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# 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.



## 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., 2017), 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,

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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 S-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., 2017). 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., 2020), 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.

## 2. Materials and methods

### 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 S-1 and S-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).

### 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, (impact/functional unit), is calculated as:

$$IS = \sum_{p,c} (m_{\mathrm{emi},p,c} \times CF_{p,c}) \tag{1}$$

where $m_{\mathrm{emi},p,c}$ ($\mathrm{kg_{emitted}}$/FU) is the total emitted mass of pesticide $p$ from the tomato production into a given environmental compartment $c$, and $CF_{p,c}$ (impact/$\mathrm{kg_{emitted}}$) 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, $m_{\mathrm{app},p}$ ($\mathrm{kg_{applied}}$/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, $mf_{p,c}$ ($\mathrm{kg_{emitted}}$/$\mathrm{kg_{applied}}$):

$$m_{\mathrm{emi},p,c} = m_{\mathrm{app},p} \times mf_{p,c} \tag{2}$$

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:

$$m_{\mathrm{emi},p,c} = m_{\mathrm{app},p} \times mf_{p,c} \times \frac{MW_{i \in p} \times n_i}{MW_p} \tag{3}$$

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 S-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. $mf_{p,c} = 1$ for $c =$ field soil, and $mf_{p,c} = 0$ 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., 2017); details are presented in SM, Section S-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 $\sum_c mf_{p,c} = 1$ for any given pesticide. Emission input data are detailed in SM (Section S-5).

Characterization factors, $CF_{p,c}$ (impact/$\mathrm{kg_{emitted}}$), 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:

$$CF_{p,c} = FF_{p,c} \times XF_{p,c} \times EF_p = iF_{p,c} \times EF_p \tag{4}$$

where $FF_{p,c}$ ($\mathrm{kg_{in\ compartment}}$ per $\mathrm{kg_{emitted}}$/d) is the fate factor denoting the increase in pesticide mass in compartment $c$ for an emission into any compartment, $XF_{p,c}$ ($\mathrm{kg_{intake}}$/d per $\mathrm{kg_{in\ compartment}}$ or $\mathrm{kg_{dissolved}}$/$\mathrm{kg_{in\ compartment}}$) 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/$\mathrm{kg_{intake}}$ or impact/$\mathrm{kg_{dissolved}}$) is the effect factor finally relating exposure to impacts. For human toxicity, fate and exposure factors can be summarized into human population intake fractions, $iF_{p,c}$ ($\mathrm{kg_{intake}}$/d per $\mathrm{kg_{emitted}}$/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 Fig. 1). 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. (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:

$$CF_{p,c} = hF_{p,c}(t) \times PF_f \times EF_p = iF_{p,c} \times EF_p \tag{5}$$

where $hF_{p,c}(t)$ ($\mathrm{kg_{in\ crop\ harvest}}$/$\mathrm{kg_{emitted}}$) 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, $PF_f$ ($\mathrm{kg_{in\ processed\ food}}$/$\mathrm{kg_{in\ crop\ harvest}}$) is a residue reduction factor due to food processing step $f$ (e.g. washing, cooking), and $EF_p$ (impact/$\mathrm{kg_{intake\ of\ processed\ food}}$) is the human toxicity effect factor as defined in eq. (4). Harvest fraction and food processing factor can be combined into

