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The significance of nitrous oxide emission from biofuel crops on arable land: a Swedish perspective

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Abstract

The current regulations governing biofuel production in the European Union require that they have to mitigate climate change, by producing >35 % less greenhouse gases (GHG) than fossil fuels. There is a risk that this may not be achievable, since land use for crop production inevitably emits the strong GHG nitrous oxide (N_2O), due to nitrogen fertilisation and cycling in the environment. We conclude that efficient agricultural crop production resulting in a good harvest and low N_2O emission can fulfill the EU standard, and is possible under certain conditions for the Swedish agricultural and refinery production systems. However, in years having low crop yields total GHG emissions can be even higher than those released by burning of fossil fuels. In general, the N_2O emission size in Sweden and northern Europe is such that there is a >50 % chance that the 35 % saving requirement will not be met. Thus ecosystem N_2O emissions have to be convincingly assessed. Here we compare Swedish emission data with values estimated by means of statistical models and by a global, top-down, procedure; the measurements and the predictions often show higher values that would fail to meet the EU standard and thus prevent biofuel production development.

1 Introduction

In June 2009 the European Union Directive “Promotion of the use of energy from renewable sources” (EC directive 98/70/EG) came into force, setting goals for 20 % use of renewable energy in 2020. Under the Directive, the transport sector will be required to use at least 10 % renewable fuel. The motives are local energy security and reduced greenhouse gas (GHG) emissions, compared with those emanating from fossil fuels. However, one problem is that cropping of the feedstocks for the first-generation liquid biofuels – mainly cereal grains and oilseeds in the temperate zone, and sugar cane and palm oil in the tropics – inevitably involves emissions of greenhouse gases, nitrous oxide in particular, and some of these fuels may actually cause more emissions than the gasoline and diesel fuels they replace (Crutzen et al., 2008; Mosier et al., 2009). Thus the Directive contains sustainability rules for biofuels; for example, the savings of greenhouse gases expressed in g CO_2-equivalents per MJ when biofuel is used must be at least 35 %, compared to fossil fuel use, and this differential will be increased to 50 % in 2017. Recent modelling suggests that these targets are not being achieved in many circumstances (Ogle et al., 2008; Mosier et al., 2009; Smeets et al., 2009) and this raises the question of how to estimate nitrous oxide emissions caused by biofuel cropping in different regions, to reduce the uncertainties.

Nitrous oxide (N_2O) is a naturally occurring and chemically stable greenhouse gas, with a global warming potential about 300 times greater than that of carbon dioxide, and a lifetime of more than 100 yr in the atmosphere (Forster et al., 2007). It is produced mainly by microbial activity in soils, and expansion of agriculture and increasing use of nitrogen (N) in synthetic fertilisers and manures has resulted in agricultural soils becoming globally the main source of N_2O – 65 % of anthropogenic emissions according to IPCC (2006) and as much as 80 % according to Crutzen et al. (2008). In recent years these changes have caused the atmospheric N_2O concentration to increase by 0.25 % yr^{-1} (Prather et al., 2001). Nitrous oxide emissions from soil have increased by 50 % during the last 150 yr due to increased nitrogen use (Crutzen et al., 2008). The discussion on the importance of N_2O emission in agricultural production in relation to the achievement of lower emissions with biofuels compared with fossil fuels has also thrown light on GHG emissions in relation to agricultural production in general. Globally, the efficiency with which N fertiliser is used by crops is only of the order of 40 %, as measured by the recovery of N in the harvested crop (Cassman et al., 2002); the figure is slightly higher for crops in northern Europe, e.g. 47 % for Sweden (Oenema et al., 2009). This inefficiency is important in agro-ecosystems with newly added nitrogen amendments and high soil fertility, increasing the likelihood of surplus nitrogen and N_2O production (Wang and Bakken 1997; Korsaeth et al., 2001).

A key question is: how much N_2O will be emitted by the cropping for biofuel production? Detailed earlier life cycle analyses (LCAs) on three different biofuel production

Title Page
Abstract Introduction
Conclusions References
Tables Figures
◀| |▶
◀ ▶
Back Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

systems showed both higher and lower GHG emissions than those from the use of fossil fuels and the estimated emission of N_2O was a decisive factor for the overall GHG emission level (Mosier et al., 2009). In each EU country, biofuel producers and regulatory authorities need to know whether or not their products will achieve the threshold set by the EU directive; our purpose here was to examine ways to estimate the size of N_2O emissions from crop-based biofuels grown under Swedish conditions.

1.1 Lifecycle assessment for Swedish conditions

The RES directive [EC directive 98/70/EG] states a need for at least 35 % savings compared to fossil fuels like petrol, emitting 83.8 g CO_{2eq} MJ^{-1}. Thus 54.5 g CO_{2eq} MJ^{-1} is the maximum allowed emission. We began by asking: how much N_2O emission can be allowed in order to meet this criterion, in production of ethanol from wheat using current farming techniques in Sweden? The calculations were made for two southern regions for which standard yields were obtained from the Swedish Agricultural Statistics (Statistics Sweden, 2011) (Fig. 1). We used information showing that only 60.8 % of the harvested energy can be converted into ethanol energy, equal to 7.9 MJ kg^{-1} grain (Bernesson et al., 2006, cited by Ahlgren et al., 2009); the other part goes into co-products such as distillers' grain. Also, in the refinery the conversion of the grains into ethanol needs energy, which according to Börjesson (2008) is equal to half the amount contained in the ethanol produced. We then assumed the same energy mix as in the overall Swedish energy system, with 43 % from renewable energy sources (Swedish Energy Agency, 2010), having low CO_2 emissions with only 1 g CO_{2eq} MJ^{-1} (a number given by the RES directive [directive 98/70/EG]). For fossil fuels the corresponding number is 83.8 gCO_{2eq} MJ^{-1}. The refinery emissions were then estimated to be 24 g CO_{2eq} MJ^{-1} ethanol-derived energy. Energy used for transportation and crop management, field operations and drying result in 3 g CO_{2eq} MJ^{-1} ethanol-derived energy (Ahlgren et al., 2009). The manufacture of N fertiliser (and to a lesser extent biocides) needs much energy and also produces N_2O. The fertiliser source category is thus shown separately because of the large total impact, and we use a low emission value for N fertiliser production, since products produced in low-emission factories with catalytic destruction of N_2O dominate the Swedish market. Also we assume a conventional rate of fertiliser addition, 120 kg N ha^{-1} yr^{-1}. Maximum acceptable direct N_2O emissions E_{N_2O} , to achieve 35 % savings compared to fossil fuels, i.e. ≤54.5 gCO_{2eq} MJ^{-1}, were estimated by:

