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ABSTRACT

The greenhouse gas (GHG) balance of biofuels largely hinges on the magnitude of nitrous oxide (N_2O) emissions from arable soils during feedstock production, which are highly variable. Here, used an agro-ecosystem model to generate these emissions at a high resolution over the Ile-de-France region in Northern France, for a range of feedstocks. The emissions were input to a life-cycle assessment of candidate biofuel pathways: bioethanol from wheat and sugar-beet, biodiesel from oilseed rape, and ethanol from miscanthus.

Compared to the widely-used methodology based on fixed emission factors, ecosystem modelling lead to 55% to 70% lower estimates for N_2O emissions, emphasizing the importance of regional factors. The life-cycle GHG emissions of 1st generation biofuels were 50% to 70% lower than fossile-based equivalents, and 85% lower for cellulosic ethanol. Indirect land-use change effects negated these savings for bio-diesel and wheat ethanol, but were offset by direct effects for cellulosic ethanol.

Keywords: Biofuels, Energy crops, Greenhouse gas emissions, Land-use change, Life-cycle assessment

1. Introduction

Controversy is still ongoing regarding the greenhouse gas (GHG) intensity of biofuels and bioenergy chains in general (Kavanagh, 2006; Crutzen et al., 2008). While there appears a consensus on the benefits of displacing fossil fuels with energy from biomass, at least when excluding indirect land-use change (iLUC) effects (Farrell et al., 2006; Chum et al., 2011), the GHG savings reported in the literature may differ widely for seemingly similar pathways (Chum et al., 2011). These results are usually based on the life cycle assessment (LCA) methodology, relying on the same international standards but different calculation hypotheses and scope, in particular regarding co-product allocation methods, system boundaries, functional unit, or impact characterization. However, correcting for these differences to make these results commensurate only reduces part of their variability (Farrell et al., 2006). The source of input data used in the inventory step of the LCA plays a major role in its final outcome, in particular regarding crop management and biomass yields per unit area (Davis et al., 2013). Also, the emissions occurring upon the cultivation of energy crops in the field have a strong influence on the energy and GHG balances of the whole chain (Smeets et al., 2009). For instance, the efflux of nitrous oxide (N_2O) resulting from fertilizer application accounted for 20 to 40% of the GHG emissions of the 1st generation biofuels consumed in France (ADEME, 2010), and depending on the methodology used this efflux could vary by a factor of 2, according to the same study. Worldwide, Smeets et al. (2009) showed that this uncertainty, when compounded with N_2O emissions related to land-use changes could alter the life-cycle GHG emissions of biofuels by more than 50%, leading to higher emissions than their fossil counterparts in some cases.

In most LCAs of bioenergy chains, N_2O emissions from soils are estimated with fixed emission factors (EFs), expressing a proportionality between N_2O efflux and fertilizer N input

rates (Cherubini, 2010). However, these factors carry a large uncertainty. For instance on a global scale, the EF for direct emissions of N_2O related to fertilizer N inputs to cropland varies between 0.3% and 3%, with a median value of 1% (Eggleston et al., 2006). This worldwide default value were recently questioned based on the gap between bottom-up inventories of fertilizer-derived field emissions and the atmospheric build-up of N_2O (Crutzen et al., 2008). The higer EFs proposed by the latter authors would negate the GHG benefits of most bioenergy pathways, including 2nd generation biofuels from lignocellulose. It thus appears crucial to obtain more reliable assessments of the level of N_2O emissions attributable to bioenergy feedstock production (Cherubini, 2010).

Emissions of N_2O from soils are difficult to assess because they vary widely across time and space, depending on environmental conditions (soil properties and climate) and agronomic characteristics such as crop yields and management, and fertilizer use efficiency (Stehfest and Bouwman, 2006). Improving their estimation implies taking these drivers explicitly into account, and applying LCA at the supply area level rather than based on notional country- or European-wide 'average' cultivation fields (Gabrielle and Gagnaire, 2008). This stresses the need for developing generic methods with a capacity to address the variability within these feedstock supply areas. Combining experimental monitoring of field-scale emissions with biophysical models appears as a promising avenue to provide reliable estimates of N_2O emissions at a scale relevant for bioenergy units (Dufossé et al., 2013). However, there has been thus far very little work on the comparison of emission maps obtained with such bottom-up estimates with top-down, integrative atmospheric measurements, such as tower fluxes. Most top-down source inversion studies from atmospheric measurements have been carried out at large scales, from continental to global, with a coarse resolution of terrestrial sources of N_2O (Thompson et al., 2011).

The objectives of this paper were i/ to improve the estimations of field emissions of bioenergy feedstocks, in particular nitrous oxide, by using ecosystem modelling at regional scale, and ii/ to examine the impact of using such estimates in lieu of fixed emission factors in the LCA of current and prospective biofuel chains for the area. The modelling also made it possible to take into accounts the effects of land-use-change scenarios on the ouctome of the LCAs. Here, we thus generated high-resolution maps of field emissions from in the Ile de France region (Northern France), using an agro-ecosystem model, for use in the LCA of representative biofuel chains for the area. The Ile de France region was selected because it was within the catchment probed by the atmospheric measurements, and is also included a set of field trials in Grignon (to the West of the domain) in which all the crops considered had been tested (Gabrielle et al., 2012; Goglio et al., 2013). The maps of N_2O emissions of were verified by using them as prior estimates in the inversion of the meso-scale atmospheric-chemistry transport model CHIMERE, against concentrations recorded at various heights in the planetary boundary layer at 2 sites to the South of Ile de France, for the year 2007 (Gabrielle et al., 2012). The maps were subsequently expanded to test various feedstocks using a series of near-term future weather data to capture the effect of inter-annual variability and the long-term effects of perennial crops over their growing cycle (typically 20 years). The resulting fluxes were input into the life-cycle inventory of the following chains: biodiesel production from oilseed rape; bioethanol production from winter wheat and sugar-beet (first-generation biofuels); and bioethanol from *Miscanthus x giganteus* (a perennial C4 grass, further referred to as miscanthus), representing a prospective second-generation pathway. The effect of feedstock production on soil C balances and dynamics were included in the LCA, relative to a reference land-use scenario in the absence of bioenergy production in Ile de France, thus account for direct LUC effects. Indirect LUC was taken into account based on the meta-analysis of De Cara et al. (2012).

