

Vertical and meridional distributions of the atmospheric CO₂ mixing ratio between northern midlatitudes and southern subtropics

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[1] The atmospheric CO₂ mixing ratio was measured using a continuous measurement system onboard a Gulfstream-II aircraft between the northern midlatitudes and the southern subtropics during the Biomass Burning and Lightning Experiment Phase A (BIBLE A) campaign in September–October 1998. The vertical distribution of CO₂ over tropical regions was almost constant from the surface to an altitude of 13 km. CO₂ enhancements from biomass burning and oceanic release were observed in the tropical boundary layer. Measurements in the upper troposphere indicate interhemispheric exchange was effectively suppressed between 2°N–7°N. Interhemispheric transport of air in the upper troposphere was suppressed effectively in this region. The CO₂ mixing ratios in the Northern and Southern Hemispheres were almost constant, with an average value of about 365 parts per million (ppm) and 366 ppm, respectively. The correlation between the CO₂ and NO_y mixing ratios observed north of 7°N was apparently different from that obtained south of 2°N. This fact strongly supports the result that the north-south boundary in the upper troposphere during BIBLE A was located around 2°N–7°N as the boundary is not necessary a permanent feature. *INDEX TERMS*: 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 0322 Atmospheric Composition and Structure: Constituent sources and sinks; *KEYWORDS*: CO₂, aircraft, meridional distribution

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1. Introduction

[2] Atmospheric CO₂ is the second most prevalent greenhouse gas, and its mixing ratio has been increasing since the 18th century [e.g., Barnola, 1999]. To predict future CO₂ levels, it is necessary to understand the global carbon cycle. Systematic measurement of the atmospheric CO₂ is one of the most promising methods for determining the distribution and magnitude of natural sources and sinks [e.g., Conway *et al.*, 1994; Keeling *et al.*, 1995; Francey *et al.*, 1995; Nakazawa *et al.*, 1997]. Most of the CO₂ monitoring has been conducted at the surface sites, but limited CO₂ measurements have been carried out in the free troposphere [Pearman and Beardmore, 1984; Nakazawa *et al.*, 1991, 1993; Matsueda and Inoue, 1996; Anderson *et al.*, 1996; Francey *et al.*, 1999; Vay *et al.*, 1999].

[3] To extract information about sources and sinks from observed CO₂ data, many kinds of three-dimensional (3-D) tracer transport models have been developed and used [Denning *et al.*, 1999]. Because the expression of vertical transport is highly important for 3-D models [Denning *et al.*, 1996], the observed information about vertical CO₂ distribution is quite useful for constraining models.

[4] Knowledge of the CO₂ spatial distribution can be a powerful tool for determining atmospheric structure and for understanding the movement of air masses. The CO₂ mixing ratio is strongly affected near the earth's surface by photosynthesis, respiration, oxidation of organic matter, biomass burning, fossil fuel burning and air-sea exchange, but in the free troposphere, CO₂ production and reduction by chemical reaction is quite small. CO₂ data have been used as an air tracer not only in the free troposphere but also in the stratosphere or in troposphere-stratosphere exchanges [Boering *et al.*, 1996; Hintsa *et al.*, 1998].

[5] To investigate the impact of biomass burning and lightning on tropospheric O₃ and O₃ precursor gases, The Biomass Burning and Lightning Experiment phase A (BIBLE A) campaign was conducted using a Gulfstream II aircraft over Indonesia and northern Australia in September and October 1998. During this campaign, atmospheric CO₂ mixing ratios were measured continuously onboard the aircraft to know the emission factors of O₃ precursor gases

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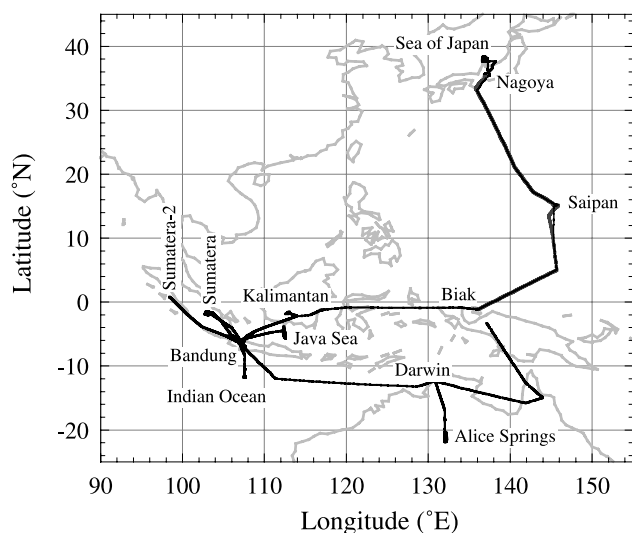


Figure 1. Flight tracks during the BIBLE A campaign.

and to classify origin of air mass. In this paper, we report the results of vertical and meridional distributions of the atmospheric CO₂ from northern midlatitudes to the southern subtropics.

