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## Effects of biomass burning and lightning on atmospheric chemistry over Australia and South-east Asia

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*This paper is derived from a presentation at the conference 'Fire and savanna landscapes in northern Australia: regional lessons and global challenges', Darwin, Australia, 8–9 July 2002*

**Abstract.** *In situ* aircraft measurements of trace gases and aerosols were made in the boundary layer (BL) and free troposphere (FT) over Indonesia and Australia during the Biomass Burning and Lightning Experiment (BIBLE)-A and B conducted in August–October 1998 and 1999. Concentrations of ozone (O<sub>3</sub>) and its precursors [CO, reactive nitrogen (NO<sub>x</sub>), non-methane hydrocarbons (NMHCs)] were measured in these campaigns to identify the sources of NO<sub>x</sub> and to estimate the effects of biomass burning and lightning on photochemical production of O<sub>3</sub>. Over Indonesia, *in-situ* production of NO<sub>x</sub> by lightning was found to be a major source of reactive nitrogen in the upper troposphere during BIBLE-A. In some circumstances, increases in reactive nitrogen were often associated with enhancements in CO and NMHCs, suggesting that the sources were biomass burning and fossil fuel combustion, followed by upward transport by cumulus convection. Over Australia the levels of O<sub>3</sub>, CO, reactive nitrogen, and NMHCs were elevated throughout the troposphere compared to those observed in the tropical Pacific. However, the mechanisms responsible for the enhanced concentrations in the BL and FT are distinctly different. The emissions from biomass burning that occurred in northern Australia were restricted to the BL because of strong subsidence in the period. In the FT over Australia, elevated concentrations of O<sub>3</sub> and its precursors result from injections of emissions as the air masses travel over Africa, South America, the Indian Ocean, and Indonesia en route to Australia. In all cases, O<sub>3</sub> levels in the biomass burning plumes were enhanced due to photochemical production.

## Introduction

Biomass burning is an important source of trace gases [such as  $\text{NO}_x$ ,  $\text{NO}_y$ ,  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{CH}_4$ , non-methane hydrocarbons (NMHCs) and  $\text{CH}_3\text{Cl}$ ] and aerosols (Crutzen and Andreae 1990; Andreae *et al.* 1996; Blake *et al.* 1996; Andreae and Marlet 2001). Generally most of the burning occurs in the tropical or subtropical regions. Emissions of trace gases such as  $\text{NO}_x$  and  $\text{CO}$  from Africa and South America constitute a large fraction of global biomass burning emissions, while those from South-east Asia, Indonesia, and Australia also make substantial contributions (Galanter *et al.* 2000).

The impacts of biomass burning on tropospheric photochemistry over tropical Africa, South America and the Asia/Pacific region have been widely studied by ground-based and airborne measurements (Harriss *et al.* 1988, 1990; Fishman *et al.* 1996; Lindsay *et al.* 1996; Blake *et al.* 1999; Thompson *et al.* 2001). However, similar studies have not been undertaken over South-east Asia and Australia. To address these issues, the Biomass Burning and Lightning Experiments (BIBLE)-A and B were conducted over the region by the Earth Observation Research Center (EORC) of the Japan Aerospace Exploration Agency (JAXA) between 1998 and 2000, in close collaboration with the International Global Atmospheric Chemistry (IGAC) program. The major aims were to:

- (1) Estimate the amount of trace species emitted by biomass burning and lightning;
- (2) Study the long-range transport and chemical transformation of the biomass burning plumes; and
- (3) Study the vertical transport of air impacted by biomass burning and evaluate the impact of  $\text{O}_3$  precursors on  $\text{O}_3$  photochemistry.

This paper summarises the major findings of the campaign.

## BIBLE-A and B experiments

Flights using a Gulfstream-II (G-II) aircraft were conducted between 24 September and 10 October 1998 and between 30 August and 14 September 1999, respectively, as shown in Fig. 1 (Kondo *et al.* 2002a). During the first BIBLE-A, the aircraft flew from Nagoya (35.3°N, 136.9°E) to Bandung (6.9°S, 107.6°E), Indonesia, via Saipan (15°N, 146°E), Biak (1.2°S, 136.1°E), and Darwin (12.4°S, 130.9°E). The return route was Bandung–Biak–Saipan–Nagoya. Coordinated balloon-borne ozonesonde measurements were carried out at Watukosek (7.5°S, 112.6°E) on Java Island, Kototabang (0.20°S, 100.3°E) on Sumatra Island, and Pontianak (0.03°N, 109.3°E) in Borneo Island in Indonesia. Nineteen ozonesonde measurements in total were made from these three stations during the period between 23 September and 9 October 1998 (Fujiwara *et al.* 2003).

During BIBLE-B, the aircraft flew from Nagoya to Darwin via Saipan, and Port Moresby (9.4°S, 147.2°E). The return route was Darwin–Biak–Saipan–Nagoya. Coordinated balloon-borne ozonesonde measurements were carried out in Darwin, Watukosek and Kototabang. Eighteen ozonesonde measurements in total were made from these three stations (six each) between 25 August and 13 September 1999 (Fujiwara *et al.* 2003).

Instruments, species and measurement characteristics are listed in Table 1.  $\text{NO}$ ,  $\text{NO}_2$ , and  $\text{NO}_y$  were measured using a chemiluminescence technique (Kondo *et al.* 1997, 2003; Ikeda *et al.* 2000; Koike *et al.* 2000).  $\text{O}_3$  was measured by using a dual-beam UV-absorption  $\text{O}_3$  photometer (Kita *et al.* 2002). The  $\text{CO}$  concentration was measured by using an automated gas-chromatograph (GC) system with a reduction gas detector (RGD) (Kita *et al.* 2002).  $\text{CO}$  was also measured by the vacuum ultraviolet resonance fluorescence technique during BIBLE-B (Takegawa *et al.* 2001). Whole air samples were collected into 2 L stainless steel sampling canisters and ~64 samples were obtained per flight. The analytical system is capable of quantifying more than 200 trace gases (Blake *et al.* 1996).  $\text{C}_2$ – $\text{C}_8$  NMHCs, halocarbons, and alkyl nitrates were quantified (Blake *et al.* 1996).  $\text{CO}_2$  was measured by a non-dispersive infrared (NDIR) analyser (Machida *et al.* 2002). Dew point was measured with two types of dew point hygrometers, a General Eastern-model 1011A and a Buck Research CR-2. The lower detectable limits are  $-50^\circ\text{C}$  for 1011A and  $-95^\circ\text{C}$  for CR-2.  $\text{H}_2\text{O}$  mixing ratios were calculated from these measurements.

