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SID 5 Research Project Final Report

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Executive Summary

7. The executive summary must not exceed 2 sides in total of A4 and should be understandable to the intelligent non-scientist. It should cover the main objectives, methods and findings of the research, together with any other significant events and options for new work.

Robust emission inventories of atmospheric pollutants are critical to understanding and predicting impacts, identifying key sources and mitigation opportunities. The objective of this study was to review the extent to which land management is accounted for in the UK National Atmospheric Emissions Inventory as a source of emissions of ammonia (NH₃), nitrous oxide (N₂O), nitrogen oxides (NO_x), organic nitrogen (N_{org}), methane (CH₄), non-methane volatile organic compounds (NMVOC), particulate matter (PM) and heavy metals (HM), in comparison with the current state of scientific knowledge; to ascertain whether there is evidence for significant gaps or that key emission sources may have been overlooked.

The first step in identifying these unknowns was to review the current (2007) UK National Atmospheric Emissions Inventory for each atmospheric pollutant, identifying current emission estimates from land management sources. Potential missing sources were then identified through a combination of literature review, expert knowledge and indications from atmospheric budgets that emission estimates were too low. Additionally, sources currently accounted for, but with large uncertainty were also highlighted. Sources were categorised broadly as soil (grassland, cropland and forest soils, cultivation operations, fertiliser and pesticide applications, land use change and riparian zones), plants (crops, forests, burning), livestock (direct emissions and from excreta) and other (including treatment and land application of organic residues). Consideration of the final category is particularly important, given the increasing emphasis on the potential of anaerobic digestion as a renewable energy source and the increasing need to recycle organic residues from a range of domestic and industrial sources back to agriculture rather than to landfill.

Results of the review can be briefly summarised for each atmospheric pollutant as follows, with estimates for the major unaccounted sources show in Table ES1:

Ammonia

More than 90% of NH₃ emissions in the UK inventory are associated with land management sources. There are not considered to be any major unknown sources of NH₃ emission from land management, although there may still be considerable uncertainties in the estimates from many of the known sources (e.g. for outdoor pig and poultry systems where little or no measurement data exist and estimates are based largely on expert opinion). Emissions due to crop senescence are not currently included in the inventory and net emissions have been reported for cereal crops between anthesis and harvest. Across all UK crops, this could potentially increase the emission inventory total by up to 5%. The treatment and land application of organic residues arising from a number of sources represents an area where activity is likely to increase but current quantification of emissions is poor.

Nitrous oxide

The UK inventory of N₂O emissions accounts for emissions from nitrogen applied to soils as fertiliser and livestock excreta and from livestock manure management (together accounting for >70% of total UK emissions) and processes leading to emission are generally well understood. However, IPCC default emission factors are applied for the majority of these sources resulting in considerable uncertainties in the estimates; the use of country-specific emission factors derived from the growing body of UK experimental data and/or through the use of process-based models could reduce these uncertainties. Nitrous oxide emissions associated with land use change are currently not reported in the UK emission inventory because the estimate produced using IPCC guidance methodology is unrealistically large and there is a lack of relevant empirical data. Further work providing empirical datasets and validated models for UK conditions for land use changes are required to provide a robust estimate for this source. There is limited knowledge regarding emissions from composting and the recycling to land of organic residues from household or industrial sources.

Nitrogen oxides

Nitrogen oxides comprise nitric oxide (NO) and nitrogen dioxide (NO₂). In the atmosphere, NO and NO₂ can readily interconvert with one another and have extremely short lifetimes and so are more often considered together as NO_x. Land management currently accounts for <1% of total UK NO_x emissions. Although there are known sources (soils, manure management), quantification is often uncertain or missing. Improved quantification is unlikely to have a significant impact on the UK NO_x emission inventory total as these sources are so small in comparison with other sectors. However, improved quantification may be important to develop a better understanding of tropospheric ozone formation in rural areas.

Organic nitrogen

There is very limited knowledge about sources and processes leading to emissions of N_{org}, and the role that these may play in local and global nitrogen budgets. Organic nitrogen compounds are overlooked components of the transfer of nitrogen from the atmosphere to the earth's surface. Recent reviews have indicated that the contribution of water-soluble organic nitrogen in precipitation to wet deposition may be up to one-third of the total, yet little is known about the chemical composition, form or sources of this material.

Methane

Emissions from land management account for >70% of total UK CH₄ emission, with enteric fermentation by ruminants representing by far the major source. This is fully accounted for in the current UK CH₄ emissions inventory, although largely using default IPCC emission factors. A move to a more country-specific methodology would improve the accuracy of the inventory and the ability to predict the impacts of differing management or mitigation scenarios. Soils can act as a net source or sink of CH₄ and, while more understanding of the processes involved is required, overall impact of including emissions from soils in the UK inventory is likely to be small. Reports from Australia of soil oxidation in grazed pastures providing a CH₄ sink which can exceed enteric emissions of the grazing livestock are not considered to apply to UK conditions, where oxidation rates of soils in productive pastures are much lower and livestock densities much greater. However, further research to provide more robust oxidation rates associated with different soil types, land uses and management practices is warranted. There is no suggestion that direct emissions from plants might be considered a significant source of CH₄ under current UK conditions. Although emissions from manure management are included in the current inventory, using IPCC methodology, recent measurements showed significant CH₄ emissions (up to 4 kg m⁻² year⁻¹) from small-scale features on farmland not currently accounted for in the UK inventory, including seepage to soil from liquid and solid manures, poached land surrounding feeding and water troughs, waterlogged areas and gateways.

Non-methane volatile organic compounds

Biogenic emissions from plants are estimated to account for 90% of total global NMVOC emissions. Emissions from managed crops, pastures and forests have not been robustly quantified for UK conditions and could have important local impacts on troposphere ozone formation. Emissions from livestock and livestock manures may represent a significantly underestimated emission source, potentially increasing the UK inventory by >10%.

Particulate matter

10% of PM₁₀ emissions are currently estimated to arise from land management sources, predominantly from housed livestock. Many uncertainties remain regarding emissions of PM from agriculture and other land management operations. Improved quantification of emission rates from different sources, and the factors influencing these, is needed, as is an improved characterisation of the types of PM from each source; the health impacts of a particulate arising from soil tillage may be quite different from those of a particulate from an intensive poultry house.

Heavy metals

Emissions of HM from land management sources are not currently included in the UK inventory. There are large uncertainties as to the role of agriculture and land management in emissions of HM to the atmosphere (potential sources include contaminated soil particulates), which, although likely to be substantially lower than from industry and traffic sources, may be significant enough to warrant inclusion in the UK inventory.

Importance in the context of Global Warming

Land management is an important source of both N₂O and CH₄, which have large global warming potentials (GWP). Short-lived chemically active gases (NO_x and NMVOC) can have an indirect effect on greenhouse gases (mainly CH₄ and ozone) and aerosols will also directly and indirectly impact on global warming, both positively and negatively. New metrics are being developed in addition to the commonly used GWP, which may better account for the different atmospheric lifetimes of the various gases and improved modelling aims to better account for interactions between gases and aerosols. This increases the importance, therefore, of robust quantification of emissions of both long- and short-lived species from land management sources and highlights the need for integrated assessment of policies targeting air quality (largely the shorter-lived species) and climate change (the longer-lived species).

Table ES1. Major unaccounted sources in the context of current (2007) emission estimates

Species	Source	Emission estimate (kt)	As % of 2007 UK land management emission	As % of 2007 total UK emission	Emission estimate as Mt CO ₂ eq
NH ₃	Crop senescence	2 - 12	1 - 4	1 - 4	
	Composting	3 - 50	1 - 19	1 - 17	
N ₂ O	Land use change	up to 7	up to 9	up to 6	up to 2.1
	Composting	0.3 - 1	<1 - 1	<1 - 1	0.1 - 0.3
NO _x	Soils	13 - 38	1300 - 3800	<1	
CH ₄	Composting	5 - 25	<1 - 1	<1 - 1	0.1 - 0.6
	On-farm 'wet patches'	> 250	>14	>10	>6.3
NMVOC	Crops	76 - 113	690 - 1030	8 - 12	<0.1 - 0.6
	Livestock	20 - 165	180 - 1500	2 - 18	<0.1 - 0.9

In conclusion, there exists a range in the degree to which emission sources of atmospheric pollutants from land management sources have been captured in the UK National Atmospheric Emissions Inventory and in the uncertainties associated with the estimated emissions. For emissions of NH₃, N₂O and CH₄, the processes leading to emission are largely understood and all major sources are thought to be captured in the current inventory, with the exception of N₂O emissions from land use change and CH₄ emissions from unmanaged on-farm 'wet patches'. The latter, in particular, may add considerably to the current greenhouse gas inventory for UK agriculture and more robust measurements and estimates of total source area are required. Quantification of uncertainties in the estimates for some of these sources is still required, as is an assessment of the improvement in accuracy of estimates through the development of country-specific emission factors for N₂O and CH₄ in particular. There is very limited knowledge about sources and processes leading to emissions of N_{org}, and the role that these may play in local and global nitrogen budgets. Land management is known to be a source of NO_x, NMVOC and PM emissions, and potentially also HM emissions. Improved quantification is required to assess the importance of land management as a source of these pollutants in comparison with other sectors and, if appropriate, to determine the potential for mitigation. There is also a need for further consideration of the interaction between chemically active short- and longer-lived atmospheric gases and aerosols and the potential implications on climate change predictions and associated policy development.

Project Report to Defra

8. As a guide this report should be no longer than 20 sides of A4. This report is to provide Defra with details of the outputs of the research project for internal purposes; to meet the terms of the contract; and to allow Defra to publish details of the outputs to meet Environmental Information Regulation or Freedom of Information obligations. This short report to Defra does not preclude contractors from also seeking to publish a full, formal scientific report/paper in an appropriate scientific or other journal/publication. Indeed, Defra actively encourages such publications as part of the contract terms. The report to Defra should include:

- the scientific objectives as set out in the contract;
- the extent to which the objectives set out in the contract have been met;

- details of methods used and the results obtained, including statistical analysis (if appropriate);
- a discussion of the results and their reliability;
- the main implications of the findings;
- possible future work; and
- any action resulting from the research (e.g. IP, Knowledge Transfer).

