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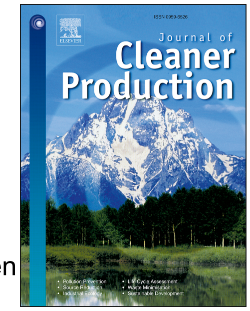

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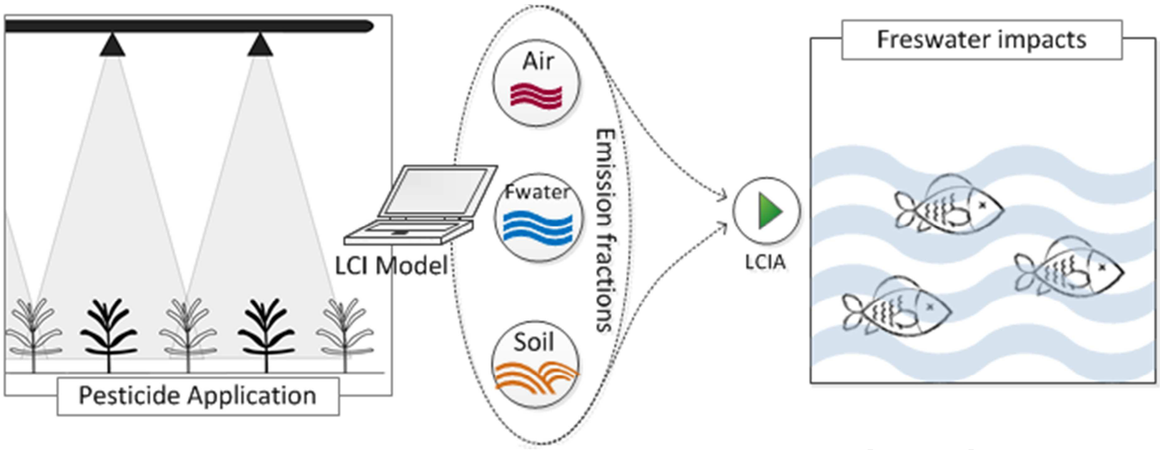

Pesticide Application
LCI Model
Air
Fwater
Soil
Emission fractions
LCIA
Freswater impacts

Freshwater ecotoxicity assessment of pesticide use in crop production: Testing the influence of modeling choices

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ABSTRACT

Pesticides help to control weeds, pests, and diseases contributing, therefore, to food availability. However, pesticide fractions not reaching the intended target may have adverse effects on the environment and the field ecosystems. Modeling pesticide emissions and the link with characterizing associated impacts is currently one of the main challenges in Life Cycle Assessment (LCA) of agricultural systems. To address this challenge, this study takes advantage of the latest recommendations for pesticide emission inventory and impact assessment and frames a suitable interface for those LCA stages and the related mass distribution of pesticide avoiding a temporal overlapping. Here, freshwater ecotoxicity impacts of the production of feed crops (maize, grass, winter wheat, spring barley, rapeseed, and peas) in Denmark were evaluated during a 3-year period, testing the effects of inventory modeling and the recent updates of the characterization method (USEtox). Potential freshwater ecotoxicity impacts were calculated in two functional units reflecting crop impact profiles per ha and extent of cultivation, respectively. Ecotoxicity impacts decreased over the period, mainly because of the reduction of insecticides use (*e.g.,* cypermethrin). Three different emission modeling scenarios were tested; they differ on the underlining assumptions and data requirements. The main aspects influencing impact results are the interface between inventory estimates and impact assessment, and the consideration of intermedia processes, such as crop growth development and pesticide application method. Impact scores for AS2 were higher than RS and AS1, but the differences in the crops ranking was less apparent. On the other hand, the influence on the estimation of impacts for individual AIs was considerable and statistical differences were found in the impact results modeled in scenarios RS and AS2. Thereby indicating the effect of inventory models on ecotoxicity impact assessment.

Keywords: Pesticide emission factors, inventory modeling, ecotoxicity characterization, life cycle impact assessment (LCIA), feed crops, agriculture.*[1]

[1] Abbreviations
AI: Active ingredient
AS: Alternative scenario
CF: Characterization factors
DK: Denmark
EF: Effect factor
FF: Fate factor
Fun: Fungicides
GAP: Good agricultural practices
Gly_agri: Total agricultural use of glyphosate
Hrb: Herbicides
Ins: Insecticides
IS: Impact scores
LAI: Leaf area index
NAP: National Action Plans
Pgr: Plant growth regulators
RS: Reference scenario
XF: Exposure factor

1 INTRODUCTION

With the increased global demand for agricultural products for food, fiber and bioenergy, and the interrelated concerns on the environmental impact hereof, there is a need to have efficient tools to evaluate the environmental performance profiles of agricultural production, to facilitate a move towards more sustainable production systems. Life Cycle Assessment (LCA) is widely applied to quantify the potential impacts of products and systems along with their entire life cycles. One of the main challenges in assessing the environmental performance of agricultural systems in LCA is modeling emissions from pesticide use and the subsequent coupling with the impact characterization model (van Zelm et al., 2014). Over the past years, a significant number of LCA studies on agricultural systems were conducted (Gasol et al., 2012; Milà et al., 2006; Noya et al., 2017; Torrellas et al., 2012). However, ecotoxicity impacts as currently modeled may lead to inconsistent results and wrong conclusions in few cases (*e.g.*, comparing conventional vs organic farming), mostly due to the lack of agreement and precise definitions on the modeling framework for this impact category (Fantke et al., 2018; Meier et al., 2015; Müller et al., 2017; Notarnicola et al., 2017; Saouter et al., 2017a, 2017b).

The development of the life cycle inventory (LCI) analysis and subsequent life cycle impact assessment (LCIA) (*e.g.*, pesticide emission quantification and related characterization of ecotoxicity impacts) are the core phases of an LCA study. The robustness and reliability of the LCA results depend mainly on the quality and representativeness of the LCI and LCIA data and models selected. Different modeling options, hence, will affect the impact profiles of a study, and this is especially relevant for agricultural systems (Anton et al., 2014).

