Life cycle assessment addressing health effects of particulate matter of mechanical versus manual sugarcane harvesting in Brazil

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Abstract

Purpose: Concerns about environmental impacts and health effects associated with particulate matter emissions of sugarcane production in Brazil have been raised, mainly due to pre-harvest burning of straw in manual harvesting. In consequence, mechanical harvesting without burning has been increasingly adopted. Life-cycle studies have assessed environmental impacts of sugarcane and sugarcane products. However, incorporating health effects of $PM_{2.5}$ in a Life Cycle Assessment focusing on evaluating the impacts of increasing use of mechanization has not been conducted. This article compares the life-cycle environmental and health impacts (with spatially differentiated characterization factors for $PM_{2.5}$) of manual and mechanical harvesting of sugarcane in Brazil, and quantifies the health benefits due to the change of harvesting operations.

Methods: An attributional LCA of manual vs. mechanical sugarcane harvesting was conducted to evaluate the impacts of one tonne of sugarcane at the distillery. ReCiPe was applied to characterize impacts at mid-point (i.e. climate change, fossil depletion, ozone depletion, terrestrial acidification, freshwater eutrophication, human toxicity, photochemical oxidant formation, and particulate matter formation) and end-point (i.e. human health, ecosystems, and resources). Impacts on climate change were compared considering different soil carbon sequestration scenarios. Characterization factors (CFs) of health effects of $PM_{2.5}$ for Brazil were calculated differentiating emission sources, population densities, and burdens of disease.

Results and discussion: At the mid-point, sugarcane production with manual harvesting has higher impacts on photochemical oxidant formation and particulate matter formation mainly due to pre-harvest burning. Mechanical harvesting system may lead to higher impacts on fossil depletion, ozone depletion and terrestrial acidification resulting from higher use of fertilizers and diesel. Differences of impacts on climate change between two systems vary depending on the soil carbon sequestration scenario. At the end-point level, manual harvesting has higher impacts on human health but lower impacts on resources use. The health effects of $PM_{2.5}$ vary considerably with population density. Changing from manual to mechanical harvesting close to urban areas, leads to a 93% reduction of health effects, while for rural only 15% and for remote areas 5%. When considering average population density, the health effects of $PM_{2.5}$ of manual harvesting were approximately six times higher than mechanical harvesting. Health effects of $PM_{2.5}$ calculated with ReCiPe are much lower and may underestimate the effects of primary $PM_{2.5}$ emissions.

Conclusions: The results of this article are an incentive to accelerate the mechanization of sugarcane harvesting in areas with lower mechanization levels (i.e. north-northeast region in Brazil and some rough terrain areas) concerning public health benefits. Meanwhile, manual harvesting with straw burning should only be performed in fields located in rural or remote areas. These results can also contribute to further studies comparing potential benefits of sugarcane culture with alternative crops and guide better decision making at regional development level. Spatially differentiated CFs of $PM_{2.5}$ calculated in this article may be applied to future studies regarding health effects in the Brazilian context.

Key words: Life cycle assessment, $PM_{2.5}$, agricultural automation, public health, air pollution, regional impacts

1. Introduction

Brazil, the world largest sugar producer and the second largest ethanol producer, has been experiencing a rapid expansion of its sugarcane sector in the past decades. In 2015, this country produced 36% of the sugarcane in the world, after an increasing production at an annual growth rate of 5.8% by weight since 2005. The harvesting area reached 10.4 million hectares, resulting in 736 million tonnes of sugarcane in 2014 (OECD-FAO 2016). Sugarcane production is concentrated in the center-south region, led by the State of São Paulo, accounting for 56% of the total production in the 2013/2014 harvest year (UNICA 2015). However, concerns about the environmental impacts and public health risks of sugarcane production have been raised in response to increasing demand for sustainable practices (Seabra et al. 2011; Cavalett et al. 2013). Sugarcane has been conventionally harvested manually, and burning the leaves and tops before harvesting has been a common practice to improve the harvest productivity, facilitate transportation and protect field workers from venomous animals. Sugarcane pre-harvest burning has been associated with increasing public health risks (Arbex et al. 2000; Cançado et al. 2006). The importance of phasing out sugarcane burning has been increasingly realized among companies, environmental groups and at the governmental level since the 1990s (Alves 2009). With incentives from the governments and the industry union, the fraction of sugarcane harvesting area with pre-harvest burning in the State of São Paulo dropped from 77% to 15% from 2005 to 2014. Meanwhile, the percentage of mechanical sugarcane harvesting without pre-harvest burning quadrupled in the same period (CTC 2014).

Regarding the health impacts of sugarcane production in Brazil, several epidemiology studies have pointed out the correlation between particulate matter emissions due to pre-harvest burning and the rising occurrence of respiratory diseases in the communities near sugarcane fields (Arbex et al. 2000; Arbex et al. 2007; Cançado et al. 2006; Mazzoli-Rocha et al. 2008; Uriarte et al. 2009; Goto et al. 2011). Arbex et al. (2007) evaluated the relation between total suspended particles generated from pre-harvest burning and asthma hospital admissions in Araraquara, São Paulo. These authors sustained that increase in total suspended particles was closely related with asthma hospital admissions. Uriarte et al. (2009) and Cançado et al. (2006) studied the impacts of particle emissions due to sugarcane burning on the respiratory health of children and the elderly. Both studies concluded sugarcane pre-harvest burning was the main cause of hospital respiratory admissions for both age groups. Other researchers studied the chemical characteristics of atmospheric particles in the areas close to sugarcane plantations during the burning

seasons. Carcinogenic components such as Polycyclic Aromatic Hydrocarbons (PAHs) were found, and significant rises of PAH concentrations during the burning seasons compared to non-burning seasons were reported (Zamperlini et al. 1997; Andrade et al. 2010; Silva et al. 2010; Cristale et al. 2012).

Life-cycle studies have been conducted to assess the environmental impacts of sugarcane ethanol in Brazil (Macedo et al. 2008; Luo et al. 2009; Ometto et al. 2009; Seabra et al. 2011; Cavalett et al. 2013; Galdos et al. 2013; Tsiropoulos et al. 2014; Chagas et al. 2016). The majority of studies have assessed environmental impacts of sugarcane ethanol, focusing on energy use and greenhouse gases emissions. However, health effects were not addressed with the exception of Galdos et al. (2013) and Tsiropoulos et al. (2014), which calculated human health impacts at the end-point level applying global characterization factors using established life cycle impact assessment methods, Ecoindicator 99 and Impact 2002+, respectively. According to previous studies, the agricultural phase (sugarcane production) contributed the most to the life-cycle environmental impacts of bioethanol. Harvesting is among the largest contributors on GHG emissions, but has been treated as a combination of manual and mechanical operations, expressed by a certain ratio or scenarios of different ratios of mechanical harvesting. Considering the importance of harvesting process on environmental and health impacts associated with sugarcane production, a life-cycle study assessing the broad environmental impacts in the conversion from manual to mechanical harvesting of sugarcane production, with a focus on health impacts of particulate matter emissions is lacking.

Fine particulate matter ($PM_{2.5}$) contributes the most to health effects associated with particulate matter emissions among all sizes (Humbert 2010). Discussions about characterizing health effects of $PM_{2.5}$ are on the rise in the LCA community (Fantke et al. 2015; Gronlund et al. 2015; Humbert et al. 2011). However, few studies were published adopting regional characterization factors (CF) for $PM_{2.5}$ for regions outside of Europe and the United States. An LCA of sugarcane production in Brazil incorporating health effects from $PM_{2.5}$ exposure calculated with regional characterization factors is needed since it can quantify the magnitude of health benefits of replacing manual by mechanical harvesting, and contribute to the assessment of health effects of $PM_{2.5}$ in LCA practices.

This article compares the life-cycle environmental and health impacts of manual and mechanical sugarcane harvesting in Brazil considering fine particulate matter emissions. The health effects associated with $PM_{2.5}$ emissions were assessed using characterization factors that differentiate geographical features of emission sources and consider different burdens of disease. Characterization factors of primary and secondary $PM_{2.5}$ for Brazil were

calculated and implemented. The results of this article can provide incentives to accelerate the mechanization of sugarcane harvesting in areas with lower mechanization rate concerning the magnitude of public health benefits. These results can also contribute to further studies comparing potential benefits of sugarcane culture with alternative crops and guide better decision making at regional development level. Characterization factors of $PM_{2.5}$ calculated in this study may also be applied to future studies regarding health effects of $PM_{2.5}$ in the Brazilian context.

