

B-16 地球温暖化抑制のための  $\text{CH}_4$ 、 $\text{N}_2\text{O}$  の対策技術開発と評価に関する研究

(1) 農耕地から放出されるメタンと亜酸化窒素の発生抑制技術に関する研究

②窒素施肥土壌から放出される亜酸化窒素などの窒素化合物の発生抑制技術の開発に関する研究  
(国際交流研究)

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[要旨]

大気中に約 310ppb 存在する亜酸化窒素( $\text{N}_2\text{O}$ )は、温室効果ガスの一つであり、また成層圏のオゾン層を破壊する物質である。農耕地からの  $\text{N}_2\text{O}$  の発生量は地球全体からの全発生量の約 20%と推定されているが、アジアでのデータが少なく不確実性は非常に大きい。さらに施肥した畑土壌からは、 $\text{N}_2\text{O}$  だけでなく、一酸化窒素( $\text{NO}$ )も大気中に放出されることがわかってきた。 $\text{NO}$  は、やはり温室効果ガスの一つである対流圏オゾンの前駆物質であり、また酸性雨の原因物質の一つである。現在、これらの窒素酸化物の施肥土壌からの発生量と発生原因、およびこれらのガス発生抑制技術を開発することは、緊急で重要な課題である。

1996 年度の研究目的は、異なる窒素肥料あるいは緩効性窒素肥料を使用することによって、日本の畑土壌（黒ボク土）からの亜酸化窒素 ( $\text{N}_2\text{O}$ )の発生量の抑制効果を、室内実験から評価することであった。室内での土壌培養実験は、硝化作用あるいは脱窒作用に好都合な範囲の水分条件（土壌孔隙の水分飽和度 (WFPS) が、40、55、70、85%）のもとで、一定温度(25℃)で 50 日間実施した。 $\text{N}_2\text{O}$  の発生は、通常の硫酸アンモニウム (AS)肥料区のほうが硝酸カルシウム(CN)肥料区よりも非常に多かったため、消化作用が  $\text{N}_2\text{O}$  の主要な発生過程であると思われた。土壌水分量が増加すると、すべての処理区からの  $\text{N}_2\text{O}$  の発生量も増大し、WFPS が 85%では 40%の場合よりも 20 倍以上も大きかった。AS あるいは燐酸アンモニウム肥料(AP)を緩効性被覆肥料の形態(S-AS と S-AP)で施用した場合、通常の AS と比較すると、 $\text{N}_2\text{O}$  の発生量は WFPS が 40%と 85%の処理区では減少したが、WFPS が 55%では差がなく、また 70%では S-AS の処理区だけが非常に少なかった。S-AS あるいは S-AP を施用すると、 $\text{N}_2\text{O}$  の発生の最大ピークは、 $\text{NH}_4^+\text{-N}$  がゆっくり放出されたために、時期が遅くなってしかも値が減少したが、放出された  $\text{NH}_4^+\text{-N}$  が硝化されるので、培養実験のほとんどの期間中  $\text{N}_2\text{O}$  は発生し続けた。CN を緩効性被覆窒素肥料(S-CN)の形態で施用すると、硝化作用に利用できるため  $\text{NH}_4^+\text{-N}$  の量が非常に少なくなるので、 $\text{N}_2\text{O}$  の発生量は ZR 区からの発生量よりも多くならなかった。これらの結果から、今回の実験の

ように硝化の起こりやすい条件の下では少なくとも、硝酸態の窒素肥料、とくに緩効性被覆窒素肥料の形態で選択的に使用することは、農耕地からの  $\text{N}_2\text{O}$  の発生ポテンシャルを非常に減少できることが、証明された。

1997 年度の研究目的は、肥料の違い（化学肥料と家畜ふん尿）、および施肥法の違い（全面全層施肥と溝施肥）によって  $\text{N}_2\text{O}$  と  $\text{NO}$  の発生がどのように異なるかを明らかにすることであった。そこで、黒ボク土の畑土壌でハクサイを栽培し、アンモニア系の化学窒素肥料と発酵豚ふんを施用して、大気中への  $\text{N}_2\text{O}$  と  $\text{NO}$  のフラックスを、1997 年 9 月～12 月に測定した。施肥法は各処理区で全面全層施肥と溝施肥とを行い、さらに対照として無窒素区を設定した。フラックス測定はクロードチャンバー法で週に 1～3 回行い、また、土壌の水分量、pH、地温、 $\text{NO}_4^-$  及び  $\text{NO}_3^-$  濃度を随時測定した。 $\text{NO}$  のフラックスは、施肥直後に最大となり数日後には急激に減少した。 $\text{NO}$  フラックスが減少し始めるとともに  $\text{N}_2\text{O}$  フラックスが大きくなり、数日後には最大となり、その後は急激に減少した。土壌中の  $\text{NO}_4^-$ 、 $\text{NO}_3^-$  濃度の分析結果から、これらのガスの発生はおもに硝化作用によると推測された。全調査期間中の  $\text{NO}$  の発生量は、化学肥料および発酵豚ふん区ともに、溝施肥区の方が全面全層施肥区よりも少なかった。しかし  $\text{N}_2\text{O}$  の発生量は、化学肥料区では  $\text{NO}$  と同様に溝施肥区のほうが少なかったが、発酵豚ふん区では、溝施肥区の方が全面全層施肥区より多かった。また、土壌水分量は  $\text{N}_2\text{O}$  と  $\text{NO}$  の発生に影響を与えていることがわかった。今回の実験から、溝施肥法は、全面全層施肥法よりも、発酵豚ふんからの  $\text{N}_2\text{O}$  を除けば、 $\text{N}_2\text{O}$  と  $\text{NO}$  の発生を抑制する方法として有効である可能性が示された。

「キーワード」 亜酸化窒素、一酸化窒素、発生抑制技術、施肥方法、緩効性被覆窒素肥料

**B-16 Studies on Development and Evaluation of the Technology for Reducing CH<sub>4</sub> and N<sub>2</sub>O Emissions to Prevent Global Warming**

