

AIR QUALITY EXPERT GROUP

Air Pollution from Agriculture



Prepared for:

Department for Environment, Food and Rural Affairs;
Scottish Government; Welsh Government; and
Department of the Environment in Northern Ireland

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This is a report from the Air Quality Expert Group to the Department for Environment, Food and Rural Affairs; Scottish Government; Welsh Government; and Department of the Environment in Northern Ireland, on air pollution from agriculture. The information contained within this report represents a review of the understanding and evidence available at the time of writing.

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Executive Summary

This report addresses the research and policy aspects of air pollutants from agriculture. The approach is to identify the pollutants emitted by agriculture and the environmental issues to which they contribute in the UK and elsewhere, the extent of knowledge and uncertainty of the sources, fate and effects of the pollutants and available options for control. The main focus is on ammonia, a pollutant largely from agricultural activity and an important contributor to air quality issues in the UK. The report refers to the wider European and global environmental issues to which nitrogen compounds contribute in recent assessments without repeating the detail they contain (European Nitrogen Assessment (Sutton et al 2011), The Global Nitrogen cycle in the 21st century (Fowler et al 2013)).

The format of the report is structured around a series of policy questions. It begins with a summary of the key points of the answers to these questions.

1.1 Q1. What pollutants are emitted by agriculture?

A: Nitrogen-containing compounds (NO_2 , NO , NH_3 , N_2O) are emitted to the atmosphere from agricultural activities. In the case of ammonia (NH_3) and nitrous oxide (N_2O), agricultural sources are the main contributors, comprising 88% and 68% respectively of annual UK emissions in 2016. In addition, agricultural soils are becoming a significant source of nitric oxide (NO) (projected to be 6% of UK NO_x emissions by 2030) as emissions from combustion sources are reduced by control measures. Methane and non-methane volatile organic compounds (VOC) are emitted by agriculture, and livestock are an important source of methane in the UK budget (51% in 2016). The pesticides/fungicides hexachlorobenzene, hexachlorocyclohexane and pentachlorophenol, which are listed in the Stockholm Convention on Persistent Organic Pollutants, are also emitted from agricultural and forestry use.

1.2 Q2. What are the impacts of ammonia upon human health and the environment?

A: The main impacts of ammonia arise through its contribution to (1) formation of particulate matter (PM) and the consequent effects on human mortality and morbidity throughout the UK, and (2) the eutrophication of the semi-natural landscape of the UK leading to marked reductions in plant biodiversity. Ammonium in particle form (NH_4^+) is a transboundary pollutant, exchanged between European countries. Therefore, UK ammonia emissions contribute to human health effects and biodiversity changes in the UK and elsewhere in Europe, while the UK is impacted by emissions from elsewhere in Europe.

1.3 Q3. How can these impacts best be addressed / targeted?

A: The most effective measures to reduce these effects are those which reduce emissions of ammonia to the atmosphere. A range of methods have been developed and field-tested at the local and country scale, by which it is possible to reduce emissions of ammonia by at least 50%. Mitigation strategies also include separation of sensitive receptors from local sources and the use of shelterbelts to enhance both dispersion through increasing turbulence and capture of ammonia close to source.

1.4 Q4: How have concentrations and the deposition of NH_3 and NH_4^+ in the UK changed following changes in emissions.

A: A 6.4% reduction in ammonia emissions has been reported in the NAEI between 2000 and 2016, a value that is smaller than the reported uncertainty in emissions ($\sim \pm 20\%$). Over

the same period concentrations of ammonia over the UK as a whole have not declined significantly, although there have been reductions in some areas. A substantial reduction in particulate NH_4^+ concentration (~40%) has been observed in the UK over the period 1999 to 2014, which is mainly due to the reduction in sulphur emissions and resulting changes in particulate matter composition.

The dry and wet deposition of ammonia and ammonium at the UK scale have not changed significantly over this period.

1.5 Q 5: How effective have controls on NH_3 elsewhere in Europe been in reducing emissions and concentrations of NH_3 and particulate matter?

A: *Control measures for ammonia from agriculture in the Netherlands have resulted in a reduction in emissions of 64% between 1990 and 2016. The reduction in ammonia emissions contributed to reductions in particulate NH_4^+ concentrations of 68% over this period, but a much smaller reduction in ambient ammonia concentration, which was not statistically significant. These different trends are consistent with a change in partitioning of gas and aerosol phase NH_x , which is largely a consequence of substantial reductions in sulphur emissions over the same time period, such that a greater fraction of NH_x is now present as gaseous ammonia. The changes in the Netherlands are further complicated by changes in emissions of NH_3 , SO_2 and NO_2 in neighbouring countries.*

1.6 Q6. Have changes in UK emissions of NO_x and NH_3 reduced the exceedances of critical loads for nutrient nitrogen? (and are both the oxidised and reduced forms of N equally important for eutrophication?).

A: *Reductions in oxidised nitrogen deposition of approx. 30% have been observed in the UK over the last decade. This has reduced total N deposition by 15% and resulted in 15% reductions in exceedance of critical loads for nutrient nitrogen. However, widespread exceedance of critical loads for eutrophication in the UK remains with ammonia and ammonium deposition being the main contributors.*

One of the main effects of nitrogen deposition is a reduction in species richness in UK vegetation. The effects have been shown experimentally and in surveys to be associated with the total annual deposition of reactive nitrogen, but the deposition of gas phase ammonia has

been shown to be more damaging to moorland and bog plant communities per unit nitrogen deposited.

1.7 Q7. What are the relative air quality benefits and costs for controls on ammonia emissions?

A: *The damage costs of ammonia over Europe have been estimated in the European Nitrogen Assessment to be in the range €2 to €20 per kg N pollution for human health and €2 to €10 per kg N pollution for biodiversity related societal costs. Combined, these provide estimated damage costs in the region of €4 to €30 per kg N pollution. These costs represent the potential benefits of control measures, and in the case of the Maximum Technically Feasible Reduction (MTFR) average estimated abatement costs are €2.4 per kg of N pollution abated.*

2 State of the Evidence

2.1 Q1. What pollutants are emitted from agricultural activities?

A range of pollutant gases, and especially nitrogen compounds (NO_2 , NO , NH_3 , N_2O) are emitted to the atmosphere from agricultural activities, including fertilizer use, farm machinery and livestock waste (Sutton et al. 2017). In addition, primary particulate matter is emitted from livestock housing and tilling activities. However, the main contribution to air pollution from agriculture in the UK is due to emissions of ammonia (NH_3). The majority of agricultural NH_3 emissions are from livestock farming, with cattle providing 49% of the annual total UK agricultural emissions (244 kT- NH_3 in 2016), and smaller contributions from other livestock and arable agriculture, as detailed in the National Atmospheric Emission Inventory (NAEI 2016, Wakeling et al. 2018) and summarised in Figure 1. Non-agricultural UK NH_3 emissions, from vehicles, human waste and industry contributes 12% of total annual emissions in 2016. It has been estimated that 80% of NH_3 emissions in Europe are from just 5% of farms (EEA, 2016). For the UK, it is still the case that a large proportion of NH_3 emissions are from a smaller fraction of farms, but it is closer to 60% of emissions from 21% of dairy farms, because the UK does not have the large number of very small farms that dominates the European picture.

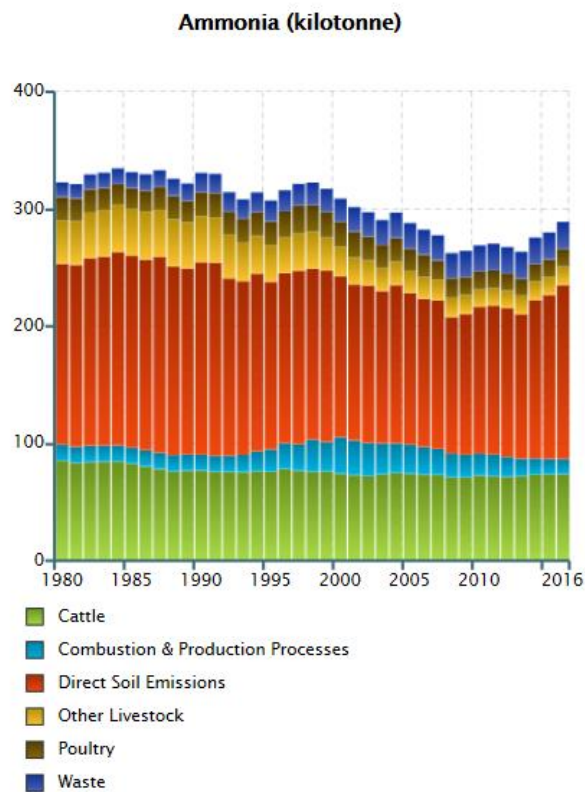


Figure 1. UK NH₃ emissions from all sectors in 2016 (NAEI, 2018)

Following emission of gaseous NH₃ to the atmosphere, NH₃ is either deposited back onto terrestrial surfaces by dry deposition or is converted to ammonium (NH₄⁺) in particulate matter through reaction with atmospheric acids, and has a short lifetime in the atmosphere of just a few hours (Fig. 2). The particulate matter has a longer lifetime in the atmosphere of a few days on average and is removed from the atmosphere primarily by rain and snow as wet deposition.