2. Experiment

[6] The BIBLE A campaign was carried out from September 21 to October 10, 1998. Fifteen observation flights were conducted during the mission and their spatial coverage is shown in Figure 1. The aircraft was equipped with a suite of instruments capable of quantifying a number of species including CO₂, NO, NO_y, CO, O₃ and aerosols. CH₄, nonmethane hydrocarbons (NMHCs) and halocarbons were measured from whole air samples collected in stainless steel canisters on the aircraft. A detailed description of the BIBLE A campaign is given by *Kondo et al.* [2001]. The CO₂ observations were carried out using a continuous measurement system with a nondispersive infrared analyzer

(NDIR) onboard the airplane. Figure 2 shows a schematic diagram of the CO₂ measurement system used in this campaign. The outside air was drawn through the inlet port mounted at the top of the fuselage and compressed by a diaphragm pump (Gast, MAA-P108-HB) up to 0.2 MPa. The inlet port was composed of 3/8-inch-diameter stainless tube extending 10 cm out from the fuselage, facing the rear of the aircraft. After passing through a Nafion drier and magnesium perchlorate, the sample air was introduced into a NDIR (LI-COR, LI-6262). The loss of CO₂ by a Nafion drier and magnesium perchlorate is determined to be negligible by introducing standard gases. The sample flow rate was kept constant at 300 standard cc per minute (sccm) by a mass flow controller (STEC, SEC4400 mark3). Standard gases of 342.26 ppm and 386.81 ppm were the CO₂-in-air mixture stored in 2-L aluminum cylinders at 10 MPa. Solenoid valves selected the flows from sample air or standard gases. A standard gas with a lower mixing ratio was continuously introduced into a reference cell at a flow rate of 5 sccm. The absolute pressure of the buffer volume attached to the outlet of two cells was actively controlled to 0.105 MPa by using a piezo valve (STEC, PV-2000) and a pressure sensor (Setra, model 270) to avoid a signal drift of the NDIR associated with changes of cabin pressure. The difference in the pressure of the buffer volume during the flight between the altitude of 13 km and 0.4 km was less than 1×10^{-4} MPa.

[7] During flight, each standard gas was introduced into the sample cell for 30 s every 15 minutes. Data from the NDIR averaged at 1-s intervals were recorded on a personal computer. The CO₂ mixing ratios of sample air were calculated post-flight by interpolating values of two standard gases before and after the sample. The influence of analyzer nonlinearity on the results was estimated to be less than 0.3 ppm.

[8] The response time of the measurement system caused mainly by the air exchange in the sample cell was determined to be about 6 s. The 1-s averaged data are used in this study to detect a short time correlation with other trace gases, although the response time is 6 s. The peak-to-peak

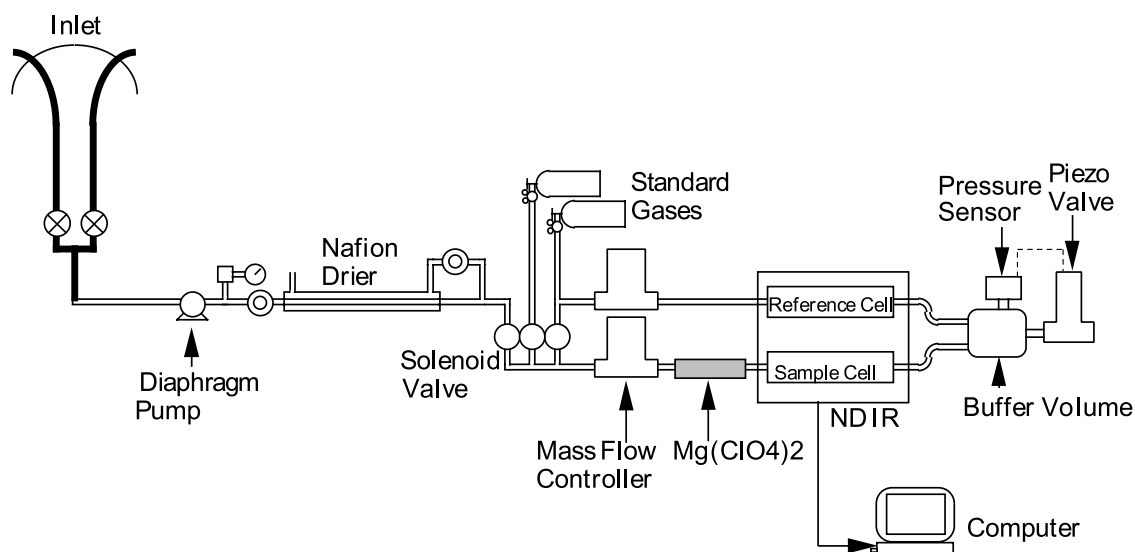


Figure 2. Schematic diagram of the CO₂ measurement system.

