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Life cycle assessment of corn-based ethanol production in Argentina

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HIGHLIGHTS

- A cradle-to-gate LCIA for corn-based ethanol production in Argentina is performed.
- The system includes from raw materials production to anhydrous ethanol by dry milling.
- Results from HI, IN and EG perspectives in Eco-indicator 99 and ReCiPe are compared.
- Corn production, supplied energy and DDGS' use are the most significant processes.

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ABSTRACT

The promotion of biofuels as energy for transportation in the world is mainly driven by the perspective of oil depletion, the concerns about energy security and global warming.
In Argentina, the legislation has imposed the use of biofuels in blend with fossil fuels (5 to 10%) in the transport sector.
The aim of this paper is to assess the environmental impact of corn-based ethanol production in the province of Santa Fe in Argentina based on the life cycle assessment methodology.
The studied system includes from raw materials production to anhydrous ethanol production using dry milling technology. The system is divided into two subsystems: agricultural system and refinery system. The treatment of stillage is considered as well as the use of co-products (distiller's dried grains with solubles), but the use and/or application of the produced biofuel is not analyzed: a cradle-to-gate analysis is presented. As functional unit, 1 MJ of anhydrous ethanol at biorefinery is chosen.
Two life cycle impact assessment methods are selected to perform the study: Eco-indicator 99 and ReCiPe. SimaPro is the life cycle assessment software used. The influence of the perspectives on the model is analyzed by sensitivity analysis for both methods.
The two selected methods identify the same relevant processes. The use of fertilizers and resources, seeds production, harvesting process, corn drying, and phosphorus fertilizers and acetamide–anillide-compounds production are the most relevant processes in agricultural system. For refinery system, corn production, supplied heat and burned natural gas result in the higher contributions. The use of distiller's dried grains with solubles has an important positive environmental impact.

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1. Introduction

Fossil fuels, mainly oil and its derivatives, have remained as the essential energy source until present. Transport system and most industry activities heavily depend on these no renewable fuels. However, nowadays, as consequence mainly of oil reserves depletion and a day-to-day more environmentally conscious society, there are strong incentives to encourage research and development projects on renewable energies.

Biofuels, mainly biodiesel and bioethanol, constitute a renewable source of primary energy and its sustainable use and production is a valuable palliative to the current global energy crisis. In Argentina, a country with fertile soils climatologically favored for cultivation of a variety of cereals and oilseeds, the legislation has imposed, like in other countries, the use of biofuels in blend with fossil fuels. Particularly, the National Law 26093 establishes that from 2010 gas oil or diesel oil and gasoline have to be blended with 5% of biodiesel and bioethanol, respectively, providing important tax benefits to promote biofuels production (Ley 26.093, 2006).

Agricultural resources for energy purposes present opportunities as well as risks. Bioenergy production almost always involves native ecosystems such as grassland or forest, pasture and protected areas, as well as intensive agriculture in crop areas. The increase of the world

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bioenergy demand has already led to undesirable socio-economic effects with respect to food production, including increases in food prices, shortage of fodder, and growing competition for land (Mol, 2007). For instance, the Secretary of Fuels of Argentina estimates that it is necessary about 330 million m^3 of ethanol and 890 million m^3 of biodiesel per year to reach the established target of 5% (Medina, 2008). To achieve these biofuels demands, it would be necessary on an agricultural frontier expansion and/or competition with food for land use.

This work aims at analyzing the environmental performance of anhydrous ethanol production from corn crop in the province of Santa Fe, in the North-East region of Argentina, applying the life cycle assessment (LCA) methodology. Santa Fe is placed in one of the two regions favored with governmental decisions and policies to promote biofuels production, and it has one of the highest corn yields of the country. The identification of the more environmentally relevant processes in anhydrous ethanol production from corn crop through LCA methodology is performed. The valorization of co-products is included into the analysis. The influence of the perspectives on the model is analyzed by sensitivity analysis for the two selected Life Cycle Impact Assessment (LCIA) methods: Eco-indicator 99 (Goedkoop et al., 2000) and ReCiPe (Goedkoop et al., 2012). A comparison of results from both methods, including critical knowledge gaps, is presented.

2. Corn crop in the province of Santa Fe

Life cycle assessment of biofuels production requires a specific analysis due to the significant importance of local conditions in estimating environmental impacts. Indeed, Argentina has a big extension with several climatic regions apt for different crops and soil uses. Therefore, it is necessary to apply an approach based on detailed local, specific input data to achieve closely representative results as much as possible.

As mentioned, the province of Santa Fe is chosen to perform the study. Santa Fe has a surface of 133,007 km^2, which represents 3.54% of the Argentine territory. It is geographically located within 28° and 34° 22′ lat S and 58° and 62° 52′ long W, comprising a region rich in natural resources and having an important productive infrastructure. The cereals and oleaginous produced in Santa Fe represent around 15% of the country's production. Its harvested surface is about 21% of the harvested surface in Argentina (Bolsa de Comercio de Santa Fe, 2009), where soybean, wheat and corn crops represent 93.6% of the total harvested surface in the province (73%, 14.1% and 6.2%, respectively). The annual production of corn in the agricultural campaigns corresponding to period 2002–2010 varied between 1,500,000 t in 2008/2009 and 4,000,000 t in 2009/2010 (Ministerio de Agricultura, Ganadería y Pesca, MAGyP, 2010). Such period was adopted because it comprises dry and humid years, includes new technologies, and considers the influence of international markets (Montico, 2009).

Direct seeding is nowadays the predominant farming practice, covering in 2009 more than 75 and 82% of Argentina's and Santa Fe's agricultural surface, respectively (Aapresid, 2009). This technique is characterized by the uniform deposition of crop residues on the soil surface without tilling, leaving the soil undisturbed (West and Marland, 2002). Crop residues contain substantial amounts of plant nutrients. Direct seeding practice reduces the fertilizer application rates and fuel use compared to conventional tillage practice. In addition, retention of crop residues on agricultural soils has numerous direct and indirect benefits with strong impacts on soil quality. Among the direct ones, crop residues retained on the soil surface as mulch moderate water and energy balance, buffer against erosive forces of raindrops and wind, recycle plant nutrients, and serve as food and habitat for soil organisms. Among the indirect benefits, residues affect soil processes through microclimate changes, soil moisture and temperature regimes, water and solute transport and erosional processes (Lal, 2009).

3. Corn-based ethanol plant via dry milling

In the world, 61% of bioethanol is obtained from sugar cane while the remaining 39% is produced from different cereals such as sorghum and corn (Copello, 2007). In Argentina, the firm Alconoa produced 13,000,000 L of ethanol per month from sugar cane in early 2011 to reach a 2% mixture with fossil fuels. Table 1 lists the corn-based ethanol projects that have been developed in Argentina during 2012, and those that will start operating during 2013 and 2014 (Anschau et al., 2009).

The predominant ethanol production technology used in those projects is the dry milling. A simplified flow diagram of the process is shown in Fig. 1. First, the grain is cleaned and grinded into a fine powder. Then, in the *liquefaction and saccharification units*, the corn mash is converted into fermentable sugars by enzymatic hydrolysis breaking the glucosidic bonds of the starch macromolecule contained in the corn. The output from the initial liquefaction step is combined with “backset”, which is a recycled stream from the liquid portion of the stillage separated by centrifugation later in the process. The “backset” provides critical nutrients for the yeast later in the fermentation step. In the *fermentation unit*, a beer at 9% (vol.) ethanol is produced. In this particular process, glucose is fermented to ethanol and carbon dioxide by yeast. The *distillation unit* separates the ethanol produced during fermentation obtaining hydrated ethanol of 95 wt.% The outlet from the bottom of the distillation column contains considerable amount of water, non-fermentable material, chemicals produced during fermentation together with the main products, as well as some compounds produced by chemical reactions that occur during distillation because of the high temperatures. The dehydration to anhydrous ethanol (99.8 wt.%) is performed by means of molecular sieves with regeneration by difference of pressure. The hydrated ethanol is overheated prior to dehydration to avoid any risk of condensation in the adsorbers. Water molecules are trapped and adsorbed inside the microporous beads, whereas the larger ethanol molecules flow around them (Kwiatkowski et al., 2006; Jungbluth and Emmenegger, 2007). Finally, anhydrous ethanol is denaturalized with 1.5% gasoline (Instituto Nacional de Vitivinicultura, 2012) to avoid its use for food and beverage alcohol. The *separation unit* mainly aims at separating insoluble dry matter (wet cake) from the soluble one contained in the stillage. This stage also allows: (1) to increase the quantities of the stillage recycled in the fermentation stage, while at the same time reducing the amount of thin stillage, and (2) to reduce the energy consumption and investment costs of the drying unit. Separation is performed by means of settling tanks (clarifiers) coupled with centrifuges producing two distinct outputs: (a) wet cake, which contains 40% of the total dry matter (DM) of the stillage remain (with a concentration of about 30% DM), and (b) thin stillage (with a dry matter concentration below 10%). Wet cake is sent to the *drying unit*, while the thin stillage is directed to the *pre-concentration unit* for further treatment. The syrup left from the concentration stage is also

Table 1
Corn-based ethanol production projects in Argentina.

