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Evaluation of the environmental impacts of agricultural systems using life cycle thinking,  
focusing on marginal changes, technological advances, and regional characteristics

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

by

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September 2015

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June 2015

Evaluation of the environmental impacts of agricultural systems using life cycle thinking,  
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\* work from a seminar class, alphabetical authorship, not necessarily reflecting contribution
- Yang Y & Suh S** “Land cover change from cotton to corn in the US relieves freshwater ecotoxicity impact but may aggravate other regional environmental impacts” *Int J LCA* 20(2) 196-203
- Yang Y & Suh S** “Marginal yield, technological advances, and emissions timing in corn ethanol’s carbon payback time” *Int J LCA* 20(2) 226-232
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- 2013 Heimpel G **Yang Y** Hill J Ragsdale D “Environmental Consequences of Invasive Species: Greenhouse Gas Emissions of Insecticide Use and the Role of Biological Control in Reducing Emissions” *PLoS ONE* 8(8): e72293.  
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## ABSTRACT

Evaluation of the environmental impacts of agricultural systems using life cycle thinking, focusing on marginal changes, technological advances, and regional characteristics

by

Yi Yang

Driven by rapid adoption and sustained improvements of genetic technologies and agronomic management practices, agricultural productivity has experienced a substantial growth worldwide since the start of the green revolution in 1960s. This growth has enabled the humanity to escape from the well-known Malthusian Trap. With the success in agricultural productivity, however, comes what is also known as “the other inconvenient truth” (Foley 2009). That is, modern agriculture has become one of the major drivers of global environmental change and is pushing the earth system beyond its safe operating boundaries (Rockström et al. 2009). Further, even more challenges lie ahead, given the growing number of population and increasing diversion of foods to fuels.

In this dissertation, three topics related to US agricultural systems are explored. In the first chapter, the environmental properties of US corn and cotton production and

implications of land cover change from cotton to corn are evaluated using state-specific data and life cycle impact assessment. The results show that U.S. cotton and corn productions per hectare on average generate roughly similar impacts for most impact categories such as eutrophication and smog formation. Life cycle water use and freshwater ecotoxicity impacts of corn per hectare on average are smaller than those of cotton. When marginal impact is analyzed, however, the results show that the shifts from cotton to corn in cotton-growing states aggravates most of the regional environmental impacts while relieving freshwater ecotoxicity impact. The differences in the two estimates are due mainly to underlying regional disparities in crop suitability that affects input structure and environmental emissions.

In the second chapter, the carbon payback time (CPT) of corn ethanol expansion is re-examined considering three aspects: (1) yields of newly converted lands (i.e., marginal yield), (2) technology improvements over time within the corn ethanol system, and (3) temporal sensitivity of climate change impacts. The results show that without technological advances, the CPT estimates for corn ethanol from newly converted Conservation Reserve Program (CRP) land exceed 100 years for all Marginal to Average (MtA) yield ratios tested except for the case where MtA yield ratio is 100 %. When the productivity improvements within corn ethanol systems since previous CPT estimates and their future projections are considered, the CPT estimates fall into the range of 15 years (100 % MtA yield ratio) to 56 years (50 % MtA yield ratio), assuming land conversion takes place in early 2000s. Incorporating diminishing sensitivity of GHG emissions to future emissions year by year, however, increases the CPT estimates to 17 to 88 years. For 60 MtA yield ratio, the CPT is

estimated to be 43 years, which is relatively close to previous CPT estimates (i.e., 40 to 48 years) but with very different underlying reasons.

In the third chapter, the trends and underlying drivers of changes in non-global environmental impacts of major crops in the U.S. are investigated. The results show that the impact per hectare corn and cotton generated on the ecological health of freshwater systems decreased by about 50% in the last decade. This change is associated with the use of genetically modified (GM) crops, which has reduced the application of insecticides and relatively toxic herbicides such as atrazine. However, the freshwater water ecotoxicity impact per hectare soybean produced increased by 3-fold, mainly because the spread of invasive species, soybean aphid, has resulted in an increasing use of insecticides. In comparison, other impact categories remained relatively stable. By evaluating the relative ecotoxicity potential of a large number of pesticides, our analysis offers new insight into the benefits associated with genetically modified (GM) crops. The finding that different impact categories show different degrees of changes suggests that agricultural inventory data can be updated selectively in LCA to maximize cost-effectiveness.

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# Chapter 1: Introduction and research questions

## *1 The success of modern agriculture*

Humanity has made giant strides toward eliminating hunger and malnutrition. Although continuous effort is needed to fight extreme poverty and hunger in some areas (Sachs 2005), today we produce more than enough food to feed the world adequately. In 2014, global cereal production reached a new record of 2.5 billion metric tons (FAO 2015). Agricultural productivity growth has made substantial contributions to these successes.

Since the start of the green revolution in the 1960s, agricultural productivity has experienced a consistent and rapid growth worldwide. For example, global land productivity, measured as an output of 185 crop and livestock commodities per harvested and pastured area, grew by a factor of 2.5 from 1961 to 2005, while labor productivity, the output per farmer, grew by a factor of 1.7 during the period (Alston et al. 2009). Global yield for maize, wheat, rice and soybean in 2007 was 2 to 3 times as large as it was in 1961 (Alston et al. 2009). These remarkable trends in productivity growth have taken place as a result of rapid adoption of, together with sustained improvements in, genetic technologies and agronomic management practices (Grassini et al. 2013). Among them are plant breeding that results in improved hybrids and varieties, application of synthetic fertilizers and pesticides, and investments in irrigation infrastructure (Doorenbos and Kassam 1979; Stewart et al. 2005; Araus et al. 2008).

## ***2 The other inconvenient truth***

Along with the successes of agriculture, however, came what Jonathan Foley terms the other inconvenient truth: “that we now face a global crisis in land use and agriculture that could undermine the health, security, and sustainability of our civilization” (Foley 2009). Indeed, agriculture has been identified as one of the major drivers of global environmental change, and is pushing the earth system beyond its safe operating boundaries (Rockström et al. 2009).

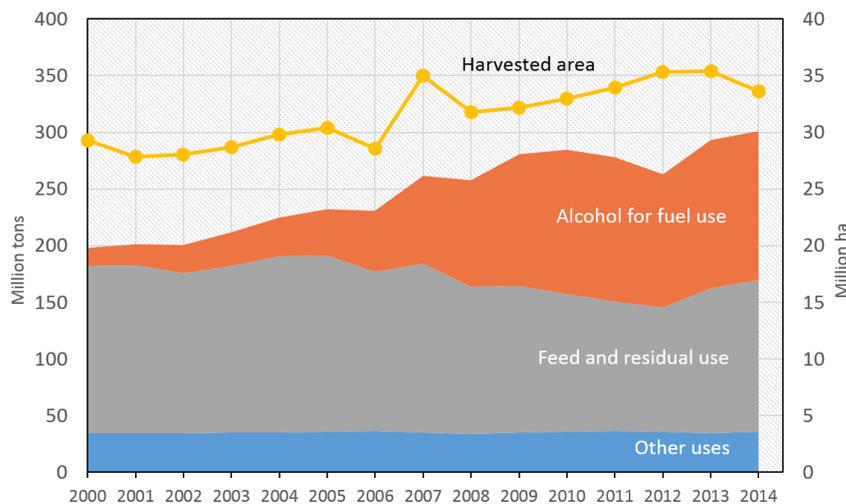
Through the intensive use of synthetic fertilizers and planation of leguminous crops, agriculture has critically disturbed the global nitrogen and phosphorus cycle, resulting in a wide range of environmental issues including eutrophication of lakes and coastal areas (Bouwman et al. 2013). Agriculture constitutes the single largest use of land, about 60 times as large as the area of all cities and suburbs combined (Foley 2009), and poses the greatest threat to ecosystems (Matson et al. 1997). Irrigation accounts for 70% of water withdraws, contributing to water shortage and scarcity in many areas of the world (Gleick et al. 2006). Further, agriculture is also the largest emitter of greenhouse gases through intensification and land conversion such as deforestation (Foley 2009). Last but not least, agriculture dominates pesticide use, which, among others, contaminates surface and ground water and leads to aquatic biodiversity loss (Gilliom 2007; Beketov et al. 2013).

## ***3 Even more challenges ahead***

Despite the severity of existing environmental impacts of agriculture, more challenges lie ahead. Global food demand is likely to double in 2050 relative to the 2005 level (Tilman et al. 2011), driven by population growth and the continuous spread of economic prosperity

in developing countries. If the current trend of agricultural practices were to continue, by 2015 about 1 billion hectare of land would be cleared globally, 250 Mt y<sup>-1</sup> of nitrogen fertilizers would be used, and 3 Gt y<sup>-1</sup> of greenhouse gases would be released (Tilman et al. 2011).

And yet the entrance of agriculture into the energy industry across the world brings more pressure to bear on land, water, and energy that are essential for the production of food for human consumption (Pimentel et al. 2009). In the U.S., for example, corn was primarily used for food and feed before the expansion of the ethanol industry, which now consumes >40% of the total production (Fig. 1.1). As a result, corn area harvested has also expanded substantially (Fig. 1.1), resulting in massive displacement of grassland as well as cropland like cotton (Wallander et al. 2011; Wright and Wimberly 2013). Rapid biofuels expansion worldwide, but primarily in the U.S. and EU, has contributed substantially to global food price hikes in the past few years (Mitchell 2008). The increases in food prices have generated dire economic and social consequences worldwide especially for the poor in developing countries (Runge and Senauer 2007).



**Fig. 1.1** Uses of corn and corn area change between 2000 and 2014 (USDA 2015a)

#### ***4 Research questions***

It is against this background that this dissertation investigates three topics related to U.S. agricultural systems. The first chapter explores the environmental implications of land use change from cotton to corn driven partly by ethanol expansion. Previous studies in this area have centered on corn ethanol's life-cycle GHG emissions (Yang et al. 2012), particularly with respect to direct and indirect conversion of natural habitats such as grassland and forest (Fargione et al. 2008; Searchinger et al. 2008). Insufficient attention has been paid to land use change between crops and associated impacts on the local environment. In the past "ethanol decade," however, substantial increases in corn prices, due in part to ethanol expansion, not only resulted in considerable conversion of grassland to corn production, but also greatly escalated the dynamics of land use change between crops (Wallander et al. 2011; Wright and Wimberly 2013). There were, for example, land use shifts from soybean, hay, and cotton to corn and from cotton to soybean. The reason to target cotton to corn, rather than other changes in land use, is as follows. Input requirements for both corn and cotton production are high, thus the environmental implications of land use shift from one to the other are much less clear than from high-input crops (e.g., corn) to low-input crops (e.g., soybean and hay) or vice versa.

The second chapter of the dissertation re-evaluates the calculation of carbon payback time (CPT) in the case of converting grassland for corn ethanol production. Previous research on the CPT of corn ethanol neglected two important elements that may substantially affect their results, namely, the actual corn yield of the newly converted land (i.e., marginal land) and technological advances of the corn ethanol system. The analysis

also tests the effect of considering emission timing on the estimates of CPT using dynamic characterization factors as proposed recently in a growing body of literature (Kendall 2012).

The third chapter explores potential changes in the environmental impacts of major crops (i.e., corn, soybean, wheat, and cotton) in the past decade. LCA has been increasingly applied to agricultural systems, as reflected in the number of agricultural LCA databases built in the past few years (CleanMetrics 2011; Blonk 2014; Nielsen et al. 2014; Quantis 2014; USDA 2014). As with LCA studies in general, agricultural LCAs often rely on static and single-year inventory data with commonly 5 to 10 years of data age. Literature suggests, however, that agricultural systems may be highly dynamic due to the increasingly changing climate and technological advances such as improved energy efficiency and deployment of genetically modified (GM) crops (Nelson et al. 2009; Shapouri et al. 2010; Fernandez-Cornejo et al. 2014). These factors may bring about substantial changes in the use of input materials and the yield of crops, hence changes in their environmental impacts.

In summary, the research questions of the dissertation are:

- *What are the environmental implications of land use change from cotton to corn?*
- *What is the CPT in converting grassland to grow corn for ethanol, taking into account the actual corn yield of the grassland and technological advances of the corn ethanol system? If considering emission timing, how large is its effect on CPT estimates?*
- *How may have the environmental impacts of major crops changed in the past decade? Do different categories change differently? In particular, has and to what extent the widespread deployment of genetically modified crops affected the pesticides-related ecotoxicity impact of these crops?*

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**Chapter 2: Land cover change from cotton to corn in the USA  
relieves freshwater ecotoxicity impact but may aggravate other  
regional environmental impacts**

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## ***Abstract***

*Purpose* Rising corn prices in the USA due partly to increasing ethanol demands have led to a substantial expansion of corn areas displacing natural vegetation and crops including cotton. From 2005 to 2009, cotton area harvested in the USA nearly halved with a reduction of 2.5 million hectares, while that of corn increased by 1.8 million hectares. However, environmental impacts of land shifts from cotton and corn have been largely neglected in literature.

*Methods* In this study, we evaluate the environmental properties of US corn and cotton production and implications of land cover change from cotton to corn using state-specific data and life cycle impact assessment. Focusing on regional environmental issues, we cover both on-farm direct emissions such as different types of volatile organic compounds and pesticides and indirect emissions embodied in input materials such as fertilizers. TRACI 2.0 is used to evaluate the environmental impacts of these emissions.

*Results and discussion* The results show that U.S. cotton and corn productions per hectare on average generate roughly similar impacts for most impact categories such as eutrophication and smog formation. For water use and freshwater ecotoxicity, corn shows a smaller impact. When land shifts from cotton to corn in cotton-growing states, however, the process may aggravate most of the regional environmental impacts while relieving freshwater ecotoxicity impact. The differences in the two estimates are due mainly to underlying regional disparities in crop suitability that affects input structure and environmental emissions.

*Conclusions* Our results highlight the importance of potential, unintended environmental impacts that cannot be adequately captured when average data are employed. Understanding

the actual mechanisms under which certain policy induces marginal changes at a regional and local level is crucial for evaluating its net impact. Further, our study calls for an attention to biofuel-induced land cover change between crops and associated regional environmental impacts.

**Keywords** Average · Biofuel · Corn expansion · Cotton · Environmental impacts · Land use change · Marginal

## ***1 Introduction***

Concerns about the negative environmental impacts of fossil fuels, particularly those on climate change and energy security, have driven the recent interest in biofuels in the USA (Hill et al. 2006). Several federal policies have been put in place to foster biofuels development, among which is the ethanol production mandate in the renewable fuel standard (RFS) (Runge and Johnson 2008). As a result of the favorable policies and gasoline prices, production of corn ethanol in the USA has expanded substantially since 2005, with an annual increase of over six billion liters (RFA 2013).

Previous research, however, has shown that biofuels policies may have caused unintended consequences that not only undermine the goal of the federal policies to reduce greenhouse gas (GHG) emissions but also degrade local environmental quality (Fargione et al. 2008; Searchinger et al. 2008; Yang et al. 2012). Increasing ethanol demand has contributed to high corn prices, incentivizing farmers to convert grassland into corn growth in the Corn Belt (Malcolm et al. 2009; Claassen et al. 2011; Wallander et al. 2011). This direct land use change (LUC) threatens wildlife habitats and creates a carbon debt that may

take up to >100 years to be paid off by replacing gasoline with corn ethanol (Gelfand et al. 2011; Wright and Wimberly 2013). Also, due to intensive use of agrochemicals and irrigation water, growing corn on grassland puts further pressure on local water quality and scarcity (Secchi et al. 2010; Yang et al. 2012).

Additionally, diversion of corn to ethanol production essentially reduces food supply, which may lead farmers worldwide to convert natural land to new cropland in order to compensate for the diverted grain (Searchinger et al. 2008). This indirect LUC would generate the same net effects as that of direct LUC as discussed above, although its estimation is much more complicated for the difficulty and uncertainty involved in quantifying the impacts of US biofuel policies on global land and agricultural commodity markets (Babcock 2009; Plevin et al. 2010). Studies continue to explore the effect of indirect LUC of biofuels with refined modeling methodologies (Hertel et al. 2010; Keeney 2010; Wang et al. 2011; Sanchez et al. 2012), improved understanding of agricultural and food systems around the world (Arima et al. 2011; Andrade de Sá et al. 2013), and extended assessment to non-GHG impacts (Tsao et al. 2012).

Yet, there is another consequence of corn ethanol expansion to which relatively less attention has been paid. In conjunction with rising corn prices, substantial land cover shift from cotton to corn has been observed—particularly between 2005 and 2009—through both direct expansion of corn into cotton and indirect expansion of corn into soybean, then of soybean into cotton (Wallander et al. 2011). This observation is supported by farm-level data, which reveal that as growing corn (and soybeans) became more profitable, some farmers reacted by reducing cotton land for growing corn (and soybeans) (Fanin et al. 2008; Wallander et al. 2011). Furthermore, the National Agricultural Statistics Service

(NASS) Cropland Data Layer (CDL) provides high-resolution maps derived from satellite imagery clearly demonstrating that land shifts from cotton to corn occurred in several states (USDA 2014a) (see Fig. S1 and S2 in Appendix A). Overall, between 2006 and 2009 when corn (and soybean) prices increased substantially relative to cotton prices (Wallander et al. 2011), cotton area harvested reduced by 40 % (i.e., by 1.8 million ha), while corn area in the cotton growing states expanded by 1.3 million ha (USDA 2014b) (see Fig. S3 in Appendix A).

Despite the potential large-scale land shift from cotton to corn, there have been few studies on associated environmental impacts. Here, we address this knowledge gap. We note that corn displacing cotton was only part of the complex land use dynamics in the past “ethanol decade” (Wallander et al. 2011) that involved also land shift from, for example, soybeans and hay to corn, cotton to soybeans, and natural vegetations to corn (Wright and Wimberly 2013). The reason we focus only on cotton to corn here is that environmental impacts of land shift between cotton to corn, both high-input crops, are less clear than that between relatively low-input crops and high-input crops (e.g., hay and soybeans to corn). In a recent study, Wallander et al. (2011) stated that “When acreage shifts from one high-input crop to another (e.g., cotton to corn), however, ethanol induced changes may be negligible or could even reduce environmental externalities.” In this study, we seek to test the validity of this statement, focusing on regional environmental issues along with a growing body of literature on the non-GHG consequences of biofuels expansion (Chiu et al. 2009; Hill et al. 2009; Tessum et al. 2012; Tsao et al. 2012; Yang et al. 2012; Yang 2013).

## ***2 Materials and methods***

### **2.1 Scope**

A land shift from one crop to the other can alter both direct, or on-site, and indirect, or offsite, environmental effects. For example, increased use of nitrogen (N) fertilizers as a result of the land shift not only can elevate N related emissions such as NO<sub>x</sub> and N runoff but also requires more energy and material inputs in the process of fertilizer production. The system boundary of the study, therefore, was drawn to cover both direct and indirect emissions. In particular, we paid a special attention to direct environmental emissions from crop production given their significance relative to indirect emissions (Yang et al. 2012).

We calculated indirect emissions embodied in input materials (e.g., fertilizers) that take place along supply chains, using the Ecoinvent database (V2.2) (Ecoinvent 2014). In our data compilation, we placed an emphasis on the crop growth and agricultural input structures at the state level, as previous studies showed that national, average data may fall short in capturing the environmental impacts of crop production at a regional level (Yang et al. 2012). This is because agricultural systems display high degrees of variability across regions in terms of input structure due primarily to differences in geography, weather patterns, soil type, and management practices (Miller et al. 2006). Also, data on major agricultural inputs such as fertilizers and pesticides collected by the US Department of Agriculture (USDA) are only available at the state level (see section 2.2.). The reference year of this study is 2005 given that cotton area experienced a substantial decline between 2005 and 2009.

