

Recent Decline of Atmospheric Concentration and Emission of Methane in Nagoya Metropolitan Area

Akinori Ito,# Shishin Yamada, Takuya Higuchi, Yuzo Ishikawa, Yoko Nagata, Koichi Chiba, and Hiroki Haraguchi*

Department of Applied Chemistry, Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603

(Received February 28, 2002)

Anthropogenic emissions of methane (CH_4) from major sources such as landfills, automobiles, and rice paddy fields in the Nagoya metropolitan area were estimated using several emission process parameters such as emission factors and activity data for the decade from 1988 to 1997. The sum of CH_4 amounts emitted from these three major sources was calculated to be 4.93 Gg of CH_4 in 1988 and 5.19 Gg of CH_4 in 1997. Methane emissions from the 12 landfill sites located around the Nagoya metropolitan area gradually increased until 1996 and then decreased in 1997, while those from automobiles and rice paddy fields continuously declined for the same decade. The total amount of CH_4 emitted from the 12 landfills was much larger than those from other two sources. In 1993, for example, the amount was estimated as 3.26 Gg of CH_4, 58% of which was generated from the largest landfill site. The excess CH_4 concentration in the urban atmosphere, which was estimated from the difference between the CH_4 concentration at the urban site and the background CH_4 concentration at the Mauna Loa observatory, Hawaii, USA, showed the long-term trends similar to the CH_4 emission from the largest landfill site. As a result, we conclude that the CH_4 emission from the largest landfill dominantly impacted on the excess CH_4 concentration due to the major sources in the Nagoya metropolitan area.

Human activities have strongly impacted on the changes in the atmospheric concentrations and distributions of greenhouse gases since the industrial revolution. In particular, the global burden of methane (CH_4) in the atmosphere, one of major greenhouse gases, has been more than doubled, from 0.7 ppmv to 1.7 ppmv, over the last 200 years.[1] In recent years, it was reported by Dlugokencky et al. that the overall growth rate of CH_4 declined since the late 1980s,[2] although the global atmospheric CH_4 concentration had been continuously increasing. This may be caused by the increase in CH_4 sink due to the increase in OH radicals in the atmosphere,[3] but there still exist large quantitative uncertainties in the estimates of CH_4 emission.

Extensive research has been conducted to evaluate the global CH_4 source strengths in order to establish a diagnosis of the CH_4 budget.[4–6] National emission inventories have been developed to evaluate the amounts of CH_4 emissions in many countries, taking their domestic affairs into consideration. Nonetheless, wide uncertainties in the estimates of CH_4 emission have still remained especially for most biotic sources, which include biomass combustion in savannas and forests as well as the biodegradation of organic matter in landfills and rice paddy fields.[7] Among these sources, the decomposition of municipal solid wastes at the landfill sites is considered as one of the major anthropogenic sources of CH_4 in most industrialized countries.[8–10]

The Ministry of the Environment (former Environmental Agency) has issued annual reports on the emissions of greenhouse gases in Japan. According to these reports, ruminants, landfills, and rice fields are major anthropogenic emission sources of CH_4, whose contributions were estimated to be about 25%, 24%, and 22% of total anthropogenic emissions, respectively. It was also reported that the total amount of CH_4 emitted from anthropogenic sources decreased from 1538 Gg of CH_4 in 1990 to 1381 Gg of CH_4 in 1997 on a national scale. Among these sources, the emission amount from the landfills decreased by ca. 8%, which resulted from the decrease in the final disposal amount at the landfill sites during the above period. Such a national inventory of CH_4 emissions includes the important indices to provide the guidelines for assessment of national strategies of CH_4 mitigation.

On the other hand, the contributions of major anthropogenic CH_4 emission sources to the regional CH_4 concentration substantially vary from region to region, reflecting the regional populations, industrial structures, landforms, and so on. The amount of CH_4 emissions from the landfills is, for instance, influenced by the human activities such as the variety of refuses and waste disposal management. It is thus desirable to evaluate the amounts and trends of regional CH_4 emissions from anthropogenic sources in order to assess effective and particular mitigation strategies for each region.

In Nagoya City, several research groups have investigated the kinetic behaviors of atmospheric CH_4 and its emission sources.[11–15] According to those reports, the atmospheric CH_4 burdens have decreased in the central area of Nagoya City

Present address: University of Michigan, 2455 Hayward, Ann Arbor, MI, 48109-2143, USA

since 1993, where CH_4 emitted from the largest landfill site predominantly enhanced the urban atmospheric CH_4 concentrations.[14,15] The Environmental Affairs Bureau (EAB) of Nagoya City reported that the final disposal amount of municipal waste has decreased since 1991. Accordingly, one may argue that the recent decrease in the CH_4 concentration in the urban area is caused by the change of CH_4 emissions from the landfill sites.