Firm	Capacity (m^3/yr)	Location
Bioetanol Río Cuarto S.A.	50,000	Córdoba
ProMaíz	140,000	Córdoba
Agroctanos	83,750	Córdoba
ACA (Asociac. Coop. Arg.)	125,000	Córdoba
ARCOR	10,000	Buenos Aires
Agroctanos	50,000	Corrientes
San José	10,000	San Luis
Soros	20,000	Santa Fe
Porta	50,000	Córdoba
Las Lajitas	50,000	Salta
Alimentos del Sur	80,000	Entre Ríos
Diaser	80,000	San Luis
Bahía Energías Renovables	100,000	Buenos Aires
Bioterai	121,000	Not defined
Green Pampas	450,000	Santa Fe

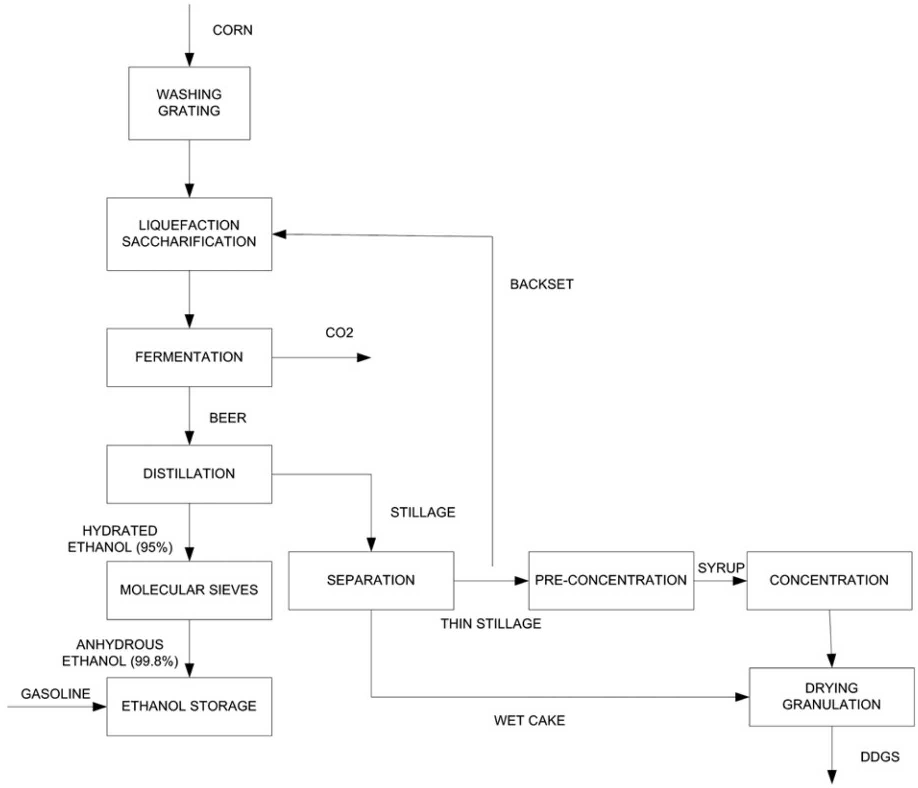

Fig. 1. Biorefinery process (Jungbluth and Emmenegger, 2007).

sent to the *drying unit*, which is a direct heating drum drier operated with natural gas. The product coming out of the drier is evacuated by pneumatic transport towards cyclones. The evaporated water vapor is washed to comply with air emission standards. The dry product falls in a granulation press, and passes through an air cooler. The obtained product is distiller's dried grains with solubles (DDGS) at 90% DM in the form of granules, and can be used as animal feed (Jungbluth and Emmenegger, 2007).

4. Life cycle assessment (LCA)

LCA has two main objectives: 1) to quantify and evaluate the environmental performance of a product, process or activity from "cradle to grave", i.e. considering the whole life cycle: extracting and processing raw materials; manufacturing, transportation and distribution; use, reuse, maintenance; recycling and final disposal; and 2) to help decision-makers to choose among alternative products or processes (Guinée et al., 1993). In addition, LCA provides a basis for assessing potential improvements in the environmental performance of a product system (Azapagic and Clift, 1999).

In this work, the Eco-indicator 99 method (Goedkoop et al., 2000) and ReCiPe method (Goedkoop et al., 2012), which are based on the ISO methodology, are used. The first is chosen because, according to Pieragostini et al. (2011), it resulted to be the most used LCIA method. However, there are more recent methods, such as ReCiPe, that provide more characterization factors and of higher quality. ReCiPe uses multi-media models for the determination of the fate factors, and provides characterization factors for specific emission compartments at different scales (Pizzol et al., 2011). The comparison among two or more methods is useful to see how vary the LCA results according to the chosen method. The main difference between them is that Eco-indicator 99 is an endpoint-oriented method while ReCiPe brings into alignment the two families of methods: the midpoint-oriented CML 2002 method (Guinée et al., 2002) and the endpoint-oriented Eco-indicator 99 method.

Eco-indicator 99 considers 11 impact categories that are aggregated in 3 endpoint categories: human health, ecosystem quality and resources (Goedkoop et al., 2000). To combine different types of damages to human health, a tool for comparative weighting of disabilities is used: the DALY (Disability Adjusted Life Years) scale, which has been developed by Murray and Lopez (1996) for the World Health Organization (WHO). Unlike human health, ecosystem quality is not an individual damage, but it is based on species diversity, as the percentage of species that are threatened or that disappear from a given area during a certain time. The unit for the damages to ecosystem quality is the Potentially Disappeared Fraction (PDF) times area times year [$m^2 \cdot yr$]. The unit of the resources damage category is the "surplus energy" in MJ per kg of extracted material; this is the expected increase of extraction energy per kg of extracted material in the future (Chapman and Roberts, 1983).

In ReCiPe, 17 impact categories are considered at the endpoint level, which are also converted and aggregated into 3 endpoint categories: human health, ecosystem quality and resources. As in Eco-indicator 99, the human health category involves 6 impact categories. Regarding to ecosystem quality, more impact categories are considered, subdividing land occupation category into urban and agricultural, and taking into account aquatic ecosystem.

At the endpoint categories, ReCiPe applies the DALY concept for human health category as Eco-indicator 99 does. Regarding to ecosystem quality, ReCiPe uses the same unit than Eco-indicator 99; and it provides a characterization factor for aquatic eutrophication (both for freshwater and marine water). The unit of this indicator is $PDF \cdot m^3 \cdot yr$, which involves an integration over volume instead of area. In the case of resources availability, the ReCiPe method is based on the geological distribution of mineral and fossil resources, and assesses how the use of these resources causes marginal changes in the efforts to extract future resources. Unlike Eco-indicator 99, it does not assess the increased energy requirement in a distant future; rather, the model is based on the marginal increase in costs due to the extraction of a resource. A function that reflects the marginal increase of the extraction cost due to the effects that result from continuing extraction is provided.

In both methods, two types of uncertainties are distinguished: (1) on the inventory data and (2) on the perspectives on the used models. The first refers to difficulties in measuring or predicting effects. This type of uncertainties is relatively easy to handle and can be expressed as a range or a standard deviation. The second type includes value choices like the choice of the time horizon in the damage model, or the question whether it should include an effect even if the scientific proof that the effect exists is incomplete.

Regarding to uncertainties about the perspectives on the model, both methods are based on the concept of cultural theory (Thompson et al., 1990), identifying three main value systems and, as consequence, three different versions of damage model (Hofstetter, 1998):

- Individualist version (IN): only proven cause–effect relations are included using the short-term perspective and accepting that a limit is not negotiable if sufficient proof is given.
- Egalitarian version (EG): it uses a precautionary principle, trying not to leave anything out and, if in doubt, it is included. This version does not accept guidance from internationally accepted scientific or political organizations. The very long time perspective is used because it does not accept that future problems can be avoided. Although it is the most comprehensive version, it has the largest data uncertainties.
- Hierarchical version (HI): it includes facts that are backed up by scientific and political bodies with sufficient recognition. The hierarchical attitude is rather common in the scientific community, and among policy makers.

The authors of the Eco-indicator 99 method (Goedkoop and Spriensma, 2001) recommend using the hierarchical version because most models work according to consensus building processes and a balanced view of long and short-term perspectives. Therefore, most models implicitly or explicitly are based on the hierarchical perspective. The other two perspectives can be used as sensitivity analysis to observe whether the conclusion drawn from LCA remains the same. In this work, reference cases of Eco-indicator 99 and ReCiPe are based on the hierarchical perspective of the model. Eco-indicator 99 and ReCiPe are available in SimaPro 7.3.3 LCA software (Goedkoop and Oele, 2008).

