

# Demonstrating an approach for including pesticide use in life-cycle assessment: Estimating human and ecosystem toxicity of pesticide use in Midwest corn farming

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## Abstract

**Purpose** This study demonstrates an approach to assess human health and ecotoxicity impacts of pesticide use by including multiple environmental pathways and various exposure routes using the case of corn grown for bio-based fuel or chemical production in US Midwestern states.

**Methods** Multiple tools including an environmental emission model (PestLCI), an impact analysis tool (USEtox), and additional databases were utilized to estimate the state-specific pesticide releases and their associated spatially explicit toxicity in Midwest states.

**Results and discussion** On average, chlorpyrifos and acetochlor exhibit the highest human toxicity potential (HTP) and the highest ecotoxicity potential (ETP) impact scores, respectively. The different ranking orders of pesticides for human health and ecosystem toxicity suggest that there are tradeoffs between these two impact categories. While the air pathway can account for 10–97 % of HTP, the water pathway is the dominating contributor for ETP for most of the

pesticides. Moreover, while chlorpyrifos, fipronil, 2,4-D-2-ethylhexyl ester, simazine, and glufosinate–ammonium together account for more than 80 % of HTP per kilogram harvested corn, acetochlor is the dominating contributor in ETP due to its high ecotoxicity characterization factor and high application rates for corn. In addition, the spatial variation analysis shows that South Dakota and Missouri are the states that have the highest HTP (per kg corn), while Kansas exhibits the highest ETP (per kg corn) among Midwest states.

**Conclusions** HTP and ETP exhibit large variations across various pesticides, US states, and application times. While chemical properties and toxicity characteristics can result in up to five orders of magnitude of variation in HTP and ETP, the rest of the parameters (such as application times, soil properties, and climate conditions) can affect the results by up to two orders of magnitude.

**Keywords** Corn · Ecotoxicity · Human health · Life-cycle assessment · Midwest · Pesticide

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## 1 Introduction

While modern agriculture relies on the use of pesticides to protect crops and ensure production, pesticide use can result in undesirable environmental and human health effects. The use of synthetic pesticides in agriculture is the most widespread method for pest and weed control. More than 50 active pesticide ingredients have been applied to crops in the last decade (USDA NASS 2012) in the USA. Meanwhile, pesticides were considered as one of the leading causes of impairment for streams included on US Clean Water Act section 303 list of impaired waters (USEPA 2009). A recent national pesticide survey detected one or more pesticides in several major rivers, streams, and aquifers in the US Midwest (Sullivan et al.

2009). Agricultural production is the first stage for a variety of bio-based fuels and chemicals. With the continuously increasing demand of fuels supported by legislation at national and state levels, the ecotoxicity and human health impacts induced by pesticide use for biofuel feedstock production calls for environmental and toxicity modeling to aid biofuel-related decision making.

Life-cycle assessment tracks environmental releases associated with a process or product from cradle to grave and quantifies a suite of environmental and human health impacts (ISO 2006). Life-cycle release inventory data (LCI) and life-cycle impact assessment (LCIA) methods have been developed for pesticides in previous studies (Pennington et al. 2005; Birkved and Hauschild 2006; Humbert et al. 2007; Landis et al. 2007; Nemecek and Kagi 2007; Juraske et al. 2009; Berthoud et al. 2011; Juraske and Sanjuán 2011; Fantke et al. 2011a, b, 2012; Dijkman et al. 2012; Ingwersen 2012; Vázquez-Rowe et al. 2012). The pesticide releases to various environmental compartments are compiled in the LCI step. During LCIA, the associated environmental and human toxicity are estimated based on impact assessment tools.

The majority of the previous pesticide life-cycle inventories have either applied a linear approach or assumed a zero value for estimating the pesticide releases to water and air environmental compartments in the USA (Landis et al. 2007; Nemecek and Kagi 2007). In addition to the incompleteness of pesticide release inventories and failure to close mass balances, previous studies have not characterized spatial variability of pesticide releases at the US state level. The majority of the reported studies provided a single value or range for pesticide release over a large spatial span such as the US Midwest, without explicitly considering the influence of soil characteristics, climatic conditions, and farming practices. The recent pesticide emission estimation model, PestLCI, is capable of calculating edge-of-field pesticide losses during and after application to multiple environmental compartments, based on pesticide, crop, climate, soil data, and farming practices under the principal of mass balance (Birkved and Hauschild 2006; Dijkman et al. 2012). However, the application of PestLCI to North America conditions has not been found yet.

LCIA approaches have been used to quantify the ecotoxicity and human health impacts of commonly used pesticides in France, Switzerland, Spain, Costa Rica, Western Europe, and other regions through USEtox and its predecessors (Pennington et al. 2005; Humbert et al. 2007; Juraske et al. 2009; Berthoud et al. 2011; Juraske and Sanjuán 2011; Vázquez-Rowe et al. 2012). Berthoud et al. (2011) assessed freshwater ecotoxicity of pesticide releases in wheat production by combining the French agricultural practices database and the USEtox model. Juraske et al. (2009) compared cumulative health effects from ingestion of pesticides in fruits and vegetables in Switzerland and the USA. Juraske and Sanjuán (2011) compared 25 pesticides used

for orange production in the Valencia region of Spain and found that preselection of pesticides could mitigate human health and ecotoxicity impacts. Using Impact 2002, Pennington et al. (2005) presented a spatially resolved multimedia and multipathway exposure model for Western Europe and found that exposure models without considering local specifics could result in significant underestimation or overestimation of chemical intake and associated human health impacts. Recently, Fantke et al. (2011a, b, 2012) developed a dynamic model (called dynamiCROP) to quantify environmental fate and toxicity of pesticides for various crops, vegetables, and fruits. Distinct from the USEtox model, the dynamiCROP model is capable of estimating the pesticide toxicity due to the exposure of pesticide residue on foods. However, this study focuses on corn farming for bio-based fuel or chemical production and not on food crop production.