2. Materials and Methods

2.1 Biofuel feedstocks and pathways

The biofuel value-chains considered included: bio-diesel from oilseed rape, ethanol from sugar-beet and winter wheat, and cellulosic ethanol obtained by enzymatic hydrolysis from miscanthus biomass. The baseline for the life-cycle inventory data was taken from a recent study carried out by the French Environment Agency on biofuels and their fossile equivalents in the context of France (ADEME, 2010). System boundaries included feedstock production to a regional storage after conversion in a biorefinery, but included CO_2 emissions upon combustion for fossile fuels.

The selected feedstock supply area was Ile de France, for which comparisons between ecosystem modelling and top-down atmospheric measurements were available to infer N_2O fluxes. Simple assumptions were made for direct land-use changes (LUC) related to feedstock production. A recent study based on historical land-use data (In Numeri, 2012) concluded that in France 1st generation biofuel crops mostly diverted existing crops from food to energy end-uses, or displaced similar crops (eg, oilseed rape substituted pea crops). Only 13% of the cropland area used for biofuel production was taken on set-aside land, and this occurred mostly in regions outside of Ile de France. Therefore, direct LUC effects were neglected for arable crops. For the perennial lignocellulosic crop (miscanthus), the assumption was that the latter either displaced annual crops or was established on set-aside land. The associated direct LUC effects were simulated by considering the differences between the emissions occurring on the modelled miscanthus fields and those occurring in either the reference rotation of arable crops or a continuous fallow. Compared to rotational fallow, the latter represented 40% of the overall set-aside area in 2007, and were considered as a reference, conservative scenario for conversion of set-aside cropland to miscanthus. For lack of an adequate

simulation model, continuous fallow was assumed to have the same C and N dynamics as the miscanthus fields.

Indirect land-use changes at the global scale (ie the cascading effects related to the diversion of food crops to fuel purposes) were accounted for in the GHG balance of biofuels based on the median values reported by De Cara et al. (2012) in their meta-analysis of the literature on this topic. The analysis accounts for biofuel type, feedstock type and production area, and we used the set of values that were the most relevant to the biofuel chains evaluated here.

2.2.2. Model simulations

Crop yields, soil C dynamics and emissions of reactive N (Nr), including N_2O in particular were simulated with the agro-ecosystem model CERES-EGC (Goglio et al., 2013) over the Ile de France region. Briefly, CERES-EGC was adapted from the CERES family of soil-crop models, with a focus on the simulation of environmental outputs such as nitrate leaching and gaseous emissions of ammonia and nitrogen oxides. It contains submodels for the major processes governing cycles of water, carbon and nitrogen in agro-ecosystem models. A physical module simulates the transfer of heat, water and nitrates down the soil profile as well as soil evaporation, plant water uptake, and transpiration in relation to climatic conditions. A microbiological module simulates the turnover of organic matter in the plough layer, involving both mineralization and immobilization of mineral N (denitrification and nitrification). CERES-EGC includes a submodel that simulates the production of NO and N_2O through the nitrification and denitrification pathways. The model was adapted to miscanthus by explicitly introducing a rhizome pool and biomass translocation processes specific to perennial crops from one growing season to the next (Picoche, 2012).

Ile de France may be approximated as a 150 km x 150 km square area surrounding Paris, with 55 % cropland. A GIS database was constructed with available geo-referenced data on this

region, including administrative borders, land-cover type, crop management practices, soil properties and climate. The corresponding layers of spatial information were mostly in vector format, and overlaid to delineate elementary spatial units representing unique combinations of soil types, weather data, and agricultural management. These units were subsequently used in the CERES-EGC simulations at the field-scale, in a bottom-up approach to map the emissions, using the procedure detailed by Dufosssé et al. (2013).

Since miscanthus is a perennial crop with a lifespan of up to 20 years, it was necessary to run long-term simulations, and also to capture inter-annual variability effects and soil organic C dynamics. We thus used weather data predicted for the 2010-2030 time slice by the DRIAS project in France (Lémond et al., 2011), using the IPSL-CM4 model with the A1B GHG emission scenario from IPCC, which appeared as an intermediate scenario for air temperature and rainfall among the range of models and forcings tested by this project. Compared to the 1961-1990 period, air temperature would rise by 1.35 °C, with a average of 11.7 °C for the 2010-2031 time slice, and rainfall would remain level at an average of 641 mm yr^{-1}. CERES-EGC was run for the current land-use (2010) and the various feedstocks and land-uses hypothesized for biofuel production. For the verification year (2007), we used gridded weather data from the MCRU data base (Chen et al., 2009), with a 0.25° x 0.25° resolution, and cropland areas from the last available survey (pertaining to the year 2000).

An alternative, reference methodology for N_2O emissions was based on the 2006 IPCC guidelines (Eggleston et al., 2006), as implemented in the baseline LCA (ADEME, 2010), using the Tier 1 approach (ie default emission factors). Indirect emissions of N_2O due to leaching were calculated as 0.75 % of the nitrate losses, simulated either with CERES-EGC or expert knowledge (ADEME, 2010), while N_2O emissions due to emissions of ammonia and nitric oxide were calculated as 1% of the latter fluxes, as simulated by CERES-EGC. In the ADEME methodology, nitrate leaching was given the same value for all crops except winter

oilseed rape, for which specific field references were available. Regarding emissions related to the N content of crop residues, average values were derived from expert knowledge and field surveys, to which a 1% emission factor was applied, in accordance with Eggleston et al. (2006).