In the present paper, the long-term trends of the anthropogenic CH_4 emissions from the major sources were investigated in association with the atmospheric CH_4 concentration monitored in the Nagoya metropolitan area from April, 1988, to March, 1998. In particular, estimation of annual CH_4 emissions from the landfills as well as from automobiles and rice paddy fields was carried out to evaluate the impacts of their source strengths on the CH_4 concentration in the urban area. The CH_4 emissions from automobiles were taken into consideration in the following discussion because they might give large impacts to the atmospheric CH_4 concentrations in the large cities. However, since the livestock farming is negligibly small in the Nagoya metropolitan area, the CH_4 emissions from ruminants were not considered here. Finally, interannual variations of excess CH_4 concentrations in the urban atmosphere were compared with those of CH_4 emissions from the major sources.

Materials and Methods

Continuous Monitoring of Atmospheric CH_4 Concentration in Nagoya City. Continuous monitoring of the atmospheric CH_4 concentration has been performed since 1990 at the campus of Nagoya University. The instrument for the CH_4 concentration monitoring was a gas chromatograph with a flame ionization detector (GC/FID) (model HCM-4A from Shimadzu, Kyoto). The experimental or operating conditions of the GC/FID system for CH_4 measurement were described in detail in the precious paper.[11] The instrument was calibrated at 0:00 a.m. every day by using the CH_4 standard gas. The CH_4 standard gas used for calibration was purchased from Nippon Sanso Co. (Tochigi). The CH_4 concentration of the standard gas was 2.07 ppmv, which was prepared by diluting high-purity CH_4 gas with high-purity N_2 gas.

Continuous monitoring of the atmospheric CH_4 concentration at Higashi-sakura, the central part of Nagoya City, has also been performed since 1983 by the EBA of Nagoya City,[15] where the same instrumental system as described above was used. The main purpose of the monitoring by the EBA of Nagoya City was to measure the concentration of non-methane hydrocarbons (NMHCs), but the concentration of CH_4 was also observed as the additional data.[11]

The annual changes of the atmospheric CH_4 concentrations observed at Nagoya University and Higashi-sakura are shown in Fig, 2. The long-term trends of the CH_4 concentrations at Nagoya University and Higashi-sakura are quite similar to each other, although some parts of them are slightly different because of their different locations. Therefore, in order to examine the trends of the atmospheric CH_4 concentration for the longer period (1988–1998), the monitoring data observed at Higashi-sakura were employed in the present paper.

Atmospheric CH_4 Concentration Data in Nagoya City for 1988–1997. The metropolitan area of Nagoya is one of the most urbanized and modernized areas in Japan. As is shown in Fig. 1, Nagoya is located midway between Tokyo and Osaka, and lies in the southern edge of the Nobi plain, where rice cultivation is the major agricultural activity. Around Nagoya, rice paddy fields expand into inland areas, even in the southeastern area of the city. The major and active landfill sites are located in the northeastern area of the city, marked as "A" and "B", in which 65% and 23% of the total amount of municipal wastes, respectively, were dumped during the past decade from 1988 to 1997. According to the previous paper[15] concerning the long-term trends of CH_4 emissions in Nagoya metropolitan area, it was found that the CH_4 concentrations observed at Higashi-sakura were predominately influenced by CH_4 emissions from landfill "A". Thus, only the data monitored at Higashi-sakura were used to discuss the long-term trends of the atmospheric CH_4 concentrations in the following

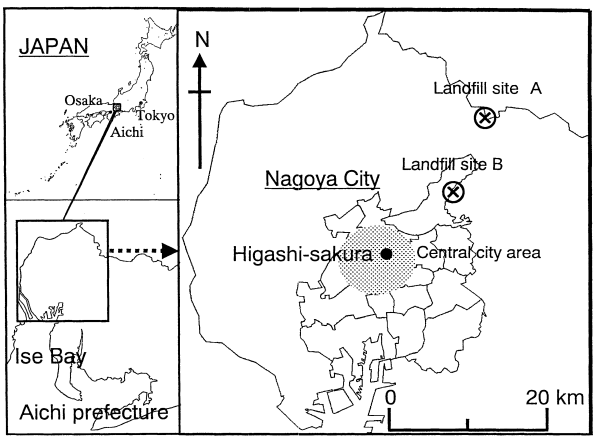

Fig. 1. Location of the Higashi-sakura observatory for the atmospheric CH_4 concentration in Nagoya City, Japan. Major landfill sites are locatted at "A" and "B", marked as ⊗.

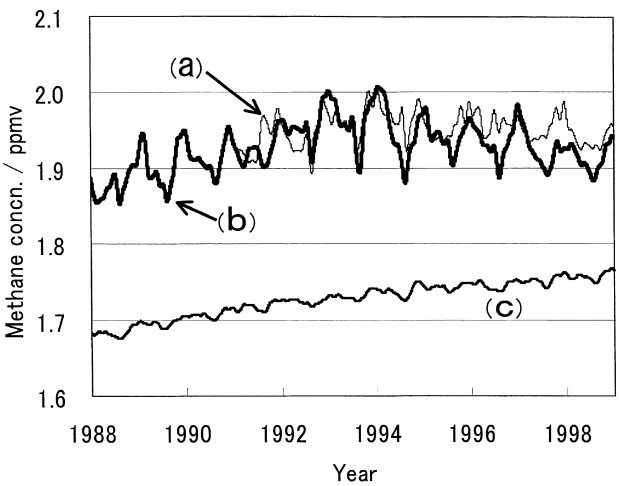

Fig. 2. Annual changes of atmospheric concentrations observed at Nagoya University, Higashi-sakura (central Nagoya), and Mauna Loa observatories. (a) Nagoya University, (b) Higashi-sakura, (c) Mauna Loa.