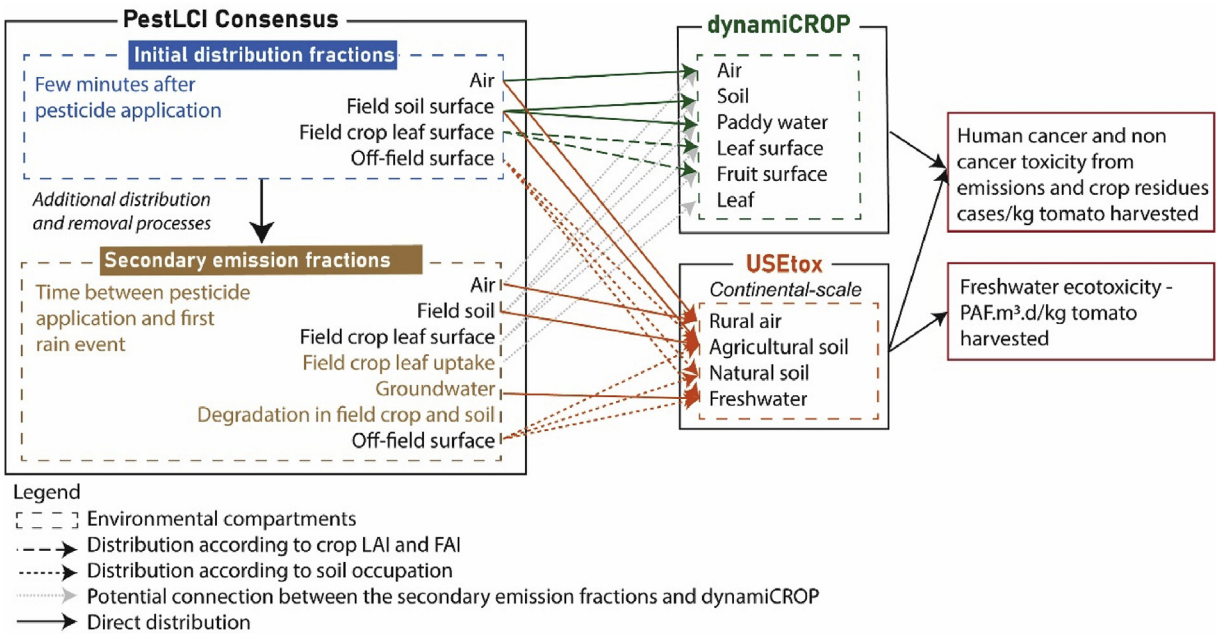


**Fig. 1.** Connection of the emission compartments of PestLCI to those of dynamiCROP and USEtox according to the pesticide mass applied per kg of the harvested crop; adapted from Fantke (2019).

residue-related intake fractions, $iF_{p,c}$ ($kg_{intake}/kg_{emitted}$), consistent with intake fractions from USEtox (see eq. (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. (1). We therefore adapted $hF_{p,c}(t)$ to relate to initial distributions and emissions, thereby consistently coupling dynamiCROP with PestLCI Consensus results:

$$hF_{p,c}(t) = \frac{\sum_h m_{\mathrm{res},p,h}(t)}{m_{\mathrm{app},p} \times mf_{p,c}} = \frac{\sum_h m_{\mathrm{res},p,h}(t)}{m_{\mathrm{emi},p,c}} \tag{6}$$

where $m_{\mathrm{res},p,h}(t)$ ($kg_{\text{in crop harvest}}/FU$) 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 $m_{\mathrm{res},p,h}(t)$, we realized the adaptation by transforming the input mass vector (i.e. $m_{\mathrm{emi},p,c}$ 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 $m_{\mathrm{res},p,h}(t)$. For details about the underlying matrix solution, see Fantke et al. (2013). Mass applied, $m_{\mathrm{app},p}$ ($kg_{applied}/FU$), distribution fractions, $mf_{p,c}$ ($kg_{emitted}/kg_{applied}$), emitted to a given environmental compartment matched between PestLCI Consensus (emission output) and dynamiCROP (input for residue calculations), and emitted mass, $m_{\mathrm{emi},p,c}$ ($kg_{emitted}/FU$) are defined in eq. (2). The air compartment (PestLCI Consensus) is assigned to air (dynamiCROP), field soil surface is assigned to soil, and field crop surface is assigned to leaf surface and fruit surface (see Fig. 1) according to their total crop surface area contributions:

$$mf_{p,c,\mathrm{leaf}} = \frac{LAI}{LAI + FAI} \text{ for leaf surfaces}$$
$$mf_{p,c,\mathrm{fruit}} = \frac{FAI}{LAI + FAI} \text{ for fruit surfaces} \tag{7}$$

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

Combining residue-related characterization factors (dynamiCROP) 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 Fig. 1). Connecting compartments between PestLCI Consensus and dynamiCROP for secondary emission fractions requires further research as there are currently potential overlaps in modeled plant uptake processes (light gray arrows in Fig. 1). Occupational exposure of farmers to pesticides are currently not considered within the existing models.