Quantifying the chemical emissions to the environment in the LCI phase is typically based on generic assumptions, often based on standard emission factors (*e.g.*, expressed in percentages of applied mass) or dynamic models based on specific application scenarios that describe the emission distribution of organic pesticides. The consensus effort on the delimitation between pesticide emission inventory and impact assessment for LCA already provides guidelines on what should be quantified in those LCA steps but explicitly exclude how to do it avoiding recommendations on specific models (Rosenbaum et al., 2015). The implications of choosing different emission models in the LCA of crop production have been discussed for some agricultural systems (Goglio et al., 2018; Schmidt Rivera et al., 2017; van Zelm et al., 2014). However, no studies are addressing the influence of the pesticide emission modeling approach, nor the evaluation of recent developments in impact assessment methods to determine pesticide ecotoxicity impacts in different crop production systems.

Thus, there is a need to test different choices on how to quantify pesticide emission fractions (i.e., different modeling approaches) and the recent developments on the recommended method for freshwater ecotoxicity characterization in the production of feed crops.

The purpose of the present study is to contribute to the evaluation of the ecotoxicological burden on freshwater ecosystems from pesticide use in crop production using the pesticide use in Denmark (DK) as a case study. It is focused on assessing the influence of pesticides on the environmental impact profiles of the cultivation of feed crops during the period 2013-2015, testing the effects of modeling choices in the inventory analysis as well as in the impact characterization.

2 MATERIALS AND METHODS

This study followed the LCA methodology to evaluate the potential ecotoxicity impacts on freshwater ecosystems from pesticide use in DK's crop production. This bottom-up analysis focuses on the evaluation and influence of pesticide application on the environmental impact profiles of maize, winter wheat, grass, spring barley, rapeseed, and peas during the period 2013-2015.

2.1 Definition of ecotoxicity impact scores

The quantification of ecotoxicity impact scores for freshwater ecosystems includes i) Detailed LCI reporting on the pesticide active ingredient (AI); application methods, time and mass, location, agricultural practices and crop stage development; ii) quantified AI emission fractions for both on-field and off-field; and iii) measures to avoid double counting of multimedia transfers considered in the quantification of emission fractions and the impact assessment fate modeling (Rosenbaum et al., 2015). Accordingly, the freshwater ecotoxicity impact scores (IS) can be described as:

$$IS = \sum_{i,x}(CF_{i,x} \cdot m_{i,x}) \tag{1}$$

Where $CF_{i,x}$ is the characterization factor for freshwater ecotoxicity [PAF m^3 d $kg_{emitted}^{-1}$], and $m_{i,x}$ is the mass of AI x emitted to compartment i per area treated [$kg_{emitted}$ ha^{-1}]. Potential freshwater ecotoxicity impacts (IS_{crop_ha}) [PAF m^3 d ha^{-1}] were determined in relation to 1 hectare [ha] of crop in a given year t within 2013 and 2015. Additionally, freshwater ecotoxicity impact profiles at country or regional level (IS_{crop}) [PAF m^3 d $crop^{-1}$] from pesticide use were derived from the product of crop impact scores and the total crop area in a given year in DK.

The interface between LCI and LCIA and related mass distribution for pesticide application in crop production are presented in Figure 1. This approach follows the proposed framework for pesticide inventory and impact assessment (Rosenbaum et al., 2015; van Zelm et al., 2014).

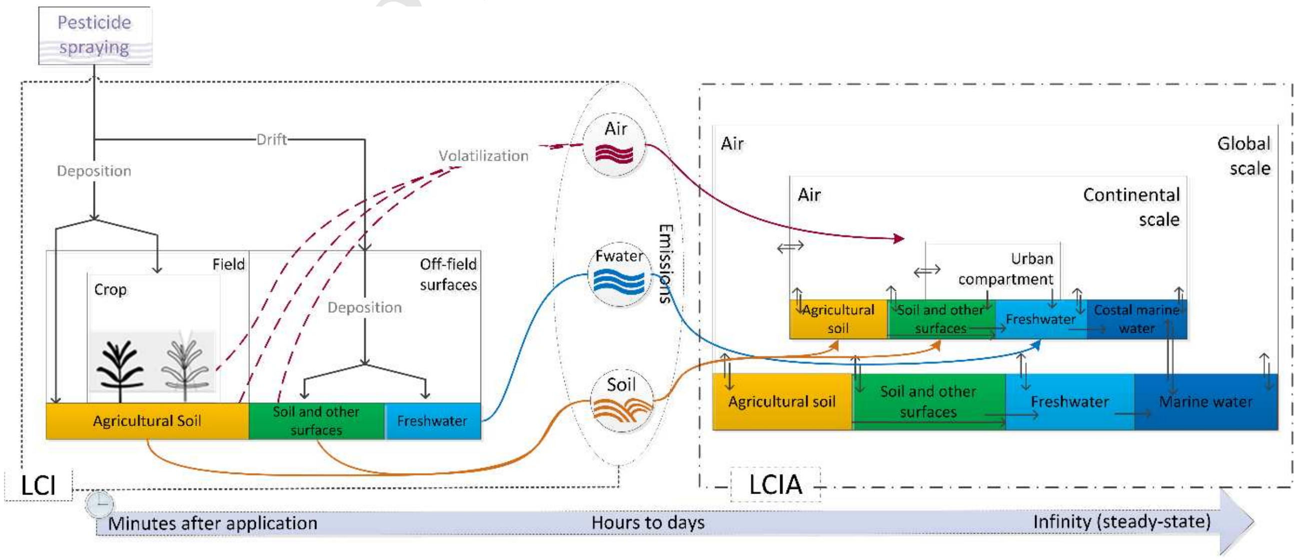

Figure 1 Interface between LCI and LCIA for pesticide application in crop production

This interface considers the boundaries between the emission inventory and impact assessment, setting also spatial and time dimensions, to quantify the AI emission fractions (in air, freshwater, and soil) and characterize ecotoxicity impacts, avoiding any overlap or double counting of the chemical fate process. Furthermore, the emission flows, both on and off the field, are clearly indicated and their link to the characterization factors for the impact pathway (i.e., freshwater ecotoxicity).

