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#### UK emissions of the greenhouse gas nitrous oxide

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#### **Abstract**

Signatories of the Kyoto Protocol are obliged to submit annual accounts of their anthropogenic greenhouse gas emissions, which include nitrous oxide (N<sub>2</sub>O). Emissions from the sectors industry (3.8 Gg), energy (14.4 Gg), agriculture (86.8 Gg), waste water (4.4 Gg), land use and land use change (2.1 Gg) can be calculated by multiplying activity data (i.e. amount of fertiliser applied, animal numbers) with simple emission factors (Tier 1 approach), which are generally applied across wide geographical regions. The agricultural sector is the largest anthropogenic source of N<sub>2</sub>O in many countries, responsible for 75% of UK N<sub>2</sub>O emissions. Microbial N<sub>2</sub>O production in nitrogen fertilised soils (27.6 Gg), nitrogen enriched waters (24.2 Gg) and manure storage systems (6.4 Gg) dominate agricultural emission budgets.

For the agricultural sector, the Tier 1 emission factor approach is too simplistic to reflect local variations in climate, ecosystems and management, and is unable to take into account some of the mitigation strategies applied. This paper reviews deviations of observed emissions from those calculated using the simple emission factor approach for all anthropogenic sectors, briefly discusses the need to adopt specific emission factors

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that reflect regional variability in climate, soil type and management, and explains how

bottom up emission inventories can be verified by top-down modelling.

Keywords: emission factors, nitrogen fertiliser, manure, atmospheric deposition,

leaching, land use change

**Short title**: UK N<sub>2</sub>O inventory

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1 Introduction

25-40% below 1990 levels by 2020 (1). The EU-27, responsible for about 10% of the global annual greenhouse gas emissions, have agreed to reduce emissions by 20% by 2020 and by 80% by 2050. They predict that this reduction can be achieved by improved efficiency, new low carbon technologies, renewable energy and abatement strategies (2). To monitor progress, greenhouse gas emissions are reported annually and are submitted to the UNFCCC (United Nations Framework Convention on Climate Change) by individual countries and the EU-27. Emissions of CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O and the fluorinated greenhouse gases are reported for all anthropogenic sources, which fall into the categories (a) energy, (b) industrial processes and product use, (c) agriculture, (d)

To keep the projected global average temperature increases within 2°C the EU of pre-

industrial levels, developed countries need to reduce their greenhouse gas emissions by

internationally agreed protocols often using simple equations, represented by a 'Tier 1'

forestry and other land use, (e) waste and (f) other sources (this includes indirect

emissions as a result of atmospheric N deposition). The reporting follows

methodology (1;3). The UK in common with most Annex one countries uses IPCC's

1996 reporting guidelines (3;4), to prepare emission inventories for the first Kyoto

commitment period (2008-2012). It is likely that there will be a general move to the

slightly modified 2006 guidelines (5), after the 2012 inventory reporting.

The basic Tier 1 approach is useful to compare anthropogenic emissions from different countries, but does not capture the well documented variations across climate regions and in agricultural management or combustion technologies, the potential effects of mitigation practices (e.g. 6)). Specific methodologies, country/regional, technology specific emission factors (Tier 2) and a range of simple to complex process-based models (Tier 3) have been developed to address this problem (e.g. (7)). The IPCC recommends such improved methods to be used alongside the Tier 1 methodologies, as long as these methods are transparent and documented. Tier 2 methodology is applied to some of the IPCC categories by some EU countries. For example the Netherlands apply a country-specific emission factor for CH<sub>4</sub> emissions from animal production, UK CH<sub>4</sub> emission factors are related to animal live weight, milk production and milk fat content, and Sweden uses a mixture of default and national emission factors for different sub-sources of direct N<sub>2</sub>O emissions.

Nitrous oxide is one of the main greenhouse gases and contributes 10% (0.16 W m $^{-2}$ ) of the total global anthropogenic radiative forcing (1). Due to the decline in CFCs and HCFCs emissions  $N_2O$  is also now the most important ozone depleting molecule in the stratosphere (8). The annual global  $N_2O$  budget is currently estimated at 27.8 Tg  $N_2O$  y $^{-1}$  (range 13.4 - 43.5); microbial processes in soils and aquatic systems are responsible for 89% of the annual global  $N_2O$  emissions. Of this total, natural soils and oceans make up the largest components (37% and 21%, respectively) (1); and various estimates suggest that tropical forest soils are globally the largest natural source of  $N_2O$  (2.11 Tg  $N_2O$  y $^{-1}$  (range 1.38 -3.72) (9), due to high turnover rates and the wet, warm environment conducive to  $N_2O$  production. The anthropogenic emissions from agricultural soils are the third most important source (15.8%, or 4.4 Tg  $N_2O$  y $^{-1}$ ; range 2.7-7.5) and those from rivers, estuaries and coastal zones next (9.6%, or 2.7 Tg  $N_2O$ -N y $^{-1}$ ; range 0.8 - 4.6).

