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Nitrogen Compounds Emission from Agricultural Lands with Hevey Application of Nitrogen Fertilizer －Nitrous Oxide Emission from Upland Fields and Leaching of Nitrogen from Forage Rice Paddies with Heavy Application of Cattle Manure－

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Summary

Nitrous oxide (N_2O) is a greenhouse gas that has a high global warming potential and a long atmospheric lifetime. The Intergovernmental Panel on Climate Change (IPCC) pointed out that the main cause of the increase in N_2O concentration in the troposphere is agriculture. Therefore, it is necessary to find ways to decrease N_2O emission from agricultural fields. However, N_2O emission has large temporal and spatial variations. The third report from the IPCC illustrated the large range of N_2O emission (from 2 to 21 Tg-N yr^{-2}). Long-term field monitoring and elucidation of the cause of N_2O emission are required for accurate estimations of global N_2O emission. In this study, we measured N_2O fluxes in Gray Lowland soil and Andosol during the snow-free season (from April to November) for either three or six years. The objectives of this study were to evaluate the seasonal patterns and amounts of N_2O emission, and to compare our results with values obtained in previous studies.

In Gray Lowland soil from 1995 to 2000, N_2O and nitric oxide (NO) fluxes from the soil to the atmosphere ranged from 0.00 to 1.86 mg-N m^{-2} h^{-1} and from 0.00 to 3.30 mg-N m^{-2} h^{-1}, respectively. In the fertilized plot of the Gray Lowland soil, the highest N_2O emissions were observed around harvesting time, from August to October with a high rainfall frequency, as well or better immediately after fertilization in May. In contrast, the NO flux increased immediately after fertilizer application. In the non-fertilized plots of Gray Lowland soil, NO and N_2O flux did not increase immediately after fertilizer application, but only N_2O flux did increase around harvesting time. The seasonal patterns of soil nitrate (NO_3^-) and ammonium (NH_4^+) levels and the ratio of N_2O/NO flux indicated that the main process responsible for N_2O production after fertilization was nitrification and that the main process responsible for N_2O production around harvest time was denitrification. The increase in N_2O flux was enhanced by the addition of water from rainfall and of organic matter from onion planting. A significant correlation could be observed between N_2O and carbon dioxide (CO_2) flux. The cumulative N_2O flux during the snow-free season for six years ranged from 0.35 to 1.56 g-N m^{-2}, and about 70% of this flux occurred near harvesting time, from August to October. Therefore, it is necessary to monitor N_2O flux during the entire growing season in order to estimate the annual N_2O emission.

In Andosol from 1998 to 2000, the N_2O and NO fluxes ranged from 0.00 to 6.42 and from 0.00 to 0.94 mg-N m^{-2} h^{-1}, respectively. N_2O flux increased markedly after only the first heavy rainfall each year, and it was higher than the N_2O flux that occurred immediately after fertilizer application. This seasonal pattern of N_2O flux from row was similar to the pattern from the furrow, even though no chemical fertilizer was applied to the furrow. The highest N_2O flux was observed after heavy rain, and an increase

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in NO flux was recognized only from the row. Seasonal fluctuations in NO_3^- and NH_4^+ concentrations in soil and in the ratio of N_2O/NO flux suggested that N_2O and NO fluxes occurring after fertilizer application (mid-May to early July) were mainly produced by nitrification and that the N_2O emitted after heavy rain was mainly produced by denitrification. The cumulative N_2O flux during the snow-free season ranged from 0.83 to 2.33 g-N m^{-2} over a three-year period. This flux was relatively high compared with those reported worldwide. In contrast, reported cumulative N_2O fluxes from agricultural Andosols in Japan are typically lower than those from other agricultural soils in Japan and around the world. Therefore, the results of our study suggest that high N_2O emissions may occur from Japanese agricultural Andosols with a shallow ground water level.