Aerosol measurements were obtained with three instruments: a Multiple-Angle Aerosol Spectrometer Probe (MASP), a Condensation Nucleus Counter (CNC), and a Particle Soot/Absorption Photometer (PSAP) (Liley *et al.* 2002). MASP is an optical particle counter that determines the concentration by size of particles from ~0.3 to 40  $\mu\text{m}$  in diameter. It is mounted below and forwards of the (port) wing tip to minimize prior disturbance of the sample airstream. The CNC and PSAP are rack-mounted within the aircraft, jointly aspirated and vented through the skin of the fuselage. By growing particles in supersaturated *n*-butyl alcohol vapour, the CNC system counts particles above 10 nm in diameter, but detection efficiency is high only for particles above 20 nm diameter. The PSAP measures attenuation of light by accumulated soot on a filter through which sample air is aspirated.

The rate coefficients for  $\text{NO}_2$  and  $\text{O}_3$  photolysis,  $J(\text{NO}_2)$  and  $J(\text{O}^1\text{D})$ , were measured by filter radiometers (Kita *et al.* 2002). The radiometers consist of upward-looking and downward-looking optical units, which were installed on the top and bottom of the fuselage.

In addition to the concentration measured by the instruments on the G-II, a photochemical model was used to calculate quantities along the flight track (Ko *et al.* 2002). The model uses measured concentrations of  $\text{NO}$ ,  $\text{O}_3$ ,  $\text{H}_2\text{O}$ ,  $\text{CO}$ ,  $\text{CH}_4$ , and NMHCs along the flight tracks to calculate the

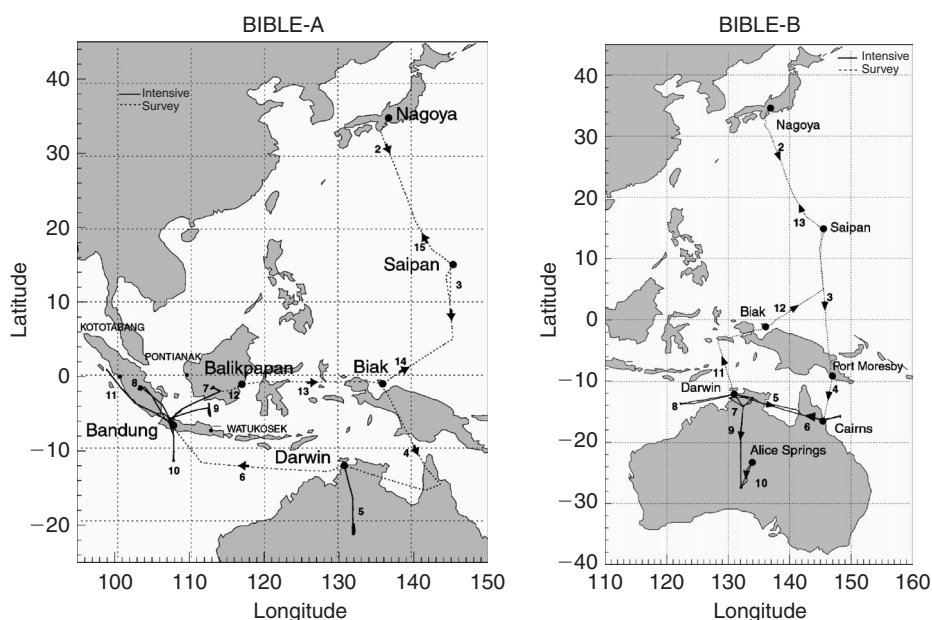


Fig. 1. Flight paths for the G-II aircraft during BIBLE-A and B campaigns.

Table 1. Instruments used aboard the G-II aircraft during BIBLE-A and B

Species/parameter	Technique	Averaging time (s)	Accuracy	Precision	Limit of detection (pptv)
NO	Chemiluminescence	1	8%	6 pptv	13 pptv
NO <sub>y</sub>	Chemiluminescence	1	17%	16 pptv	28 pptv
O <sub>3</sub>	UV absorption	1	5%	0.6 ppbv	1.2 ppbv @500 hPa
CO	Resonance fluorescence	1	5%	1.4 ppbv	2.2 ppbv
CO	GC/HgO-reduction	20	5%	2 ppbv	4 ppbv
CO <sub>2</sub>	IR absorption	1	0.3 ppmv	0.1 ppmv	–
H <sub>2</sub> O	Cryogenic chilled mirror hygrometer	60	±0.2–0.5°C	±0.2–0.5°C	–100°C to +20°C
H <sub>2</sub> O	Thermoelectric hygrometer	60	±0.5–1.0°C	±0.5–1.0°C	–75°C to +50°C
NMHCs (C <sub>2</sub> –C <sub>10</sub> )	Grab sample/GC	60	5–10%	1–3%	3 pptv
Halocarbons (C <sub>1</sub> –C <sub>2</sub> )	Grab sample/GC	60	2–20%	1–10%	0.02–50 pptv
Alkyl nitrates (C <sub>1</sub> –C <sub>4</sub> )	Grab sample/GC	60	10–20%	1–10%	0.02 pptv
Aerosol size distribution	MASP	1	30%	<1%	0.1 cm <sup>–3</sup>
CN	CN counter	1	5√N	0.04 cm <sup>–3</sup>	0.04 cm <sup>–3</sup>
Black carbon	Aethalometer	10	40%	0.1 μg m <sup>–3</sup>	0.1 μg m <sup>–3</sup>
J(NO <sub>2</sub> )	Filter radiometer	1	8%	1.5(–4) s <sup>–1</sup>	3.0(–4) s <sup>–1</sup>
J(O <sup>1</sup> D)	Filter radiometer	1	15%	0.8(–6) s <sup>–1</sup>	2.5(–6) s <sup>–1</sup>

concentrations of the O<sub>x</sub> radicals, the HO<sub>x</sub> radicals, and the nitrogen species at the sampling points. The calculations make use of the measurements from the radiometer to scale the clear sky photolysis rates to account for cloud cover and ground albedo at the sampling point. The concentrations of the nitrogen species are computed assuming they are in equilibrium with the measured NO. The results from the box model are then used in a diurnal model to estimate the diurnally averaged production and removal rates for O<sub>3</sub> at the sampling point.

