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**STABLE ISOTOPE RATIOS OF THE ATMOSPHERIC  
CH<sub>4</sub>, CO<sub>2</sub> AND N<sub>2</sub>O IN TOKAI-MURA**

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Stable Isotope Ratios of the Atmospheric CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O in Tokai-mura

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This report presents the results and interpretation of stable isotope ratios of the atmospheric CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O from a variety of sources in Tokai-mura. The seasonal changes of δ<sup>13</sup>CH<sub>4</sub>, δ<sup>13</sup>CO<sub>2</sub> and δ<sup>15</sup>N<sub>2</sub>O were determined under in-situ conditions in four sampling sites and one control site. Such measurements are expected to provide a useful means of estimating the transport mechanisms of the three trace gases in the environment. These isotopic signatures were analyzed by Isotope Ratio Mass Spectrometer (IRMS, Micromass Isoprime). Our data showed the significant seasonal fluctuation in the Hosoura rice paddy during the entire growing season in 1999. Possible causes for the variation are postulated. Additional measurements on soil properties and on organic δ<sup>13</sup>C in rice plant are suggested. Cited outstanding original papers are summarized in the references.

Keywords: Stable Isotope Ratio, Isotopic Signature, Methane, Carbon Dioxide, Nitrous Oxide, Rice Paddy

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東海村における大気中 CH<sub>4</sub>, CO<sub>2</sub>, N<sub>2</sub>O の  
炭素、酸素、窒素安定同位体比

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(2000年8月25日受理)

本報告書は、環境条件の異なる東海村内数地点において、大気中 CH<sub>4</sub>, CO<sub>2</sub>, N<sub>2</sub>O の炭素、酸素、窒素安定同位体比を測定した結果及び考察をまとめたものである。δ<sup>13</sup>CH<sub>4</sub>、δ<sup>13</sup>CO<sub>2</sub>、δ<sup>15</sup>N<sub>2</sub>O の季節変動は、東海村の 4カ所のサンプリング地点とコントロールサイト 1 地点で調べられた。これらの測定により得られたデータは、上記 3 つの大気中微量ガスの環境中挙動研究に役立てることができる。安定同位体比の測定は、安定同位体比質量分析装置を用いて行われた。1999 年の稻栽培時期に水田において測定したこれらのガスの安定同位体比は、明瞭な季節変動を示し、施肥や湛水、排水、収穫等の作業と関連している。より進んだ研究のためには、稻の有機物中炭素の δ<sup>13</sup>C の測定が必要である。

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## 1. INTRODUCTION

The increasing trend of trace gases in the atmosphere, over the past decades is believed to cause the greenhouse effect in the near future. The greenhouse effect causes the changes in global climate, sea level, and ecosystems [1,2]. The increasing concentrations of atmospheric methane, carbon dioxide, and nitrous oxide are expected to alter the Earth's climate. Therefore, substantial researches have been carried out on measurement of these three radiatively active trace gases to obtain an accurate prediction on the Earth's atmosphere.

Methane is an atmospheric trace gas and one of the important greenhouse gases, has a long-lived residence time of 8-10 years. It is involved in the chemistry of the atmosphere and the Earth's radiation balance [e.g.3]. It is estimated that methane contributes about 15% of the global warming due to trace gases in the atmosphere [4]. Past atmospheric CH<sub>4</sub> observed in air bubbles of polar ice cores dating back to 160,000 years before present was about 0.8 part per million by volumes (ppmv). The current concentration of CH<sub>4</sub> in the atmosphere is about 1.8 ppmv. It is increasing at the rate of about 1% per year which raises the Earth's temperature of about 0.4<sup>0</sup>K over the next 40-50 years [3,5,6,7]. This is evident that population growth and intensified human activities such as fossil fuel used and cultivation have been linked to the increasing rate of the CH<sub>4</sub> flux [7]. The human activities release about 70% of total methane emission [8,9]. Recently, irrigated rice cultivation is considered to be one of the dominant CH<sub>4</sub> sources because the floodwater soils provide suitable conditions for methanogenic bacteria in producing CH<sub>4</sub>. Scheme of methane emission from rice paddy is showed in Fig. 1 [10].

The current CO<sub>2</sub> concentrations in the atmosphere since the industrial revolution have been increased about 80 ppm due to anthropogenic emission. [11,12]. Major anthropogenic emissions come from fossil fuel combustion, changes in land use, and biomass burning. However, about 50% of the CO<sub>2</sub> released by human activity remains in the atmosphere. This indicates that half of the emitted CO<sub>2</sub> are currently absorbed at the surface of the Earth. Both the oceans and the terrestrial ecosystem can absorb CO<sub>2</sub> and store the large quantities of carbon [2,11]. CO<sub>2</sub> is dissolved in the ocean through air-sea exchange processes and is stored in deep water. In terrestrial ecosystem, the transfer of CO<sub>2</sub> from atmosphere is a major component of the global carbon cycle. The storage of carbon in biosphere occurs only if plant photosynthesis exceeds its respiration process, which release carbon dioxide to the atmosphere. Hence, seasonal changes in the global CO<sub>2</sub> flux are resulted from the uptake and release of CO<sub>2</sub> by terrestrial vegetation and soil during photosynthesis and respiration.

The ocean will probably absorb CO<sub>2</sub> for a long time but the balancing of emitted CO<sub>2</sub> may not meet. The biospheric carbon can be re-released through deforestation and represent an important feedback in the complex of global climate. Carbon dioxide stored in the oceans is not likely to reenter the atmosphere as rapidly as storage in the wood and soil of terrestrial ecosystem. The latter is more temporary. However, the oceans have limited capacity to take up carbon.

It is known that the carbon dioxide in oceans that enter from the atmosphere do not have a  $\delta^{13}\text{C}$  composition significantly different from the atmosphere. The transfer of  $\text{CO}_2$  between atmosphere and oceans proceeds with only small fractionation. The exchange involves about 2‰ isotopic fractionation. Whereas in plant tissues, around 20‰ depleted  $^{13}\text{C}/^{12}\text{C}$  relatively to atmosphere typically exist. This occurs as plant takes up carbon dioxide via photosynthesis in which process  $^{12}\text{CO}_2$  is preferable. The isotopically light carbon or an isotopically enriched carbon is returned from the biosphere to the atmosphere during plant respiration process. It is found that fossil fuel also carries the  $^{13}\text{C}$  signature of photosynthesis because it derives from plant material. As a consequence, observations of the  $^{13}\text{C}/^{12}\text{C}$  ratio of atmospheric  $\text{CO}_2$  help to determine the global carbon budget and a better understanding of the global carbon cycle [2,11,13].

Nitrous oxide also is one of the important greenhouse gases, accounting for 6-8% of the current increase in global warming. It is an absorbent of infrared radiation that is approximately 20 times more effective than  $\text{CO}_2$ . In addition, nitrous oxide involves in the ozone destruction in the stratosphere. Over last two decades, it was found that the  $\text{N}_2\text{O}$  concentration in the atmosphere has increased in the rate of about 0.25% per year. It has risen from 275 ppbv in the preindustrial time to about 311 ppbv at the beginning of 1993 [5,14]. The atmospheric  $\text{N}_2\text{O}$  emission is attributed to many sources such as agricultural soils, natural vegetation soils, aquatic sources, biomass burning, fossil fuel combustion, industrial sources, and traffic. Recently, other sources have been identified including waste incineration, sewage treatment, animal manure and land conversion from forest to agriculture. It is believed that the production and increasing use of N fertilizers in the cultivated soils is one of the main global  $\text{N}_2\text{O}$  emissions. **Figure 2** shows a simplified diagram of nitrogen transfer, removals and losses of the agro-ecosystem [15].  $\text{N}_2\text{O}$  emission from soils come from two microbial processes that are nitrification and denitrification. Many studies confirmed that these processes were accelerated by the input of mineral nitrogen fertilizers including nitrogen input from animal manure and from the cultivation of nitrogen fixing leguminous plants. However, soil water content and temperature, soil texture, aeration, organic carbon and pH are also keys variable factors [5,16,17,18,19,20].

Increased emissions of atmospheric  $\text{CH}_4$ ,  $\text{CO}_2$ , and  $\text{N}_2\text{O}$  are of great concern because these gases change the physical and chemical properties of the atmosphere. However, their global budgets are complex and not well analysed, mainly because of a wide variety of natural and anthropogenic sources. Emissions of  $\text{CH}_4$ ,  $\text{CO}_2$ , and  $\text{N}_2\text{O}$  from region to region are different due to latitudinal location, regional environment and human activities in the area. Isotopic analysis has been proved to be helpful for improving estimation of their behavior and mechanisms occurring within their sources, sinks, and transport. The reason is that various sources have characteristic isotopic ratios and that their reactions processes are associated with isotopic fractionation [ e.g.11,13,21]. Considerable amounts of research involving isotopic analysis of these three atmospheric gases have been carried out. However, current available data are not sufficient to fully understand their processes.

The purposes of this study are (1) to provide the data of stable isotope ratios of atmospheric CH<sub>4</sub>, CO<sub>2</sub>, and N<sub>2</sub>O from a variety of sources in Tokai-mura, especially from Hosoura wetland rice paddy, Ibaraki prefecture where is one of the Japanese rice-growing area; and (2) to determine the transportation mechanism of the three atmospheric trace gases in the sources.

## 2. EXPERIMENTAL METHODS

### 2.1 Studies sites and sampling methods

Sampling was performed on five sites which are, JAERI's main gate, pine wood at JAERI's site, Hosoura rice paddy, near shore the Pacific ocean, and Lab 041 building 3 (control point). Map of Tokai-mura where samples were collected is in Fig. 3.

Gas collector of 100 ml volume was previously evacuated at pressure  $1 \times 10^{-3}$  Torr by vacuum pump (ulvac G50D). Three air samples were collected at the ground level of each site, date and time were recorded. Temperature of ambient air and soil at 5-cm depth at Hosoura rice paddy were measured.

### 2.2 Analysis

The samples were analyzed for isotopic ratios of CH<sub>4</sub>, CO<sub>2</sub>, and N<sub>2</sub>O individually using Isotopic Ratio Mass Spectrometer (IRMS Micromass Isoprime).

## 3. RESULTS AND DISCUSSION

Results of isotope ratios were expressed in the common  $\delta$  notation by  $\delta = (R_{\text{sample}}/R_{\text{reference}} - 1) \times 1000$  ‰ with relative to V-PDB (Vienna Pee Dee Belemnite) for  $\delta^{13}\text{C}$  and V-SMOW (Vienna Standard Mean Ocean Water) for  $\delta^{18}\text{O}$ . A reference value for  $^{13}\text{C}$  and  $^{18}\text{O}$  are 30.04 ‰ and 19.95‰, respectively. Precision for repeated 6 times of the same sample were < 2.0‰.

Summarized data of  $\delta^{13}\text{CH}_4$ ,  $\delta^{13}\text{CO}_2$ , and  $\delta^{15}\text{N}_2\text{O}$  in five sampling sites are in **Tables 1, 2, and 3**, respectively. Note that the  $\delta^{15}\text{N}_2\text{O}$  data have not converted to  $\delta^{15}\text{N}$  reference yet.

The sampling sites in this study are divided into three different categories of environments which are biological (rice paddy and pine forest), oil gas combustion (JAERI's main gate), and marine environment (near shore the Pacific ocean) included one control point (Lab041 bldg. 3 which is in a concrete building). The isotopic compositions of  $\delta^{13}\text{CH}_4$ ,  $\delta^{13}\text{CO}_2$ , and  $\delta^{15}\text{N}_2\text{O}$  at each source types are clearly different and therefore are useful for identification of processes occurring within the sources including the cross-link affects.

The discussion is divided according to the individual sampling sites

### 3.1 Hosoura rice paddy

Discussion is made separately on the observations of seasonal variation and diurnal variation.

### 3.1.1 Seasonal variation

The purpose of this study was to determine and interpret the  $\delta^{13}\text{CH}_4$ ,  $\delta^{13}\text{CO}_2$ , and  $\delta^{15}\text{N}_2\text{O}$  in rice field, in terms of seasonal variations during the completed vegetation period.

#### Details of the cultivation practices (Fig. 4)

##### Study area

Hosoura rice paddy, Tokai-mura, Ibaraki prefecture

Total area of 25 are (1 are = 100 m<sup>2</sup>)

##### Study duration

April 1999 through January 2000 (the entire rice growing season)

##### Name of cultivar

Chiyonishiki

##### Sowing period

40 days

##### Total growth period in the paddy field

114 days (transplanted on May 14<sup>th</sup> and harvested on Sep 4<sup>th</sup>)

##### Plant density

~ 50 plants/m<sup>2</sup>

##### Cultivation practice

No crop rotation during winter (single cropping regime). The cultivar was seeded for ~ 40 days before it was transplanted into the paddy field. Rice stubble from the previous growing season has been plowed back into the field after harvest when it was around September to October 1998. Flood - water was first introduced to the paddy on May 5<sup>th</sup> and was maintained at 5-8 cm depth. Field baking which was carried out twice, on July 10<sup>th</sup> for one-week period (wet soil) and on August 18<sup>th</sup> when floodwater was drained until harvest. "Field baking" is the common practice of keeping rice field dry for a certain period of time in order to enhance the development of the root and the reproductive (flowering) system of the rice plant. This practice is the same as in the Wali -village, about 25 km north of Beijing China [20,21].

##### Fertilization

Broadcasting of chemical fertilizer. The fertilizer treatments were

- (i) Broadcasting and incorporation of PO<sub>4</sub> and nitric lime in land. Amount of 60 kg/10 are have been prepared together with rice straws and animal manures.
- (ii) Broadcasting and incorporation of N, K, and P fertilizers in April. Amount of 20 kg/10 are have been made before transplanting the rice. .
- (iii) Broadcasting of N and K fertilizer (ratio 2:8). Amount of 10 kg/10 are has been applied to the wet soil around 10 days before flowering (in July).

## I Methane

The methane formation and its transportation in the paddy soil are very complex. The observations were started before the rice transplantation and continued throughout the entire vegetation cycle including after harvest. A strong seasonal variation was found, illustrated in Fig. 5.

The phenomena could be explained as follows:

- (1) **Land preparing stage** The CH<sub>4</sub> precursor should be organic matter from previous vegetation (rice straws which were incorporated into the soil) and from a previous seasons input of root exudates and litter produced by the rice plant [21,24].
- (2) **Tillering stage** The precursor was a fermentation of acetate, same sources as in the land preparing stage. During this time, the paddy field was flooded with water and the rice was in its early stage. Major pathway of CH<sub>4</sub> was directly from natural ebullition of gas bubbles. It was a partial oxidation process occurred. The highest <sup>13</sup> $\delta$ CH<sub>4</sub> values found in this stage ranged from -18.0 to -12.8‰. Emitted methane was enriched <sup>13</sup> $\delta$ CH<sub>4</sub>. The same variation was also found in the study of *Bergamasch P.* [19].
- (3) **Flowering stage** The land still had floodwater at 5-10 cm. The rice was in its reproductive level when the growth as well as the root system increased. Precursors of CH<sub>4</sub> were fermentation of acetate and root exuded substrates i.e., carbohydrate, organic acid and amino acid. Transportation of CH<sub>4</sub> was via the rice plants where preferred light isotope <sup>13</sup> $\delta$ CH<sub>4</sub>; therefore, the emitted CH<sub>4</sub> into the atmosphere was depleted <sup>13</sup> $\delta$ CH<sub>4</sub>. A well-developed root systems provided substrates for oxidizing bacteria which affected the oxidation processes to be occurred.[21,22]. Emitted methane thus was depleted <sup>13</sup> $\delta$ CH<sub>4</sub>.
- (4) **Ripening stage until harvest** Plant-mediated transport became important pathway as the stems grew larger [7,21,22,25,26,27]. Rice, the same as other march plants, has a spiral hollow structure which provide a transportation path for the methane formed by methanogenic bacteria under the water. *Dacey and Klug* [28] reported that the substantial amounts of methane escaping from a eutrophic lake in Michigan rose through water lily, *Nuphar luteum*, rather than by ebullition (bubble release). The floodwater was drained although water still provided to the rice paddy (wet soil). Oxidation process decreased as root substrates and oxidizing bacteria reduced. Emitted methane thus was slightly enriched <sup>13</sup> $\delta$ CH<sub>4</sub>.
- (5) **Post-harvest** There was neither rice plants nor flooding water to provide a transport path. Soil-entrapped methane transported through soil macro pores when the field fell dry. It was observed that stable isotopic ratios of methane were enriched in this phase.

This distinctive feature appears very similar to the other studies [7,21,27]. However, *Tyler et al.*, [29] found no significant variation in <sup>13</sup> $\delta$ CH<sub>4</sub> of rice CH<sub>4</sub> throughout the growth cycle in Kenya rice fields. It should be noted that the previous studies covered only a growing period, but this study contained both land preparing and post harvest stages.

## II Carbon dioxide

Mechanisms concerning carbon dioxide in the rice paddy environment are categorized in three processes[2,11,13]:

- (1) *Respiration process* -emits light isotope (enriched  $\delta^{13}\text{CO}_2$  in the atmosphere)
- (2) *Transpiration process* -emits light isotope (enriched  $\delta^{13}\text{CO}_2$  in the atmosphere)
- (3) *Photosynthesis process* – emits heavy isotope (depleted  $\delta^{13}\text{CO}_2$  in the atmosphere)

These processes are relatively consistent with time through plant's life [30]. Therefore, the seasonal variation was not obviously observed as was found in methane. However,  $\delta^{13}\text{CO}_2$  were slightly decreased with time through the vegetation cycle during reproductive stage i.e., flowering and ripening (until harvested). This can be explained that rice in its flowering and ripening stage is used to be active on photosynthesis process, especially in flowering stage (days 62-86 of transplanting). After that, isotopic ratios of atmospheric  $\text{CO}_2$  became enrichment, illustrated in Fig. 6. Following post harvested, weeds were found wildly grown in the paddy field which resulted in the rising peak of enriched  $\delta^{13}\text{CO}_2$ . This may be explained by two assumptions that respiration process in that weeds exceeded photosynthesis resulted in enriched  $\delta^{13}\text{CO}_2$  in the atmosphere. Other possible explanation is weeds found in the rice paddy are a C4 plant type while rice is a C-3 plant type (C-3 plants follow a 3 carbon photosynthesis pathway and are depleted in  $^{13}\text{C}$  in comparison to C-4 plants which follow a 4 carbon pathway.  $\delta^{13}\text{CO}_2$  in the atmosphere reflect the  $\delta^{13}\text{CO}_2$  of the plant carbon) [29]. However, at the moment these two assumptions can not be confirmed, it is clear that further measurements are needed.