2. Materials and methods

2.1 Life cycle model and inventory

A comparative cradle-to-gate Life Cycle Assessment was conducted, addressing sugarcane cultivation, harvesting and transportation in the center-south region of Brazil. Two sugarcane product systems were investigated and compared: one harvested manually with pre-harvest burning and the other in which this operation occurs mechanically without pre-harvest burning. The functional unit chosen is 1 tonne of sugarcane at the distillery. A simplified diagram of the product system is shown in Fig.1. A life cycle inventory based on the database of Brazilian Bioethanol Science and Technology Laboratory (CTBE) was collected and implemented (Bonomi et al. 2016). Detailed inventory presented by functional unit were included in Online Resource (Appendix 1). The inventory includes average data representing current technologies and operations of manual and mechanical sugarcane harvesting in the center-south region of Brazil. The sugarcane yield per hectare of manual harvesting system is slightly higher than in mechanical harvesting. It is because in manual harvesting, sugarcane sets are semi-mechanically planted requiring 12 tonne setts per hectare, while in mechanical harvesting the planting process is fully mechanized, requiring 20 tonne setts per hectare to compensate for inefficiency of the planting machine (CONAB 2011). Transport of raw materials and final products was also included in the product system. The average transportation distance of sugarcane stalks from the field to the mill is assumed as 25 km (Chagas et al. 2016). Production and field emissions of raw materials including organic and inorganic fertilizers, agrochemicals, diesel used in agricultural machineries were also considered. Vinasse, the liquid effluent of ethanol distillation, boiler ashes and filter cake, were used together with inorganic fertilizer (urea, SSP, and KCl) to supply the nutritional requirements of the sugarcane culture. The emissions of transporting vinasse from the industrial plant to the field (25 km), and operations of pumping, storage and aspersion were included. Capital goods including harvesters, tractors and agricultural machineries were also accounted for.

Regarding estimations of field emissions, for inorganic N fertilizer, 30% of the total N applied as urea was considered to be emitted as ammonia, and 1% of the ammonia was converted as N_2O. 1% of the total N applied directly emitted as N_2O, and 0.75% of the nitrogen leached were assumed to be emitted as N_2O (Costa 2003; Nemecek et al. 2007). Estimations of emissions from organic fertilizers (vinasse and filter cake) and sugarcane residues (straw and roots) followed IPCC (2006), emission factor for direct and indirect N_2O emissions was established as 1.22%, nitrogen content was assumed as 0.595 kgN/m3 for vinasse and 12.5 kgN/tonne for filter cake (Macedo 2007; Chagas et al. 2016). We assumed 4.77 gN/kg of sugarcane straw and 5.1 gN/kg of sugarcane roots, with a root:shoot ratio (defined as the weight of all biomass below the ground surface divided by the weight of all biomass above the ground surface, on a dry basis) of 0.2 (Smith et al. 2005; Hassuani 2005). Quantification of climate change impacts followed the concept of neutral biogenic carbon, thus emissions of biogenic CO_2 from burning sugarcane residues and straw, as well as the capture of CO_2 by sugarcane were not accounted for. Emission factors of sugarcane straw (leaves and tops) burning were based on GREET (2009) and França et al. (2012). Details of assumptions and emission factors applied are in Online Resource (Appendix 2).

Sugarcane residues left on the ground from mechanical harvesting may result in an increase in soil organic carbon thus reducing CO_2 emissions depending on the level of soil carbon saturation of sugarcane fields. Carbon accumulation rates ranging from 1.1 – 1.5 tonne C/ha/year in sugarcane fields in São Paulo at the time span of 4 – 16 years and the soil depth of 20 – 60 cm have been reported (Galdos et al. 2009; Cerri et al. 2011; Carvalho et al. 2013; Segnini et al. 2013). Research on integrating soil organic carbon sequestration (SOC) in LCA is on the rise, but there is lack of consensus on how to proceed at the methodological level (Bosco et al. 2013; Brandão et al. 2013; Petersen et al. 2013). We calculated SOC following the IPCC guidelines (IPCC 2006). The IPCC method is based on the assumptions that soil carbon stock in a certain field will be saturated at some point, and soil carbon changes over time are linear. The default values of the time dependence of stock change and the soil depth in the IPCC method are 20 years and 30 cm respectively. The lands currently adopting mechanical harvesting of sugarcane have mostly experienced the transition from manual harvesting with pre-harvest burning, which was a dominant agricultural practice of sugarcane sector throughout Brazil for decades. To understand the contribution of SOC on climate change, two SOC scenarios were considered in mechanical harvesting without pre-harvesting burning of straw: (i) soil carbon saturated (no SOC increase); and (ii) SOC saturated in a 20-year span. The two SOC scenarios

were selected because they represent the extreme scenarios for soil carbon change when a sugarcane field changes from manual to mechanical harvesting. In the second scenario, a total of 5.2 tonne of carbon was sequestrated per hectare in 20 years, considering conditions of sugarcane plantation in Brazil (temperature zone: tropical moist; Soil type: low activity clay) (IPCC 2006). This scenario projects an average rate of 260 kg carbon sequestered per hectare per year from the sugarcane residues. Detailed assumptions and calculations of soil organic carbon change can be found in Online Resource (Appendix 3).

2.2 Life Cycle Impact Assessment extended with health effects of $PM_{2.5}$

Life cycle impact assessment (LCIA) was carried out for ReCiPe mid-point impacts (climate change, ozone depletion, terrestrial acidification, freshwater eutrophication, human toxicity, photochemical oxidant formation, particulate matter formation, and fossil depletion) and end-point damage categories (human health, ecosystem and resources) (Goedkoop et al. 2013). Hierarchist perspective was adopted since it is the default one in ReCiPe and follows the most common policy principles with regards to time-frame and other issues. ReCiPe method was chosen among other LCIA methods due to its feature of consistent use of midpoints and endpoints in the same environmental mechanism (ILCD 2010), and because it is a widely used method. Impact categories were selected considering the importance of environmental issues for sugarcane production. Water depletion was not addressed because water needs of sugarcane cultivation in the Centre-South of Brazil mostly relied on rainfall with no rainwater storage, and there are no expected differences on water needs between manual and mechanical product systems. To characterize the health effects of fine particulate matter in LCIA, models based on Humbert et al. (2011) and Gronlund et al. (2015) were applied. Two groups of characterization factors for $PM_{2.5}$ were calculated for Brazil, as described below.

2.2.1 Health effects of particulate matter in LCIA

Particulate matter (PM) can be categorized by various sizes and compositions. PM_{10} (PM with aerodynamic diameters lower than or equal to 10 μm) and $PM_{2.5}$ (a subset of PM_{10} with aerodynamic diameters lower than or equal to 2.5 μm) are the size ranges widely monitored at emission sources and in ambient air. PM can be emitted directly, referred as primary PM, or be referred as secondary PM, when formed in the atmosphere from chemical reactions involving primary gaseous emissions (e.g. SO_2, NO_x, and NH_3). $PM_{10\text{-}2.5}$ (a subset of PM_{10} with

aerodynamic diameters between 2.5 μm and 10 μm) is composed largely of primary PM. On the other hand, a much greater portion of $PM_{2.5}$ contains secondary PM (Humbert et al. 2015). Particulate matter (PM) has been widely recognized for its adverse human health effects with $PM_{2.5}$ being the main contributor among all PM sizes (Humbert 2010). A number of epidemiological studies showed that $PM_{2.5}$ is related to heart disease, lung cancer, reduced life expectancy, and low birth weight (Laden et al. 2000; Pope et al. 2002; Bell et al. 2008; Pope et al. 2009). Health effects of PM are evaluated in most life-cycle studies under the mid-point impact categories of particulate matter/respiratory inorganics, respiratory effects or human toxicity, and attributable to the end-point area of protection of human health, usually expressed by disability-adjusted life year (DALY) (Humbert et al. 2015). Widely applied LCIA methods such as CML, ReCiPe, Ecoindicator99 and IMPACT 2002+, do not differentiate the health effects of PM based on its size nor the geographical characteristics of emission sources (Notter 2015). Global efforts have been carried out to provide recommendations and guidance on evaluating the health effects of PM in LCAs, such as the flagship project launched by UNEP/SETAC Life Cycle Initiative and the study performed for the Joint Research Center of the European Commission (JRC). Both projects have contributed to identify the best practices for characterization modeling in LCA (Hauschild et al. 2013; Fantke et al. 2015).