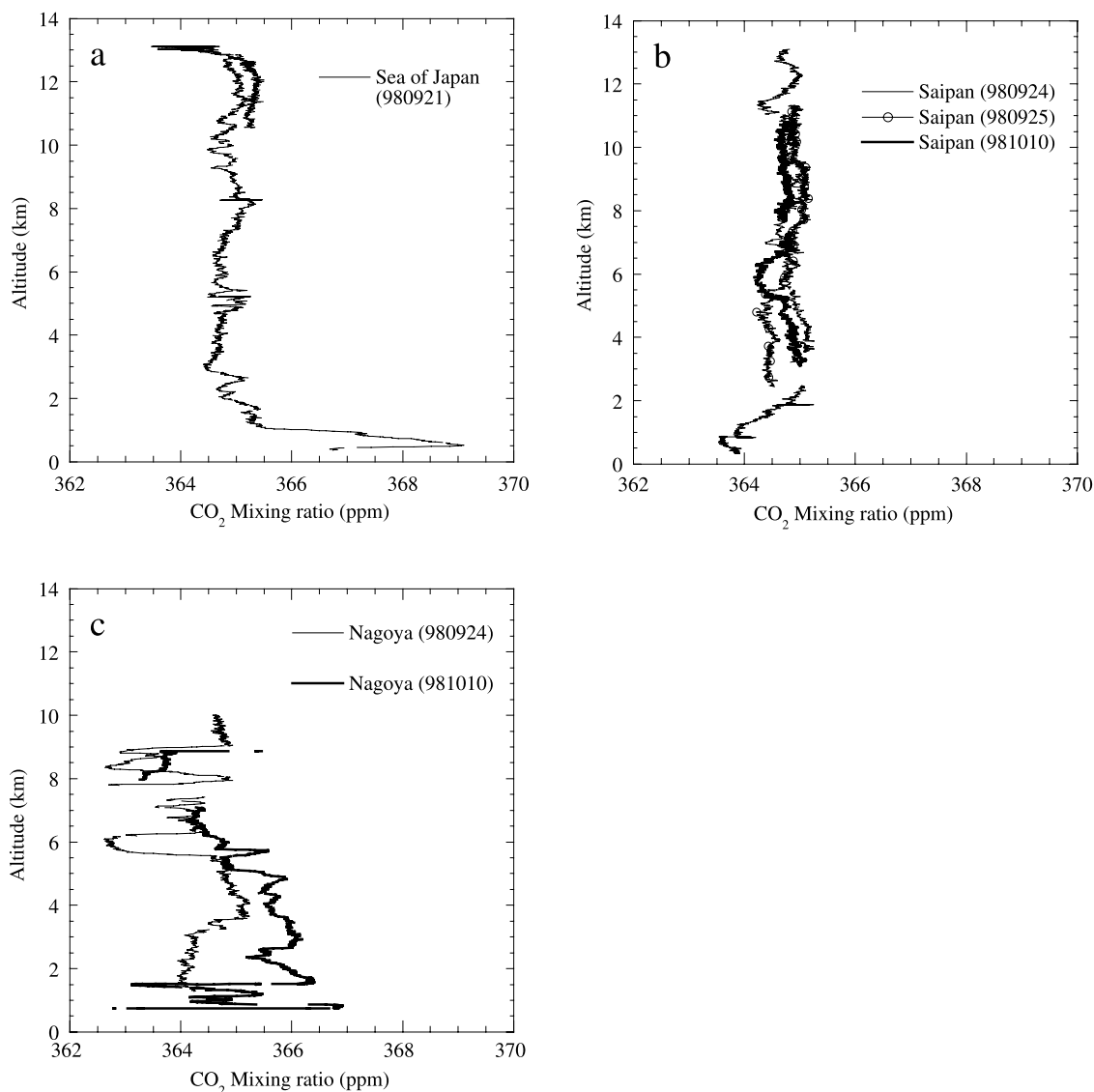


Figure 3. Vertical distributions of the CO₂ mixing ratio over (a) Sea of Japan, (b) Saipan, and (c) Nagoya in the Northern Hemisphere.

noise of data averaged at 1-s intervals was less than 0.2 ppm. The standard gases were calibrated before and after the campaign against the CO₂ standard scale at the National Institute for Environmental Studies (NIES), prepared in 1995 by the gravitational method (NIES95 scale). The concentration differences in 2-L cylinders between before and after the campaign were less than 0.3 ppm. The NIES95 scale was compared with the CO₂ standard scale at the National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostic Laboratory (NOAA/CMDL) in 1996 [Peterson *et al.*, 1997]. The differences in the scales between the two laboratories were within 0.12 ppm in a range between 343 and 372 ppm.

3. Results and Discussion

3.1. Vertical Distribution

[9] The BIBLE A campaign was conducted from late September to early October, which coincided with the seasonal minimum of CO₂ mixing ratios at the surface in

northern middle to low latitudes. The seasonal minimum appears about one month earlier in northern middle to high latitudes [Nakazawa *et al.*, 1997].

[10] The vertical distributions of the CO₂ mixing ratio observed in the Northern Hemisphere during the BIBLE A campaign are presented in Figures 3a–3c. The CO₂ mixing ratios in the free troposphere were almost constant over the Sea of Japan and Saipan, values being 364.5–365 ppm, and indicated little influence from local pollution sources. But layers with low CO₂ mixing ratios (<364 ppm) were seen at 6 km and 8–9 km over Nagoya (Figure 3c) and 13 km over the Sea of Japan (Figure 3a). NO_y mixing ratios were relatively higher (300–500 parts per trillion (ppt)) in those layers suggesting a stratospheric influence. However, CO₂ mixing ratios should be higher in the stratosphere than troposphere in the late summer/early fall in the northern middle or high latitudes [Anderson *et al.*, 1996]. Because of the fact that seasonal CO₂ cycle of the lower stratosphere shows a maximum in September, whereas CO₂ in the upper troposphere shows minimum in summer at the northern