### Biomass burning and lightning activities

Global distribution of biomass burning (hot spots) is available from the Along Track Scanning Radiometer (ATSR) World

Fire Atlas (<http://shark1.esrin.esa.it/FIRE/AF/ATSR/>). For this study, we used information for September and October 1998, August–September 1999, and August–October 1997. The last period corresponds to higher than normal burning activities in 1997 (Fujiwara *et al.* 1999; Kita *et al.* 2000). Biomass burning activity over Indonesia was much lower than average in October 1998. The amount of available fuel in 1998 was much smaller because there was not enough time for biomass to regenerate after the large fires in 1997. In addition, La Niña prevailed during this period, causing higher convective activity and leading to higher humidity and higher precipitation. La Niña prevailed through 1998 but was significantly weaker by September 1999. Although there was some influence of biomass burning or urban pollution on CO,

NMHCs, and  $\text{NO}_x$  concentrations, the enhancements were much less compared with those in 1997 (Kita *et al.* 2002). During BIBLE-B, biomass burning activity was high over northern Australia as observed by ATSR.

Lightning activity in September–October 1998 was observed by the Optical Transient Detector (OTD) on board the MicroLab satellite (<http://thundr.msfc.nasa.gov/otd/>) (Nesbitt *et al.* 2000) and Lightning Imaging Sensor (LIS) on board TRMM (<http://thundr.msfc.nasa.gov/lis/>). Generally, lightning activity was high over South-east Asia, Central Africa, South America, and southern North America during the BIBLE-A and B periods.

### Findings from BIBLE-A and B

The summary of the results will be given in terms of four types of air-masses:

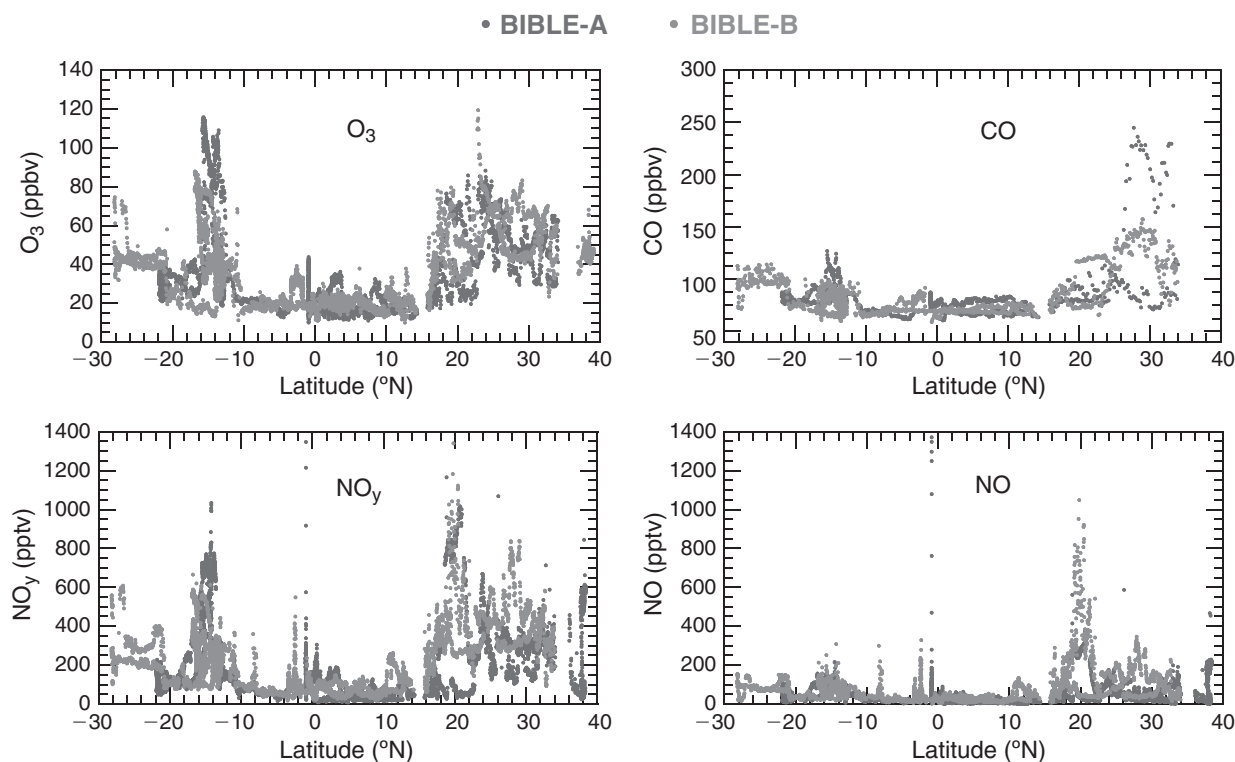
- (1) The tropical Pacific air masses, which are typically unaffected by recent emissions from land;
- (2) Air masses over Indonesia, which are typical of regions with convective activities where emissions from the ground are quickly transported to the free troposphere (FT; altitude 3–13.5 km);
- (3) The air masses in the boundary layer (BL; altitude 0–3 km) over northern Australia, which are strongly affected by biomass burning in Australia; and

- (4) The air masses in the FT over Australia, which are affected by biomass burning in regions away from Australia, such as Africa and South America.

