TAO, Vol.6, No.3, 367-377, September 1995

Methane Emission in Large Cities

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(Manuscript received 5 July 1994, in final form 25 September 1994)

ABSTRACT

The mixing ratio of atmospheric methane (CH_4) over the two large cities of Osaka and Kyoto achieves its maximum in the central part of the city and decreases toward the suburbs, with the same distribution patterns for non-methane hydrocarbons (NMHC) and carbon monoxide (CO).

It is shown that CH_4 is emitted from the same sources as NMHC and CO, whose contribution amounts to about 4% of the yearly mean of observed methane. The emission ratio of CH_4 to total hydrocarbons is estimated to be 8.3±1.0%, calculated in terms of carbon number. The emission ratio of CH_4 to CO is estimated to be 0.034±0.006 in volume.

(Key words: Atmospheric methane, Urban emission, Distribution Kyoto, Japan)

1. INTRODUCTION

Atmospheric methane (CH_4), though its mixing ratio is about 1.8 ppmv, plays important roles in the chemistry of the troposphere and stratosphere and in the climate of the earth. In the troposphere, CH_4 and CO control the concentration of hydroxyl radical (OH) (Levy, 1972). Hydroxyl radicals remove many chlorofluoromethanes which, if transported into the stratosphere, destroy stratospheric ozone. Oxidation of CH_4 in the presence of NO can be the source of ozone (Crutzen, 1973). In the stratosphere, methane mitigates the ozone destroying effect of chlorofluoromethanes by terminating the odd chlorine cycle (Molina and Rowland, 1974) and is the source of water vapor. Methane contributes to global warming by absorbing outgoing terrestrial radiation (Wang *et al.*, 1976).

The major sources of atmospheric CH_4 are enteric fermentation, natural wetlands, rice paddies, biomass burning, coal mining and natural gas exploration and transmission (Cicerone and Oremland, 1989). The mixing ratio of CH_4 in the atmosphere shows a significant increase which amounts to $1\%\mathrm{yr}^{-1}$ (Ehhalt, 1988). A quantitative explanation of the contributions of individual sources to the increase in atmospheric CH_4 is required. Increasing human activities which bring about a simultaneous increase in the utilization of fossil fuels should have an influence on methane emission.

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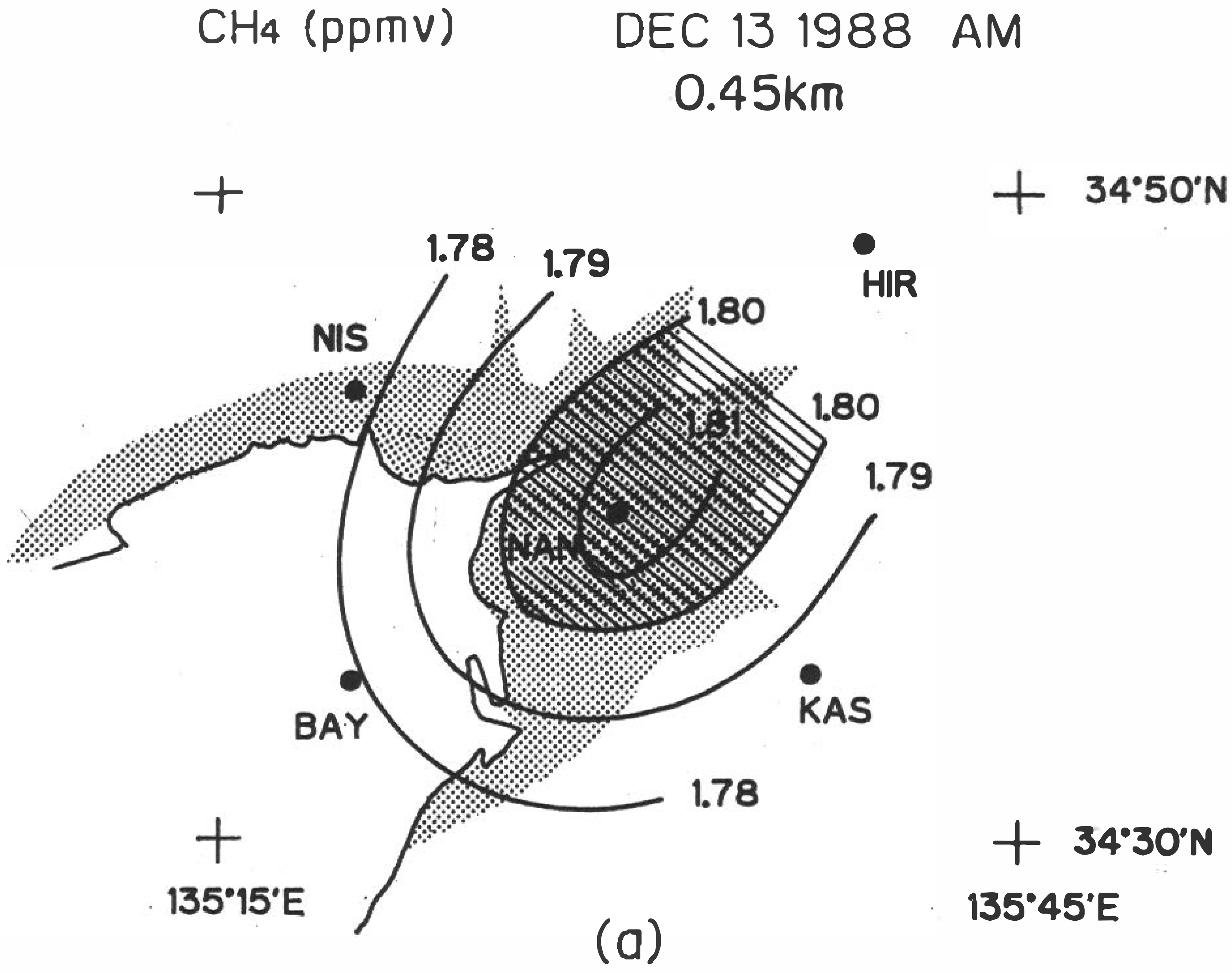