**B-16.1 Studies on Development of Reduction Techniques for Methane and Nitrous Oxide Emissions from Agricultural Fields**

**B-16.1.2 Development of Techniques for the Reduction of Nitrous Oxide Emissions from Nitrogen Fertilized Upland Soils**

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**Abstract:**

(1) A laboratory experiment was performed to evaluate the effectiveness of altering the form of applied fertilizer nitrogen and/or the use of controlled-release fertilizers in reducing emissions of nitrous oxide (N<sub>2</sub>O) from an upland Japanese soil incubated in the laboratory for 50 days at 25 °C. Nitrification appeared to be the main source of N<sub>2</sub>O with much greater emissions from ammonium sulphate (AS) than from calcium nitrate (CN). Applying ammonium sulphate or ammonium phosphate in the form of controlled release fertilizers (S-AS and S-AP) reduced emissions in the 40% and 85% wfps treatments compared with normal AS, but at 55% wfps there was no difference while at 70% only the S-AS treatment was significantly lower. The results demonstrate that, at least under conditions similarly conducive to nitrification, the selective use of nitrate fertilizer, especially in a controlled release form can greatly reduce potential emissions of N<sub>2</sub>O from agricultural soil.

(2) A field experiment on N<sub>2</sub>O and NO emission was performed in agricultural plots where Hakusai was cultivated, applying organic fertilizer (swine manure) and chemical fertilizer (ammonium phosphate) in two different ways, i.e. by broadcasting and band application. The measurements were

carried out from the day prior to fertilizer application until one month after harvest. A number of soil parameters like pH, soil moisture, soil temperature, soil ammonium and soil nitrate have also been measured. The experiment resulted in significantly reduced NO emissions in both band applied organic and chemical fertilizer plots. The N<sub>2</sub>O emissions also seemed to be retarded in band applied chemical fertilizer plot compared to the broadcast applied chemical fertilizer plots. However, in organic fertiliser plots, N<sub>2</sub>O emission was higher in band applied organic fertilizer plot than in that of broadcast applied organic fertilizer plot. The soil moisture also seems to have affected the N<sub>2</sub>O and NO emissions.

**Keywords:** nitrous oxide, nitric oxide, application method of fertilizer, controlled-release fertilizer, reduction of emission

## **1. Introduction:**

Nitrous oxide (N<sub>2</sub>O) makes a significant contribution to global change. It is a “greenhouse gas”, accounting for approximately 5% of the total greenhouse effect<sup>1)</sup> and it is also involved in the destruction of stratospheric ozone. Soils are the major source of N<sub>2</sub>O, accounting for 65% of total global emissions<sup>2)</sup> the microbial processes of nitrification and denitrification are responsible, and emissions increase with increasing availability of NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> substrates, e.g. following the application of fertiliser nitrogen.

Nitric oxide (NO) in the atmosphere is another important constituent of global nitrogen cycle and soil is among the major sources of its emission into the atmosphere. Nitric oxide has a short residence time in the atmosphere but has a significant bearing in the atmospheric chemistry. It is a precursor of tropospheric ozone, which is a greenhouse gas and has deleterious effects on human and vegetation. The same microbial activities are responsible for the production of NO in the soil.

## **2. Objectives:**

### **2.1 Effects of the use of different forms of fertiliser N on N<sub>2</sub>O emission**

The aims of this laboratory study were to determine the effect of varying the form of nitrogen applied to a local Japanese andosol soil under a range of moisture conditions likely to favour either nitrification or denitrification. In addition NH<sub>4</sub>-N and NO<sub>3</sub>-N were applied in easily soluble and controlled release fertiliser forms to investigate whether N<sub>2</sub>O emissions can be reduced by controlling the release of mineral nitrogen to the soil, and whether the emissions of N<sub>2</sub>O from controlled release fertiliser are also significantly influenced by soil moisture.

### **2.2 Effects of different application modes of organic and chemical fertilizers to agricultural fields on N<sub>2</sub>O and NO emissions**

The modification in agricultural practices can also be a potential abatement technology for reduction of N<sub>2</sub>O emissions, however, this option has not been addressed so far sufficiently. The purpose of this field study is to make clear the effects of different application modes of organic and chemical fertilizers in agricultural field on N<sub>2</sub>O and NO emissions.

## **3. Effects of the use of different forms of fertiliser N on N<sub>2</sub>O emission**

### **3.1 Materials and Methods**

An upland andosol soil was collected from a field site at the National Institute of Agro-Environmental Sciences in Tsukuba, Japan. A bulk sample from the surface 15 cm of this soil was

sieved (2-mm mesh) and air-dried before storage at 5°C until needed. The following six fertiliser nitrogen treatments were studied: three “Nutricote Type 40” controlled-release fertilisers with the nitrogen in the form of ammonium sulphate (S-AS), ammonium phosphate (S-AP) and calcium nitrate (S-CN) [Chisso Asahi Co. Ltd. Tokyo, Japan], also easily soluble ammonium sulphate (AS) and calcium nitrate (CN) and an unfertilised control (ZR). The “Type 40” controlled-release fertilisers means that at a constant temperature of 25 °C 80% of the applied nitrogen will be released over 40 days. Each treatment was studied at 4 moisture contents corresponding to soil water filled pore space (wfps) values of 40%, 55%, 70% and 85%, with a total of 15 replicate samples for each treatment. Due to the large number of samples the incubation experiment was carried out in two batches; in the first run soils were incubated at 40% and 70% water-filled pore space (wfps), followed by a second run with soils incubated at 50% and 85% wfps. All soils were incubated at 25°C for 50 days, to allow a calculated release of approximately 90% of the total nitrogen applied in the 3 controlled-release fertiliser treatments. Gas samples were taken from the headspace of each of 3 replicate flasks every 3-4 days and analysed for N<sub>2</sub>O by electron capture gas chromatography (all treatments) and for CO<sub>2</sub> by thermal conductivity gas chromatography (55% and 85% wfps only). Periodic measurements were made on 3 replicate samples for soil moisture, soil mineral N (by continuous flow colorimetric analysis) and soil pH<sub>(CaCl2)</sub> with samples then discarded. A more detailed account of the experimental procedure has been reported elsewhere<sup>3</sup>.