2.2 Pesticide emission inventory

Pesticide application practices in DK for the selected crops were determined. Concrete AI was used throughout the study, meaning, that the chemical that is the biologically active part of any pesticide was assessed (European Commission, 2017). The mass applied per AI was derived from the annual statistical report on pesticide use by crop in DK for 2013 (Ørum and Samsøe-Petersen, 2014), 2014 (Ørum and Hossy, 2015) and 2015 (Ørum and Holtze, 2017). We addressed nearly 60 different AIs from four distinct target classes, herbicides (Hrb), plant growth regulators (Pgr), fungicides (Fun), and insecticides (Ins). Additionally, glyphosate (CAS-RN107-83-6) use is not allocated to any specific crop cultivation, and it was assessed as the total agricultural use of the AI per 1 hectare [ha] in a given year, hereafter identified as ($Gly_{\_agri}$). All AI identification (CAS registry numbers-RN and names), and classes are reported in Supporting Information (SI), Table S1.

2.3 Pesticide emission quantification

Crops are treated by foliar spray application (typically boom sprayers), and the reported DK statistics on pesticide treatments were used as a proxy for agricultural practices. The agricultural field is considered as part of the ecosphere. The total emission fraction of an AI [kg kg^{-1}] is quantified as the sum of the fractions initially emitted to the different environmental compartments:

$$f_{em} = \frac{m_{em}}{m_{app}} = f_{em\_air} + f_{em\_fw} + f_{em\_soil.agri} + f_{em\_soil.other} + f_{em\_crop} \qquad (2)$$

Where f_{em} is the fraction of the applied mass of pesticide that becomes an emission to the environment, m_{em} the mass emitted, m_{app} the mass of pesticide applied, f_{em_air} the fraction of applied mass that is emitted to air, f_{em_fw} the fraction of applied mass that is emitted to freshwater, $f_{em\_soil.agri}$ the fraction of applied mass that is emitted to on-field soil, $f_{em\_soil.other}$ the emission fraction reaching off-field soil and other surfaces, and f_{em_crop} is the fraction reaching crop surfaces. These pesticide emissions were modeled in two sequential steps, initial distribution (primary processes) and secondary emission transfers.

Primary distribution

The primary distribution processes between compartments occur during the initial minutes after pesticide application. These primary processes are emission by wind drift (f_{d_lost}), pesticide deposition and the fraction intercepted by the crop or weed (further details are presented in SI, Table S2). Since the fractions from initial distribution to environmental media should sum up to 100% of the applied mass, considering losses via degradation during the initial minutes negligible, the aggregated emission fractions will be equal to one (Fantke et al., 2011a; Juraske et al., 2007). Consequently, the crop/weed interception fraction (f_{int_crop}) of an AI directly after the application will be given by:

$$f_{\text{int\_crop}} = 1 - \left(f_{d\_lost} + f_{dep\_soil.agri}\right) \quad (3)$$

The fraction lost by wind drift f_{d_lost} [kg kg^{-1}], depends on the application method, i.e., the spray equipment and elevation, and wind speed. Based on models for conventional spray equipment on field crops and deposition curve parameters assuming good agricultural practices (GAP), the f_{d_lost} was fixed to a value of 0.1 (Gil et al., 2014; Gil and Sinfort, 2005; Gyldenkrne et al., 1999; van de Zande et al., 2007). The soil deposition $f_{dep\_soil.agri}$ [kg kg^{-1}], depends on crop-specific leaf area index (LAI), thereby affecting fractions reaching soil surfaces of the treated field area (Fantke et al., 2011b). With an exponential model (Gyldenkærne et al., 2000; Juraske et al., 2007), based on crop growth stage and capture efficacy, the fraction reaching the soil surface is described as:

$$f_{dep\_soil.agri} = e^{-k_p \times LAI} \quad (4)$$

Where kp is the capture coefficient [-] and set to 0.55 for pesticide spray solutions prepared with adjuvants (Gyldenkrne et al., 1999). Pesticide target class and specific application time were used to define crop-specific growth stages in the selected crops. The LAI was derived for plant growth regulators, insecticides and fungicides distinctly as a value dependent on the target class/crop growth stage/application time combination, (Fantke et al., 2011b; Itoiz et al., 2012; Olesen and Jensen, 2013); for herbicide application on weeds the corresponding LAI of 0.5 is used. This value is based on the reported leaf cover factor for fallow lands (Panagos et al., 2015).

Secondary distribution

The subsequent secondary emission transfers include re-volatilization after deposition and off-field emissions allocation. The volatilization from fractions deposited in the different compartments is derived from the default Tier 1 emission factors per AI from their vapor pressures (Webb et al., 2016) see Table S1 and S3 in SI. The emission factor emF was calculated for each AI (see, SI Table S1), the inter-media transfer and the final emission factors are presented in SI, SI-1, and SI-2. Finally, the water to soil area ratio for DK (0.016) was used to allocate the off-field emissions (i.e., drift fraction deposited in off-field surfaces) see SI, Table S2. This value is based on reported data of the Danish ministry of environment (Stockmarr and Thomsen, 2009).

2.4 Freshwater ecotoxicity characterization

For assessing the ecotoxicity of pesticides on freshwater ecosystems, we followed the LCIA emission-to-damage framework that links emissions to impacts through environmental fate, exposure and effects (Jolliet et al., 2004). According to (Hauschild and Huijbregts, 2015; Rosenbaum et al., 2008) characterization factors CF for freshwater ecotoxicity of chemical emissions can be expressed as:

$$\mathrm{CF}_{i,x} = FF_{i\rightarrow fw,x} \times XF_{fw,x} \times EF_{fw,x} \quad (5)$$

Where $FF_{i\rightarrow fw,x}$ is the fate factor in [$kg_{in\_compartment}$ / ($kg_{emitted}$ d^{-1})] describing the mass transport, distribution and degradation in the environment. The ecosystem exposure factor, $XF_{fw,x}$, is defined as the bioavailable fraction of a chemical in freshwater; and an effect factor ($EF_{fw,x}$) expressing the ecotoxicological effects associated with the bioavailable fraction, in the exposed ecosystems integrated over the surrounding water volume. CFs were estimated with USEtox 2.02 as characterization model, with the specific European landscape dataset (i.e., representing DK conditions) (Fantke et al., 2017; Westh et al., 2015). New CFs for 10 additional AIs, following the procedure in Fantke et al. (2017) were derived. A detailed description of the resulting CF and the data used can be found in SI, SI-3.

Recent developments for the estimation of CFs, such as the improvements in the calculation of the fate factors now accounting for the influence of pH on partitioning processes ionizing organic chemicals, were introduced in USEtox version 2.02 (Fantke et al., 2017). The differences in the potential freshwater ecotoxicity impact results, coming from the different model versions (USEtox 1.01 and 2.02) were tested. This evaluation was performed taking into account AIs available in both USEtox versions.