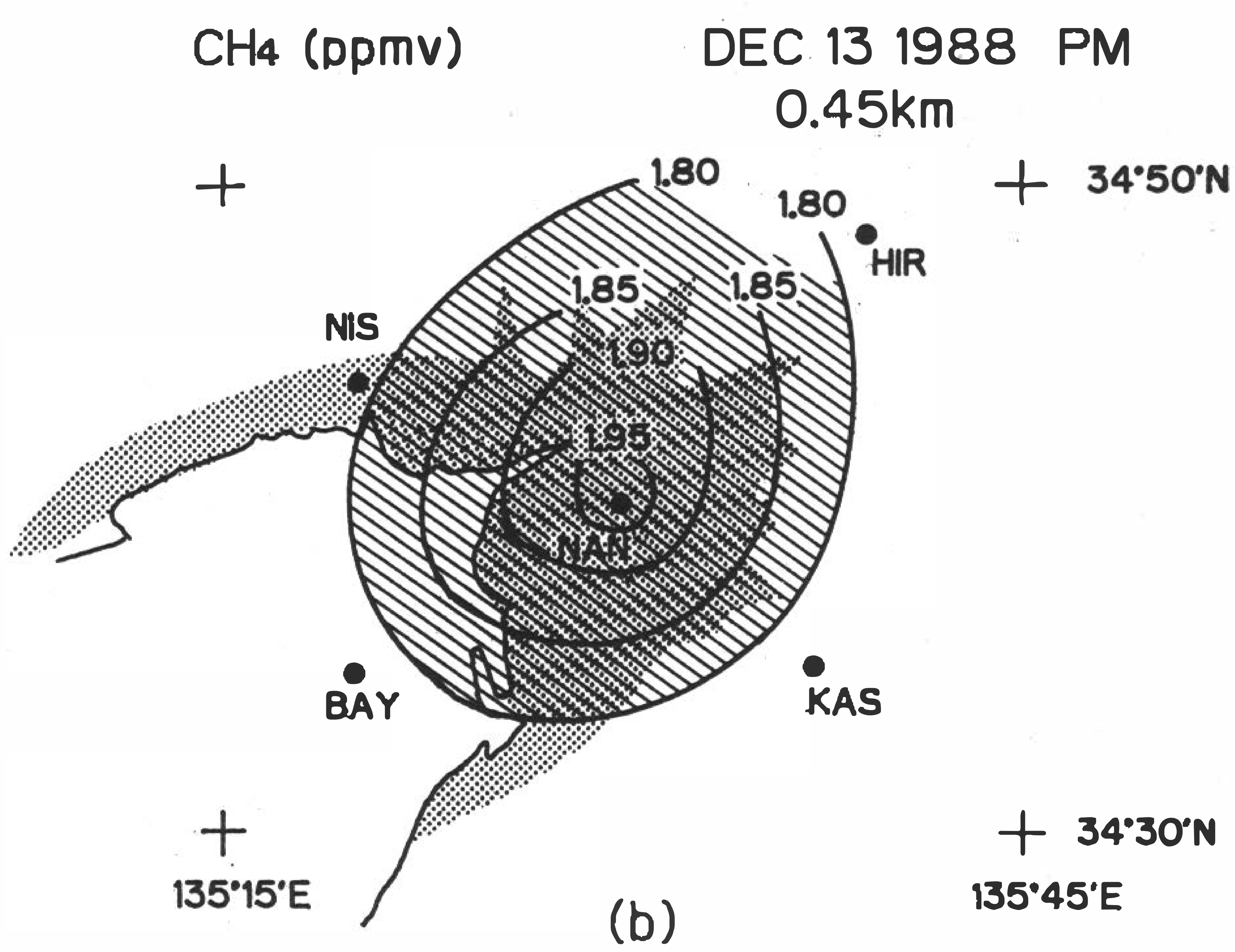

Fig. 1. **Methane distributions over Osaka and Kobe in the morning** (a), and afternoon (b) of December 13, 1988. The areas where CH_4 mixing ratios are higher than 1.80 ppmv are shown by slanting lines. The most densely populated areas are stippled (from Muramatsu, 1989).

In order to estimate the emission related to human activities, this author observed the distribution of CH_4 over Osaka and Kobe (Muramatsu, 1989). The horizontal distributions of CH_4 at the altitude of 0.45 km are shown in Figure 1. Here the maximum CH_4 is at the central part of Osaka both in the morning (Figure 1(a)) and afternoon (Figure 1(b)), and the mixing ratio of CH_4 is higher in the afternoon than in the morning. The vertical distributions of CH_4 over the central part of Osaka (Nanba) are shown in Figure 2. The CH_4 mixing ratio increases from morning to afternoon below the inversion layer. When upward transport through the inversion layer and horizontal transport were neglected, the release rate of 0.011g $CH_4m^{-2}hr^{-1}$ was estimated and was found to be comparable to the most intense flux from such ecosystems as wetlands or rice fields. In that report, the sources of CH_4 from the observations in Osaka could not be specified.

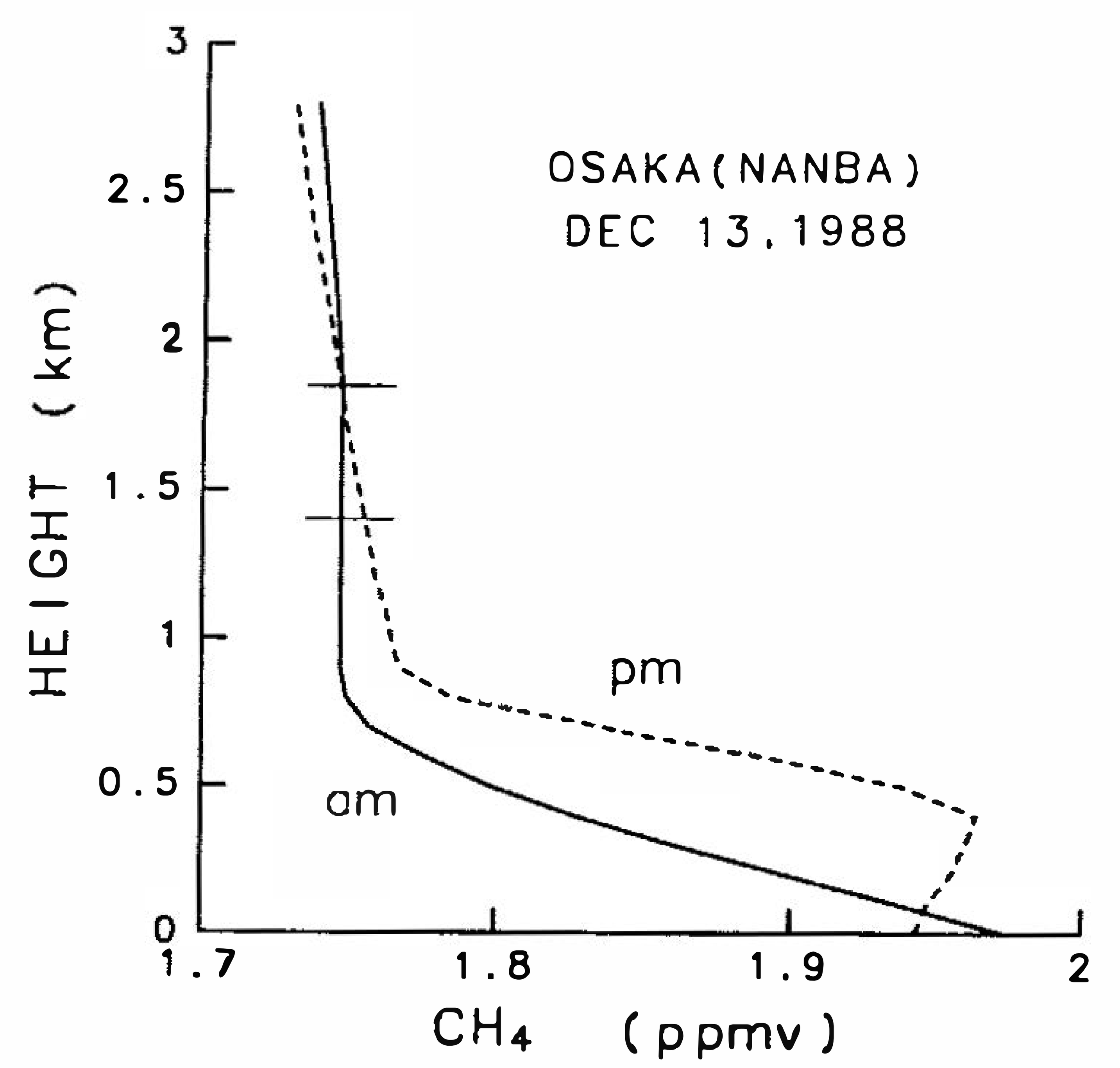

Fig. 2. Vertical distributions of CH_4 over the central part of the City of Osaka in the morning (solid curve) and afternoon (broken curve). The horizontal lines crossing the curves show the boundaries of the temperature inversion (from Muramatsu, 1989).

The aim of this report is to specify the sources of methane in the city. The data of CH_4, NMHC and CO obtained by the Environmental Conservation Office of the Public Health Bureau of the Kyoto City Government are used here. The hourly mean values for seven stations in Kyoto City are employed. In this report, the enhanced mixing ratio of CH_4 in the atmosphere in Kyoto City, the variation of which is closely related to fossil fuel combustion by automobiles is described.

2. DISTRIBUTION OF CH_4, NMHC AND CO IN KYOTO

In order to obtain the general distribution of methane in the Kyoto City, the yearly mean methane distribution in the urban area is shown in Figure 3(a). The observational stations are shown by C_1-C_5 and A_1 and A_2. The maximum of CH_4 mixing ratio is seen in the central part of the densely populated area which is also the district with heavy traffic. The distributions of NMHC and CO for the same period are shown in Figures 3(b) and 3(c), respectively. The maxima of both species are seen at the same location as CH_4. As for CO, a more complete concentric pattern can be obtained with the aid of another observational station, C_6.

Essentially, similar patterns of distribution are obtained for the monthly, daily and hourly means of these three species. Examples of the daily mean distributions of CH_4, NMHC and CO on December 10, 1986 are shown in Figure 4. The locations of the maximum CH_4 and NMHC shift southward from those of the yearly mean, but the maxima are seen at the same place as those of the yearly means when these species attain the highest mixing ratio of that day (Figure 5).