In the UK and similar densely populated countries in temperate climates,  $N_2O$  emissions are dominated by anthropogenic sources. Biological emissions from the agriculture sector are the main anthropogenic source of  $N_2O$  and in 2008 accounted for approximately 75% of the annual  $N_2O$  emissions, both in the EU-27 (877.0 Gg) and in the UK (87.6 Gg) (10). The remaining source categories: energy, industry, LULUCF (Land Use, Land-Use Change and Forestry) and waste are similarly proportioned in the

UK (13.2%, 6.6%, 1.8%, 3.9%) and the EU-27 (9.5%, 10.9%, 0.9%, 3.8%) (10). Overall the UK emits about  $1/10^{th}$  of the EU-27 annual N<sub>2</sub>O emissions.

Total UK emissions have steadily declined from  $210 \text{ Gg N}_2\text{O}$  in  $1990 \text{ to } 111 \text{ Gg N}_2\text{O}$  in 2009 (Figure 1). This is mainly due to a sharp decline in industrial emissions by 90%, and a decline in agricultural emissions by 23% (Table 1). Below we discuss the anthropogenic emission inventory in detail, using the UK situation as an example.

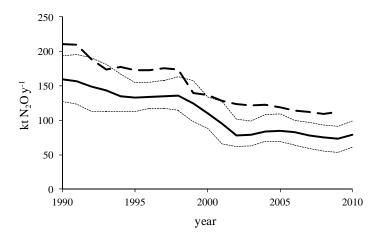


Figure 1: Nitrous oxide emission estimates for the UK, calculated by the NAME-Inversion model from atmospheric concentration measurements at Mace Head on the west coast of Ireland (solid line, dotted lines represent the 5% and 95% uncertainty estimates) and by the bottom –up approach using IPCC Tier 1 emission factors (dashed line) (11-Annex 10), (12).

# 2 Industrial processes

Industrial production of nitric acid and adipic acid contributed 37% (79 Gg) to the UK's annual  $N_2O$  emissions in 1990, but since then have declined by 90% and now only contribute 3% (3.8 Gg) of the total UK annual  $N_2O$  emission. This reduction was achieved by lowering production rates and removing  $N_2O$  from adipic acid production by thermal decomposition, and by lowering the temperature during nitric acid production. Similar reductions were achieved by other EU countries (13). Currently nitric acid is manufactured at two locations in the UK and adipic acid production in the UK stopped in April 2009 (11). Industrial emissions are monitored by the producers and Environment Agency before being reported to the Pollution Inventory. The emission factor uncertainty for adipic acid production is 15% and for nitric acid production 230% (11-Annex 7).

#### **3** Fuel combustion

The energy sector (stationary and mobile fuel combustion) contributed 13% of the total UK  $N_2O$  emissions in 2009, and has declined by 28% since 1990 due to effective abatement measures and improved combustion efficiencies. Emissions are calculated from fuel combustion statistics and emission factors. The emission factor uncertainty ranges from 33 to 170% for fuel combustion processes and 195% for other combustion processes (11-Annex 7).

### 4 Biological sources

Microbial production in soils and aquatic systems (including waste waters) primarily via nitrification and denitrification processes is the largest source of  $N_2O$ . These processes are ubiquitous and many microbial species have the necessary suite of enzymes to produce  $N_2O$  (14). The rate of  $N_2O$  production and emission to the atmosphere depends on climate (rainfall and temperature), soil type, availability of mineral nitrogen, the redox potential and pH of the soil/aquatic environment and for denitrifiers and heterotrophic nitrifiers the availability of simple carbon compounds ((15;16;17). The combined requirements of the above variables results in highly variable emissions, both in space and time. Consequently biological emissions calculated by the IPCC Tier 1 methodology are very uncertain. The combined uncertainty range as a percentage of source category is 424% for agricultural soils, 414% for manure handling and 401% for waste water handling (11-Annex 8).

# 4.1 Waste water handling

Waste water handling (i.e. sewage treatment plants) contributes 4% of the UK  $N_2O$  emissions, produced in activated sludge by nitrification and denitrification processes as discussed in detail by (18). Emissions are calculated from per capita protein consumption, using the assumption that 1% of sewage N produced is emitted as  $N_2O$  (3), but measurements from treatment plants have shown that emission factors range from 0 to 15% (15). Nitrous oxide emissions can be minimised by maintaining high  $O_2$  and small nitrite concentrations during the aerobic part of the waste water treatment,

and by maintaining a high C/N ratio during the anaerobic stage (15).  $N_2O$  emissions also occur when sewage sludge is applied to land (11). The emission factor uncertainty for waste water handling is 401% (11-Annex 7).