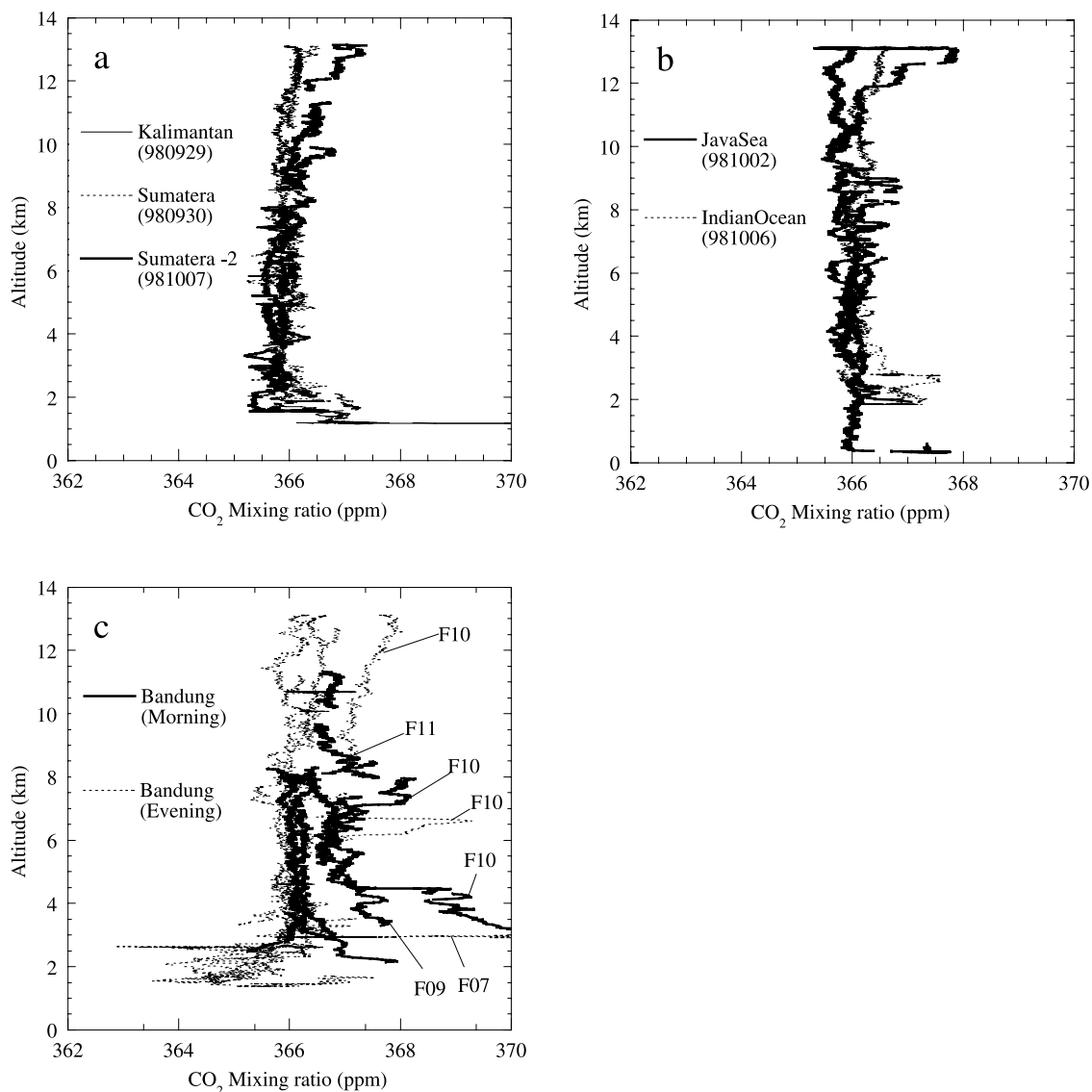


Figure 4. Vertical distributions of the CO₂ mixing ratio over (a) Kalimantan and Sumatera, (b) Java Sea and Indian Ocean, and (c) Bandung in the tropical region.

mid/high-latitudes [Nakazawa *et al.*, 1991]. Therefore these CO₂-depleted layers aren't likely attributable to troposphere/stratosphere exchange.

[11] The land surface acts as a strong CO₂ sink in the summer season because of the active photosynthesis by the land biosphere. Therefore CO₂-depleted air masses may be formed near the land surface and transported to the middle or upper troposphere by convective activities. Anderson *et al.* [1996] also found such structures with the lower CO₂ layer in the northern midlatitudes (30°–40°N) in September 1991 and explained that CO₂-depleted layers were lifted by convective activity over central and northern China and advected to the experiment area (over the western Pacific) by rapid horizontal transport. The enhanced NO_y mixing ratios observed in these layers could therefore result from the biogenic emissions from soils that are known major source of tropospheric NO_y [Yienger and Levy, 1995]. During PEM-West B aircraft observation, the NO_y mixing

ratio in the continental air masses increased significantly between the surface and 4 km, the median value reaching 700–900 ppt in the lower troposphere [Kondo *et al.*, 1997].

[12] Over Saipan, lower CO₂ mixing ratios were found near the surface. Back trajectory analysis using the European Center for Medium-Range Weather Forecasts (ECMWF) suggests that the air mass at 1 km altitude over Saipan had been transported over the western subtropical Pacific Ocean, its origin 5 days before was the middle of the subtropical Pacific (around 21°N, 175°E). The lower mixing ratios of 363.5–364 ppm were due partly to the CO₂ assimilation by local vegetation and partly to CO₂ uptake by the western subtropical Pacific Ocean.

[13] The high CO₂ mixing ratio below 1 km over the Sea of Japan probably had an anthropogenic origin. The NO_y mixing ratio showed a similar structure, that is, lower values (<200 ppt) above 2 km, extremely high values (>2000 ppt) between 1 and 0.5 km and somewhat high values (~450

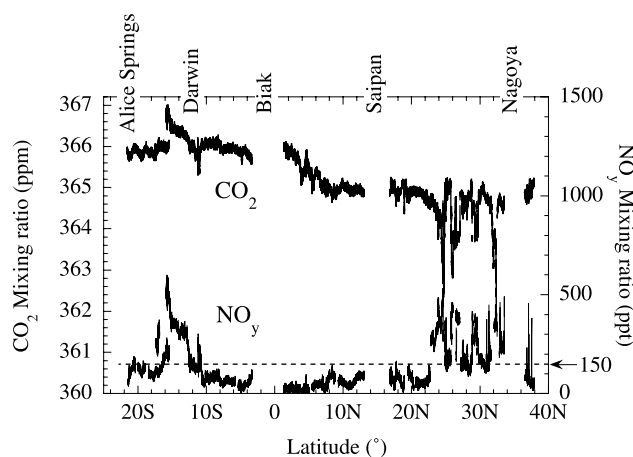


Figure 5. Meridional distributions of CO₂ and NO_y mixing ratios in the upper troposphere observed on the way from Japan.

ppt) at 0.4 km. A high mixing ratio of C₂Cl₄, which is an indicator of industrial activities, was 5.4 ppt at 0.4 km, while upper level mixing ratios were <3 ppt.