It is concluded that the mixing ratios of CH_4, NMHC and CO reach their highest in the central part of Kyoto City, and they decrease toward the suburbs. This characteristic is seen with the distributions of the yearly, monthly, daily and hourly means. However, the

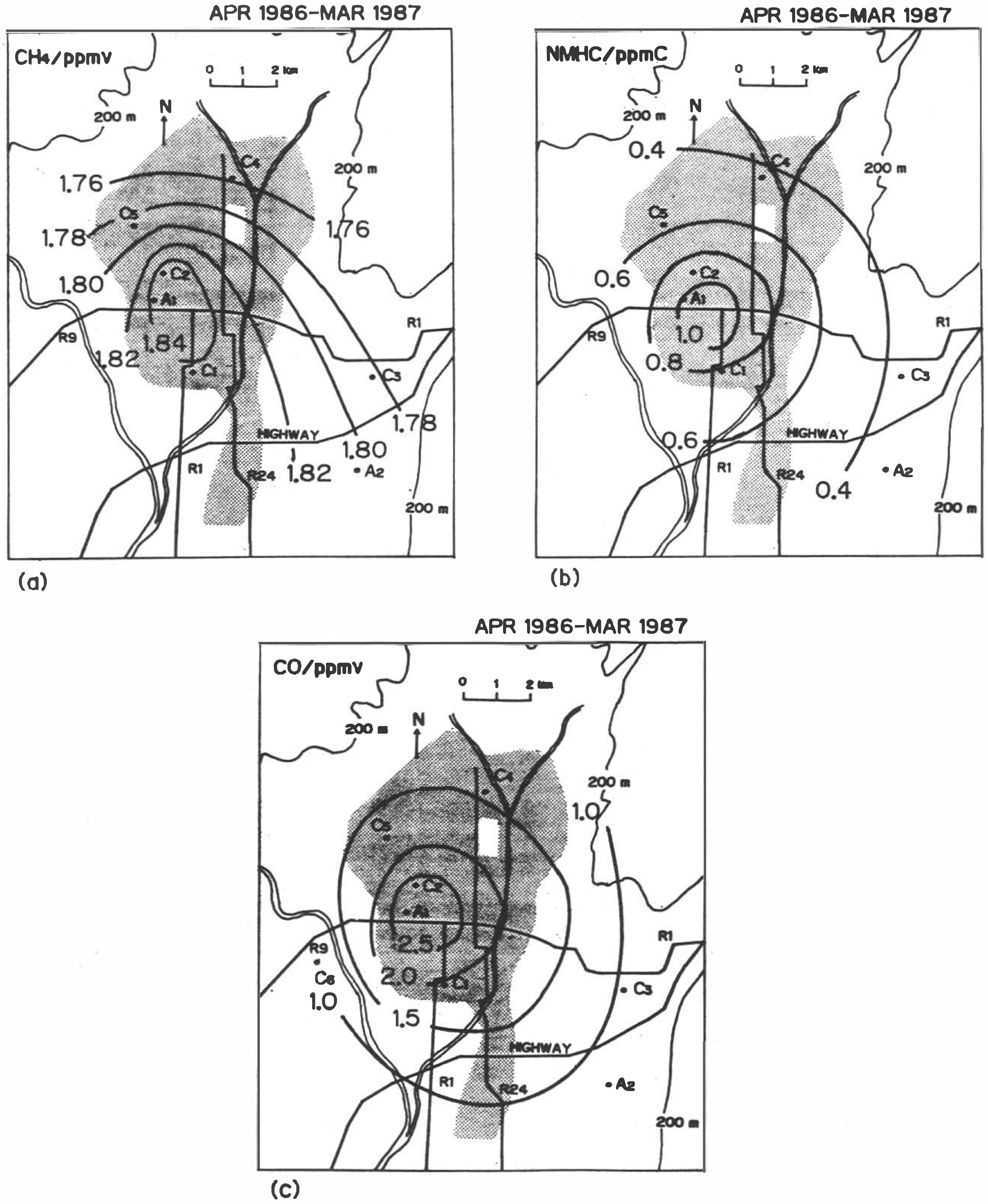

Fig. 3. Yearly mean **distributions in Kyoto for the period** of April 1986 to March 1987 for (a) CH_4, (b) **NMHC and** (c) CO. Symbols A_1, A_2, C_1-C_6 indicate the observational stations. The most densely populated area is stippled. The main national roads (R1, R9, R24 and a highway) are shown.

distortion and shift of the location of the maximums occur with the distributions of the daily and hourly means due to meteorological influences.

3. ADDITIONAL METHANE IN THE CITY

Such high correlations are recognized not only between the spatial distributions of CH_4, NMHC and CO, as shown before, but also between the temporal variations of these species. The observed mixing ratio of CH_4 increases linearly with NMHC as shown in Figure 6(a). When the January 1987 daily mean values for Yamashina (Station C_3) are plotted, the

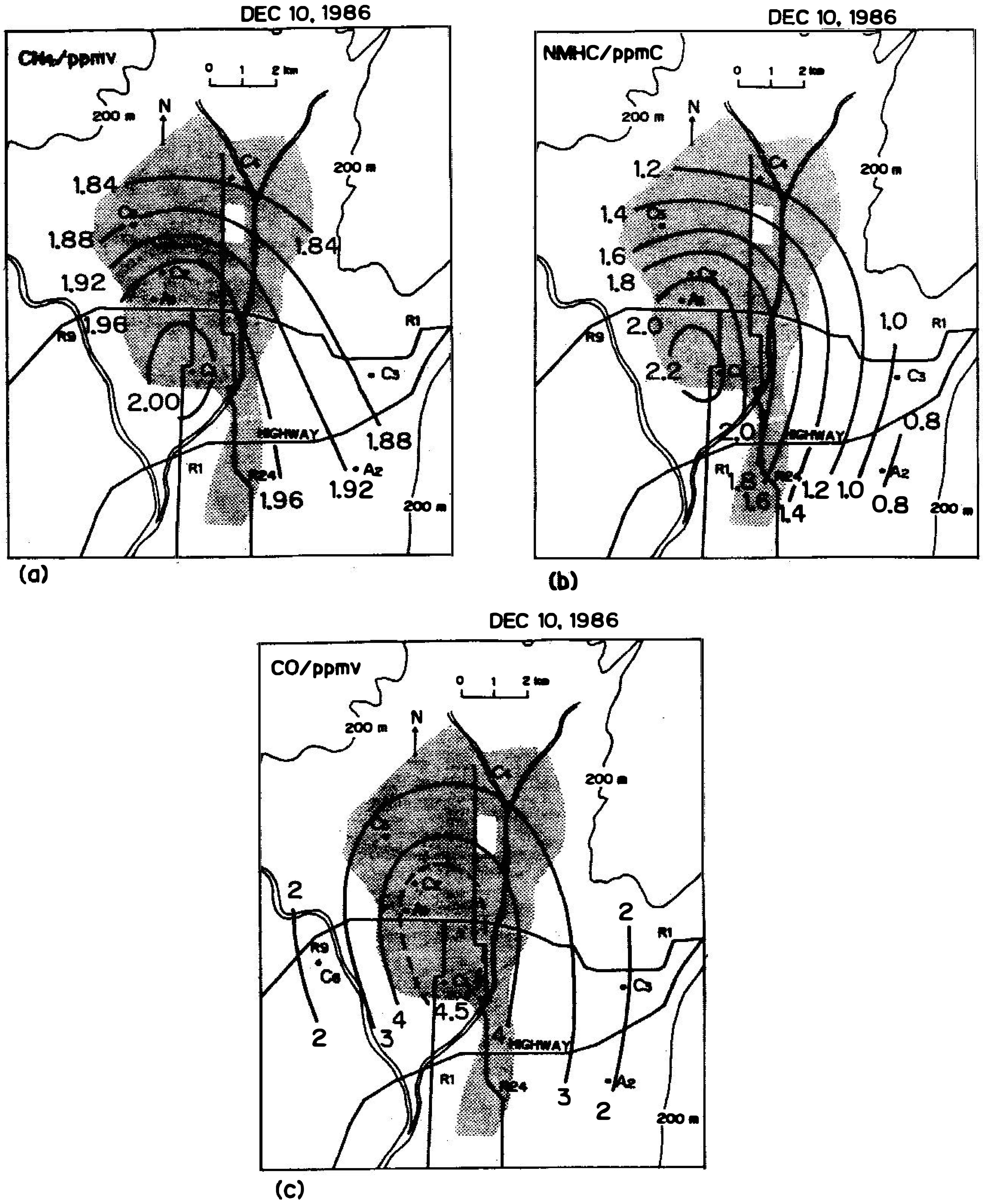

Fig. 4. Daily mean distributions on December 10, 1986 for (a) CH_4, (b) NMHC, and (c) CO.

correlation coefficient is 0.92. The monthly mean background methane $(CH_4)_0$ is defined as the intercept of the linear regression line. In this case, $(CH_4)_0$ is 1.759 ppmv. From this procedure, the observed methane $(CH_4)_{ob}$ is separated into two parts, $(CH_4)_0$ and ΔCH_4, or $(CH_4)_{ob} = (CH_4)_0 + \Delta CH_4$. The additional methane, ΔCH_4, shows the part of the methane emitted from the same sources as NMHC, and it increases linearly with the mixing ratio of NMHC. From the data of CO, another $(CH_4)_0$ and ΔCH_4 which increases with the mixing ratio of CO is obtained, as shown in Figure 6(b). From this plot, $(CH_4)_0$ =1.754 ppmv is obtained, and this agrees reasonably well with that obtained from NMHC. With this method, the monthly mean background methane $(CH_4)_0$ and daily mean additional methane ΔCH_4 corresponding to the daily mean NMHC or CO for individual stations are obtained.