Table 1: The UK N<sub>2</sub>O emission inventory in 1990 and 2009 (Gg N<sub>2</sub>O) (11)

		2009	
Non biological sources			
Industrial processes	79.52	3.82	
Fuel combustion inc. waste incineration <sup>1</sup>	19.89	14.35	
Biological sources			
Waste water handling	4.04	4.44	
Agriculture: direct emissions			
Manure storage	8.67	6.44	
Agricultural soils:			
Synthetic fertilisers	28.89	18.98	
Animal manure & sewage sludge applied to	9.41	8.65	
Pasture range and paddock manure	16.07	13.29	
Crop residue	7.17	8.46	
N-fixing crops	0.85	0.92	
Cultivation of histosols	0.49	0.49	
Improved grassland	0.54	0.58	
Agriculture: indirect emissions			
Atmospheric deposition	6.42	4.77	
N leaching and runoff	33.69	24.21	
LULUCF			
Forests	0.02	0.01	
Land use change to cropland <sup>2</sup>	2.54	2.05	
Total	218 <sup>3</sup>	$112^{3}$ .	

waste incineration = 0.14 & 0.15 Gt in 2009 & 1990; <sup>2</sup> for 1990 this included 0.02 Gt land use change to wetland and settlement; <sup>3</sup> the overall estimate has been rounded to avoid unjustifiable precision.

### 4.2 Agricultural activities

Since the decline in industrial emissions the agricultural sector has been the largest source of  $N_2O$  in the UK and is responsible for more than  $^{3}4$  of the total annual emissions (78% in 2009) (11). The main agricultural sources are fertiliser and manure application to soils (23%) and indirect emissions to the atmosphere of  $NH_3$  and  $NO_x$ , and to waters of  $NO_3^-$ , which downstream or when redeposited can be nitrified or denitrified and produce  $N_2O$  (26%). Nitrogen excretion onto pasture, range and

paddocks accounted for 12%, crop residue incorporation for 8% and manure storage systems for 6% of the UK's  $N_2O$  emissions in 2009 (Table 1). The spatial distribution of  $N_2O$  emissions across the UK clearly shows largest emissions in regions dominated by livestock production, the grazed grassland regions in the high rainfall areas in the west of GB and in Ireland, beef production in northeast Scotland and intensive poultry and pig production in eastern England (Yorkshire, Lincolnshire and East Anglia) (Figure 2). Overall agricultural emissions have decreased steadily by ~23% since 1990, mainly due to reduced N fertiliser application rates and a decrease in livestock numbers (19).

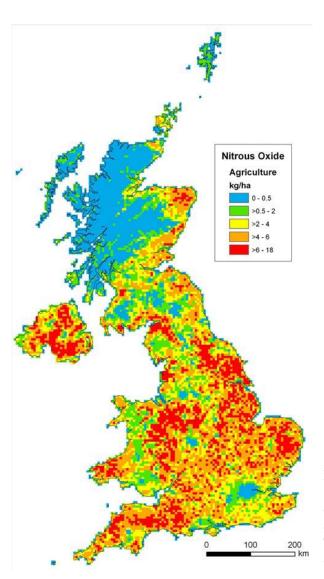


Figure 2: UK N<sub>2</sub>O emissions from agriculture (soils, direct and indirect emissions, and manure management), calculated for every 5 km<sup>2</sup> using the IPCC Tier 1 methodology and using 2009 activity data (20).

#### 4.2.1 Agricultural soils: direct emissions

Nitrous oxide emissions as a result of mineral fertiliser, animal manure and sewage sludge applied to soils, incorporated crop residues, biological N fixation in legumes and improved grassland are calculated assuming that 1% of the N applied, incorporated or fixed is emitted as  $N_2O$ ) (3). Only the organic rich histosols have a much larger emission factor of 8%. In these, high mineralisation rates provide a constant supply of mineral N and carbon ideal for denitrification. However, only a small area of histosols is farmed in the UK and emissions from these accounted for less than 1% of the total  $N_2O$  emissions in 2009.

Under optimal conditions, fertiliser application stimulates a rapid rise in  $N_2O$  emissions, which usually only lasts for 1 - 3 weeks (21) (Figure 3). However, in addition to fertiliser rate, which is the only factor accounted for by the Tier 1 emission inventory, the onset, magnitude and length of fertiliser induced emissions depends on rainfall, particularly the timing of fertiliser application in relation to rainfall and temperature, soil type, organic matter content in mineral soils, drainage and fertiliser type ((16;22;23), . Consequently  $N_2O$  emissions show large seasonal and interannual variations (6;24). An example of such variations is reported here from long-term monitoring of greenhouse gases from an intensively managed, mostly sheep grazed, grassland (25;26). The grassland was fertilised three times during spring/early summer, each time with  $50 - 70 \text{ kg NH}_4\text{NO}_3\text{-N}$  ha  $\text{y}^{-1}$ . Weekly  $N_2O$  fluxes were measured using static chambers throughout the year, with daily measurements immediately after fertiliser application, between August 2006 - November 2010 (26). Results for 2007 are shown in Figure 3.

