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Abstract. \textit{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\textsubscript{3}) and its precursors [CO, reactive nitrogen (NO\textsubscript{x}), non-methane hydrocarbons (NMHCs)] were measured in these campaigns to identify the sources of NO\textsubscript{x} and to estimate the effects of biomass burning and lightning on photochemical production of O\textsubscript{3}. Over Indonesia, \textit{in-situ} production of NO\textsubscript{x} 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\textsubscript{3}, 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\textsubscript{3} 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\textsubscript{3} levels in the biomass burning plumes were enhanced due to photochemical production.
Introduction

Biomass burning is an important source of trace gases [such as NO\textsubscript{x}, NO\textsubscript{y}, CO\textsubscript{2}, CO, CH\textsubscript{4}, non-methane hydrocarbons (NMHCs) and CH\textsubscript{3}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 NO\textsubscript{x} and 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 O\textsubscript{3} precursors on O\textsubscript{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 (15.0°N, 146.0°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 ozoneonde 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 ozoneonde 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 ozoneonde measurements were carried out in Darwin, Watukosek and Kototabang. Eighteen ozoneonde 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. NO, NO\textsubscript{2}, and NO\textsubscript{y} were measured using a chemiluminescence technique (Kondo et al. 1997, 2003; Ikeda et al. 2000; Koike et al. 2000). O\textsubscript{3} was measured by using a dual-beam UV-absorption O\textsubscript{3} photometer (Kita et al. 2002). The CO concentration was measured by using an automated gas-chromatograph (GC) system with a reduction gas detector (RGD) (Kita et al. 2002). 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). C\textsubscript{2}–C\textsubscript{8} NMHCs, halocarbons, and alkyl nitrates were quantified (Blake et al. 1996). CO\textsubscript{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°C for 1011A and ~95°C for CR-2. H\textsubscript{2}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 µm 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 NO\textsubscript{2} and O\textsubscript{3} photolysis, J(NO\textsubscript{2}) and J(O\textsubscript{1}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 NO, O\textsubscript{3}, H\textsubscript{2}O, CO, CH\textsubscript{4}, and NMHCs along the flight tracks to calculate the
concentrations of the $O_3$ radicals, the $HO_x$ 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 $O_3$ 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 NO\textsubscript{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 O\textsubscript{3}, CO, NO\textsubscript{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 O\textsubscript{3}, CO, NO\textsubscript{y}, and NO mixing ratios were \( \sim 5–15 \) ppbv, \( 60–75 \) ppbv, \( 20–70 \) pptv, and \( 3–5 \) pptv, respectively, with the smallest O\textsubscript{3} and NO\textsubscript{y} values observed near the surface. The O\textsubscript{3}, CO, NO\textsubscript{y}, and NO mixing ratios at 8–13 km are shown in Fig. 2. Over the tropical Pacific, the NO and NO\textsubscript{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
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 NOx and NOy 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, NOx and NOy 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 NOx (dNOx) and NOy (dNOy) 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 dNOy/dCO and dNOx/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 NOx 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 O3 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 O3, NO, NOy, CO, C2H6, C2H4, C2H2, and C3H8 in the upper troposphere, including short-lived C2H4, 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 O3 precursor gases from the surface to the upper troposphere in recent convection. Convection also caused NO production by lightning as discussed above.

When the dNOy/dCO and dNOx/dCO ratios for the upper troposphere are compared to the values found in the lower troposphere, the dNOx and dNOy values are higher by 40–60 pptv (80% of NOx) and 70–100 pptv (50% of NOy). This difference is attributed to in-situ production of NOx and NOy 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 NOx values, while convection over the ocean generally lowers the NOx concentration. As a consequence of the high NOx concentrations, net photochemical O3 production rates of 1–2 ppbv/day were found in both air masses in the upper troposphere. These results suggest that NOx production by lightning over the Indonesian islands had a significant impact on the O3 budget over the western Pacific.