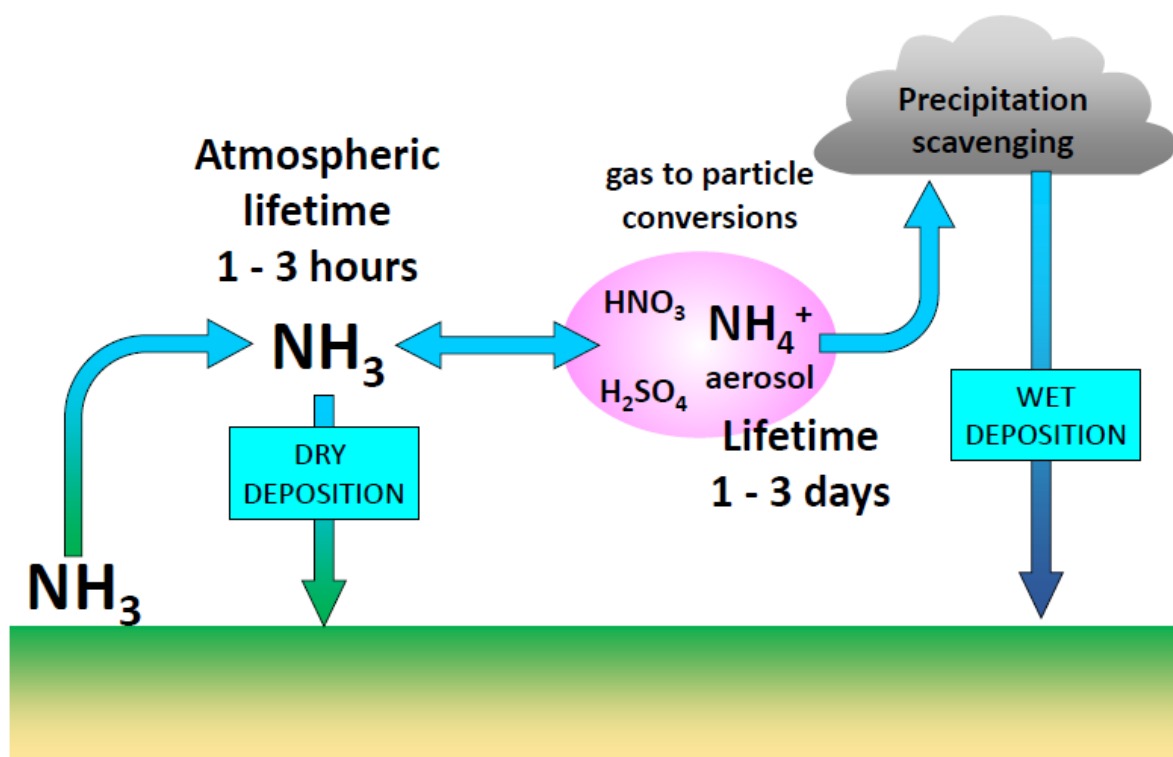


Figure 2. NH_3 release to the atmosphere, transformation to particulate matter and removal by dry and wet deposition

Before the turn of the 21st century, the main acid in the UK (and European) atmosphere was sulphuric acid (H_2SO_4), from emissions of SO_2 . The particulate matter produced through reaction with NH_3 was largely ammonium sulphate ($(\text{NH}_4)_2\text{SO}_4$), which was the form in which most of the long-range transport of sulphur and acidity throughout the countries of Europe took place. Now that European emissions of SO_2 have declined by nearly 90% from their peak in the 1970s, the atmospheric acid forming particulate matter with NH_3 is mainly nitric acid (HNO_3) and the resulting compound is ammonium nitrate (NH_4NO_3). The contribution of NH_4NO_3 to particulate matter in the UK atmosphere varies with time between a few percent and tens of percent in episode conditions, which often occur in spring (Twigg et al., 2015; Young et al., 2015).

Some of the nitrogen used in agriculture is chemically transformed in soils to nitric oxide (NO) which is emitted to the atmosphere and subsequently oxidized to NO_2 and HNO_3 . While NH_3 is the priority agricultural air pollutant, many studies have shown the relevance of nitric oxide (NO) emissions from agricultural soils (e.g. Skiba and Ball 2002). The contribution of NO emissions from agricultural soils has hitherto not been a major focus due to the dominance of vehicles and power generation and other industry as NO sources in the UK. However, as the

combustion sources are projected to decrease the agricultural share of UK total NO_x emissions through soil NO emissions is expected to increase (currently estimated at 4% and projected to increase to 6% by 2030 (Sutton et al., 2017)). A recent study suggest that soil NO emissions may be larger than previously thought, being estimated to account for 20-32% of NO_x emissions in California (Almaraz et al. 2018). Recognising uncertainties in these estimates, it has been decided that soil NO_x emissions from agriculture are to be excluded when assessing compliance with the emissions reduction commitments set out in the revised Gothenburg Protocol (2012) and the revised National Emissions Ceilings Directive (2016)¹. However, when reporting national emissions to demonstrate on-going compliance with emissions ceilings in the original Gothenburg Protocol and the National Emissions Ceilings Directive, the UK will need to include soil NO_x emissions from agriculture. As there is currently no strategy to reduce soil NO emissions it is anticipated that these will not decrease as fast as combustion source emissions, and are expected to contribute a significant share in future decades. The uncertainty associated with estimating NO emissions from soils is particularly high, and further work is required to improve the reliability and sophistication of methodologies for estimating emissions. This will allow UNECE countries to consider the potential for their mitigation in future. It should be noted that the link between soil NH₃ and soil NO emissions provides an opportunity for significant synergy as methods to improve nitrogen use efficiency in agriculture should have the potential to reduce NH₃ and NO emissions simultaneously.

Soil nitrogen compounds are also reduced to nitrous oxide (N₂O), a powerful greenhouse gas. In this regard, the use of nitrogen fertilizers and livestock wastes in agriculture is the main contributor to UK N₂O emissions, accounting for 80% of N₂O emissions in the UK national atmospheric emissions inventory. Thus a wider role of agriculture in air pollution generally, and in greenhouse gas emissions and climate change, also needs to be considered (Pilegaard, 2013; Skiba et al., 2014). To achieve significant emission reductions of UK N₂O, mitigation action in the agricultural sector would be required.

2.1.1 Other gaseous emissions from agriculture

2.1.1.1 Methane and Non-methane volatile organic compounds (NMVOC)

Methane is emitted from agriculture, mainly from ruminants through enteric fermentation contributing 51% of UK methane emissions in 2016. NMVOC emissions from manure management are particularly uncertain, and as a result have only recently been included in national emissions inventories. The speciation of emissions from manure management is very

¹ This is the commonly used name for referring to the Directive (EU) 2016/2284 on the reduction of national emissions of certain atmospheric pollutants..

different to emissions from e.g. combustion or solvent use. So whilst emissions from agriculture may account for a large fraction of the national total in mass terms, it does not necessarily follow that agricultural sources of NMVOC have a significant impact on ozone formation. At present little is known on whether NMVOC emissions from livestock manures have significant ozone forming potential

2.1.1.2 Pesticides

Hexachlorobenzene (HCB): Pesticides that contain HCB as an impurity are imported into the UK, and their use contributes approximately a third of the UK's total emissions of HCB to air. HCB is listed in the Stockholm Convention on Persistent Organic Pollutants and the CLRTAP requires the reporting of HCB emissions.

The UK emissions inventory programme also estimates emissions of both hexachlorocyclohexane (HCH) and pentachlorophenol (PCP), both also listed in the Stockholm Convention on Persistent Organic Pollutants. HCH has been used in the past as an insecticide, fungicide and wood preservative; PCP has been used in the past as a wood preservative. Emissions of both are declining as previously treated wood is gradually removed from use with time.

2.2 Q2. What are the impacts of ammonia upon human health and the environment?

There are two main effects of NH_3 emissions. First, through the contribution of NH_3 to the formation of secondary inorganic particulate matter in the atmosphere (secondary inorganic aerosol (SIA)), i.e. ammonium nitrate and ammonium sulphate, the NH_3 emissions are indirectly responsible for effects on human health, increasing mortality and morbidity throughout the UK, and, following long-range transport, also in neighbouring countries of continental Europe. The health effects of particulate matter have been reviewed by COMEAP (COMEAP 2009, 2010).

Secondary inorganic aerosol is a major contributor to PM_{10} or $\text{PM}_{2.5}$ in the UK, estimated at ~40% of total $\text{PM}_{2.5}$ (AQEG 2012). The main contributions to secondary inorganic aerosol within the UK are NO_3^- , SO_4^{2-} and NH_4^+ , with NO_3^- as the dominant contributor by mass (AQEG, 2012). The contribution of NH_4^+ within SIA varies in time and space but is generally between a few percent and 20% (Twigg et al., 2015). Part of the variability arises due to seasonal variations in emissions due to fertilizer and manure spreading, but the effects of weather are also very important as temperature strongly influences the partitioning between the gas and condensed (aerosol) phase (Ellis et al 2011). Some of the largest contributions of NH_3 to PM

occur in spring, when emissions are high and temperatures are cool, (in warm summer conditions ammonium nitrate sublimates to its component gases, HNO_3 and NH_3). In Paris during an Air Quality episode in spring 2014, 62% of $\text{PM}_{2.5}$ was estimated to originate from NH_3 (CNRS, 2005).

It has been estimated that a global halving of agricultural emissions could reduce the mortality attributed to $\text{PM}_{2.5}$ by ~250,000 globally and by 52,000 across Europe (Pozzer et al., 2017).

The other main environmental effect of NH_3 emissions is its contribution to eutrophication due to nitrogen deposition on the semi-natural landscape. Nitrogen deposition has been estimated to be in excess of critical loads for eutrophication over 63% of sensitive habitats of the UK. The main effect of excessive nitrogen deposition is a reduction in plant biodiversity, which has been demonstrated at a range of sites throughout the UK (Hall et al., 2017, Stevens et al., 2010). While the effect of reactive nitrogen deposition on the species composition of semi-natural plant communities is clear, it is important to appreciate the different forms of nitrogen deposited, not all of which have the same effects on vegetation (Shepherd et al., 2014). The different forms include dry deposition of NO_2 , NH_3 , and HNO_3 as well as wet deposition of NO_3 and NH_4^+ . For some widespread ombrotrophic (rain-fed) ecosystems in upland UK, dry deposited NH_3 has been shown to be much more damaging per unit N deposited than wet deposited NO_3^- and NH_4^+ (Shepherd et al., 2011; APIS, 2017). Reductions in emissions of NH_3 are therefore considered one of the most effective measures to reduce effects on biodiversity.

As the NH_3 deposited to the landscape also contributes to eutrophication of freshwaters and marine ecosystems, it is recognised that a range of co-benefits of control measures to reduce emissions exist (Sutton et al., 2011).

Finally, it is also of note is that ammonia is an odorous gas, so odour reduction is another benefit of control measures.

