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Reactive nitrogen over the tropical western Pacific: Influence from lightning and biomass burning during BIBLE A

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[1] The Biomass Burning and Lightning Experiment phase A (BIBLE A) aircraft campaign was carried out over the tropical western Pacific in September and October 1998. During this period, biomass burning activity in Indonesia was quite weak. Mixing ratios of NO_x and NO_y in air masses that had crossed over the Indonesian islands within 3 days prior to the measurement (Indonesian air masses) were systematically higher than those in air masses originating from the central Pacific (tropical air masses). Sixty percent of the Indonesian air masses at 9–13 km (upper troposphere, UT) originated from the central Pacific. The differences in NO_y mixing ratio between these two types of air masses were likely due to processes that occurred while air masses were over the Islands. Evidence presented in this paper suggests convection carries material from the surface, and NO is produced from lightning. At altitudes below 3 km (lower troposphere, LT), typical gradient of NO_x and NO_y to CO (dNO_y/dCO and dNO_x/dCO) was smaller than that in the biomass burning plumes and in urban areas, suggesting that neither source has a dominant influence. When the CO-NO_x and CO-NO_y relationships in the UT are compared to the reference relationships chosen for the LT, the NO_x and NO_y values are higher by 40–60 pptv (80% of NO_x) and 70–100 pptv (50% of NO_y). This difference is attributed to in situ production of NO by lightning. Analyses using air mass trajectories and geostationary meteorological satellite (GMS) derived cloud height data show that convection over land, which could be accompanied by lightning activity, increases the NO_x values, while convection over the ocean generally lowers the NO_x level. These processes are found to have a significant impact on the O_3 production rate over the tropical western Pacific. *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; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); *KEYWORDS:* reactive nitrogen, ozone, biomass burning, lightning, western Pacific

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1. Introduction

[2] The oxidizing power of the tropical troposphere is considered to be a critical parameter in the budgets of various long–lived gases. Both UV radiation intensity and humidity are high in the tropics; thus ozone (O_3) plays a key role in controlling the OH level and therefore the oxidizing power of the tropics. Recent three-dimensional global model calculations showed that photochemical production and loss in the troposphere were the dominant source and sink of O_3 in the tropics [*Wang et al.*, 1998]. Model calculations also showed that lightning NO production contributes more than 70% to the zonal mean NO_x (= NO + NO_2) source in the tropical free troposphere, while biomass burning, fossil fuel combustion, and soil emissions contribute 10 to 30% [*Lamarque et al.*, 1996; *Levy et al.*, 1999]. Convection in the tropical western Pacific is significant, injecting NO_x into the middle and upper troposphere from surface sources, and

causing lightning NO production. Consequently, O_3 production in that region can be particularly important for the O_3 budget in the tropics [*Jacob et al.*, 1996].

[3] During the Pacific Exploratory Mission phase A (PEM-West A) experiment carried out over the western Pacific in September and October 1991, a net decreasing tendency was found in the column-integrated O_3 value in the tropical western Pacific [*Davis et al.*, 1996]. A net decrease tendency of O_3 was also found over the South Pacific during the PEM-Tropics A experiment conducted in September and October 1996 [*Schultz et al.*, 1999]. In the latter case, NO_x concentrations in that region were likely maintained by the long range transport of reactive nitrogen, which were likely produced by biomass burning and lightning over the tropical continent. During the PEM-West B experiment carried out in February and March 1994, a net decreasing tendency was also found in low-NO_x regimes, while a small net increasing tendency was found in the high-NO_x regimes [*Crawford et al.*, 1997]. *Kawakami et al.* [1997] showed that the level of NO_x in the tropics was greatly affected by convective activity because convection over the ocean generally lowers NO_x levels, while convection over the land, which causes lightning, increases NO_x levels. However, observations to evaluate individual NO_x sources are still limited and thus the budget of O_3 in the tropics remains poorly understood.

[4] The Biomass Burning and Lightning Experiment phase A (BIBLE A) was carried out over the tropical western Pacific between September 21 and October 10 of 1998, using a Gulfstream II (G-II) aircraft. NO and NO_y (= $NO + NO_2 + NO_3 + HNO_2 + HNO_3 + HNO_4 + 2N_2O_5$ + PAN + other organic nitrates + aerosol nitrates) were measured together with O_3, CO, CO_2, H_2O, CH_4, various nonmethane hydrocarbons (NMHCs), selected halocarbons, condensation nuclei (CN), aerosol size distribution, and actinic flux for NO_2 photolysis ($J(NO_2)$) and O_3 photolysis to produce $O(^1D)$ ($J(O^1D)$). Measurements were made during five intensive flights from Bandung, Indonesia and a series of ferry flights between Nagoya, Japan and Bandung. During this experiment, relatively large amounts of data were obtained in the upper troposphere at altitudes between 9 and 13 km. In this study, a possible contribution to the reactive nitrogen budget in the upper troposphere from biomass burning, fossil fuel combustion, and lightning is examined.

