 5.2% at early stage and 8.2% at late stage. Differences between early and late stages are mainly due to crop height and canopy interception capacity.

Boom sprayer applications

For boom sprayer application, the drift deposition curves defined in tropical conditions were estimated with means and 90th percentiles of observed data according to the Dutch fitting model applied on bean and soybean production in Brazil (Bueno et al. 2016, 2017). These drift deposition curves were compared with four curves of boom sprayer: “Boom sprayer - standard flat fan – low crop < 20 cm” – using medians or 90th percentile observed data and “boom sprayer - standard flat fan - field crops that are > 20 cm” – using medians or 90th percentile observed data from van de Zande et al. 2015; with respectively a total drift deposition to off-field surfaces of 1.1% and 1.2% for low crop < 20 cm, and 1.3% and 1.6% for crops > 20 cm. Drift deposition fractions to off-field surfaces were slightly higher on bean production compared to soybean production, regardless of the type of boom sprayer, with respectively "2.7-2.9%" and "2.5-2.6%". Drift deposition fractions from boom sprayer in tropical conditions (bean and soybean) were around twice as high as in temperate conditions, regardless of the type of nozzle. Nevertheless, we observed a slight overestimation of drift deposition fractions using the 90th percentiles of observed data; this emphasizes that median observed data should be in priority used for comparative assessments, such as LCA.

In conclusion, drift deposition fractions close to the field edge are generally higher with non-aerial applications than with aerial ones. Conversely, drift deposition fractions are higher with aerial applications with off-field emissions far from the field edge measured up to 2000 m (Baio et al. 2019), while drift deposition fractions are measured up to 20 m with hand-operated applications (García-Santos et al. 2016). The drift deposition fractions to off-field surfaces vary from less than 1% with a knapsack sprayer on cotton up to 20% with aerial application on soybean, with intermediate values for air blast sprayers around 3% on coffee and 5-8% on papaya, and less than 3% on bean and soybean with a boom sprayer. Overall, drift deposition fractions to off-field surfaces are generally higher in temperate conditions compared to that in tropical ones, except with the boom sprayer. However, due to different crop-application method pairs in these different regions, a more systematic comparisons of drift deposition curves developed for tropical conditions with those developed for temperate conditions is required to arrive at justifiable conclusions.

Table 14: Drift deposition functions proposed to quantify drift-related pesticide emissions in LCA with their lower and upper validity limits.

Application method	Equation	Data used	R ²	Lower validity limit [m]	Upper validity limit [m]	Reference
Air blast sprayer - without air induction - nozzle ATR	$f(x)= 33.4101x^{(-1.1958)}$	90 th percentile	0.99	2.5	50	Alves et al 2014, da Cunha et al 2015
Air blast sprayer - air induction - nozzle TVI	$f(x)= 8.7734x^{(-0.9129)}$	90 th percentile	0.81	2.5	50	Alves et al 2014, da Cunha et al 2015
Air blast sprayer - Turbo fan driven sprayer	$f(x)=1/(0.01+7.82E-04x^2)$	90 th percentile	0.99	0	30	Hernández-Hernández et al 2007
Air blast sprayer - Turbo fan driven sprayer	$f(x)=1/(0.01+2.80E-04x^2)$	90 th percentile	0.95	0	30	Hernández-Hernández et al 2007
Boom sprayer - flat-fan deflector standard - nozzle TT (medium)	$f(x)= 20.2565e^{(-0.5923x)} + 0.6914e^{(-0.0077x)}$	90 th percentile	0.99	2.5	50	Bueno et al 2016
Boom sprayer - flat-fan standard - nozzle XR (fine)	$f(x)= 30.7647e^{(-0.4949x)} + 1.8279e^{(-0.0347x)}$	90 th percentile	0.99	2.5	50	Bueno et al 2016
Boom sprayer - flat-fan venture - nozzle AIXR (coarse)	$f(x)= 7.5039e^{(-0.5222x)} + 0.5485e^{(-0.0046x)}$	90 th percentile	0.99	2.5	50	Bueno et al 2016
Boom sprayer - flat-fan deflector venture - nozzle TTI (very coarse)	$f(x)= 6.77e^{(-0.5943x)} + 0.5482e^{(-0.0047x)}$	90 th percentile	0.99	2.5	50	Bueno et al 2016
Boom sprayer - flat-fan deflector standard - nozzle TT (medium)	$f(x)= 12.5031e^{(-0.6532x)} + 0.2362e^{(-0.0353x)}$	90 th percentile	0.90	2.5	50	Bueno et al 2017
Boom sprayer - flat-fan standard - nozzle XR (fine)	$f(x)= 19.276e^{(-0.7277x)} + 0.2732e^{(-0.0353x)}$	90 th percentile	0.99	2.5	50	Bueno et al 2017
Boom sprayer - flat-fan air-induction - nozzle AXIR (coarse)	$f(x)= 4.2753e^{(-0.6477x)} + 0.2092e^{(-0.0305x)}$	90 th percentile	0.99	2.5	50	Bueno et al 2017
Air blast sprayer – without air induction - nozzle ATR	$f(x)= 15.2104x^{(-1.0759)}$	Means	0.98	2.5	50	Alves et al 2014, da Cunha et al 2015
Air blast sprayer - without air induction - nozzle TVI	$f(x)= 5.0738x^{(-0.8128)}$	Means	0.82	2.5	50	Alves et al 2014, da Cunha et al 2015
Knapsack sprayer (Spraying at 1m)	$f(x)= -0.562\ln(x) + 2.7812$	Means	0.74	0.5	16	Gouda et al 2018
Knapsack sprayer (Spraying at 1.5m)	$f(x)= -0.723\ln(x) + 2.233$	Means	0.83	0.5	16	Gouda et al 2018

Application method	Equation	Data used	R²	Lower validity limit [m]	Upper validity limit [m]	Reference
Centrifugal cane ultra-low volume sprayer (spraying at 1m)	$f(x) = -3.169 \ln(x) + 11.901$	Means	0.76	0.5	16	Gouda et al 2018
Centrifugal cane ultra-low volume sprayer (spraying at 1.5m)	$f(x) = -3.591 \ln(x) + 17.242$	Means	0.60	0.5	16	Gouda et al 2018
Boom sprayer - flat-fan deflector standard - nozzle TT (medium)	$f(x) = 9.1207e^{(-0.5426x)} + 0.5741e^{(-0.0161x)}$	Means	0.99	2.5	50	Bueno et al 2016
Boom sprayer - flat-fan standard - nozzle XR (fine)	$f(x) = 14.9906e^{(-0.4052x)} + 0.9266e^{(-0.0252x)}$	Means	0.99	2.5	50	Bueno et al 2016
Boom sprayer - flat-fan deflector standard - nozzle TT (medium)	$f(x) = 6.3761e^{(-0.6381x)} + 0.1435e^{(-0.0284x)}$	Means	0.99	2.5	50	Bueno et al 2017
Boom sprayer - flat-fan standard - nozzle XR (fine)	$f(x) = 11.6064e^{(-0.7251x)} + 0.1559e^{(-0.028x)}$	Means	0.99	2.5	50	Bueno et al 2017
Boom sprayer - flat-fan air-induction - nozzle AXIR (coarse)	$f(x) = 2.1988e^{(-0.5481x)} + 0.1297e^{(-0.0284x)}$	Means	0.99	2.5	50	Bueno et al 2017
Aerial Nozzle 90°	$f(x) = 15.647x^{(-1.019)}$	Means	0.99	20	320	da Cunha et al 2017a
Aerial Nozzle 30°	$f(x) = 3.897x^{(-0.877)}$	Means	0.97	20	320	da Cunha et al 2017a
Aerial Atomizer 65°	$f(x) = 2.205x^{(-1.135)}$	Means	0.94	20	320	da Cunha et al 2017a
Aerial Atomizer 55°	$f(x) = 0.331x^{(-0.389)}$	Means	0.83	20	320	da Cunha et al 2017a
Aerial electrostatic system	$f(x) = 0.237x^{(-0.238)}$	Means	0.87	20	320	da Cunha et al 2017b
Aerial rotary atomizer 55°	$f(x) = 0.716x^{(-0.431)}$	Means	0.94	20	320	da Cunha et al 2017b
Aerial rotary atomizer	$f(x) = 0.1532x^{(-0.361)}$	Means	0.72	10	2000	Baio et al 2019

4.3.3 Pesticide emission fractions for crops grown in tropical conditions

Table 15 presents the set of pesticide emission fractions for the considered application methods, spraying technique and crops grown in tropical conditions (according to the previously selected studies) with the emitted fraction to air (f_{air}), to off-field surfaces (f_{dep}), to field crop ($f_{\text{field} \rightarrow \text{crop}}$) and to field soil ($f_{\text{field} \rightarrow \text{soil}}$). The ranges of values are presented for each application method, highlighting the variations related to the specific characteristics of the application methods (e.g. nozzle type).

The fractions emitted to air (f_{air}) do not vary across the crops and their growth stages, since they were estimated from the existing airborne emission fractions in PestLCI Consensus.

Within a given application method, drift deposition fractions (f_{dep}) do not differ across crops, with slight variations only, which are mainly due to differences in e.g. tested nozzle types. Drift is influenced by the nozzle type and its dimension (van de Zande et al. 2012). In particular, the coarser the size of the droplets, the less the drift to off-field surfaces (e.g. boom sprayer application on soybean and bean (Bueno et al. 2016, 2017)). Indeed, coarser droplets are heavier and less prone to drift compared to fine droplets (van de Zande et al. 2012; Chen et al. 2020). Nevertheless, fine droplets allow a better coverage of leaves with pesticides, which is important for contact pesticides such as fungicides (Franke et al. 2010). For an aerial application on soybean, the drift was reduced using rotary atomizers (da Cunha et al. 2017a) and hydraulic nozzles associated with the electrostatic system (da Cunha et al. 2017b). For the air blast application on coffee, the drift was reduced with air-induction nozzles (Alves and da Cunha 2014). The drift is higher with the ultra-low volume centrifugal than with the knapsack sprayer in the cotton production (Gouda et al. 2018), due to smaller droplets prone to drift. The fraction reaching the crop leaves ($f_{\text{field} \rightarrow \text{crop}}$) is directly dependent on the foliar interception fraction ($f_{\text{intercept,crop}}$), which is dependent on the growth stage (LAI) and the spraying technique. For the pesticide applications where the pesticide has to reach the crop itself, i.e. the applications over the canopy with the aerial application, boom sprayer and air blast sprayer and the plant-by-plant application with a hand-operated sprayer, the higher the foliar interception fraction, the higher the fraction emitted to the crop ($f_{\text{field} \rightarrow \text{crop}}$) and the lower the fraction emitted to field soil ($f_{\text{field} \rightarrow \text{soil}}$). When $f_{\text{intercept,crop}} = 1$, the fraction emitted to field soil is zero. For the applications under the crop canopy, the fraction emitted to the crop is less than 5% regardless of the crop growth stage. For the soil application, the pesticides are assumed to be 100% incorporated to soil, and with that the initial distribution is assumed to be $f_{\text{field} \rightarrow \text{soil}} = 1$, hence $f_{\text{air}} = f_{\text{dep}} = f_{\text{field} \rightarrow \text{crop}} = 0$.

Table 15: Set of pesticide emission fractions for the application methods (according to the previous selected studies) with the emitted fractions to air (f_{air}), off-field surfaces (f_{dep}), field crop ($f_{\text{field} \rightarrow \text{crop}}$) and field soil ($f_{\text{field} \rightarrow \text{soil}}$) according to crop growth stages (0: installation; 1: development; 2: reproductive phase) and the pesticide spraying technique.

Crop/crop class	Application method	Pesticide spraying technique	Crop stage	f_{air}	f_{dep}	$f_{\text{field} \rightarrow \text{crop}}$	$f_{\text{field} \rightarrow \text{soil}}$
Bean	Boom sprayer	Over the canopy	0	0.10	[0.0273; 0.0288]	[0.1751; 0.1754]	[0.6961; 0.6973]
			1			[0.7142; 0.7154]	[0.1571; 0.1573]
			2			[0.8712; 0.8727]	0
Soybean	Boom sprayer	Over the canopy	0	0.10	[0.0254; 0.0256]	0.1758	[0.6986; 0.6988]
			1			[0.7265; 0.7266]	0.1479
			2			[0.8744; 0.8746]	0
Soybean	Aerial	Over the canopy	0	0.25	[0.1030; 0.2040]	[0.1098; 0.1301]	[0.4362; 0.5170]
			1			[0.4536; 0.5376]	[0.0924; 0.1094]
			2			[0.5460; 0.6470]	0
Cotton	Aerial	Over the canopy	1	0.25	0.1030	0.4821	0.1649
			2			0.6470	0
Cotton	Knapsack, ULV*	Soil application	1; 2	0	0	0	1
Cotton	knapsack	Plant-by-plant application	1; 2	0.06	[0.0063; 0.0077]	[0.8857; 0.8870]	[0.0466; 0.0467]
						ULV*	[0.0142; 0.0212]
Cotton	knapsack	Under the canopy	1	0.06	[0.0063; 0.0077]	[0.0347; 0.0348]	[0.8976; 0.8989]
			ULV*			1	[0.0142; 0.0212]
Cotton	knapsack	Under the canopy	2	0.06	[0.0063; 0.0077]	[0.0466; 0.0467]	[0.8857; 0.8870]
			ULV*			2	[0.0142; 0.0212]
Coffee	Air blast	Over the canopy	0	0.10	[0.0274; 0.0288]	[0.2390; 0.2394]	[0.6322; 0.6332]
			1			[0.6376; 0.6386]	[0.2336; 0.2339]
			2			[0.8712; 0.8726]	0
Papaya	Air blast	Over the canopy	0	0.20	0.0519	0.2362	0.5119
			1; 2	0.08	0.0821	0.8379	0

*ULV: ultra-low volume centrifugal cane

Details of all scenarios are presented in SM table S4.5.

4.4 Discussion

4.4.1 Applicability and limitations of the presented approach

The development of foliar interception and drift deposition fractions for a set of common crops grown in tropical conditions allows to estimate related pesticide emissions for several crops grown in tropical regions. For this purpose, we proposed to define foliar interception fractions not only as a function of crop growth stage (as done in e.g. Linders et al. 2000), but also as a function of spraying technique. This is particularly important for hand-operated applications, which allow to apply pesticides either under crop canopy or plant-by-plant, leading to very different foliar interception fractions. Compared to foliar interception defined by Linders et al. (2000) (Table 16), our approach allows to consider the precision of hand application sprayers when spraying plant-by-plant. When information on the spraying technique is missing, for hand operated applications, herbicide applications can by default be considered as applications under the canopy, while insecticide and fungicide applications can by default be considered as plant-by-plant applications. Nevertheless, by grouping the 10 main growth stages (from the BBCH scale, Meier 2018) of crops into 3 stages, the foliar interception fractions are less specific. When applying pesticides with known growth stage and LAI, more specific foliar interception fractions can be calculated using eq. 4.6 to derive the fraction of field area covered by crop canopy, and by selecting the corresponding spraying technique.

Table 16: Comparison of foliar interception fractions for a plant-by-plant application with a hand application method for vegetable fruits and cotton

Foliar interception fraction – plant-by-plant application		Installation (0)	Development (1)	Reproductive phase (2)
Vegetable fruits	Linders et al 2000	0.25	0.50	0.70
	This study	0.20	0.95	0.95
Cotton	Linders et al 2000	0.25	0.60	0.70
	This study	n/a	0.95	0.95

We introduced 29 drift deposition curves in PestLCI Consensus for crop-application method pairs commonly found in tropical regions, with a set of diverse nozzle types that have generally a marginal influence on the off-field surfaces deposition fractions. However, several crop-application method pairs relevant for tropical regions are currently missing and require further research, such as boom sprayer on pineapple, and more generally application methods on banana, pineapple, sugar cane, vegetables and tubers. Usually, aerial application is less sensitive to crop types than other application methods. This is in line with earlier studies (Baio et al. 2019) that have developed a generic drift deposition curve for aerial application

that is also applicable to other crops (cotton, millet, sorghum, soybean and corn). This drift deposition curve is likely to be also applicable to several tropical crops where aerial applications are frequently used and respective crop-specific drift deposition curves are missing, such as banana. Drift deposition fractions to off-field surfaces with knapsack sprayers are very low, less than 1% in both considered studies, on cotton in tropical conditions (Gouda et al. 2018) and on potato in temperate conditions (García-Santos et al. 2016). Due to the proximity between the sprayer and the crop, the climatic conditions have no substantial influence on drift deposition, as opposed to the height of the crop. Consequently, knapsack sprayer drift deposition curves might be extrapolated to other crops with similar crop height. In most of the considered drift deposition curves, the crop growth stage does not influence drift-related off-field emissions, whereas it may be relevant in particular for fruit tree production (e.g. on papaya in Hernández-Hernández et al. 2007 and for orchards (Ganzelmeier and Rautmann 2000; Holterman and van de Zande 2003; Holterman et al. 2018). Our drift deposition estimates are mainly limited by the scope of available experimental drift functions. Hence, drift fractions that were extrapolated across crops will come with higher uncertainty. Depending on the application method and crop height, drift deposition curves might be carefully extrapolated, particularly for hand application methods and boom sprayers. All considered drift deposition curves are defined for specific experimental conditions, considering upper and lower validity limits of the fitted curve.

In pesticide emission modeling in LCA, drift experiment results are used to estimate pesticide drift to off-field surfaces, nevertheless they are not based on a mass balance approach. Indeed, the drift experiments are not designed for a specific field size, but rather for a single sufficiently large field area to estimate drift without further increase of drift off-field surfaces. Further experiments are required to validate that the default square field of one hectare well represents pesticide drift deposition to off-field surfaces using drift experiments results in a mass balance approach.

Overall, the new foliar interception fractions for crops grown in tropical conditions and related drift deposition curves allow to improve the estimation of pesticide emissions and related impacts in LCA studies with a focus on tropical regions.

4.4.2 Providing emission fractions for a set of crops grown in tropical conditions

We provided a set of emission fractions for crops grown in tropical conditions, considering the respective foliar interception fractions and application methods to identify the related drift deposition curves. A diversity of specific application scenarios (e.g. different nozzles types) was proposed for use where such information is available and relevant. Our emission fractions are directly available for LCA practitioners with little access to data on phytosanitary practices, without using over-simplifying assumptions, such as all of the applied pesticide mass being emitted to field soil.

Nevertheless, emission fractions for pesticide applications with air blast sprayer must be carefully extended to other crop productions. Indeed, the proposed approach is valid for fruit trees with a continuous canopy, which is often not the case in reality. Orchards are generally treated by application methods such as air blast sprayers, requiring to pass between the rows of the crops (i.e. inter-row) to reach vertical and horizontal leaves. Foliar interception fractions for applications with air blast sprayer on orchards should therefore be improved to consider this specificity of air blast application of orchards, as well as standard protocol (ISO 22866:2005) to be more adapted to orchards specificities (Grella et al. 2017b). For an inter-row application with air blast sprayers, the foliar interception fraction of insecticides or fungicides on orchard fruits and leaves is highly variable according to the application method, the LAI, the main row width (i.e. tractor path), the adjacent row width, the space between trees in the same row, etc. (Soheilifard et al. 2020), and cannot be defined using only the LAI. Furthermore, ground deposit inside the field (tractor path and adjacent row) is also highly variable and can reach up to 25% in an apple orchard (van de Zande et al. 2008), 55% in a citrus orchard (Soheilifard et al. 2020) and almost 80% in a papaya plantation (Hernández-Hernández et al. 2007). In direct relation to foliar interception fractions, pesticide fractions reaching field soil must be detailed. Furthermore, airborne emissions are highly variable depending on the crop-air blast sprayer method, with emission fractions to air on citrus of 6 to 14% (Salyani et al. 2007) and 17 to 23% (Garcerá et al. 2017), and on apple orchards of 8 to 20% (Ganzelmeier and Rautmann 2000). Similar data are needed to refine airborne emission fractions. In general, we recommend for the purpose of LCA to report airborne emission fractions in a way that they can be used in a mass balance approach.

The airborne emission estimations for these identified (Table 12) crop-application method studies/pairs in tropical conditions were not available, which represents a limitation of our proposed approach. Airborne emissions can reach up to 25% with aerial applications, and their estimation and modeling are limited (Gil and Sinfort 2005). Indeed, airborne drift is dependent on the application method and its specific characteristics (e.g. nozzle size), on the spray physical properties and formulation and on climatic conditions (Gil and Sinfort 2005).

Overall, we present for the first time a set of emission fractions as a suitable starting point for consistently deriving emission fractions in LCA under tropical conditions, a very useful data set for LCA practitioners.

4.4.3 Future research needs

Our results highlight the need for additional drift experiments, for example for pneumatic cannon sprayers in tropical crops. Indeed, further experimental studies are required on pesticide drift losses to off-field surfaces. Important crop-application method pairs in tropical conditions are missing (e.g. on banana

production, on vegetable fruits or on orchards). Further research is needed particularly on orchard/ fruit tree production and associated pesticide emission to off-field soil, to field soil and also to define the crop foliar interception fraction. For an application done with a passage between the rows of crops without spraying only the top of the canopy, more experimentations or modeling of pesticide emissions are required in tropical conditions. Generally speaking, further research and experimental studies are necessary to assess properly the initial emission fractions of pesticide application with an air blast sprayer. For the purpose of LCA, these experimentations and drift deposition curve developments should report new drift deposition curves using median observed values, and provide the fitting coefficients and final fitting equation for deposition that is either expressed in kg deposited/m² deposition area per kg applied/m² treated area or in units that can be converted into this unit. During initial distribution, bystander exposure pathway should be added to the inventory (Ryberg et al. 2018), and particularly with hand-operated applications (García-Santos et al. 2016). Furthermore, measurements of airborne emissions in tropical conditions are necessary and more generally in all agricultural contexts.

Finally, drift functions used and recommended in our study should be flexibly updated in line with revised or new drift experiments for both temperate and tropical conditions. This will also help to increase crop coverages and account for regional differences in drift.

4.5 Conclusions

In the present study, we provided a set of pesticide initial distribution fractions to various environmental compartments for a diversity of crops and application methods, based on implementing drift deposition curves and deriving foliar interception fractions for various crops grown under tropical conditions. With that, our approach helps to consistently deriving emission fractions for several crops commonly grown in tropical regions.

Drift deposition depends on the combination of crop and application method, particularly on crop height. Application method characteristics, such as nozzle types, have limited influence on the overall emission mass balance. Emission fractions vary for each combination of crop and application method, and part of combinations differs substantially from emission fractions obtained under temperate conditions. The drift deposition curves selected in this study might be carefully extended to other crops with similar characteristics and height, considering on which crops the drift deposition curves were initially developed. The foliar interception fraction method can be extended to other crops by adding the LAI of the corresponding crop growth stages. With that, the emission fractions developed for a set of crop-application

method pairs are directly available for use by LCA practitioners with little access to data on phytosanitary practices.

The selected drift deposition curves allow to estimate off-field surfaces emissions, while estimating the associated airborne drift fractions requires further research. Furthermore, some relevant crop-application method combinations are still missing and require additional experiments to derive drift deposition estimates, such as boom sprayer on pineapple. Specifically, pesticide application with air blast sprayer are highly dependent on trees' organization in the field and crop growth stages, so crop-air blast application pairs need to be further refined.

Overall, our proposed foliar interception fractions for crops grown in tropical conditions and related drift curves allow to estimate pesticide emissions in support of assessing the environmental performance of agrifood systems in LCA with focus on tropical regions. Based on our initial emission distribution fractions for tropical regions, secondary processes could also be estimated integrating climate, soil and farming practices, to better assess pesticide emission of diverse farming systems under tropical conditions.

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References Chapter 4

- ASABE 2009 S561.1—Procedure for Measuring Drift Deposits from Ground, Orchard, and Aerial Sprayers. St. Joseph, MI: ASABE. pp. 1
- Allen RG, Pereira LS (2009) Estimating crop coefficients from fraction of ground cover and height. *Irrig Sci* 28:17–34. doi.org/10.1007/s00271-009-0182-z
- Alves GS, da Cunha JPAR (2014) Field data and prediction models of pesticide spray drift on coffee crop. *Pesquisa Agropecuária Brasileira* 49:622–629. doi.org/10.1590/S0100-204X2014000800006
- Arias-Estévez M, López-Periago E, Martínez-Carballo E, et al (2008) The mobility and degradation of pesticides in soils and the pollution of groundwater resources. *Agri Ecosyst Environ* 123:247–260. doi.org/10.1016/j.agee.2007.07.011

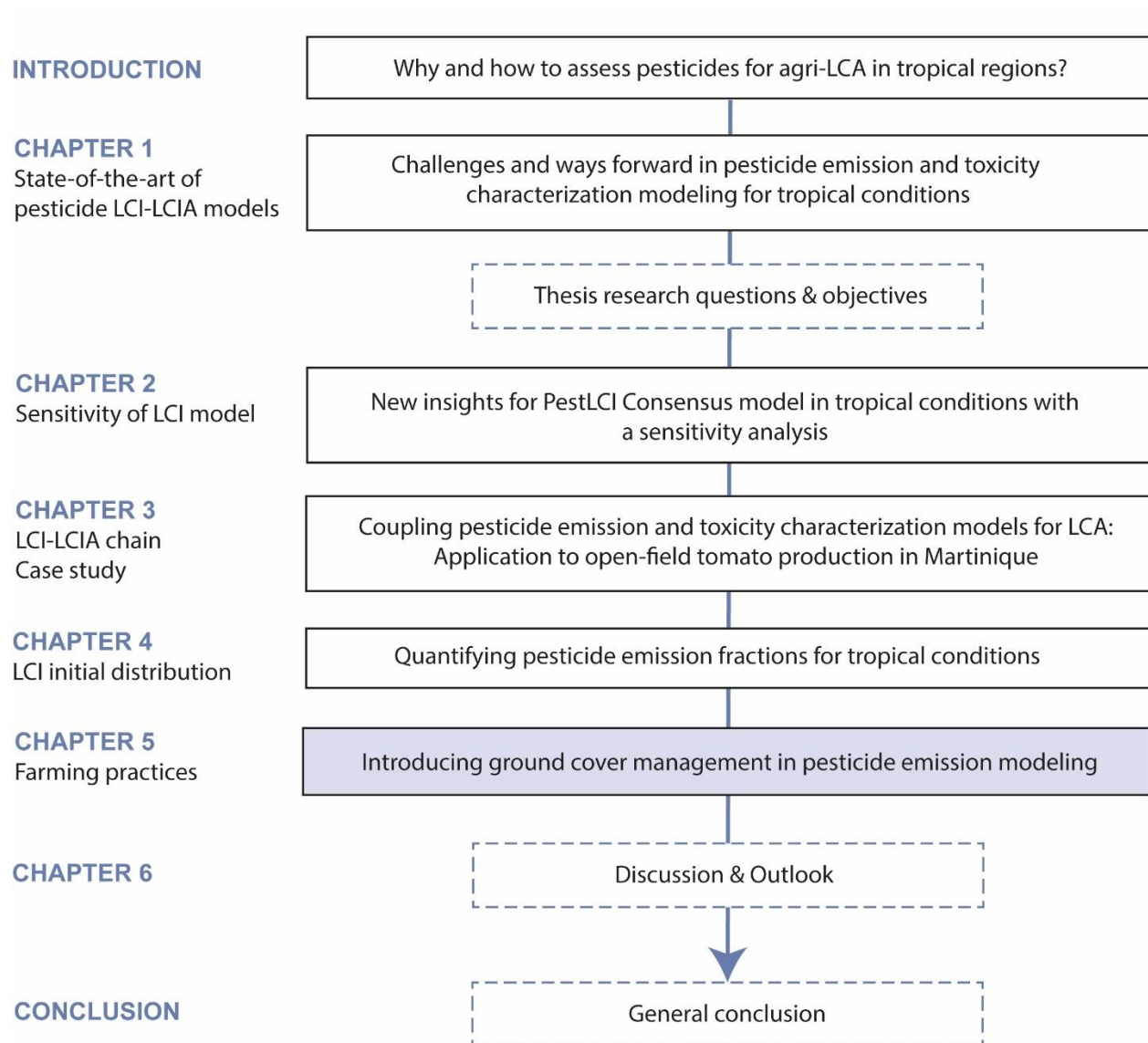
- Baio FHR, Antuniassi UR, Castilho BR, et al (2019) Factors affecting aerial spray drift in the Brazilian Cerrado. *PLOS ONE* 14(2):e0212289. doi.org/10.1371/journal.pone.0212289
- Beck HE, Zimmermann NE, McVicar TR, et al (2018) Present and future Köppen-Geiger climate classification maps at 1-km resolution. *Scientific Data* 5:180214. doi.org/10.1038/sdata.2018.214
- Birkved M, Hauschild MZ (2006) PestLCI—A model for estimating field emissions of pesticides in agricultural LCA. *Ecol Model* 198:433–451. doi.org/10.1016/j.ecolmodel.2006.05.035
- Bueno MR, Cunha JPAR da, Santana DG de, et al (2016) Drift curves from spray applications on common bean crop. *Ciência e Agrotecnologia* 40:621–632. doi.org/10.1590/1413-70542016406016716
- Bueno MR, da Cunha JPAR, de Santana DG (2017) Assessment of spray drift from pesticide applications in soybean crops. *Biosyst Eng* 154:35–45. doi.org/10.1016/j.biosystemseng.2016.10.017
- Cadersa Y, Govinden N (1999) Relationship between canopy cover and light interception in potato in a tropical climate. pp.137-144
- Charlier J-B, Cattan P, Voltz M, Moussa R (2009) Transport of a nematicide in surface and groundwaters in a tropical volcanic catchment. *J Environ Qual* 38:1031–1041. doi.org/10.2134/jeq2008.0355
- Chen S, Lan Y, Zhou Z, et al (2020) Effect of Droplet Size Parameters on Droplet Deposition and Drift of Aerial Spraying by Using Plant Protection UAV. *Agronomy* 10:195. doi.org/10.3390/agronomy10020195
- Combella JH, Westen NM, Richardson RG (1996) A comparison of the drift potential of a novel twin fluid nozzle with conventional low volume flat fan nozzles when using a range of adjuvants. *Crop Protection* 15:147–152. https://doi.org/10.1016/0261-2194(95)00089-5
- Cuhra M (2015) Review of GMO safety assessment studies: glyphosate residues in Roundup Ready crops is an ignored issue. *Environmental Sciences Europe* 27:20. doi.org/10.1186/s12302-015-0052-7
- da Cunha JPAR, Barizon RRM, Ferracini VL, et al (2017a) Spray drift and pest control from aerial applications on soybeans. *Engenharia Agrícola* 37:493–501. doi.org/10.1590/1809-4430-eng.agric.v37n3p493-501/2017
- da Cunha JPAR, Barizon RRM, Ferracini VL, Assalin MR (2017b) Spray drift and caterpillar and stink bug control from applications with electrostatic charge and atomizer on soybean crop. *ENGENHARIA AGRICOLA* 37:1163–1170. doi.org/10.1590/1809-4430-Eng.Agric.v37n6p1163-1170/2017
- Daam MA, van den Brink PJ (2010) Implications of differences between temperate and tropical freshwater ecosystems for the ecological risk assessment of pesticides. *Ecotoxicology* 19:24–37. doi.org/10.1007/s10646-009-0402-6
- de Schampheleire M, Spanoghe P, Steurbaut W, et al (2005) The assessment of spray drift damage for ten major crops in Belgium. *Commun Agric Appl Biol Sci* 70:1037–1042
- Dijkman TJ, Birkved M, Hauschild MZ (2012) PestLCI 2.0: a second generation model for estimating emissions of pesticides from arable land in LCA. *Int J Life Cycle Assess* 17:973–986. doi.org/10.1007/s11367-012-0439-2