$$E_{N_2O} = (E_{CO2M} - E_{EM}) \times y \times 3/4/298 \times 28/44 \quad (1)$$

where E_{CO2M} is the maximum allowed emissions for 35 % savings (54.5 gCO_{2eq} MJ^{-1}) and E_{EM} is the sum of emissions caused by energy use and manufacturing (management, transportation, ethanol refinery and emission caused in N manufacturing), y is the crop yield converted into ethanol (MJ ha^{-1}), direct emissions are 3/4 and indirect emissions are 1/4 of the total N_2O emissions, division by 298 (GWP) converts CO_2 into N_2O and multiplication by 28/44 converts N_2O into N_2O-N.

Summing the emissions, we concluded that for the two Swedish regions N_2O emission from agricultural fields cannot exceed in total 2 kg N_2O-N ha^{-1} yr^{-1} (1.5 for direct emission and 0.5 for indirect), in order to achieve the required saving of 35 % (Fig. 1). Measurements on unfertilised organic soils in Sweden have shown much larger emissions from barley production (a direct emission of 10 kg N_2O-N ha^{-1} yr^{-1}) (Kasimir Klemedtsson et al., 2009); use of such land would not be possible if the "sustainability goal" were to be achieved. Therefore in the remainder of our investigation we only include mineral soils, and determine the magnitude of the emissions we may expect on the basis of flux measurements and estimation models.

2 Method

2.1 Data from measurements

To estimate the size of agricultural N_2O emissions as a consequence of cropping of the biofuel feedstock, we need measurements. The simplest and most common measurement method is by use of flux chambers: frames inserted permanently (apart from having to be removed briefly during seeding and soil management operations) in the soil surface. The rate of accumulation of N_2O in the chambers is measured by gas chromatography. This gives data on direct emissions, i.e. those from the surface of the agricultural field. Chamber measurements, in spite of their limitations, are still the staple method for obtaining emission data, and are the only technique readily available to most researchers. In principle, better measurements can be made by micrometeorological techniques, which make it possible to measure the emission while disturbing neither the soil nor the crop (Wagner-Riddle et al., 2007). The emissions are often characterised by large temporal variations; thus it is important to detect sudden increases that commonly occur after fertiliser N additions or rainfall/irrigation events. This makes field measurements complicated and prolonged – they should run for at least a 12-month cycle – and consequently costly. However, since the 1980s, data have been collected in this way in many different countries and locations. Swedish data used here include measurements during winter periods when soils were frozen, since emissions can then be as great as during other seasons, with large emission peaks often being found during thawing events (Jungkunst et al., 2006).

Emission measurements in Sweden on crops appropriate for producing biofuels such as ethanol or biodiesel include wheat, rye, barley and oilseed rape. Here we report data on emissions from two experimental farms, both on mineral soils: Mellby gård in Halland, south west Sweden, and Logården in Västra Götaland, west Sweden. Both farms have conventional plant production systems with no livestock, and in both cases the crop was spring wheat. The first farm has coarse sandy soil, with 5 % organic matter and 5–10 % clay. Calcium ammonium nitrate (CAN), 120 kg N ha^{-1} yr^{-1}, was added to two plots by broadcasting and drilling and one was maintained without fertiliser addition. The second farm has a light clay soil with 30–40 % clay in the topsoil and more than 50 % in the subsoil, and an organic matter content of >4 %: a soil type identified as one providing a risk of high emissions, due to small soil pores having high water-holding capacity, increasing the risk of lack of oxygen. Here the fertiliser (NH_4NO_3, Axan) additions were 117 and 128 kg N ha^{-1} yr^{-1}.

Published data for similar systems elsewhere were taken from the large data set compiled by Stehfest and Bouwman (2006) (which also is the basis for the new IPCC (2006) direct emission factor – see below), comprising 1008 measurements of N_2O from agricultural systems, of which 223 measurements were made on the crops wheat, rye, barley and rape. We selected only data measured in northern Europe (>46° N) and Canada, with the quality criterion of at least a one-year measurement period. Moreover, data from organic soils were omitted. This selection provided 28 values, and we have added three data points from Kavdir et al. (2008) and our five values obtained from the measurements at Mellby and Logården, Thus the total number of measurements comes to 36, and the annual emissions are plotted against N application rates in Fig. 2. The full dataset can be obtained in Supplementary data A.

2.2 Data from models

IPCC emission factor

The Intergovernmental Panel on Climate Change (IPCC) has agreed on a relatively simple estimation method which can be used by all nations to estimate their national emissions of nitrous oxide. Originally the emission factor of 1.25 % for the direct nitrous oxide emission from soil and fertiliser additions (IPCC, 1997) was based on 20 data points (Bouwman 1996), measured during one year mainly from mineral soils in USA and UK, which had different crops and types of N additions. A linear increase in nitrous oxide emission with the amount of nitrogen added to the field was obtained. More data have been obtained since then and the correlation has become weaker, especially where <100 kg N ha^{-1} has been added to the field (Stehfest and Bouwman, 2006). This is also evident in Fig. 2. In the 2006 updating of the IPCC ”Guidelines” the emission factor for nitrous oxide from arable fields has been modified to 1 % (IPCC, 2006) based on Bouwman et al. (2002), Stehfest and Bouwman (2006) and Novoa and Tejeda (2006). The IPCC report points out that reporting countries should use the most detailed and appropriate estimation method available, preferably their own equations and emission factors (Tier 2) and, where possible, process modelling (Tier 3). If no such methods are available the reporting country has to use Tier 1, based on the 1996 Guidelines, as the 2006 revised version has not yet come into force. Sweden uses a modified emission factor for agricultural land receiving only inorganic fertilisers, 0.8 % of added N (Kasimir Klemedtsson, 2001).