Trajectory analyses show that air masses transported from western Indonesia were observed south of 9°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 O3 precursors except for C2H4 were similar to those in western Indonesia. However, the median O3 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 O3 proceeded in the air masses transported from Indonesia, and the average net O3 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 O3 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 O3 mixing ratio (40 ppbv) in the Indonesian outflow air was also significantly higher than the average O3 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...
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$_2$H$_4$) and propene (C$_3$H$_6$) 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$_x$ 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...
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 CO, CH₄ and NMHCs showed significant variation among different vegetation types, while those of CO₂ 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 km² 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 km² 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 km² 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 CO₂, CO, CH₄, 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
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 O₃ 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 O₃ due to the increase in O₃ precursors emitted by biomass burning. The linear regression slope of the O₃–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 O₃–CO slope between these two regions is probably due to the subsequent O₃ 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 O₃ of 33 ppbv was found. In contrast, median values of O₃ over Arnhem Land and the Timor Sea were 45 ppbv and 49 ppbv, respectively. Median O₃ over the Timor Sea was ~1.5 times larger than that over the South Pacific Ocean. The change in the O₃ level is associated with the change in the slope of the O₃–CO correlation. These results are consistent, indicating that biomass burning in northern Australia significantly impacts the regional O₃ budget in 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 O₃, CO, NOₓ, NO, and C₂H₆ values above 8 km in all the types of air except for the south midlatitude air, were similar at ~44 ± 6 ppbv, 80 ± 2 ppbv, 230 ± 30 pptv, 72 ± 5 pptv, and 370 ± 35 pptv, respectively, which are much higher than those in the tropical Pacific air (Kondo et al. 2002b). The high O₃, NO, NOₓ, CO, and C₂H₆ 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.

Correlation of NOₓ with CO was used to investigate the removal process of NOₓ in biomass burning plumes (Takegawa et al. 2003a). By comparing the NOₓ–CO correlation observed over the Timor Sea to that over the Arnhem Land region, it is estimated that ~60% of the NOₓ molecules emitted from fires were removed within 2–3 days. The decrease in NOₓ can be explained by the dry deposition of HNO₃ on the surface, as schematically shown in Fig. 3. The deposition of HNO₃ 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 10⁴ cm⁻³, exceeding the BIBLE-A median for the tropical BL by more than an order of magnitude. Aerosol surface area, in the range 100–600 µm² 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 CO₂, CO, NOₓ, CH₄, 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 CO₂ (~92%) and CO (~7%), exceeding the CH₄ 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.
The relatively high C\textsubscript{3}H\textsubscript{8}/C\textsubscript{2}H\textsubscript{6} and n-C\textsubscript{4}H\textsubscript{10}/C\textsubscript{2}H\textsubscript{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\textsubscript{x} 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\textsubscript{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\textsubscript{3}, NO\textsubscript{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\textsubscript{y}, CO, O\textsubscript{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\textsubscript{y} and O\textsubscript{3} with CO indicate that NO\textsubscript{y} and O\textsubscript{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\textsubscript{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\textsubscript{3} was correlated with NO\textsubscript{y} more tightly than with CO, mainly because of the strong dependence of the net O\textsubscript{3} production rate on NO abundance, irrespective of the NO and NO\textsubscript{y} sources (biomass burning or lightning). The NO\textsubscript{y}-catalysis of O\textsubscript{3} production, relatively small loss of NO\textsubscript{y} in the upper troposphere, together with mixing/dilution probably led to a steady state characterised by the observed NO\textsubscript{y}–O\textsubscript{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\textdegree–150\textdegree E is distinguished from the rest of the region by the fact that concentrations of O\textsubscript{3}, CO, NO\textsubscript{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 traces species in the tropics, and preclusion of mid-latitude air and air from the west by the presence of active convection throughout the troposphere.

During BIBLE-A, concentrations of NO and NO\textsubscript{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\textsubscript{3} precursors did not lead to significant increases in O\textsubscript{3} in the vicinity of the Indonesian Islands because of the limited time for photochemical O\textsubscript{3} production. O\textsubscript{3} was produced at a rate of \sim 2 ppbv/day in air masses with moderately high O\textsubscript{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\textsubscript{y}–CO correlation observed over the Timor Sea to that over the Arnhem Land region, it is estimated that \sim 60% of the NO\textsubscript{y} molecules emitted from fires were removed through dry deposition of HNO\textsubscript{3} within 2–3 days. The median value of O\textsubscript{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\textsubscript{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\textsubscript{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\textsubscript{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\textsubscript{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 O3 precursors produced by biomass burning and lightning were rapidly transported from the BL to the FT due to convection, leading to O3 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 O3 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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References