A generic framework for assessing health effects of PM can be expressed by Equation 1 (Fantke et al. 2015),

$$\mathrm{IS} = \mathrm{m} \times \mathrm{CF} = \mathrm{m} \times \mathrm{iF} \times \mathrm{ERF} \times \mathrm{SF} \quad (1)$$

Where IS is the impact score (usually expressed by DALY); m is the mass emitted of PM; iF stands for intake fraction (fraction of the mass of PM inhaled by the affected population over the mass of primary PM or secondary PM precursors emitted). Exposure-response factor, ERF, links the health effects in the affected population to the ambient PM concentration. ERF is commonly derived from epidemiological studies and expressed by disease rate or risks per unit mass concentration. SF stands for severity factor, which is usually expressed by DALY per disease case or unit of risk. The product of iF, ERF and SF represents the characterization factor (CF; DALY per mass emitted). Humbert et al. (2011) developed a set of intake fractions considering emission release height (high stack, low stack, ground level) and archetypal environment (indoor, outdoor: urban, rural, and remote). As pointed out in the two abovementioned international projects, Humbert et al. (2011) is considered to be probably one of the most comprehensive existing models on deriving iF for PM characterization.

Compared to the development of iF, less consensus has been reached on the development of exposure-response assessment (Fantke et al. 2015). In most methods (Pope et al. 2002; WHO 2006; Van Zelm et al. 2008), a linear, no-threshold exposure-response curve is often assumed; however, when the concentration of PM is not within the range (~10-35 μg/m^3 for $PM_{2.5}$) of ambient PM concentration observed in the epidemiological studies often conducted in the European or American conditions, the linearity assumption may not hold (Lim et al. 2012; Burnett et al. 2014; Humbert et al. 2015). With respect to the health effects associated with PM exposure, the field is under development and more consensus need to be achieved among the scientific community. Van Zelm et al. (2008) considered chronic and acute mortality, and acute respiratory and cardiovascular morbidity due to exposure to PM_{10}. Gronlund et al. (2015) accounts for cardiopulmonary and lung cancer mortality attributable to chronic exposure to $PM_{2.5}$. Humbert et al. (2010) also proposed a set of health effects should be considered and corresponding effect factors for PM_{10} and $PM_{2.5}$. Fantke et al. (2015) pointed out the potential of the Global Burden of Disease (GBD) 2010 study as a starting point for calculating health effects of $PM_{2.5}$ exposure. $PM_{2.5}$ as one of the 67 risk factors in the GBD study is related to five adverse health effects, including ischemic heart disease (IHD), cerebrovascular disease (stroke), chronic obstructive pulmonary disease among adults (COPD), trachea, bronchus and lung cancer (LC), and lower respiratory infections among young (Lim et al. 2012). To current knowledge, $PM_{2.5}$ is primarily responsible for human health effects related to PM emissions (Humber et al. 2015). For this reason, merely health effect of primary and secondary $PM_{2.5}$ were assessed in this article.

2.2.2 Calculating characterization factors of $PM_{2.5}$ for Brazil

We have first calculated intake fractions for Latin America based on methods implemented in Humbert et al. (2011). Secondly, we calculated two groups of effect factors considering different burdens of disease: the first group adopted the dose-response factors estimated by Gronlund et al. (2015) and the severity factors calculated based on Global Burden of Disease for Brazil (WHO 2004); while the second group followed Humbert (2010).We chose these two groups of effect factors because they represent the latest methods of exposure-response assessment in the literature; meanwhile, it is worth noting how the magnitude of effect factors vary depending on the burdens of disease considered. Finally, the intake fractions were multiplied with the two groups of effect factors respectively to generate the characterization factors.

We calculated intake fractions of primary and secondary $PM_{2.5}$ for Latin America (Table 1) based on the emission-weighted average iF recommended for Latin America and the methods to differentiate the intake fractions based on emission heights and population densities from Humbert et al. (2011). Equations and values used for calculation can be found in Online Resource (Appendix 4). For secondary $PM_{2.5}$, stack height has limited importance in affecting iF. Distance from the affected population to emission locations is a critical factor influencing the magnitude of the health effects of $PM_{2.5}$. When distance to the affected population is unknown, population-weighted iF can be employed, which is a weighted sum of iF for urban, rural and remote with its corresponding fraction of population in the region. In Brazil, the distances between sugarcane fields and populated areas varied significantly from one place to another. For instance, according to CANASAT, a project developed by National Institute for Space Research (INPE) aiming at mapping the sugarcane cultivation and harvest activities in São Paulo State, some municipalities such as Ribeirão Preto are closely surrounded by sugarcane plantation, while other municipalities are hundreds of kilometers away. Due to this reason, CFs calculated using population-weighted iF was applied to characterize the health effects.

Regarding effect factors, the first group (Table 2) was calculated for Brazil based on Gronlund et al. (2013). Mortalities due to cardiopulmonary diseases and lung cancer were considered, and the total effect factor is 45.6 DALY/kg $PM_{2.5}$ inhaled. Data of Global Burden of Disease for Brazil were collected and implemented. Ischemic heart disease, cerebrovascular disease and chronic obstructive pulmonary disease were considered under cardiopulmonary disease (GBD disease code: W107, W108, W112 respectively). For lung cancer, the analysis took into account trachea, bronchus and lung cancer (GBD disease code: W067). The second set of effect factors were calculated based on the values proposed in Humbert (2010). A wider range of diseases were considered including chronic mortality, acute respiratory and cardiovascular morbidity, chronic bronchitis for children and adults, asthma attacks for children and adults and restricted activity days. Assuming $PM_{2.5}$ is 1.67 times as toxic as PM_{10} (European Commission 2005), the effect factor for $PM_{2.5}$ was calculated to be 137 DALY/kg $PM_{2.5}$ inhaled. For clarification, CF calculated with effect factor considering cardiovascular diseases and lung cancer is referred as Cardio.& Lung, and CF calculated with effect factor based on Humber (2010) is referred as Humbert.

The two groups of CFs of $PM_{2.5}$ calculated are presented in Table 3. Inventory of primary $PM_{2.5}$ and secondary $PM_{2.5}$ precursors of all the unit processes were then aggregated and multiplied with the relevant CFs

according to different sources and emission heights. Heights of $PM_{2.5}$ emissions from production of raw materials such as fertilizers, pesticides and diesel were unknown, thus emission-weighted CFs were applied. $PM_{2.5}$ emissions from transportation and field emissions from fertilizer use and residue burning are considered to be at the ground-level, and CFs at the ground-level were used. A sensitivity analysis was conducted to discuss the influence of distance between emission sources and population (urban, rural and remote) on health impacts.

3. Results and discussion

3.1 Mid-point impacts

Table 4 showed LCA results at the mid-point level and relative difference (Δ) between two systems. Mechanical harvesting had much lower impacts for photochemical oxidant formation (Δ = - 88%) and particulate matter formation (Δ = - 61%). Manual harvesting presented slightly better performances on fossil depletion, ozone depletion and terrestrial acidification (Δ = 17% to Δ = 19%). Differences of two product systems on freshwater eutrophication and human toxicity were very small (Δ <5%). The contributions for impacts from different processes were detailed in Figure 2, and results for each mid-point impact category were described in the following paragraphs.

Fossil depletion: Mechanical harvesting lead to 17% higher impacts on fossil depletion than manual harvesting. Diesel use in the sugarcane fields was related with 25% and 28% of the impacts in the manual and mechanical harvesting systems respectively, followed by diesel production and fertilizer production as main contributors on this impact category. Among fertilizer production, nitrogen fertilizer was responsible for more than 75% of the impacts resulting from fertilizer production. The worse performance of mechanical harvesting in this category is mainly due to the higher use of fertilizer and diesel compared to manual harvesting. Higher use of potassium in mechanical harvesting system is related to the lower recycling rate of this nutrient when sugarcane straw is not burnt. Similarly, higher use of nitrogen is explained by the need of additional amount to make up for the decreased efficiency of this fertilizer when applied over the straw mulch.

Ozone depletion: Mechanical harvesting system had 19% higher impacts due to higher inputs of diesel and fertilizer. Production of nitrogen fertilizer was the largest contributor for both systems, accounting for 34% of the impacts for the mechanical harvesting system, and 25% for manual harvesting.

Terrestrial acidification: Manual harvesting had 17% lower impacts on terrestrial acidification than mechanical harvesting. Fertilizer field emissions contributed the most mainly due to emissions of ammonia, accounting for 83% and 96% of the impacts respectively for the manual and mechanical harvesting systems. For the manual harvesting system, sugarcane residue burning was another main contributor on this category, presenting 13% of impacts. It is worth mentioning that ammonia emitted from fertilizer use was calculated based on the IPCC method assuming the same soil conditions and NH_3 emission factor for both systems, which is a simplification that we acknowledge may be revised with future data and models. However, it is important to mention that a higher amount of nitrogen fertilizer (and consequently higher emissions from NH_3 volatilization) is used in mechanized harvesting systems for compensating higher nitrogen volatilization losses due to fertilization in the presence of sugarcane straw on the soil.

