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Nitrous oxide emissions from fertilised UK arable soils: Fluxes, emission factors and mitigation

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A B S T R A C T

Cultivated agricultural soils are the largest anthropogenic source of nitrous oxide (N_2O), a greenhouse gas approx. 298 times stronger than carbon dioxide. As agricultural land covers 40–50% of the earth's surface agricultural N_2O emissions could significantly influence future climate. The timing, amount and form of manufactured nitrogen (N) fertiliser applied to soils are major controls on N_2O emission magnitude, and various methods are being investigated to quantify and reduce these emissions. A lack of measured N_2O emission factors (EFs) means that most countries report N_2O emissions using the IPCC's Tier 1 methodology, where an EF of 1% is applied to mineral soils, regardless of soil type, climate, or location. The aim of this research was to generate evidence from experiments to contribute to improving the UK's N_2O agricultural inventory, by determining whether N_2O EFs should vary across soil types and agroclimatic zones. Mitigation methods were also investigated, including assessing the impact of the nitrification inhibitor (NI) dicyandiamide (DCD), the application of more frequent smaller doses of fertiliser, and the impact of different rates and forms of manufactured N fertiliser. Nitrous oxide emissions were measured at one cropland site in Scotland and two in England for 12 months in 2011/2012, along with soil and environmental variables. Crop yield was also measured, and emission intensities were calculated for the contrasting fertiliser treatments. The greatest mean annual cumulative emissions from a range of ammonium nitrate (AN) fertiliser rates were measured at the Scottish site (2301 g N_2O-N ha^{-1}), which experienced 822 mm rainfall compared to 418 mm and 472 mm at the English sites, where cumulative annual emissions were lower (929 and 1152 g N_2O-N ha^{-1}, respectively). Climate and soil mineral N influenced N_2O emissions, with a combination of factors required to occur simultaneously to generate the greatest fluxes. Emissions were related to fertiliser N rate; however the trend was not linear. EFs for AN treatments varied between sites, but at both English sites were much lower than the 1% value used by the IPCC, and as low as 0.20%. DCD reduced AN- and urea-generated N_2O emissions and yield-scaled emissions at all sites. AN application in more frequent smaller doses reduced emissions at all sites, however, the type of fertiliser (AN or urea) had no impact. A significant difference in mean annual cumulative emissions between sites reflected differences in rainfall, and suggests that location specific or rainfall driven emission estimates could be considered.

1. Introduction

With a global warming potential 298 times greater than carbon dioxide (CO_2), nitrous oxide (N_2O) is a powerful greenhouse gas (GHG), additionally responsible for destruction of stratospheric ozone (Bhatia et al., 2010). Its potential impact on future climate is large, highlighting a requirement to identify the source and extent

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of N_2O emissions, and to mitigate emissions to limit our impact on climate change (Kavdir et al., 2008). Agriculture is responsible for approximately 60% of global (Bhatia et al., 2010) and 75% of the UK's (Skiba et al., 2012) anthropogenic N_2O emissions. In the UK this is a large increase on the 47% estimated in 1990 (Brown et al., 2002), mostly due to a decrease in non-agricultural (industrial sector) N_2O emissions over this time (Defra, 2015; Skiba et al., 2012). It is clear that agriculture is not matching other sectors in emission reduction performance, and covering 40–50% of the earth's surface (Flynn and Smith, 2010) has the potential to influence future climate. Nitrous oxide emissions in agriculture originate from many sources including manure storage and agricultural soils (Skiba et al., 2012). One of the greatest sources of emissions is 'cultivated agricultural soil' (Regina et al., 2013; Van Groenigen et al., 2010), which includes direct and indirect N_2O emissions from applied manures, synthetic fertiliser application, urine deposition and crop residues. Globally these emissions are estimated to contribute to 37% of total agricultural GHG emissions, and synthetic fertiliser direct + indirect N_2O emissions are estimated to be the third largest source of GHG emissions in agriculture (Tubiello et al., 2013). In the UK, total N_2O emissions from soils are estimated to contribute between 60% (Cardenas et al., 2013) and 92% (Buckingham et al., 2014; Salisbury et al., 2014) to total agricultural N_2O emissions, with synthetic fertilisers being the largest source of these emissions (Skiba et al., 2012). Although in the UK N_2O emissions from agriculture are decreasing (Salisbury et al., 2014), with emissions from synthetic fertilisers witnessing a 17% decline on 1990 levels in 2013 (Defra, 2015), they are increasing globally (Tubiello et al., 2013).

A reported 17% increase in global N_2O emissions from agriculture between 1990 and 2005 (Flynn and Smith, 2010) corresponds with increased nitrogen (N) fertiliser application in attempts to increase food production (Qin et al., 2012). It is estimated that in 2030 there will be global demand for 135 million tons of fertiliser N (Tilman et al., 2002; Van Groenigen et al., 2010). Manufactured fertiliser N applications are essential to optimise crop yields and quality. However over-application can lead to increased N_2O emissions (Archer and Halvorson, 2010; Qin et al., 2012; Snyder et al., 2014) and potential exponential emission responses (Hoben et al., 2011). Mitigation strategies often involve reducing N supply to reduce losses to the environment (Petersen et al., 2010); however without fertilisation global food supplies would decline. With global food security a major priority the focus is on increasing the efficiency of crop production, but typically less than half of N used in agriculture is currently utilised (Oenema et al., 2009). It is apparent that a balance must be attained (Sutton et al., 2011), and methods to reduce N_2O emissions are sought. Uptake of these methods will require confidence in their performance and quantification of the reductions achievable.