- Fantke P, Antón A, Grant T, Hayashi K (2017) Pesticide emission quantification for life cycle assessment: A global consensus building process. *J Life Cycle Assess* 13:245–251
- Fantke P, Juraske R, Antón A, et al (2011) Dynamic multicrop model to characterize impacts of pesticides in food. *Environ Sci Technol* 45:8842–8849. doi.org/10.1021/es201989d
- Fantke P, Wieland P, Juraske R, et al (2012) Parameterization models for pesticide exposure via crop consumption. *Environ Sci Technol* 46:12864–12872. doi.org/10.1021/es301509u
- FAOSTAT (2020) FAOSTAT. In: FAOSTAT. <http://www.fao.org/faostat/en/#home>. Accessed 15 Jan 2019
- Franke AC, Kempenaar C, Holterman HJ, van de Zande JC (2010) Spray drift from knapsack sprayers, A study conducted within the framework of the Sino-Dutch Pesticide Environmental Risk Assessment Project PERAP. Plant Research International B.V., Wageningen
- Ganzelmeier H, Rautmann D (2000) Drift, drift reducing sprayers and sprayer testing. *Aspects of Applied Biology* 1–10
- Ganzelmeier H, Rautmann D, Spangenberg R, et al (1995) Studies on the spray drift of plant protection products. Federal Biological Research Centre for Agriculture and Forestry, Berlin
- Garcerá C, Moltó E, Chueca P (2017) Spray pesticide applications in Mediterranean citrus orchards: Canopy deposition and off-target losses. *Science of The Total Environment* 599–600:1344–1362. doi.org/10.1016/j.scitotenv.2017.05.029
- García-Santos G, Feola G, Nuyttens D, Diaz J (2016) Drift from the use of hand-held knapsack pesticide sprayers in Boyacá (Colombian Andes). *J Agric Food Chem* 64:3990–3998. doi.org/10.1021/acs.jafc.5b03772
- Gentil C, Fantke P, Mottes C, Basset-Mens C (2020) Challenges and ways forward in pesticide emission and toxicity characterization modeling for tropical conditions. *Int J Life Cycle Assess* 25:1290–1306. doi.org/10.1007/s11367-019-01685-9
- Gil E, Llorens J, Gallart M, et al (2018) First attempts to obtain a reference drift curve for traditional olive grove's plantations following ISO 22866. *Science of The Total Environment* 627:349–360. <https://doi.org/10.1016/j.scitotenv.2018.01.229>
- Gil Y, Sinfort C (2005) Emission of pesticides to the air during sprayer application: A bibliographic review. *Atmospheric Environment* 39:5183–5193. doi.org/10.1016/j.atmosenv.2005.05.019
- Gouda AI, Mehoba MHL, Toko II, et al (2018) Comparison of drift of two types of sprayers used in cotton production in Benin. *Biotechnol Agron Soc* 22:94–105. doi.org/10.25518/1780-4507.16431
- Grella M, Gallart M, Marucco P, et al (2017) Ground Deposition and Airborne Spray Drift Assessment in Vineyard and Orchard: The Influence of Environmental Variables and Sprayer Settings. *Sustainability* 9:728. doi.org/10.3390/su9050728
- Grella M, Gil E, Balsari P, et al (2017b) Advances in developing a new test method to assess spray drift potential from air blast sprayers. *Span J Agric Res* 15:e0207. <https://doi.org/10.5424/sjar/2017153-10580>

- Haverkort AJ, Uenk D, Veroude H, Van De Waart M (1991) Relationships between ground cover, intercepted solar radiation, leaf area index and infrared reflectance of potato crops. *Potato Res* 34:113–121. doi.org/10.1007/BF02358105
- Hernández-Hernández CNA, Valle-Mora J, Santiesteban-Hernández A, Bello-Mendoza R (2007) Comparative ecological risks of pesticides used in plantation production of papaya: Application of the SYNOPS indicator. *Science of The Total Environment* 381:112–125. doi.org/10.1016/j.scitotenv.2007.03.014
- Hewitt AJ, Solomon KR, Marshall EJP (2009) Spray droplet size, drift potential, and risks to nontarget organisms from aerially applied glyphosate for coca control in Colombia. *J Toxicol Environ Health Part A* 72:921–929. doi.org/10.1080/15287390902929667
- Holterman HJ, van de Zande JC (2003) IMAG Drift Calculator v1.1: user manual
- Holterman HJ, van de Zande JC, Huijsmans JFM, Wenneker M (2018) Development of a spray drift model for spray applications in fruit orchards. Stichting Wageningen Research, Wageningen Plant Research, Business Unit Agrosystems Research, Wageningen
- ISO 22866 (2005) Equipment for crop protection - Methods for field measurement of spray drift. International Standards Organization, 22866
- Knudsen MT, Dorca-Preda T, Djomo SN, et al (2019) The importance of including soil carbon changes, ecotoxicity and biodiversity impacts in environmental life cycle assessments of organic and conventional milk in Western Europe. *J Clean Prod* 215:433–443. doi.org/10.1016/j.jclepro.2018.12.273
- Kottek M, Grieser J, Beck C, et al (2006) World Map of the Köppen-Geiger climate classification updated. *Meteorol Z* 15:259–263. doi.org/10.1127/0941-2948/2006/0130
- Lammoglia S-K, Moeys J, Barriuso E, et al (2016) Sequential use of the STICS crop model and of the MACRO pesticide fate model to simulate pesticides leaching in cropping systems. *Environ Sci Pollut Res* 24:6895–6909. doi.org/10.1007/s11356-016-6842-7
- Langenbach T, Caldas LQ (2018) Strategies for reducing airborne pesticides under tropical conditions. *Ambio* 47:574–584. doi.org/10.1007/s13280-017-0997-4
- Larbi PA, Salyani M (2012) Model to predict spray deposition in citrus airblast sprayer applications: part 2, spray deposition. *TRANSACTIONS OF THE ASABE* 55:41–48
- Lesueur Jannoyer M, Cattan P, Woignier T, Clostre F (2016) Crisis management of chronic pollution: contaminated soil and human health, 1st Edition. CRC Press
- Lewis SE, Silburn DM, Kookana RS, Shaw M (2016) Pesticide behavior, fate, and effects in the tropics: an overview of the current state of knowledge. *J Agric Food Chem* 64:3917–3924. doi.org/10.1021/acs.jafc.6b01320
- Li S, Huang B, Zhao F, et al (2019) Environmental impact assessment of agricultural production in Chongming ecological island. *Int J Life Cycle Assess* 24:1937–1947. doi.org/10.1007/s11367-019-01614-w

- Linders J, Mensink H, Stephenson G, et al (2000) Foliar interception and retention values after pesticide application: a proposal for standardised values for environmental risk assessment. *Pest Manag Sci* 58:315–315. doi.org/10.1002/ps.448
- Meier MS, Stoessel F, Jungbluth N, et al (2015) Environmental impacts of organic and conventional agricultural products – Are the differences captured by life cycle assessment? *J Environ Manag* 149:193–208. doi.org/10.1016/j.jenvman.2014.10.006
- Meier U (2018) Growth stages of mono- and dicotyledonous plants: BBCH Monograph. Open Agrar Repository
- Mottes C (2013) Evaluation des effets des systèmes de culture sur l'exposition aux pesticides des eaux à l'exutoire d'un bassin versant. Proposition d'une méthodologie d'analyse appliquée au cas de l'horticulture en Martinique. AgroParisTech
- Mottes C, Lesueur Jannoyer M, Le Bail M, et al (2017) Relationships between past and present pesticide applications and pollution at a watershed outlet: The case of a horticultural catchment in Martinique, French West Indies. *Chemosphere* 184:762–773. doi.org/10.1016/j.chemosphere.2017.06.061
- Mottes C, Lesueur-Jannoyer M, Bail ML, Malézieux E (2014) Pesticide transfer models in crop and watershed systems: a review. *Agron Sustain Dev* 34:229–250. doi.org/10.1007/s13593-013-0176-3
- Nemecek T, Schnetzer J (2011) Methods of assessment of direct field emissions for LCIs of agricultural production systems. Data v3.0. Swiss Center for Life Cycle Inventories, Dübendorf, Switzerland
- Nuyttens D, De Schamphelleire M, Steurbaut W, et al (2006a) Experimental study of factors influencing the risk of drift from field sprayers, Part 1: Meteorological conditions. *Aspects of Applied Biology* 77:
- Nuyttens D, De Schamphelleire M, Steurbaut W, et al (2006b) Experimental study of factors influencing the risk of drift from field sprayers Part 2: Spray application technique. *Aspects of Applied Biology* 77:331–339
- Oliquino-Abasolo A (2015) Agro-environmental sustainability of conventional and organic vegetable production systems in Tayabas, Quezon, Philippines. FAO University Library, University of the Philippines at Los Baños:
- Perrin A, Basset-Mens C, Gabrielle B (2014) Life cycle assessment of vegetable products: a review focusing on cropping systems diversity and the estimation of field emissions. *Int J Life Cycle Assess* 19:1247–1263. doi.org/10.1007/s11367-014-0724-3
- Racke KD, Skidmore MW, Hamilton DJ, et al (1997) Pesticides report 38. Pesticide fate in tropical soils - (Technical report). *Pure Appl Chem* 69:1349–1371. doi.org/10.1351/pac199769061349
- Rautmann D, Strelake M, Winkler R (2001) New basic drift values in the authorization procedure for plant protection products. *Mitteilungen aus der Biologischen Bundesanstalt für Land* 388:133–141
- Ryberg MW, Rosenbaum RK, Mosqueron L, Fantke P (2018) Addressing bystander exposure to agricultural pesticides in life cycle impact assessment. *Chemosphere* 197:541–549. doi.org/10.1016/j.chemosphere.2018.01.088
- Salyani M, Farooq M, Sweeb RD (2007) Spray Deposition and Mass Balance in Citrus Orchard Applications

- Soheilifard F, Marzban A, Ghaseminejad Raini M, et al (2020) Chemical footprint of pesticides used in citrus orchards based on canopy deposition and off-target losses. *Science of The Total Environment* 732:139118. doi.org/10.1016/j.scitotenv.2020.139118
- van de Zande JC, Huijsmans JFM, Porskamp HAJ, et al (2008) Spray techniques: how to optimise spray deposition and minimise spray drift. *Environmentalist* 28:9–17. doi.org/10.1007/s10669-007-9036-5
- van de Zande JC, Rautmann D, Holterman HJ, Huijsmans JFM (2015) Joined spray drift curves for boom sprayers in The Netherlands and Germany. *Plant Research International B.V., Wageningen*
- van de Zande JC, Wenneker M, Michielsen JGP, et al (2012) Nozzle classification for drift reduction in orchard spraying. *Aspects of Applied Biology* 114:253–260
- van der Werf HMG, Knudsen MT, Cederberg C (2020) Towards better representation of organic agriculture in life cycle assessment. *Nature Sustainability* 3:419–425. doi.org/10.1038/s41893-020-0489-6
- Wang F, Liu Y, Ouyang X, et al (2018) Comparative environmental impact assessments of green food certified cucumber and conventional cucumber cultivation in China. *Renewable Agriculture and Food Systems* 33:432–442. doi.org/10.1017/S1742170517000229
- Zhang L, Hu Z, Fan J, et al (2014) A meta-analysis of the canopy light extinction coefficient in terrestrial ecosystems. *Front Earth Sci* 8:599–609. doi.org/10.1007/s11707-014-0446-7



CHAPTER 5

INTRODUCING GROUND COVER MANAGEMENT IN PESTICIDE EMISSION MODELING

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Abstract

Ground cover management (GCM) is an important agricultural practice used to reduce weed growth, erosion and runoff, and improve soil fertility. In the present study, an approach to account for GCM is proposed in the modeling of pesticide emissions to evaluate the environmental sustainability of agricultural practices. As starting point, we include a cover crop compartment into the mass balance of calculating initial (within minutes after application) and secondary (including additional processes) pesticide emission fractions. The following parameters were considered: (i) cover crop occupation between the rows of main field crops, (ii) cover crop canopy density, and (iii) cover crop family. Two modalities of cover crop occupation and cover crop canopy density were tested for two crop growth stages, using scenarios without cover crops as control. From that, emission fractions and related ecotoxicity impacts were estimated for

pesticides applied to tomato in Martinique and to grapevine in the Loire Valley (France). Our results show that the presence of a cover crop decreased on average the pesticide emission fraction reaching field soil by a factor of 3 compared to bare soil, independently of field crop and its growth stage, and cover crop occupation and density. When considering cover exported from the field, ecotoxicity impacts were reduced by around 65% and 90% compared to bare soil, respectively for grapevine and tomato, regardless of the emission distribution used. Since additional processes may influence emission distributions under GCM, such as run-off, leaching or preferential flow, further research is required to incorporate these processes consistently in our proposed GCM approach. Considering GCM in pesticide emission modeling highlights the potential of soil cover to reduce pesticide emissions to field soil and related freshwater ecotoxicity. The consideration of GCM as common farming practice furthermore opens the path towards the modeling of pesticide emissions in intercropping systems.

Keywords: Active ingredient, environmental modeling, cover crop, Life Cycle Assessment, farming practices, pest management

5.1 Introduction

A transition towards more agro-ecological cropping systems is urgently needed to ensure sustainable food production systems and consequently sustainable food security, based, for example, on biological pest control, reduced tillage, intercropping, cover crops or agroforestry (Tschamntke et al. 2012; Wezel et al. 2014). This corresponds to the goal number 2 of the Sustainable Development Goals (SDGs) proposed by the United Nations (Griggs et al. 2013; HLPE 2019). One central key practice in agro-ecological cropping systems is ground cover management (GCM). It is defined as the field soil occupation between crop rows and under the crop canopy. GCM can be provided by several farming practices, using either living cover (i.e. spontaneous or planted cover crops) or dead cover (e.g. mulch composed of crop residues and residues from the previous crop, impervious mulch such as plastic mulch) (Mottes et al. 2014).

These ground covers (GC) are used to provide several agricultural and environmental benefits. They limit weeds and consequently herbicide applications, they improve soil fertility and bearing capacity (CIRAD 2009; Wezel et al. 2014). They also reduce erosion (Durán Zuazo and Rodríguez Pleguezuelo 2008) and runoff as well as pesticide (i.e. the active ingredient in a pesticide formulation, hereafter referred to as pesticide) transfer to surface water (Reichenberger et al. 2007; Alletto et al. 2010; Mottes et al. 2014). Among these multiple benefits, the reduction of pesticide applications and transfers is particularly important in tropical conditions, where the use of pesticides occurs all year round, due to high pest pressures and to crop all along the year (Lewis et al. 2016; Mottes et al. 2017b). GC practices exist in all cultivated areas, in

temperate and tropical conditions. For example in 2011, 49% of French vineyards were cover cropped (Ambiaud 2012), with e.g. spontaneous vegetation, oats, clover, fescue (Renaud-Gentié et al. 2015a). In the tropical conditions of Martinique (French West Indies), 23.2% of banana farmers were using cover crops in 2015 (DAAF Martinique 2018), composed of diverse species mixed or separated, e.g. *Brachiaria decumbens*, *Stylosanthes guianensis* (Tixier et al. 2011).

Life Cycle Assessment (LCA), an ISO-standardized methodology to evaluate the environmental performance of product systems, is widely applied to agri-food systems (Andersson 2000; Roy et al. 2009; Poore and Nemecek 2018), to quantify the environmental performance of farming management practices (Bessou et al. 2013; Rouault et al. 2016; Renaud-Gentié et al. 2020). In the context of agro-ecological transition, there is an urgent need to address these practices, such as GCM, in LCA studies to compare different farming systems, such as conventional, integrated and organic farming (Meier et al. 2015) as well as to better inform eco-design (Rouault et al. 2020). As a prerequisite to consider GCM in LCA, pesticide emission distributions need to be properly modeled. However, while many LCA studies do not yet consider pesticide emissions and related-impacts (Perrin et al. 2014), other approaches assume that 100% of pesticides applied are emitted to agricultural soil (e.g. Nemecek and Schnetzer 2011). More detailed approaches exist, such as a generic distribution of pesticides applied between air and soil (e.g. Neto et al. 2013; Oliquino-Abasolo 2015) or modeling pesticides emissions based on a consistent mass balance, as in the PestLCI model (Birkved and Hauschild 2006; Dijkman et al. 2012) and its subsequent PestLCI Consensus version (Fantke et al. 2017a). However, none of these approaches currently considers GCM practices. Renaud-Gentié et al. (2015) developed an initial approach to consider the effect of living GCM for the cover crops between vine rows, by replacing the single crop intercepted fraction by a combination of vine and cover crop interceptions according to their respective development stages. However, this first accounting of a cover crop for grapevine has never been extended to other types of GCM or crops. Therefore, the influence of GCM on the environmental performance of different cropping systems can currently not be properly evaluated. To fill this gap, GCM needs to be integrated into state-of-the-art pesticide emission models.

To extend the modeling of GCM to other types of GC and crops in pesticide emission modeling, the definitions of the cover crop occupation and its canopy development were first proposed. In the present study, GCM composed of spontaneous or planted cover crops without being a cash crop were considered. The fraction intercepted by the cover leaves was estimated and processes occurring on those leaves were simulated (volatilization, degradation and uptake), considering as far as possible the cover family (e.g. *Fabaceae*). Furthermore, pesticide plant uptake modeled by the crop exposure model dynamiCROP (Fantke et al. 2011b), was recently coupled with the initial distribution of PestLCI Consensus (Gentil et al. 2020a).

Consequently, the pesticide intercepted fraction by the crop has been distinguished from that intercepted by the cover and other living plants not harvested in the field (e.g. buffer zone).

The main goal of the present study is to consistently introduce GCM in pesticide emission modeling. To achieve this goal, three specific objectives were defined: (i) to propose an approach for considering GCM in a state-of-the-art pesticide emission model, (ii) to analyze the effect of GCM on distributions of emissions and related freshwater ecotoxicity impacts, and (iii) to test our approach in a case study using different GCM types under temperate and tropical climate conditions.

5.2 Materials and methods

5.2.1 Ground cover management modeling

The ground cover management (GCM) was introduced in PestLCI Consensus, a mass balance model that calculates fractions of pesticide mass reaching different plant-environment compartments (air, crop leaves, off-field surfaces, field soil, groundwater) with two distributions (Birkved and Hauschild 2006; Dijkman et al. 2012). The initial (or primary) distribution of PestLCI considers processes occurring few minutes after pesticide application, with a pesticide distribution to air, off-field surfaces, field soil and field crop. Initial distribution is followed by the secondary emission processes occurring over a default duration of 24 hours (Fantke et al. 2017a).

To integrate GCM in PestLCI Consensus, a new compartment in the model for the cover crop was created. Data required to define the cover crop are: i) cover crop occupation fraction between the rows and the stems of the main crop, below the crop canopy (i.e. area fraction of crop-free field soil that is occupied by cover crop), $f_{\text{soil} \rightarrow \text{cover}}$; ii) cover crop canopy fraction (i.e. area fraction of cover crop that is covered by leaves), $f_{\text{intercept,cover}}$ (Renaud-Gentié et al. 2015a); and iii) cover family (e.g. grass composed of *Pooideae*). The cover crop occupation fraction, $f_{\text{soil} \rightarrow \text{cover}}$, is dependent on the farmer's goals for applying GCM and is also dependent on the crop, its growth and field characteristics (e.g. slope, crop family, soil). The fraction intercepted by the cover, $f_{\text{intercept,cover}}$, is a function of the leaf area index (LAI) defined as the ratio of the leaf surface area (one side) per unit of ground surface area ($\text{m}^2 \text{m}^{-2}$) and it is derived in analogy to the fraction intercepted by field crop leaves ($f_{\text{intercept,crop}}$). In the current PestLCI Consensus model, they were defined based on Linders et al. (2000) using the growth stages with BBCH-scale (Meier 2018).

Initial distribution fractions

As a starting point, few minutes after pesticide application, a fraction of pesticides is deposited to off-field surfaces (f_{dep}). It is derived from drift deposition functions specific to each application method.

Another fraction goes to the air by wind drift (f_{air}) as a default fraction per application method and crop, and a last fraction goes to the field (f_{field}):

$$1 = f_{\text{field}} + f_{\text{air}} + f_{\text{dep}} \quad (5.1)$$

Figure 19 presents the pesticide distribution to the various compartments.

After re-arranging eq. 5.1, the fraction reaching the field surface area (f_{field}) was obtained:

$$f_{\text{field}} = 1 - (f_{\text{air}} + f_{\text{dep}}) = f_{\text{field} \rightarrow \text{crop}} + f_{\text{field} \rightarrow \text{rest}} \quad (5.2)$$

The fraction reaching the field surface area (f_{field}) is partially deposited on crop leaves ($f_{\text{field} \rightarrow \text{crop}}$) according to crop intercepted fraction $f_{\text{intercept,crop}}$ and calculated as:

$$f_{\text{field} \rightarrow \text{crop}} = f_{\text{field}} \times f_{\text{intercept,crop}} \quad (5.3)$$

Then, the fraction left on the field after crop interception ($f_{\text{field} \rightarrow \text{rest}}$) is calculated as:

$$f_{\text{field} \rightarrow \text{rest}} = f_{\text{field}} \times (1 - f_{\text{intercept,crop}}) = f_{\text{field} \rightarrow \text{cover}} + f_{\text{field} \rightarrow \text{soil}} \quad (5.4)$$

The fractions of pesticide reaching the cover leaves ($f_{\text{field} \rightarrow \text{cover}}$) and the field soil ($f_{\text{field} \rightarrow \text{soil}}$) are then calculated according to the effective area fraction of crop-free field that is covered by cover crop ($f_{\text{eff,cover}}$):

$$f_{\text{field} \rightarrow \text{cover}} = f_{\text{field} \rightarrow \text{rest}} \times f_{\text{eff,cover}} \quad (5.5)$$

$$f_{\text{field} \rightarrow \text{soil}} = f_{\text{field} \rightarrow \text{rest}} \times (1 - f_{\text{eff,cover}}) \quad (5.6)$$

Where $f_{\text{eff,cover}}$ is calculated by multiplying the area fraction of crop-free field that is cover crop ($f_{\text{soil} \rightarrow \text{cover}}$) and the area fraction of cover crop that is covered by leaves ($f_{\text{intercept,cover}}$):

$$f_{\text{eff,cover}} = f_{\text{soil} \rightarrow \text{cover}} \times f_{\text{intercept,cover}} \quad (5.7)$$

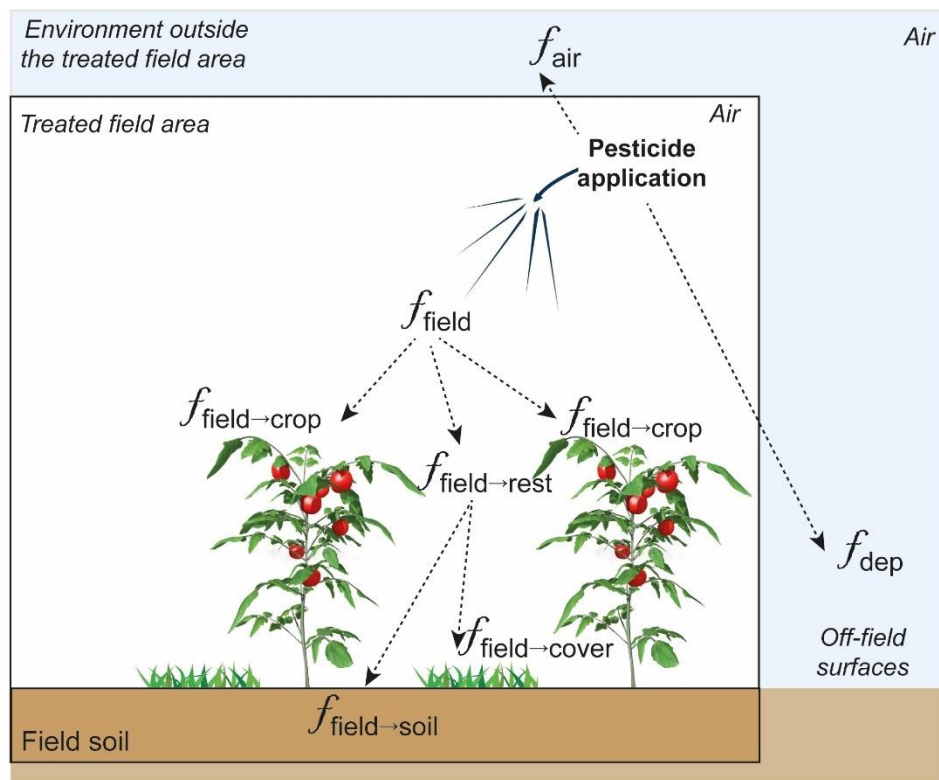


Figure 19: Illustration of initial pesticide distribution fractions to air (f_{air}), off-field surfaces (f_{dep}), field crop leaves ($f_{field \rightarrow crop}$), cover crop leaves ($f_{field \rightarrow cover}$) and field soil surface ($f_{field \rightarrow soil}$)

Secondary emission fractions

From the initial distribution fractions, secondary emissions processes occur. All processes defined in PestLCI for the main crop leaves were also applied to cover crop leaves, i.e. degradation, uptake and volatilization. The fraction degraded and volatilized were aggregated with the respective processes occurring on the main crop, ultimately affecting fractions emitted to air and to field soil. A new emission output fraction, $f_{cover-sec}$, was created to consider the uptake fraction by the cover leaves and the fraction remaining on cover leaves and not yet taken up by the cover leaves.

The degradation on cover crop leaves was integrated in the total degraded fraction of pesticide. To consider the cover family (e.g. *Pooideae*), the degradation rate constant for pesticides on cover leaves ($k_{degCover}$) in (d^{-1}) was calculated. Based on a database of measured dissipation half-lives (Fantke and Juraske 2013), the pesticide dissipation half-life (d) on leaf cover at the average temperature in the month of application ($DT50_{cover,T}$) was used as a proxy. From these data, a dissipation half-life model was derived, using a temperature correction coefficient (Fantke et al. 2014) to correct for actual field temperature as:

$$DT50_{cover,T} = 10^{\log_{10}(DT50_{cover,T_{ref}}) - 0.01995 \times (T - T_{ref})} \quad (5.8)$$

Where $DT50_{cover,T_{ref}}$ the pesticide half-life on cover leaf at reference temperature (20 °C) (d), T the average temperature in the month of application (°C) and T_{ref} the temperature at which the dissipation half-life is reported (20 °C). $DT50_{cover,T_{ref}}$ is selected from a list of coefficients for crops that correspond to one of the relevant cover family (e.g. *Pooideae*).

Then, the degradation rate constant for pesticides on cover leaves at average temperature in the month of application (d^{-1}) was calculated as:

$$k_{degCover} = \frac{\ln(2)}{DT50_{cover,T}} \quad (5.9)$$

Finally, the overall rate constant for the cover leaves k_{OCover} (d^{-1}) was calculated as:

$$k_{OCover} = k_{volat} + k_{uptake} + k_{degCover} \quad (5.10)$$

Where the rate constant for volatilization from the leaf surface, k_{volat} (d^{-1}), and the rate constant for leaf uptake, k_{uptake} (d^{-1}), are both modeled independently of the crop or cover family, and where the degradation rate constant for pesticides on cover leaves, $k_{degCover}$ (d^{-1}), is modeled according to eq. 5.9.

The processes of degradation, volatilization and uptake on cover leaves are calculated until the time defined for the secondary emissions to occur after pesticide application (default: $t_{assess} = 1$ d).

Considering the crop family of the cover, the degradation on cover leaves ($f_{degCover}$) is different from that on the main crop, and was calculated as:

$$f_{degCover} = f_{field \rightarrow cover} \times \frac{k_{degCover}}{k_{OCover}} \times (1 - e^{(-k_{OCover} \times t_{assess})}) \quad (5.11)$$

Where $f_{field \rightarrow cover}$ the fraction reaching cover leaves after pesticide application (eq. 5.5), $k_{degCover}$ the degradation rate constant for pesticides on leaf at average temperature in the month of application (d^{-1}) (eq. 5.9) and k_{OCover} the overall rate constant dissipation on cover leaves (d^{-1}) (eq. 5.10).

The volatilization on cover leaves ($f_{volCover}$) was calculated as:

$$f_{volCover} = f_{field \rightarrow cover} \times \frac{k_{volat}}{k_{OCover}} \times (1 - e^{(-k_{OCover} \times t_{assess})}) \quad (5.12)$$

Where $f_{field \rightarrow cover}$ the fraction reaching cover leaves after pesticide application (eq. 5.5), k_{volat} the rate constant for volatilization from the leaf surface (d^{-1}) and k_{OCover} the overall rate constant dissipation on cover (d^{-1}) (eq. 5.10).