Freshwater eutrophication: The difference between two systems was less pronounced on this category. Production of nitrogen and phosphate fertilizers accounted for approximately one third of the impacts; while production of capital goods including agricultural machinery, tractors and harvesters represented another one third.

Human toxicity: Difference of two product systems on this impact category was unclear. For both harvesting systems, the processes that presented the highest impacts for human toxicity were fertilizer production and field application. Production of capital goods was another major source of impacts for this category, representing 22-23% of the impacts for both systems.

Photochemical oxidant formation: Manual harvesting had much higher impacts than mechanical harvesting due to emissions of carbon monoxide and nitrogen oxides from pre-harvest burning. For mechanical harvesting, around 46% of the impacts occurred in the sugarcane field from fertilizer use, diesel burning, and vinasse application.

Particulate matter formation: Mechanical harvesting appeared to have 61% less impacts, while 90% of the impacts came from the field emissions of ammonia due to fertilizer use. However, fertilizer use only contributed to 25% of the impact in the manual harvesting system. $PM_{2.5}$ emissions from pre-harvest burning was the largest source, accounting for 70% of the impacts.

3.1.1 Climate change considering two SOC scenarios

Figure 3 compared climate change impacts of manual and mechanical harvesting systems considering the two scenarios of soil carbon sequestration (SOC) previously mentioned: (i) soil carbon saturated (no SOC increase); and

(ii) SOC saturated at a time span of 20 years. Manual harvesting resulted in 38.3 $kgCO_{2eq}$/tonne sugarcane. Changing the harvesting operation to a mechanized system lead to an increase of 6% on climate change impacts when not considering the contribution of SOC, whilst a decrease of nearly 25% is observed when considering SOC.

Fertilizer application was the largest contributor on climate change in all the scenarios, accounting for approximately 55% of the total impact in the mechanical harvesting system without SOC, and 40% in the manual harvesting system. Diesel burning in agricultural operations was another important contributor to climate change, representing 12% and 10% of the total for mechanical (no SOC) and manual harvesting systems, respectively. GHG emissions from pre-harvest burning corresponded to 18% of the total GHG emissions of the manual harvesting system. In both scenarios, more than 70% of the impacts occurred in the sugarcane field, mainly due to the emissions of N_2O and CO_2.

3.2 End-point damage impacts

Environmental impacts at damage (end-point) level were presented in Fig 4. Mechanical harvesting had lower impacts (Δ = -43% to Δ = -51%) on human health for both SOC scenarios compared to manual harvesting. This is mainly due to the elimination of pre-harvest burning practices. On the other hand, because of higher fertilizer and diesel use, mechanical harvesting had higher impacts on resources increasing by 17%. However, as pointed out in the LCIA literature (e.g. ILCD 2011), there is important uncertainty associated with end-point results, which should be taken into consideration when discussing LCA results. Regarding the impacts on ecosystems, the difference between manual and mechanical harvesting is unclear, although when considering SOC increase, it may suggest a slightly lower impact of mechanical harvesting system.

3.3 Health effects of $PM_{2.5}$

Fig 5 compares the health effects of $PM_{2.5}$ of manual and mechanical harvesting, calculated with population-weighted CFs together with a sensitivity analysis for different population densities (urban, rural and remote) and applying ReCiPe. The results show that population density is a key factor when assessing the health effects of $PM_{2.5}$. Health effects for manual harvesting in urban and population-weighted conditions were much larger than for rural and remote conditions. Mechanical harvesting showed lower health impacts than manual harvesting in all conditions, but important differences were only observed when applying urban and population-weighted CFs. Comparing our

results with those calculated using ReCiPe, it shows ReCiPe underestimates health effects for population-weighted condition (1.5-6.6 timers lower), whilst showing comparable results with rural condition.

With regards to the two groups of CFs considering different burdens of disease, when applying population-weighted CFs, producing one tonne of sugarcane in manual harvesting resulted in a loss of 3.7 x 10^{-4} DALY and 1.1 x 10^{-3} DALY respectively, due to the different burdens of disease considered in the CFs. When a wider range of diseases was considered, health effects were two times higher than when only considering cardiovascular diseases and lung cancer. This difference highlighted the needs for more transparency and consensus regarding effect factors when characterizing health effects of $PM_{2.5}$.

Health effects of mechanical harvesting do not vary with population density as much as manual harvesting, because more than 90% of the health effects are due to secondary $PM_{2.5}$ from NH_3 associated with fertilizer field application; while for manual harvesting, primary $PM_{2.5}$ contributed the most to health effects. The CFs of NH_3 calculated in this article were much smaller than the CFs of primary $PM_{2.5}$, and fairly comparable with CF of NH_3 in ReCiPe. The CFs of primary $PM_{2.5}$ calculated were higher than the one in ReCiPe. When applying population-weighted CFs, manual harvesting presented six times higher health effects of $PM_{2.5}$ than mechanical harvesting regardless the effect factors chosen. To put it in perspective, from 2005 to 2014 (data for the harvest season of 2006/2007 were missing) and considering population-weighted CFs, if the sugarcane harvested mechanically without pre-harvest burning were harvested manually with pre-harvest burning, it would have resulted in a potential loss of 479 000 - 1 440 000 DALYs. Considering average life expectancy of Brazil in 2014, this is equivalent to 6 438 - 19 355 life losses.

3.4 Comparison with previous studies and limitations

Macedo et al. (2008) found higher energy consumption with increasing percentage of mechanical harvesting, and this is in consensus with our findings on fossil depletion. Galdos et al. (2013) used generic characterization factors from Ecoindicator 99 for particulate matter to assess the health impacts, without differentiating emission sources, specifying burdens of disease and including effects of secondary $PM_{2.5}$ from precursor SO_2. Chagas et al. (2016) compared six sugarcane bioethanol production systems and concluded similar findings of increased impacts on eutrophication, ozone depletion and human toxicity, and lower impacts on photochemical oxidant formation comparing mechanical to manual harvesting system. However, health effects and SOC scenarios were not evaluated,

and higher acidification and global warming impacts from manual harvesting were found, due to different choices on emission factors of sugarcane burning. Factors for generic agricultural residues burning from GREET (2009) were adopted, while in this article we employed emission factors for sugarcane burning based on laboratory experiments (França et al. 2012). Chagas et al. (2016) carried out an uncertainty analysis, based on a similar inventory (also from CTBE database), to assess how parameter uncertainty affects economic and environmental impacts, and reported relatively low standard deviations (SD) of ethanol GHG (SD from 20 to 23 g CO_2 eq/L for a mean value of 518 to 478 gCO_2 eq/L). Thus, this type of parameter uncertainty is not expected to affect the ranking of product systems in this article. Regarding the limitations of our article, characterization of health effects of $PM_{2.5}$ was based on exposure-response factors from Gronlund et al. (2015) calculated for the USA, and intake fractions were calculated for the scale of Latin America, due to lack of specific data for Brazil.

4. Conclusions

This article compared the life-cycle environmental and health impacts of sugarcane produced with manual and mechanical harvesting in Brazil. The results showed that the transition from manual to mechanical sugarcane harvesting systems in Brazil clearly reduces impacts on photochemical oxidant formation and particulate matter formation, mainly due to the elimination of pre-harvest burning practices. However, mechanical harvesting may increase the impacts on fossil depletion, ozone depletion, and terrestrial acidification resulting from higher use of fertilizer and diesel. Differences of impacts on freshwater eutrophication and human toxicity were not significant. In terms of climate change, the difference between two systems depended on the soil organic carbon sequestration scenario considered. When considering soil carbon increase at a 20-year time span, reduction of CO_2 emissions offset the contribution from higher use of diesel and fertilizers, and mechanical harvesting showed lower impacts on climate change. Whereas when not considering the contribution of soil carbon sequestration, the difference between manual and mechanical harvesting systems was small. At the end-point level, manual harvesting presented higher impacts on human health, but lower impacts on resources. The health effects of $PM_{2.5}$ vary considerably with population density. Changing from manual to mechanical harvesting close to urban areas leads to a drastic reduction of impacts, while for rural and remote areas, reductions are less important. When considering average population density, health effects of $PM_{2.5}$ of manual harvesting were approximately six times higher than mechanical

harvesting. Health effects of $PM_{2.5}$ calculated with ReCiPe are much lower and may underestimate the effects of primary $PM_{2.5}$ emissions.