2.5 Sensitivity analysis

Two types of local sensitivity tests were conducted. First, a scenario sensitivity analysis was performed testing the sensitivity on IS results of different modeling scenarios in the impact profiles of feed crop cultivation in DK.

There are very different approaches and assumptions in order to provide emission estimates for quantifying lifecycle emission inventories of pesticides in any LCA study involving agricultural systems (Fantke, 2018). The most simplified approaches are based on generic assumptions regarding varying percentages for pesticide application. The most frequently used approach and the more simplified is the assumption that all pesticides remain in the soil (i.e., 100% emitted to soil) (Nemecek and Kagi, 2007). Following this line of fixed percentages, there are several approaches that distribute pesticide emissions on more than one environmental compartment (Berthoud et al., 2011; Margni et al., 2002; Neto et al., 2013). A different approach is the more complex emission modeling as in PestLCI model. This model estimates emissions to three environmental compartments: air, surface water, and groundwater. It considers the agricultural field down to 1 m depth into the soil and up 100 m into the air as part of the technosphere, thus excluding emissions to soil on-field and off-field (Birkved and Hauschild, 2006; Dijkman et al., 2012). The main differences between the methods are the underlining assumptions (*e.g.*, boundaries between ecosphere and technosphere), the level of sophistication (fixed percentages vs. modeling), the data requirements and applicability for quantifying pesticides emissions. All these different approaches, may offer inconsistent results, which in some cases are partially overlapping (spatially or temporally) with the impact pathways for pesticides. Therefore, IS results tend to be not compatible or comparable.

For the present study, modeling approaches that allowed the inclusion of agricultural soil in the assessment and that involve simplified assumptions for application methods were selected. Three scenarios were considered, the above proposed simplified estimation routine (section 2.3), was selected as a reference scenario (RS) and two alternative scenarios (AS1-AS2) that represent different modeling approaches to quantify emissions from pesticide use. The alternative scenario AS1 followed Margni et al. (2002), which represents a usually used pesticide emission modeling, and furthermore is one of the first approaches that account for pesticide emission distribution in different environmental media in LCA studies for agricultural systems. In this approach, the pesticide emissions are distributed in environmental media based on fixed share percentages. They assume that the fraction of AI emitted to the soil will be 85% of the total application, 5% will stay on leaves and the remaining 10% is lost into the air across crops and pesticides. The second tested scenario AS2 represents fixed emission fractions dependent on the foliar spray application and drift distributions for field crops. This approach was chosen to represent a modeling framework where the initial distribution (i.e., application method and crop relation) is taken into account but also allowing the inclusion of field emissions in the assessment (Balsari et al., 2007; Felsot et al., 2010; Gil and Sinfort, 2005). Table 1 displays the emission fractions in the three scenarios considered.

Table 1 Comparison of pesticide emission fractions (*f*em) calculated by the RS (reference scenario), AS1 (Margni et al. 2002) and AS2 (application method and crop relation)

Emission scenarios	Average fraction emitted [kg kg^{-1}]	Standard deviation on fractions
RS		
f_{em_air}	1.16x10^{-1}	2.03x10^{-1}
f_{em_fw}	1.60x10^{-3}	N/A
$f_{em\_soil.agri}$	3.75x10^{-1}	3.11x10^{-1}
$f_{em\_soil.other}$	8.70x10^{-2}	2.01x10^{-2}
AS1		
f_{em_air}	1.00x10^{-1}	N/A
f_{em_crop}	5.00x10^{-2}	N/A
f_{em_soil}	8.50x10^{-1}	N/A
AS2		
f_{em_air}	1.70x10^{-1}	N/A
f_{em_fw}	1.00x10^{-2}	N/A
f_{em_soil}	4.50x10^{-1}	N/A
f_{em_crop}	3.70x10^{-1}	N/A

Note: f_{em} is the fraction emitted. Indices air, fw (freshwater), soil and crop denote environmental compartment were the emission occurs.

Second, input parameter sensitivity analysis was performed to test the sensitivity of the RS by evaluating the change in the impact scores (propagated from the change in emission fractions) as a function of the variation of several input parameters by a factor of 2 larger of their initial values, one at the time. Local sensitivity to input S_{in} [-] was further expressed as the effect on the model output due to a change in an input parameter (for further details see SI, SI-5). Finally, a statistical analysis between impact results of the different scenarios was conducted.

3 RESULTS AND DISCUSSION

3.1 Pesticides use in Danish crop production (2013-2015)

The AIs considered in the study cover 98.3% of the total pesticide applications in relation to the mass applied for the selected crops: maize, winter wheat, grass, spring barley, rapeseed, peas and the agricultural use of glyphosate ($Gly_{\_agri}$). The total pesticide use was 3165 tons in 2013, 1438 tons in 2014 and 2105 tons in 2015. The average pesticide application rates per crop vary between 2 and 3 orders of magnitude (SI, Table S6). Grass is the crop with the lowest application rates and pesticide use; together, fungicides and insecticides represent nearly 20% of the total use in grass-2013; additionally, in 2014-2015, there was no use of insecticides, and fungicides use was reduced by less than 2.5%. Gly_agri sum up to 2722 tons in the 3 years and represents near 40% of the total use of pesticides in DK. Winter wheat (2672 tons) is the crop with higher pesticide use followed by spring barley (748 tons) (SI, Table S7). The most used pesticide target class is Herbicides, and prosulfocarb is the most used AI after Gly_agri within this target class. Regarding pesticide use it is important to mention that the farmer's choice for an AI or another can be influenced by many different external factors, such as climate variations or the emergence of pests and diseases, crop rotations, market needs and many others (Steingrímsdóttir et al., 2018). For annual crops, a three year assessment period could balance somehow the fluctuations in the crop growing conditions (*e.g.,* as recommended for the assessment of greenhouse gas emissions) and the variations of the factors mentioned above (BSI, 2012; Knudsen et al., 2014).

