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**N₂O release from
fertilizer use in
biofuel production**

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N₂O release from agro-biofuel production negates global warming reduction by replacing fossil fuels

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Abstract

The relationship, on a global basis, between the amount of N fixed by chemical, biological or atmospheric processes entering the terrestrial biosphere, and the total emission of nitrous oxide (N_2O), has been re-examined, using known global atmospheric removal rates and concentration growth of N_2O as a proxy for overall emissions. The relationship, in both the pre-industrial period and in recent times, after taking into account the large-scale changes in synthetic N fertiliser production and deforestation, is consistent, showing an overall conversion factor of 3–5%. This factor is covered only in part by the ~1% of “direct” emissions from agricultural crop lands estimated by IPCC (2006), or the “indirect” emissions cited therein. This means that the extra N_2O entering the atmosphere as a result of using N to produce crops for biofuels will also be correspondingly greater than that estimated just on the basis of IPCC (2006). When the extra N_2O emission from biofuel production is calculated in “CO₂-equivalent” global warming terms, and compared with the quasi-cooling effect of “saving” emissions of fossil fuel derived CO₂, the outcome is that the production of commonly used biofuels, such as biodiesel from rapeseed and bioethanol from corn (maize), can contribute as much or more to global warming by N_2O emissions than cooling by fossil fuel savings. Crops with less N demand, such as grasses and woody coppice species have more favourable climate impacts. This analysis only considers the conversion of biomass to biofuel. It does not take into account the use of fossil fuel on the farms and for fertilizer and pesticide production, but it also neglects the production of useful co-products. Both factors partially compensate each other. This needs to be analyzed in a full life cycle assessment.

1 Introduction

N_2O , a by-product of fixed nitrogen application in agriculture, is a “greenhouse gas” with a 100-year average global warming potential (GWP) 296 times larger than an equal

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mass of CO₂ (Prather et al., 2001). As a source for NO_x, i.e. NO plus NO₂, N₂O also plays a major role in stratospheric ozone chemistry (Crutzen, 1970). The increasing use of biofuels to reduce dependence on imported fossil fuels and to achieve “carbon neutrality” will further cause atmospheric N₂O concentrations to increase, because of N₂O emissions associated with N-fertilization. Here we propose a global average criterion for the ratio of N to dry matter in the plant material, which indicates to what degree the reduced global warming (“saved CO₂”) achieved by using biofuels instead of fossil fuel as energy sources is counteracted by release of N₂O. This study shows that the use of several agricultural crops for biofuel production and climate protection can readily lead to enhanced greenhouse warming by N₂O emissions.

2 A global factor to describe N₂O yield from N fertilization

We start this study by deriving the yield of N₂O from fresh N input, based largely on data compiled by Prather et al. (2001) and Galloway et al. (2004). The pre-industrial, natural N₂O sink and source at an atmospheric mixing ratio of 270 nmol/mol is calculated to be equal to 10.2 Tg N₂O-N/year (Prather et al., 2001), which includes marine emissions. By the start of the present century, at an atmospheric volume mixing ratio of 315 nmol/mol, the stratospheric photochemical sink of N₂O was about 11.9 Tg N₂O-N/year. The total N₂O source at that time was equal to the photochemical sink (11.9 Tg N₂O-N/year) plus the atmospheric growth rate (3.9 Tg N₂O-N/year), together totalling 15.8 Tg N₂O-N/year (Prather et al., 2001). The anthropogenic N₂O source is the difference between the total source strength, 15.8 Tg N₂O-N/year, and the current natural source, which is equal to the pre-industrial source of 10.2 Tg N₂O-N/year minus an uncertain 0–0.9 Tg N₂O-N, with the latter number taking into account a decreased natural N₂O source due to 30% global deforestation (Klein Goldewijk, 2001). Thus we derive an anthropogenic N₂O source of 5.6–6.5 Tg N₂O-N/year. To obtain the agricultural contribution, we subtract the estimated industrial source of 0.7–1.3 Tg N₂O-N/year (Prather et al., 2001), giving a range of 4.3–5.8 Tg N₂O-N/year. This

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is 3.3–4.6% of the anthropogenic “new” fixed nitrogen input of 127 Tg N/year for the early 1990s (Galloway et al., 2004). In an earlier study (Mosier et al., 1998) the source of N₂O from agriculture was estimated to be even larger, 6.3 Tg N₂O-N, giving an N₂O yield of 5%.