1. Introduction

Inventories of UK anthropogenic emissions of air pollutants and greenhouse gases are compiled annually (e.g. Dore et al., 2008; www.naei.org.uk) as part of the UK obligation under international legislation for monitoring and reporting (e.g. the United Nations Framework Convention on Climate Change, the UNECE Convention on Long Range Transboundary Air Pollution, The EC National Emission Ceilings Directive). Such inventories are also important in providing information to policy makers on the key polluting sources, so that informed decisions can be made in developing mitigation strategies. Significant uncertainties or omissions from such inventories may result in significant errors in the prediction of potential policy impacts and the implementation of inappropriate measures. For example, the suggestion by Keppler et al. (2006) that living plants are a significant source of methane (CH₄), although since widely refuted (Nisbet et al., 2009; Dueck et al., 2007), would have had important implications for the global CH₄ budget and the role of natural sources of CH₄ in climate change. This review focuses on air pollution and greenhouse gas emissions arising from land management (predominantly agriculture and forestry) in the UK, attempting to identify key unknowns and uncertainties and the implications these may have for policy development and future research requirements.

The atmospheric pollutants considered in this review are ammonia (NH₃), nitrous oxide (N₂O), nitrogen oxides (NO_x), organic nitrogen (N_{org}), methane (CH₄), non-methane volatile organic compounds (NMVOC), particulate matter (PM) and heavy metals (HM). Ammonia is of concern because of potential damage to sensitive ecosystems through acidification and eutrophication, and also because of its role in the formation of secondary PM (through reaction in the atmosphere with sulphate or nitrate) and their implications regarding human health (Erisman et al., 2007). Nitrous oxide and CH₄ are powerful greenhouse gases, with global warming potentials of 298 and 25 times that of carbon dioxide, respectively (Forster et al., 2007). Nitrous oxide is now also considered as the major anthropogenic emission causing stratospheric ozone depletion (Ravishankara et al., 2009). Nitrogen oxides are associated with human health impacts (respiratory diseases), contribute to the production of photochemical smog in the presence of sunlight and in the formation of tropospheric ozone (with similar human health effects and also leading to crop yield reductions and ecosystem damage) if VOC are also present (Pandis et al., 1991). The sources, composition and deposition of N_{org} in the atmosphere are important in understanding local and global nitrogen budgets and their environmental impacts (e.g. Neff et al., 2002). Atmospheric deposition of HM to aquatic and terrestrial ecosystems can have direct toxic effects on plants and animals, as well as wider impacts, as they accumulate through the food chain.

2. Scientific objectives

The objective of this study was to review the extent to which land management is accounted for as a source of emission of these various atmospheric pollutants and greenhouse gases according to the current state of scientific knowledge and to ascertain whether there is evidence for significant gaps, uncertainties or key emission sources that may have been overlooked.

3. Approach

'Unknowns' were defined according to 4 categories:

- a) Unknown source with unquantified emission;
- b) Known source with unquantified emission;
- c) Known source with a rough estimate, but unknown uncertainty;
- d) Known from estimates for other parts of the world, but no specific data for the UK.

The first step in identifying these unknowns was to review the current (2007) UK emissions inventory for each atmospheric pollutant (Table 1, excepting N_{org}, which is not reported). Data for Table 1 were taken from the UK National Atmospheric Emissions Inventory web-site (<http://www.naei.org.uk/>), where emission sources are categorised according to the EMEP/CORINAIR Nomenclature for Reporting (NFR) categories. Those considered as land management for the purposes of this review include Agriculture (NFR 4), Forest and Grassland Conversion (NFR 5) and some elements of Waste Management (NFR 6). In the current UK inventory of atmospheric pollutants, therefore, land management sources account for the majority of NH₃, N₂O and CH₄ emissions, are a significant source for emissions of PM₁₀, but could be considered insignificant for NO_x, NMVOC and HM.

Table 1. Total UK emission of selected atmospheric pollutants in 2007 and the proportion arising from land management (Dore et al., 2008)

	Total anthropogenic UK emission (kt)	Emission from UK land management sources [†] (kt)	Proportion from land management sources (%)
NH ₃	289	267	92
N ₂ O	110	81	73
NO _x	1486	1	<1
CH ₄	2327	1840	79
NMVOG	942	11	1
PM ₁₀	135	14	10
HM	Varies with metal	unknown	unknown

[†]Agriculture, forest and grass conversion, solid waste disposal to land, composting and anaerobic digestion

Potential missing sources were then identified through a combination of literature review, expert knowledge and indications from atmospheric budgets that emission estimates were too low. Sources were categorised broadly as soil (grassland, cropland and forest soils, cultivation operations, fertiliser and pesticide applications, land use change and riparian zones), plants (crops, forests, burning), livestock (direct emissions and from excreta) and other (including treatment and land application of organic residues other than livestock excreta). Consideration of the final category is particularly important, given the increasing emphasis on the potential of anaerobic digestion as a renewable energy source and the increasing need to recycle organic residues from a range of domestic and industrial sources back to agriculture rather than to landfill.

4. Results and Discussion

A summary of the current state of knowledge regarding potential UK land management emission sources for each of the considered atmospheric pollutants is given in Table 2.

4.1 Ammonia

Detailed emission inventories for agricultural (Misselbrook et al., 2000) and non-agricultural (Sutton et al., 2000) sources revealed emissions to be lower than expected from atmospheric budget studies (Sutton et al., 2000). This gap was closed to a large extent through further refinement of inventory estimates and, in particular, the inclusion of emissions from outdoor yards used by livestock which had previously not been accounted for (Misselbrook et al., 2006; Misselbrook et al., 2001). There are not considered, therefore, to be any major unknown sources of NH₃ emission from land management, although there may still be considerable uncertainties in the estimates from many of the known sources (e.g. for outdoor pig and poultry systems where little or no measurement data exist and estimates are based largely on expert opinion). Direct emissions from livestock via, e.g., sweat have not been discretely quantified, but are accounted for in measurements of emission from livestock housing or grazing.

Decomposition of litter on forest floors might be expected to be associated with NH₃ emission, although reports of measurements specifically from this source were not found. (Razgulín and Stepanov (2009) reported a small flux from the soil under a Russian bilberry-birch forest). More generally, forests and other extensively land areas are associated with net deposition of NH₃, although there can be short periods of emission (e.g. Horvath, 2004; Sutton et al., 1997; Sutton et al., 1993a). Non-inclusion of these sources in the UK NH₃ emission inventory is therefore of little significance, although inclusion of bidirectional NH₃ exchange from such land areas is important in subsequent transport and deposition modelling.

Burning biomass, whether above-ground vegetation or organic soils, produces ammonia (NH₃) gas. An estimate of NH₃ emissions from controlled biomass burning in the UK can be made on the assumption that this is dominated from muirburn of heather moorlands. There are no data available for measurements of emissions from heather burning specifically, so estimates are based on the proportion of 'fuel' N emitted as NH₃ or the ratio of NH₃ emission to 'fuel' dry matter derived from a number of biomass burning studies (e.g. Bertschi et al., 2003; Lee and Atkins, 1994; Allen, 1964). Although considerable uncertainty exists in the estimate of emission for this source, a top of the range estimate would still represent <1% of total UK NH₃ emission.

Bidirectional exchange of NH₃ in fertilised agricultural grassland and crops has been relatively well studied in recent years and it is known that there are periods of net emission associated with fertiliser applications and periods of crop senescence (Milford et al., 2009; Nemitz et al., 2000; Sutton et al., 1993b; Whitehead et al., 1988). The current UK inventory estimate of NH₃ emissions from fertiliser applications to crops and grassland acknowledges that the emission derives from the combined soil and plant system (Misselbrook et al., 2004). However, emissions due to senescence are not currently included in the inventory. Schjoerring and Mattsson

Table 2. Current quantification of atmospheric pollutants from land management sources in the UK inventory: X = unlikely source; Y = a known source, quantified with known uncertainty; U1 = unknown source, unknown quantity; U2 = known source, unquantified; U3 = known source, basic quantification, but unquantified uncertainty; U4 = known source not in UK inventory

Soil	NH ₃	N ₂ O	NO _x	N _{org}	CH ₄	NMVOC	PM	HM
Cultivation, management	X	U2	U3	U1	U3	X	U3	U4
Fertilizers, pesticides	Y	Y	Y	U2	X	U2	X	U2
Forests and extensively managed land - soils	U1	U4	U4	U1	U4	X	X	X
Land use change	X	U2	U3	U2	U4	X	U2	U1
Riparian zones and water bodies (incl. fish farming)	X/U2	U3	X	X	U4	X	X	X
Plants								
Plants crops and grass	U3/U4	U2	U2	U2	U2	U3	U3/U4	X
Forests and extensively managed land - plants	U4	U2	U2	U1	U2	Y/U4	U3/U4	X
Land burning	U3	U3	U2	U2	U3	U3	U3	U2/U3
Livestock								
Livestock - direct	U2	U2	X	X	Y	U2	U3	X
Livestock - excreta	Y	Y	U3	U3	Y	U3	U2/3	U1
Other								
Anaerobic digestion	U2	U1	U1	U1	U4	U2	U1	X
Composting	Y/U3	U3	U2	U2	U3	U2	U2	X
Application of organic residues to land	U3	Y/U3	U3	U2	U3	U3	U2	U1

(2001) reported seasonal emissions of 1 - 5 kg ha⁻¹ from fertilised arable crops in Denmark, representing 1 - 4% of applied fertiliser nitrogen, with 50% or more occurring between anthesis and harvest. Across UK crops, this could potentially increase the emission inventory total by up to 12 kt NH₃.