[14] Figures 4a–4c show vertical CO₂ distributions observed over a tropical region. The CO₂ mixing ratio was almost constant (Figures 4a and 4b) from the lower to upper troposphere, implying that the seasonal change in biospheric activities in a tropical region is small and/or the air was well mixed by strong vertical convection. The CO₂ values (around 366 ppm) are slightly higher than those obtained in the free troposphere in the Northern Hemisphere. Constant vertical profiles of CO₂ were also found over the tropical region in October, 1991 during the PEM-West A aircraft campaign [Anderson *et al.*, 1996].

[15] Extremely high mixing ratios of CO₂ were observed lower than 1.2 km over Kalimantan (Figure 4a) and the Java Sea (Figure 4b). At lowest altitude of 1.2 km over Kalimantan, NO_y and CO mixing ratios as well as CH₄ and NMHCs had enhanced concentrations, whereas no enhancement of CFCs and C₂Cl₄ was observed. This suggests that biomass burning played a dominant role in the increased CO₂ near the surface over Kalimantan. On the other hand, no enhanced mixing ratios of NO_y, CH₄, NMHCs, CFCs or C₂Cl₄ and slightly decreased O₃ were observed at 0.3–0.6 km over the Java Sea. Hashida [1996] indicated that CO₂ was released from the ocean around the Java Sea by measuring the partial pressure of CO₂ (pCO₂) in the seawater. Therefore observed high CO₂ mixing ratios over the Java Sea were likely caused by CO₂ released from the ocean and accumulated in the marine boundary layer.

[16] Relatively increased CO₂ was observed above 12 km over Sumatera (Figure 4a) and Java Sea (Figure 4b). CO and NO_y mixing ratios also show higher values in these layers. Kita *et al.* [2002] indicated that atmospheric convection was active over Indonesia during the BIBLE A period and the increase of CO in the upper troposphere was explained as the convective transport of surface air influenced by urban pollution and biomass burning. The CO₂ enhancement in the upper troposphere was also possibly caused by the vertical transport of surface air. Lightning associated with convection activity mainly contributed to the observed increase of NO_y [Koike *et al.*, 2002].

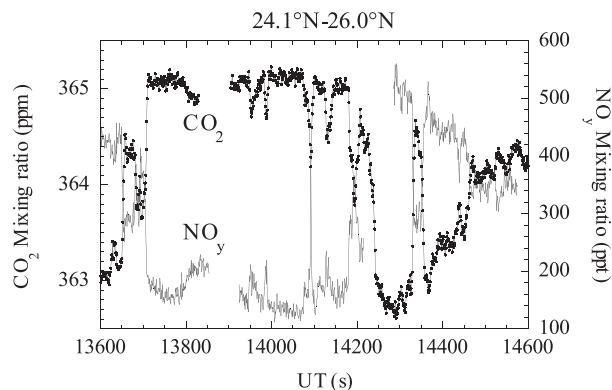


Figure 6. Temporal variations of the CO₂ and NO_y mixing ratios observed from 33°N to 24°N.

[17] Twelve CO₂ profiles were obtained by flight number 06–12 (27 September to 9 October) over Bandung, Indonesia (Figure 4c). Sporadic CO₂ enhancement was observed above 4 km in flight number 09, 10 and 11. NO_y and CO mixing ratios increased simultaneously with CO₂ in these layers. Therefore urban pollution and/or biomass burning also caused CO₂ enhancement in middle troposphere. If we exclude the polluted air (typically >150ppt of NO_y and >80 ppb of CO), CO₂ mixing ratios above 4 km over Bandung are almost constant, being about 366 ppm. At the altitudes lower than 3 km over Bandung, higher mixing ratios were observed early in the morning and lower values were observed in the evening. In the nighttime, large amount of CO₂ is released from land biosphere by respiration and accumulated in the nocturnal inversion layer. On the other hand, land biomass absorb large amount of atmospheric CO₂ by photosynthesis in the daytime. The differences in CO₂ mixing ratio at the lower altitudes were likely created by diurnal changes in activities in the land biosphere around Bandung.

3.2. Meridional Distribution

[18] The meridional distributions of the CO₂ mixing ratio obtained during the level flight from Nagoya to Alice Springs via Saipan, Biak, and Darwin in the upper tropo-

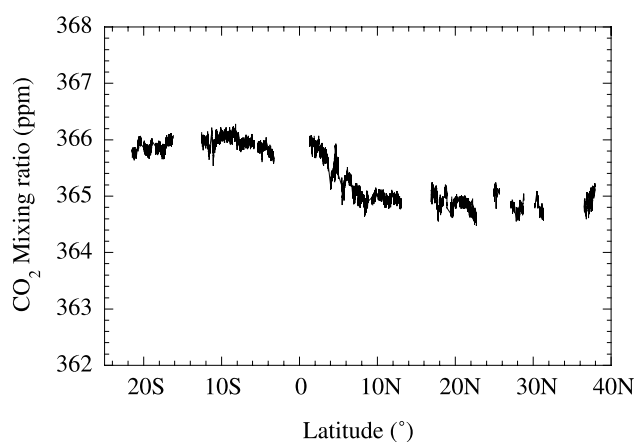


Figure 7. Meridional distributions of the CO₂ mixing ratio in background air in the upper troposphere.