## **2.2 Data on agricultural inputs**

Major inputs in crop growth include fertilizers, pesticides, energies, and irrigation water. We obtained relevant state-level data from several USDA surveys and censuses (USDA 2004, 2014b, c; Shapouri et al. 2010) reflecting cotton and corn farming practices around 2005 and then compiled a set of state-specific inventories. Not all inputs data, however, are available for every state that grows cotton and corn. The USDA Farm and Ranch Irrigation survey, for example, includes more states than surveys of energy and agricultural use. Nevertheless, the states for which all inputs data are available capture the majority of US cotton and corn production. Specifically, the inventories we compiled cover 19 corn-growing states, which account for 95 % of domestic corn production in 2005, and 9 cotton-growing states, which account for 88 % of domestic cotton production in 2005.

## **2.3 Estimation of environmental emissions**

Due to use of agricultural inputs like fertilizers and pesticides, crop production contributes to an array of environmental impacts from acidification, eutrophication, water scarcity to human and ecological toxicity (Socolow 1999; Yang et al. 2012). To best capture these impacts associated with US cotton and corn growth, we estimated all potential onsite environmental emissions based on various databases, models, and literature (see Table 2.1). The emissions data compiled cover >100 different substances, the majority of which are pesticides and volatile organic compound (VOC) emissions. Numerical information on all emission factors used in this study can be found in the Table S1–S6 (Appendix A).

**Table 2.1** Estimation of on-farm environmental emissions

Sources	Environmental emissions	Ref/Note
N fertilizers	NH <sub>3</sub> to air	(Krauter et al. 2002; Goebes et al. 2003)
	NO <sub>x</sub> to air	(Yienger and Levy 1995)
	N runoff and leaching <sup>a</sup>	(USDA 2006)
P fertilizers	P runoff and leaching <sup>a</sup>	(USDA 2006)
	Heavy metals to soil	(Mortvedt 1995; EPA 1999)
Pesticides	Pesticides to air <sup>b</sup>	(Herner 1992; Berthoud et al. 2011)
	Pesticides runoff and leaching	(USDA 2000)
	Pesticides to soil <sup>c</sup>	(Berthoud et al. 2011)
Farm equipment	NO <sub>x</sub> , SO <sub>x</sub> , PM <sub>2.5</sub> , PM <sub>10</sub> , CO	(Wang 2013)
	Speciated VOCs	(EPA 1995)
Irrigation	Water withdraw <sup>d</sup>	(USDA 2014c)

a. State nutrient runoff and leaching rates were derived from regional rates estimated in (USDA 2006).  
b. The fraction of pesticides emitted to air was estimated based on their vapor pressure (VP) (Berthoud et al. 2011), data on pesticide VP mostly from the USDA Pesticide Properties Database (Herner 1992).  
c. Fraction to soil = 1 – fraction to air – fraction to water (capped at 85%).  
d. Irrigation data for 2003 and 2008 were averaged to represent the 2005 situation.

## 2.4 Impact assessment

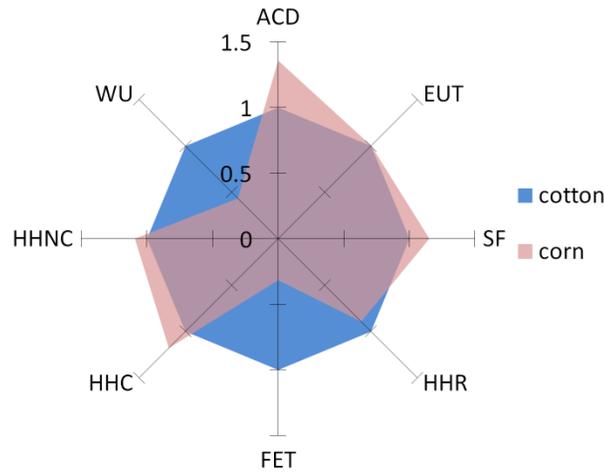
After compiling emissions data for cotton and corn, we evaluated their environmental impacts using characterization factors from life cycle impact assessment (LCA) (ISO 2006). Reflecting the relative significance of an emission or resource, characterization factors are used to aggregate emission results, usually including a large number of different substances, into a dozen of impact category scores that enable better comparison between alternatives (Guinee et al. 2002).

In this study, we focused on regional environmental aspects of cotton and corn, and based on our previous study (Yang et al. 2012), we selected eight impact categories to which cotton and corn production potentially contribute. These impact categories are acidification, eutrophication, smog formation, freshwater ecotoxicity, and water use as well as human health cancer, noncancer, and respiratory effects. Characterization factors for all categories

except water use are taken from the Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI 2.0) developed for the USA by the EPA (Bare 2011). Characterization factors for water use were based on the ReCiPe model (Goedkoop et al. 2009). Note that TRACI 2.0, compared with its original version (Bare et al. 2003), has incorporated the recently developed USEtox model for the ecotoxicity, human health cancer, and noncancer impact categories (Rosenbaum et al. 2008).

### 3 Results and discussion

#### 3.1 Average environmental impacts of cotton and corn in the USA



**Fig. 2.1** Comparison of *average* cotton and corn by impact category on the basis of per hectare crop produced in 2005, weighted by state crop area harvested. Each impact category result of corn is normalized to that of cotton. *ACD* acidification, *EUT* eutrophication, *SF* smog formation, *HHR* human health respiratory effect, *FET* freshwater ecotoxicity, *HHC* human health cancer, *HHNC* human health noncancer, and *WU* water use

For comparison between the two crops, results are organized on the basis of per hectare produced. Figure 2.1 shows the average environmental impacts, weighted by state area harvested, of corn relative to that of cotton in 2005 in the USA. For most impact categories,

corn and cotton per hectare show roughly similar environmental impacts, with relative magnitude (corn over cotton) ranging from 1.4 for acidification and 0.9 for human health cancer. For freshwater ecotoxicity, however, corn shows about one third of impact by cotton per hectare, and corn's water use is less than half that of cotton.

Above all, most of the environmental impacts associated with cotton and corn production are due to on-site environmental emissions rather than that embodied in input materials like fertilizers and pesticides. Their acidification effect is due in large part to application of nitrogen (N) fertilizers (about 60–80 %) and diesel combustion (5–20 %). Although N intensity of corn is much larger than that of cotton (159 versus 91 kg ha<sup>-1</sup>), corn farming uses much less diesel (47 versus 172 kg ha<sup>-1</sup>). Overall, the acidification impact of corn per hectare is 1.4 times that of cotton. The same can be said about smog formation. Not surprisingly, the two crops' eutrophication impact is caused mainly by use of N and phosphate (P) fertilizers. Although corn has higher nutrient application intensities than cotton, its average N and P leaching and runoff rates are lower (21 and 14 versus 39 and 24 %, respectively); thus, the two crops have a comparable eutrophication impact.

Water use by cotton and corn comes primarily from irrigation: about 400 m<sup>3</sup> is applied per hectare corn produced as opposed to 940 m<sup>3</sup> applied per hectare cotton produced. Freshwater ecotoxicity for both crops is due in large part to pesticide use, and cotton per hectare has a freshwater ecotoxicity about three times that of corn. This is partly because pesticide application intensity of cotton is approximately twice as much as that of corn (5.2 kg ha<sup>-1</sup> versus 2.7 kg ha<sup>-1</sup>). Also, many of the pesticides such as cyfluthrin, lambda-cyhalothrin, and cypermethrin used in cotton growth generally show higher toxicity-related characterization factors than the major ones used in corn growth.

The two crops' potential human health respiratory impacts are comparable, although that of cotton is slightly higher. The respiratory effect is mainly caused by diesel combustion, application of N fertilizers, and emissions embodied in P fertilizers. Human health cancer and noncancer impacts of corn per hectare are slightly larger than that of cotton. Heavy metals contained in phosphate constitute the major contributor to both crops' noncancer effect, but use of acephate, an insecticide, is also another important source of noncancer impact for cotton. This is why corn's relative magnitude of noncancer effect (1.1 times) is not as large as that of phosphate application intensity (1.6 times). The two crops' potential human health cancer impact is due to a number of factors including diesel combustion and heavy metals brought about by phosphate as well as the cancer impact embodied in fertilizers.

The results above (Fig. 2.1) indicate that corn and cotton grown per hectare in the USA on average generate roughly comparable impacts for most of the impact categories except for water use and freshwater ecotoxicity, where cotton shows lower impacts. The results seem consistent with the view of a recent USDA study (Wallander et al. 2011), "When acreage shifts from one high-input crop to another (e.g., cotton to corn), however, ethanol-induced changes may be negligible or could even reduce environmental externalities."

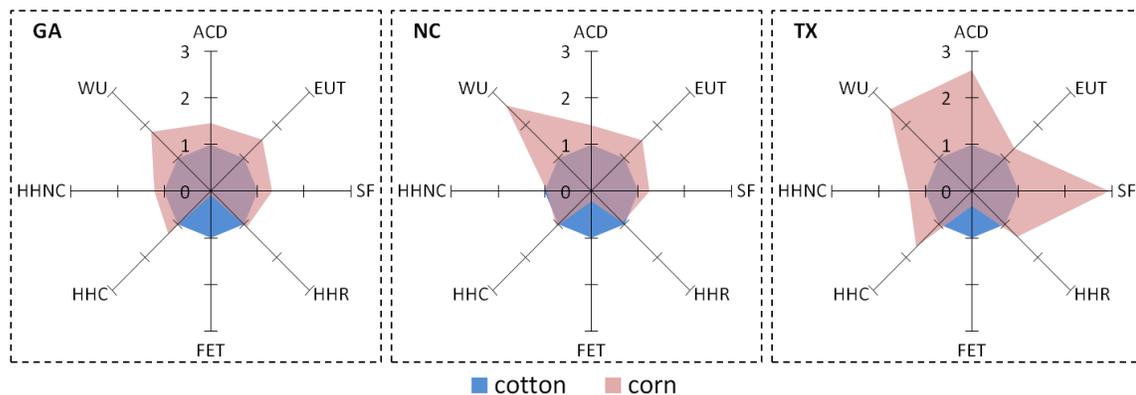
We argue that, however, the average results as shown in Fig. 2.1 are inadequate to capture the net environmental impacts associated with land cover change from cotton to corn that took place in the USA. First, Fig. 2.1 is largely a portrait of corn and cotton growth in different regions (north versus south) and, weighted by state crop area, mainly represents the major crop-growing states where respective crops are likely the most suitable to grow. But, when land shifts from cotton to corn growth, it happens in cotton-growing areas in the

South. Lands in these areas can be by and large considered marginal lands for corn in both geographic and economic senses as they are generally less suitable for corn growth than the Corn Belt.

Further confounding the issue is the existence of large spatial variability among corn- and cotton-growing states (see Fig. S4 in the Appendix A). The range of spatial variation in cotton growth is two to threefold for acidification, smog formation, eutrophication, human health noncancer, and respiratory effects and four to sixfold for freshwater ecotoxicity and human health cancer effect. The range of spatial variation in corn growth is about two to threefold for acidification, smog formation, human health cancer, noncancer, and respiratory effects and fourfold for eutrophication. Water use can vary by orders of magnitude for both crops as some states use little irrigation water (e.g., Tennessee with about 20 m<sup>3</sup> applied per hectare cotton produced) while some rely heavily on irrigation (e.g., California with over 2000 m<sup>3</sup> applied per hectare cotton produced). In short, the results for average corn and cotton as reflected in Fig. 2.1 fall short of representing the environmental performance of marginal corn in cotton-growing states and, therefore, should not be used for evaluating environmental impacts of land use change from cotton to corn or vice versa.

### **3.2 State-specific environmental impacts and implications of land use shift from cotton to corn**

Although the USDA surveys are centered on major crop growing states (see section 2.2), there are three states for which all inputs data on both crops are available (see Fig. 2.2). They are Georgia (GA), North Carolina (NC), and Texas (TX), and judging from crop area changes in Fig. S3 in Appendix A land shift from cotton to corn might have likely occurred during 2005 to 2009 in these three states.



**Fig. 2.2** Comparison of the environmental impacts between cotton and corn grown in Georgia (GA), North Carolina (NC), and Texas (TX) in 2005 on the basis per hectare crop produced. Each impact category result of corn is normalized to that of cotton

Comparing Fig. 2.2 with Fig. 2.1 reveals that corn and cotton growths in 2005 at the state level can be quite different from the average situation. Land shift from cotton to corn in Georgia and Texas would likely aggravate all of the impact categories except freshwater ecotoxicity. For North Carolina, however, the land shift would increase water withdraw and aggravate eutrophication impact, but would not cause substantial changes to human health effects. For TX, land shift from cotton to corn would especially aggravate acidification and smog formation impacts. This is because TX, as the major producer of cotton in the US, applies far less nutrients per hectare cotton produced than per hectare corn produced there ( $62 \text{ kg ha}^{-1}$  versus  $171 \text{ kg ha}^{-1}$  for nitrogen and  $29 \text{ kg ha}^{-1}$  versus  $45 \text{ kg ha}^{-1}$  for phosphate). It is worth pointing out that that while using nutrients much more intensively, corn growth in TX does not generate a substantially larger eutrophication impact than cotton. This is because nutrient runoff and leaching rates of corn in TX are generally smaller than that of cotton (see Table S7). For all states, as with the average situation in Fig. 2.1, land shift from cotton to corn would relieve freshwater water ecotoxicity impact.

#### ***4 Implications of the study***

In summary, our study calls for an attention to policy-induced land cover change from cotton to corn and associated environmental issues. In doing so, we demonstrate that average data reflecting national situations are inadequate to capture the likely environmental impacts of corn expansion into cotton on marginal land at regional level. Our results for three states North Carolina, Georgia, and Texas show that corn expansion into cotton in the South relieves freshwater ecotoxicity but may aggregate many other regional environmental impacts. Overall, our study confirms the earlier studies that demonstrated the importance of understanding “marginal” impacts in LCA (Weidema et al. 1999; Schmidt and Weidema 2008; Mathiesen et al. 2009): environmental consequences of the policies that encourage converting cotton to corn cultivation in the regions where corn is generally less suitable to grow cannot be understood by comparing average environmental profiles of cotton and corn. Our results also favor “consequential thinking,” as an analytical paradigm, in biofuel LCA, while our study is not intended to demonstrate how to perform a “consequential LCA,” as an operational model (Plevin et al 2014; Suh and Yang 2014; Brandão et al 2014; Dale and Kim 2014; Hertwich 2014).

Corn ethanol, supported by several federal policies as a means of reducing GHG emissions by displacing gasoline (Keeney 2008), has been a point of heavy dispute in the last decade (Babcock 2009). However, it has become increasingly clear that although corn ethanol may have the potential to combat climate change (Liska et al. 2009), its large-scale expansion is reported to generate adverse environmental consequences including, notably, direct, and indirect land use changes (Searchinger et al. 2008; Wright and Wimberly 2013).

These adverse consequences, first, undermine the climate objectives of the public policies. Second, for intensive use of agrochemicals and irrigation water, corn expansion adds to the pressure on local water quality and scarcity issues (Yang et al. 2012).

Our study focused on yet another consequence related to ethanol expansion, namely, land cover change from cotton to corn, and analyzed the potential implications of such change for local environments. Contrary to the previous view that land shift between cotton and corn, both high-input crops, may cause negligible net environmental impacts (Wallander et al. 2011), our study revealed a more complex picture. Although land switch from cotton to corn relieves ecotoxicity, it likely aggravates other various environmental problems depending on where the crops are grown. Note that our study only covers part of the effects biofuels policies have generated on crop conversions. To understand the overall environmental impacts of biofuel policies through crop conversions, further research is needed to estimate the environmental aspects of other crops affected, particularly soybean (Wallander et al. 2011), and the magnitude of land shifts between the crops.

Our results highlight the importance of potential, unintended consequences that cannot be adequately captured when average data are employed. Understanding the actual mechanisms under which certain policy induces marginal changes at a regional and local level is crucial for evaluating its net impact. Our results also show the importance of recognizing potential trade-offs between environmental objectives in policy making. Climate policies focusing narrowly on carbon, for instance, could shift burden to regional issues like water scarcity and eutrophication (Jackson et al. 2005; Yang et al. 2012). Therefore, environmental policymaking should attend to not only unintended effects within

its targeted problems like the indirect LUC effect (Searchinger et al. 2008), but also those across impact categories to avoid or minimize burden shifting across impact categories.

Also, our study reinforces previous research with respect to spatial variability in agricultural systems (Miller et al. 2006; Yang et al. 2012). Unlike industrial systems, agricultural systems are subject to the influence of weather patterns, soil type, geography, and management practices. Even the same agricultural product may have drastically different input structures, hence environmental impacts, in different regions. Therefore, average data with generic descriptions of material and energy fluxes are hardly adequate to capture the high degree of system variability of agricultural products. With the rising interests in biofuels as a means to combat climate change across the world, we strongly recommend future studies in this area to take into consideration the spatial variability of biomass growth.

Just as technological and environmental variability exists across states, there is probably certain variability within a state, too, that may not be precisely captured by state average data. This does not mean, however, that state-level data should be dismissed for the research question at hand because they are still likely more reflective of local or farm-level practices than national averages. In addition, state average data are especially valuable and representative, more so than farm-level data, in situations in which massive land shift between crops takes place within a state. Nevertheless, we encourage finer-scale, more detailed studies into land shift between cotton and corn and associated environmental impacts, which could not have been conducted in our analysis due to the data limitation and resources constraints.

Additional research is needed to paint a more complete picture on the impact of cropland conversion to corn: In 2005, 41 states grew corn and 17 states grew cotton, among which only 19 of the corn-growing states and 7 of the cotton-growing states had data on major inputs that can be used to generate LCIs (see section 2.2). Among these states, only three overlap, namely, North Carolina, Georgia, and Texas. Therefore, this study does not quantify the environmental impact and their trade-offs in other cotton-growing states where conversion to corn might have happened. Nevertheless, environmental implications of cotton-to-corn land shift in these other states are probably worse than that indicated by Fig. 2.1 and closer to that indicated in Fig. 2.2 because cropland in southern states are generally less suitable for corn growth than the Corn Belt. Future studies pursuing this line of research may make the effort to quantify the magnitude of land shift in each cotton-growing state when relevant data on agricultural inputs, environmental outputs, and acreage of conversion become available.

Furthermore, it is worth noting that spatially detailed data are often unavailable or incomplete, although such data can improve the environmental relevance of an LCA study. In this case, one may rely on assumptions or spatially generic data to fill the gaps, and this may increase the uncertainty of the LCA results (Mathiesen et al. 2009). In our study, data on agricultural inputs such as fertilizers and pesticides were available at the state level, but we often relied on spatially generic emission factors to estimate their emissions (except for N and P runoff and leaching rates). Further, the LCA results for corn and cotton were found to be moderately sensitive to the emission factors which are likely to vary across regions (see Table S7 and S8 in the Appendix A for sensitivity analysis). Future spatially explicit

LCAs on agricultural systems may take this into account and direct efforts to estimate spatially differentiated emission factors.

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## **Chapter 3: Marginal yield, technological advances, and emissions timing in corn ethanol's carbon payback time**

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## ***Abstract***

*Purpose* Previous estimates of carbon payback time (CPT) of corn ethanol expansion assumed that marginal yields of newly converted lands are the same as the average corn yield, whereas reported marginal yields are generally lower than the average yield (47–83 % of average yield). Furthermore, these estimates assumed that the productivity of corn ethanol system and climate change impacts per unit greenhouse gas (GHG) emissions remain the same over decades to a century. The objective of this study is to re-examine CPT of corn ethanol expansion considering three aspects: (1) yields of newly converted lands (i.e., marginal yield), (2) technology improvements over time within the corn ethanol system, and (3) temporal sensitivity of climate change impacts.

*Methods* A new approach to CPT calculation is proposed, where changes in productivity of ethanol conversion process and corn yield are taken into account. The approach also allows the use of dynamic characterization approach to GHGs emitted in different times, as an option. Data are collected to derive historical trends of bioethanol conversion efficiency and corn yield, which inform the development of the scenarios for future biofuel conversion efficiency and corn yield. Corn ethanol's CPTs are estimated and compared for various marginal-to-average (MtA) yield ratios with and without considering technology improvements and time-dependent climate change impacts.