Riparian zones would not generally be associated with direct nitrogen inputs as part of water quality management plans and significant NH₃ emissions would not be expected. Measurements from a grazed riparian ecosystem in the USA (Walker et al., 2002) were of the same order as emissions from conventionally grazed pasture systems, so there would be no justification for adding this as a discrete source to the UK inventory. Inland fish farming enterprises may be associated with NH₃ emissions (Aubin et al., 2006; Gross et al., 1999), although the scale and magnitude of these is likely to be small.

Composting of organic residues is associated with NH₃ emissions, although for green waste (grass, woodchips, prunings) these emissions might be considered biogenic and may even be lower from composting than from natural degradation (Chou and Buyuksonmez, 2006). Ammonia emissions from composting of household waste, which contain significant quantities of food waste, can be up to 30% of the initial nitrogen content, depending on operating conditions, although some of this may be recovered in condensate (Jarvis et al., 2009; Eklind et al., 2007; Beck-Friis et al., 2001). Assuming that approximately 10 million tonnes of UK household waste is composted, with a total N content of between 0.5 - 2% and an emission of between 5 - 20% of initial total N, NH₃ emission from this source could be in the range 3 - 50 kt NH₃. Active composting of livestock manure (i.e. regular turning of the manure heap) increases NH₃ emissions compared with conventionally stored manure (Parkinson et al., 2004), although this is not currently widely practised in the UK. Anaerobic digestion of livestock slurries results in a material with a lower total solids content and a higher pH (Sommer, 1997; Sommer and Husted, 1995), potentially increasing NH₃ emissions from post-digestion slurry stores if left uncovered. However, the counteracting influences of the pH and total solids content changes may result in little overall difference in NH₃ emissions following land spreading of digested compared with undigested slurry (Pain et al., 1990). Co-digestion with food waste or other organic residues can increase the amount of nitrogen in the system and potentially increase NH₃ emissions depending on operating and storage conditions. Subsequent application of the digestate to land also represents an emission source. The treatment and land application of organic residues arising from a number of sources therefore represents an area where activity is likely to increase but current quantification of emissions is poor.

4.2 Nitrous oxide

Nitrous oxide emissions arise predominantly from the processes of nitrification and denitrification, with soils being the major source. In general, N₂O production increases with increasing concentrations of ammonium and nitrate, available organic C (carbon) content and with increasing soil wetness and soil density (Skiba and Smith, 2000). However, when the soil becomes too wet, N₂O is further reduced to N₂, therefore N₂O emissions decrease (Davidson, 1991). The UK inventory of N₂O emissions accounts for emissions from nitrogen applied to soils as fertiliser and livestock excreta and from livestock manure management. However, IPCC default emission factors are applied for the majority of these sources, resulting in considerable uncertainties in the estimates; the use of country-specific emission factors derived from the growing body of UK experimental data and/or through the use of process-based models with detailed activity data could reduce these uncertainties substantially.

The influences of soil cultivation, irrigation and drainage on N₂O emissions depend on soil conditions. Ploughing and drainage of a heavy soil, in which denitrification to N₂ dominates over N₂O, would increase rates of N₂O production and emission. For a less heavy soil, the opposite may be the case. Irrigation would promote the anaerobicity required for denitrification. Depending on the soil characteristics, this may increase N₂O emissions or, for a heavier soil, may promote denitrification to N₂, thereby reducing N₂O emissions. While some data and models are available (e.g. Chen et al., 2008), significant uncertainties exist in our ability to quantify the effect of these processes on N₂O emissions.

Nitrous oxide is also produced in soils during nitrate ammonification, but the contribution of this process to emissions from soils is unknown. While likely to be a small source in comparison with denitrification, little is known about the environmental conditions conducive to this process. Current research (Defra project AC0105) seeks to address this and provide information on the potential magnitude of this source and for finding conditions conducive to the mitigation of N₂O production during ammonification, which are likely to differ from those for denitrification.

Nitrous oxide concentrations in the soil are much larger than in the atmosphere, and consequently the soil water may be supersaturated with N₂O. This can result in plant mediated emissions, with emissions of N₂O through the plant leaves (e.g. Chang et al., 1998). For most short vegetation, plant mediated emissions are accounted for by the typical chamber measurements. However the importance of this transport in larger crops and trees, in particular on soils likely to have large concentrations of N₂O has yet to be determined.

Nitrous oxide emissions associated with land use change are currently not reported in the UK emission inventory because the estimate produced using IPCC guidance methodology (c. 7 kt N₂O) is very uncertain and there is a lack of relevant empirical data (Jackson et al., 2009). Land use change is generally associated with net mineralization (e.g. conversion of forests or grassland to cropland) or immobilization (e.g. conversion of cropland to grassland) of soil nitrogen, thereby increasing or decreasing the potential for N₂O emissions, respectively. Further work providing empirical datasets and validated models for UK conditions for land use changes is required to provide a robust estimate for this source. Emissions due to changes in fertiliser use associated with land use change are accounted for in fertiliser use statistics as input to the UK emission inventory. The total area in the UK converted to wetlands is restricted to small insignificant areas of newly created riparian zones, along rivers in nitrogen vulnerable zones (NVZ). This activity has a potential to decrease N₂O emissions, if the soil water filled pore space (WFPS) can be maintained above 90% (Skiba and Smith, 2000). Anaerobic conditions and the accumulation of soil organic matter content favour denitrification to proceed to N₂ rather than stop at N₂O production, which is generally the case in more aerobic soils.

Composting is a source of N₂O. A study on household waste showed emissions ranging from 1–1464 mg N₂O m⁻² day⁻¹. This large range in emissions indicates the importance of compost management (Beck-Friis et al., 2000). Composting of green waste was shown to have an emission factor of 0.5% of the total nitrogen content of the initial material (Hellebrand, 1998). Assuming 10 million tonnes of UK household waste is composted, with a total N content of between 0.5 – 2%, this would increase the estimate of total UK N₂O emissions by 0.3 – 1.5 kt N₂O. Organic amendments to land containing carbon and nitrogen will be a potential source of N₂O emissions. Apart from livestock manures, these are not currently accounted for in the UK N₂O emissions inventory. Further work is required to characterize the composition of such amendments and predict the subsequent N₂O emissions for given application rates, timings and conditions.

4.3 Nitrogen oxides

Nitrogen oxides comprise nitric oxide (NO) and nitrogen dioxide (NO₂). In the atmosphere, NO and NO₂ can readily interconvert with one another and have extremely short lifetimes and so are more often considered together as NO_x. As for N₂O, NO emissions from soils are a product of microbial nitrification and denitrification, with the ratio of NO and N₂O emission to the atmosphere being controlled by soil physical and chemical characteristics, soil management and climate (Skiba et al., 1997).

Soil N₂O fluxes from the dominant terrestrial ecosystems in Europe have been studied extensively, but similar research on NO is limited to many fewer studies. Reasons for this are that it is more complex to measure NO fluxes and, whereas soil microbial processes are the main source of N₂O, they only contribute to less than 10% of total NO_x emissions in industrialised nations and only 3% in the UK (Stohl et al., 1996).

Measurements of NO in livestock breath have been made in relation to detecting respiratory diseases (Roller et al., 2007), but concentrations are extremely small and this can be considered an insignificant source. Livestock excreta spread onto fields are a known source of NO (e.g. Vallejo et al., 2005; Paul et al., 1993). Emissions from other organic amendments applied to soils are likely to be of the same order, related to N input, although the C:N ratio of the amendment is also likely to be an influencing factor. Groenestein and Faassens (1996) showed that deep litter housing for pigs was only a very small source of NO emission. However, there is little information on NO emissions from manure storage systems. Anaerobic and wet manure storage systems are an unlikely source of significant rates of NO, but dry storage systems and dung heaps may be, especially during low rainfall periods when the surface layer may be well aerated and promote nitrification and NO release.

Nitric oxide emissions from fertiliser applications to soils have been better quantified and modelled than from other land management sources, with data suggesting emission factors broadly similar to those for N₂O (Stehfest and Bouwman, 2006; Bouwman et al., 2002). Soil tillage reduces the WFPS and increase the air filled porosity of the soil, thereby increasing aerobic microbial processes, including mineralisation and nitrification, and has been shown to significantly increase NO emissions (Pinto et al., 2004; Skiba and Ball, 2002; Yamulki and Jarvis, 2002; Skiba et al., 1997). Based on the limited data available, Skiba et al. (1997) estimated an emission from UK agricultural soils of between 13 – 38 kt NO. Information on NO emissions from forest soils also exists. A forest specific version of the DNDC model has been developed (Butterbach-Bahl et al., 2001) and applied to simulate NO emissions from all European forests (Kesik et al., 2005). Butterbach-Bahl et al. (2009) used combined agricultural and forest versions of DNDC to estimate European-wide soil NO emission, estimating that it contributed 4–6% of total NO_x, but that the contribution may increase to >10% in early summer, when fertilizer-induced emissions peak.

Plants can act as a significant sink for NO_x (Fowler et al., 1998; Davidson and Kingerlee, 1997) but also as a direct source of NO. It has been shown that senescing leaves emit NO together with ethylene (Leshem et al., 1998). Nitric oxide emission rates from intact leaves are generally low and only account for 0.01–0.1% of the nitrate reductase activity in the tissue (Rockel et al., 2002). Wildt et al. (1997) showed a direct relationship between NO emissions and CO₂ uptake by a range of plants. From this relationship he deduced that globally

plants emit 0.23 Tg N y^{-1} , which is less than 0.5 % of the total global NO_x emission. For short vegetation, chamber measurements will account for direct plant emissions in addition to soil flux, but direct plant emissions may not have been fully considered for taller crops and forests.

Processes associated with land use change that influence N_2O emissions are also likely to influence NO emissions in a similar way, although little or no data exist for this source. Similarly, no data exist for NO emissions from riparian zones, but these are unlikely to represent a significant source.

Literature values for NO emissions from composting could not be identified. However, Hellebrand (1998) showed that N_2O and CH_4 emissions from the composting of green waste could be reduced by aeration, suggesting that under those conditions NO emissions are likely to be the dominant source of N release. If aeration of composts becomes a commonly used practice to mitigate possible N_2O and CH_4 emissions, then the importance of NO should be investigated.