## 3.2 Results and Discussion

### (1) Effect of fertiliser form and soil moisture on cumulative N<sub>2</sub>O emissions

Emissions from the controlled-release ammonium fertilisers (S-AS and S-AP) were similar except at 70% wfps where emissions from S-AP were higher. The use of these two controlled-release fertilisers reduced emissions compared with those from the easily soluble NH<sub>4</sub>-N treatment (AS) at 40% and 85% wfps. At 70% wfps only S-AS was significantly lower, while there was no difference at 55% wfps. Emissions from CN and S-CN were low, never significantly higher than ZR, except CN at 85% wfps. This indicated that nitrification was the main source of emissions. Also the period of high N<sub>2</sub>O emissions from AS matched the time required to complete the nitrification of the applied NH<sub>4</sub>-N (see section 3. 2. (3)) also indicating nitrification as the main source of emissions.

### (2) Changes in emissions of N<sub>2</sub>O with time

More than 90% of all N<sub>2</sub>O emissions from AS occurred in the first 17 days, before emissions declined rapidly to near background levels (Figure 2a). Maximum emission rates at 70% and 85% wfps were >7 times higher than those at 55% wfps. There was a similar rapid increase in emissions with CN, but only at 85% wfps. In all the fertiliser treatments at 85% wfps there was an initial rapid increase in N<sub>2</sub>O emissions, including the unfertilised control (ZR), indicating that some emissions were derived from mineral nitrogen already present in the soil at the start of the experiment. Only in AS and CN, where easily soluble N was immediately available, were the emissions significantly higher than from ZR. With the exception of the initial peak at 85% wfps, emissions from S-AS and S-AP increased steadily over time reaching a maximum after 14-17 days (Figure 2c, d). Emissions then declined, but more slowly than in AS, with significant emissions occurring until at least day 30 in all treatments, except for 40% wfps where emissions remained low throughout. The use of S-AS and S-AP reduced the rate of maximum emissions and also tended to delay it for a few days. However, particularly at 55% wfps, it

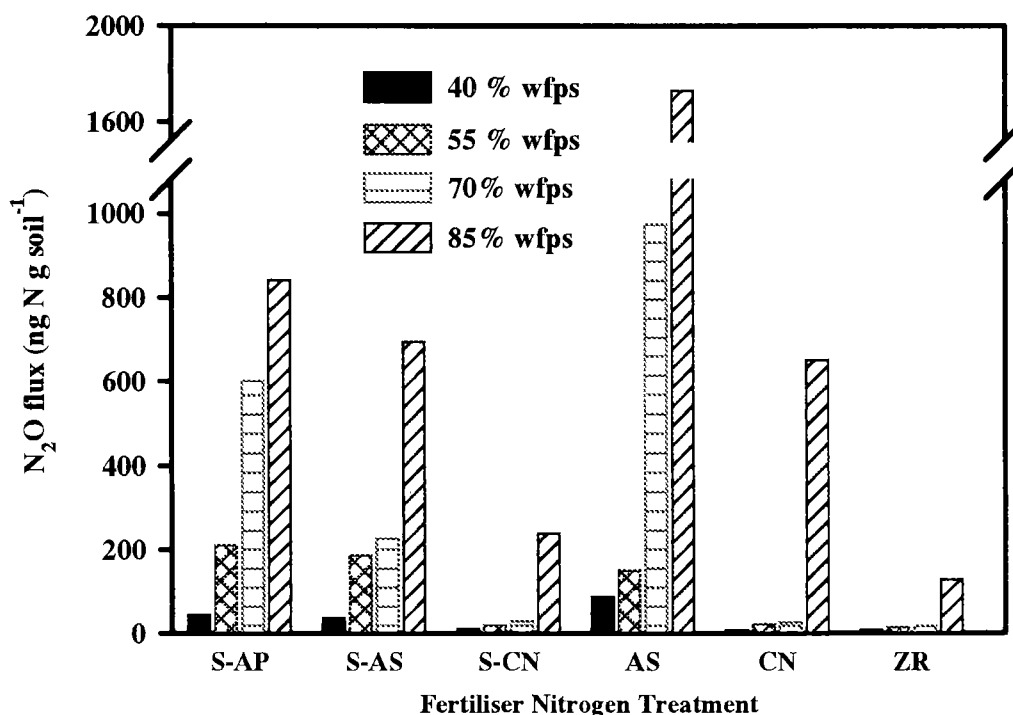


Figure 1. Cumulative emissions of N<sub>2</sub>O 49 days after the application of different forms of fertiliser nitrogen to an upland soil under different soil moisture conditions; S-AS: controlled-release ammonium sulphate, S-AP: controlled-release ammonium phosphate, S-CN: controlled-release calcium nitrate, AS: ammonium sulphate, CN: calcium nitrate, ZR: unfertilised control, wfps: water filled pore space (Note break in y-axis and change of scale).

appeared that as the NH<sub>4</sub>-N was released into the soil it was quickly nitrified and therefore the cumulative flux over the whole period was not reduced.

Tsuruta<sup>4</sup> reported that controlled-release urea reduced emissions of N<sub>2</sub>O by 25% compared with normal urea. However, apart from an initial high flux immediately after fertiliser application, Tsuruta reported that emissions from normal urea were no higher than from controlled-release urea. Other results showing a similar pattern have found no reduction in total N<sub>2</sub>O emissions after applying controlled-release urea<sup>3</sup>.