5 Because of good knowledge of the chemical processing of N₂O in the atmosphere and its tropospheric concentrations, obtained from air enclosure in ice cores, its natural sources and sinks are well known and can be calculated with models. Thus, pre-industrial, natural conditions provide additional information on the yield of N₂O from fixed N input. For that period, the global source and sink of N₂O was 10.2 Tg N₂O-N/year with 6.2–7.2 Tg N₂O-N/year coming from the land and coastal zones (Prather et al., 2001), derived from a fresh fixed N input of 141 Tg N/year (Galloway et al., 2004), giving an N₂O-N yield of 4.4–5.1%. Supported by the above information, we accept a ratio of 3–5% for the past, present and also future yield of N₂O from fixed nitrogen input. The main uncertainty in our analysis is the fixed N input. Galloway et al (2004) only give single values for the annual inputs of new fixed N for the year 1860 and the early 1990s.

15 An evaluation of hundreds of field measurements has shown that N fertilization causes a release of N₂O in agricultural fields that is highly variable but averages close to 1% of the fixed nitrogen input from mineral fertilizer or biologically fixed N (Bouwman et al., 2002; Stehfest and Bouwman, 2006), and a value of 1% for such direct emissions has recently been adopted by IPCC (2006). There is an additional emission from agricultural soils of 1 kg N₂O-N/ha/year, which does not appear to be directly related to recent fixed N-input. The in-situ fertilizer-related contribution from agricultural fields to the N₂O flux is thus 3-5 times smaller than our adopted global average N₂O yield of 4±1% of the fixed N input. The large difference between the low yield of N₂O in agricultural fields, compared to the much larger average value derived from the global N₂O budget, implies considerable “background” N₂O production occurring beyond agricultural fields, but, nevertheless, related to fertilizer use, from sources such as rivers, estuaries and coastal zones, animal husbandry and the atmospheric deposition

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of ammonia and NO_x (Kroeze et al., 1999).

3 N₂O release versus CO₂ saved in biofuels

As a quick indicator to describe the consequence of this “background” N₂O production we compare its global warming with the cooling due to replacement of fossil fuels by biofuels. Here we will only consider the climatic effects of conversion of biomass to biofuel and not a full lifecycle, leaving out for instance the input of fossil fuels for biomass production, on the one hand, and the use of co-products on the other hand.

We assume that the fixed nitrogen, which is co-harvested with the biofuels, wherever it may occur, must be replenished over time in the fields with new fixed nitrogen. Thus we estimate the fixed nitrogen input from the nitrogen content of the harvested biomass. We also obtain the fossil CO₂ emissions avoided from the carbon processed in the harvested biomass to yield the biofuel. With these assumptions, we can compare the climatic gain of fossil fuel-derived CO₂ “savings”, or net avoided fossil CO₂ emissions, with the counteracting effect of enhanced N₂O release resulting from fixed N input. Our assumptions lead to expressions per unit mass of dry matter harvested in biofuel production to avoid fossil CO₂ emissions, “saved CO₂”, (*M*), and for “equivalent CO₂”, (*Meq*), the latter term accounting for the global warming potential (GWP) of the N₂O emissions:

$$M = r_C \times \mu_{CO_2} / \mu_C \times cv \quad (1)$$

$$Meq = r_N \times y \times \mu_{N_2O} / \mu_{N_2} \times GWP / e \quad (2)$$

In these formulae *r_C* is in g carbon per g dry matter in the feedstock; *r_N* is the mass ratio of N to dry matter in gN/kg; *cv* is the mass of carbon in the biofuel per mass of carbon in feedstock biomass (corn, rapeseed, sugar cane); *e* is the uptake efficiency of the fertilizer by the plants; *y*=0.03–0.05, the range of yields of N₂O-N from fixed N application; GWP=296; $\mu_{CO_2}/\mu_C=44/12$, $\mu_{N_2O}/\mu_{N_2}=44/28$, where the μ terms are the molar weights of N₂O, N₂, CO₂, and C.