In addition, recognizing the disconnection between tools for detailed pesticide life-cycle inventory and impact assessment, recent efforts have been directed at coupling the detailed pesticide emission estimation model and an impact assessment tool (Vázquez-Rowe et al. 2012; Ingwersen 2012). The studies presented by Vázquez-Rowe et al. (2012) and Ingwersen (2012) have used the USEtox model coupled with life-cycle pesticide inventory models to evaluate human health and ecotoxicity impacts of pesticide use in fruit production. In our literature search, we did not find examples assessing ecotoxicity and human health impacts of pesticide use in US crop production through combining state-specific pesticide release inventory and life-cycle impact assessment. Life-cycle studies on the state-wide pattern of pesticide releases and associated human health impacts in US corn production were not found in our literature search.

In this study, we modeled the pesticide releases and assessed their human health and ecotoxicity impacts for corn farming in the Midwest states, defined here as Illinois, Indiana, Iowa, Kansas, Minnesota, Missouri, Nebraska, Ohio, and South Dakota. The Midwest states produced more than 80 % of the total corn crop in the US in 2010 (USDA NASS 2012). The pesticides researched in this study include both herbicides and insecticides. Managing weeds and pests in corn requires a relatively larger amount of pesticide application than any other US staple crop (USDA NASS 2012). In 2006, corn growers applied 458 million kg of pesticide for corn production (USDA NASS 2012). A large amount of corn (42 %) was used for animal feed, and 13 % of corn was used for food, seed, and industrial uses in 2012 (USDA ERS 2012). The rest of the corn (45 %) was used as feedstock for producing ethanol. The US domestic use of corn for fuel production has increased by 120 % from 2006 to 2012 due to the continuously increasing demand of fuels. This study was conducted as part of an effort to understand the impacts associated with corn production for use in ethanol production. The Energy Independence and Security Act of 2007 provides incentives

for achieving an annual production rate of 56.8 billion L of conventional ethanol (15 billion gallon/year) by 2015 (USEPA 2012). At present, nearly all of this is produced from corn grain. Properly accounting for the effects of corn production on human health and ecosystems is important for properly managing pesticide use in corn farming and minimizing environmental effects and public health risks of corn-based fuel.

This study aims to assess human health and ecotoxicity impacts of pesticide use by including multiple environmental pathways and various exposure routes using the case of corn grown for bio-based fuel or chemical production in US Midwestern states. The contribution of this study is to demonstrate an approach to assess potential human health and ecotoxicity impacts across a suite of exposure routes of pesticides in a manner consistent with LCA approaches. Additionally, this study adds value to existing studies by (a) compiling transparent environmental release inventories of pesticide use for corn farming for bio-based fuel or chemical production in the Midwest, (b) addressing the influence of application times, soil properties, and climate conditions on environmental releases and toxicity impacts for multiple pesticide types in the Midwest, and (c) evaluating the human health and ecotoxicity impacts of pesticide use in corn farming for bio-based fuel or chemical production.

## 2 Methods

### 2.1 Overview of the approach

PestLCI was used in conjunction with USEtox to characterize human health effects and ecotoxicity of pesticides applied to Midwest corn for bio-based fuel or chemical production. The PestLCI model was modified as detailed below to better represent US conditions and farming practices, and then the edge-of-field flows from PestLCI were used as inputs to USEtox for characterization of toxicity. USEtox is not spatially explicit and only considers far-field exposure (not exposures through direct use or ingestion of a product containing a toxin), but as the consensus model for toxicity, provides a common standard for comparing toxicity effects of emissions. Use of a standard exposure and toxicity characterization model was judged to be more important than using other LCIA toxicity models (such as Impact 2002) that allow spatially explicit characterization of human and ecological exposure once emissions have been estimated, but that report vast differences between other impact characterization models (Rosenbaum et al. 2008). The USEtox database was amended to include the pesticides considered in this study when found missing. Additional model details are provided in the sections that follow.

Results represent five basic perspectives on pesticide use: (1) results per kilogram pesticide applied address questions related to the relative toxicity of pesticides, (2) results per

hectare of treated land (i.e., only hectares treated with the specific pesticide) provide a means for characterizing results in terms of efficacy (as no more direct metric was available for incorporation in our functional unit), (3) results per kilogram of corn production provide results from the perspective of the final product, (4) state-specific results provide a first step toward addressing the issue of geographic variability, and (5) impact potentials due to various chemical properties and state-specific parameters address variations in these properties and parameters.