The original purpose of the IPCC emission factors was not to estimate nitrous oxide emission for every field, crop or year, but only to give a method to estimate the possible scale of emissions from a country to include in reports to UNFCCC, and to indicate trends from year to year. Inevitably, as long as the Tier 1 methodology is employed, there will be countries or regions with greater-than-average emissions, and others with lower-than-average emissions, depending on environmental and local agricultural management factors. The IPCC emission factors have led to a general acceptance of a clear connection between N-addition and N_2O emission, despite increasing data collection showing a weak connection to inorganic N-additions up to 400 kg N ha^{-1} yr^{-1} (Novoa and Tejeda 2006). It is important to recognise that even if no fertiliser nitrogen is added the nitrous oxide emission will still continue since agricultural soils contain much labile nitrogen (Freibauer et al., 2004). Moreover, factors other than nitrogen have an influence on the emission, in particular soil wetness, temperature and carbon availability. This leads into more complex models:

Statistical models

Statistical models that try to include other influencing factors in addition to the nitrogen input have been made by Freibauer (2003), Freibauer and Kaltschmitt (2003) and Stehfest and Bouwman (2006). The first two aimed for a regionalized estimation procedure and compiled data on soil emissions from European measurement and analyzed it by a stepwise multivariate method. The very south of Sweden was included into their region with a "mild westerly climate", but most of Sweden was categorized as being in the sub-boreal region. In the south, determining factors for direct N_2O emission were, besides nitrogen fertiliser addition, the topsoil carbon and sand contents. For the boreal region two important parameters deciding emissions were fertiliser addition and nitrogen content of the topsoil. The procedure gave the following statistical relation for N_2O emission from mineral soils in the south:

$$E_{N2O} = 0.6(\pm 0.5) + (0.002(\pm 0.002) \times N_{fertiliser}) + (1.27(\pm 0.28) \times C_{soil}) - (0.024(\pm 0.005) \times \text{sand});$$

and for the sub-boreal region:

$$E_{N2O} = -1.3(\pm 2.1) + (0.033(\pm 0.0008) \times N_{fertiliser}) + (28(\pm 13) \times N_{soil})$$

where E_{N2O} = Nitrous oxide emission in kg
N_2O-N ha^{-1} yr^{-1} $N_{fertiliser}$ = Added nitrogen fertiliser in kg N ha^{-1} yr^{-1}
C_{soil} = Carbon content in the topsoil, % of soil weight
N_{soil} = Nitrogen content in the topsoil, % of soil weight
Sand= Sand content in topsoil, % of soil weight.

The other statistical model was constructed by Stehfest and Bouwman (2006), who applied different parameter values (F) for different conditions to the relation Log $E_{N2O} = A + \Sigma F_i$, where different conditions receive different values on F; examples of important parameters are given in the box below:

A = constant (-1.5)
F_1 = $0.0038 * N_{\text{fertiliser}}$,
F_2 = 0 if $<1\,\%$ C and $=0.05$ if 1–3 % C,
F_3 = -0.069 if pH 5.5–7.3,
F_4 = 0 if sandy soil and $=0.43$ if clay soil,
F_5 = 0.02 if coastal temperate climate,
F_6 = 0 if cereal crop and $= -0.35$ if grass and $= 0.44$ if other crop as rapeseed,
F_7 = 1.99 if data is obtained during year-long measurements.

Estimation by use of process-based models

Estimation by use of process-based models has been suggested since the production of nitrous oxide is complex and depends on many soil physical, chemical and biological factors. Also, interactions with the crop or other plants where the nitrogen uptake efficiency can be important. This is why process-based models have been and are being developed, aiming at estimating nitrous oxide emission accurately. One example is the DAYCENT model which is currently in use for estimation of nitrous oxide in the USA's national reporting of greenhouse gas emissions (Del Grosso et al., 2008). Another widely used process model is the Denitrification-Decomposition model, DNDC, to which several sub-models have been attached (Li et al., 1992, 2000), and which can use geographic databases connected to modelling of soil processes to simulate N_2O emission from different ecosystems (Kesik et al., 2005; Beheydt et al., 2008; Giltrap et al., 2010; Smith et al., 2010b). Yet another model platform is COUP (Coupled heat and mass transfer model for soil-plant-atmosphere systems), which originally was a soil physical model based on Swedish soil physical data for calculating processes in the soil-plant system, and has been further developed to be applicable to whole crop rotations (Jansson and Karlberg 2004). A module for estimating nitrification/denitrification processes via the PnET-N-DNDC model has been connected to the Coup model (Norman et al., 2008). This combined model has so far been used for research purposes to understand soil processes.

Top-down estimation

A top-down approach based on calculations by Crutzen et al. (2008) can be used to give a global average figure, if no local or regional possibilities exist. The method is based on the fact that atmospheric nitrous oxide concentration before industrialization was fairly stable (as evidenced by ice-core data), with the rate of addition to the atmosphere balanced by the rate of loss by stratospheric decomposition (Prather et al., 2001). At present, however, the N_2O concentration in the atmosphere is increasing at about 0.7 ppb yr^{-1} and the annual addition is 50 % higher than in the pre-industrial period (Crutzen et al., 2008). They argued an overall connection between the N_2O addition to the atmosphere and the global creation of reactive nitrogen by fixation, which also has increased by 50 %. Nitrogen fixation in this context means biological fixation and fixation by the Haber-Bosch process. Thus 3 to 5 % of the newly fixed nitrogen is emitted as nitrous oxide – similar to the proportion of the N that Galloway et al. (2004) estimated to have been newly fixed in natural ecosystems in the pre-industrial era.