5. Background on LCA studies of corn-based bioethanol

There have been published several works on LCA of crop-based bioethanol in the last decade. Regarding to corn-based ethanol, which is the focus in this work, different aspects were reviewed such as the defined system boundaries, selected functional units, used software, and considered environment impacts. Kim and Dale (2005) analyzed corn-based ethanol via dry and wet milling using 1 kg of biofuel as functional unit and considering a well-to-ethanol analysis for the USA context. The mass basis allocation method was applied, and GHG emissions and net energy value (NEV) were the impact indicators evaluated. The GREET model (Wang, 2000) was used to compute the energy requirements and GHG emissions for the upstream processes, except for N fertilizer. The DAYCENT model (Del Grosso et al., 2001) was the computation tool used to simulate dynamics of soil organic carbon and related N containing species. Kim and Dale (2008) considered 1 kg of ethanol used in an E10 fuelled vehicle as functional unit considering well-to-tank analysis. The authors assessed the overall environmental and economic performance of corn-based ethanol production in a dry mill to estimate local effects due to farming sites and to determine the effects of possible scenarios for reducing N losses from soil during corn cultivation. Eutrophication and photochemical smog formation categories from TRACI method were added to GHG emissions and energy requirements. Farrell et al. (2006) studied the potential effects of increased biofuel use, evaluating six representative analyses of bioethanol. The authors explained studies with negative results for ethanol by the lack of impact allocation to co-products and the use of old data. The functional unit used was 1 MJ of ethanol. The authors analyzed three scenarios related to current US corn ethanol industry, lignite-powered ethanol plant, and cellulosic ethanol from switchgrass. Wang et al. (2007) studied new designs for corn ethanol plants and their associated energy and GHG emission effects using the GREET model for a well-to-wheels analysis, and their comparison to 2007 and 2010 gasoline production and use, and cellulosic ethanol production from switchgrass in the future. The designs for corn ethanol plants are related to fuel used (natural gas, coal, wood chips), incorporation of CHP systems and production of wet DGS instead of DDGS. Wakeley et al. (2009) analyzed the large-scale ethanol production and distribution in USA. The functional unit chosen was 1 L ethanol on E85. They studied different scenarios through the policy analysis system (POLYSYS) model, a national simulation model for the U.S. agriculture sector, which estimates impacts resulting from policy, economic, resource, or environmental changes (De La Torre and Ray, 2000). The economic input–output life cycle assessment model was used to quantify total life cycle emissions from truck and rail transportation (Sangwon, 2004). Liska et al. (2009) analyzed six common types of corn-ethanol biorefineries and two improved technologies for crop production (high-yield, progressive crop and soil management) or biorefinery operation and co-product use (closed loop). The authors considered GHG emissions and energy efficiencies for 1 MJ of corn-ethanol, taking into account well-to-tank analysis, cattle open feedlot, and anaerobic digestion as used in a closed-loop biorefinery. BESS software was used to calculate dynamic co-product energy and GHG credit. Factors that determine the magnitude of this credit include the percentage of inclusion in cattle diets, transportation distance from the ethanol plant to the feedlot, and cattle performance. In Hsu et al. (2010), three conversion technologies are assessed: advanced dry mill (corn grain), biochemical (switchgrass, corn stover, wheat straw), and thermochemical (forest residues). One km traveled by a light-duty passenger car operated on E85 in the year 2022 was used as functional unit considering a well-to-tank analysis. Avoided impacts are accounted for using product displacement (boundary expansion). For products that share inputs (e.g., corn grain and corn stover), burdens are allocated between products based on a “product-purpose” approach. SimaPro v7.1 was the LCA software used. GHG emissions and NEV were evaluated and compared with gasoline in 2005. For the first, 100-year global warming potentials for CO_2, CH_4 and N_2O were used (Solomon et al., 2007). The authors used the multivariate analysis technique of partial least-squares regression modeling (Brereton, 2007) to identify which input variables are most influential. A reference case is compared against results from Monte Carlo uncertainty analysis. Feng et al. (2010) performed the assessment of the GHG impacts of ethanol from different corn sources: (a) additional corn produced on land area that, if it were not used for ethanol production, would had been left idle; (b) additional corn produced on land area that, if it were not used for ethanol production, would had used for others crops; and (c) existing corn that was not used to ethanol production. The authors applied two methodologies: LCA and system wide approach. They evaluated the degree to which corn ethanol reduces GHG emissions depends on how corn is produced, how corn is processed into ethanol, and what emissions would be without corn ethanol. Kauffman et al. (2011) proposed a LCA on the basis of a hectare of corn, where the corn grain is used for production of ethanol and the corn stover is subjected to fast pyrolysis yielding biochar and bio-oil. A portion of the biochar was used for pyrolysis energy requirements and the remainder was applied to the agricultural soils from which the corn stover was initially harvested. Corn yields estimated for 2022 are taken from the EPA analysis, which considers land use change, fuel and feedstock transport, farm inputs and fertilizer N_2O, livestock, rice CH_4, and tailpipe emissions. All domestic economic modeling utilized the FASOM model while international modeling utilized FAPRI models combined with Winrock satellite data. Emission factors were taken from GREET, DAYCENT, and IPCC for the relevant emissions categories. Tailpipe emissions were based on MOVES model results. Xunmin et al. (2009) performed a LCA of China's current six biofuel pathways, which are: corn-derived ethanol, cassava-derived ethanol, sweet sorghum-derived ethanol, soybean-derived bio-diesel, jatropha fruit-derived bio-diesel, and used cooking oil-derived bio-diesel. WTW module of Tsinghua-CA3EM model was used. The comparison with conventional petroleum-based gasoline and diesel pathways was performed. Only energy consumption and GHG emissions were analyzed.

To the authors' knowledge, there have not been published works regarding to LCA of corn-based ethanol production in Argentina by applying the LCA methodology. However, there exists some works addressing environmental issues or aspects such as Lavado et al. (2001), Fabrizzi et al. (2005), Apezteguía et al. (2009), Domínguez et al. (2009), Martinello and Giner (2010), Cisneros et al. (2011) and Timilsina et al. (2013). Mele et al. (2011) and Acreche and Valeiro (2013) presented works on LCA of ethanol production for the Argentina context from sugar cane, and Asal et al. (2006), Panichelli et al. (2009), Tomei and Upham (2009), van Dam et al. (2009) and Emmenegger et al. (2011) on LCA of biodiesel production in Argentina.

6. System description and inventory data

6.1. Goals and scope definition

The aim is to assess the environmental impact of corn-based ethanol production in the province of Santa Fe in Argentina. The studied system includes from raw materials production to anhydrous ethanol production using dry milling technology. The treatment of stillage is considered as well as the use of co-products (distiller's dried grains with solubles), but the use and/or application of the produced biofuel is not analyzed, i.e. a “cradle to gate” analysis is performed. Sensitivity analysis to evaluate the influence of the perspectives on the model is performed, but not the uncertainties on the inventory data.

6.2. System definition and boundaries

The analyzed system is divided into two subsystems: agricultural system (S1) and refinery system (S2). In S1, soil under continuous direct seeding for 40 years was assumed. Therefore, direct and indirect carbon emissions originated by land use changes were not included (Searchinger et al., 2008). Even if the carbon content in soil was considered stable, it was included in the analysis because it depends on the used farming practice. Note that land used for food is devoted to bioenergy feedstock production and the demand for food still remains. Although greenhouse gas emissions from this land use change are deemed to be even more important than emissions from direct land use change, no methodological standards exist on this issue (Cherubini and Strømman, 2011); therefore, it is not considered in this study.

In S2, all stages until anhydrous ethanol production are considered: milling, liquefaction, saccharification, distillation, dehydration and stillage treatment, including the use of DDGS, but without taking into account the use of biofuel. According to the Instituto Nacional de Vitivinicultura (2012), a denaturalization with 1.5% of gasoline is assumed to allow anhydrous ethanol transportation as fuel instead of food. The drying unit is assumed to be an emission-controlled unit. 1 MJ of anhydrous ethanol at biorefinery plant was chosen as functional unit for the system. System boundaries are shown in Fig. 2. Regarding to transport, the arrow that goes from S1 to S2 refers to corn transport while the other transport arrow entering the refinery refers to chemical compounds transport. Finally, there is a transport arrow entering the agricultural subsystem related to seeds, pesticides and fertilizers.

6.3. Inventory analysis

The used agricultural data are specific to Argentina. The dry milling input data are based on national projects and the USA reference case (Jungbluth and Emmenegger, 2007) adapted to the Argentine context. Transport distances are based on average distances in the province of Santa Fe.