2. Measurements

[5] Both NO and NO_y were measured using an NO-O_3 chemiluminescence technique. NO_y compounds were catalytically converted to NO on the surface of a heated gold tube (300°C) with the addition of CO. The NO and NO_y instrument used for BIBLE A was very similar to that used during NASA's Subsonic Assessment (SASS) Ozone and NOx Experiment (SONEX) conducted in 1997 [*Koike et al.*, 2000; *Kondo et al.*, 1997a]. The main difference was the way in which the gold tube NO_y converter was mounted. During BIBLE A, the NO_y converter unit was placed outside the aircraft cabin so as to minimize the length of the inlet Teflon tube for air sampling upstream of the gold tube converter. The inlet tube, which was heated to 40°C, faced rearward, discriminating against particles of diameter larger than about 1 μm. Data were recorded every 1-s, however 10-s averaged data were used in this study. The precision of the 10-s NO and NOy measurements at 10 km estimated from the photon count fluctuations (1 σ) was 2 and 3 parts per trillion by volume (pptv) for NO and NO_y values of 100 and 200 pptv, respectively. The absolute accuracy was estimated to be 8 and 17% for these NO and NO_y values.

[6] Carbon monoxide (CO) concentrations were measured using an automated gas chromatograph (GC) system with a reduction gas detector developed at the University of Tokyo [*Kita et al.*, 2002]. The precision of the 10-s data (1 σ) and absolute accuracy were 3 parts per billion by volume (ppbv) and 5%, respectively. Measurements of O_3 were made using a dual beam UV absorption photometer, which was also developed at the University of Tokyo [*Kita et al.*, 2002]. The precision of the 10-s data (1 σ) and absolute accuracy were 0.9 ppbv and 5%, respectively. The concentrations of CH_4, various NMHCs, selected halocarbons, and C_1-C_4 alkyl nitrates were measured by collecting whole air samples and analyzing them using gas chromatography with flame ionization, electron capture, and mass spectrometer detection. Sample filling time ranged from 0.5 to 3 minutes and samples were collected every 2–5 minutes on average. The detection limit of the whole air measurements is summarized by *Simpson et al.* [2000]. The values of $J(NO_2)$ and $J(O^1D)$ were measured using two sets of the filter radiometers; one set looked upward and the other looked downward from the aircraft (Meteorologie Consult, Glashutte, Germany) [*Kita et al.*, 2002]. The photostationary state NO_2 mixing ratios were calculated using a box model developed at Atmospheric and Environmental Research Inc. [*Ko et al.*, 2002] from the observed values of atmospheric temperature, pressure, NO, O_3, CO, CH_4, H_2O, NMHCs, and $J(NO_2)$ for periods when the solar zenith angles were lower than 80°.

3. Meteorological Conditions and Air Mass Classification

[7] For air masses encountered by the G-II aircraft, 10-day kinematic back trajectories were calculated, using the method developed by *Matsuzono et al.* [1998] and the European Centre for Medium-Range Weather Forecasts (ECMWF) data on a 2.5° × 2.5° latitude-longitude grid. For the present analyses, we classified air masses as Indonesian when they were sampled after having crossed any Indonesian islands within the previous three days. Indonesian air masses were sampled over and around the Indonesian islands at latitudes between the equator and 15°S (Figure 1).

[8] In addition to these air masses, we selected two other typical air masses, which were sampled between 20°N and 22°S. When air masses had originated from the central Pacific and had been transported by the dominant easterly winds without crossing any continent or islands for ten days, they were classified as tropical air masses. When air masses had passed over Indonesian islands five to ten days prior to the measurement, they were classified as aged Indonesian air masses. Tropical air masses were sampled at latitudes between 20°N and 5°N (Figure 1) and had likely been strongly affected by deep convection over the ocean [*Kondo*

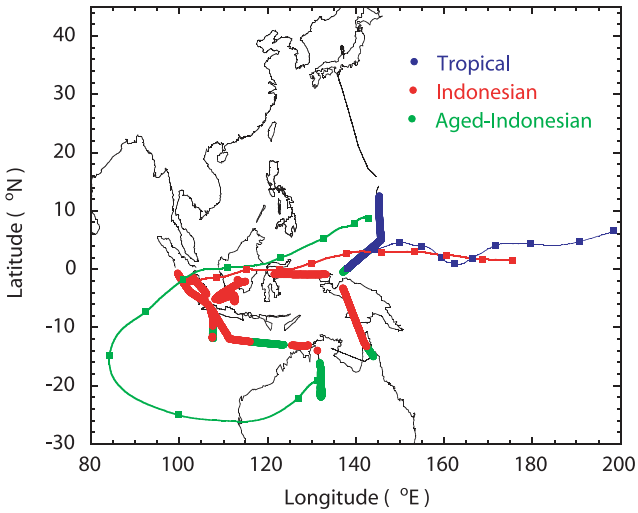

Figure 1. Locations where measurements were made during BIBLE A in the UT (9–13 km, black thin solid lines). Locations where Indonesian, tropical, and aged Indonesian air masses were sampled are shown by red, blue, and green circles. A typical back trajectory of each air mass (10 days) is also shown. Symbols on the trajectories denote the location of air masses every 24 hours prior to the measurement.