Land management is known to be a source of NO_x emissions, although quantification from discrete sources is often uncertain or missing. Improved quantification is unlikely to have a significant impact on the UK NO_x emission inventory total, as these sources are so small in comparison with other sectors. However, improved quantification may be important to develop a better understanding of tropospheric ozone formation in rural areas.

4.4 Organic nitrogen

Organic nitrogen compounds are overlooked components of the transfer of nitrogen from the atmosphere to the earth's surface. Recent reviews (Cornell et al., 2003; Keene et al., 2002; Neff et al., 2002) have indicated that the contribution of water-soluble organic nitrogen (WSON) in precipitation to wet deposition may be up to one-third of the total, yet little is known about the chemical composition, form or sources of this material. Initial scepticism about the nature of WSON has to some extent been dispelled (Cape, 2001), but the broad range of possible composition and emission sources means that the transfer pathways are still somewhat uncertain. It is known, for example, that both biological and chemical processes interconvert inorganic and organic nitrogen (Zhang and Anastasio, 2003; Calvert, 2002), but it is not clear how much biological and chemical activity may occur in the atmosphere or on the surfaces of sampling equipment. The presence of both gaseous and particulate WSON in the atmosphere implies that dry deposition is an important but yet unquantified pathway for transfer of organic nitrogen to the earth's surface.

Recent studies in the UK have shown that the careful application of quality control measures can result in the unequivocal measurement of N_{org} in precipitation (Cape, 2001). The use of a biocide as a general principle prevents microbial degradation of a rain sample both during and after sampling, although in practice, microbial losses may be small at low ambient temperatures. Application of these techniques to a network of sites across the United Kingdom over a period of up to 2 years showed a consistent pattern of N_{org} in rainfall across the country. Expressed as a percentage of total water-soluble nitrogen, the contribution of N_{org} was least in the north and east of the country and greatest in the south and west, and ranged from 24 to 40% (Cape et al., 2004). Although there appeared to be a positive correlation between annual average N_{org} and ammonium (NH_4^+) concentrations across the sites, the seasonal patterns showed that N_{org} and NH_4^+ were not strongly correlated through the year.

The chemical composition of the water-soluble N_{org} is not known. The sources of these compounds in air and rain are also unknown; possibilities include urea and amines (from agriculture), hydrogen cyanide and methyl cyanide (from biomass burning), organic nitrates (from NO_x and NMVOC emissions) and biological debris (resuspended/emitted). The quantities of emissions of water-soluble N_{org} are likely to be comparable to those of NH_3 .

Although the contribution of organic N to both wet and dry N deposition is very uncertain, it is likely that some if not all water-soluble organic N compounds such as amino-acids are readily assimilated by plants (e.g. Gallet-Budynek et al., 2009) and therefore may contribute directly to eutrophication. Moreover, some components such as nitrophenols are thought to be phytotoxic (Natangelo et al., 1999). The difficulty in measuring and quantifying such compounds has meant that our estimates of N deposition are systematically low by a significant proportion over much of the UK.

4.5 Methane

Enteric fermentation by ruminants represents the major source of CH_4 emissions from land management activities. This is fully accounted for in the current UK CH_4 emissions inventory, although largely using default IPCC Tier 1 emission factors. A move to a more country-specific methodology (Tier 2 or Tier 3) would improve the accuracy of the inventory and the ability to predict the impacts of differing management or mitigation scenarios.

The net CH₄ emission from soil is influenced by the activity of two microbial communities, the methanogens and CH₄ oxidisers. Methane production requires strict anaerobic conditions and in the UK the wettest parts of moorlands are the largest source of CH₄, but even the contribution of these to the total national emission is less than 5%. Most of the CH₄ produced in the deeper anaerobic layers of a soil by the methanogens is oxidised by the CH₄ oxidisers in the aerated upper parts of the same soil. Methane oxidation rates vary widely across different land use types, with rates of 1 – 200 µg m⁻² h⁻¹ being reported (Smith et al., 2000). Conversion of natural land to managed (agricultural) land results in significant decreases in soil CH₄ oxidation rates, typically by 70%, with very slow recovery rates on reversion (Smith et al., 2000). Willison et al. (1995), in common with others, reported CH₄ oxidation rates in soils to follow the order woodland > grassland > arable. Nitrogen fertiliser has been shown to reduce CH₄ oxidation, especially when applied in the ammonium form (Ojima et al., 1993; Powlsen et al., 1997; Smith et al., 2000). The inhibiting action of N additions is due in the short term to ammonium interfering with the methanotrophic enzyme system (Boeckx and van Cleemput, 1996) and in the longer term to changes in the microbial community. Organic N additions (e.g. livestock manure) have much less effect than mineral fertilisers on oxidation rates (Powlsen et al., 1997; Seghers et al., 2003). Tillage is often associated with a decrease in soil CH₄ oxidation rate, but Suwanwaree and Robertson (2005) argued that the effects of tillage and increased N availability were often confounded and in a controlled experiment reported no effect of soil disturbance (simulated tillage) per se. Increasing soil moisture is associated with decreasing CH₄ oxidation rate (Mosier et al., 1997). Various agrochemicals (herbicides and pesticides) have also been shown to inhibit CH₄ oxidation (e.g. Boeckx et al., 1998).

Recent studies in Australia have suggested that the CH₄ oxidation in grazed pastures may exceed enteric emissions by grazing cattle. Prof Mark Adams of the University of Sydney has measured CH₄ oxidation rates of 100 µg m⁻² h⁻¹ in relatively high organic matter soils (5 – 7%) in the Snowy Mountains region of Australia, under extensive grazing. Li and Kelliher (2007) compared oxidation rates in freely and poorly drained grassland soils in New Zealand, with and without additions of cattle urine, reporting mean rates of 20 and 7 µg m⁻² h⁻¹ for the freely and poorly drained soils, respectively, and showed significant short-term reductions (40 – 60%) following the addition of urine. Oxidation rates for soils under grazed pastures in the UK would more typically be in the range 0 – 10 µg m⁻² h⁻¹ (Powlsen et al., 1997) and the much higher stocking rates than for Australia (and therefore greater urine additions) would mean that enteric emissions from grazing livestock farms in the UK would far exceed the CH₄ oxidation sink of grazed pastures. Changes in farming practices aimed at increasing soil CH₄ oxidation rates are unlikely to have significant effects on total net CH₄ emissions for the UK, although further research to provide more robust oxidation rates associated with different soil types, land uses and management practices is warranted.

That plants can act as a conduit for CH₄, particularly in wetland, has long been known (e.g. Sebacher et al., 1985). However, more recent observations have suggested that direct plant emissions of CH₄ may be significant under some circumstances (e.g. Keppler et al., 2006). Although there is still some debate regarding the mechanism and significance of this (Nisbet et al., 2009; Beerling et al., 2008; Dueck et al., 2007), Qaderi and Reid (2009) have suggested that such emissions may increase under climatic stress conditions (higher temperatures and drought). There is no suggestion that this might be considered a significant source of CH₄ under current UK conditions.

Composting of household waste and green waste is a source of CH₄, although emission rates can vary greatly according to compost management (; Andersen et al., 2010; Beck-Friis et al., 2000). Applying reported emission factors of between 0.5 and 2.4 g CH₄ kg⁻¹ fresh material (Andersen et al., 2010; Beck-Friis et al., 2000) to the 10 million tonnes of household waste in the UK that is composted gives an estimated emission of between 5 – 25 kt CH₄.

Methane emissions from livestock manure management, including emissions from manure applications and grazing returns to soil, are accounted for in the UK CH₄ emission inventory, using IPCC default emission factors specific to manure management systems. However, recent measurements showed significant CH₄ emissions (up to 4 kg m⁻² year⁻¹) from small-scale features on farmland not currently accounted for in the UK inventory, including seepage to soil from liquid and solid manures, poached land surrounding feeding and water troughs, waterlogged areas and gateways (Matthews et al., 2009). Although small in area (<5% of total farm area), accounting for emissions from these sources greatly increased the estimate of CH₄ emission from the total farm area (by between 15 – 150%) as calculated according to default IPCC methodology. These results are from only 4 livestock farms and further data (emission measurements and source area estimates) are required to provide a robust basis for scaling to national impact, but applying the lower end of this estimate to the current inventory total would increase UK CH₄ emissions by 250 kt.

4.6 Non-methane volatile organic compounds

Biogenic plant sources of NMVOC are estimated to account for 90% of total global NMVOC emissions (Guenther et al., 1995). An estimate of biogenic NMVOC emission is reported in the UK inventory, but not included in the UK total. Isoprene is a major component of plant emissions, particularly from trees, where it may have evolved as a

mechanism to protect leaves from sunlight-induced rapid heating (Sharkey et al., 2008). Oxygenated hydrocarbons (ethanol, methanol) are more dominant in emissions from pasture, with lower overall emission rates than from forests (Kirstine et al., 1998), although cutting and drying are associated with elevated fluxes (de Gouw et al., 1999; Kirstine et al., 1998). Kirstine and Galbally (2004) estimated that NMVOC from grass and grass cutting in cities could account for up to 5% of total NMVOC and that grass cutting behaviour may contribute to weekday/weekend ozone concentration differences. Elevated emissions of NMVOC have been measured during the flowering period of oil seed rape (Muller et al., 2002), an important crop in the UK. Karl et al. (2009) estimated NMVOC emissions from agricultural crops in Great Britain to be 113 kt, somewhat larger than an earlier estimate by Simpson et al. (1999) of 76 kt. Land use and management may therefore have important local or regional impacts on tropospheric ozone formation through the emission of biogenic NMVOC, but this has not been well characterised for the UK.