6.3.1. Agricultural subsystem (S1)

The inventory data for the agricultural subsystem S1 are presented in Table 2. The average corn yield in the studied period is 7726 kg/ha (Ministerio de Agricultura, Ganadería y Pesca, MAGyP, 2010). The corn cycle lasts for 7 months (Jungbluth and Emmenegger, 2007) and the predominant crop rotation is corn-wheat/soybean, i.e. three crops in two years (García, 2002). As the USA reference case implemented in Ecoinvent database, this analysis takes into account seeds, fertilizers and pesticides production (without considering production and waste treatment of catalysts), diesel fuel consumption, raw material transport, emissions to the air from combustion, and the emission to the soil from tire abrasion during the work process of agricultural machinery

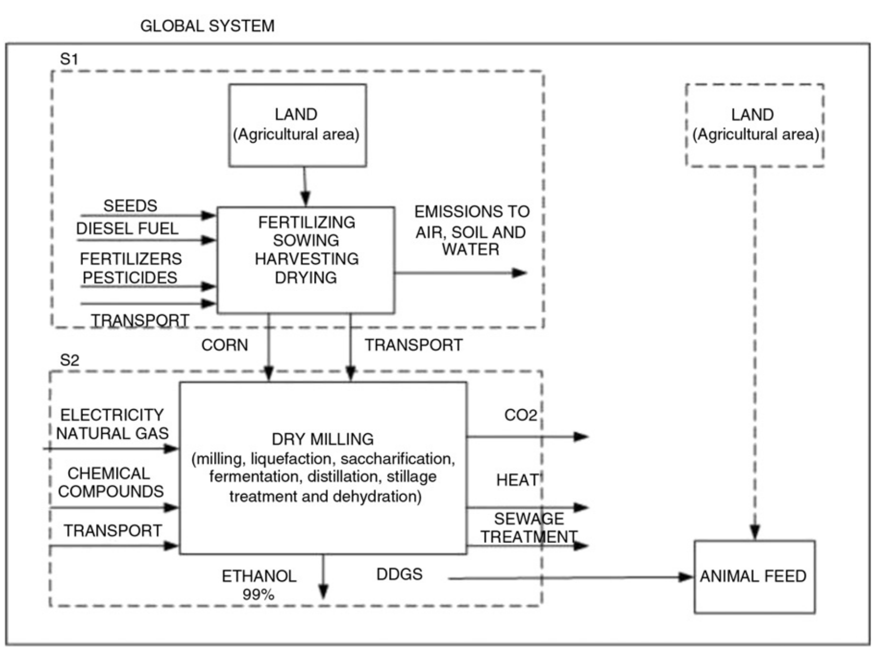

Fig. 2. System boundaries.

Table 2
Inventory data for agricultural subsystem S1.

Products		Materials/fuels	
Corn [kg]	1	Appl. of plant protection products [ha]	1.446E-04
Resources		Combine harvesting [ha]	1.294E-04
Carbon, in organic matter, in soil [kg]	0.148	Maize drying [kg]	1.780E-01
Energy, gross calorific value, in biomass [Mj]	15.910	Transport, tractor and trailer [tkm]	3.002E-02
Occupation, arable, conservation tillage [ha]	9.599E-05	Transport, lorry 28 t [tkm]	6.953E-03
Transformation, from arable [ha]	1.646E-04	Emissions to air	
Transformation, to arable [ha]	1.646E-04	Ammonia [kg]	7.607E-04
Carbon dioxide, in air [kg]	1.350	Dinitrogen monoxide [kg]	3.628E-04
Materials/fuels		Nitrogen oxides [kg]	2.107E-04
Maize seed [kg]	5.430E-03	Methane [g]	1.170E-01
Urea Production[kg]	6.704E-03	Emissions to water	
Urea ammonium nitrate Production [kg]	4.999E-03	Phosphorus to river [kg]	8.667E-05
Diammonium phosphate Production [kg]	3.982E-03	Phosphorus to groundwater [kg]	6.747E-06
Monoammonium phosphate production [kg]	4.502E-03	Nitrate to groundwater [kg]	3.745E-03
Atrazine Production [kg]	7.232E-06	Emissions to soil	
Acetamide–anillide-compounds production [kg]	1.409E-04	Acetamide [kg]	1.409E-04
Glyphosate production [kg]	5.034E-04	Atrazine [kg]	7.232E-06
Pyridine-compound production [kg]	3.824E-05	Pyridine [kg]	3.824E-05
Fertilizing, by broadcaster [ha]	1.446E-04	Glyphosate [kg]	5.034E-04
Sowing [ha]	1.646E-04		

(combine harvesting, sowing, fertilizing, application of plant protection products and irrigating). Regarding to the irrigated area, data reported by Abraham and Gramicci (2007) are considered. In Argentina, gravity-based irrigation systems are the most used irrigation schemes. In this work, an irrigation rate of 1200 m^3 of water per hectare per year is assumed. Corn is dried with direct air heaters that raise the air temperature to 110–120 °C, allowing for a more efficient drying process and a lower fuel consumption per kg of water extracted. The air heaters have a nominal power of 4 MW. In this process, emissions and heat waste to the air from combustion are taken into account, but waste and other emissions such as noise and dust are not. For transportation, trucks' production, maintenance, operation and final disposal are included, as well as construction, maintenance and disposal of roads.

Pesticide, fertilizer and seed amounts are based on data from the Center Roundup Ready Plus (Monsanto Argentina SAIC, 2012), DEKALB Maíz (Monsanto Argentina S.A.I.C., 2010) and the Agricultural Department of Argentina (Frana and Ramuno, 1998). Biogenic CO_2 captured by photosynthesis during plant growth and biomass energy are estimated according to the USA case, as well as the energy content in corn. Carbon sequestration in organic matter by farming practices is considered according to Montico (2009) and Kim and Dale (2005). The applied pesticides are calculated as emissions to soil; heavy metal emissions to soil are not included due to lack of data. NO_x, N_2O and ammonia (NH_3-N) emissions to air are estimated according to NREL (NREL National Renewable Energy Laboratory, 2006). Nitrate and phosphorous emissions to groundwater and phosphorous emissions to surface water are estimated according to Nemecek and Kägi (2007), being both emissions considered in Jungbluth and Emmenegger (2007). As estimated in Cherubini and Ulgiati (2010), CH_4 emissions are also included. Seeds, pesticides and fertilizers are transported over 300 km in 28 t trucks from regional storage to the local area, and 30 km by tractor-trailer to the field. Corn is transported 30 km by tractor to regional storage (SAGPyA, 2006).

6.3.2. Refinery subsystem (S2)

Dry milling technology is chosen for ethanol production, which is more accessible than wet mill for cooperatives due to it demands lower capital requirements for construction and plant operation costs (Vergagni, 2004). Corn, electricity, water and natural gas consumption, and ethanol yield values correspond to a project for producing 74,300 t/yr in Córdoba, Argentina (Agroctanos, 2013), while the rest of process data is based on average international technology provided by Ecoinvent. The transport distances are specific to the Argentine context. The inventory data for the refinery subsystem S2 are shown in Table 3.

Raw materials are transported over 150 km in 28 t trucks, while 80% of corn is transported by 28 t trucks and 20% by train in a radius of 300 km.

Economic allocation is chosen for common stages and carbon balance allocation is chosen for CO_2 emissions, as the USA reference case in Ecoinvent database (Jungbluth and Emmenegger, 2007). The electricity production was based on the Argentina's energy matrix; its transmission and distribution is also taken into account. The heat for process is produced by natural gas in an industrial furnace (>100 kW). The process "heat, natural gas" refers to the heat necessary for the stages of the bioethanol process (such as saccharification and fermentation), while "natural gas, burned in industrial furnace" refers to the natural gas needed for the drying unit. The latter is all allocated to DDGS while CO_2 emissions in fermentation stage are all allocated to ethanol. The considered process raw materials include sulfuric acid, soda (powder), and N-based nutrients in the form of ammonium sulfate and diammonium phosphate. Although enzymes and yeast are needed in this process, their production processes have not been taken into consideration in Ecoinvent database; so, they are not included in this study. Mill infrastructure is included in the same way as in the USA

Table 3
Inventory data for refinery subsystem S2.

Products	
Anhydrous ethanol [MJ]	1
DDGS 90% [kg]	0.033
Materials/fuels	
Corn direct seeding [kg]	0.110
Tap water, at user [kg]	0.247
Sulfuric acid (liquid) production [kg]	8.760E-04
Soda (powder) production [kg]	1.314E-03
Ammonium sulfate production [kg]	3.518E-04
Diammonium phosphate production [kg]	3.518E-04
Transport, freight, rail [tkm]	0.007
Transport, lorry 28 t [tkm]	0.027
Transport, barge [tkm]	6.668E-04
Electricity, medium voltage, at grid [kW h]	0.011
Heat, natural gas [MJ]	0.281
Ethanol fermentation plant [p]	2.018E-11
Natural gas, burned in industrial furnace [MJ]	0.272
Gasoline, unleaded [kg]	5.435E-04
Emissions to air	
Carbon dioxide, biogenic [kg]	0.092
Heat, waste [MJ]	0.385
Waste to treatment	
Treatment, sewage [m^3]	1.761E-04

reference case. Regarding to dehydration stage, as the processes of fermentation and distillation are not physically separated in the plant, molecular sieves are included in the fermentation infrastructure. It is assumed that 100% of the electricity consumed in the dehydration stage is converted to waste heat and released to the air. The natural gas needed for the molecular sieves operation represents about the 6% of the natural gas used in the whole process, while the electricity represents 3%. As for trucks, construction, maintenance, operation and railway structure disposal are considered for train transport.