The seasonal variations of the monthly mean background $(CH_4)_0$ and observed methane averaged over seven stations are shown in Figure 7 for the April 1986 to March 1987 period.

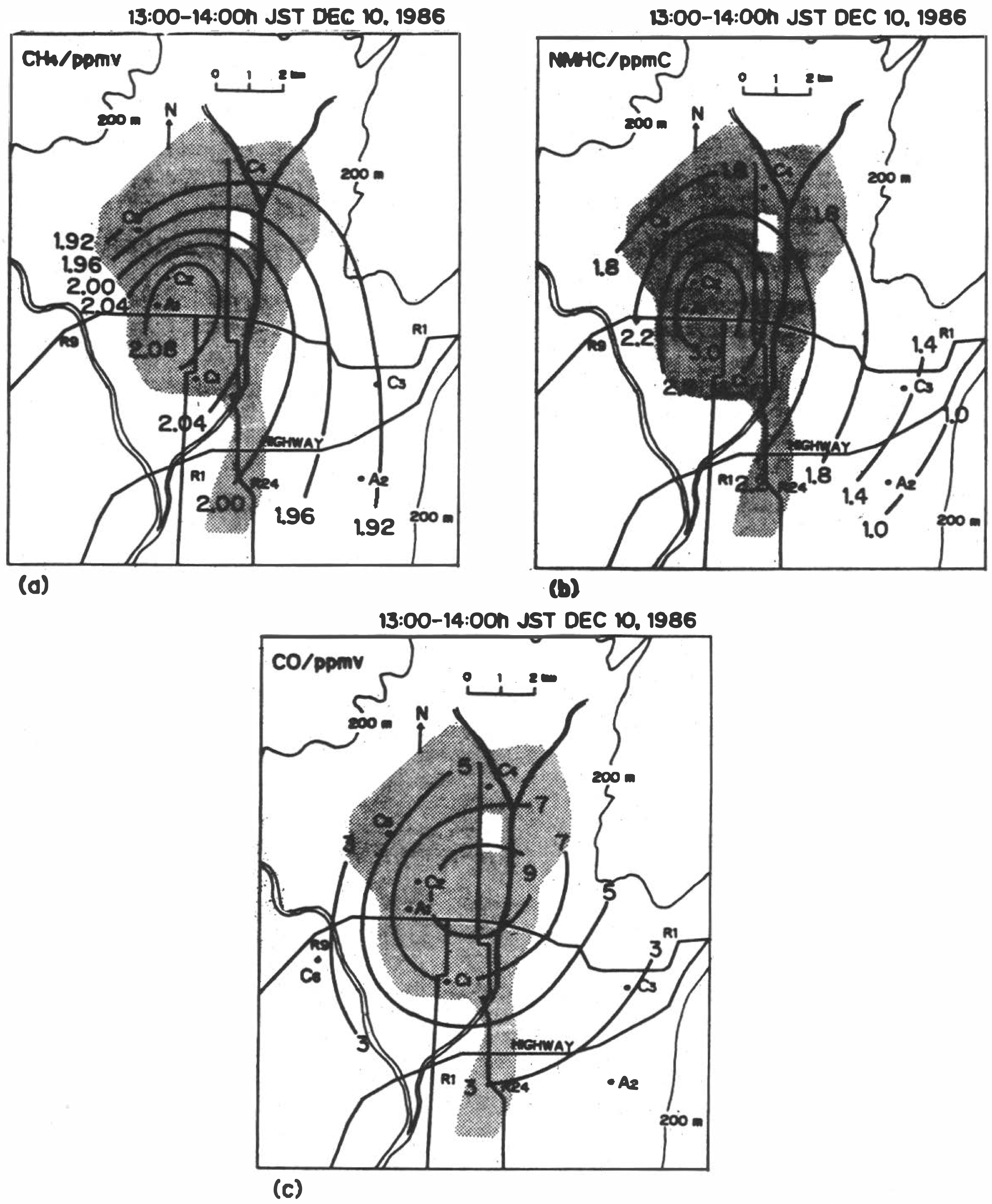

Fig. 5. Hourly mean distributions for 13:00-14:00 hrs JST (Japan Standard Time) on December 10, 1986 for (a) CH_4, (b) NMHC and (c) CO.

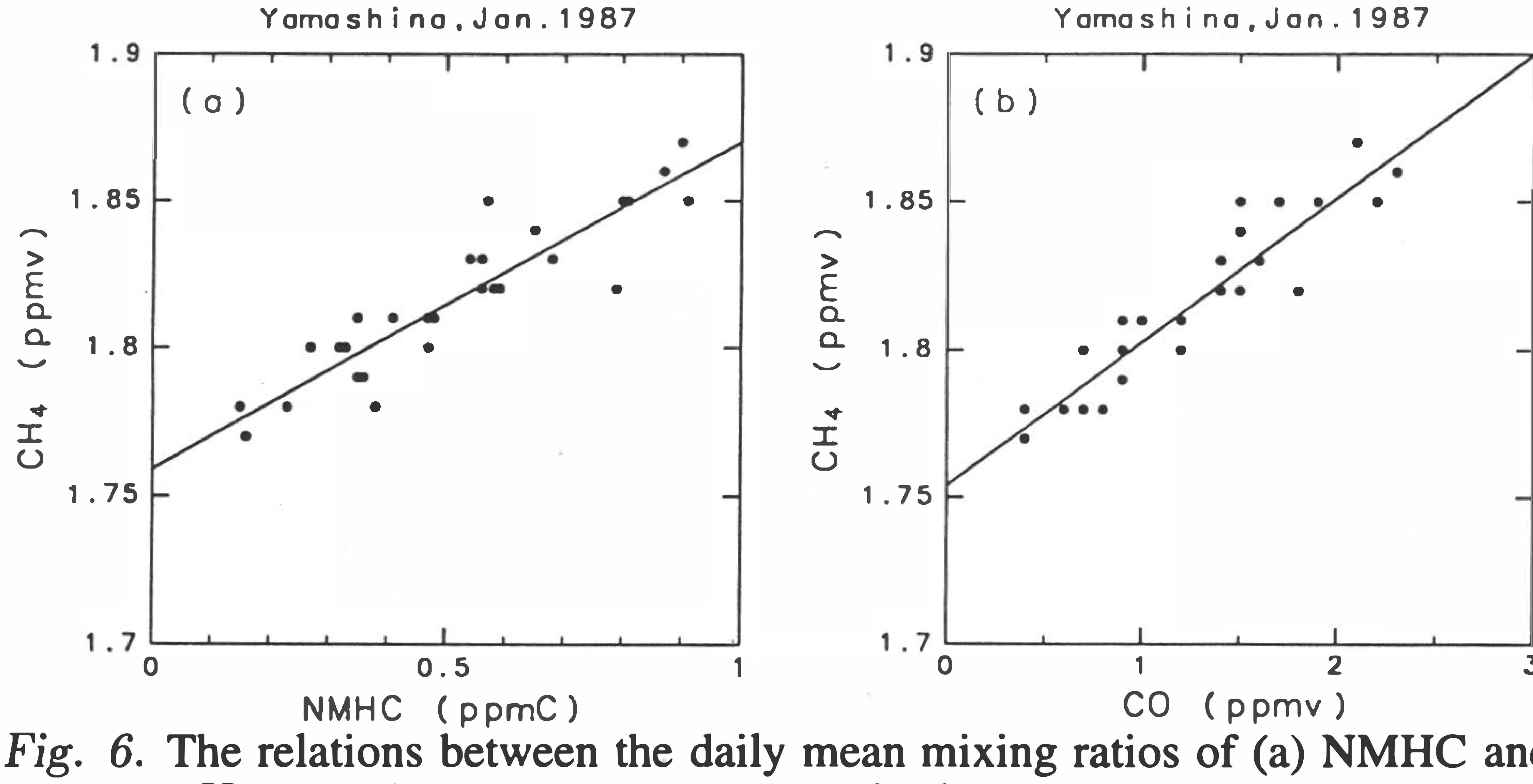

Fig. 6. The relations between the daily mean mixing ratios of (a) NMHC and CH_4, and (b) CO and CH_4. The solid lines show the linear regression fit to the data.