The cover leaves uptake ($f_{uptakeCover}$) was calculated as:

$$f_{\text{uptakeCover}} = f_{\text{field} \rightarrow \text{cover}} \times \frac{k_{\text{uptake}}}{k_{\text{OCover}}} \times (1 - e^{(-k_{\text{OCover}} \times t_{\text{assess}})}) \quad (5.13)$$

Where $f_{\text{field} \rightarrow \text{cover}}$ the fraction reaching cover leaves after pesticide application (eq. 5.5), k_{uptake} the rate constant for leaf uptake (d^{-1}) independent of the crop or cover family and k_{OCover} the overall rate constant dissipation on cover leaves (eq. 5.10).

The fraction remaining on cover leaves ($f_{\text{Leaves-cover}}$) that was not degraded, taken up, or volatilized at $t_{\text{assess}} = 1 \text{ d}$, was calculated as:

$$f_{\text{Leaves-cover}} = f_{\text{field} \rightarrow \text{cover}} \times e^{(-k_{\text{OCover}} \times t_{\text{assess}})} \quad (5.14)$$

Where $f_{\text{field} \rightarrow \text{cover}}$ the fraction reaching cover leaves after pesticide application (eq. 5.5) and k_{OCover} the overall rate constant for removal from cover leaves (d^{-1}) (eq. 5.10).

The total fraction in and on cover leaves ($f_{\text{cover-sec}}$) is calculated by summing the fraction uptake from cover leaves ($f_{\text{uptakeCover}}$) and the fraction remaining on cover leaves ($f_{\text{Leaves-cover}}$), as:

$$f_{\text{cover-sec}} = f_{\text{uptakeCover}} + f_{\text{Leaves-cover}} \quad (5.15)$$

Other processes not affected by the presence of a cover crop (e.g. processes related to crop leaves) are detailed elsewhere (Dijkman et al. 2012).

5.2.2 Propagation of GCM to impact score level

To evaluate how the introduction of GCM influences final LCA results, we calculated impact scores for freshwater ecotoxicity impacts of pesticide emissions with USEtox (Rosenbaum et al. 2008), considering the application of 1 kg of pesticide on a tomato or grapevine field of 1 ha. The impact score (IS in $\text{PAF m}^3 \text{ d/kg}_{\text{applied}}$) was calculated as:

$$IS = \sum_{p,c} (m_{\text{emi},p,c} \times CF_{p,c}) \quad (5.16)$$

where $m_{\text{emi},p,c}$ ($\text{kg}_{\text{emitted}}/\text{kg}_{\text{applied}}$) is the total emitted mass of pesticide p from the crop production into a given environmental compartment c , and $CF_{p,c}$ ($\text{PAF m}^3 \text{ d/kg}_{\text{emitted}}$) is the corresponding characterization factor for freshwater ecotoxicity. Related impact characterization factors for freshwater ecotoxicity of the two considered pesticides (mancozeb and pyriproxyfen) are presented in Supplementary Material (SM), Table S5.2.

As recommended in Fantke et al. (2018) and applied in Gentil et al. (2020), emissions to air were assigned in the impact assessment model USEtox to continental rural air, emissions to field soil were

assigned to continental agricultural soil, and emissions to groundwater were assigned to continental freshwater. Off-field surfaces emissions were assigned to continental agricultural soil, natural soil and freshwater according to the typical area share of each compartment in a given region. Cover and crop uptake fractions and degradation were not linked to the impact assessment, in our case study as they are not relevant for ecotoxicity.

5.2.3 Definition of illustrative case study under different conditions

To evaluate the proposed GC modeling approach, pesticide applications were simulated for grapevine (*Vitis vinifera*) in Loire Valley in France (Figure 20a) and an open-field tomato crop (*Solanum lycopersicum*) in Martinique in French West Indies (Figure 20b). With that, different conditions of crop, climate, soil, application method and cover crop were considered (see Table 17). A default situation was defined for field characteristics and farming practices (i.e. field length and width of 100m, no slope, no drift reduction system on the application method, no drainage system, no irrigation and no tillage). Initial emissions to air (f_{air}) and to off-field surfaces (f_{dep}) were defined according to pesticide drift caused by the selected application method, i.e. with a knapsack sprayer on tomato (García-Santos et al. 2016) and with an air assisted sprayer side-by-side flat fan nozzles on grapevine (Codis et al. 2011). For estimating secondary emission fractions, a default time assessed of 1 day was considered.

Table 17: Main characteristics of the case study and the 2 scenarios, grapevine and tomato production

Crop	Grapevine	Tomato in open-field
Location	Loire Valley (France)	Martinique (French West Indies)
Climate type and weather station	Marine West Coast Climate (Cfb)* - Beaucouzé weather station	Tropical savanna climate (Aw)* - Le Prêcheur, Météo France weather station
Soil type	Sand on calcareous formation	Vitric andosol (FAO soil data)
Application method	Air assisted sprayer side-by-side flat fan nozzles	Knapsack sprayer
Cover crop family	Grass (<i>Pooideae</i>), Clover (<i>Fabaceae</i>)	Grass (<i>Panicoideae</i>), <i>Stylosanthes guianensis</i> (<i>Fabaceae</i>)
$f_{\text{eff,cover}}$		0.35 and 0.7

* from Köppen-Geiger climate classification (Beck et al. 2018)

To assess the variability of pesticide distribution on cover crop, different growth stages of the main crop ($f_{\text{intercept,crop}}$) were considered, as well as the effective area fraction of crop-free field that is cover cropped ($f_{\text{eff,cover}}$). For each scenario, two main crop growth stages were considered, namely the leaf development and the flowering stage/development of fruit/ripening; and defined as $f_{\text{intercept,crop}} = 0.3$ for leaf

development and $f_{\text{intercept,crop}} = 0.8$ for flowering (Linders et al. 2000). Two types of cover crop were considered, a planted cover crop on the row of the main crop, $f_{\text{soil} \rightarrow \text{cover}} = 0.5$, with a high pesticide interception fraction of $f_{\text{intercept,cover}} = 0.7$ (Renaud-Gentié et al. 2015a), and a spontaneous cover crop covering all field soil between the main crop with $f_{\text{soil} \rightarrow \text{cover}} = 1$ and a high pesticide interception fraction of $f_{\text{intercept,cover}} = 0.7$. Therefore, the effective area fractions of cover crop are respectively $f_{\text{eff,cover}} = 0.35$ and $f_{\text{eff,cover}} = 0.7$. Each of the cover crop was composed either of grass from *Pooideae* or clover from *Fabaceae* family for the grapevine production, and grass from *Panicoideae* or *Stylosanthes guianensis* from *Fabaceae* family for the tomato production. Covers from *Pooideae* family are mainly present in temperate and dry climates, whereas covers from *Panicoideae* family are dominating tropical and sub-tropical areas (Zuloaga et al. 2007). Cover crops of *Panicoideae* and *Pooideae* families are grass, either planted or spontaneous, while cover crops from the *Fabaceae* family are legumes planted. The cover crop family is required to estimate the pesticide degradation on leaves (see eq. 5.8). Pesticide dissipation from leaves is generally faster than from soil (Juraske et al. 2008) and varies according to plant family (Fantke and Juraske 2013). Two pesticides homologated on tomato and grapevine were tested with contrasted dissipation half-lives on soil and plant, namely the insecticide pyriproxyfen and the fungicide mancozeb. Their relevant physicochemical properties are detailed in SM, Table S5.1.

Pesticide emissions were assigned in the impact assessment model USEtox. The off-field deposition fraction was shared according to prevailing surface compartments in the region of each scenario. In Martinique, the off-field deposition fraction was allocated as follows: 29% to agricultural soil, 70% to natural soil and 1% to freshwater and in Loire Valley, 67% to agricultural soil, 31.2% to natural soil and 1.8% to freshwater.

Based on the calculation framework for emission distributions under GCM conditions, and the related calculation of ecotoxicity impacts, in the case study, we analyzed the effect of GCM on emission and related freshwater ecotoxicity impact results.

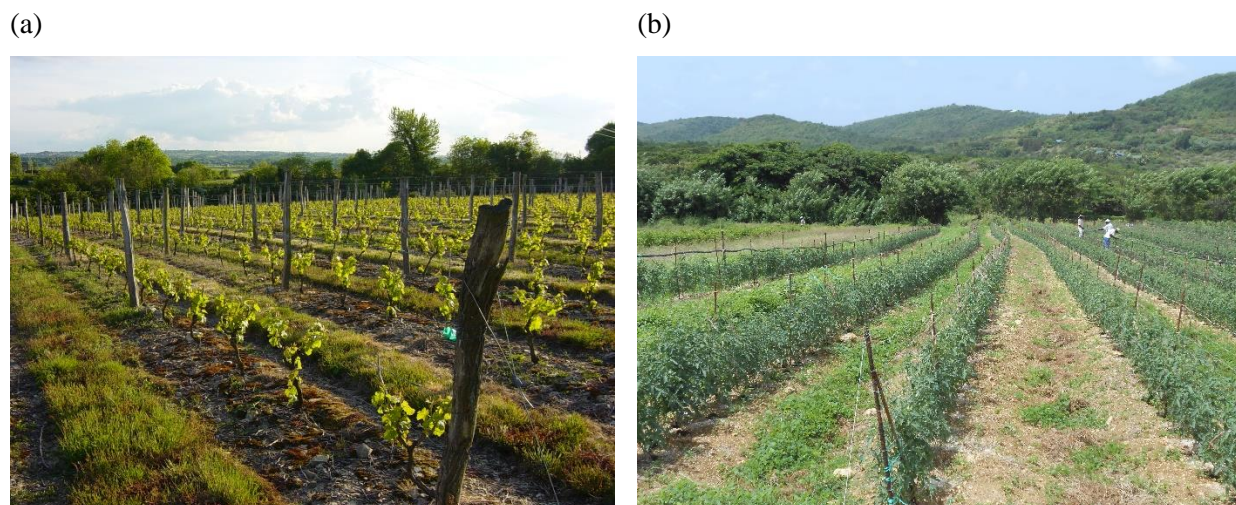


Figure 20: Cover crop on (a) grapevine in Anjou, in Loire Valley (Credits: Christel Renaud-Gentié) and (b) tomato production in Martinique, in French West Indies (Credits: Sarah Manteaux)

5.2.4 Analysis of GCM effect on pesticide emissions and related impacts

To assess cover crops effects on initial and secondary distributions, control scenarios without cover (i.e. with bare soil) for each combination of pesticide, crop and intercepted fractions (i.e. 8 control scenarios with $f_{\text{eff,cover}} = 0$) were designed and simulated. Distribution fractions are presented as a function of continuously increasing cover crop area fractions. After an analysis of the influence of the effective area fraction of cover crop ($f_{\text{eff,cover}}$) on initial distribution fractions, a second analysis was carried out on the secondary emission fractions and particularly on processes involved on cover leaves, namely degradation, volatilization and uptake.

At the impact level, scenarios with and without a cover crop were compared by calculating the percentage of change of impact scores (IS) for freshwater ecotoxicity ($\text{PAF m}^3 \text{ d} / \text{kg}_{\text{applied}}$), for each considered environmental emission compartment. The percentage of change (% – change) was calculated as:

$$\% \text{ – change} = \left[\frac{\text{IS without cover} - \text{IS with cover}}{\text{IS without cover}} \right] \times 100 \quad (5.17)$$

When pesticide emissions to cover leaves are assigned to the agricultural soil compartment as field soil surface emissions, this corresponds for initial distribution fractions to a scenario without considering any

cover. Therefore, the initial emission fraction reaching cover leaves, $f_{\text{field} \rightarrow \text{cover}}$, was not linked to the impact assessment but considered as a removal process as for the pesticide emission on crop leaves. For secondary emissions, two agronomic situations were considered for the fraction taken up by the cover and left on cover leaves ($f_{\text{cover-sec}}$). One situation considered that the cover crop was exported from the actual field crop, i.e. mowed and transferred outside the field. Hence, $f_{\text{cover-sec}}$ was not linked to the impact assessment. The second situation considered the cover buried into the field soil by e.g. a tillage practice, so $f_{\text{cover-sec}}$ was assumed to reach agricultural soil as emission. In summary, 3 situations were analyzed: (i) initial distribution with cover exported from the field, (ii) secondary emissions with cover exported from the field and (iii) secondary distribution with cover buried into the field soil.

5.3 Results – model application on the case study

5.3.1 Emission results

Across a range of effective area fractions of cover ($f_{\text{eff,cover}}$) for the tomato and grapevine scenarios, the variability of initial and secondary distribution fractions is presented in Figure 21. Emission fractions for the two cover crops ($f_{\text{eff,cover}} = 0.35$ and $f_{\text{eff,cover}} = 0.7$) are detailed in Table 18.

At initial distribution, air-assisted sprayer in grapevine shows higher off-field surfaces emissions, $f_{\text{dep}} = 0.04$ and higher emission to air, $f_{\text{air}} = 0.08$, than the knapsack sprayer used in tomato production with $f_{\text{dep}} = 0.02$ and $f_{\text{air}} = 0.06$.

The initial pesticide distributions to soil and cover crop depend on the main crop intercepted fraction and the effective area fraction of cover. The higher the fraction intercepted by the crop (reproductive-flowering phase), the lower the fraction distributed under the crop canopy (i.e. to the field soil and cover crop). Indeed, in our case study, when the main crop is starting to grow, the fraction of pesticide distributed between the soil and the cover crop was 3.5 times higher than when the main crop is at the flowering stage (Table 18). The presence of a cover crop decreased on average the fraction reaching field soil by a factor of 3 compared to a bare soil regardless of the field crop, its growth stage, and the effective area fraction of cover. The variation of the fraction reaching field soil among scenarios was mainly due to different effective area fractions of cover, driven by the area fraction of crop-free field that is cover crop and the area fraction of cover crop that is covered by leaves corresponding to the cover leaves density. The higher the effective area fraction of cover, the lower the fraction distributed to field soil. The effect of cover crop on pesticide initial distribution fractions to field soil and cover crop was propagated to the secondary emission fractions.

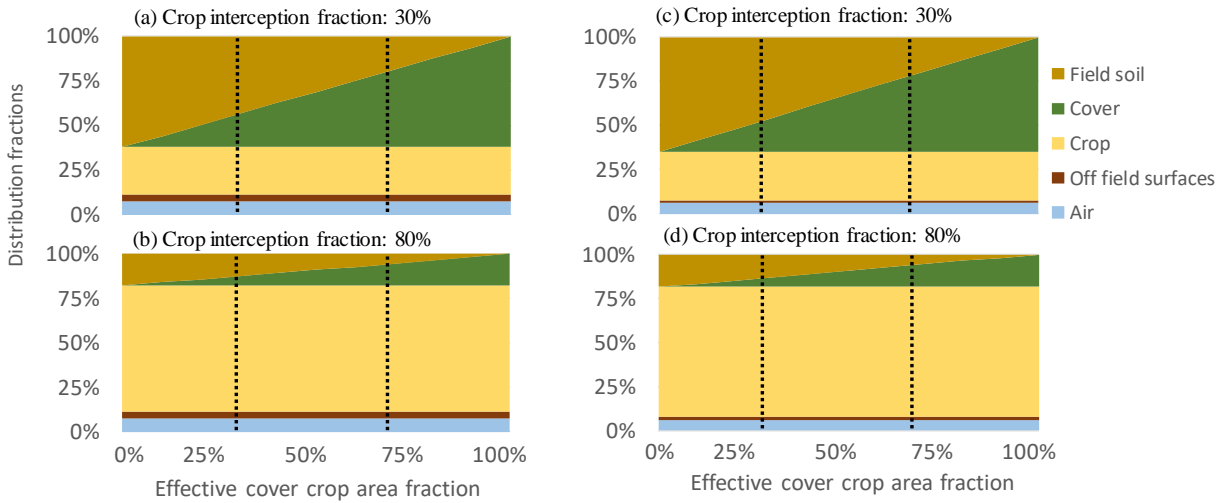
When including secondary emission distribution processes, cover crop leaf uptake contributed with more than 99% to cover crop-related processes, yielding an average leaf uptake fraction of 0.22 across all

scenarios. The total degraded fractions (on soil, on field crop and cover leaves) in our case study were highest when there was no cover crop. This is explained by the fact that despite faster degradation in crops and cover crops compared to soil, the even faster plant uptake into both crop and cover crop drives the overall pesticide distribution within 24 hours that are considered in the emission model. Consequently, most of the pesticide is taken up into crop, while degradation processes will drive subsequent fate processes that are modeled as part of the impact assessment. The degraded fractions on cover leaves showed only slight differences between the cover crop families, i.e. *Pooideae* vs *Fabaceae* for grapevine scenarios and *Panicoideae* vs *Fabaceae* for tomato scenarios. Indeed, degradation on *Fabaceae* leaves was lower than on *Pooideae* and *Panicoideae*, due to longer dissipation half-lives on *Fabaceae* (see detail of DT50 in SM, Table S5.3). On leaves, the only process considering crop and cover family is degradation. The fraction degraded on cover leaves was on average below 0.1%, but could reach a maximum of 0.65% for mancozeb application with full coverage of *Fabaceae* (i.e. $f_{\text{eff,cover}} = 1$). The fraction taken up by the cover that depends on the initial distribution fraction on cover leaves reached up to around 18% when the crop growth stage was on maturation ($f_{\text{intercept,crop}} = 0.8$), and up to around 60% when the crop growth stage was on installation ($f_{\text{intercept,crop}} = 0.3$) for both scenarios.

The presence of a cover crop did not modify initial distribution fractions to air, crop and off-field surfaces. It did not modify either secondary emission fractions to off-field surfaces, crop uptake and fraction left on crop leaves. For secondary distribution scenarios, processes that are by default initiated after the first day after application were not included in our study but require further analysis to ensure consistency with our proposed GCM processes and to evaluate emissions eventually reaching groundwater or surface water. With that, the fractions emitted to off-field surfaces did not vary with the presence or absence of a cover crop in our case study.

Overall, during initial and secondary distributions, the introduction of a cover crop with its own leaf surface area reduced the fraction reaching the field soil.

A. Initial distribution fractions



B. Secondary distribution fractions

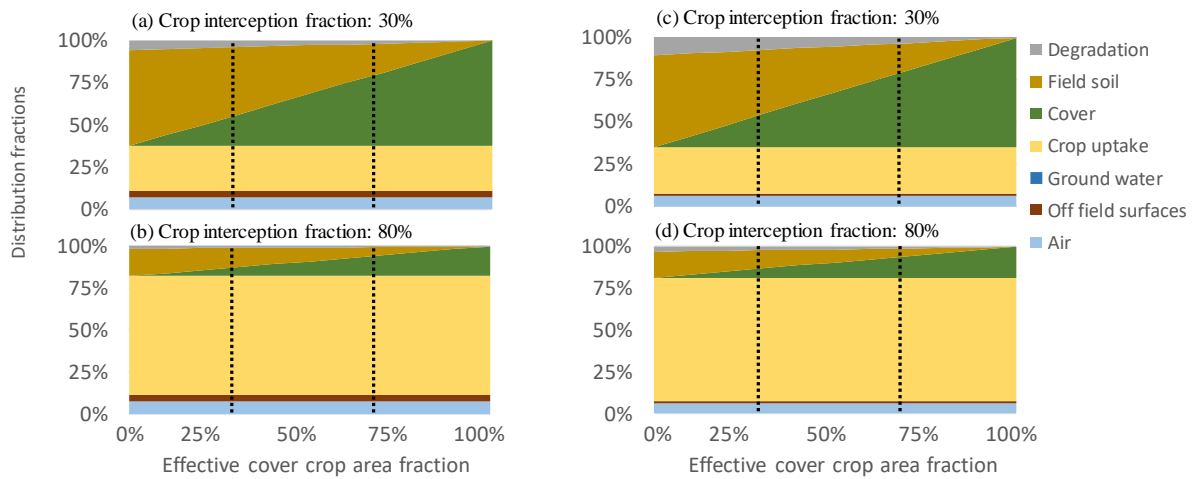


Figure 21: Initial (A) and secondary (B) emission distribution fractions for two crop growth stages, $f_{\text{intercept,crop}} = 0.3$ and $f_{\text{intercept,crop}} = 0.8$, respectively corresponding to the installation (a, c) and flowering stage (b, d) for the grapevine (a, b) and tomato (c, d) for a range of effective area fractions of crop-free field that is covered by cover crop ($f_{\text{eff,cover}}$). Vertical lines represent the cover crop setup of the case study with $f_{\text{eff,cover}} = 0.35$ and $f_{\text{eff,cover}} = 0.7$.

Table 18: Initial and secondary emission fractions for the tomato and grapevine production, for two crop growth stages, $f_{\text{intercept,crop}} = 0.3$ and $f_{\text{intercept,crop}} = 0.8$, for the effective area fraction of crop-free field that is covered by cover crop representing the two cover crop types with $f_{\text{eff,cover}} = 0.35$ and $f_{\text{eff,cover}} = 0.7$ (mean across scenarios).

		Crop →		Tomato				Grapevine			
		$f_{\text{intercept,crop}} \rightarrow$		0.3		0.8		0.3		0.8	
↓ Distribution	$f_{\text{eff,cover}} \rightarrow$ Compartment ↓	0.35	0.7	0.35	0.7	0.35	0.7	0.35	0.7	0.35	0.7
Initial	Air	0.06	0.06	0.06	0.06	0.08	0.08	0.08	0.08	0.08	0.08
	Off field surfaces	0.02	0.02	0.02	0.02	0.04	0.04	0.04	0.04	0.04	0.04
	Crop leaves	0.28	0.28	0.74	0.74	0.27	0.27	0.71	0.71	0.71	0.71
	Field soil	0.42	0.19	0.12	0.06	0.40	0.19	0.12	0.05	0.12	0.05
	Cover leaves	0.23	0.45	0.07	0.13	0.22	0.44	0.06	0.12	0.06	0.12
Secondary	Air	0.06	0.06	0.06	0.06	0.08	0.08	0.08	0.08	0.08	0.08
	Off field surfaces	0.02	0.02	0.02	0.02	0.04	0.04	0.04	0.04	0.04	0.04
	Groundwater	0	0	0	0	0	0	0	0	0	0
	Field soil	0.35	0.16	0.10	0.05	0.37	0.17	0.11	0.05	0.11	0.05
	Degradation	0.07	0.03	0.02	0.01	0.04	0.02	0.01	0.01	0.01	0.01
	Crop uptake	0.28	0.28	0.74	0.74	0.27	0.27	0.71	0.71	0.71	0.71
	Cover	0.23	0.45	0.07	0.13	0.22	0.43	0.06	0.12	0.06	0.12

5.3.2 Impact results

The impact scores for the scenarios with a cover crop and the percentage of change compared to the scenarios without a cover (i.e. bare soil) are presented in Table 19. The contribution of the USEtox environmental compartments to the total impact scores calculated for the scenarios with and without cover crop for tomato and grapevine are presented in Figure 22 according to the distribution used (initial or secondary).

Pesticide initial distribution fractions were assigned to the environmental compartments of the impact assessment model (i.e. USEtox), except the emission fractions to crop and cover leaves, because the plant compartment does not exist in the model. Only the impacts due to agricultural soil emissions were affected by the consideration of a cover crop, with a reduction of around 100% regardless of crop type. As a consequence, the total impact scores using initial distribution fractions were reduced by 65% for grapevine and by 90% for the tomato crop with the use of a cover crop.

Two connections were assessed between secondary emission fraction to cover and USEtox compartments. First, as for the connection with the initial distribution, no link to any USEtox compartment

was assumed for the emission fraction to cover, considering cover exported from the field. Second, the fraction taken up by and present on cover leaves was assigned to agricultural soil, considering the cover buried into the field soil.

Using either initial or secondary distribution fractions, impact scores due to emissions to the natural soil were not affected by cover crops, they are unrelated to emissions from off-field surfaces. In our two scenarios, the variation of impact scores to air with or without a cover was very small (% – change = 10^{-5} %). A variation of one order of magnitude was observed on the impact score due to emissions to air across scenarios due to the vapor pressure of the two pesticides tested (see details of pesticide characteristics in SM, table S5.1). Impact scores for emissions to continental freshwater (including emissions to ground and surface water) were not affected by the presence of a cover crop. In all scenarios, impact results based on secondary emission fractions, were mainly due to emissions to the continental agricultural soil compartment (Figure 22).

In all scenarios with and without cover crop, the total impact scores were higher on grapevine than on tomato. This is mainly due to the higher emission fractions to off-field surfaces for grapevine, with higher off-field surfaces water area fraction in Loire Valley (1.8%) than in Martinique (1%), and with higher characterization factors (CF) on emissions directly into the freshwater compartment than into other compartments (detail of CFs in SM, table S5.2).

Overall, across the two scenarios, the modeling of GCM demonstrated the potential to reduce emissions to field soil and related freshwater ecotoxicity impacts. Moreover, when considering the cover exported from the field and consequently not assigned to any environmental compartment in the impact assessment, total impact scores were reduced by around 65% (grapevine) and 90% (tomato) compared to bare soil scenarios for initial and secondary emissions. Conversely, when considering the cover buried into the field soil, total impact scores were similar compared to bare soil, reduced by only 1% (grapevine) and 3% (tomato).

Table 19: Impact score (IS in PAF m³ d/kg_{applied}) of the average scenarios with a cover crop for each environmental compartment (continental: rural air, freshwater, natural soil and agricultural soil), for the two scenarios: tomato and grapevine and %-change (eq. 5.17) when not using a cover crop (i.e. bare soil), for the application of two pesticides (mancozeb and pyriproxyfen). Calculation of %-change = [(IS_{without_cover} – IS_{with_cover})/IS_{with_cover}] \times 100

Emission distribution and cover crop fate	Crop	IS Rural air	IS Freshwater	IS Natural soil	IS Agricultural Soil	TOTAL
Initial distribution - with cover exported from the field*	Tomato	21 (<1%)	18 (-2%)	10 (1%)	201 (113%)	251 (91%)
	Grapevine	26 (<1%)	80 (-1%)	11 (1%)	214 (103%)	331 (66%)
Secondary emissions – cover exported from the field*	Tomato	21 (<0.1%)	18 (-)	10 (-)	189 (111%)	238 (88%)
	Grapevine	26 (<0.1%)	79 (-)	11 (-)	209 (100%)	326 (64%)
Secondary distribution - cover buried into the field soil**	Tomato	21 (<0.1%)	18 (-)	10 (-)	414 (-3%)	463 (-3%)
	Grapevine	26 (<0.1%)	79 (-)	11 (-)	424 (-2%)	541 (-1%)

* $f_{field \rightarrow cover}$ and $f_{cover \rightarrow sec}$ were not linked to the impact assessment model used (i.e. USEtox).

** $f_{cover \rightarrow sec}$ was assigned to continental agricultural soil in the impact assessment model used (i.e. USEtox).

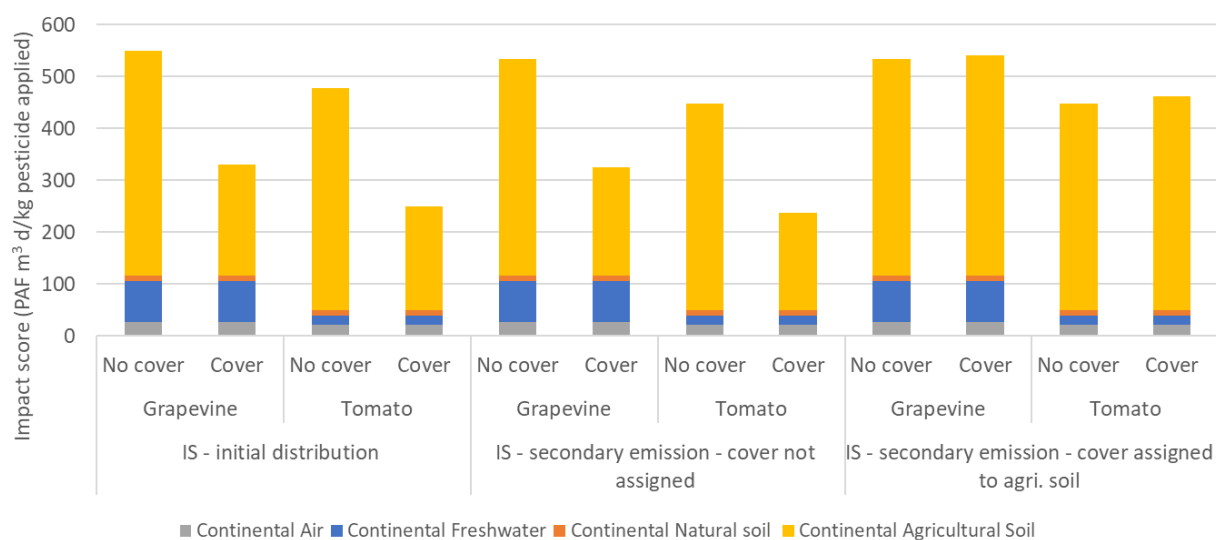


Figure 22: Contribution to total impact score in PAF m³ d/ kg pesticide applied, from each environmental compartment, for the tomato and grapevine scenarios, considering the presence of cover, using either the

initial or secondary distribution, and indicating whether the fraction emitted to cover is assigned to agricultural soil or not (results are mean results obtained with the two pesticides).

5.4 Discussion and outlook

5.4.1 Applicability and limitations of the presented approach

The modeling of GCM in pesticide emission analysis was considered, by defining an effective area of cover and cover family, for initial and secondary pesticide distributions as derived with the PestLCI Consensus model. Within 1 day of assessment time after pesticide application, processes on cover leaves, i.e. degradation, uptake and volatilization were modeled. The modeling of GCM fills a gap in the consideration of common farming practices, and demonstrated the potential of GCM to reduce emissions to field soil and related freshwater ecotoxicity impacts. Following our proposed approach for considering GCM processes, our emission results reproduce well the effect of GC on pesticide distribution as observed in cocoa farm systems by Vaikosen et al. (2019), where the reduction of bare soil surface with a GC of fallen leaves of the main crop decreased emission to the top soil. The higher the fraction of effective area of cover (composed of living or dead cover), the lower the fraction distributed to field soil.