In this study, we selected a representative sample of pesticides based on their usage in Midwest corn farming. The most common active ingredients in pesticides for Midwest corn production are acetochlor, chlorpyrifos, glyphosate, atrazine, S-metolachlor, fipronil, alachlor, dimethenamid-*P*, 2,4-D-2-ethylhexyl ester, glufosinate-ammonia, 2,4-D-dimethylammonium, and simazine (USDA NASS 2012). This set includes two insecticides and ten herbicides, which account for more than 95 % of total pesticide use by mass in the US Midwest in 2006. Year 2006 was selected because it was the most recent year when the USDA pesticide application datasets were available.

### 2.2 PestLCI and model modifications

Based on pesticide, crop, climate, soil data, and information regarding farming practices, PestLCI estimates the fractions of the applied amount taken up by the plant, degraded, and released to air, surface water, and groundwater (Birkved and Hauschild 2006). Pesticide pathways modeled in PestLCI include direct emissions to air by wind drift, volatilization from leaves and top soil, photochemical oxidation on leaves, plant uptake, biodegradation, runoff to surface water, and leaching of pesticide through soil to groundwater. PestLCI integrates degradation, loss, and uptake processes in soils and on leaf surfaces at half hour and hourly time steps, respectively. In prior studies for European conditions, PestLCI has been used to quantify pesticide releases (Hellweg and Geisler 2003; Ometto et al. 2009; Cerutti et al. 2011; Mattila et al. 2011; Vázquez-Rowe et al. 2012), and the standard PestLCI model includes pesticides commonly used in European and Danish soil and climate conditions. Birkved and Hauschild performed a parameter sensitivity analysis (Birkved and Hauschild 2006) and found that the PestLCI edge-of-field release fractions are sensitive to soil properties and meteorological conditions such as relative humidity, solar radiation, and monthly rainfall, which are not commonly gathered in LCA studies. While PestLCI has been developed and applied to understand European conditions, to date, it has only had limited application outside Europe. To adapt PestLCI to US conditions, we made three modifications to PestLCI: (1) added characteristics of ten pesticides commonly applied to corn in the USA, (2) added meteorological data, and (3) added soil characteristics

specific to each of the Midwestern states where the majority of US corn is grown. In addition, we modified the PestLCI spreadsheet model to create a version capable of performing calculations for up to 50 scenarios simultaneously to allow for efficient variability analysis.

Data was added to the PestLCI substance database for ten of the 12 pesticides considered with the exceptions being glyphosate and fipronil, which were already available in the original model. Physiochemical properties including molecular weight, molecular volume, solubility, vapor pressure,  $pK_a$ ,  $K_{ow}$ ,  $K_{oc}$ , and Henry's Law constant were gathered from the USEtox database (Rosenbaum et al. 2008, 2011; Henderson et al. 2011). The remaining datasets including half-life of soil degradation and atmospheric hydroxyl oxidation rate were collected from the Hazardous Substance Data Bank (Toxnet 2011). We extracted and implemented climate data from the North America Regional Reanalysis database (NARR 2011), including daily maximum and minimum temperatures, precipitation, solar radiation, wind speed, and relative humidity. Evapotranspiration values for each US state were calculated using the Penman–Monteith equation (FAO 2011). We used average state-level Soil Survey Geographic data from the USDA to provide soil physical and chemical properties such as slope gradient, number of soil layers, depth of soil layers and bulk density, sand fraction, silt fraction, clay fraction, organic carbon content, and pH (SSURGO 2011).

In order to facilitate the efficient evaluation of a large number of scenarios, we modified PestLCI by duplicating the results displayed across multiple spreadsheet columns and then propagating these changes throughout the linked calculations. This allowed us to model pesticide releases under various application time scenarios for each state and to test the variability of results to multiple parameters. Since the USDA survey does not specify application time, we included scenarios to capture the release variability due to the theoretical choice of application time. The examined scenarios represent pesticide application during leaf development, stem elongation, inflorescence emergence, and development of fruits/ripening phases. The simulation results for all scenarios are reported in the [Electronic Supplementary Material \(ESM\)](#).

### 2.3 Calculation of pesticide characterization factors

USEtox is a model for comparative assessment of human health and ecotoxicity impacts of chemicals in conjunction with LCA modeling which was developed under the task force on toxic impacts under the UNEP-SETAC Life Cycle Initiative (Rosenbaum et al. 2008, 2011). USEtox models successive calculation steps for factors including fate in days, exposure in  $\text{day}^{-1}$ , and effect in cases/kilogram intake for human toxicity to obtain the product human health characterization factors (CF) expressed in cases per kilogram emitted, which are intended for use in connection with LCA studies.

Ecotoxicity is calculated in terms of PAF·cubic meter·day per kilogram (where PAF is the potentially affected fraction of species). The full details of the USEtox model are publicly available on <http://usetox.org>. USEtox v1.01 contains characterization factors (CF) for nearly 4000 chemicals and includes two main exposure routes: ingestion and inhalation. Since  $ED_{50}$  values (defined as lifetime daily dose resulting in a probability of effect of 0.5) of researched chemicals are not available in the present version, we followed the extrapolation procedure for adding chemicals described in the USEtox documentation (Rosenbaum et al. 2008, 2011) to estimate both carcinogenic and noncarcinogenic characterization factors of the pesticides. The added chemicals include dimethenamid-*P*, 2,4-D-dimethylammonium, glufosinate–ammonium, 2,4-D-2-ethylhexyl ester, alachlor, fipronil, *S*-metolachlor, atrazine, glyphosate, and acetochlor (Table S3, ESM). NOEL values (defined as no observed effect level) obtained from the IRIS databases were used in the extrapolation. The justification of this extrapolation approach is discussed in the USEtox reports (Rosenbaum et al. 2008, 2011).

