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Inventories and scenarios of nitrous oxide emissions

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Abstract

Effective mitigation for N₂O emissions, now the third most important anthropogenic greenhouse gas and the largest remaining anthropogenic source of stratospheric ozone depleting substances, requires understanding of the sources and how they may increase this century. Here we update estimates and their uncertainties for current anthropogenic and natural N₂O emissions and for emissions scenarios to 2050. Although major uncertainties remain, ‘bottom-up’ inventories and ‘top-down’ atmospheric modeling yield estimates that are in broad agreement. Global natural N₂O emissions are most likely between 10 and 12 Tg N₂O-N yr⁻¹. Net anthropogenic N₂O emissions are now about 5.3 Tg N₂O-N yr⁻¹. Gross anthropogenic emissions by sector are 66% from agriculture, 15% from energy and transport sectors, 11% from biomass burning, and 8% from other sources. A decrease in natural emissions from tropical soils due to deforestation reduces gross anthropogenic emissions by about 14%. Business-as-usual emission scenarios project almost a doubling of anthropogenic N₂O emissions by 2050. In contrast, concerted mitigation scenarios project an average decline of 22% relative to 2005, which would lead to a near stabilization of atmospheric concentration of N₂O at about 350 ppb. The impact of growing demand for biofuels on future projections of N₂O emissions is highly uncertain; N₂O emissions from second and third generation biofuels could remain trivial or could become the most significant source to date. It will not be possible to completely eliminate anthropogenic N₂O emissions from agriculture, but better matching of crop N needs and N supply offers significant opportunities for emission reductions.

Keywords: climate change, greenhouse gases, N₂O, nitrogen cycle, ozone depleting substance, representative concentration pathways, RCPs

1. Introduction

As the third most important anthropogenic greenhouse gas and the largest remaining anthropogenic stratospheric ozone depleting substance currently emitted, nitrous oxide (N₂O) is one of the most important forms of nitrogen (N) pollution (Ravishankara *et al* 2009, Ciais *et al* 2013). Excess N pollution has been identified as one of the three global environmental issues whose ‘planetary boundary’ has been surpassed (Rockström *et al* 2009). Once an N atom is in a reactive form, it

can contribute to a number of cascading environmental problems as it is transported through terrestrial and aquatic ecosystems and into the atmosphere (Galloway *et al* 2003). Effective mitigation for N₂O emissions requires understanding of the sources and how they may increase this century.

N₂O is a by-product of several fundamental natural reactions of the N cycle: nitrification, denitrification, and chemo-denitrification (Firestone and Davidson 1989). Humans began altering the natural N cycle as they expanded agricultural land, used fire as a land clearing and management tool, and cultivated leguminous crops that carry out biological N fixation. Human alteration accelerated dramatically with the discovery of the Haber–Bosch process, the chemical process that synthetically transforms atmospheric dinitrogen (N₂) gas into ammonia (NH₃) (Erisman *et al* 2008). The



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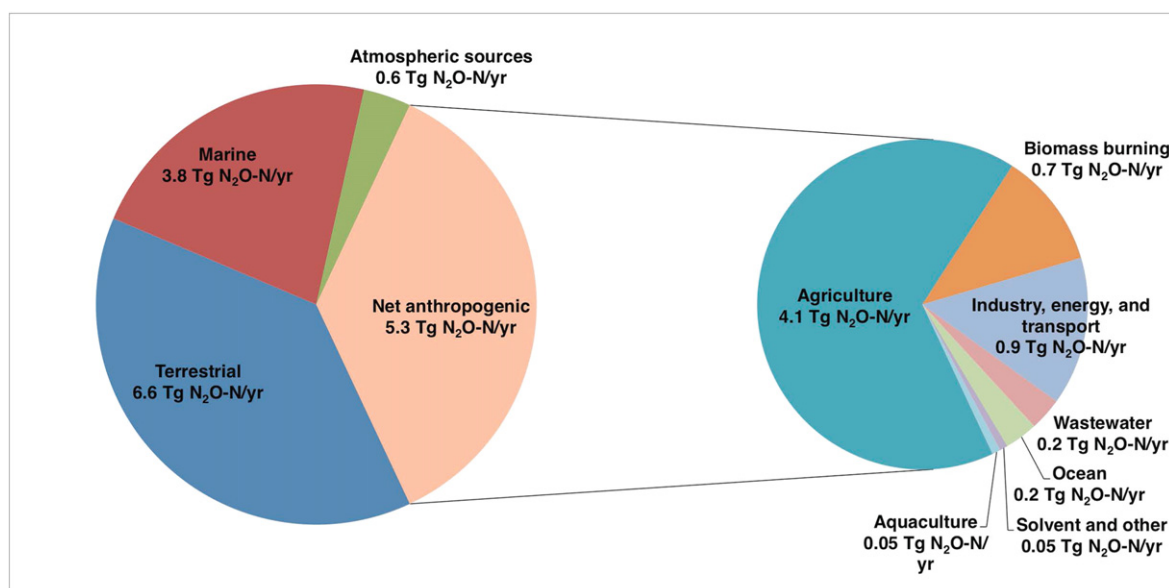


Figure 1. Natural versus anthropogenic N₂O emissions in 2005. The values for natural emissions (terrestrial, marine, and atmospheric chemistry in the pie chart on the left) are taken from Ciais *et al* (2013), while the anthropogenic values are the best estimate values by sector from the 2013 UNEP report (Bouwman *et al* 2013, Oenema *et al* 2013, van der Werf *et al* 2013, Wiesen *et al* 2013), as summarized in table 1 of this paper. The net anthropogenic estimate in the left pie chart takes into account the effect of land use change on reducing net anthropogenic emissions (about 0.9 Tg N₂O-N yr⁻¹, see section 3.2). Reprinted, with permission, from Davidson *et al* (2013), (figure 3.1).

industrial production of NH₃ led to the development synthetic N fertilizers, which play a central role in feeding the world's rapidly increasing population. Without the Haber–Bosch process, about half of the world's population today would likely not be adequately nourished (Erisman *et al* 2008). This growth in anthropogenically-fixed N has simultaneously led to an unintended increase in global N pollution, including N₂O emissions, driven largely by the fact that mismatch between crop N demand and soil N supply frequently leads to N losses. With the possible exception of certain industrial point sources, it is impossible to completely eliminate global N pollution, particularly from agriculture—its largest source.