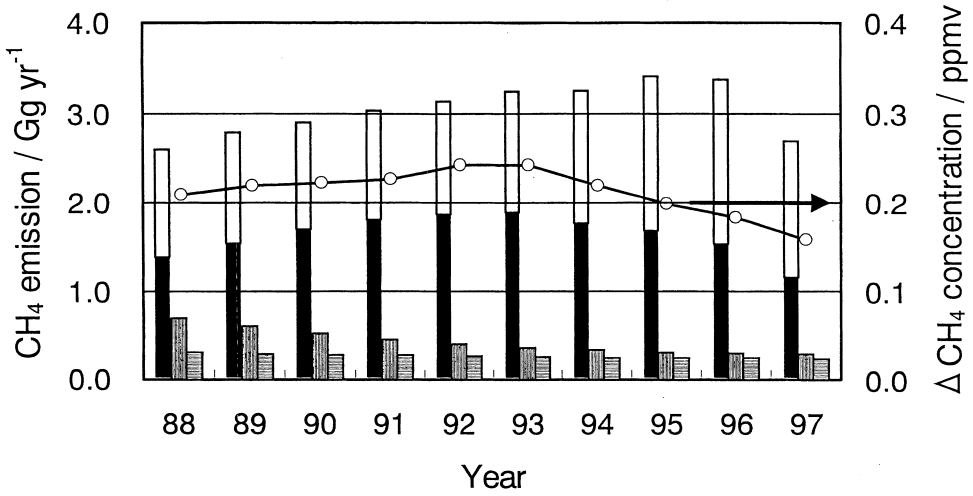

Fig. 3. Urban excess CH_4 concentrations (open circles) at the Higashi-sakura observatory from April of 1988 to March of 1997 and long-term evolutions of the anthropogenic CH_4 emissions in Nagoya metropolitan area. ■: Landfill A. □: Other landfills. ▥: Automobiles. ▤: Rice paddy fields.

discussion.

The 10-year (from April, 1988, to March, 1998) records of the atmospheric CH_4 concentrations monitored at the Higashi-sakura observatory in the central part of Nagoya and at the Mauna Loa observatory in Hawaii, USA, are shown in Fig. 2; data were obtained by the EAB of Nagoya City and NOAA/CMDL,[16] respectively. It should be noted here that the atmospheric CH_4 concentration observed at Higashi-sakura has been significantly decreasing since 1993. Among the data shown in Fig. 2, two data sets of atmospheric CH_4 concentrations at Higashi-sakura and Mauna Loa were used as the representatives of the urban and background trends to elucidate quantitatively the recent decline of CH_4 emissions due to anthropogenic sources in Nagoya metropolitan area. Then, the excess CH_4 concentrations were estimated as the differences between the CH_4 concentrations observed at Higashi-sakura and Mauna Loa observatories. The results are shown in Fig. 3, the details of which will be discussed later.

Estimation of CH_4 Emissions from Major Emission Sources. Annual CH_4 emissions in Nagoya metropolitan area from April, 1988, to March, 1998, were calculated in the present research, where landfills, automobiles, and rice paddy fields were considered as the major emission sources, as described in the introductory section. All statistic data were quoted from the official reports of municipal statistics from Nagoya City. The emission models and emission factors were retrieved from the recent papers and reports of the national inventory.[17]

Results and Discussion

Estimates of CH_4 Emission from Landfills. In order to estimate the CH_4 emission from the landfills for municipal solid wastes, the following data were used as the parameters: the amount of wastes dumped at the 12 landfill sites in Nagoya metropolitan area, the percentages of organic materials in municipal waste, rates of carbon contents in organic materials, gasification rates of decomposed organic materials, percentage of CH_4 in generated gases, and temporal profiles of gas generation from the landfills.

In Nagoya City, the amounts of municipal wastes gradually increased from 882 Gg (dry weight) in 1988 to 1037 Gg in 1997. In order to reduce the amounts of final disposal at landfill sites, the municipal wastes were primarily sorted into combustible refuses, noncombustible ones, large-size ones, and recycling resources by civilian cooperation, and then most of combustible municipal refuses was incinerated to reduce their volumes and masses. As a result, the incineration fly and bottom ashes were dumped at the landfill sites. As for large-size refuses such as cupboards, bookshelves, desks, and so on, more than half of which were made of organic materials, they had been also dumped into the landfills directly. However, the incineration systems for large-size refuses made of organic materials were installed at the landfills "A" and "B" in July, 1993, and in March, 1979, respectively. Among noncombustible refuses, organic materials, such as yard wastes, paper products, woods, leathers, and so on, had often been mixed in the wastes to be dumped. In 1997, the new waste treatment facility started to sort noncombustible refuses into combustible ones, noncombustible ones, and metals automatically. Since then, much lower amounts of organic materials were dumped into landfills.