Estimates of CFs for human health and ecotoxicity are presented in Table S3 in the ESM. While the highest ecotoxicity value is  $2.99 \times 10^8$  PAF·cubic meter·day per kilogram for acetochlor, the lowest freshwater ecotoxicity value is  $1.25 \times 10^{-2}$  PAF·cubic meter·day per kilogram for simazine. The human health CFs of pesticides vary by up to 12 orders of magnitude due to differences in their environmental fate, human exposure routes, and inherit toxicity characteristics. Chlorpyrifos, one of the most popular insecticides in Midwest corn farming, has the highest human health CF value. In contrast, acetochlor, a widely used herbicide for multiple crops, has lower human health CF values among those studied here. In the case of freshwater ecotoxicity, the characterization factors vary by up to ten orders of magnitude.

### 2.4 Calculation of human health and ecotoxicity impacts

The toxicity impact score per kilogram of pesticide is calculated by multiplying the mass of substance released to a given environmental compartment as provided by the output of PestLCI with the corresponding toxicity characterization factor provided by USEtox,

$$S_x = \sum_i M_{x,i} CF_{x,i}$$

where  $S$  is the ecotoxicity or human toxicity impact score,  $M_{x,i}$  (kg) is the amount of substance  $x$  released to compartment  $i$ , and  $CF_{x,i}$  (cases per kilogram or PAF·cubic meter·day per kilogram) is the characterization factor (CF) of substance  $x$  released to compartment  $i$ . The integration of model results and the spatial resolution of model outputs are described in Fig. S1 in the ESM.

## 2.5 Alternative functional units and pesticide application rates

The choice of functional unit is important for the outcome of any LCA study. For example, Matheys et al. (2007) discussed the effect of functional unit on the outcomes of LCA of batteries. In the case of pesticides, while kilograms applied provides the most easily measured quantity, it fails to address relative effectiveness in mitigating insects or weeds or the ultimate implications for corn yields. As we were unable to identify a pesticide effectiveness metric and as the effect of pesticide on corn yield is confounded by many other factors, we calculated results on the basis of three different functional units to provide multiple perspectives. The simplest functional unit based on kilograms pesticide is useful for translating application rates into associated impact potentials. In order to address effectiveness, we provide results based on the functional unit of area of treated corn farmland to reflect differences in application rates. Finally, a functional unit based on corn produced incorporates the influence of harvesting ratio of corn fields and relates to the purpose of agricultural pesticide use. This final functional unit based on corn produced also allows for incorporation of our results into broader assessments involving corn production for bio-based fuel or chemical production.

The application rate of active pesticide ingredient can be calculated as an overall regional average per unit of corn production or with respect to only the land area treated with a specific active ingredient. The application rates per hectare and per kilogram corn were calculated using USDA application rates for corn production in 2006. The mean value of application rates of studied pesticides is  $0.021 \text{ mg}(\text{kg corn})^{-1}$  or  $170 \text{ mg}(\text{treated hectare})^{-1}$  with values ranging from  $0.013$  to  $0.37 \text{ mg}(\text{kg corn})^{-1}$  or  $51$  to  $330 \text{ mg}(\text{treated hectare})^{-1}$ . Application rates associated with the functional units used in developing the results are provided in Table S1 in the ESM.

## 3 Results

The results presented below are organized in the following sections: (1) pesticide releases based on PestLCI model, (2) human toxicity (HTP) and ecotoxicity potential (ETP) for the three different functional units based on the combination of PestLCI and USEtox, and finally, (3) state-by-state total impact potentials per kilogram corn produced.

### 3.1 Pesticide releases to various environmental compartments

The release fractions of pesticides to environmental compartments are shown in Fig. 1. The average fractions emitted to the air compartment range from 9 to 59 %, while the fraction