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Inserting these values in Eqs. 1 and 2 we thus obtain, with expressions in parentheses representing ranges,

$$M=3.667 \cdot cv \cdot r_C \quad (3)$$

$$Meq=(14-23.2)r_N/e \quad (4)$$

$$5 \quad Meq/M=(3.8-6.3)r_N/(e \cdot cv \cdot r_C) \quad (5)$$

The latter term is the ratio between the climate warming effect of N₂O emissions and the cooling effect due to the displacement of fossil fuels by biofuels.

These equations are valid for all above-ground harvested plant material, and separately also for the products and residues, which are removed from the agricultural fields. If Meq>M, there will be net climate warming, the greenhouse warming by increased N₂O release to the atmosphere then being larger than the quasi-cooling effect from “saved fossil CO₂”. There will neither be net climate warming nor cooling by biofuel production when Meq=M, which occurs for

$$r_N=(0.158-0.263) \cdot (e \cdot cv \cdot r_C) \quad (6)$$

15 Under current agricultural practices, worldwide, the average value for e ≈ 0.4 (Cassman et al., 2002; Galloway et al., 2003; Balasubramanian et al., 2004). The data (and their sources) used to calculate the carbon contents, r_C, and the conversion efficiency factors, cv, and the calculations themselves, are given in Appendix A. As r_C we use 0.61, 0.44 and 0.43 for rapeseed, corn, and sugar cane, respectively. We derive values of cv=0.58 for rapeseed bio-diesel, 0.37 for corn bio-ethanol, and cv=0.30 for sugar cane ethanol production. Consequently,

$$r_N=22.3-37.2 \text{ g N/kg dry matter for rapeseed bio-diesel,}$$

$$r_N=10.3-17.1 \text{ g N/kg dry matter for corn bio-ethanol}$$

$$r_N=8.1-13.6 \text{ g N/kg dry matter for sugar cane bio-ethanol.}$$

25 For each of these biofuels, a larger value of r_N in the plant matter than this range implies that use of the fuel causes a net positive climate forcing.

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Note that our analysis only considers the conversion of biomass to biofuels, emphasizing the role of N₂O emissions. It does not take into account the supply of fossil fuel for farm machinery or fertilizer production; on the other hand it also neglects the production of useful co-products, which partially compensate for each other (see for instance Hill et al., 2006, for corn ethanol).

4 Discussion

4.1 Nitrogen content in biofuels

Data on r_N for several agricultural products, in g(N)/kg dry matter (Velthof and Kuikman, 2004; Biewinga and van der Bijl, 1996), are presented in Table 1. They show net climate warming, or considerably reduced climate cooling, by fossil fuel “CO₂ savings”, due to N₂O emissions. The r_N value for corn is equal to 15 g N/kg dry matter, leading to a relative climate warming of 0.9–1.5 compared to fossil fuel CO₂ savings. The effect of the high nitrogen content of rapeseed is particularly striking; it offsets the advantages of a high carbon content and energy density for biodiesel production. World-wide, rapeseed is the source of >80% of bio-diesel for transportation, and has been particularly promoted for this purpose in Europe. For bio-diesel derived from rapeseed, this analysis indicates that the global warming by N₂O is on average about 1.0–1.7 times larger than the quasi-cooling effect due to “saved fossil CO₂” emissions. For corn/ ethanol the relative warming due to N₂O emissions is very similar: 0.9–1.5, while for sugar cane/ethanol the relative warming is 0.5–0.9, based on a r_N value of 7.3 g N/kg dry matter (Isa et al., 2005).

Agricultural plant residues can also be used for bio-fuel production. Also for these materials, high r_N values cause unfavourable or low gain impacts on climate (Table 1).

Although there are possibilities for improvements by increasing the efficiency, e.g. for the uptake of N fertilizer by plants (Cassman et al., 2002) – which is much needed in regular agriculture as well – on a globally averaged basis the use of agricultural crops

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for energy production can readily be detrimental for climate due to the accompanying N_2O emissions, as indicated here for the common biofuels: rapeseed/bio-diesel, and corn/ethanol.