### Tropical Pacific air masses

The mixing ratios of  $\text{O}_3$ , CO,  $\text{NO}_y$ , NO, and NMHCs over the western Pacific Ocean at 120°–150°E in August–October 1998 and 1999 showed large latitudinal variations from northern midlatitudes to the equatorial region, with the lowest values at 15°N–5°S throughout all altitudes (Kondo *et al.* 2002b). Between 0.5 and 4 km, the median  $\text{O}_3$ , CO,  $\text{NO}_y$ , and NO mixing ratios were ~5–15 ppbv, 60–75 ppbv, 20–70 pptv, and 3–5 pptv, respectively, with the smallest  $\text{O}_3$  and  $\text{NO}_y$  values observed near the surface. The  $\text{O}_3$ , CO,  $\text{NO}_y$ , and NO mixing ratios at 8–13 km are shown in Fig. 2. Over the tropical Pacific, the NO and  $\text{NO}_y$  values reaching 300–1000 pptv were often observed at 10–12 km, due to the NO production near New Guinea, as expected from the frequent lightning flashes observed by LIS and high convective activity in October–March. Similar conclusions were reached for observations during Pacific Exploratory Mission-West (PEM-W)-B (Kawakami *et al.* 1997).

Active convection in the ITCZ and SPCZ prevailed in the tropical Pacific. Trajectory analysis demonstrates that the tropical Pacific air was transported from the BL at 15°N–5°S east of the sampling locations by extensive convection in the



**Fig. 2.** Latitudinal variations in the mixing ratios of  $\text{O}_3$ , CO,  $\text{NO}_y$ , and NO above 8 km during BIBLE-A and B. Units are parts per billion/trillion by volume (ppbv; pptv).

ITCZ, indicating that the tropical air was nearly completely isolated from midlatitude air. Similarly low values were observed in the upper troposphere over the same region during PEM-W-A in September–October and in convective air masses during PEM-W-B in February (not shown). Anthropogenic hydrocarbons are mostly emitted at mid-latitudes. Since the tropical Pacific air masses were isolated from mid-latitude influence, the concentrations of the long-lived anthropogenic halocarbons in the upper troposphere should be typical of the global background values. This was the case and, in fact, several halocarbons showed decreases in concentrations compared to several years ago and attest to the success of international environmental treaties (Elliott *et al.* 2002).

#### Indonesian air masses

Mixing ratios of  $\text{NO}_x$  and  $\text{NO}_y$  in air masses that had crossed over the Indonesian islands within 3 days before the measurement (Indonesian air masses) were systematically higher than those in air masses originating from the tropical Pacific (Fig. 2) (Koike *et al.* 2002). In Indonesian air masses,  $\text{NO}_x$  and  $\text{NO}_y$  mixing ratios at 12 km were as high as 80 and 200 pptv [these data are not included in Fig. 2]. This is surprising since 60% of the Indonesian air masses at 9–13 km originated from the Central Pacific. The differences in  $\text{NO}_x$  ( $\text{dNO}_x$ ) and  $\text{NO}_y$  ( $\text{dNO}_y$ ) mixing ratios between Indonesian and tropical air masses (70 and 130 pptv) were likely due to processes that occurred while air masses were over the Islands.

At altitudes below 3 km, typical  $\text{dNO}_y/\text{dCO}$  and  $\text{dNO}_x/\text{dCO}$  ratios were smaller than those in the biomass burning plumes and in urban areas. This suggests that neither source has a dominant influence and that the  $\text{NO}_y$  concentration in the lower troposphere resulted from various land processes averaged over a large area. This is consistent with the low biomass burning activity during BIBLE-A and BIBLE-B and strong convective activity (primarily due to the La Niña condition) during the BIBLE-A period. During BIBLE-A and -B, mixing ratios of  $\text{O}_3$  precursor gases were higher than the marine background values in the upper troposphere above 8 km over Indonesia (Kita *et al.* 2002). Median mixing ratios of  $\text{O}_3$ ,  $\text{NO}$ ,  $\text{NO}_y$ ,  $\text{CO}$ ,  $\text{C}_2\text{H}_6$ ,  $\text{C}_2\text{H}_4$ ,  $\text{C}_2\text{H}_2$ , and  $\text{C}_3\text{H}_8$  in the upper troposphere, including short-lived  $\text{C}_2\text{H}_4$ , increased along the prevailing easterlies over these three regions (i.e. tropical Pacific Ocean < eastern Indonesia < western Indonesia), indicating that this increase was caused by upward transport of  $\text{O}_3$  precursor gases from the surface to the upper troposphere in recent convection. Convection also caused  $\text{NO}$  production by lightning as discussed above.

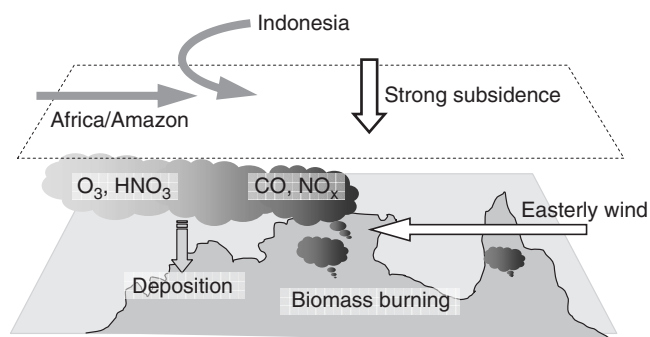
When the  $\text{dNO}_y/\text{dCO}$  and  $\text{dNO}_x/\text{dCO}$  ratios for the upper troposphere are compared to the values found in the lower troposphere, the  $\text{dNO}_x$  and  $\text{dNO}_y$  values are higher by 40–60 pptv (80% of  $\text{NO}_x$ ) and 70–100 pptv (50% of  $\text{NO}_y$ ). This difference is attributed to *in-situ* production of  $\text{NO}_x$  and

$\text{NO}_y$  from lightning. Analyses using air mass trajectories and Geostationary Meteorological Satellite (GMS) derived cloud height data show that convection over the land, which could be accompanied by lightning activity, increases the  $\text{NO}_x$  values, while convection over the ocean generally lowers the  $\text{NO}_x$  concentration. As a consequence of the high  $\text{NO}_x$  concentrations, net photochemical  $\text{O}_3$  production rates of 1–2 ppbv/day were found in both air masses in the upper troposphere. These results suggest that  $\text{NO}_x$  production by lightning over the Indonesian islands had a significant impact on the  $\text{O}_3$  budget over the western Pacific.