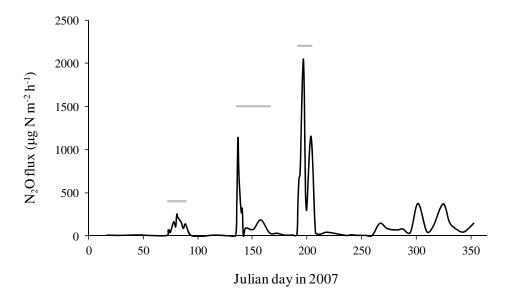


Figure 3:  $N_2O$  fluxes from a sheep grazed grassland in SE Scotland. Data are median emissions from 8 chambers. The fertiliser induced emission peaks are identified by grey bars, the start of these bars mark the addition of  $NH_4NO_3$  (69, 52, 52 kg N ha<sup>-1</sup> y<sup>-1</sup> on Julian days 73 (14<sup>th</sup> March), 136 (16<sup>th</sup> May), and 192 (11<sup>th</sup> July), respectively) (Jones & Skiba, unpublished data).

Fertiliser induced emissions were short-lived (< 15 days) and varied in magnitude. Collectively the three emission events were responsible for 52% of the total annual emission (7.5% (14-30 March) + 16.3% (16-31 May) + 30% (11-23 July) of the total annual emissions. In 2008, fertiliser induced emission peaks accounted for 65% and in 2009 for 27% of the annual flux. For 2007-2009 these seasonal and interannual variations could be modelled reasonable well by variations in the rainfall amount from 5 days before fertilisation until the end of the fertiliser induced emission peak, plus the average air temperature during the emission period (Figure 4).

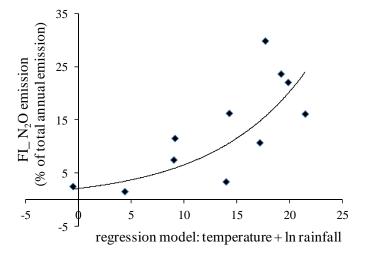


Figure 4: The dependence of the magnitude of the  $N_2O$  emission peaks induced by N fertiliser application (FI  $N_2O$  emissions) on the synergistic influence of rainfall (cumulative rainfall (mm) 5 days before and during the fertilised induced  $N_2O$  emission peaks) and temperature (average air temperature during the fertilised induced  $N_2O$  emission peaks), calculated using a multi-linear regression model (% of total annual flux = -8.81 + 3.18 ln rain + 0.983 temp,  $r^2 = 55.9\%$ , p < 0.05) (x-axis). The data are from weekly flux measurements, 2007-2009, at the sheep grazed grassland (see Figure 3) (Jones & Skiba, unpublished data).

The IPCC Tier 1 emission factor approach assumes a linear relationship between N fertiliser application rate and emissions, but there is good evidence to suggest that this is not the case, with emissions rising more steeply beyond optimum fertiliser application rates (22;27;28). A better understanding of this relationship will allow us to determine the difference between the currently applied economic optimum fertiliser rate and environmentally optimum rates. In some circumstances it may be more appropriate to report emissions on a unit yield basis rather than unit area. (29). It is also apparent that the chemical form of fertiliser N (28) and crop type (7) can influence emissions. The latter study used smaller emission factors for NH<sub>4</sub><sup>+</sup> compared to NO<sub>3</sub><sup>-</sup> based fertilisers, and smaller emission factors for arable land compared to grassland to calculate N<sub>2</sub>O emissions for European agricultural land.

### 4.2.2 Agricultural soils: application of slurry, sewage sludge and manure

Most of the  $N_2O$  generated from the manure management continuum (viz; livestock buildings, manure stores, manure treatment and land spreading) occurs after it has been spread onto soils (30). Resulting emissions depend on the soil and environmental factors discussed previously, but also on the C and N content and N forms of the manure, especially the decomposability of the organic material and the ratio of ammonium to total N. These factors vary, depending on the origin of the manure, storage conditions, treatment process that the manure may be subjected to, and the climate conditions during storage (31). The Tier 1 IPCC methodology uses the same  $N_2O$  emission factor for manure spreading as for mineral fertilisers: 1% of the N content of manure is emitted as  $N_2O$  (3). This approach does not reflect the large differences in the manure's chemical composition, its C/N ratio and method of application. For example, the emission factor from pig slurry (7-14%) was larger than from cattle slurry (2-3%) (32). This was a result of the larger content of ammoniacal N in the pig manure. Comparing  $N_2O$  emission factors from mineral fertilisers with those from animal manures has

provided contrasting results. For example, (33) has shown that  $N_2O$  emissions from a clay loam soil cropped with potatoes were significantly smaller (by 23%) when fertilised with pig than with urea. However, (34) reported no conclusive differences when applications of cattle slurry with and without calcium nitrate to grasslands on sandy soils were compared.