3 Results and discussion

3.1 Swedish measurements

Measured emission from the Mellby farm was on average 2–3 kg N_2O-N ha^{-1} yr^{-1} where the fertilised plots did not differ from the control. The emission peaks in spring and early summer (June) (Fig. 3) made the difference between the median emission and the average. The control plot had a lower yield, which can be important when expressing emission per unit yield, as in this case of ethanol production. For the Logården farm, in spite of the clay soil and fertiliser addition, the emission was low – but had a few emission peaks, in early spring and after harvest (Fig. 4). This resulted in the average emission being two times higher than the median (Table 1). The lower emission in the second year may have been due to lack of measurements during soil thawing in early spring when we frequently have observed higher emissions. The emission from Mellby farm was double that from Logården, in spite of Mellby having a sandy soil, which was unexpected, as was the fact that the fertiliser addition at the Mellby farm did not increase the emission compared with the plot without fertilisation. At the Logården farm the design did not include a zero addition plot but a parallel organic rotation had even lower emissions (half the size) than the conventional wheat. Our interpretation is that preceding management actions and crops also have influence on the emissions. But the important issue here is the overall size of the emissions, affecting the sustainability of the conversion of agricultural produce into liquid fuels.

3.2 Collected data

It is common practice to present emissions in relation to fertiliser addition. Figure 2 shows the compiled literature data together with the Swedish data presented above. Average emission for the whole dataset is 2.8 kg N_2O-N ha^{-1} yr^{-1}, with a median of 1.9 kg N_2O-N ha^{-1} yr^{-1}. Sites receiving fertiliser have higher emissions compared to those with no fertiliser addition, averaging 3.0 ± 0.6 compared to 1.2 ± 0.6 kg N_2O-N ha^{-1} yr^{-1}. It is also possible to see that the Swedish data are in the same range as other data from the northern European region. The nitrogen addition explains less than 50 % of the emissions shown in Fig. 2, estimated from linear regression. Thus, other environmental factors have an important influence on the emissions. Like natural ecosystems, fertile arable fields contain tons of nitrogen per hectare, which has accumulated after many years of N fixation and/or addition in manure or fertiliser. Varying proportions of this organic N can be mineralized, and contribute to the N_2O production, depending on environmental influences and management actions. Effective cropping systems that assimilate most of the available nitrogen may give low nitrous oxide emissions, at moderate fertiliser nitrogen additions (Snyder et al., 2009). This can be seen in Fig. 2, with a more pronounced tendency for high emissions to follow after high N addition but a less clear connection at low N addition. Since the flux could not exceed

1.5 kg N_2O-N ha^{-1} yr^{-1} if the goal of a 35 % saving compared to fossil fuels were to be reached and the median of the emissions shown in Fig. 2 was 1.9 kg N_2O-N ha^{-1} yr^{-1} for studies performed during similar conditions as prevailing in Sweden, the risk of exceeding this limit is of the order of >50 %.

3.3 Comparing measured data with model estimates

The need for estimation models that can match actual emissions is why both IPCC's emissions factors and the more complex models were developed. By using the data from measurements on Mellby and Logården where auxiliary data are available we were able to validate the different methods. Table 2 shows the evident inability of the IPCC 2006 method to estimate emission for cases where no N-additions were made. Comparing the use of IPCC emission factors with data from fertilised plots the Swedish results shows an underestimation for Mellby and overestimation for Logården. The statistical method by Freibauer and Kaltschmitt estimates an emission 3–14 times as large as what was measured, and also the uncertainty range is above measured emissions. We found that the most important factors in the equations deciding the emission are the soil carbon content for Mellby and the soil nitrogen content for Logården, and the nitrogen addition is of less importance. In contrast we found the Stehfest and Bouwman estimation method to agree, but only for the control plot at Mellby that had no N-addition; however, for all cases receiving fertilisers the emissions was predicted to be 2–8 times higher compared with measured values. In this method, the nitrogen addition and duration of field measurements were the most decisive factors for the estimation. The Stehfest and Bouwman method shows the largest uncertainty and it is somewhat surprising that the length of measurement period was such an important factor for the result.

Both measured and estimated emissions presented in Table 2 are all within the range of emissions measured in northern Europe, but the average emission of compiled measurement data from (inorganically) fertilised arable land in the region, presented in

Fig. 2, was found to be 2.8 ± 0.5 kg N_2O-N ha^{-1} yr^{-1}, which is smaller than that estimated by both the Freibauer and Kaltschmitt and the Stehfest and Bouwman calculation methods but is in line with our measurements in Sweden. The Crutzen et al. (2008) method would, for the fertilised plots, result in a similar emission to the two statistical ones, with a total emission around 3–6 kg N_2O-N ha^{-1} yr^{-1}. However, with this global method, all subsequent emissions caused by introducing new reactive N into the ecosystem are included, such as indirect emissions following N loss to air and water.