The Gray Lowland soil and the Andosol, as described previously, had a different soil structure, especially with regard to the distribution of macropores and cracks. The influence of this difference on the production and emission of N_2O was investigated. N_2O concentration profiles were measured in two soils during the snow-free season, and N_2O flux in the soil through to a depth of 0.3 m was calculated using the gradient method (using Fick's law). This flux was compared with the N_2O flux from the soil to the atmosphere using the chamber method. In the Gray Lowland soil, the N_2O concentration above 0.4 m increased with an increase in soil depth. In the Andosol, there were no distinctive N_2O concentration gradients in the topsoil when the N_2O flux did not increase. However, the N_2O concentration at a depth of 0.1 m increased significantly, and this concentration was higher than the concentration below 0.2 m when the N_2O flux increased significantly. The N_2O concentration profiles were thus different between these two soils. The contribution ratios of the N_2O produced in the top soil (0–0.3 m depth) to the total N_2O emitted from the soil to the atmosphere in the Gray Lowland soil and the Andosol were 0.86 and 1.00, respectively. This indicates that the N_2O emitted from the soil to the atmosphere was produced mainly in the top soil. However, the contribution ratio of the subsoil to the N_2O emitted from the Gray Lowland soil was higher than that of the Andosol. There was a significant positive correlation between the N_2O flux in the soil through to a 0.3 m depth and the flux from the soil to the atmosphere in only the Gray Lowland soil. These results suggest that N_2O production in the subsoil of the Gray Lowland soil could have been activated by NO_3^- leaching through macropores and cracks, and subsequently, the N_2O produced in the subsoil might have been rapidly emitted to the atmosphere through those macropores and cracks. The soil carbon content of subsoil in the Gray Lowland soil was higher than that in the Andosol. Denitrification was prompted by an increase in the soil organic carbon; therefore, it is believed that the high carbon content and macropores in the Gray Lowland soil caused the high concentration of N_2O in the subsoil.

In Japan, the annual N_2O emission from upland fields for various periods ranged from 0.01 to 0.87 g-N m^{-2}, with most measured values being less than 0.1 g-N m^{-2}. About 80 % of the measured N_2O emission worldwide was less than 0.5 g-N m^{-2}. The cumulative emissions in this study from the Gray Lowland soil and the Andosol were relatively high, compared with those reported worldwide. This suggests that the increases of the N_2O flux in the study fields after heavy rain and harvesting were equal to or higher than the increase that occurred immediately after fertilizer application.

The conclusion of this study was showed in the following text. In the Gray Lowland soil and the Andosol, the N_2O emission derived from denitrification, from summer to autumn, was larger than that from the nitrification occurring immediately after fertilizer application. This might be due to the seasonal pattern of rainfall, i.e., no distinct rainy season in the early summer (immediately after fertilizer application) and most rain occurring in September (nearly harvesting). The differences in the seasonal patterns and the amount of N_2O emission between the Gray Lowland soil and the Andosol might be due

to the differences in the depth of the N_2O production in the soil and the N_2O mobility from the soil to the atmosphere. The annual N_2O emissions from both types of soils were relatively high, compared with those reported worldwide.

We investigated the effects of heavy application of composted cattle manure on the leaching of nitrogen from small lysimeter paddies, where forage rice was cultivated from April 2003 to March 2007. Nitrogen leaching increased with manure application when adequate rainfall occurred after the application of cattle manure during investigated period. The amount of nitrogen leaching from the paddy to which 18 kg m^{-2} (18M-plot) manure was applied was higher than this amount from the paddy without manure application (0M-plot). Although the dry matter yield of forage rice increased in the 18M-plot, the losses of nitrogen was high, and the excessive input caused nitrogen to accumulate in the soil. It was determined that heavy application of manure increased the environment load.