This paper first updates constraints on estimates and their uncertainties for anthropogenic and natural components of N₂O emissions by biome and by anthropogenic sector. We then consider a suite of emissions scenarios for N₂O, including those of the recent Representative Concentration Pathways (RCPs) of the Intergovernmental Panel on Climate Change (IPCC, van Vuuren *et al* 2011a) and a recent special United Nations report on N₂O (UNEP 2013), and the magnitude of mitigation efforts that would be needed to stabilize atmospheric N₂O by 2050. Future potential emissions from biofuels are discussed separately given particularly high levels of uncertainty for this sector. This paper integrates the authors' contributions to that UNEP report, other chapters in that report, the IPCC fifth assessment (AR5), and other recent literature for the most recent information on N₂O.

2. Natural emissions

The first approach to emission estimation is called 'bottom-up,' because it sums up emission inventories from field

measurements, organized according to ecosystem type or by geographic region. Using the 'bottom-up' approach, published central estimates of current natural emissions of N₂O from terrestrial, marine and atmospheric sources based on several inventories range from 10 to 12 Tg N₂O-N yr⁻¹ (Mosier *et al* 1998, Galloway *et al* 2004, Crutzen *et al* 2008, Syakila and Kroeze 2011). The IPCC AR5 (Ciais *et al* 2013) estimated that current natural sources of N₂O add up to roughly 11 (range 5.4–19.6) Tg N₂O-N yr⁻¹, which is the sum of emissions from terrestrial (6.6; range 3.3–9.0), marine (3.8; range 1.8–9.4) and atmospheric sources (0.6; range: 0.3–1.2; see figure 1). Note that the indicated uncertainty ranges from each bottom-up estimate are added together to produce the large range about the AR5 global estimate. Combining estimates of natural and anthropogenic emissions from Ciais *et al* 2013 and the 2013 UNEP report (see section 3) respectively, we estimate that natural emissions account for approximately two thirds of total global N₂O emissions (figure 1).

A second approach is called 'top-down,' because it is based on atmospheric measurements and an inversion model. Prather *et al* (2012) provide a spreadsheet model; here we employ the one-box mixing model of Daniel *et al* (2007):

$$E = \frac{m_{i+1} - m_i e^{-t/\tau}}{f\tau(1 - e^{-t/\tau})}, \quad (1)$$

where E is annual emissions (Tg N₂O-N yr⁻¹), m_{i+1} and m_i are the observed atmospheric mixing ratios (ppb N₂O) at the start of consecutive years, τ is the lifetime, t is 1 year, and f relates the mass burden to the mixing ratio (0.21 ppb N₂O/Tg N; Prather *et al* 2012).

Estimates of atmospheric N_2O mixing ratios (specifically, the tropospheric mean mole fraction) prior to the industrial revolution are from ice cores measurements, which were at an approximate steady state from 1730 to 1850 (Machida *et al* 1995). An important source of uncertainty in the top-down approach is the estimated atmospheric lifetime of N_2O . Most estimates range between the IPCC-AR4 assumption of 114 years (Forster *et al* 2007) to 131 (Prather *et al* 2012). The Stratosphere-troposphere Processes And their Role in Climate report (SPARC 2013) suggests 123 years as the ‘recommended’ estimate, with the ‘most likely range’ between 104 and 152 years. Because of this large range in estimates from a number of respected sources, we choose here to use only two significant figures. We round the SPARC recommended estimate to 120 years, with an uncertainty range 110–130 years, which encompasses most of the range of central estimates used commonly in the literature. We also assume that the lifetime has not changed substantially, due to a lack of compelling evidence to the contrary. With these assumptions, pre-industrial emissions are estimated to be about $11 \text{ Tg N}_2\text{O-N yr}^{-1}$, with an uncertainty range of $10\text{--}12 \text{ Tg N}_2\text{O-N yr}^{-1}$. The sensitivity of the estimate is a change of about $1 \text{ Tg N}_2\text{O-N yr}^{-1}$ of pre-industrial emissions for every 10 years change in assumed lifetime. Hence, if the lifetime was as long as about 140 years in the pre-industrial period, the emissions estimate would be $9 \text{ Tg N}_2\text{O-N yr}^{-1}$ (Prather *et al* 2012). The central estimates of both top-down and bottom up approaches for pre-industrial natural emissions are in agreement at $11 \text{ Tg N}_2\text{O-N yr}^{-1}$, although both have considerable uncertainties.

Uncertainty in pre-industrial natural emission estimates also arises from the lack of complete understanding about the influence of anthropogenic changes prior to the industrial revolution (such as the expansion of agriculture) and from temporal variability of natural emissions. Between 1730 and 1850, N_2O concentrations in the atmosphere varied slightly from year to year and decade to decade, but showed little or no consistent long term trend (Machida *et al* 1995). Although Syakila and Kroeze (2011) estimated that average net anthropogenic emissions were around $0.5 \text{ Tg N}_2\text{O-N yr}^{-1}$ during the 18th and early 19th centuries, these possible anthropogenic emissions were sufficiently low that their signal cannot be distinguished from the effects of climatic variation on natural emissions.

Despite the uncertainties, both bottom-up and top-down approaches suggest that natural emissions were and probably still are between 10 and $12 \text{ Tg N}_2\text{O-N yr}^{-1}$. We will concentrate the rest of our analyses on anthropogenic effects since 1850.

3. Anthropogenic emissions

3.1. Top-down atmospheric modeling constraints

Modern anthropogenic emissions of N_2O can be calculated using the same top-down method described above (equation (1)). In this case, the changes in atmospheric concentrations from 1850 to the present (Machida *et al* 1995,

NOAA 2014) are assumed to be entirely anthropogenic, assuming relative stability of natural emissions over the same period and the same atmospheric N_2O lifetime of $120 (\pm 10)$ years. The natural emission estimate ($11 \text{ Tg N}_2\text{O-N yr}^{-1}$) is subtracted from the total modern emissions calculated from equation (1) to yield modern net anthropogenic emissions. We averaged the emission estimates for the period 2000–2007 to avoid artifacts of short-term interannual variation, yielding an estimate for net anthropogenic emissions of $5.3 \text{ Tg N}_2\text{O-N yr}^{-1}$ (range $5.2\text{--}5.5$) for that period. This estimate includes all anthropogenic activities that have contributed to changing atmospheric N_2O , including any decrease in emissions from forest soils because of deforestation and increases in emissions from expanded activity in agriculture and other sectors.