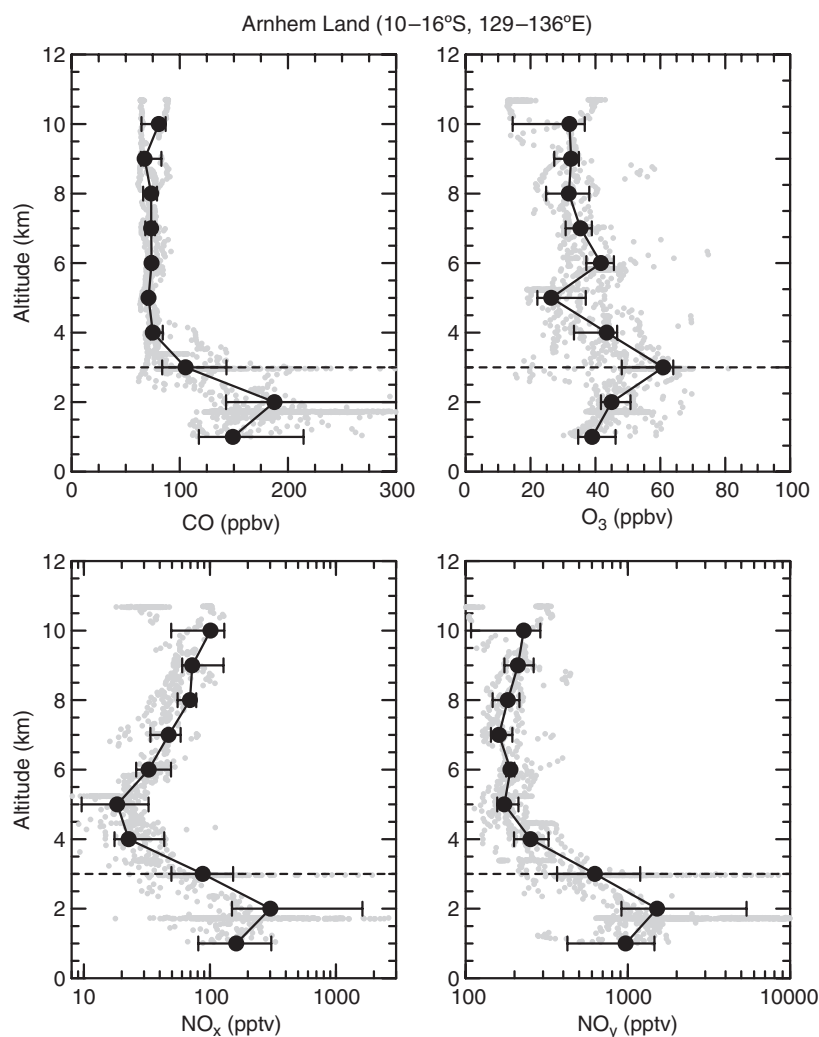
Trajectory analyses show that air masses transported from western Indonesia were observed south of  $9^\circ\text{S}$  over the ocean south of Indonesia and northern Australia (Kita *et al.* 2002). In these air masses (Indonesian outflow air), the mixing ratios of  $\text{O}_3$  precursors except for  $\text{C}_2\text{H}_4$  were similar to those in western Indonesia. However, the median  $\text{O}_3$  concentration (33 ppbv) was higher than that found over western Indonesia (22 ppbv) and tropical Pacific (16 ppbv). This result indicates that photochemical production of  $\text{O}_3$  proceeded in the air masses transported from Indonesia, and the average net  $\text{O}_3$  production rate is estimated to be 1.8 ppbv/day, being consistent with that estimated by using a photochemical box model (Ko *et al.* 2002). A similar increase of  $\text{O}_3$  and its precursors in the air mass transported from Indonesia was observed in the upper troposphere over northern Australia during BIBLE-B. In this case, the median  $\text{O}_3$  mixing ratio (40 ppbv) in the Indonesian outflow air was also significantly higher than the average  $\text{O}_3$  mixing ratio (25 ppbv) obtained from ozonesonde observations at Kototabang, western Indonesia (Kita *et al.* 2002; Fujiwara *et al.* 2003).

#### Australian air masses

Due to stable high pressure over northern Australia, downward motion was predominant both in the upper and lower troposphere for the duration of BIBLE-B as shown schematically in Fig. 3 (Kondo *et al.* 2002b; Takegawa *et al.* 2003a). Temperature profiles obtained by radiosonde over Darwin



**Fig. 3.** Schematic representation of the typical transport pattern of biomass burning plumes over northern Australia during the BIBLE-B period.



**Fig. 4.** Altitude profiles of CO, O<sub>3</sub>, NO<sub>x</sub>, and NO<sub>y</sub> over the Arnhem Land region (10–16°S, 129–136°E). Shaded dots show 10-s averaged data. Solid circles and bars represent median values and central 67 percentile, respectively. The altitude of the upper boundary of the BL is indicated as dashed lines.