## III Nitrous oxide

Nitrous oxide emission is mainly controlled by mineral fertilizer, soil temperature and soil water content [26,17,18,19,20]. It is distinctively increased after broadcasting N fertilizer 20 days before flowering (indicated in the Figure 7). In addition, it was rainy season (July) and soil water content is one of the key factors. The increasing of soil water affects the emission of  $\text{N}_2\text{O}$ . Isotopic ratios of nitrous oxide were increasing with time, illustrated in Fig. 7. The peak fell sharply after harvested because of no N fertilizer input and it was a dry season.

### 3.1.2 Diurnal variation

The purpose of this study was to investigate the influence of time of day, soil temperature and season on the isotopic signatures of all three atmospheric trace gases  $\text{CH}_4$ ,  $\text{CO}_2$ , and  $\text{N}_2\text{O}$  in the rice paddy, Hosoura Tokai-mura.

Diurnal measurement of stable isotopic ratio measurements in atmospheric trace gases of  $\text{CH}_4$ ,  $\text{CO}_2$ , and  $\text{N}_2\text{O}$  were carried out at three stages of rice at the Hosoura rice paddy, Tokai-mura. The

diurnal observations were performed during the “2-3 leaf” stage (May15<sup>th</sup> and May22<sup>nd</sup>), the tillering stage (June20<sup>th</sup>) and at the end of the reproductive stage (August15<sup>th</sup>), respectively. All three atmospheric trace gases showed strong diurnal variation during the entire growing season although the amplitude varied. The daily variation of the stable isotopic ratios of CH<sub>4</sub> were likely dependent on soil temperature whilst the diurnal variation of CO<sub>2</sub> and N<sub>2</sub>O may be due to stages of rice growth and fertilizer input, as illustrated in **Fig 8**. Except for the CH<sub>4</sub>, the isotopic signatures showed the diurnal variation pattern during the early tillering stage (May15<sup>th</sup>, May22<sup>nd</sup>, and June20<sup>th</sup>) but did not show during the reproductive stage (August15<sup>th</sup>). Summarized data are in **Table 4** and are illustrated in **Figs. 9 and 10**. The lowest stable isotopic ratios were always found in the morning and late evening, on the contrary the maximum values were observed in the afternoon. It is evident that  $\delta^{13}\text{CH}_4$  are strongly dependent on temperature within a certain period of vegetation cycle. Observations made in May15<sup>th</sup> and May22<sup>nd</sup> fell into a short interval, a few days after transplanting. The measurement in June 20<sup>th</sup> showed the correlation between soil temperature and  $\delta^{13}\text{CH}_4$ . During these days the growth of rice is relatively stable and no large-scale agricultural management was practiced. Floodwater remained almost unchanged. The soil temperature therefore appears to be the dominant factor during the period when the other factors remain stable. In some stage of rice growth the other factors may be more significant than soil temperature. These observations were agreed well with the other studies [21,24].

### 3.2 JAERI's main gate and Pine wood

Measurement of the  $\delta^{13}\text{CH}_4$ ,  $\delta^{13}\text{CO}_2$ , and  $\delta^{15}\text{N}_2\text{O}$  in air samples collected at JAERI's main gate and pine stand (in JAERI) were also carried out. Gas sampling was conducted once a day in varied time during April 1999 through January 2000. The  $\delta^{13}\text{CH}_4$  in samples from JAERI's main gate and pine stand did not show diurnal pattern corresponding to either time of the day or Julian day, illustrated in **Figs. 11 and 14**. It was as well similar to the  $\delta^{15}\text{N}_2\text{O}$  observed on the JAERI's main gate,  $\delta^{15}\text{N}_2\text{O}$  were scattered randomly (**Fig. 13**). Although there are data indicated that cars with catalytic converters produce more N<sub>2</sub>O than cars without converters. This can not be clearly evaluated the pattern found. Diurnal pattern of  $\delta^{15}\text{N}_2\text{O}$  in pine wood air showed peaks corresponding to the season (Julian day) but not to the time of the day, illustrated in **Figs 16 and 17**. It is suggested that this is due to the soil water content rather than the solar radiation. Diurnal cycles in  $\delta^{13}\text{CO}_2$  in both sampling sites indicated the maximum values in the early afternoon and minimum values in the morning and late afternoon (**Fig. 12 and 15**). As samples were collected in automobiles travelling on crowded road near traffic junction in front of JAERI's main gate, the most likely main source would be commuter traffic although the surroundings are agricultural land and the Pacific ocean is close to the site. However, the diurnal pattern of  $\delta^{13}\text{CO}_2$  did not show any peaks corresponding to the morning and evening rush hours, as would be expected in this case. Neither is there any systematic variation in the pattern with day of the week. Within the pine stand, plant and

soil respiration was the primary  $\delta^{13}\text{CO}_2$  sources. Photosynthesis uptake of  $\text{CO}_2$  during the early afternoon increased the  $\delta^{13}\text{C}$  of atmospheric  $\text{CO}_2$  the same as found in the rice paddy (see above discussion).

### 3.3 The place near shore of the Pacific ocean

The relatively similar pattern of  $\delta^{13}\text{CH}_4$ ,  $\delta^{13}\text{CO}_2$ , and  $\delta^{15}\text{N}_2\text{O}$  with biological environment (rice paddy and pine wood) along the elapsed time were observed in near shore of the Pacific ocean, illustrated in Figs. 18-20. This can be explained by the effect from terrestrial activities. As mentioned above, rice paddy is the main sources of not only  $\text{CH}_4$  itself but also changing  $\delta^{13}\text{CH}_4$ ; therefore, the rice cultivation area might has caused the  $\delta^{13}\text{CH}_4$  pattern found in this site. However, the place near shore of the Pacific ocean is a sampling site where may be effected as well from the windy marine [31,32]. The wind generally takes part in either minimize or maximize the influence of anthropogenic and terrestrial activities on atmospheric studied. But it was unlikely the cause in this case.

In addition, there is a large area covered with pine trees close to the sampling site and those pines have been in their growing stages. The ammonium and nitrate concentrations in soil and other soil conditions such as pH, soil texture, aeration, amount of dry organic matter, soil water content and soil temperature principally affect the  $\text{N}_2\text{O}$  emission [15,17,18,19,20]. The pine wood sampling site and the pine area where in the proximity of near shore of the Pacific ocean are possibly different in the contribution factors. This is the reason that both sites did not have exactly the same patterns.

In near shore of the Pacific ocean, the metabolic activity of terrestrial plants primarily caused the variation of  $\text{CO}_2$  same as resulting in the  $\text{N}_2\text{O}$ . The other mechanisms such as air-sea exchange and the wind transport of air with different isotope fractionation to the study area are possible; however, it was unlikely to be a significant cause at the sampling site. This is based on the assumption that air-sea exchange produces negligible fractionation [30,31].

However, *Mook et al* [31] reported the seasonal variation in  $\delta^{13}\text{C}$  of atmospheric  $\text{CO}_2$  in the Southern Hemisphere and tropics. It was partially a result of oceanic  $\text{CO}_2$  exchange while the seasonal variation at the three northernmost stations (La Jolla, Mauna Loa Observatory, Cape Kumukahi) was almost entirely owing to land plant activity. This agrees with the studies of *Nakazawa and Morimoto* [32]. They also suggested that the seasonal variation in the region between  $10^0\text{N}$  and the equator (northern low latitudes) were probably due to a northward transport of Southern Hemisphere air with isotopically heavier  $\text{CO}_2$  in association with the monsoon circulation in Southeast Asia.

## 4. SUMMARY

Measurements of the seasonal changes of the stable isotopic ratios of  $\delta^{13}\text{CH}_4$ ,  $\delta^{13}\text{CO}_2$ , and  $\delta^{15}\text{N}_2\text{O}$  under in-situ conditions from a variety of sources in Tokai-mura are presented. Each process occurring and its effect on the stable isotopic ratio of  $\delta^{13}\text{CH}_4$ ,  $\delta^{13}\text{CO}_2$ , and  $\delta^{15}\text{N}_2\text{O}$  in the sampling sites are discussed. Measured data indicate a strong seasonal variations in biological environment (rice paddy and pine wood).

It was observed that the land plant activity was the main cause of the seasonal variation found in the marine environment (near shore of the Pacific ocean). However, either seasonal or diurnal cycles were not found in the JAERI's main gate sampling site. .

Our data indicate that a prolonged period of study is required before annual flux can be obtained. Results showed that the fluctuations of the weather have a dominating influence on their isotopic fractionations. Therefore, short-term study of these three atmospheric trace gases lead to high uncertainty. This study confirms that the isotopic signatures could provide clues as to the mechanisms of atmospheric methane, carbon dioxide, and nitrous oxide, especially in the rice paddy.

In the rice paddy, studies on organic  $^{13}\text{C}$  in rice plant on each stage of growing period and each part of the mature rice plant are suggested. Additionally, measurements of soil type and  $\delta\text{D}$  of  $\text{CH}_4$  are necessary to support the estimating transport mechanisms.

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Table 1 Isotopic ratios of atmospheric methane

Station	collected date	time	Analysed date	time	C-13
JAERI	14-Apr	1100	24-Apr	1112	-21.36
	19-Apr	0845	26-Apr	1346	-44.00
	22-Apr	0850	26-Apr	1430	-20.57
	26-Apr	0855	27-Apr	1452	-13.90
	27-Apr	0850	27-Apr	1557	-14.34
	28-Apr	0850	10-May	1112	-16.88
	30-Apr	0850	10-May	1000	-39.96
	2-Jun	1130	17-Jun	0850	-21.73
	22-Jun	1330	22-Jun	1422	-38.85
	23-Jun	1140	6-Jul	1717	-15.77
	7-Jul	1030	7-Jul	1153	-13.90
	8-Jul	0820	8-Jul	1004	-12.54
	12-Jul	1330	12-Jul	1404	-38.92
	15-Jul	1345	5-Aug	1106	-31.55
	16-Jul	1100	5-Aug	1127	-31.71
	27-Jul	0850	5-Aug	1148	-31.88
	30-Jul	1050	5-Aug	1309	-32.02
	6-Aug	0840	6-Aug	0853	-22.72
	12-Aug	0955	12-Aug	1029	-40.59
	18-Aug	1445	1-Sep	0925	-29.68
	28-Aug	1555	1-Sep	0950	-30.43
	31-Aug	1535	1-Sep	1010	-30.78
	2-Sep	1610	30-Sep	0931	-35.05
	3-Sep	1550	1-Oct	1602	-28.05
	9-Sep	1530	30-Sep	1037	-35.95
	12-Sep	1600	30-Sep	1015	-33.47
	1-Sep	1455	1-Oct	1750	-40.46
	24-Sep	1405	7-Oct	1518	-37.87
	30-Sep	1555	7-Oct	1457	-29.48
	1-Oct	1610	1-Oct	1729	-33.61
	9-Dec	1335	13-Jan	1414	-23.74
	10-Dec	1320	13-Jan	1435	-24.72
	13-Dec	1415	16-Jan	1208	-15.88
	14-Dec	1310	13-Jan	1044	-14.81
	16-Dec	1330	27-Jan	1633	-27.69
	14-Jan	1535	17-Jan	0953	-10.66
	17-Jan	1350	17-Jan	1435	-27.59
	18-Jan	1600	27-Jan	1331	-21.82
	19-Jan	1010	27-Jan	1612	-24.06
	21-Jan	1010	27-Jan	1552	-21.08
	24-Jan	1340	27-Jan	1154	-20.56
	25-Jan	0945	27-Jan	1132	-23.27
	26-Jan	0950	27-Jan	1102	-23.68
	27-Jan	0845	27-Jan	0909	-29.16
	3-Feb	1915	8-Feb	1122	-23.46
	3-Feb	2040	7-Feb	1658	-24.28
	6-Feb	1240	8-Feb	10.32	-23.40

Table 1 Isotopic ratios of atmospheric methane(cont'd)

Station	collected date	time	Analysed date	time	C-13
L 041 Bldg3	14-Apr	1115	15-Apr	1442	-14.76
	15-Apr	1550	15-Apr	1554	-13.37
	27-Apr	1620	27-Apr	1626	-14.47
	22-Jun	1405	22-Jun	1442	-15.09
	7-Jul	1045	7-Jul	1100	-14.09
	8-Jul	1020	8-Jul	1026	-13.68
	19-Jul	1535	19-Jul	1536	-45.09
	19-Jul	1600	19-Jul	1620	-43.83
	4-Aug	1620	4-Aug	1627	-22.97
	5-Aug	1020	5-Aug	1024	-30.51
	5-Aug	1325	5-Aug	1330	-33.96
	5-Aug	1655	5-Aug	1659	-35.78
	13-Aug	1500	27-Aug	1620	-34.07
	27-Aug	1620	1-Sep	1034	-33.79
Pine wood	26-Apr	1305	27-Apr	1516	-16.06
	30-Apr	1005	10-May	1318	-42.99
	30-Apr	1345	10-May	1339	-41.82
	7-May	1430	10-May	1400	-44.98
	10-May	1305	10-May	1421	-24.38
	18-May	0915	18-May	1049	-46.59
	31-May	1320	17-Jun	0911	-17.18
	1-Jun	1320	17-Jun	1045	-16.65
	2-Jun	1045	17-Jun	0934	-17.02
	3-Jun	0935	17-Jun	1106	-16.82
	17-Jun	1310	17-Jun	1426	-15.07
	21-Jun	1340	22-Jun	1045	-12.60
	23-Jun	1310	7-Jul	0953	-16.32
	24-Jun	1035	7-Jul	1038	-13.66
	7-Jul	0940	7-Jul	1017	-13.76
	8-Jul	0920	8-Jul	0942	-12.28
	12-Jul	1140	12-Jul	1203	-33.10
	15-Jul	0940	5-Aug	1403	-32.71
	16-Jul	0950	5-Aug	1430	-34.64
	27-Jul	1005	5-Aug	1451	-33.72
	28-Jul	1035	5-Aug	1512	-35.08
	29-Jul	1145	5-Aug	1533	-35.31
	1-Aug	1355	5-Aug	1554	-35.07
	11-Aug	1405	11-Aug	1543	-39.66
	12-Aug	0910	12-Aug	1049	-41.12
	13-Aug	1400	31-Aug	1548	-33.93
	18-Aug	1315	31-Aug	1723	-32.49
	24-Aug	1345	31-Aug	1609	-28.69
	25-Aug	1545	31-Aug	1633	-29.85
	28-Aug	1615	31-Aug	1744	-31.57
	31-Aug	1540	31-Aug	1653	-30.30
	1-Sep	1450	7-Oct	0917	-26.70
	2-Sep	1605	6-Oct	1340	-28.01

Table 1 Isotopic ratios of atmospheric methane(cont'd)

Station	collected date	time	Analysed date	time	C-13
Pine wood	3-Sep	1545	7-Oct	0938	-33.57
	9-Sep	1535	7-Oct	1324	-33.39
	12-Sep	1605	7-Oct	0959	-29.57
	23-Sep	1100	7-Oct	1345	-34.38
	30-Sep	1600	7-Oct	1303	-31.58
	1-Oct	1615	7-Oct	1539	-37.87
	8-Oct	1000	8-Oct	1012	-35.28
	23-Nov	1735	2-Dec	1346	-29.13
	10-Dec	1325	13-Jan	1352	-21.32
	13-Dec	1420	13-Jan	1022	-17.97
	16-Dec	1335	16-Dec	1431	-19.75
	14-Jan	1530	17-Jan	1041	-28.84
	17-Jan	1355	17-Jan	1404	-35.93
	18-Jan	1555	27-Jan	1539	-19.47
	19-Jan	0955	27-Jan	1714	-24.81
	21-Jan	1005	27-Jan	1653	-25.09
	24-Jan	1335	27-Jan	1508	-22.92
	25-Jan	0940	27-Jan	1447	-21.73
	26-Jan	0955	27-Jan	1414	-23.75
	27-Jan	0850	27-Jan	1352	-20.95
	3-Feb	1920	7-Feb	1719	-23.05
	3-Feb	2045	8-Feb	0918	-20.81
	6-Feb	1245	8-Feb	1011	-22.10
Rice paddy	14-Apr	1045	15-Apr	1508	-36.30
	18-Apr	1705	26-Apr	1408	-25.59
	22-Apr	0700	27-Apr	1430	-11.28
	26-Apr	0830	27-Apr	1409	-21.07
	27-Apr	0705	27-Apr	1537	-19.22
	28-Apr	0720	10-May	1031	-45.24
	3-May	0825	10-May	1053	-45.44
	5-May	0810	10-May	1116	-40.37
	5-May	1615	10-May	1137	-38.50
	8-May	1005	10-May	1443	-44.53
	8-May	1530	10-May	1504	-46.00
	15-May	0940	17-May	1253	-46.59
		1340	17-May	1316	-46.33
		1650	17-May	1338	-46.18
		1925	17-May	1359	-46.71
		2055	17-May	1421	-43.88
	18-May	0755	18-May	0959	-46.64
	20-May	0830	24-May	1704	-48.73
	22-May	2100	24-May	1449	-15.19
		1200	24-May	1516	-17.24
		1500	24-May	1428	-46.65
		1700	24-May	1536	-47.79
		1900	24-May	1558	-47.52
		2050	25-May	0915	-46.50

Table 1 Isotopic ratios of atmospheric methane(cont'd)