2.2 3. Life-cycle inventories and impact characterization

Table 1 summarizes the sources of data used for the various steps of the biofuel chains. Crop management data were taken from surveys carried out by the French Ministry of Agriculture in 2006 for arable crops (Gabrielle et al., 2012), and expert knowledge for miscanthus in Northern France (Bessou, 2009; Table 2). The main agricultural inputs are listed in Table 3. Miscanthus was considered unfertilized, but the N exports in harvested biomass were compensated for by adding a corresponding consumption of synthetic fertilizer N to the feedstock production system. Grain yields were obtained from regional simulations by CERES-EGC for arable crops, while miscanthus yields were calculated as 90% of the biomass simulated in late winter, correspond to a late harvest (Strullu et al., 2011). The remaining 10% was returned to the soil as harvest residues.

Direct emissions of reactive N (Nr), whether gaseous or leaching, were simulated at regional scale with CERES-EGC over the 2010-2030 time period, along with C and water balances. The predicted Nr losses included: nitrate leaching, emissions of ammonia, nitric oxide and nitrous oxide. Annual arable crops were included in the single following rotation, representative of current crop successions in the area: oilseed rape – winter wheat – sugar-beet - winter wheat - winter barley. For each crop in the rotation, daily simulated fluxes were accumulated from its sowing to the sowing of the following crop. Miscanthus was simulated as a single cycle from planting in 2010 to removal or re-establishment in 2030. Emission fluxes were averaged over the various cropping periods and expressed on a yearly basis for all

crops. Fluxes were spatially averaged on the basis of the land area occupied by each crop or fallow, and a 5%-95% inter-quartile range was calculated from the distribution of emissions per ha in the relevant spatial simulation units, to derive a confidence interval in the LCA.

Data on biomass conversion to biofuels were taken from various sources relevant to France or Europe (Table 1). Feed co-products of 1st generation biofuels were handled by system expansion using data from Lehuger et al. (2009) on soybean meal from Brazil, as a substitute for either rapeseed meal or dried distillers grains with solubles (DDGS) from wheat, and silage maize, as a substitute for sugar-beet pulp. Other co-products were dealt with by economic allocation (Jungbluth et al., 2007).

Rape methyl-ester (or bio-diesel) was produced from the esterification of rapeseed oil with fossile-based methanol, involving glycerin and potassium sulfate as co-products. Data for the conversion of sugar-beet to ethanol were taken from 2 plants located in the Champagne-Ardennes and Picardy regions about 200 kms to the North of Ile de France, considered representative of current best technologies (Bessou, 2009). The conversion of wheat to ethanol involved dry-milling of wheat grains, saccharification and fermentation, and distillation to produce hydrated ethanol (95% basis). The production of the DDGS co-product from stillage included draff separation, concentration, drying and pelletization. The conversion of lignocellulose to ethanol involved steam pre-treatment of biomass, simultaneous saccharification and fermentation of the resulting mash, and distillation to hydrated ethanol. The vinasses produced by the fermentation process were digested to produce biogas and process heat and power. The data originally applied to a pilot plant using grass feedstock but we considered the latter could be replaced by miscanthus biomass due to the relative close chemical compositions of both feedstocks. In addition to ethanol, the process produced proteins and fibres and an economic allocation was made between the 3

end-products (Jungbluth et al., 2007).

Environmental impacts were characterized using the CML 2000 at mid-point level (Guinée et al., 2002). The following impact categories were analyzed: eutrophication, acidification, photochemical ozone formation, global warming, and depletion of abiotic resources. The sensitivity of the global warming indicators to the spatial and temporal variations of N_2O emissions in Ile de France was examined by calculating the GHG emissions of biofuels with the median and 5-95% inter-quartile range of these fluxes. LCA calculations were carried out with the Sima Pro software package (v7.1, Pré Consultants, Amersfoort, NL).

3. Results & Discussion

3.1 Field emissions of reactive N and soil C variations

Table 3 lists the field emissions of reactive N (Nr) and soil organic C (SOC) variations for the various feedstocks obtained with the ecosystem model CERES-EGC. Crop yields predicted in 2007, the reference year for the verification of N_2O fluxes (Gabrielle et al., 2012) are compared to statistics reported by the Ministry of Agriculture for the same year. Modelled and observed yields differed by less than for 5% for winter wheat and oilseed rape, but CERES-EGC over-estimated the yields of sugar-beet by 22%. However, the sugar-beet yields averaged over the 2010-2030 simulations were closer to the values reported in 2007 (87 t fresh tuber weight (FW) ha^{-1}vs. 82 t FW ha^{-1}), which mitigates the possibility of a model bias for sugar-beet. For comparison, sugar-beet yields ranged between 85 and 100 t FW ha^{-1} over the 2007-2012 period according to the regional agricultural statistics. For winter wheat, the 2010-2030 yields predicted by CERES-EGC were similar to the 2007 value (at 7.53 vs. 7.87 t grain dry matter (DM) ha^{-1}), while they were 30% higher for oilseed rape. This increase for rapeseed is 3 times larger than the projections made by a set of agro-ecosystem models for 2030 in one site in Ile de France (Brisson and Levrault, 2011), while for wheat yields the

latter predicted a 1 t DM ha^{-1} increase. These differences with our may due to the fact that no beneficial effects of rising CO_2 concentrations were included in the photosynthesis component of CERES-EGC for either crop, and to differences in the soil properties between our regional map and the particular site modelled by Brisson and Levrault (2011).

According to CERES-EGC, the highest direct emissions of N_2O occurred with sugar-beet (Table 2), due to its being a spring crop with fertilizer applications later into spring than the winter-sown crops (wheat and oilseed rape), when soil conditions, in particular its temperature are more conducive to denitrification. The high rate of N returned as crop residues by sugar-beet also explains these large fluxes. The emissions of miscanthus were 2- to 5-fold lower than those of the annual crops, in agreement with Drewer et al. (2012), while winter crops had intermediate emission levels. Except for sugar-beet, the direct emissions of N_2O simulated by CERES-EGC were 2- to 4-fold lower than output by the ADEME methodology. Such pattern was already noted in the verification of 2007 over France, for which the atmospheric inversion suggested emission factors (EF) half lower than the default 1% value of the IPCC guidelines for cropland (Gabrielle et al., 2012). Deviations between modelled and IPCC Tier 1 estimates for N_2O have been reported in both directions, depending on local climatic and management conditions. An EF of 0.80% was derived for cropland in China with the DNDC model (Li et al., 2001), while Leip et al. (2011) reported values similar to the IPCC Tier 1 range in Europe, with values around the 1% median in France over the 1990-2000 time slice. Here, the annual amounts and seasonal distribution of rainfall were similar between the current and near-future climates in Ile de France, which should not have altered the denitrification patterns. Therefore, the low emission levels simulated over 2010-2030 in Ile de France compared to 2007 or other estimates were probably not due to changes in climate forcing.