The compositions of municipal wastes and the portions of organic materials in the municipal wastes from 1978 to 1997 were derived from the data in the publications from the EAB of Nagoya City. Then the annual amounts of organic materials dumped into the landfills were calculated from the amount of municipal wastes and their portions of organic materials. The results are shown in Fig. 4. As is seen in Fig. 4, the amounts of both total wastes and organic materials were reduced during the two decades from 1978 to 1997, such reductions mostly resulted from the employment of the incineration and recycling systems.

The carbon content rates of organic materials were estimated to be 45% for paper products and 44% for the others, such rates were calculated on the average from wood textiles and garden/park wastes.[17] About 50% of organic materials was as-

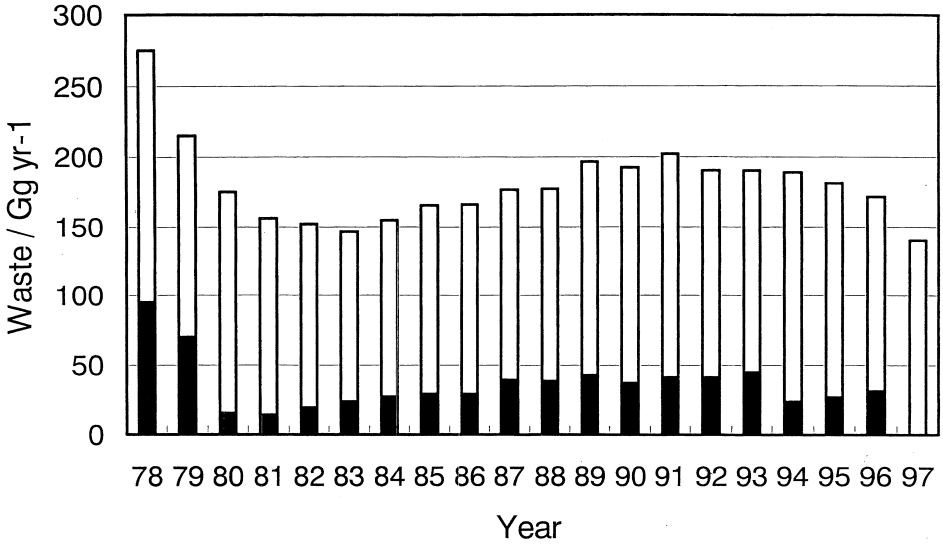

Fig. 4. Annual amounts of municipal wastes dumped at 12 landfill sites from 1978 to 1997. ■: Organic materials. □: Other wastes.

sumed to be biochemically degraded to fermented gases, which were emitted to the atmosphere through the ventilation pipes.[18] It was reported that the percentage of CH_4 was about 20% of generated gas from a semiaerobic type of landfill and about 50% from an anaerobic one.[19] In Japan, the semiaerobic landfills have mainly been constructed since 1977, while most of the landfills constructed before 1977 were the anaerobic ones.[19] In the present cases, the landfill "B" constructed in 1974 was an anaerobic type of landfill, whereas the landfill "A" constructed in 1982 was a semiaerobic one. Neither of them have any facilities to flare the biogas emitted from the landfills. Consequently, the rates of CH_4 content were assumed to be 20% and 50% of generated gas from the landfill "A" and "B", respectively.

According to the direct survey of gas-generation period from active landfills, biochemically-degraded gas was generated for about 6 years after the wastes were buried in the landfills.[20] It is known that the composition of generated gas varies during the decomposition process.[21] Thus, it was assumed that CH_4 generation from organic materials linearly increased up to a steady state for the first year, kept constant for the following 4 years, and finally decreased exponentially. The temporal profile of CH_4 in our estimate was in good agreement with the temporal change of the CH_4 concentrations measured on the top of the ventilation pipe at the landfill "A" by the EAB of Nagoya City. The CH_4 emissions from the landfills "A", "B" and others were estimated from the above data and parameters in Nagoya metropolitan area.

After the above consideration and data collection, the amounts of CH_4 emissions from the landfills from April, 1988, to March, 1997, were estimated on the annual basis. The long-term evolution of CH_4 emission are shown as bar graphs in Fig. 3. These show that the CH_4 emissions from the 12 landfills gradually increased from 2.60 Gg of CH_4 in 1988 to 3.39 Gg of CH_4 in 1996 and then rapidly decreased to 2.70 Gg of CH_4 in 1997. Among the 12 landfills, 42–59% of the total CH_4 was emitted from the largest landfill "A" in the recent decade, where the CH_4 emission gradually increased from 1.38 Gg of CH_4 in 1988 to 1.89 Gg of CH_4 in 1993 and then rapidly decreased to 1.15 Gg of CH_4 in 1997. The decrease in CH_4 emission from the landfill "A" may reflect the improvement in the waste disposal management such as sorting, recycling, and incineration of the wastes, as mentioned earlier.