Estimating pesticide interception by the crop or the cover crop is important to estimate pesticide losses to the environment (Lammoglia et al. 2016). Defining $f_{\text{eff,cover}}$ with the cover crop occupation fraction and the cover crop canopy fraction according to the growth stage of the cover allows estimating the pesticide interception by the cover crop, which could reach up to 60% during the installation phase of the main crop in our case study. Defining the crop intercepted fraction according to the leaf area index and the pesticide spraying technique for each crop family allows defining the intercepted fraction separately for the crop and the cover along the whole crop cycle and pesticide applications. As a consequence, the more precisely the fraction intercepted by the crop and the cover is defined, the better the estimate of the pesticide distribution for a living cover. The effective area fraction of cover should preferably be defined by observation. However, LCA practitioners rarely know the GCM and its characteristics. If the studied field cannot be visited, for crops grown in rows (e.g. orchards, grapevine) an effective area fraction of cover crop of $f_{\text{eff,cover}} = 0.35$ can be assumed for a planted cover at its maximum development and an effective area fraction of $f_{\text{eff,cover}} = 0.7$ for a spontaneous cover crop. Furthermore, a set of common living cover crops should be developed according to the main crop characteristics (e.g. on banana production at leaf development stage), defining the cover effective area fraction and the cover family.

After pesticide application, processes of degradation, uptake and volatilization occur on the crop and cover leaves. To estimate the dissipation half-lives on leaves, which are crop dependent (Fantke et al. 2014), the cover family (mostly composed of *Pooideae*, *Panicoideae* or *Fabaceae*) has to be defined. For multispecies cover (e.g. weeds development of multiple species), the dominant cover crop family should be selected as reference.

In our two scenarios, the assignment of the emissions on the cover crop (uptake and left on leaves) to agricultural soil (considering the cover crop buried into the field) was compared with no assignment to any compartment (considering the cover exported from the field). The importance of the differences obtained with these two calculations (impact score reduced by 65% for grapevine and by 90% for tomato when cover not assigned to any environmental compartments as emission compared to assigned to field soil) needs to be relativized compared to the high uncertainty of the pesticide characterization factors for which uncertainties of one to three orders of magnitude apply (Rosenbaum et al. 2008). This is notably due to the absence of modeling of the impact on soil life and pesticide metabolites (Notarnicola et al. 2017; van der Werf et al. 2020).

In the absence of complete sets of measured emission fractions to the environment, our proposed approach can only be qualitatively discussed, notably exploring other models design. Most pesticide transfer models at field scale (e.g. GLEAMS, MACRO, PEARL, PRZM) do not consider cover crop, and rather consider GC as plant residues (e.g. GLEAMS, PRZM) (Mottes et al. 2014). Few models consider cover crops and associated pesticide transfer (i.e. R-pest, WATPPASS) as proposed in this study. The R-pest model coupled with SIMBA allows simulating banana and diverse cover crops and defining an indicator of pesticide risk exposure to surface and groundwater (Tixier et al. 2007, 2011). The WATPPASS model, an hydrological model for small tropical volcanic catchments, allows to consider various types of ground cover, modifying environmental characteristics and thus pesticide transfers (Mottes 2013; Mottes et al. 2015). As in those two models, the consideration of GCM in our proposed approach allows considering an important farming practice and its effects on pesticide transfer to the environment. Nevertheless, the outputs from those models cannot be compared to our emission fractions, mainly because they are not based on an emission mass balance as PestLCI Consensus.

To generalize our conclusions on GCM pesticide emissions and related impacts, a sensitivity analysis should be performed notably testing the GCM modeling with a wide range of pesticides with diverse characteristics (e.g. DT50) and including additional environmental processes, climate conditions and field crop as well as cover crop types (see 4.2). Overall, our proposal for modeling GCM raises awareness of the fraction of pesticides reaching living cover inside the field crop, which can affect the distribution of pesticides to the environment with its possible (eco-)toxicity impacts through agricultural soil emissions.

5.4.2 Future research needs

Additional research efforts are required on the modeling of processes occurring few days after pesticide application (secondary distribution), with minimum redundancy with the impact assessment fate model. Modeling secondary distribution processes requires the accounting of local field, farming practices, soil and climate conditions, which is of particular interest to assess farming systems diversity. Further research is required to consistently include additional processes, such as run-off, as well as to account for the influence of GCM on pesticide residues in edible field crops and on remaining pesticide fractions in unharvested cover crops.

More specifically, GC may affect water processes, such as run-off, leaching or preferential flow (Reichenberger et al. 2007; Alletto et al. 2010; Mottes et al. 2014), hence have a potential influence on impacts. At emission level, GCM should reduce emissions to off-field surfaces and to groundwater (Potter et al. 2007), and consequently at impact level, total impact scores might be reduced for scenarios with a GC. Further research is therefore required to incorporate such processes consistently in our proposed GCM approach.

As recommended in Fantke et al. (2018), pesticide emissions were assigned to continental rural air, freshwater, agricultural soil and natural soil compartments of the impact model USEtox. Recently, PestLCI initial distributions and the plant uptake and crop residue exposure model dynamiCROP (Fantke et al. 2011b), which was recently adjusted for LCA (Fantke and Jolliet 2016), were coupled (Gentil et al. 2020a), allowing to consider human toxicity due to pesticides present in the harvested and consumed part of field crops. The pesticide deposited on non-harvested living or dead plant, inside (e.g. cover crops) or outside the field (e.g. hedge tree), might have (eco-)toxicity impacts (Sharma et al. 2019) and requires further research to be fully considered. Indeed, if crop residues and unharvested cover crop remain on the field, there could be further emissions to field soil and air. The modeling of plant root uptake would be required to assess pesticide emissions to crop and cover crop residues (on the non-harvested stem, roots and leaves). Whenever cover crops are removed from the field, related impacts will depend on the subsequent processes (e.g. burning).

As for the modeling of pesticide degradation on crop and cover leaves, volatilization and plant uptake should be refined to be modeled as function of crop family, considering in particular leaf characteristics.

Further improvement of GCM should be considered for modeling dead GC. Indeed, mulch or crop residues left on field (e.g. stripping of banana while growing) are largely used in tropical conditions to keep moisture or reduce soil erosion (Lewis et al. 2016). At the initial distribution stage, all types of GC (living or dead mulch) can be modeled, by defining the effective area fraction of cover ($f_{\text{eff,cover}}$). At the secondary distribution stage, specificities of the dead cover must be considered and modeled. Mulch is generally

modeled as the first soil layer (Mottes et al. 2015) with its own characteristics notably of composition and degradation rate (Cassigneul et al. 2015). Plastic mulches are also used on vegetable production or on pineapple production for example, generating specific water flows and consequently requiring a specific modeling (Dusek et al. 2010). The inclusion of dead covers would be an important future step forward for modeling different types of GCM in the estimation of pesticide emissions in LCA and elsewhere.

Overall, the modeling of GC opened the path towards the modeling of pesticide emissions in intercropping systems. This is particularly important since these systems are widely conducted in market gardening in tropical conditions to ensure income stability (Malézieux et al. 2009), increase crop yield per hectare and optimize field conditions for growing certain crops (e.g. banana/cocoa).

5.5 Conclusions

The inclusion of GCM in pesticide emission modeling was proposed, defining an effective area of cover and the cover family through initial and secondary pesticide distributions. Across the two scenarios on tomato in Martinique and on grapevine in Loire Valley, the modeling of GCM allowed highlighting the potential of soil cover to reduce pesticide emissions and related freshwater ecotoxicity impacts. Including, a new fraction on and in cover crop leaves, the emissions to field soil decrease by a factor of 3. During the secondary distribution, over the three processes occurring on the cover leaves, the fraction taken up by the cover leaves was predominant with more than 99% contribution to these processes. At both initial and secondary distribution levels, considering the cover exported from the field and consequently not assigned to any environmental emission compartment, total impact scores were reduced by around 65% and 90% compared to bare soil, respectively for grapevine and tomato. Additional processes, such as run-off, should be considered in future efforts, along with accounting for the influence of GCM on pesticide residues in edible field crops and on remaining pesticide fractions in unharvested cover crops. Indeed, if crop residues and unharvested cover crop remain on the field, there could be further emissions to soil and air. The inclusion of dead cover would be an important step forward to achieve the modeling of various types of GCM.

From the initial work on vine of Renaud-Gentié et al. (2015), the consideration of GCM as common farming practice opened the possibility to model it more widely for all crops with living cover crop. The modeling of living cover crop also opened the path towards the modeling of pesticide emissions in intercropping systems, widely conducted in market gardening in particular in tropical regions. With that, our proposed approach constitutes a valuable starting point for addressing GCM practices in emission and impact assessments applied in LCA and elsewhere.

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References Chapter 5

- Alletto L, Coquet Y, Benoit P, et al (2010) Tillage management effects on pesticide fate in soils. A review. *Agron Sustain Dev* 30:367–400. doi.org/10.1051/agro/2009018
- Ambiaud E (2012) Moins de désherbants dans les vignes. *Agreste Primeur*:8 pages
- Andersson K (2000) LCA of food products and production systems. *Int J LCA* 5:239. doi.org/10.1007/BF02979367
- Beck HE, Zimmermann NE, McVicar TR, et al (2018) Present and future Köppen-Geiger climate classification maps at 1-km resolution. *Scientific Data* 5:180214. doi.org/10.1038/sdata.2018.214
- Bessou C, Basset-Mens C, Tran T, Benoist A (2013) LCA applied to perennial cropping systems: a review focused on the farm stage. *Int J Life Cycle Assess* 18:340–361. doi.org/10.1007/s11367-012-0502-z
- Birkved M, Hauschild MZ (2006) PestLCI—A model for estimating field emissions of pesticides in agricultural LCA. *Ecol Model* 198:433–451. doi.org/10.1016/j.ecolmodel.2006.05.035
- Cassigneul A, Alletto L, Benoit P, et al (2015) Nature and decomposition degree of cover crops influence pesticide sorption: Quantification and modelling. *Chemosphere* 119:1007–1014. doi.org/10.1016/j.chemosphere.2014.08.082
- CIRAD (2009) *Mémento de l'agronome*. Ed. Quae
- Codis S, Bos C, Laurent S (2011) Réduction de la dérive, 8 matériels testés sur vigne. *Phytoma* 640:1–5
- DAAF Martinique (2018) *Pratiques culturales en 2015 : Kou d'zyé sur celles de la banane en Martinique*. Dossiers Agreste Martinique
- Dijkman TJ, Birkved M, Hauschild MZ (2012) PestLCI 2.0: a second generation model for estimating emissions of pesticides from arable land in LCA. *Int J Life Cycle Assess* 17:973–986. doi.org/10.1007/s11367-012-0439-2
- Durán Zuazo VH, Rodríguez Pleguezuelo CR (2008) Soil-erosion and runoff prevention by plant covers. A review. *Agron Sustain Dev* 28:65–86. doi.org/10.1051/agro:2007062

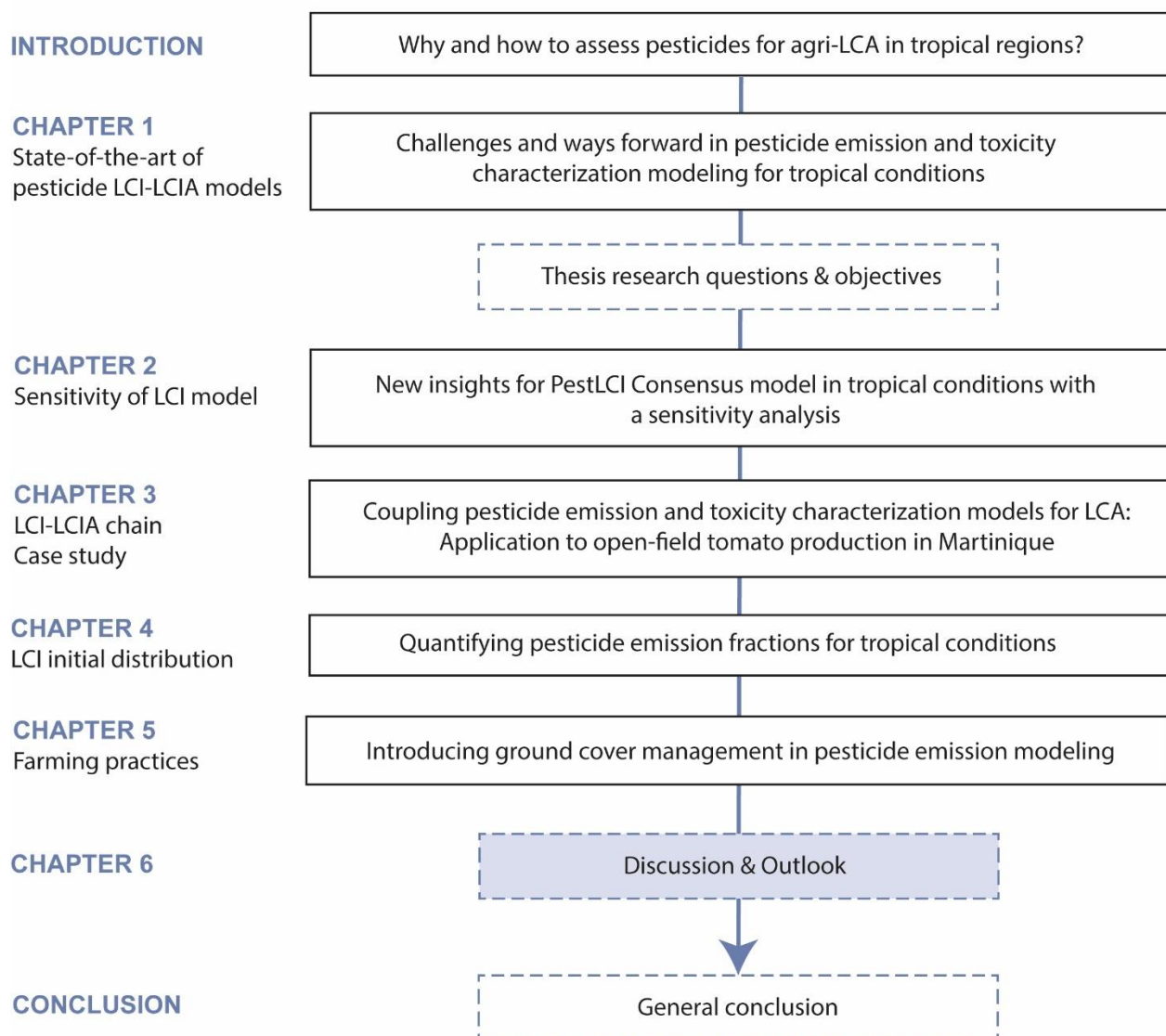
- Dusek J, Ray C, Alavi G, et al (2010) Effect of plastic mulch on water flow and herbicide transport in soil cultivated with pineapple crop: A modeling study. *Agricultural Water Management* 97:1637–1645. doi.org/10.1016/j.agwat.2010.05.019
- Fantke P, Antón A, Grant T, Hayashi K (2017) Pesticide emission quantification for life cycle assessment: A global consensus building process. *J Life Cycle Assess* 13:245–251
- Fantke P, Aurisano N, Bare J, et al (2018) Toward harmonizing ecotoxicity characterization in life cycle impact assessment. *Environ Toxicol Chem* 37:2955–2971. doi.org/10.1002/etc.4261
- Fantke P, Gillespie BW, Juraske R, Jolliet O (2014) Estimating half-lives for pesticide dissipation from plants. *Environ Sci Technol* 48:8588–8602. doi.org/10.1021/es500434p
- Fantke P, Jolliet O (2016) Life cycle human health impacts of 875 pesticides. *Int J Life Cycle Assess* 21:722–733. doi.org/10.1007/s11367-015-0910-y
- Fantke P, Juraske R (2013) Variability of Pesticide Dissipation Half-Lives in Plants. *Environ Sci Technol* 47:3548–3562. doi.org/10.1021/es303525x
- Fantke P, Juraske R, Antón A, et al (2011) Dynamic multicrop model to characterize impacts of pesticides in food. *Environ Sci Technol* 45:8842–8849. doi.org/10.1021/es201989d
- García-Santos G, Feola G, Nuyttens D, Diaz J (2016) Drift from the use of hand-held knapsack pesticide sprayers in Boyacá (Colombian Andes). *J Agric Food Chem* 64:3990–3998. doi.org/10.1021/acs.jafc.5b03772
- Gentil C, Basset-Mens C, Manteaux S, et al (2020) Coupling pesticide emission and toxicity characterization models for LCA: Application to open-field tomato production in Martinique. *Journal of Cleaner Production* 277:124099. doi.org/10.1016/j.jclepro.2020.124099
- Griggs D, Stafford-Smith M, Gaffney O, et al (2013) Sustainable development goals for people and planet. *Nature* 495:305–307. doi.org/10.1038/495305a
- HLPE (2019) Agroecological and other innovative approaches for sustainable agriculture and food systems that enhance food security and nutrition. A report by the High Level Panel of Experts on Food Security and Nutrition of the Committee on World Food Food Security. 163
- Juraske R, Antón A, Castells F (2008) Estimating half-lives of pesticides in/on vegetation for use in multimedia fate and exposure models. *Chemosphere* 70:1748–1755. doi.org/10.1016/j.chemosphere.2007.08.047
- Lammoglia S-K, Moeys J, Barriuso E, et al (2016) Sequential use of the STICS crop model and of the MACRO pesticide fate model to simulate pesticides leaching in cropping systems. *Environ Sci Pollut Res* 24:6895–6909. doi.org/10.1007/s11356-016-6842-7
- Lewis SE, Silburn DM, Kookana RS, Shaw M (2016) Pesticide behavior, fate, and effects in the tropics: an overview of the current state of knowledge. *J Agric Food Chem* 64:3917–3924. doi.org/10.1021/acs.jafc.6b01320
- Linders J, Mensink H, Stephenson G, et al (2000) Foliar interception and retention values after pesticide application: a proposal for standardised values for environmental risk assessment. *Pest Manag Sci* 58:315–315. doi.org/10.1002/ps.448

- Malézieux E, Crozat Y, Dupraz C, et al (2009) Mixing plant species in cropping systems: concepts, tools and models. A review. *Agronomy for Sustainable Development* 29:43–62. doi.org/10.1051/agro:2007057
- Meier MS, Stoessel F, Jungbluth N, et al (2015) Environmental impacts of organic and conventional agricultural products – Are the differences captured by life cycle assessment? *J Environ Manag* 149:193–208. doi.org/10.1016/j.jenvman.2014.10.006
- Meier U (2018) Growth stages of mono- and dicotyledonous plants: BBCH Monograph. Open Agrar Repository
- Mottes C (2013) Evaluation des effets des systèmes de culture sur l'exposition aux pesticides des eaux à l'exutoire d'un bassin versant. Proposition d'une méthodologie d'analyse appliquée au cas de l'horticulture en Martinique. AgroParisTech
- Mottes C, Lesueur Jannoyer M, Le Bail M, et al (2017) Relationships between past and present pesticide applications and pollution at a watershed outlet: The case of a horticultural catchment in Martinique, French West Indies. *Chemosphere* 184:762–773. doi.org/10.1016/j.chemosphere.2017.06.061
- Mottes C, Lesueur-Jannoyer M, Bail ML, Malézieux E (2014) Pesticide transfer models in crop and watershed systems: a review. *Agron Sustain Dev* 34:229–250. doi.org/10.1007/s13593-013-0176-3
- Mottes C, Lesueur-Jannoyer M, Charlier J-B, et al (2015) Hydrological and pesticide transfer modeling in a tropical volcanic watershed with the WATPPASS model. *Journal of Hydrology* 529:909–927. doi.org/10.1016/j.jhydrol.2015.09.007
- Nemecek T, Schnetzer J (2011) Methods of assessment of direct field emissions for LCIs of agricultural production systems. Data v3.0. Swiss Center for Life Cycle Inventories, Duebendorf, Switzerland
- Neto B, Dias AC, Machado M (2013) Life cycle assessment of the supply chain of a Portuguese wine: from viticulture to distribution. *Int J Life Cycle Assess* 18:590–602. doi.org/10.1007/s11367-012-0518-4
- Notarnicola B, Sala S, Antón A, et al (2017) The role of life cycle assessment in supporting sustainable agri-food systems: A review of the challenges. *J Clean Prod* 140:399–409. doi.org/10.1016/j.jclepro.2016.06.071
- Oliquino-Abasolo A (2015) Agro-environmental sustainability of conventional and organic vegetable production systems in Tayabas, Quezon, Philippines. FAO University Library, University of the Philippines at Los Baños:
- Perrin A, Basset-Mens C, Gabrielle B (2014) Life cycle assessment of vegetable products: a review focusing on cropping systems diversity and the estimation of field emissions. *Int J Life Cycle Assess* 19:1247–1263. doi.org/10.1007/s11367-014-0724-3
- Poore J, Nemecek T (2018) Reducing food's environmental impacts through producers and consumers. *Science* 360:987–992. doi.org/10.1126/science.aaq0216
- Potter TL, Bosch DD, Joo H, et al (2007) Summer Cover Crops Reduce Atrazine Leaching to Shallow Groundwater in Southern Florida. *Journal of Environmental Quality* 36:1301–1309. doi.org/10.2134/jeq2006.0526

- Reichenberger S, Bach M, Skitschak A, Frede H-G (2007) Mitigation strategies to reduce pesticide inputs into ground- and surface water and their effectiveness; a review. *Sci Total Environ* 384:1–35. doi.org/10.1016/j.scitotenv.2007.04.046
- Renaud-Gentié C, Dieu V, Thiollot-Scholtus M, Mérot A (2020) Addressing organic viticulture environmental burdens by better understanding interannual impact variations. *Int J Life Cycle Assess* 25:1307–1322. doi.org/10.1007/s11367-019-01694-8
- Renaud-Gentié C, Dijkman TJ, Bjørn A, Birkved M (2015) Pesticide emission modelling and freshwater ecotoxicity assessment for Grapevine LCA: adaptation of PestLCI 2.0 to viticulture. *Int J Life Cycle Assess* 20:1528–1543. doi.org/10.1007/s11367-015-0949-9
- Rosenbaum RK, Bachmann TM, Gold LS, et al (2008) USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment. *Int J Life Cycle Assess* 13:532. doi.org/10.1007/s11367-008-0038-4
- Rouault A, Beauchet S, Renaud-Gentie C, Jourjon F (2016) Life Cycle Assessment of viticultural technical management routes (TMRs): comparison between an organic and an integrated management route. *OENO One* 50:. doi.org/10.20870/oeno-one.2016.50.2.783
- Rouault A, Perrin A, Renaud-Gentié C, et al (2020) Using LCA in a participatory eco-design approach in agriculture: the example of vineyard management. *Int J Life Cycle Assess* 25:1368–1383. doi.org/10.1007/s11367-019-01684-w
- Roy P, Nei D, Orikasa T, et al (2009) A review of life cycle assessment (LCA) on some food products. *Journal of Food Engineering* 90:1–10. doi.org/10.1016/j.jfoodeng.2008.06.016
- Sharma A, Kumar V, Thukral AK, et al (2019) Responses of Plants to Pesticide Toxicity: an Overview. *Planta Daninha* 37:. doi.org/10.1590/s0100-83582019370100065
- Tixier P, Lavigne C, Alvarez S, et al (2011) Model evaluation of cover crops, application to eleven species for banana cropping systems. *European Journal of Agronomy* 34:53–61. doi.org/10.1016/j.eja.2010.10.004
- Tixier P, Malézieux E, Dorel M, et al (2007) Rpest—An indicator linked to a crop model to assess the dynamics of the risk of pesticide water pollution. *European Journal of Agronomy* 26:71–81. doi.org/10.1016/j.eja.2006.08.006
- Tscharntke T, Clough Y, Wanger TC, et al (2012) Global food security, biodiversity conservation and the future of agricultural intensification. *Biological Conservation* 151:53–59. doi.org/10.1016/j.biocon.2012.01.068
- Vaikosen EN, Olu-Owolabi BI, Gibson LT, et al (2019) Kinetic field dissipation and fate of endosulfan after application on *Theobroma cacao* farm in tropical Southwestern Nigeria. *Environ Monit Assess* 191:196. doi.org/10.1007/s10661-019-7293-7
- van der Werf HMG, Knudsen MT, Cederberg C (2020) Towards better representation of organic agriculture in life cycle assessment. *Nature Sustainability* 3:419–425. doi.org/10.1038/s41893-020-0489-6
- Wezel A, Casagrande M, Celette F, et al (2014) Agroecological practices for sustainable agriculture. A review. *Agron Sustain Dev* 34:1–20. doi.org/10.1007/s13593-013-0180-7

Zhang N, Zhang Q, Li Y, et al (2020) Effect of Groundcovers on Reducing Soil Erosion and Non-Point Source Pollution in Citrus Orchards on Red Soil Under Frequent Heavy Rainfall. *Sustainability* 12:1–16

Zuloaga F, Morrone O, Davidse G, Pennington S (2007) Classification and Biogeography of Panicoideae (Poaceae) in the New World. *Aliso: A Journal of Systematic and Evolutionary Botany* 23:503–529. doi.org/10.5642/aliso.20072301.39



CHAPTER 6

DISCUSSION AND OUTLOOK

This last chapter is dedicated to the wider discussion and outlook of the thesis work. The main goal of the thesis was to advance emission and impact modeling for agricultural pesticides under tropical conditions for improving scientific foundation of the environmental evaluation of tropical agri-food systems.

6.1 Progress made to assess pesticide emissions and related impacts for agri-LCA in tropical conditions – Discussion and context of main thesis results

In Figure 23, the main thesis results to assess pesticides applied on crops grown in tropical conditions are synthesized along the LCI-LCIA chain.

6.1.1 State-of-the-art of pesticide emissions and impacts modeling in LCA for crops grown under tropical conditions – identification of ways of improvements

Chapter 1 and 2 highlighted that current LCI and LCIA models are not fully suitable for evaluating pesticide emissions and impacts for crops cultivated in tropical regions. Indeed, the models commonly reflect temperate conditions for climate, soil, application methods and crops; whereas processes driving pesticide emission and impact patterns under tropical conditions are specific in relation to these characteristics (Figure 7 in Chapter 1). To address emissions and impacts of pesticides applied under tropical conditions, a first outline of how the models could be adapted was provided (Chapter 1) and the sensitivity of emission results and impact scores to PestLCI Consensus input variables was tested in relation to tropical conditions (Chapter 2). The most important inputs and processes on which focusing adaptations to tropical conditions were identified: (i) the characteristics of tropical crops and related growth stages with the foliar interception fraction, (ii) the application method and its drift, and (iii) soil processes influencing emissions

to water bodies. The scenario analysis highlighted that secondary emissions to freshwater contributed on average to 75% of the freshwater ecotoxicity (based on an insecticide application on tomato in tropical conditions as a reference scenario). Therefore, the rain-related processes modeling according to local soil and climate conditions need to be further improved. Moreover, farming practices such as ground cover management influencing pesticide transfer has been identified as key to estimate pesticide emissions for agri-LCA. Furthermore, due to ambiguous boundaries between LCI and LCIA stages (van Zelm et al. 2014; Fantke 2019), reflection and consensus on the conceptual framework of the LCI-LCIA calculation chain were recommended. Besides, a consistent inclusion of plant uptake and crop residue exposure modeling in LCA appeared to be essential. Indeed, food crop consumption was identified as the main pesticide exposure pathway for the general human population (Fantke and Jolliet 2016). Moreover, in the scenario analysis almost 50% of pesticides were distributed to crop leaves with the potential to end up as residues in harvested crop components for human or animal consumption.

Therefore, the thesis work was mainly focused on: (i) the LCI stage, i.e. on pesticide emission modeling, for crops grown in tropical conditions, considering all compartments to which pesticides are emitted after their application; and (ii) on model coupling, in order to fully assess pesticide emissions and related impacts in tropical conditions, including those from plant uptake and crop pesticide residues exposure.

6.1.2 A coupling of pesticide emission and impact models for LCA – consideration of pesticide residues exposure in crop on human toxicity

In the chapter 3, the state-of-the-art LCI and LCIA models were coupled for assessing the emissions and (eco-)toxicity impacts due to pesticide applications. Their combined use was illustrated with a case study on tomato production in Martinique (Figure 11 in Chapter 3). In response to the current separate use of the models (presented in Figure 5 in the general introduction), the thesis work provided a consistent coupling of the three models. These improvements are presented in Figure 23. An operational assessment of pesticide (eco-)toxicity impacts was developed, including the consideration of the main route of exposure for human health, namely crop pesticide residues ingestion (Fantke and Jolliet 2016), with the dynamiCROP model. This formalization will help practitioners to use PestLCI Consensus, dynamiCROP and USEtox consistently and evaluate the (eco-)toxicity impacts in their LCA studies in all conditions, not only tropical. Moreover, this approach, using pesticide initial distribution, is suitable for LCA studies where the farm stage is either in the background or in the foreground part of the system (consideration of application method and its drift, and crop characteristics of interception). For LCA studies where the farm stage is part of the foreground system, the use of secondary emission fractions seems more appropriate to compare farming systems, practices and/or soil and climate conditions. Nevertheless, secondary emission calculations are

more demanding in input data and not fully satisfactory yet regarding the modeling of rain-related processes. In the tomato case study, freshwater ecotoxicity impacts were higher using secondary emission fractions than when using the initial distribution. Furthermore, freshwater ecotoxicity and human toxicity impacts for organic and metal-based substances were evaluated separately for best possible transparency according to current recommendations (Frischknecht and Jolliet 2019) and to allow highlighting dominating contributors in both substance categories. The cradle-to-farm-gate LCA study of tomato production in Martinique highlighted that the major contributor to human toxicity was pesticide residues in crop and freshwater ecotoxicity impacts were dominated by pesticide field emissions from emission to agricultural soil and freshwater. This confirms the need in LCA to assess pesticide emissions and impacts, including plant uptake and crop pesticide residues exposure modeling.

6.1.3 Pesticide initial distribution fractions for crops grown in tropical conditions

In support of evaluating pesticide emissions under tropical conditions, the initial distribution has been refined to crops grown in tropical conditions, providing a set of foliar interception fractions as function of the crop, the growth stage and the spraying technique (i.e. application over and under canopy, plant-by-plant and soil application) (in Chapter 4). This approach considering the spraying technique, i.e. the part of the field receiving the pesticide application related to the application method, is particularly important for hand operated application. Indeed, these applications are either under crop canopy or plant-by-plant for knapsack sprayer, producing different foliar interception fractions according to the spraying technique. Furthermore, knowing the LAI of the crop during pesticide application, the foliar interception fraction can be adapted to the studied field and growth stage. As the most influent input variable in pesticide emission modeling, the refinement of foliar interception fractions of a set of crops grown in tropical regions (banana, bean, coffee, cotton, pineapple, soybean, sugar cane, tropical fruit trees, tubers and vegetables fruits), helps to consistently evaluating emission fractions for these crops in tropical regions. Furthermore, drift deposition fractions were introduced for a set of common crop-application method combinations in tropical conditions, with 29 drift deposition curves for boom sprayer, knapsack sprayer, air blast sprayer, ultra-low volume and aerial application. According to the development of foliar interception fractions and application methods for tropical crops, initial distribution fractions were provided for a set of crop-application methods with all crop growth stages and possible spraying techniques. It is particularly useful for LCA practitioners with little access to data on phytosanitary practices replacing the default over-simplifying assumption of 100% of pesticide applied being emitted to soil.