3.2. Bottom-up inventory estimates

Protocols have been developed by the IPCC (2006) for countries to estimate their N_2O emissions. The IPCC Tier 1 Protocol multiplies metrics of activity in agriculture, energy generation, transportation, and other sectors, by emission factors (EFs), the amount of N_2O emitted per unit of activity. For example, the direct emissions of N_2O from agricultural soils are estimated as a 1% EF applied to synthetic-N fertilization application activity rates. Additional EFs are used to calculate the amount of fertilizer N leached into surface and groundwaters and volatilized as ammonia or nitrogen oxide gases, and the subsequent indirect N_2O emissions from downstream and downwind ecosystems, which often are substantial. For example, emissions from coastal, estuarine and riverine waters are estimated to be about 9% of total anthropogenic sources (Ciais *et al* 2013), although the original source of most of this N was from agricultural field applications. The EFs have been derived from the literature and are periodically revised as warranted. By necessity, they are averages across a broad range of conditions and often do not yield accurate estimates for individual sites. Nevertheless, there is evidence that errors on the small scale are largely canceled when aggregated to larger scales (Del Grosso *et al* 2008).

Another source of inaccuracy in the use of Tier 1 EFs is that they assume a linear relationship between N application rates and N_2O emissions. A growing number of studies demonstrate nonlinear (usually exponential) relationships between N application rate and N_2O emissions (Shcherbak *et al* 2014). The nonlinear relationship is likely the result of large increases in N_2O emissions once N application rates are in excess of plant demands. This has important implications for targeting mitigation where N application rates are higher than N harvested in crop export and for not discouraging additional N application in N-deficient regions where mining of soil nutrients is common, such as sub-Saharan Africa. The implications of nonlinearity are not yet clear for global N_2O budgets. The differences between linear and nonlinear models for estimating N_2O emissions are more likely to be important at the farm scale compared to the global scale, because the biases of the linear model (probably overestimation of fluxes where N applications are low and underestimation where N

application rates are high) at least partially cancel as the spatial scale increases.

The United Nations Food and Agriculture Organization (FAO) estimates agricultural N_2O emissions by applying IPCC Tier 1 EFs to their country data gathered from national publications and questionnaires. The Emissions Database for Global Atmospheric Research (EDGAR) database uses a blend of private and public data, applying IPCC Tier 1 EFs to estimate both non-agricultural and agricultural N_2O emissions (with the exception of biomass burning, where they apply EFs described in Andreae and Merlet (2001).

A variant of the bottom-up global inventory approach involves a combination of ‘top-down’ constraints, based on a global atmospheric budget, and ‘bottom-up’ inventory estimates of minor N_2O sources from biomass burning, industry, energy, and transportation sectors, and from statistical correlations at the global scale using data on fertilizer use, manure production, and land-use change (Crutzen *et al* 2008, Davidson 2009, Smith *et al* 2012). These approaches yield EFs based on newly fixed N (either Haber–Bosch or biological N fixation) and N remobilized from tillage of soils (Smith *et al* 2012) or through production of manure by livestock (Davidson 2009). They implicitly include both direct and indirect emissions (i.e., on the farm and downwind and downstream) from these N fluxes, so comparison with the IPCC EFs is not straightforward. Nevertheless, the estimates from Davidson (2009) of 2.0% of manure-N and 2.5% of fertilizer-N converted to N_2O are not far off of the sums of IPCC EFs for direct and indirect agricultural emissions and human sewage. The estimate from Smith *et al* (2012) that 4% of newly fixed N is converted to N_2O may not be far off of the sum of IPCC EFs when the cascading effects of newly fixed N moving through croplands, livestock operations, downwind and downstream ecosystems, and human sewage are considered.

Countries that have sufficient data to calculate EFs more specific to their particular situations are allowed to use them under IPCC’s Tier 2 Protocol, which presumably yields more accurate estimates for those specific regions and management practices (IPCC 2006). Under the Tier 3 Protocol, countries with access to validated biogeochemical models and sufficient input data are allowed to use these models to calculate N_2O emissions (IPCC 2006). This presumably yields even more accurate estimates if the models skillfully account for spatial and temporal variation of the most important factors affecting emissions.

With the advent of new laser technologies for measurements of N_2O fluxes (e.g., Savage *et al* 2014) there is likely to be continued improvement in estimating emission factors for the Tier 1 and Tier 2 Protocols and for developing and validating the biogeochemical models used with the Tier 3 Protocol. However, it will remain difficult to fully account for the large spatial and temporal variation of N_2O emissions. Improvement in the quality of activity data for each country, such as its fertilizer application rates, livestock production, and manure handling procedures, is also necessary for improved emission estimates. New EFs are also needed for new cropping systems, such as second generation biofuel

crops. Indeed, fertilizer application rates and EFs for biofuel production are among the largest uncertainties for projections of future N_2O emissions (see section 5.2).

Table 1 summarizes recent efforts at partitioning anthropogenic emissions from bottom-up inventories and from integrated bottom-up and top-down analyses. Here we adopt the recent estimates from UNEP (2013) for total net anthropogenic N_2O emissions of $5.3 \text{ Tg } \text{N}_2\text{O-N yr}^{-1}$, which is equal to the top-down estimate (section 3.1). The ‘best estimate’ from the UNEP report is lower than the estimates from other inventories shown in table 1, partly because of some lower updated sectoral estimates and partly due to including the effect of lower tropical forest soil emissions resulting from historic and on-going deforestation, which is neglected in many other inventories. The best estimate of gross anthropogenic emissions is $6.2 \text{ Tg } \text{N}_2\text{O-N yr}^{-1}$. Because tropical forest soils are a large natural source of N_2O emissions, tropical deforestation should be considered as a significant human-induced decrease in emissions. Soil N_2O emissions from recently converted tropical forests may initially increase, but the long-term trend is for emissions from the pasture soils and degraded land soils to be lower than those from intact, mature tropical forests (Davidson *et al* 2001, Melillo *et al* 2001), resulting in current estimates of a decreased source of $0.9 \text{ Tg } \text{N}_2\text{O-N yr}^{-1}$ (Davidson 2009). Subtracting the effect of tropical deforestation from the estimate of gross anthropogenic emissions yields a best estimate of $5.3 \text{ Tg } \text{N}_2\text{O-N yr}^{-1}$ for net anthropogenic emissions, which is 15% below the gross anthropogenic emission estimate. Without this adjustment, the bottom-up and top-down approaches would not agree, although the apparent exact agreement to a tenth of a teragram is probably partly fortuitous.

3.3. Anthropogenic emissions by sector

Significant uncertainties remain for activity data and especially for several of the emission factors in each sector. Brief summaries of expert analyses from each sector from chapters 4–7 of the UNEP (2013) report are presented here.