Concerning public health benefits, the results of this article recommend to accelerate mechanization of sugarcane harvesting in areas with lower mechanization levels (i.e. north-northeast region in Brazil and some rough terrain areas), and manual harvesting should only be performed in fields located in rural or remote areas. To reduce environmental impacts, measures such as removing sugarcane residues on the fields before applying fertilizers to increase use efficiency and using biodiesel in agricultural machineries should be considered.

Acknowledgments

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Fig. 1

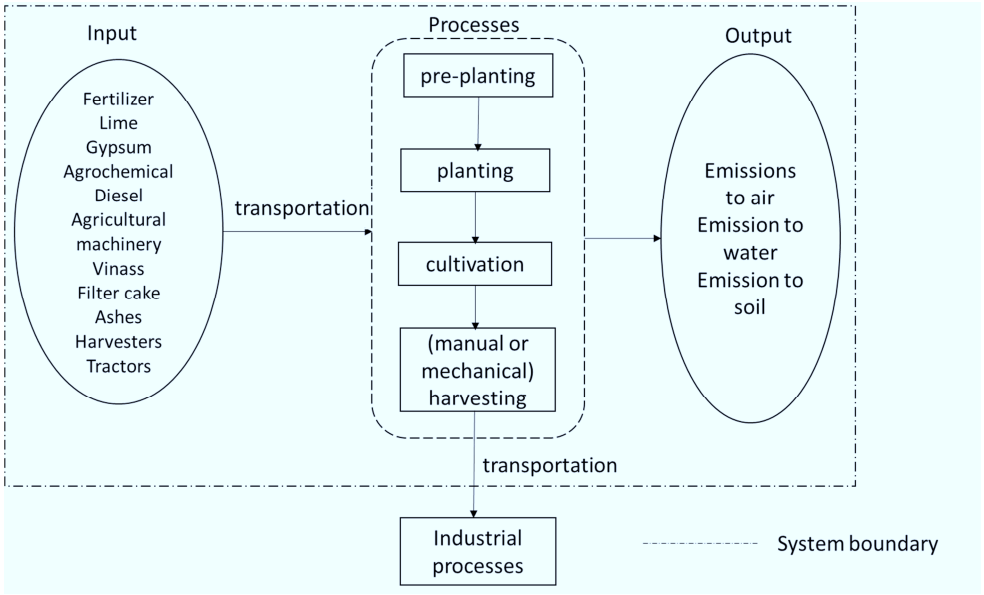

Input
Fertilizer
Lime
Gypsum
Agrochemical
Diesel
Agricultural machinery
Vinass
Filter cake
Ashes
Harvesters
Tractors
transportation
Processes
pre-planting
planting
cultivation
(manual or mechanical) harvesting
transportation
Industrial processes
Output
Emissions to air
Emission to water
Emission to soil
System boundary

Fig. 2

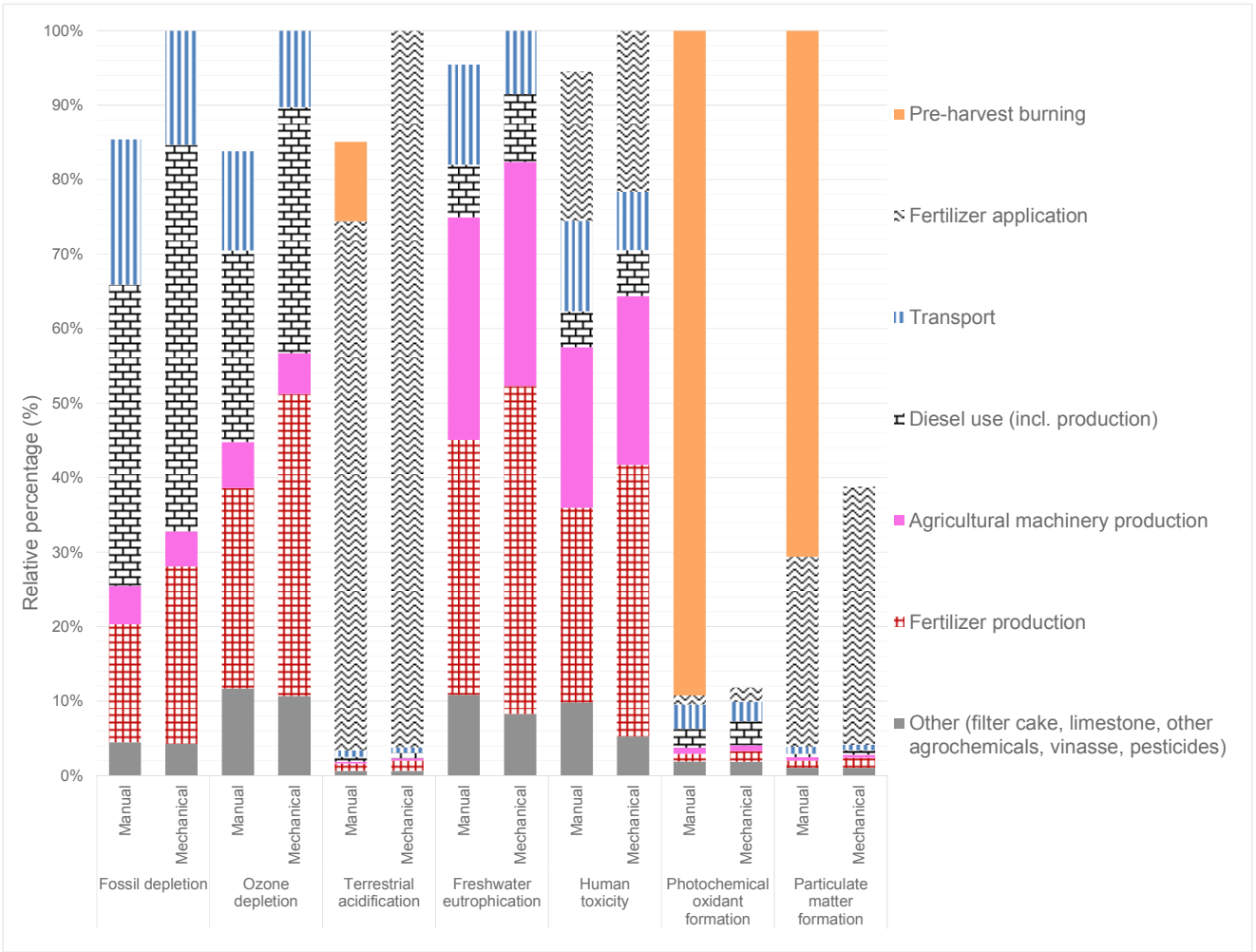

Relative percentage (%)
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
Manual
Mechanical
Fossil depletion
Ozone depletion
Terrestrial acidification
Freshwater eutrophication
Human toxicity
Photochemical oxidant formation
Particulate matter formation
Pre-harvest burning
Fertilizer application
Transport
Diesel use (incl. production)
Agricultural machinery production
Fertilizer production
Other (filter cake, limestone, other agrochemicals, vinasse, pesticides)

Fig. 3

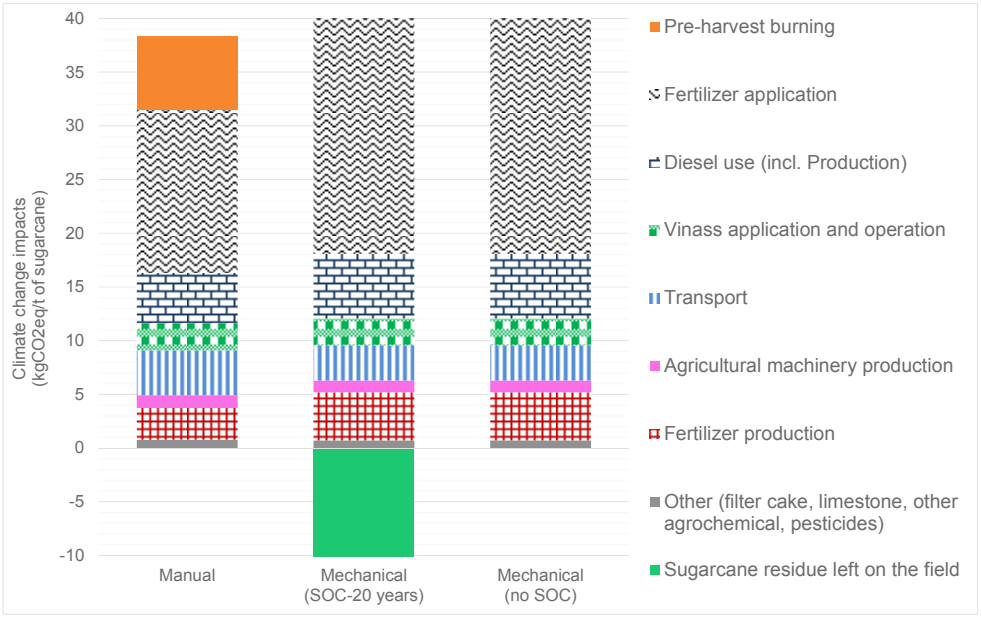

Climate change impacts (kgCO2eq/t of sugarcane)
40
35
30
25
20
15
10
5
0
-5
-10
Manual
Mechanical (SOC-20 years)
Mechanical (no SOC)
Pre-harvest burning
Fertilizer application
Diesel use (incl. Production)
Vinass application and operation
Transport
Agricultural machinery production
Fertilizer production
Other (filter cake, limestone, other agrochemical, pesticides)
Sugarcane residue left on the field