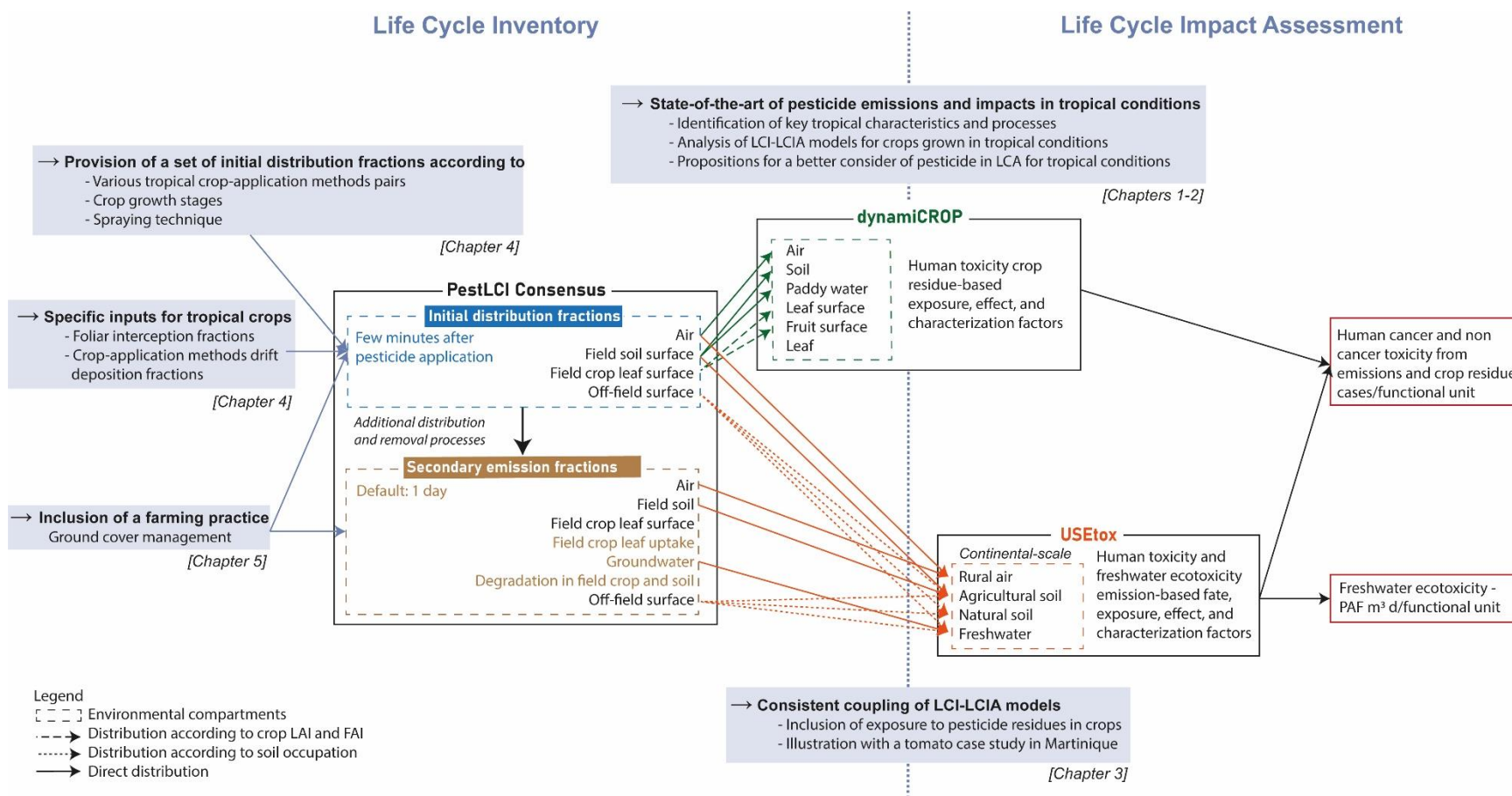


Figure 23: Synthesis of the main thesis results along the LCI-LCIA chain for pesticide assessment and proposed connection of the emission compartments of PestLCI to those of dynamiCROP and USEtox

6.1.4 Modeling of ground cover management in pesticide emission modeling

Finally, chapter 5 proposed a framework to introduce a common agricultural practice, i.e. the ground cover management (GCM), in the modeling of pesticide emissions, through initial distribution and secondary emissions. The development was performed for a living cover, defining an effective area fraction of cover and the cover family, for all crop types and conditions. The modeling of this practice for crops grown in tropical conditions is particularly important, because there are often no bare soil areas in the cultivated plots in tropical agriculture. Indeed, a common characteristic of tropical crop farming is the possibility to grow crops almost all year round without interruption by a cold season, as it is the case in temperate climates. In a context of agro-ecological transition (Wezel et al. 2014; HLPE 2019), the modeling of GCM allows to better compare different farming systems, such as conventional, integrated and organic farming (Meier et al. 2015) and to support eco-design of new agricultural practices (Rouault et al. 2020). This will allow to better inform stakeholders and decision-makers on agricultural practices and related (eco-)toxicity impacts in LCA. The proposed approach also constitutes a step forward to the modeling of pesticide emissions in intercropping systems, widely conducted in market gardening in particular in tropical regions (Malézieux et al. 2009).

In **conclusion**, this thesis produced methodological, scientific and operational advances for assessing pesticide emissions and impacts of crops grown in tropical conditions, in particular by developing foliar interception fractions for tropical crops and by including drift deposition curves for a set of crop-application method combinations defined in tropical areas. More generally for all agricultural contexts, a consistent coupling of LCI-LCIA models was proposed, including human toxicity due to exposure to pesticide residues in crops. A framework to account for ground cover management in the modeling of pesticide emissions was proposed to evaluate the environmental sustainability of agricultural practices.

However, this thesis work has limitations and some points initially identified could not be developed. The second part of the discussion will present the main limitations of the thesis work and further development on pesticide emission and impact modeling in tropical conditions will be proposed.

6.2 Limitations and recommendations for future developments on pesticide emission and impact modeling in tropical conditions

6.2.1 Refinements of initial pesticide emissions distribution

Providing foliar interception fractions and application methods for crops grown in tropical conditions helps to enhance the estimation of pesticide emissions in these conditions. However, certain margins for further improvements remain. By grouping the 10 main growth stages (from the BBCH scale (Meier 2018)) of crops into 3 stages (installation, development and reproductive phase), the foliar interception fractions are less specific. Furthermore, the proposed crop-application method combinations and related drift deposition fractions are likely to be applicable to several tropical crops. This is in particular the case for aerial application drift deposition fraction from Baio et al. (2019) which was defined for various crops (i.e. cotton, millet, sorghum, soybean and corn). Nevertheless, in relation to the application method, drift deposition to off-field surfaces is highly dependent on crop height and should be carefully extrapolated to other situations of application in which crop characteristics are unknown. Specifically, pesticide application with air blast sprayer is highly dependent on trees' organization in the field and on crop growth stages. These aspects should ideally be accounted for in the estimation of drift deposition.

The sensitivity analysis (chapter 2) showed that field width was influent on the estimation of emissions to off-field surfaces, but not the field length. The use of field dimensions as input variables is questioned since it does not seem relevant in relation to drift experiment results used to estimate pesticide drift to off-field surfaces. Indeed, drift deposition measurements are not based on a mass balance approach and they are not designed for a specific field size or shape. Instead, they are designed for a single sufficiently large field area to estimate drift without further increase of drift to off-field surfaces in relation to an increase or change of plot size. For using drift experiments results in a mass balance approach (i.e. in PestLCI Consensus), further experiments would be required to validate that the default square field of one hectare that was defined, well represents pesticide drift deposition to off-field surfaces. This approach using a default square field of 1 ha would also be relevant with common LCA functional unit (per surface in ha or per yield in kg/ha) and primary data known by the LCA practitioners (rather the pesticide doses per hectare than the field characteristics). In addition, for the purpose of LCA, these experimentations and drift deposition curve development should report new drift deposition curves using median observed values. As one of the most influent parameters in pesticide emissions modeling (chapter 2), pesticide application methods are associated with pesticide drift losses to off-field surfaces and more drift experiments are required, in particular for tropical crops. Some relevant crop-application method combinations are still missing and require additional experiments to derive drift deposition estimates, such as boom sprayer on pineapple and more generally application methods on banana, pineapple, sugar cane, vegetables and tubers.

In all agricultural contexts, further research and experimental studies are also necessary to assess airborne emissions during pesticide application, because their estimation and modeling are limited (Gil and Sinfort 2005), in particular in tropical regions. To assess pesticide emissions of flooded crops such as rice, specific developments are necessary as recently proposed by (Inao et al. 2018; Tang et al. 2020).

6.2.2 Need to fully adapt rain-related processes in PestLCI

Tropical frequent and intense rainfalls were identified to drive emissions to surface water and groundwater (Figure 7– chapter 1). However, water flows modeling in secondary distribution estimates was initially questioned in the bibliographic review and then along all thesis chapters, and not only for tropical conditions. The following paragraphs present how rain-related processes are considered in the current PestLCI Consensus model (illustrated in Figure 24), and what are the resulting issues. Then, suggestions for improvement are provided.

In PestLCI, initial distribution is followed by the secondary emission processes occurring over few days, and by default within 24 hours after pesticide application (Fantke et al. 2017a). The model uses the occurrence of one rain event (i.e. according to the monthly average rain frequency) as boundary condition for related environmental processes. According to the climate and the month of application, the first rain event after application can occur beyond 1 day and up to several days or even weeks after pesticide application. Consequently, by considering by default one day after pesticide application for secondary emission processes, rain-related processes have generally not yet begun and are omitted. Furthermore, even using several days of simulation, the model considers only one rain event and related processes (i.e. one runoff, one preferential flow) regardless of the water status of the soil and without accounting for any threshold effect. To evaluate these processes, the amount of precipitation per rain event is averaged along all rain events occurring during the month of application, while in tropical conditions, intense rainfall events are responsible for high runoff and leaching leading to pesticide distributions to surface water and groundwater compartments (Sanchez-Bayo and Hyne 2011). By simulating a sufficient number of days for one rain event to occur, the modeling of rain-related processes can result in pesticide losses to surface and groundwater, with either an overestimation (i.e. by considering rain-related processes occurring necessarily after each pesticide application) or an underestimation (i.e. by omitting the high rain intensity responsible for more runoff notably). As discussed in Lammoglia et al. (2016), the use of an average rainfall intensity may be inappropriate to estimate runoff. Furthermore, in the model, water processes are modeled successively, whereas certain water processes occur simultaneously in the soil, such as the infiltration into the soil and the runoff (Alletto et al. 2010).

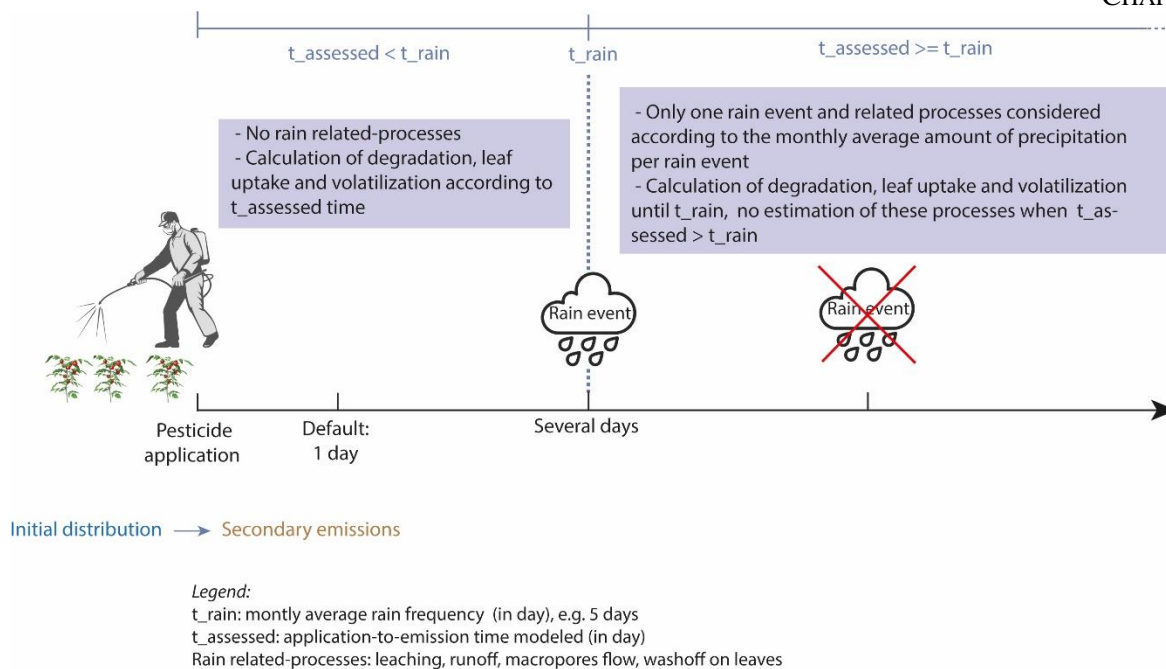


Figure 24: Rain-related processes consideration in current PestLCI Consensus model according to the application-to-modeled time

PestLCI was designed and parametrized for pesticide applications under temperate conditions, consequently certain equations to model water flow processes are parametrized for temperate climate and soil conditions only. For instance, the calculation of the length of a rainfall event in case of a macropore flow to estimate the leached fraction, is based on Danish climate characteristics. Moreover, the field capacity equation was based on temperate soil and was invalid for tropical soils. Therefore, an approach to calculate field capacity considering tropical soil characteristics was proposed and applied in the tomato case study (chapter 3). Details of the approach are presented in the Supplementary Material in S6.1. Furthermore, the current modeling of runoff in PestLCI Consensus (based on Reus et al. (1999)) neither considers hydrologic conditions, crop, field and soil characteristics, nor farming practices such as ground cover. Overall, the water flux modeling in PestLCI Consensus is questioned for tropical conditions, but more generally for all conditions.

A modified framework of PestLCI Consensus with a full rain-based mass balance considering all related processes (run-off, leaching, preferential flow, and even root uptake) could be an interesting development. Furthermore, the approach could consider the averaged processes according to the climatic characteristics related to the month of application, instead of using the average rainfall to estimate rain-related processes. This approach could allow to better estimate rain-related processes in pesticide emission modeling and could avoid the dependency on a minimum simulation time to generate rain-related processes with the apparition of a rain event. To guarantee a consistent use of the model in LCA, the model should not offer any choice in simulation time. Scenarios could be consistently compared to each other according to their own climate

characteristics, regardless of the rain frequency (and related number of days required to have a rain event for a given climate). As part of this rain-based mass balance approach, a more specific runoff estimation could be proposed by using the curve number approach considering the farming practice, the cropping systems, the soil texture and the surface water conditions (Ponce and Hawkins 1996). Furthermore, it is the only approach including tropical cropping system archetypes (i.e. for purely tropical crops such as sugar cane, pineapple, coffee, and for crop archetypes as minor crops (garden or truck crops) and grass (Cooley and Lane 1982; Sartori et al. 2011; USDA 2012). Lastly, the modeling of rain-related processes would make it possible to consider processes occurring with a GCM, which is of particular interest, since GCM can reduce runoff and consequently pesticide transfer to surface water (Reichenberger et al. 2007; Alletto et al. 2010; Mottes et al. 2014).

6.2.3 A full coupling of emission and impact models with secondary emission fractions

Pesticide LCI and LCIA models were designed separately with different system boundaries and considered compartments leading to potential overlaps and gaps between LCI and LCIA phases (van Zelm et al. 2014; Rosenbaum et al. 2015). As a consequence, combining these models in a consistent way constitutes a challenge for practitioners (Fantke 2019). In this thesis (chapter 3), a consistent coupling of these LCI-LCIA models (i.e. PestLCI, dynamiCROP and USEtox) was provided using initial distribution, i.e. pesticide distribution few minutes after application, considering only crop characteristics and application method. Nevertheless, when estimating secondary emission fractions, PestLCI considers some fate processes (e.g. degradation, rain-related processes such as runoff). These potential overlaps with impact assessment should be quantified to propose a complete alignment of fate processes in PestLCI and in USEtox models. A possible way to quantify the importance of overlaps between models' processes would be to make USEtox' fate factor dynamic. This would make it possible to compare the pesticide fate to the environmental compartments within the first days or weeks after pesticide application from USEtox and calculated with the modified framework of rain-related processes in PestLCI. Furthermore, the proper coupling of the secondary emission fractions of PestLCI Consensus with dynamiCROP requires further research also in relation to potential overlaps in plant uptake processes.

6.2.4 Providing tools and guidance for LCA practitioners

According to the previous limitations presented, for current LCA studies where the agricultural stage is in background, to assess pesticide emissions, LCA practitioners might use the initial distribution modeling available on the web-tool or the default initial distribution fractions provided for a set of common crop-

application method pairs in tropical conditions. For LCA studies where the agricultural stage is in foreground and where the purpose is to support eco-design, secondary emission fractions should be used to compare different farming practices, soil and climatic characteristics. To this end, they will need many primary data (i.e. soil and climate characteristics with the month of application, farming practices (e.g. irrigation, tillage, GCM) and the active ingredient) that are not always easy to obtain. These recommendations have been recently presented in a stakeholder's workshop of the OLCA-Pest scientific project to which this thesis was a support. The interest of using secondary emission fractions will be further increased for background LCA applications, when rain-related processes will be properly accounted for and the GCM modeling will be operationalized in the PestLCI web-tool. Concerning the consistent use of dynamiCROP and PestLCI initial distribution fractions, for each pesticide application the dynamiCROP's inputs can be replaced manually by the PestLCI's outputs. A common interface for PestLCI and dynamiCROP would simplify the consistent use of both models and the inclusion of impacts from pesticide residues in crop ingested. Finally, the inclusion of these advances on pesticide emission and impact modeling in the most common LCA software should be considered (e.g. in SimaPro software).

6.3 Additional research needs to improve pesticide assessment in LCA

Future research needs to assess in agricultural LCA pesticide emissions and impacts are presented for tropical conditions and more generally for all agricultural regions.

6.3.1 The inclusion of inorganic pesticide and metabolite emissions and related impacts in LCA

To assess metal-based pesticide emission, currently only initial distributions can be derived (presented in chapter 3), since secondary emissions processes need to be adapted to consider metal speciation and other metal-relevant characteristics (Dong et al. 2014), which might additionally depend on the emission location (Peña et al. 2018). This is of particular concern since metal-based pesticides are authorized and widely used in organic farming, but also in agro-ecological production systems. To properly assess and compare diverse farming cropping systems in LCA, emissions and related-impacts of these inorganic pesticides is required.

Metabolites of active ingredients may be widely found in the environment as a result of the degradation of applied active ingredients (e.g. AMPA metabolite from glyphosate) (Mottes et al. 2017b) and may have even higher toxic effects than their parent compounds (e.g. metabolites of diuron) (Oturán et al. 2008), notably on freshwater ecosystems (van Zelm et al. 2010). However, the current state of knowledge on their emissions and impacts does not allow their inclusion in LCA.

Chemical mixtures and their “cocktails effects” are also omitted in LCA. A framework for environmental risk assessment has been proposed to consider them (Backhaus and Faust 2012) and could constitute a relevant base for their consideration in LCA.

6.3.2 Consideration of all non-target exposure of organisms and humans in LCA

Future models should consider all different exposed populations including workers and bystanders, and all types of pesticide application not only at field plot level, but also at post-harvest and seed treatment levels. Some pesticides exposure pathways in agriculture are omitted in LCA as occupational exposure (when preparing and applying pesticides) (Ingwersen 2012), residential bystanders, and family of exposed workers (Ryberg et al. 2018). A framework for assessing residential bystander exposure to field pesticide applications (potatoes) in LCIA has been recently presented by Ryberg et al. (2018). Further adaptation of this approach is required to include additional tropical crops and application methods, in particular to hand-operated spraying (García-Santos et al. 2016), which is one of the main application methods used in tropical agriculture, particularly for horticulture production (Charlier et al. 2009). As non-applied in the field, post-harvest treatments are generally omitted whereas they can have environmental and human health impact (Bajwa and Sandhu 2014).

Furthermore, ecotoxicity assessment must consider all non-target organisms (i.e. freshwater, marine, and terrestrial organisms, pollinators, and birds). Efforts are required on on-field impacts and missing impact categories including terrestrial and pollinators’ ecotoxicity (Fantke 2019). The impacts of insecticide and fungicide toxicity on pollinating insects were recently modeled for LCA (Crenna et al. 2020), considering new measures of pesticide exposures through ingestion and bee contact. To apply the model to a wider range of pesticides, gaps in data on residues in pollen and nectar need to be filled and more exposure pathways need to be included. Terrestrial ecotoxicity should be considered and advances has already been proposed for metal-based substances (Owsianiak et al. 2015). Finally, specific acute toxicity data with tropical natives species (earthworm) has been presented (Arias-Andrés et al. 2018), as well as different toxicity responses between shrimps from temperate and tropical regions (Daam and Rico 2018). To fully consider tropical species’ sensitivity to ecotoxicity in LCIA, further research is required on indigenous species in tropical conditions and organisms’ sensitivity to pesticides from tropical origins (Lacher and Goldstein 2009).

References of Discussion and Outlook

Alletto L, Coquet Y, Benoit P, et al (2010) Tillage management effects on pesticide fate in soils. A review. *Agron Sustain Dev* 30:367–400. doi.org/10.1051/agro/2009018

- Arias-Andrés M, Rämö R, Torres FM, et al (2018) Lower tier toxicity risk assessment of agriculture pesticides detected on the Río Madre de Dios watershed, Costa Rica. *Environ Sci Pollut Res* 25:13312–13321. doi.org/10.1007/s11356-016-7875-7
- Backhaus T, Faust M (2012) Predictive Environmental Risk Assessment of Chemical Mixtures: A Conceptual Framework. *Environ Sci Technol* 46:2564–2573. doi.org/10.1021/es2034125
- Baio FHR, Antuniassi UR, Castilho BR, et al (2019) Factors affecting aerial spray drift in the Brazilian Cerrado. *PLOS ONE* 14(2):e0212289. doi.org/10.1371/journal.pone.0212289
- Bajwa U, Sandhu KS (2014) Effect of handling and processing on pesticide residues in food- a review. *J Food Sci Technol* 51:201–220. doi.org/10.1007/s13197-011-0499-5
- Charlier J-B, Cattani P, Voltz M, Moussa R (2009) Transport of a nematicide in surface and groundwaters in a tropical volcanic catchment. *J Environ Qual* 38:1031–1041. doi.org/10.2134/jeq2008.0355
- Cooley KR, Lane LJ (1982) Modified runoff curve numbers for sugarcane and pineapple fields in Hawaii. *Journal of Soil and Water Conservation* 37:295–298
- Crenna E, Jolliet O, Collina E, et al (2020) Characterizing honey bee exposure and effects from pesticides for chemical prioritization and life cycle assessment. *Environment International* 138:105642. doi.org/10.1016/j.envint.2020.105642
- Daam MA, Rico A (2018) Freshwater shrimps as sensitive test species for the risk assessment of pesticides in the tropics. *Environ Sci Pollut Res* 25:13235–13243. doi.org/10.1007/s11356-016-7451-1
- Dong Y, Gandhi N, Hauschild MZ (2014) Development of Comparative Toxicity Potentials of 14 cationic metals in freshwater. *Chemosphere* 112:26–33. doi.org/10.1016/j.chemosphere.2014.03.046
- Fantke P (2019) Modeling the environmental impacts of pesticides in agriculture. In: Weidema, B.P. (Ed) *Assessing the environmental impact of agriculture*, Burleigh Dodds Science Publishing. Cambridge, United Kingdom. doi.org/10.19103/AS.2018.0044.08
- Fantke P, Antón A, Grant T, Hayashi K (2017) Pesticide emission quantification for life cycle assessment: A global consensus building process. *J Life Cycle Assess* 13:245–251
- Fantke P, Jolliet O (2016) Life cycle human health impacts of 875 pesticides. *Int J Life Cycle Assess* 21:722–733. doi.org/10.1007/s11367-015-0910-y
- Frischknecht R, Jolliet O (2019) *Global Guidance for Life Cycle Impact Assessment Indicators. Volume 2. United Nations Environment Programme*
- García-Santos G, Feola G, Nuyttens D, Diaz J (2016) Drift from the use of hand-held knapsack pesticide sprayers in Boyacá (Colombian Andes). *J Agric Food Chem* 64:3990–3998. doi.org/10.1021/acs.jafc.5b03772
- Gentil C, Basset-Mens C, Manteaux S, et al (2020) Coupling pesticide emission and toxicity characterization models for LCA: Application to open-field tomato production in Martinique. *Journal of Cleaner Production* 277:124099. doi.org/10.1016/j.jclepro.2020.124099
- Gil Y, Sinfort C (2005) Emission of pesticides to the air during sprayer application: A bibliographic review. *Atmospheric Environment* 39:5183–5193. doi.org/10.1016/j.atmosenv.2005.05.019

- HLPE (2019) Agroecological and other innovative approaches for sustainable agriculture and food systems that enhance food security and nutrition. A report by the High Level Panel of Experts on Food Security and Nutrition of the Committee on World Food Food Security. 163
- Inao K, Iwafune T, Horio T (2018) An improved PADDY model including uptake by rice roots to predict pesticide behavior in paddy fields under nursery-box and submerged applications. *J Pestic Sci* 43:142–152. doi.org/10.1584/jpestics.D17-084
- Ingwersen WW (2012) Life cycle assessment of fresh pineapple from Costa Rica. *J Clean Prod* 35:152–163. doi.org/10.1016/j.jclepro.2012.05.035
- Lacher T, Goldstein M (2009) Tropical ecotoxicology: Status and needs. *Environmental Toxicology and Chemistry* 16:100–111. doi.org/10.1002/etc.5620160111
- Lammoglia S-K, Moeys J, Barriuso E, et al (2016) Sequential use of the STICS crop model and of the MACRO pesticide fate model to simulate pesticides leaching in cropping systems. *Environ Sci Pollut Res* 24:6895–6909. doi.org/10.1007/s11356-016-6842-7
- Malézieux E, Crozat Y, Dupraz C, et al (2009) Mixing plant species in cropping systems: concepts, tools and models. A review. *Agronomy for Sustainable Development* 29:43–62. doi.org/10.1051/agro:2007057
- Meier MS, Stoessel F, Jungbluth N, et al (2015) Environmental impacts of organic and conventional agricultural products – Are the differences captured by life cycle assessment? *J Environ Manag* 149:193–208. doi.org/10.1016/j.jenvman.2014.10.006
- Meier U (2018) Growth stages of mono- and dicotyledonous plants: BBCH Monograph. Open Agrar Repository
- Mottes C, Lesueur Jannoyer M, Le Bail M, et al (2017) Relationships between past and present pesticide applications and pollution at a watershed outlet: The case of a horticultural catchment in Martinique, French West Indies. *Chemosphere* 184:762–773. doi.org/10.1016/j.chemosphere.2017.06.061
- Mottes C, Lesueur-Jannoyer M, Bail ML, Malézieux E (2014) Pesticide transfer models in crop and watershed systems: a review. *Agron Sustain Dev* 34:229–250. doi.org/10.1007/s13593-013-0176-3
- Oturan N, Trajkovska S, Oturan MA, et al (2008) Study of the toxicity of diuron and its metabolites formed in aqueous medium during application of the electrochemical advanced oxidation process “electro-Fenton.” *Chemosphere* 73:1550–1556. doi.org/10.1016/j.chemosphere.2008.07.082
- Owsianiak M, Holm PE, Fantke P, et al (2015) Assessing comparative terrestrial ecotoxicity of Cd, Co, Cu, Ni, Pb, and Zn: The influence of aging and emission source. *Environmental Pollution* 206:400–410. doi.org/10.1016/j.envpol.2015.07.025
- Peña N, Antón A, Kamilaris A, Fantke P (2018) Modeling ecotoxicity impacts in vineyard production: Addressing spatial differentiation for copper fungicides. *Sci Total Environ* 616–617:796–804. doi.org/10.1016/j.scitotenv.2017.10.243
- Pollacco JAP (2008) A generally applicable pedotransfer function that estimates field capacity and permanent wilting point from soil texture and bulk density. *Can J Soil Sci* 88:761–774. doi.org/10.4141/CJSS07120

- Ponce VM, Hawkins RH (1996) Runoff Curve Number: Has It Reached Maturity? *Journal of Hydrologic Engineering* 1:11–19. doi.org/10.1061/(ASCE)1084-0699(1996)1:1(11)
- Reichenberger S, Bach M, Skitschak A, Frede H-G (2007) Mitigation strategies to reduce pesticide inputs into ground- and surface water and their effectiveness; a review. *Sci Total Environ* 384:1–35. doi.org/10.1016/j.scitotenv.2007.04.046
- Reus J, Leendertse P, Bockstaller C (1999) Comparing environmental risk indicators for pesticides: results of the European CAPER project. CLM (Netherlands)
- Rosenbaum RK, Antón A, Bengoa X, et al (2015) The Glasgow consensus on the delineation between pesticide emission inventory and impact assessment for LCA. *Int J Life Cycle Assess* 20:765–776. doi.org/10.1007/s11367-015-0871-1
- Rouault A, Perrin A, Renaud-Gentié C, et al (2020) Using LCA in a participatory eco-design approach in agriculture: the example of vineyard management. *Int J Life Cycle Assess* 25:1368–1383. doi.org/10.1007/s11367-019-01684-w
- Ryberg MW, Rosenbaum RK, Mosqueron L, Fantke P (2018) Addressing bystander exposure to agricultural pesticides in life cycle impact assessment. *Chemosphere* 197:541–549. doi.org/10.1016/j.chemosphere.2018.01.088
- Sanchez-Bayo F, Hyne RV (2011) Comparison of environmental risks of pesticides between tropical and nontropical regions. *Integr Environ Assess Manag* 7:577–586. doi.org/10.1002/ieam.189
- Sartori A, Hawkins RH, Genovez AM (2011) Reference Curve Numbers and Behavior for Sugarcane on Highly Weathered Tropical Soils. *Journal of Irrigation and Drainage Engineering* 137:705–711. doi.org/10.1061/(ASCE)IR.1943-4774.0000354
- Sharma A, Kumar V, Thukral AK, et al (2019) Responses of Plants to Pesticide Toxicity: an Overview. *Planta Daninha* 37:. doi.org/10.1590/s0100-83582019370100065
- Suárez LA (2005) PRZM-3, a model for predicting pesticide and nitrogen fate in the crop root and unsaturated soil zones: User's manual for release 3.12.2., U.S. Environmental Protection Agency, Washington DC, USA
- Tang L, Hayashi K, Inao K, et al (2020) Developing a management-oriented simulation model of pesticide emissions for use in the life cycle assessment of paddy rice cultivation. *Science of The Total Environment* 716:137034. doi.org/10.1016/j.scitotenv.2020.137034
- USDA (2012) NRCS National Engineering Handbook - Part 630 / Hydrology. USDA, National Resource Conservation Service
- van Zelm R, Huijbregts MAJ, van de Meent D (2010) Transformation products in the life cycle impact assessment of chemicals. *Environmental Science & Technology* 44:1004–1009. doi.org/10.1021/es9021014
- van Zelm R, Larrey-Lassalle P, Roux P (2014) Bridging the gap between life cycle inventory and impact assessment for toxicological assessments of pesticides used in crop production. *Chemosphere* 100:175–181. doi.org/10.1016/j.chemosphere.2013.11.037
- Wezel A, Casagrande M, Celette F, et al (2014) Agroecological practices for sustainable agriculture. A review. *Agron Sustain Dev* 34:1–20. doi.org/10.1007/s13593-013-0180-7

INTRODUCTION

Why and how to assess pesticides for agri-LCA in tropical regions?