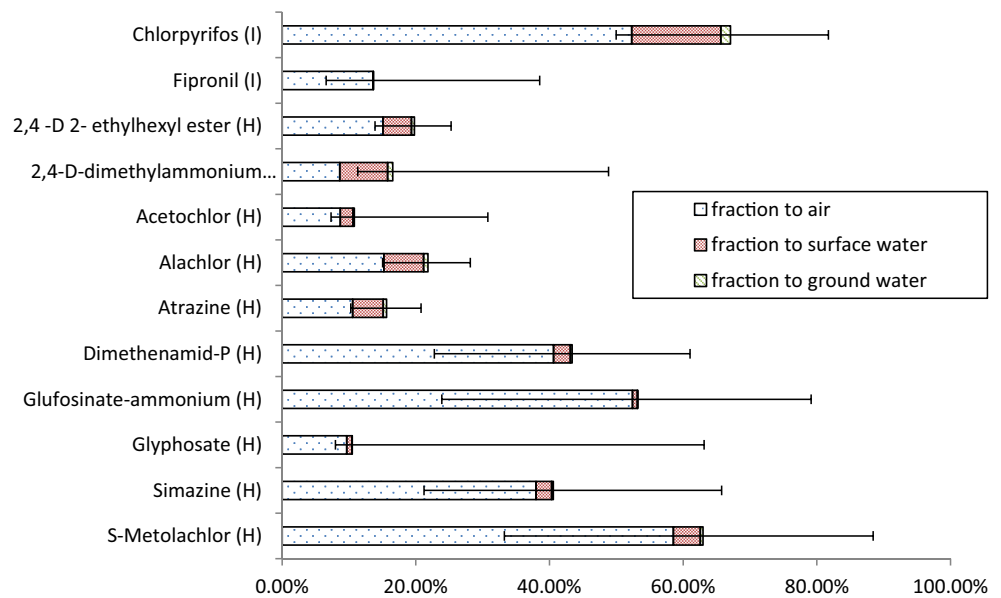
reaching the ground and surface water compartments varies from 0.8 to 15 %. Comparing our study with a set of standard air emission factors (USEPA 1994), we find that our air emission estimates for atrazine, acetochlor, and metolachlor are around 5 % higher than the previously reported standard emission factors. The difference is likely due to the inclusion of substance dispersion by wind drift, which is not considered in the previously reported emission factor (USEPA 1994). Water releases from atrazine and metolachlor use in the Midwest were reported previously (Ng et al. 1995; Gaynor et al. 2001) based on multiple field and modeling studies, which found 2.5–15 % of atrazine and metolachlor applied together in the Midwest was discharged into the combined runoff. This overlaps with our results for atrazine and *S*-metolachlor of 1.4–16 %.

The pesticide emissions vary with chemical physiochemical properties, application times, and soil characteristics. 2,4-D-Dimethylammonium and alachlor, with relatively high solubilities, exhibit high water release fractions. *S*-Metolachlor, with relatively high volatility, showed the highest air release fraction of 58 %. PestLCI estimates for pesticides applied during the postemergence phase result in higher air emission fractions and lower water release fractions relative to earlier growth phases such as preemergence and stem-elongation. The air release fraction for acetochlor applied during the post-emergence growth phase can be eight times higher than the fraction for the same chemical applied during the pre-emergence growth phase (Fig. S2 in the ESM). This difference is associated with leaf interception and subsequent volatilization. PestLCI model results suggest that higher leaf interception occurred during postemergence phase than pre-emergence phase. For most of the studied pesticides, the release to groundwater in sandy soil conditions is likely higher than the release in silty and loamy soil conditions (Fig. S4 in the ESM).

### 3.2 Pesticide human toxicity and ecotoxicity potentials in the US Midwest by amount of pesticide applied, corn production, and treated area

Figure 2 presents the average human health and ecotoxicity impacts on the basis of three relevant functional units each of which provides a distinct perspective. As shown in Fig. 2a, chlorpyrifos, an insecticide with the highest human health CF, presented the highest HTP per kilogram pesticide. Simazine and glufosinate-ammonium exhibit higher HTP when compared to other herbicides due to the combined effect of relatively higher CFs and corresponding release rate to air. From Fig. 2b, acetochlor has the highest ecotoxicity potential, followed by chlorpyrifos. The comparison between Fig. 2a, b suggests a tradeoff between human health and ecotoxicity rankings of pesticides. While chlorpyrifos ranks highly for both HTP and ETP per kilogram, many of

**Fig. 1** The fraction of pesticides applied in the US Midwest released to environmental compartments. *Whiskers* represent the overall minimum and maximum values of release fractions to environmental compartments. The remainder of pesticides applied are taken up by plants, degraded by bacteria, or fixed in soil on the field. *I* insecticide, *H* herbicide



the other pesticides are ranked much higher or lower in one impact category. One can examine the pathways by which these pesticides generate potential impacts. For instance, releases to the air compartment account for 10 to 97 % of HTP. Releases to the water compartment contribute more than 80 % to ETP and are two orders of magnitude larger than the air compartment contribution in all cases except glyphosate and 2,4-D-dimethylammonium. The noncarcinogenic impact accounts for 7 to 100 % of pesticide human health impacts for the assessed pesticides. While noncarcinogenic impact contributes more than 80 % of HTP for simazine and glufosinate–ammonium, the noncarcinogenic impact contributes less than 30 % of HTP for the rest of the assessed pesticides. Figure S5 in the ESM illustrates the relative contribution of each environmental compartment to HTP and ETP.

Figure 2c, d presents impacts of pesticide use in Midwest corn cultivation per kilogram corn. Chlorpyrifos, fipronil, simazine, and glufosinate–ammonium exhibit higher HTP than the rest of the pesticides, owing to the combined effect of high application rates and high CFs. Similarly, acetochlor ranks as the largest contributor to ETP due to a combination of high application rate and CF. Comparing the impacts of acetochlor along various pathways indicates that its release to water is the main contributor for ecotoxicity impacts (Fig. S5, ESM).

Figure 2e, f presents results per hectare treated corn farmland. Chlorpyrifos exhibits the highest human toxicity and acetochlor the highest ecotoxicity. The ranking differentiation between results per hectare treated farmland and results per kilogram corn reflects various treated rates of corn farmland for pesticides. For example, a relatively lower percent of corn farmland has received simazine and glufosinate–ammonium compared with the rest of the herbicides; therefore, their relative impacts per hectare treated corn farmland are higher than their relative impacts per kilogram corn.