Emissions of NMVOC are known to occur directly from livestock (Hobbs and Mottram, 2000) and livestock manure (Ngwabie et al., 2008; Hobbs et al., 2004), although Shaw et al. (2007) reported that the dominant NMVOC emitted from a dairy house in California had ozone formation potentials of approximately 10% of typical combustion or biogenic NMVOC. Sun et al. (2008) found alcohols (methanol and ethanol) to be the major NMVOC from dairy cows (volatile fatty acid concentrations were largely below detection limits), with most of the emission arising from the manure rather than directly from the animal. They estimated mean flux of alcohols to exceed $0.25 \text{ g cow}^{-1} \text{ h}^{-1}$ (Sun et al., 2008). Applying this emission factor to the UK cattle population results in a total emission of c. 20kt NMVOC. Hobbs et al. (2004) estimated total NMVOC emission of 165 kt from UK livestock, which, if included in the UK inventory, would increase the UK total emission by >10%.

Biomass burning is associated with NMVOC emissions (Bertschi et al., 2003; Goode et al., 1999) and burning of heather moorland might therefore be locally important as an emission source, although contributing little to the national total.

4.7 Particulate matter

Approximately 10% of primary PM₁₀ emissions (i.e. directly emitted, not including the formation of secondary PM from atmospheric processes) reported in the UK inventory are from land management sources, based largely on initial estimates by Phillips et al. (2002), with housed livestock (predominantly poultry) as the major source. However, it is acknowledged that there are large uncertainties in the estimated emission factors. At the European scale (EU15), agriculture is estimated to account for 25% of PM₁₀ and 5% of PM_{2.5}, although there is a large gap between measured atmospheric PM levels and those modelled, suggesting that emissions from agricultural sources, amongst others, may have been underestimated (Erisman et al., 2008). Mitigation of point sources of PM emission within other sectors is likely to increase the overall importance of PM emissions from the more diffuse sources within the agricultural sector (Klimont and Amann, 2002), increasing the requirement for more robust estimates.

Emissions of PM from intensive livestock housing are influenced by feeding practices, animal type and environmental factors, resulting in a broad range in reported emission rates (Cambra-Lopez et al., 2010). In pig housing, feed and faecal matter are cited as being the major source of PM, while in poultry housing the major sources are litter, down feathers and mineral crystals from excreta (Cambra-Lopez et al., 2010). Straw-bedded cattle and pig housing systems, common in the UK, might also be expected to be associated with PM emissions, but there has been little quantification to date. Additionally, there has been no quantification of PM emissions from slurry and solid manure spreading operations.

Many uncertainties still remain, therefore, regarding emissions of PM from agriculture and other land management operations. Improved quantification of emission rates from different sources, and the factors influencing these, is needed, as is an improved characterisation of the types of PM from each source; the health impacts of a particulate arising from soil tillage may be quite different from those of a particulate from an intensive poultry house.

4.8 Heavy metals

Agriculture and land management are not considered as sources of HM emissions to the atmosphere in the current UK inventory (Table 1). However, results of a study in Taiwan indicated that soil was implicated as the source for 20 – 40% of particulate metals measured at a farm sampling site, with industry and traffic being the other sources (Fang et al., 2003). Heavy metal inputs to agricultural soils in England and Wales have been quantified, showing livestock manures (c. 30% of Zn and Cu) and inorganic fertilisers (20% of Cd and 10% of Ni) to be important sources (Nicholson and Chambers, 2006), with that in manures deriving from the metal content of feed (Nicholson et al., 1999). Additional sources were identified as atmospheric deposition, sewage sludge, agrochemicals, industrial wastes, green wastes compost, canal and river dredgings, metal corrosion, lead shot and irrigation water (Nicholson and Chambers, 2006). As with PM emissions, manure spreading and the application of pesticides, herbicides and fungicides may represent a currently unquantified direct source of HM

emissions to the atmosphere, while re-suspension (e.g. during soil cultivation) of HM deposited on agricultural land may represent an unquantified secondary source.

There are therefore large uncertainties as to the role of agriculture and land management in emissions of HM to the atmosphere, which, although likely to be substantially lower than from industry and traffic sources, may be significant enough to warrant inclusion in the UK inventory.

4.10 Importance in the context of Global Warming

An important consideration with potentially highly relevant policy implications are the impacts of the atmospheric pollutants being reviewed on global warming. Greenhouse gases contribute to climate change by absorbing the Earth's infrared radiation that would otherwise escape to space. The major greenhouse gases are water vapour, carbon dioxide, CH₄, N₂O and tropospheric ozone. As discussed above, land management is an important source of both N₂O and CH₄, which have large global warming potentials (GWP, relative to a value of 1 for carbon dioxide) and long atmospheric lifetimes (Table 3).

Short-lived chemically active gases (NO_x and NMVOC) can have an indirect effect on greenhouse gases (mainly CH₄ and ozone) and aerosols will also directly and indirectly impact on global warming (e.g. Shindell et al., 2009). Nitrogen oxides react in the troposphere to cause a decline in CH₄ concentrations, and have short-term (increasing) and longer term (decreasing) effects on ozone concentration. The net effect on radiative forcing depends on the cancellation of these effects and generally results in a net global cooling effect, although there is strong dependence on the magnitude, location and season of NO_x emission (Derwent et al., 2008). Non methane VOC in the atmosphere are precursors of tropospheric ozone and therefore have a positive global warming effect. Aerosols can directly alter the radiative balance of the Earth-atmosphere system by scattering and absorbing shortwave and longwave radiation. The indirect effect of aerosols is the mechanism by which they modify the microphysical and hence the radiative properties, amount and lifetime of clouds. The net effect of the aerosol is dependent on the nature of the aerosol and can be very strongly positive (e.g. black carbon) or negative (e.g. organic carbon, sulphate), with a range of GWP₁₀₀ values reported (Fuglestedt et al., 2009), adding importance to improving the speciation of PM emissions from land management sources.

Although GWP₁₀₀ values (GWP over a 100 year time horizon) can be computed for short-lived species (Table 3), uncertainties in current chemical transport models and the markedly different climatic effects for the same mass emissions from different locations mean it is difficult to adopt these within current policy making. However, recent work by Shindell et al. (2009) has shown the impact that short-lived species can have on GWP values of greenhouse gases, particularly taking account of the direct and indirect effects of aerosols (Table 3). Although subject to large uncertainties, the estimated impact on the GWP value of CH₄, in particular, is significant and perhaps should be accounted for in optimisation of climate change mitigation policies.

Table 3. Global warming potential (100 year time horizon) and atmospheric lifetime for relevant species without and with inclusion of direct and indirect effect of aerosols (after Shindell et al., 2009)

Species	Atmospheric lifetime	GWP ₁₀₀			UK 2007 emission (Mt CO ₂ eq) [‡]
		Not incl. aerosol effects [†]	Incl. direct aerosol	Incl direct+indirect aerosol	
N ₂ O	114 years	298			32.8
CH ₄	12 years	25	24 to 33	26 to 41	58.2
NH ₃	10 days	0	-19	-15	
NO _x	<1 day	-10 to -30	-60 to -125	-75 to -240	
NMVOC	Typically <1 day	0.5 to 5.5	2 to 5	3 to 8	0.5 to 5.2
PM	4 – 7 days	-70 to 460			

[†]From IPCC 4th assessment report

[‡]Based on reported emission of gas and GWP₁₀₀ from this table

Recent discussions have been examining the use of GWP₁₀₀ values against other metrics (such as the time-dependent global temperature change potential), particularly with respect to the relative importance of shorter- and longer-lived atmospheric species (e.g. Fuglestedt et al., 2009; Shine et al., 2007). The development of improved atmospheric modelling and the potential benefits of a move to a temperature change metric for greenhouse gases (Shine et al., 2007) increase the importance of robust quantification of emissions of both long- and short-lived species from land management sources. This highlights the need for integrated assessment of policies targeting air quality (largely the shorter-lived species) and climate change (the longer-lived species).

4.10 Estimating the magnitude and significance of the key unaccounted sources

Table 4 provides estimates for the major unaccounted sources identified in this review, in the context of current UK total emission and that from land management. These values must be treated with caution because of the large uncertainties associated with them (as they are based on very limited data not necessarily representative of UK-wide conditions), but they are useful in providing an indication of the areas where more data are required.

Table 4. Major unaccounted sources in the context of current (2007) emission estimates (as shown in Table 1)

Species	Source	Emission estimate (kt)	As % of 2007 UK land management emission	As % of 2007 total UK emission	Emission estimate as Mt CO ₂ eq
NH ₃	Crop senescence	2 - 12	1 - 4	1 - 4	
	Composting	3 - 50	1 - 19	1 - 17	
N ₂ O	Land use change	up to 7	up to 9	up to 6	up to 2.1
	Composting	0.3 - 1	<1 - 1	<1 - 1	0.1 - 0.3
NO _x	Soils	13 - 38	1300 - 3800	<1	
CH ₄	Composting	5 - 25	<1 - 1	<1 - 1	0.1 - 0.6
	On-farm 'wet patches'	> 250	>14	>10	>6.3
NMVOC	Crops	76 - 113	690 - 1030	8 - 12	<0.1 - 0.6
	Livestock	20 - 165	180 - 1500	2 - 18	<0.1 - 0.9

In terms of their significance with respect to total emissions from UK land management, each of the categories in Table 4 is important, while in terms of significance compared to the UK totals inventories, N₂O and CH₄ emissions from composting and NO_x emissions from soils are less important. With respect to global warming potential, an improved estimate of CH₄ emissions from on-farm 'wet patches' is extremely important and improved data on N₂O emissions from land use change are also warranted.