Station	collected date	time	Analysed date	time	C-13
Rice paddy	26-May	0800	17-Jun	1130	-14.77
	29-May	1515	17-Jun	1002	-18.03
	30-May	1500	17-Jun	1151	-15.21
	2-Jun	0800	17-Jun	1318	-15.15
	3-Jun	0825	17-Jun	1339	-15.00
	4-Jun	1040	17-Jun	1401	-15.85
	19-Jun	1510	22-Jun	1107	-15.52
	20-Jun	0820	22-Jun	1129	-16.50
		1200	22-Jun	1142	-15.57
		1500	22-Jun	1207	-12.68
		1700	22-Jun	1250	-14.93
		1900	22-Jun	1311	-15.19
		2100	22-Jun	1327	-17.24
	22-Jun	0810	22-Jun	1344	-14.89
	23-Jun	0825	5-Jul	1414	-12.77
	24-Jun	0815	5-Jul	1352	-16.59
	25-Jun	0720	6-Jul	1529	-15.20
	27-Jun	0910	6-Jul	1443	-15.63
	28-Jun	0810	6-Jul	1550	-16.13
	7-Jul	0750	7-Jul	1121	-13.89
	10-Jul	1015	12-Jul	1258	-36.05
	11-Jul	1045	12-Jul	1318	-38.61
	15-Jul	0825	6-Aug	1259	-29.92
	16-Jul	0730	6-Aug	1344	-33.99
	17-Jul	1330	6-Aug	1436	-35.21
	24-Jul	1450	6-Aug	1415	-35.43
	25-Jul	0900	11-Aug	1358	-40.60
	27-Jul	0835	11-Aug	1419	-40.81
	28-Jul	0825	11-Aug	1440	-40.14
	29-Jul	0820	11-Aug	1016	-38.55
	30-Jul	0730	11-Aug	1500	-39.55
	31-Jul	1335	11-Aug	1037	-37.99
	1-Aug	0755	11-Aug	1100	-39.99
	7-Aug	0920	11-Aug	1522	-39.24
	8-Aug	1630	11-Aug	1146	-38.61
	9-Aug	0830	11-Aug	1121	-40.37
	12-Aug	0840	12-Aug	1111	-38.50
	13-Aug	0835	31-Aug	0954	-29.00
	15-Aug	0830	27-Aug	1353	-30.26
		1035	27-Aug	1415	-30.19
		1330	27-Aug	1435	-30.67
		1515	27-Aug	1456	-30.89
		1715	27-Aug	1517	-31.13
		1900	27-Aug	1537	-32.47
		2050	27-Aug	1559	-32.91
	18-Aug	0700	31-Aug	1014	-28.91
	19-Aug	0640	31-Aug	1035	-30.72
	24-Aug	0655	31-Aug	1055	-31.79

Table1 Isotopic ratios of atmospheric methane (cont'd)

Station	collected date	time	Analysing date	time	C-13
Rice paddy	25-Aug	1325	31-Aug	1117	-31.42
	27-Aug	0845	31-Aug	1137	-31.76
	28-Aug	1350	31-Aug	1158	-32.55
	29-Aug	0925	31-Aug	1321	-28.10
	31-Aug	0840	31-Aug	1342	-29.05
	1-Sep	0835	30-Sep	1100	-27.89
	2-Sep	0715	30-Sep	1121	-31.59
	3-Sep	0720	30-Sep	1144	-33.34
	4-Sep	0925	30-Sep	1204	-34.60
	5-Sep	0720	30-Sep	1316	-28.44
	6-Sep	0720	30-Sep	1341	-29.24
	7-Sep	0710	30-Sep	1403	-30.40
	9-Sep	0800	30-Sep	1424	-32.36
	11-Sep	1005	30-Sep	1445	-39.15
	12-Sep	0820	30-Sep	1507	-38.36
	13-Sep	0820	30-Sep	1528	-37.99
	15-Sep	1320	30-Sep	1550	-40.13
	18-Sep	0950	30-Sep	1611	-34.82
	19-Sep	1335	30-Sep	1632	-43.99
	23-Sep	1045	30-Sep	1653	-39.72
	28-Sep	0805	30-Sep	1713	-31.44
	2-Oct	0735	1-Dec	1533	-31.68
	3-Oct	1110	1-Dec	1554	-30.80
	9-Oct	1650	1-Dec	1615	-33.93
	10-Oct	1400	1-Dec	1635	-35.26
	11-Oct	1130	1-Dec	1657	-34.69
	16-Oct	1535	1-Dec	1720	-32.54
	1-Nov	0830	1-Dec	1825	-30.75
	12-Nov	0745	1-Dec	1748	-32.27
	13-Nov	0800	1-Dec	1809	-29.94
	14-Nov	1500	2-Dec	0854	-25.76
	15-Nov	0705	2-Dec	0915	-24.58
	16-Nov	0700	2-Dec	1119	-27.94
	18-Nov	0700	2-Dec	0950	-29.81
	19-Nov	1510	2-Dec	1011	-25.58
	21-Nov	0755	1-Dec	1830	-31.44
	23-Nov	0705	2-Dec	1037	-30.18
	8-Dec	0740	16-Dec	1554	-21.17
	9-Dec	0755	16-Dec	1615	-23.68
	10-Dec	0735	16-Dec	1636	-28.56
	14-Dec	1155	16-Dec	1657	-27.21
	15-Dec	0755	16-Dec	1516	-26.33
	17-Dec	0820	14-Jan	1720	-28.15
	18-Dec	0725	14-Jan	1741	-27.46
	15-Jan	0840	17-Jan	1314	-28.98
	16-Jan	0755	17-Jan	1335	-28.30

Table1 Isotopic ratios of atmospheric methane (cont'd)

Station	collected date	time	Analysing date	time	C-13
Ocean	14-Apr	1035	15-Apr	1530	-34.72
	14-May	0730	14-May	0938	-45.57
	18-May	1125	18-May	1137	-46.30
	19-May	0945	24-May	1643	-47.80
	20-May	0925	24-May	1342	-43.18
		1400	24-May	1140	-45.06
		1900	25-May	0852	-45.73
		2100	24-May	1406	-44.32
	25-May	1545	17-Jun	1447	-15.58
	26-May	1415	17-Jun	1512	-14.51
	27-May	1045	17-Jun	1534	-16.33
	28-May	1030	17-Jun	1600	-15.10
	31-May	1635	17-Jun	1620	-15.34
	1-Jun	1020	17-Jun	1641	-15.06
	3-Jun	1010	17-Jun	1025	-17.47
	4-Jun	0940	17-Jun	1702	-15.20
	19-Jun	1510	22-Jun	1154	-15.89
	20-Jun	0820	22-Jun	1301	-14.93
	21-Jun	1355	22-Jun	1322	-14.81
	23-Jun	1420	6-Jul	1507	-15.58
	25-Jun	0740	6-Jul	1611	-15.47
	26-Jun	1310	6-Jul	1632	-15.39
	27-Jun	0940	6-Jul	1653	-17.00
	7-Jul	1140	7-Jul	1215	-13.56
	11-Jul	1110	12-Jul	1343	-34.73
	16-Jul	1020	5-Aug	1616	-34.87
	17-Jul	1550	5-Aug	1637	-33.40
	18-Jul	0815	6-Aug	0937	-32.59
	24-Jul	1855	6-Aug	0958	-32.99
	25-Jul	0935	6-Aug	1040	-35.59
	28-Jul	0920	5-Aug	1046	-28.25
	29-Jul	0945	6-Aug	1019	-33.92
	30-Jul	0925	6-Aug	1102	-36.46
	31-Jul	1420	6-Aug	1143	-33.55
	6-Aug	0925	6-Aug	1203	-36.22
	9-Aug	0930	11-Aug	1249	-37.61
	11-Aug	0915	11-Aug	1208	-34.63
	24-Aug	0920	27-Aug	0945	-28.16
	25-Aug	0945	27-Aug	1006	-29.07
	26-Aug	1300	27-Aug	1331	-26.05
	27-Aug	0935	31-Aug	1403	-29.97
	28-Aug	1600	31-Aug	1424	-30.99
	29-Aug	0935	31-Aug	1445	-34.07
	30-Aug	0905	31-Aug	1505	-31.77
	31-Aug	1310	31-Aug	1526	-32.05
	1-Sep	1300	7-Oct	1559	-37.48
	2-Sep	1315	7-Oct	1626	-36.77
	3-Sep	0935	7-Oct	1647	-40.51

Table1 Isotopic ratios of atmospheric methane (cont'd)

Station	collected date	time	Analysing date	time	C-13
Ocean	4-Sep	1525	7-Oct	1708	-36.10
	5-Sep	0740	7-Oct	1728	-40.92
	6-Sep	1315	7-Oct	1749	-39.14
	8-Sep	1245	8-Oct	0840	-33.81
	10-Sep	1335	7-Oct	1406	-36.41
	12-Sep	1615	8-Oct	0901	-34.50
	14-Sep	0920	8-Oct	0929	-37.87
	17-Sep	1337	8-Oct	0950	-36.60
	23-Sep	1130	7-Oct	1427	-36.94
	9ocr	1615	2-Dec	1143	-23.02
	10-Oct	1430	2-Dec	1325	-31.51
	17-Oct	1100	2-Dec	1228	-30.49
	21-Oct	1020	2-Dec	1304	-31.03
	14-Nov	1355	2-Dec	1207	-25.29
	13-Dec	1435	16-Dec	1343	-16.44
	16-Dec	0940	17-Jan	1126	-31.63
	19-Jan	1025	27-Jan	1310	-21.81

Table 2 Isotopic ratios of atmospheric carbon dioxide

Station	collected date	time	Analysing date	time	C-13	O-18
JAERI	21-Apr	0850	22-Apr	1449	-13.80	7.55
	22-Apr	0850	22-Apr	1531	-13.77	5.12
	27-Apr	0850	28-Apr	1105	-14.18	7.09
JAERI	6-Jun	1130	2-Jun	1216	-9.99	9.59
	22-Jun	1330	23-Jun	1606	-7.68	8.93
	23-Jun	1140	23-Jun	1653	-7.87	10.56
	7-Jul	1030	9-Jul	1517	-10.99	8.95
	8-Jul	0820	9-Jul	1505	-11.51	9.29
	12-Jul	1330	12-Jul	1516	-10.46	9.85
	14-Jul	1525	15-Jul	1625	-11.97	8.66
	15-Jul	1345	15-Jul	1636	-9.56	10.39
	16-Jul	1100	16-Jul	1329	-9.71	9.53
	27-Jul	0850	27-Jul	1135	-11.14	9.25
	30-Jul	1050	30-Jul	1134	-13.79	7.88
	6-Aug	0840	12-Aug	1137	-11.73	9.37
	12-Aug	0955	12-Aug	1149	-12.07	8.32
	18-Aug	1445	19-Aug	1128	-11.85	9.58
	25-Aug	1555	9-Sep	1629	-9.39	9.37
	31-Aug	1534	9-Sep	1640	-9.61	10.21
	1-Sep	1455	20-Sep	1757	-13.02	10.62
	2-Sep	1610	6-Sep	1500	-9.40	9.11
	3-Sep	1550	6-Sep	1255	-10.19	8.79
	9-Sep	1530	21-Sep	0955	-9.98	8.36
	12-Sep	1600	21-Sep	1047	-11.54	9.44
	24-Sep	1405	24-Sep	1423	-11.87	9.46
	9-Dec	1335	14-Dec	1630	-8.10	7.23
	10-Dec	1320	21-Jan	1517	-11.49	6.29
	13-Dec	1415	14-Dec	1649	-8.59	7.23
	14-Dec	1310	21-Jan	1712	-11.05	5.96
	14-Jan	1535	21-Jan	1143	-9.00	6.95
	16-Jan	1330	21-Jan	1130	-10.55	10.89
	17-Jan	1350	19-Jan	1401	-9.26	7.13
	18-Jan	1600	19-Jan	1737	-8.84	7.31
	19-Jan	1000	21-Jan	0918	-8.82	7.73
	21-Jan	1010	21-Jan	1118	-7.94	7.54
	24-Jan	1340	24-Jan	1358	-6.33	8.57
	26-Jan	0950	2-Feb	1136	-9.34	8.89
	27-Jan	0845	2-Feb	1148	-11.81	7.96
	3-Feb	1915	8-Feb	1122	-11.43	8.09
	3-Feb	2040	8-Feb	1134	-20.34	4.96
	6-Feb	1240	8-Feb	1055	-10.04	8.42
L 041 Bldg3	23-Apr	1015	23-Apr	1019	-11.65	8.65
	30-Apr	1545	30-Apr	1548	-13.67	7.83
	20-May	1410	20-May	1414	-14.40	8.07
	20-May	1710	20-May	1715	-13.41	8.42
	4-Jun	1300	4-Jun	1304	-11.07	4.85
	22-Jun	1505	23-Jun	1408	-14.39	7.43

Table 2 Isotopic ratios of atmospheric carbon dioxide(cont'd)

Station	collected date	time	Analysing date	time	C-13	O-18
L o41 Bldg3	9-Jul	1620	9-Jul	1656	-12.53	8.08
	15-Jul	1635	15-Jul	1647	-13.80	7.90
	16-Jul	1035	16-Jul	1340	-13.11	7.32
	23-Jul	1050	23-Jul	1055	-15.36	5.11
	26-Jul	1445	26-Jul	1451	-13.25	7.79
	29-Jul	1350	29-Jul	1352	-13.47	8.15
	30-Jul	1100	30-Jul	1123	-13.73	8.30
	13-Aug	1530	13-Aug	1702	-14.49	7.63
	18-Aug	1520	19-Aug	1151	-11.76	8.91
	19-Aug	1440	19-Aug	1444	-13.48	8.41
	2-Sep	1600	6-Sep	1333	-16.51	6.02
	22-Sep	1015	22-Sep	1318	-14.62	6.54
	24-Sep	1320	24-Sep	1323	-8.07	7.83
Pine wood	22-Apr	0930	23-Apr	1051	-10.66	8.73
	26-Apr	1305	28-Apr	1012	-11.92	8.31
	30-Apr	1005	30-Apr	1627	-13.42	7.59
	10-May	1305	10-May	1339	-7.63	9.89
	13-May	1620	13-May	1639	-10.85	9.14
	18-May	0915	21-May	1027	-11.79	9.56
	31-May	1320	31-May	1409	-23.45	7.89
	1-Jun	1320	1-Jun	1449	-9.74	9.80
	2-Jun	1045	2-Jun	1237	-9.81	8.94
	3-Jun	0935	3-Jun	1126	-11.64	8.51
	17-Jun	1310	23-Jun	1641	-8.32	10.45
	21-Jun	1340	23-Jun	1551	-8.39	7.78
	23-Jun	1310	24-Jun	1112	-7.29	10.35
	24-Jun	1035	24-Jun	1124	-7.71	9.61
	8-Jul	0925	9-Jul	1531	-8.74	10.48
	12-Jul	1140	12-Jul	1505	-14.40	7.71
	14-Jul	1620	15-Jul	1547	-10.35	8.92
	15-Jul	0940	15-Jul	1559	-9.41	10.24
	16-Jul	0950	16-Jul	1159	-13.84	7.20
	17-Jul	1605	23-Jul	1110	-12.27	7.56
	27-Jul	1005	27-Jul	1147	-10.55	9.24
	28-Jul	1035	28-Jul	1109	-12.59	7.36
	29-Jul	1145	29-Jul	1315	-10.05	10.63
	1-Aug	1355	12-Aug	1340	-13.72	7.93
	11-Aug	1405	12-Aug	1317	-10.23	9.73
	12-Aug	0910	12-Aug	1329	-12.39	7.73
	13-Aug	1400	13-Aug	1650	-9.97	9.82
	18-Aug	1315	19-Aug	1139	-10.03	10.34
	19-Aug	1355	19-Aug	1431	-8.55	11.25
	24-Aug	1345	24-Aug	1424	-9.10	10.17
	25-Aug	1545	2-Sep	1603	-9.20	9.33
	28-Aug	1615	2-Sep	1552	-9.97	8.83
	31-Aug	1540	2-Sep	1617	-9.39	10.30
	2-Sep	1605	6-Sep	1511	-11.52	8.12

Table 2 Isotopic ratios of atmospheric carbon dioxide (cont'd)

Station	collected date	time	Analysing date	time	C-13	O-18
Pine wood	3-Sep	1545	6-Sep	1539	-13.21	8.25
	9-Sep	1535	21-Sep	1020	-9.09	8.37
	12-Sep	1605	21-Sep	1058	-8.53	8.80
	23-Sep	1100	24-Sep	1359	-9.56	7.19
	23-Nov	1735	2-Dec	1521	-11.71	6.11
	10-Dec	1325	14-Dec	1715	-6.26	7.74
	13-Dec	1420	21-Jan	0956	-11.12	6.41
	16-Dec	1335	24-Jan	1130	-8.14	8.17
	14-Jan	1530	21-Jan	1725	-8.72	7.65
	17-Jan	1355	21-Jan	1105	-8.59	7.29
	18-Jan	1555	21-Jan	0944	-9.19	7.32
	19-Jan	0955	21-Jan	0930	-8.14	7.55
	21-Jan	1005	21-Jan	1017	-7.54	7.34
	24-Jan	1335	24-Jan	1445	-5.63	8.88
	26-Jan	0955	2-Feb	1111	-8.81	9.19
	27-Jan	0850	2-Feb	1123	-9.60	8.83
	3-Feb	1915	8-Feb	1149	-11.75	8.94
	3-Feb	2045	8-Feb	1201	-12.17	7.63
	6-Feb	1245	8-Feb	1110	-11.30	8.02
Rice paddy	22-Apr	0700	23-Apr	1037	-12.88	7.61
	27-Apr	0705	28-Apr	1149	-13.91	7.80
	28-Apr	0720	28-Apr	1201	-11.60	9.30
	5-May	0810	6-May	1721	-13.57	7.70
	8-May	1005	11-May	1432	-8.88	9.36
	14-May	0700	14-May	0956	-9.82	8.95
	15-May	0940	18-May	1346	-12.90	8.66
		1340	18-May	1358	-9.43	9.58
		1650	18-May	1413	-9.21	9.91
		1925	18-May	1333	-11.25	9.65
		2055	18-May	1427	-8.31	9.10
	18-May	0755	21-May	1003	-10.88	10.15
	20-May	0830	21-May	1015	-12.88	8.68
	22-May	0815	25-May	1215	-12.12	9.45
		1200	25-May	1319	-10.54	9.50
		1500	25-May	1411	-9.59	8.71
		1700	25-May	1331	-11.06	9.43
		1900	25-May	1343	-11.51	9.21
		2050	25-May	1355	-10.72	9.88
	26-May	0800	31-May	1356	-9.24	10.90
	29-May	1515	1-Jun	1312	-10.93	8.50
	30-May	1500	1-Jun	1527	-11.81	7.53
	1-Jun	0745	1-Jun	1540	-9.67	9.52
	2-Jun	0800	2-Jun	1417	-12.78	9.33
	3-Jun	0825	3-Jun	1138	-10.40	9.46
	4-Jun	1040	4-Jun	1157	-10.53	10.27
	19-Jun	1510	23-Jun	1707	-14.30	7.58
	20-Jun	0820	23-Jun	1718	-14.72	7.20