LCA practitioners who are performing an LCA that includes corn farming may find the extensive results tabulated in the SI valuable. The user can find application rates, soil properties, and climate conditions that are similar or conservative to use as a first approximation in other studies. The approximation can save significant time and effort and be used to compare to toxicity results from other life-cycle stages. For those considering corn as food, it will still be necessary to add this exposure route into the analysis.

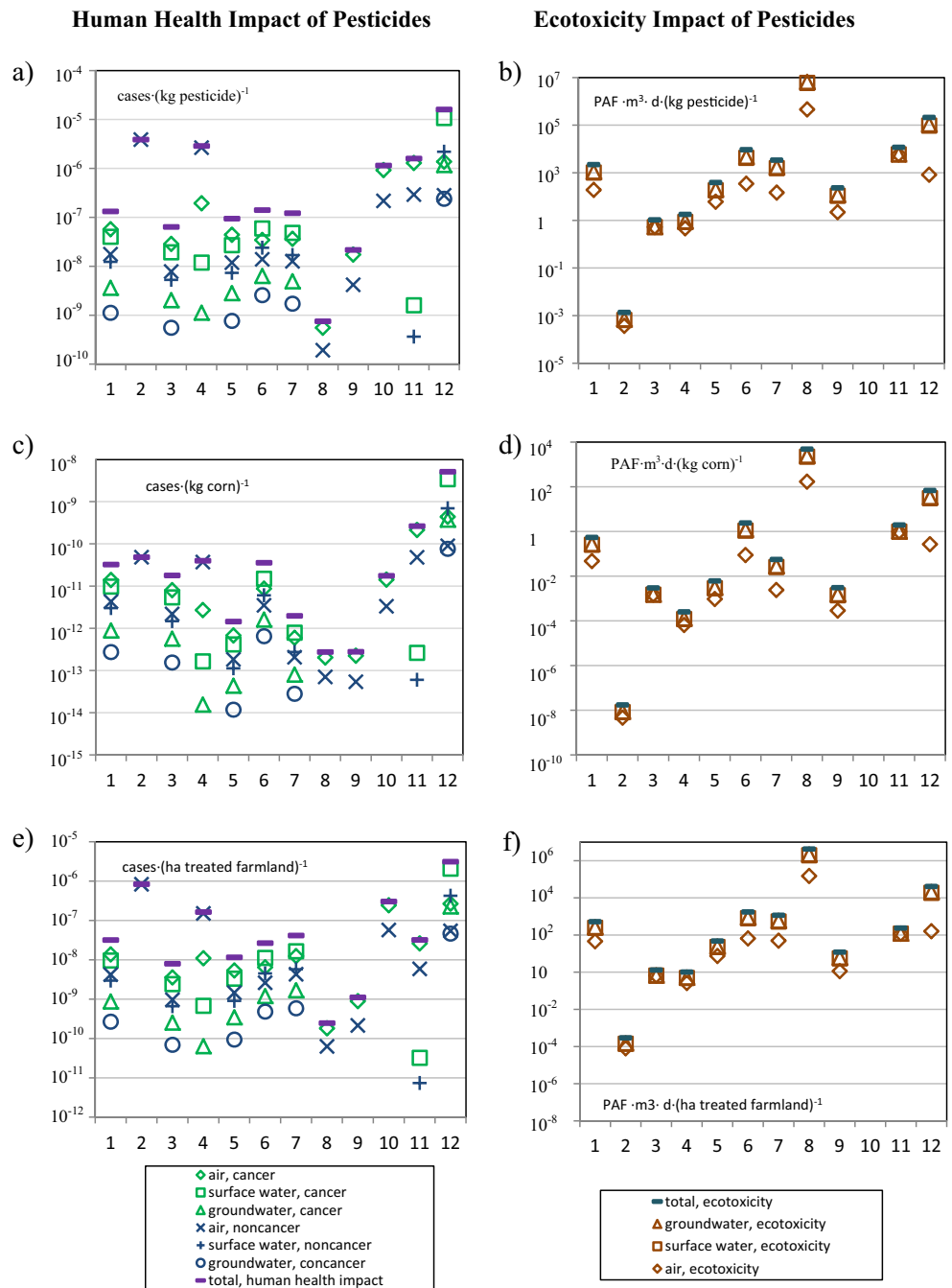
### 3.3 State-specific impact potentials per unit corn production across states

The aggregated HTP and ETP per unit corn production in each state are plotted in Fig. 3a, b based on pesticide usage in 2006. The upper and lower bounds represent the maximum and minimum values of impact potentials, respectively. The whiskers in Fig. 3a, b represent variations in on-field partitioning caused by application time, local soil types, and climate conditions within the Midwest states. South Dakota, Missouri, Iowa, and Illinois are the states which have the highest human health risk due to their large applications of chlorpyrifos with chlorpyrifos having the highest human health CF. In contrast, Indiana, Kansas, and Ohio have the lowest HTP because of significantly lower application rates of insecticides including chlorpyrifos and fipronil. Kansas exhibits the highest ETP among Midwest states because of its relatively high usage of acetochlor.