2.3 Q3. How can these impacts best be addressed / targeted?

Model simulations have shown that reductions in NH_3 emissions are effective in reducing concentrations of particulate matter (Megaritis et al., 2013; Vieno et al., 2016a; Pozzer et al., 2017). Experience from other European countries shows that it is possible to achieve substantial reductions in NH_3 emissions. The countries with the most ambitious NH_3 policies are the Netherlands and Denmark, both of which are estimated to have reduced their national NH_3 emissions by 64% and 40% respectively between 1990 and 2016 (Bleeker et al., 2009, Netherlands 2018, Denmark 2018). In the case of the Netherlands, a reduction of agricultural

emissions has been claimed of at least 60%, though an independent review concluded that the available data presented was more consistent with a reduction of around 50% (40-60%) (Sutton et al., 2015).

In both the examples of the Netherlands and Denmark, the reductions in NH₃ emissions were achieved through strict programmes of national regulations that focused primarily on manure management, including land application of manure, manure storage and animal housing. Technically, all the measures were achievable by farmers and in the Netherlands have now been in routine use for over 20 years.

Considering the country case studies together, the main measures adopted have been:

- *Prohibiting the free surface spreading of liquid manure* (i.e. prohibiting use of the “splash plate spreader”). In both countries a high ambition approach was taken requiring the use of shallow injection of liquid manure, and where surface “band-spreading” was also considered insufficient, apart from exceptions:
 - A trailing shoe band-spreading approach was specifically designed and accepted in the Netherlands for use on grass swards over vulnerable peat soils, so as not to break the coherency of the root mat.
 - A relaxation to permit the use of bandspreading methods (trailing hose or trailing shoe) has been approved in Denmark where the manure is acidified immediately prior to application (a proprietary system is in use on many farms who wanted to avoid manure injection).
- *Requirement to use covered manure stores.* A wide range of covered manure storage approaches have appeared, from tanks, to covered lagoons to slurry bag approaches. The costs of using covered manure stores are to some extent compensated by the higher nitrogen content, consistency and lower water content of the stored manure product.
- *Requirement to use low emission housing.* A wide range of animal housing systems are in place in the Netherlands and Denmark. In particular, pig and poultry buildings with controlled ventilation are used both to optimize indoor climate for the animals and to allow cleaning of the exhaust air, using either chemical or biological methods. In the Netherlands, it is reported that Smart Metering is increasingly being installed to allow central administrative monitoring of the air purification systems.

Overall, it appears that the largest gains in reducing emissions, with the lowest cost to farmers have been associated firstly with low-emission manure spreading, and secondly with low-emission manure storage. In the case of large scale animal production (> 40,000 poultry, 2,000 pigs), the Industrial Emissions Directives (IED) of the European Union apply to regulate emissions. Thus local emission control is already applied at some sites in the UK as part of permitting near designated conservation areas or as planning conditions for odour control; in these cases keeping manure/litter dry until it is incorporated in the soil is the key to reducing emissions. The largest livestock sector of NH₃ emissions in the UK and Europe is cattle. Here, there are major opportunities for cost effective NH₃ emission reduction from manure storage and land spreading. Currently, the naturally ventilated nature of most cattle housing in the UK can make it harder to control emissions from this source. However, there are opportunities to reduce emissions by restricting the area of soiled surfaces and cleaning by scraping and washing down. There is the need for research to improve housing design for cattle that simultaneously reduces NH₃ emissions, while considering opportunities for animal welfare, energy and water savings.

One of the most cost effective ways to reduce part of the NH₃ emissions is through low emission mineral fertilizers. In the UK, most nitrogen is applied as ammonium nitrate (NH₄NO₃) which is associated with low NH₃ emissions, while emissions from urea are much higher. While urea is typically cheaper to purchase, around 5 to 40% of the N applied may be lost as NH₃ emissions. This loss can be minimized, for example by incorporating urea into the soil (as widely practiced in Canada), by using chemicals that slow the break-down of urea ('urease inhibitors') or by using NH₄NO₃ (Bittman et al., 2014). To date, it appears that strict regulatory requirements in relation to the use of urease inhibitors for fertilizer application to reduce these losses have not been adopted in either the Netherlands or Denmark. However, under the Revised German Fertilizer Regulations, from 1 February 2020, urea as fertilizer "*may only be applied if it has been given a urease inhibitor or is incorporated immediately, but no later than four hours after application*" (Bundesrat 2017, Section 6 (2)).

At present the extent of Europe-wide adoption of NH₃ mitigation methods can be well illustrated by Annex IX of the Gothenburg Protocol (1999). The only firm mandatory requirement was to establish a National Ammonia Code of good agricultural practice (NAC). Recent review under the LRTAP Convention showed that less than 10 parties out of 25 ratified signatories had so far established such a code (UNECE, 2017). The Annex IX requirements for manure and fertilizer application while formulated as mandatory, either do not go beyond

existing EU legislation (for pig and poultry housing) or are each associated with caveats that soften any hard requirement.

2.3.1 Pollution Swapping

Because there are risks of pollution swapping (i.e. increasing emissions of one pollutant as a consequence of controlling another pollutant), the challenge is to look at strategies that lead to overall pollution reduction (pollution synergies). A core strategy here is improving nitrogen use efficiency across the agricultural chain of nitrogen flows. This approach allows a larger fraction of input nitrogen compounds (e.g. in fertilizers and feeds) to reach products with a smaller fraction becoming losses to the environment. In this way, multiple forms of nitrogen pollution to air and water may be reduced simultaneously to improving agricultural productivity.

A key principle to achieve pollution reduction synergies is delivered when increases in efficiency are accompanied by reduced fertilizer inputs or increased production outputs. This provides opportunities for increased profits to farmers.

An example which is often discussed is the potential for pollution swapping when applying liquid manure using surface band-spreading or injection methods as compared with the traditional broadcast surface spreading. The UNECE estimates that these methods typically reduce NH_3 emissions by around 70% (injection), 30% to 60% (trailing shoe) or 30% (trailing hose) (Bittman et al., 2014). These reductions in emissions provide the opportunity to increase nitrogen use efficiency, which is further helped by the more precise and even application of the manure fertilizer resource in these methods. Such an NH_3 reduction leads to the risk that the saving is exchanged for an increase in nitrification and denitrification emissions as nitric oxide (NO), nitrous oxide (N_2O), or as nitrate and other leaching losses to water. To avoid such trade-offs and to fully realise the benefits of the lower NH_3 emissions it is necessary to match the manure inputs to the crop demands. This requires appropriate timing of manure application to match crop needs, combined with either an accompanying reduction in fertilizer inputs or strategies to increase the harvest outputs. In this way, low NH_3 emission manure spreading can become part of an overall strategy to improved nitrogen management, which improves efficiency, and either reduces costs or increases harvest revenue.

2.3.2 Mitigating emissions using shelter belts

Ammonia emissions from small area sources such as livestock buildings rapidly disperse into the surrounding lower atmosphere and are subject to deposition and gas to particle conversion. Deposition rates onto vegetation close to the source can be enhanced using shelter-belts of trees around the source. The fraction of ammonia recaptured varies with

characteristics of the source, meteorological and surface conditions. Studies by Fowler et al. (1998) showed that between 3% and 8% of NH_3 emissions from a livestock building were deposited in woodland within 300 m of the source. Further work by Bealey et al. (2016) focussed on a range of planting strategies showed that woodlands designed to recapture NH_3 from a range of livestock sources could recapture a substantially greater fraction of NH_3 emissions, in the range 20% to 40%, the latter representing housing of poultry under a woodland canopy. Rough landscape features such as shelter-belts can also help disperse ammonia, reducing concentrations near sources and therefore decreasing the most extreme impacts on nearby sensitive receptor ecosystems. Dragosits et al. (2006) contrasted the opportunities of two strategies: With a source-oriented landscape approach, trees planted around farms both capture and help disperse ammonia. With a receptor-oriented landscape approach, trees and other buffer zones around nature areas contribute to reducing ammonia concentrations on the most sensitive locations. Both approaches were found to contribute to the effectiveness of the mitigation measure, depending on amount of trees, distance between source and receptor and on the regional baseline ammonia concentration. A useful guide to near field dispersion (and measurement/model comparison) from agricultural sources is also provided by ADMLC (2016).

2.4 Q4: How have concentrations and the deposition of NH_3 and NH_4^+ in the UK changed following changes in emissions.

The UK emission inventory for NH_3 (NAEI, 2018) of 289 kt in 2016 is smaller than the peak emissions in 1998 of 322.5 kt shown in Figure 1. Annual emissions of NH_3 were relatively constant between 1990 and 1997. The reduction of 33.5 kt between 1998 and 2016 represents a 10 % reduction in emissions over the last 18 years. However, emission inventories for NH_3 are highly uncertain due to the complexity and very large range of sources and agricultural practices that give rise to NH_3 emissions (Wakeling et al. 2018). Overall uncertainty in total NH_3 emissions in the UK has been estimated at $\pm 20\%$. Close links between emissions and environmental conditions which are not fully incorporated in inventory methodology also introduce substantial uncertainty and the emission from the same activity can differ significantly between years, an effect not captured by the current emission inventory methodology (Sutton et al., 2013). The uncertainty in emissions is therefore of a greater magnitude than the reduction in emissions reported in the UK National Atmospheric Emissions Inventory (NAEI, 2016).

2.4.1 Reduced nitrogen in gas, liquid and particulate phases

The atmospheric behaviour of gaseous NH_3 illustrated in Figure 2 identifies the chemical components which need to be monitored to quantify changes in emissions and concentrations of reduced-N species over the UK. Measurements of NH_3 cannot be analysed and interpreted in isolation, and its interaction with NH_4^+ in the aerosol phase needs to be considered. Any sulphuric acid (H_2SO_4) formed from the oxidation of SO_2 reacts with any available gaseous NH_3 to form $(\text{NH}_4)_2\text{SO}_4$ which will stay in the aerosol form until it is deposited, mainly through wet deposition. Any remaining NH_3 will, together with HNO_3 , form an equilibrium with NH_4NO_3 aerosol, which can be deposited but also re-evaporate into NH_3 and HNO_3 if gas-phase concentrations drop or temperatures increase. Thus, the overall rate of NH_3 conversion to NH_4^+ , but also the type of NH_4^+ aerosol formed, dictate the gas/aerosol partitioning and the atmospheric lifetime and transport distance of the total reduced nitrogen ($\text{NH}_x = \text{NH}_3 + \text{NH}_4^+$).