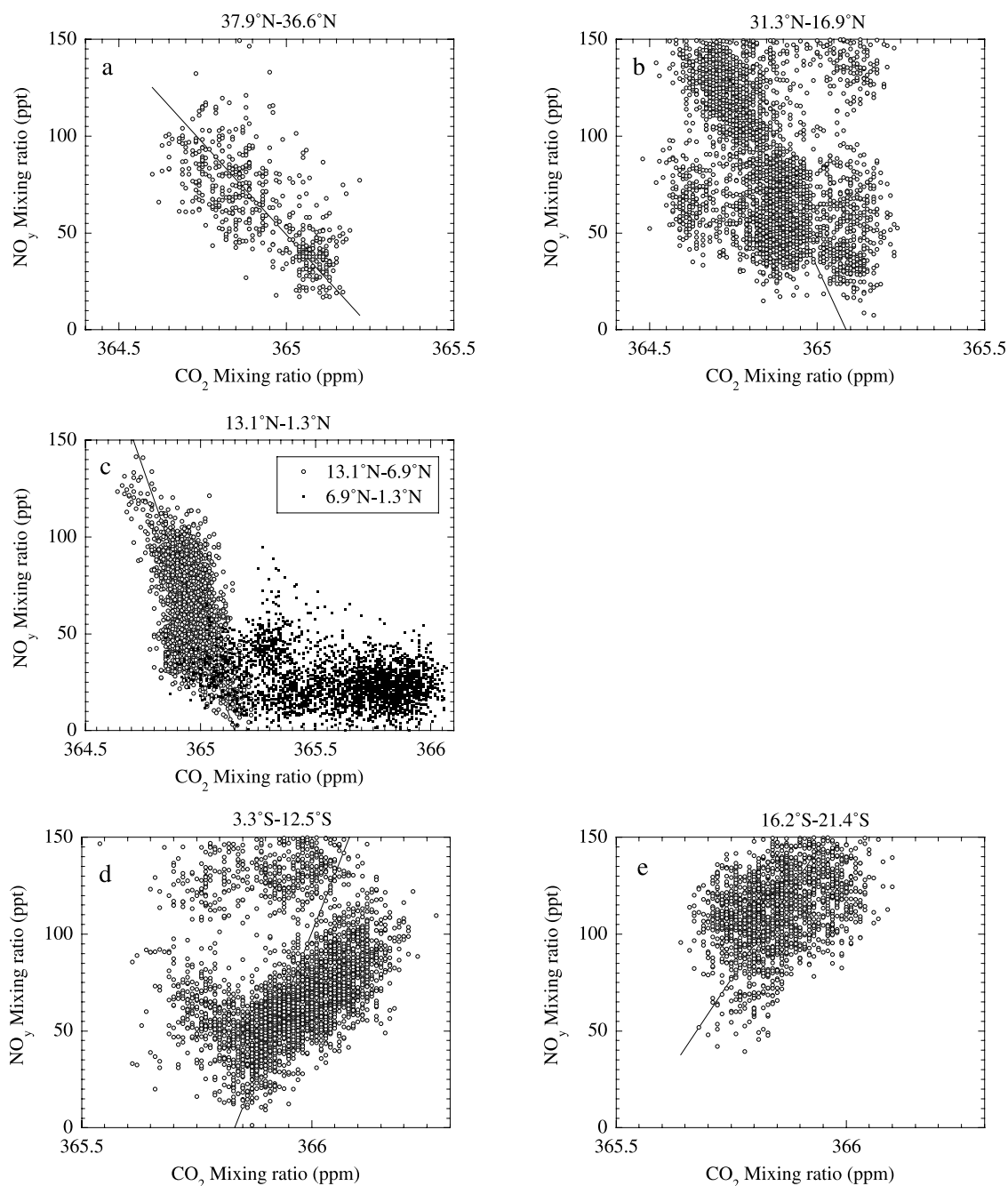


Figure 8. Correlation plot between CO₂ and NO_y in background air in the upper troposphere at the latitude between (a) 37.9°N and 36.6°N, (b) 31.3°N and 16.9°N, (c) 13.1°N and 1.3°N, (d) 3.3°S and 12.5°S, and (e) 16.2°S and 21.4°N. The solid line represents the least squares fitting to the data.

sphere (11–13 km) are presented in Figure 5. The CO₂ mixing ratios in the Northern Hemisphere were generally lower than those in the Southern Hemisphere. Extremely low levels of CO₂ were observed occasionally in the latitude range from 23°N–34°N. The meridional distributions of NO_y mixing ratio are also presented in Figure 5 for reference. In the Northern Hemisphere, the NO_y mixing ratio showed large spatial variations at 23°N–34°N. CO₂ and NO_y were anticorrelated during this period. Higher NO_y mixing ratios were observed where the CO₂ is at lower levels. Variations of the CO₂ mixing ratio in this region

corresponded negatively with NO_y variations (Figure 6), which indicates that changes in CO₂ and NO_y were associated with varying air masses. As mentioned in section 3.1, the air mass with high NO_y and low CO₂ mixing ratios could be from the lower troposphere rather than the stratosphere at northern midlatitudes in the summer season. In addition, higher levels of CO, CH₄ and NMHCs were observed in air with lower CO₂ mixing ratios at 23°N–34°N. Therefore the air mass with low CO₂ and high NO_y mixing ratios was considered to be strongly affected by the land surface and transported to the upper troposphere within

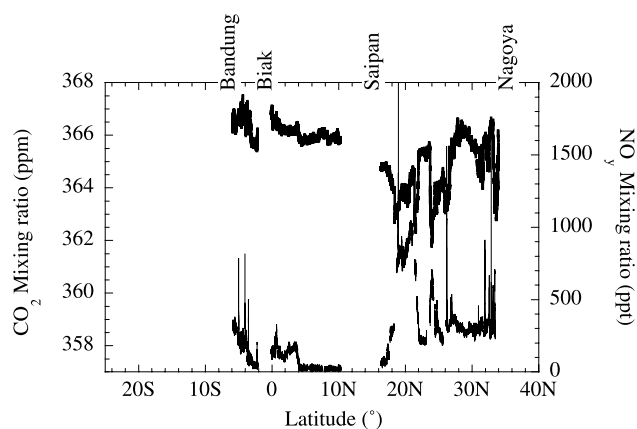


Figure 9. Meridional distributions of the CO₂ mixing ratio in the upper troposphere observed on the way to Japan.

a couple of days. *Matsueda and Inoue* [1996] reported that low mixing ratios of CO₂, which largely exceeded the seasonal variation, with high CH₄ mixing ratios were observed in the upper troposphere around 15°N in October 1993 between Sydney, Australia and Tokyo, Japan. They indicated that the continental surface air was transported upward and affected the sampling sites.