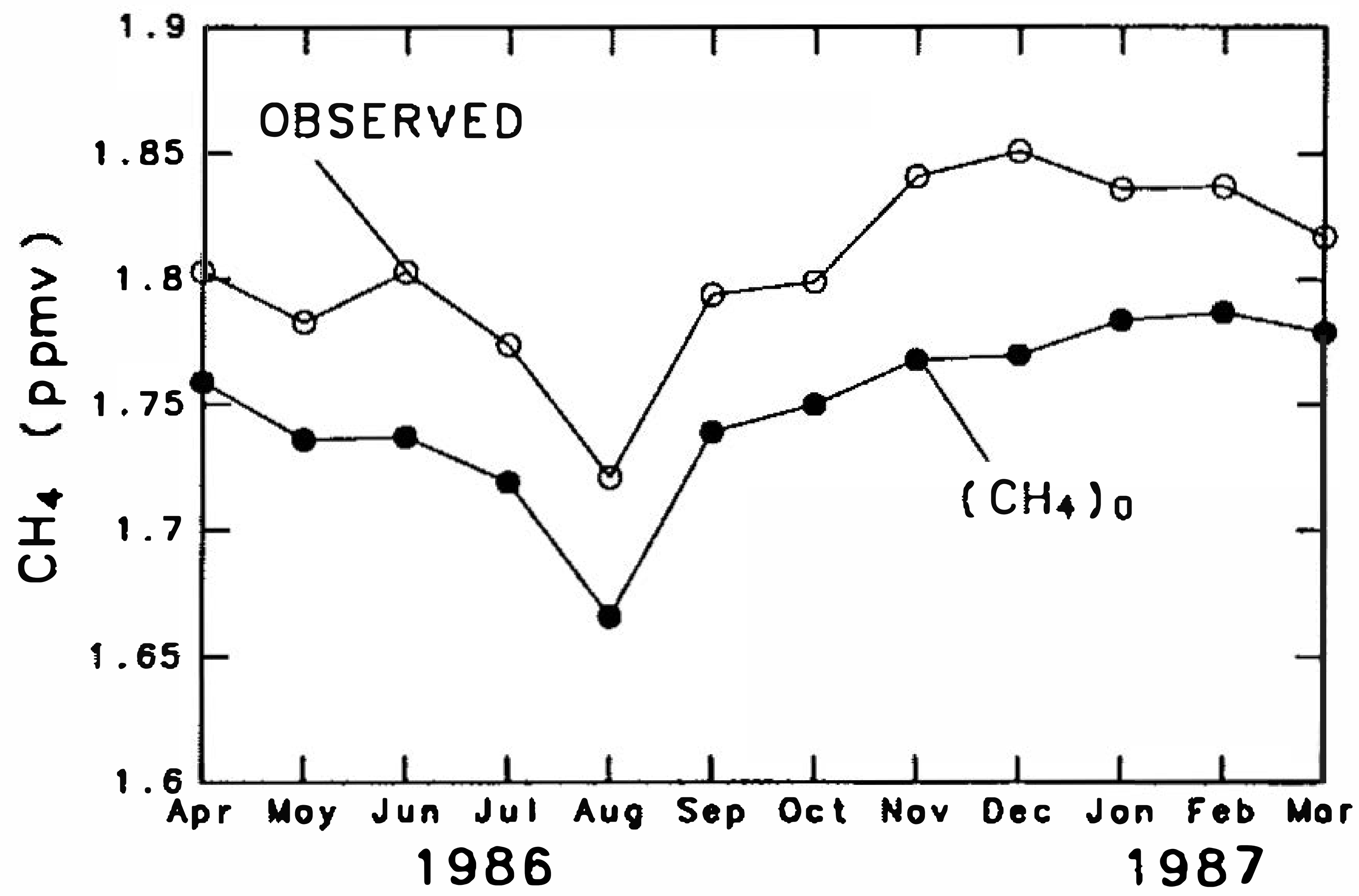

Fig. 7. Seasonal variations of background $(CH_4)_0$ (filled circles) and observed methane (open circles).

The background methane $(CH_4)_0$ (filled circles) shows the minimum in August and the broad maximum in winter. The observed methane $(CH_4)_{ob}$ (open circles) shows the same seasonal variation except that it is higher than $(CH_4)_0$ by 0.03 ppmv to 0.08 ppmv, which corresponds to the additional methane, ΔCH_4. The minimum in August is due to the low methane mixing ratio in the tropical maritime air mass which extends to this region in summer. The spatial distribution of the yearly mean additional methane, ΔCH_4, is shown in Figure 8. The maximum is at the same location as the observed methane (Figure 3(a)). The yearly mean additional methane, ΔCH_4, amounts to 4% of the observed methane in the central part of the city. An example of the distribution of the daily mean additional methane, ΔCH_4, on December 10, 1986 is shown in Figure 9. In this case, the maximum additional methane, ΔCH_4, amounts to 11% of the observed methane.

4. ADDITIONAL METHANE IN TOTAL HYDROCARBONS

The content of carbon emitted as CH_4 in total hydrocarbons (THC) estimated from the daily means of NMHC and ΔCH_4 is shown in Figure 10. The monthly means over the densely populated area (5 stations excluding A_2, C_3 and C_6 in Figure 3) are plotted. The observed mixing ratio of NMHC is given by the unit ppmC which is calculated in terms of carbon number (or in terms of methane). Accordingly, the mixing ratio of total hydrocarbons in terms of carbon number is given by the sum of the mixing ratios of NMHC and ΔCH_4. The yearly mean ΔCH_4 content is 8.3% with the standard deviation of 1.0%, but the weak seasonal variation is seen with the maxima in summer and winter. The day-to-day variation of the daily mean ΔCH_4 content in January 1987 is shown in Figure 11. The monthly mean ΔCH_4 content is 8.5% with the standard deviation of 1.5%.

5. RATIO OF ADDITIONAL METHANE TO CO

The observed mixing ratio of CH_4 increases with CO as shown in Figure 6(b). Methane emitted from the same sources as CO is estimated from the regression coefficients for individual stations. The monthly mean ratios of ΔCH_4 to CO averaged over the densely populated area (5 stations excluding A_2, C_3 and C_6 in Figure 3) are shown in Figure 12. This ratio