Fig. 4

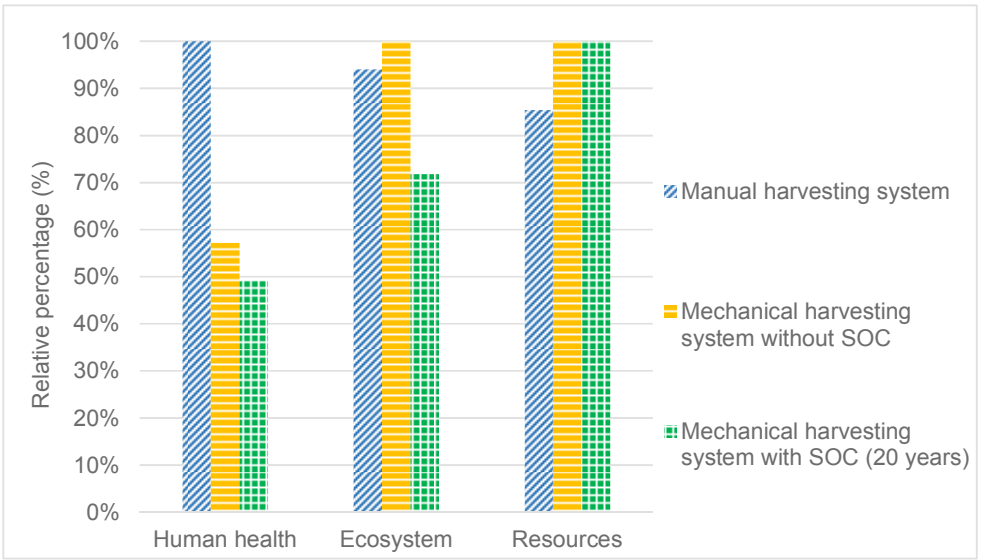
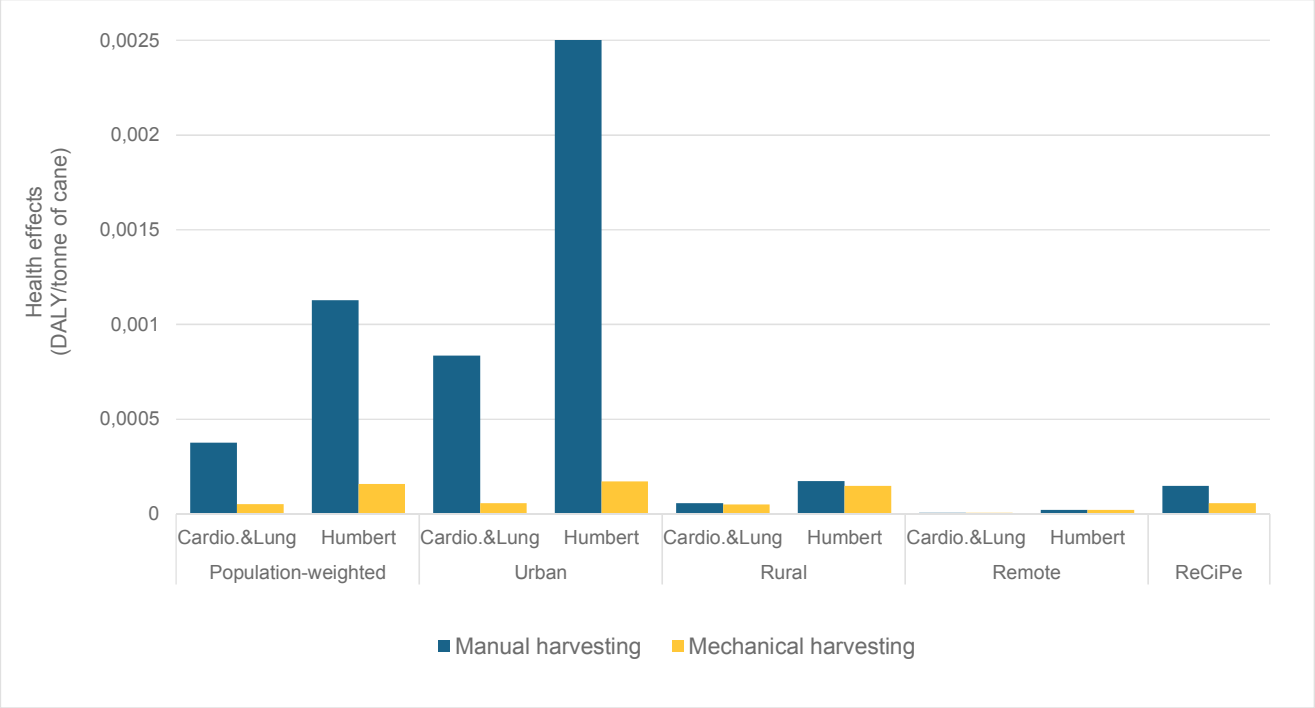

Relative percentage (%)
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
Human health
Ecosystem
Resources
Manual harvesting system
Mechanical harvesting system without SOC
Mechanical harvesting system with SOC (20 years)

Fig. 5



*Cardio.&Lung stands for health effects considering cardiopulmonary diseases and lung cancer

*Humbert stands for health effects considering a wider range of diseases based on Humbert et al. (2010)

Table 1 Intake fraction (iF) of primary $PM_{2.5}$ and secondary $PM_{2.5}$ (ppm - parts per million, representing mg PM inhaled per kg PM emitted) for Latin America calculated based on the recommended values and methods by Humbert et al. (2011)

Pollutant and stack height	Urban	Rural	Remote	Population-weighted average
Primary $PM_{2.5}$				
high-stack	13	0.48	0.1	5.6
low-stack	17	0.58	0.1	7.3
ground-level	49	1.1	0.1	20.7
emission-weighted average	29	0.75	0.1	12
Secondary $PM_{2.5}$				
SO_2	0.99	0.79	0.11	0.86
NO_x	0.2	0.17	0.02	0.18
NH_3	1.7	1.7	0.23	1.7

Table 2 Parameters and effect factors applied to calculate CFs considering cardiopulmonary diseases and lung cancer

	Cardiopulmonary				Lung cancer	Total
	IHD[a]	Stroke	COPD[b]	Total death	LC[c]	
Death[e] (thousands)	140.8	129.2	50.5	320.5	22.3	342.8
DALY[d, f] (thousands)	1427	1279	796	3502	223	3725
Severity factor (DALY/death)	10.1	9.9	15.8	10.9	10	10.9
Exposure-response factor[g] (death/kg $PM_{2.5}$ inhaled)				3.9	0.35	4.2
Effect factor (DALY/kg $PM_{2.5}$ inhaled)				42.6	3.5	45.6

[a]: Ischemic heart disease; [b]: Chronic obstructive pulmonary disease; [c]: Lung cancer; [d]: Disability adjusted life years; [e,f]: Data from WHO Global Burden of Disease 2004 statistics; [g]: (Gronlund et al., 2015)

Table 3 Two groups of CFs (DALY/kg $PM_{2.5}$ or secondary $PM_{2.5}$ precursors emitted) of $PM_{2.5}$

Pollutant and stack height	CFs considering cardiopulmonary diseases and lung cancer (Cardio.& Lung)				CFs considering a wider range of diseases (Humbert)			
	Urban	Rural	Remote	Population-weighted	Urban	Rural	Remote	Population-weighted
Primary $PM_{2.5}$								
High stack	5.93E-04	2.19E-05	4.56E-06	2.56E-04	1.78E-03	6.58E-05	1.37E-05	7.67E-04
Low stack	7.76E-04	2.65E-05	4.56E-06	3.33E-04	2.33E-03	7.95E-05	1.37E-05	1.00E-03
Ground level	2.24E-03	5.02E-05	4.56E-06	9.45E-04	6.71E-03	1.51E-04	1.37E-05	2.84E-03
Emission-weighted average	1.32E-03	3.42E-05	4.56E-06	5.48E-04	3.97E-03	1.03E-04	1.37E-05	1.64E-03
Secondary $PM_{2.5}$								
SO_2	4.52E-05	3.61E-05	5.02E-06	3.92E-05	1.36E-04	1.08E-04	1.51E-05	1.18E-04
NO_x	9.13E-06	7.76E-06	9.13E-07	8.21E-06	2.74E-05	2.33E-05	2.74E-06	2.47E-05
NH_3	7.76E-05	7.76E-05	1.05E-05	7.76E-05	2.33E-04	2.33E-04	3.15E-05	2.33E-04