Emissions from S-CN followed a similar pattern to S-AS and S-AP at 85% wfps, but at a much lower rate; at all other moistures the emissions remained very low. The lowest total emission rate was from S-CN where all the applied N was in the form of NO<sub>3</sub>-N. Therefore by minimising the NH<sub>4</sub>-N substrate available to microbial nitrifiers emissions were reduced by 90% compared with those from AS. This reduction was maintained even at 85% wfps, where some denitrification might have been expected. It was only at 85% wfps that emissions from CN were significantly higher than from S-CN, due almost entirely to the initial brief period of high emissions. The high emissions in the CN treatment indicated that some denitrification did occur, but by using S-CN such emissions were reduced due to the controlled release of the applied N. Tsuruta et al<sup>6</sup>, Minami<sup>7</sup> reported that the use of S-CN reduced emissions by nearly 80% compared with AS, mainly by preventing large peaks in emissions immediately after fertiliser application.

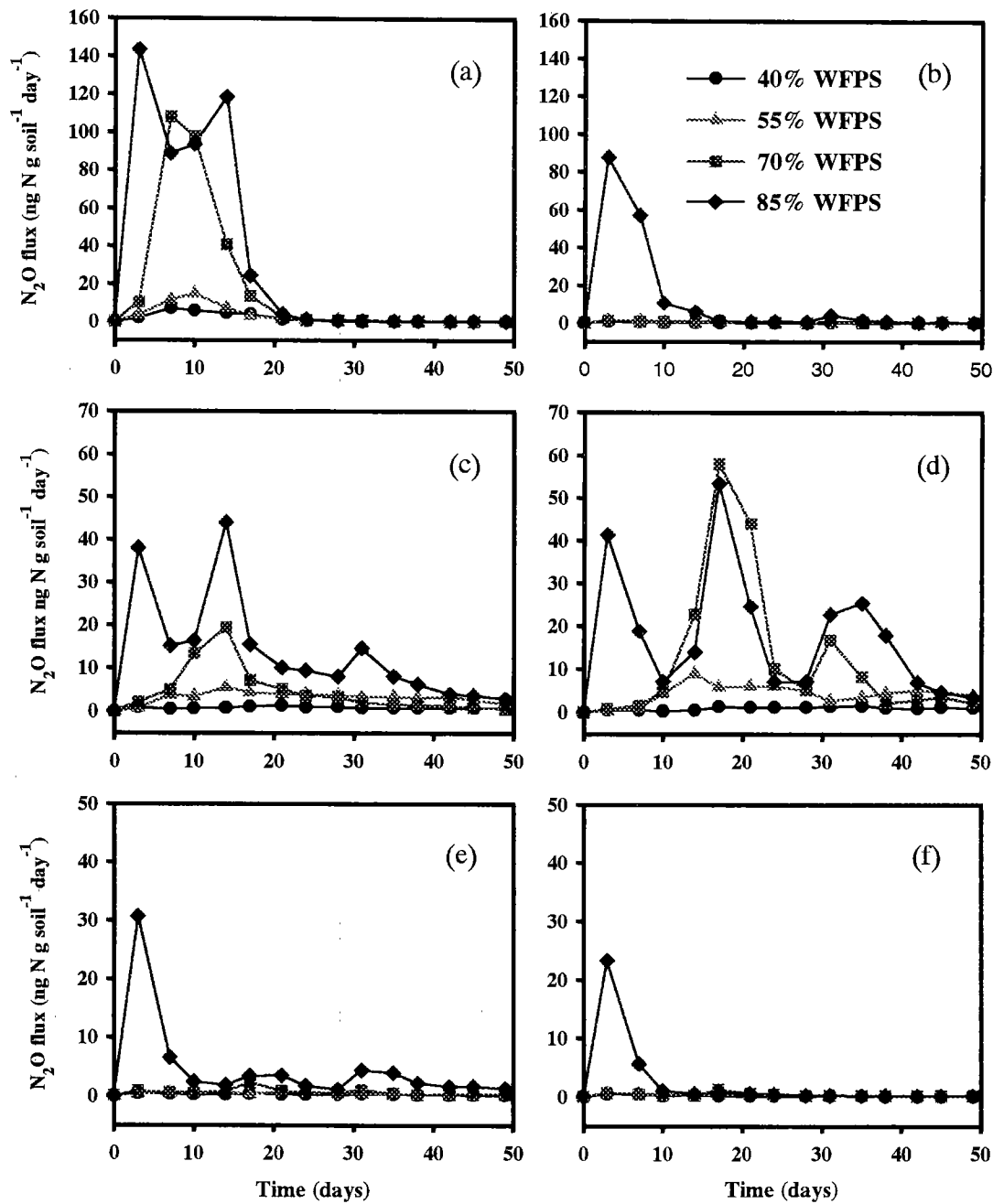


Figure 2. Daily emissions of  $N_2O$  following the application of different forms of fertiliser nitrogen to an upland soil under different soil moisture conditions; (a) ammonium sulphate, (b) calcium nitrate, (c) controlled release ammonium sulphate, (d) controlled release ammonium phosphate, (e) controlled release calcium nitrate, (f) unfertilised control. (Note different scales on y-axes).