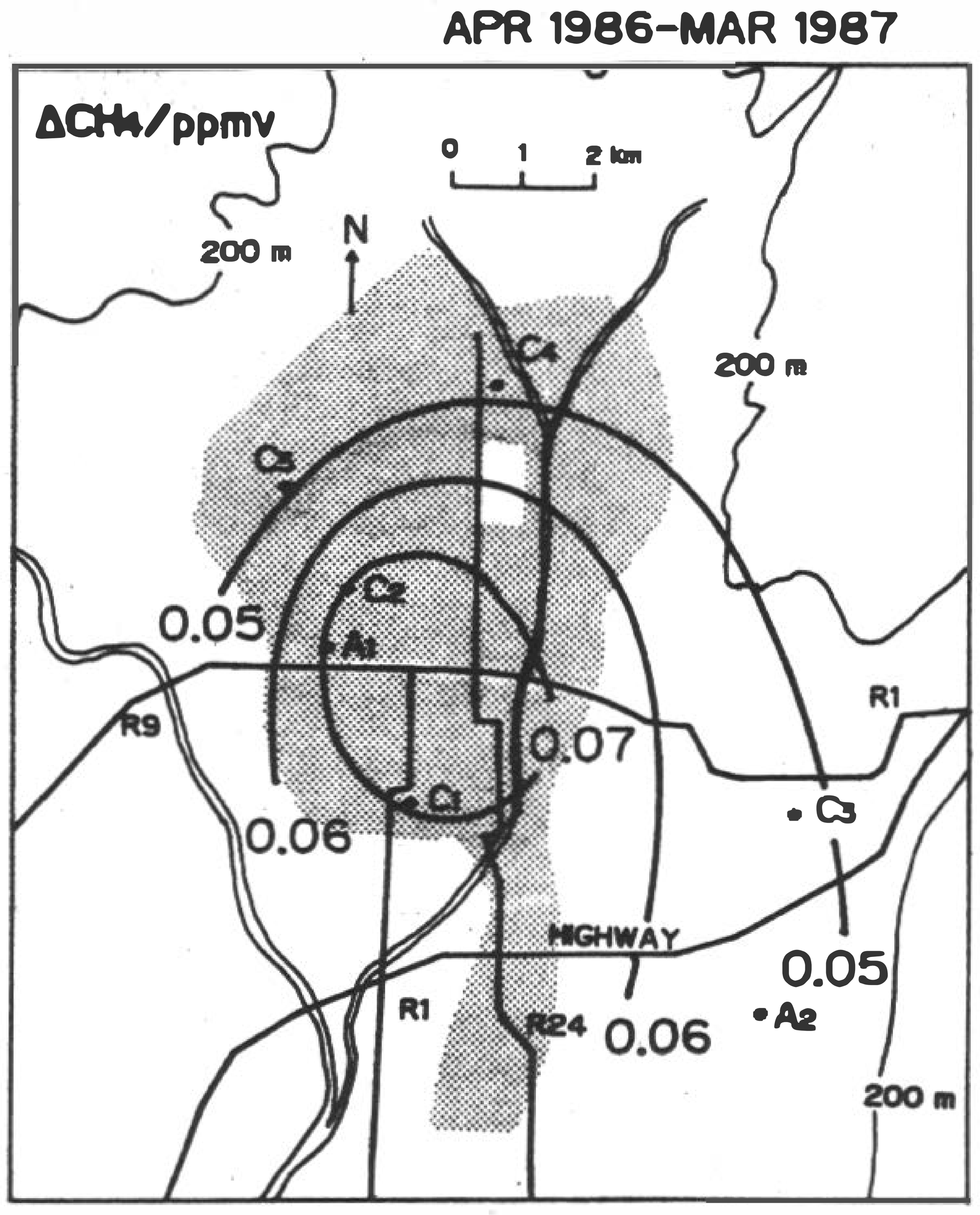

Fig. 8. The distribution of yearly mean additional methane ΔCH_4 for April 1986 to March 1987.

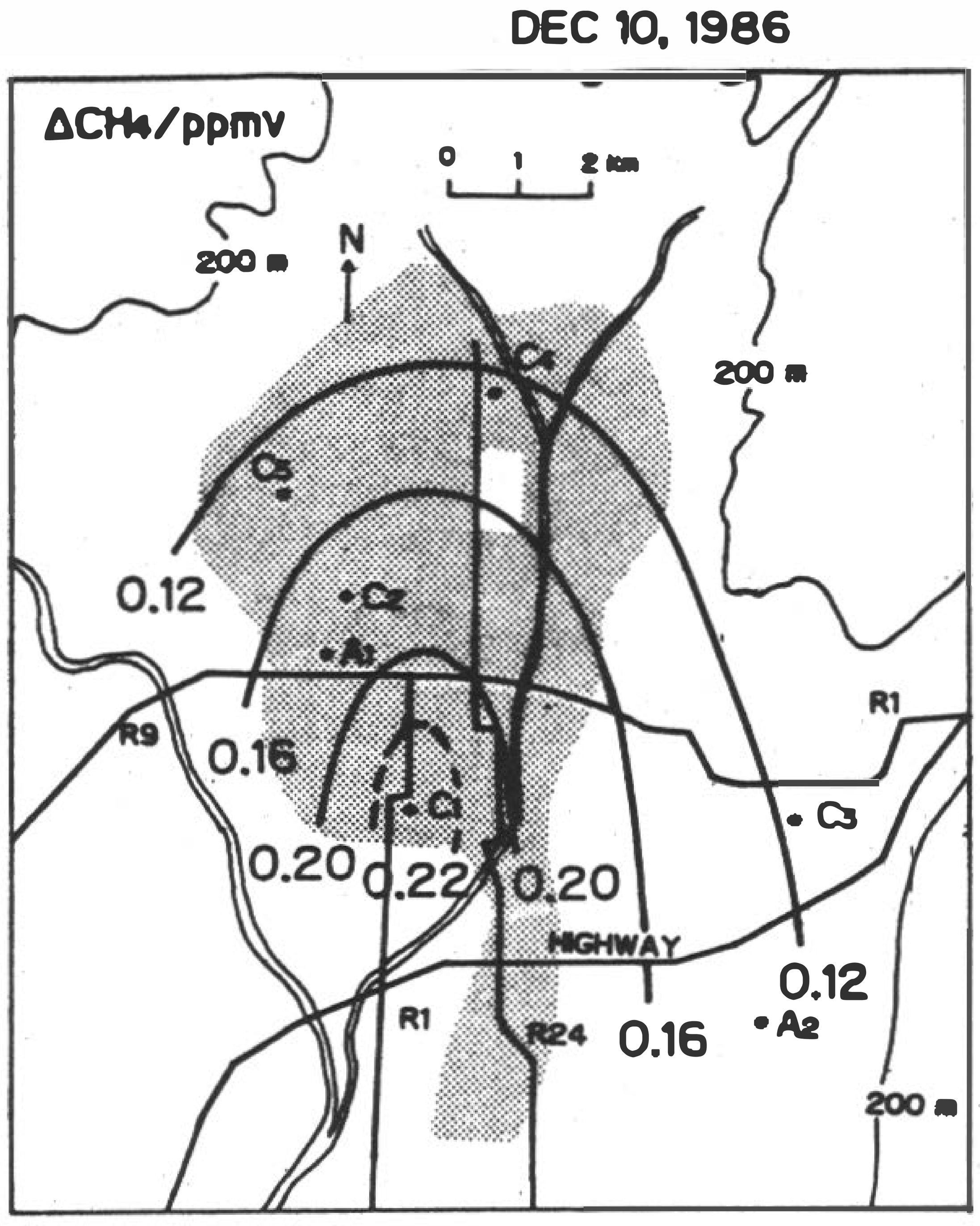

Fig. 9. The distribution of daily mean additional methane ΔCH_4 on December 10, 1986.

shows the seasonal variation with the maxima in summer and winter. The yearly mean with the standard deviation is 0.034$\pm$0.006(v/v) in volume. The January 1987 day-to-day variation of the ratio of ΔCH_4/CO averaged over the densely populated area is shown in Figure 13. The monthly mean with the standard variation is 0.031$\pm$0.005(v/v).

6. SUMMARY AND DISCUSSION

It is shown that the atmosphere in the large cities of Osaka and Kyoto contains more CH_4 than in the surrounding areas. The maximum CH_4 is located in the central part of the cities for the mean distributions of the shorter (hour) to the longer (year) period. The emission