5. Conclusions

Robust emission inventories of atmospheric pollutants are critical to understanding and predicting impacts, identifying key sources and mitigation opportunities and are required to fulfil the reporting obligations of the UK to international conventions. While significant advances in knowledge have been made concerning the role of land management for some of these pollutants, there is still a considerable lack of knowledge for others. For emissions of NH₃, N₂O and CH₄, the processes leading to emission are largely understood and all major sources are thought to be captured in the current inventory, with the exception of N₂O emissions from land use change and CH₄ emissions from unmanaged on-farm 'wet patches'. The latter, in particular, may add considerably to the current greenhouse gas inventory for UK agriculture and more robust measurements and estimates of total source area are required. Quantification of uncertainties in the estimates for some of these sources is still required, as is an assessment of the improvement in accuracy of estimates through the development of country-specific emission factors for N₂O and CH₄ in particular. There is limited knowledge about sources and processes leading to emissions of N_{org}, and the role that these may play in local and global nitrogen budgets. Land management is known to be a source of NO_x, NMVOC and PM emissions, and potentially also HM emissions. Improved quantification is required to assess the importance of land management as a source of these pollutants in comparison with other sectors and, if appropriate, to determine the potential for mitigation. There is also a need for further consideration of the interaction between chemically active short- and longer-lived atmospheric gases and aerosols and the potential implications on climate change predictions and associated policy development.

References

- Allen, S.E., 1964. Chemical aspects of heather burning. *Journal of Applied Ecology* 1, 347-367.
- Andersen J.K., Boldrin A., Samuelsson J., Christensen T.H., Scheutz C. 2010. Quantification of Greenhouse Gas Emissions from windrow composting of garden waste. *Journal of Environmental Quality* 39, 713-724.
- Aubin, J., Papatryphon, E., Van der Werf, H.M.G., Petit, J., Morvan, Y.M., 2006. Characterisation of the environmental impact of a turbot (*Scophthalmus maximus*) re-circulating production system using Life Cycle Assessment. *Aquaculture* 261, 1259-1268.
- Beck-Friis, B., Pell, M., Sonesson, U., Jonsson, H., Kirchmann, H., 2000. Formation and emission of N₂O and CH₄ from compost heaps of organic household waste. *Environmental Monitoring and Assessment* 62, 317-331.
- Beck-Friis, B., Smars, S., Jonsson, H., Kirchmann, H., 2001. Gaseous emissions of carbon dioxide, ammonia and nitrous oxide from organic household waste in a compost reactor under different temperature regimes. *Journal of Agricultural Engineering Research* 78, 423-430.

- Beerling, D.J., Gardiner, T., Leggett, G., McLeod, A., Quick, W.P., 2008. Missing methane emissions from leaves of terrestrial plants. *Global Change Biology* 14, 1821-1826.
- Bertschi, I., Yokelson, R.J., Ward, D.E., Babbitt, R.E., Susott, R.A., Goode, J.G., Hao, W.M., 2003. Trace gas and particle emissions from fires in large diameter and belowground biomass fuels. *Journal of Geophysical Research-Atmospheres* 108, Article number 8472.
- Boeckx P., VanCleemput O. 1996. Flux estimates from soil methanogenesis and methanotrophy: Landfills, rice paddies, natural wetlands and aerobic soils. *Environmental Monitoring and Assessment* 42, 189-207.
- Boeckx P., Van Cleemput O., Meyer T. 1998. The influence of land use and pesticides on methane oxidation in some Belgian soils. *Biology and Fertility of Soils* 27, 293-298.
- Bouwman, A.F., Boumans, L.J.M., Batjes, N.H., 2002. Emissions of N₂O and NO from fertilized fields: Summary of available measurement data. *Global Biogeochemical Cycles* 16, Article number 1058.
- Butterbach-Bahl, K., Kahl, M., Mykhayliv, L., Werner, C., Kiese, R., Li, C., 2009. A European-wide inventory of soil NO emissions using the biogeochemical models DNDC/Forest-DNDC. *Atmospheric Environment* 43, 1392-1402.
- Butterbach-Bahl, K., Stange, F., Papen, H., and Li, C.S., 2001. Regional inventory of nitric oxide and nitrous oxide emissions for forest soils of southeast Germany using the biogeochemical model PnET-N-DNDC. *Journal of Geophysical Research-Atmospheres* 106, 34155-34166.
- Calvert, P., 2002. Nitrogen transformations in forest throughfall. MSc, Edinburgh University, Edinburgh.
- Cambra-Lopez, M., Aarnink, A.J.A., Zhao, Y., Calvert, S., Torres, A G., 2010. Airborne particulate matter from livestock production systems: a review of an air pollution problem. *Environmental Pollution* 158, 1-17.
- Cape, J.N., Anderson, M., Rowland, A.P., Wilson, D., 2004. Organic nitrogen in precipitation across the United Kingdom. *Water Air Soil Pollution: Focus* 4, 25-35.
- Cape, N.J., 2001. Organic nitrogen in precipitation: real problem or sampling artefact? *The Scientific World* 1, 230-237.
- Chang, C., Janzen, H.H., Cho, C.M., Nakonechny, E.M., 1998. Nitrous oxide emission through plants. *Soil Science Society of America Journal* 62, 35-38.
- Chen, D.L., Li, Y., Grace, P., Mosier, A.R., 2008. N₂O emissions from agricultural lands: a synthesis of simulation approaches. *Plant and Soil* 309, 169-189.
- Chou, C.H., Buyuksonmez, F., 2006. Biogenic emissions from green waste and comparison to the emissions resulting from composting (Part 1: Ammonia). *Compost Science & Utilization* 14, 16-22.
- Cornell, S.E., Jickells, T.D., Cape, J.N., Rowland, A.P., Duce, R.A., 2003. Organic nitrogen deposition on land and coastal environments: a review of methods and data. *Atmospheric Environment* 37, 2173-2191.
- Davidson, E.A., 1991. Fluxes of nitrous oxide and nitric oxide from terrestrial ecosystems. In: Rogers, J.E., Whitman, B.E. (Eds.), *Microbial Production and Consumption of Greenhouse Gases: Methane, Nitrogen Oxides and Halomethanes* pp. 219 – 235, American Society of Microbiology, Washington, DC.
- Davidson, E.A., Kinglerlee, W., 1997. A global inventory of nitric oxide emissions from soils. *Nutrient Cycling in Agroecosystems* 48, 37-50.
- de Gouw, J.A., Howard, C.J., Custer, T.G., Fall, R., 1999. Emissions of volatile organic compounds from cut grass and clover are enhanced during the drying process. *Geophysical Research Letters* 26, 811-814.
- Derwent, R.G., Stevenson, D.S., Doherty, R.M., Collins, W.J., Sanderson, M.G., Johnson, C.E. (2008). Radiative forcing from surface NO_x emissions: spatial and seasonal variations. *Climatic Change* 88, 385-401.
- Dore, C.J., Murrels, T.P., Passant, N.R., Hobson, M.M., Thistlethwaite, G., Wagner, A., Li, Y., Bush, T., King, K.R., Norris, J., Coleman, P.J., Walker, C., Stewart, R.A., Tsagatakis, I., Conolly, C., Brophy, N., Hann, M.R., 2008. UK Emissions of Air Pollutants 1970 to 2006. Didcot, Oxon., UK.
- Dueck, T.A., de Visser, R., Poorter, H., Persijn, S., Gorissen, A., de Visser, W., Schapendonk, A., Verhagen, J., Snel, J., Harren, F.J.M., Ngai, A.K.Y., Verstappen, F., Bouwmeester, H., Voesenek, L., van der Werf, A., 2007. No evidence for substantial aerobic methane emission by terrestrial plants: a C-13-labelling approach. *New Phytologist* 175, 29-35.
- Eklind, Y., Sundberg, C., Smars, S., Steger, K., Sundh, I., Kirchmann, H., Joensson, H., 2007. Carbon turnover and ammonia emissions during composting of biowaste at different temperatures. *Journal of Environmental Quality* 36, 1512-1520.
- Erisman, J.W., Bleeker, A., Galloway, J., Sutton, M.S., 2007. Reduced nitrogen in ecology and the environment. *Environmental Pollution* 150, 140-149.
- Erisman, J.W., Bleeker, A., Hensen, A., Vermeulen, A., 2008. Agricultural air quality in Europe and the future perspectives. *Atmospheric Environment* 42, 3209-3217.
- Fang, G.C., Chang, C.N., Chu, C.C., Wu, Y.S., Fu, P.P.C., Yang, I.L., Chen, M.H., 2003. Characterization of particulate, metallic elements of TSP, PM_{2.5} and PM_{2.5-10} aerosols at a farm sampling site in Taiwan, Taichung. *Science of The Total Environment* 308, 157-166.
- Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D.W., Haywood, J., Lean, J., Lowe, D.C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., van Dorland, R., 2007. Changes in Atmospheric Constituents and in Radiative Forcing. In: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K.B., Tignor, M., Miller, H.L. (Eds.), *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 996 pp.