[19] In the Southern Hemisphere, higher CO₂ and NO_y mixing ratios were observed at 13°S–16°S. In this region, enhancement of CO, CH₄ and NMHCs mixing ratios were observed ($\Delta\text{CO}/\Delta\text{CO}_2 = 0.041$ ppm/ppm, $\Delta\text{CH}_4/\Delta\text{CO}_2 = 0.022$ ppm/ppm), whereas CFCs and C₂Cl₄ levels showed no increase. It appears that relatively high CO₂ mixing ratios at 13°S–16°S were due mainly to CO₂ released by biomass burning.

[20] In order to see background levels of the CO₂ mixing ratio in the upper troposphere, CO₂ results affected by the land surface were excluded from the meridional distributions. In this study, air masses with NO_y mixing ratios higher than 150 ppt were considered recently influenced by surface sources (Figure 5). The selected meridional variations of the CO₂ are shown in Figure 7. A clear north-south difference of atmospheric CO₂ mixing ratio in the upper troposphere is evident, with the boundary lying between 2°N and 7°N. *Nishi and BIBLE Science Team* [2001] showed that higher cloud top was seen around 5–10°N along our flight track during 23 September to 12 October 1998 using outgoing longwave radiation. It is almost consistent with the result that the boundary lying between 2°N and 7°N. The CO₂ mixing ratios from 7°N to 38°N and from 2°N to 22°S were almost constant, with average values of 365 ppm and 366 ppm, respectively. This suggests that, if we exclude the air influenced by the land surface, the upper tropospheric air in each hemisphere was mixed well at latitude. Previous studies conducted at similar longitudes indicated that CO₂ mixing ratios in the Northern Hemisphere were lower than those in the Southern Hemisphere in the upper troposphere during Northern Hemispheric summer [*Nakazawa et al.*, 1991; *Matsueda and Inoue*, 1996].

[21] *Nakazawa et al.* [1991] found that the South Pacific Convergence Zone (SPCZ) largely suppressed the interhemispheric air exchange and made a clear discontinuity of CO₂ mixing ratio at 10–12 km around 0°–10°S of observed

longitudes from December to April. The CO₂ discontinuity found in the present study was located at 2°N–7°N in a different season. The continuous CO₂ measurements carried out during the PEM-West A aircraft campaign in 1991 did not show a clear interhemispheric difference in the upper troposphere, even though they flew in September at similar longitudes [*Anderson et al.*, 1996]. Aircraft measurements of CO₂ during the PEM-T campaign found that SPCZ acted as an effective barrier to meridional transport only at lower altitudes over the south Pacific, in August–October 1996 [*Vay et al.*, 1999]. In the upper troposphere, they showed evidence of Northern Hemispheric air being transported to the Southern Hemisphere. This suggests that the air suppression by the interhemispheric boundary in the upper troposphere is not always sufficient.

[22] In background air shown in Figure 7, small CO₂ variations with peak-to-peak amplitudes of 0.5–0.6 ppm along the latitude are seen. Because noise levels of the CO₂ measurement system used in this study are less than 0.2 ppm, the variations seen in Figure 7 are regarded as actual spatial CO₂ changes, with a scale of 10–50 km, in the upper troposphere. To ascertain the characteristics of air in the upper troposphere, 1-s averaged data of CO₂ mixing ratio in background air were compared with the NO_y mixing ratios, Figures 8a–8e. A negative correlation between CO₂ and NO_y is seen in Figures 8a and 8b and to the north of 6.9°N in Figure 8c. On the other hand, a positive correlation is shown in Figures 8d and 8e, and there is no relation to the south of 6.9°N in Figure 8c. The NO_y-CO₂ correlations in each hemispheres are similar to the relation obtained from the air influenced by lower troposphere. It appears that air masses in background conditions maintain characteristics from when the air was in the source/sink region (near the surface). These correlations strongly support the result that the north-south boundary lies around 2°N–7°N along the observed region.

[23] CO₂ latitudinal distributions in the upper troposphere observed during the level flight from Bandung to Nagoya via Biak and Saipan on October 1998 are presented in Figure 9. The CO₂ mixing ratio in the northern midlatitudes changes from 362 ppm to 367 ppm, which is similar to the results obtained during the flight to Australia. Figure 10 shows the meridional CO₂ variations in background air

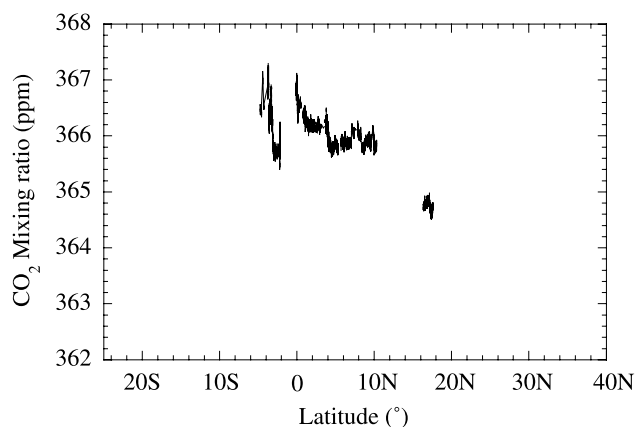


Figure 10. Meridional distributions of the CO₂ mixing ratio in background air observed on the way to Japan.