Table 2 Isotopic ratios of atmospheric carbon dioxide (cont'd)

Station	collected date	time	Analysing date	time	C-13	O-18
Rice paddy	22-Jun	0810	23-Jun	1730	-9.96	8.47
	23-Jun	0825	23-Jun	1742	-8.26	11.15
	24-Jun	0815	24-Jun	1136	-10.60	7.83
	25-Jun	0720	25-Jun	0839	-8.91	9.34
	26-Jun	1240	9-Jul	1618	-9.04	9.68
	27-Jun	0910	9-Jul	1631	-13.75	7.11
	28-Jun	0810	9-Jul	1642	-13.50	7.79
	10-Jul	1015	12-Jul	1430	-9.21	10.14
	11-Jul	1045	12-Jul	1442	-13.18	8.06
	15-Jul	0825	15-Jul	1613	-10.25	9.68
	16-Jul	0730	16-Jul	1317	-12.68	8.05
	18-Jul	0750	23-Jul	1122	-18.41	6.05
	20-Jul	0910	23-Jul	1134	-13.62	6.24
	24-Jul	1450	26-Jul	1503	-13.65	7.82
	25-Jul	0900	26-Jul	1515	-12.43	7.78
	27-Jul	0835	27-Jul	1159	-13.75	7.91
	28-Jul	0825	28-Jul	1132	-9.31	10.59
	29-Jul	0820	29-Jul	1340	-17.00	6.19
	30-Jul	0730	30-Jul	1157	-10.40	9.89
	31-Jul	1335	12-Aug	1439	-12.13	9.17
	1-Aug	0755	12-Aug	1451	-15.28	6.87
	7-Aug	0920	12-Aug	1537	-14.82	7.23
	8-Aug	1630	12-Aug	1502	-17.86	6.36
	9-Aug	0830	12-Aug	1514	-16.11	6.58
	12-Aug	0840	12-Aug	1526	-15.49	7.13
	13-Aug	0835	13-Aug	0901	-14.27	8.06
	15-Aug	0830	18-Aug	1014	-17.32	6.67
		1035	18-Aug	1033	-17.64	6.05
		1330	18-Aug	1050	-16.61	6.20
		1515	18-Aug	1101	-10.51	9.25
		1715	18-Aug	1113	-14.38	6.87
		1900	19-Aug	1015	-12.16	9.70
		2050	18-Aug	1137	-15.24	7.01
	18-Aug	1900	19-Aug	1027	-11.74	9.75
	19-Aug	0640	19-Aug	1116	-15.92	6.91
	24-Aug	0655	24-Aug	1436	-13.31	7.69
	25-Aug	1325	2-Sep	1326	-9.81	9.14
	27-Aug	0845	2-Sep	1337	-14.07	7.15
	28-Aug	1350	2-Sep	1349	-12.36	7.11
	29-Aug	0925	2-Sep	1401	-11.16	8.34
	31-Aug	0840	2-Sep	1413	-12.56	6.54
	1-Sep	0835	6-Sep	1552	-9.50	10.26
	2-Sep	0715	6-Sep	1603	-9.27	9.48
	3-Sep	0720	6-Sep	1242	-14.06	6.95
	4-Sep	0925	6-Sep	1322	-9.42	9.11
	5-Sep	0720	6-Sep	1346	-16.57	6.19
	6-Sep	0720	6-Sep	1358	-15.80	5.81
	7-Sep	0710	8-Sep	1545	-10.23	7.73

Table 2 Isotopic ratios of atmospheric carbon dioxide (cont'd)

Station	collected date	time	Analysing date	time	C-13	O-18
Rice paddy	9-Sep	0800	20-Sep	1632	-13.35	8.04
	11-Sep	1005	20-Sep	1644	-15.43	6.89
	12-Sep	1530	20-Sep	1655	-11.56	7.11
	13-Sep	0820	20-Sep	1710	-10.67	6.65
	15-Sep	1320	20-Sep	1721	-13.30	7.68
	18-Sep	0950	20-Sep	1733	-11.65	9.73
	19-Sep	1335	20-Sep	1745	-14.65	7.84
	23-Sep	1045	24-Sep	1347	-14.18	9.84
	28-Sep	0805	18-Nov	1611	-12.33	8.37
	2-Oct	0735	18-Nov	1558	-15.03	12.07
	3-Oct	1110	18-Nov	1545	-13.90	13.41
	9-Oct	1650	18-Nov	1533	-14.32	12.51
	10-Oct	1400	18-Nov	1520	-12.86	13.30
	11-Oct	1130	18-Nov	1508	-13.58	12.86
	16-Oct	1535	18-Nov	1457	-15.50	11.86
	1-Nov	0830	18-Nov	1631	-15.37	12.84
	12-Nov	0745	18-Nov	1444	-14.98	13.26
	13-Nov	0800	18-Nov	1329	-13.53	12.75
	14-Nov	1500	18-Nov	1316	-14.14	14.06
	16-Nov	0700	18-Nov	1114	-10.28	10.09
	18-Nov	0700	18-Nov	10.14	-12.42	14.28
	19-Nov	1510	2-Dec	1533	-8.45	8.35
	21-Nov	0755	2-Dec	1545	-12.61	5.50
	23-Nov	0705	2-Dec	1600	-10.01	8.30
	8-Dec	0740	24-Jan	1500	-10.62	6.27
	10-Dec	0735	24-Jan	1432	-9.86	7.64
	14-Dec	1155	24-Jan	1528	-7.98	8.02
	15-Dec	0755	24-Jan	1542	-12.73	6.10
	17-Dec	0820	24-Jan	1554	-9.51	8.50
	18-Dec	0725	24-Jan	1606	-9.11	7.91
	15-Jan	0840	24-Jan	0916	-11.08	8.46
	16-Jan	0735	2-Feb	10.59	-11.06	7.98
Ocean	12-May	0715	12-May	1110	-8.46	10.01
	14-May	0730	14-May	1022	-7.59	9.97
	18-May	1125	21-May	0931	-10.34	10.09
	19-May	0945	21-May	1951	-9.31	10.26
	20-May	0925	21-May	0841	-30.37	-17.11
		1400	21-May	0854	-10.32	9.67
		1905	21-May	0906	-12.61	9.88
		2100	21-May	0918	-12.68	9.40
	25-May	1545	31-May	1147	-9.33	10.19
	26-May	1415	31-May	1159	-10.91	9.31
	27-May	1045	31-May	1326	-11.46	8.02
	28-May	1030	31-May	1343	-10.94	8.76
	31-May	1635	31-May	1719	-8.60	11.20
	1-Jun	1020	1-Jun	1503	-9.02	9.88
	3-Jun	1010	3-Jun	1114	-9.41	10.41

Table 2 Isotopic ratios of atmospheric carbon dioxide (cont'd)

Station	collected date	time	Analysing date	time	C-13	O-18
Ocean	4-Jun	0940	4-Jun	1317	-5.50	10.05
	19-Jun	1510	24-Jun	1157	-11.76	7.93
	20-Jun	0820	24-Jun	1209	-11.89	7.63
	21-Jun	1355	24-Jun	1250	-11.06	8.34
	23-Jun	1420	24-Jun	1302	-8.11	10.34
	25-Jun	0740	25-Jun	0851	-8.13	9.15
	26-Jun	1310	9-Jul	1543	-11.92	7.44
	27-Jun	0940	9-Jul	1555	-11.88	8.02
	7-Jul	1140	9-Jul	1607	-9.85	9.58
	11-Jul	1110	12-Jul	1453	-11.73	9.50
	16-Jul	1020	16-Jul	1305	-10.97	8.86
	17-Jul	1550	23-Jul	1145	-10.68	8.34
	18-Jul	0815	23-Jul	1157	-11.32	7.75
	24-Jul	1855	26-Jul	1527	-17.33	4.97
	28-Jul	0920	28-Jul	1120	-9.11	10.62
	29-Jul	0945	29-Jul	1328	-9.16	11.00
	30-Jul	0925	30-Jul	1146	-13.81	8.13
	31-Jul	1420	12-Aug	1404	-15.03	6.89
	6-Aug	0925	12-Aug	1416	-13.63	8.15
	9-Aug	0930	12-Aug	1352	-12.73	8.17
	11-Aug	0915	12-Aug	1427	-11.73	8.79
	24-Aug	0920	24-Aug	1448	-11.02	9.17
	25-Aug	0945	2-Sep	1436	-10.05	8.67
	26-Aug	1300	2-Sep	1424	-10.00	8.88
	27-Aug	0935	2-Sep	1448	-10.37	8.75
	28-Aug	1600	2-Sep	1541	-17.55	5.51
	29-Aug	0935	2-Sep	1514	-19.44	7.72
	30-Aug	0905	2-Sep	1529	-10.89	8.27
	31-Aug	1310	2-Sep	1502	-10.54	8.95
	1-Sep	1300	6-Sep	1615	-9.55	10.20
	2-Sep	1315	6-Sep	1641	-9.22	9.60
	3-Sep	0935	6-Sep	1448	-12.49	7.68
	4-Sep	1525	6-Sep	1413	-14.85	7.11
	5-Sep	0740	6-Sep	1425	-14.50	6.59
	6-Sep	1315	6-Sep	1437	-9.35	9.74
	8-Sep	1245	21-Sep	1118	-11.31	10.04
	10-Sep	1335	21-Sep	1034	-10.04	7.48
	12-Sep	1615	21-Sep	1130	-10.67	11.19
	14-Sep	0920	21-Sep	1153	-11.24	6.84
	17-Sep	1337	21-Sep	1142	-7.22	9.30
	23-Sep	1130	24-Sep	1411	-7.28	8.43
	14-Nov	1355	18-Nov	0919	-4.11	13.90
	13-Dec	1435	14-Dec	1703	-7.34	7.77
	16-Dec	0940	24-Jan	1152	-8.30	7.64
	19-Jan	1025	24-Jan	1312	-5.79	9.03

Table 3 Isotopic ratios of atmospheric nitrous oxide

Station	collected date	time	Analysing date	time	N-15	O-18
JAERI	14-Apr	1100	23-Apr	1127	-311.58	-490.57
	19-Apr	0845	23-Apr	1357	-339.44	-488.23
	26-Apr	0855	28-Apr	1419	-338.53	-490.01
L 041 Bldg3	2-Jun	1130	2-Jun	1152	-343.42	-491.55
	22-Jun	1330	22-Jun	1550	-340.79	-490.42
	23-Jun	1140	23-Jun	1153	-341.73	-490.64
	7-Jul	1030	12-Jul	1532	-342.22	-489.31
	8-Jul	0820	12-Jul	1554	-337.90	-491.95
	12-Jul	1330	12-Jul	1618	-329.94	-492.62
	14-Jul	1525	15-Jul	1334	-335.43	-490.11
	15-Jul	1345	15-Jul	1358	-320.17	-490.75
	16-Jul	1100	16-Jul	1137	-351.82	-492.80
	27-Jul	0850	27-Jul	1028	-332.95	-490.14
	30-Jul	1050	30-Jul	1100	-325.45	-491.80
	6-Aug	0840	13-Aug	1353	-321.32	-491.93
	12-Aug	0955	13-Aug	1415	-318.58	-491.96
	18-Aug	1445	18-Aug	1623	-312.28	-490.79
	25-Aug	1555	2-Sep	1036	-329.13	-491.24
	31-Aug	1535	2-Sep	1058	-327.40	-490.39
	1-Sep	1455	21-Sep	1219	-339.83	-491.14
	2-Sep	1610	7-Sep	1035	-328.36	-491.50
	3-Sep	1550	21-Sep	1322	-336.21	-489.51
	9-Sep	1530	10-Dec	1249	-344.67	-502.94
	12-Sep	1600	10-Dec	1316	-344.86	-504.13
	30-Sep	1555	10-Dec	1338	-347.20	-509.36
	1-Oct	1610	10-Dec	1537	-342.58	-501.85
	9-Dec	1335	13-Dec	1341	-349.49	-505.78
	10-Dec	1320	13-Dec	1407	-345.94	-501.28
	13-Dec	1415	14-Dec	1031	-352.77	-505.54
	16-Dec	1330	17-Dec	1044	-335.28	-500.54
	14-Jan	1535	17-Jan	1609	-349.84	-508.43
	17-Jan	1350	17-Jan	1501	-349.22	-505.91
	18-Jan	1600	18-Jan	1736	-346.17	-502.44
	19-Jan	1000	19-Jan	1147	-347.85	-499.30
	21-Jan	1010	26-Jan	0937	-350.82	-500.62
	24-Jan	1340	26-Jan	1300	-352.20	-503.09
	25-Jan	0940	25-Jan	1118	-351.50	-505.49
	26-Jan	0950	26-Jan	1323	-353.25	-505.11
	27-Jan	0845	2-Feb	1341	-350.02	-486.74
	3-Feb	1910	9-Feb	0941	-357.29	-497.41
	3-Feb	2040	9-Feb	1007	-349.31	-488.18
	6-Feb	1240	9-Feb	1209	-350.59	-490.34

Table 3 Isotopic ratios of atmospheric nitrous oxide (cont'd)

Station	collected date	time	Analysing date	time	N-15	O-18
L 041 Bldg3	23-Jul	1515	23-Jul	1517	-337.82	-492.46
	26-Jul	1150	26-Jul	1201	-336.27	-492.63
	29-Jul	1105	29-Jul	1108	-328.03	-490.62
	30-Jul	1035	30-Jul	1031	-326.98	-490.46
	13-Aug	1025	13-Aug	1628	-317.22	-486.96
	18-Aug	1520	18-Aug	1601	-322.13	-491.55
	2-Sep	1600	21-Sep	1417	-341.41	-484.04
	7-Sep	1035	7-Sep	1057	-329.11	-491.14
Pine wood	14-Apr	1105	23-Apr	1335	-340.09	-491.64
	22-Apr	0930	23-Apr	1600	-338.39	-490.47
	30-Apr	1005	7-May	1403	-317.38	-490.17
	10-May	1305	11-May	1603	-344.97	-491.31
	12-May	1040	12-May	1152	-341.93	-490.27
	13-May	1120	13-May	1124	-345.05	-490.66
	18-May	0915	21-May	1027	-343.08	-489.57
	1-Jun	1320	1-Jun	1420	-342.60	-490.69
	2-Jun	1045	2-Jun	1056	-342.01	-490.63
	3-Jun	0935	3-Jun	0951	-343.34	-490.99
	17-Jun	1310	22-Jun	1504	-338.73	-490.33
	21-Jun	1340	22-Jun	1527	-339.93	-491.12
	23-Jun	1310	23-Jun	1339	-341.63	-489.67
	24-Jun	1035	24-Jun	1050	-343.87	-495.27
	7-Jul	0940	9-Jul	1735	-340.03	-481.31
	8-Jul	0925	9-Jul	1714	-280.10	-405.80
	12-Jul	1140	12-Jul	1639	-327.07	-491.09
	14-Jul	1620	14-Jul	1627	-336.88	-491.36
	15-Jul	0940	15-Jul	1139	-331.68	-491.81
	16-Jul	0950	16-Jul	1001	-332.46	-490.22
	17-Jul	1605	23-Jul	1210	-336.95	-490.36
	27-Jul	1005	27-Jul	1112	-326.05	-491.97
	28-Jul	1035	28-Jul	1045	-338.60	-491.11
	29-Jul	1145	29-Jul	1202	-327.52	-491.75
	1-Aug	1355	13-Aug	1545	-321.27	-486.32
	11-Aug	1405	13-Aug	1436	-322.68	-487.87
	12-Aug	0910	13-Aug	1502	-323.40	-489.71
	13-Aug	1400	13-Aug	1524	-321.93	-488.90
	18-Aug	1315	18-Aug	1538	-319.02	-492.09
	19-Aug	1355	19-Aug	1409	-326.25	-490.41
	24-Aug	1345	2-Sep	1120	-327.56	-490.91
	25-Aug	1545	2-Sep	1141	-317.33	-490.46
	28-Aug	1615	2-Sep	1203	-328.21	-490.92
	31-Aug	1540	2-Sep	1251	-328.99	-492.00
	2-Sep	1605	7-Sep	1139	-316.85	-489.49
	1-Sep	1450	21-Sep	1438	-348.19	-498.89
	3-Sep	1545	21-Sep	1500	-335.12	-489.78
	23-Sep	1100	10-Dec	1202	-342.59	-498.58
	30-Sep	1600	10-Dec	1622	-343.96	-507.96

Table 3 Isotopic ratios of atmospheric nitrous oxide (cont'd)