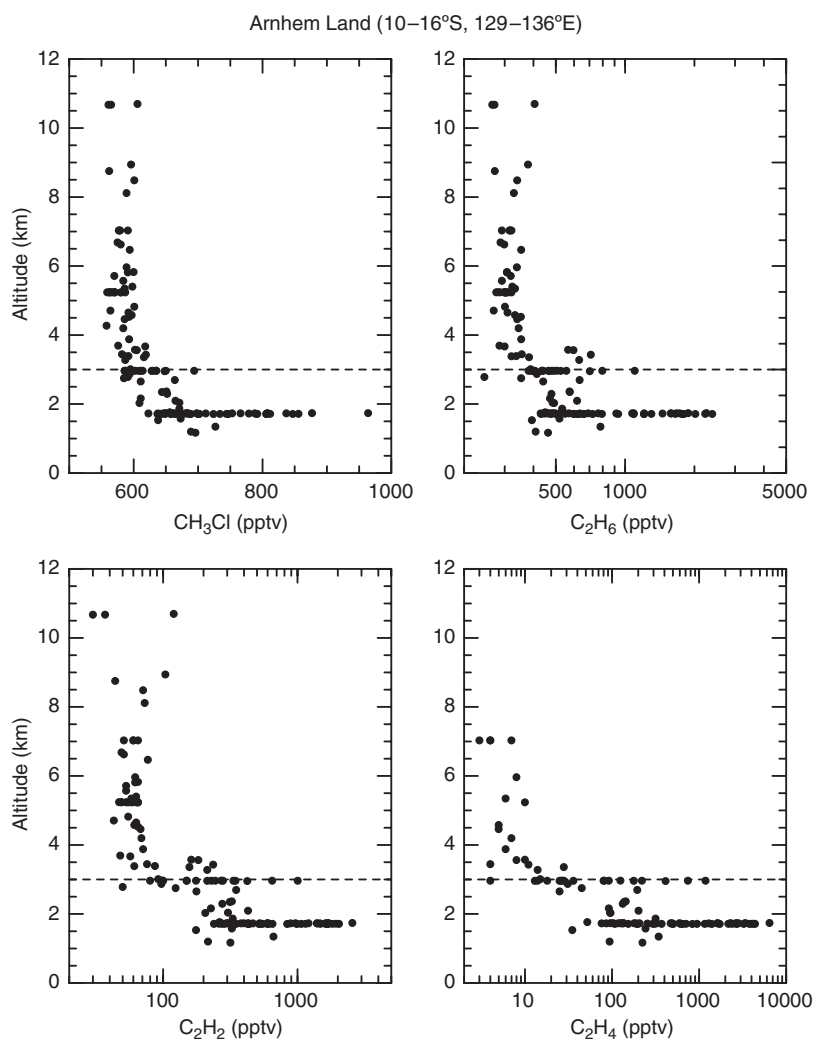
showed that distinct and stable inversion layers repeatedly appeared at 2–4 km. The well defined inversion layers and strong downward motion prevented the biomass burning emissions from going up to the FT during BIBLE-B, as shown in Figs 4 and 5. Thus, air in the BL and the FT are affected by different processes. The results will be discussed separately below.

#### *Biomass burning emissions*

During BIBLE-B local flights, significant enhancements of trace gases such as CO and NMHCs were observed in the BL (<3 km) over the region of intensive fire activity. The air masses with CO > 800 ppbv were coincident with the locations of biomass burning hot spots observed by Advanced Very High Resolution Radiometer (AVHRR), as shown in Fig. 6. Concentrations of short-lived NMHCs such as ethene

(C<sub>2</sub>H<sub>4</sub>) and propene (C<sub>3</sub>H<sub>6</sub>) were very high in these air masses, indicating that they were strongly influenced by recent biomass burning emissions (less than 1 day residence time).

Emission ratios (relative to CO) were determined for NO<sub>x</sub> and NMHCs based on the measured BL enhancements over the Arnhem Land region. Tight correlations of these species with CO were obtained for this region, indicating the homogeneity of the source, which apparently represents the local savanna burning emissions (Shirai *et al.* 2003). The emission factors were derived from the emission ratios and the assumed emission factor for CO. The emission factors depend heavily on the combustion efficiencies of the fire and emission characteristics of each compound. Higher combustion efficiency and a lower emission factor of methane compared to forest fires were observed, which agreed well with savanna fires



**Fig. 5.** Altitude profiles of  $\text{CH}_3\text{Cl}$ ,  $\text{C}_2\text{H}_6$ ,  $\text{C}_2\text{H}_2$ , and  $\text{C}_2\text{H}_4$  over the Arnhem Land region (10–16°S, 129–136°E). The altitude of the upper boundary of the BL is indicated as dashed lines.

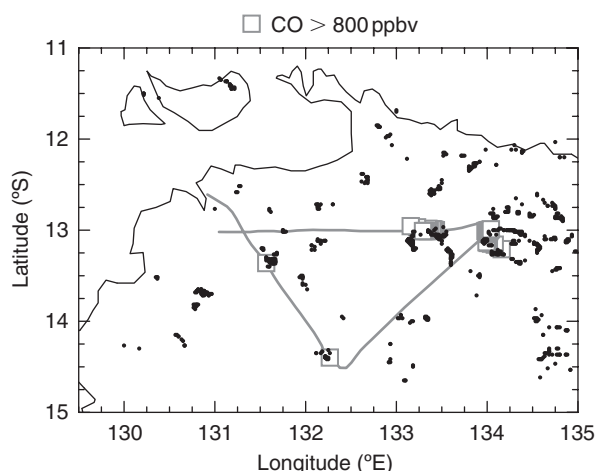
of other tropical regions (Shirai *et al.* 2003). The emission pattern of Australian savanna fires in the local late dry season (September–October) was mainly from the flaming phase, and lower emission ratios were obtained for Australian savanna fires compared to previous measurements for Brazil and Africa. Emission factors of  $\text{CO}$ ,  $\text{CH}_4$  and NMHCs showed significant variation among different vegetation types, while those of  $\text{CO}_2$  varied little.