3.2 Ecotoxicity impact profiles of feed crops (2013-2015)

The IS_{crop} from pesticide use decreased over the three years (Figure 2). The reduction of the IS_{crop} was more apparent in 2014 (59%) than in 2015 (33%) with respect to the base year (2013). Most of the decrease in the IS_{crop} was due to the non-use of a single substance: cypermethrin. This insecticide was the major contributor to IS_{crop} in 2013 across crops (*e.g.,* 87% in maize, 60% in spring barley and 47% in winter wheat) and was no longer used in 2014-2015 (see Table S8 in SI). Furthermore, the fact that maize and grass did not require the use of insecticides in 2015 also contributes to the reduction of IS_{crop}. However, it is essential to note that this may be the result of unfavorable climatic conditions for the emergence of pests, different insect population cycles (affecting abundance), competition and predation (*e.g.,* natural enemies) among many other different reasons.

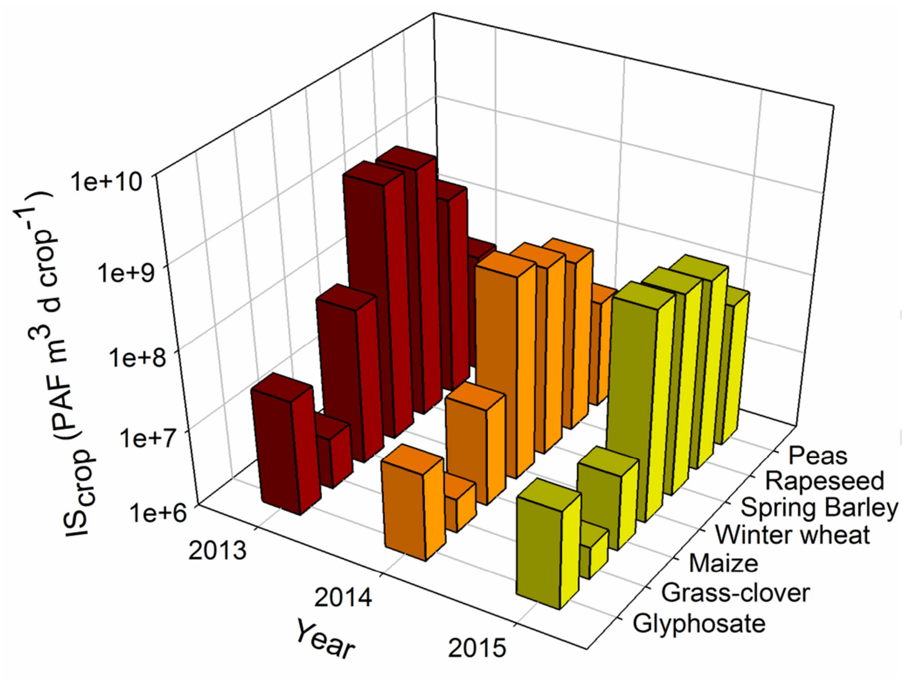

Figure 2 Freshwater ecotoxicity impact profiles for crop production (2013-2015), impact scores IS_{crop} expressed in [PAF m^3 d $crop^{-1}$]. *Glyphosate (CAS RN-107-83-6) assessed as the total agricultural use in Denmark

As shown in figure 2, winter wheat-2013 ($1.6x10^9$ PAF m^3 d $crop^{-1}$), spring barley-2013 ($1.4x10^9$ PAF m^3 d $crop^{-1}$) and rapeseed-2013 ($3.3x10^8$ PAF m^3 d $crop^{-1}$) present the higher IS_{crop}. The larger IS_{crop} in those crops is associated with the use of insecticides (*e.g.*, cypermethrin, pendimethalin, and lambda-cyhalothrin) and fungicides (*e.g.*, pyraclostrobin, azoxystrobin, and folpet), AIs with relatively high CFs, and also because these are some of the crops more extensively cultivated in DK (i.e., cultivated area). Consequently, substance prioritization by LCA helps to identify potentially harmful AI for ecosystems and, with the restriction of their use or the implementation of more sustainable practices, significant changes in the impact profiles of the crops can be made more apparent (*e.g.*, cypermethrin). In this sense, if farmers choose to use pesticides AIs causing lower impacts, the load on agricultural systems will decline, even if they continue to spray their fields as usual for pests and disease control. Moreover, linking this decision with integrated pest management (IPM) will further contribute to lowering the ecotoxicological burden on freshwater ecosystems from pesticide use.

3.3 Pressure of pesticide impacts by hectare and class (2013-2015)

When calculating the potential ecotoxicity impacts on freshwater ecosystems per 1 hectare of crop per year (IS_{crop_ha}) [PAF m^3 d ha^{-1}] the interaction of agricultural systems and practices is more apparent. The variations in pesticide use (almost 3 orders of magnitude) and impact scores for individual AIs (up to 9 orders of magnitude) are significant (see Table S9 in SI). Therefore, in the same year, the trends for the two indicators can move in different directions (Figure 3), meaning that pesticide use or application rates are not an adequate indicator of potential impacts (*e.g.*, Gly_agri and rapeseed), since toxicity potentials might be higher for pesticides that are applied in lesser amounts (Fantke and Jolliet, 2016).

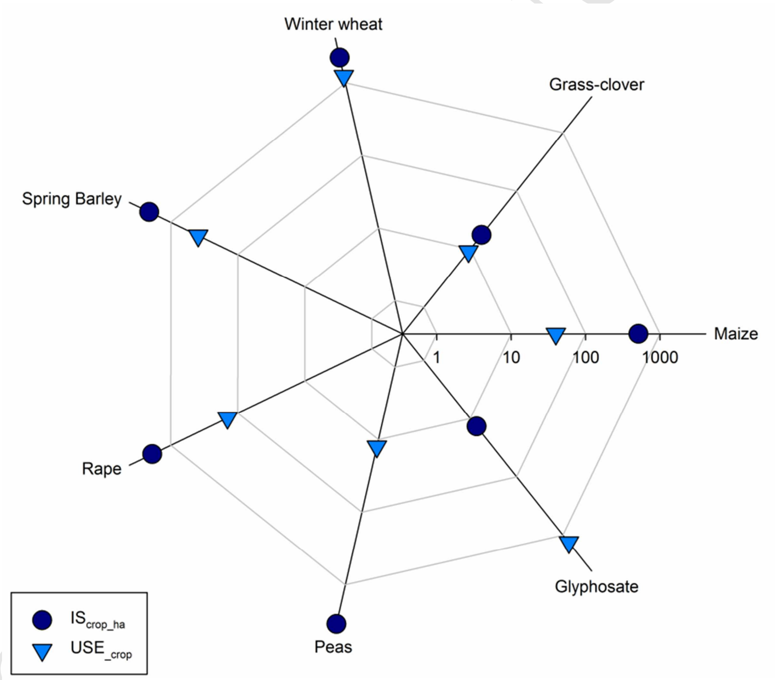

Figure 3 Comparison between use of pesticide active ingredient (USE_crop) [tonnes] and potential freshwater ecotoxicity impacts (IS_{crop_ha}) [PAF m3 d ha^{-1}] for 5 analyzed crops 2013 and *Glyphosate (CAS107-83-6) assessed as the total agricultural use in Denmark in logarithmic scale