3.3.1. Agriculture. Agriculture is the largest source of anthropogenic N_2O emissions, responsible for $4.1 \text{ Tg } \text{N}_2\text{O-N yr}^{-1}$ ($3.8\text{--}6.8 \text{ Tg } \text{N}_2\text{O-N yr}^{-1}$; Oenema *et al* 2013) or 66% of total gross anthropogenic emissions (table 1). Emission estimates include direct soil emissions from synthetic N fertilizer and manure application and indirect emissions that occur from downstream or downwind water bodies and soils after nitrate leaches away from croplands and after N emitted from croplands as ammonia or nitrogen oxide gases fall back to earth as atmospheric N deposition. Also included are N_2O emissions resulting from crop residues, manure management, cultivation of organic soils, and crop biological N fixation (C-BNF). The central factor responsible for agricultural N_2O emissions is a lack of synchronization between crop N demand and soil N supply, with, on average, around 50% of N applied to soils not being taken up by the crop (Snyder *et al* 2009, Oenema *et al* 2013, Venterea *et al* 2012). Inputs of N to agricultural soils are mainly from synthetic N fertilizer

Table 1. Published inventories of N₂O emissions by sector.

All units in Tg N ₂ O-N yr ⁻¹	FAO ^a	EDGAR ^b	EPA (2012) ^c	Syakila and Kroeze (2011)	Davidson (2009) ^d	Del Grosso <i>et al</i> (2008) ^e	Crutzen <i>et al</i> (2008) ^f	Denman (2007) ^g	Mosier <i>et al</i> (1998) ^h	UNEP report ⁱ	Estimate range
Agriculture	4.1	3.8	4.6	5.1	5	4.9	4.3–5.8	5.1	6.8	4.1^j	3.8–6.8
Fertilizer	1.4						—	—		—	
Direct	1.1				2.2		—	—		—	
Indirect	0.3	3.6		2.3			—	—	3.9	—	
Manure	1.8						—	—		—	
Direct	1.4		4.2		2.8	4.5	—	—		—	
Indirect	0.4						—	—		—	
Organic soils	0.2	—		0.1	—		—	—	0.1	—	
Crop residues	0.3	—		0.3	—		—	—	0.4	—	
C-BNF	—	—		0.1	—		—	—	0.1	—	
Manure management	0.3	0.2	0.4	2.3	—	0.4	—	—	2.3	—	
Biomass burning	—	1.1^k	1.7	0.7	0.5	—	—	0.7	—	0.7	0.5–1.7
Residue burning	0.01	—	1.6	—	—	—	—	—	—	0.04	
Savanna burning	—	—	—	—	—	—	—	—	—	0.3	
Other ^l	—	—	0.1	—	—	—	—	—	—	0.4	
Industry, energy and transport^m	—	1.7	0.9	—	0.8	—	0.7–1.3	0.7	1.3	0.9	0.7–1.7
Wastewater	—	0.2	0.2	0.3	—	—	—	0.7	0.3	0.2	—
Aquaculture	—	—	—	—	—	—	—	—	—	0.05	—
Solvent and other product use	—	—	0.2	—	—	—	—	—	—	0.05	—
Land-use change ⁿ	—	—	—	−0.6	−0.9	—	0–0.9	—	—	−0.9	—
Ocean	—	—	—	1.0	—	—	—	—	—	0.2 ^o	—
Total	—	6.8	7.6	6.5	5.4	—	5.6–6.5	6.7	8.4	5.3	5.3–8.4

^a 2010 estimates; UN Food and Agriculture Organization (FAO) estimates agricultural N₂O emissions by applying IPCC Tier 1 EFs to their country data gathered from national publications and questionnaires.

^b 2008 estimates; the Emissions Database for Global Atmospheric Research (EDGAR) category 'Indirect N₂O from non-agricultural NH₃ and NO_x' is partitioned between 'Biomass Burning' and 'Industry, Energy, Transport' and weighted by their total direct emissions. The EDGAR database uses a blend of private and public data, applying IPCC Tier 1 EFs to estimate both non-agricultural and agricultural N₂O emissions (with the exception of biomass burning, where they apply EFs described in Andreae and Merlet 2001).

^c 2006 estimates.

^d 2005 estimates; agriculture estimates include human waste emissions.

^e Mix of data from 2000 and 1994.

^f 2000 estimates; agriculture estimates include human waste, aquaculture, and biomass burning emissions.

^g Effects of atmospheric deposition are included in the agriculture sector, although a portion of the deposition comes from other sectors.

^h 2006 estimates for agriculture and waste (adapted by Syakila and Kroeze 2011), 1989 estimates for industrial/energy.

ⁱ The UNEP estimates for agriculture, biomass burning, wastewater, aquaculture, and land use change are from Bouwman *et al* (2013), Oenema *et al* (2013), van der Werf *et al* (2013), Wiesen *et al* (2013).

^j The FAO estimates are adopted by UNEP (Oenema *et al* 2013). This estimate does include indirect emissions from downwind and downstream ecosystems, but does not include sewage wastewater emissions.

^k Includes category 'large-scale biomass burning' which denotes savanna burning, forest fires, peat fires, grassland fires, decay of wetland/peatland and post burn decay after forest fires, agricultural residue burning, and other vegetation fires.

^l 'Other' biomass burning includes tropical, temperate and boreal forest fires, tropical peat fires, and fuelwood fires.

^m Several literature sources combine emissions from industry and/or energy and transport into one overall estimate.

ⁿ The S&K estimate of reduced natural emissions is for pre-industrial land use change only. Crutzen *et al* provide only a range, so we use Davidson's (2009) estimate for post-industrial tropical deforestation.

^o Emissions from the ocean due to anthropogenic N deposition should be included in indirect emission factors for agriculture and other sectors, but are probably underestimated, so we include this estimate from Suntharalingam *et al* (2012).

and manure application, with additional supply from legume N fixation, crop residues, and N deposition.

3.3.2. Industry and fossil fuel combustion. The industry sector plus fossil fuel combustion (stationary combustion and transportation) together are responsible for about 0.9 Tg N_2O -N yr^{-1} (0.7–1.6 Tg N_2O -N yr^{-1}) or 15% of total gross anthropogenic N_2O emissions (Wiesen *et al* 2013). Nitric and adipic acid production are the major industrial sources. Nitric acid is mainly used as a feedstock in the production of explosives and N fertilizer, particularly ammonium nitrate, with N_2O emitted during the ammonia oxidation process (Lee *et al* 2011). Adipic acid is a key feedstock in synthetic fiber production, with N_2O resulting from the use of nitric acid to oxidize several organic chemicals (Schneider *et al* 2010). Stationary combustion (mainly coal power plants) is the principal source of N_2O from the energy sector. Emissions of N_2O from this sector arise via the oxidation of both atmospheric N_2 and organic N in fossil fuels. Emissions vary with the amount of organic N in the fuel, the operating temperature, and the oxygen levels during combustion (EPA 2012). N_2O from transport is released primarily by catalytic converters used to control NO_x , carbon monoxide, and hydrocarbons in tailpipe emissions, with older technologies responsible for significantly higher emission rates per kilometer than more advanced technologies (IPCC 2006).