Estimates of the 2-daily extent of savanna burning from calibrated coarse-resolution AVHRR data, and associated consumption of fine (<6 mm diameter) grass and litter fuels derived from published fuel accumulation relationships, were made for an extensive 230 000 km<sup>2</sup> continental region in the vicinity of Darwin over the time period in which local BIBLE-B flights were undertaken (Russell-Smith *et al.* 2003). Over the 25 day study period, it is estimated that 43 000 km<sup>2</sup> were burnt, consuming 18.8 Mt dry matter (DM) of fine fuels. For

the tropical savannas regions of Australia as a whole, it is estimated from uncalibrated AVHRR data that 417 500 km<sup>2</sup> burned in 1999. Using generalized fine fuel accumulation equations for this broader region, it is estimated that such biomass burning consumed 212 Mt DM in 1999 (Russell-Smith *et al.* 2003); no reliable data are available for coarse fuels.

The annual trace gas emissions of Australian savanna fires were deduced from the emission factors and the above estimates of burned fuel derived for the year 1999. This is the most comprehensive inventory for Australian savanna fires beyond what was reported previously for  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{CH}_4$ , and a few NMHCs based on measurements by Hurst *et al.* (1994a, 1994b). The potential contribution of Australian savanna fire emissions to the global biomass burning emissions was estimated to be ~3% for 1999. While a reliable dataset of emission parameters (i.e. emission ratios and





**Fig. 6.** Locations of biomass burning hot spots (dots) observed by AVHRR on 4 September 1999. The G-II flight track at 1350 LT–1520 LT on 4 September is shown as a shaded line. Locations where CO mixing ratios exceeding 800 ppbv are indicated as shaded squares.

emission factors) was obtained for Australian savanna fires in this study, significant uncertainty still remains in the estimation of total biomass burned. The development of more reliable methods for estimating burned fuels, especially coarse fuels, is required for determining the contribution of Australian biomass burning emissions to global emissions budgeting.

#### *Boundary layer over Australia*

The correlation of  $O_3$  with CO observed in the BL (1–3 km) over the Arnhem Land region (11°–15°S, 129°–136°E) and that over the Timor Sea (14°S, 122°–124°E) was investigated (Takegawa *et al.* 2003b). Clear positive correlations were found in both air masses, indicating the active photochemical production of  $O_3$  due to the increase in  $O_3$  precursors emitted by biomass burning. The linear regression slope of the  $O_3$ –CO correlation was calculated to be 0.12 ppbv/ppbv for the Arnhem Land air and 0.15 ppbv/ppbv for the Timor Sea air. Considering that the air masses observed over the Timor Sea were transported from Arnhem Land by easterly wind within 2–3 days, the difference in the  $O_3$ –CO slope between these two regions is probably due to the subsequent  $O_3$  production downwind of the Arnhem Land region.

At 1–3 km over the South Pacific Ocean, which is upwind of Arnhem Land, a median value for  $O_3$  of 33 ppbv was found. In contrast, median values of  $O_3$  over Arnhem Land and the Timor Sea were 45 ppbv and 49 ppbv, respectively. Median  $O_3$  over the Timor Sea was  $\sim 1.5$  times larger than that over the South Pacific Ocean. The change in the  $O_3$  level is associated with the change in the slope of the  $O_3$ –CO correlation. These results are consistent, indicating that biomass burning in northern Australia significantly impacts the regional  $O_3$  budget in the BL.

Correlation of  $NO_y$  with CO was used to investigate the removal process of  $NO_y$  in biomass burning plumes (Takegawa *et al.* 2003a). By comparing the  $NO_y$ –CO correlation observed over the Timor Sea to that over the Arnhem Land region, it is estimated that  $\sim 60\%$  of the  $NO_y$  molecules emitted from fires were removed within 2–3 days. The decrease in  $NO_y$  can be explained by the dry deposition of  $HNO_3$  on the surface, as schematically shown in Fig. 3. The deposition of  $HNO_3$  originating from biomass burning should be taken into account in assessing total acid deposition on the Arnhem Land region.

On the BIBLE-B flights through the Northern Territory BL, where high concentrations of gaseous products of biomass burning were observed, aerosol measurements were also greatly enhanced. Concentrations of condensation nuclei were greater than  $10^4 \text{ cm}^{-3}$ , exceeding the BIBLE-A median for the tropical BL by more than an order of magnitude. Aerosol surface area, in the range  $100\text{--}600 \mu\text{m}^3 \text{ cm}^{-3}$  for dry air, was similarly an order of magnitude greater than typical BL values in BIBLE-A, and in high humidity (smoke cloud) it was more than an order of magnitude greater again. On the same flights the PSAP measured very high concentrations of black carbon, strongly correlated with estimates from the MASP data of total particle mass and with  $CO_2$ , CO,  $NO_x$ ,  $CH_4$ , and NMHC measurements. From the MASP data the black carbon represents as much as one-quarter of the dry aerosol mass. The correlations with gas species show that black carbon represented 0.5% of total airborne carbon, which was dominated by  $CO_2$  ( $\sim 92\%$ ) and CO ( $\sim 7\%$ ), exceeding the  $CH_4$  fraction and the combined total of all other NMHCs. The ratios for dry air are comparable to published data for measurements close to the source, indicating that, once airborne, the black carbon is mixed efficiently through the BL.

#### *Free troposphere over Australia*

As discussed above, the air impacted by biomass burning that occurred in northern Australia was mostly confined to the BL due to strong subsidence. Free tropospheric air masses sampled over Australia at 10°–28°S were classified according to their origins determined using 14-day back trajectories: Indonesia, Indian Ocean-A, Indian Ocean-B, Africa, and south midlatitude, as schematically shown in Fig. 3. Indonesian air constituted 55% of the all air masses sampled in the upper troposphere. The median values of  $O_3$ , CO,  $NO_y$ , NO, and  $C_2H_6$  values above 8 km in all the types of air except for the south midlatitude air, were similar at  $44 \pm 6$  ppbv,  $80 \pm 2$  ppbv,  $230 \pm 30$  pptv,  $72 \pm 5$  pptv, and  $370 \pm 35$  pptv, respectively, which are much higher than those in the tropical Pacific air (Kondo *et al.* 2002b). The high  $O_3$ , NO,  $NO_y$ , CO, and  $C_2H_6$  values in the south midlatitude air were most pronounced in the upper troposphere. The south midlatitude air should have fewer chances of mixing with clean tropical air than the other air masses.