*Results and discussion* The results show that CPT estimates are highly sensitive to both MtA yield ratio and productivity of ethanol system. Without technological advances, our CPT estimates for corn ethanol from newly converted Conservation Reserve Program (CRP) land exceed 100 years for all MtA yield ratios tested except for the case where MtA yield ratio is 100 %. When the productivity improvements within corn ethanol systems since

previous CPT estimates and their future projections are considered, our CPT estimates fall into the range of 15 years (100 % MtA yield ratio) to 56 years (50 % MtA yield ratio), assuming land conversion takes place in early 2000s. Incorporating diminishing sensitivity of GHG emissions to future emissions year by year, however, increases the CPT estimates by 57 to 13 % (from 17 years for 100 % MtA yield ratio to 88 years for 50 % MtA yield ratio). For 60 MtA yield ratio, CPT is estimated to be 43 years, which is relatively close to previous CPT estimates (i.e., 40 to 48 years) but with very different underlying reasons.

*Conclusions* This study highlights the importance of considering technological advances in understanding the climate change implications of land conversion for corn ethanol. Without the productivity improvements in corn ethanol system, the prospect of paying off carbon debts from land conversion within 100 years becomes unlikely. Even with the ongoing productivity improvements, the yield of newly converted land can substantially affect the CPT. The results reinforce the importance of considering marginal technologies and technology change in prospective life cycle assessment.

**Keywords** Biofuels · Carbon payback time · Corn ethanol · Dynamic characterization · Marginal yield · Prospective LCA · Technological change

## ***1 Introduction***

For the potential to mitigate climate change, reduce dependence on oil imports, and invigorate rural economic development, biofuel development in the USA has been supported by an array of policy measures (Keeney 2008; Runge and Johnson 2008). Among them is the federal Renewable Fuel Standard (RFS), a mandate that requires 140 billion liters biofuels to be produced annually from different sources by 2022. Corn ethanol is currently the primary biofuel and is likely to continue dominating US biofuels market as cellulosic and other advanced biofuels are far from mass production (Schnoor 2011). Driven by the favorable policies and high oil prices, corn ethanol production has increased eight-fold since 2000, to the current level of about 50 billion liter per year.

Early Life Cycle Assessment (LCA) research on corn ethanol (Farrell et al. 2006; Hill et al. 2006; Wang et al. 2007) was largely in support of the policies aiming partly at reducing greenhouse gas (GHG) emissions. As is typically done in LCA, these studies quantified GHG emissions generated at each stage of corn ethanol's life cycle, summed them up, and then compared the results against that of gasoline. Corn ethanol was found to have 10–20 % lower life cycle GHG emissions than gasoline and, therefore, concluded to provide a modest carbon benefit in replacing gasoline (Farrell et al. 2006; Hill et al. 2006; Wang et al. 2007).

However, the conclusion was later called into question, when the land use change (LUC) effects of corn ethanol expansion emerged in the literature (Fargione et al. 2008; Searchinger et al. 2008). Converting natural vegetation or forestland to corn field for ethanol production releases a substantial amount of carbon from soil and plant biomass, creating a “carbon debt” that could not be repaid in dozens of years or even a century (Fargione et al. 2008; Piñeiro et al. 2009; Gelfand et al. 2011). Similarly, diversion of existing cropland for

ethanol could generate indirect LUC (iLUC) effect through market-mediated mechanisms (Searchinger et al. 2008). In this scenario, corn ethanol expansion reduces food supply, which could lead to conversion of natural vegetation or forestland elsewhere in the world to compensate for the diverted grains. While the concept of iLUC has become widely accepted in academic and policy arenas (Tilman et al. 2009; EPA2010), quantification of iLUC emissions is known to be difficult and highly uncertain (Babcock 2009). Plevin et al. (2010), for example, estimated the range from 10 to 340 CO<sub>2</sub>e MJ<sup>-1</sup> y<sup>-1</sup>. This wide range is due in large part to a lack of quality data and detailed understanding as to how the global agricultural market would respond to biofuels expansion (Babcock 2009).

In contrast, the direct land use change (dLUC) emissions can be relatively accurately quantified (Gelfand et al. 2011). Previous studies used the concept of carbon payback time (CPT) to measure the magnitude of dLUC effect of corn ethanol. While the initial carbon debt due to land conversion may be large, it can be repaid over time by the annual carbon savings corn ethanol yields in displacing gasoline because corn ethanol has lower life cycle GHG emissions. The first dLUC study estimated that 48 years would be required for corn ethanol to pay back its carbon debt if the Conservation Reserve Program (CRP) land is converted and that 93 years would be required if central grassland is converted (Fargione et al. 2008). Subsequent studies have largely confirmed these initial estimates. Gelfand et al. (2011) conducted a field experiment on CRP land conversion to measure its carbon loss. They found that approximately 40 years would be required for the use of corn ethanol to pay back this carbon loss with the converted land under no-till management. In another study, Piñeiro et al. (2009) arrived at a similar estimate of approximately 40 years for the payback time for CRP land conversion to corn ethanol.

However, these studies were based on several oversimplifications that may substantially affect their results. First, these studies assumed that newly converted land has the same yield as existing cornfields, neglecting the potential yield differences of newly converted land. In particular, CRP land is generally less fertile than cornfields that have been in continuous use (Lubowski et al. 2006; Varvel et al. 2008). Thus, corn ethanol from CRP land generates lower annual carbon savings, hence a longer payback time. Land with extremely low yield may even fail to provide any carbon savings, in which case the carbon loss due to land conversion is permanently lost.

Second, the dLUC studies relied primarily on life cycle assessments (LCAs) based on early biofuel conversion processes (Farrell et al. 2006; Hill et al. 2006; Wang et al. 2007) that did not reflect the productivity improvements that have occurred in the past decade due to yield and energy efficiency increases at both the corn growing and ethanol conversion stages (Mueller 2010; Shapouri et al. 2010; Mueller and Kwik 2013). Recent studies have shown that corn ethanol's carbon benefit has increased to up to 50 % (Liska et al. 2009; Wang et al. 2011; Yang et al. 2012; Wang et al. 2012; Chum et al. 2013), compared with earlier estimates of 10–20 % (Farrell et al. 2006; Hill et al. 2006; Wang et al. 2007). The productivity of the gasoline production system over the same period of time has been fairly steady (Wang 2013). The productivity improvements in the corn ethanol system result in greater amounts of annual carbon savings that, if considered, would yield a shorter payback time than previously estimated.

Finally, the dLUC studies used the global warming potential (GWP) 100 (Forster et al. 2007) to assess the global warming impact of corn ethanol, gasoline, and dLUC emissions. This approach assumes equal weights to GHGs emitted at different times. More recent

literature explores the application of different weights to GHG emissions emitted in different times. First, from a scientific point of view, increasing background GHG concentrations in the atmosphere result in a diminishing marginal radiative forcing for a unit GHG emission (Reisinger et al. 2011). The rate at which the relative radiative forcing effect of a unit GHG emission diminishes depends on future atmospheric GHG concentrations. Reisinger et al. (2011), for example, estimated that the 100-year Absolute Global Warming Potential (AGWP) of CO<sub>2</sub> from 2000 to 2100 could decrease by 2 to 36 % under various GHG concentration scenarios. Second, a series of articles have attempted to synchronize the temporal system boundary under which life cycle emissions are taken into account and the time horizon under which characterization factors are derived. For example, if GWP100 is to be used, one can set the temporal system boundary to the next 100 years and account for the radiative forcing effects that occur within that time horizon (Kendall et al. 2009; O'Hare et al. 2009; Levasseur et al. 2010; Schwietzke et al. 2011; Kendall 2012). One of the rationales is that the efforts to reduce GHG emissions today is perhaps more valuable than those in the future because climate change may bring about irreversible damages to the planet (Schwietzke et al. 2011). In this class of literature, simple climate-carbon cycle model like Bern model or simple first-order decay model is used to calculate atmospheric load of GHGs over time, and corresponding radiative forcing (see, e.g., Levasseur et al. 2010). Background concentrations of GHGs are, however, generally assumed to be constant in the literature. Third, some argues that future climate change impacts should be discounted at certain rates using the net present value approach (Delucchi 2011). These approaches use different rationales and involve varying degrees of subjectivity in, e.g., the choice of emission

scenarios and discount rates. For the sake of simplicity, however, these approaches are collectively referred to as dynamic characterization method in this paper.

The objective of this study is to re-examine corn ethanol's CPT, taking into account the potential yield differences of converted land and technological advances within the corn ethanol system. We also examine how dynamic characterization of GHG emissions changes the CPT using one particular approach as an example. We focus on conversion of CRP land primarily for ease of comparison with previous studies (Fargione et al. 2008; Piñeiro et al. 2009; Gelfand et al. 2011) and also because there is evidence indicating that conversion of CRP land to cornfield has occurred with the expansion of corn production in the past decade (Wallander et al. 2011; Wright and Wimberly 2013).

## 2 *Methods*

### 2.1 Estimation of carbon payback time

Following Fargione et al. (2008), CPT can be calculated using Eq. (1),

$$\text{CPT} = \frac{a \cdot CD}{Y_l \cdot Y_r (CF_f - CF_b)} \quad (1)$$

where CD ( $\text{Mg ha}^{-1}$ ) represents the carbon debt, which is the amount of carbon lost due to land conversion, including foregone soil carbon sequestration;  $a$  represents the portion of carbon debt attributable to biofuel in the presence of coproducts; and  $Y_l$  ( $\text{Mg ha}^{-1} \text{ y}^{-1}$ ) and  $Y_r$  ( $\text{MJ Mg}^{-1}$ ) represent biomass yield from the converted land and biofuel yield at the refinery plant, respectively.  $CF_b$  ( $\text{Mg MJ}^{-1}$ ) and  $CF_f$  ( $\text{Mg MJ}^{-1}$ ) represent the life cycle GHG emissions (i.e., carbon footprint) of biofuels and fossil fuels per MJ, respectively. The denominator is the annual repayment, which represents the amount of carbon savings resulting from the use of biofuels from the converted land each year. Both the carbon debt

and the annual repayment depend on the type of land converted. Clearing forest, for example, incurs a larger carbon debt than clearing grassland. For annual repayment, the type of land affects the biomass yield ( $Y_l$ ) and the farming practices used, which, together with the biofuel conversion technology ( $Y_r$ ), determine the biofuel's carbon footprint.

Naturally, land with higher yield produces a larger carbon benefit per unit of biofuel and a larger volume of biofuel to displace equivalent fossil fuel and hence a higher annual repayment and shorter CPT. In summary, biofuel's CPT hinges on the type of land converted, the farming practices used, the biomass yield of the converted land and the biorefining technology at the time of the land conversion and how this technology advances in the future.

As discussed earlier, however, the CPT calculation in Eq. (1) treats annual carbon repayment as a fixed quantity and gives equal weights to emissions occurring in different times. To enhance the accuracy of corn ethanol's CPT estimates, we propose the following approach in which CPT is calculated as the minimum  $t$  value that satisfies the following inequality:

$$a \times CD \leq \sum_t Y_{l,t} \times Y_{r,t} s_t (CF_{f,t} - CF_{b,t}), \quad t = \{0, 1, 2, \dots\} \quad (2)$$

where subscript  $t$  stands for the number of years since land conversion and  $s_t$  is the temporal GHG emission sensitivity at year  $t$  relative to year  $t=0$ . By incorporating a time dimension ( $t$ ) to crop and ethanol yields and the fuels' carbon footprint, Eq. (2) allows the productivity of the fuel systems to change over time (see 2.2.). Further, the addition of  $s_t$  accounts for the varying cumulative climate impacts of GHG emissions and savings occurring in different times (see 2.3.).

## **2.2 Annual carbon savings from the Conservation Reserve Program-corn ethanol system**

We start with estimating the amount of annual carbon savings that can be generated by corn ethanol from an average cornfield and how the amount changes over time. For this analysis, we use the Biofuel Analysis Meta-Model (EBAMM) with several modifications (Farrell et al. 2006). Specifically, because the base year of EBAMM is 2001 (Shapouri et al. 2004), we incorporate into the model historical data on the process inputs and outputs of corn growth and ethanol conversion for 2005 and 2010 to reflect the system's productivity improvements in the past decade (detailed information is provided in Appendix B). We project further productivity improvements to 2020 using projections in the Greenhouse gases, Regulated Emissions and Energy use in Transportation (GREET) model (Wang 2013). We assume that technology advancement stabilizes after 2020 (Wang 2013). Detailed information is provided in Appendix B.

We then incorporate yield differences into the model to approximate the amount of annual carbon savings that the CRP-corn ethanol system provides. The CRP program, established by the Food Security Act of 1985, is intended to retire highly erodible and environmentally sensitive cropland from production (Sullivan et al. 2004). Because highly erodible land is less productive in general, the program enrolls land with lower productivity indirectly (Lubowski et al. 2006). Additionally, due in part to the early payment scheme—the maximum acceptable rental rates—farmers tended to offer their low-quality land for CRP consideration while retaining productive land for continuous cultivation (Sullivan et al. 2004). As a result, CRP land appears less productive than other types of cropland, including land that shifts into or out of the cultivated cropland from less, other intensive uses (e.g., hay

to corn) (Lubowski et al. 2006). Direct measurements of crop yield on CRP land are scarce, but measurements of crop yield on marginal land, including CRP and shifting land, can be used as indications of the relative yield differences between CRP land and average croplands (Keeney 2010). Estimates of the ratio of marginal to average (MtA) yield for US corn based on different methods range from 47 to 82 % (Hertel et al. 2010; Keeney 2010). This range is consistent with estimates on the global scale (Taheripour et al. 2012). In our analysis, we test how different MtA yield ratios (i.e., 50, 60, 70 and 80% of the yield of existing cornfields) affect ethanol's CPT. We also identify the best scenario for CRP land with comparable fertility (100 %) because the CRP program can sometimes retire highly productive land (Sullivan et al. 2004).

For the carbon debt caused by CRP land conversion, we use the field measurement by Gelfand et al. (2011), estimated at  $68 \text{ Mg CO}_2\text{e ha}^{-1}$ , with the assumption that no-till practices are used for corn farming after land conversion. This estimate is similar to that by Fargione et al. (2008) for CRP land conversion, i.e.,  $69 \text{ Mg CO}_2\text{e ha}^{-1}$ . Following Fargione et al., 83 % of the total carbon debt is allocated to ethanol and 17 % to coproducts, primarily distiller grains with solubles (DGS), based on their economic values (see Appendix B for sensitivity analysis using the system expansion method).

### **2.3 Accounting for emissions timing**

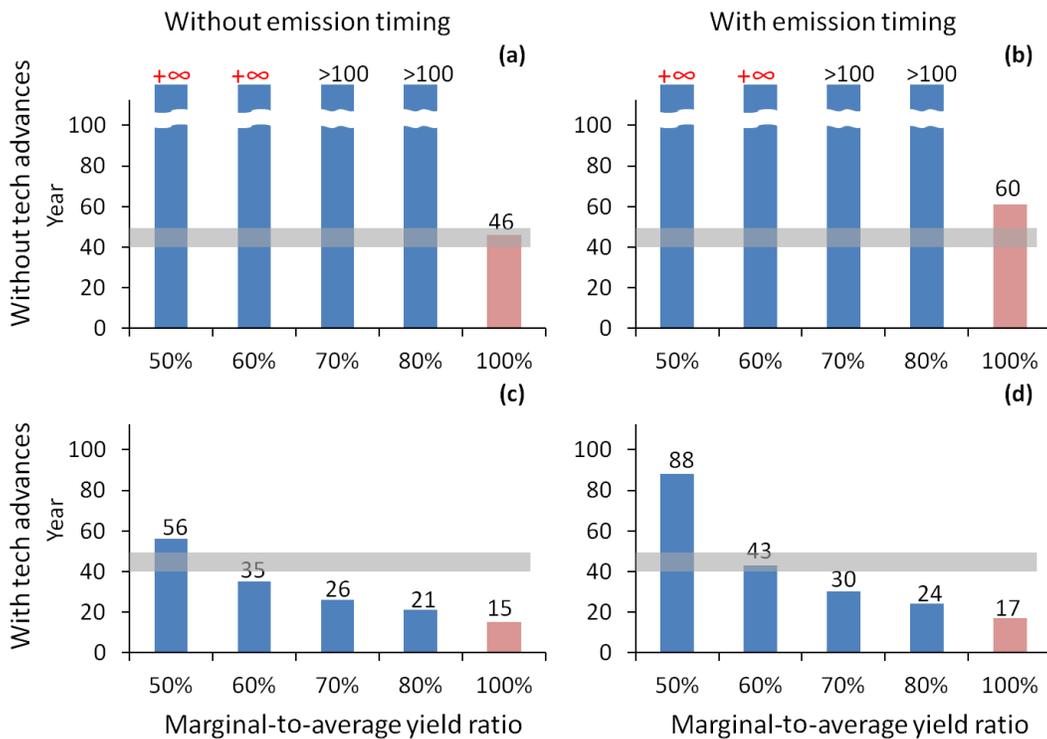
As discussed in the Introduction section, a number of approaches to accounting for emission timing effects in LCA and carbon accounting are reported in the literature with various rationales. These approaches are referred to as dynamic characterization in this paper. Dynamic characterization uses temporally specific emissions and characterization factors instead of using time-integrated life cycle inventory (LCI) and characterization

factors (Kendall 2012). In general, emissions and characterization factors are calculated for each annual time-step and summed up to produce a cumulative impact over a certain period of time. Just like any other characterization methods, dynamic characterization approaches are not free from subjective choices. In this study, we select 100 years (following land conversion) as the time horizon, and following the approach by Levasseur et al. (2010), we then calculate the cumulative radiative forcing (CRF), over the 100-year time horizon, for 1 kg CO<sub>2</sub> emitted in different years. Similar to Kendall (2012), we further normalize the CRF results of different years by that of the year when the carbon debt occurs ( $t=0$ ). This step yields a set of weights of decreasing value (temporal GHG emission sensitivity,  $s_t$ ) from 1 for year 0, 0.5 for year 60, to nearly 0 towards year 100. Finally, we assume that carbon debt occurs all at once in the year of land conversion ( $t=0$ ). The assumption of instantaneous carbon loss is somewhat unrealistic but can be considered as a worst-case scenario to indicate the impact of considering emissions timing. Following the argument of Hellweg et al. (2003), we do not further discount future emissions, a practice that is common in economics to account for time value of money. The general approach to CPT calculation outlined in this paper could, however, employ other dynamic characterization approaches discussed earlier; we select the approach by Levasseur et al. (2010) with 100-year time horizon only for the purpose of illustration.

### **3 Results**

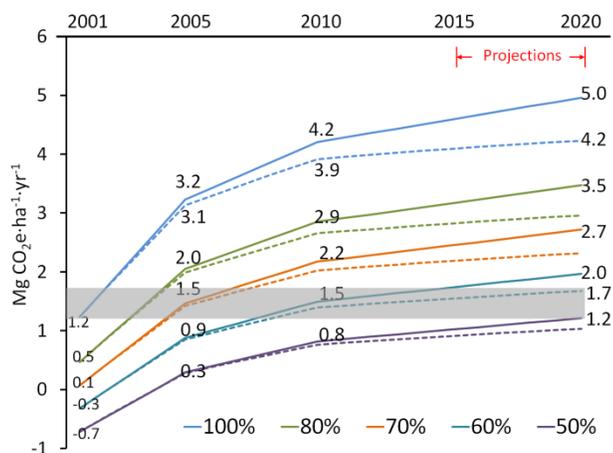
Taking into account potential corn yield differences, productivity improvements within the corn ethanol system, and emissions timing, our analysis shows that the CPT ranges from 43 to 24 years for CRP land with MtA yield ratios from 60 to 80 % (Fig. 3.1d), taking 2001

as the base or land conversion year ( $t=0$ ). In other words, these lands would start to generate carbon benefits from year 24 to 43 after land conversion. For CRP land of low soil fertility with 50 % of average yield, however, the CPT is estimated to be 88 years, which is more than double the CPT estimate for CRP land with 60% of average yield. If the converted land, on the other hand, is highly productive with MtA yield ratio of 100 %, the payback time would be as short as 17 years. Previous estimates of 40 and 48 years of payback time (Fargione et al. 2008; Piñeiro et al. 2009; Gelfand et al. 2011) are close to our estimate for 60 % MtA yield ratio, which is 43 years (Fig. 3.1d), but with very different underlying reasons.