7. Results and discussion

7.1. Classification and characterization

The inventory data are classified for the all 11 impact categories considered in Eco-indicator 99 and the all 17 in ReCiPe endpoint level. The characterization allows seeing the percentage contribution of each process to the total environmental impact of a given system. Both subsystems S1 and S2 and the global system GS (S1 coupled to S2) are characterized according to the chosen LCIA methods: Eco-indicator 99 (Fig. 3) and the endpoint level of ReCiPe (Fig. 4). Regarding to characterization for the refinery subsystem S2, all environment impacts of ethanol production without allocation are discussed. The results are based on the hierarchical (HI) perspective of both methods.

Fig. 3 shows that the use of fertilizers and resources, seeds production, harvesting process, corn drying, and phosphorus fertilizers and acetamide–anillide-compounds production are the most relevant processes in S1 according to Eco-indicator 99. The first one has the most relevant impacts in the categories of acidification/eutrophication (66%), climate change (35%), and respiratory inorganics (28%). The impact in the climate change category is caused by CO_2 and CH_4 emissions while the contributions to the rest are mainly due to nitrogen and ammonia emissions. The high impact of seed production in the land use category (67%) can be attributed to conventional tillage, which is used instead of direct seeding. Corn drying has important impacts (about 30%) in different categories due to its high energy demand, specifically in the fossil fuels, ecotoxicity, ionizing radiation and land use categories. Harvesting process has the same contribution (30%) in respiratory organics category due to benzoalphapyreno and PAH (polycyclic aromatic hydrocarbons) emissions, and a less impact (21%) in minerals category caused by the steel needed for harvester production. Finally, diammonium and monoammonium phosphate production has a high impact in the category of carcinogens (72%) due to the phosphoric acid production, while acetamide–anillide-compounds production, which is the only pesticide production that has a significant impact, has a contribution of 48% in ozone layer category because of trichloromethane production.

Regarding to the refinery subsystem S2, corn production is the most relevant process among all involved processes, with more than 50% of impact in 7 of the 11 categories: land use (91%), carcinogens (80%), acidification/eutrophication (69%), ionizing radiation and minerals (about 63% in both), respiratory inorganics and ecotoxicity (55% in both). Supplied heat and burned natural gas are the other processes with important contributions, particularly in the rest of categories due to their high energy demand and emissions to the air from combustion. Together they represent 47% of impact in the category of respiratory organics, 50% in climate change, and about 58% in fossil fuel and ozone layer categories.

Finally, characterization for the global system GS (S1 coupled to S2) is performed according to allocation mentioned in Section 6. In this study, DDGS' use has only beneficial effects on the environment since they are only considered as animal feed production avoided, which does not affect negatively any of the impact categories. According to Ecoinvent database (Jungbluth and Emmenegger, 2007), a ratio of 0.77 kg animal feed/kg DDGS is assumed. It can be observed in a positive environmental impact of the use of DDGS. The most relevant impact of DDGS' use is in land use category (−100%) because of land use for

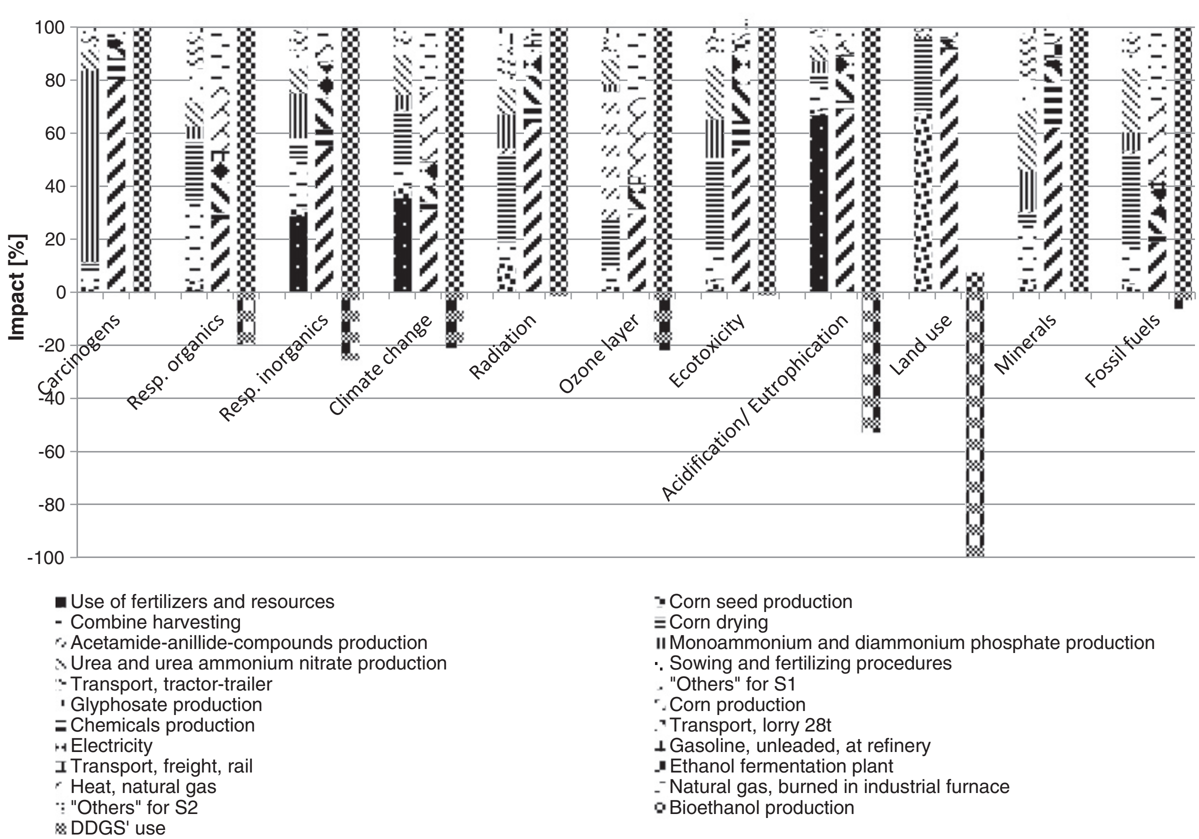

Fig. 3. Characterization by Eco-indicator 99: agricultural subsystem S1 (left bar), refinery subsystem S2 (center bar), and global system GS (right bar).

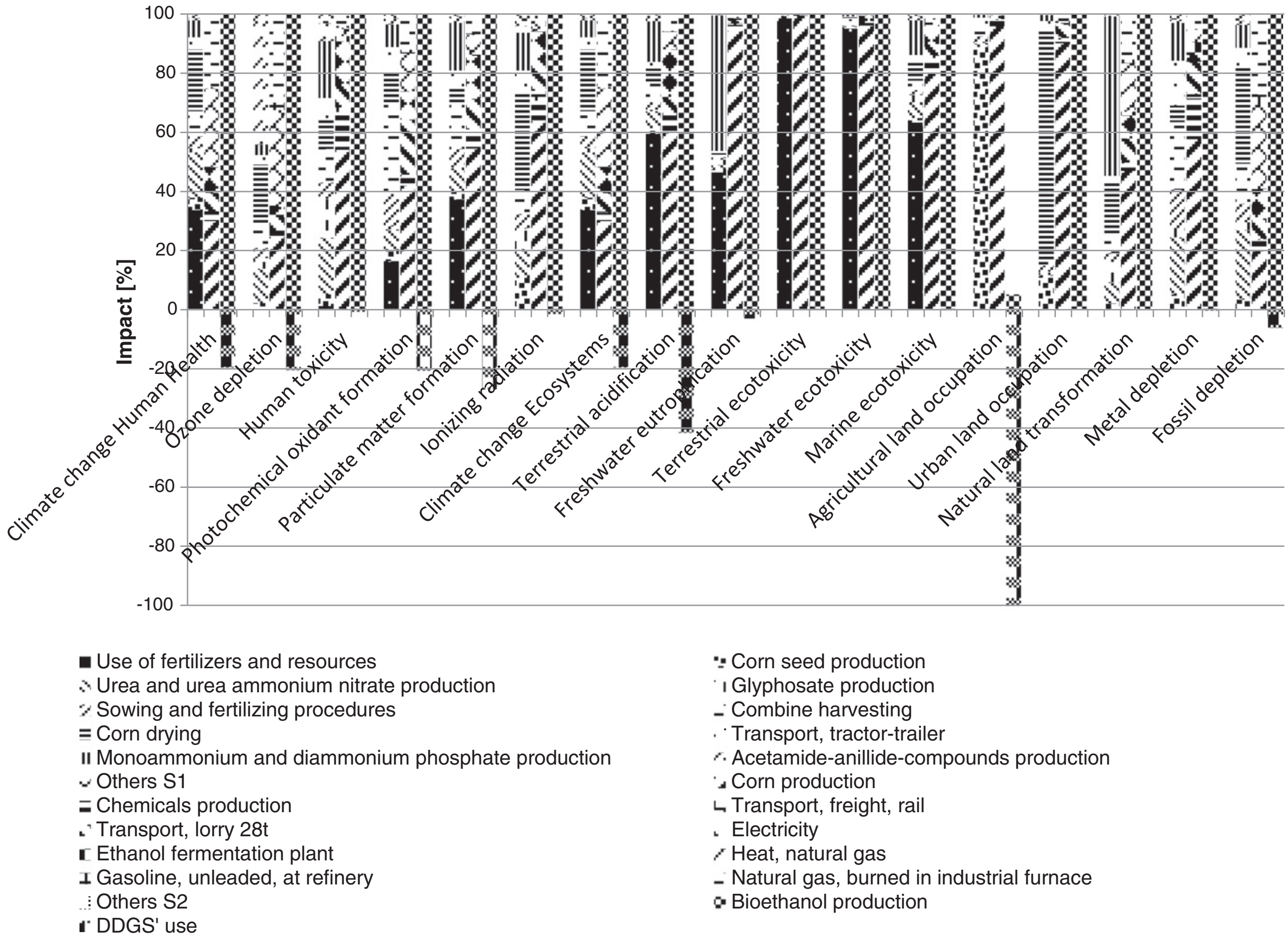

Fig. 4. Characterization by ReCiPe: agricultural subsystem S1 (left bar), refinery subsystem S2 (center bar), and global system GS (right bar).