Table 4 Mid-point Life Cycle Impact Assessment (per tonne of sugarcane)

Impact category	Unit	Manual harvesting product system (A)	Mechanical harvesting product system (B)	Relative Difference (Δ = (B-A)/A)
Fossil depletion	kg oil eq	6.66	7.80	+ 17.1%
Ozone depletion	kg CFC-11 eq	1.47E-06	1.75E-06	+ 19%
Terrestrial acidification	kg SO_2 eq	1.34	1.57	+ 17.2%
Freshwater eutrophication	kg P eq	1.90E-03	1.99E-03	+ 4.7%
Human toxicity	kg 1,4-DB eq	4.22	4.38	+ 3.8%
Photochemical oxidant formation	kg NMVOC	6.83E-01	8.05E-02	- 88.2%
Particulate matter formation	kg PM_{10} eq	5.76E-01	2.24E-01	- 61.1%

Online Resource

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Life cycle assessment addressing health effects of particulate matter of mechanical versus manual sugarcane harvesting in Brazil

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Appendix 1 Life-Cycle Inventory for two sugarcane production systems (per tonne of sugarcane), including cultivation, harvesting and transportation from field to industry

	Unit	Manual harvesting	Mechanical harvesting	
Inputs[a]				
Vinasse	m3	0.88	0.88	
Filter cake	kg	7.95	7.95	
Urea, as N	kg	0.74	1.18	
Single superphosphate, as P_2O_5	kg	0.15	0.16	
Potassium chloride, as K2O	kg	1.0	1.21	
Limestone	kg	4.84	4.94	
Gypsum	kg	2.42	2.47	
Pesticide unspecified	g	3.7	3.3	
Glyphosate	g	3.1	3.2	
Diuron	g	1.2	1.2	
Carbofuran	g	5.1	0	
Harvester, production	kg	0	0.055	
Tractor, production	kg	0.097	0.063	
Agricultural machinery, production	kg	0.15	0.12	
Diesel, farm operation	kg	1.22	1.57	
Diesel, sugarcane transportation[b]	kg	0.80	0.58	
Diesel, input transportation[c]	kg	0.10	0.11	
Diesel, vinasse transportation[b]	kg	0.20	0.20	
Emission to air[d]				
Dinitrogen monoxide	g	39.9	62.1	Fertilizer, residues (vinasse, filtercake, straw and roots) and soil amendments field emissions
Ammonia	g	454.2	615.1	
Nitrogen oxides	g	8.4	13.0	
CO2, fossil	kg	3.29	4.03	
VOC	kg	0.95	0	Straw burning (only apply to manual harvesting system)
Nitrogen oxides	kg	0.2	0	
$PM_{2.5}$	kg	0.35	0	
Sulfur oxides	kg	0.05	0	
Dinitrogen monoxide	kg	0.01	0	
Methane	kg	0.13	0	
Carbon monoxide, biogenic	kg	8.81	0	
Emissions to water (groundwater)[e]				
Nitrate	kg	1.67	2.64	Fertilizer and residues (vinasse, filtercake, straw and roots)
Emissions to water (river)[f]				
Carbofuran	g	0.076	0	Pesticides
Diuron	g	0.018	0.018	
Fiproni	g	0.007	0	
Glyphosate	g	0.047	0.048	
Hexazinone	g	0.005	0.005	
Pesticides, unspecified	g	0.025	0.026	
Tebuthiuron	g	0.018	0.019	
Emissions to soil[e,f]				
Zinc	g	0.26	0.26	Emissions of tire (machinery)
Lead	g	0.04	0.04	
Cadmium	g	0.01	0.01	
Cadmium	g	0.005	0.005	Fertilizers
Copper	g	0.09	0.1	
Zinc	g	0.31	0.36	
Lead	g	0.11	0.11	
Nickel	g	0.05	0.06	
Chromium	g	0.1	0.11	
Carbofuran	g	5.01	0	Pesticides
Diuron	g	1.16	1.19	
Fiproni	g	0.48	0	
Glyphosate	g	3.09	3.15	
Hexazinone	g	0.34	0.35	
Imazapic	g	1.67	1.7	
Tebuthiuron	g	1.19	1.22	

a. Inputs and outputs were based on data from Brazilian Bioethanol Science and Technology Laboratory (CTBE), which represents the average technology and agricultural operations in the centre-south of Brazil. (Bonomi et al. 2016)

b. For sugarcane and vinasse transportation, it was considered a transport distance of 25 km between field and mill.

c. Inputs included are seed cane, limestone, gypsum, fertilizers and agrochemicals, and filter cake.
d. Breakdowns of emission sources and assumptions can be found in Appendix 2.
e. It was assumed that 5% of the total nitrogen applied as urea or as ammonia leach to groundwater, being converted into nitrate. All the heavy metals contained in mineral fertilizers, limestone and gypsum were considered as emissions to soil (Trivelin and Franco 2011; Renouf et al 2010). No phosphorous or potassium is assumed to leach to groundwater because Brazilian soils have, in general, acid pH (CTBE 2012).
f. 98.5% of pesticides were considered to be emitted to agricultural soil and 1.5% to superficial water (Renouf et al 2010).

Appendix 2 Emission factors applied to calculate sugarcane field emissions

Emission	Unit	Emission factor
Emission to air		
Diesel combustion in agricultural machinery[a]		
Carbon dioxide, fossil	kg/kg diesel	3.14
Carbon monoxide, fossil	kg/kg diesel	1.14E-02
Dinitrogen monoxide	kg/kg diesel	1.20E-04
Heat, waste	MJ/kg diesel	45.58
Methane, fossil	kg/kg diesel	1.61E-04
Nickel	kg/kg diesel	7.05E-08
Fertilizer field emissions[b]		
Ammonia	kg/kg urea	3.60E-01
Nitrogen oxides	kg/kg urea	4.40E-03
Dinitrogen monoxide	kg/kg urea	2.10E-02
Carbon dioxide, fossil	kg/kg urea	1.58
Vinasse field emissions[c]		
Nitrogen oxides	kg/m^3 vinasse	3.00E-03
Dinitrogen monoxide	kg/m^3 vinasse	1.40E-02
Filter cake field emissions[c]		
Nitrogen oxides	kg/kg filter cake	5.05E-05
Dinitrogen monoxide	kg/kg filter cake	2.41E-04
Sugarcane straw[d]		
Nitrogen oxides	kg/t straw	2.70E-03
Dinitrogen monoxide	kg/t straw	1.27E-02
Sugarcane roots[d]		
Nitrogen oxides	kg/t roots	0.01
Dinitrogen monoxide	kg/t roots	0.05
Limestone[b]		
Carbon dioxide, fossil	kg/limestone	0.48
Sugarcane burning before manual harvesting[e]		
VOC	kg/t residues	7
Nitrogen oxides	kg/t residues	1.5
$PM_{2.5}$	kg/t residues	2.6
Sulfur oxides	kg/t residues	0.4
Dinitrogen monoxide	kg/t residues	0.1
Methane	kg/t residues	0.93
Carbon monoxide, biogenic	kg/t residues	65
Emission to water (groundwater)		
Fertilizer: urea[f]		
Nitrate	kg/kg of fertilizer	0.22
Emission to water (river)		
Pesticides[g]		
Carbofuran	g/g carbofuran	0.015
Diuron	g/g diuron	0.015
Fiproni	g/g fiproni	0.015
Glyphosate	g/g glyphosate	0.015
Hexazinone	g/g hexazinone	0.015
Imazapic	g/g imazapic	0.015
Tebuthiuron	g/g tebuthiuron	0.015
Emission to soil		
Agricultural machinery[a]		
Zink	g/kg tire	8.96
Lead	g/kg tire	1.456
Cadmium	g/kg tire	0.336
Pesticides[h]		
Carbofuran	g/g carbofuran	0.985
Diuron	g/ g diuron	0.985
Fiproni	g/g fiproni	0.985
Glyphosate	g/g glyphosate	0.985