Peas appeared as the crop with the highest pressure by hectare cultivated in the entire period, with the maximum value (6440 PAF m^3 d ha^{-1}) in 2015. In 2013 rapeseed, spring barley and winter wheat showed IS_{crop_ha} between 64% and 54% lower than peas, in 2014 the difference for the same crops was among 70% and 85% lower and for 2015 all crops showed IS_{crop_ha}

80% lower than peas (see Figure 4). The IScrop_ha for the study varies up to 3.5 orders of magnitude, and the substances cypermethrin (Ins), aclonifen (Hrb), pendimethalin (Hrb) and lambda-cyhalothrin (Ins) present the most significant contribution to IScrop_ha, which his nearly 70% (see Table S9 in SI).

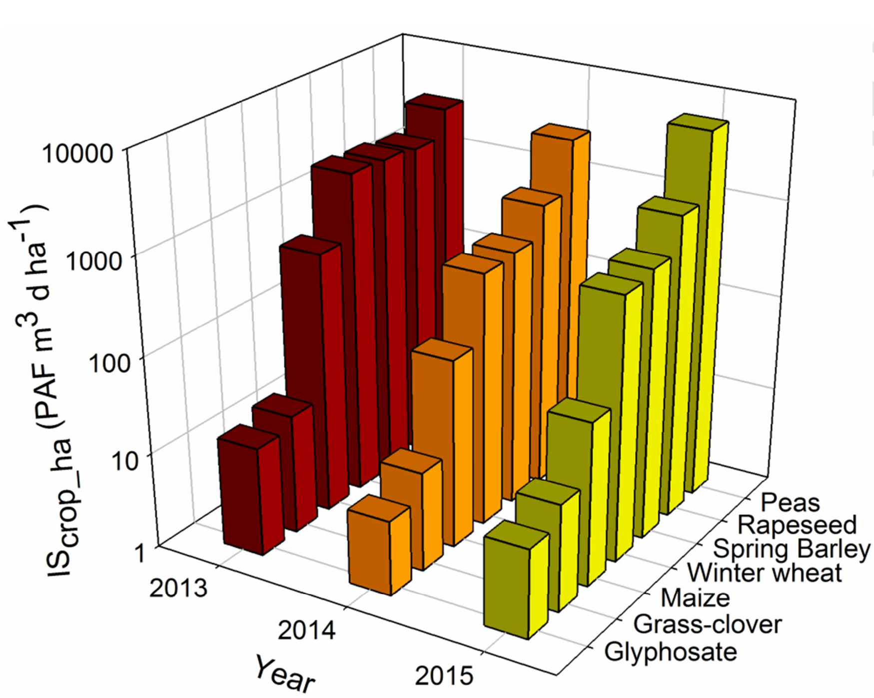

Figure 4 Pressure of pesticide impact scores by hectare of crop cultivated for Danish crop production (2013-2015), impact scores IS_{crop_ha} in [PAF m^3 d ha^{-1}]. *Glyphosate (CAS RN-107-83-6) assessed as the total agricultural use in Denmark

The large IScrop_ha for peas-2015, almost double than precedent years, is mainly explained by the bloated use of aclonifen (Hrb). This intensification of herbicide treatments in 2015 could be potentially associated with the emergence of weed infestation in pea's productions fields. Moreover, the sharp increment on IScrop_ha in part is explained by the dose increment by hectare and the relatively high CF for direct emissions to surface water of aclonifen (SI, Table S5), which is driven by a significant EF ($1.3\times10^{+4}$ PAF m3 kg^{-1}). Furthermore, it is important to note that even if some substances have a high CF, their use could be justified at low doses, because of their agronomic importance and effectiveness of pest or disease control. The contribution by pesticide target class to freshwater IScrop_ha can be observed in Figure 5. Insecticides are the class that contributes the most (56%) to impact scores, followed by herbicides (36.4%) and fungicides (7%); plant growth regulators were not included in Figure 5 as their contribution to IScrop_ha and IScrop_DK was lower than 1%.