5. 結論

灰色低地土と黒ボク土において，施肥後よりも降雨頻度が高まる夏から秋の収穫期前後に大きい土壌から大気へのN_2O放出の季節推移は同様であり，これは梅雨がなく夏から秋に降雨頻度が高まる北海道中央部の降雨パターンの影響を強く受けていた．施肥直後のN_2O放出は主に施肥窒素の硝化に由来し，降雨による水供給や作物残渣による有機物の供給が増加する夏から秋の収穫期前後のN_2O放出は主に脱窒に由来すると考えられた．灰色低地土と黒ボク土ともに，土壌から大気へ放出されたN_2Oの大部分は深さ0.3 mより上で生成されたが，灰色低地土では下層で生成されたN_2Oも粗孔隙を通して大気へ放出されたと推察された．N_2O生成に影響を与える土壌の炭素含有率は作土では灰色低地土よりも黒ボク土で高く，下層では灰色低地土の方が高かった. 脱窒は有機炭素が多いほど進みやすいため，粗孔隙の存在とともに下層の有機炭素含有率が高かったことが，灰色低地土の下層のN_2O濃度を高めた一因と考えられた．本調査2圃場からのN_2O放出は日本国内や世界の測定事例と比べて高い値だった．これは，両圃場ともに排水性が悪い土壌であり，脱窒が起こりやすい環境だったことが原因と考えられた．このように，土壌中の水とガスの移動を支配する土壌構造と排水性はN_2Oの生成・放出に大きな影響を与えたことが明らかとなった．本調査圃場のように施肥後で無い時期に著しいN_2O放出が起こる場合は，施肥後を中心とした短期間の測定ではN_2O放出量を過小評価する危険が高く，N_2O放出量を推定するためには栽培期間を通した測定が必要である．また，畑や水田において，化成窒素肥料の施用や牛ふん堆肥の多量施用は作物の収量を向上させるが，同時に農地からの窒素負荷を増加させる可能性も高いことが明らかとなった．

Ⅸ 摘　要

20世紀において，窒素施肥量の増加に伴い，単位面積当りの収量は飛躍的に向上したが，農地では施肥窒素に由来する一酸化二窒素（N_2O）や一酸化窒素（NO）といったガス態の窒素化合物の発生量の増加や硝酸態窒素等の流出に伴う地下水・河川・湖沼などの水質汚染等の様々な環境問題が浮上している．N_2Oは温室効果ガスの一種であり，長い寿命と高い地球温暖化係数を持つ．気候変動に関する政府間パネル（IPCC）は，N_2O濃度上昇の最大要因は農業と指摘し，農耕地由来のN_2O放出を早急に低減させる必要がある．しかしながら，時間・空間的変動が非常に大きいN_2O放出量の推定は難しく，IPCCの3次報告書で示された人為起源のN_2O放出量は2–21 Tg-N yr^{-2}の大きな幅があり，N_2O放出量の実測値の積み上げとN_2O放出要因の解明を進めて，推定精度を上げる必要がある．また，我が国では，食料自給率向上のため水田での飼料用イネの作付けが奨励されており，水田への牛ふん堆肥の還元量が増えている．従って，牛ふん堆肥の多量施用による水田からの窒素流出の増大が懸念される．さらに，土壌から溶脱する窒素はN_2Oの間接放出量の増加につながる．これより本研究では，北海道中央部の灰色低地土と黒ボク土の畑地において，無積雪期の土壌から大気へのN_2Oフラックス等を3-6年間測定し，N_2O放出の季節推移やこれに対する気象および土壌環境の影響を明らかにし，無積雪期のN_2O放出量を見積もることと牛ふん堆肥を多量施用した飼料用イネ栽培水田からの窒素浸透流出量を明らかにすることを目的とした．

1995-2000年の無積雪期（4-11月）に，灰色低地土においてN_2Oフラックスと関連要因を測定した．土壌から大気へのN_2Oフラックスは0.0–1.9 mg-N m^{-2} h^{-1}の範囲で，施肥区では毎年5月の施肥直後と同等かそれ以上のN_2Oが，降雨頻度が高まる収穫期前後に放出され，一酸化窒素（NO）フラックスは施肥直後のみに上昇した．無施肥区ではNOおよび施肥直後のN_2Oフラックスの上昇はなく，収穫期前後にN_2Oフラックスが上昇した．土壌中の無機態窒素濃度およびフラックスのN_2O/NO比より，施肥直後のN_2O生成は主に硝化，収穫期前後のN_2O生成は主に脱窒由来だと考えられた．収穫期前後のN_2Oフラックスはタマネギの植栽によって上昇し，N_2Oと二酸化炭素（CO_2）フラックスの間には時系列および空間変動ともに有意な正の関係

が得られたため，降雨による土壌への水供給と，タマネギ由来の有機物が N_2O 生成を促進したと考えられた．各年の無積雪期の N_2O 放出量は 0.4–1.6 g-N m^{-2} であり，この 70 % は 8–10 月に放出された．従って，施肥後中心の N_2O フラックスの測定では放出量を過少評価する可能性がある．