3.4 Possibility of achieving required GHG savings in Swedish ethanol production

The refinery in our Swedish LCA releases only 24 g CO_2equiv MJ^{-1}, which is low compared with the Mosier et al. (2009) study where 43–64 g CO_2equiv MJ^{-1} ethanol-derived energy was reported for refinery emissions in UK and USA, but this is due to a greater potential to use renewable energy in Sweden, which helps to reduce GHG emissions in this part of the production chain. Despite use of low N_2O-emitting fertiliser production and high proportion of renewable energy used in the refinery, the LCA shows that a biofuel based on the Mellby data does not reach the sustainability goal of 35 % savings (Table 3), since yields are low and field emissions high, and this ethanol production would give more GHG emission than fossil fuels. But the Logården case shows a low emission, <0.5 kg N_2O-N ha^{-1} yr^{-1} and despite a low yield it is possible to achieve the 35 % GHG savings (Table 3). With a higher yield and still fairly low emission, 1.1 kg N_2O-N ha^{-1} yr^{-1}, 40 % savings can be obtained. Even if refinery operation uses solely renewable energy, the results still show too high an emission for Mellby, with only 25 % GHG savings. But the same test for Logården shows it may be possible to gain 70 % savings if both a good yield, normal for the region, and fairly low emissions are achieved. The problem is that these low emissions are often not the case since there is more than a 50 % risk for emissions higher than 1.5 kg N_2O-N ha^{-1} yr^{-1} (Fig. 2), when ethanol production will not reach the sustainability rules.

Three main factors have been shown to be important for the possibility of reaching the threshold of 35 % savings of GHG compared with fossil fuels; these are harvest size, energy use in the refinery, and nitrous oxide emissions (direct and indirect). These indirect emissions are most often calculated by use of the IPCC emission factors. The 2006 IPCC default EFs for these indirect emissions are 1 % (uncertainty range 0.2–5 %) for volatilised N and 0.75 % (0.05–2.5 %) for leached N. At default volatilization fractions of 10 % (mineral fertiliser) or 20 % (animal manure), and the default leaching fraction of 30 %, indirect emissions can be negligibly small, 0.04 % of the added N using the numbers at the lower end of these ranges, but up to more than the direct emission (1.2 %), at the upper end. In most calculations the default EFs are used, resulting in an estimated emission of 0.3–0.4 % of the N applied to the land. More detailed discussion of indirect emissions and the likely EFs can be found in IPCC (2006) and Well and Butterbach-Bahl (2010). Here we have assumed the indirect emissions to be 1/3 of the direct emissions, based on the IPCC emission factors. Experiments for similar systems in Sweden to Logården and Mellby have shown N leaching to be lower from clay soil than from sandy soil, 2–22 and 15–53 kg N ha^{-1} yr^{-1}, respectively (Aronsson et al., 2011). Leaching at Logården the same year as emission was measured showed values of 18–22 kg N ha^{-1} yr^{-1}, which confirms a low leaching from clay soil (Stenberg et al., 2011; Wessén et al., 2011). The low leaching from the clay soil compared to the IPCC default may indicate a rather low indirect emission from clay soils, making the overall picture somewhat better than estimated in Table 3, but the N-leaching from sandy soil may be similar to the default values. We have shown the LCA to be very sensitive to the size of the N_2O emissions, both direct and indirect, where a small change can make it possible or impossible to reach the goal.

3.5 N_2O as a consequence of agriculture in general

The expected future population growth needs agricultural production to increase by 70 % to fulfil demands for food over the next 50 yr (FAO, 2009) raising the question: how much more N will be required to meet both increased food needs and biofuel production. And how much N_2O emission will result? Can agriculture management be designed for an overall low N_2O emission? One attempt to answer this was by van Groenigen et al. (2010), who showed that fertiliser addition rates below 200 kg N ha^{-1} give minimum N_2O emissions per unit of yield (8 g N_2O-N kg^{-1} crop N). Attempts to increase the yields with higher N additions will decrease the nitrogen use efficiency and inevitably increase N_2O emissions. However, there should be no departure from attempting to achieve the optimum level of yield and minimum level of N_2O per unit of yield, consequently the option to increase the yields of food and feed crops, thus releasing land for biofuel cropping, is limited. But even the best management options leave reactive nitrogen behind, which can possibly be converted into N_2O over time, providing some explanation for the Crutzen et al. (2008) emission factor of 3–5 % of the new N being converted into N_2O-N.

Due to the transfer of reactive N from agricultural land to surrounding ecosystems indirect emissions will also take place in the year of application and in following years (IPCC, 2006; Well and Butterbach-Bahl, 2010), as mentioned above. These emissions are included in the estimate produced by the method of Crutzen et al. (2008), but on a regional or country scale the overall emissions can be both lower and higher than the global average emission factor of 3–5 % of the newly fixed nitrogen.

Arguments have been raised that the atmospheric N_2O increase depends also on other factors besides the nitrogen fixation, e.g. the roles of an increased livestock production and mineralisation of soil nitrogen as substantial additional sources of liberated N (Davidson, 2009). We agree that taking virgin land into cultivation has been, and will continue to be a source, through the release of nitrogen stored in the soil organic matter, but N_2O from livestock production is already implicitly included in the Crutzen et al. (2008), concept, because the reactive N has entered the ruminant N cycle either as fertiliser on grass, or by BNF by fodder legumes and soya beans used in feed supplements.

The large overall emission calculated by the Crutzen et al. (2008) method may not always be welcomed by biofuel producers and authorities, who have argued that a lower direct emission factor is more appropriate, since only emissions directly associated with the cropping for bio-ethanol should be included and emissions that anyhow should result from the land, so-called background emission, can be subtracted (Ahlgren et al., 2009). A similar way to lower the emission is to use a reference case: an alternative land use for which hypothetical emissions can be subtracted from the actual production emissions [EC directive 98/70/EG]. However, we argue that a major part of such emissions is anthropogenic, resulting from past agricultural management (IPCC, 2006), and like present land use will influence future emissions. Thus the emissions are an integral part of agriculture and cannot be subtracted. Therefore we have not made any background subtraction in our simple LCA, which shows three factors having a major influence on the possibility of reaching the goal of 35 % GHG savings: harvest size, refinery emissions and soil N_2O emissions.