5 More favourable conditions for bio-energy production, with much lower nitrogen to dry matter ratios, resulting in smaller N_2O emissions, exist for special “energy plants”, for instance perennial grasses (Christian et al., 2006) such as switch grass (*Panicum virgatum*) and elephant grass (*Miscanthus* × *giganteus* hybrid), with a r_N of 7.3 g N/kg dry matter. The production of biofuel from oil palm (Wahid et al., 2005), with a r_N of 6.4 g N/kg dry matter, may also have moderately positive effects on climate. Other
10 favourable examples are ligno-cellulosic plants, e.g. eucalyptus, poplar and willow. However, in all cases, a complete life cycle analysis, including the effect of nitrogen, is necessary.

The importance of N_2O emissions for climate also follows from the fact that the agricultural contribution of 4.3–5.8 Tg N_2O -N/year gives the same climate radiative forcing
15 as that provided by 0.55–0.74 Pg C/year, that is 8–11% of the greenhouse warming by fossil fuel derived CO_2 . Increased emissions of N_2O will also lead to enhanced NO_x concentrations and ozone loss in the stratosphere (Crutzen, 1970). Further, NO is also produced directly in the agricultural N cycle. Adopting the relative yield of NO to N_2O of 0.8 (Mosier et al., 1998), and the agricultural contribution to the N_2O growth rate
20 of 4.3–5.8 Tg N_2O -N/year, the global NO production from agriculture is equal to 3.4–4.6 Tg N/year, about 20% of that caused by fossil fuel burning (Prather et al., 2001), affecting tropospheric chemistry in significant ways.

4.2 Application in life cycle analysis

An abridged analysis as presented above, yielding N/C ratios to indicate whether bio-
25 fuels are GHG-positive or GHG-negative, can not replace a full life cycle assessment. In recent years, a number of such assessments have become available (Adler et al., 2007; Kaltschmitt et al., 2000; von Blottnitz et al., 2006; Farrell et al., 2006; Hill et al., 2006). At this stage, we can not discuss the differences between these respective

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approaches, which also affect conclusions. But we may look into the release rate of N_2O -N used, presented as a function of applied fertilizer N. In these life cycle studies, release rates typically are based on the rates recommended by IPCC (2006) for “direct” emissions which were derived from plot-scale measurements (1% of the fertilizer N applied, or, in a previous version, 1.25%). Only a few studies (Adler et al., 2007) fully account for the “indirect” emissions also specified by IPCC (which, together with the direct emissions, add up to almost 2% of fertilizer N), whereas our global analysis indicates a value of 3–5%. Clearly, all past studies have severely underestimated the release rates of N_2O to the atmosphere, with great potential impact on climate warming. The effect of applying higher N_2O yields can be assessed using the openly accessible EBAMM model (Farrell et al., 2006). As N_2O release is a significant item in life cycle assessment, it is obvious that a strong increase may also shift the overall balance. This will be the subject of further studies.

5 Conclusions

As release of N_2O affects climate and stratospheric ozone chemistry by the production of biofuels, much more research on the sources of N_2O and the nitrogen cycle is urgently needed. Here we have shown that the yield of N_2O from fixed nitrogen application in agro-biofuel production is 3–5% N_2O -N, 3–5 times larger than assumed in current life cycle analyses, with great importance for climate. We have also shown that the replacement of fossil fuels by biofuels may not bring the intended climate cooling due to the accompanying emissions of N_2O . There are also other factors to consider in connection with the introduction of biofuels. Here we concentrated on the climate effects due only to required N fertilization in biomass production and we have shown that, depending on N content, the use of several agricultural crops for energy production can readily lead to N_2O emissions large enough to cause climate warming instead of cooling by “saved fossil CO_2 ”. What we have discussed is one important step in a life cycle analysis, i.e. the emissions of N_2O , which must be considered in addition to

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the fossil fuel input and co-production of useful chemicals in biofuel production. We have not yet considered the extent to which the high percentage of N-fertilizer which is not taken up by the plants, and the organic nitrogen in the harvested plant material, may stimulate CO₂ uptake from the atmosphere; estimates for this effect are very uncertain (Nadelhoffer et al., 1999; Townsend et al., 1996; Magnani et al., 2007). We conclude, however, that the relatively large emission of N₂O exacerbates the already huge challenge of getting global warming under control.