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CHAPTER 3

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CHAPTER 4

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CHAPTER 5

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CONCLUSION

General conclusion

GENERAL CONCLUSION (IN FRENCH)

La demande en ACV de produits agricoles et alimentaires, notamment en provenance des régions tropicales, est aujourd'hui très forte à la fois pour soutenir l'éco-conception des systèmes de production et pour nourrir l'affichage environnemental. Cependant, la prise en compte des impacts liés aux pesticides n'est pas satisfaisante et constitue un enjeu méthodologique majeur pour permettre la comparaison fiable de systèmes conventionnels, labellisés AB ou simplement dits « agroécologiques ». En effet, l'utilisation des pesticides en agriculture a des impacts sur l'environnement et sur la santé humaine, en particulier dans les régions tropicales. Si des modèles d'estimation des émissions au champ et d'impact de la toxicité humaine et de l'écotoxicité existent aujourd'hui pour l'ACV, ceux-ci demeurent trop peu utilisés, leur usage combiné étant à la fois complexe et partiellement incohérent. L'ambition de cette thèse était de faire progresser la prise en compte des meilleures connaissances disponibles sur les émissions de pesticides en conditions tropicales et la mise en cohérence des modèles d'inventaire et d'impact pour améliorer l'évaluation environnementale par l'ACV des systèmes agricoles tropicaux.

Sur la base d'une revue bibliographique (chapitre 1), les processus et les paramètres qui déterminent les émissions de pesticides et les impacts associés dans les conditions tropicales ont été identifiés, afin d'évaluer dans quelle mesure les modèles actuels dédiés en ACV doivent être adaptés pour mieux prendre en compte ces conditions. Les questions de recherches qui en ont découlées étaient :

- Quels sont les paramètres plus spécifiques de la phase d'inventaire qui déterminent la variabilité des résultats d'émission et d'impact des pesticides ?
- Est-il possible d'évaluer de façon cohérente les émissions et les impacts d'(éco-)toxicité des applications de pesticides en conditions tropicales en ACV en prenant en compte l'impact des résidus de pesticides dans les cultures consommées ?
- Comment quantifier les fractions initiales d'émissions de pesticides pour les systèmes de cultures en conditions tropicales ?
- Comment prendre en compte les pratiques agroécologiques dans la modélisation des émissions de pesticides ?

Tout d'abord, grâce à une analyse de sensibilité du modèle d'émission des pesticides, PestLCI Consensus, nous avons identifié les variables d'entrée qui devaient être définies avec le plus de soin (chapitre 2). Puis, ce travail de thèse a principalement été axé sur : (i) le couplage des modèles d'émission et d'impact des pesticides pour les conditions tropicales en considérant les impacts liés à l'ingestion de résidus de pesticides présents dans la partie récoltée de la culture et (ii) la phase d'inventaire par la modélisation des pesticides utilisés sur des cultures en conditions tropicales incluant les émissions vers la plante.

Un couplage cohérent des modèles d'émission et d'impact des pesticides valable pour tous les contextes agricoles a été proposé (chapitre 3), pour permettre une évaluation opérationnelle des impacts d'(éco-)toxicité des pesticides, incluant la principale voie d'exposition pour la santé humaine, l'ingestion directe de résidus de pesticides dans la partie consommée de la plante. Notre formalisation pour coupler correctement les modèles aidera les praticiens à évaluer de façon cohérente ces impacts dans leurs études ACV. Cela permettra une estimation plus fiable des émissions au champ de pesticides et une prise en compte plus facile des résidus de pesticides dans les cultures dans les études ACV des produits agricoles. La proposition a été testée sur l'ACV de la production de tomate plein champ en Martinique (Antilles françaises) en paramétrant le modèle aux conditions locales. Ce cas d'étude a montré la contribution majeure des émissions des pesticides au champ à l'ensemble des impacts d'écotoxicité eau douce, ainsi que la contribution prépondérante des pesticides liés à l'ingestion de produit récolté sur la toxicité humaine. Cette étude ACV a également montré que malgré une pression phytosanitaire importante localement, les impacts d'(éco-)toxicité par kg de tomate récoltée étaient modérés par rapport aux impacts d'autres tomates plein-champ tropicales grâce au respect par les producteurs des doses homologuées et des délais avant récolte. Cette étude approfondie des impacts d'(éco-)toxicité pour la tomate plein-champ de Martinique a constitué un point fort d'une étude ACV plus globale comparant les systèmes de production de tomate locale plein-champ et sous serre.

La distribution initiale des émissions a ensuite été affinée (chapitre 4) en développant un ensemble cohérent de fractions d'interception foliaire en fonction des caractéristiques de la culture (selon l'indice de surface foliaire) et des techniques de pulvérisation pour les cultures tropicales. De plus, des mesures de dérive due aux méthodes d'application utilisées ont été extraites d'expérimentation spécifiquement menées dans des conditions tropicales, pour une large gamme de méthodes d'application (pulvérisation à rampe et à jet porté, application aérienne et application manuelle) et de cultures (dont café, coton, soja, haricot, papaye). Ainsi, un jeu de fractions d'émission de pesticides a été produit pour les praticiens d'ACV ayant peu d'information sur les pratiques phytosanitaires de la culture étudiée. Certaines méthodes d'application sur des cultures tropicales font encore défaut et nécessitent des expérimentations supplémentaires pour

obtenir des estimations de dérive plus spécifiques, comme le pulvérisateur à rampe sur l'ananas et plus généralement les méthodes d'application sur la banane, l'ananas, la canne à sucre, les légumes et les tubercules.

Dans un contexte de transition agroécologique, la prise en compte de la couverture vivante d'un sol dans le modèle d'émission des pesticides a également été proposée (chapitre 5). Dès son implémentation dans la version en ligne du modèle d'émission PestLCI Consensus, différents systèmes agricoles, tels que l'agriculture conventionnelle, intégrée ou biologique pourront plus facilement être comparés selon leurs pratiques de gestion de la couverture du sol. L'inclusion de couvert tel que le mulch serait un développement intéressant pour étendre la modélisation de la gestion de la couverture du sol. Même si les processus liés à la pluie n'ont pas pu être considérés dans cette thèse, l'approche que nous proposons constitue un point de départ solide pour aborder les pratiques de gestion de la couverture du sol dans les évaluations d'émissions et d'impact associés dans l'ACV. Des premières pistes de travail ont d'ailleurs pu être ébauchées pour une meilleure prise en compte des processus liés à la pluie dans la modélisation des émissions de pesticides, en particulier dans les conditions tropicales.

De manière plus générale, de nombreux défis demeurent pour permettre une évaluation exhaustive des impacts liés aux applications de pesticides en ACV. L'exposition des travailleurs agricoles mais également des riverains demeure non prise en compte en ACV, de même que la modélisation des émissions et impacts des métabolites des pesticides parfois plus dangereux que la substance mère. Un certain nombre d'impacts d'écotoxicité sur des organismes vivants ne sont pas considérés, comme sur les abeilles et les autres organismes terrestres. Une grande partie de ces carences est cependant avant tout due à un manque de connaissances fondamentales sur les mécanismes impliqués.

Les avancées méthodologiques, scientifiques et opérationnelles de la thèse permettent de faciliter dès à présent pour les praticiens d'ACV et les agronomes l'estimation fiable des émissions de pesticides au champ, y compris vers la partie consommée de la plante, et leur conversion cohérente en impacts, afin d'améliorer l'évaluation des performances environnementales des systèmes agroalimentaires par l'ACV, en particulier dans les régions tropicales.

PUBLICATION OF THE THESIS RESULTS

Peer-reviewed publications

- ✓ **Gentil-Sergent C.**, Basset-Mens C., Gaab J., Mottes C., Melero C., Fantke P. **Quantifying pesticide emission fractions for tropical conditions.** *Chemosphere* 275:130014. <https://doi.org/10.1016/j.chemosphere.2021.130014>
- ✓ **Gentil C.**, Basset-Mens C., Manteaux S., Mottes C., Maillard E., Biard Y., Fantke P. 2020 **Coupling pesticide emission and toxicity characterization models for LCA: Application to open-field tomato production in Martinique.** *Journal of Cleaner Production* 277:124099. <http://doi.org/10.1016/j.jclepro.2020.124099>
- ✓ **Gentil C.**, Fantke P., Mottes C., Basset-Mens C. 2020. **Challenges and ways forward in pesticide emission and toxicity characterization modeling for tropical conditions.** *The International Journal of Life Cycle Assessment* 25:1290–1306. <https://doi.org/10.1007/s11367-019-01685-9>
- ✓ Basset-Mens C., Edewa A., **Gentil C.** 2019. **An LCA of French beans from Kenya for decision-makers.** *Indonesian Journal of Life Cycle Assessment and Sustainability* 3(1), 10p.

Presentations in conferences

- ✓ **Gentil C.**, Renaud-Gentié C., Fantke P., Launay A., Basset-Mens C. **Towards consideration of ground cover management in pesticide emission modelling in LCA.** International Conference on Life Cycle Assessment of Food 2020 (LCAfood), 13-16 October 2020. Virtual Conference.
- ✓ **Gentil C.**, Gaab J., Nemecek T., Fantke P., Basset-Mens C. **New insights for PestLCI Consensus model: sensitivity and scenario analysis for pesticide emissions and impacts.** Society of Environmental Toxicology and Chemistry (SETAC) Europe 30th Annual Meeting, 3-7 May 2020. Virtual Conference.
- ✓ Nemecek T., Basset-Mens C., **Gentil C.**, Renaud-Gentié C., Roux P., Peña N., Antón A., Melero C., Fantke P. **Emission and toxicity modelling for pesticides: operationalizing pesticide consensus.** Society of Environmental Toxicology and Chemistry (SETAC) Europe 30th Annual Meeting, 3-7 May 2020. Virtual Conference.
- ✓ **Gentil C.**, Manteaux S., Fantke P., Mottes C., Maillard E., Basset-Mens C. **Modelling pesticide emission and impact in LCA in tropical regions: a case study of tomato production in Martinique (French West Indies).** Conferencia Internacional Analisis de Ciclo de Vida 2019 (CILCA), 15-20 July 2019. Cartago, Costa Rica.
- ✓ **Gentil C.**, Basset-Mens C., Fantke P., Mottes C. **Pesticide emission and toxicity models in LCA need to be adapted for tropical regions.** International Conference on Life Cycle Assessment of Food 2018 (LCAfood), 17-19 October 2018. Bangkok, Thailand.
- ✓ Basset-Mens C., Edewa A., **Gentil C.** **An LCA of French beans from Kenya with a critical analysis of impacts due to pesticide applications.** International Conference on Life Cycle Assessment of Food 2018 (LCAfood), 17-19 October 2018. Bangkok, Thailand.

ANNEXES

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Chapter 2 – Supplementary Material – New insights for PestLCI Consensus model in tropical conditions with a sensitivity analysis

Table S2.1 Definition, range of values or default values and units of input variables of PestLCI Consensus model

Input variable	Definition	Range of value or default value	Unit
Pesticide	Active ingredient in plant protection formulations indicated with CAS number	1070 organic pesticides	-
Crop type	16 crop types are defined according to their characteristics.	e.g. Vegetable fruits, <i>Pooideae</i>	-
Foliar interception fraction	Fraction that the crop leaves can intercept from an applied pesticide amount	[0;1]	kg/kg
Soil type	Soil characteristics and composition (e.g. pH, bulk density)	75 soils from FAO	-
Climate type	Climate characteristics (e.g. average monthly temperature)	30 climates from Köppen-Geiger	-
Month	Month of application	-	-
Application method	Application method associated to a crop type	28 application methods	-
Application drift reduction	Drift reduction factor according to the application method	-	-
Buffer zone, outside the field	Stripe of vegetated land non-harvested on the downside of the treated field, outside the field	Yes/No	-
Buffer zone, inside the field	Stripe of vegetated land non-harvested on the downside of the treated field, inside the field	Yes/No	-
Buffer zone interception fraction	Fraction of the pesticide deposited in the buffer zone that is intercepted by the plant in the buffer zone	0.4	-
Buffer zone width	Width of the buffer zone considered present on the downside of the field	-	m
Field width	Width of the treated field	-	m
Field length	Length of the treated field	-	m
Field slope	Slope of the treated field	-	%
Drainage depth	Depth of drainage tubes or tiles dug down below the field surface	< 1 m	m
Fraction drained	Fraction of the field drained	[0;1]	-
Annual irrigation	Annual irrigation	-	mm
Tillage type	Conventional (sub-soiling), reduced (tillage depth between 0 and 15 cm) and no tillage	-	-
Soil material density	Density of solid materials in soil	2.65	kg/l
Fraction macropores	Fraction of the soil volume which are continuous macropores	0.002	-
Reference soil moisture content	Reference soil moisture content at which soil biodegradation rate has been measured	0.5	-
Response factor for biodegradation rate	Exponential response factor for temperate correction	0.7	-
Q-value	Temperature correction factor to estimate soil biodegradation	2.58	-
Air boundary layer	Thickness of air boundary layer for evaporation	0.00475	m
D(lam)	Thickness of the laminar air boundary layer	0.005	m
A(p,ref)	Reference areic mass of pesticide on plants	0.0001	kg/m ²

Table S2.2 Soil and climate scenarios details

Soil abbreviation	Soil name	Climate abbreviation	Climate name
Nd	Dystric Nitisols	Af ¹	Equatorial rainforest, fully humid
Bk	Calcic Cambisols	Am ¹	Equatorial monsoon
Bv	Vertic Cambisols	As ¹	Equatorial savannah with dry summer
Fh	Humic Ferralsols	Aw ¹	Equatorial savannah with dry winter
Je	Eutric Fluvisols	Cfb ¹	Warm temperate climate, without dry season, warm summer
Q	Arenosols	Dfc ¹	Snow climate, without dry season, cold summer
Th	Humic andosols	SVn ²	South Martinique – Vauclin, closed to Af
Tv	Vitric andosols	SSa ²	South Martinique – Sainte-Anne, closed to Af
Vp	Pellic Vertisols	NPr ²	North Martinique – Prêcheur, closed to Af

Ref: FAOSTAT 2020 *Ref: ¹Kottek et al. 2006; Beck et al. 2018, ²Météo France*

Table S2.3 Details of ecotoxicity and human health characterization factors

Human health characterization factor non-cancer [cases/kg _{emitted}]		Emission to cont. rural air	Emission to cont. freshwater	Emission to cont. natural soil	Emission to cont. agric. Soil
CAS Number	Pesticide				
71751-41-2	Abamectin	1.68E-05	1.81E-05	2.74E-08	1.84E-05
135410-20-7	Acetamiprid	2.31E-06	1.24E-06	1.07E-08	1.59E-07
131860-33-8	Azoxystrobin	6.29E-07	1.01E-06	2.04E-07	7.84E-07
120116-88-3	Cyazofamid	3.45E-07	7.38E-07	1.62E-09	1.66E-08
52918-63-5	Deltamethrin	1.31E-06	7.52E-06	6.21E-09	2.01E-07
155569-91-8	Emamectin benzoate	3.58E-04	3.68E-05	2.65E-05	3.17E-03
77182-82-2	Glufosinate-ammonium	3.07E-06	2.47E-06	1.75E-08	5.68E-08
1071-83-6	Glyphosate	3.99E-08	1.60E-07	3.58E-08	1.16E-07
173584-44-6	Indoxacarb	2.52E-06	3.59E-05	9.99E-09	9.83E-07
91465-08-6	Lambda-cyhalothrin	4.84E-06	1.10E-04	8.30E-09	2.14E-07
8018-01-7	Mancozeb	1.08E-07	2.16E-06	8.04E-08	2.08E-07
108-62-3	Metaldehyde	2.22E-07	3.76E-06	4.05E-07	1.34E-06
123312-89-0	Pymetrozine	5.93E-06	1.33E-05	2.74E-06	7.86E-06
95737-68-1	Pyriproxyen	1.99E-07	2.17E-06	1.14E-09	2.02E-08
168316-95-8	Spinosad	3.67E-06	3.34E-06	2.93E-11	4.77E-09

Ecotoxicity characterization factor [PAF m³ d/kg_{emitted}]		Emission to cont. rural air	Emission to cont. freshwater	Emission to cont. natural soil	Emission to cont. agric. soil
CAS Number	Pesticide				
71751-41-2	Abamectin	1.94E+02	1.01E+05	1.40E+02	1.40E+02
135410-20-7	Acetamiprid	6.07E+01	1.02E+04	8.79E+01	8.79E+01
131860-33-8	Azoxystrobin	1.19E+04	7.70E+04	1.56E+04	1.56E+04
120116-88-3	Cyazofamid	1.85E+02	4.37E+04	5.75E+01	5.75E+01
52918-63-5	Deltamethrin	1.32E+04	3.43E+06	7.96E+02	7.96E+02
155569-91-8	Emamectin benzoate	2.39E+03	6.61E+04	1.06E+04	1.06E+04
77182-82-2	Glufosinate-ammonium	5.95E+00	5.78E+02	4.10E+00	4.10E+00
1071-83-6	Glyphosate	9.59E+00	3.21E+02	7.17E+01	7.18E+01
173584-44-6	Indoxacarb	1.13E+03	1.56E+05	4.33E+01	4.33E+01
91465-08-6	Lambda-cyhalothrin	5.46E+05	1.39E+08	9.29E+03	9.29E+03
8018-01-7	Mancozeb	2.93E+02	5.26E+04	1.96E+03	1.96E+03
108-62-3	Metaldehyde	2.89E+00	2.46E+02	2.52E+01	2.52E+01
123312-89-0	Pymetrozine	8.63E+01	8.07E+02	1.66E+02	2.17E+02
95737-68-1	Pyriproxyfen	4.05E+02	1.90E+05	3.97E+01	3.96E+01
168316-95-8	Spinosad	3.60E+02	1.30E+04	3.49E+03	3.49E+03
3878-19-1	Fuberidazole	2.43E+01	5.75E+03	1.59E+01	3.35E+01

Table S2.4 Input variables having an influence on PestLCI Consensus outputs for the initial distribution and secondary emission fractions

Input variable	Initial distribution				Secondary emissions				
	Air	Off-field surfaces	Field crop	Field soil	Air	Off-field surfaces	Ground-water	Field soil	Degradation and field crop uptake + crop leaf
Pesticide					X	X	X	X	X
Foliar interception fraction			X	X		X	X	X	X
Soil type						X	X	X	X
Climate type						X	X	X	X
Month of application						X	X	X	X
Application method	X	X	X	X	X	X	X	X	X
Application drift reduction	X	X	X	X	X	X	X	X	X
Buffer zone width, outside the field		X	X	X		X	X	X	X
Buffer zone width, inside the field		X	X	X		X	X	X	X
Buffer zone interception fraction, outside the field		X	X	X		X	X	X	X
Buffer zone interception fraction, inside the field		X	X	X		X	X	X	X
Field width		X	X	X		X	X	X	X
Field length									
Field slope						X	X	X	
Drainage depth									
Fraction drained									
Annual irrigation									
Tillage type							X	X	
Soil material density*						X	X	X	X
Fraction macropores*							X	X	
Reference soil moisture content*									
Response factor for biodegradation rate*									
Q-value*						X	X	X	X
Air boundary layer*									
D(lam)*									
A(p,ref)*									

* default parameters (user adjustable)

Chapter 3 – Supplementary Material – Coupling pesticide emission and toxicity characterization models for LCA: Application to open-field tomato production in Martinique

Table S3.1 Model input data for characterization factors' calculation

CAS number	Name	MW (g/mol)	Chemical Class	Kow (L/L)	Henry constant (Pa m ³ /mol)	Pv (Pa)	Solubility (mg/L)	kdeg air (1/s)	kdeg water (1/s)	kdeg sediment (1/s)	kdeg soil (1/s)	kdiss tomato (1/s)
71751-41-2	Abamectin	873.1	neutral	2.5E+04	2.7E-03	3.7E-06	1.2E+00	1.9E-04	3.3E-06	9.0E-08	2.8E-07	N/A
135410-20-7	Acetamiprid	222.68	neutral	6.3E+00	5.3E-08	1.7E-07	3.7E+03	5.7E-05	1.7E-06	1.9E-07	2.8E-06	N/A
120116-88-3	Cyazofamid	324.8	neutral	1.6E+03	4.0E-02	1.3E-05	1.1E-01	3.1E-05	1.0E-06	5.9E-07	1.2E-06	N/A
155569-91-8	Emamectin benzoate	1008.3	neutral	3.0E+05	1.3E-02	4.0E-06	1.1E+02	1.8E-05	9.2E-07	6.7E-08	1.0E-07	N/A
168316-95-8	Spinosyn A: (131929-60-7)	731.98	neutral	4.1E+04	1.9E-07	3.0E-08	1.6E+02	5.8E-04	3.7E-07	3.0E-08	8.0E-06	N/A
168316-95-8	Spinosyn D: (31929-63-0)	746	neutral	5.5E+04	2.3E-05	2.0E-08	7.4E+00	6.1E-04	4.0E-07	1.5E-08	8.0E-06	N/A
173584-44-6	Indoxacarb	527.84	neutral	1.6E+05	4.9E-04	1.6E-08	1.7E-02	3.1E-05	4.5E-08	5.0E-09	4.0E-07	2.3E-06
91465-08-6	Lambda- cyhalothrin	449.86	acid	1.0E+07	1.5E-01	2.0E-07	5.0E-03	2.4E-05	4.5E-08	5.0E-09	3.2E-07	2.4E-06
123312-89-0	Pymetrozine	217.23	amphoter	6.6E-01	1.4E-06	1.8E-06	2.9E+02	7.3E-06	2.1E-07	2.4E-08	2.3E-07	2.4E-06
131860-33-8	Azoxystrobin	403.4	neutral	3.2E+02	7.4E-09	1.1E-10	6.0E+00	3.5E-05	1.3E-07	1.5E-08	4.4E-08	2.0E-06
108-62-3	Metaldehyde	176.21	neutral	1.3E+00	7.1E-02	9.0E-02	2.2E+02	2.2E-05	5.3E-07	5.9E-08	1.6E-06	3.0E-06
52918-63-5	Deltamethrin	505.21	acid	1.6E+06	1.2E-04	2.0E-06	2.0E-03	1.8E-05	1.3E-07	1.5E-08	6.7E-08	1.9E-06

CAS number	Name	avlogEC50	ED50 inhalation, non-cancer	ED50 ingestion, non- cancer	ED50 inhalation, cancer	ED50 ingestion, cancer	BAFfish
		(log(mg/L))	(kg/lifetime)	(kg/lifetime)	(kg/lifetime)	(kg/lifetime)	(L/kg)
71751-41-2	Abamectin	-1.8	1.2E+00	2.9E-01	N/A	N/A	6.9E+01
135410-20-7	Acetamiprid	-0.5	1.1E+01	6.3E+00	N/A	N/A	2.1E+01
120116-88-3	Cyazofamid	-0.94	5.2E+01	4.7E+01	N/A	N/A	2.9E+02
155569-91-8	Emamectin benzoate	-1.1	5.5E+00	5.4E-01	N/A	N/A	8.2E+01
168316-95-8	Spinosyn A: (131929-60-7)	-0.17	4.9E+01	1.0E+01	N/A	N/A	1.1E+02
168316-95-8	Spinosyn D: (31929-63-0)	-0.17	4.9E+01	1.0E+01	N/A	N/A	1.2E+02
173584-44-6	Indoxacarb	-0.72	5.2E+01	2.8E+00	N/A	N/A	7.5E+01
91465-08-6	Lambda-cyhalothrin	-4.5	5.6E-01	2.7E+00	N/A	N/A	3.2E+03
123312-89-0	Pymetrozine	1.4	1.7E+04	3.1E+00	N/A	N/A	9.1E-01
131860-33-8	Azoxystrobin	-0.46	1.1E+01	6.9E+01	N/A	N/A	3.0E+01
108-62-3	Metalddehyde	1.6	3.2E+04	5.0E+00	N/A	N/A	1.0E+00
52918-63-5	Deltamethrin	-2.5	1.1E+01	1.1E+01	N/A	0	3.2E+02

Table S3.2 Mid-point characterization factors for ecotoxicity and human toxicity (* calculated)

Active ingredient	Freshwater ecotoxicity [PAF m ³ d/kgemitted]				Human toxicity, non-cancer [cases/kgemitted]				Human toxicity, cancer [cases/kgemitted] (For all compartments)
	Emission to cont. air	Emission to cont. freshwater	Emission to cont. natural soil	Emission to cont. agricultural soil	Emission to cont. air	Emission to cont. freshwater	Emission to cont. natural soil	Emission to cont. agricultural soil	
Abamectin	1.9E+02*	1.0E+05*	1.4E+02*	1.4E+02*	1.7E-05*	1.8E-05*	2.7E-08	1.8E-05*	N/A
Acetamiprid	6.1E+01*	1.0E+04*	8.8E+01*	8.8E+01*	2.3E-06*	1.2E-06*	1.1E-08	1.6E-07*	N/A
Azoxystrobin	1.2E+04	7.7E+04	1.6E+04	1.6E+04	6.3E-07*	1.0E-06*	2.0E-07	7.8E-07*	N/A
Copper	3.7E+06	9.9E+06	5.2E+06	5.2E+06	3.5E-05	1.4E-07	7.2E-08	1.1E-04	N/A
Cyazofamid	1.8E+02*	4.4E+04*	5.8E+01*	5.8E+01*	3.5E-07*	7.4E-07*	1.6E-09*	1.7E-08*	N/A
Deltamethrin	1.3E+04	3.4E+06	8.0E+02	8.0E+02	1.3E-06	7.5E-06	6.2E-09	2.0E-07	0.0E+00
Emamectin benzoate	2.4E+03*	6.6E+04*	1.1E+04*	1.1E+04*	3.6E-04*	3.7E-05*	2.7E-05*	3.2E-03*	N/A
Glufosinate-ammonium	5.9E+00	5.8E+02	4.1E+00	4.1E+00	3.1E-06	2.5E-06	1.8E-08	5.7E-08	N/A
Glyphosate	9.6E+00	3.2E+02	7.2E+01	7.2E+01	4.0E-08	1.6E-07	3.6E-08	1.2E-07	N/A
Indoxacarb	1.1E+03*	1.6E+05*	4.3E+01*	4.3E+01*	2.5E-06*	3.6E-05*	1.0E-08*	9.8E-07*	N/A
Lambda-cyhalothrin	5.5E+05	1.4E+08	9.3E+03	9.3E+03	4.8E-06*	1.1E-04*	8.3E-09*	2.1E-07*	N/A
Mancozeb	2.9E+02	5.3E+04	2.0E+03	2.0E+03	1.1E-07	2.2E-06	8.0E-08	2.1E-07	N/A
Metaldehyde	2.9E+00	2.5E+02	2.5E+01	2.5E+01	2.2E-07*	3.8E-06*	4.0E-07*	1.3E-06*	N/A
Pymetrozine	8.6E+01	8.1E+02	1.7E+02	2.2E+02	5.9E-06*	1.3E-05*	2.7E-06*	7.9E-06*	N/A
Pyriproxyfen	4.1E+02	1.9E+05	4.0E+01	4.0E+01	2.0E-07	2.2E-06	1.1E-09	2.0E-08	N/A
Spinosad	3.6E+02*	1.3E+04*	3.5E+03*	3.5E+03*	3.7E-06*	3.3E-06*	2.9E-11*	4.8E-09*	N/A

Human toxicity via residues ingestion, non-cancer [cases/kgemitted]				
Active ingredient	Emission to air	Emission to soil	Emission to leaf surface	Emission to fruit surface
Abamectin	7.7E-05*	1.0E-04*	1.4E-07*	1.1E-03*
Acetamiprid	7.9E-05*	2.1E-04*	2.1E-04*	1.2E-03*
Azoxystrobin	8.2E-07*	1.3E-05*	5.4E-07*	4.0E-05*
Copper	N/A	N/A	N/A	N/A
Cyazofamid	2.3E-05*	5.9E-06*	2.4E-05*	6.0E-04*
Deltamethrin	2.7E-05	3.0E-09	7.9E-07	4.6E-04
Emamectin benzoate	1.6E-08*	4.5E-06*	6.0E-14*	9.4E-09*
Glufosinate-ammonium	2.7E-04	2.6E-05	1.9E-05	7.3E-03
Glyphosate	1.2E-07	4.1E-08	2.1E-07	0
Indoxacarb	6.4E-07*	7.4E-07*	1.7E-07*	7.1E-05*
Lambda-cyhalothrin	3.4E-07*	3.6E-10*	4.9E-10*	3.1E-05*
Mancozeb	1.6E-06	1.5E-06	1.1E-05	1.2E-04
Metaldehyde	0	1.1E-05*	1.9E-09*	0
Pymetrozine	1.4E-05*	4.6E-05*	8.8E-06*	6.1E-05*
Pyriproxyfen	1.5E-05	3.0E-08	2.3E-07	1.6E-03
Spinosad	1.2E-05*	2.5E-06*	1.1E-06*	2.2E-03*

S3.3 Metal-based substances impact calculation example

An example is basic copper chloride used as fungicide (CAS: 1332-40-7, molecular formula: $\text{Cl}_2\text{Cu}_4\text{H}_6\text{O}_6$, $MW_p = 427.14$ g/mol), which contains $n_i = 4$ molecules of Cu(II) (CAS: 15158-11-9, $MW_{i \in p} = 63.55$ g/mol ((US EPA 2020))). This gives a mass contribution of Cu(II) on basic copper chloride emission of $(63.55 \text{ g/mol} \times 4)/427.14 \text{ g/mol} = 0.6$.