### 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 S-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.

### 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 1 summarizes the characteristics of the different scenarios, with details on the study system and applied pesticides in SM (Sections S-7 and S-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, and the active ingredient and its applied dose for the impact assessment, including crop characteristics for residue calculations.

## 3. Results

### 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 Fig. 2. 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%).

In Fig. 3, 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 Fig. 3, 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 surface 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).

### 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

**Table 1**
Characteristics of the considered scenarios of tomato production in Martinique.

| Scenario ID (farm) | | A | B | C | D | E | F |
|---|---|---|---|---|---|---|---|
| **Climate**[a] | | Npr | Npr | Svn | Svn | Ssa | Ssa |
| **Soil** | | | Vitric andosol (TV) | | | Vertic cambisol (BV) | |
| **Practices** | Conventional tillage | yes | yes | yes | yes | yes | yes |
| | Irrigation[c] | no | no | no | yes | yes | yes |
| | Buffer zone width (m)[c] | 2 | 2 | 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**[b] | 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 |

[a] 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.
[b] Considered pesticides are detailed in SM (Section S-8).
[c] Practices not assessed in the present study.

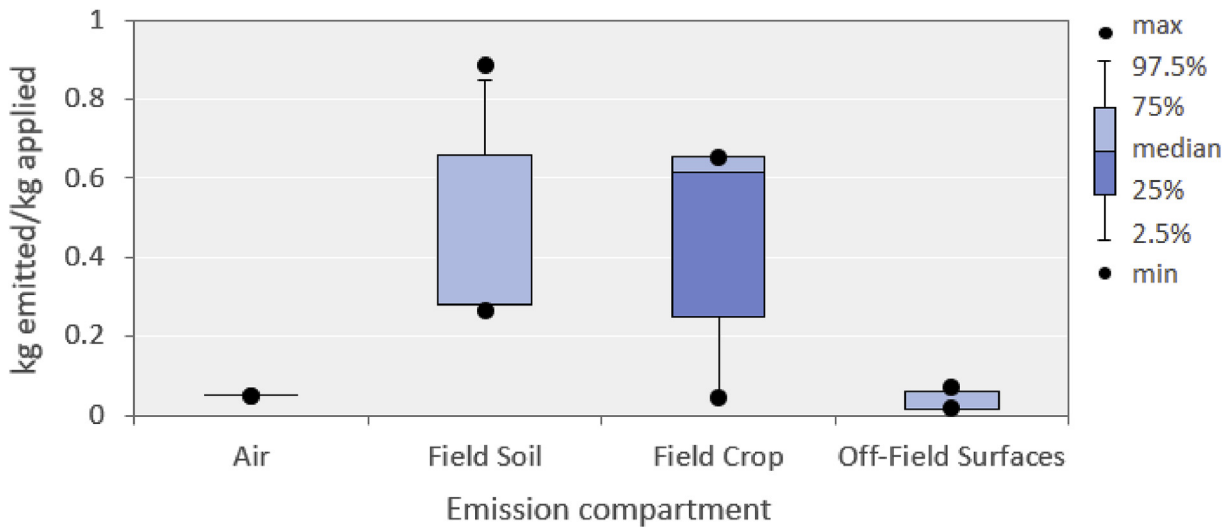


**Fig. 2.** Box-and-Whisker plot indicating the variability of initial pesticide distribution fractions to the different environmental compartments across 80 considered pesticide-field combinations.

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). Fig. 4 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 $m^3$ d/kg tomato for organic substances and 92 PAF $m^3$ 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 Fig. 3 and SM, Section S-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 (5–23 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 $m^3$ 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.

### 3.3. Human toxicity impacts across tomato production scenarios

Fig. 4 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 $2 \times 10^{-7}$ cases/kg tomato for organic substances to $3 \times 10^{-8}$ 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.