We now turn to the question of whether we can put numbers on the N_2O emission at a regional scale. The most important conclusion is that emission measurements in northern systems on mineral soils cropped with cereals and oilseed rape span a yearly nitrous oxide emission range from nearly zero to $>10\,\mathrm{kg\,N_2O\text{-}N\,ha^{-1}\,yr^{-1}}$, but for 75 % of the cases the emission is below $4\,\mathrm{kg\,ha^{-1}\,yr^{-1}}$ with an average $2.8\,\mathrm{kg\,N_2O\text{-}N\,ha^{-1}\,yr^{-1}}$ for fertilised agricultural mineral soils (Fig. 2). The variability between different investigations can be due to shortcomings in the intermittently performed measurements. But since the data were collected in different systems there are also different influencing factors and biological variation in soil may be huge. An emission below $1.5\,\mathrm{kg\,N_2O\text{-}N\,ha^{-1}\,yr^{-1}}$, needed to fulfil a GHG saving of 35 %, was found for less than 50 % of the data collected. And also the Logården clay soil had this necessary low emission. But because of the low yields it was still barely possible to fulfil the 35 % rule, and not possible to achieve a 50 % GHG reduction as will be required in 2017. The Mellby farm LCA estimations resulted in an increased GHG emission compared with fossil fuel, due to both low yields and “high” emission size.

We here included only mineral soils since this are the typical soils used for arable cropping across Sweden, but unless adequate account is taken of the contribution of organic soils, the average emissions will be underestimated. Of the arable land in Sweden, 9 % is classified as having organic soils, where stored carbon and nitrogen is released by mineralisation, causing high emissions: 10 kg N_2O-N ha^{-1} yr^{-1} when cultivated for cereal crops (Kasimir Klemedtsson et al., 2009), and 28 % of the organic soils in Sweden are used in this way (Berglund and Berglund, 2008). If there is no regulation controlling which soil type can be used for biofuel production, these emissions should be added and included in the typical soil emission values for the region, resulting in an average increase of almost 1 kg N_2O-N ha^{-1} yr^{-1}, which we can conclude would make biofuel production from arable crops impossible under the rules of 35 % (and 50 %) GHG savings.

3.6 Prediction of N_2O emission

To predict the size of the N_2O emission from an area of land, it is necessary to know which environmental factors have the most impact. The IPCC revised Tier 1 method, basing emissions solely on N inputs, is a convenient way to estimate the emissions, as it is easy to obtain the data, but as Fig. 2 shows, the influence of the fertiliser on the emission can be quite small. However, this concept has been used in many life cycle analyses (Smeets et al., 2009), even though it does not work well in some circumstances. Problematic cases are where soil organic matter contributes to the emission, from the release of nitrogen accumulated into the ecosystem long ago. Thus in principle we need attempts like those of Freibauer and Kaltschmitt (2003) and Stehfest and Bouwman (2006), where other influencing factors besides nitrogen addition were also included. Unfortunately, these methods resulted in estimates that were much higher than the Swedish measurements. Important factors in the Freibauer and Kaltschmitt method were the C and N content of the soil; this could be an indication of turnover of organic matter and release of N, but the contents themselves do not predict the rates of N turnover processes such as mineralisation, nitrification, denitrification and assimilation, that have a major influence on the actual N_2O emissions. There is a need for continued method development for nitrous oxide estimations at a local level, including different soils and crops in the whole agricultural system, and where indirect emissions are also included. This would make emission estimates more reliable and make it easier to adjust towards lower-emission systems. Process-based models can be a significant help in the evaluation process. We are convinced that models will be important in the future as tools for estimating nitrous oxide emission from local ecosystems and possibly to use for upscaling, but so far we have not estimated N_2O by use of a process model for Mellby and Logården; this will be the next step, by use of the CoupModel.

4 Conclusions

It can be concluded that it is possible to produce biofuels having lower GHG emissions than fossil fuels but only for systems with small N-surpluses in the soil, but still giving good yields. However, soils having ordinary levels of emissions (>1.5 kg N_2O-N ha^{-1} yr^{-1}) and high-emitting organic soils cannot be used for biofuel crop production. Also important is energy efficiency and low GHG emissions in the production of fertilisers and in ethanol refineries. Overall, the probability of not reaching the goal of 35 % GHG savings with the present agricultural system is as large as 50 %. To be able to avoid the high-emission systems in an explicit and reliable way will need better methods than those available today, but the methods discussed here could not give reliable estimates of N_2O emission for regions in Sweden that would help to steer towards those sites and systems with low emissions. Process-based models can be a help in achieving this objective in the future.

Supplementary material related to this article is available online at: http://www.biogeosciences-discuss.net/8/6743/2011/ bgd-8-6743-2011-supplement.pdf.

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Table 1. Results of Swedish field N_2O measurements.

Farm	Field/fertilisation method	N_2O emission,		Yield,
		Average	Median	
		kg N_2O-N ha^{-1} yr^{-1}		kg ha^{-1}
Mellby	Control (no N)	2.5	1.9	2400
	Broadcasting	2.9	2.1	3300
	Drilling	1.9	1.3	3300
Logården	Field C6[a]	1.1	0.6	5800
	Field C2[b]	0.5	0.2	3100

[a] April 2005 – March 2006
[b] April – December 2006

Table 2. Comparison between measured and calculated direct nitrous oxide emission. Measured data are from Mellby and Logården in Sweden, having mineral soils cropped with cereals, with the addition of mineral fertilisers.

Site and year	Fertiliser addition	Swedish field data[a]	IPCC [b] (1997)	IPCC (2006)[c]	Freibauer and Kaltschmitt 2003[d]	Stehfest and Bouwman 2006[e]
	kg N ha^{-1}	N_2O emission, kg N_2O-N ha^{-1} yr^{-1}				
Mellby, 1996–1997	0	2.5 (1.4–3.7)	1	0 (0)	6.6 (5.9–7.3)	2.9 (1.4–6.1)
Mellby, 1996–1997	120	2.4 (1.1–3.7)	2.5 (1.3–3.8)	1.2 (0.4–3.6)	6.9 (6.2–7.6)	4.7 (2.3–9.7)
Logården, 2005	117	1.1 (0.9–1.3)	2.5 (1.3–3.6)	1.2 (0.4–3.5)	6.5 (5.9–7.3)	3.8 (1.9–7.9)
Logården, 2006	128	0.5 (0.4–0.6)	2.6 (1.3–3.9)	1.2 (0.4–3.8)	6.9 (6.2–7.6)	4.0 (1.9–8.2)

[a] Measured direct emission, average from measurements all year round is shown together with standard error of the mean.