1998–2000 年の無積雪期において，黒ボク土からの N_2O フラックスと関連要因を調査した．N_2O フラックスの範囲は 0.0–6.4 mg-N m^{-2} h^{-1} であった．化学肥料の有無に関わらず，N_2O フラックスは毎年最初の大雨直後に一度だけ大きく上昇し，これは施肥直後よりも大きかった．NO フラックスは施肥後に化学肥料を施用した株間でのみ上昇した．土壌の無機態窒素濃度とフラックスの N_2O/NO 比から，施肥直後に放出された N_2O は硝化由来，大雨後に放出された N_2O は脱窒由来と推定された．各年の無積雪期の N_2O 放出量は 0.8–2.3 g-N m^{-2} であった．これまで比較的 N_2O 放出が小さいと言われている日本の黒ボク土でも，本圃場のように地下水位が高い条件では，大きな N_2O 放出が起こる可能性がある．

これまで検討してきた灰色低地土と黒ボク土は土壌中の水やガスの移動に大きく関与する粗孔隙の発達程度が異なる土壌であり，この違いが N_2O の生成放出メカニズムに与える影響を検討した．無積雪期に土壌中の N_2O の濃度分布を調査し，深さ 0.3 m を通過する N_2O フラックスを拡散法により求め，チャンバー法で測定した土壌から大気へのフラックスと比較した．下層の N_2O 濃度は黒ボク土よりも灰色低地土で高く，これが二つの土壌から大気への N_2O 放出の推移と量の違いの原因だと考えられた．両土壌ともに，土壌から大気へ放出された N_2O の大部分は深さ 0.3 m より上で生成されていたが，灰色低地土では下層で生成された N_2O も粗孔隙を通して大気へ放出されたと推察された．灰色低地土の下層の N_2O 濃度上昇の原因には，降雨時に粗孔隙を通って水とともに下層に輸送された NO_3^- が脱窒を受けたことが考えられる．脱窒は有機炭素が多いほど進みやすく，灰色低地土では粗孔隙の存在とともに下層の土壌炭素含量が黒ボク土よりも高かったことが，下層の高い N_2O 濃度の一因と考えられた．

既往の作物栽培期間の積算 N_2O 放出量の測定事例（測定期間が 90 日以上 1 年以内）と比較すると日本の事例の大部分は 0.1 g-N m^{-2} 以下，世界の測定事例の 80 % 程度は 0.5 g-N m^{-2} 以下で，これらと比べて本圃場の N_2O 放出量は高かった．これは，両圃場ともに，多くの事例で測定期間の最大 N_2O 放出が認められた施肥後以外の時期に，施肥後と同等かそれ以上の N_2O が放出されたためだと考えられた．

飼料イネ栽培水田では牛ふん堆肥 18 kg m^{-2} の施用により，堆肥散布後から 7 月頃まで牛ふん堆肥由来の窒素が浸透水経由で流出したと推察された．その後は堆肥施用の有無に関わらず，窒素浸透流出の季節推移は同様であったが，流出量は調査期間を通して無施用区よりも牛ふん堆肥施用区で大きかった．牛ふん堆肥 18 kg m^{-2} を施用すると窒素収支は投入過剰となり土壌の全窒素および可給態窒素濃度の推移から，牛ふん堆肥に由来し土壌に蓄積した窒素が浸透水経由で 4 年間に渡り流出したと考えられた．

以上のように，北海道中央部の灰色低地土と黒ボク土では，北海道の降雨パターンの影響を受けたため，施肥直後の硝化よりも夏から秋の脱窒由来の N_2O 放出が大きく，粗孔隙の有無による土壌中の N_2O 生成深度と大気への移動性の違いが，二つの土壌からの N_2O 放出の季節推移や量の違いの原因であったこと，本調査地の無積雪期の N_2O 放出量は世界的にみても大きかったこと，化成肥料や牛ふん堆肥による多量の窒素施肥は土壌から大気への N_2O・NO 放出や浸透水経由での窒素流出を増加させたことを明らかとした．