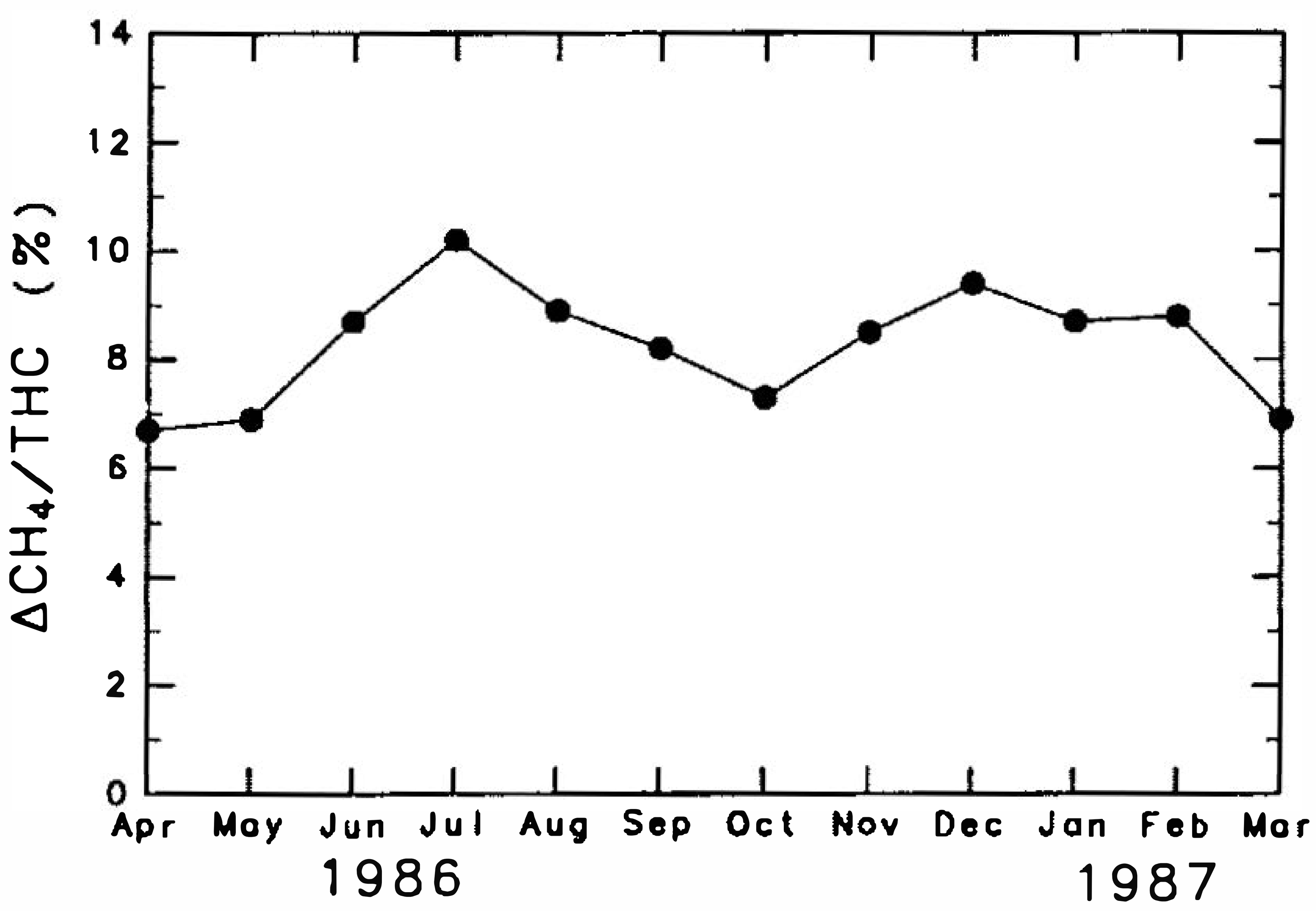

Fig. 10. The seasonal variation of the ratio of additional methane ΔCH_4 to total hydrocarbons calculated in tenns of carbon number (or in terms of CH_4).

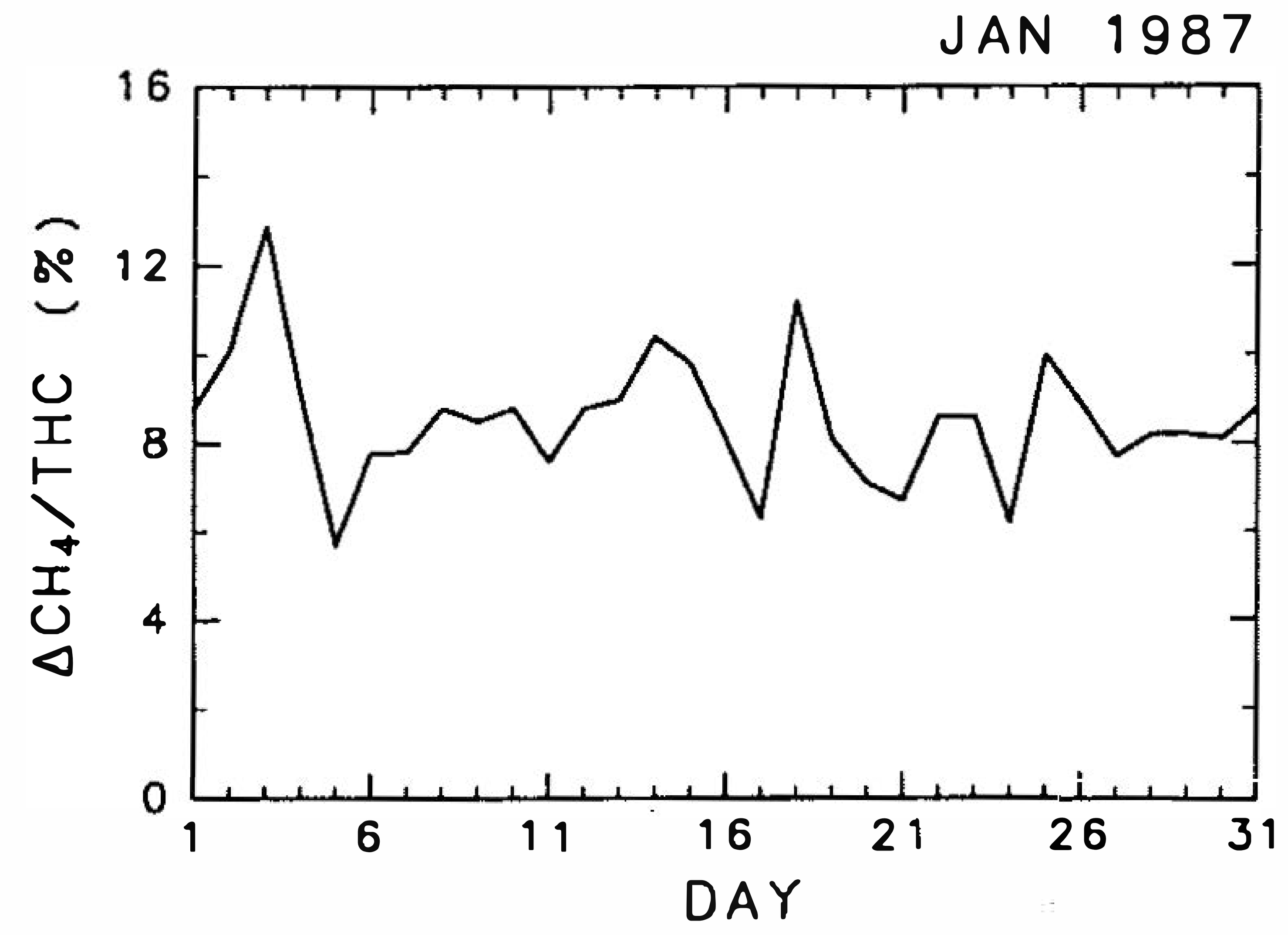

Fig. 11. The day-to-day variation of the ratio of additional methane ΔCH_4 to total hydrocarbons (THC) in January 1987, calculated in terms of carbon number.

