Denitrification potential in subsoils: A mechanism to reduce nitrate leaching to groundwater

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ABSTRACT

Understanding subsurface denitrification potential will give greater insights into landscape nitrate (NO_3^-) delivery to groundwater and indirect nitrous oxide (N_2O) emissions to the atmosphere. Potential denitrification rates and ratios of $N_2O/(N_2O + N_2)$ were investigated in intact soil cores collected from 0–0.10, 0.45–0.55 and 1.20–1.30 m depths representing A, B and C soil horizons, respectively from three randomly selected locations within a single intensively managed grazed grassland plot in south eastern Ireland. The soil was moderately well drained with textures ranging from loam to clay loam (gleysol) in the A to C horizon. An experiment was carried out by amending soils from each horizon with (i) 90 mg NO_3^-–N as KNO_3, (ii) 90 mg NO_3^-–N + 150 mg glucose-C, (iii) 90 mg NO_3^-–N + 150 mg DOC (dissolved organic carbon, prepared using top soil of intensively managed grassland) kg^{-1} dry soil. An automated laboratory incubation system was used to measure simultaneously N_2O and N_2, at 15 °C, with the moisture content raised by 3% (by weight) above the moisture content at field capacity (FC), giving a water-filled pore space (WFPS) of 80, 85 and 88% in the A, B and C horizons, respectively. There was a significant effect ($p < 0.01$) of soil horizon and added carbon on cumulative N_2O emissions. N_2O emissions were higher from the A than the B and C horizons and were significantly lower from soils that received only nitrate than soils that received NO_3^- + either of the C sources. The two C sources gave similar N_2O emissions. The N_2 fluxes differed significantly ($p < 0.05$) only between the A and C horizons. During a 17-day incubation, total denitrification losses of the added N decreased significantly ($p < 0.01$) with soil depth and were increased by the addition of either C source. The fraction of the added N lost from each horizon were A: 25, 61, 45%; B: 12, 29, 28.5% and C: 4, 20, 18% for nitrate, nitrate + glucose-C and nitrate + DOC, respectively. The ratios of N_2O to $N_2O + N_2$ differed significantly ($p < 0.05$) only between soil horizons, being higher in the A (0.58–0.75) than in the deeper horizons (0.10–0.36 in B and 0.06–0.24 in C), clearly indicating the potential of subsoils for a more complete reduction of N_2O to N_2. Stepwise multiple regression analysis revealed that N_2O flux increased with total organic C and total N but decreased with NO_3^-–N which together explained 88% of the variance ($p < 0.001$). The N_2 flux was best explained ($R^2 = 0.45$, $p < 0.01$) by soluble organic nitrogen (SON) (positive) and with NO_3^-–N (negative). Stepwise multiple regression revealed a best fit for total denitrification rates which were positive for total C and negative for NO_3^-–N with the determination coefficient of 0.76 ($p < 0.001$). The results suggest that without C addition, potential denitrification rate below the root zone was low. Therefore, the added C sources in subsoils can satisfactorily increase nitrate depletion via denitrification where the mole fraction of N_2O would be further reduced to N_2 during diffusional transport through the soil profile to the atmosphere and/or to groundwater. Subsoil denitrification can be accelerated either through introducing C directly into permeable reactive barriers and/or indirectly, by irrigating dirty water and manipulating agricultural plant composition and diversity.

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1. Introduction

An excess of N in the environment is viewed as an escalating global threat, due to its impacts on groundwater quality and the atmosphere (Stark and Richards, 2008). Soils under grazed

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grassland often have high concentrations of nitrate (NO_3^-), arising from the application of mineral fertilizers, slurries, animal excreta and from the native soil organic matter (Foster, 2000). Large amounts of N transferred within the soil system increase the potential and the opportunities for NO_3^- losses (Davies, 2000). The average leaching losses of NO_3^- from terrestrial ecosystems in central Europe is 15 kg N ha^{-1} y^{-1} (Werner, 1994). Nitrate transformation in the root zone is well documented (Ibendahl and Fleming, 2007), but its movement and transformations in prevailing geochemical conditions below the root zone are less well understood (Jarvis and Hatch, 1994). The added NO_3^- can be transported through percolating water and transformed to gaseous forms, thereby leaving agricultural systems, or may be lost through leaching and runoff (Clough et al., 2005). Substantial quantities of dissolved inorganic N, particularly NO_3^-, are exported through low order streams (Alexander et al., 2000). Nitrate contamination of surface water and groundwater is common in watersheds dominated by agricultural activities (Townsend et al., 2003), primarily because of diffuse pollution from intensive farming (Foster and Young, 1980). Denitrification is one of the most important processes that can control the quantity of nitrate available for leaching from soil to water (Jarvis, 2000).