### (3) Soil mineral nitrogen

Nitrification in AS rapidly converted all applied  $NH_4-N$  to  $NO_3-N$  except at 40% wfps where complete nitrification was delayed by more than one week longer (Figure 3). The release of mineral nitrogen from S-AS and S-AP increased steadily with time at all moisture contents, but the rate was slower at 40% wfps. The percentage of released N which was nitrified at 40% wfps increased gradually

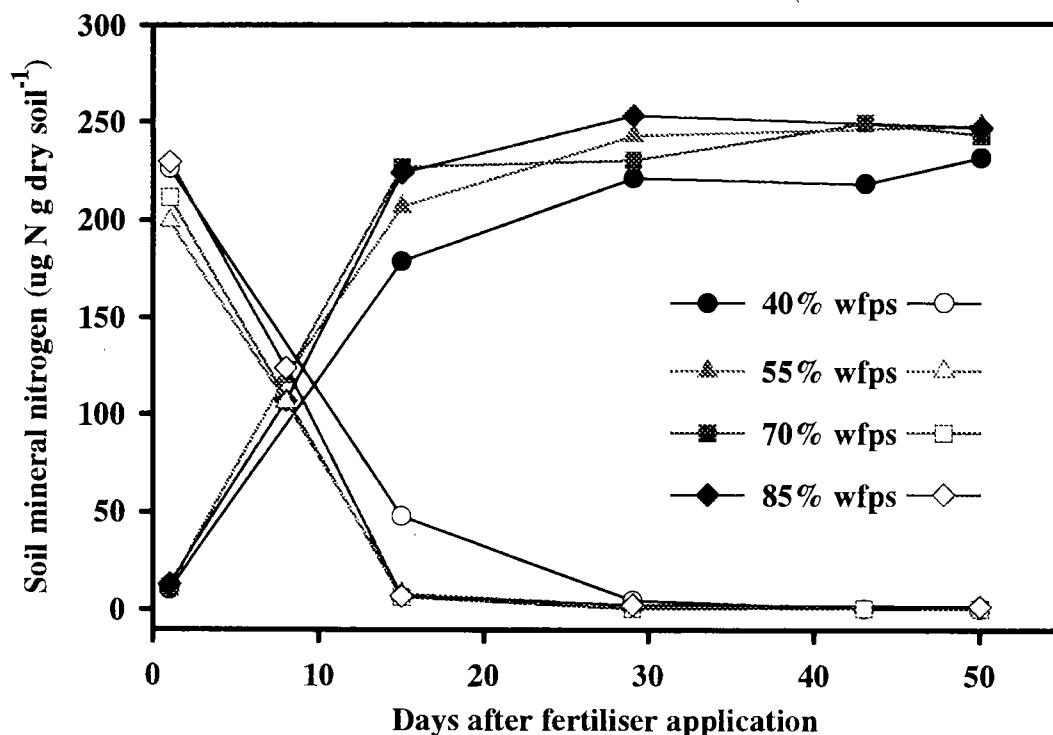


Figure 3. Changes in soil mineral nitrogen over 50 days following the application of ammonium sulphate to an upland soil under different moisture conditions. (closed symbols represent nitrate-N, open symbols represent ammonium-N).

with time reaching around 70% of the released  $\text{NH}_4\text{-N}$  by day 50 compared with values of over 90% by day 29 in the other moisture treatments. The release of  $\text{NO}_3\text{-N}$  from S-CN increased more quickly than in S-AS and S-AP. One potential problem with the use of  $\text{NO}_3\text{-N}$  fertiliser is the risk of significant leaching of  $\text{NO}_3\text{-N}$  down the soil profile. However, given the rapid nature of the nitrification of applied  $\text{NH}_4\text{-N}$  and the controlled release of  $\text{NO}_3\text{-N}$  from S-CN it is unlikely that this problem would be increased.

### 3.3 Conclusions

These results show that where nitrification is the main source of  $\text{N}_2\text{O}$ , emissions can be effectively reduced by using  $\text{NO}_3\text{-N}$  fertiliser, especially in a controlled-release form. Controlled-release  $\text{NH}_4\text{-N}$  fertilisers did not reduce  $\text{N}_2\text{O}$  emissions compared with normal  $\text{NH}_4\text{-N}$  fertiliser under the moisture conditions most likely to be encountered under field conditions in Japan. However, further research will be required to determine if the effects would be the same under different soil and environmental conditions where denitrification could be a more important source of  $\text{N}_2\text{O}$  emissions.

## 4. Effects of different application modes of organic and chemical fertilizers to agricultural fields on $\text{N}_2\text{O}$ and NO emissions

### 4.1 Materials and Methods

A field experiment was carried out on five plots in an agricultural field of National Agricultural



Research Center, Tsukuba. For the preparation of plots, two different modes of fertilizer application, i.e. broadcast and band applications were used. In the organic fertilizer plots and chemical fertilizer plots, swine manure and ammonium phosphate, respectively were applied. Out of the five plots, non-nitrogen fertilizer was applied in one plot (CNTR plot). In the second plot, organic fertilizer was applied by broadcasting (BROF plot), while in third plot organic fertilizer was applied in band application mode (BAOF plot). Similarly, the fourth and fifth plots were prepared by applying chemical fertilizer in broadcast (BRCF plot) and band (BACF plot) modes, respectively. However, an equal amount of nitrogen ( $25\text{gN m}^{-2}$ ) was applied to all the fertilizer plots. In the broadcast plots, the fertilizer was spread uniformly over the surface, and then mixed with the underlying soil using mechanical device capable of mixing the soil down to a depth of 10cm. In the band plots, initially trenches were prepared of 10cm depth and a width of 8cm at the bottom and 25cm at the top. The distance between the edges of two adjacent trenches was 45cm. In these trenches, fertilizer was applied which was then covered with soil. After that, soil and fertilizer in the trench areas were mixed together using the same mechanical device. The fertilizers were applied to the plots on September 11, 1997 and on the next day (i.e. September 12, 1997) the plants of Hakusai (Chinese Cabbage) were transplanted in these plots. The distance between two rows of plants was 70cm while the distance between two adjacent plants in the same row was about 30cm. After the plantation on September 12, 1997, two frames made of white PVC (poly-vinyl chloride) with 3cm deep edges were fixed almost in the center of each plot. These were fixed in such a way that each of them covered two plants. These frames remained fixed in the plots throughout the measurement period and were used as bases for chambers. The method of sampling and analysis has been reported elsewhere<sup>8</sup>.