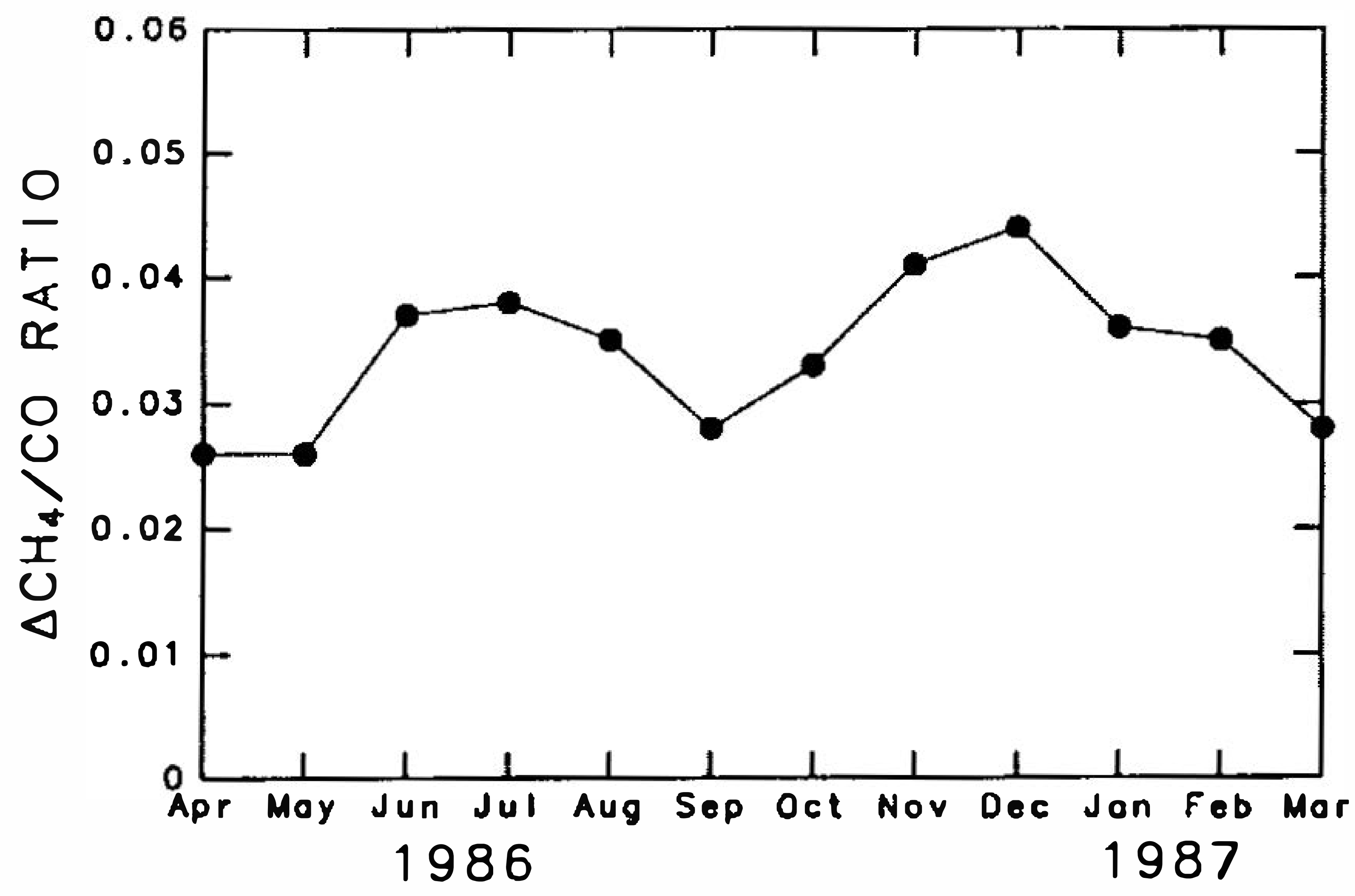

Fig. 12. The seasonal variation of the ratio of additional methane ΔCH_4 to CO in volume.

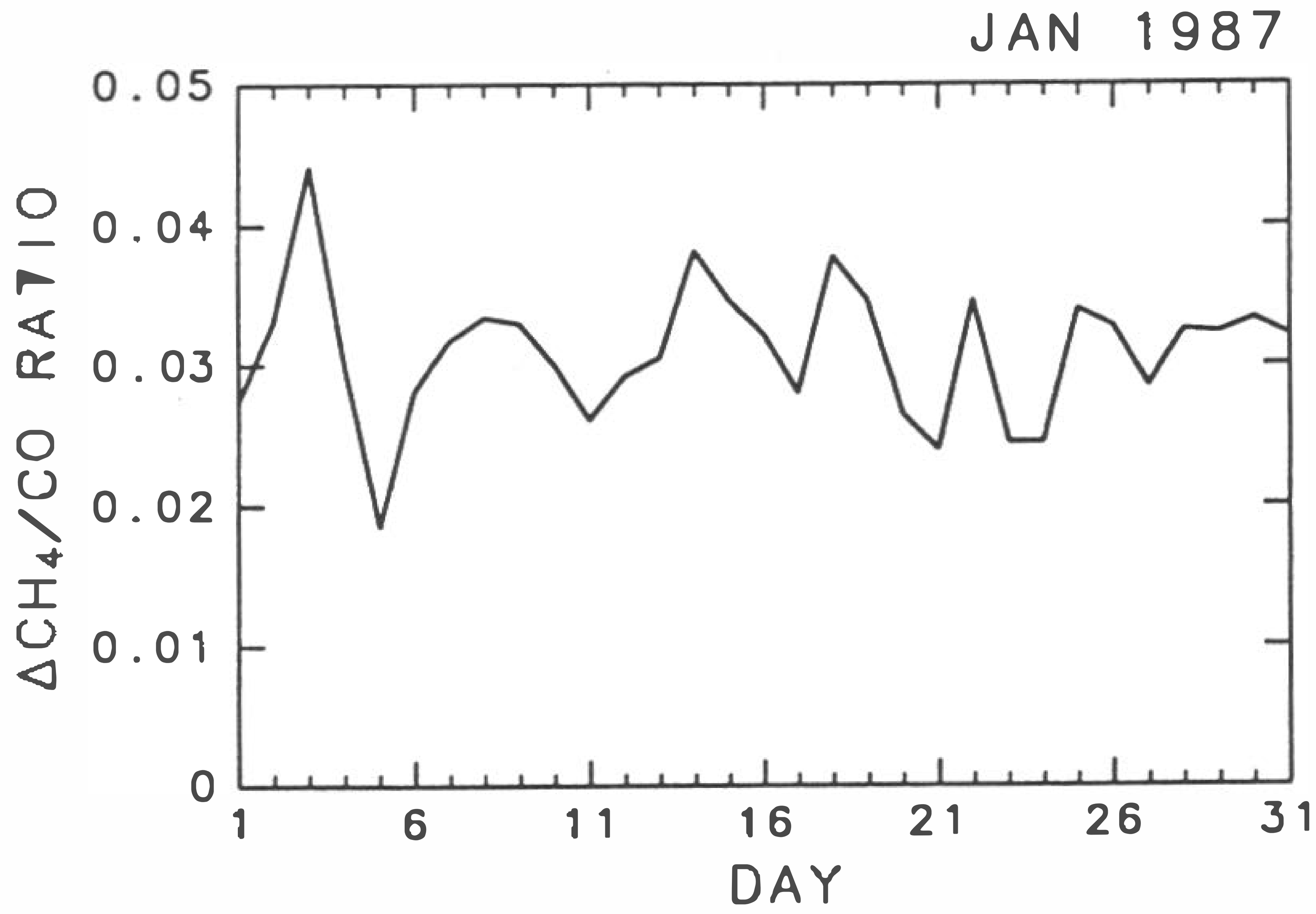

Fig. 13. The day-to-day variation of the ratio of additional methane ΔCH_4 to CO in volume in January, 1987.

of methane is closely related to CO and NMHC, and it is concluded that the methane in the city, amounting to 4% of the observed methane for the yearly mean in the central part of Kyoto, is emitted from the same sources as CO and NMHC. The content of carbon emitted as methane in total hydrocarbons is 8.3%, while the emission ratio of $\Delta CH_4/CO$ is 0.034 in volume.

It is known that the main source of CO and NMHC is the exhaust from automobiles in Kyoto (Kyoto City Government,1992). In the same way, the main source of the additional methane in Kyoto City is the exhaust from automobiles. The CH_4 emission strength in the city can be estimated using the ratios $\Delta CH_4/CO$ or $\Delta CH_4/THC$ obtained above, if the emission strengths of CO or THC, albeit not yet seized quantitatively, are known.

Acknowledgments The author wishes to thank the Environmental Conservation Office of the Public Health Bureau of the Kyoto City Government for granting permission to use the data of CH_4, NMHC and CO in the City of Kyoto. Thanks are also due to the research staff of the Institute of Hygiene and Environmental Pollution of Kyoto City and the staff of our research section of the Disaster Prevention Research Institute for arranging and analyzing the data.

REFERENCES

Cicerone, R. J., and R.S.Oremland, 1988: Biogeochemical aspects of atmospheric methane. *Global Biogeochem. Cycles.*, **2**, 299-327.

Crutzen, P. J., 1973: A discussion of the chemistry of some minor constituents in the stratosphere and troposphere. *Pure Appl. Geophys.*, **106-108**, 1385-1399.

Ehhalt, D. H., 1988: How has the atmospheric concentration of CH_4 changed? In: F. S. Rowland and I. S. A. Isaksen, (Eds.), The Changing Atmosphere, John Wiley & Sons Ltd., Chichester, 25-32.

Kyoto City Government, 1992: Environment of Kyoto City-1992, 208pp. (in Japanese)

Levy, H., 1972: Photochemistry in the lower troposphere. *Planet. Space Sci.*, **20**, 919-935.

Molina, M. J., and F. S. Rowland, 1974: Stratospheric sink for chlorofluoromethanes: chlorine atom-catalyzed destruction of ozone. *Nature*, **249**, 810-812.

Muramatsu, H., 1989: Distribution and flux of methane over Hanshin District. Proc. of the Japan-China (Taipei) Joint Seminar on Natural Hazard Mitigation, Kyoto, Japan, 211-216.

Wang, W. C., Y. L. Yung, A. A. Lacis, T. Mo, and J. E. Hansen, 1976: Greenhouse effects due to man-made perturbations of trace gases. *Science*, **194**, 685-690.