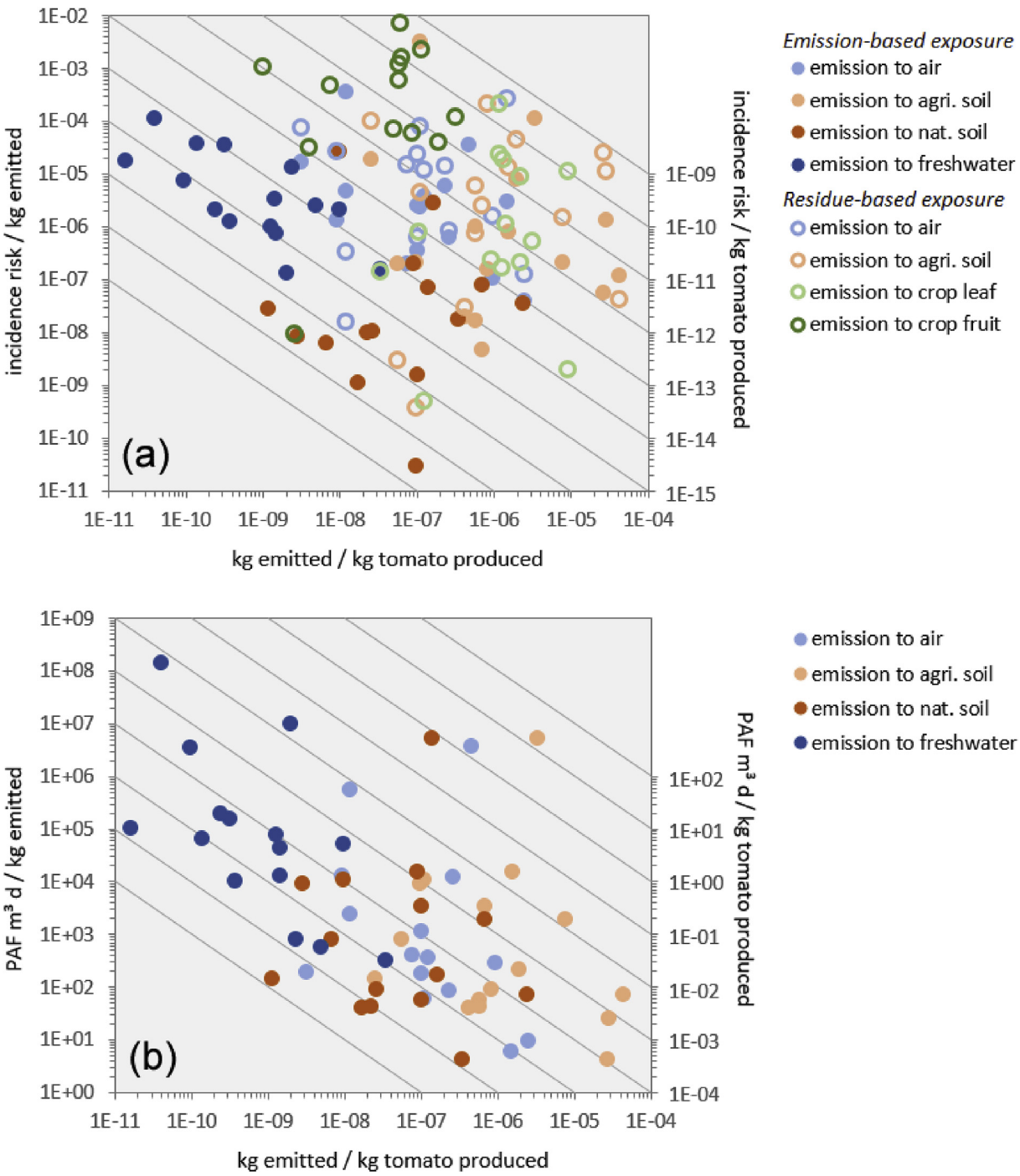


**Fig. 3.** Characterization factors (left-side y-axes) for (a) human toxicity and (b) freshwater ecotoxicity plotted as function of pesticide initial distribution fractions for six tomato fields in Martinique (x-axis). Diagonal equi-impact lines (right-side y-axes) show the respective impact scores.