Denitrification is the mainly microbial reduction of NO_3^- to the gaseous products nitric oxide (NO), nitrous oxide (N_2O) or dinitrogen (N_2). This process is an important mechanism for nitrate removal in a variety of suboxic environments (Seitzinger et al., 2006). Some studies have shown that the highest rates of denitrification occur in the upper soil horizon (Clement et al., 2002; Cosandey et al., 2003; Kustermann et al., 2010), the extent of which depends on moisture levels (Khalil and Baggs, 2005). Recently, researchers have found microbial 'hot spots' with significant denitrification activity in patches of organic rich subsoils at depths of several meters (Hill et al., 2004) and in urine treated subsoils (Dixon et al., 2010). Subsoil denitrification has been suggested as an important mechanism for the removal of excess NO_3^- before leaching to groundwater, transport within saturated subsoil zones, or discharge to surface aquifers via subsurface drainage (Fenton et al., 2009; Sotomayor and Rice, 1996). Denitrification not only serves as a natural pathway for the elimination of excess NO_3^- in soil and water (Ellis et al., 1975), but also contributes to the emissions of N_2O, a potent greenhouse gas (Knowles, 1982) and an indirect contributor to the depletion of ozone (O_3) in the stratosphere (Crutzen, 1970). An interesting feature of denitrification in subsurface soils is that it is likely to be overlooked as a contributor to global atmospheric N_2O concentrations, due to the possible further reduction of N_2O to N_2 under O_2 limited conditions during upward diffusion through the soil profile, if adequate sources of organic carbon (C) are present (Elmi et al., 2003; Castle et al., 1998).

The beneficial effect to the environment of NO_3^- removal by denitrification depends on the partitioning of its end-products into N_2O and N_2. Knowledge of the denitrification gaseous end-products and the $N_2O/(N_2O + N_2)$ ratio is necessary to assess accurately the environmental consequences of the denitrification process (Elmi et al., 2003), with emphasis on the subsoil environment (Bergsma et al., 2002). The lack of information on N_2 emissions from terrestrial ecosystems not only limits our understanding of its significance as a sink for reactive N, but also impedes the quantification of the process as a whole (Davidsson and Seitzinger, 2006; Groffman et al., 2006) so that N budgets in biogeochemical models are incomplete (Boyer et al., 2006). To date, only a few estimates of denitrification in the subsoils of riparian wetlands and peat soils have been reported (Casey et al., 2001; Dhondt et al., 2004; Hill et al., 2000, 2004; Well et al., 2001). Depending on the environmental conditions, the mechanisms and magnitude of denitrification losses in subsoils of grazed grassland may however, deviate considerably from those of other sites warranting further investigation under

grassland ecosystems. The relative importance of the denitrification process depends strongly on certain environmental conditions including O_2 concentration, NO_3^- content and C availability (Tiedje, 1988), though their influences on the mole fractions of N_2O and N_2 in agricultural soils are still under debate, with little consensus (Venterea et al., 2005). Where organic C is added, a significant denitrifying potential may be revealed at depths as great as 7 m (Jarvis and Hatch, 1994; McCarty and Bremner, 1992).

A lack of organic C to provide energy to denitrifiers is usually identified as the major factor limiting denitrification rates (Devito et al., 2000; Pabich et al., 2001). More precisely, the quality and quantity of the C source is most often more important than total organic C due to its variable availability to microbes (Ciarlo et al., 2007). The specific contribution of the different C sources available to denitrifying micro-organisms has not been defined (Beauchamp et al., 1989). Therefore, knowledge of the factors controlling the denitrification process and, more specifically the $N_2O/(N_2O + N_2)$ ratios, are crucial to improve our understanding of the extent of complete reduction of NO_3^- via denitrification occurring in subsoil environments. Concerning health and environmental hazards of NO_3^- and the global warming potential of N_2O, we hypothesized that the addition of a readily available source of C (glucose) would enhance the reduction of N_2O to N_2 in subsoils and show a lower $N_2O/(N_2O + N_2)$ ratio in amended soils than in unamended soils. The main objectives of this research were (a) to measure the potential denitrification rates in subsoils under optimized substrate and moisture conditions and (b) to investigate the relationships between soil properties and maximum denitrification rates and ratios of $N_2O/N_2O + N_2$.

2. Materials and methods

2.1. Study site characteristics

Soil samples were collected in January 2008 (winter) from three randomly selected locations within a single intensively managed grazed grassland plot at the dairy farm of Teagasc Environment Research Centre, Johnstown Castle, Wexford, Ireland (52.3342°N, −6.4575°W). The soil textures of a profile up to 1.3 m depth varied from loam to clay loam (Brown Earth) overlying Ordovician sediments of sandstone and shale. Soil physical and chemical properties including the initial nitrate content of three horizons at the experimental site are presented in Table 1. The average groundwater table is below 1.2 m during winter and below 2.0 m during summer. On a yearly average, 24 cows graze the land for a total of 50 days and about 375 kg N ha^{-1} y^{-1} is harvested in total by one silage cut and offtake by grazing animals. The total annual N inputs are about 450 kg N ha^{-1} from inorganic fertilizers, animal excrement and N deposition.

2.2. Soil sampling

Intact soil cores (45) were collected from three depths (0–0.10, 0.45–0.55 and 1.20–1.30 m), representing the A, B and C horizons, of the soil profile. Stainless steel cylinders (0.12 m × 0.15 m) were manually inserted up to 0.02 m below the top rim of the core (with a 0.02 m wooden spacer inside the corer) using a percussion hammer into the soil after trimming off the swards to sample the surface/upper horizon (0–0.10 m) and then a hole was dug around the cylinder to assist removal, giving each core a size of 0.1 m × 0.15 m. The two other (deeper) horizons were sampled from the same locations by first removing the soil from the upper horizons. Fine mesh netting was placed over the top and bottom of the cylinders to contain the soil and kept in place using rubber bands at both ends. Soil samples were stored immediately after collection in a cold room at 4 °C and transported to Rothamsted Research, North Wyke, UK, in