Station	collected date	time	Analysing date	time	N-15	O-18
Pine wood	23-Nov	1735	10-Dec	1141	-349.86	-513.60
	10-Dec	1325	13-Dec	1443	-344.29	-501.15
	13-Dec	1420	14-Dec	1007	-356.96	-509.63
	16-Dec	1335	17-Dec	1129	-333.79	-497.38
	14-Jan	1530	17-Jan	1630	-346.02	-503.09
	17-Jan	1355	17-Jan	1524	-347.09	-503.64
	18-Jan	1555	18-Jan	1714	-346.04	-499.99
	19-Jan	0955	19-Jan	1124	-351.80	-507.41
	21-Jan	1005	26-Jan	0912	-355.48	-505.99
	24-Jan	1335	26-Jan	1121	-352.31	-503.36
	25-Jan	0940	26-Jan	1144	-357.62	-510.45
	26-Jan	0955	26-Jan	1000	-355.96	-509.81
	27-Jan	0850	2-Feb	1320	-352.71	-490.34
	3-Feb	1915	9-Feb	1048	-349.08	-487.33
	3-Feb	2045	9-Feb	1125	-370.27	-482.62
	6-Feb	1245	9-Feb	1147	-350.20	-489.85
Rice paddy	14-Apr	1045	23-Apr	1312	-346.10	-492.36
	5-May	0810	7-May	1510	-338.18	-489.86
	5-May	1615	7-May	1534	-341.37	-494.68
	8-May	1005	11-May	1520	-348.64	-492.75
	8-May	1530	11-May	1541	-341.84	-489.86
	12-May	0700	12-May	1214	-341.94	-490.67
	13-May	0700	13-May	1031	-344.31	-489.78
	14-May	0700	12-May	1102	-345.26	-489.70
	15-May	0940	18-May	1533	-345.44	-492.91
		1340	18-May	1555	-346.35	-493.53
		1650	18-May	1617	-344.39	-487.69
		1925	18-May	1507	-347.56	-498.01
		2055	18-May	1442	-346.29	-496.75
	18-May	0755	21-May	1003	-342.80	-489.25
	20-May	0830	21-May	1015	-343.58	-490.53
	22-May	0815	22-May	0948	-340.13	-490.06
		1200	25-May	1012	-340.97	-490.34
		1500	25-May	1036	-342.86	-490.17
		1700	25-May	1100	-343.14	-482.99
		1900	22-May	1123	-341.39	-489.75
		2050	25-May	1149	-341.06	-488.36
	26-May	0800	31-May	1446	-342.10	-488.90
	29-May	1515	1-Jun	1313	-343.03	-489.00
	30-May	1500	1-Jun	1334	-343.03	-489.37
	1-Jun	0800	1-Jun	1356	-342.34	-489.50
	2-Jun	0800	2-Jun	1120	-341.46	-488.90
	3-Jun	0825	3-Jun	1028	-339.73	-511.64
	4-Jun	1040	4-Jun	1109	-348.20	-492.74
	19-Jun	1510	23-Jun	1047	-341.53	-490.27
	20-Jun	0820	23-Jun	1109	-341.46	-490.52
	22-Jun	0810	22-Jun	1638	-341.67	-490.21

Table 3 Isotopic ratios of atmospheric nitrous oxide (cont'd)

Station	collected date	time	Analysing date	time	N-15	O-18
Rice paddy	23-Jun	0825	23-Jun	1131	-340.61	-489.16
	24-Jun	0815	24-Jun	1027	-344.00	-490.53
	25-Jun	0720	25-Jun	0816	-342.03	-490.65
	26-Jun	1240	14-Jul	1432	-331.74	-491.00
	10-Jul	1015	14-Jul	1605	-332.16	-490.76
	11-Jul	1045	14-Jul	1649	-336.85	-491.25
	15-Jul	0825	15-Jul	1120	-338.89	-492.44
	16-Jul	0730	16-Jul	1031	-335.55	-489.97
	17-Jul	1330	23-Jul	1326	-337.51	-491.60
	18-Jul	0750	23-Jul	1347	-334.46	-481.66
	20-Jul	0910	23-Jul	1410	-337.14	-490.71
	24-Jul	1450	26-Jul	1317	-332.83	-490.21
	25-Jul	0900	26-Jul	1340	-324.93	-484.14
	27-Jul	0835	27-Jul	1050	-337.99	-493.01
	28-Jul	0825	28-Jul	1023	-334.29	-489.53
	30-Jul	0730	30-Jul	1008	-338.05	-512.37
	31-Jul	1335	13-Aug	1013	-324.38	-492.07
	1-Aug	0755	13-Aug	1034	-328.19	-486.75
	7-Aug	0920	13-Aug	0951	-328.12	-490.36
	8-Aug	1630	13-Aug	1056	-327.88	-491.24
	9-Aug	0830	13-Aug	1606	-324.09	-489.56
	12-Aug	0840	13-Aug	1118	-324.33	-492.39
	13-Aug	0835	13-Aug	0930	-326.17	-489.64
	15-Aug	0830	18-Aug	1151	-325.17	-489.24
		1035	18-Aug	1327	-323.32	-489.06
		1330	18-Aug	1349	-317.99	-491.01
		1515	18-Aug	1411	-323.77	-487.76
		1715	18-Aug	1433	-314.70	-490.32
		1900	18-Aug	1455	-321.19	-489.72
		2050	18-Aug	1517	-322.00	-489.87
	18-Aug	1900	19-Aug	1326	-322.65	-490.63
	19-Aug	0640	19-Aug	1348	-327.37	-491.83
	24-Aug	0655	1-Sep	1056	-319.50	-490.87
	25-Aug	1325	1-Sep	1118	-309.25	-490.25
	27-Aug	0945	1-Sep	1140	-321.85	-491.48
	28-Aug	1350	1-Sep	1202	-322.27	-491.21
	29-Aug	0925	1-Sep	1242	-338.78	-492.59
	31-Aug	0840	1-Sep	1316	-318.45	-491.20
	1-Sep	0835	7-Sep	1558	-311.63	-493.53
	2-Sep	0715	7-Sep	1624	-320.07	-489.60
	3-Sep	0720	7-Sep	1216	-326.45	-491.19
	5-Sep	0720	7-Sep	1309	-314.57	-489.49
	6-Sep	0720	7-Sep	1350	-323.11	-490.40
	7-Sep	0710	7-Sep	1416	-321.11	-489.51
	12-Sep	1530	21-Sep	1548	-335.45	-489.05
	13-Sep	0820	21-Sep	1624	-339.86	-491.53
	15-Sep	1320	21-Sep	1646	-337.14	-489.69
	18-Sep	0950	21-Sep	1718	-334.10	-490.11

Table 3 Isotopic ratios of atmospheric nitrous oxide (cont'd)

Station	collected date	time	Analysing date	time	N-15	O-18
Rice paddy	19-Sep	1335	21-Sep	1739	-338.26	-490.34
	23-Sep	1045	4-Dec	1538	-336.25	-499.46
	28-Sep	0805	6-Dec	1131	-341.24	-501.45
	2-Oct	0735	6-Dec	1040	-345.47	-506.88
	3-Oct	1110	6-Dec	1109	-338.40	-498.63
	9-Oct	1650	6-Dec	1153	-340.82	-502.59
	10-Oct	1400	6-Dec	1312	-341.64	-503.78
	11-Oct	1130	6-Dec	1336	-345.31	-509.30
	16-Oct	1535	6-Dec	1358	-342.85	-506.74
	1-Nov	0830	6-Dec	1700	-338.20	-500.41
	12-Nov	0745	6-Dec	1433	-336.85	-501.48
	14-Nov	1500	6-Dec	1539	-343.55	-508.42
	15-Nov	0705	6-Dec	1602	-340.20	-505.58
	18-Nov	0700	6-Dec	1629	-336.71	-498.80
	19-Nov	1510	6-Dec	1651	-341.43	-506.51
	21-Nov	0755	6-Dec	1713	-339.34	-504.10
	23-Nov	0705	6-Dec	1735	-339.80	-503.54
	8-Dec	0740	13-Dec	1534	-345.19	-501.11
	9-Dec	0755	13-Dec	1510	-347.99	-508.54
	10-Dec	0735	13-Dec	1557	-350.68	-509.17
	14-Dec	1150	14-Dec	1455	-347.82	-508.27
	15-Dec	0755	17-Jan	1653	-350.64	-508.52
	16-Dec	0755	19-Jan	1039	-350.17	-504.16
	17-Dec	0820	17-Dec	1215	-345.38	-495.94
	18-Dec	0725	17-Dec	1546	-347.08	-503.19
	15-Jan	0840	19-Jan	0926	-350.27	-503.24
Ocean	14-Apr	1035	23-Apr	1149	-318.67	-492.10
	12-May	0715	12-May	1235	-347.94	-476.09
	14-May	0730	14-May	1126	-344.97	-489.50
	18-May	1125	21-May	0931	-342.98	-489.11
	19-May	0945	21-May	0951	-342.99	-489.34
	20-May	0925	21-May	1130	-345.46	-491.49
		1400	21-May	1046	-344.53	-491.17
		1905	21-May	1108	-343.63	-489.55
		2100	21-May	1151	-344.10	-489.56
	25-May	1545	31-May	1510	-342.58	-489.69
	26-May	1415	31-May	1532	-343.23	-491.05
	27-May	1045	31-May	1555	-342.97	-490.35
	28-May	1030	31-May	1617	-343.20	-490.86
	31-May	1635	31-May	1650	-343.08	-490.56
	1-Jun	1020	1-Jun	1225	-341.23	-488.76
	3-Jun	1010	3-Jun	1050	-343.66	-491.16
	19-Jun	1510	23-Jun	0940	-342.31	-490.47
	20-Jun	0820	23-Jun	1003	-342.59	-491.69
	21-Jun	1355	23-Jun	1025	-342.26	-491.05
	23-Jun	1420	23-Jun	1441	-341.77	-490.53
	25-Jun	0740	25-Jun	0754	-342.36	-491.72

Table 3 Isotopic ratios of atmospheric nitrous oxide (cont'd)

Station	collected date	time	Analysing date	time	N-15	O-18
Ocean	26-Jun	1310	15-Jul	1420	-338.29	-490.98
	27-Jun	0940	15-Jul	1442	-338.77	-492.51
	7-Jul	1140	15-Jul	1503	-338.71	-491.80
	11-Jul	1110	15-Jul	1524	-333.16	-490.98
	16-Jul	1020	16-Jul	1053	-329.37	-489.51
	17-Jul	1550	23-Jul	1432	-329.30	-491.65
	18-Jul	0815	23-Jul	1454	-335.64	-492.02
	24-Jul	1855	26-Jul	1402	-334.12	-489.84
	25-Jul	0935	26-Jul	1425	-334.73	-490.61
	28-Jul	0920	28-Jul	0956	-319.72	-489.49
	29-Jul	0945	29-Jul	1130	-319.13	-489.46
	30-Jul	0925	30-Jul	0946	-328.72	-493.02
	31-Jul	1420	13-Aug	1139	-326.94	-489.30
	6-Aug	0925	13-Aug	1311	-322.63	-488.53
	9-Aug	0930	13-Aug	1332	-323.80	-490.31
	11-Aug	0915	13-Aug	1249	-325.21	-491.39
	24-Aug	0920	1-Sep	1339	-321.91	-490.29
	25-Aug	0945	1-Sep	1400	-320.38	-489.85
	26-Aug	1300	1-Sep	1422	-306.46	-490.61
	27-Aug	0935	1-Sep	1444	-321.75	-491.92
	28-Aug	1600	1-Sep	1506	-312.10	-489.21
	29-Aug	0935	1-Sep	1527	-320.61	-490.12
	30-Aug	0905	1-Sep	1549	-321.75	-491.74
	31-Aug	1310	1-Sep	1611	-321.69	-491.69
	3-Sep	0935	7-Sep	1436	-320.82	-489.71
	4-Sep	1525	7-Sep	1514	-321.46	-489.35
	5-Sep	0740	7-Sep	1536	-323.80	-490.02
	1-Sep	1300	21-Sep	1526	-332.06	-489.04
	2-Sep	1315	6-Dec	1801	-315.55	-516.08
	3-Sep	0935	7-Sep	1436	-320.82	-489.71
	4-Sep	1525	7-Sep	1514	-321.46	-489.35
	5-Sep	0740	7-Sep	1536	-323.80	-490.02
	6-Sep	1315	6-Dec	1845	-338.88	-503.26
	8-Sep	1245	6-Dec	1823	-339.89	-493.53
	10-Sep	1335	10-Dec	0953	-347.57	-507.66
	12-Sep	1615	10-Dec	1015	-348.41	-507.08
	14-Sep	0920	9-Dec	1607	-341.98	-496.53
	23-Sep	1130	10-Dec	1056	-348.16	-508.88
	9-Oct	1615	9-Dec	1545	-349.00	-508.89
	10-Oct	1430	10-Dec	1225	-346.16	-509.02
	17-Oct	1100	9-Dec	1653	-350.10	-509.22
	21-Oct	1020	9-Dec	1714	-343.26	-496.72
	14-Nov	1355	9-Dec	1737	-345.02	-501.89
	16-Dec	0940	17-Dec	1106	-333.19	-496.19
	19-Jan	1025	19-Jan	1102	-346.70	-499.81

Table 4 Diurnal variation of isotopic ratio of methane, carbon dioxide and nitrous oxide observed during the three stages of rice growth.

Time(local)	CH4 dC13	CO2 dC13	dO18	N2O dN15	dO18	soil temp deg C
5/15/99 (2nd day of rice transplanting)						
940	-46.59	-12.90	8.66	-345.44	-492.91	12
1340	-46.33	-9.43	9.58	-346.35	-493.53	14
1650	-46.18	-9.21	9.91	-344.39	-487.69	10
1925	-46.71	-11.25	9.65	-347.56	-498.01	6
2055	-43.88	-8.31	9.10	-346.29	-496.75	6
5/22/99 (9th day of rice transplanting)						
815	-47.72	-12.12	9.45	-340.13	-490.06	12
1200	-48.22	-10.54	9.50	-340.97	-490.34	15
1500	-46.65	-9.59	8.71	-342.86	-490.17	20
1700	-47.79	-11.06	9.43	-343.14	-482.99	20
1900	-47.52	-11.51	9.21	-341.39	-489.75	16
2100	-46.50	-10.72	9.88	-341.06	-488.36	14
6/20/99_38th day of rice transplanting)						
820	-16.50	-14.72	7.20	-341.46	-490.52	18
1200	-15.57	-12.35	8.61	-339.10	-488.11	23
1500	-12.68	-10.04	9.77	-335.27	-485.74	23
1700	-14.93	-11.08	8.83	-340.79	-489.21	20
1900	-15.19	-13.31	8.72	-342.56	-491.55	18
2100	-17.24	-15.49	7.14	-344.08	-491.89	16
8/15/99 (93th day of rice transplanting)						
830	-30.26	-17.32	6.67	-325.17	-489.24	26
1035	-30.19	-17.64	6.05	-323.32	-489.06	27
1330	-30.67	-16.61	6.20	-317.99	-491.01	29
1515	-30.89	-10.51	9.25	-323.77	-487.76	29
1715	-31.13	-14.38	6.87	-314.70	-490.32	28
1900	-32.47	-12.16	9.70	-321.19	-489.72	27
2050	-32.91	-15.24	7.01	-322.00	-489.87	26

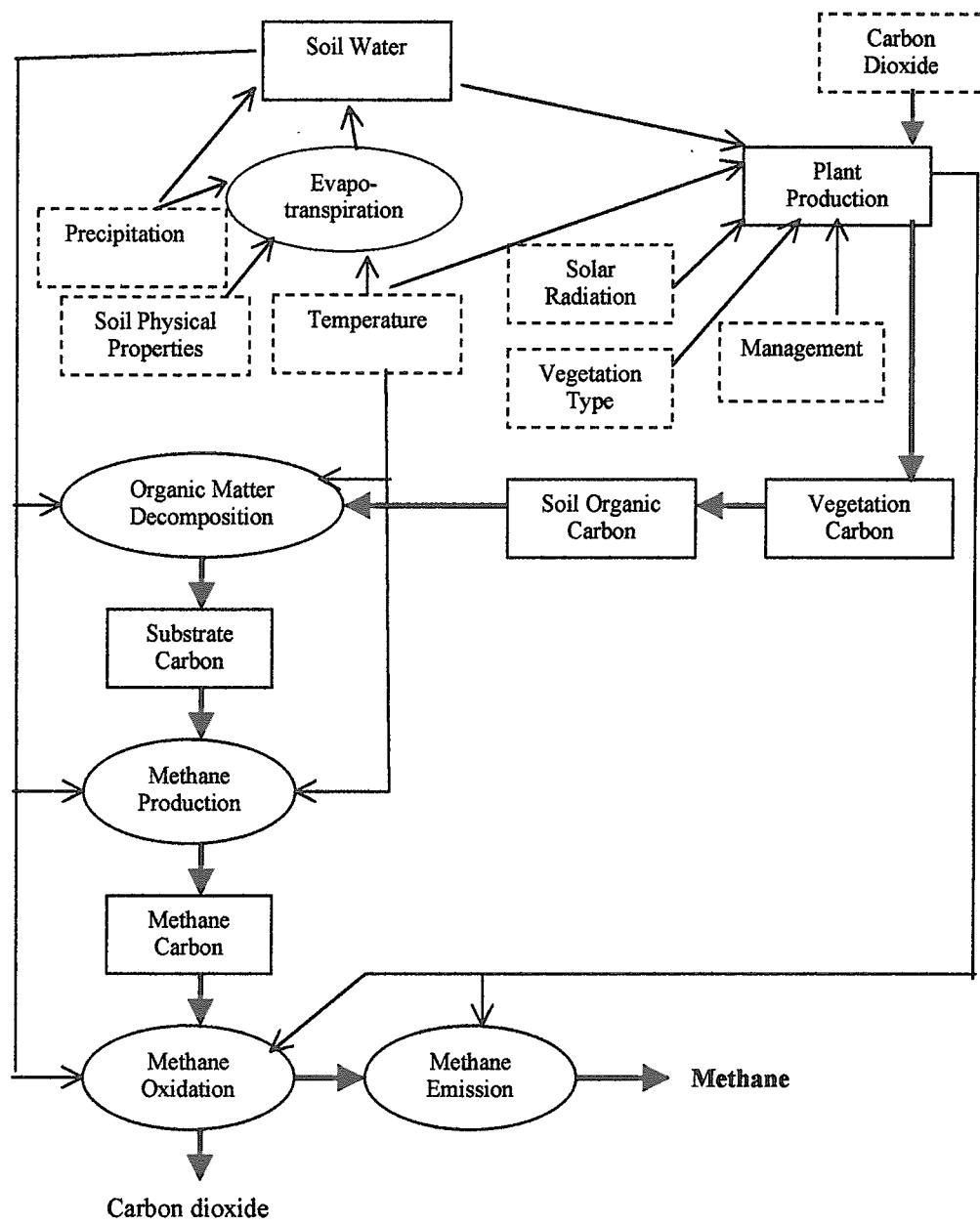


Fig 1 Schematic representation of the methane emission [10]