NH_3 in the gas phase requires a large number of monitoring sites to identify the spatial variability and to be operated with consistent methodology for many years to allow the inter-year effects of meteorology to be quantified. The short atmospheric lifetime is illustrated by the large geometric relative standard deviation of measured concentrations, typically a factor of 3 at a rural location based on half-hourly measurements (Flechard and Fowler, 1997). Concentrations of secondary inorganic aerosol in the countryside show much smaller temporal and spatial variability than gas phase NH_3 (RoTAP, 2012; Tang et al., 2018), and a network to quantify the spatial and temporal trends in aerosol and rainfall composition requires fewer sites than NH_3 (RoTAP, 2012).

2.4.2 Ammonia in the gas phase

The UK national ammonia monitoring network (NAMN) has ~80 sites providing monthly data, consisting of 49 sites with passive (alpha) samplers (Tang et al, 2018) and 59 sites with active denuder samplers, and two sites (the super-sites) at which NH_3 is continuously measured at hourly resolution using an automated wet chemistry technique based on denuder sampling of gas phase NH_3 (Twigg et al., 2013). The different techniques are co-located at a range of sites to allow the methods to be compared.

The annual mean NH_3 concentration field is heterogeneous, with several areas of large concentration (>4 ppb) in East Yorkshire, Norfolk, the Welsh-English border counties and in SW England (Figure 3). In upland areas throughout the UK NH_3 concentrations average < 1 ppb.

The longest running sites were established in 1996-7. The long-term trend in measured NH_3 averaged across all sites shows a small downward trend (Figure 4), but which is not statistically significant at the country scale using a Mann-Kendall (MK) test (Tang et al., 2018). In areas in which emissions of NH_3 are dominated by pig and poultry farming, a statistically significant 22% reduction (MK) in annual NH_3 concentrations has been observed since 1997. By contrast, in areas with NH_3 emissions dominated by cattle there was a small, non-statistically significant increase in concentrations, despite a decrease in emissions from this sector as estimated by the NAEI (Tang et al., 2018).

Overall, therefore, the NH_3 monitoring data show only very small changes in NH_3 concentrations over the period 1999 to 2015 and do not show the 16% decline reported in the NAEI emissions.

2.4.3 Particulate NH_4^+ concentrations in the air

Concentrations of particulate NH_4^+ have been monitored since 1999. This provides a 15 year period to examine spatial and temporal trends. The annual mean spatial concentration field is similar to that for concentrations in precipitation with a general NW to SE increase in concentration from the smallest values in NW Scotland (Figure 3).

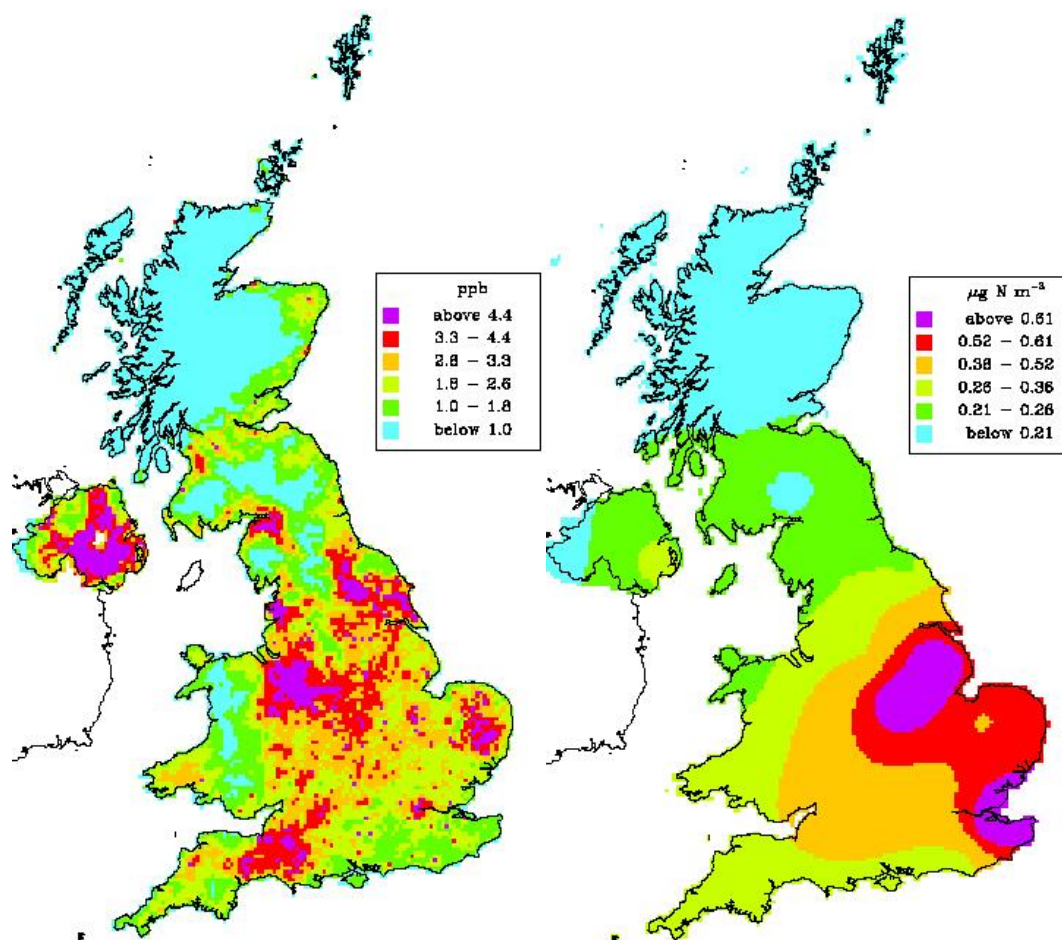


Figure 3. Annual mean concentrations of gaseous NH_3 (left), using the FRAME model, calibrated with the UK ammonia network measurements and particulate NH_4^+ (right) interpolated from the UK measurement network, for in 2015.

Over the 15 years, particulate NH_4^+ concentrations averaged over all measurement sites declined by 44% (Fig. 4), along with declines in particulate SO_4^{2-} and NO_3^- (Tang et al., 2018). The longer-term declines in SO_4^{2-} are much larger, as reflected in trends of non-marine SO_4^{2-} in precipitation from 1986 onwards. The declines in particulate SO_4^{2-} have been important in modifying the atmospheric behaviour and lifetime of NH_3 , in part because NH_4NO_3 exists in equilibrium with gaseous NH_3 and HNO_3 , which therefore sublimates as the HNO_3 and NH_3 are dry deposited at the surface, but also because an important sink for NH_3 (its reaction with H_2SO_4) has been removed from the atmosphere. By contrast, when the NH_3 was sequestered by SO_4^{2-} , the NH_4^+ was retained within particulate matter by the SO_4^{2-} throughout its atmospheric lifetime.

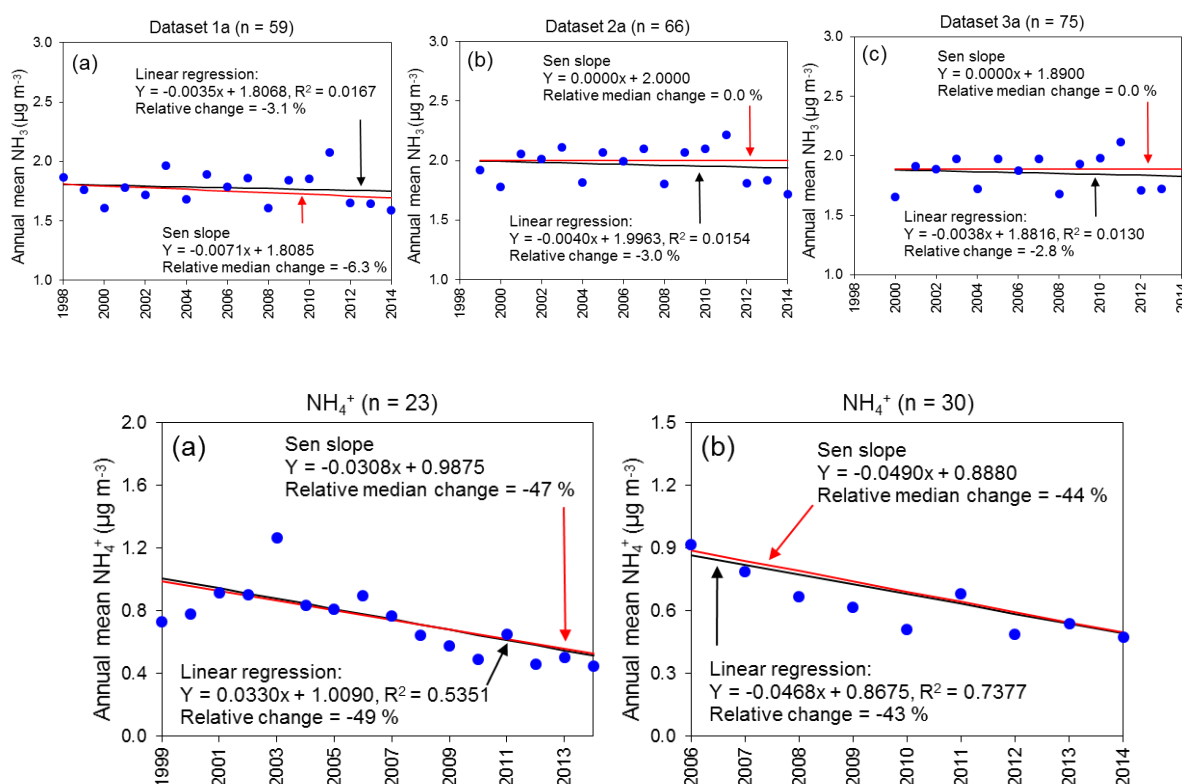


Figure 4. Upper panel: Long-term trends in annual NH_3 concentrations over the UK for (a) the 59 sites operational 1998 to 2013, (b) the 66 sites operational 1999 to 2014 and (c) the 75 sites operational 2000-2014 (Tang et al., 2018). Lower panel: Long term trends in annual particulate NH_4^+ concentrations between 1999 and 2014 over the UK for (a) the original 23 site network and (b) since the network was expanded to 30 sites in 2006 (from Tang et al., 2018, Supplement). The high value of NH_4^+ for 2003 can be explained by long-range transport associated with particular meteorological conditions, as explored in detail by Vieno et al. (2016b).