3.3.3. Biomass burning. Biomass burning is currently responsible for about 0.7 Tg N_2O -N yr^{-1} (0.5–1.7 Tg N_2O -N yr^{-1} ; van der Werf *et al* 2013) or 11% of total gross anthropogenic emissions. This includes crop residue burning, forest fires (resulting from both natural and human activities), and prescribed savannah, pasture, and cropland burning. It also includes N_2O emissions from household biomass stoves. N_2O is released via the oxidation of organic N in biomass during combustion. Although some wildfires are ignited naturally by lightning, all emissions from biomass burning have been attributed as anthropogenic emissions, because it is impossible to separate out which wildfires are ignited by humans. Furthermore, anthropogenic climate change may also be increasing fire frequency and severity (Pechony and Shindell 2010).

3.3.4. Wastewater, aquaculture, and other sources. N_2O emissions from wastewater were 0.2 Tg N_2O -N yr^{-1} in 2010 (Bouwman *et al* 2013), or 3% of total gross anthropogenic emissions. This includes N_2O emitted either directly from wastewater effluent or from bioreactors removing N in biological nutrient removal plants (Law *et al* 2012). A small amount of N_2O is also emitted in aquaculture (<0.1 Tg N_2O -N yr^{-1} in 2010). Various human-related changes to the oceanic environment have affected the amount of N_2O emissions produced by the oceans. Increased N deposition onto the ocean has been estimated to have increased the oceanic N_2O source by 0.2 Tg N_2O -N yr^{-1} (0.08–0.34 Tg N_2O -N yr^{-1}) or 3% of total gross anthropogenic emissions

(Suntharalingam *et al* 2012). In principle, increased oceanic emission due to N deposition should be included in the indirect emission estimates from agricultural, energy, and transportation sources, but it is included here as a separate category because the oceans may have been under-represented in calculations of emissions from downwind and downstream ecosystems.

4. Trends in emissions over the last 20 years

Figure 2 illustrates how N_2O emissions from three of the most important sectors of the EDGAR (2009) database have changed from 1990 to 2008. The dominance of emissions by agricultural soils is clear, with the importance of South Asia, parts of Latin America and especially East Asia growing in the last two decades. Large-scale biomass burning emissions are most important in tropical savannah regions. Industrial emissions are most important in developed countries and are growing in South and East Asia.

5. Emission projections

5.1. Synthesis of published scenarios

Projections of future emissions depend upon assumptions about changes in:

- Population growth rates.
- Per capita consumption of calories and protein.
- Relative sources of vegetable versus animal products for meeting food demands.
- Rates of wastage/loss of food from production to consumption.
- Nutrient use efficiency in crop and animal production systems.
- Production of newly fixed N for agriculture (including biofuels) and aquaculture.
- Emissions of NH_3 and NO_x from all sectors, which contribute to N deposition on native soils and oceans.
- Fire frequency, including household biomass burning, slash-and-burn agriculture, pasture clearing, and wildfire.
- Industrial and energy sectors (such as fertilizer manufacturers and industries using coal combustion) that can reduce emissions.
- Land-use change.
- Energy sector technology and demand for biofuels.
- Climate and its effects on N cycling processes.

Climate change can affect N_2O emissions from water bodies and soils under native vegetation, but this effect is not well represented in current models (Pinder *et al* 2012) and it is not dealt with here. Most published projections of future emissions focus on assumptions about changes in emissions from agriculture, biomass burning, energy, transportation, and industry, which vary widely among the scenarios considered here (table 2) and elsewhere (e.g. Bodirsky *et al* 2014). Here, four sets of published N_2O emission scenarios were