[b] IPCC (1997) emission factor 1.25 % (0.25–2.25 %) of N-addition in fertiliser plus a background emission of 1 kg N_2O-N ha^{-1} yr^{-1}. Crop residues and indirect emissions are not included.

[c] IPCC (2006) emission factor 1 % (0.3–3 %) of N-addition in fertiliser. Crop residues and indirect emissions are not included.

[d] For Mellby the function for temperate western climate was used and for Logården the function for alpine/sub-boreal climate.

[e] For Mellby, factor values for temperate oceanic climate are used together with a coarse soil texture and a high soil carbon content >3 %. For the Logården case, factor values for temperate continental climate, fine soil texture and soil carbon content between 1 and 3 % were used.

Table 3. LCA applied to Mellby and Logården data, showing GHG saving possibilities compared to fossil fuels[a].

Site	Yield	Measured emission	N addition	Energy in refinery	Energy use[d]	Industrial use[e]	Direct N_2O	Indirect N_2O	Total emission	Difference compared, to fossil fuels
	kg ha^{-1} yr^{-1}	kg N_2O-N ha^{-1} yr^{-1}	kg N ha^{-1}	g CO_{2eq} MJ^{-1}						%
Refinery using average Swedish energy mix										
Mellby	2400	2.5	0	24[b]	3	0	62	20	109	+30
Mellby	3300	1.9	120	24	3	13	34	11	85	+3
Logården	5800	1.1	117	24	3	7	11	4	50	−40
Logården	3100	0.5	128	24	3	15	10	3	55	−34
Refinery using only renewable energy										
Mellby	3300	1.9	120	0.5[c]	3	13	34	11	62	−25
Logården	5800	1.1	117	0.5	3	7	11	4	26	−69

[a] fossil fuels emit 83.8 g CO_{2eq} MJ^{-1}.

[b] Assuming the same energy mix in refinery as in the overall Swedish energy system, with 43 % renewable energy sources, having low CO_2 emissions with 1 g CO_{2eq}/MJ and 83.8 g CO_{2eq}/MJ for fossil fuels.

[c] Assuming renewable energy used in refinery.

[d] Energy use in transport and management.

[e] Emissions connected with industry production of fertiliser and biocides. Major part is from fertiliser production using catalytic conversion of N_2O into N_2, with 90 % efficiency (Yara), giving 2.9 kg CO_{2eq} kg^{-1} N.

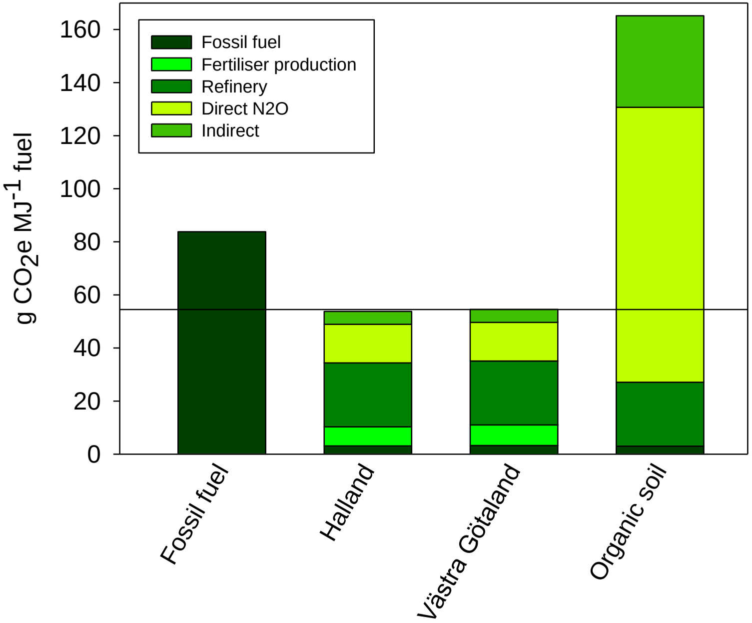

Fig. 1. LCA of greenhouse gas emissions due to production of ethanol from wheat, expressed as carbon dioxide equivalents per MJ ethanol-derived energy. For Halland and Västra Götaland, maximum N_2O-N emissions of 1.5 and 0.5 kg N ha^{-1} yr^{-1} for direct and indirect emissions, respectively, are allowed to achieve 35 % savings (horizontal line). Wheat production for bioethanol on unfertilised organic soils with emissions of CO_2 (not included here) and 10 kg N_2O-N ha^{-1} yr^{-1} gives higher emissions than fossil fuels and is shown for comparison. Standard yields for winter wheat were 6097 and 5676 kg ha^{-1} for Halland and Västra Götaland respectively (14 % water content) (Statistics Sweden, 2011). Fertiliser addition: 120 kg N ha^{-1} yr^{-1}. Emissions connected with industry production of fertiliser production using catalytic conversion of N_2O into N_2, with 90 % efficiency (Yara), giving 2.9 kg CO_{2eq} kg^{-1} N.