2.4.4 NH_4^+ in rain

The concentrations of NH_4^+ in rain, originating from the washout of both NH_3 and NH_4^+ , show W-E and N-S gradients of increasing concentration with the largest concentrations in the East and South of the UK and a region of large concentrations between North Yorkshire and Kent, centred on Lincolnshire (Fig. 5). In these areas, the annual mean concentration exceeds those on the west coast by approximately a factor of three. Across all sites there is a small (statistically non-significant) reduction in wet deposition of NH_4^+ , but with considerable spatial variability over the period 1986 to 2015 (some individual sites report statistically significant trends and some do not). For comparison with the measurements of NH_3 , which begin in the

late 1990s, there is no significant trend in wet NH_4^+ deposition over the 1999-2015 period, and the inter-annual variability conceals any trend in the data at the UK scale.

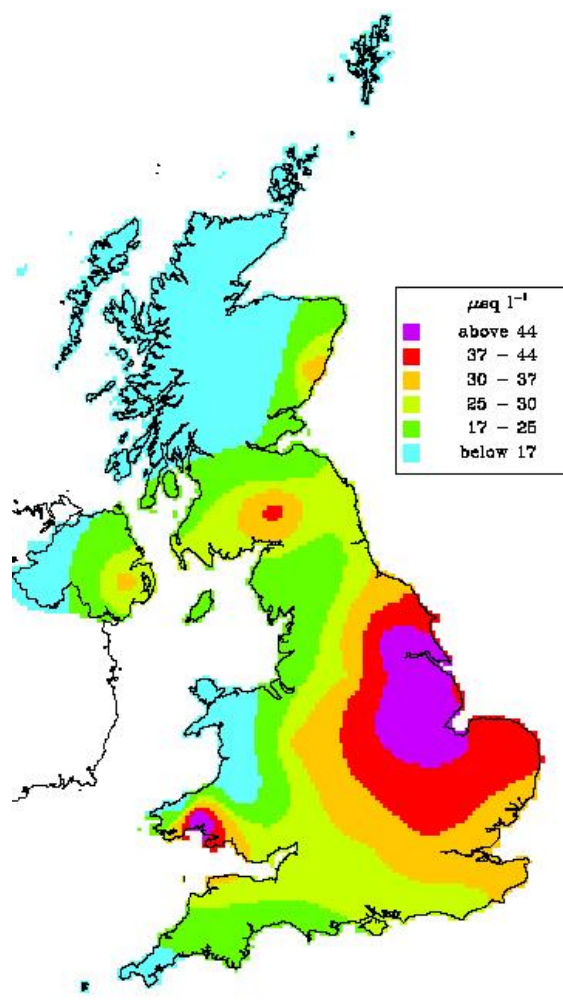


Figure 5. Annual precipitation weighted mean concentrations of NH_4^+ in rain over the UK in 2015. The map is interpolated from measurements of UK precipitation chemistry as described in RoTAP 2012.

2.4.5 Interpretation

At first glance, it appears inconsistent that, firstly, NH_3 emissions are thought to have declined, whilst NH_3 concentrations have remained more or less constant, and, secondly, that NH_4^+ concentrations have declined whilst NH_4^+ wet deposition has shown variability but not a clear trend. One reason is that the effective lifetime (and transport distance) of NH_3 has increased with decreasing SO_2 emissions because less is tied up as non-volatile $(\text{NH}_4)_2\text{SO}_4$ and more forms the dynamic equilibrium with volatile NH_4NO_3 . This is consistent with observations of NH_3 concentration increases in remote areas and those dominated by sheep emissions, suggesting that more of the emitted NH_3 remains in the gas phase as the acidic aerosol sinks

(SO_4^{2-} , and NO_3^-) have declined. In addition, NH_3 deposition rates are thought to have decreased due to leaf surfaces having become increasingly alkaline (Fowler et al., 2009). Both these effects appear to have balanced the reported decrease in emission, and it follows that in the absence of a decrease in NH_3 emission, NH_3 concentrations would have gone up over the same period due to the reduction in SO_2 and NO_x emissions.

The NH_4^+ concentrations have decreased over this period because the emissions of all precursor gases for the NH_4^+ compounds have decreased, less NH_3 is (irreversibly) converted to particulate NH_4^+ and because the lifetime of NH_4^+ has decreased due to the shift from sulphate to nitrate. In addition, as a longer-lived compound particulate NH_4^+ strongly reflects emission changes elsewhere in Europe. From measurements alone it is impossible to attribute how much of the NH_4^+ reduction is attributable to the reduction in NH_3 emission and how much to the reduction in SO_2 and NO_x emission. The change of the dominant aerosol form from $(\text{NH}_4)_2\text{SO}_4$ to NH_4NO_3 , also reduces the average atmospheric lifetime of reduced N overall and consequently the long-range transport out of the country.

It is unclear at present, why NH_4^+ wet deposition has not decreased in line with NH_4^+ concentrations, but the observed trends in NH_3 and NH_4^+ concentrations, and in NH_x wet deposition can successfully be reproduced with an atmospheric chemistry transport model (Fig. 6).

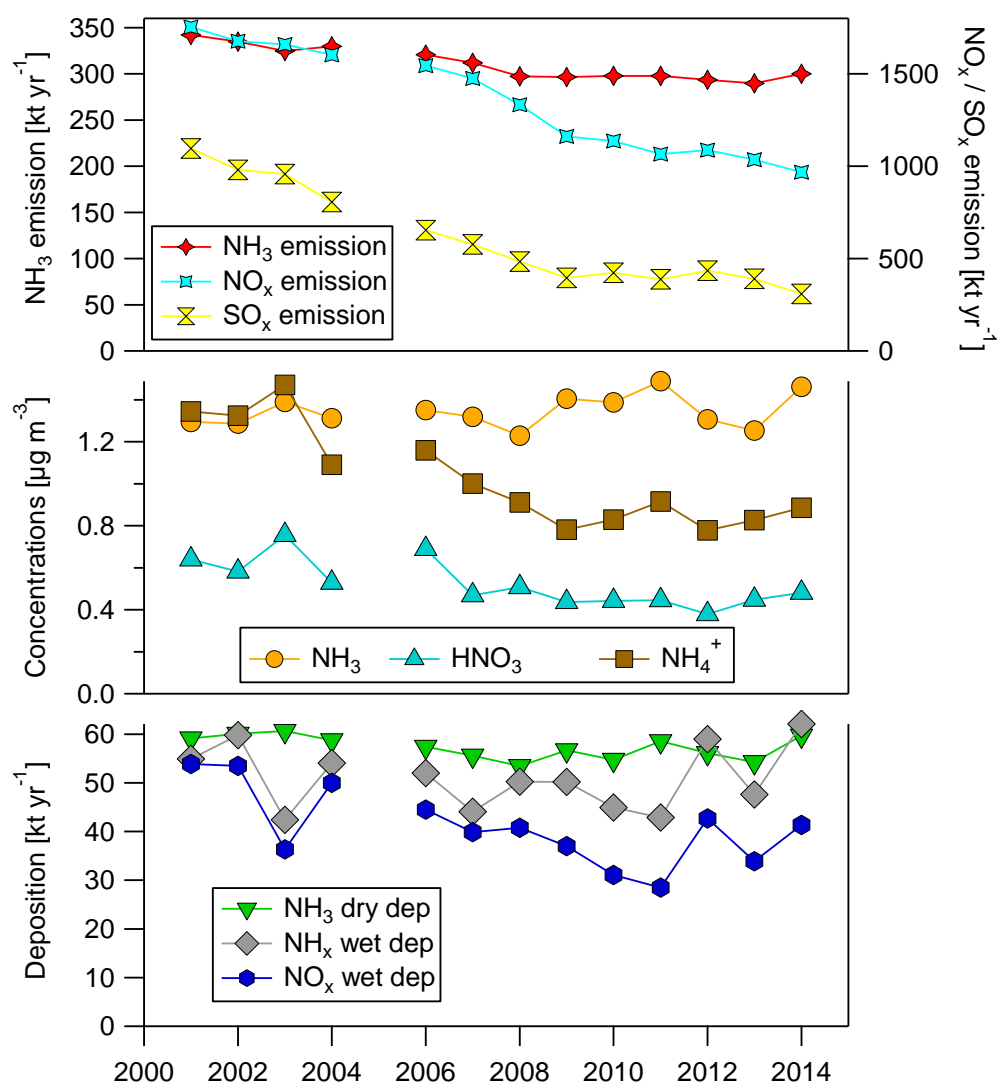


Figure 6a. Modelled trends in UK average concentrations and UK total deposition in response to emission changes as simulated with the EMEP4UK modelling system (Massimo Vieno, pers. commun.). 2003 was a particularly dry year where the annual mean was also affected by long-range transport events in the spring (Vieno et al. 2016b), while 2012 and 2014 were wet years.