**Fig. 3.1** Carbon payback time for the CRP-corn ethanol system under different MtA yield ratios, with or without consideration of technological advances and emission timing. Panel (a): payback time without consideration of technological advances nor emission timing. Panel (b): payback time considering emission timing but not technological advances. Panel (c): payback time considering technological advances but not emission timing. Panel (d): payback time considering both technological advances and emission timing. Grey bars indicate previous CPT estimates, namely, 40 to 48 years

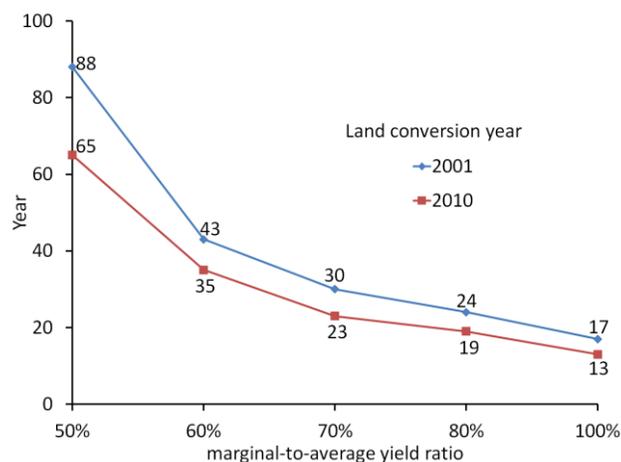
The effect of considering emissions timing increases corn ethanol's CPT by 2–32 years (difference between Fig. 3.1c, d). The more productive the CRP land, the less substantial the impact of emissions timing is. The most substantial changes in CPT are associated with technological improvements. Without technological advances, CRP land with  $\leq 80\%$  average yield would not produce any carbon benefits over the 100- year time horizon (Fig. 3.1b), and CRP land as productive as average cornfield would need as long as 60 years (Fig. 3.1b) to pay back the carbon debt from land conversion as opposed to 17 years when technological advances are considered.



**Fig. 3.2** Increasing annual carbon savings due to technological advances within the corn ethanol system, with and without consideration of emissions timing, indicated by dashed and solid lines, respectively. The shaded area reflects previous estimates ( $1.2$  to  $1.7$   $\text{Mg CO}_2\text{e}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ). Values are generated for 2001, 2005, 2010, and 2020, with a linear increase of annual carbon savings assumed for the years in between

Figure 3.2 illustrates the opposing effects of emissions timing and technological advances on annual carbon savings. Considering temporal sensitivity of GHG emissions

reduces the climate benefit of carbon savings (dashed lines), and the farther into the future, the lower the benefit per Mg carbon savings provide over the 100-year time horizon. For example, using the conventional GWP approach without considering temporal sensitivity, CRP land as productive as average cornfield would provide a saving of 5.0 Mg CO<sub>2</sub>e ha<sup>-1</sup> in 2020, which is the same for all years. But given the diminishing climate effect of later carbon emissions (or savings), saving 5.0 Mg CO<sub>2</sub>e in 2020 would be equivalent to saving 4.2 Mg CO<sub>2</sub>e in 2001. On the other hand, technological advances could substantially increase the amount of annual carbon savings the CRP-corn ethanol system provides (Fig. 3.2). By 2020, the amount of carbon savings provided by CRP land with ≥60 % of average yield would all exceed previous estimates of 1.2–1.7 Mg CO<sub>2</sub>e ha<sup>-1</sup>, with highly productive CRP land providing 2.5–3.5 times as much. It is worth noting that CRP land with only 50 to 60 % of average yield would provide negative carbon savings in the first years after land conversion.



**Fig. 3.3** Comparison of carbon payback time for CRP land converted to corn ethanol production in 2001 and 2010, with technological advances and emission timing taken into account

Lastly, the influence that the choice of the base year has on CPT estimates is tested. As Fig. 3.2 illustrates, annual carbon savings produced by the CRP-corn ethanol system are determined by corn farming and ethanol conversion technologies at the time. Therefore, as the technologies change over time, so does the amount of annual carbon savings. To adequately reflect the effect of technology change on the CPT, the carbon savings need to be calculated dynamically, a point that was also overlooked in previous studies (Fargione et al. 2008; Piñeiro et al. 2009; Gelfand et al. 2011). The payback times shown in Figs. 3.1 and 3.2 are calculated based on the assumption that land conversion occurs in 2001. However, if land is converted later at a time when the corn ethanol system is more productive, the CPT would be further shortened. Figure 3.3 shows that if CRP land were converted in 2010, the payback time would be 65 to 13 years, in contrast to 88 to 17 years for land conversion occurring in 2001.

#### ***4 Discussion and conclusions***

We have re-examined corn ethanol's carbon payback time (CPT) in the case of converting CRP land for corn ethanol production taking into account three factors that were neglected in previous studies: (1) yield differences on newly converted land, (2) productivity improvements within the corn ethanol system, and (3) emissions timing (dynamic characterization). Our results show that CPT estimates for converting low-fertility CRP land with 50 % marginal-to-average yield ratio ranges from 65 to 88 years (Fig. 3.3). For highly productive CRP land, the payback time could be reduced to less than 20 years. For CRP lands with 60–80 % of marginal-to-average yield ratio, which is considered to be a more typical case, the payback time range from 19 to 43 years (Fig. 3.3). Previous estimates of 40

and 48 years of payback time are near the upper bound of our estimates. Technological advances within the corn ethanol system are the key for the CRP-corn ethanol system to be able to generate positive climate impacts. Without technological advances, CRP land with  $\leq 80\%$  of marginal-to-average yield ratio would fail to provide any carbon benefits over the 100 years after the land conversion. Note that our study does not consider the reversion of land use, which, if included, would further shorten our estimates of CPT for the CRP-corn ethanol system (Delucchi 2011).

Overall, our study confirms the importance of understanding marginal technologies and efficiency changes in LCA (see also Gavankar et al. 2014); LCAs based on a static productivity assumption may fail to recognize the long-term benefits of the technology as it matures. Also, our study demonstrates the relevance of considering the actual yield of the converted land rather than the average yield, as direct corn expansion will most likely bring marginal, less-fertile land (e.g., CRP land) into production.

One of the key questions in biofuel policies is whether *additional* corn ethanol production would reduce GHG emissions. Therefore, LCA studies based on *average* data from existing corn ethanol systems fall short of offering adequate insights for the policy questions at hand. Ideally, such policy questions can be answered using a prospective model that embraces the complex dynamics between and within marginal technologies, marginal impacts, displacement mechanisms and behavioral changes. Our analysis highlights the importance of taking the underlying dynamics into account in understanding the implications of a technology, which can be referred to as “consequential thinking” as an *analytical paradigm* (see also Sandén and Karlström 2007). However, we acknowledge and admit that our analysis neglects many other factors that would influence the system. That is

one of the main reasons why we believe that the term, “consequential LCA”, which implies the existence of a well-defined, *operational model* (as opposed to an *analytical paradigm*) that is capable of showing the future trajectories of human-nature complexity, can be misleading (Suh and Yang 2014). Instead, our study employs a scenario approach to answer what-if questions focusing on marginal yield and ethanol system productivity. Needless to say, our results shall be interpreted only under the assumptions employed as well as the limitations associated with them.

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## **Chapter 4: Changes in environmental impacts of major crops in the USA**

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## ***Abstract***

As with life cycle assessment (LCA) studies in general, agricultural LCAs often rely on static and outdated inventory data, but literature suggests that agricultural systems may be highly dynamic. Here, we applied life cycle impact assessment (LCIA) methods to investigate the trends and underlying drivers of changes in non-global environmental impacts of major crops in the U.S. The results show that the impact per hectare corn and cotton generated on the ecological health of freshwater systems decreased by about 50% in the last decade. This change is mainly due to the use of genetically modified (GM) crops, which has reduced the application of insecticides and relatively toxic herbicides such as atrazine. However, the freshwater water ecotoxicity impact per hectare soybean produced increased by 3-fold, mainly because the spread of invasive species, soybean aphid, has resulted in an increasing use of insecticides. In comparison, other impact categories remained relatively stable. By evaluating the relative ecotoxicity potential of a large number of pesticides, our analysis offers new insight into the benefits associated with genetically modified (GM) crops. Our finding that different impact categories show different degrees of changes suggests that agricultural inventory data may be updated selectively in LCA.

**Keywords** Agriculture · Life cycle assessment · Ecotoxicity · Genetically modified organism · Pesticides · Invasive species

## ***1 Introduction***

Agriculture is essential for feeding a majority of the global population, but it has also been identified as one of the major drivers behind various global environmental degradations (Socolow 1999; Rockström et al. 2009; Yang and Suh 2011). For example, due to a quintupling of global fertilizer use in the past decades, agriculture has greatly disturbed the global nitrogen and phosphorus cycles (Foley et al. 2011). This results in a wide range of environmental issues from release of N<sub>2</sub>O, formation of photochemical smog over large regions of earth, to accumulation of excessive nutrients in estuaries and coastal oceans (Socolow 1999). Agriculture dominates pesticide use (Grube et al. 2011), which contaminates surface and ground water and threatens human and ecological health (Pimentel 2005; Gilliom 2007). So also does agriculture dominate freshwater withdrawal worldwide (Gleick et al. 2006), adding stresses where there are competing needs for water (Rosegrant et al. 2009). Despite the severity of existing environmental impacts of agriculture, the challenge of addressing them is compounded by increasing global food demand (Godfray et al. 2010). Continuous global population growth and spread of economic prosperity (Braun 2007), mainly in developing countries, will likely drive the global food demand to double by 2050 (Tilman et al. 2011).

Over the past decade, life cycle assessment (LCA) has been increasingly applied to agricultural and food products (Mourad et al. 2007; Roy et al. 2009), with a number of agricultural LCA databases developed worldwide recently (CleanMetrics 2011; Blonk 2014; Nielsen et al. 2014; Quantis 2014; USDA 2014). LCA is a tool that quantifies products' environmental emissions and resource use throughout the life cycle and evaluates the potential impacts they generate on human and ecological health (ISO 2006a). Impact

categories evaluated in LCA span a wide range, from global warming, ozone depletion, acidification, eutrophication, to ecotoxicity, human health cancer, and non-cancer (Finnveden et al. 2009). Applications of LCA in agriculture include comparing the environmental performance of alternative products or technologies (Pelletier et al. 2010), such as organic versus conventional farming (Thomassen et al. 2008), and identifying hotspots and improvement opportunities (Blengini and Busto 2009). In particular, LCA has played an active and important role in assessing the environmental benefits of bioenergy (Cherubini and Strømman 2011) and contributed to the making of public climate policies (EPA 2010).

As with LCA studies in general, agricultural LCAs often rely on static and single-year inventory data with commonly 5 to 10 years of data age. In Ecoinvent (version 2.2) database, for example, the data year for *U.S. Corn Farming* is around 2005 and for *Swiss Corn Farming* is around 2000 (Ecoinvent 2014). Literature suggests, however, that agricultural systems may be highly dynamic due in part to the increasingly changing climate (USDA 2013a) and technological advances such as improved yield and energy efficiency (Fuglie et al. 2007). These factors may bring about substantial changes in the use of input materials and the yield of crops, hence substantial changes in the environmental impacts. For example, direct energy inputs per ha corn produced in the U.S. declined by about 40% between 1996 and 2005 and in the meantime corn yield increased by about 30% (Shapouri et al. 2010).

In this study, we seek to evaluate if on-going changes in input use and structure of four major crops in the U.S. might have resulted in substantial changes in their environmental impacts over the past decade, focusing on regional issues such as eutrophication,

acidification, and ecological toxicity. The crops studied are corn, soybean, wheat, and cotton, which together account for around 70% of total harvested area domestically (USDA 2012). The main objectives of the study are to understand the extent to which different environmental impacts might have changed and to identify major drivers behind such changes.

## ***2 Materials and methods***

### **2.1 Method**

Following previous LCA studies (Kim and Dale 2005; Fargione et al. 2008; Kim et al. 2009) we analyzed the cradle-to-gate life cycle environmental impacts of 1 ton and 1 hectare (ha)-year of crop production. The system boundary covers both direct and indirect emissions associated with crop cultivation and harvest. Direct emissions, such as nutrient leaching and runoff, result from the use of agricultural inputs, and indirect emissions that occur along the supply chain, including emissions from production and transportation of agricultural inputs like synthetic fertilizers. We focused on the estimation of direct emissions for their substantial contribution to the overall life-cycle environmental impacts of crops (Yang et al. 2012; Yang and Suh 2015a), and used the Ecoinvent database (version 2.2) to calculate indirect emissions (Ecoinvent 2014).

We began with collecting data on the use of agricultural inputs in different years, and then estimated associated emissions based on emission statistics and models. The emission data compiled were next aggregated using characterization models from Life Cycle Impact Assessment (LCIA) (ISO 2006b) to quantify their relative magnitudes of environmental impact. Equation 1 summarizes this calculation:

$$E_{i,k} = \sum_j C_{i,j} (m_{j,k}^D + m_{j,k}^I) \quad (1)$$

where  $i$  denotes impact category,  $k$  crops, and  $j$  environmental emissions.  $m^D$  and  $m^I$  represent direct and indirect emissions, respectively. And  $C$  represents characterization factors used to aggregate emissions  $j$  into characterized environmental impact scores  $E$ .

A characterization factor in LCA reflects the potency of an environmental exchange relative to that of a reference exchange for a given impact category (Bare et al. 2003). Global warming potentials, for example, are commonly used characterization factors in LCA for the impact category of climate change. Characterization factors used in this study are from the Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI) version 2.0 developed by the U.S. Environmental Protection Agency (EPA) (Bare 2011). As our study targeted non-global impacts, the impact categories selected from TRACI 2.0 are acidification (air), eutrophication (water), smog formation, freshwater ecotoxicity, and human health criteria (air), cancer, and non-cancer (e.g., reproductive, developmental, and neurotoxic effects). Categories excluded from TRACI 2.0 are global warming, ozone depletion, and eutrophication (air). We also included irrigation water use as an indicator of the stress crops place on the environment. We excluded water use embodied in inputs partly because of a mismatch between the data years for irrigation water and other inputs like fertilizers (see sections 2.2.) and partly because embodied water in agricultural inputs is generally negligible relative to irrigation water use (Chiu et al. 2009; Yang and Suh 2015a).

## 2.2 Data on agricultural inputs

Major agricultural inputs include fertilizers, pesticides, irrigation water, and energy (Yang et al. 2012). Data on fertilizer and pesticide use are from the U.S. Department of Agriculture (USDA) (USDA 2013b), which surveys farmers in top-producing states annually on a rotating basis (table 4.1). We selected the years with the largest number of states covered for each crop to best represent U.S. national situations. We found that top-producing states were consistently surveyed in the years selected for each crop, which ensures comparability across years. For example, the same 19 states were covered for corn and they accounted for around 95% of total corn area harvested in each of the years selected. Similarly, the same 9, 19, and 15 states were covered for cotton, soybean, and wheat, and these states accounted for around 92%, 96%, and 88% of total area harvested, respectively.

**Table 4.1** Number of states surveyed by USDA between 2000 and 2012<sup>a</sup>

	2000	11	12	12	14	15	16	17	18	19	20	21	2012
Corn	18	19	7	18		19					19		
Cotton	11	7		12		9		11			7		
Soybeans	18	7	20		11	17	19						19
Wheat													
Durum	1		1		2		2			3			2
Spring	4		3		7		6			7			4
Winter	16		10		14		14			15			13

<sup>a</sup> Values in shade indicate years selected for analysis for each crop.

Irrigation water use data are from the Farm and Ranch Irrigation surveys conducted also by USDA (USDA 2015b), and the most recent three surveys for 2002, 2007, and 2012 were used for our analysis. State-level energy use data were also compiled from the USDA (USDA 2004), but the data are somewhat outdated as they reflect crops planted in late 1990s

or early 2000s. USDA has unfortunately ceased to update such data for most crops except for corn, which was updated to the year 2005 (Shapouri et al. 2010). On the other hand, farms have become more efficient in response to rise in fuel and fertilizer prices in the last decade (USDA 2008). For example, on-farm energy use in corn production reduced by >20% between 2001 and 2005 (Shapouri et al. 2010). To reflect the trend of farm energy efficiency gains, we adopted the estimates from the widely used GREET model (Wang 2013), which shows an efficiency increase of about 30% for corn and soybean growth over the last decade. Few studies exist on cotton and wheat on-farm energy change, thus we assumed a similar 30% efficiency gain for them over the timescale investigated. Note that we did not consider nitrogen from manure considering that it is small relative to other nitrogen sources (USDA 2006). We estimated nitrogen input from biological fixation for soybean (see Appendix C).

### **2.3 Direct environmental emissions**

Building on our previous studies (Yang et al. 2012; Yang and Suh 2015a), we estimated a large number of emissions (>100) from all the agricultural inputs applied based on emission factors from various models and references (see table 4.2). Most of the emissions are pesticides and speciated Volatile Organic Compounds (VOCs). Estimation of pesticide emissions was slightly more complicated than that of other emissions, thus a detailed explanation is in order. Several approaches to pesticides emissions have been applied in literature and LCA databases. For example, the Ecoinvent (v2.2) database assumes that all pesticides remain in soil after application (Ecoinvent 2014). The PestLCI model, on the other hand, treats agricultural soil as part of the technosphere and excludes the impacts of

pesticides on ecosystems in the soil (Birkved and Hauschild 2006). And yet there is another approach that estimates pesticide emissions to different compartments (i.e., air, soil, and water) (Berthoud et al. 2011; Laurent et al. 2012; Yang et al. 2012). We adopted the third approach here. Following Berthoud et al. (2011), we used a pesticide’s vapor pressure to approximate its air emissions, assumed a generic factor of 0.5% of the total applied for pesticides lost to water systems through runoff and leaching, and assumed the remaining fraction, capped at 85% of the total applied, for pesticides emitted to soil.