## **4.2 Results and discussion**

### **(1) Daily fluxes of NO and N<sub>2</sub>O in a cultivating period**

The average values of replicated measurements of NO and N<sub>2</sub>O fluxes for CNTR, BROF, BAOF and BACF plots have been shown in Figures 4A to 4E and 5A to 5E. The difference between replicate measurements have been shown as error bars in these figures. In general, the NO fluxes were highest on the day after the application of fertilizers in all the plots and dropped down to near background levels quickly after one to two weeks. After that, the NO fluxes remained very low in almost all the plots throughout the measurement period. The N<sub>2</sub>O fluxes in all the four fertilized plots showed increasing trends after the fertilizer application and attained maximum values after 7 days in both BROF and BAOF plots and after 9 days in BRCF plots. After that, the N<sub>2</sub>O fluxes gradually decreased to near background levels after about one month in all these four plots. The N<sub>2</sub>O fluxes showed a small increase after harvesting for a few weeks and then returned back to near background levels. This increase in fluxes is to be expected due to the decomposition of plant residues in the soil and soil disturbance caused by harvesting. The CNTR plot showed very small fluxes of NO and N<sub>2</sub>O.

### **(2) Fluxes of NO and N<sub>2</sub>O in different application modes of fertilizer**

The NO fluxes were substantially less in the plots having band mode of fertilizer application compared to the corresponding broadcasting mode application. This also occurred with N<sub>2</sub>O in the chemical fertilizer plots, but in case of organic fertilizer plots, the BAOF plot showed higher fluxes than the BROF plot. This is primarily due to the higher N<sub>2</sub>O flux shown by the BAOF plot in the first few days after fertilizer application. The higher soil moisture during that time could be responsible for these

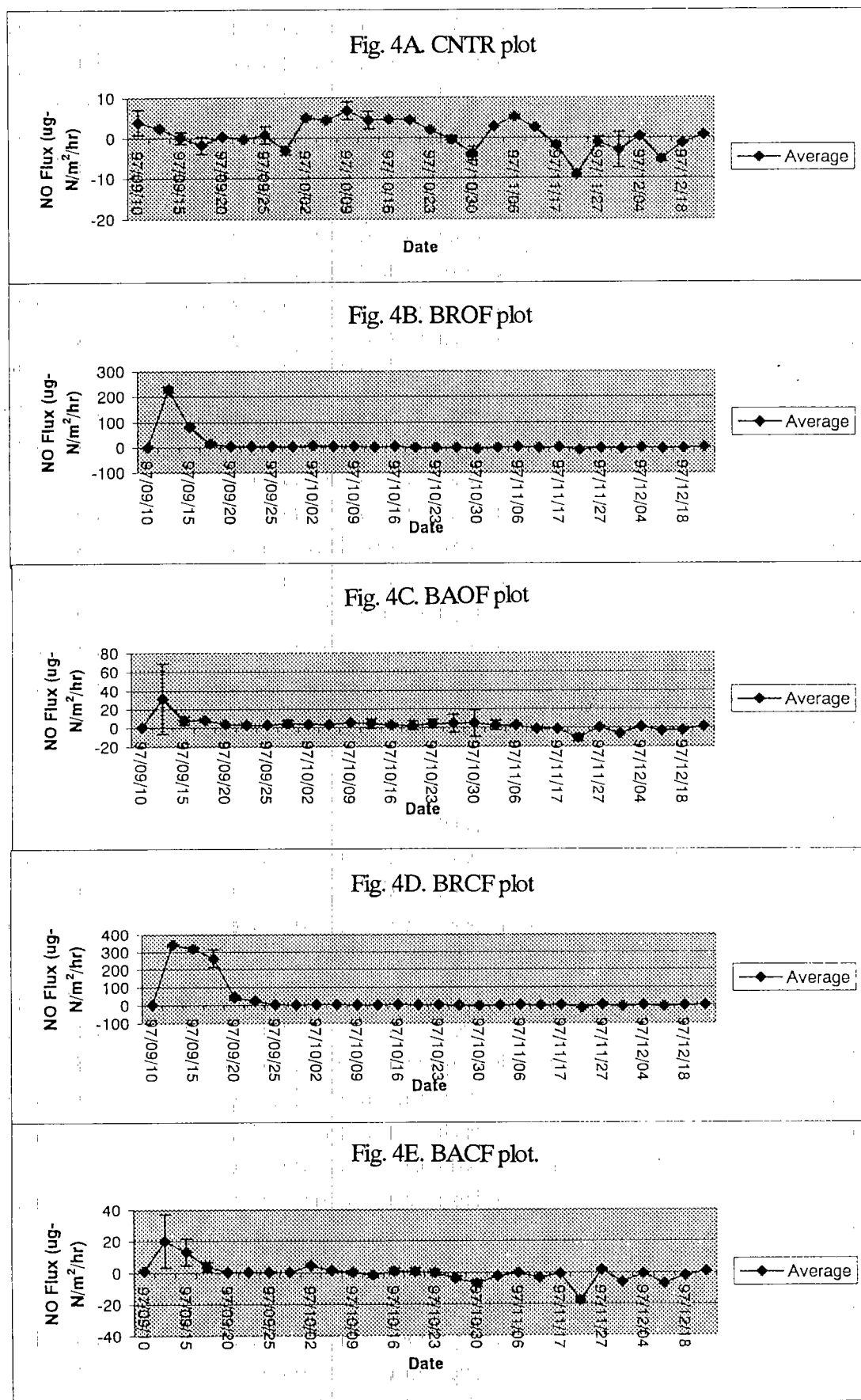


Figure 4. Variation in NO fluxes during a measurement period in (A) CNTR plot, (B) BROF plot, (C) BAOF plot, (D) BRCF plot, and (E) BACF plot.