Indirect emissions of N_2O due to leaching or gaseous losses of Nr, as well as the fate of crop

281 residues, differed even more markedly between the 2 estimation methodologies (Table 4). The
282 overall ranges across crops were larger with the ADEME than with the model-based
283 methodology: 0.71 – 2.80 kg N_2O-N ha^{-1} yr^{-1} vs. 0.30-1.78 kg N_2O-N ha^{-1} yr^{-1}, respectively.
284 Most of the variations arose from the contribution of crop residues, which was included in the
285 direct emissions estimated with CERES-EGC (since the model simulates the fate of harvest
286 residues), while this term reached up to 1.32 kg N_2O-N ha^{-1} yr^{-1} for sugar-beet due to the large
287 N content of the green leaves returned to soils at harvest. This was also captures in the model
288 simulations, as mentioned above. Indirect emissions due to nitrate leaching were similar
289 across the 2 approaches, with the exception of oilseed rape for which the model simulated
290 extremely low nitrate losses.

291 As a consequence, the ranking of crops varied according to the N_2O flux estimation methods:
292 according to ADEME, annual crops had very similar emission rates, around 2.8 kg N_2O-N ha^{-1}
293 yr^{-1}, while miscanthus emitted 4 times less. Conversely, the modelled efflux from oilseed rape
294 was close to that of miscanthus (less than 0.5 kg N_2O-N ha^{-1} yr^{-1}), while the other 2 annual
295 crops emitted about 3 times more N_2O, with sugar-beet in the upper end. The same pattern
296 applied to nitrate leaching rates, with winter wheat resulting in the highest losses (at 37 kg
297 NO_3-N ha^{-1} yr^{-1}). The leaching rate from wheat was close to the 40 kg NO_3-N ha^{-1} yr^{-1} estimate
298 given by ADEME (2010) as a flat rate for all annual crops except sugar-beet (with a value of
299 18 kg NO_3-N ha^{-1} yr^{-1} derived from lysimeter studies). The low leaching rates (under 10 kg
300 NO_3-N ha^{-1} yr^{-1}) simulated by CERES-EGC with winter oilseed rape arose from the capacity
301 of this crop to take up N in fall. Similarly, the sowing of catch crops or the regeneration of
302 volunteers, in particular before the sowing of the sugar-beet crop could have reduced the
303 losses of nitrate overwinter several-fold (McDonald et al., 2005).

304 Gaseous emissions of NO were in the in the 0.4 – 1.0 kg NO-N ha^{-1} yr^{-1} range for annual crops
305 and twice lower for miscanthus (Table 3). Similarly to N_2O, these values are under the 1-2 kg
25

NO-N ha^{-1} yr^{-1} range reported by Stehfest and Bouwman (2006) for cropland in Ile de France, using regionalized emission factors and a large data set of field observations. However, in previous tests against field data in the same area, the CERES-EGC model did not reveal any systematic bias in the prediction of NO emissions (Rolland et al., 2008). Regarding ammonia (NH_3), the model simulated net emissions in the 2.3 – 5.1 kg NH_3-N ha^{-1} yr^{-1} range for annual crops, and a very small net deposition rate (-0.2 kg NH_3-N ha^{-1} yr^{-1}) for miscanthus. Overall, simulated gaseous losses of ammonia and nitric oxide were several-fold lower than the IPCC Tier 1 estimate, making up 10% of the fertilizer N inputs (for synthetic fertilizers). However, the modelled estimates are in line with the lower end of this emission factor, at 3% (Eggleston et al., 2006).

While there were no significant trends in terms of soil organic carbon (SOC) storage for annual crops, with rates in the -34 to +41 kg C ha^{-1} yr^{-1} range, miscanthus had a large capacity to sequester C, with rates around 575 kg C ha^{-1} yr^{-1}. The latter is in line with multiple long-term studies on miscanthus stands and the 680 kg C ha^{-1} yr^{-1} value reported in the review of Don et al. (2011) under stands more than 10 years old. This increase mainly reflected a build-up of soil organic matter due to large rates of litter inputs, combined with the protective effect of reduced tillage.

3.2 Life-cycle impacts of feedstock production

Figure 1 compares the impacts of the various candidate energy crops on a hectare (ha) and MJ of feedstock energy content basis. Direct land-use change effects were excluded for miscanthus in this analysis. The annual food crops (wheat, oilseed rape and sugar-beet) had the highest impacts per ha due to their high requirements in agricultural inputs compared to miscanthus, resulting in 60% to 85% lower impacts for the latter. Such pattern was noted by

330 Monti et al. (2009) in Northern Italy, although to a lower extent, with 50% to 60% lower LCA
331 impacts with miscanthus compared to a wheat-maize rotation on a ha basis.

332 The ranking among the 3 annual crops varied according to the indicator considered. The
333 abiotic depletion (AD) and ozone formation potential (OFD) indicators were within a 10%
334 relative range for all crops, while winter wheat and oilseed rape had 15% lower global
335 warming potentials (GWP) compared to sugar-beet. Oilseed rape had 25% and 60% lower
336 impacts than the other 2 annual crops due to its much low nitrate leaching rates in the field.
337 Overall, most of the differences in LCA impacts on a ha basis arose from the variations in Nr
338 emission patterns across feedstocks.