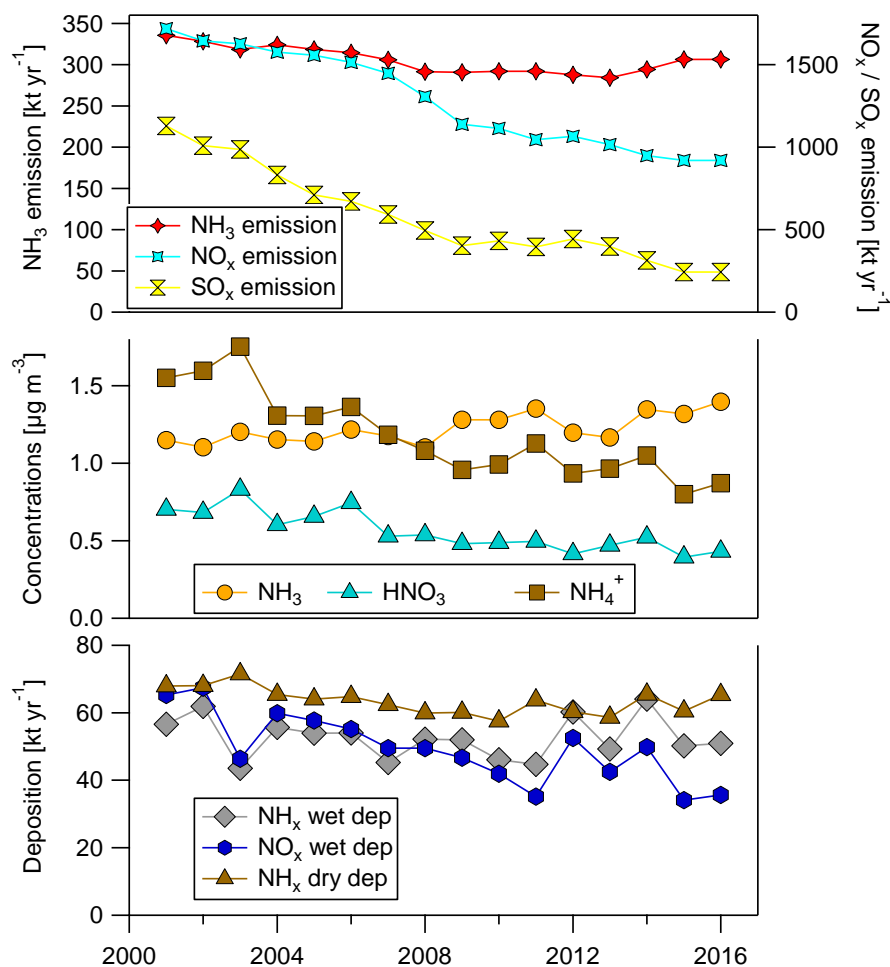


Figure 6b. Modelled trends in UK average concentrations and UK total deposition in response to emission changes as simulated with the EMEP4UK modelling system (Massimo Vieno, pers. commun.). 2003 was a particularly dry year where the annual mean was also affected by long-range transport events in the spring (Vieno et al. 2016b), while 2012 and 2014 were wet years.

2.5 Q 5: How effective have controls on NH₃ elsewhere in Europe been in reducing emissions and concentrations of NH₃ and particulate matter?

As noted above, the most effective regulatory controls on NH₃ emission in Europe have been applied in the Netherlands and Denmark since the late 1990s. In the Netherlands, a reduction in NH₃ emissions of about 50% has been estimated to be achieved between 1993 and 2014 (Figure 7) through reducing emissions from manure (van Zanten et al., 2017; Wichink Kruit et al., 2017). There has been much debate in these countries on the “ammonia gap” where NH₃ concentrations were found not to have reduced as quickly as anticipated (Erisman et al., 2001; Bleeker et al., 2009).

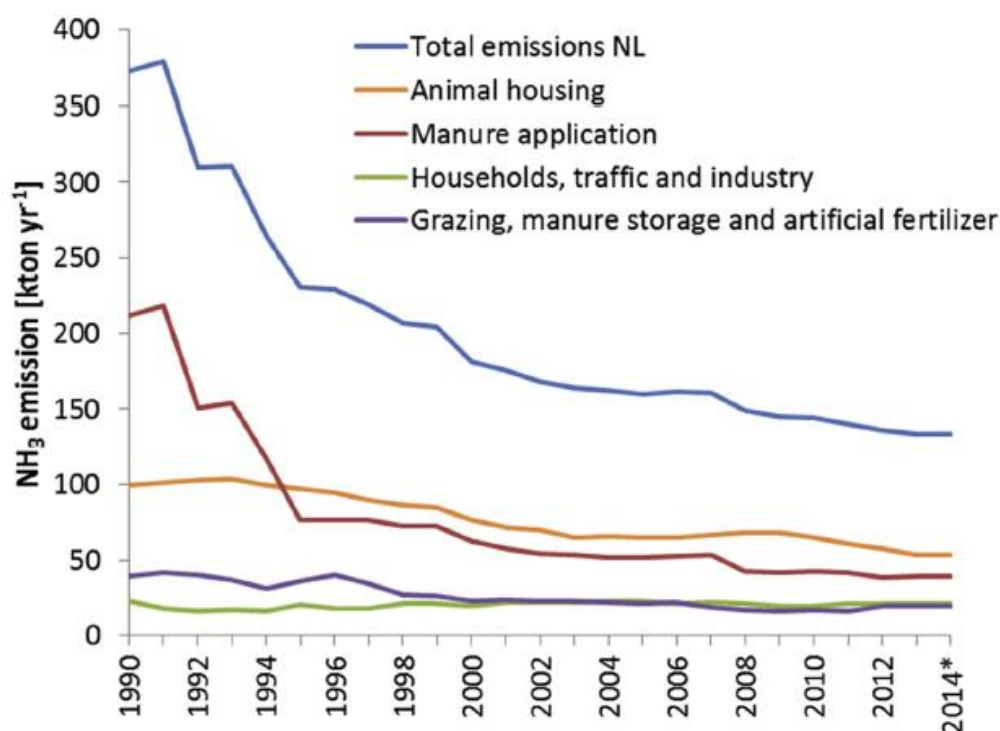


Figure 7: Estimated contributions to the changes in NH₃ emission from agriculture in the Netherlands 1990 to 2014 (Wichink-Kruit et al., 2017).

The corresponding trends in ambient NH₃, NO₂ and SO₂ concentrations in the Netherlands are shown in Figure 8. These show marked declines in SO₂ and NO₂, but much smaller changes in NH₃, despite the large reduction in reported emissions. Similar to the emission responses in the UK, no significant trend in NH₃ concentration was observed for the entire period. However, if broken up into two periods, there was an initial statistically significant decrease (-36%) in NH₃ during 1993-2004, followed by an increase (+19%) in 2005-2014 (van

Zanten et al., 2017). At the same time, NH_4^+ concentrations decreased by 68%, i.e. by more than the reduction in NH_3 emission.

Unlike in the UK, however, total NH_4^+ wet deposition also decreased, by 47%.

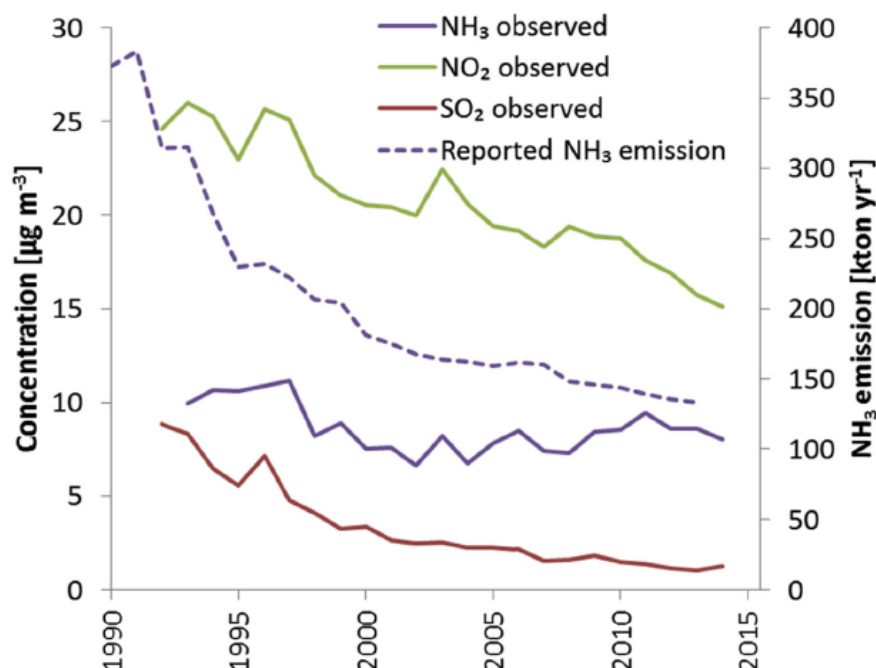


Figure 8. Trends in NH_3 emission and observed annual average NH_3 , NO_2 and SO_2 in the Netherlands between 1990 and 2014 (from Wichink Kruit et al., 2017)

Overall, analysis of the monitoring networks in the Netherlands, the UK, other European countries such as Hungary (Horvath et al., 2009) and Denmark (Bleeker et al., 2009), but also the US shows that the reduction in NH_3 emission since 1990 primarily resulted in reduction in particulate NH_4^+ concentrations rather than in gaseous NH_3 concentrations, which can be attributed to the simultaneous reduction in atmospheric acid concentrations and the change from sulphate to nitrate chemistry. This in turn maintained higher gaseous NH_3 concentrations in areas where NH_3 emissions have reduced, compensated by a substantial reduction in particulate NH_4^+ concentrations and, in the case of both the Netherlands and Denmark, a substantial reduction in wet deposited NH_4^+ (47% for Netherlands, van Zanten et al., 2017; 28% for Denmark, Bleeker et al. 2009).

The fact that the observations of a weak response in NH_3 concentration and a much stronger response in NH_4^+ concentrations is due to the reduction in emissions of NH_3 coupled with simultaneous reductions in NO_x and SO_2 emissions is strongly supported by the ability of atmospheric chemistry transport models to reproduce the observed trends in the Netherlands and the UK. The modelled trends for the UK have been presented above (Fig. 6). The modelled Dutch trends for the period 1990 to 2014 are shown in Figure 9, alongside the measurement trends (Wichink Kruit et al., 2017). The modelled data reproduce the trends in particulate matter quite well. The fact that the OPS model of Wichink Kruit et al. (2017) does not exactly match the gaseous NH_3 concentrations may be due to remaining uncertainties in capturing the chemical interactions. These findings are consistent with those also found for Hungary, where emissions decreased due to reductions in animal numbers and fertilizer use after 1990 (Horvath and Sutton, 1998; Horvath et al., 2009), and with the reductions in NH_3 and NH_4^+ concentrations observed following an active programme of NH_3 mitigation activities in Denmark (Bleeker et al. 2009).

In summary, the close scrutiny of responses to NH_3 emission reductions in the Netherlands raised early concerns over the effectiveness of the control measures, but it is clear now that temporal trends in gaseous NH_3 concentrations should not be seen in isolation from concurrent trends in particulate NH_4^+ and acid gas concentrations, especially SO_2 and HNO_3 . As outlined in answer to Question 3, a range of simple and well-validated control measures are available to reduced NH_3 emissions, which have been deployed in both the Netherlands and Denmark. In the case of the Netherlands, the results are consistent with an NH_3 emission reduction of approximately 50%. While the resulting reductions in concentrations of NH_3 in the Netherlands have been much smaller and statistically non-significant over the entire time-frame, particulate NH_4^+ reductions have been larger at 68% and NH_4^+ deposition reductions in between these two measures at 47%.