**Table 4.2** Estimation of direct environmental emissions from agricultural inputs

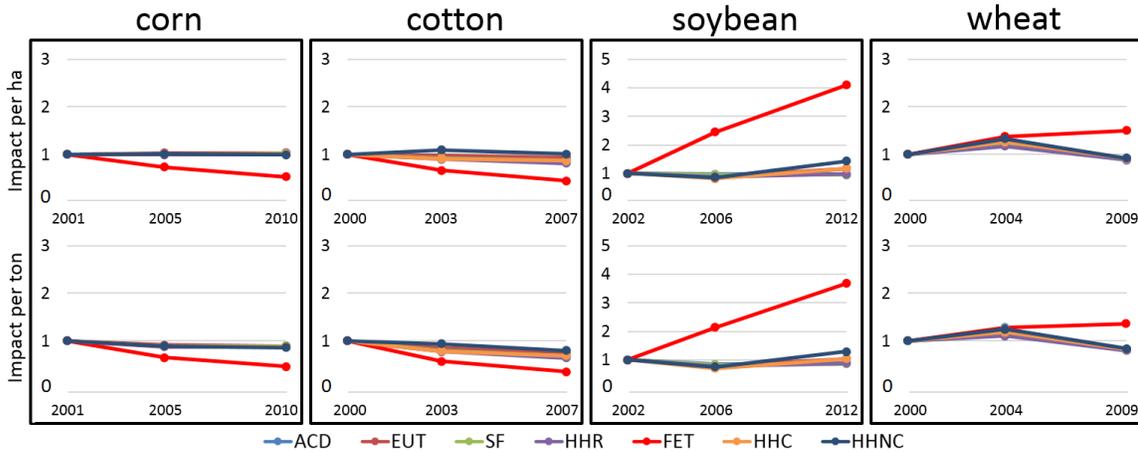
<b>Sources</b>	<b>Environmental emissions</b>	<b>References</b>
N input	NH <sub>3</sub> to air	(Krauter et al. 2002; Goebes et al. 2003)
	NO <sub>x</sub> to air	(Yienger and Levy 1995)
	N runoff and leaching	(USDA 2006)
P fertilizers	P runoff and leaching	(USDA 2006)
	Heavy metals to soil	(Mortvedt 1995; Yang 2013)
	Pesticides	(Herner 1992; Berthoud et al. 2011)
Pesticides	Emissions to air	(USDA 2000)
	Runoff and leaching	(Berthoud et al. 2011)
	Emissions to soil	(Berthoud et al. 2011)
Farm equipment	NO <sub>x</sub> , SO <sub>x</sub> , PM <sub>2.5</sub> , PM <sub>10</sub> , CO	(Wang 2013)
	Speciated VOCs	(EPA 1995)

Last, the data we compiled are at the state level, but given our emphasis on the change of environmental impacts of U.S. agriculture on average we aggregated the state-level results to present totals. We also aggregated the three different types of wheat (winter, spring durum, and spring other) into one “wheat” by adding up their annual agricultural inputs and outputs. In deriving the impacts per ton of crop produced, we followed previous studies (Shapouri et al. 2002; Shapouri et al. 2010) and used 3-year average yield data to reduce annual variation caused by possible extreme weathers such as droughts and floods. For

example, 2001 impact per ton for corn was calculated by dividing 2001 impact per ha by the average corn yield of 2000, 2001, and 2002.

### 3 Results and discussion

#### 3.1 Changes in environmental impacts of U.S. major crops



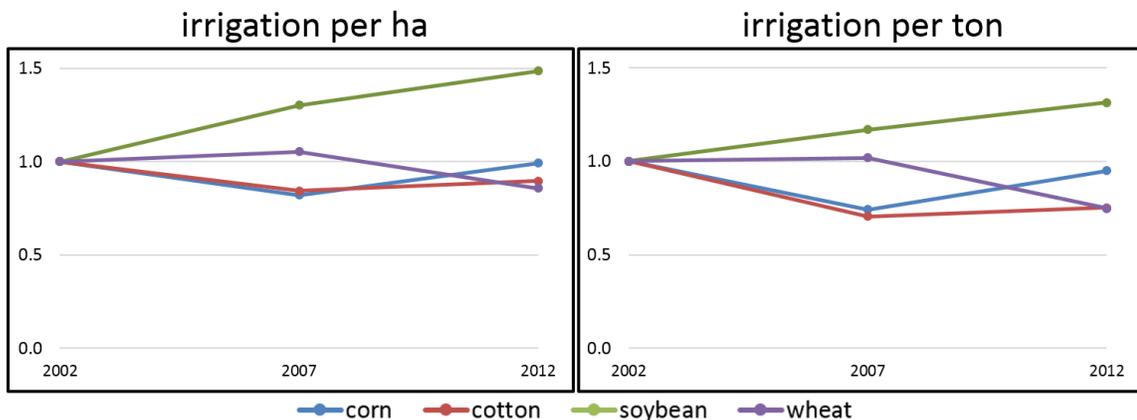
**Fig. 4.1** Changes in environmental impacts of U.S. corn, cotton, soybean, and wheat. Impacts for different years were normalized to that for the base-year and expressed on the basis of impact per ha and per ton. ACD = acidification, EUT = eutrophication, SF = smog formation, HHR = human health respiratory, FET = freshwater ecotoxicity, HHC = human health cancer, and HHNC = human health non-cancer.

Figures. 4.1 and 4.2 present our main results; because irrigation data span a different time frame, it is presented in a separate figure. The major finding of our analysis is that freshwater ecotoxicity is the most dynamic of all impact categories, while the change is not unidirectional across the crops studied. We elaborate on this impact category, including its major contributors and probable drivers in the next subsection (3.2.). Here we focus on other impact categories.

Non-ecotoxicity categories were relatively stable over the past decade. Mostly, they changed 10-20% for each crop within the timescale studied. This is mainly because nutrient

inputs – particular nitrogen – are the major contributor for many of these impact categories. The use of nutrients result in both direct emissions, such as NH<sub>3</sub>, NO<sub>x</sub>, and nitrogen and phosphorus runoff, and indirect emissions embodied in the inputs, such as NO<sub>x</sub> released from the production of fertilizers. Also, fertilizers, particularly phosphate, introduce heavy metals into agricultural soils (Mortvedt 1995). The total amount of nitrogen and phosphonate inputs remained largely unchanged for all of the crops over the periods investigated, and this is the main reason that most of the non-ecotoxicity impacts do not show a substantial change.

For corn, soybean, and wheat, nutrients in general account for >75% of the non-ecotoxicity impacts (i.e., acidification, smog formation, eutrophication, human health cancer, non-cancer, and respiratory impacts). For cotton, energy use was intensive, about 2 times that of corn. As a result, nutrients account for around 50-80% for the non-ecotoxicity categories, while energy use contributes 25%, 40%, and 50% for acidification, smog formation, and human health respiratory impacts. For all crops, heavy metals introduced by phosphate fertilizers were identified to be the major contributor (60%-90%) to human health non-cancer effect.



**Fig. 4.2** Changes in average on-farm irrigation water use per ha and per ton crop produced, with results for

different years normalized to that for 2002. Results for irrigation water per ha were derived from (*irrigation intensity for irrigated area* × *area irrigated*) / *total area harvested*.

As Fig. 4.2 reflects, changes in the average irrigation water use from 2002 to 2012 were also moderate for corn, cotton, and wheat, with variations <20% between 2002 and 2007 or between 2002 and 2012. In contrast, a noticeable upward trend can be observed for soybean. Average irrigation water use per ha soybean produced increased by around 50%, from 180 m<sup>3</sup> in 2002 to 270 m<sup>3</sup> in 2012. On a per ton basis, the percentage increase is 30%, from 4300 to 5600 m<sup>3</sup>, due to yield increase over the period. Behind this upward trend are several factors, including the slightly increasing irrigation intensity for irrigated area, but the major contributor is the growth in area irrigated (from 2.2 to 3.0 million ha) and its share in the total area harvested (from 7% to 10%). What led to the growth in soybean area irrigated is unclear, however, and further research is needed. Here, we offer a possible explanation. In the past “ethanol decade,” soybean and corn areas substantially expanded, into other cropland and also grassland (Wallander et al. 2011; Wright and Wimberly 2013). Because such marginal land as grassland is on average not as fertile as existing corn or soybean land (Yang and Suh 2015a), irrigation might have been applied to boost or maintain yield. Consequently, as total soybean and corn areas expanded, so also did the area irrigated. In the case of corn, however, although area irrigated grew from 4.0 to 5.4 million ha between 2002 and 2012, its share in the total area harvested only slightly increased (from 14% to 15%). Additionally, irrigation intensity for area irrigated decreased from 1480 to 1234 m<sup>3</sup> ha<sup>-1</sup>. As a result, average irrigation use per ha or per ton corn produced barely changed from 2002 to 2012.

### 3.2 Changes in freshwater ecotoxicity impact of U.S. major crops

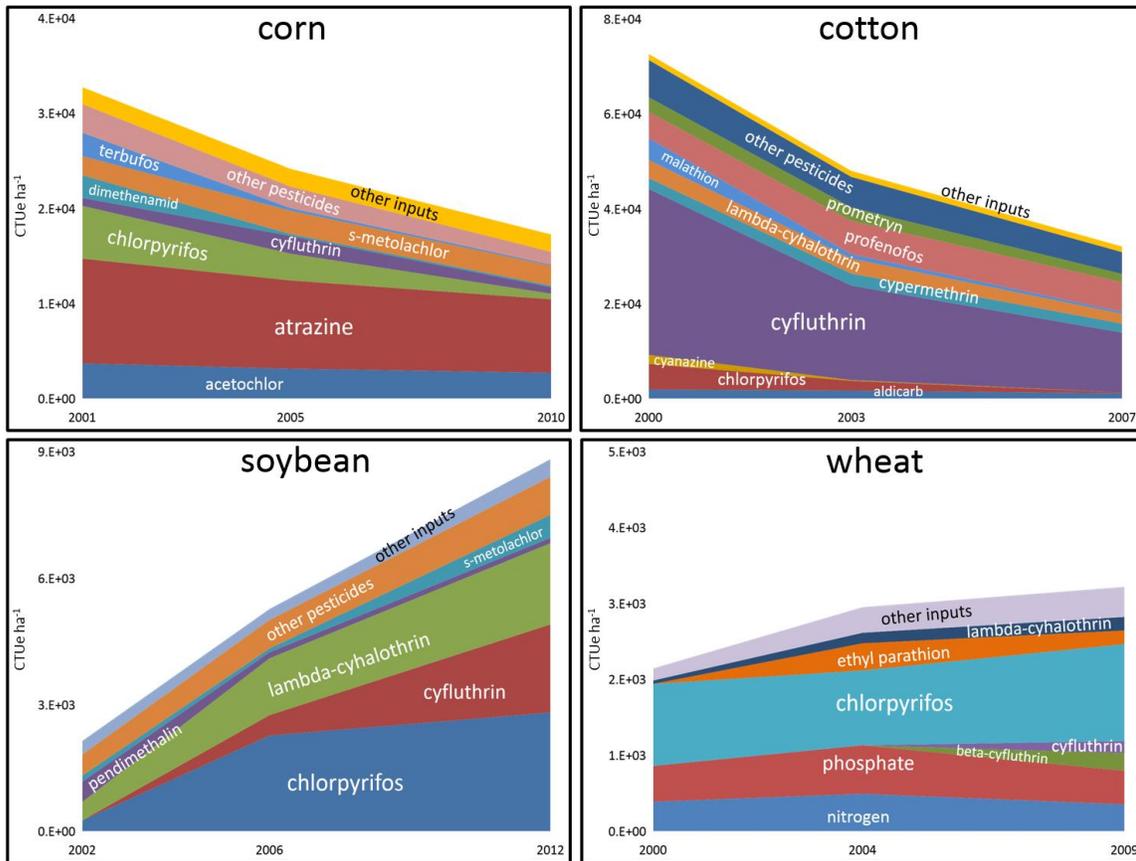


Fig. 4.3 Changes in the composition of the cradle-to-gate life cycle freshwater ecotoxicity impact per ha corn, cotton, soybean, and wheat produced over the past decade, expressed in comparative toxic units (CTU), which is a measure of the potentially affected fraction of species integrated over time and volume per unit mass of a chemical emitted.

As reflected in Fig. 4.1, freshwater ecotoxicity impact of corn decreased by around 50% from 2001 to 2010. Major contributors include reduced use of herbicides *atrazine* and *acetochlor*, and of insecticides *terbfos*, *dimethenamid*, and, especially, *chlorpyrifos* (Fig. 4.3). The downward trend is likely due to the continuous expansion of herbicide resistant (HR) and insect-resistant corn, particularly *glyphosate* tolerant and *Bt* (*Bacillus thuringiensis*) corn. Since its introduction in 1996, HR corn has now expanded to over 70%

of cornfield (Benbrook 2012), resulting in increasing use of *glyphosate* compounds in place of conventional herbicides like *atrazine* and *acetochlor*. In fact, *glyphosate* and related compounds had gradually surpassed *atrazine* and other herbicides over the past decade to become the most commonly applied pesticide (Yang 2013). As *glyphosate* compounds are relatively less toxic to ecosystems compared with the replaced herbicides like *atrazine* and *acetochlor* (Rosenbaum et al. 2008), the overall ecotoxicity impact of corn attributable to herbicides decreased moderately between 2001 and 2010. Meanwhile, *Bt* corn has also dominated U.S. cornfield now (Benbrook 2012), offering both economic and environmental benefits by protecting yield and reducing handling and use of insecticides (Hellmich and Hellmich 2012). This likely further contributed to the downward trend of corn's freshwater ecotoxicity impact.

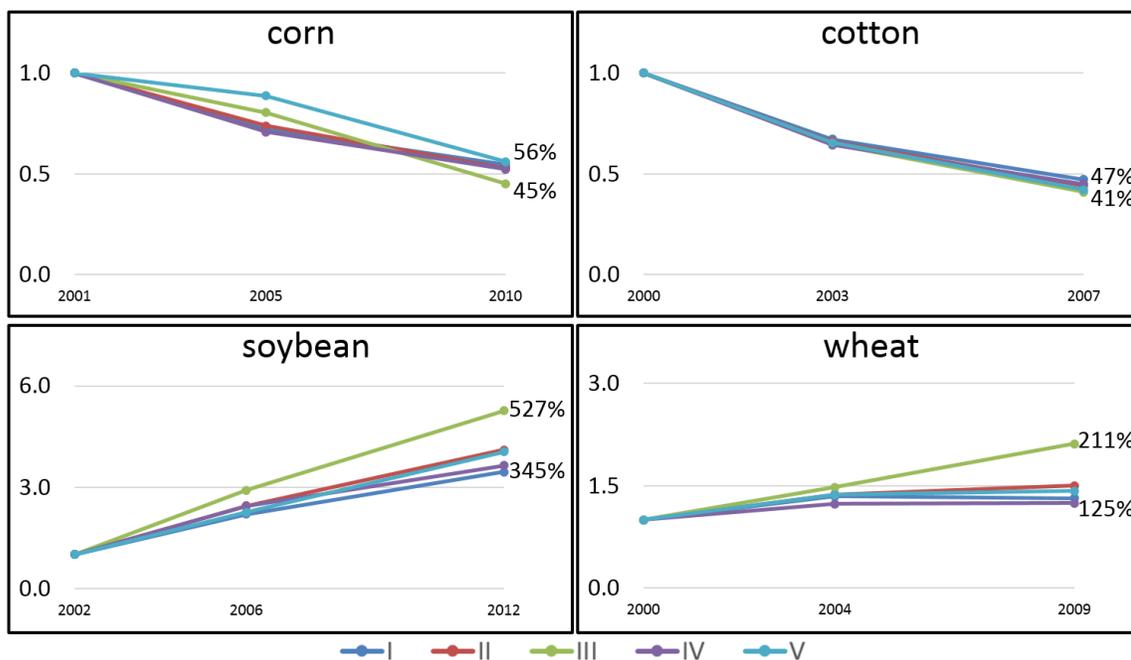
Similar to corn, the freshwater ecotoxicity impact of cotton decreased by 60% from 2000 to 2007, due to the reduced use of herbicides *chlorpyrifos*, *lambda-cyhalothrin*, and particularly *cyfluthrin* (Fig. 4.3). Application of *cyfluthrin* reduced from 11 g ha<sup>-1</sup> in 2000 to 4 g ha<sup>-1</sup> in 2007. Similar to corn, the downward trend in cotton's freshwater ecotoxicity impact was attributable to the expansion of HR and *Bt* varieties, which are now planted 95% and 75% of U.S. cotton field respectively (Benbrook 2012). Our result on decreasing freshwater ecotoxicity impact of corn and cotton due to changes in pesticide use and patterns reinforces previous findings (Duke et al. 2012; Hellmich and Hellmich 2012; Brookes and Barfoot 2012).

Unlike corn and cotton, soybean's freshwater ecotoxicity impact quintupled between 2002 and 2012. HR soybean has also expanded dramatically in the US, now planted on 95% of soybean field (Benbrook 2012). Along with the expansion, application of *glyphosate*

compounds per ha has increased by over 60% between 2002 and 2012, and now they account for 80% of total pesticides applied in soybean growth. However, the benefits of HR soybean seem to have been entirely offset by the increasing use of insecticides *lambda-cyhalothrin*, *cyfluthrin*, and *chlorpyrifos* (Fig. 4.3). This is due to the invasion of soybean aphid, a species native to eastern Asia and first detected in North America in 2000, and application of insecticides has been the primary means of pest management (Heimpel et al. 2013). Since its first detection, soybean aphid had rapidly spread to 30 states in the U.S. by 2009 and become a major source of economic loss in soybean production (Ragsdale et al. 2011). As a result, the total quantity of insecticides applied to soybean quadrupled between 2002 and 2012, resulting in a 3-fold increase in soybean's freshwater ecotoxicity impact.

The freshwater ecotoxicity impact of wheat increased by about 40% from 2000 to 2009, attributable partly to increased use of several insecticides including *chlorpyrifos*, *cyfluthrin*, *beta-cyfluthrin*, and *lambda-cyhalothrin*. Also, pesticide application rate in general increased from 0.45 kg ha<sup>-1</sup> in 2000 to 0.88 kg ha<sup>-1</sup> in 2009. Unlike the other major crops, however, there is not a clear explanation for the upward trend. One possible reason may be the growing resistance of pests as a result of increasing pesticide use. Further research is needed in this area.

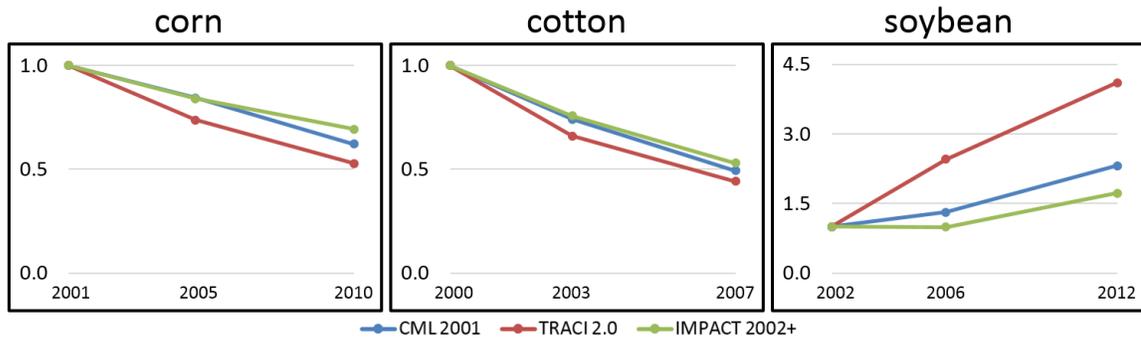
### **3.3 Sensitivity analyses**



**Fig. 4.4** Changes in freshwater ecotoxicity impact per ha crop produced based on different scenarios reflecting different pesticide runoff and leaching rates and approaches to pesticide emissions. Results for different years were normalized to that for the base-year. Shown in each panel are maximal and minimal ratios, as an indicator of the variation of the scenarios tested. Scenario I = low runoff and leaching rate (0.1%); II = moderate runoff and leaching rate (0.5%, adopted in this study); III = high runoff and leaching rate (5%); IV = pesticides assumed to remain all in soil (100%); V = pesticides to soil considered as part of the technosphere and excluded from the overall ecotoxicity impact (with moderate runoff and leaching rate (0.5%) and estimation of pesticides to air based on their vapor pressure).

We conducted sensitivity analysis to test the robustness of the changes in freshwater ecotoxicity impact, considering that it is our major finding and that large uncertainty is involved in the estimation of pesticide emissions and assessment of their ecotoxicity impact (Rosenbaum et al. 2008; Nordborg et al. 2014). First, the proportion in which pesticides are emitted to water systems was identified as the major contributor to crops' freshwater ecotoxicity. Literature also shows it may vary greatly, from 5% (USDA 2000) to 0.1% or even less (Laurent et al. 2012; Nordborg et al. 2014) (0.5% used in this study). We thus built 3 scenarios to test the sensitivity of the ecotoxicity result to different leaching and runoff rates. Additionally, we also tested the sensitivity of the trends to other analytical approaches

to pesticide emissions (see section 2.3.), with one assuming all pesticides to remain in soils and the other excluding the impact of pesticides on agricultural soils. All 5 scenarios are presented in Fig. 4.4, which reinforces the trends identified of freshwater ecotoxicity impact regardless of different runoff and leaching rates and analytical approaches to pesticide emissions.