- Fowler, D., Sutton, M.A., Smith, R.I., Pitcairn, C.E.R., Coyle, M., Campbell, G., Stedman, J., 1998. Regional mass budgets of oxidized and reduced nitrogen and their relative contribution to the nitrogen inputs of sensitive ecosystems. *Environmental Pollution* 102, 337-342.
- Fuglestedt, J.S., Shine, K.P., Bernsten, T., Cook, J., Lee, D.S., Stenke, A., Skeie, R.B., Velders, G.J.M., Waitz, I.A. 2010. Transport impacts on atmosphere and climate: Metrics. *Atmospheric Environment in press (available online)*.
- Gallet-Budynek A., Brzostek E., Rodgers V.L., Talbot J.M., Hyzy S., Finzi A.C., 2009. Intact amino acid uptake by northern hardwood and conifer trees. *Oecologia* 160, 129-138.
- Goode, J.G., Yokelson, R.J., Susott, R.A., Ward, D.E., 1999. Trace gas emissions from laboratory biomass fires measured by open-path Fourier transform infrared spectroscopy: Fires in grass and surface fuels. *Journal of Geophysical Research-Atmospheres* 104, 21237-21245.
- Groenestein, C.M., VanFaassen, H.G., 1996. Volatilization of ammonia, nitrous oxide and nitric oxide in deep-litter systems for fattening pigs. *Journal of Agricultural Engineering Research* 65, 269-274.
- Gross, A., Boyd, C.E., Wood, C.W., 1999. Ammonia volatilization from freshwater fish ponds. *Journal of Environmental Quality* 28, 793-797.
- Guenther, A., Hewitt, C.N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., McKay, W.A., Pierce, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J., Zimmerman, P., 1995. A global model of natural volatile organic compound emissions. *Journal of Geophysical Research-Atmospheres* 100, 8873-8892.
- Hellebrand, H.J., 1998. Emission of nitrous oxide and other trace gases during composting of grass and green waste. *Journal of Agricultural Engineering Research* 69, 365-375.
- Hobbs, P., Mottram, T., 2000. New Directions: Significant contributions of dimethyl sulphide from livestock to the atmosphere. *Atmospheric Environment* 34, 3649-3650.
- Hobbs, P.J., Webb, J., Mottram, T.T., Grant, B., Misselbrook, T.M., 2004. Emissions of volatile organic compounds originating from UK livestock agriculture. *Journal of the Science of Food and Agriculture* 84, 1414-1420.
- Horvath, L., 2004. Determination of the nitrogen compound balance between the atmosphere and a Norway spruce forest ecosystem. *Nutrient Cycling in Agroecosystems* 70, 143-146.
- Jackson, J., Choudrie, S., Thistlethwaite, G., Passant, N., Murrells, T., Watterson, J., Mobbs, D., Cardenas, L., Thomson, A., Leach, A., 2009. UK Greenhouse gas inventory, 1990 to 2007. Annual report for submission under the Framework Convention for climate Change. AEA, Didcot, Oxon, UK.
- Jarvis, A., Sundberg, C., Milenkovski, S., Pell, M., Smars, S., Lindgren, P.E., Hallin, S., 2009. Activity and composition of ammonia oxidizing bacterial communities and emission dynamics of NH₃ and N₂O in a compost reactor treating organic household waste. *Journal of Applied Microbiology* 106, 1502-1511.
- Karl M., Guenther A., Koble R., Leip A., Seufert G. 2009. A new European plant-specific emission inventory of biogenic volatile organic compounds for use in atmospheric transport models. *Biogeosciences* 6, 1059-1087.
- Keene, W.C., Montag, J.A., Maben, J.R., Southwell, M., Leonard, J., Church, T.M., Moody, J.L., Galloway, J.N., 2002. Organic nitrogen in precipitation over Eastern North America. *Atmospheric Environment* 36, 4529-4540.
- Keppler, F., Hamilton, J.T.G., Brass, M., Rockmann, T., 2006. Methane emissions from terrestrial plants under aerobic conditions. *Nature* 439, 187-191.
- Kesik, M., Ambus, P., Baritz, R., Bruggemann, N.B., Butterbach-Bahl, K., Damm, M., Duyzer, J., Horvath, L., Kiese, R., Kitzler, B., Leip, A., Li, C., Pihlatie, M., Pilegaard, K., Seufert, G., Simpson, D., Skiba, U., Smiatek, G., Vesala, T., Zechmeister-Boltenstern, S., 2005. Inventories of N₂O and NO emissions from European forest soils. *Biogeosciences* 2, 353-375.
- Kirstine, W., Galbally, I., Ye, Y.R., Hooper, M., 1998. Emissions of volatile organic compounds (primarily oxygenated species) from pasture. *Journal of Geophysical Research-Atmospheres* 103, 10605-10619.
- Kirstine, W.V., Galbally, I.E., 2004. A simple model for estimating emissions of volatile organic compounds from grass and cut grass in urban airsheds and its application to two Australian cities. *Journal of the Air & Waste Management Association* 54, 1299-1311.
- Klimont, Z., Amann, M., 2002. European control strategy for fine particles: the potential role of agriculture. In: Hinz, T., Rönnpagel, B., Linke, S., (Eds.), *Particulate matter in and from agriculture*. Braunschweig, Germany, pp. 29-35.
- Lee, D.S., Atkins, D.H.F., 1994. Atmospheric ammonia emissions from agricultural waste combustion. *Geophysical Research Letters* 21, 281-284.
- Leshem, Y.Y., Wills, R.B.H., Ku, V.V.V., 1998. Evidence for the function of the free radical gas - nitric oxide (NO center dot) - as an endogenous maturation and senescence regulating factor in higher plants. *Plant Physiology and Biochemistry* 36, 825-833.
- Li Z., Kelliher F.M. 2007. Methane oxidation in freely and poorly drained grassland soils and effects of cattle urine application. *Journal of Environmental Quality* 36, 1241-1248.
- Matthews, R.A., Chadwick, D.R., Retter, A.L., Yamulki, S., 2009. Nitrous oxide and methane emissions from small-scale farmland features in livestock systems. In: Grignani, M.A.C., Zavattaro, L., Bechini, L., Bertora, C., Marino Gallina, P., Sacco, D. (Eds.), *Proceedings of the 16th Nitrogen Workshop: Connecting different scales of nitrogen use in agriculture*. Turin, Italy, pp. 15-16.

- Milford, C., Theobald, M.R., Nemitz, E., Hargreaves, K.J., Horvath, L., Raso, J., Dammgren, U., Neftel, A., Jones, S.K., Hensen, A., Loubet, B., Cellier, P., Sutton, M.A., 2009. Ammonia fluxes in relation to cutting and fertilization of an intensively managed grassland derived from an inter-comparison of gradient measurements. *Biogeosciences* 6, 819-834.
- Misselbrook, T.H., Sutton, M.A., Scholefield, D., 2004. A simple process-based model for estimating ammonia emissions from agricultural land after fertilizer applications. *Soil Use and Management* 20, 365-372.
- Misselbrook, T.H., van der Weerden, T.J., Pain, B.F., Jarvis, S.C., Chambers, B.J., Smith, K.A., Phillips, V.R., Demmers, T.G.M., 2000. Ammonia emission factors for UK agriculture. *Atmospheric Environment* 34, 871-880.
- Misselbrook, T.H., Webb, J., Chadwick, D.R., Ellis, S., Pain, B.F., 2001. Gaseous emissions from outdoor concrete yards used by livestock. *Atmospheric Environment* 35, 5331-5338.
- Misselbrook, T.H., Webb, J., Gilhespy, S.L., 2006. Ammonia emissions from outdoor concrete yards used by livestock - quantification and mitigation. *Atmospheric Environment* 40, 6752-6763.
- Mosier A.R., Delgado J.A., Cochran V.L., Valentine D.W., Parton W.J. 1997. Impact of agriculture on soil consumption of atmospheric CH₄ and a comparison of CH₄ and N₂O flux in subarctic, temperate and tropical grasslands. *Nutrient Cycling in Agroecosystems* 49, 71-83.
- Muller, K., Pelzing, M., Gnauk, T., Kappe, A., Teichmann, U., Spindler, G., Haferkorn, S., Jahn, Y., Herrmann, H., 2002. Monoterpene emissions and carbonyl compound air concentrations during the blooming period of rape (*Brassica napus*). *Chemosphere* 49, 1247-1256.
- Natangelo M., Mangiapan S., Bagnati R., Benfenati E., Fanelli R., 1999. Increased concentrations of nitrophenols in leaves from a damaged forestal site. *Chemosphere* 38, 1495-1503
- Neff, J.C., Holland, E.A., Dentener, F.J., McDowell, W.H., Russell, K.M., 2002. The origin, composition and rates of organic nitrogen deposition: A missing piece of the nitrogen cycle? *Biogeochemistry* 57, 99-136.
- Nemitz, E., Sutton, M.A., Gut, A., San Jose, R., Husted, S., Schjoerring, J.K., 2000. Sources and sinks of ammonia within an oilseed rape canopy. *Agricultural and Forest Meteorology* 105, 385-404.
- Ngwabie, N.M., Schade, G.W., Custer, T.G., Linke, S., Hinz, T., 2008. Abundances and flux estimates of volatile organic compounds from a dairy cowshed in Germany. *Journal of Environmental Quality* 37, 565-573.
- Nicholson, F.A., Chambers, B.J., 2006. Quantifying heavy metal inputs to agricultural soils in England and Wales. In: Petersen, S.O. (Eds.), *Technology for recycling of manure and organic residues in a whole-farm perspective, Proceedings of the 12th RAMIRAN International conference*, Danish Institute of Agricultural Sciences Report No 123, pp.41-43.
- Nicholson, F.A., Chambers, B.J., Williams, J.R., Unwin, R.J., 1999. Heavy metal contents of livestock feeds and animal manures in England and Wales. *Bioresource Technology* 70, 23-31.
- Nisbet, R.E.R., Fisher, R., Nimmo, R.H., Bendall, D.S., Crill, P.M., Gallego-Sala, A.V., Hornibrook, E.R.C., Lopez-Juez, E., Lowry, D., Nisbet, P.B.R., Shuckburgh, E.F., Srisankantharajah, S., Howe, C.J., Nisbet, E.G., 2009. Emission of methane from plants. *Proceedings of the Royal Society B-Biological Sciences* 276, 1347-1354.
- Ojima D.S., Valentine D.W., Mosier A.R., Parton W.J., Schimel D.S. 1993. Effect of land-use change on methane oxidation in temperate forest and grassland soils. *Chemosphere* 26, 675-685.
- Pain, B.F., Misselbrook, T.H., Clarkson, C.R., Rees, Y.J., 1990. Odor and ammonia emissions following the spreading of anaerobically-digested pig slurry on grassland. *Biological Wastes* 34, 259-267.
- Pandis, S.N., Paulson, S.E., Seinfeld, J.H., Flagan, R.C., 1991. Aerosol formation in the photooxidation of isoprene and beta-pinene. *Atmospheric Environment Part a-General Topics* 25, 997-1008.
- Parkinson, R., Gibbs, P., Burchett, S., Misselbrook, T., 2004. Effect of turning regime and seasonal weather conditions on nitrogen and phosphorus losses during aerobic composting of cattle manure. *Bioresource Technology* 91, 171-178.
- Paul, J.W., Beauchamp, E.G., Zhang, X., 1993. Nitrous and nitric oxide emissions during nitrification and denitrification from manure-amended soil in the laboratory. *Canadian Journal of Soil Science* 73, 539-553.
- Phillips, R., Brush, S., Sneath, R., Simon, H.A., Wathes, C., 2002. Creating an inventory of agricultural PM emissions. In: Hinz, T., Ronnpagel, B., Linke, S., (Eds.), *Particulate matter in and from agriculture*. Braunschweig, Germany pp. 21-28.
- Pinto, M., Merino, P., del Prado, A., Estavillo, J.M., Yamulki, S., Gebauer, G., Piertzak, S., Lauf, J., and Oenema, O., 2004. Increased emissions of nitric oxide and nitrous oxide following tillage of a perennial pasture. *Nutrient Cycling in Agroecosystems* 70, 13-22.
- Powlson D.S., Goulding K.W.T., Willison T.W., Webster C.P., Hutsch B.W. 1997. The effect of agriculture on methane oxidation in soil. *Nutrient Cycling in Agroecosystems* 49, 59-70.
- Qaderi, M.M., Reid, D.M., 2009. Methane emissions from six crop species exposed to three components of global climate change: temperature, ultraviolet-B radiation and water stress. *Physiologia Plantarum* 137, 139-147.
- Ravishankara, A.R., Daniel, J.S. and Portmann, R.W., 2009. Nitrous Oxide (N₂O): The dominant ozone-depleting substance emitted in the 21st Century. *Science* 326,123-125.
- Razgulin, S.M., Stepanov, A.L., 2009. Ammonia emissions from soddy-podzolic soils under different phytocenoses. *Eurasian Soil Science* 42, 793-796.
- Rockel, P., Strube, F., Rockel, A., Wildt, J., and Kaiser, W.M., 2002. Regulation of nitric oxide (NO) production by plant nitrate reductase in vivo and in vitro. *Journal of Experimental Botany* 53, 103-110.