With respect to impacts related to ingestion of pesticide residues in crops, we find a wide variation across scenarios (see Fig. 4c, 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

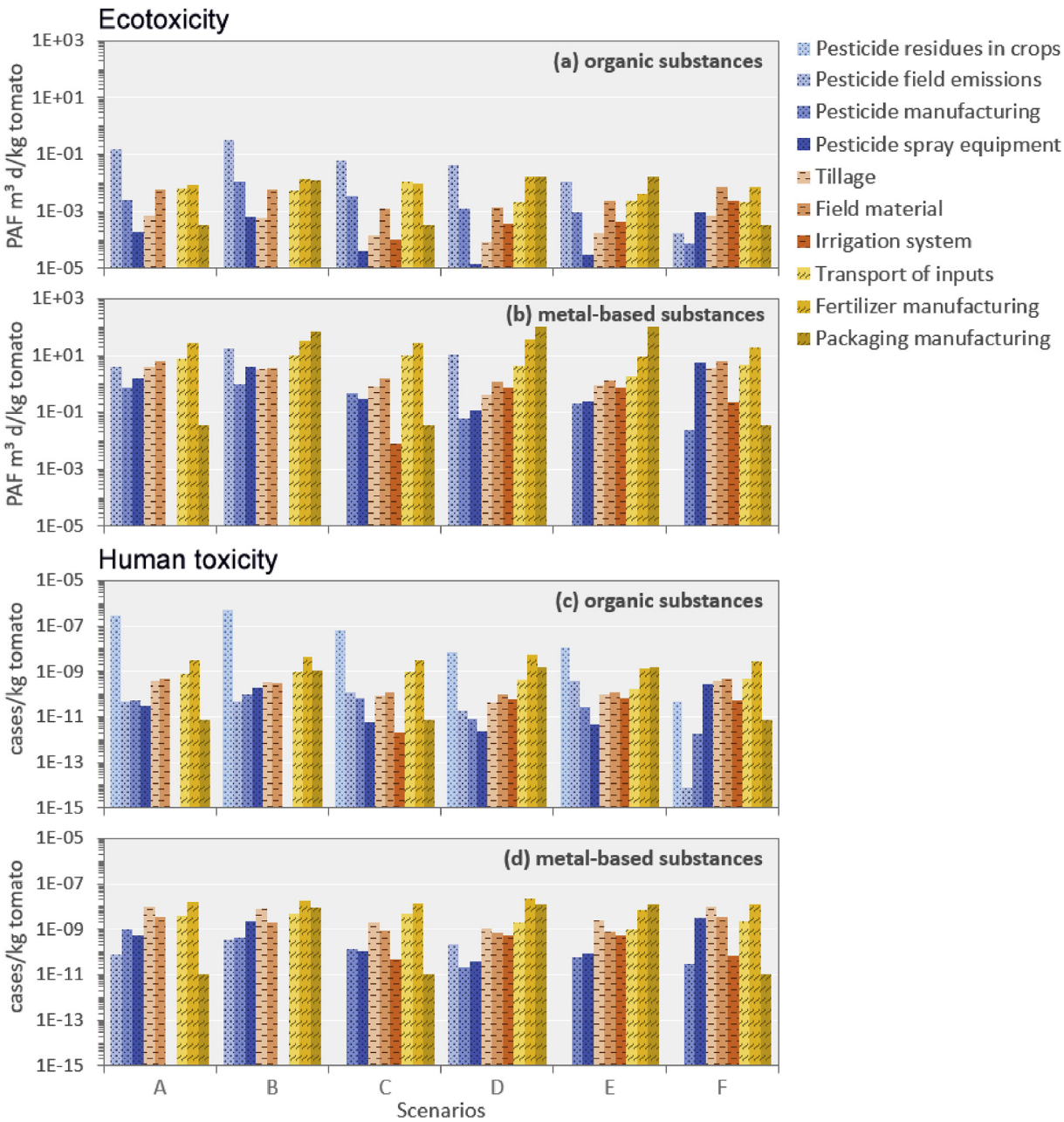


**Fig. 4.** Contribution of cradle-to-farm gate stages to freshwater ecotoxicity and human toxicity impacts per kg tomato harvested for six scenarios, based on initial emission distributions for organic substances (a, c) and for metal-based substances (b, d). Dotted processes belong to pesticides, horizontally 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 1).