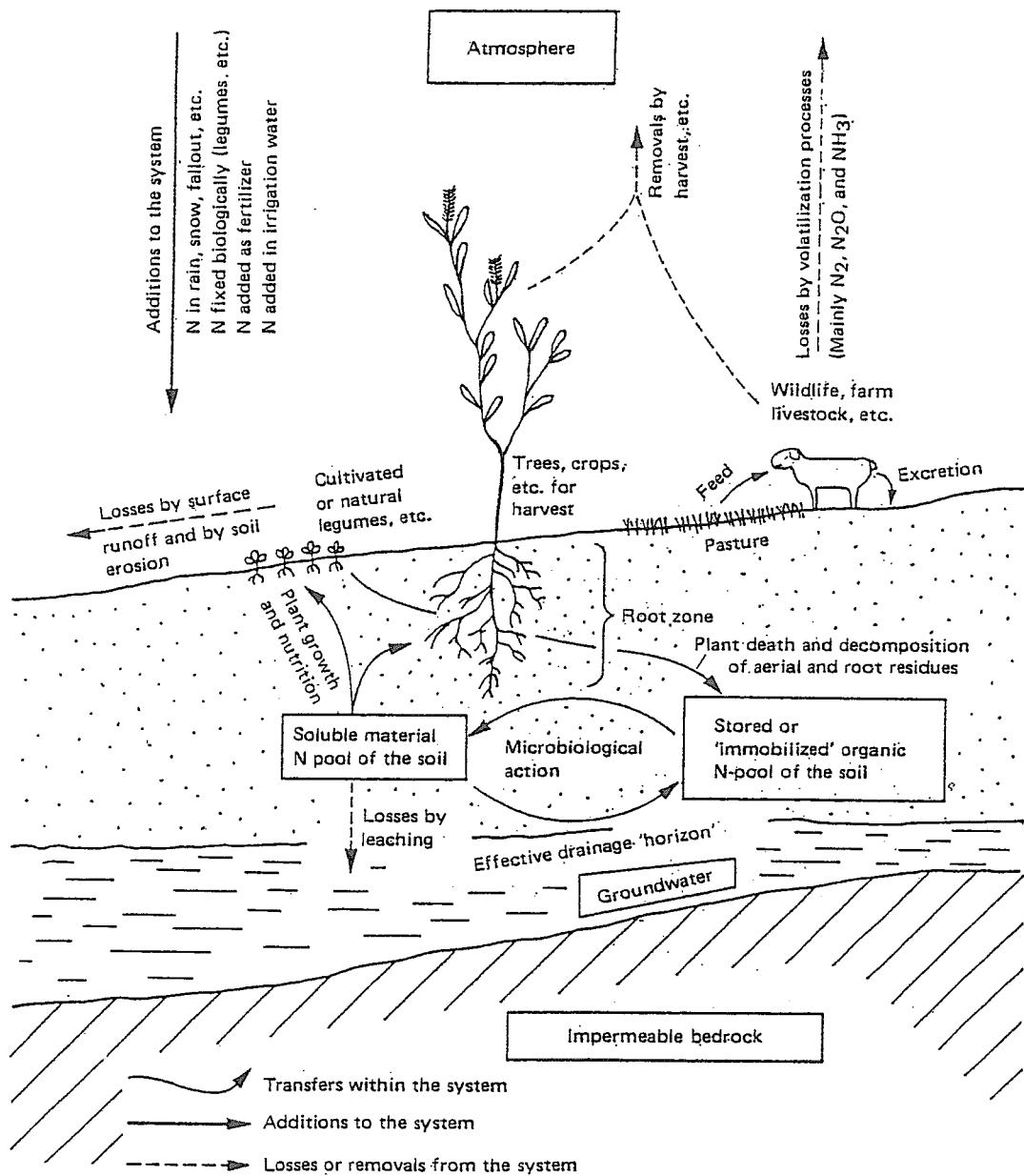
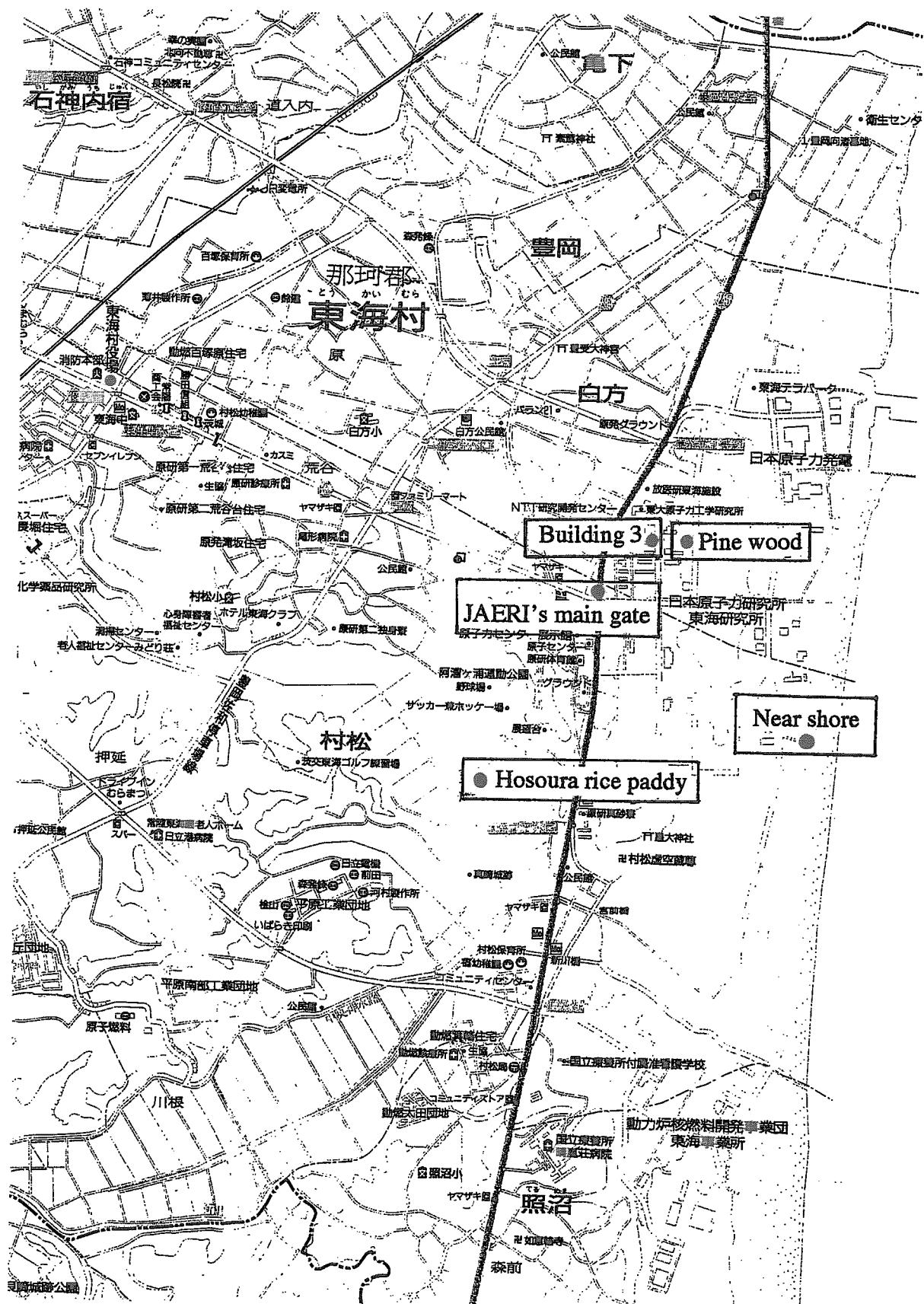


Fig 2 Simplified diagram of nitrogen transfer, removals and losses of the agro-ecosystem [15]



**Fig. 3** Sampling sites at Tokai-mura

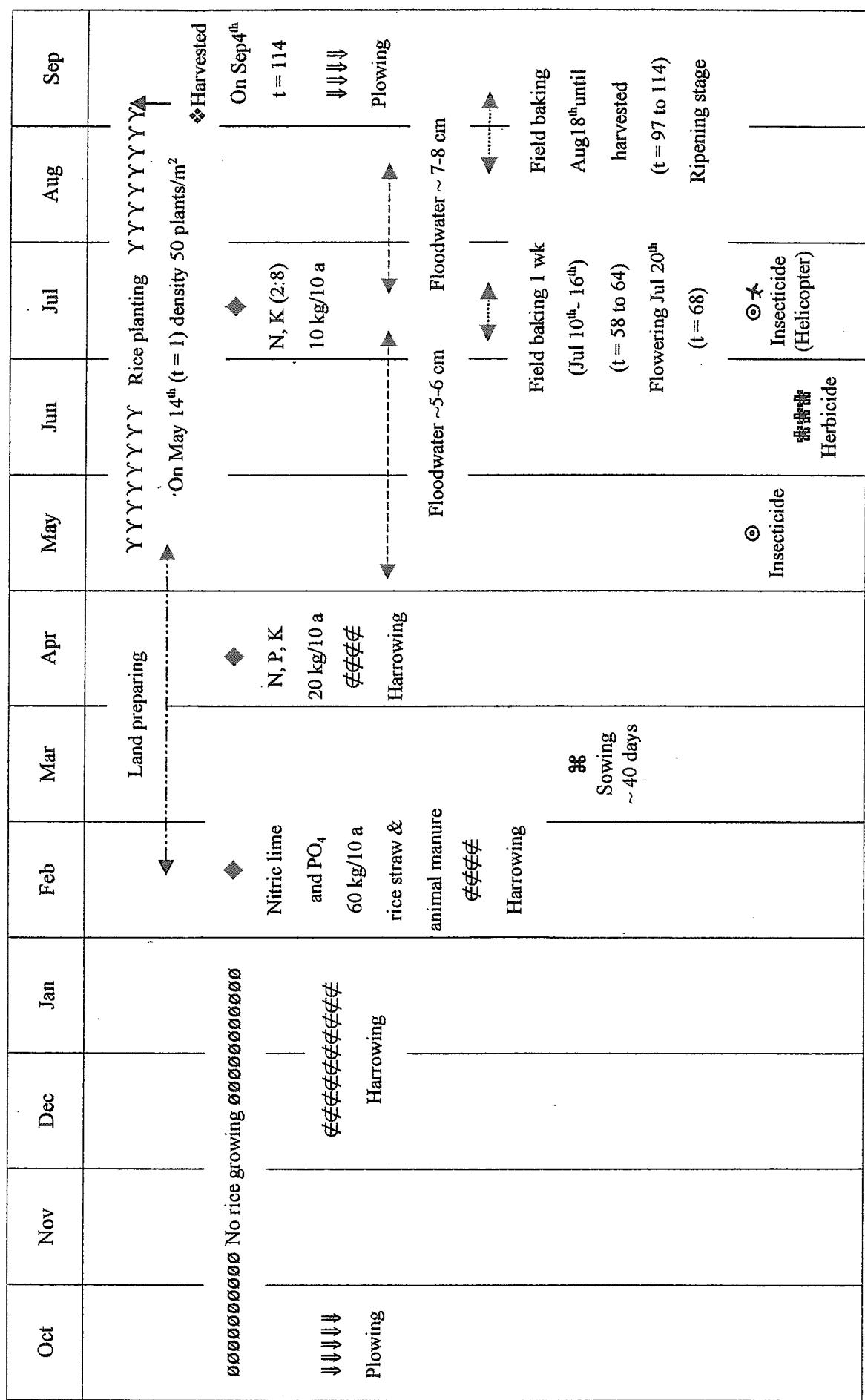
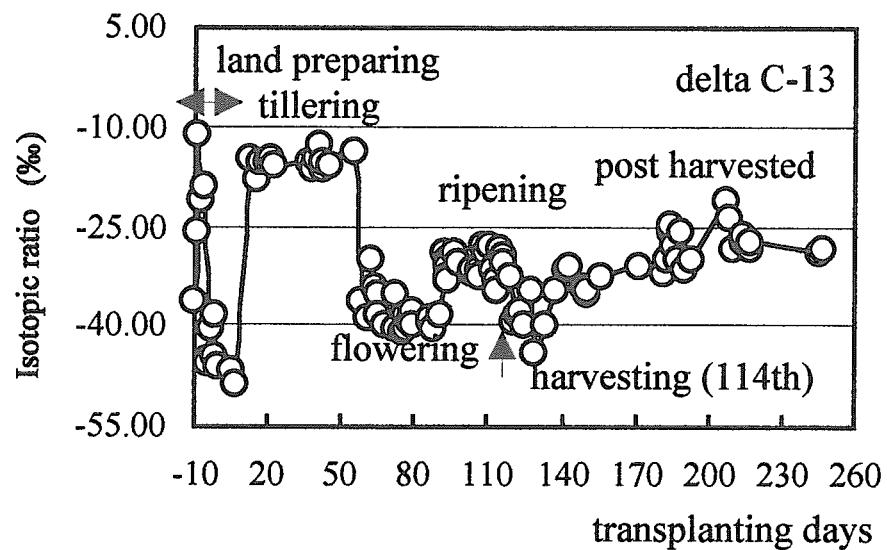
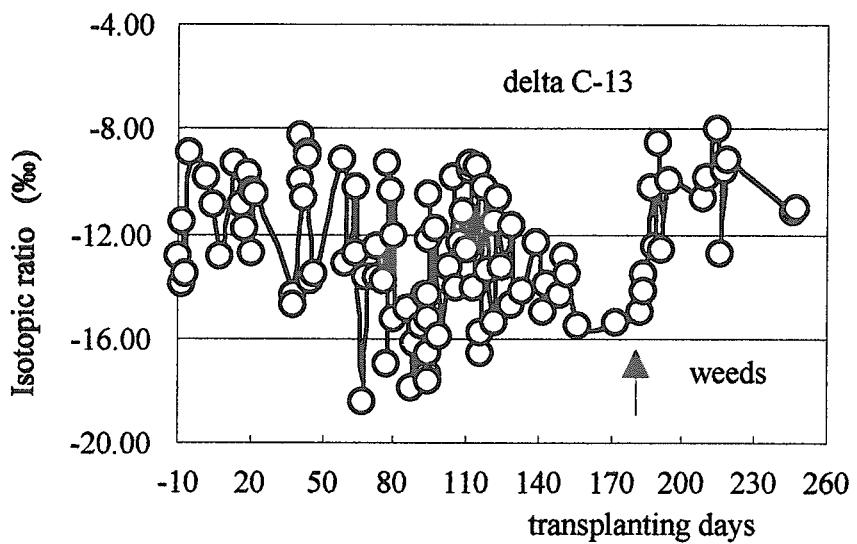
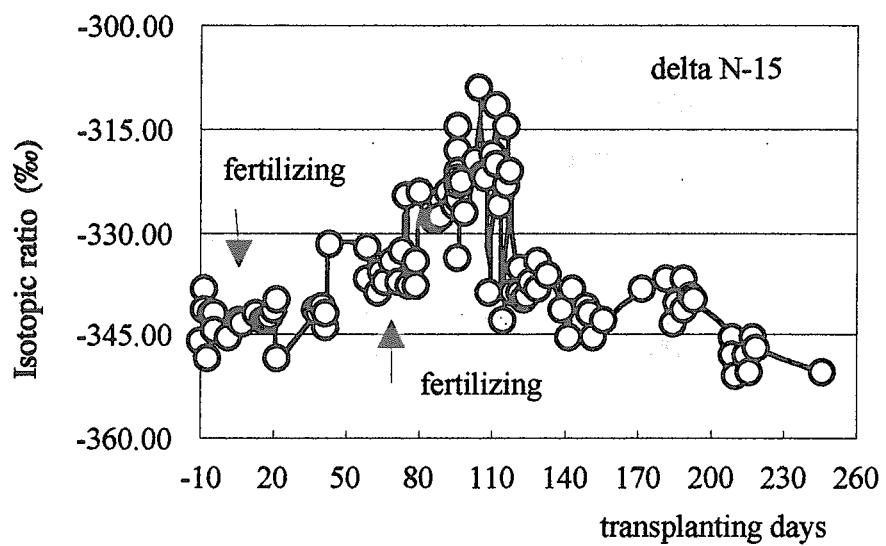
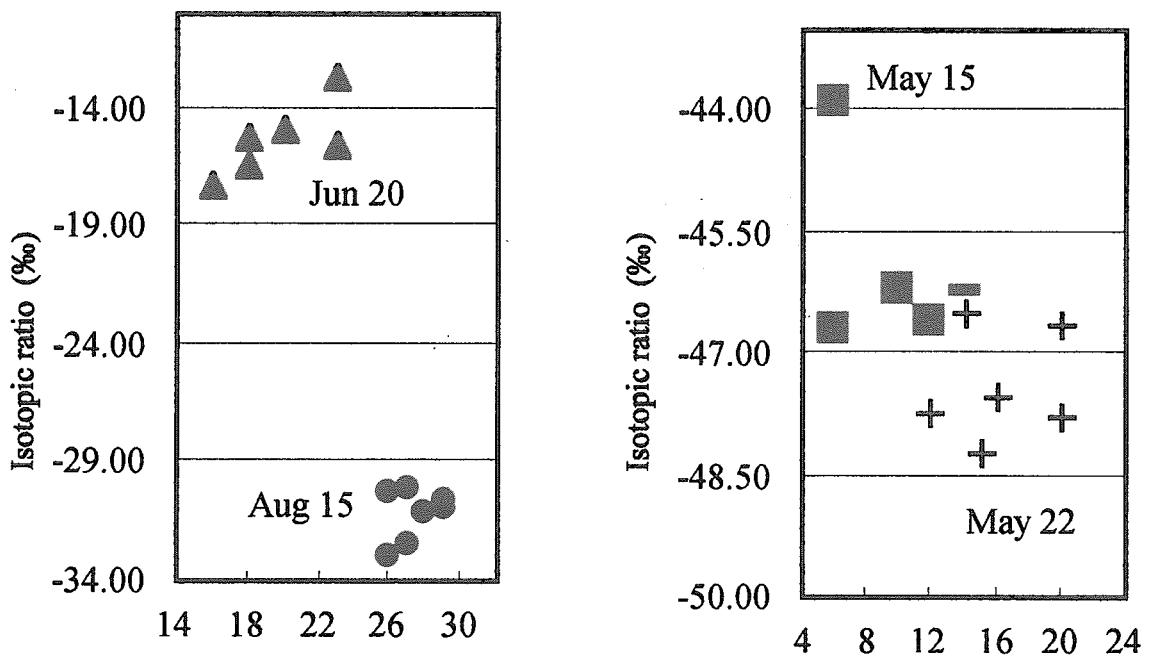


Fig 4 Cultivation practice of the year 1999 rice growing period on Hosoura rice paddy.