Estimates of CH_4 Emission from Automobiles. Methane is also emitted from automobiles to the atmosphere by the process of incomplete combustion of fuels. The Ministry of the Environment of Japan reported the emission factors of CH_4 from automobiles according to vehicle types in 1996. However, the actual values of the emission factors are generally dependent on the performance and mileages of automobiles. In the present research, thus, the emission factor of each vehicle type was estimated on the basis of the emission factors in 1996 as a function of model year and fleet age of automobiles.

The road traffic census data for the different types of vehicles were available in Nagoya City in 1987, 1990, 1994, and 1997. The data gaps of the traveled distances during the period investigated were interpolated with a linear method. Total traveled distance for all automobiles in Nagoya City increased from 8.8×10^9 km yr^{-1} to 10.8×10^9 km yr^{-1} for the recent decade. In 1997, passenger gasoline cars shared 65% of total traveled distance, small-sized gasoline cars 4%, passenger small-sized gasoline cars 4%, passenger diesel cars 8%, freight small-sized diesel vehicles 5%, and freight large-sized diesel vehicles 14%.

The emission factors of CH_4 from automobiles have reduced year by year due to the development of clean technology for exhaust gases. Thus, the improvement in emission performance for later-model cars was taken into consideration to include temporal variations of the emission factors. As for the emission standards from automobiles, the emission of total hydrocarbons (THCs) is regulated by the law, although there is no standard for only CH_4 emission. It is known that the emission of THCs provides good correlation with that of CH_4.[23] Therefore, the improvement factors of reduction in CH_4 emission from each model-year car were calculated on the basis of the emission standards of THCs from vehicles.

On the contrary, the three-way catalysts (TWCs) for reduction of air pollutants in exhaust gases are gradually deteriorated with accumulation of mileages due to the physical attrition, such as the thermal alteration and the adsorption of catalyst poisons.[23] From the remote-sensing measurements of carbon monoxide (CO) emission from gasoline light-duty vehicles passing on a freeway ramp, the annual deterioration rate of TWC cars was estimated to be 17–30% per year.[24] In the present research, thus, the deterioration factor of the catalyst was assumed to be 20% per year.

As a result, the emission factors in each year were estimated from the factors mentioned above and the motor vehicle registration records. The records tell us that the average fleet age was about 5 years and most vehicles were scrapped in 10 years. Figure 5 represents the annual CH_4 emission factors of automobiles according to the vehicle types during the period from 1988 to 1997. It is noted here that the emission factors of all kinds of vehicles have gradually decreased for the recent decade due to the improvement in performances of automobiles. Among them, the emission factor of freight small-sized gasoline cars was the largest in similarity to the actual emission factors reported by the Ministry of the Environment.

The estimates of CH_4 emissions from automobiles for 10 years (1988–1997) are also shown in Fig. 3. As is seen in Fig. 3, the CH_4 emissions from automobiles decreased from 0.69 Gg of CH_4 to 0.28 Gg of CH_4 during the period examined, despite the increase in the traveled distances in Nagoya City. These results are certainly due to the reduction of the emission factors from automobiles owing to the improvement in the performances of engines and catalysts, as discussed above.

Evaluation of the Emission Model for Automobiles. In order to evaluate the present CH_4 emission model for automobiles, it was applied to the estimation of CO emissions from automobiles, because the CO emissions from automobiles mainly contribute to the atmospheric CO concentration in the downtown area of Nagoya City.[26] Therefore, the CO emissions from automobiles were calculated in the same manner as the CH_4 emissions mentioned above.

Long-term evolutions of CO emissions estimated by the present method and by atmospheric CO concentrations observed at the Higashi-sakura observatory are shown in Fig. 6. This shows that the temporal variation of CO emission provided rather good correlation with that of CO concentration, both of which gradually decreased from 1988 to 1992, and then remained almost constant till 1997. These results suggest that the emission model developed here may be reasonable to estimate the long-term trends of CO emissions as well as of CH_4 emissions from automobiles.

Estimates of CH_4 Emission from Rice Paddy Fields. The annual emission rates of CH_4 from paddy fields widely vary from 8 to 27 g of CH_4 m^{-2} yr^{-1} for gley soil in Japan, which depends on the application of organic fertilizers.[25] The highest CH_4 emission rate was observed in the fields with rice straw application as a fertilizer. Since rice straws were successively applied to paddy fields in the most southeastern part of Nagoya, the emission rate from paddy fields was assumed to be 27 g of CH_4 m^{-2} yr^{-1} in the present study.

The estimates of CH_4 emissions from rice paddy fields from 1988 to 1997 are also shown in Fig. 3. Methane emissions from rice cultivation were decreased from 0.30 Gg of CH_4 in 1988 to 0.23 Gg of CH_4 in 1997. The reduction of CH_4 emission from rice cultivation may be explained by the fact that the cultivated area of rice paddy fields in Nagoya was gradually decreased from 11.0 km^2 to 8.6 km^2 during the past decade.