#### 4.2.3 Manure management: storage of animal waste

Part of the livestock manures require containment and storage prior to spreading, to ensure they can be spread when there is a crop demand for the nutrients they contain. This is especially important in those areas designated in the UK as Nitrate Vulnerable Zones (NVZs). Liquid livestock manure, i.e. slurries, is stored in lagoons or tanks, whilst solid manures are stacked in heaps prior to spreading. The anaerobic conditions and high C and N concentrations of lagoons, slurry tanks and dung heaps provide perfect conditions for methanogenesis and denitrification of a suitable N source (35;36). In the UK, manure storage systems are responsible for 15% of the total agricultural CH<sub>4</sub> emissions, but only for 6% (6.4 Gg y<sup>-1</sup>) of agricultural N<sub>2</sub>O emissions (11). Nitrous oxide emissions are very variable (37) and depend on the C and N composition of the manure (which itself depends on the animal species and diet), the temperature and storage method and length of storage. The IPCC emission inventory assumes no N2O emissions from slurry based livestock buildings and slurry stores, because the slurry remains in an anaerobic state and there is little opportunity for NH<sub>4</sub><sup>+</sup> to be nitrified (untreated slurry contains no or very little NO<sub>3</sub><sup>-</sup>) (30). However, as reviewed by Chadwick(30), crust development can provide a zone of nitrification and hence a source of NO<sub>3</sub> which can subsequently by denitrified, so the crust may be a source of N<sub>2</sub>O (38). Amon(35) reported the effects of cattle slurry treatment on N<sub>2</sub>O emissions during storage. Untreated slurry emitted 24 g N<sub>2</sub>O m<sup>-3</sup> slurry, anaerobically digested slurry emitted 31.2 g N<sub>2</sub>O m<sup>-3</sup> and aerated slurry 54.2 g N<sub>2</sub>O m<sup>-3</sup>.

The IPCC emission factor for solid storage systems is 0.02 kg N<sub>2</sub>O/kg N in the manure (3). In reality emissions from solid manure stores are very variable. Typical emissions range between 1% and 4.3% of the total N stored in cattle and pig farmyard manure heaps were reported by Chadwick(30). Emissions depend on the C/N ratio, dry matter content at the start of storage (39) and storage conditions (31;40;41). Although solid

manure stores are only a small component of the total agricultural  $N_2O$  emission budget, they are  $N_2O$  hotspots in the landscape, easily detectable by fast response high resolution laser systems (42).

#### 5. 1 Indirect N<sub>2</sub>O emissions: atmospheric deposition

The rise in rates of atmospheric N deposition is directly linked to the rise in population growth and demand for food and industrialisation (43). Over the last century a total of 29 Mt of N was deposited to the UK, which is an equivalent of 1.2 t N deposited to every ha in the UK (44). Recent deposition rates were about 400 Gg N  $y^{-1}$  in 2006 with approximately 50% each of reduced (NH $_x$ ) and oxidised (NH $_x$ ) N (45). Agriculture is by far the largest source of NH $_x$ , predominately as NH $_3$  and NH $_4^+$  emitted from livestock, manure storage and N fertilisers. Combustion processes are the main source of NOx and 10-20% of the global total is emitted from microbial nitrification in soils (46). These gases are deposited downwind of the original source as a gas, aerosol (dry deposition) or through precipitation (wet deposition) and unintentionally fertilise 'natural' ecosystems. Consequences are changes in species composition (47) and increased rates of NO and N $_2$ O emissions (48;49).

Agriculture related indirect  $N_2O$  emissions from atmospheric deposition are calculated by the IPCC Tier 1 methodology assuming that 10% (range 3 – 30%) of mineral fertiliser and 20% (range 5 – 50%) of the organic N fertiliser or excreta from grazing livestock are volatilised and 1% (range 0.2 - 5%) of the deposited N is emitted as  $N_2O$  (3). In the UK this sector accounts for 4.8 Gg  $N_2O$  in 2009 (11). Indirect emissions due to  $NO_x$  and  $NH_3$  emissions from non-agricultural sources do not need to be included in national inventories, but can be calculated assuming that 1% of the  $NO_x$ -N and  $NH_3$ -N emitted is deposited as  $N_2O$  (3). This is likely to overestimate this source as only a third of the UK's NOy emissions (580 Gg in 2005) are deposited to the UK (45); the rest is deposited to other countries and the seas. The more accurate approach would be to calculate N deposition induced  $N_2O$  emissions directly from atmospheric N deposition rates.