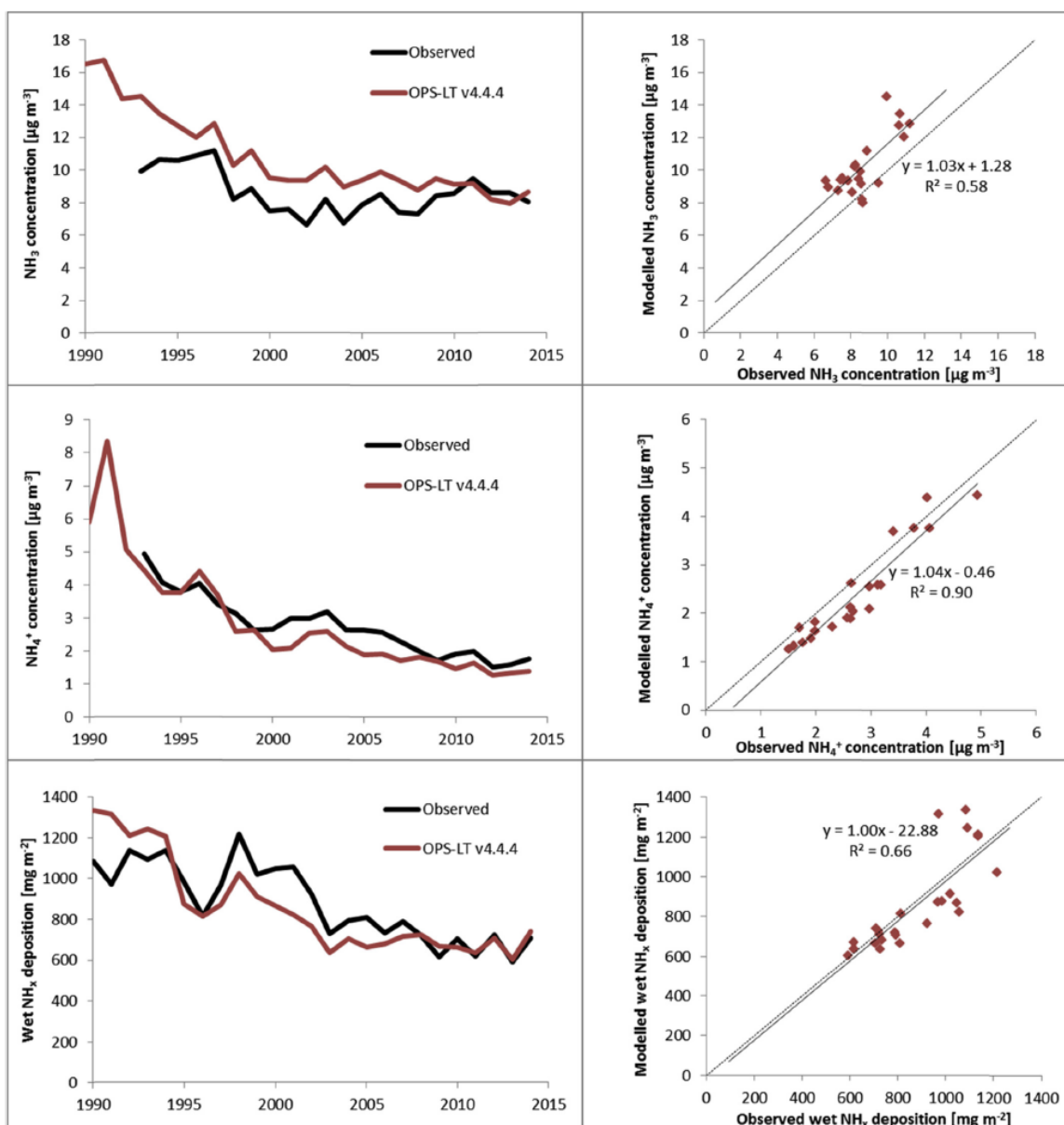


Figure 9. Annual average measured and modelled concentrations of NH_3 (upper panels), particulate NH_4^+ (middle panels) and average accumulated wet deposited NH_4^+ in the Netherlands between 1990 and 2014, (Kruit et al 2017)

2.6 Q6. Have changes in UK emissions of NO_x and NH₃ reduced the exceedances of critical loads for nutrient nitrogen? (and are both the oxidised and reduced forms of N equally important for eutrophication?).

The exceedance of critical loads for nutrient nitrogen are quantified by comparing the spatial distribution of total N deposition, including all oxidized and reduced forms of reactive nitrogen deposition with the critical loads (Hall et al., 2017). For these calculations, no distinction is made between the oxidized and reduced forms of nitrogen. However, it is clear that NH₃ in the gas phase is more damaging to some ecosystems per unit N deposited than oxidized N (Sheppard et al., 2010), at least for many moorland as well as moss and lichen species. The recognition of differential responses to different forms of reactive nitrogen deposited from the atmosphere led to a major revision of the UNECE Critical Levels for NH₃ which were reduced from a previous value of 8 µg m⁻³ (annual mean) to 1 µg m⁻³ (annual mean, habitats with lichens and bryophytes) and 3 (2-4) µg m⁻³ (annual mean, higher plants) (Cape et al., 2009; Sutton et al., 2009). The importance for the UK can be gauged by the findings of Hallsworth et al. (2010), who determined the extent to which Special Areas of Conservation (SACs) are subject to NH₃ concentrations larger than critical levels. For the 1 µg m⁻³ threshold, 69% of UK SACs were estimated to be exceeded, with the regional figures being 97% (England), 92% (Wales), 30% (Scotland) and 85% (Northern Ireland) and recent updates (Hall et al., 2017) with differing methodology have shown little change over time.

Total nitrogen deposition in the UK (Fig. 10) has declined by approximately 15% since 2000 in response to reductions in the wet and dry deposition of oxidized nitrogen (of about 30%) . The deposition of reduced nitrogen in the UK has not changed significantly since 2000, as detailed in answers to Questions 4 and 5, but inter-year variability in wet and dry deposition is large, typically 10 to 20 kt-N in totals of 100 to 150 kt-N. The trend of declining acid deposition has continued as a result of declines in sulphur and oxidized nitrogen emissions and deposition and the exceedances of critical loads for nitrogen (eutrophication) are now the main focus of air pollution impacts on the natural environment. Recent reported trends in Critical Load exceedances for the UK show (Hall et al 2017) :

- The area of acid-sensitive habitats in the UK with exceedance of acidity critical loads has fallen by over a third, from 72.6% in 1995 to 44.1% in 2015.

- The magnitude of the acidity exceedance (expressed as the Average Accumulated Exceedance) for all UK habitats combined more than halved, from 0.78 to 0.28 keq ha⁻¹ year⁻¹ between 1995 and 2015.
- The area of nitrogen-sensitive habitats in the UK with exceedance of nutrient nitrogen critical loads has fallen from 75% in 1995 to 63.4% in 2015, (i.e., overall decline of about 15%).
- The magnitude of nutrient nitrogen exceedance (expressed as the Average Accumulated Exceedance) for all UK habitats combined has decreased by one-third, from 9.5 kg N ha⁻¹ year⁻¹ in 1995 to 6.2 kg N ha⁻¹ year⁻¹ in 2015.

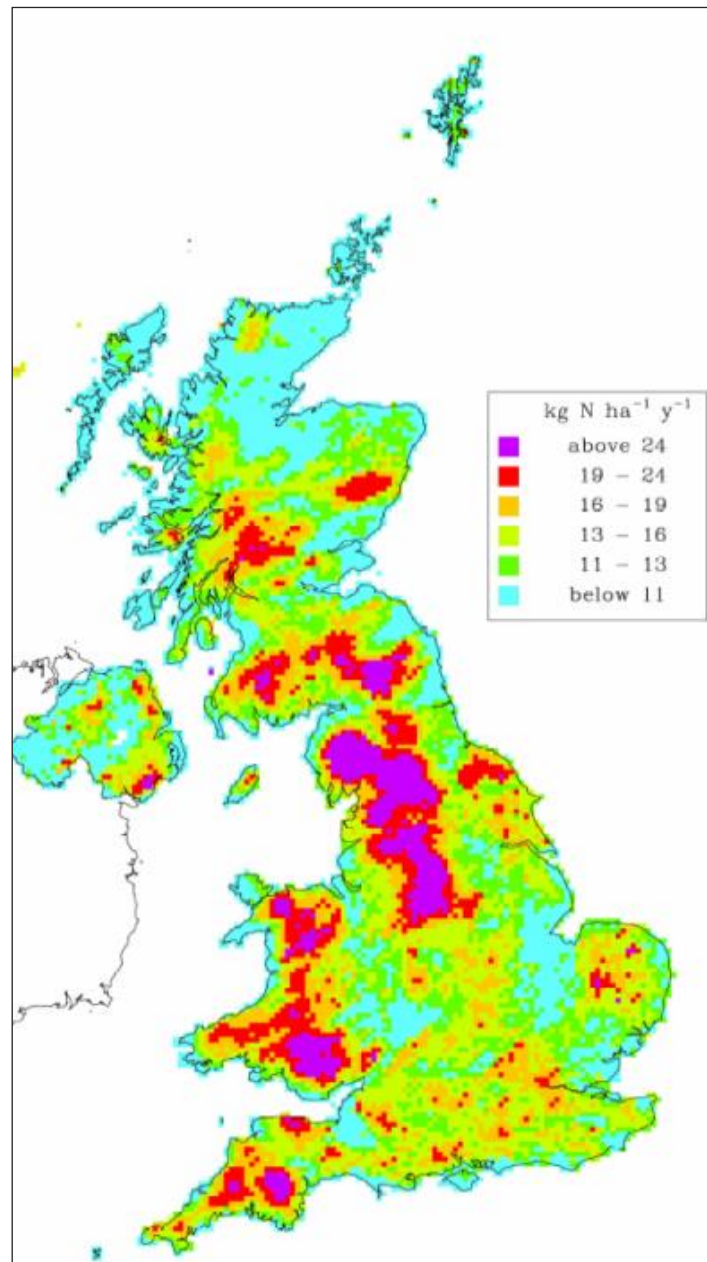


Figure 10. Total deposition of N in the UK 2015. The map comprises wet deposition of NH_4^+ , NO_3^- and dry deposition of NO_2 , HNO_3 and NH_3 . Estimates from CBED (concentration based estimates of deposition) as described in RoTAP 2012.