Atmospheric CH_4 Concentration in the Urban Area. Since CH_4 is a stable trace greenhouse gas in the lower atmosphere, whose lifetime is estimated about 12 years, the trends of background CH_4 concentrations and the CH_4 flux from the local emission sources would be reflected in the atmospheric CH_4 concentrations in the urban area. Thus, the urban excess

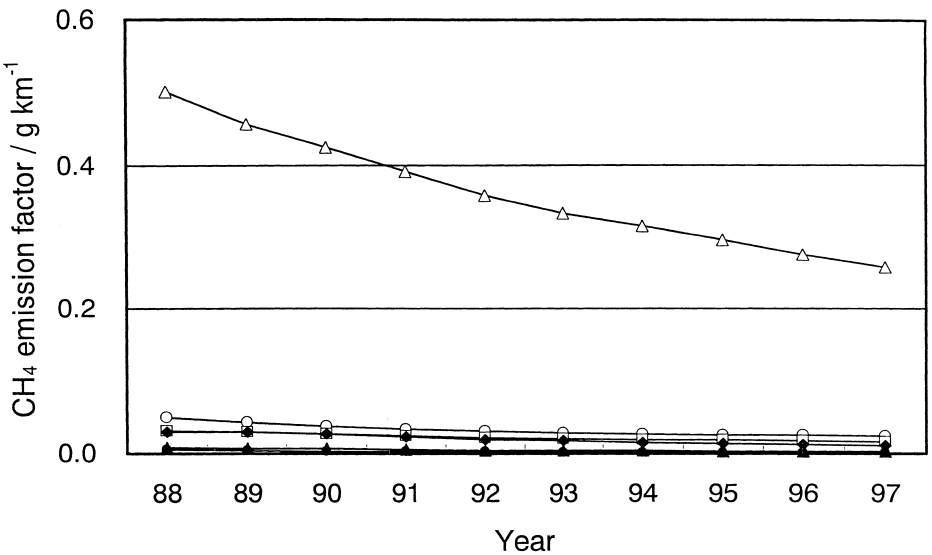

Fig. 5. Changes in annual CH_4 emission factors for various types of automobiles from 1988 to 1997. ○: Passenger gasoline cars. △: Freight small-sized gasoline cars. □: Passenger small-sized gasoline cars. ●: Passenger diesel cars. ▲: Freight small-sized diesel cars. ◆: Freight large-sized diesel cars.

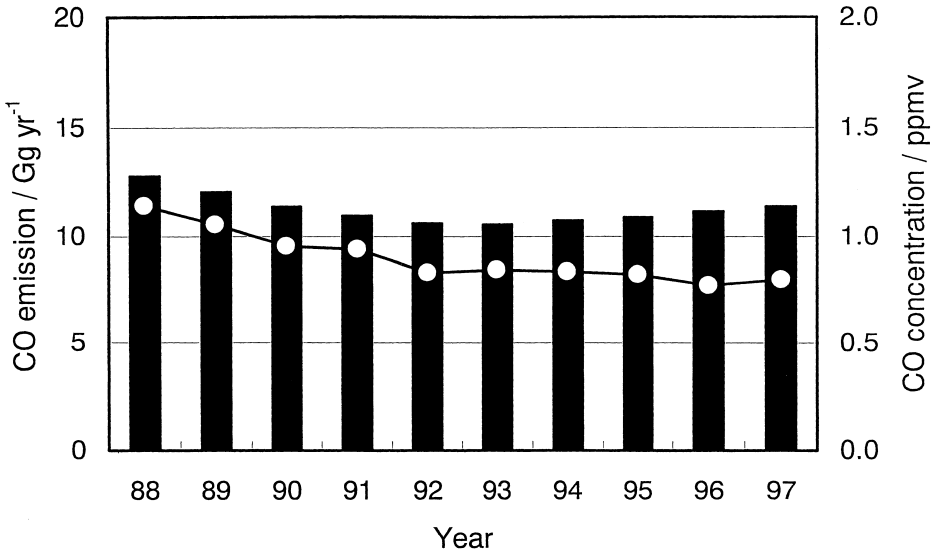

Fig. 6. Temporal variations of CO emissions in Nagoya City and CO concentrations (open circles) at the Higashi-sakura observatory.

CH_4 concentration was considered to elucidate the relationship with the major emission sources of CH_4 in Nagoya City.

The urban excess CH_4 concentrations were obtained as the differences between the CH_4 concentrations observed at Higashi-sakura and Mauna Loa, which are shown in Fig. 2. The results for urban excess CH_4 concentrations are plotted as open circles in Fig. 3. The CH_4 emissions from the major emission sources examined here are also shown as bar graphs in Fig. 3. The urban excess CH_4 concentration gradually increased from 0.21 ppmv in 1988 to 0.24 ppmv in 1993, and then significantly decreased to 0.16 ppmv in 1997. It should also be noted here that the long-term trends of the urban excess CH_4 concentration provided fairly good correlation with that of CH_4 emissions from the landfill "A". These results suggest that the urban excess CH_4 concentration is mainly influenced by the regional CH_4 emissions from the landfill "A". In the previous paper,[15] we reported that the atmospheric CH_4 concentration in the central city of Nagoya was significantly enhanced by the air flow from the "NE" sector (from 0° to 90°), where the largest landfill site "A" was located on the path of northeastern wind blown into the central city. Thus, it is noted here that the present result concerning the urban excess CH_4 concentration is consistent with the kinetic behaviors of CH_4 observed in the Nagoya metropolitan area.