Fig 5. Seasonal fluctuation of stable isotope ratios of  $\delta^{13}\text{CH}_4$  in Hosoura rice paddyFig 6. Seasonal fluctuation of stable isotope ratios of  $\delta^{13}\text{CO}_2$  in Hosoura rice paddy

Fig. 7 Seasonal fluctuation of stable isotope ratios of  $\delta^{15}\text{N}_2\text{O}$  in Hosoura rice paddyFig. 8 Diurnal cycle of  $\delta^{13}\text{CH}_4$  in Hosoura rice paddy with dependence on soil temperature at 5 cm depth

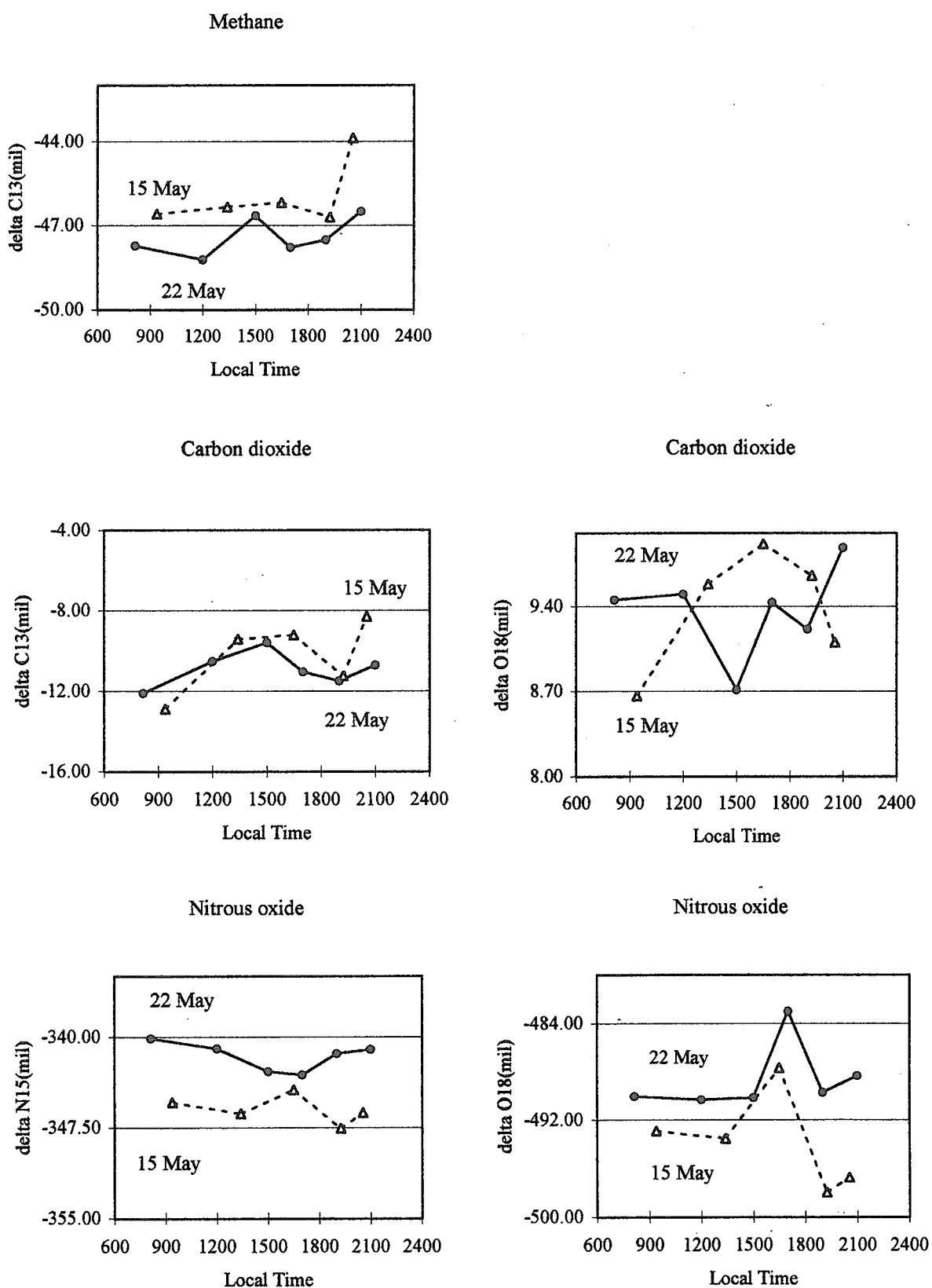


Fig 9 Diurnal variation of methane, carbon dioxide and nitrous oxide observed on May 15 and May 22, 1999 at the rice paddy.

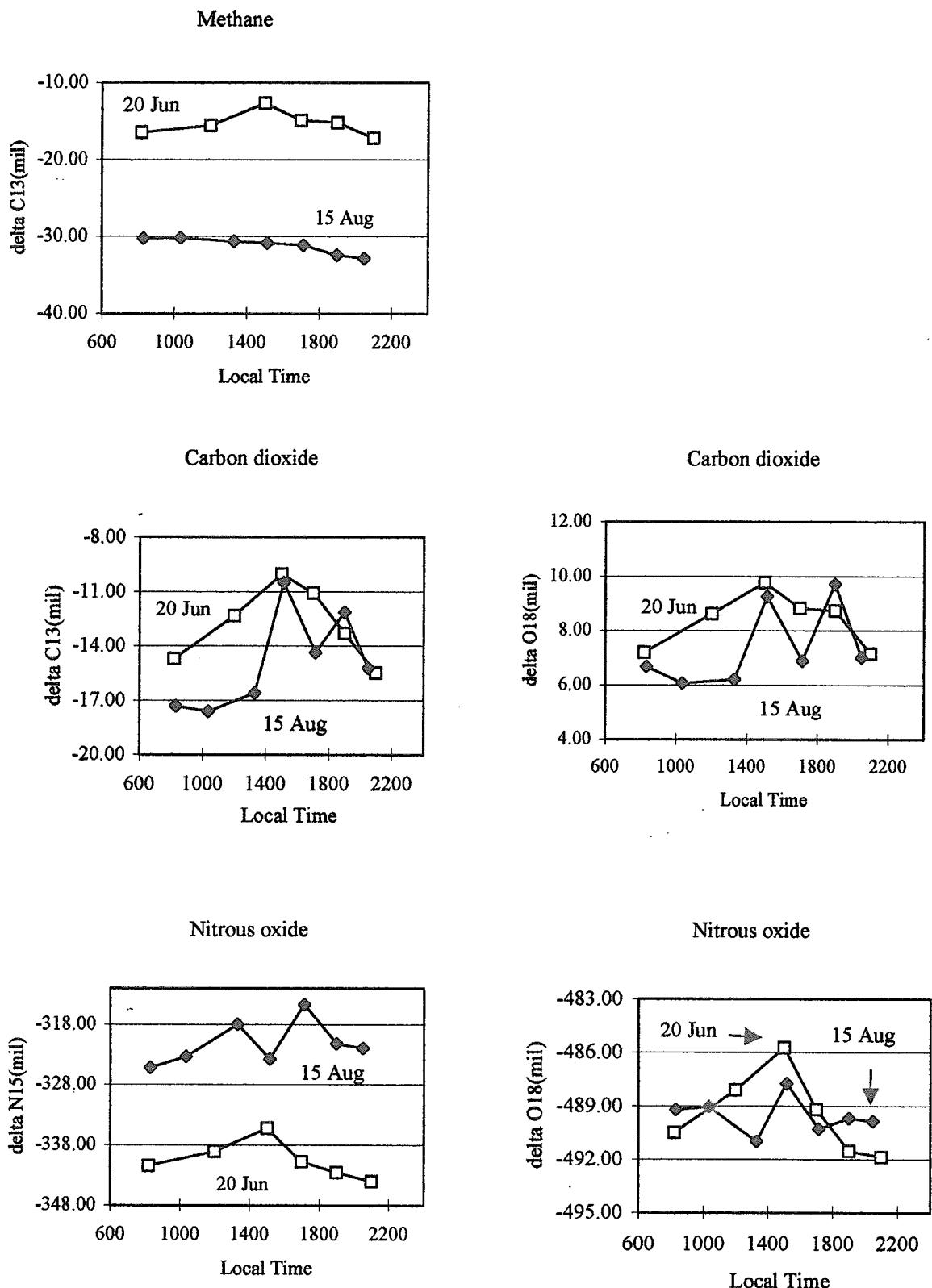


Fig. 10 Diurnal variation of methane, carbon dioxide and nitrous oxide observed on June 20 and August 15, 1999 at the rice paddy.

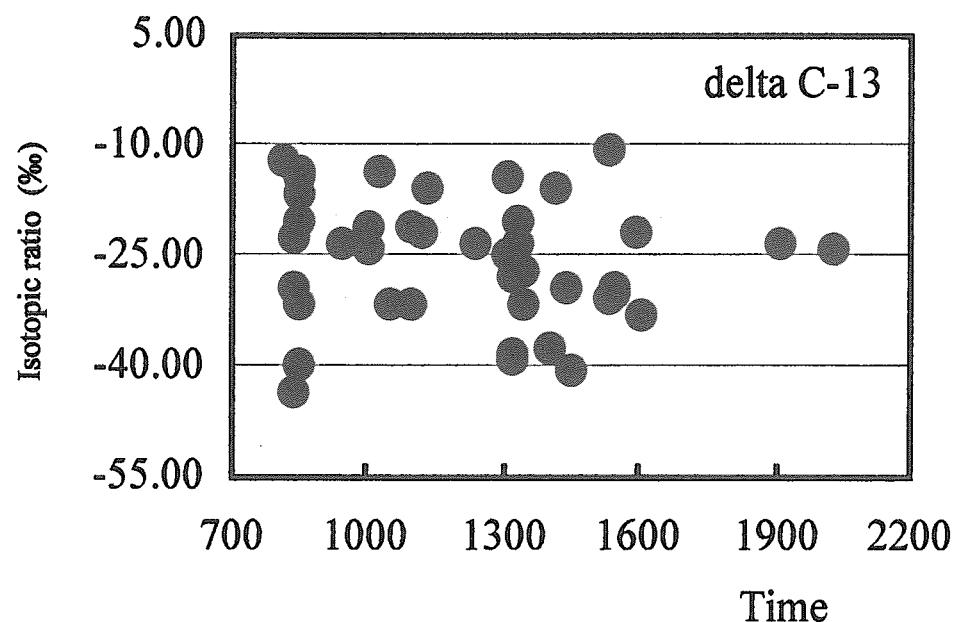


Fig. 11 Seasonal fluctuation of stable isotope ratios of  $\delta^{13}\text{CH}_4$  in JAERI's main gate

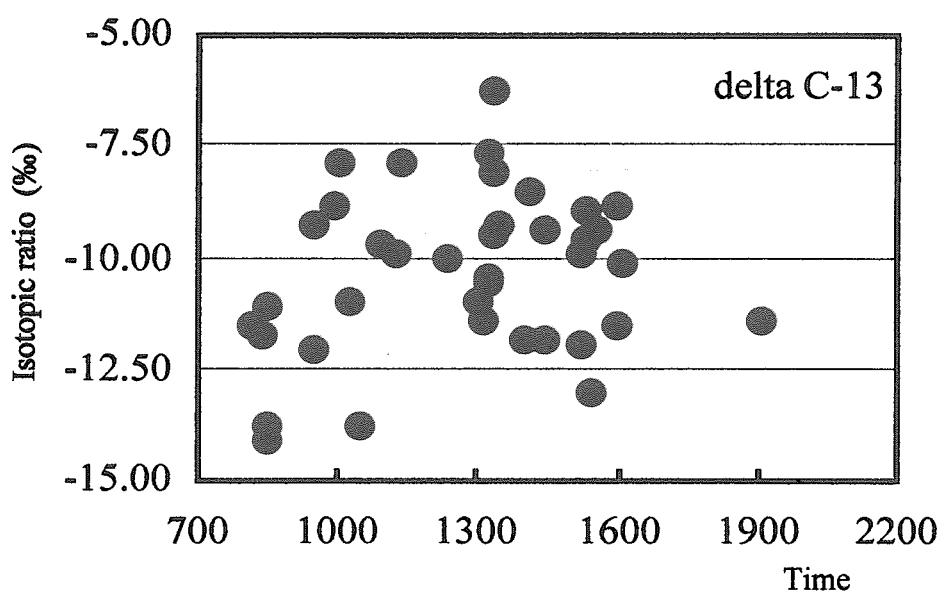


Fig. 12 Seasonal fluctuation of stable isotope ratios of  $\delta^{13}\text{CO}_2$  in JAERI's main gate

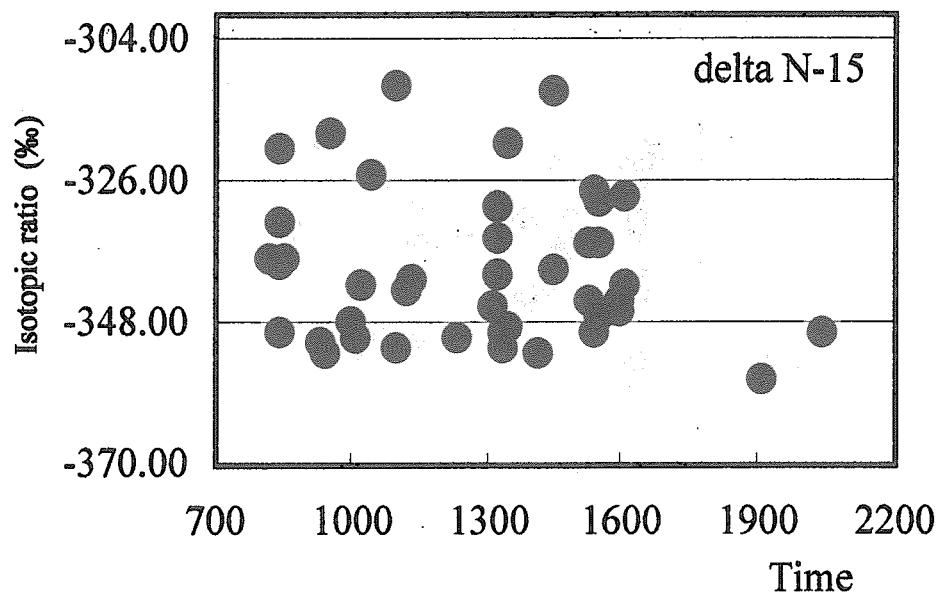


Fig. 13 Seasonal fluctuation of stable isotope ratios of  $\delta^{15}\text{N}_2\text{O}$  in JAERI's main gate

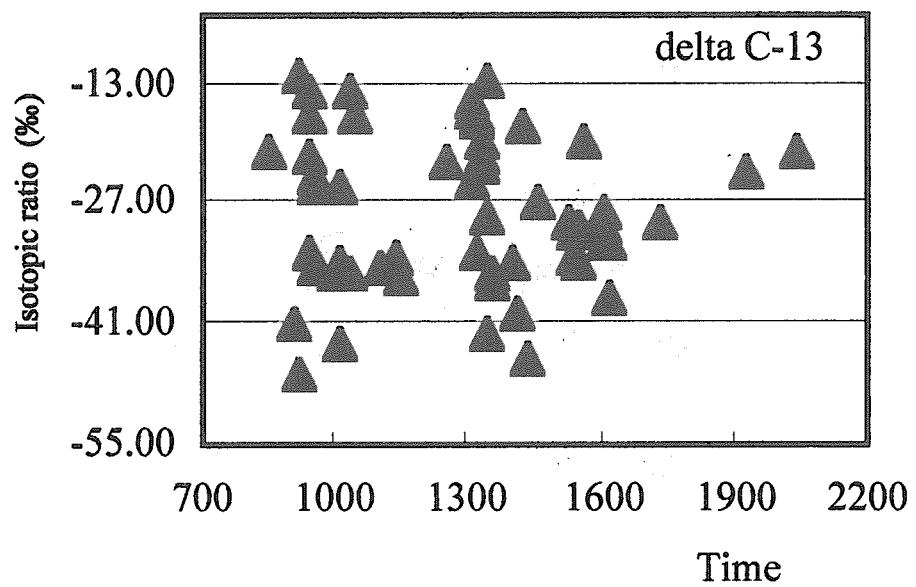


Fig. 14 Seasonal fluctuation of stable isotope ratios of  $\delta^{13}\text{CH}_4$  in Pine wood