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.

### 3.4. 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). Fig. 5 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

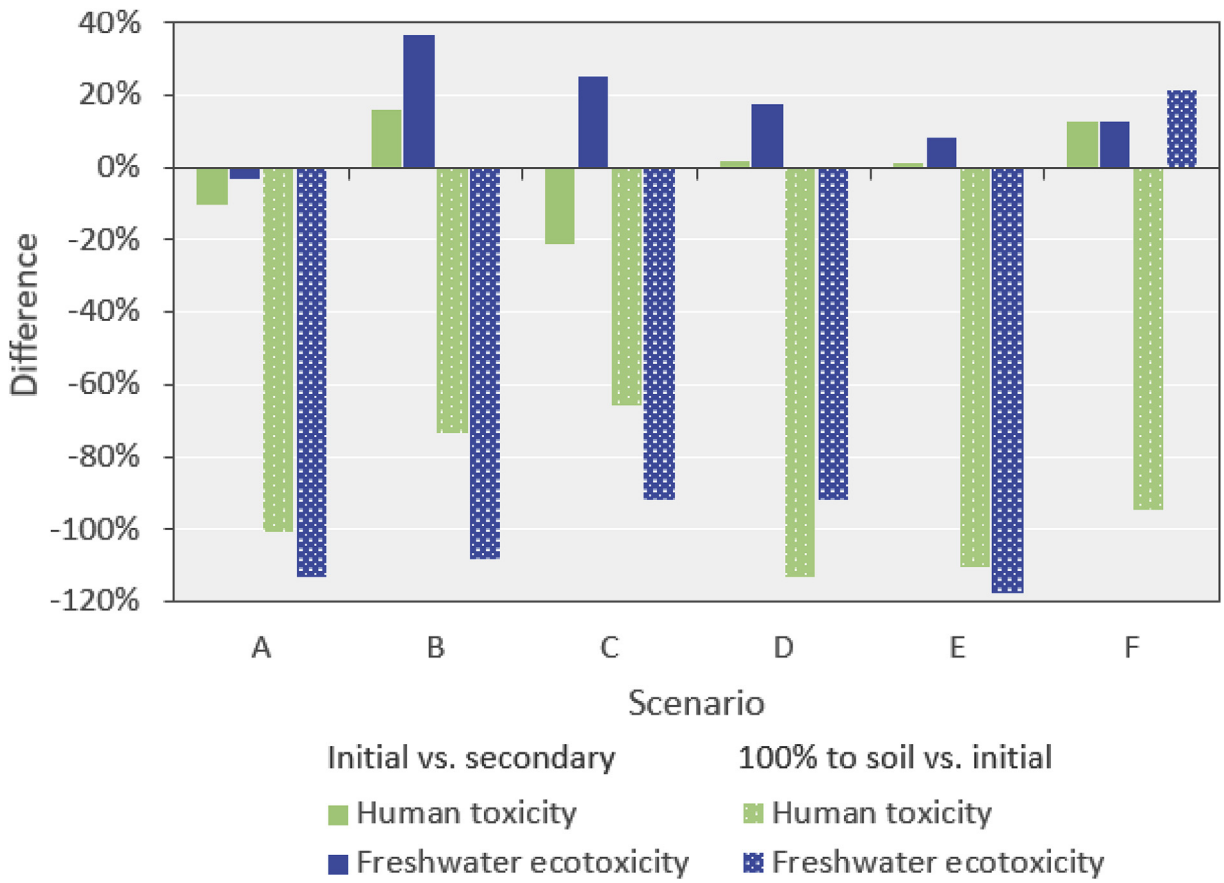


**Fig. 5.** Difference for freshwater ecotoxicity and human toxicity impacts for organic substances contrasting initial distribution fractions, secondary emission fractions, and the common assumption of 100% of applied pesticide being emitted to agricultural soil as underlying approach. Scenarios A-F indicate different combinations of climate, soil and agricultural practice (see Table 1). For the comparison of initial and secondary, the difference was calculated as: variation(%)= (sec − initial)/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: variation(%)=(initial-100%soil)/initial where '100%soil' is for (eco-)toxicity using an emission fractions of 100% to soil.