Soil microbes do not distinguish between N applied via fertilisation or atmospheric deposition so one can assume that the emission factor applied to calculate fertiliser

induced emissions is applicable also to atmospheric deposited N. Thus 1% of the 400 Gg N annually deposited (4 Gg N) is emitted as  $N_2O-N$  (6.3 Gg  $N_2O$ ). Similar to fertilised soils, there are large variations in emissions in relation to atmospheric deposition due to variations in soil type, vegetation, climate and rate of N deposition; the relationship may not be linear (Figure 5), and should be further investigated.

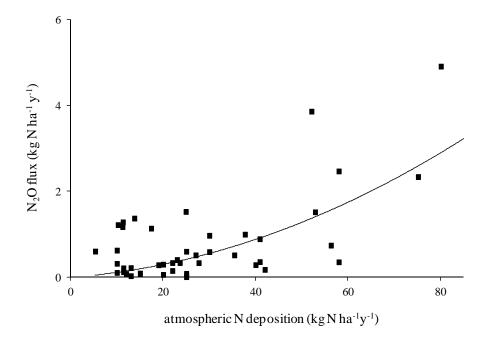


Figure 5: The relationship between  $N_2O$  and N deposition from forests, heather and grass moorlands on mineral soil in Great Britain. Locations with N deposition rates  $> 40 \text{ kg N ha}^{-1} \text{ y}^{-1}$  were close to NH<sub>3</sub> emitting livestock farms. These measurements were made by CEH between 1991 and 1997 (48;49).

#### 5. 2 Indirect N<sub>2</sub>O emissions: NO<sub>3</sub> leaching and runoff

A proportion of fertiliser, manure, sewage sludge and excreta N is lost through leaching and surface runoff to groundwater, rivers and estuaries, where  $N_2O$  is produced via nitrification and denitrification in sediments and the water columns ((50)(51) . Nitrous oxide is highly soluble in water (0.15 g/100 ml (15 °C), and often supersaturated by several orders of magnitude of ambient atmospheric  $N_2O$  concentrations. Emissions to the atmosphere are highly variable and depend on the solubility, the water to air pressure difference and transfer velocity (which increases with windspeed and turbulence). For example maximum  $N_2O$  emissions were measured where field drains feed into a river (52). In general,  $N_2O$  emissions from aqueous systems are directly

related to N concentrations in the water (53). The Tier 1 IPCC approach assumes that 30% (range 10-80%) of the fertiliser or manure N applied to soils is lost to the water bodies when rainfall > 0.5\* pan evaporation (616 Gg in 2009) and that 0.75% (range 0.05 -2.5%) of the leached N is emitted as N<sub>2</sub>O, 0.25% each from groundwater and surface drainage, rivers and estuaries (3). The IPCC has reduced the emission factor for these sources from 2.5%, based on experimental evidence (50). However, (54) et al (2011) estimated that 0.75% (range 0-0.9%) of the dissolved inorganic nitrogen input to rivers is emitted as N2O, based on measurements in the USA. The UK still uses the IPCC 1996 default value of 2.5% and N<sub>2</sub>O emissions due to leaching and runoff amount to 24.2 Gg y<sup>-1</sup> (Table 1). The new IPCC 2006 emission factor (0.75%) calculates indirect N<sub>2</sub>O emissions of aqueous bodies as only 4.6 Gg y<sup>-1</sup> (2009), with 1.5 Gg emitted from each the groundwaters, rivers and estuaries. Studies are needed to identify the most appropriate emission factor for the UK. An annual emission of 1.5 Gg N<sub>2</sub>O from estuaries, calculated with the newer 0.75% emission factor is in good agreement with a recent measurement based estimate by (51) (1.9  $\pm$  1.2 Gg N<sub>2</sub>O y<sup>-1</sup>; the uncertainty is 64%).

# 6 Land use and Land use change (LULUCF)

Nitrous oxide and CH<sub>4</sub> emissions from forests, and land use change from and to forests, have only very recently been included in the LULUCF inventory. Annual emissions from the LULUCF sector are very small (<2% of UK 2009 emissions). Anthropogenic activities leading to N<sub>2</sub>O emissions considered are N fertilisation to newly planted forest and emissions due to soil drainage, but reporting on this latter activity is currently not mandatory. Direct fertiliser-induced N<sub>2</sub>O emissions from forests are calculated using the same emission factor as used for fertilised agricultural soils. Fertiliser is rarely applied to forests, and so far the annual inventory has been set at zero. Emissions due to forest management, such as felling and thinning are not considered, although these activities could potentially change N<sub>2</sub>O emission rates by altering the soil water content due to the absence of trees (felling) or reduction of shading (thinning). Indirect emissions due to atmospheric N deposition is not included either, in spite of observations that deposition rates to forests can be 2 to 3 fold larger than to shorter vegetation (44) and can be especially large to small forests in intensively managed agricultural areas (48;49).