Hexazinone	g/g hexazinone	0.985
Imazapic	g/g imazapic	0.985
Tebuthiuron	g/g tebuthiuron	0.985
Fertilizer field emissions: Urea[i]		
Cadmium	mg/kg of urea	0.034
Lead	mg/kg of urea	0.73
Nickel	mg/kg of urea	1.32
Copper	mg/kg of urea	3.96
Zinc	mg/kg of urea	29.04
Chromium	mg/kg of urea	1.32
Fertilizer field emissions: Single superphosphate[i]		
Cadmium	mg/kg of input	0.64
Lead	mg/kg of input	9.59
Nickel	mg/kg of input	3.23
Copper	mg/kg of input	2.43
Zinc	mg/kg of input	16.32
Chromium	mg/kg of input	4.63
Cadmium	mg/kg of input	0.64
Fertilizer field emissions: Potassium chloride[i]		
Cadmium	mg/kg of input	0.05
Lead	mg/kg of input	4.79
Nickel	mg/kg of input	1.83
Copper	mg/kg of input	4.35
Zinc	mg/kg of input	40
Chromium	mg/kg of input	1.74
Fertilizer field emissions: Limestone[i]		
Copper	mg/kg of input	6
Zinc	mg/kg of input	7
Chromium	mg/kg of input	9.9
Fertilizer field emissions: Gypsum[i]		
Cadmium	mg/kg of input	0.8
Lead	mg/kg of input	9.9
Nickel	mg/kg of input	4.9
Copper	mg/kg of input	10
Zinc	mg/kg of input	5
Chromium	mg/kg of input	9.9

a. Based on data from Nemececk et al. 2007, with updates to represent Brazilian conditions.

b. 30% of the total N applied as urea was considered to be emitted as ammonia, and 1% of the ammonia was converted as N_2O. 1% of the total N applied directly emitted as N_2O, and 0.75% of the nitrogen leached were assumed to be emitted as N_2O. All carbon content in urea and limestone is emitted as carbon dioxide (Costa 2003; Nemecek et al. 2007; IPCC 2006).

c. 1.225% of N in vinasse and filter cake is converted to direct and indirect N_2O emissions (IPCC 2006). Nitrogen content was assumed as 0.595 kgN/m^3 for vinasse and 12.5 kgN/tonne for filter cake (Macedo 2007; Chagas et al. 2016).

d. Direct and indirect N_2O emissions are considered based on the IPCC method (IPCC 2006). Nitrogen content in straw and roots is assumed as 4.77 gN/kg of sugarcane straw and 5.1 gN/kg of sugarcane roots (Smith et al. 2005; Hassuani 2005).

e. Emission factors for VOC and sulfur oxides are based on GREET (2009); and NO_x, $PM_{2.5}$, N_2O, CH_4, and CO (biogenic) are estimated based on França et al. (2012).

f. It is assumed that 5% of the nitrogen content in organic and inorganic fertilizers, and in sugarcane biomass residues leach to groundwater and are converted to nitrate (Trivelin and Franco 2011). No phosphorous or potassium is assumed to leach to groundwater because Brazilian soils have, in general, acid pH (CTBE 2012).

g. We considered that 1.5% of applied agrochemicals were emitted to surface water. The assumption is based on Renouf et al. (2010) on sugarcane production in Australia, and due to lack of data of specific pesticide emissions to water in Brazilian sugarcane systems, we adopted this value.

h. We assume that the remainder 98.5% of pesticides are emitted to soil. The degradation and absorption of pesticides are not considered.

i. Heavy metal contained in mineral fertilizers, limestone and gypsum were assumed to emit to soil. Emission factor for each substance is calculated based on the heavy metal content of Brazilian and imported agricultural products, considering the proportions of Brazilian and imported fertilizers in 2010 in Brazil (ANDA 2011; Gabe and Rodella 1999; Nemecek 2007).

Appendix 3 Calculation of soil carbon changes transferring from manual to mechanical harvesting

Location: São Paulo

Temperature zone: Tropical moist

Soil type: Low activity clay

SOC_{REF} = 47 tC/ha

Mechanical harvesting product system (with residues)

$F_{LU\_residue}$ = 0.48 (*long-term cultivated*) $F_{MG\_residue}$ = 1.15 (*reduced tillage*)

$F_{I\_resiude}$ = 1 (*medium input*)

According to IPCC *Equation 2.25* in calculating annual change in organic carbon stocks in mineral soils,

$$SOC_{residue} = 47 \times 0.48 \times 1.15 \times 1 = 25.94 \text{ tC/ha}$$

Manual harvesting product system (without residues)

$F_{LU\_noresidue}$ = 0.48 (*long-term cultivated*) $F_{MG\_noresidue}$ = 1 (*full tillage*)

$F_{I\_noresiude}$ = 0.92 (*low input*)

$SOC_{noresidue} = 47 \times 0.48 \times 1 \times 0.92 = 20.75$ tC/ha

Thus,

Soil carbon sequestrated = 5.19 tC/ha

Soil carbon sequestrated at a 20 year perspective:

5.19 tC ha^{-1}/20 year = 259.5 kgC $ha^{-1}yr^{-1}$

CO_2 reduction: 259.5 × 44 / 12 = 951.5 $kgCO_2$ $ha^{-1}yr^{-1}$

Per functional unit, avoided CO_2 is - 11.75 $kgCO_2$/t cane.

Appendix 4 Equations and values applied to calculate Intake Fraction

This study adopted the recommended values and method by Humbert et al. (2011) to calculate Intake Fraction. Recommended emission-weighted average iF for Latin America is applied to calculate the iF with respect to primary and secondary $PM_{2.5}$ under the conditions of different emission heights and population densities. Values for iF of primary $PM_{2.5}$ for Latin America are as follows: Urban – 29 ppm; Rural – 0.75 ppm; Remote – 0.1 ppm; and Population-weighted average – 12 ppm (ppm stands for parts per million, representing mg PM inhaled per kg PM emitted). These recommended values of iF are for unknown stack height emissions. In order to differentiate emission heights, following equations are applied,

$$iF_{high\text{-}stack} = iF_{unknown\text{-}stack} / (f_{e,\ high\text{-}stack} + Y \times f_{e,\ low\text{-}stack} + X \times Y \times f_{e,\ ground\text{-}level})$$

$$iF_{low\text{-}stack} = Y \times iF_{unknown\text{-}stack} / (f_{e,\ high\text{-}stack} + Y \times f_{e,\ low\text{-}stack} + X \times Y \times f_{e,\ ground\text{-}level})$$

$$iF_{ground\text{-}level} = X \times Y \times iF_{unknown\text{-}stack} / (f_{e,\ high\text{-}stack} + Y \times f_{e,\ low\text{-}stack} + X \times Y \times f_{e,\ ground\text{-}level})$$

where $f_{e,\ high\text{-}stack}$, $f_{e,\ low\text{-}stack}$ and $f_{e,\ ground\text{-}level}$ are the fractions of total emissions from high-stack (>100m), low-stack (>25m) and ground-level respectively. Values applied in this study is in consistent with Humbert et al. (2011), which is based on American conditions, with $f_{e,\ high\text{-}stack}$= 41%, $f_{e,\ low\text{-}stack}$ = 17%, $f_{e,\ ground\text{-}level}$ = 42%. X and Y are the intake fraction ratios of ground-level to low-stack and low-stack to high-stack emissions respectively. In the Humbert method, X and Y values from RiskPoll were applied, in which X equals to 1.9 for rural and 2.9 for urban conditions, and Y equals to 1.2 for rural and 1.3 for urban conditions. To calculate population-weighted average iF, the population fractions for urban, rural and remote conditions are assumed to be 41%, 57% and 2%. In terms of secondary $PM_{2.5}$, the Humbert method adopted the regressions of Greco et al. (2007) and Van Zelm et al. (2008). For secondary $PM_{2.5}$, stack height has limited importance in affecting iF. Equations applied to calculate secondary $PM_{2.5}$ are shown in the table below.

	Urban	Rural	Remote
SO_2	Based on Greco et al. (2007) with adjusting breathing rate to 13 m^3 $person^{-1}day^{-1}$, the values are equal to ones in Humbert et al. (2011).		= iF ($SO_{2\ rural}$) × (iF ($PM_{2.5\ remote}$) / iF($PM_{2.5\ rural}$))
NO_x			= iF ($NO_{x\ rural}$) × (iF ($PM_{2.5\ remote}$) / iF($PM_{2.5\ rural}$))
NH_3	Based on Van Zelm et al. (2008), $iF_{urban} = iF_{rural}$		= iF ($NH_{3\ rural}$) × (iF ($PM_{2.5\ remote}$) / iF($PM_{2.5\ rural}$))

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