animal feed is avoided. The second most favored category is acidification/eutrophication (−53%) since the use of fertilizers is also avoided.

Fig. 4 shows the characterization results according to the ReCiPe method. Although the impact categories are different, the most relevant processes are the same as from the Eco-indicator 99 method. The use of fertilizers and resources has the most significant impact in the categories of terrestrial, freshwater and marine ecotoxicity (98, 95 and 63%, respectively); terrestrial acidification (59%); freshwater eutrophication (46%); climate change (33%); and particulate matter formation (37%). Seeds production has a relevant impact in the categories of agricultural land occupation (89%), while corn drying has a contribution of 78% in urban land occupation and 33% in ionizing radiation and fossil depletion. Harvesting process has an impact of 31% in photochemical oxidant formation. Diammonium and monoammonium phosphate production has relevant impacts (54%) in the categories of natural land transformation because of provision of stubbed land in the phosphoric acid production, and freshwater eutrophication (46%), while acetamide–anillide-compounds have an impact of 42% in ozone depletion category. Since both methods compute contributions of pyridine-compounds and atrazine production, irrigation and transport by lorry lower than 2% for all impact categories, are aggregated in Figs. 3 and 4 as “others for S1” processes.

Regarding the characterization results for S2 according to the ReCiPe method, the most relevant processes are the same as for Eco-indicator 99. Corn production has more than 50% contribution in 10 of the 17 categories: terrestrial, freshwater and marine ecotoxicity (99, 96 and 80%, respectively); agricultural and urban land occupation (96 and 90%, respectively); freshwater eutrophication (94%); ionizing radiation (64%); terrestrial acidification and fossil fuels (about 60%); and particulate matter formation (54%). Supplied heat and burned natural gas contribute together to categories of climate change (51%), ozone depletion (62%), fossil depletion (56%), natural land transformation (31%), and photochemical oxidant formation (24%). Since both methods compute contributions of barge transport, tap water and waste treatment lower than 2% for all impact categories, they are aggregated in Figs. 3 and 4 as “others for S2” processes. Finally, similarly to Eco-indicator 99, it can be observed for GS a positive environmental impact of the use of DDGS. The most affected categories by DDGS' use computed by ReCiPe are agricultural land occupation (−100%) and terrestrial acidification (−42%) categories. Land use category in Eco-indicator 99 and agricultural land occupation category in ReCiPe do not reach 100% as the beneficial effects on the impact category are bigger than that of the negative effects. It should be noted that only the percentages related to impact categories in which the impact of DDGS' use is negligible have a total impact equal to 100%; the rest of impact categories presents different total impacts depending on the influence of DDGS' use.

7.2. Damage assessment

This step also allows seeing the percentage contribution of each sub process to the total environmental burden, but in terms of damage instead of impact categories. As both LCIA methods have the same damage categories (human health, ecosystem quality and resources), their results for subsystems S1 and S2 and the global system GS are compared in Fig. 5.

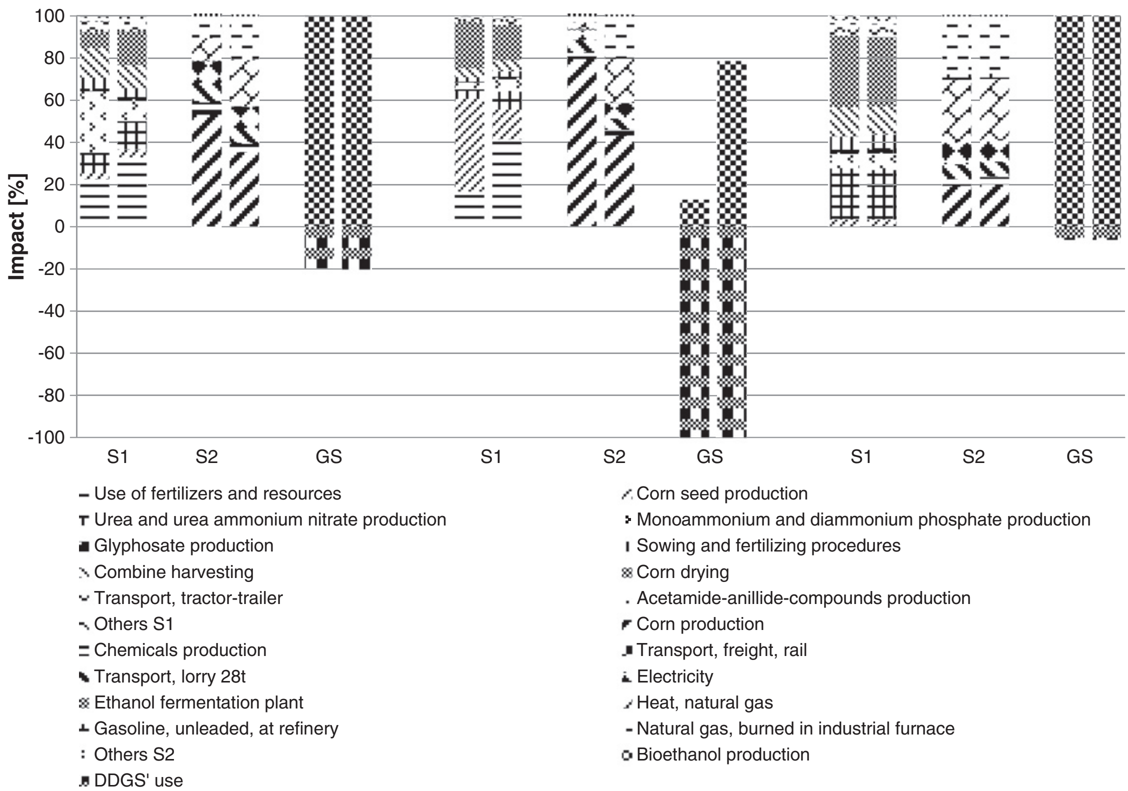

Fig. 5. Damage assessment for S1, S2 and GS by Eco-indicator 99 (left) and ReCiPe (right).

The same predominant processes as in the characterization step are identified, except for acetamide–anillide-compounds production, which is less important.

For subsystem S1, Fig. 5 shows that there are no significant differences between both methods in the resources damage category (less than 2% in the most relevant processes) because the same categories (fossil fuels and minerals depletion) are considered in both methods. However, in human health category, the differences between them are bigger, particularly in phosphorus fertilizers (29% for Eco-indicator 99 vs. 10% for ReCiPe), corn drying (7% vs. 16%) and use of fertilizers and resources (22% vs. 32%). With respect to the first one, the more impact computed by Eco-indicator 99 is due to that phosphorus fertilizers production has a high impact in carcinogens category (included in human health damage category); while, according to ReCiPe, it has more impact in ecosystem quality damage category. Regarding to corn drying, both methods compute similar main impact contributions to radiation category; however, its contributions to human toxicity and photochemical oxidant formation categories, which also correspond to human health damage category, computed by ReCiPe are higher than those of Eco-Indicator 99. Finally, the use of fertilizers and resources has similar contributions to climate change category, which is related to this damage category in both methods; but the impact in particulate matter formation is higher in ReCiPe than that of the impact in respiratory inorganic effects in Eco-indicator 99.

Ecosystem quality damage category shows big differences, particularly in the use of fertilizers and resources (16% vs. 41%) and corn seeds production (45% vs. 14%). In the first case, the difference between both methods is because of the use of fertilizers and resources in Eco-indicator 99 has relevant impacts only in one impact category (acidification), which is related to ecosystem quality damage category; while in ReCiPe this process has significant impact in six impact categories (terrestrial, freshwater and marine ecotoxicity, terrestrial acidification and freshwater eutrophication) of this damage category. In the second case (corn seeds production), although both methods have a relevant impact in land use, the less contribution to the ecosystem quality damage category computed by ReCiPe is due to the land damage is divided in three impact categories (agricultural and urban land occupation, and natural land transformation); then, the impact of corn seeds production has less influence on ecosystem quality damage category.