On the contrary, it is seen from Fig. 3 that the long-term trends of CH_4 emissions from automobiles had no correlation with those of the urban excess CH_4 concentration. These results indicate that the CH_4 emissions from automobiles do not much contribute to the excess CH_4 concentration in the urban atmosphere. In order to confirm the influences of automobiles on trace gases in the urban atmosphere, the effects of the traffic flows on the atmospheric CO and CH_4 concentrations were further investigated. Since the human activities are generally larger on weekdays than on weekends, the data sets of the traffic flows, CO, and CH_4 were investigated separately for weekdays and weekends. In Fig. 7 (a), the data set of the traffic flow census, which was obtained from 4 roads around the Higashi-sakura observatory on Tuesdays and Sundays in October, 1997, is shown separately for weekdays and weekends. As is seen in Fig. 7 (a), the traffic flows on the weekdays and weekends are substantially different from each other, particularly in the morning rush-hour time. The daily changes of annually-averaged CO and CH_4 concentrations at the Higashi-sakura observatory from April, 1997 to March, 1998, which are the concentration values averaged through the year at the same daily time, are also shown in Figs. 7 (b) and (c). The error bars represent the standard deviations of the averages. Figures 7 (b) and (c) show that the CO concentration significantly varied on weekdays and weekends, while the atmospheric CH_4 concentration did not. These facts indicate that the CO concentration in the urban atmosphere is clearly affected by the traffic flows, while the urban CH_4 concentration is not substantially influenced by the CH_4 emissions from automobiles.

Rice paddy fields are another major source for CH_4 emission, especially during the flooded period in early summer. However, in the summer season, the vertical mixing of air most strongly occurs, so that the air mass of the high CH_4 concentration near the rice paddy fields is easily diluted to the background concentration level. Therefore, the CH_4 emissions from rice paddy fields exerted low influences on its concentrations measured at the Higashi-sakura observatory, although the high CH_4 concentration was clearly observed at Tomita surrounded by the paddy fields in July.

Conclusion

The atmospheric CH_4 concentration in 1988–1997 was examined in relation with CH_4 emissions from the major sources, which were estimated from some statistical data sets in Nagoya metropolitan area. The results indicate that the landfills are the most dominant emission sources, generating 2.60–3.42 Gg of CH_4 for 1988–1997. The annual CH_4 emissions from automobiles and rice paddy fields were 9–27% and 7–11% of those from landfills, respectively. It was also found that the long-term evolution of major CH_4 emissions from the

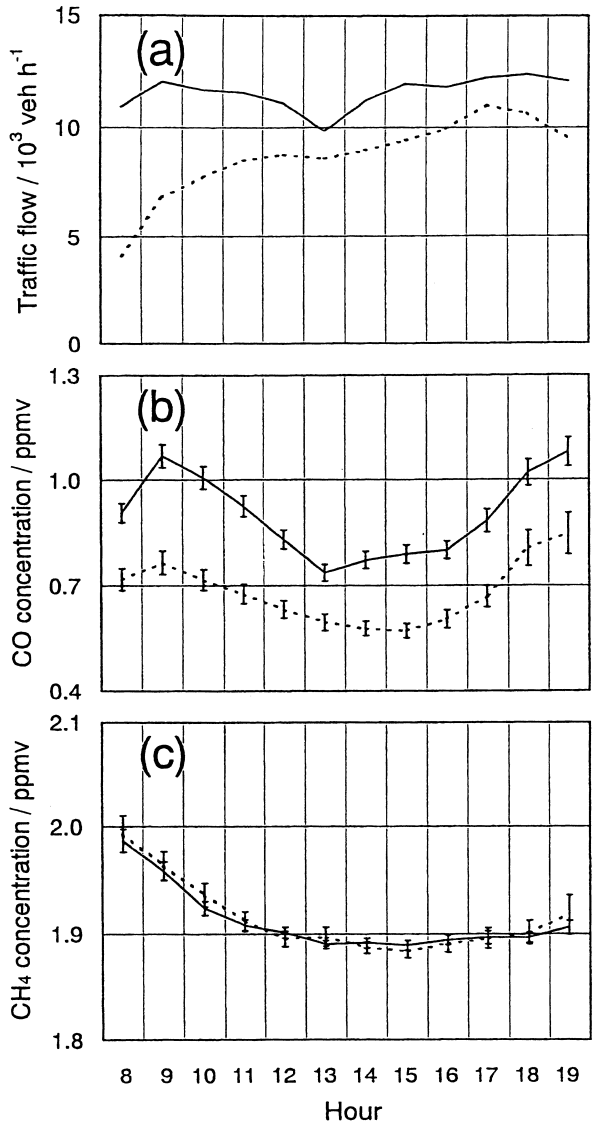

Fig. 7. Daily changes of (a) traffic flows, (b) atmospheric CO concentration, and (c) atmospheric CH_4 concentration at the Higashi-sakura observatory for weekdays (continuous line) and weekends (dashed line).