**Fig. 4.5** Changes in freshwater ecotoxicity impact per ha crop produced using different LCIA characterization models, with a moderate runoff and leaching rate (i.e., 0.5%) assumed. Results for different years were normalized to that for the base-year.

Second, impact assessment of freshwater ecotoxicity is also highly uncertain, with the uncertainty range for TRACI 2.0 being likely 1-2 orders of magnitude (Rosenbaum et al. 2008). However, detailed information on the distribution of each characterization factor is not available yet, thus a full uncertainty analysis is not feasible at this stage. To further test the robustness of the ecotoxicity results, we applied two other characterization models (i.e., IMPACT 2002+ and CML 2001) (Jolliet et al. 2003; CML 2015) to evaluate the aquatic ecotoxicity impact of pesticide emissions. For corn, cotton, and soybean, the other two models confirm the directionality of the changes but generally show a lower magnitude of change (Fig. 4.5). This is due in part to differences in the number of pesticides covered by the three models and in part to differences in the relative ecotoxicity potential they assign to

each pesticide. Generally, IMPACT 2002+ and CML 2001 cover a smaller number of pesticides than TRACI 2.0, thus they may not capture all the changes in pesticide use and patterns that are captured by TRACI 2.0. For wheat, however, the three characterization models seem to disagree on the directionality as well as the magnitude of changes. A detailed comparison, together with contribution analysis, is provided in the Appendix C.

### **3.4 Implications**

In this study, we evaluated several non-global environmental impacts of U.S. corn, cotton, soybean, and wheat, and analyzed how they changed in the past decade. Due likely to the increasing adoption of genetically modified varieties, freshwater ecotoxicity impact per ha corn produced declined by around 50% from 2001 to 2010 and per ha cotton produced declined by 60% from 2000 to 2007. Due to the invasion of alien species (aphid) and increasing use of insecticides, freshwater ecotoxicity impact per ha soybean produced increased by 3-fold from 2002 to 2012. In the meantime, on-farm irrigation water use per ha soybean harvested increased by about 50%. In comparison, other non-global impacts were relatively stable.

The major implication of our study is that identifying the underlying drivers of the dynamical mechanisms in agricultural systems would be essential for making informed agricultural decisions and policies, prioritizing LCA data update needs, and interpreting LCA results. By evaluating the relative ecotoxicity potential of a large number of pesticides, we found that the use of GM crops have contributed to substantial declines in corn and cotton's freshwater ecotoxicity impact. This finding provides an opportunity for better assessing the tradeoffs between the potential impacts of GM and conventional crops, as

opposed to comparisons based mainly on the total quantity of pesticides applied (Benbrook 2012). Additionally, our results suggest that updates on agricultural inventory data can be done selectively, with regular updates needed for impact categories that are highly dynamic, such as pesticide related ecotoxicity. Studies relying on single-year and outdated data may inaccurately portray a crop's ecotoxicity impact; even just a few years of data age may under or overestimate the ecotoxicity impact. This also implies that we should exercise caution when interpreting an LCA study in which ecotoxicity impact of agricultural processes plays an important role in the overall conclusion. Broadly, our study highlights the importance of understanding the dynamics in the input and output structure of a process or a technology in LCA (Gavankar et al. 2014; Yang and Suh 2015b).

The focus of our study was to evaluate how environmental impacts of agriculture might have changed in the past decade. Our results that show decreasing freshwater ecotoxicity impacts for corn and cotton are not intended to prove that GM crops are overall more ecologically friendly than conventional crops. Other impacts of GM crops that could not have been evaluated due to the limitations of the current LCIA methods should also be taken into consideration in such comparisons. Current LCIA methods, for example, are not able to properly evaluate potential adverse effects of *Bt* toxin on populations of non-target species and elevated risk of species invasiveness through genetic modifications (Wolfenbarger and Phifer 2000). In addition, it should be noted that the trend of decreasing ecotoxicity impact is unlikely to continue for cotton and corn. Due to the dominant use of HR and *Bt* crops, pests and weeds have evolved to be increasingly resistant (Powles 2008; Tabashnik et al. 2013). As a result, farmers may need to resort to earlier pest control practices that rely more on conventional pesticides, hence increasing crops' freshwater ecotoxicity impact.

Nevertheless, the dynamics of pest management, and associated ecological impacts, further corroborates the importance of understanding the dynamics of agricultural systems.

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## Chapter 5: Answers to research questions and discussion

This chapter summarizes answers to the research questions proposed in the first chapter of the dissertation, and discusses some other important implications that were not covered in the chapters above.

### *1. Answers to research questions*

- *What are the implications of land use change from cotton to corn, both high-input crops, for local environments?*

For many of the impact categories studied, the environmental impacts of US corn and cotton on average are roughly comparable on a per hectare basis, while cotton consumes more water and generates much higher freshwater ecotoxicity impact. However, the average results, mainly reflecting corn produced in the Midwest and cotton produced in the South, are inadequate to capture the likely environmental consequences of corn expansion into cotton, which has taken place in cotton-growing states in the South. The state-level results show that a land use shift from cotton to corn relieves freshwater ecotoxicity but may aggregate many other regional environmental impacts. Due to the limitation of data, a definitive conclusion may not be drawn for other Southern states where the cotton-to-corn land use change has also occurred. But our finding of tradeoffs based on the three states is *probably* generalizable for these other states considering that cropland there is generally less suitable for corn production than in the Midwest.

- *What is the carbon payback time (CPT) in converting grassland to grow corn for ethanol, taking into account the actual corn yield of the grassland and technological advances of the corn ethanol system? If considering emission timing, how large is its effect on CPT estimates?*

Taking into account marginal yield and technological advances, the CPT for converting the CRP grassland for corn ethanol production in early 2000s, when the ethanol industry begun to grow, ranges from 15 years for highly productive grassland with average corn yield to 56 years for infertile grassland with only 50% of average corn yield. Considering the diminishing climate effect of later GHG emissions within a 100-year timeframe, the CPT estimates would increase to 17 to 88 years. Understandably, the shorter the payback time, the less strongly it would be affected by the consideration of emission timing. Technological advances are crucial to the climate benefit of the CRP-corn ethanol system. In the no technological advances scenario, most of the grassland would not produce any climate benefit within a 100-year timeframe. Even for the highly productive grassland, it would take up to 46 years before the system could start generate carbon savings. Last, because the technology and productivity of the corn ethanol system changes over time, the timing of land conversion also plays a part in the CPT estimation. If land conversion took place in 2010, the CPT estimates would be 13 to 65 years, as opposed to 17 to 88 years for land conversion occurring in early 2000s.

- *How have the environmental impacts of major crops changed in the past decade? Did different categories change differently? In particular, has and to what extent*

*the widespread deployment of genetically modified crops affected the pesticides-related ecotoxicity impact of these crops?*

The environmental impacts per hectare crop harvested for most of categories studied were relatively stable in the past decade. This is because these impact categories are dominated by the direct and indirect emissions of nutrient, particularly nitrogen fertilizers, and the amount of nutrient inputs did not change much in the past decade. In contrast, the freshwater ecotoxicity impact per ha corn harvested declined by around 50% from 2001 to 2010 and per ha cotton produced declined by 60% from 2000 to 2007. These downward trends are due in large part to the increasing adoption of genetically modified organisms (GMO), which have resulted in reduced use of insecticides and replacement of some conventional herbicides with more benign ones, particularly glyphosate and compounds. Soybean production in the USA has also adopted GMOs widely, and this should have also led to a decline in soybean's freshwater ecotoxicity as with corn and cotton. But because of the invasion of soybean aphid, a native of Asia, which have resulted in a substantial increase in insecticides use, the freshwater ecotoxicity impact per hectare soybean harvested increased by a factor of 4 from 2002 to 2012. In the meantime, on-farm irrigation water use per ha soybean harvested increased by about 50%. This increase is due *probably* to the expansion of soybean into marginal land where intensive irrigation is needed.

## ***2. Discussion***

Implications of the above findings have been extensively discussed in individual chapters. Discussed below is the implication of considering marginal yield in the case of

direct land use change (dLUC) for studies of indirect land use change (iLUC) and consequential LCA modelling. Some of the points, such as the importance of additional corn and carbon, have been somewhat touched upon in (Searchinger 2010), but the discussion here is more detailed and from a methodological point of view.

## **2.1 Land use change and consequential LCA**

Early LCA estimates differed with respect to whether corn ethanol offers carbon benefits in displacing gasoline (Farrell et al. 2006; Babcock 2009). Notably, the findings of the Cornell Professor David Pimentel were all negative (Pimentel 2003; Pimentel and Patzek 2005), leading him to strongly oppose the use of corn ethanol (also due to concern over soil, air, and water pollution associated with corn production) (Pimentel 2008). But subsequent LCA studies, with updated data and ethanol coproducts correctly accounted for, seemed to converge on that corn ethanol has a moderately smaller carbon footprint than gasoline, thus contributes to climate goals (Farrell et al. 2006; Hill et al. 2006; Wang et al. 2007; Kim and Dale 2008; Liska et al. 2009).

However, a core factor was neglected in all these LCA studies, that is, land use change (Fargione et al. 2008; Searchinger et al. 2008). The reason land use change did not come into play in these LCA studies is that they were basically a portrayal of *existing* corn ethanol with corn grown on *long-standing cornfield*. But with increasing ethanol demand driven by federal policies like the renewable fuel standard (RFS2) aimed partly at mitigating climate change (Schnepf and Yacobucci 2010), what mattered was not the carbon footprint of *existing* corn ethanol but of *additional* corn ethanol.

The key issue then became the supply of *additional* corn. Yield increase through intensification could produce more corn in the long run, but was hardly enough, and too

uncertain, to meet annual ethanol expansion. The pressure was on land resources (Malcolm et al. 2009). Higher corn prices between 2005 and 2008 were driving farmers to bring new cornfield into production by converting natural habitats or to reallocate existing cropland to growing more corn (Wallander et al. 2011; Johnston 2013; Wright and Wimberly 2013; Yang and Suh 2015a). Either way, however, has dire carbon consequences that run counter to the initial climate goal of the federal policies.

Direct conversion of forest or grassland to grow corn for ethanol production (i.e., dLUC) would release a substantial amount of carbon stored in soil and plant biomass, creating a “carbon debt” that may take dozens of years to be repaid by carbon savings from substituting corn ethanol for gasoline (Fargione et al. 2008). Similarly, reallocation of existing cropland to growing more corn could generate similar net effects through market-mediated mechanisms (i.e., iLUC) (Searchinger et al. 2008). For example, if the extra corn came at the expense of reduced soybean production, this could drive up global soybean prices and led farmers across the world to produce more soybeans by converting forest and grassland, resulting in loss of large amounts of carbon as well.

In hindsight, that the majority of LCA studies failed to take account of land use change has a lot to do with the methodology they took, namely, attributional LCA (ALCA). In these studies, corn ethanol’s carbon footprint was quantified in the simple accounting manner. They first estimated carbon emissions at different life-cycle stages based on *existing*, *average* corn farming practices and ethanol conversion technologies, and then summed them up and compared the total against the carbon footprint of gasoline. If they found that corn ethanol has a lower carbon footprint, they would conclude that corn ethanol offers carbon benefits in displacing gasoline. Underneath the conclusion was the implicit assumption that

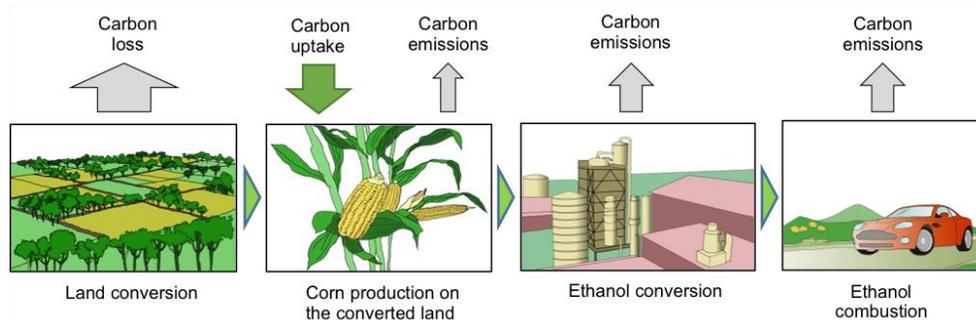
the finding based on *existing, average* technologies would hold true for any amounts of *additional* corn ethanol.

As argued above, however, the assumption is invalid. Because of land constraints, carbon emissions associated with *additional* corn ethanol would be much different from that associated with *existing* corn ethanol based on corn from *long-standing cornfield* (Fargione et al. 2008; Searchinger et al. 2008). And it is the *additional* corn ethanol and associated carbon emissions that ultimately matter from both a policy perspective and in terms of reducing greenhouse gas (GHG) emissions. In a word, consequential LCA (CLCA) looking into changes and effects is more relevant and better suited for addressing policy questions with potentially large economic and environmental consequences (Plevin et al. 2014). But it should be noted that which specific methods to use for consequential modelling needs further research (Suh and Yang 2014).

## **2.2 Direct land use change: the importance of marginal yield**

The core to consequential modelling is the consideration of marginal changes, or processes actually to be affected by decisions at hand (Weidema 2003). In the case of dLUC, marginal changes include land conversion, *additional* corn production on the converted land, and *additional* ethanol produced and used. Particularly, the *additional* corn grown on the converted land sequesters *additional* carbon from the atmosphere. Without the *additional* carbon uptake, corn ethanol's carbon benefits would not be possible as rightly pointed out by Searchinger (2010). In short, it is everything that takes place on the converted land, together with *additional* ethanol production and use, that should serve as the basis for calculating corn ethanol's total life-cycle carbon emissions in the case of dLUC (Fig. 1).

Although Fargione *et al.* rightly considered land conversion and associated carbon loss, they relied on prior LCA studies (Farrell et al. 2006; Hill et al. 2006; Wang et al. 2007), which were based on corn from *long-standing cornfield*, to estimate everything else. In so doing, they failed to recognize that newly converted land (i.e., *marginal* land) is generally not as fertile as cornfield persisting in cultivation (Keeney 2010) and that corn ethanol originating from low-fertility land would provide smaller carbon benefits than corn ethanol originating from *long-standing cornfield*. Accounting for the actual yield of the converted land (i.e., *marginal yield*), as demonstrated by Yang and Suh (2015b), could substantially increase the time it takes for the use of corn ethanol to repay the carbon debt created by the initial land conversion.



**Fig. 5.1** Carbon consequences in the case of direct land use change: converting natural habitats to grow corn for ethanol production as a consequence of ethanol. Marginal emissions in this case include carbon loss from land conversion, carbon uptake and emissions from corn production on the converted land, and carbon emissions from ethanol conversion and combustion

### 2.3 Indirect land use change: the importance of new cropland brought into production

Exiting iLUC studies calculate corn ethanol’s total carbon emissions in the same way as do previous dLUC studies (Fargione et al. 2008; Piñeiro et al. 2009; Gelfand et al. 2011) by adding carbon loss from land conversion to the carbon footprint of corn ethanol. When

exposed with the same consequential reasoning, however, the iLUC literature commits the same error as committed in previous dLUC studies. But for iLUC effect it is beyond the actual yield or fertility of the converted land; what and how new crops are produced following land conversion matters.

To drive home, let us consider a simple, hypothetical example of iLUC. Suppose, in response to increasing ethanol demand, part of U.S. corn was diverted to ethanol production at the expense of reduced exports to China. Total U.S. corn production and areas thus remained unchanged. This drove up Chinese corn prices and subsequently led Chinese people in rural areas to eat more rice, which drove up rice prices there and led Chinese subsistence farmers to convert reforested land to rice cultivation. What are the carbon consequences of corn ethanol expansion in this example? Indirect LUC studies (see, e.g., (Searchinger et al. 2008; Hertel et al. 2010)) would calculate it in such a way that only adds land conversion related carbon loss to U.S. corn ethanol's carbon footprint:

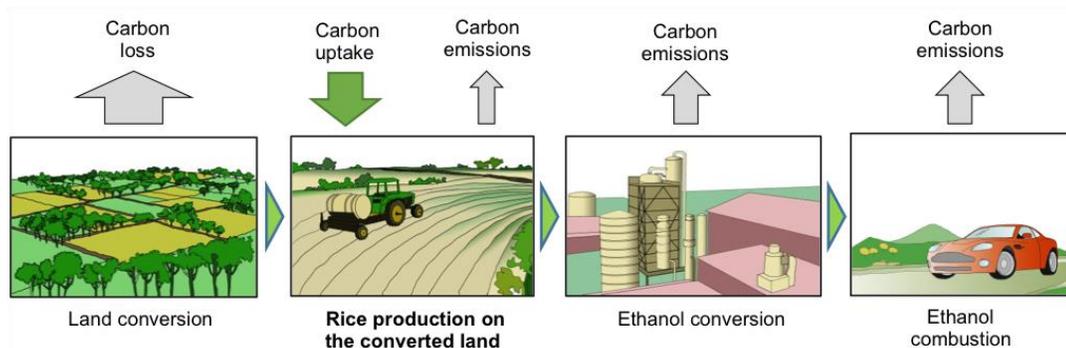
carbon consequences of ethanol expansion  
= carbon loss from converting reforested land to rice + carbon uptake from corn production  
+ carbon emissions from corn production, ethanol conversion, and ethanol combustion

However, because U.S. corn production did not change or was not affected in this example, it is irrelevant to corn ethanol's carbon consequences, as is the corn from *long-standing cornfield* in the case of dLUC. There was no *additional* carbon uptake from corn growth, nor were there *additional* carbon emissions from the use of agricultural inputs (e.g., nitrogen and fuels) in corn production. What matters, instead, is the *additional* rice cultivation in China – which took place to compensate for the U.S. corn diverted to ethanol

production – and associated carbon uptake and emissions (Fig. 2). Therefore, the carbon consequences of corn ethanol expansion in this simplified example of iLUC should be:

carbon consequences of ethanol expansion

= carbon loss from converting reforested land to rice + carbon uptake from *rice* production + carbon emissions from *rice* production, ethanol conversion, and ethanol combustion carbon emissions



**Fig. 5.2** Carbon consequences in a hypothetical indirect land use change example: converting reforested land in China to grow rice to compensate for reduced corn imports as a consequence of ethanol expansion in the U.S. (i.e., indirect land use change). Marginal emissions in this example include carbon loss from land conversion, carbon uptake and emissions from *rice production on the converted land*, and carbon emissions from ethanol conversion and combustion.

Of course, this is an extremely simplified example. Real-world consequences of U.S. corn ethanol expansion could be much more complicated, involving conversion of assorted natural habitats and different croplands brought into production in different countries. In any case, carbon uptake and emissions associated with whatever cropland being brought into production worldwide – including, likely, additional corn – should count towards the carbon consequences of ethanol expansion. Simply adding carbon loss from indirect land conversion across the world to the carbon footprint of U.S. corn ethanol is not meaningful from both theoretical and empirical perspectives. In addition to estimation of carbon loss from indirect land conversion (Plevin et al. 2010), future studies need also direct efforts to

account for what and how crops would be grown following land conversion and associated carbon uptake and emissions.

### ***3. Limitations and future work***

#### **3.1 Toward a more realistic displacement ratio**

In the chapter on carbon payback time (CPT), we assumed a perfect 1:1 displacement ratio between corn ethanol and gasoline on an energy basis, an assumption also used in previous carbon payback time studies (Farrell et al. 2006; Hill et al. 2006; Wang et al. 2007). That is to say, 1 additional MJ of corn ethanol is assumed to take the place of 1 MJ of gasoline. For example, suppose gasoline production is 500 MJ this year and is predicted to reach 600 MJ next year to keep up with rising demand under business-as-usual (i.e., without corn ethanol), and then comes 100 MJ of corn ethanol in the second year. If gasoline production remains 500 MJ in the second year, with the other 100 MJ of demand met by corn ethanol, this is considered a perfect 1:1 displacement ratio.