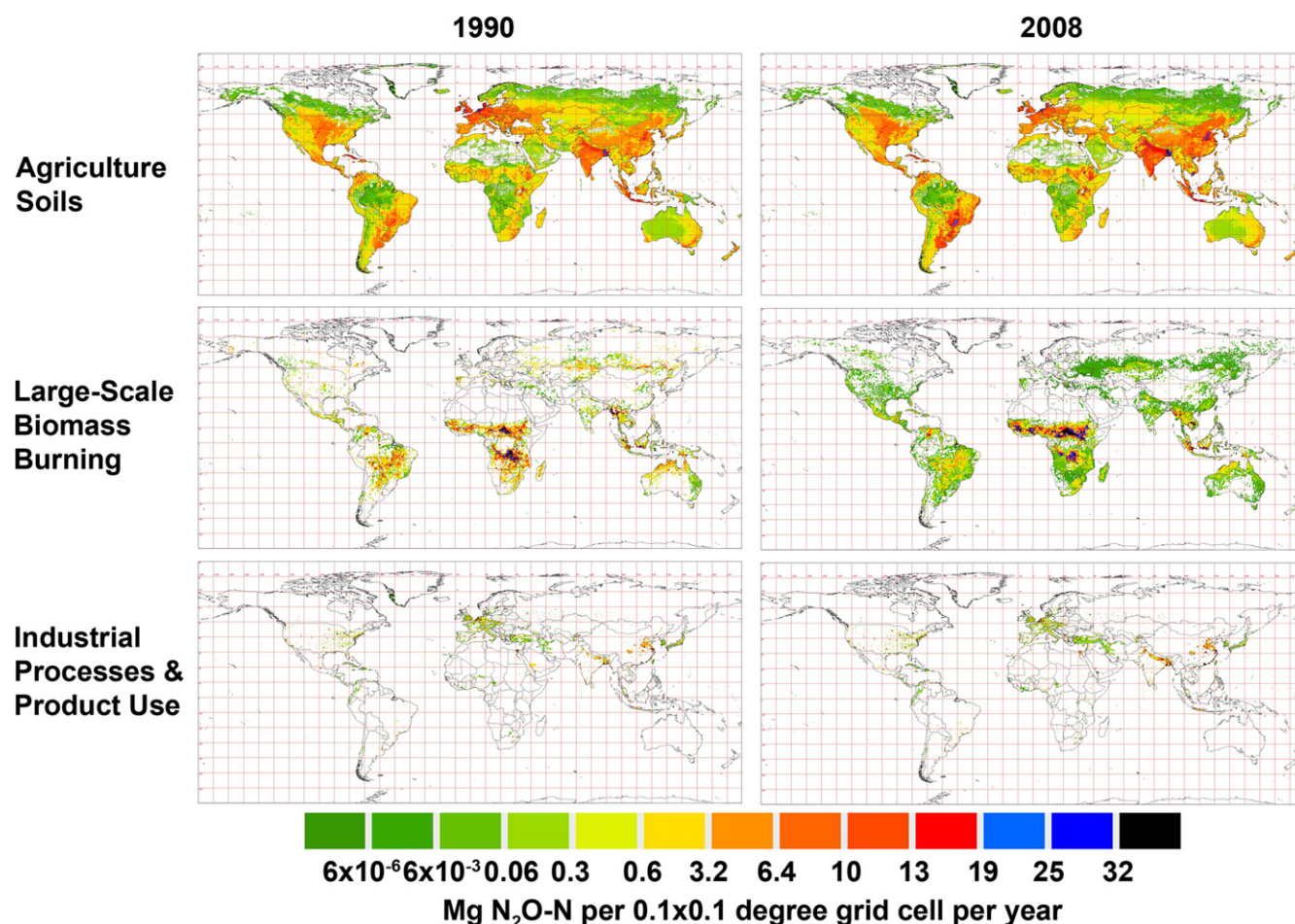


Figure 2. Global maps of direct emissions from agricultural soils, large scale biomass burning, and industrial process (mostly adipic and nitric acid production) for 1990 and 2008 from the Emissions Database for Global Atmospheric Research (EDGAR), version 4.0 (2009) (<http://edgar.jrc.ec.europa.eu>). Units are tons of $\text{N}_2\text{O-N}$ per 0.1×0.1 degree grid cell per year. Note that this size grid cell is about 123 km^2 at the equator and declines with increasing latitude, approaching zero near the poles. Reprinted, with permission, from Davidson *et al* (2013), (figure 3.2).

aggregated to characterize the potential range of future anthropogenic emissions:

- The Special Report on Emissions Scenarios (SRES) (Nakicenovic *et al* 2000) created four major global greenhouse gas emissions scenarios (A1, A2, B1 and B2) based on the degree of globalization versus regionalization and the priority given to economic versus social and environmental objectives. These were used in the IPCC's Third and Fourth Assessment Reports.
- The RCPs (Van Vuuren *et al* 2011a) are used in the IPCC AR5, with four scenarios based on differing radiative forcing levels rather than emissions (RCP 2.6, 4.5, 6.0 and 8.5, with the numbers referring to different radiative forcing levels in Wm^{-2} in the year 2100).
- Davidson (2012) used FAO projections of population and dietary demands to estimate fertilizer and manure demands and subsequent N_2O emissions, including five variants (S1-S5) of mitigation and dietary habits.
- Five scenarios of a new UNEP report (Sutton *et al* 2013) based on expert analyses of feasible mitigation options in

each sector: TR1: Business-As-Usual; TR2: Mitigation of Industry, Fossil Fuel Combustion, and Biomass Burning; TR3: Efficiency of Agricultural Production; TR4: Efficiency of Agricultural Production and Consumption; and TR5: Combined Mitigations.

These studies have different base years and employ different inventory sources. In order to make their results comparable, all emission estimates were normalized to the best estimate of 2005–2010 average net anthropogenic emissions from the UNEP report ($5.3 \text{ Tg N}_2\text{O-N yr}^{-1}$). The scenarios of annual emissions are presented graphically in figure 3. The numerous scenarios are organized into three groups and means calculated for each group:

5.2. Business-as-usual scenarios (BAU)

The RCP 8.5, SRES A2, Davidson's S1, and UNEP1 scenarios have no or little mitigation. On average, the emissions of these scenarios increase to $9.7 \text{ Tg N}_2\text{O-N yr}^{-1}$ by 2050, which is nearly double their level in 2005 (83% increase).

Table 2. Summary of assumptions of published scenarios for future N₂O emissions.

Source	Scenario	Description
SRES (Nakicenovic <i>et al</i> 2000)	A1	A world of increased regional, economic, social and cultural convergence drives rapid economic growth and the dissemination of new technologies, with global population peaking at nine billion in 2050 and declining thereafter.
	A2	A fragmented world with more regionally focused economic development leads to slower per capita economic growth and technological change than other scenarios. Continuous population growth due to slow convergence of regional fertility patterns.
	B1	A global approach to economic, social and environmental sustainability leads to the swift creation of a service and information economy, with a rapid expansion of clean technologies and less resource use. Similar population growth to A1.
	B2	A similar commitment to sustainability as in B1, but with an emphasis on local solutions. Continuous population growth (at a slower rate than A2), moderate levels of economic development, with slower and more varied technological change than A1 and B1.
RCPs (van Vuuren <i>et al</i> 2011a)	RCP8.5	A high emissions pathway—representing a scenario where little is done to limit climate change—leading to radiative forcing of 8.5 Wm ⁻² in 2100.
	RCP6	An emissions pathway that eventually leads to a stabilization of radiative forcing at 6 Wm ⁻² after 2100.
	RCP4.5	An emissions pathway that eventually leads to a stabilization of radiative forcing at 4.5 Wm ⁻² after 2100.
	RCP2.6	A low emissions pathway—representing ambitious international action to limit climate change—that leads to a peak in radiative forcing at 3 Wm ⁻² before 2100, dropping to 2.6 Wm ⁻² by 2100.
Davidson (2012)	S1	Future fertilizer and manure use scaled to FAO projections of population growth, per capita caloric intake and meat consumption.
	S2	Developed countries reduce per capita meat consumption to 50% of 1980 levels by 2030, remaining constant to 2050. Results in a 21% reduction in fertilizer use and manure production by 2030 and 2050 relative to Scenario 1.
	S3	Improved efficiency of fertilizer use and manure production reduces N ₂ O emission factors 50% by 2050.
	S4	The same as Scenario 3, with the addition of emission reductions from industry, energy, transport, and biomass burning of 50% by 2050.
	S5	A scenario combining the mitigation actions of Scenarios 2 through 4.
UNEP (Sutton <i>et al</i> 2013)	TR1	Emissions are projected to increase according to the ‘business-as-usual’ assumptions presented in Chapters 4 to 7 of UNEP (2013).
	TR2	Combined emissions from industry, energy, transport and biomass burning are reduced by 58% by 2050 relative to Case 1.
	TR3	Improved efficiency of fertilizer use reduces fertilizer demand by 15% and the N ₂ O emission factor for fertilizer 20% by 2050 relative to Case 1. Improvements in manure management reduce N excretion per unit animal product by 30% and the N ₂ O emission factor for manure production 10% by 2050 relative to Case 1.
	TR4	The efficiency improvements in Case 3 are combined with 50% reductions in global food waste and developed country meat consumption relative to Case 1.
	TR5	A combination of all the mitigation actions in Cases 2 through 4.