339 Differences across crops were less pronounced when using one MJ of feedstock energy
340 content (including grains and straw for annual crops) as a functional unit (Figure 1). The
341 highest impacts generally occurred with winter wheat, except for the OFP which was slightly
342 larger with winter wheat. Sugar-beet had overall lower impacts than oilseed rape and wheat,
343 due its 40% higher energy yield per ha (Table 3). Similarly to the results per ha, the impacts of
344 miscanthus per MJ were 80 to 98% lower than the annual crops, given that its energy output
345 per ha was in the upper range of the crops. Overall, crops with a high energy output per ha
346 (such as sugar beet and miscanthus) had comparably lower impacts per MJ than the others,
347 showing that energy yields predominated in the trade-off between high productivity, fertilizer
348 requirements and N_2O emission rates. Miscanthus performed best because it minimzed this
349 trade-off. The compensation of N exports by synthetic fertilizers accounted for 10 to 50% of
350 the LCA impacts. Many miscanthus fields in France or the UK are currently unfertilized but is
351 likely to lead to the depletion of soil nutrients in the long-run (Strullu et al., 2011). Prior land
352 use (cropland versus fallow) played a negligible role, bearing in mind that the effects on soil
353 C dynamics were not included at this stage and thus the differences between the two scenarios
354 only reflected variations in terms of soil distribution over Ile de France.
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356 *3.3 Life-cycle impacts of biofuel chains*

357 Figure 2 reports the GHG emissions of the various biofuel chains investigated here, compared
358 to their fossile equivalents. When excluding land-use change effects (ie on an attributionnal
359 basis), all biofuel chains emitted less GHG than the fossile fuels, with savings ranging from
360 50% (ethanol from winter wheat) to 80% (cellulosic ethanol). For the 1st generation biofuels,
361 the figures based on modelled field emissions (referred to as CERES) were slightly lower than
362 those based on default emission factors (ADEME). The lower field N_2O emission rates with
363 the former methodology were in a large part compensated for by the differences in co-product
364 handling: system expansion with CERES vs. energy-based allocations for ADEME. The latter
365 generally results in a lower environmental burden on biofuels than the former (Cherubini,
366 2010). The GHG emissions of miscanthus-based ethanol were only 60% lower than those of
367 sugar-beet ethanol (the 1st generation chain with the lowest emissions), and 75% lower than
368 the other biofuels. This gap is larger than with the feedstock data (Figure 1) because of the
369 relatively high biomass-to-ethanol yield hypothesized in the conversion process due to the
370 economic allocation with the other end-products. This yield is around 0.60 kg ethanol kg^{-1}
371 feedstock DM, higher than the theoretical yield and the 0.24 value proposed by a recent blueprint
372 design for corn stover (Humbird et al., 2011). Thus, changing the allocation system and process design
373 with fewer co-products would probably have resulted in larger impacts for miscanthus ethanol.
374 The GHG balances presented here fell in the middle or lower end of the ranges given by
375 Chum et al. (2011) worldwide: 22-44 g CO_2 eq./MJ for sugar-beet ethanol, 16-70 eq./MJ for
376 wheat ethanol, 5-75 g CO_2 eq./MJ for bio-diesel from plant oils, and -7 to 47 g CO_2 eq./MJ for
377 cellulosic ethanol. Aside from the low Nr emission rates specific to the CERES methodology,
378 this pattern is probably due to the use of recent technologies for conversion (representative of
379 the last decade), and biomass yields higher than world averages for arable crops.
31

Accounting for direct LUC effects for miscanthus grown on cropland offset most of the life-cycle emissions due to the high capacity of this perennial grass to sequester C with respect to arable crops (Table 2), resulting in a net value close to the bottom end of the range from Chum et al. (2011). The inclusion of ecosystem C pools in LCA and in particular the effects of land-use and management practices on soil organic C dynamics has been shown to exert a large influence on the GHG abatements of bioenergy. A case-study for miscanthus in the UK concluded to a potential for negative emissions under -40 g CO_2 eq. MJ^{-1} (Davis et al., 2013). Including indirect LUC effects dramatically altered the GHG savings of all biofuels, including cellulosic ethanol from feedstock grown on cropland (Figure 2). It resulted in negative values for bio-diesel and wheat ethanol, and divided the savings by 2 for sugar-beet and miscanthus-based ethanol grown on cropland, compared to attributional values. Such patterns are common in the literature on consequential LCA, whether based on macro-economic sectoral modelling or simpler substitution and displacement schemes (De Cara et al., 2012; Silalertruksa et al., 2009). For cassava-based ethanol in Thailand, the latter authors cited LUC-related GHG emissions in the 20% to 90% range compared to attributional values, depending on the crops displaced and crop production regions.

The uncertainty on N_2O emissions due to their temporal and spatial variability in Ile de France resulted in asymetric confidence intervals (5%-95%) for the net GHG balance of biofuels, with a magnitude ranging from 2 to 25 g CO_2 eq./MJ. This value was largest with wheat ethanol and bio-diesel. The fact that the confidence intervals of these 2 biofuels overlapped indicate that their net GHG emissions were not significantly different. Conversely, the differences between cellulosic ethanol, sugar-beet ethanol and the other 2 biofuels were significant ($p>0.9$). In their uncertainty analyses on the influence of N_2O emissions on the GHG balances of biofuels, Smeets et al. (2009) reported much larger confidence intervals, similar to life-cycle totals for ethanol from wheat or bio-diesel from oilseed rape. This arose

405 from the large uncertainties on field emissions (ranging from 1 to 4 kg kg N_2O-N ha^{-1} yr^{-1} for
406 winter wheat in Europe), and emphasizes the benefits of regional modelling to reduce the
407 magnitude of this uncertainty.
408 Figure 3 shows the LCA results of biofuel chains for a broader range of impact categories. As
409 could be expected from the feedstock comparison, miscanthus ethanol had the lowest impacts,
410 whether established on cropland or fallow. Compared to gasoline, cellulosic ethanol had an 8-
411 fold lower acidification potential but a 5-fold larger impact for eutrophication (ADEME,
412 2010). The LCA results based on ecosystem modelling (CERES) were generally lower than
413 those obtained with the fixed emission factors (ADEME), by a factor of up to ten. These gaps
414 were much larger than for the GHG emissions because feedstock production was the top
415 contributor to these impacts, and the relative differences between ADEME and CERES were
416 in a similar range for Nr emissions (Table 3). Differences between 1st generation biofuels were
417 more pronounced with the ADEME methodology.