Due to the complexity of economic systems and human behaviour, however, it is more likely less than one unit of gasoline will be displaced by corn ethanol (York 2012). The introduction of corn ethanol into the market will put downward pressure on gasoline prices, leading to a higher demand for the fuel. To continue with our example, because of the higher demand, suppose 550 MJ of gasoline and 100 MJ of corn ethanol are produced and consumed in the second year, all else being equal. Thus the net result is that 50 MJ of gasoline is displaced by 100 MJ of corn ethanol (i.e., a 1:0.5 displacement ratio).

Displacement ratio is an important factor in the calculation of carbon payback time (Table 5.1). A 10% decrease from the perfect displacement ratio would increase the CPT by

63% for unproductive land (i.e., 50% marginal to average (MoA) yield) to 27% for highly productive land (i.e., 100% MoA yield). If only 0.6 MJ of gasoline is displaced, most of the marginal land would fail to provide any carbon benefits within the 100-year time horizon studied. If only 0.5 MJ of gasoline is displaced, even the most productive land (i.e., 100% MoA yield) would fail to yield any carbon benefits within the time horizon studied. These results suggest that whether corn ethanol provides carbon benefits depends importantly on the extent to which gasoline can be displaced by additional corn ethanol production. In future research, effort may be directed to estimate a more realistic displacement ratio that takes into account such market mechanisms as supply-demand price changes than the perfect ratio assumed in this and previous CPT studies. Models such as the partial equilibrium analyses (PEA) can be used to derive such market-mediated displacement ratios (Zinc et al. 2015).

**Table 5.1** Carbon payback time based on different displacement ratios under the technological advances without emission timing scenario

	50%	60%	70%	80%	100%
<b>1 : 1</b>	<b>56</b>	<b>35</b>	<b>26</b>	<b>21</b>	<b>15</b>
1 : 0.9	91	49	34	27	19
1 : 0.8	>100	82	50	36	24
1 : 0.7	>100	>100	94	58	34
1 : 0.6	>100	>100	>100	>100	60
1 : 0.5	>100	>100	>100	>100	>100

### 3.2 Data gaps

Concern has been raised over the eco-toxicity impact of emerging pesticides and the lack of characterization models to evaluate them. This is a general question of data gap. In fact,

in addition to emerging pesticides, there are also pesticides whose usage data are withheld by the USDA (to avoid disclosing information for individual farms). However, the ecotoxicity impact of these ‘undocumented’ pesticides is likely small as a large majority of the pesticides applied to the crops studied are covered by both usage and characterization data. Specifically, such data are available for 50 to 90 different types of pesticides; they generally account for 90% to 95% of the total amount of all pesticides applied; and they include the key pesticides that contribute the largest toxicity impacts identified by recent research (Nordborg et al 2014; Xue et al. 2015; Yang et al. 2012). It is worth noting that in terms of the number of pesticides covered, our analyses in chapters 2 and 4 are by far the most comprehensive in comparison to similar studies, which evaluated at most a dozen of pesticides (Nordborg et al 2014; Xue et al. 2015).

Nevertheless, our analyses may benefit from evaluating the possible ecotoxicity impact of the “uncovered” pesticides. For emerging pesticides, their characterization factors may be derived from models such as the USEtox based on their physicochemical properties and ecotoxicity effect data if available. For pesticides without usage data, their total usage is in fact aggregated in the total amount of pesticides applied (USDA 2015c; Yang et al. forthcoming) and can be derived by subtracting the pesticides with usage data. Next, sensitivity analysis can be carried out to compute the possible range of their total ecotoxicity impact by assuming different amounts for individual pesticides subject to the total usage derived.

### **3.3 Pesticide emissions**

Following the approach developed in previous studies (Berthoud et al. 2011), we assumed a generic factor for the fraction of pesticides in aquatic systems through leaching and runoff. However, this factor is likely to vary by pesticide – due to differences in their intrinsic physio-chemical properties – and by location – due to differences in local topographic, climatic, and soil conditions. To better estimate pesticide emissions after application, future studies may conduct field experiments – at least for the key pesticides identified – or rely on more sophisticated models than used in this dissertation, such as the PestLCI, that take into consideration pesticides’ properties, environmental factors, and application methods (Birkved and Hauschild 2006; Nordborg et al. 2014).

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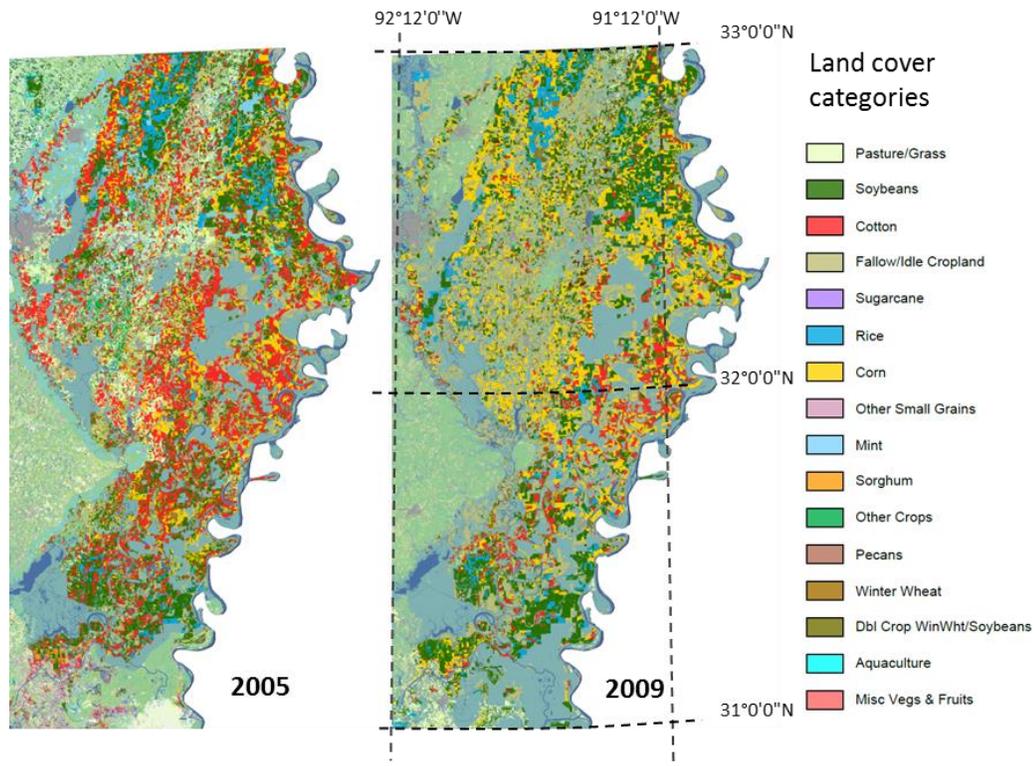
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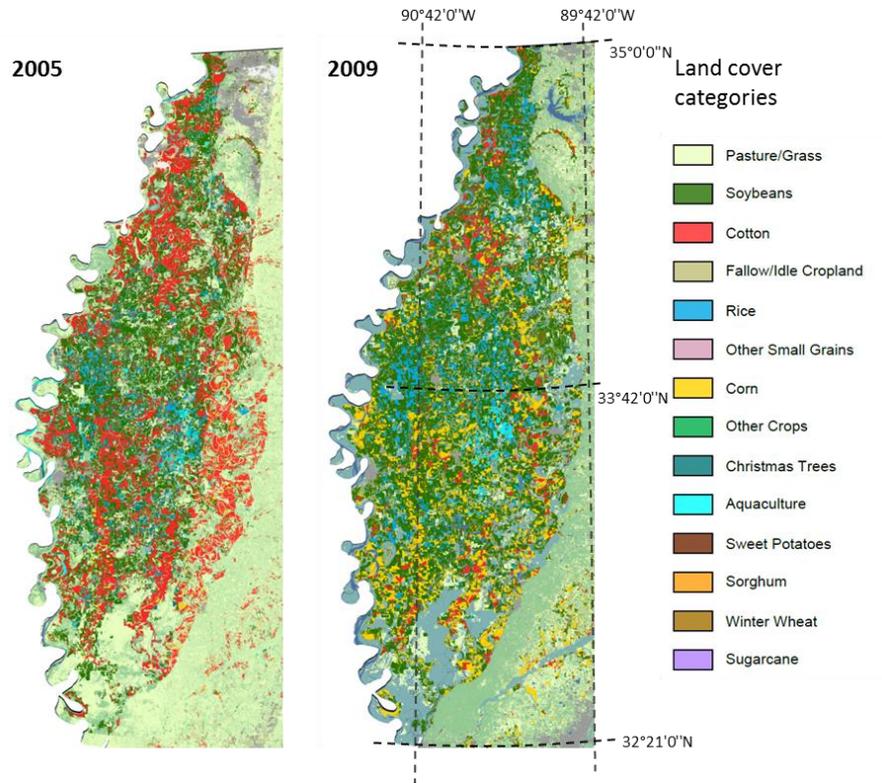
# Appendix A

## 1. Spatial evidence of land shift from Cropland Data Layer (CDL)

Based on satellite imagery, National Agricultural Statistics Service (NASS) Cropland Data Layer (CDL) maps crop land cover at a 56-m spatial resolution with high crop-type specificity [1-3]. For most states covered in CDL, however, crop data are only available for the year 2008 and thereafter. This data gap limits our observation of land shift between crops that occurred primarily before 2008. Nevertheless, crop data for three states, Louisiana (LA), Mississippi (MS), and Arkansas (AS), are available for earlier years. Fig. S1 and S2, derived from the images created by the CDL, demonstrates a massive land shift from cotton to corn in Northeast LA and Northwest MS.



**Fig S1.** Crop growth in Northeast Louisiana in 2005 and 2009. Red denotes cotton and yellow corn. (Source: USDA NASS [1])



**Fig S2.** Crop growth in Northwest Mississippi in 2005 and 2009. Red denotes cotton and yellow corn. (Source: USDA NASS [1])

## 2. Change of corn and cotton areas in cotton-growing states

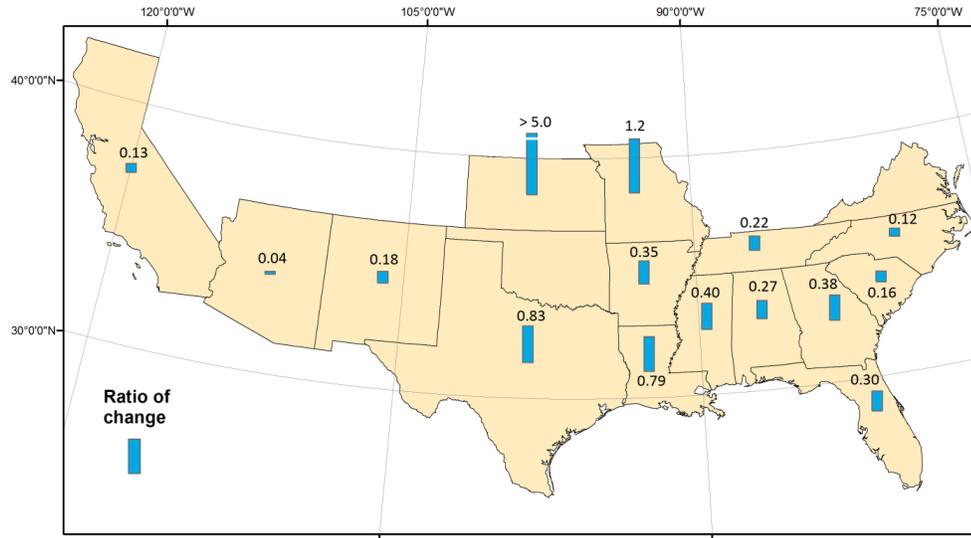


Fig S3. Change of cotton and corn area harvested from 2006 and 2009, expressed in ratios of corn area increase (ha) to cotton area decrease (ha). States with columns imposed upon indicate both increase in corn area and decrease in cotton area over the period. Column ratios indicate the extent to which corn expansion might have come at the expense of cotton. A ratio close to 1 suggests that a large majority of corn expansion might have come from cotton, and larger than 1 suggests that corn *might* have expanded into other types of cropland. (Source: USDA [14])

### 3. Regional variability of cotton and corn life-cycle environmental impacts

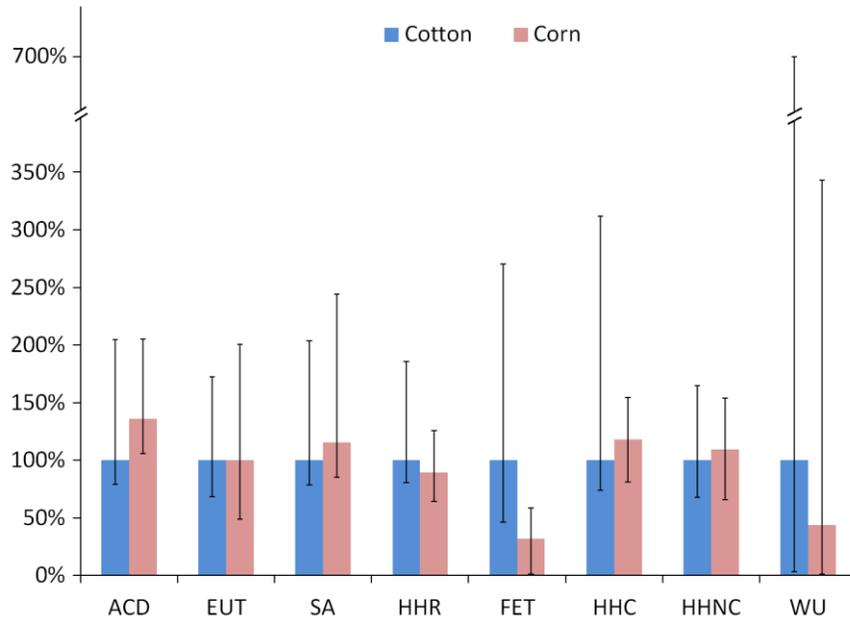


Fig S4. Spatial variability for different environmental impacts, per ha crop produced. Bar heights denote weighted average results for each impact category, with error bars reflecting the range of variability. The upper bounds of error bars represent the states with the highest impacts, lower bounds states with the lowest impacts.

All results for a given impact category are normalized by the average impacts of cotton.

#### 4. Emission factors

**Table S1** Estimation of pesticide emissions into different compartment

Compartment	Fraction	Note	Ref
Air	95%	p>10 (mPa)	[4]
	50%	1<p<10	
	15%	0.1<p<1	
	5%	0.01<p<0.1	
Water	1%	p<0.01	[5]
	5%	Generic runoff and leaching rate	
Soil	1-air-water	Capped at 85%	[4]

**Table S2** Heavy metal concentration in phosphate fertilizer [6]

	Heavy metal concentration, mg kg <sup>-1</sup>						
	As	Cd	Cr	Pb	Hg	Ni	V
Phosphate rock	12	11	109	12	0.05	37	82
P <sub>2</sub> O <sub>5</sub> fertilizers	34	32	310	35	0.1	105	235

**Table S3** Emission factors for fuel combustion (mg Mj<sup>-1</sup>)

	Diesel	Gasoline	LPG	NG
	Farming Tractor	Farming Tractor	Commercial Boiler	Stationary Reciprocating Engine
CO	344.2	516.4	10.2	324
NO <sub>x</sub>	649.3	454.0	80.6	1137
PM10	58.8	23.7	2.3	5.2
PM2.5	52.8	21.7	2.3	5.2
SO <sub>x</sub>	7.6	1.1	0	0.3
CH <sub>4</sub>	0.6	4.9	1.0	350
N <sub>2</sub> O	0.9	1.1	4.6	1.4
CO <sub>2</sub>	73371	71697	64485	54719

Data source: [7]

**Table S4** Nutrient runoff and leaching rates by state

	N		P	
	Cotton	Corn	Cotton	Corn
AL	42.5%		35.1%	
AR	42.5%		35.1%	
CA	30.7%		2.3%	
CO		12.7%		
<b>GA</b>	<b>47.4%</b>	<b>47.5%</b>	<b>14.5%</b>	<b>13.2%</b>
IA		21.2%		15.2%
IL		21.2%		15.2%
IN		21.2%		15.2%
KS		18.2%		12.3%
KY		38.3%		16.8%
LA	42.5%		35.1%	
MI		21.2%		15.2%
MN		21.2%		15.2%
MO		25.2%		17.8%
MS	42.5%		35.1%	
<b>NC</b>	<b>47.4%</b>	<b>47.5%</b>	<b>14.5%</b>	<b>13.2%</b>
ND		12.7%		9.1%
NE		12.7%		9.1%
NY		32.8%		21.7%
OH		27.0%		18.4%
PA		32.8%		21.7%
SD		12.7%		9.1%
TN	42.5%		35.1%	
<b>TX</b>	<b>33.6%</b>	<b>19.4%</b>	<b>22.0%</b>	<b>12.5%</b>
WI		21.2%		15.2%

a. Nutrient loss rates for each state are derived from regional (multistate) nutrient loss rates according to their relative locations [8]

**Table S5** Emission factors for NH<sub>3</sub> and NO<sub>x</sub>

Emission factor <sup>a</sup>	NH <sub>3</sub>	Ref	NO <sub>x</sub>	Ref
		3.5%	[9, 10]	2.5%

a. NO<sub>x</sub> emissions from soil consist primarily of NO [12, 13]

**Table S6** Speciated VOC emissions from natural gas and diesel combustion

Substances <sup>a</sup>	Natural Gas <sup>b</sup> (kg/Mj)			Diesel
	2-stroke lean-burn	4-stroke lean-burn	4-stroke rich-burn	kg/kg
1,1,2,2-Tetrachloroethane	2.9E-08	1.7E-08	1.1E-08	0
1,1,2-Trichloroethane	2.3E-08	1.4E-08	6.6E-09	0
1,1-Dichloroethane	1.7E-08	1.0E-08	4.9E-09	0
1,2,3-Trimethylbenzene	1.5E-08	9.9E-09	0	0
1,2,4-Trimethylbenzene	4.8E-08	6.2E-09	0	0
1,2-Dichloroethane	1.8E-08	1.0E-08	4.9E-09	0
1,2-Dichloropropane	1.9E-08	1.2E-08	5.6E-09	0
1,3,5-Trimethylbenzene	7.7E-09	1.5E-08	0	0
1,3-Butadiene	3.5E-07	1.2E-07	2.9E-07	7.2E-07
1,3-Dichloropropene	1.9E-08	1.1E-08	5.5E-09	0
2,2,4-Trimethylpentane	3.6E-07	1.1E-07	0	0
2-Methylnaphthalene	9.2E-09	1.4E-08	0	0
Acenaphthene	5.7E-10	5.4E-10	0	2.6E-08
Acenaphthylene	1.4E-09	2.4E-09	0	9.3E-08
Acetaldehyde	3.3E-06	3.6E-06	1.2E-06	1.4E-05
Acrolein	3.3E-06	2.2E-06	1.1E-06	1.7E-06
Anthracene	3.1E-10	0	0	3.4E-08
Benz(a)anthracene	1.4E-10	0	0	3.1E-08
Benzene	8.3E-07	1.9E-07	6.8E-07	1.7E-05
Benzo(a)pyrene	2.4E-12	0	0	0
Benzo(b)fluoranthene	3.7E-12	7.1E-11	0	1.8E-09
Benzo(e)pyrene	1.0E-11	1.8E-10	0	3.5E-09
Benzo(g,h,i)perylene	1.1E-11	1.8E-10	0	9.0E-09
Benzo(k)fluoranthene	1.8E-12	0	0	2.9E-09
Biphenyl	1.7E-09	9.1E-08	0	0
Butane	2.0E-06	2.3E-07	0	0
Butyr/Isobutyraldehyde	1.9E-07	4.3E-08	2.1E-08	0
Carbon Tetrachloride	2.6E-08	1.6E-08	7.6E-09	0
Chlorobenzene	1.9E-08	1.3E-08	5.6E-09	0
Chloroethane	0	8.0E-10	0	0
Chloroform	2.0E-08	1.2E-08	5.9E-09	0
Chrysene	2.9E-10	3.0E-10	0	6.5E-09

**Table S6** Speciated VOC emissions from natural gas and diesel combustion (continued)

Cyclohexane	1.3E-07	9.8E-08	0	0
Cyclopentane	4.1E-08	0	0	0
Dibenz(a,h)anthracene	0	0	0	1.1E-08
Ethane	3.1E-05	4.5E-05	3.0E-05	0
Ethylbenzene	4.6E-08	1.7E-08	1.1E-08	0
Ethylene Dibromide	3.2E-08	1.9E-08	9.2E-09	0
Fluoranthene	1.6E-10	4.8E-10	0	1.4E-07
Fluorene	7.3E-10	2.4E-09	0	5.4E-07
Formaldehyde	2.4E-05	2.3E-05	8.8E-06	2.2E-05
Indeno(1,2,3-c,d)pyrene	4.3E-12	0	0	6.9E-09
Isobutane	1.6E-06	0	0	0
Methanol	1.1E-06	1.1E-06	1.3E-06	0
Methylcyclohexane	1.5E-07	5.3E-07	0	0
Methylene Chloride	6.3E-08	8.6E-09	1.8E-08	0
n-Hexane	1.9E-07	4.8E-07	0	0
n-Nonane	1.3E-08	4.7E-08	0	0
n-Octane	3.2E-08	1.5E-07	0	0
n-Pentane	6.6E-07	1.1E-06	0	0
Naphthalene	4.1E-08	3.2E-08	4.2E-08	1.6E-06
Perylene	2.1E-12	0	0	0
Phenanthrene	1.5E-09	4.5E-09	0	5.4E-07
Phenol	1.8E-08	1.0E-08	0	0
Propane	1.2E-05	1.8E-05	0	0
Propylene	0	0	0	4.8E-05
Pyrene	2.5E-10	5.9E-10	0	8.8E-08
Styrene	2.4E-08	1.0E-08	5.1E-09	0
Tetrachloroethane	0	1.1E-09	0	0
Toluene	4.1E-07	1.8E-07	2.4E-07	7.5E-06
Vinyl Chloride	1.1E-08	6.4E-09	3.1E-09	0
Xylene	1.2E-07	7.9E-08	8.4E-08	5.2E-06

a. Emission factors for gasoline seem not available (EPA, 1995)

b. For natural gas, average emission factors of different modes are used for calculation in this study.