Before mitigation of N_2O emissions can be introduced, quantification is required to identify key emission sources, which can vary both spatially and temporally (Desjardins et al., 2010; Regina et al., 2013), making them difficult to estimate at various scales (Regina et al., 2013). Uncertainties in N_2O emission estimates have led many countries to adopt the IPCC Tier 1 methodology. In the UK, the emission factor (EF1) of 1% is applied to mineral soils (IPCC, 2006), assuming that 1% of N input as fertiliser is directly emitted, regardless of soil, climate, form of fertiliser N or rate of application. Nitrous oxide emission estimates are reported as the major source of uncertainty in national GHG inventories (Leip et al., 2011); with half of all published N_2O emission inventories using this approach (Philibert et al., 2012). Although Tier 1 EFs are easy to apply, they do not reflect spatial and temporal variability, or the impact of climate (Kim et al., 2014; Laville et al., 2011). A further concern is the assumed linear increase in N_2O emissions with N application. Much evidence suggests an exponential response (Cardenas et al., 2010; Hoben et al., 2011; McSwiney and Robertson, 2005; Shcherbak et al., 2014), indicating that EFs should depend on N input rate, but the processes behind a linear/non-linear relationship are not completely understood (Kim et al., 2013).

Annual emission estimates constructed from measured data are often calculated from 15 to 30 measurement dates (Bouwman et al., 2002; Laville et al., 2011), but large temporal variation over an annual period means that peaks of emission can be missed and interpolation between distant points is necessary. Both of these factors mean that the annual emission based on limited measurement points has a high degree of uncertainty. Quantification of emissions is further complicated by variations dependent on the type of fertiliser applied (Regina et al., 2013). Some studies report greater emissions from nitrate- than ammonium- and urea-based fertilisers (Dobbie and Smith, 2003a; Smith et al., 2012), but there is uncertainty, with Bouwman et al. (2002) reporting greater emissions from ammonium-based fertilisers, and Li et al. (2013) reporting conflicting results. There is also evidence that N_2O emissions vary with soil N and C content (Ding et al., 2013; Miller et al., 2008; Tiemann and Billings, 2008), soil texture (Maag and Vinther, 1996), soil pH, climate, and crop type (Jones et al., 2007; Regina et al., 2013). Measurements of N_2O emissions from different climates and management systems are scarce (Laville et al., 2011; Rees et al., 2013), but if emissions vary with soil and climate then location based EFs may improve the accuracy of N_2O emission inventories. Smith et al. (2012) considered use of the IPCC default EF to be an important weakness in emission inventories. There is a clear requirement for improved emission estimates before large-scale mitigation can be implemented. Yield-scaled emission estimates are especially important, because not only is there a need to decrease N_2O emissions, but to simultaneously increase food production (Qin et al., 2012).

In attempts to limit emissions several methods have been investigated, including applications of more frequent but smaller doses of fertiliser to avoid unutilised surplus N (Maidl et al., 1996). As denitrification and nitrification are controlled by external soil and climatic factors the time of application can also influence N_2O emissions. With maximum denitrification in temperate climates most likely to occur in late autumn to early spring (when rainfall and soil moisture are high and oxygen status low) (Cameron et al., 2013), it is often advised to avoid fertiliser application at this time. The impact of temperature, however, creates a complex situation and more research into these interactions and their influence on emissions is required. Other research has investigated reducing emissions using nitrification inhibitors (NIs) (Pfab et al., 2012), which can also potentially improve crop yields (Cui et al., 2011; Di and Cameron, 2012; Weiske et al., 2001). Reported results have been mixed (*e.g.* Menendez et al., 2012), although a recent meta-analysis (Abalos et al., 2014) of worldwide (predominantly) field studies concluded that *N*-(*n*-butyl)thiophosphoric triamide (NBPT, a urease inhibitor), dycandiamide (DCD, a NI) and DCD + NBPT increased crop productivity and N use efficiency (NUE) compared to the control, but with varying degrees of success for crop productivity. The study also concluded that DMPP (3,4-dimethylepyrazole phosphate) increased NUE but not crop productivity. Before they can be advocated as a robust mitigation option for use on arable land there is a requirement for more research into the environmental impact of NIs, and for N_2O emission estimates over a complete annual cycle (Liu et al., 2013; Pfab et al., 2012).

The aim of this research was to assess whether N_2O emissions are controlled by location specific soil and environmental factors, and whether the use of a universal EF of 1% for applications of N fertiliser to mineral soils is appropriate for UK conditions. In addition, the research was designed to evaluate a number of strategies to reduce N_2O emissions; *viz*, the effect of the