5.3. Moderate mitigation scenarios

The scenarios RCP 4.5, RCP 6.0, SRES A1, SRES B1, Davidson’s S2 and S3, UNEP2 and UNEP3 have moderate mitigation, defined here as scenarios showing emission trends that are higher than 2005 emissions in 2050 but below BAU. On average, emissions grow to 6.7 Tg N₂O-N yr⁻¹ by 2050, an increase of 26% relative to 2005.

5.4. Concerted mitigation scenarios

The RCP 2.6, SRES B2, Davidson’s S4 and S5, UNEP4, and UNEP5 mitigation scenarios are concerted, because they lead to emissions in 2050 that are below the 2005 level. On

average, emissions decline to 4.2 Tg N₂O-N yr⁻¹ by 2050, a decrease of 22% relative to 2005.

The concerted mitigation scenarios result in near stabilization of atmospheric concentrations of N₂O between 340 and 350 ppb by 2050 (Davidson 2012, Davidson *et al* 2013), whereas N₂O concentration continues rising beyond 2050 for the BAU and moderate mitigation scenarios.

By 2020 the average concerted mitigation scenario reduces emissions by 1.8 Tg N₂O-N yr⁻¹ or 25% below the average BAU scenario, equivalent to 0.8 Gt CO₂ eq yr⁻¹ less than BAU (table 3). This is approximately 10% of the emissions gap that needs to be bridged by 2020 for it to be ‘likely’ that average global warming stays below a 2 °C

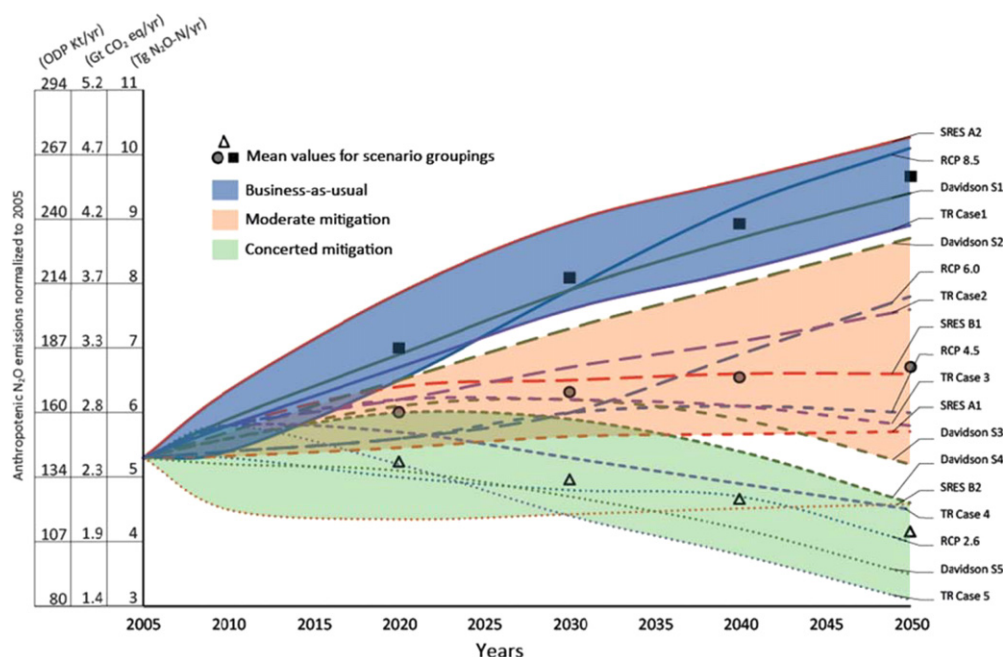


Figure 3. Projections of anthropogenic N_2O emissions according to groupings of published business-as-usual, moderate mitigation, and concerted mitigation scenarios (Nakicenovic *et al* 2000, Van Vuuren *et al* 2011a, Davidson 2012, Sutton *et al* 2013; see table 2). The ‘TR’ cases 1–5 refer to ‘this report’, being the 2013 UNEP report (Sutton *et al* 2013). The mean for each grouping of scenarios is shown by square, circle, and triangle markers. All projections have been adjusted to a common emission estimate baseline in 2005 consistent with the UNEP report’s best estimate of net anthropogenic emissions of $5.3 \text{ Tg N}_2\text{O-N yr}^{-1}$. Concerted scenarios include aggressive mitigation in all sectors and most include reduced per capita meat consumption in the developed world. Y-axis units have been converted to CO_2 equivalents for a 100-year global warming potential and to Ozone Depletion Potential (ODP; Ravishankara *et al* 2009). Reprinted, with permission, from Sutton *et al* (2013), (figure 8.1).

Table 3. Projected annual anthropogenic N_2O emissions for three emission scenario groupings, given in units of N, CO_2 equivalents, and ozone depletion potential.

	2020	2030	2040	2050
Units: nitrogen equivalents ($\text{Tg N}_2\text{O-N yr}^{-1}$) ^a				
Business-as-usual	7.0	8.1	8.9	9.7
Moderate mitigation	6.0	6.3	6.5	6.7
Concerted mitigation	5.2	5.0	4.7	4.2
Units: equivalents of carbon dioxide ($\text{Gt CO}_2\text{-eq yr}^{-1}$) ^b				
Business-as-usual	3.3	3.8	4.2	4.5
Moderate mitigation	2.8	3.0	3.1	3.1
Concerted mitigation	2.5	2.3	2.2	1.9
Units: ozone depletion potential (ODP kt yr^{-1}) ^c				
Business-as-usual	187	216	238	258
Moderate mitigation	160	169	175	178
Concerted mitigation	140	133	125	111

^a The values are the mean of four sets of scenarios according to SRES (Nakicenovic *et al* 2000), RCP (van Vuuren *et al* 2011a), Davidson (2012), and UNEP (2013) and grouped as described in the main text.

^b Calculated using a 100-year global warming potential of 298 for N_2O .

^c Calculated using an ozone depleting potential of 0.017 for N_2O .

threshold (UNEP 2012). By 2050, the average concerted mitigation scenarios are 57% lower than the average BAU scenario—around $5.5 \text{ Tg N}_2\text{O-N yr}^{-1}$ ($2.6 \text{ Gt CO}_2 \text{ eq yr}^{-1}$). The avoided emissions between 2014 and 2050 sum to $22 \text{ Tg N}_2\text{O-N}$ ($57 \text{ Gt CO}_2\text{eq}$). To put this figure in context, it is equal to about ten years of the CO_2 emissions of all of the passenger cars currently on the road.