2.7 Q7. What are the relative air quality benefits and costs for controls on ammonia emissions?

The cost benefit analysis needs to consider the costs to farmers of taking action to reduce NH_3 emissions, the benefits to farmers of improving nitrogen use efficiency and the benefits to society of improving air quality.

2.7.1 Benefits

2.7.1.1 PM concentrations

The greatest benefits of control measures to reduce NH_3 emissions to date have been the reductions in particulate NH_4^+ , which (as detailed in answers to Questions 4 and 5) have declined more rapidly than NH_3 concentrations in air. However, from the measurements alone it is unclear how much of the NH_4^+ reduction observed can be attributed to decreases in NH_3 emission and how much to the simultaneous decrease in emissions of NO_x and SO_2 . Such attribution is only possible through model scenario runs. Because the formation of NH_4NO_3 approximately forms an equilibrium with the concentration product of NH_3 and HNO_3 , there are areas where NH_4NO_3 formation is NH_3 limited (i.e. it responds more sensitively to changes in NH_3 concentration) and areas that are HNO_3 limited. A modelling study, using the same EMEP4UK modelling framework that has successfully reproduced past trends (Fig. 6 above), suggested that a 30% reduction in UK NH_3 emissions would reduce $\text{PM}_{2.5}$ concentrations by 0.3 to 0.5 $\mu\text{g m}^{-3}$ over most of England and Wales, for the reference year 2010 (Figure 11; Vieno et al., 2016a). At the same time, the model suggested that about 50% of the particulate NH_4^+ related PM in the UK originated from gases emitted elsewhere in Europe. Thus, additional NH_3 emission reduction across Europe would result in a larger reduction in PM.

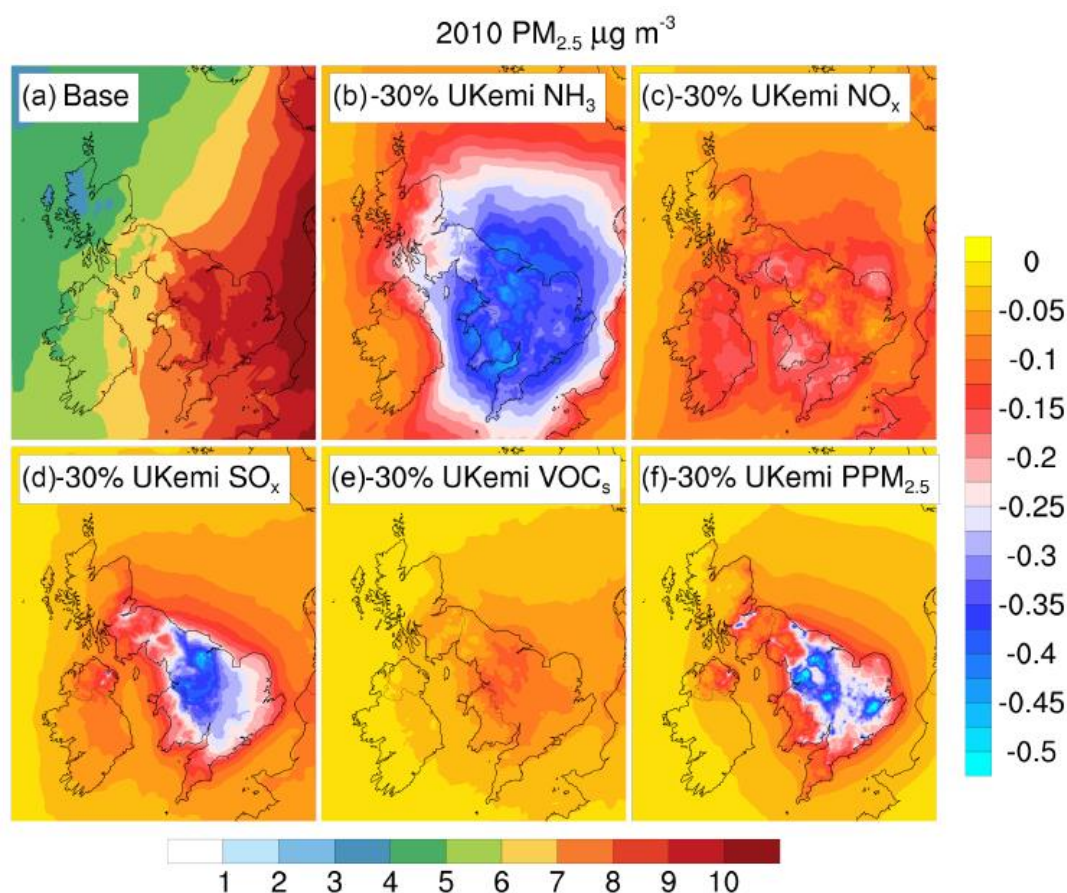


Figure 11. (a) PM_{2.5} concentrations simulated for 2010 (bottom colour scale) and changes in PM_{2.5} concentrations in response to 30% emission reductions of (b) NH₃, (c) NO_x, (d) SO_x, (e) volatile organic compounds and (f) primary PM_{2.5} (right-hand-side colour scale) (from Vieno et al., 2016a).

A global modelling study (Pozzer et al., 2017) calculated that a 50% reduction in European NH₃ emissions would result in a reduction of the European average PM_{2.5} concentration by about 1 µg m⁻³. This would be associated with a reduction of the UK PM_{2.5}-related premature mortality by 21%. It should be noted that this analysis of premature mortality assumes all chemical components of PM_{2.5} to be equally detrimental to human health. There is currently no conclusive evidence to either support or disprove this assumption.

A similar sensitivity of PM to changes in NH₃ emission was derived in a further European modelling study (Megaritis et al., 2013). By contrast, two studies (Redington et al., 2009; Harrison et al., 2013) derived much smaller sensitivities to NH₃ emission reductions, but both these studies did not use fully coupled 3-dimensional Eulerian chemistry and transport models and derived their conclusions from the modelling of short time-periods which increases uncertainty of being representative of the full year. Some of the high NH₄NO₃ episodes in the

UK are dominated by UK emissions, whilst others are controlled by European emissions outside the UK (Vieno et al., 2014, 2016b), and some high concentration episodes (as investigated by Redington et al. (2009)) may be particularly insensitive to NH_3 changes because they are particularly HNO_3 limited.

2.7.1.2 Deposition

The effect of ammonia emission abatement on the area of critical loads exceedance can also be simulated using atmospheric chemistry transport models. For 2020 emissions as predicted in 2012, further NH_3 emission reduction scenarios of 15 to 26% has been suggested to result in an additional protection of 26 to 72% of unprotected area of the Spatial Areas of Conservation sites in the UK (CEH, 2012). Available evidence suggests that that ecosystems tend to recover following a reduction in ammonia deposition, although there has been much debate on the timescales needed for recovery (ROTAP, 2012). For example, Mitchell et al. (2004) found recovery of epiphytic species one year after transplanting from a polluted to a clean site, while for forest ground flora, trees and soils cycling of available nitrogen pools may require several decades for full recovery. Field observations from a peatland ecosystem showed substantial recovery in moss and lichen (*Sphagnum* and *Cladonia*) populations within 5 years of cessation of a local point source of NH_3 emissions (M. Sutton and N. van Dijk, pers. commun.).

2.7.2 Costs

Most effort has been placed on estimating the NH_3 mitigation costs. In early studies during the 2000s, assessments tended to estimate very high costs of NH_3 mitigation. This seemed to be based on a precautionary approach to the costs and lack of widespread experience, as well as the potential for improved experience and scaling up to reduce costs. A major review and re-evaluation of the mitigation costs led to substantial re-valuing of the mitigation costs, which led to much smaller estimates (Reis et al., 2015). At the same time it was agreed that the value of the nitrogen saved in reducing NH_3 emissions should be included in regional cost estimates.

The resulting cost estimates calculated for the European Commission under the review of the National Emissions Ceilings Directive included these revisions. These included cost curves, such that the first measures taken could be estimated to be profitable for farmers, while increasing ambition would be associated with larger costs. The report of Amann et al. (2014) estimated that maximum technically feasible reduction in emissions (MTFR) would reduce EU emissions by around 1000 kt NH_3 per year, almost all of which was associated with agricultural

mitigation, at a cost of around 2000 million € per year, which is equivalent to around €2 per kg NH_3 abated, or about €2.40 per kg N abated.

By comparison the European Nitrogen Assessment estimated the damage costs of different forms of nitrogen pollution at 70-320 billion € per year (Sutton et al., 2011), subsequently updated by van Grinsven et al. (2013) to 75-485 billion € per year. Embedded in these estimates were estimated costs of NH_3 pollution to human health and biodiversity protection. According to the ENA approach, these were estimated at €2 to €20 per kg N pollution for human health and €2 to €10 per kg N pollution for biodiversity related societal costs. Combined these provide estimates in the region of €4 to €30 per kg N pollution as NH_3 , with the wide uncertainty ranges reflective of both the uncertainty in scientific knowledge and the challenge of applying a willingness to pay approach to different societal costs.

Overall, this comparison shows that the mitigation costs (even with a MFTR scenario) are much smaller than the estimated societal benefits based on the current estimates. The costs to farmers would also be expected to reduce substantially through further upscaling and technological refinement as circular economy approach emerges more strongly in the next years offering profits to farmers when combining the most cost-effective approaches as part of a strategy to improve nitrogen use efficiency. Here innovative approaches to improving nitrogen management may make a substantial contribution to cost savings and profitability on farms. For example, it has been calculated that total nitrogen losses from European farms amount to 18 billion tonnes per year (Sutton et al., 2011, ENA). Based on a fertilizer value of €0.80 per kg N, Sutton et al. (2017) have shown that these losses represent a cash loss of 25% of the entire EU Common Agricultural Policy (CAP) budget of €56 billion per year. Recognition of the scale of this financial loss should provide a huge motivation for farmers to reduce NH_3 and other nitrogen emissions. A study by Pinder et al (2007) of the options for reducing $\text{PM}_{2.5}$ concentrations in Eastern United States of America suggested that controlling ammonia emissions provided a more cost effective approach relative to further controls on SO_2 and NO_x emissions.

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