### 3.4 Variation of impact potentials

The human health and ecotoxicity impacts per kilogram of applied pesticides exhibit significant variability (Fig. S6 in the ESM). The HTP varies by up to five orders of magnitude

**Fig. 2** The average human health and ecotoxicity impacts of pesticide use in Midwest corn farming. The graphs in the left column represent the human health impacts per kilogram pesticide, per kilogram corn, and per hectare corn farmland, respectively. The graphs in the right column represent the ecotoxicity impacts per kilogram pesticide, per kilogram corn, and per hectare corn farmland, respectively. *S*-Metolachlor (1), simazine (2), glyphosate (3), glufosinate-ammonium (4), dimethenamid-*P* (5), atrazine (6), alachlor (7), acetochlor (8), 2,4-D-dimethylammonium (9), 2,4-D-2-ethylhexyl ester (10), fipronil (11), and chlorpyrifos (12)

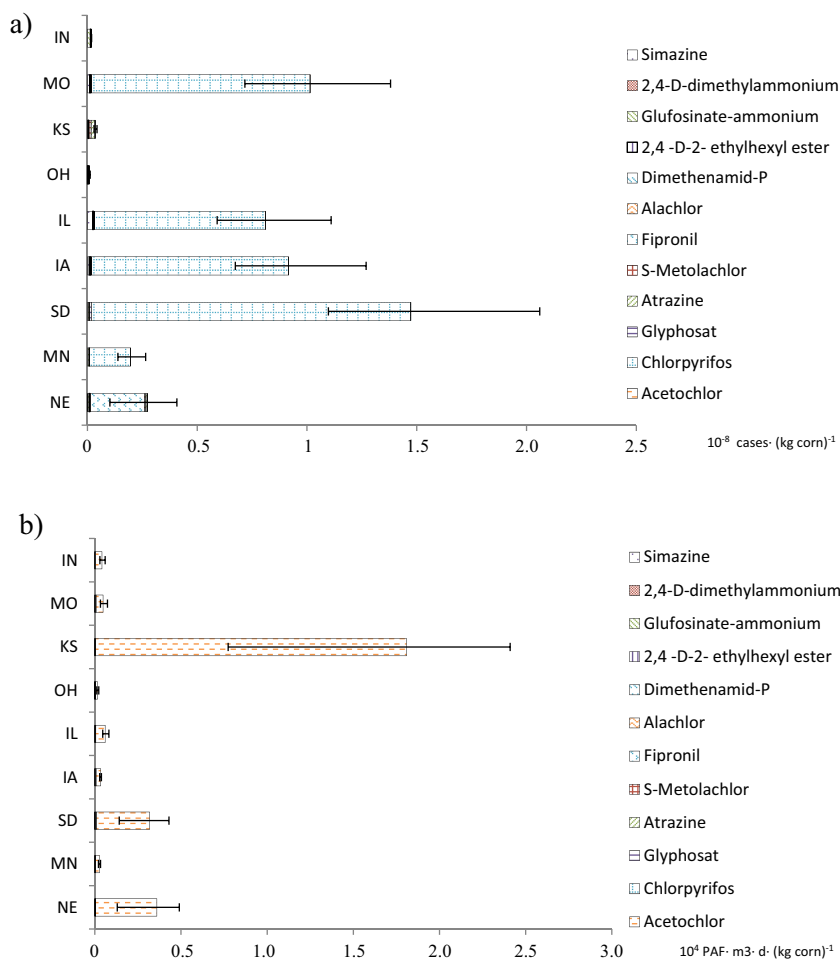


within a state for different pesticides and up to two orders of magnitude for a given pesticide across the nine states. The ETP per kilogram pesticide ranges from  $10^{-12}$  to  $10^7$  PAF·cubic meter·day per kilogram across pesticides and states, while variation for a given pesticide across states is within two orders of magnitude. Among the researched pesticides, chlorpyrifos exhibited the greatest variability for HTP. In contrast, acetochlor showed the greatest variability for ETP. The variability originates from various physiochemical properties, climate parameters, soil properties, and application times. Our results indicate pesticide characteristics such as Henry’s Law

constant and effective dose contribute more to variation than geographic factors such as soil type and weather conditions (Table S5, ESM).

Due to the causal link between chemical environmental fate and human exposure in the USEtox model, the pesticide environmental releases are positively correlated with the ingestion and inhalation rates of pesticides. The HTPs of chemicals with low Henry’s Law constants, such as atrazine, alachlor, and chlorpyrifos, are proportional to their aqueous releases (Pennington et al. 2005). For example, the high aqueous release rate of chlorpyrifos causes high

**Fig. 3** Aggregated **a** HTP and **b** ETP of pesticide use for corn farming in each Midwest state. *Whiskers* represent the minimum and maximum values associated with variations in on-field partitioning within each state. The variability reflects the influence of location-specific parameters on pesticide releases



ingestion rates of chlorpyrifos, consequently resulting in relatively high human health impact potentials of applied chlorpyrifos within Midwest states (as shown in Fig. S6, ESM). In contrast, for organic chemicals with high Henry's Law constants, which volatilize to a greater extent, such as 2,4-D-2-ethylhexyl ester, the exposure pathway is predominantly associated with inhalation. Thus, the high HTPs of chemicals with high Henry's law constants are associated with their relatively large emissions and subsequent inhalation exposure.