Similarly to S1, the differences between both LCIA methods for S2 are not significant in resources category, but the differences are big in the other categories. In human health damage category, corn production contributions computed by Eco-indicator 99 and ReCiPe are 53% and 35%, respectively, and the contributions of the sum of supplied heat and burned natural gas are 20% and 42%, respectively. Regarding to corn production, the impacts in ionizing radiation and respiratory inorganic categories in Eco-indicator 99 are similar to the impacts in radiation and particulate matter formation categories in ReCiPe (all categories correspond to human health damage in each method). However, in Eco-indicator 99 this process has a relevant impact in carcinogens category causing a higher impact in the damage category. Therefore, supplied heat and burned natural gas, the second important process, has more importance in ReCiPe. In ecosystem quality damage category, the differences between the contributions of these processes are bigger: 80% vs. 43% for corn production and 5% vs. 41% for supplied heat and burned natural gas, for Eco-indicator 99 and ReCiPe, respectively. Unlike the previous damage category, in this one, ReCiPe shows relevant impacts of supplied heat and burned natural gas in two categories (climate change ecosystem and natural land transformation), while in Eco-indicator 99 these processes do not have any important impact in this damage category. As a result, corn production has more relative impact when using Eco-indicator 99 than ReCiPe.

Finally, the (positive) impacts of the DDGS' use computed by both methods for the global system GS are similar; only the impact of bioethanol production in ecosystem quality damage category is different (12.5% vs. 78.6% for Eco-indicator 99 and ReCiPe, respectively). Although the DDGS' use has equal positive impact in land use category in both methods, in ReCiPe this positive impact results only in one of the three categories related to land damage, what results in less influence. Moreover, there is less positive impact in acidification terrestrial category, resulting in a more negative impact in ecosystem quality damage category.

7.3. Normalization

In the normalization step, a better understanding of the relative proportion or magnitude for each category of a product system under study is performed (ISO, 14040, 2006). The ReCiPe method provides both European and World normalization, while Eco-indicator 99 has only European normalization. The normalization results for damage assessment by applying the World normalization from ReCiPe and the European normalization from Eco-indicator are compared in Figs. 6, 7 and 8 for S1, S2 and GS, respectively.

Most affected categories in the three systems are the same regardless the selected method; human health and resources damage categories have similar importance in both methods. For S1 and S2, the ecosystem quality damage category is more affected by Eco-indicator 99 than that of ReCiPe (around 2.4 times higher). However, for GS, the (positive) impact of the DDGS' use in ecosystem quality damage category computed by Eco-indicator 99 is more than 8 times higher than that of the value computed by ReCiPe. The ecosystem quality category has more negative impacts for ReCiPe than those for Eco-indicator 99. However, according to both methods, ecosystem quality is the least affected damage category in the three examined systems. For both methods, ethanol fermentation plant and transport by freight are added in the category “others” due to their small contributions.

7.4. Sensitivity analysis

Following, the individualist (IN) and egalitarian (EG) perspectives are considered and compared to the hierarchical (HI) perspective. More precisely, the damage assessment step results for the agricultural subsystem S1, refinery subsystem S2 and the global system GS obtained from the three perspectives on both methods are compared. As the tables showing the comparison results are too large, a file containing such comparisons is supplied as supplementary material.

Results obtained from the perspectives comparison for the refinery subsystem S2 for Eco-indicator 99 and ReCiPe are depicted in Fig. 9. Only graphical representations for S2 has been included because this subsystem comprises all processes related to the ethanol production that may lead to improvements in the environmental performance of the studied system.

In the following, only processes with significant contribution to the total impact showing differences higher than 30% between IN or EG perspectives and HI perspective (the reference perspective) are discussed.

7.4.1. Agricultural subsystem S1

- *Eco-indicator 99*. There are significant differences in resources category for the IN perspective of the Eco-indicator 99. The contributions of corn seeds production, phosphorus fertilizers production, transport by tractor-trailer and combine harvesting are, respectively, 46%, 103%, 163% and 51% higher than the reference perspective, while corn drying has an impact of 88% lower than it. Regarding to the EG perspective, no significant differences are observed for the relevant processes in any category.
- *ReCiPe*. The ReCiPe method computes large differences in the human health and ecosystem quality categories but no significant difference in the resources category. The IN perspective presents changes only in the human health category, where the impact of the use of fertilizers and resources is 138% higher than the HI perspective, but the impacts of corn seeds production, phosphorus fertilizers production, combine harvesting and corn drying are around 70% lower than it for all processes. Regarding to the EG perspective, the use of fertilizers and resources has less impact in both damage categories, being 72% and 34% lower than the HI perspective, respectively, but the contributions of phosphorus fertilizers production are 231% and 73% higher than the reference perspective in the human health and ecosystem quality categories, respectively. The impact of combine harvesting in ecosystem quality category is 48% higher.

7.4.2. Refinery subsystem S2

- *Eco-indicator 99*. The IN perspective shows differences in the resources category as in the previous case, while the EG perspective

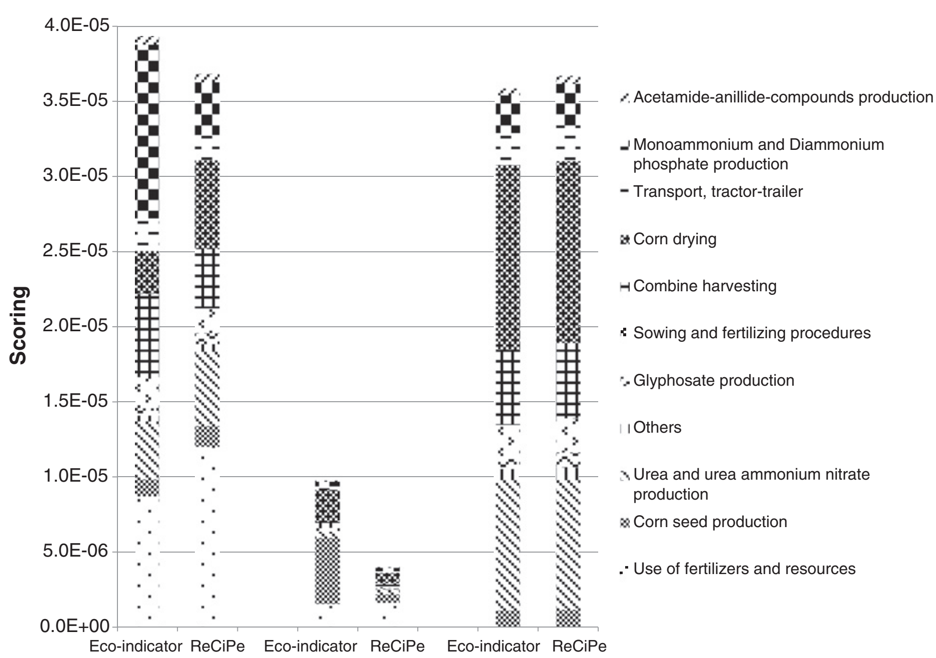

Fig. 6. Normalization results for agricultural subsystem S1: European Eco-indicator 99 (left) vs. World ReCiPe (right) normalization.

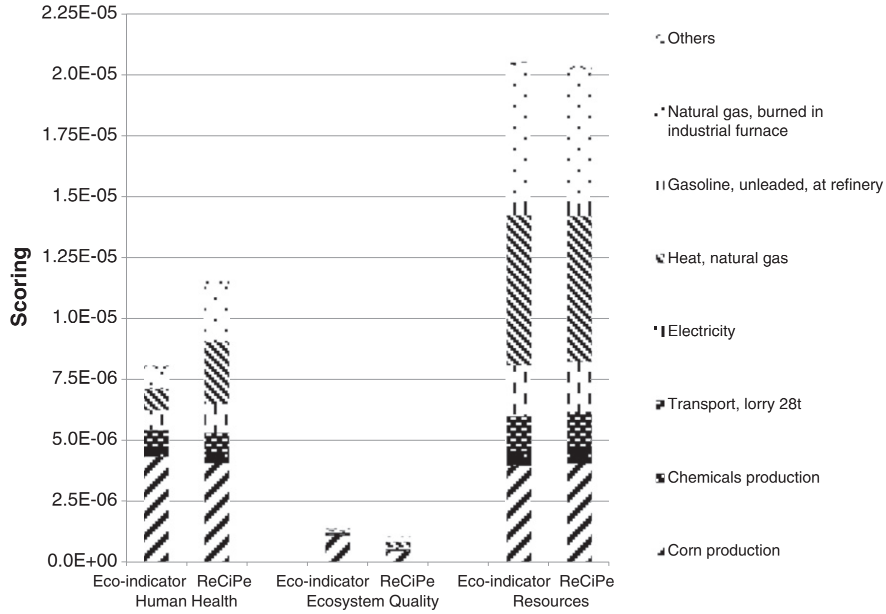

Fig. 7. Normalization results for refinery subsystem S2: European Eco-indicator 99 (left) vs. World ReCiPe (right) normalization.

affects the human health category. For the IN perspective, the impact of corn production and chemicals production is 222% and 445% higher than that of the HI perspective but the contributions of supplied heat and burned natural gas are 92% lower. According to the EG perspective, the contributions of corn production, supplied heat and burned natural gas are lower than the reference perspective (51%, 35% and 35%, respectively) while supplied electricity has an impact of 424% higher.