## 5. Sensitivity analysis

**Table S7** Emission factors/assumptions to which different impact categories are most sensitive for the corn system<sup>a</sup>

Model parameters	Change in parameter values	Change in impact				
		ACD	EUT	SF	FET	HHNC
Pesticide runoff & leaching	+10%				+7%	
NH <sub>3</sub> from N-fertilizer	+10%	+4%				
NO <sub>x</sub> from N-fertilizer	+10%	+2%		+5%		
N runoff and leaching	+10%		+5%			
P runoff and leaching	+10%		+4%			
NO <sub>x</sub> from fuels	+10%	+1%		+2%		
Cadmium concentration in P fertilizers	+10%					+5%

a. HHR and HHC are not included in the table considering that indirect emissions embedded in input materials were found to be the major contributor in the two impact categories. WU is excluded because it is dominated by irrigation water withdrawal and no assumptions were used for the impact category.

**Table S8** Emission factors/assumptions to which different impact categories are most sensitive for the cotton system

Model parameters	Change in parameter values	Change in impact					
		ACD	EUT	SF	HHR	FET	HHNC
Pesticide runoff & leaching	+10%					+9%	
NH <sub>3</sub> from N-fertilizer	+10%	+3%					
NO <sub>x</sub> from N-fertilizer	+10%	+2%		+3%			
N runoff and leaching	+10%		+5%				
P runoff and leaching	+10%		+4%				
NO <sub>x</sub> from fuels	+10%	+2%		+4%	+1%		
PM10 from fuels	+10%				+2%		
PM2.5 from fuels	+10%				+2%		
Cadmium concentration in P fertilizers	+10%						+4%

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## Appendix B

### 1. Data on corn production and ethanol conversion

**Table S1.** Data on key inputs for 9-state average corn production in the US

		2001 <sup>a</sup>	2005 <sup>b</sup>	2010 <sup>c</sup>	2020 <sup>d</sup>
<b>Yield</b>	Kg/ha	8744	10024	10281	10922
<b>Fertilizer</b>					
Nitrogen	Kg/ha	149.7	149.5	162.4	162.4
Potash	Kg/ha	63.7	60.9	57.8	57.8
Phosphate	Kg/ha	98.9	68.7	64.5	64.5
Lime <sup>e</sup>	Kg/ha	448.3	621.4	231.9	231.9
<b>Pesticide</b>					
Herbicide	Kg/ha	2.8	2.3	2.5	2.5
Insecticide	Kg/ha	0.2	0.1	0.02	0.02
<b>Energy</b>					
Gasoline	Mj/ha	1277	722	722	722
Diesel	Mj/ha	2719	2308	2308	2308
NG gas	Mj/ha	670	570	570	570
LPG	Mj/ha	765	717	717	717
Electricity	Mj/ha	820	498	498	498

a. Default data in the EBAMM model, originally from Shapouri 2004 [1].

b. Data from Shapouri et al. 2010 [2], 9-state weighted averages.

c. Fertilizer, yield, and pesticide data for 2010 from USDA [3], 9-state weighted averages; see Table S2. Energy inputs are assumed to be the same as in 2005.

d. Yield rate projected using the GREET model [4], which is similar to the USDA corn long-term projection [5]. All other values are assumed to be the same as in 2010.

e. The 2010 lime use is an average of 1991, 1995, 2001, and 2005 lime uses [1, 2, 6].

**Table S2.** Data on yield and key chemicals for corn growth in 9 major corn-growing states in 2010.

		IL	IN	IA	MI	MN	NE	OH	SD	WI	Ave <sup>a</sup>
<b>Yield</b>	Mg/ha	10.2	9.9	10.9	9.4	10.6	10.5	10.3	8.7	9.8	10.3
<b>Fertilizer</b>											
Nitrogen	kg/ha	186	204	155	154	128	161	165	154	120	162.4
Phosphate	kg/ha	89	72	53	38	43	32	68	52	44	57.8
Potash	kg/ha	98	120	63	100	50	6	88	12	58	64.5
<b>Pesticide</b>											
Herbicide	kg/ha	2.65	2.95	2.25	2.41	1.78	2.59	3.14	2.25	3.14	2.5
Insecticide	kg/ha	0.04	0.02	0.01	0	0.02	0.03	0.03	0.001	0.02	0.02

a. Weighted averages by state corn areas harvested.

**Table S3.** Data on ethanol conversion technologies, from GREET (2012)<sup>a</sup>.

		2005			2010			2020		
		Dry	Wet	Ave	Dry	Wet	Ave	Dry	Wet	Ave
Total energy	Mj/L	8.7	13.2	<b>9.5</b>	7.5	13.2	<b>8.1</b>	7.5	13.2	<b>8.0</b>
Yield (anhydrous)	L/Kg	0.40	0.39	<b>0.40</b>	0.42	0.39	<b>0.41</b>	0.44	0.41	<b>0.43</b>
Market share	%	81%	19%		88.6%	11.4%		91.1%	8.9%	
Fuel share										
NG	%	72.6%	60%		83.3%	72.5%		83.3%	72.5%	
Coal	%	18.1%	40%		7.3%	27.5%		7.3%	27.5%	
Electricity	%	9.3%			9.4%			9.4%		

a. Values in shade are adjusted based on the results of a survey by Wu 2008 [7], as GREET (2012) [4] seems to overestimate the yield of dry-mill ethanol plants and underestimate wet mill yield for 2005.

## 2. Sensitivity analysis based on the system expansion method

Following Fargione et al. (2008) [8], we allocate 83% of the carbon debt to ethanol and 17% to distiller grains with soluble (DGS) based on their 2007 economic values. In a more recent study, Yang et al. 2012 [9] arrived at similar allocation ratios for ethanol and DGS after analyzing their market values over the past few years. Below, we apply the system expansion method to test the robustness of the payback time estimates based on economic allocation.

First, Table S4 and S5 present the carbon footprint of corn ethanol using the system expansion method, exclusive of carbon uptake during corn growth and carbon emissions during vehicle operation because they roughly cancel each other out (Table S4 and S5).

**Table S4.** Carbon emissions of corn ethanol from highly productive CRP land.

		100% marginal-to-average yield			
		2001	2005	2010	2020
Agriculture	g CO <sub>2</sub> e/Mj	36.8	31.0	28.3	25.3
Biorefinery	g CO <sub>2</sub> e/Mj	63.8	38.9	31.8	31.1
Coproduct credits	g CO <sub>2</sub> e/Mj	-24.8	-15.1	-13.9	-13.1
Ethanol distribution	g CO <sub>2</sub> e/Mj	1.4	1.4	1.4	1.4
Net GHG emissions	g CO <sub>2</sub> e/Mj	77.2	56.2	47.6	44.7
Gasoline GHG emissions	g CO <sub>2</sub> e/Mj	94.0			
GHG reduction	%	17.8%	40.2%	49.4%	52.5%

**Table S5.** Carbon emissions of corn ethanol from CRP land with different yield ratios<sup>a</sup> (in units of g CO<sub>2</sub>e Mj<sup>-1</sup>).

	<b>50% yield</b>				<b>60% yield</b>			
	2001	2005	2010	2020	2001	2005	2010	2020
Agriculture	73.1	62.0	56.6	50.55	60.9	51.7	47.2	42.13
Net GHG emissions	113	87	76	70	101	77	66	62
GHG reduction (%)	-20.7%	7.2%	19.2%	25.6%	-7.8%	18.2%	29.3%	34.6%
	<b>70% yield</b>				<b>80% yield</b>			
	2001	2005	2010	2020	2001	2005	2010	2020
Agriculture	52.2	44.3	40.5	36.1	45.7	38.8	35.4	31.60
Net GHG emissions	93	69	60	56	86	64	55	51
GHG reduction (%)	1.5%	26.1%	36.4%	41.0%	8.4%	32.0%	41.8%	45.8%

a. Emissions from biorefinery, coproduct credits and ethanol distribution are the same as those for CRP land with 100% yield in Table S4.

Coproduct credits are estimated as follows. In this study, we assume that the CRP land converted is dedicated to ethanol production, thus the coproduct DGS will displace crops from existing land. According to the GREET model [4], the amount of DGS generated per Mj ethanol displaces around 24.8 g corn, 9.8 g soybean meal, and 0.7 urea for the dry mill conversion technology and 40.3 g corn, 0.6 g soybean meal, and 5.5 g soy oil for the wet mill. Combined with their market shares and carbon emissions embodied in corn, soybean meal, soy oil, and urea, the amount of coproduct credits is estimated to range from 24.8 CO<sub>2</sub>e/Mj in 2001 to 13.1 g CO<sub>2</sub>e/Mj (Table S4).

That coproduct credits decrease from 2001 to 2020 reflects the displaced product systems, primarily corn and soybeans, becoming more efficient over time and thus generating smaller amounts of carbon emissions. Data on coproduct credits for 2001 are directly from the default model EBAMM used in this study, and for 2005, 2010 and 2020 are from the GREET model [4]. The credits (per Mj ethanol produced), however, do not vary between different CRP-corn ethanol systems in a given year, because DGS, be it from high- or low-fertility CRP land systems, is assumed to displace the same products.

Second, we calculate the proportions of coproduct credits in the total carbon emissions of different CRP-corn ethanol systems (Table S6). Note that for a given year the total carbon emissions of different CRP-corn ethanol systems are different, so are the proportions of coproduct credits.

**Table S6.** The proportion of coproduct credits in the total carbon emissions of different CRP-corn ethanol systems in different years.

marginal-to-average yield	Year					
	2001	2005	2010	2015	2020	<b>Ave</b>
50%	24%	22%	23%	23%	23%	<b>23%</b>
60%	18%	15%	16%	15%	16%	<b>16%</b>
70%	20%	16%	17%	17%	18%	<b>18%</b>
80%	21%	18%	19%	19%	19%	<b>19%</b>
100%	22%	19%	20%	20%	20%	<b>20%</b>

Last, we allocate part of the carbon debt to the coproduct DGS based on the average proportions in Table S6 for different CRP-corn ethanol systems, in the same way economic allocation is done as described in the manuscript. The results of carbon payback time estimated using both economic allocation and system expansion are quite similar, as shown in Table S7.

Table S7. Carbon payback time (year) based on economic allocation and system expansion, taking into account technological advances and the effect of emissions timing (land conversion year: 2001).

marginal-to-average yield ratio	economic allocation	system expansion	difference
100%	17	16	-1
80%	24	23	-1
70%	30	30	0
60%	43	43	0
50%	88	91	3

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## Appendix C

### 1. Estimation of biological nitrogen fixation for soybean

Nitrogen input through biological nitrogen fixation (BNF), due to nitrogen-fixing bacteria hosted symbiotically by the legume crops, can also be a source of nitrogen pollution [1]. Here, we estimated the total amount of BNF for soybean production in the major states covered, based on a USDA study that quantified nutrient inputs from multiple sources to major U.S. crops in a single year.<sup>1</sup> Their estimates of BNF ranged from 91 to 243 kg ha<sup>-1</sup> for 7 geographic regions into which the study classified the continental United States. We first derived state-level estimates from the regional estimates based on the location of the state relative to the regions classified in the USDA study. We assumed that the rates remained unchanged over the period investigated given that there is no strong evidence that the environmental factors [2] affecting BNF have changed substantially in the past decade or. Based on the state rates, we further calculated the average rate for per ha soybean in the U.S., which is about 185 kg ha<sup>-1</sup>. The total amount of BNF calculated for the 19 to 20 states major soybean-producing states covered in this study is around 5.4 Tg yr<sup>-1</sup>, which is close to a recent estimate of 5.7 Tg calculated for the entire country [3].

### 2. Comparison between TRACI 2.0, IMPACT 2002+, and CML 2001

To test the robustness of our result on freshwater ecotoxicity impact, we applied two additional characterization models, IMPACT 2002+ and CML 2001, to evaluating the inventory data compiled. Because pesticide emissions were identified as the major contributor to freshwater ecotoxicity (see figure 3 in the manuscript), the other two models were used to characterize pesticide emissions only. The impact categories in IMPACT 2002+ and CML 2001 that correspond to freshwater ecotoxicity in TRACI 2.0 are aquatic ecotoxicity. Estimation of pesticide emissions to different compartments was kept the same for all three methods (see section 2.4. in the manuscript).

Results are presented below. Two factors lead to the differences in the magnitude of change calculated by the three characterization models (see also figure 5 in the manuscript). First, different models cover different numbers of pesticides. TRACI 2.0, whose freshwater ecotoxicity impact category is adopted from USEtox [4], covers the largest number of pesticides, with 63/84 for corn, 89/113 for cotton, 55/81 for soybean, and 48/65 for wheat. Compare this with CML 2001 (19/84 for corn, 23/113 for cotton, 16/81 for soybean, and 13/65 for wheat) and IMPACT 2002+ (63/84 for corn, 89/113 for cotton, 55/81 for soybean, and 48/65 for wheat). Second, different characterization models give different ecotoxicity potentials to different pesticides. In TRACI 2.0, for example, *cyfluthrin* shows the largest freshwater ecotoxicity potential, which is 300 times that of *atrazine*. In IMPACT 2002+, however, *lambda-cyhalothrin* shows the largest aquatic ecotoxicity potential, and that of *cyfluthrin* is only 17 times that of *atrazine*.

Tables S13-S26 present impact results based on CML 2001 and contributions of key pesticides identified.

Table S13. Aquatic ecotoxicity impact per ha corn produced and contributions of key pesticides (unit: kg 1,4-dichlorobenzene eq.)

	2001	2005	2010
atrazine	22%	22%	25%
chlorpyrifos	14%	9%	3%
metolachlor	15%	4%	1%
permethrin	7%	4%	3%
simazine	4%	6%	7%
s-metolachlor	36%	51%	60%
other pesticides	2%	4%	1%
<b>total</b>	<b>1599</b>	<b>1348</b>	<b>993</b>

Table S14. Aquatic ecotoxicity impact per ha cotton produced and contributions of key pesticides (unit: kg 1,4-dichlorobenzene eq.)

	2000	2003	2007
aldicarb	66%	78%	83%
cypermethrin	6%	9%	10%
malathion	23%	7%	3%
other pesticides	5%	6%	4%
<b>total</b>	<b>25935</b>	<b>19058</b>	<b>12729</b>

Table S15. Aquatic ecotoxicity impact per ha cotton produced and contributions of key pesticides (unit: kg 1,4-dichlorobenzene eq.)

	2002	2006	2012
chlorpyrifos	11%	71%	50%
metolachlor	10%	0%	2%
permethrin	7%	1%	0%
s-metolachlor	23%	10%	38%
trifluralin	25%	6%	3%
zeta-cypermethrin	16%	10%	2%
other pesticides	9%	1%	6%
<b>total</b>	<b>347</b>	<b>456</b>	<b>804</b>

Table S16. Aquatic ecotoxicity impact per ha cotton produced and contributions of key pesticides (unit: kg 1,4-dichlorobenzene eq.)

	2002	2006	2012
chlorpyrifos	66%	33%	68%
ethyl parathion	0%	18%	0%
methanone	0%	0%	8%
triallate	22%	1%	1%
trifluralin	5%	1%	0%
zeta-cypermethrin	0%	42%	23%
other pesticides	7%	5%	0%
<b>total</b>	<b>67</b>	<b>125</b>	<b>78</b>

Tables S17-20 present impact results based on IMPACT 2002 + and contributions of key pesticides identified.

Table S17. Aquatic ecotoxicity impact per ha corn produced and contributions of key pesticides (unit: kg triethylene glycol. eq.)

	2001	2005	2010
atrazine	98%	98%	99%
other pesticides	2%	2%	1%
<b>total</b>	<b>962290</b>	<b>808538</b>	<b>667197</b>

Table S18. Aquatic ecotoxicity impact per ha cotton produced and contributions of key pesticides (unit: kg triethylene glycol. eq.)

	2000	2003	2007
aldicarb	18%	21%	21%
chlorpyrifos	5%	2%	0%
cyanazine	4%	0%	0%
cyfluthrin	4%	3%	3%
diuron	5%	10%	13%
lambda-cyhalothrin	4%	4%	4%
malathion	3%	1%	0%
methomyl	4%	3%	0%
prometryn	17%	21%	19%
tribufos	17%	16%	24%
trifluralin	4%	5%	5%
other pesticides	16%	13%	10%
<b>total</b>	<b>69240</b>	<b>51911</b>	<b>36533</b>

Table S19. Aquatic ecotoxicity impact per ha soybean produced and contributions of key pesticides (unit: kg triethylene glycol. eq.)

	2002	2006	2012
chlorpyrifos	5%	42%	30%
glyphosate	32%	1%	2%
lambda-cyhalothrin	9%	28%	23%
metribuzin	13%	13%	11%
pendimethalin	7%	2%	1%
s-metolachlor	9%	5%	19%
trifluralin	13%	4%	2%
other pesticides	13%	4%	12%
<b>total</b>	<b>3273</b>	<b>3250</b>	<b>5658</b>

Table S20. Aquatic ecotoxicity impact per ha wheat produced and contributions of key pesticides (unit: kg triethylene glycol. eq.)

	2002	2006	2012
atrazine	47%	71%	60%
bromoxynil	19%	8%	0%
chlorpyrifos	16%	9%	23%
triallate	7%	1%	0%
other inputs	12%	11%	17%
<b>total</b>	<b>4173</b>	<b>6890</b>	<b>3307</b>

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