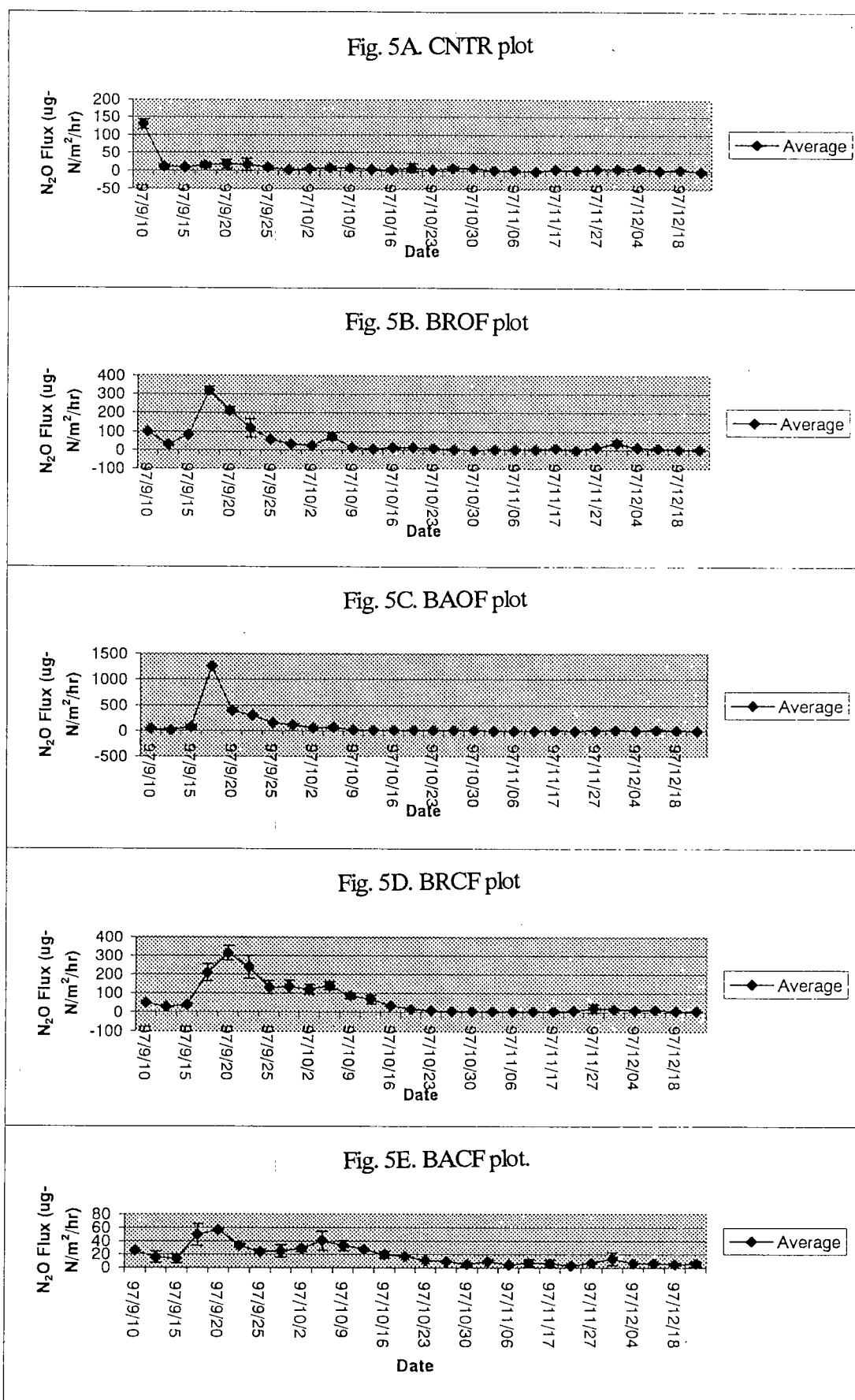


Figure 5. Variation in  $\text{N}_2\text{O}$  fluxes during a measurement period in (A) CNTR plot, (B) BROF plot, (C) BAOF plot, (D) BRCF plot, and (E) BACF plot.

higher fluxes in BAOF plot. In addition the soil temperature in the BAOF plot was also higher than that of the BROF plot, might also have influenced N<sub>2</sub>O emissions.

### (3) Emission ratios of NO-N and N<sub>2</sub>O-N to the total fertilizer N

As the amount of fertilizer covered by the chambers was more in the plots of band application mode compared to those in the plots of broadcast mode application, the ratios of NO-N/total-N and N<sub>2</sub>O-N/total-N for swine manure and chemical fertilizer plots were calculated and shown in Figures 6A and 6B, and 7A and 7B. Figures 6A and 6B clearly show that the NO emissions were much lower in the organic and chemical band applied fertilizer plots compared to the broadcast fertilizer plots. This is in concurrence with the model study made by Hosen et al.<sup>9)</sup>, which shows that NO emissions are retarded if fertilizers are placed below the soil surface. However, the N<sub>2</sub>O emissions also seemed to be retarded in BACF plot compared to BRCF plot (Fig. 7B) which is in contrast to the model calculation which shows no effect of the depth of fertilizer application on N<sub>2</sub>O emissions. A few references are available which show either no effect of the depth of calcium nitrate fertilizer application on the N<sub>2</sub>O production rate<sup>10)</sup> or increased N<sub>2</sub>O production with increased application depth of anhydrous ammonia<sup>11)</sup>. The present study shows, however, a different trend indicating that probably other soil factors are responsible for the emission rate of N<sub>2</sub>O to the atmosphere in BACF plot. In organic fertilizer plots, the emissions of N<sub>2</sub>O were larger in BAOF plot than in the BROF plot.

### 4.3 Conclusions

The emission of NO was significantly reduced in both organic and chemical band applied fertilizer plots compared to broadcasting. The N<sub>2</sub>O emissions also decreased in band applied chemical fertilizer plots compared with broadcasting application. In organic fertilizer plots, however, the N<sub>2</sub>O emission was higher in band application than in broadcast application mode. These results showed that the band application mode has the potential to reduce trace gas emissions to the atmosphere. However, further research is needed to determine if the modification in agricultural practices would reduce not only N<sub>2</sub>O and NO emissions to the atmosphere, but also leaching of nitrogen into groundwater.