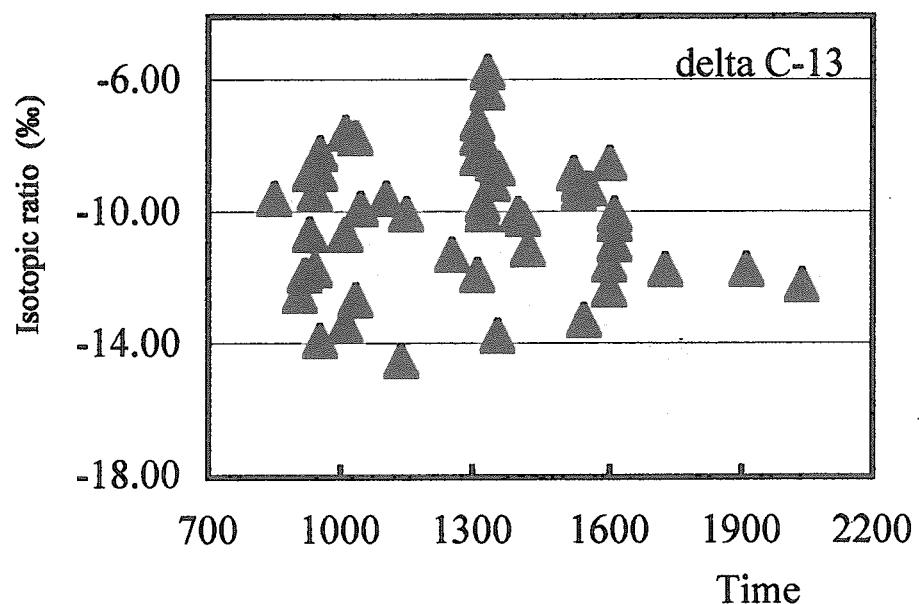


Fig. 15 Seasonal fluctuation of stable isotope ratios of  $\delta^{13}\text{CO}_2$  in Pine wood

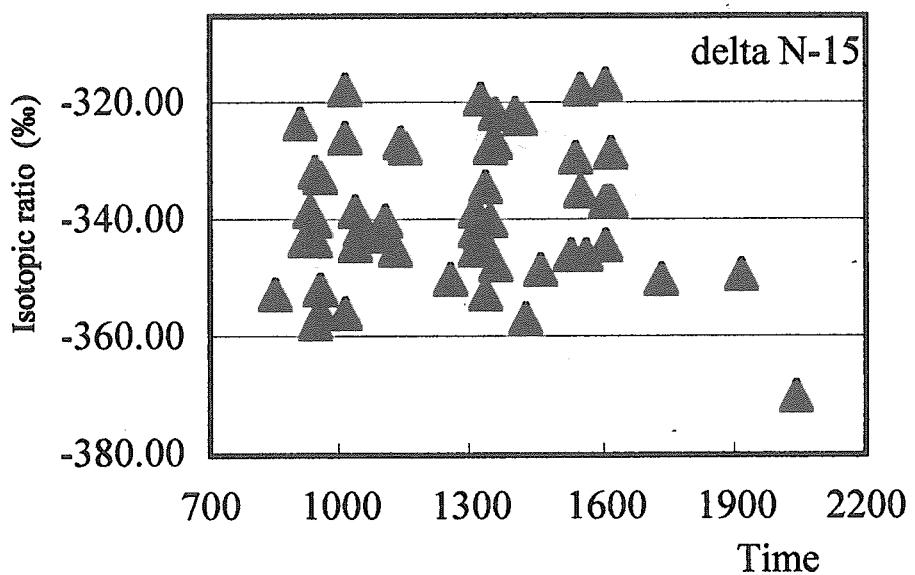
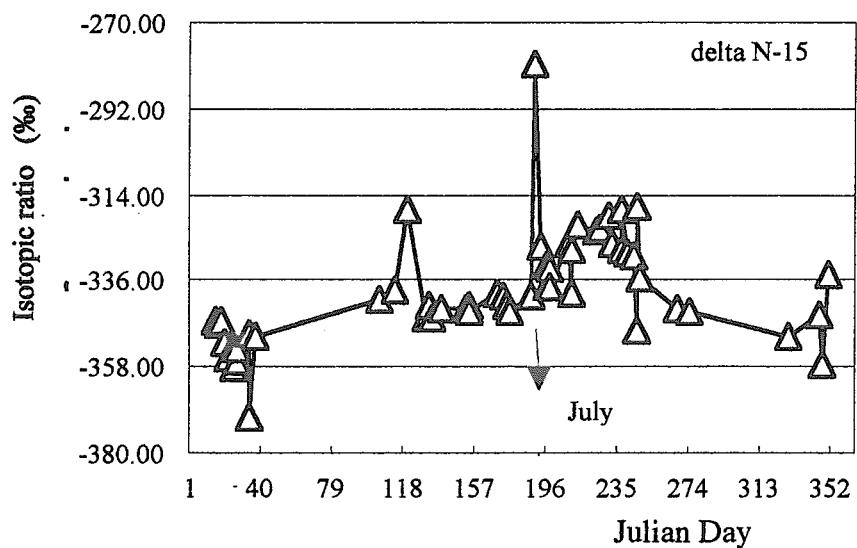
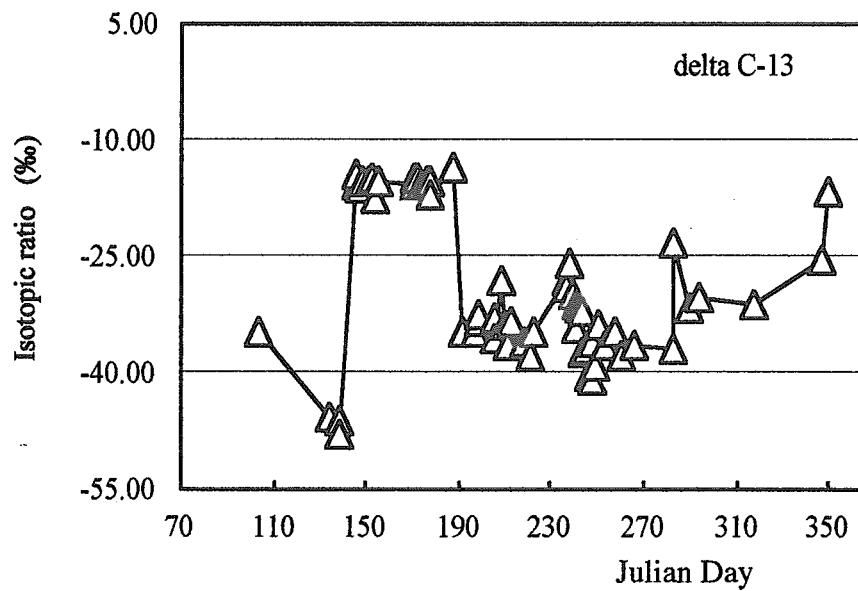


Fig. 16 Seasonal fluctuation of stable isotope ratios of  $\delta^{15}\text{N}_2\text{O}$  in Pine wood

Fig. 17 Seasonal fluctuation of stable isotope ratios of  $\delta^{15}\text{N}_2\text{O}$  in Pine woodFig. 18 Seasonal fluctuation of stable isotope ratios of  $\delta^{13}\text{CH}_4$  in near shore the Pacific ocean

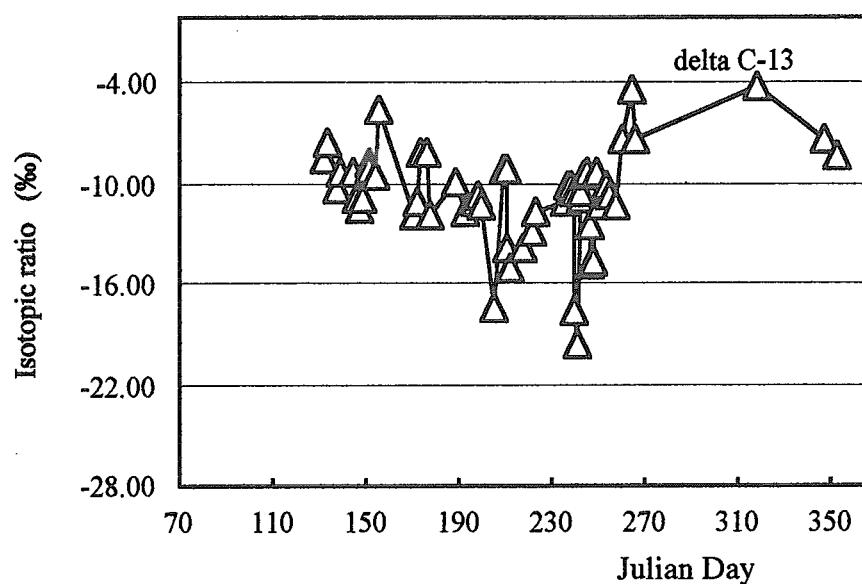


Fig. 19 Seasonal fluctuation of stable isotope ratios of  $\delta^{13}\text{CO}_2$  near shore the Pacific Ocean

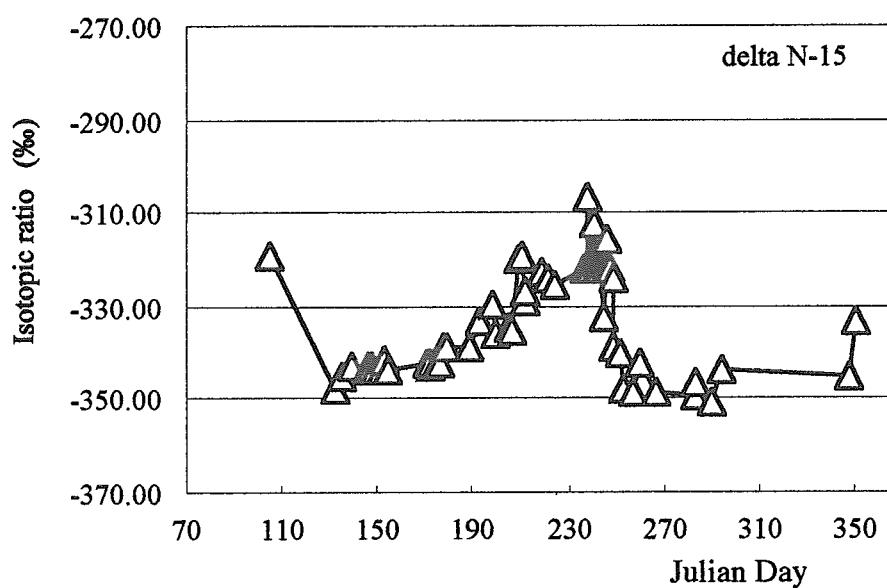


Fig. 20 Seasonal fluctuation of stable isotope ratios of  $\delta^{15}\text{N}_2\text{O}$  near shore the Pacific ocean

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## 国際単位系(SI)と換算表

表1 SI基本単位および補助単位

量	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd
平面角	ラジアン	rad
立体角	ステラジアン	sr

表3 固有の名称をもつSI組立単位

量	名称	記号	他のSI単位による表現
周波数	ヘルツ	Hz	$s^{-1}$
圧力	ニュートン	N	$m \cdot kg/s^2$
圧力、応力	パスカル	Pa	$N/m^2$
エネルギー、仕事、熱量	ジユール	J	$N \cdot m$
工率、放射束	ワット	W	$J/s$
電気量、電荷	クーロン	C	$A \cdot s$
電位、電圧、起電力	ボルト	V	$W/A$
静電容量	フアラード	F	$C/V$
電気抵抗	オーム	$\Omega$	$V/A$
コンダクタンス	ジーメンス	S	$A/V$
磁束密度	ウエーバ	Wb	$V \cdot s$
磁束密度	テスラ	T	$Wb/m^2$
インダクタンス	ヘンリー	H	$Wb/A$
セルシウス温度	セルシウス度	°C	
光束度	ルーメン	lm	$cd \cdot sr$
照度	ルクス	lx	$lm/m^2$
放射能	ベクレル	Bq	$s^{-1}$
吸収線量	グレイ	Gy	$J/kg$
線量等量	シーベルト	Sv	$J/kg$

表2 SIと併用される単位

名 称	記 号
分、時、日	min, h, d
度、分、秒	°, ', "
リットル	L, L
トン	t
電子ボルト	eV
原子質量単位	u

$$1 \text{ eV} = 1.60218 \times 10^{-19} \text{ J}$$

$$1 \text{ u} = 1.66054 \times 10^{-27} \text{ kg}$$

表5 SI接頭語

倍数	接頭語	記号
$10^{18}$	エクサ	E
$10^{15}$	ペタ	P
$10^{12}$	テラ	T
$10^9$	ギガ	G
$10^6$	メガ	M
$10^3$	キロ	k
$10^2$	ヘクト	h
$10^1$	デカ	da
$10^{-1}$	デシ	d
$10^{-2}$	センチ	c
$10^{-3}$	ミリ	m
$10^{-6}$	マイクロ	μ
$10^{-9}$	ナノ	n
$10^{-12}$	ピコ	p
$10^{-15}$	フェムト	f
$10^{-18}$	アト	a

表4 SIと共に暫定的に維持される単位

名 称	記 号
オングストローム	Å
バーン	b
バール	bar
ガル	Gal
キュリー	Ci
レントゲン	R
ラド	rad
レム	rem

$$1 \text{ Å} = 0.1 \text{ nm} = 10^{-10} \text{ m}$$

$$1 \text{ b} = 100 \text{ fm}^2 = 10^{-28} \text{ m}^2$$

$$1 \text{ bar} = 0.1 \text{ MPa} = 10^5 \text{ Pa}$$

$$1 \text{ Gal} = 1 \text{ cm/s}^2 = 10^{-2} \text{ m/s}^2$$

$$1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$$

$$1 \text{ R} = 2.58 \times 10^{-4} \text{ C/kg}$$

$$1 \text{ rad} = 1 \text{ cGy} = 10^{-2} \text{ Gy}$$

$$1 \text{ rem} = 1 \text{ cSv} = 10^{-2} \text{ Sv}$$

(注)

1. 表1~5は「国際単位系」第5版、国際度量衡局1985年刊行による。ただし、1eVおよび1uの値はCODATAの1986年推奨値によった。

2. 表4には海里、ノット、アール、ヘクタールも含まれているが日常の単位なのでここでは省略した。

3. barは、JISでは流体の圧力を表わす場合に限り表2のカテゴリーに分類されている。

4. EC閣僚理事会指令ではbar、barnおよび「血圧の単位」mmHgを表2のカテゴリーに入れている。

## 換 算 表

力	N(=10 <sup>5</sup> dyn)	kgf	lbf
1	0.101972	0.224809	
9.80665	1	2.20462	
4.44822	0.453592	1	

$$\text{粘度 } 1 \text{ Pa} \cdot \text{s} (\text{N} \cdot \text{s}/\text{m}^2) = 10 \text{ P} (\text{ポアズ})(\text{g}/(\text{cm} \cdot \text{s}))$$

$$\text{動粘度 } 1 \text{ m}^2/\text{s} = 10^4 \text{ St} (\text{ストークス})(\text{cm}^2/\text{s})$$

圧力	MPa(=10bar)	kgf/cm <sup>2</sup>	atm	mmHg(Torr)	lbf/in <sup>2</sup> (psi)
1	10.1972	9.86923	7.50062 × 10 <sup>3</sup>	145.038	
0.0980665	1	0.967841	735.559	14.2233	
0.101325	1.03323	1	760	14.6959	
1.33322 × 10 <sup>-4</sup>	1.35951 × 10 <sup>-3</sup>	1.31579 × 10 <sup>-3</sup>	1	1.93368 × 10 <sup>-2</sup>	
6.89476 × 10 <sup>-3</sup>	7.03070 × 10 <sup>-2</sup>	6.80460 × 10 <sup>-2</sup>	51.7149	1	

エネルギー・仕事・熱量	J(=10 <sup>7</sup> erg)	kgf·m	kW·h	cal(計量法)	Btu	ft·lbf	eV
1	0.101972	2.77778 × 10 <sup>-7</sup>	0.238889	9.47813 × 10 <sup>-4</sup>	0.737562	6.24150 × 10 <sup>18</sup>	
9.80665	1	2.72407 × 10 <sup>-6</sup>	2.34270	9.29487 × 10 <sup>-3</sup>	7.23301	6.12082 × 10 <sup>19</sup>	
3.6 × 10 <sup>6</sup>	3.67098 × 10 <sup>5</sup>	1	8.59999 × 10 <sup>5</sup>	3412.13	2.65522 × 10 <sup>6</sup>	2.24694 × 10 <sup>25</sup>	
4.18605	0.426858	1.16279 × 10 <sup>-6</sup>	1	3.96759 × 10 <sup>-3</sup>	3.08747	2.61272 × 10 <sup>19</sup>	
1055.06	107.586	2.93072 × 10 <sup>-4</sup>	252.042	1	778.172	6.58515 × 10 <sup>21</sup>	
1.35582	0.138255	3.76616 × 10 <sup>-7</sup>	0.323890	1.28506 × 10 <sup>-3</sup>	1	8.46233 × 10 <sup>18</sup>	
1.60218 × 10 <sup>-19</sup>	1.63377 × 10 <sup>-20</sup>	4.45050 × 10 <sup>-26</sup>	3.82743 × 10 <sup>-20</sup>	1.51857 × 10 <sup>-22</sup>	1.18171 × 10 <sup>-19</sup>	1	

1 cal = 4.18605 J (計量法)  
= 4.184 J (熱化学)  
= 4.1855 J (15°C)  
= 4.1868 J (国際蒸気表)  
仕事率 1 PS(仏馬力)  
= 75 kgf·m/s  
= 735.499 W

放射能	Bq	Ci	吸収線量	Gy	rad
1	2.70270 × 10 <sup>-11</sup>	1	1	100	
3.7 × 10 <sup>10</sup>	1		0.01	1	

照射線量	C/kg	R
1	3876	
2.58 × 10 <sup>-4</sup>	1	

線量当量	Sv	rem
1	100	
0.01	1	