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 S-9).

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 S-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.

## 4. Discussion

### 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., 2020). 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).

### 4.2. LCA comparisons and limitations

Various LCA studies focusing on one or more aspects of tomato production exist (Bojacá et al., 2013; Khoshnevisan et al., 2014; Ingrao et al., 2019; Payen et al., 2015; Romero-Gámez et al., 2017; Zarei et al., 2019). Detail on existing LCA studies are presented in SM, Section S-11). Out of these studies, all but one, namely Bojacá et al. (2013), 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., 2016), 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 (Fig. 6) 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.

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

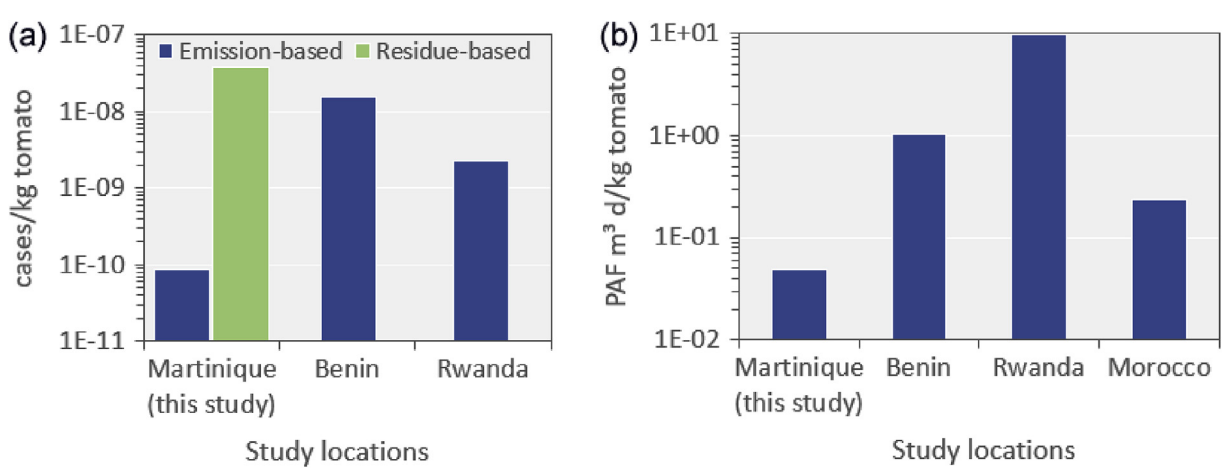


**Fig. 6.** Comparison of (a) human non-cancer toxicity and (b) freshwater ecotoxicity impacts for organic substances for cradle-to-farm gate tomato LCA studies in open-field production in Martinique (our study), Bénin in open-field (Perrin et al., 2015), Rwanda in open-field (Basset-Mens et al., 2016), and Morocco in cold greenhouse (Payen et al., 2015).

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.

### 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., 2020), 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., 2017).

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).

## 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

(Kirchhübel and Fantke, 2019).

## CRediT authorship contribution statement

**Céline Gentil:** Conceptualization, Methodology, Software, Formal analysis, Writing - original draft, Visualization. **Claudine Basset-Mens:** Investigation, Validation, Writing - review & editing, Supervision. **Sarah Manteaux:** Investigation. **Charles Mottes:** Methodology. **Emmanuel Maillard:** Investigation. **Yannick Biard:** Software, Data curation. **Peter Fantke:** Conceptualization, Methodology, Software, Validation, Data curation, Writing - review & editing, Visualization, Supervision.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2020.124099.

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