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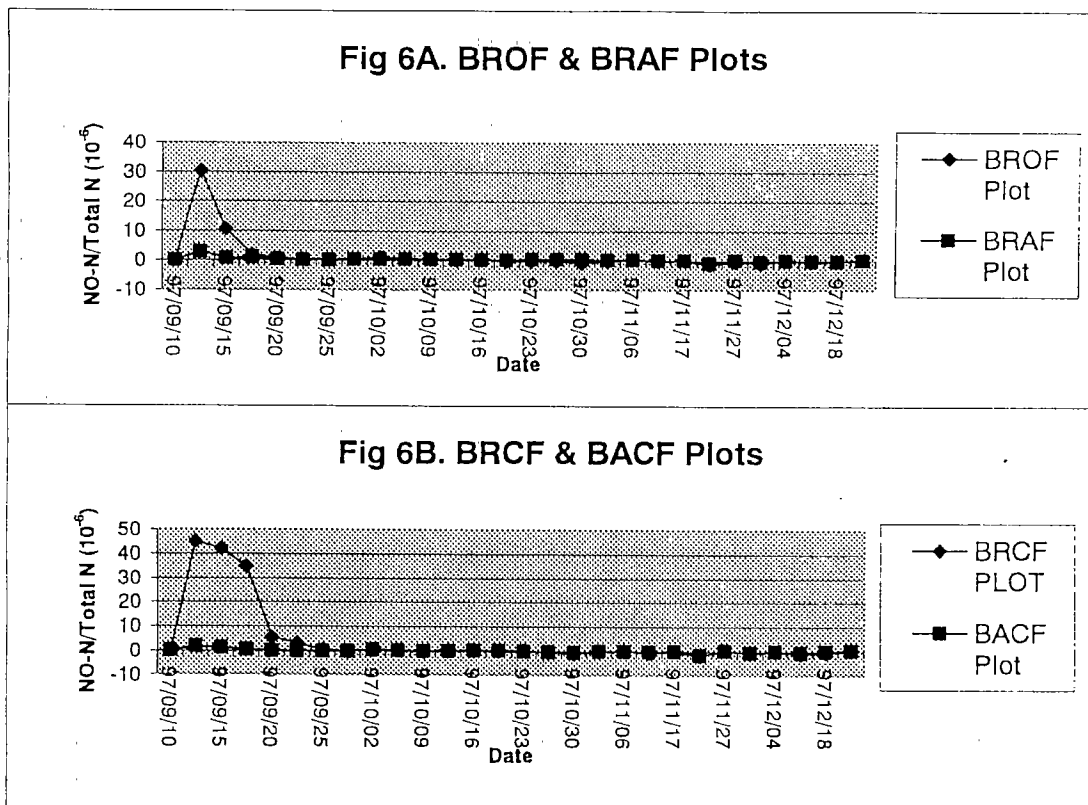


Figure 6. Variation in the ratio of NO-N/total N during a measurement period in (A) BROF and BACF plots, (B) BRCF and BACF plots.

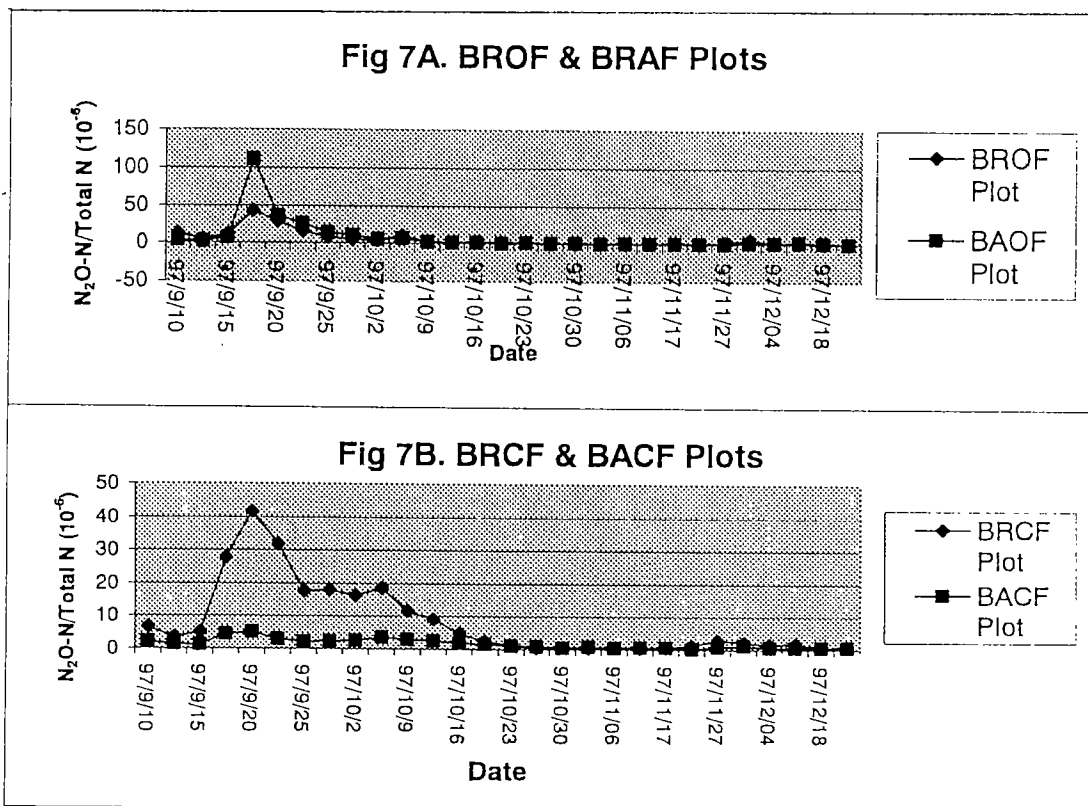


Figure 7. Variation in the ratio of N<sub>2</sub>O-N/total N during a measurement period in (A) BROF and BACF plots, (B) BRCF and BACF plots.

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