- Roller, C.B., Holland, B.P., McMillen, G., Step, D.L., Krehbiel, C.R., Namjou, K., McCann, P.J., 2007. Measurement of exhaled nitric oxide in beef cattle using tunable diode laser absorption spectroscopy. *Applied Optics* 46, 1333-1342.
- Schjoerring, J.K., Mattsson, M., 2001. Quantification of ammonia exchange between agricultural cropland and the atmosphere: Measurements over two complete growth cycles of oilseed rape, wheat, barley and pea. *Plant and Soil* 228, 105-115.
- Sebacher, D.I., Harriss, R.C., Bartlett, K.B., 1985. Methane emissions to the atmosphere through aquatic plants. *Journal of Environmental Quality* 14, 40-46.
- Seghers D., Top E.M., Reheul D., Bulcke R., Boeckx P., Verstraete W., Siciliano S.D. 2003. Long-term effects of mineral versus organic fertilizers on activity and structure of the methanotrophic community in agricultural soils. *Environmental Microbiology* 5, 867-877.
- Sharkey, T.D., Wiberley, A.E., Donohue, A.R., 2008. Isoprene emission from plants: Why and how. *Annals of Botany* 101, 5-18.
- Shaw, S.L., Mitloehner, F.M., Jackson, W., Depeters, E.J., Fadel, J.G., Robinson, P.H., Holzinger, R., Goldstein, A.H., 2007. Volatile organic compound emissions from dairy cows and their waste as measured by proton-transfer-reaction mass spectrometry. *Environmental Science & Technology* 41, 1310-1316.
- Shindell, D. T., Faluvegi, G., Koch, D.M., Schmidt, G.A., Unger, N., Bauer, S.E. 2009. Improved attribution of climate forcing to emissions. *Science* 326, 716-718.
- Shine, K. P., Berntsen, T.K., Fuglestedt, J.S., Skeie, R.B., Stuber, N. 2007. Comparing the climate effect of emissions of short- and long-lived climate agents. *Philosophical Transactions of the Royal Society a-Mathematical Physical and Engineering Sciences* 365, 1903-1914.
- Simpson D., Winiwarter W., Borjesson G., Cinderby S., Ferreira A., Guenther A., Hewitt C.N., Janson R., Khalil M.A.K., Owen S., Pierce T.E., Puxbaum H., Shearer M., Skiba U., Steinbrecher R., Tarrason L., Oquist M.G. 1999. Inventorying emissions from nature in Europe. *Journal of Geophysical Research-Atmospheres* 104, 8113-8152.
- Skiba, U., Ball, B., 2002. The effect of soil texture and soil drainage on emissions of nitric oxide and nitrous oxide. *Soil Use and Management* 18, 56-60.
- Skiba, U., Fowler, D., Smith, K.A., 1997. Nitric oxide emissions from agricultural soils in temperate and tropical climates: sources, controls and mitigation options. *Nutrient Cycling in Agroecosystems* 48, 139-153.
- Skiba, U., Smith, K.A., 2000. The control of nitrous oxide emissions from agricultural and natural soils. *Chemosphere* 2, 379-386.
- Smith, K.A., Dobbie, K.E., Ball, B.C., Bakken, L.R., Sitaula, B.K., Hansen, S., Brumme, R., Borken, W., Christensen, S., Prieme, A., Fowler, D., Macdonald, J.A., Skiba, U., Klemmedtsson, L., Kasimir-Klemmedtsson, A., Degorska, A., Orlanski, P., 2000. Oxidation of atmospheric methane in Northern European soils, comparison with other ecosystems, and uncertainties in the global terrestrial sink. *Global Change Biology* 6, 791-803.
- Sommer, S.G., 1997. Ammonia volatilization from farm tanks containing anaerobically digested animal slurry. *Atmospheric Environment* 31, 863-868.
- Sommer, S.G., Husted, S., 1995. The chemical buffer system in raw and digested animal slurry. *Journal of Agricultural Science* 124, 45-53.
- Stehfest, E., Bouwman, L., 2006. N₂O and NO emission from agricultural fields and soils under natural vegetation: summarizing available measurement data and modeling of global annual emissions. *Nutrient Cycling in Agroecosystems* 74, 207-228.
- Stohl, A., Williams, E., Wotawa, G., KrompKolb, H., 1996. A European inventory of soil nitric oxide emissions and the effect of these emissions on the photochemical formation of ozone. *Atmospheric Environment* 30, 3741-3755.
- Sun, H., Trabue, S.L., Scoggin, K., Jackson, W.A., Pan, Y., Zhao, Y., Malkina, I.L., Koziel, J.A., Mitloehner, F.M., 2008. Alcohol, volatile fatty acid, phenol, and methane emissions from dairy cows and fresh manure. *Journal of Environmental Quality* 37, 615-622.
- Sutton, M.A., Dragosits, U., Tang, Y.S., Fowler, D., 2000. Ammonia emissions from non-agricultural sources in the UK. *Atmospheric Environment* 34, 855-869.
- Sutton, M.A., Fowler, D., Moncrieff, J.B., 1993a. The exchange of atmospheric ammonia with vegetated surfaces. 1. Unfertilized vegetation. *Quarterly Journal of the Royal Meteorological Society* 119, 1023-1045.
- Sutton, M.A., Fowler, D., Moncrieff, J.B., Storetonwest, R.L., 1993b. The exchange of atmospheric ammonia with vegetated surfaces. 2. Fertilized vegetation. *Quarterly Journal of the Royal Meteorological Society* 119, 1047-1070.
- Sutton, M.A., Perthue, E., Fowler, D., StoretonWest, R.L., Cape, J.N., Arends, B.G., Mols, J.J., 1997. Vertical distribution and fluxes of ammonia at Great Dun Fell. *Atmospheric Environment* 31, 2615-2624.
- Suwanwaree P., Robertson G.P. 2005. Methane oxidation in forest, successional, and no-till agricultural ecosystems: Effects of nitrogen and soil disturbance. *Soil Science Society of America Journal* 69, 1722-1729.
- Vallejo, A., Garcia-Torres, L., Diez, J.A., Arce, A., Lopez-Fernandez, S., 2005. Comparison of N losses (NO₃⁻, N₂O, NO) from surface applied, injected or amended (DCD) pig slurry of an irrigated soil in a Mediterranean climate. *Plant and Soil* 272, 313-325.

- Walker, J.T., Geron, C.D., Vose, J.M., Swank, W.T., 2002. Nitrogen trace gas emissions from a riparian ecosystem in southern Appalachia. *Chemosphere* 49, 1389-1398.
- Whitehead, D.C., Lockyer, D.R., Raistrick, N., 1988. The volatilization of ammonia from perennial ryegrass during decomposition, drying and induced senescence. *Annals of Botany* 61, 567-571.
- Wildt, J., Kley, D., Rockel, A., Rockel, P., Segschneider, H.J., 1997. Emission of NO from several higher plant species. *Journal of Geophysical Research-Atmospheres* 102, 5919-5927.
- Willison T.W., Webster C.P., Goulding K.W.T., Powison D.S. 1995. Methane oxidation in temperate soils - effects of land-use and the chemical form of nitrogen fertilizer. *Chemosphere* 30, 539-546.
- Yamulki, S., Jarvis, S.C., 2002. Short-term effects of tillage and compaction on nitrous oxide, nitric oxide, nitrogen dioxide, methane and carbon dioxide fluxes from grassland. *Biology and Fertility of Soils* 36, 224-231.
- Zhang, Q., Anastasio, C., 2003. Conversion of fogwater and aerosol organic nitrogen to ammonium, nitrate, and NO_x during exposure to simulated sunlight and ozone. *Environmental Science & Technology* 37, 3522-3530.



References to published material

9. This section should be used to record links (hypertext links where possible) or references to other published material generated by, or relating to this project.

Publications arising from the project

Misselbrook, T.H., Cape, J.N., Cardenas, L.M., Chadwick, D.R., Dragosits, U., Hobbs, P.J., Nemitz, E., Reis, S., Skiba, U. and Sutton, M.A. Key unknowns in estimating atmospheric emissions from UK land management. Submitted to *Atmospheric Environment*