418 Overall, sugar-beet ethanol performed better than wheat ethanol and bio-diesel, except for
419 eutrophication for which bio-diesel and miscanthus ethanol were on a par according to the
420 CERES estimation, due to similar nitrate leaching rates. Only a few published LCAs of
421 biofuels include impacts other than GHG emissions or fossile energy consumption. They
422 generally conclude to the same pattern as evidenced here, with short-range impacts such as
423 eutrophication, ozone formation or acidification potentials being larger with biofuels than
424 their fossile counterparts due to Nr emissions upon feedstock cultivation (Gabrielle and
425 Gagnaire, 2008). However, in the case of lignocellulosic plants such as miscanthus, co-
426 benefits in terms of control soil erosion or nutrients filtering have also been pointed out
427 (Chum et al., 2011). At the field- and landscape scales, management practices such as cover
428 crops and buffer zones may thus be introduced to mitigate these local impacts and optimize
429 biofuel chains (Davis et al., 2013; Bessou, 2009). These options may be investigated using the
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regional modelling systems presented here at the plot-scale, and with the use of integrated landscape models (Dufossé et al., 2013).

Overall, sugar-beet ethanol thus emerged as the 1st generation biofuel with the best performance due to its high energy output per unit area, and miscanthus ethanol was still more efficient at abating GHG emissions while minimizing local impacts. Establishing miscanthus on fallow land proved the best option, but is limited in practice by the availability of fallow land in Ile de France (which only makes up 4% of the utilizable arable area in the region). It should also be borne in mind that the data on miscanthus conversion to ethanol is based on an extrapolation of laboratory-scale processes, thus still fraught with a large uncertainty and more prospective in nature. Its results should therefore be considered as indicative rather than representative of a commercial plant. Results from currently-operating pilots should help improve life-cycle inventory data for this process in a near future.

4. Conclusion

We assessed the environmental impacts of several biofuel feedstocks and pathways at the regional scale, based on either annual or perennial crops. The use of an ecosystem model to estimate for the emissions of reactive N, as opposed to fixed emission factors, lead to a 5% to 15% reduction in the overall GHG emissions of biofuels, due to local soil and climatic characteristics. The GHG savings incurred by the substitution of fossile fuels with biofuels hinged on land-use change scenarios and biofuel pathways, being highest for cellulosic ethanol and lowest for bio-diesel from oilseed rape.

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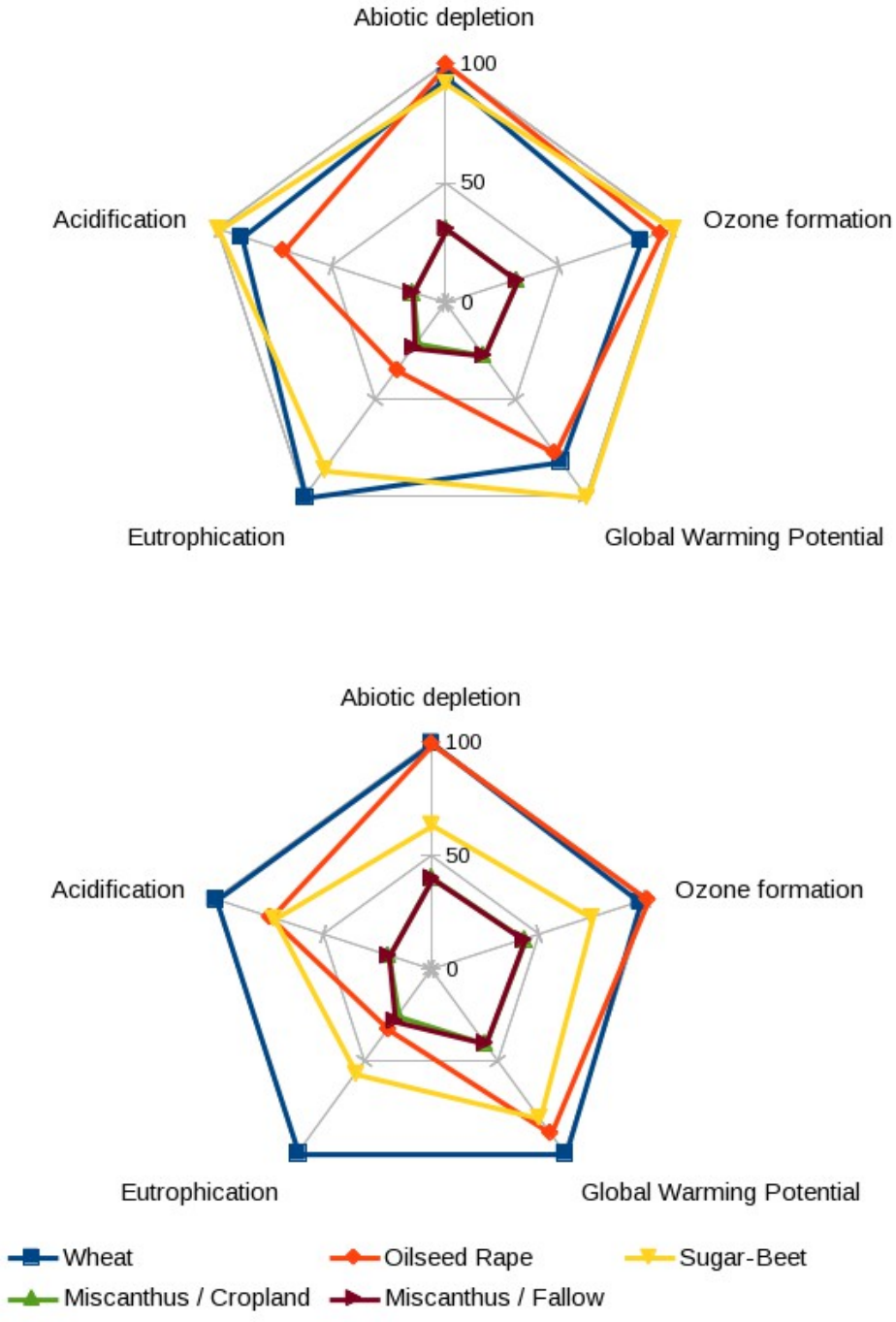
Figure captions

Figure 1. Compared cradle-to-farm-gate LCA results for the various feedstocks per ha (top) and MJ of biofuel feedstock energy content (bottom). Two scenarios were implemented for miscanthus : conversion from cropland or fallow land. Direct effects of land-use changes are excluded here.