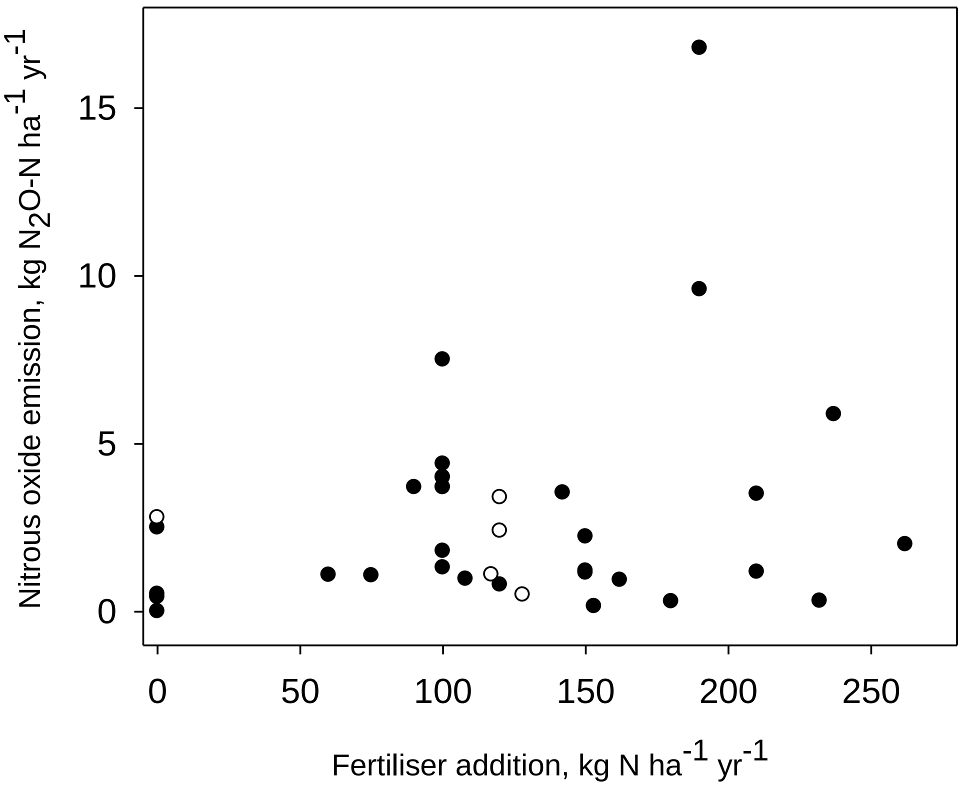

Fig. 2. Compiled measurement data on nitrous oxide emissions from cereal and rape crops in northern Europe and Canada, plotted against N fertiliser addition. White spots denote Swedish data.

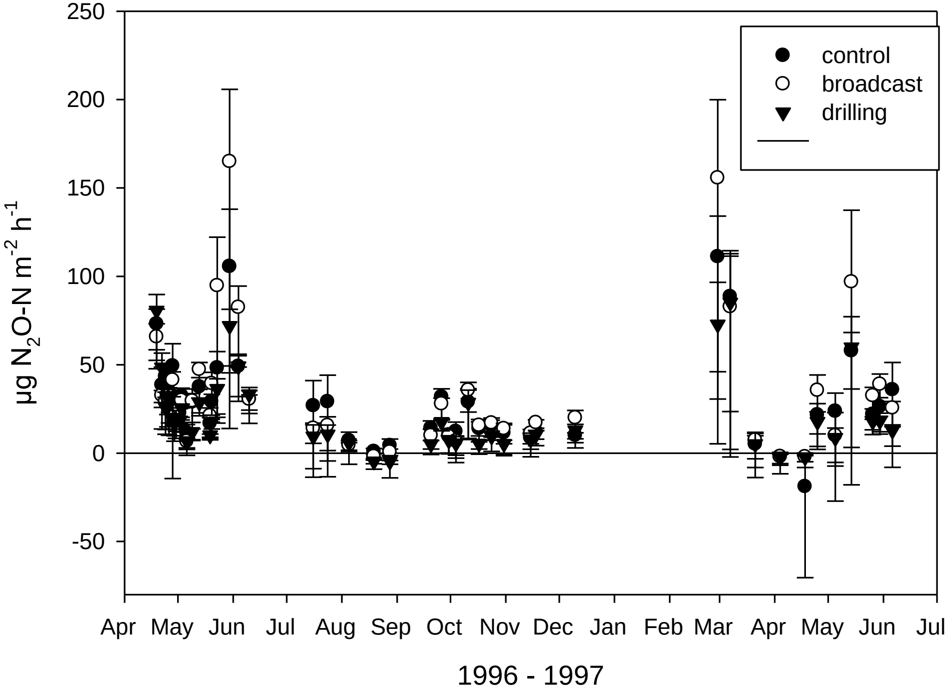

Fig. 3. N_2O emission at Mellby, comparing fertiliser spreading techniques; site had one control plot without N-additions. Average emission and standard error of eight static chambers per plot.

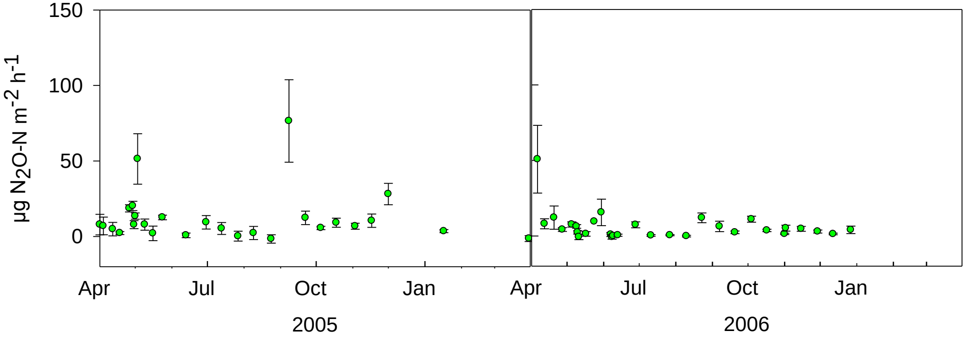

Fig. 4. N_2O emission at Logården, for two different plots spring wheat (C6 in 2005 and C2 in 2006); average and standard error of six chambers.