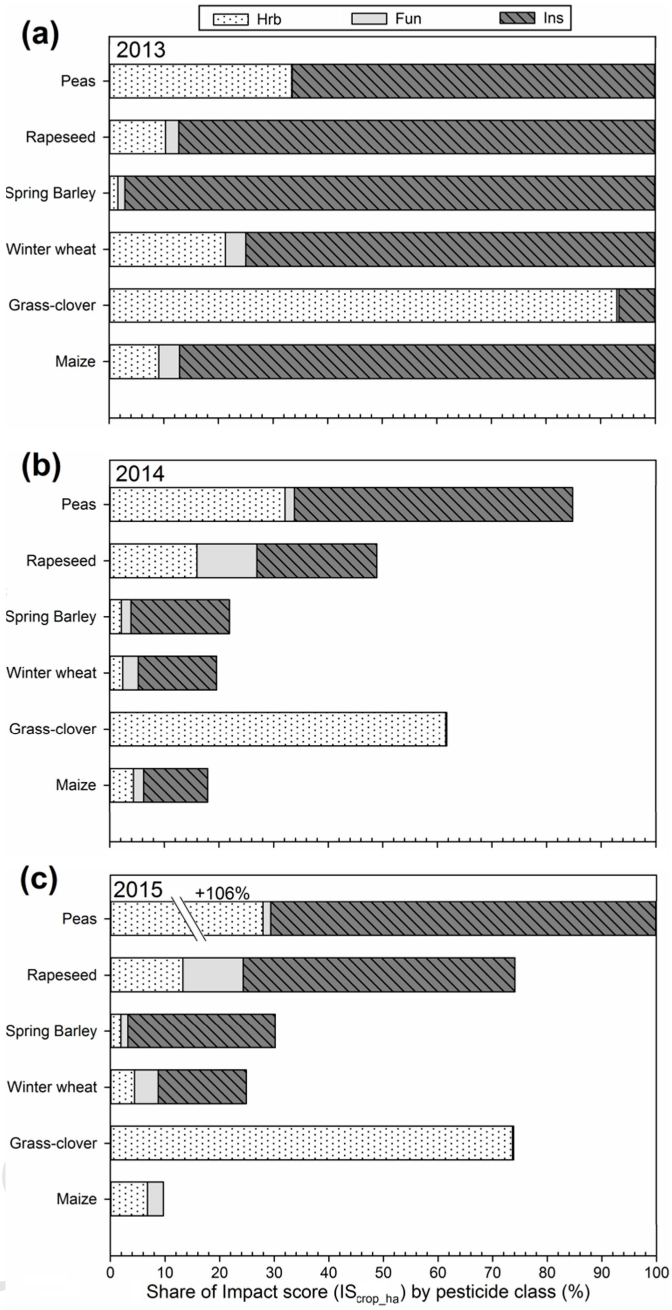

Figure 5 Share of freshwater ecotoxicity impact scores IS_{crop_ha} in [%] for a) 2013, b) 2014 and c) 2015; by pesticide class herbicides (Hrb), insecticides (Ins) and fungicides (Fun) taking IS_{crop_ha} - 2013 as the reference year.

It is well known that pesticide treatments are a highly dynamic activity that varies year by year. It could be more static for herbicides than for the other pesticide classes (i.e., insecticides and fungicides) that are more closely correlated with the specific climatic conditions on the area and year of study and thus also the emergence of any specific pest or disease. If these dynamics are to be considered, the relevant data (on, *e.g.,* pesticide treatment and crop characteristics) have to be consistently reported (Fantke et al., 2016). Furthermore, the assessment period should also reflect these fluctuations in the crop growing conditions, that is why it should also carefully designated (Knudsen et al., 2014).

Some other authors have found similar results than here (unallocated values by hectare and year). For example, similar trends are found by Nordborg et al. (2014) for the cultivation of maize, rapeseed and winter wheat for biofuel feedstock production; Parajuli et al. 2017 for grass, maize and winter wheat straw for bio-refinery, and Schmidt Rivera et al. 2017 for barley production in Italy and Denmark. The studies mentioned above use PestLCI (version 1 or 2) as inventory model and USEtox 1.01 as characterization method for the impact assessment. Therefore, using a fewer data demanding and a simplified approach could lead to the same results for substance prioritization. Despite the similarities in the trends of IS_{crop_ha}, when comparing the results with the absolute values of AI use per 1 ha in a given crop (in the same studies), the IS_{crop_ha} are up to 2.2 orders of magnitude higher; considering the uncertainty range of the characterization method (between 1-2 orders of magnitude) this difference might be moderately significant, and more probably associated with differences in the LCI and the pesticide emission model.

3.4 Effects of modeling choices on ecotoxicity impact score results

3.4.1 Sensitivity of different pesticide emission modeling scenarios

The selected methodologies are described in section 2.5, and the results of the three scenarios (RS, AS1, and AS2) were compared between the five crops in the 3-year period. The mean values for f_{em} into air, freshwater and soil in the RS are ~1 order of magnitude higher than the alternative AS2 and greater than AS1 but within the same order of magnitude. The major difference between RS and AS2 comes from the fact that in the latter 37% of the mass applied is considered retained by the crop, degraded or taken up, and thus not been considered as an emission. When modeling soil emissions the mean values present lower differences in comparison with the variations of freshwater and air emissions. Significant differences were found between RS and AS2 for freshwater emissions, and across the three scenarios for air emissions. All these variations in the emission fractions lead to further changes (propagated) in the resulting IS.

Results for IS_{crop_ha} in [PAF m^3 d ha^{-1}] in the scenarios RS, AS1 and AS2 are summarized in Table 2. AS1 presented the lowest impact results across crops and years; the highest impact results appear in AS2, whereas RS showed higher impacts than AS1 but in the same order of magnitude.

Table 2 Comparison of scenarios to test different emission modeling approaches. Results for potential freshwater ecotoxicity impact scores IS_{crop_ha} expressed as [PAF m3 d ha^{-1}] in the reference scenario (RS) and alternative scenarios AS1 and AS2

Crop	RS			AS1			AS2		
	2013	2014	2015	2013	2014	2015	2013	2014	2015
Maize	513	92	50	182	35	45	3041	475	138
Grass	17	11	13	17	7	9	51	31	37
Winter wheat	2210	434	551	883	221	256	12410	2502	3154
Spring Barley	2086	458	631	669	167	182	13514	2701	3808
Rape	1880	921	1394	1532	1312	1243	12244	4144	7267
Peas	3454	2928	6440	2641	1275	3564	23547	14653	26057

High variability in IS_{crop_ha} results within RS and AS2 approaches were observed. Also, the Tukey's range test was conducted to determine statistical differences in the impact assessment of the three modeling approaches. AS2 estimated impact scores were significantly higher than the scores in RS, which did not differ significantly from the AS1 values.

The different modeling approaches to calculate pesticide emission inventory from a given applied quantity (i.e. elementary flows from the product system to the ecosphere) and the connection to the impact characterization have also previously shown to have considerable influence on the estimation of IS for individual AIs and the impact profiles of crop production (Rosenbaum et al., 2015; van Zelm et al., 2014). The consideration of inter-media transfer processes, crop growth development and application method allow for a more accurate estimation of the real phenomena, which are also the aspects that usually have the highest influence on LCI and LCIA models (Dijkman et al., 2012; Fantke et al., 2012).

On the other hand, the consistency showed for trend results (for substance prioritization) of others studies using PestLCI (a more sophisticated emission modeling approach) compared to the RS are satisfactory (see section 3.3). Keeping in mind that such a model is much more data demanding and since IS_{crop_ha} represent potential impacts rather than actual damages, the substance prioritization with a simplified method as the RS may serve as a first proxy in LCA studies when more detailed data are lacking.