Figure 2. Life-cycle emissions of greenhouse gases per MJ of of biofuels and their fossile equivalents. EtOH : ethanol, RME: rape methyl-ester; W: wheat; SB: Sugar-Beet. ADEME refers to the ADEME 2010 study and methodology, and CERES to the modelled emissions of Nr. Emissions related to direct and indirect land-use changes are noted dLUC and iLUC, respectively. The error bars correspond to the 5%-95% confidence intervals with respect to the field emissions of N2O.

Figure 3. Relative life-cycle impacts of biofuels in Ile de France using either regional ecosystem modelling (CERES) or IPCC Tier 1 emission factors (ADEME), per MJ of biofuel energy content. EtOH : ethanol, RME: rape methyl-ester; SB: Sugar-Beet; Misc: miscanthus.



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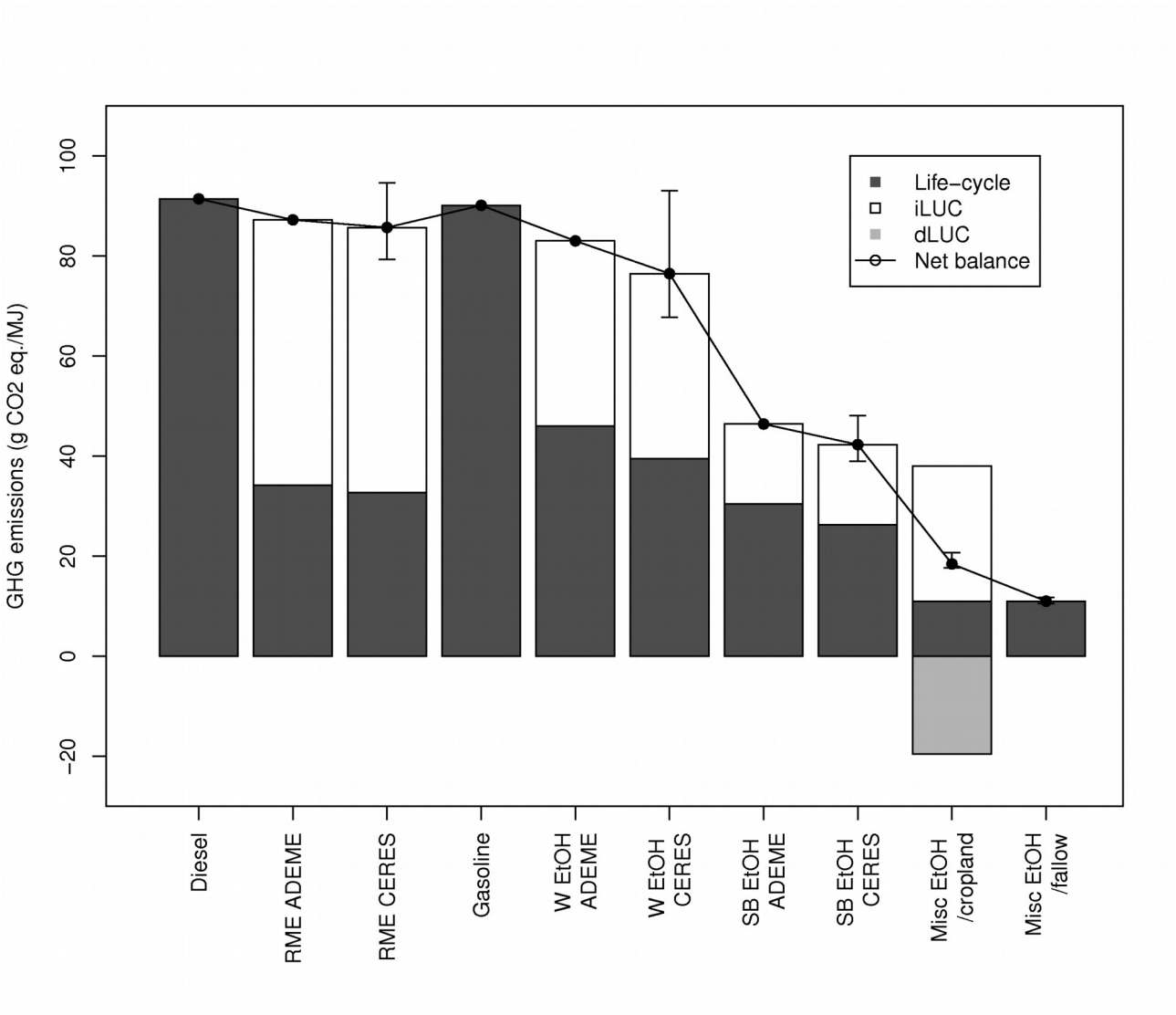