Conversion of one land use to another invariably requires ploughing and perhaps drainage, and forest converted to cropland or grassland needs to be clear felled. These activities stimulate soil organic matter mineralization and increase soil mineral N concentrations. In the absence of plants there is no competition for this newly available N, thereby maximising substrate availability for microbial nitrification and denitrification and release of N<sub>2</sub>O. The few studies on the effect of clear-felling on greenhouse gas fluxes revealed that clear-felling resulted in a pulse of N<sub>2</sub>O, NO and CO<sub>2</sub> emissions (55;56;57) Morison *et al.* (58) have compiled the data relevant to the UK, which demonstrates that N<sub>2</sub>O emissions from forests are influenced by soil type and that clear-felling may increase N<sub>2</sub>O emissions in the short term (Figure 6).

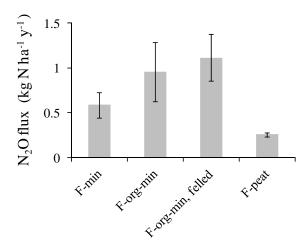


Figure 6: Comparison of  $N_2O$  emissions from British forests (F) grown on different soil types. Average and standard error of the mean for F-min = 7 forests/woodland plantations on mineral soil, F-org-min = 5 forests on peaty podzols and peaty gleys, F-org-min, felled = 2 clear-felled plantations, F-peat = 3 plantations on peat (58)(Morison *et al.* 2011).

Growing interest in perennial bioenergy crops is likely to increase areas undergoing land use change. Perennial bioenergy crops are favoured over annual crops because they sequester carbon over 15-20 year periods and have very low N fertiliser requirements, thus much lower  $N_2O$  emissions (59;60). However, they are economically productive for a finite period only, at the end of which they will be removed. Removal by clear-felling is one option, but as shown in Figure 6, there are too few studies to predict the rate and length of increased emissions of  $N_2O$  and other greenhouse gases triggered by clear-felling. There is urgent need to understand clear-

felling effects on greenhouse gas emissions, and account for these in life cycle analyses (60).

The total area in the UK converted to wetlands is restricted to small insignificant areas of newly created riparian zones, along rivers in NVZs, or reinstating peat wetlands. This activity has a potential to decrease  $N_2O$  emissions if the soil water-filled pore space can be maintained above 90% (16). Under such conditions anaerobic conditions and accumulation of soil organic matter content will favour denitrification to proceed to  $N_2$  rather than stop at  $N_2O$  production, which is generally the case in more aerobic soils. The uncertainty of this source is very high, and depends on maintaining a high water table. In spite of lack of data to calculate  $N_2O$  emissions from the LULUCF sector, surprising small levels of uncertainty of 20% were calculated for all LULUCF categories, except for the conversion to croplands, which have an uncertainty of 50% (11-Annex 7)

#### 7 Natural emissions

Nitrous oxide emissions from 'natural' ecosystems are not included in the UK's anthropogenic emission inventory. Nitrogen inputs and losses, including N<sub>2</sub>O to and from 'natural' environments tend to be small (26). For a country like the UK, where only about 20% of the land area is not used for agriculture or settlements (12% is in forest, the rest are heaths, moorlands, bogs and montane ecosystems) it is difficult to separate natural emissions from those resulting from enhanced N deposition rates. In a review of greenhouse gas fluxes from natural ecosystems Dalal and Allen(61) calculated average emissions of 1.57 kg N<sub>2</sub>O ha<sup>-1</sup> y<sup>-1</sup> for temperate forests and 0.55 kg N<sub>2</sub>O ha<sup>-1</sup> y<sup>-1</sup> for temperate grasslands. Based on the above mentioned emission rates natural emissions in the UK may contribute an extra 5.8 Gg of N<sub>2</sub>O to the anthropogenic emission inventory.

### 8 Developing Tier 2 emission factors for the UK for agriculture

The current largely Tier 1 based  $N_2O$  inventory methodology used in the UK is a fairly blunt tool. For the main  $N_2O$  source, the agricultural sector, there is now a requirement

to produce a reporting tool that better reflects the climatic and soil variability and production systems (including N management) throughout the UK. Importantly, any new approach should explicitly account for mitigation practices so that their uptake is fully reflected in emission estimates. An improved inventory approach will not only be used for reporting UK agricultural N<sub>2</sub>O emissions to the UNFCCC, but will also allow the UK Government to track progress against the challenging targets it has published in its low carbon transition plan, i.e. an 80% reduction in greenhouse gas emissions (CO<sub>2</sub> equivalents) by 2050 (2), and assist the agricultural industry to monitor progress against the sector roadmaps. Tier 2, country specific emission factors will need to be generated through a carefully co-ordinated approach to modelling, reviews of existing literature and experiments. These activities will provide temporally and spatially disaggregated direct and indirect N<sub>2</sub>O emissions factors for the major sources of N (fertiliser, dung and urine, and livestock manure applications to land). The efficacy of key potential mitigation methods will need to be tested and introduced into the new inventory structure, e.g. use of nitrification inhibitors. The complexity of the new inventory will, by necessity, be increased, not only through the increased level of disaggregation, but also through our growing level of understanding of the controls on N<sub>2</sub>O emissions, and how these are influenced by on-farm management decisions.