largest landfill sites showed fairly good correlation with that of the urban excess CH_4 concentration. These results clearly showed the recent decline of CH_4 emissions in the Nagoya metropolitan area since 1993. It was also elucidated that the CH_4 emissions from the largest landfill site provided the greatest impacts on the urban excess CH_4 concentration among the major emission sources in Nagoya. The significant decrease in the CH_4 emissions from the landfills was caused by the reduction in the amount of organic wastes directly dumped into the landfills owing to the facilities for crushing and incineration as well as for sorting into flammable and inflammable refuses and recycling resources. It is thus concluded from these results that the anthropogenic CH_4 emissions may be substantially reduced by the mitigation efforts for waste management in a short term.

The present authors express their sincere thanks to the Environmental Affairs Bureau of Nagoya City and to the National Oceanic and Atmospheric Administration's Climate Monitoring and Diagnostics Laboratory for the kind supplies of data used in the present research.

References

1 D. M. Etheridge, L. P. Steel, K. J. Francey, and R. L. Langenfelds, *J. Geophys. Res.*, **103**, 15679 (1998).
2 E. J. Dlugokencky, K. A. Masarie, P. M. Lang, and P. P. Tans, *Nature*, **393**, 447 (1998).
3 R. Prinn, D. Cunnold, P. Simmonds, F. Alyea, R. Boldi, A. Carwford, P. Fraser, D. Gutzler, D. Hartley, R. Rosen, and R. Rasmussen, *J. Geophys. Res.*, **97**, 2445 (1992).
4 I. Fung, J. John, E. Matthews, M. Prather, L. P. Steel, and P. J. Frazer, *J. Geophys. Res.*, **96**, 13033 (1991).
5 R. Hein, P. J. Crutzen, and M. Heimann, *Global Biogeochem. Cycles*, **11**, 43 (1997).
6 S. Houweling, F. Dentener, J. Lelieveld, B. Walter, and E. J. Dlugokencky, *J. Geophys. Res.*, **105**, 8981 (2000).
7 S. Subak, *Environ. Sci. Poll.*, **2**, 229 (1999).
8 D. Blaha, K. Bartlett, P. Czepiel, R. Harriss, and P. Crill, *Atmos. Environ.*, **33**, 243 (1999).
9 D. Y. C. Leung and Y. T. Lee, *Atmos. Environ.*, **34**, 4487 (2000).
10 I. Levin, H. Glatzel-Mattheier, T. Marik, M. Cuntz, M. Schmidt, and D. E. Worthy, *J. Geophys. Res.*, **104**, 3447 (1999).
11 M. Aikawa, K. Yoshikawa, M. Tomida, and H. Haraguchi, *Environ. Sci.*, **9**, 201 (1996).
12 J. Moriizumi, K. Nagamine, T. Iida, and Y. Ikebe, *Atmos. Environ.*, **32**, 2947 (1998).
13 H. Otsuka, H. Inoue, K. Yoshida, and K. Hayakawa, *Chikyukagaku* (Geochemistry), **29**, 75 (1995).
14 A. Ito, I. Takahashi, Y. Nagata, K. Chiba, and H. Haraguchi, *Sci. Total Environ.*, **263**, 37 (2000).
15 A. Ito, I. Takahashi, Y. Nagata, K. Chiba, and H. Haraguchi, *Atmos. Environ.*, **35**, 3137 (2001).
16 E. J. Dlugokencky, L. P. Steele, P. M. Lang, and K. A. Masarie, *J. Geophys. Res.*, **100**, 23101 (1995).
17 Japanese Report Based on the Intergovernmental Panel on Climate Change (IPCC)/Framework Convention on Climate Change (FCCC), Japanese government (1994).
18 M. Tanaka, M. Miyazaki, and I. Watanabe, "Industrial Sources," ed by K. Minami et al., NIAES (1994).
19 Y. Matsufuji, and A. Tachifuji, *Waste Manag. Res.*, **8**, 438 (1997).
20 J. Fukuyama, K. Yamada, and A. Honda, Report of Osaka City Institute of Environmental Sciences, **44**, 36 (1981).
21 G. J. Farquhar and S. A. Rovers, *Water, Air, Soil, Pollut.*, **2**, 493 (1973).
22 "Report of Investigation on the Greenhouse Gases Emitted from the Automobile," Environmental Agency of Japan (1992).
23 K. Yokota, S. Matsunaga, and K. Ohsawa, *Catalyst*, **41**, 43 (1999).
24 A. Sjödin, K. Andréasson, *Atmos. Environ.*, **34**, 4657 (2000).
25 K. Yagi and K. Minami, *Soil Sci. Plant Nutr.*, **36**, 599 (1990).
26 K. D. M. Karim, H. Matsui, and R. Guensler, *Transp. Res., Part D4*, **1998**, 3.