- *ReCiPe*. It presents the most significant differences in human health category for both EG and IN perspectives. The contributions of supplied heat and burned natural gas computed by them are about 38% lower than those of the HI perspective. The impact of corn production from the IN perspective is 76% higher than that of the reference perspective; while supplied electricity contribution from the EG perspective is 84% higher than it.

7.4.3. Global system GS

When comparing the EG and IN perspectives with respect to HI for the global system GS, ReCiPe computes more differences than Eco-indicator 99.

- *Eco-indicator 99*. There are only two significant differences among perspectives, and are related to the DDGS' use; in the human health category, the DDGS' use reduces 50% the positive impact in the EG perspective, and 96% in the IN perspective. However, the total impact computed by the different perspectives for all damage categories is similar.

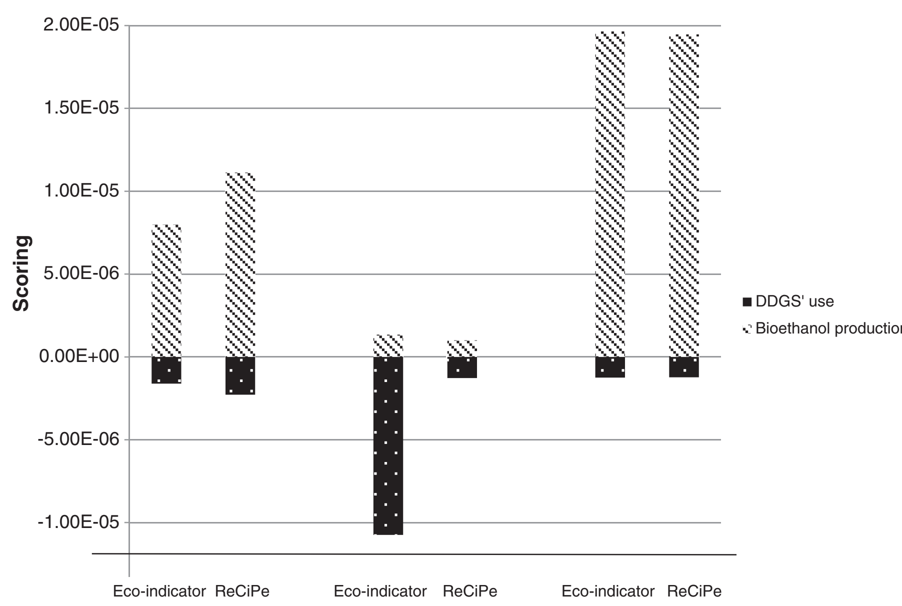

Fig. 8. Normalization results for global system GS: European Eco-indicator 99 (left) vs. World ReCiPe (right) normalization.

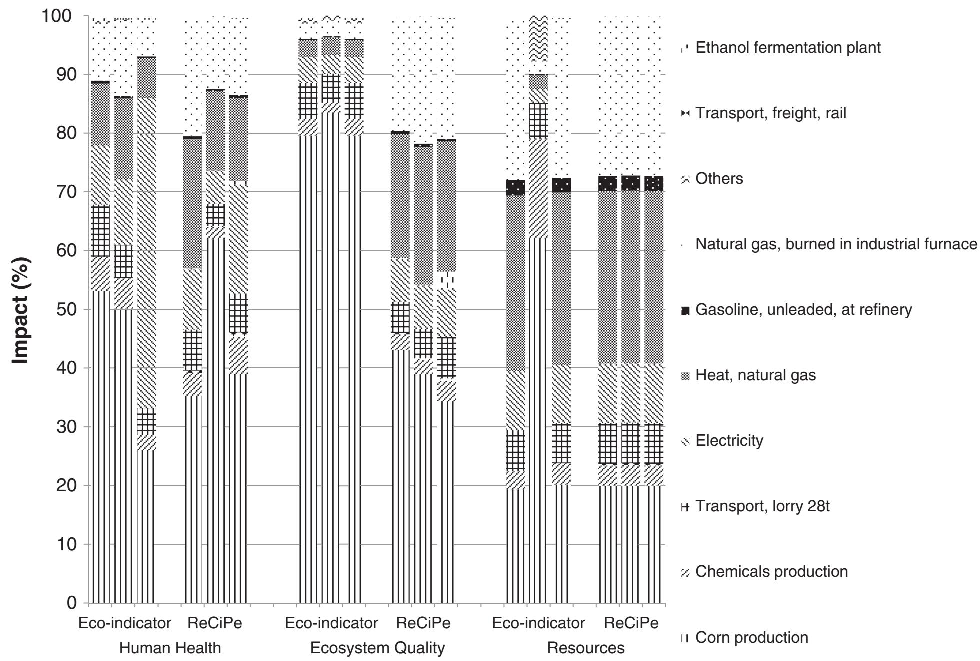

Fig. 9. Perspectives comparison for damage assessment by Eco-indicator 99 and ReCiPe for S2: HI (left bar), IN (center bar) and EG (right bar).

- *ReCiPe*. It has significant differences in the total impact, particularly in the ecosystem quality category, computing a positive impact from the IN and EG perspectives 95% and 261% lower, respectively, than the HI perspective. Regarding to the DDGS' use contribution, 47% lower positive impact in human health category is computed from the IN perspective, while there are lower positive impacts in two damage categories from the EG perspective: 57% and 34% for human health and ecosystem quality categories, respectively. The total impact computed by the IN and EG perspectives for the ecosystem quality category is 2.6 times higher than that of the HI perspective.

8. Conclusions

The environmental performance of corn-based ethanol production was evaluated through the LCA methodology. Most LCA studies reported in literature correspond to the European or North American geographical and economical context. This work is intended to provide, as much as possible, an Argentina-specific LCA study for corn-based ethanol production based on a standard life cycle inventory database such as Ecoinvent. As the impact assessment concerns, besides GWP and energy consumption, other impacts were considered to evaluate in the most complete way as possible the environmental burdens of the process.

The opportunities for environmental improvements of the global system as well as the individual agricultural and refinery subsystems were identified. In fact, the valorization of DDGS determines significant positive impacts; although no-till practice was assumed, corn production has a high negative impact.

Regarding to corn production (agricultural subsystem), the use of fertilizers and resources has the most relevant impact without yet considering the environmental problems related to a safe handling of pesticide containers in Argentina, which pose high risk for human health, water and soil. Corn dry process has also significant impacts, which can be decreased using renewable energy sources instead of natural gas to supply energy. The acetamide–anillide-compounds production is the only pesticide production process that has significant impacts; then, it should be better replaced with atrazine.

Among all involved processes, corn production and supplied energy are the most relevant processes in the refinery subsystem. A cogeneration system can lead to an important environmental performance improvement of this subsystem.

The selected LCIA methods Eco-indicator 99 and ReCiPe identified the same relevant processes but with some different contributions to the total environmental impact. Although it is difficult to discern which method is the most appropriate, ReCiPe is newer and considers more aspects than Eco-indicator 99 such as water ecotoxicity, and includes more metals; in addition, it provides World normalization, which is more suitable for the Argentina's context than the European normalization. In fact, human health and resources damage categories have similar normalization results, but ecosystem quality damage category is more affected by the European normalization than the World normalization, overestimating the impacts in this category. However, according to both methods, it can be concluded that, although human health category is very affected in agricultural subsystem, resources is the most affected category in the refinery subsystem and global system.

In Eco-indicator 99, the individualist perspective underestimates the importance of fossil fuels depletion; then, corn drying in the agricultural subsystem and supplied heat and natural gas burned in the refinery subsystem lose importance, being of significance other processes such as transport by tractor-trailer in the agricultural subsystem and chemicals' production in the refinery subsystem. Regarding to the egalitarian perspective, the electricity process becomes the most relevant one in the human health category for the refinery subsystem. In the case of ReCiPe method, there are no significant changes regarding the relevance of the involved processes, remaining the same ones regardless of the selected perspective. Finally, for the global system, the hierarchical perspective from both methods shows the least environmental impacts. However, Eco-indicator 99 does not predict significant differences in the total impact for all categories while ReCiPe computes big differences, particularly in ecosystem quality category.

The indirect impacts of land use change should be considered since land used for food is devoted to bioenergy feedstock production. Then, as demand for food increases, agricultural production is shifted to other places. Therefore, an exhaustive study to estimate the agricultural expansion is an interesting and necessary challenge for a complete environmental evaluation of the crop-based bioethanol production in Argentina.

Conflict of interest

No conflict of interest.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2013.11.012.

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