The relatively high  $C_3H_8/C_2H_6$  and  $n-C_4H_{10}/C_2H_6$  ratios in Indonesian air indicate rapid transport of the continental BL air to the upper troposphere where the lifetimes of the short-lived NMHCs are longer. Lightning is likely to have increased the levels of NO and  $NO_y$  in the Indonesian air considering that the NO levels over Indonesia were strongly impacted by lightning during BIBLE-A as discussed above. Photochemical production in the high NO environment elevated the  $O_3$  values by 15 ppbv during transport from Indonesia to Australia, as discussed above and in Kita *et al.* (2002). The NMHC ratios were lower in the Indian Ocean air that was transported over long distances from continental sources. The NO levels in the Indian Ocean air were probably influenced by lightning over South-east Asia, as for the Indonesian air.

African air in the upper troposphere was strongly influenced by convection and lightning over central Africa, consistent with the high NMHC ratios. However, the median  $O_3$ ,  $NO_y$ , and NO values were not distinctly high as compared with those in the Indian Ocean and Indonesian air. The south midlatitude air masses sampled during BIBLE-A were significantly aged, suggesting that they were not recently impacted by biomass burning over South America and Africa. Efficient accumulation of  $NO_y$ , CO,  $O_3$ , and NMHCs from these sources, without being diluted by the relatively clean tropical air, likely augmented the values of these species in the south midlatitude air.

The enhancements in CO above the background levels in the FT over Australia were mainly due to the emissions from biomass burning, which is the dominant anthropogenic source of CO in the southern hemisphere. The positive and reasonably tight correlations of  $NO_y$  and  $O_3$  with CO indicate that  $NO_y$  and  $O_3$  were impacted by biomass burning. The signature of biomass burning was strong in south midlatitude air, where the CO values sometimes exceeded 100 ppbv. Although lightning is likely an important source of NO in Indonesian air, separating the effects of lightning and biomass burning on the observed NO and  $NO_y$  was difficult for African and south midlatitude air, because biomass burning occurred in areas adjacent to the intensive lightning regions over Africa and South America.

$O_3$  was correlated with  $NO_y$  more tightly than with CO, mainly because of the strong dependence of the net  $O_3$  production rate on NO abundance, irrespective of the NO and  $NO_y$  sources (biomass burning or lightning). The  $NO_x$ -catalysis of  $O_3$  production, relatively small loss of  $NO_y$  in the upper troposphere, together with mixing/dilution probably led to a steady state characterised by the observed  $NO_y$ – $O_3$  correlation.

## Summary

*In situ* aircraft measurements of trace gases and aerosols were made over the western Pacific Ocean, Indonesia, and Australia during the 1998 BIBLE-A and 1999 BIBLE-B

missions. With the exception of occasional enhancements in reactive nitrogen seen over New Guinea associated with lightning activities, the tropical Pacific region at  $120^\circ$ – $150^\circ$ E is distinguished from the rest of the region by the fact that concentrations of  $O_3$ , CO,  $NO_y$ , NO, and NMHCs are smaller. This can be explained in terms of the absence of surface sources over the ocean, lack of stratospheric intrusion, rapid loss of trace species in the tropics, and preclusion of midlatitude air and air from the west by the presence of active convection throughout the troposphere.

During BIBLE-A, concentrations of NO and  $NO_y$  in the upper troposphere over Indonesia were enhanced primarily by lightning. In some circumstances, increases in reactive nitrogen were accompanied by enhancements in CO and NMHCs. This indicates that these air masses were influenced by biomass burning and fossil fuel combustion in Indonesia in the BL and transported to the FT by cumulus convection. Enhanced concentrations of the  $O_3$  precursors did not lead to significant increases in  $O_3$  in the vicinity of the Indonesian Islands because of the limited time for photochemical  $O_3$  production.  $O_3$  was produced at a rate of  $\sim 2$  ppbv/day in air masses with moderately high  $O_3$  precursors during transport from the Indonesian region to the Indian Ocean.

The effect from biomass burning on trace gases and aerosols that occurred in northern Australia was limited to within the BL because of strong regional subsidence over the mission period. Emission rates of NMHCs from savanna burning in Arnhem Land in northern Australia have been estimated from the BIBLE-B data obtained in the BL. The emission factors depend heavily on the combustion efficiencies of the fire and emission characteristics of each compound.

By comparing the  $NO_y$ –CO correlation observed over the Timor Sea to that over the Arnhem Land region, it is estimated that  $\sim 60\%$  of the  $NO_y$  molecules emitted from fires were removed through dry deposition of  $HNO_3$  within 2–3 days. The median value of  $O_3$  at altitudes of 1–3 km over the South Pacific Ocean, which is upwind of Arnhem Land, was found to be 33 ppbv. In contrast, median values of  $O_3$  over the Arnhem Land and Timor Sea were 45 ppbv and 49 ppbv, respectively, indicating that the biomass burning in northern Australia significantly impacts the regional  $O_3$  budget in the BL. On the BIBLE-B flights through the Northern Territory BL that observed high concentrations of gaseous products of biomass burning, aerosol measurements were also greatly enhanced.

In the FT over Australia, elevated concentrations of  $O_3$  and its precursors above those observed over the tropical Pacific air were found in air masses originating further away over Africa, South America, the Indian Ocean, and Indonesia during BIBLE-A and B. The median mixing ratios of  $O_3$  and its precursors were highest in air masses that stayed mainly at southern midlatitudes for 2 weeks without being diluted by the relatively clean tropical air.

## Overall conclusion

Over South-east Asia, the O<sub>3</sub> precursors produced by biomass burning and lightning were rapidly transported from the BL to the FT due to convection, leading to O<sub>3</sub> formation in the upper troposphere. By contrast, over northern Australia, the majority of the trace species emitted by biomass burning remained in the BL due to strong regional subsidence, leading to O<sub>3</sub> formation in the BL. These trace species either deposited over northern Australia or were transported westward within the BL. Elevated concentrations of the trace species in the FT over Australia were due to long range transport from other regions.

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