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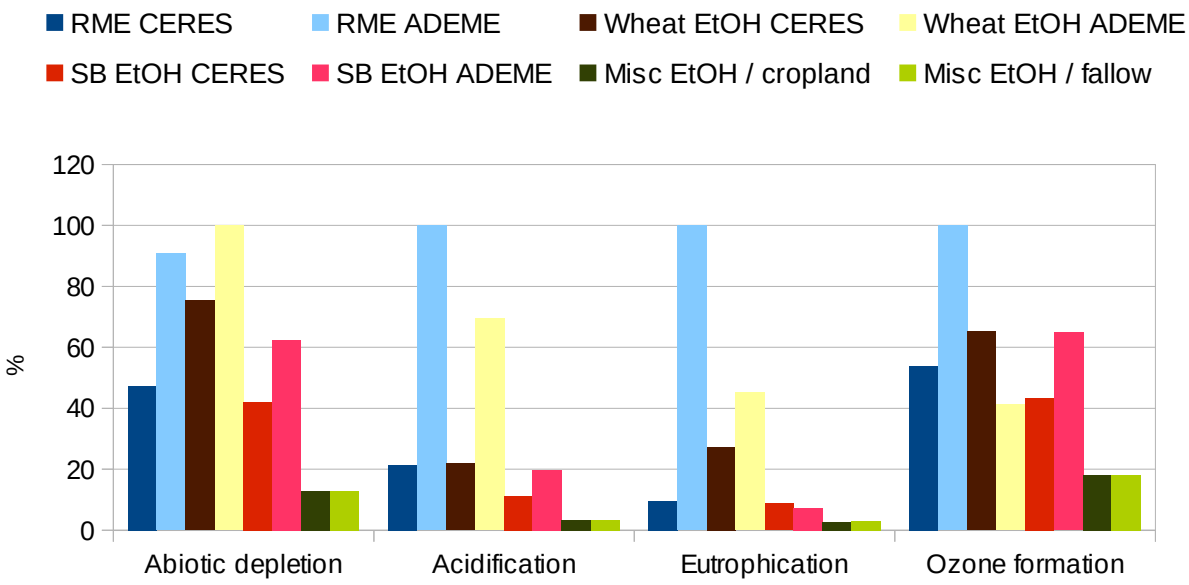

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Processes / stage	Source
Feedstock production	
Crop management data:	
- miscanthus	Expert knowledge[1]
- winter wheat	AGRESTE survey[2]
- oilseed rape	AGRESTE survey[2]
- sugar-beet	AGRESTE survey[2]
Crop yields	CERES-EGC simulations
Machinery & inputs production (fertilizers, pesticides, fuel)	EcoInvent database (v2.0)[3]
Field emissions	
N_2O emissions	IPCC 2006 guidelines, CERES-EGC or O-CN simulations
Other direct Nr losses	CERES-EGC simulations
Soil C dynamics	IPCC guidelines or CERES-EGC simulations
Transport	EcoInvent database
Conversion to biofuel	
1st generation biofuels	EcoInvent database, refs 1 and 4
2nd generation ethanol	EcoInvent database, ref. 1

Table 1. Sources of data for the life-cycle inventory of the pathways investigated. AGRESTE refers to the Statistical Bureau of the French Ministry of Agriculture. References: 1:Bessou, 2009; 2: Gabrielle et al., 2012; 3: Jungbluth et al., 2007 ; 4: ADEME, 2010

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	Winter wheat	Sugar Beet	Oilseed Rape	Miscanthus on cropland	Miscanthus on fallow
	Fertilizer input rates (kg N-P-K ha^{-1})				
Mineral N	200	133	182	0	0
P	28	48	33	2.7	2.7
K	26	160	44	4	4
	Pesticide input rates (kg a.i. ha^{-1})				
	1.11	1.42	2.48	0.10	0.10
	Regional crop yields in 2007 (t DM ha^{-1})				
Observed	7.70[a]	8.50[a]	3.20[a]		
CERES-EGC predictions	7.87 (grains)	10.92 (tubers)	3.09 (grains)	14.9 (aerial DM)	14.4 (aerial DM)

a : from agricultural statistics for 2007

Table 2. Crop management data for the energy crops considered here. DM: dry matter.

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	Winter wheat	Sugar Beet	Oilseed Rape	Miscanthus on cropland	Miscanthus on fallow
N_2O (direct):					
CERES-EGC	1.04 (0.11-2.98)	1.49 (0.11 – 3.97)	0.42 (0.10–0.87)	0.23 (0.10 – 0.60)	0.23 (0.14 – 0.35)
NO	0.80	0.99	0.86	0.41	0.42
NH_3	4.10	5.14	2.31	-0.16	-0.16
Nitrate	36.8	30.4	9.4	8.9	10.3
Soil C stock variations (kg C ha^{-1} yr^{-1})					
	5	-34	41	575	577
Crop yields (t DM or fresh tuber ha^{-1})					
	7.53	82.2[a]	4.19	13.1	13.2
Energy yield, whole-plant basis (GJ ha^{-1})					
	257	392	274	210	211

a: fresh tuber weight ha^{-1}

Table 3. Nr losses, soil C changes and dry matter (DM) and energy yields for the biofuel feedstocks (area-weighted mean and 5%-95% inter-quartile range in brackets for direct N_2O emissions). Variations in soil C stocks are reported over the 21-year simulation period, as averaged on an annual basis.

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	Winter wheat	Sugar Beet	Oilseed Rape	Miscanthus on cropland	Miscanthus on fallow
CERES-EGC :					
- direct	1.04	1.49	0.42	0.23	0.23
- indirect (leaching + gaseous)	0.32	0.29	0.10	0.07	0.08
TOTAL	1.36 (0.33-3.30)	1.78 (0.40-4.26)	0.52 (0.20-0.97)	0.30 (0.17-0.67)	0.31 (0.22-0.43)
ADEME (2010)					
- direct	2.00	1.33	1.82	0.5	0.5
- indirect :					
harvest residues	0.50	1.32	0.60	0.06[c]	0.06[c]
nitrate leaching	0.30	0.13	0.30	0.15[b]	0.19[b]
TOTAL	2.80	2.78	2.72	0.71	0.71

599 a : nitrate leaching rates from ADEME (2010)

600 b : nitrate leaching rates from CERES-EGC simulations

601 c : residues left after removal of the miscanthus stand (below-ground N stock estimated at 100

602 kg N ha^{-1} in late spring ; Strullu et al., 2011).

Table 4. Estimates of N_2O emissions (kg N_2O-N ha^{-1} yr^{-1}) for the various feedstocks obtained with the agro-ecosystem model CERES-EGC or the IPCC 2006 guidelines (ADEME, 2010).

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