Appendix A

A1 Bio-ethanol production from corn

Yield=2.66 U.S. gallons per U.S. bushel (mean of values for wet and dry milling processes) (USDA 2002, cited in UK Dept for Transport, 2006)

$$\begin{aligned} &= 2.66 \times 3.785 = 10.07 \text{ litres ethanol/25.4 kg corn} \\ &\equiv 7.945 \text{ kg ethanol/25.4 kg corn} \\ &= 0.313 \text{ kg ethanol/kg corn.} \end{aligned}$$

C content of ethanol (C₂H₅OH, mol. wt. 46) by weight=24/46=522 g/kg.

C content of corn (r_C) $\cong 0.44$ g/g $\cong 440$ kg/tonne.

$$cv = (0.313 \times 522) / 440 = 0.37.$$

A2 Bio-diesel production from rapeseed

(i) the average oil yield is 45% (450 kg/tonne rapeseed) (E. Booth, SAC Aberdeen, personal communication)

(ii) the average composition of the oil is adequately represented by the triglyceride of the dominant fatty acid, erucic acid, i.e. (C₂₂H₄₁O₂)₃(C₃H₅), mol. wt. 1052, then

C content of the oil by weight=828/1052=0.787 kg/kg.

Thus the C content of the oil=(450×0.787)=354 kg/tonne rapeseed.

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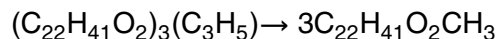
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The conversion to bio-diesel involves conversion to the methyl ester:



but the C content of the bio-diesel is almost unchanged from that of the natural oil:

mol. wt. of methyl ester=352, and

C content=(276/352)×450=353 kg/tonne rapeseed

Oil content of original rapeseed=45% (450 kg/tonne),

and non-oil components ≅550 kg/tonne, of which

– protein is 40% (≅220 kg/tonne original rapeseed), with a C content of 510 g/kg;

– the remainder (60%, ≅330 kg/tonne original rapeseed) is dominantly carbohydrate, (Colin Morgan, SAC Edinburgh, personal communication)

Thus the C content of the protein fraction in the original rapeseed=220×510/1000=112 kg/tonne; and the C content of the carbohydrate fraction (for which a C content of 440 g/kg can be adopted, as for grains)=330×440/1000=145 kg/tonne.

The overall C content of the original rapeseed ($r_C = C_{oil} + C_{protein} + C_{CHO}$)=354+112+145=612 kg/tonne.

$cv = 353/612 = 0.58$.

A3 Bio-ethanol production from sugar cane

Yield is 86 l dry ethanol (density 0.79 kg/l) per tonne sugar cane harvested at a water content of 72.5%, or 247 kg ethanol per tonne dry sugar cane (Macedo et al., 2004, as cited by JRC, 2007).

C content of ethanol (C₂H₅OH, mol. wt. 46) by weight=24/46=522 g/kg.

C content of dry sugar cane is determined by its structural material, cellulose, and its sugar content (polysaccharides: 440 g/kg; saccharose: 420 g/kg), we use $r_C = 430$ g/kg $cv = (0.247 \times 522) / 430 = 0.30$.

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Table 1. Relative warming derived from N₂O production for crops, crop residues, and forages used in the production of biofuel.

Crop	r _N (gN/kg dry matter)	relative warming (Meq/M)	type of fuel produced
Rapeseed	39	1.0–1.7	Bio-diesel
Wheat	22	1.3–2.1	Bio-ethanol
Barley, Oat	19	1.1–1.9	Bio-ethanol
Maize	15	0.9–1.5	Bio-ethanol
Sugar cane	7.3	0.5–0.9	Bio-ethanol
Residue			
Sugar beet leaves	25	1.5–2.4	Bio-ethanol
Root crops	16	0.9–1.6	Bio-ethanol
Forages, low N	15	0.9–1.5	Bio-ethanol
Forages, high N	27	1.6–2.6	Bio-ethanol

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