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Indirect emissions of N₂O, CO₂ and CH₄ via groundwater in contrasting agricultural systems

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Introduction

The dynamics of dissolved carbon (C) and nitrogen (N) in groundwater is a key “missing piece” in our understanding of global C and N balances. In addition, quantification of dissolved nitrous oxide (N₂O), carbon dioxide (CO₂) and methane (CH₄) in groundwater beneath an agricultural system is of huge importance for global greenhouse gas (GHG) budget estimation. This research aimed to (i) measure the amount of dissolved C and N losses from two terrestrial ecosystems to the aquatic ecosystems via groundwater, and (ii) estimate the contribution of indirect emissions of GHG to the atmosphere.

Materials and Methods

The investigation was carried out at two low permeability (L: L1, Johnstown Castle; L2, Solohead) and two high permeability (H: H1, Oak Park; H2, Dairygold) sites in Ireland. Of the sites, L1, L2 and H2 were grassland and H1 was arable. Groundwater sampling was carried out monthly between Feb, 2009 and Jan, 2011 for hydrochemistry and dissolved gases. For dissolved N₂O, CO₂ and CH₄, samples were degassed using *He* headspace extraction technique (*He*: water =3:10; Jahangir et al., 2012) and analysed by gas chromatography. Denitrified N₂ was measured using Membrane Inlet Mass Spectrometer (MIMS). Prior to groundwater sampling water table (WT) depth was measured using an electronic dip meter. A water balance was used to calculate the effective rainfall (ER). Analysis was performed using the General Analysis of Variance (GenStat, Version 13). For each dissolved gas, the effects of location and time were examined along with their interactions using repeated measures analysis. Where significant differences were found, Tukey–Kramer HSD multiple comparison test were used to distinguish differences between individual locations. Differences between the two study years (2009 and 2010) were tested using a paired *t* test.

Results and Discussion

Total N input was 300, 213, 150 and 297 kg N ha⁻¹, respectively at L1, L2, H1 and H2 sites (Jahangir et al., 2013). Among the grassland sites, the number of livestock units (LU) was lower at L2 (2.0 LU) than L1 (2.2 LU) and H2 (2.2 LU). Rainfall was well above average (130–140%) in 2009 and below average (87–90%) in 2010 across sites (Table 1 and 2). The annual WT fluctuation ranges were 1.9, 0.7, 3.5 and 5.3 m below ground level. Mean N₂O-N conc. over the two years differed significantly between sites (*p*<0.001) (Table 3 and 4). Dissolved CO₂ conc. was significantly higher at grassland than arable sites. Mean CH₄ conc. was higher at the L sites than the H sites. Mean dissolved N (DN=NO₃⁻-N+NO₂⁻-N+N₂-N+N₂O-N+NH₄⁺+dissolved organic N) loads in groundwater over the two years accounted for 12, 8, 38, and 27% of

the surface N input. The major fraction of DN was NO₃⁻ N (81–92%) at H sites and N₂ (46–77%) at L sites. Indirect N₂O emissions via groundwater denitrification accounted for 0.03–0.12% of N input. Loads of dissolved C (dissolved organic C (DOC)+CO₂+CH₄) discharged ranged from 78–344 kg ha⁻¹ at L and 30–217 kg C ha⁻¹ at H sites.

Table 1. Annual rainfall (P), potential evapotranspiration (PET), actual evapotranspiration (AET) and effective rainfall (ER) at four sites during 2009 and 2010

| Hydrologic events | Year | L1 | L2 | H1 | H2 |
|-------------------|------|------|------|------|------|
| P (mm) | 2009 | 1452 | 1403 | 1167 | 1293 |
| | 2010 | 947 | 879 | 759 | 869 |
| PET (mm) | 2009 | 632 | 681 | 713 | 694 |
| | 2010 | 633 | 686 | 718 | 700 |
| AET (mm) | 2009 | 615 | 643 | 630 | 620 |
| | 2010 | 562 | 553 | 518 | 543 |
| ER (mm) | 2009 | 836 | 759 | 537 | 673 |
| | 2010 | 385 | 326 | 241 | 326 |

Table 2. N₂O, CO₂ and CH₄ concentrations in groundwater at four sites in 2009 and 2010 (mean ± SE, n=12)

| Dissolved gases | Year | L1 | L2 | H1 | H2 |
|---|------|-------------|-------------|--------------|-------------|
| N ₂ O (mg NL ⁻¹) | 2009 | 0.037±0.001 | 0.033±0.002 | 0.039±0.002 | 0.049±0.001 |
| | 2010 | 0.015±0.006 | 0.012±0.001 | 0.018±0.0061 | 0.046±0.002 |
| CO ₂ (mg CL ⁻¹) | 2009 | 37.5±4.5 | 30.8±7.1 | 14.7±2.8 | 34.7±3.3 |
| | 2010 | 33.7±4.2 | 22.4±4.7 | 9.5±1.4 | 30.9±3.1 |
| CH ₄ (mg CL ⁻¹) | 2009 | 0.206±0.011 | 0.013±0.011 | 0.007±0.003 | 0.002±0.000 |
| | 2010 | 0.279±0.013 | 0.042±0.017 | 0.003±0.002 | 0.001±0.001 |

Conclusions

Substantial loads of C and N lost from the terrestrial agricultural ecosystems to surface water via groundwater. Groundwater CO₂-C export was an important part of the farm C balance accounting for loads up to 314 kg C ha⁻¹ year⁻¹. Methane occurrence in groundwater from the terrestrial agricultural ecosystem was episodic, with low emissions. Mean indirect N₂O emissions via groundwater denitrification accounted for approximately 0.03–0.12% of farm N input, which seems to be an important component of atmospheric N₂O emissions.

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