## 4 Discussion

### 4.1 Strengths and limitations of tools and methods

The results presented here provide insight into the strengths and limitations of the combined use of PestLCI and USEtox for life-cycle assessment. Both tools are flexible, transparent, user friendly, and publicly available for further adaptation. PestLCI provides plausible estimates of pesticide releases to air and water environmental compartments while deliberately simplifying certain processes to keep the estimation

manageable. Although PestLCI captures the releases to multiple compartments, it excludes some possible environmental pathways, including leaf wash-off contribution to surface water and groundwater releases, and uptake by plant roots. However, considering the uncertainty of toxicity estimates in impact assessment tools, PestLCI is appropriate for estimating release inventories and represents a significant improvement over static emission factors. In addition, we verified our estimates of pesticide releases through comparison with data sets published by EPA and peer-reviewed studies (USEPA 1994; Ng et al. 1995; Gaynor et al. 2001). The user should be aware that the analysis represents pesticide use for corn farming in 2006, and appropriate modifications and validation are required for other years. We acknowledge that findings are subject to specific model and data uncertainties and recommend field experiments to further test the model robustness for various pesticides across Midwest farms. In addition, this study did not include fungicides due to data limitations. It is recommended to expand the fungicide database for the future toxicity assessment of corn farming.

The USEtox method provides holistic impact analysis through providing multiple environmental media and incorporating a large number of chemicals at a moderate level of



complexity within a single framework. On the other hand, the current USEtox model only considers ingestion and inhalation exposure pathways while excluding certain potentially important exposure pathways, such as ingestion of directly applied pesticide residue on foods, increased exposure due to proximity to the source (i.e., farmers handling and spraying pesticides), and dermal exposure. These could be dominant exposure pathways in specific situations for fruits and vegetables or for special populations such as farm workers. Tailoring human exposure models for specific conditions might help to estimate the health impacts for farmers and other populations, but customization of USEtox applies to the continental/national level scale which was not relevant for showing the state level distinctions here. USEtox quantifies the aggregated human carcinogenic and noncarcinogenic impacts. The tool does not calculate the risk of specific illness (such as lung cancer and others) associated with pesticides. The USEtox model estimates the impacts of a single chemical at a steady state and does not consider the synergistic impacts of the occurrence of multiple chemicals. As our understanding of multipollutant impacts increases, this is an aspect of USEtox which could certainly be improved. Overall, USEtox is a stable transparent tool offering reliable comparative analyses of toxicity impacts.

While in this study, we considered three alternative functional units, kilogram pesticide, hectare treated farmland, and kilogram corn, to compare human and ecosystem toxicity, the most appropriate functional unit could be equivalent units of pesticide efficacy. The development of an efficacy metric, which could be used to compare across pesticides, would be a valuable contribution for future research. Such an efficacy factor is difficult to determine, as the effectiveness of pesticides is influenced by a number of factors including application level, application time, targeted pests or weeds, and environmental conditions (such as soil pH value).

#### 4.2 Implications for pesticide use management

The historical trend of pesticide usage indicates that pesticides with high human health impacts have been gradually substituted with pesticides with similar function and lower human health impacts. From 1990 to 2000, the triazine herbicide atrazine has alone accounted for about 30 % of all corn herbicide-treated area and about 35 % of total mass applied. The acetanilide herbicides alachlor, acetochlor (alachlor was largely replaced by acetochlor in 1994–1995), metolachlor, and *S*-metolachlor (metolachlor was replaced by *S*-metolachlor in 1998–2000) have together accounted for another approximately 30 % of total acres treated and over 40 % of total mass applied (USDA NASS 2012). Together, atrazine and the acetanilides have been applied to approximately 60 % of herbicide-treated area and account for over 75 % of use when measured as mass applied for corn production.

In order to minimize the adverse impacts of pesticide use, both human health and ecosystem impacts need to be considered. The possible tradeoffs between health and ecotoxicity impact categories might result in difficulties to achieve risk reduction goals for both impacts. For example, acetochlor has the lowest human health and highest ecotoxicity impact scores among the studied pesticides. Thus, replacing another pesticide with acetochlor reduces HTP at the detriment of ecosystems.

In conclusion, we find that the use of PestLCI and USEtox in series offers an approach to evaluate pesticide release inventory and toxicity impact, while taking into account multipollutant effects, pesticide efficacy, additional environmental pathways, and exposure routes provides important opportunities for further development of these tools and LCA approaches. HTP and ETP across various pesticides, states, and application times exhibit significant variation. The chemical and toxicity characteristics of pesticides play a more influential role than application times, soil properties, climate conditions, and farming locations. While our results may imply undesirable consequences that might be avoided through selecting less toxic formulations, the expectation for reduced impacts should be tempered by other agronomic and ecological factors such as cost, efficacy, and pesticide resistance and weighed against the potential for tradeoffs between human and ecosystem effects. The decision to use a formulation for a given application is often based on a range of factors such as pest biology, pesticide resistance, efficacy, applicator safety, availability of pesticides and application equipment, cost, and environmental and health considerations, where often, cost, efficacy, availability, and resistance are the primary considerations. In the future, pesticide LCA, coupled with risk assessment at farm and watershed levels, and field agronomic experiments are suggested for multi-criteria decision making regarding pesticide use.

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