S3.4 General description of PestLCI and associated assumptions

PestLCI Consensus provides ‘initial distribution fractions’ (i.e. pesticide mass reaching the environment as emissions within minutes after application) for compartments $c \in \{\text{air, field crop surface, field soil surface, off-field surfaces}\}$. Initial distribution fractions are mainly influenced by growth stage and physiology of treated field crops defining the fraction of applied mass that is intercepted by crop surfaces, and by the drift deposition function for a given pesticide application method defining the fraction reaching off-field surfaces. PestLCI Consensus furthermore provides ‘secondary emission fractions’ (i.e. pesticide mass reaching the environment at the end of a defined time, e.g. 5 days after application) for compartments $c \in \{\text{air, field crop surface, field crop leaf uptake, field soil, off-field surfaces}\}$, also considering degradation in field crop and soil. Emission fractions are likewise a function of crop characteristics and application method, but additionally depend on field and pesticide properties.

To run the model, some data are mandatory and some are optional. For the initial distribution, the mandatory data are: crop type, applied fraction intercepted by crop, application method and the optional data are: presence or not of a drift reduction method during application, presence or not of a buffer zone, width of the buffer zone and fraction deposited on buffer zone leaves, field width perpendicular to wind direction. For the secondary distribution, the mandatory data are the same as for the initial distribution plus soil type, climate, month of application, and the optional data are the same as for the initial distribution plus application-to-emission time modeled, application rate of active ingredient (a.i.), field length, field slope, depth of field drainage, fraction of the field which is drained, annual irrigation on the cultivated crop and tillage type.

Table S3.5 Input data for the unit-scenario in PestLCI

Input variable	Values
Name pesticide	Abamectin, acetamiprid, azoxystrobin, copper sulfate, cyazofamid, deltamethrin, emamectin benzoate, glufosinate ammonium, glyphosate, indoxacarb, lambda-cyhalothrin, mancozeb, metaldehyde, pymetrozine, pyriproxyfen, spinosad
Crop type	Vegetables fruit
Fraction intercepted by leaf	According to the stage of development [0.25 ; 0.7]
Climate	Prêcheur, Vauclin, Sainte-Anne
Month of application	All months
Soil type	BV, TV
Application-to-emission time modeled (day)	5
Application method	Knapsack sprayer (García-Santos et al. 2016)
Drift reduction	No
Application rate (kg/ha)	According to the pesticide application
Buffer zone	No
Buffer zone width (m)	According to the plot, [2 ; 3]
Pesticide fraction on leaves in buffer zone (default 0.4)	0.4
Field width (m)	According to the plot, [20 ; 100]
Field length (m)	According to the plot, [17 ; 150]
Slope (%)	According to the plot, [5 ; 25]
Drainage depth (m)	No
Fraction of field drained	No
Annual irrigation (mm)	According to the plot, [0 ; 1280]
Tillage type	Conventional tillage

S3.6 Details of the parameterization and coupling of PestLCI and dynamiCROP

PestLCI

The model was parametrized to the humid tropical conditions of Martinique. First, the climate database was completed with local meteorological data from “Météo France” (<https://donneespubliques.meteofrance.fr/>), using the 3 closest weather stations to the studied plots. The required data are the mean, minimal and maximal temperature (°C) per month, the amount of rainfall (mm) per month, the number of days with >1 mm of rainfall (day), the average rainfall on rainy day per month (mm), the rain frequency per month (day) and the annual potential evaporation (mm). Weather data were mostly averaged between 1992 and 2018. The annual potential evaporation data were not available for the 3 weather stations and came from Reguieg (1986). According to the definition of soil type in Martinique by Colmet-Daage (1969) and Cheignard et al. (1987), the world soil classification from FAO was used (soil profiles available on <http://www.fao.org/soils-portal/en/>). In the FAO classification, the soil called “TV” for Vitric Andosol was selected for the vitric soils of the plots in the

North and the soil called “BV” for Vertic Cambisol for the vertic soils in the South. The required data for each soil layer (up to 1m) are the pH, the fraction of clay, sand, silt and organic carbon, and the soil bulk density. Due to the use of an equation developed in temperate conditions, the calculation of field capacity was causing miscalculations for our local tropical soils (TV and BV). So the field capacity equation was corrected with an equation estimated with tropical soil from Pollacco (2008).

Some active ingredients data were missing in the model’s own database such as e.g. solubility, molecular weight, so the PestLCI pesticide database was completed with data from international database (PPDB, BPDB, AGRITOX, Efsa, HSBDB, SAgE pesticides, PAN pesticide database, Ineris) for 11 active ingredients. Furthermore, the emissions and impacts of a bio-insecticide, *Bacillus thuringiensis*, could not be estimated, due to missing effect factors.

The CropWat model (version 8.0) from FAO (available on <http://www.fao.org/land-water/databases-and-software/cropwat/en/>) was used to estimate the quantity of irrigated water (according to the crop water demand and the quantity of rain) for irrigated plots for which farmers could not provide the amount of water used.

dynamiCROP

The dynamiCROP model was used to estimate the human exposure to pesticide residues in tomato (Fantke et al. 2011a, b). The parametrization of climate and soil data was respectively done using local data from Météo France and FAO soil. Overall, 10 parameters were adapted to the local conditions of tomato crop in Martinique. For the climatic data, it was: mean wind speed at 10m above surface, mean temperature in air, precipitation rate during wet period (intensity of rain event), interval between 2 consecutive rains, average relative humidity in air and annual average precipitation rate. For soil data, it was: pH, soil organic carbon content and depth of soil. Except for copper-sulfate (inorganic substance) and *Bacillus thuringiensis* (bacterial insecticide), for each unit-scenario the active ingredient, the applied mass per m², the dates of planting, harvesting, application and fruit growth beginning (15 days after plantation) and the yield were entered into the model, with a food processing factor for washing (corresponding to a simulation of raw eaten tomato, washed with water).

The dynamiCROP model version that is adapted for LCA, calculating all results factors per kg emitted into different environmental compartments, is further described and can be requested from <http://dynamicrop.org/model.php>.

S3.7 System studied

The study has been carried out in Martinique, a small tropical volcanic island in the Caribbean ocean (1128 km²) with a maritime tropical climate. Temperatures vary slightly during the year with monthly averages between 25 and 28 °C. The relative humidity is high at 70-80% and rainfalls are abundant, with annual rainfall average between 970 mm and over 6000 mm (MétéoFrance). These conditions are favorable to pests (e.g. in tomato production: *Trialeurodes vaporariorum*, *Ralstonia solanacearum*) and

weeds. In Martinique in 2018, based on local experts' advice, we estimated that 6 000 tonnes of tomatoes were marketed. Among these 6000 tonnes consumed locally, local production supplied 1800 tonnes including 2/3 produced in 4 greenhouse farms (1100 tonnes) and 1/3 in open-field farms (700 tonnes) by around 100 farmers. In open-field systems, tomatoes (mainly variety Heatmaster) are produced in small plots (in our sample, between 0.04 to 1.5 ha) with market gardening production including intercropping systems and sometimes even livestock.

Primary field data were collected in 6 sampled farms distributed as follows, 3 in the North Caribbean (Municipality of Le Prêcheur) and 3 in the South Atlantic (Municipalities of Vauclin and Sainte-Anne) for the years 2017 and 2018. These two regions of production feature by contrasted soil and climate conditions. In this small sample of 6 farms, 6 plots with one production cycle each.

Table S3.8 List of active ingredients (a.i.) and quantity applied

CAS number	Pesticide Name	Target	Amount a.i. applied per unit harvest [kg a.i./kg tomato]	Amount a.i. applied per unit area [kg a.i./ha]
71751-41-2	Abamectin	Insecticide	[6.6E-11 ; 4.2E-07]	[6.6E-07 ; 2.2E-02]
135410-20-7	Acetamiprid	Insecticide	[6.7E-07 ; 3.3E-06]	[1.3E-02 ; 1.0E-01]
131860-33-8	Azoxystrobin	Fungicide	[9.1E-10 ; 1.8E-05]	[9.1E-06 ; 2.5E-01]
7758-99-8	Copper sulfate	Fungicide	[7.3E-06 ; 1.5E-04]	[5.3E-02 ; 4.0E+00]
120116-88-3	Cyazofamid	Fungicide	[2.0E-06 ; 2.0E-06]	[8.0E-02 ; 8.0E-02]
52918-63-5	Deltamethrin	Insecticide	[5.0E-08 ; 3.0E-07]	[1.0E-03 ; 1.2E-02]
155569-91-8	Emamectin benzoate	Insecticide	[2.4E-07 ; 2.4E-07]	[8.4E-03 ; 8.4E-03]
77182-82-2	Glufosinate-ammonium	Herbicide	[1.5E-05 ; 3.0E-05]	[3.0E-01 ; 3.0E-01]
1071-83-6	Glyphosate	Herbicide	[4.0E-06 ; 7.2E-05]	[9.6E-02 ; 2.9E+00]
173584-44-6	Indoxacarb	Insecticide	[1.0E-06 ; 2.0E-06]	[2.0E-02 ; 2.0E-02]
91465-08-6	Lambda-cyhalothrin	Insecticide	[3.6E-10 ; 6.7E-07]	[3.6E-06 ; 1.3E-02]
7775-09-9	Mancozeb	Fungicide	[2.9E-09 ; 3.2E-05]	[2.9E-05 ; 2.9E-01]
108-62-3	Metaldehyde	Molluscicide	[3.8E-05 ; 3.8E-05]	[1.5E+00 ; 1.5E+00]
123312-89-0	Pymetrozine	Insecticide	[2.5E-06 ; 7.5E-06]	[5.0E-02 ; 2.0E-01]
95737-68-1	Pyriproxyfen	Insecticide	[6.7E-07 ; 1.7E-06]	[1.3E-02 ; 1.7E-02]
168316-95-8	Spinosad	Insecticide	[2.4E-06 ; 2.4E-06]	[8.5E-02 ; 8.5E-02]

In the 6 tomato farms surveyed, a variety of pesticides were used: 10 insecticides, 4 fungicides, 2 herbicides and 1 molluscicide, to control a wide range of pests and diseases (whiteflies, mites, caterpillars, mildew, powdery mildew etc.). Each farmer has his own pest management scheme and most of them use mixtures of several active ingredients (up to 4 a.i.), in particular a combination of one insecticide and one fungicide.

Table S3.9 Sensitivity analysis – PestLCI emission fractions – heat maps

The three tables present the sum of the freshwater ecotoxicity (PAF m³ d/kg tomato harvested) and human toxicity (cases/kg tomato harvested) for each scenario (A to F) and each active ingredient using the initial distribution fractions of PestLCI (1), the secondary emission fractions of PestLCI (2) and the hypothesis of 100% of pesticide emitted to agricultural soil (3). The color gradient from lightest to red classifies by scenario the active ingredients with the least impact towards the most impact in cases/kg tomato harvest.

The active ingredients with the highest (eco-)toxicity per scenario are nearly the same according to the approach chosen, with copper as the most toxic substance in the scenarios where it is used.

For all scenarios, freshwater ecotoxicity and human toxicity impacts are overestimated with the hypothesis of 100% of pesticide emitted to agricultural soil than using PestLCI distribution fractions. Then variation between initial and secondary distribution are less significant due to high characterization factors uncertainties.

1 ↓ Substance Scenario →	cases/kg tomato (initial distribution)						PAF m ³ d/kg tomato (initial distribution)					
	A	B	C	D	E	F	A	B	C	D	E	F
abamectin		2.6E-12	1.2E-13	2.5E-12	2.0E-12	3.3E-15		2.6E-05	2.2E-06	3.0E-05	2.6E-05	3.4E-08
acetamiprid		8.4E-13		3.1E-13				1.9E-04		6.0E-05		
azoxystrobin		9.4E-12	2.1E-12	1.2E-12		2.0E-15		1.9E-01	4.5E-02	2.5E-02		4.0E-05
copper (sulphate) - Cu(II)	8.0E-11	3.4E-10		2.2E-10			3.9E+00	1.7E+01		1.1E+01		
cyazofamid			4.5E-14						1.2E-04			
deltamethrin	3.8E-14	2.9E-14	7.2E-14	3.0E-14			8.8E-04	3.1E-04	1.9E-03	3.7E-04		
emamectin benzoate					3.6E-10						1.3E-03	
glufosinate-ammonium		1.2E-11						2.5E-04				
glyphosate	1.5E-12		1.3E-11		6.3E-12		9.5E-04		8.3E-03		4.0E-03	
indoxacarb		8.2E-13						1.9E-04				
lambda-cyhalothrin		5.7E-13		1.0E-13		7.4E-16		8.9E-02		1.5E-02		1.2E-04
mancozeb	1.5E-11	3.1E-12				1.6E-15	1.5E-01	2.8E-02				1.5E-05
metaldehyde			7.5E-11						1.4E-03			
pymetrozine	3.0E-11	2.0E-11	3.5E-11	1.6E-11			8.4E-04	5.4E-04	9.9E-04	4.2E-04		
pyriproxyfen		4.8E-14						1.9E-04				
spinosad					9.0E-13						5.6E-03	
Total/scenario	1.3E-10	3.9E-10	1.3E-10	2.4E-10	3.6E-10	7.7E-15	4.1E+00	1.7E+01	5.8E-02	1.1E+01	1.1E-02	1.7E-04
Total/scenario (organic subst.)	2.7E-07	5.0E-07	6.7E-08	6.6E-09	1.1E-08	4.9E-11	1.5E-01	3.1E-01	5.8E-02	4.1E-02	1.1E-02	1.7E-04
Total/scenario (metal-based subst.)	8.0E-11	3.4E-10	0.000E+00	2.2E-10	0.0E+00	0.0E+00	3.9E+00	1.7E+01	0.0E+00	1.1E+01	0.0E+00	0.0E+00
Field emissions (organic subst.)	4.7E-11	5.0E-11	1.3E-10	2.0E-11	3.6E-10	7.7E-15						
Crop residues (organic subst.)	2.7E-07	5.0E-07	6.6E-08	6.6E-09	1.1E-08	4.9E-11						

2 ↓ Substance Scenario →	cases/kg tomato (secondary emissions)						PAF m ³ d/kg tomato (secondary emissions)					
	A	B	C	D	E	F	A	B	C	D	E	F
abamectin		4.3E-12	2.0E-13	4.4E-12	1.7E-12	4.1E-15		4.0E-05	2.8E-06	4.5E-05	2.5E-05	4.2E-08
acetamiprid		6.9E-13		2.3E-13				1.2E-04		1.8E-05		
azoxystrobin		1.8E-11	3.2E-12	1.7E-12		2.4E-15		3.6E-01	6.7E-02	3.4E-02		4.8E-05
copper (sulphate) - Cu(II)	8.0E-11	3.4E-10		2.2E-10			3.9E+00	1.7E+01		1.1E+01		
cyazofamid			4.2E-14						1.1E-04			
deltamethrin	3.5E-14	2.7E-14	6.9E-14	2.6E-14			8.6E-04	3.0E-04	1.9E-03	3.6E-04		
emamectin benzoate					3.6E-10						1.3E-03	
glufosinate-ammonium		1.2E-11						2.2E-04				
glyphosate	1.2E-12		1.2E-11		3.3E-12		7.6E-04		7.4E-03		2.1E-03	
indoxacarb		1.1E-12						2.2E-04				
lambda-cyhalothrin		5.6E-13		9.3E-14		6.9E-16		8.9E-02		1.4E-02		1.1E-04
mancozeb	1.3E-11	2.9E-12				1.6E-15	1.3E-01	3.1E-02				2.1E-05
metaldehyde			5.5E-11						1.0E-03			
pymetrozine	2.8E-11	2.0E-11	3.5E-11	1.4E-11			7.8E-04	5.5E-04	9.6E-04	3.7E-04		
pyriproxyfen		4.1E-14						1.8E-04				
spinosad					9.2E-13						8.6E-03	
Total/scenario	1.2E-10	4.0E-10	1.0E-10	2.4E-10	3.7E-10	8.7E-15	4.0E+00	1.8E+01	7.8E-02	1.1E+01	1.2E-02	1.8E-04
Total/scenario (organic subst.)	2.7E-07	5.0E-07	6.7E-08	6.6E-09	1.1E-08	4.9E-11	1.4E-01	4.8E-01	7.8E-02	4.9E-02	1.2E-02	1.8E-04
Total/scenario (metal-based subst.)	8.0E-11	3.4E-10	0.0E+00	2.2E-10	0.0E+00	0.0E+00	3.9E+00	1.7E+01	0.0E+00	1.1E+01	0.0E+00	0.0E+00
Field emissions (organic subst.)	4.3E-11	5.9E-11	1.0E-10	2.0E-11	3.7E-10	8.7E-15						
Crop residues (organic subst.)	2.7E-07	5.0E-07	6.6E-08	6.6E-09	1.1E-08	4.9E-11						

3 ↓ Substance Scenario →	cases/kg tomato (100% soil)						PAF m ³ d/kg tomato (100% soil)					
	A	B	C	D	E	F	A	B	C	D	E	F
abamectin		5.8E-12	2.8E-13	7.7E-12	2.8E-12	6.9E-15		4.4E-05	2.1E-06	5.8E-05	2.1E-05	5.2E-08
acetamiprid		7.4E-13		3.1E-13				4.1E-04		1.7E-04		
azoxystrobin		2.9E-11	4.9E-12	3.8E-12		4.1E-15		5.7E-01	9.8E-02	7.5E-02		8.1E-05
copper (sulphate) - Cu(II)	9.3E-10	5.3E-09		2.2E-09			4.3E+01	2.4E+02		1.0E+02		
cyazofamid			3.3E-14						1.2E-04			
deltamethrin	6.1E-14	4.0E-14	1.1E-13	4.8E-14			2.4E-04	1.6E-04	4.5E-04	1.9E-04		
emamectin benzoate					7.6E-10						2.5E-03	
glufosinate-ammonium		3.4E-12						2.5E-04				
glyphosate	1.7E-12		1.5E-11		7.0E-12		1.0E-03		9.1E-03		4.3E-03	
indoxacarb		2.0E-12						8.7E-05				
lambda-cyhalothrin		3.6E-13		5.2E-14		4.5E-16		1.5E-02		2.2E-03		1.9E-05
mancozeb	3.3E-11	6.1E-12				3.5E-15	3.1E-01	5.7E-02				3.3E-05
metaldehyde			1.0E-10						1.9E-03			
pymetrozine	5.9E-11	3.9E-11	8.8E-11	3.0E-11			1.6E-03	1.1E-03	2.4E-03	8.3E-04		
pyriproxyfen		6.1E-14						1.2E-04				
spinosad					2.3E-14						1.7E-02	
Total/scenario	1.0E-09	5.4E-09	2.1E-10	2.3E-09	7.7E-10	1.5E-14	4.3E+01	2.5E+02	1.1E-01	1.0E+02	2.4E-02	1.3E-04
Total/scenario (organic subst.)	2.7E-07	5.0E-07	6.7E-08	6.7E-09	1.2E-08	4.9E-11	3.2E-01	6.5E-01	1.1E-01	7.9E-02	2.4E-02	1.3E-04
Total/scenario (metal-based subst.)	9.3E-10	5.3E-09	0.0E+00	2.2E-09	0.0E+00	0.0E+00	4.3E+01	2.4E+02	0.0E+00	1.0E+02	0.0E+00	0.0E+00
Field emissions (organic subst.)	9.4E-11	8.6E-11	2.1E-10	4.2E-11	7.7E-10	1.5E-14						
Crop residues (organic subst.)	2.7E-07	5.0E-07	6.6E-08	6.6E-09	1.1E-08	4.9E-11						

Table S3.10 Sensitivity analysis – dynamiCROP - heat maps

The two tables present the sum of human toxicity due to pesticide residues (cases/kg tomato harvested) for each scenario (A to F) and each active ingredient using the dynamiCROP standard version (1) and dynamiCROP adapted version using initial distribution fractions and parametrized to local tropical conditions (2). The color gradient from lightest to red classifies by scenario the substances with the least impact towards the most impact in cases/kg tomato harvest.

The active ingredients with the highest (eco-)toxicity per scenario are nearly the same according to the approach chosen, with various active ingredients with the highest impact per scenario.

Parametrizing to local conditions and using the initial distribution fractions of PestLCI show up to 2 order of magnitude higher impacts from crop residues in cases/kg tomato harvested than the standard version of dynamiCROP using generic distribution fractions and default climate and soil data.

1 ↓ Substance	Scenario→	cases/kg tomato (dynamiCROP standard version)					
		A	B	C	D	E	F
abamectin			4.0E-11	2.6E-11	7.2E-11	5.4E-11	2.9E-13
acetamiprid			2.9E-10		4.9E-10		
azoxystrobin			4.3E-10	1.8E-10	7.5E-11		3.0E-14
cyazofamid				2.8E-10			
deltamethrin		5.2E-11	1.3E-12	1.5E-10	2.9E-11		
emamectin benzoate						2.1E-11	
glufosinate-ammonium			1.9E-08				
glyphosate		1.3E-11		2.9E-11		6.1E-11	
indoxacarb			2.8E-12				
lambda-cyhalothrin			8.3E-10		9.3E-15		7.1E-13
mancozeb		5.1E-09	3.7E-11				4.1E-13
metaldehyde				5.4E-10			
pymetrozine		5.0E-12	1.9E-12	1.3E-09	1.2E-11		
pyriproxyfen			1.5E-10				
spinosad						5.5E-09	
Total/scenario		5.2E-09	2.1E-08	2.5E-09	6.8E-10	5.7E-09	1.4E-12

2 ↓ Substance	Scenario→	cases/kg tomato (dynamiCROP adapted version)					
		A	B	C	D	E	F
abamectin			1.9E-09	3.5E-10	7.1E-10	1.8E-11	2.1E-11
acetamiprid			1.1E-08		5.4E-09		
azoxystrobin			1.1E-08	9.4E-10	2.4E-10		1.1E-12
copper (sulphate) - Cu(II)							
deltamethrin		2.5E-09	4.2E-11	1.4E-09	1.2E-10		
emamectin benzoate						1.1E-12	
glufosinate-ammonium			4.6E-07				
glyphosate		4.9E-10		5.8E-11		2.6E-12	
indoxacarb			1.4E-10				
lambda-cyhalothrin			5.6E-09		9.1E-16		6.9E-12
mancozeb		2.7E-07	1.3E-09				2.1E-11
metaldehyde				8.4E-10			
pymetrozine		7.4E-10	6.3E-11	6.2E-08	1.8E-10		
pyriproxyfen			4.9E-09				
spinosad						2.5E-09	
Total/scenario		2.7E-07	5.0E-07	6.6E-08	6.6E-09	2.5E-09	4.9E-11

Table S3.11 Synthetic review of LCA studies on tomato considering pesticide emissions and impacts

Reference	Type of production and localization	LCI approach	LCIA approach	Distinction between organic and metal-based substances	LCI and LCIA adapted to tropical conditions	Pesticide crop residues assessment
Our LCA study	Open-field (OF) - Martinique	PestLCI consensus	USEtox	Yes	LCI: Yes LCIA: No (can be done)	Yes, dynamiCROP
Bojacá et al. 2014	Greenhouse (GH) - Colombia	Model from Anton et al 2004 for GH	USES-LCA	No	LCI- LCIA: No	No
Khoshnevisan et al. 2014	GH - Iran	Fixed emission to air according to (Sahle and Potting 2008), ecoinvent	USES-LCA	No	LCI- LCIA: No	No
Payen et al. 2015	GH - Morocco	100% soil, ecoinvent	USES-LCA	No	LCI- LCIA: No	No
Romero-Gómez et al. 2017	OF, GH, greenhouse - Spain	Fixed emission to air according to bibliography	USEtox	No	LCI: No LCIA: No (can be done)	No
Zarei et al. 2019	OF, GH – Iran	Fixed emission to air according to (Sahle and Potting 2008), ecoinvent	USES-LCA	No	LCI- LCIA: No	No
Ingrao et al 2019	GH – Italy	ecoinvent	Impact 2002	No	LCI- LCIA: No	No
Basset-Mens et al 2016	OF, GH - Rwanda	100% soil, ecoinvent	USEtox	No	LCI: No LCIA: No (can be done)	No
Perrin et al 2015	OF - Morocco	No (TFI calculation)	No	No	No	No

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- Aktar MdW, Sengupta D, Chowdhury A (2009) Impact of pesticides use in agriculture: their benefits and hazards. *Interdiscip Toxicol* 2:1–12. doi.org/10.2478/v10102-009-0001-7
- Arias-Estévez M, López-Periago E, Martínez-Carballo E, et al (2008) The mobility and degradation of pesticides in soils and the pollution of groundwater resources. *Agri Ecosyst Environ* 123:247–260. doi.org/10.1016/j.agee.2007.07.011
- Chevignard T, Feller C, Andreux F, Quantin P (1987) Le “remodelage” des terres en Martinique : modification des propriétés de “ferrisols” et d’andosols cultivés en canne à sucre. *Cahiers ORSTOM Série Pédologie* 23:223–236
- Colmet-Daage F (1969) Carte pédologique de la Martinique 1/20 000
- Daam MA, van den Brink PJ (2010) Implications of differences between temperate and tropical freshwater ecosystems for the ecological risk assessment of pesticides. *Ecotoxicology* 19:24–37. doi.org/10.1007/s10646-009-0402-6
- Dijkman TJ, Birkved M, Hauschild MZ (2012) PestLCI 2.0: a second generation model for estimating emissions of pesticides from arable land in LCA. *Int J Life Cycle Assess* 17:973–986. doi.org/10.1007/s11367-012-0439-2
- Fantke P (2019) Modeling the environmental impacts of pesticides in agriculture. In: Weidema, B.P. (Ed) *Assessing the environmental impact of agriculture*, Burleigh Dodds Science Publishing, Cambridge, United Kingdom. doi.org/10.19103/AS.2018.0044.08
- Fantke P, Bijster M, Guignard C, et al (2017) USEtox® 2.0 Documentation (Version1)
- Fantke P, Charles R, de Alencastro LF, et al (2011a) Plant uptake of pesticides and human health: Dynamic modeling of residues in wheat and ingestion intake. *Chemosphere* 85:1639–1647. doi.org/10.1016/j.chemosphere.2011.08.030
- Fantke P, Juraske R, Antón A, et al (2011b) Dynamic multicrop model to characterize impacts of pesticides in food. *Environ Sci Technol* 45:8842–8849. doi.org/10.1021/es201989d
- García-Santos G, Feola G, Nuyttens D, Diaz J (2016) Drift from the use of hand-held knapsack pesticide sprayers in Boyacá (Colombian Andes). *J Agric Food Chem* 64:3990–3998. doi.org/10.1021/acs.jafc.5b03772
- Lesueur Jannoyer M, Cattan P, Woignier T, Clostre F (2016) *Crisis management of chronic pollution: contaminated soil and human health*, 1st Edition. CRC Press
- Lewis SE, Silburn DM, Kookana RS, Shaw M (2016) Pesticide behavior, fate, and effects in the tropics: an overview of the current state of knowledge. *J Agric Food Chem* 64:3917–3924. doi.org/10.1021/acs.jafc.6b01320
- Mottes C, Lesueur Jannoyer M, Le Bail M, et al (2017) Relationships between past and present pesticide applications and pollution at a watershed outlet: The case of a horticultural catchment in Martinique, French West Indies. *Chemosphere* 184:762–773. doi.org/10.1016/j.chemosphere.2017.06.061
- Pollacco JAP (2008) A generally applicable pedotransfer function that estimates field capacity and permanent wilting point from soil texture and bulk density. *Can J Soil Sci* 88:761–774. doi.org/10.4141/CJSS07120

Racke KD, Skidmore MW, Hamilton DJ, et al (1997) Pesticides report 38. Pesticide fate in tropical soils - (Technical report). *Pure Appl Chem* 69:1349–1371. doi.org/10.1351/pac199769061349

Reguieg (1986) L'évapotranspiration et bilan hydrique en Martinique. Document Météo-France

Rosenbaum RK, Bachmann TM, Gold LS, et al (2008) USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment. *Int J Life Cycle Assess* 13:532. doi.org/10.1007/s11367-008-0038-4

US EPA (2020) Chemistry Dashboard | Home. <https://comptox.epa.gov/dashboard>. Accessed 26 Mar 2020

Chapter 4 – Supplementary Material – Quantifying pesticide emission fractions for tropical conditions

Table S4.1 Leaf area index (LAI) by crop and growth stages with references

Crop / crop class	Stage	Details on stage	LAI	Reference
Banana	0	1-2 months	0.2	Mottes 2013
Banana	1	2-3 months	1.89	
Banana	2	4-6 months	3.77	
Sugar cane	0	1-1.5 months	0.3	
Sugar cane	1	2-3 months	3	
Sugar cane	2	> 6months	6	
Pineapple	0	2-3 months	0.4	
Pineapple	1	4 months	4.3	
Pineapple	2	9 months - 2 years	8.6	
Vegetable fruits	0		0.2	
Vegetable fruits	1		2	
Vegetable fruits	2		4	
Tropical fruit trees	0		0.5	
Tropical fruit trees	1		5	
Tropical fruit trees	2		10	
Tubers	0		0.1	
Tubers	1		1.5	
Tubers	2		3	
Soybean	0		0.3	Haboudane et al. 2004
Soybean	1	Development of the grain	1.95	da Cunha et al. 2017
Soybean	2	Fully developed pods and beginning of grain filling	4.1	Bueno et al. 2017
Cotton	1	Early bloom	1.6	Pettigrew 2004
Cotton	2	Late bloom	3.29	
Coffee	0	<18 months	0.425	Favarin et al. 2002
Coffee	1	[1.5; 3] years	1.552	
Coffee	2	> 3 years	3.895	
Bean	0		0.3	Haboudane et al. 2004
Bean	1		1.9	Bueno et al. 2016
Bean	2	Filling pods and maturation	4	