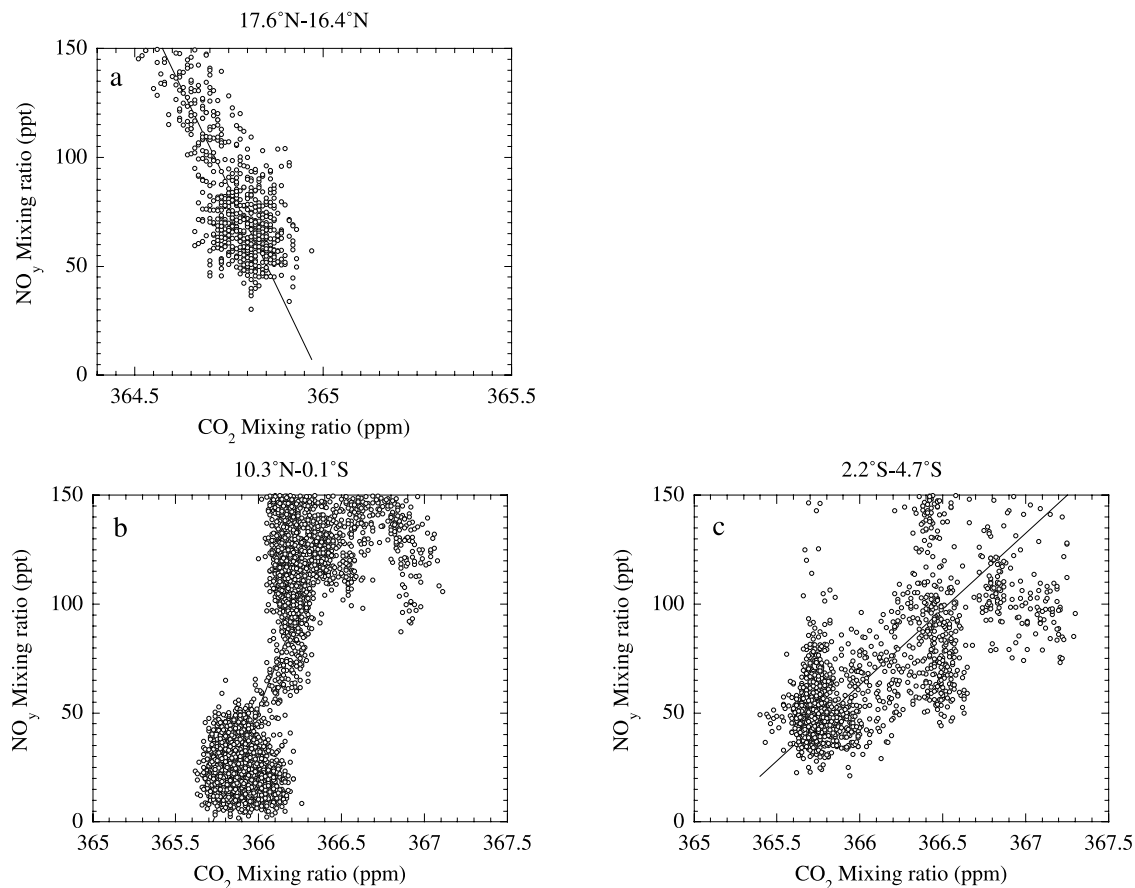


Figure 11. Correlation plot between CO₂ and NO_y in background air in the upper troposphere on the way from Bandung to Nagoya at the latitude between (a) 17.6°N and 16.4°N, (b) 10.3°N and 0.1°S, and (c) 2.2°S and 4.7°S. The solid line represents the least squares fitting to the data.

selected by the same procedures used for Figure 7. As Figure 10 shows, typical background air was rarely found north of 16°N. The CO₂ mixing ratio in the background air is lower in the northern latitudes, but the atmospheric boundary between the Northern and Southern Hemispheres is not evident in Figure 10. The CO₂ mixing ratios presented in Figure 10 are compared with the NO_y mixing ratio in Figures 11a–11c. The characteristics of the air mass at each latitudinal area can be distinguished clearly; a negative correlation between the two mixing ratios can be seen to the north of 16.4°N and a positive correlation can be seen to the south of 10.3°N. As can be seen, Figure 11, the north-south boundary of the upper troposphere appears to lie between 16.4°N and 10.3°N and appears to have moved northward from September to October 1998.

4. Summary and Conclusions

[24] The atmospheric CO₂ mixing ratio was measured to determine the CO₂ spatial distributions between the northern midlatitudes and the southern subtropics. The background CO₂ mixing ratios were about 365 ppm in the Northern Hemisphere and 366 ppm in the Southern Hemisphere, in September and October 1998. These mixing ratios extended from the lower to the upper troposphere. In addition to these levels, relatively high or low mixing ratios that were affected by strong sources or sinks were

sometimes observed. In the upper troposphere at 23°N–34°N, extremely low mixing ratios were observed. This result indicates that air masses previously in contact with the land surface can be transported to the upper troposphere in a wide range of latitudes. Using CO₂ spatial distributions, we showed a clear boundary of air between the Northern and Southern Hemispheres in the upper troposphere. Measuring CO₂ spatial distribution is useful not only for understanding the global carbon cycle but also for understanding the atmospheric structure and the transport of air masses.

[25] **Acknowledgments.** The authors express their appreciation to the staff at the Diamond Air Service Co., Ltd., for operating the aircraft in this campaign. We also thank Y. Tohjima, NIES, Japan for his helpful comments in preparing the manuscript.

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