The development of Tier 2 emission factors is an interesting scientific endeavour *per se*, but represents only one part of the inventory calculation. Activity data, e.g. livestock numbers, annual N excretion by different livestock, knowledge of the grazing season length, application rates of different N fertiliser forms etc., are just as important in determining the total source of  $N_2O$  from any region and at any point in time. Thus, a hugely important component of the development of a Tier 2 approach is to ensure that this information is available at the most suitable level of disaggregation (in both time and space). Indeed, the spatial and temporal availability of appropriate activity data is the limiting factors in the ability to generate robust estimates of  $N_2O$  emissions at a given spatial scale.

Given the increased complexity of producing a reporting tool that better reflects the range of soil, climate and farming systems in the UK (that a Tier 2 approach requires), it is not necessarily the case that the overall uncertainty value for the annual UK agricultural  $N_2O$  emission total will be less than the current Tier 1 estimate, i.e. +-250%, although it is very much hoped that it will. What is of key importance, is that the

revised approach will allow the uncertainty to be apportioned to the different steps and parts of the inventory, allowing us to focus future resources on research (perhaps improved emission factors or /and improved spatial and temporal activity data) to reduce those uncertainties. Section 9 describes how the new (bottom up) inventory could be verified, by a top down modelling approach.

### 9 Verification of the UK N<sub>2</sub>O emission inventory

In order to provide verification of the UK greenhouse gas inventory the UK government maintains a high-quality remote observation station at Mace Head on the west coast of Ireland. Mace Head reports high-frequency concentrations of the key greenhouse gases (62). A Lagrangian dispersion model NAME (Numerical Atmospheric dispersion Modelling Environment) (63;64) driven by three-dimensional modelled meteorology is used to interpret the observations. NAME determines the history of the air arriving at Mace Head at the time of each observation. Deviation from the baseline is used to estimate the N<sub>2</sub>O source strength of the UK and regions of NW Europe. This NAME-inversion methodology uses an iterative best-fit technique which searches a set of random emission maps to determine the one that most accurately mimics the Mace Head observations (65). The 'top-down' NAME-inversion estimates of UK emissions 1990-2010 are compared to the 'bottom-up' greenhouse gas inventory estimates and are shown in Figure 1. The median NAME-inversion estimates are approximately 30-40 kt lower than the greenhouse gas inventory estimates throughout the whole time period. The trends in the time-series are in good agreement with both showing declining UK totals. The greenhouse gas inventory estimates show a sharp decline (40 Gg) between 1998 and 1999 in line with the introduction of the clean technology at an adipic acid plant in Wilton, north east England. The NAME-inversion estimates, with a longer averaging period, show a more gradual decline from 1998 to 2003 but the overall reduction is similar.

More direct measurements of  $N_2O$  and other greenhouse gas emissions from aircraft have also used inversion methods to deduce the UK source strength and its spatial distribution (66). Improved validation of the UK greenhouse gas emission inventories could be provided by a network of tall towers monitoring greenhouse gas concentrations across the UK and aircraft measurements.

#### 10 Conclusions

The above account of the deviations of observed N<sub>2</sub>O emissions from those calculated using the Tier 1 emission factor approach clearly shows that this methodology is too simplistic to reflect regional variations of biologically produced N<sub>2</sub>O emissions or provide the best estimate of the UK source strengths. The reasonably good agreement of the bottom-up emission factor and top down inverse modelling approaches imply that total UK N<sub>2</sub>O emissions may be accounted for adequately, but the attribution to individual sources using the bottom up methodology within the agricultural sector needs to be refined taking into consideration regional variations in climate, soil properties and seasonal agricultural management. Developing a methodology that can account for such variations is not a trivial task. In spite of the wealth of N<sub>2</sub>O emission measurements made in the last 20 years, there are still not enough long-term data sets to provide the information needed to develop emission factors for the range of combinations of different climate zones or soil types and N sources. Modelling is required to aid the interpolation between measured scenarios. Adopting Tier 2 methodologies requires detailed knowledge of variations of emissions in relation to easily measureable activity data. All sectors (energy, industry, wastewater, agriculture, LULUCF) would benefit from adopting Tier 2 emission accounting. But in the first instance, Tier 2 methods need to be developed for the largest N<sub>2</sub>O emitter, the agricultural sector. A Tier 2 approach should provide a more transparent and accurate picture of N<sub>2</sub>O emissions, capable of reflecting changes in soil and N management and take explicit account of mitigation strategies. Improved verification of the new approach to inventory reporting could be obtained by monitoring atmospheric N<sub>2</sub>O concentrations from a more extensive tower network across the UK.

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