Regarding the input parameter sensitivity analysis (performed on several input parameters of the RS and using Maize 2013 and Peas in 2015 as example), the primary sources of uncertainty in the RS are identified as i) the application method and the drift fractions, and ii) the allocation for the off-field emission, specifically the water to soil ratio as shown in Figure 6 (further details, see SI-5). However, the uncertainty range associated with pesticide emissions have not yet been quantified and is beyond the scope of the present study.

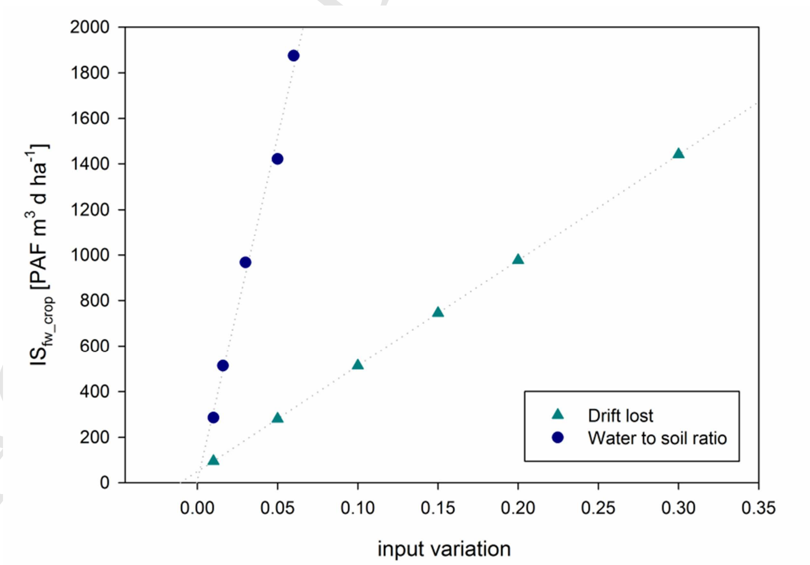

Figure 6 Sensitivity to model input parameters of the reference scenario (RS). Variation for ecotoxicity impact scores (IScrop_ha) expressed as [PAF m3 d ha^{-1}] for the example of Maize in 2013

3.4.2 Influence of choice of LCIA characterization method

The range of variation for the CF of all AI in the study with USEtox 2.02 was almost 9 orders of magnitude. FF and XF vary by near 2 orders of magnitude, while EF varies up to 7 orders of magnitude indicating substantial differences in pesticide-specific ecotoxicity potential. The variation in the CF for direct emissions to surface water, continental air or agricultural soil was near to 10 orders of magnitude, but CF for direct emissions to continental air and agricultural soil was lower than the CF for direct emissions to freshwater (3 and 2 orders of magnitude, respectively). From which, the importance of modeling the impacts of the dose applied, with a coherent coupling of the LCI to the LCIA model results (i.e., characterized results).

Results for IS_{crop_ha} in the RS using USEtox versions 1.0 and 2.02 are summarized in Table 3. Improvements and scientific consensus have been achieved for the new features introduced in the USEtox 2.02 among which substances and updated substance data and continent-specific landscape parameters contribute to further improving the accuracy in the quantification of CFs, given that respective spatial emission data are available. The more substantial differences in the impact results might come from the updated EF or by the influence of considering ionization (acid/bases) in USEtox 2.02.

Table 3 Comparison of scenarios to test developments of LCIA characterization method. Results for potential freshwater ecotoxicity impact scores IS_{crop_ha} in [PAF m3 d ha^{-1}] in the reference scenario (RS) and USEtox version 1.0 and 2.02

Crop	**RS - USEtox 1.0**			**RS - USEtox 2.02**		
	2013	**2014**	**2015**	**2013**	**2014**	**2015**
Maize	246	63	146	491	80	32
Grass	24	12	14	17	11	13
Winter wheat	1349	445	1223	2139	387	508
Spring Barley	758	267	390	2068	435	622
Rape	776	563	702	1880	921	1393
Peas	1483	1893	6080	3454	2928	6440
Glyphosate Agri-use	28	12	17	14	6	8

The uncertainty of CFs (USEtox 2.02) due to emissions to air, freshwater and agricultural soil is 176, 18 and 103 GSD^2 (Rosenbaum, 2016). The major sources of uncertainty are substances half-lives and ecotoxicity EF (Henderson et al., 2011). Without considering winter wheat-2015, in general, ISs characterized with the improved USEtox 2.02 are higher than the results obtained with the previous version. Furthermore, in comparison with the FF and XF, the EF shows a substantial variation among the AI in this study, explaining a large part of the variations in the CFs for the AI after emissions to freshwater.

4 CONCLUSIONS

The combination of the emission modelling scenario RS, shown in the present study, and the characterization model USEtox 2.0 has allowed to recognize trends of different pesticides treatments and burdens on freshwater ecosystems, thus accounting for interactions between different compartments and a defined clear interface between LCI and LCIA. LCI modeling options do affect the ecotoxicological burden on freshwater ecosystems from pesticide use, and directly affects substance prioritization in LCA studies. Furthermore, the updated CF with the continent-specific landscape parameters contributes to a broader assessment. In the case of scenario and sensitivity analysis, application method and allocation for the off-field emission were identified as the main descriptors for modeling emissions of pesticides. The use of the modeling framework presented in this study allows for delivering more robust results and accurate evaluation of ecotoxicity impacts. Finally, to provide consumers and policymakers with more reliable information on the environmental performances of agricultural systems, LCA studies need to include all relevant emission outputs; therefore, a final consensus needs to be reached with a specific emission model recommendation.

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APPENDIX A. SUPPORTING INFORMATION

The following is the supplementary material related to this article. Detailed information of scenarios, physicochemical properties, and data on pesticide active ingredients, further annotations on pesticide emission quantification, data and sources for the derivation of new CFs, as well as supporting materials for results and sensitivity analysis included in the study are provided in the Supporting information.

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HIGHLIGHTS FOR:

FRESHWATER ECOTOXICITY ASSESSMENT OF PESTICIDE USE IN CROP PRODUCTION: TESTING THE INFLUENCE OF MODELING CHOICES

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- Pesticide use is not an apt indicator for impact as toxicity may be higher for pesticides applied in low rates
- A suitable interface between LCI and LCIA and related mass distribution for pesticides is framed
- Crop growth development and application method have major influence on the emission model
- Substance ranking with a simplified modeling framework serve as proxy in LCA studies when detailed data lacks