The impact of N_2O on stratospheric ozone depletion has been estimated using an ozone-depletion potential (Ravishankara *et al* 2009; ODP—a measure of its ozone destructiveness relative to CFC-11, which is defined as an ODP of 1). Although the use of ODPs is controversial because of complex interactions of various anthropogenic gases and stratospheric temperature (Fleming *et al* 2011), we employ it here to place the potential impact of N_2O mitigation on stratospheric ozone in context with previous efforts to mitigate CFCs. By 2050, the difference between the concerted average mitigation scenario and the average BAU scenario ($147 \text{ ODP kt yr}^{-1}$ is equivalent to a 13% decrease in chlorofluorocarbon (CFC) emissions from their peak in the late 1980s, approximately halving ODP-weighted emissions in 2050 compared to BAU (table 3). The sum of the avoided emissions between 2014 and 2050 is 3270 ODP kt . These reductions would be 40%–110% greater than the potential reductions from the destruction of the remaining recoverable stocks of other ozone depleting substances, which has been identified as the most substantive remaining action that could be taken to accelerate ozone layer recovery (UNEP 2010).

An important caveat of these projections is that they all begin in 2005, and significant differences are already apparent in their trajectories by 2013. So far (up to 2013), actual global N₂O emissions have been closer to BAU trajectories than the mitigation trajectories.

5.5. Projecting N₂O emissions from biofuel production

Another caveat of these projections is that the highly uncertain impact of expansion of biofuel production is not considered (Davidson *et al* 2013). In addition to uncertainties about total biofuels produced, the N fertilization rates needed for producing second- or third-generation fuel stocks and the N₂O EFs for those cultivation practices are not known. Fertilization rates and EFs for rapidly growing trees and native grasses, forbs, and shrubs may be much lower than for most current food and fiber crops. To put this uncertainty into perspective, two methods are offered here to bound the range of future N₂O emissions from biofuels—one based on the potential for energy production and the other based on total land available for biofuel crops.

For the first method, Edenhofer *et al* (2011) estimate a bioenergy deployment range of 100–300 EJ yr⁻¹ by 2050, which takes into account soil conservation and biodiversity goals, as well as potential water scarcity and the use of land for subsistence farming (Edenhofer *et al* 2011, Creutzig *et al* 2012). For this calculation, it was assumed that by 2050 all bioenergy demand will be supplied by second-generation biofuels. Given data constraints, the estimation focuses on jatropha (*Jatropha curcas*), miscanthus (*Miscanthus x giganteus*), eucalyptus (*Eucalyptus cinera*) and switchgrass (*Panicum virgatum* L). To estimate emissions, a range of published N₂O EFs for these biofuels (0.2 to 27.1 g N₂O-N kJ⁻¹; Hoefnagels *et al* 2010) was used. This approach generates estimates of 0.02–8.1 Tg N₂O-N yr⁻¹ from biofuels by 2050, depending on the fuel source and the total amount of bioenergy deployed, with a central estimate of 2.1 Tg N₂O-N yr⁻¹ based on the combined means of the bioenergy deployment range and the published emission factors for second-generation biofuels.

The second method focuses on the amount of land potentially available to cultivate biofuel crops. Estimates range from 60–3700 Mha, covering 0.4%–28% of the Earth's land surface, excluding Greenland and Antarctica, with several estimates clustering between 240–500 Mha (Creutzig *et al* 2012). In comparison, Melillo *et al* (2009) estimated that 2000 Mha of biofuel crop cultivation will be needed by 2100 to stabilize atmospheric CO₂ concentrations at 550 ppm, while van Vuuren *et al* (2011b) estimated that 3000–4000 Mha will be needed by 2100 in the RCP 2.6 scenario. We assume an average fertilizer application rate of 100 kg N ha⁻¹ yr⁻¹ for land devoted to biofuel crops, as was done by Erisman *et al* (2008), and use the IPCC (2006) direct and indirect default EFs. Using this approach, N₂O emissions were estimated to be 0.08–4.9 Tg N₂O-N yr⁻¹, depending on the amount of land devoted to biofuel crop cultivation, with a

central estimate of 0.5 Tg N₂O-N yr⁻¹ based on the mean of the cluster of land-use estimates cited in Creutzig *et al* (2012).

These estimates are considerably lower than the 16.1–18.6 Tg N₂O-N yr⁻¹ estimated by Melillo *et al* (2009) for 2100. These data illustrate the huge uncertainty that still remains in future estimates of N₂O emissions from biofuels. Comparing these estimates from <1 to 18 Tg N₂O-N yr⁻¹ to the range of the aggregated RCP, SRES and Davidson (2012) scenarios (4.4 to 9.9 Tg N₂O-N yr⁻¹, table 3) demonstrates that biofuels could either remain a relatively trivial source or become the most significant source of anthropogenic N₂O emissions at some point this century. Energy and climate policy decisions in the coming decades as well as the pace of technical innovation will be among the major determinants of future N₂O emissions from biofuels.

6. Conclusions

- Natural N₂O emissions are most likely between 10 and 12 Tg N₂O-N yr⁻¹.
- Both bottom-up and top-down analyses suggest that net anthropogenic N₂O emissions are now (2005–2010) about 5.3 Tg N₂O-N yr⁻¹.
- Agriculture currently accounts for 56–81% of gross anthropogenic N₂O emissions. Some N₂O emissions associated with food production is inevitable, but future N₂O emissions from agriculture will be determined by several factors, including population, dietary habits, and agricultural management to improve N use efficiency.
- The BAU emission scenarios project almost a doubling of anthropogenic N₂O emissions, from 5.3 Tg N₂O-N yr⁻¹ in 2005 to 9.7 Tg N₂O-N yr⁻¹ in 2050. In contrast, the concerted mitigation scenarios result in an average decline to 4.2 Tg N₂O-N yr⁻¹ by 2050, a decrease of 22% relative to 2005, which would lead to a near stabilization of atmospheric concentration of N₂O at about 350 ppb.
- The impact of growing demand for biofuels on future N₂O emissions is highly uncertain, depending on the types of plants grown, their nutrient management, the amount of land dedicated to their cultivation, and the fates of their waste products. N₂O emissions from second and third generation biofuels could remain trivial or could become the most significant source to date. Research is needed to reduce the uncertainty of the future impact of biofuels on N₂O.

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