Bibliographic references table S4.1

Alves GS, da Cunha JPAR (2014) Field data and prediction models of pesticide spray drift on coffee crop. *Pesquisa Agropecuária Brasileira* 49:622–629. doi.org/10.1590/S0100-204X2014000800006

- Bueno MR, Cunha JPAR da, Santana DG de, et al (2016) Drift curves from spray applications on common bean crop. *Ciência e Agrotecnologia* 40:621–632. doi.org/10.1590/1413-70542016406016716
- Bueno MR, da Cunha JPAR, de Santana DG (2017) Assessment of spray drift from pesticide applications in soybean crops. *Biosyst Eng* 154:35–45. doi.org/10.1016/j.biosystemseng.2016.10.017
- da Cunha JPAR, Barizon RRM, Ferracini VL, Assalin MR (2017) Spray drift and caterpillar and stink bug control from applications with electrostatic charge and atomizer on soybean crop. *ENGENHARIA AGRÍCOLA* 37:1163–1170. doi.org/10.1590/1809-4430-Eng.Agric.v37n6p1163-1170/2017
- Favarin JL, Dourado Neto D, García y García A, et al (2002) Equações para a estimativa do índice de área foliar do cafeeiro. *Pesq agropec bras* 37:769–773. doi.org/10.1590/S0100-204X2002000600005
- Haboudane D, Miller JR, Pattey E, et al (2004) Hyperspectral vegetation indices and novel algorithms for predicting green LAI of crop canopies: Modeling and validation in the context of precision agriculture. *Remote Sensing of Environment* 90:337–352. doi.org/10.1016/j.rse.2003.12.013
- Mottes C (2013) Evaluation des effets des systèmes de culture sur l'exposition aux pesticides des eaux à l'exutoire d'un bassin versant. Proposition d'une méthodologie d'analyse appliquée au cas de l'horticulture en Martinique. AgroParisTech
- Pettigrew WT (2004) Physiological Consequences of Moisture Deficit Stress in Cotton. *Crop Science* 44:1265–1272. doi.org/10.2135/cropsci2004.1265

Table S4.2 Bibliographic research on drift deposition under tropical conditions

	KEYWORDS	PUBLICATIONS
#1	TS=(drift)	145 198
#2	TS=(air or aerial or sediment* or spray)	1 548 954
#3	TS=(pesticide* or insecticide* or herbicide* or fungicide* or acaracide* or nematocide*)	227 031
#4	TS=(tropic* or "southern?afrika" or "south?afrika" or botswana or lesotho or namibia or swaziland or libya or "east?afrika" or djibouti or ethiopia or eritrea or sudan or somalia or kenya or rwanda or uganda or burundi or tanzania or malawi or zambia or mozambique or zimbabwe or madagascar or "reunion?island" or mauritius or mayotte or seychelles or comoros or "central?afrika" or angola or cameroon or "central?african?republic" or centrafrique or congo or gabon or "equatorial?guinea" or "sao?tome" or chad or zaire or "west?afrika" or benin or "burkina?faso" or ghana or mali or mauritania or niger or senegal or togo or "western?sahara" or "cape?verde" or gambia or guinea or "guinea?bissau" or liberia or nigeria or "sierra?leone" or "south?asia" or bangladesh or india or maldives or pakistan or "sri?lanka" or "south?east?asia" or brunei or burma or cambodia or lao or vietnam or "viet nam" or thailand or kampuchea or indonesia or bali or java or sumatra or timur or singapore or malaysia or philippines or "far?east?asia" or china or "hong?kong" or macao or taiwan or caribbean or "anguilla?island" or antigua or bahamas or barbados or cuba or dominica or "dominican?republic" or grenada or guadeloupe or haiti or jamaica or martinique or montserrat or "netherlands?antilles" or "puerto?rico" or "santa?lucia" or "trinidad?and?tobago" or aruba or bonaire or curaco or "turks?and?caicos" or "leeward?islands" or "virgin?islands" or "windward?islands" or "cayman?islands" or "central?america" or belize or "costa?rica" or "el?salvador" or guatemala or honduras or nicaragua or panama or mexico or "south?america" or bolivia or brazil or colombia or ecuador or guyana or "french?guyana" or paraguay or peru or surinam or venezuela or australia or "papua?new?guinea" or malanesia or micronesia or polynesia or "new?caledonia" or fiji or vanuatu or tonga or samoa or tahiti or "salomon?islands" or "gambier?islands" or "society?islands" or "wallis?and?futuna")	2 736 119
#5	#1 AND #2 AND #3 AND #4	84

Table S4.3 Considered drift deposition functions for comparisons to those selected in tropical conditions to quantify drift-related pesticide emissions in LCA with their lower and upper validity limits

Application method	Equation	Data used	R²	Lower validity limit [m]	Upper validity limit [m]	Reference
Boom sprayer - standard flat fan	$f(x) = 0.9681x^{(-0.951)}$	Medians	n/a	1	100	Ganzelmeier and Rautmann 2000
Boom sprayer - standard flat fan - field crops that are > 20 cm	$f(x) = 176.568e^{(-1.8551x)} + 1.7010e^{(-0.1420x)}$	Medians	n/a	1	100	Ganzelmeier and Rautmann 2000; Holterman and van de Zande 2003
Air blast sprayer - perennial crop - late stage (in leaf)	$f(x) = 59.5405e^{(-1.0524x)} + 6.7072e^{(-0.1379x)}$	Medians	n/a	3	50	Holterman and van de Zande 2003
Air blast sprayer	$f(x) = 55e^{(-3.4341x)} + 12.9178e^{(-0.1556x)}$	Medians	n/a	3	50	Ganzelmeier and Rautmann 2000
Knapsack sprayer	$f(x) = 29e^{(-6.82x)} + 18.35e^{(-0.44x)}$	Means	n/a	0.5	20	Garcia-Santos et al 2016

Table S4.4 Experimental protocol of the drift deposition curves selected

Crop	Application method	Experimental protocol: collectors	Experimental protocol: treatment	Reliability of measurements	Type of results	Reference
Cotton	Knapsack sprayer	Collectors (cotton patches) at the ground, at 1 m and 1.5 m high	Extraction in water with colorimeter to dose out the tartrazine	Non-compliance with ISO standard and 5 replications	Calculation of drift percentage and drift deposition curves with the mean	Gouda et al. 2018
Papaya (early and late stage)	Turbo fan driven sprayer with 12 nozzles	PEP bottles filled with distilled water are the collector: located on the ground at 0, 2, 5, 10, 20 and 30 m from the edge of the field. 9 pesticides applications were done at different stages of the crop development and other vegetation in the sampled area was removed in order to avoid interception	Extraction and gas chromatography of chlorothalonil, chloropyrifos and malathion	Non-compliance with ISO 22866 standard and 3 replications	Drift deposition curves (% according to the distance) + 90th percentile of the drift percentage for each application method	Hernández-Hernández et al. 2007
Bean	Boom sprayer with fat fan deflector standard and 4 nozzle types for fine, medium, coarse and extremely coarse droplets	20 parallel distances outside of the area spaced at 2.5 m from 2.5 to 50 m from the target area	Extraction of the rhodamine B fluorescent and use of a fluorimeter	Compliance with ISO 22866 standard and 10 replications	Calculation of drift percentage and drift deposition curves with the mean and 90 th percentile	Bueno et al. 2017
Soybean	Aerial sprayer, rotary atomizer with blades at 55° and 65°, hydraulic nozzle with deflector at 30° and 90°	Collectors: 2 mm nylon strings extended for 2 m vertically, placed at 20, 40, 80, 160 and 320 m downwind from application area, counted from the borderline of each plot, with 4 replications for each	Quantification by the amount of thiamethoxam on strings analyzed by means of liquid chromatography	Non-compliance with ISO 22866 standard and 4 replications	Calculation of drift percentage and drift deposition curves with the mean	Bueno et al. 2016
Coffee	Air blast sprayer with a hollow cone with and without air induction	Vertical polyamide yarns at the external sampling points + horizontal rectangular glass plates at the internal sampling points. 2 external collectors positioned at 1.8 m from the ground and spaced at 10 m apart. Placed at distance, 10, 50, 200, 500, 1200 and 2000 m. totally 60 collectors in each application	Extraction of the rhodamine B fluorescent and use of a fluorimeter	Non-compliance with ISO 22866 standard and 25 replications		Alves and da Cunha 2014
Soybean	Aerial sprayer, electrostatic spray system and rotary atomizer					da Cunha et al. 2017a
Soybean	Aerial sprayer, 8 rotary atomizers					da Cunha et al. 2017b
Millet, sorghum, soybean, corn						Baio et al. 2019

Table S4.5 Emission fractions to air (f_{air}), off-field surfaces (f_{dep}), crop leaves ($f_{\text{field} \rightarrow \text{crop}}$) and to field soil ($f_{\text{field} \rightarrow \text{soil}}$) for a set of combinations of crop – application method – growth stage - spraying technique and field width and length

Crop	Application method, nozzle type, crop on which the drift deposition curve was developed	Growth stage	Spraying technique	Foliar interception fraction	Field width (m)	Field length (m)	f_{air}	f_{dep}	$f_{\text{field} \rightarrow \text{crop}}$	$f_{\text{field} \rightarrow \text{soil}}$
Bean	Boom/flat-fan deflector, medium nozzle, Bean	0	Application over the canopy	0.201	100	100	0.10	0.0273	0.1754	0.6973
Bean	Boom/flat-fan, fine nozzle, Bean	0	Application over the canopy	0.201	100	100	0.10	0.0288	0.1751	0.6961
Bean	Boom/flat-fan deflector, medium nozzle, Bean	1	Application over the canopy	0.820	100	100	0.10	0.0273	0.7154	0.1573
Bean	Boom/flat-fan, fine nozzle, Bean	1	Application over the canopy	0.820	100	100	0.10	0.0288	0.7142	0.1571
Bean	Boom/flat-fan deflector, medium nozzle, Bean	2	Application over the canopy	1.000	100	100	0.10	0.0273	0.8727	0.0000
Bean	Boom/flat-fan, fine nozzle, Bean	2	Application over the canopy	1.000	100	100	0.10	0.0288	0.8712	0.0000
Soybean	Boom/flat-fan deflector, medium nozzle, Soybean	0	Application over the canopy	0.201	100	100	0.10	0.0256	0.1758	0.6987
Soybean	Boom/flat-fan, fine nozzle, Soybean	0	Application over the canopy	0.201	100	100	0.10	0.0256	0.1758	0.6986
Soybean	Boom/flat-fan, air induction, coarse nozzle, Soybean	0	Application over the canopy	0.201	100	100	0.10	0.0254	0.1758	0.6988
Soybean	Boom/flat-fan deflector, medium nozzle, Soybean	1	Application over the canopy	0.831	100	100	0.10	0.0256	0.7265	0.1479
Soybean	Boom/flat-fan, fine nozzle, Soybean	1	Application over the canopy	0.831	100	100	0.10	0.0256	0.7265	0.1479
Soybean	Boom/flat-fan, air induction, coarse nozzle, Soybean	1	Application over the canopy	0.831	100	100	0.10	0.0254	0.7266	0.1479
Soybean	Boom/flat-fan deflector, medium nozzle, Soybean	2	Application over the canopy	1.000	100	100	0.10	0.0256	0.8744	0.0000
Soybean	Boom/flat-fan, fine nozzle, Soybean	2	Application over the canopy	1.000	100	100	0.10	0.0256	0.8744	0.0000
Soybean	Boom/flat-fan, air induction, coarse nozzle, Soybean	2	Application over the canopy	1.000	100	100	0.10	0.0254	0.8746	0.0000
Soybean	Aerial Nozzle 90°, Soybean	0	Application over the canopy	0.201	100	100	0.25	0.2040	0.1098	0.4362
Soybean	Aerial Nozzle 30°, Soybean	0	Application over the canopy	0.201	100	100	0.25	0.2019	0.1102	0.4380
Soybean	Aerial Atomizer 65°, Soybean	0	Application over the canopy	0.201	100	100	0.25	0.2003	0.1105	0.4392
Soybean	Aerial Atomizer 55°, Soybean	0	Application over the canopy	0.201	100	100	0.25	0.2015	0.1103	0.4382
Soybean	Aerial electrostatic system, Soybean	0	Application over the canopy	0.201	100	100	0.25	0.2022	0.1101	0.4377
Soybean	Aerial rotary atomizer 55°, Soybean	0	Application over the canopy	0.201	100	100	0.25	0.2027	0.1100	0.4373
Soybean	Aerial rotary atomizer, Soybean, millet, corn, cotton, sorghum	0	Application over the canopy	0.201	100	100	0.25	0.1030	0.1301	0.5170
Soybean	Aerial Nozzle 90°, Soybean	1	Application over the canopy	0.831	100	100	0.25	0.2040	0.4536	0.0924
Soybean	Aerial Nozzle 30°, Soybean	1	Application over the canopy	0.831	100	100	0.25	0.2019	0.4554	0.0927
Soybean	Aerial Atomizer 65°, Soybean	1	Application over the canopy	0.831	100	100	0.25	0.2003	0.4567	0.0930
Soybean	Aerial Atomizer 55°, Soybean	1	Application over the canopy	0.831	100	100	0.25	0.2015	0.4557	0.0928
Soybean	Aerial electrostatic system, Soybean	1	Application over the canopy	0.831	100	100	0.25	0.2022	0.4551	0.0927
Soybean	Aerial rotary atomizer 55°, Soybean	1	Application over the canopy	0.831	100	100	0.25	0.2027	0.4548	0.0926

Crop	Application method, nozzle type, crop on which the drift deposition curve was developed	Growth stage	Spraying technique	Foliar interception fraction	Field width (m)	Field length (m)	f_{air}	f_{dep}	$f_{field \rightarrow crop}$	$f_{field \rightarrow soil}$
Soybean	Aerial rotary atomizer, Soybean, millet, corn, cotton, sorghum	1	Application over the canopy	0.831	100	100	0.25	0.1030	0.5376	0.1094
Soybean	Aerial Nozzle 90°, Soybean	2	Application over the canopy	1.000	100	100	0.25	0.2040	0.5460	0.000
Soybean	Aerial Nozzle 30°, Soybean	2	Application over the canopy	1.000	100	100	0.25	0.2019	0.5481	0.000
Soybean	Aerial Atomizer 65°, Soybean	2	Application over the canopy	1.000	100	100	0.25	0.2003	0.5497	0.000
Soybean	Aerial Atomizer 55°, Soybean	2	Application over the canopy	1.000	100	100	0.25	0.2015	0.5485	0.000
Soybean	Aerial electrostatic system, Soybean	2	Application over the canopy	1.000	100	100	0.25	0.2022	0.5478	0.000
Soybean	Aerial rotary atomizer 55°, Soybean	2	Application over the canopy	1.000	100	100	0.25	0.2027	0.5473	0.000
Soybean	Aerial rotary atomizer, Soybean, millet, corn, cotton, sorghum	2	Application over the canopy	1.000	100	100	0.25	0.1030	0.6470	0.000
Cotton	Aerial rotary atomizer, Soybean, millet, corn, cotton, sorghum	1	Application over the canopy	0.745	100	100	0.25	0.1030	0.4821	0.1649
Cotton	Aerial rotary atomizer, Soybean, millet, corn, cotton, sorghum	2	Application over the canopy	1.000	100	100	0.25	0.1030	0.6470	0.0000
Cotton	Knapsack (Spraying at 1 m), Cotton	1	Plant-by-plant application	0.950	100	100	0.06	0.0077	0.8857	0.0466
Cotton	Knapsack (Spraying at 1.5 m), Cotton	1	Plant-by-plant application	0.950	100	100	0.06	0.0063	0.8870	0.0467
Cotton	Centrifugal cane ultra-low volume (spraying at 1 m), Cotton	1	Plant-by-plant application	0.950	100	100	0.06	0.0142	0.8795	0.0463
Cotton	Centrifugal cane ultra-low volume (spraying at 1.5 m), Cotton	1	Plant-by-plant application	0.950	100	100	0.06	0.0212	0.8728	0.0459
Cotton	Knapsack (Spraying at 1 m), Cotton	1	Application under the canopy	0.037	100	100	0.06	0.0077	0.0347	0.8976
Cotton	Knapsack (Spraying at 1.5 m), Cotton	1	Application under the canopy	0.037	100	100	0.06	0.0063	0.0348	0.8989
Cotton	Centrifugal cane ultra-low volume (spraying at 1 m), Cotton	1	Application under the canopy	0.037	100	100	0.06	0.0142	0.0345	0.8913
Cotton	Centrifugal cane ultra-low volume (spraying at 1.5 m), Cotton	1	Application under the canopy	0.037	100	100	0.06	0.0212	0.0342	0.8845
Cotton	Knapsack (Spraying at 1 m), Cotton	2	Application under the canopy	0.050	100	100	0.06	0.0077	0.0466	0.8857
Cotton	Knapsack (Spraying at 1.5 m), Cotton	2	Application under the canopy	0.050	100	100	0.06	0.0063	0.0467	0.8870
Cotton	Centrifugal cane ultra-low volume (spraying at 1 m), Cotton	2	Application under the canopy	0.050	100	100	0.06	0.0142	0.0463	0.8795
Cotton	Centrifugal cane ultra-low volume (spraying at 1.5 m), Cotton	2	Application under the canopy	0.050	100	100	0.06	0.0212	0.0459	0.8728
Cotton	Knapsack (Spraying at 1 m), Cotton	1	Soil application	0	100	100	0	0	0	1

Crop	Application method, nozzle type, crop on which the drift deposition curve was developed	Growth stage	Spraying technique	Foliar interception fraction	Field width (m)	Field length (m)	f_{air}	f_{dep}	$f_{field \rightarrow crop}$	$f_{field \rightarrow soil}$
Cotton	Knapsack (Spraying at 1.5 m), Cotton	1	Soil application	0	100	100	0	0	0	1
Cotton	Centrifugal cane ultra-low volume (spraying at 1 m), Cotton	1	Soil application	0	100	100	0	0	0	1
Cotton	Centrifugal cane ultra-low volume (spraying at 1.5 m), Cotton	1	Soil application	0	100	100	0	0	0	1
Coffee	Air blast/without air induction, nozzle ATR, Coffee	0	Application over the canopy	0.274	100	100	0.10	0.0288	0.2390	0.6322
Coffee	Air blast/air induction, nozzle TVI, Coffee	0	Application over the canopy	0.274	100	100	0.10	0.0274	0.2394	0.6332
Coffee	Air blast/without air induction, nozzle ATR, Coffee	1	Application over the canopy	0.732	100	100	0.10	0.0288	0.6376	0.2336
Coffee	Air blast/air induction, nozzle TVI, Coffee	1	Application over the canopy	0.732	100	100	0.10	0.0274	0.6386	0.2339
Coffee	Air blast/without air induction, nozzle ATR, Coffee	2	Application over the canopy	1.000	100	100	0.10	0.0288	0.8712	0.0000
Coffee	Air blast/air induction, nozzle TVI, Coffee	2	Application over the canopy	1.000	100	100	0.10	0.0274	0.8726	0.0000
Tropical fruit trees	Air blast/turbo fan, early stage, 90%ile, Papaya	0	Application over the canopy	0.316	100	100	0.20	0.0519	0.2362	0.5119
Tropical fruit trees	Air blast/turbo fan, late stage, 90%ile, Papaya	1	Application over the canopy	1.000	100	100	0.08	0.0821	0.8379	0.0000

Chapter 5 – Supplementary Material – Introducing ground cover management in pesticide emission modeling

Table S5.1 Pesticide characteristics for emission modeling

Cas number	Active ingredient	a.i. target	MW (g/mol)	Solubility (mg/L)	Vapor pressure (Pa)	Chemical category	Log Kow (-)	KOC (L/kg)
7775-09-9	mancozeb	fungicide	212.4	2.8E+01	1.0E-05	acid	1.3	5.5E+02
95737-68-1	pyriproxyfen	insecticide	321.4	1.7E-04	2.9E-04	base	5.4	1.9E+04

Cas number	DT50 soil (d)	DT50 <i>Pooideae</i> (d)	DT50 <i>Panicoideae</i> (d)	DT50 <i>Fabaceae</i> (d)	DT50 vegetable fruits (d)	DT50 grapevine (d)
7775-09-9	18	3.1	8.7	2.6	4.1	6.3
95737-68-1	4.2	18.2	51.3	15.5	24.0	37.2

Table S5.2 Mid-point characterization factor for freshwater ecotoxicity [PAF m3 d/kgemitted]

Active ingredient	Emission to continental rural air	Emission to continental freshwater	Emission to continental natural soil	Emission to continental agricultural soil
Mancozeb	2.90E+02	5.30E+04	2.00E+03	2.00E+03
Pyriproxyfen	4.10E+02	1.90E+05	4.00E+01	4.00E+01

Table S5.3 Dissipation half-lives on three crop families: *Pooideae*, *Panicoideae* and *Fabaceae* according to the review of Fantke and Juraske (2013)

	<i>Pooideae</i>	<i>Panicoideae</i>	<i>Fabaceae</i>
All crop components			
Number of data points	186	62	335
Median half-life [d]	4.56	2.68	3.60
2.5%-ile half-life [d]	0.30	0.38	0.67
97.5%-ile half-life [d]	32.97	10.49	20.98
Leaf			
Number of data points	96	34	146
Median half-life [d]	5.00	2.54	3.50
2.5%-ile half-life [d]	1.37	0.34	0.70
97.5%-ile half-life [d]	24.98	6.24	25.88

Chapter 6 – Supplementary Material – Discussion and outlook

S6.1 Field capacity extension to tropical soil

The field capacity (FC_s) equation was based on PRZM modeling (Suárez 2005) using coefficients data defined by Rawls for non-tropical, -organic and -volcanic soils, leading to aberrant values for tropical soils. For the grapevine case study, the field capacity was consistently calculated according to PRZM modeling, but not for the tomato case study in tropical conditions.

The field capacity calculation for tomato case study was modified using an equation adapted from Pollacco (2008) for tropical soils and was also previously implemented in Gentil et al. (2020), to allow the consideration of the tropical soil characteristics (fraction of clay, sand, silt, organic matter, and the bulk density). The field capacity of the vitric andosol (TV), FC_s (g g^{-1}) was calculated using the fitting parameters selected from “Vitric” soil in the pedotransfer function (PFT) model 4 in Pollacco (2008):

$$FC_s = SAT_s \left[P_{\min} + (P_{\max} - P_{\min}) \times f_{\text{clay}}^{(P_{\text{clay}} + P_{\rho} \times SAT_s^2)} \right] \times e^{\left(\frac{P_{\text{sand}} \times f_{\text{sand}}^3}{SAT_s} \right)}$$

where SAT_s (g g^{-1}) is the saturated gravimetric moisture content of soil s (here TV), P_{\min} , P_{\max} , P_{clay} , P_{sand} , and P_{ρ} are dimensionless empirical fitting parameters (respectively, 0.371, 1.000, 0.563, 0.187, -0.030), f_{clay} (g g^{-1}) is the fraction of clay in soil and f_{sand} (g g^{-1}) is the fraction of sand in soil.

The field capacity obtained for the vitric andosol in the tomato case in Martinique is: 0.281 (g g^{-1}).

We proposed to use the equation of Pollacco et al (2008) and the corresponding fitting parameters to calculate the field capacity equation according to soil characteristics (i.e. vitric, humic, tropical and typical as defined in Pollacco et al (2008)), allowing an extension of the model to tropical soils.

References S6.1

- Gentil C, Basset-Mens C, Manteaux S, et al (2020) Coupling pesticide emission and toxicity characterization models for LCA: Application to open-field tomato production in Martinique. *Journal of Cleaner Production* 277:124099. doi.org/10.1016/j.jclepro.2020.124099
- Pollacco JAP (2008) A generally applicable pedotransfer function that estimates field capacity and permanent wilting point from soil texture and bulk density. *Can J Soil Sci* 88:761–774. doi.org/10.4141/CJSS07120
- Suárez LA (2005) PRZM-3, a model for predicting pesticide and nitrogen fate in the crop root and unsaturated soil zones: User’s manual for release 3.12.2., U.S. Environmental Protection Agency, Washington DC, USA

ABBREVIATIONS LIST

CF	Characterization Factor
DALY	Disability Adjusted Life Years
EF	Effect Factor
FAI	Fruit Area Index
FAO	Food and Agricultural Organization of the United Nations
FF	Fate Factor
FU	Functional Unit
GCM	Ground Cover Management
GC	Ground Cover
hF	Harvest Fraction
iF	Intake Factor
IS	Impact Score
ISO	International Organization of Standardization
KOC	Organic carbon affinity
LAI	Leaf Area Index
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
MAD	Mean Absolute Deviation
MRL	Maximum Residue Limit
MW	Molecular Weight
OC	Organic Carbon
PAF	Potentially Affected Fraction
PF	Food Processing Factor
SDG	Sustainable Development Goals
ULV	Ultra-Low Volume
XF	Exposure Factor

ABSTRACT

The use of pesticides in agriculture leads to environmental and human health impacts, particularly in tropical regions where pedoclimatic conditions favorable to pests and diseases, encourage their use all year round. Life Cycle Assessment (LCA), an ISO-standardized methodology is widely applied to quantify the environmental performance of agri-food systems but generally do not account properly for impacts due to pesticide applications. A bibliographic review identified the characteristics that determine pesticide emissions and related impacts under tropical conditions, and assessed to what extent Life Cycle Inventory (LCI) and Life Cycle Impact Assessment (LCIA) models need to be adapted to better account for these conditions. The main goal of the thesis was to advance emission and impact modeling for agricultural pesticides under tropical conditions for improving scientific foundation of the environmental evaluation of tropical agri-food systems. First, we identified with a sensitivity analysis on the pesticide emission model PestLCI Consensus, aspects that needed be developed in priority. Then, the initial emission distribution was refined by developing a consistent set of foliar interception fractions as function of crop characteristics and spraying techniques for crops grown under tropical conditions. In addition, based on a literature review, measures of drift from application methods were extracted from experimentations specifically conducted in tropical conditions. Thanks to these developments, a consistent set of pesticide emission fractions were provided for direct use by LCA practitioners. A consistent coupling of pesticide LCI-LCIA models was proposed for all agricultural contexts including human toxicity due to exposure to pesticide residues in crops. The proposition was tested successfully on an LCA of an open-field tomato produced in Martinique (French West Indies) by parametrizing the model to the local conditions. Moreover, to evaluate the environmental sustainability of agricultural practices a framework was proposed to account for ground cover management in the modeling of pesticide emissions. These methodological, scientific and operational advances constitute an important step toward a more reliable modeling of field pesticide emissions, including emission to the consumed part of the crop, and their consistent conversion into (eco)-toxicity impacts, as part of LCA for agri-food systems in tropical regions. To improve further the assessment of agricultural pesticides in LCA, especially for tropical conditions, a better consideration of rain-related processes in pesticide emission model is needed.

Keywords: Active ingredient, models, tropical crops, farming practices, (eco-)toxicity, LCA

RESUME

L'utilisation des pesticides en agriculture a des impacts sur l'environnement et la santé humaine, en particulier dans les régions tropicales où les conditions pédoclimatiques favorables aux maladies et aux bio-agresseurs, motivent leur utilisation tout au long de l'année. L'Analyse du Cycle de Vie (ACV), une méthodologie normalisée par l'ISO, est largement utilisée pour quantifier la performance environnementale des systèmes agroalimentaires mais la prise en compte des impacts liés aux pesticides y représente un défi majeur. Une analyse bibliographique a permis d'identifier les caractéristiques qui déterminent les émissions de pesticides et leurs impacts associés dans les conditions tropicales, et d'évaluer dans quelle mesure les modèles d'inventaire et d'évaluation de l'impact doivent être adaptés pour mieux tenir compte de ces conditions. Le principal objectif de cette thèse était d'aller vers une meilleure modélisation des émissions et impacts des pesticides agricoles dans les conditions tropicales afin d'améliorer les bases scientifiques de l'évaluation environnementale des systèmes agroalimentaires tropicaux. Tout d'abord, grâce à une analyse de sensibilité du modèle d'émission des pesticides PestLCI Consensus, nous avons identifié les aspects qui devraient être développés en priorité. Ensuite, la distribution initiale des émissions a été affinée en développant un ensemble cohérent de fractions d'interception foliaire en fonction des caractéristiques de la culture et des techniques de pulvérisation pour les cultures tropicales. De plus, des mesures de la dérive due aux méthodes d'application utilisées ont été extraites d'expérimentation spécifiquement menées dans des conditions tropicales. Ainsi, un jeu de fractions d'émission de pesticides a été produit pour une utilisation directe par les praticiens de l'ACV. Un couplage cohérent des modèles d'émission et d'impact des pesticides a été proposé pour tous les contextes agricoles, incluant la toxicité humaine due à l'exposition aux résidus de pesticides dans les produits récoltés. La proposition a été testée de façon satisfaisante sur l'ACV de la production de tomate plein champ en Martinique (Antilles françaises) en paramétrant le modèle aux conditions locales. De plus, pour évaluer la durabilité environnementale des pratiques agricoles, la modélisation de la gestion de la couverture du sol dans le modèle d'émissions des pesticides a également été proposée. Ces progrès méthodologiques, scientifiques et opérationnels constituent une étape importante vers une modélisation plus fiable des émissions au champ de pesticides, y compris l'émission dans la partie consommée de la culture, et leur conversion cohérente en impacts d'(éco)toxicité, dans le cadre de l'ACV des systèmes agroalimentaires des régions tropicales. Pour améliorer encore l'évaluation des pesticides agricoles dans le cadre de l'ACV, en particulier pour les conditions tropicales, il est nécessaire de mieux prendre en compte les processus liés à la pluie dans le modèle d'émission des pesticides.

Mot clés : substance active, modèles, cultures tropicales, pratiques agricoles, (éco-)toxicité, ACV