et al., 2002]. These air masses were sampled in an area where the Intertropical Convergence Zone (ITCZ) and South Pacific Convergence Zone (SPCZ) merged. Outgoing Longwave Radiation (OLR) values over the tropical western Pacific in September 1998 and their deviation from the average values in the years between 1979 and 1995 obtained by National Oceanic and Atmospheric Administration (NOAA) satellites are shown in Figures 2a and 2b. Very high convective activity was seen in the area west of Saipan (15°N, 146°E), where the maximum of the monthly mean velocity potential appeared at 200 hPa (not shown), suggesting a large number of air masses were transported by deep convection and diverged in the upper troposphere. The higher-than-average convective activity resulted from the September 1998 La Niña condition. Wind direction at lower altitude suggests that clean air masses transported from the central Pacific generally were pumped up to higher altitudes.

[9] Aged Indonesian air masses were sampled at various locations between 5°N and 22°S (Figure 1). Most of these air masses circulated around a high pressure system over northern Australia, which persistently appeared during BIBLE A. Within air masses sampled between 20°N and 22°S during BIBLE A, 48%, 18%, and 18% of air masses were classified into Indonesian, tropical, and aged Indonesian air masses, respectively.

[10] The geographic origins of Indonesian air masses, prior to passing over the Indonesian islands, were examined. During the BIBLE A period, easterly or north-easterly winds generally dominated both at 200 and 850 hPa over and east of the Indonesian islands bringing clean maritime air masses to the Indonesian region. At altitudes between 9 and 13 km (defined as upper troposphere, UT), 62% of the Indonesian air masses had been transported by easterly winds from the central Pacific, the location where the tropical air masses had originated. Consequently, differences in the chemical characteristics between Indonesian and tropical air masses in the UT are considered to be generally due to entrainment over the Indonesian islands. At altitudes of 0–3 km (lower troposphere, LT) and 3–9 km (middle troposphere, MT), 43% and 35% of Indonesian air masses originated from the central Pacific, and 28% and 44% of the air masses in the LT and MT had stagnated within the Indonesian region. These results also allow us to use tropical air masses as a reference to assess inputs from the Indonesian islands.

4. Results and Discussion

4.1. Vertical Profiles

[11] Vertical profiles of median values for CO, NO_y, NO_x, and NO_x/NO_y are shown for the Indonesian, tropical, and aged Indonesian air masses (Figures 3a–3d). For the statistical analysis, 300–2100 10-s data were used in each of the altitude bins in the case of NO_x in Indonesian air masses. We will first examine the profiles of the individual species to obtain information on the sources. CO values in Indonesian air masses (75–90 ppbv) were systematically higher than those in tropical air masses (60–75 ppbv). As described above, the differences were likely due to the influence of emissions over the Indonesian islands. The CO values in Indonesian air masses had a "C-shaped" vertical profile. The value at 2 km was higher (90 ppbv) than at higher altitudes, presumably due to surface sources, such as biomass burning and urban pollution. Fossil fuel combustion was considered to be the predominant source of urban pollution. Relatively higher CO values in the UT, compared with those in tropical air masses, indicates a clear surface influence. This is in agreement with the conclusion from the detailed examination of the measured NMHC and their correlations with CO by *Kita et al.* [2002].

[12] NO_y mixing ratios were systematically higher at all altitudes in Indonesian air masses than those in tropical air masses, 80–220 pptv and 20–60 pptv. In tropical air masses, the NO_y values were low, likely because of very small source strength, frequent removal by dry and wet deposition, and frequent vertical mixing. In Indonesian air masses, the NO_y values showed a "C-shaped" vertical profile, similar to that of CO; the values below 3 km and above 9 km were slightly enhanced over those between 3 and 9 km. However, in contrast to the CO case, NO production by lightning activity over the Indonesian region could be a potential source of NO_y, in addition to the input from surface sources, i.e. biomass burning and urban pollution.

[13] NO_x mixing ratios in Indonesian air masses generally increased with altitude and reached 80 pptv at 12 km. These values were much higher than those in tropical air masses in which NO_x values were generally 5–20 pptv. During the PEM-West B experiment, very different levels of NO_x were observed over the tropical western Pacific, and they were attributed to lightning NO production and convective transport of clean marine boundary air [*Crawford et al.*, 1997; *Kawakami et al.*, 1997]. The median values of these two air masses are also shown in Figure 3c. The NO_x values in BIBLE A tropical air masses were similar to those in the PEM-West B low-NO_x regime. As described above, the

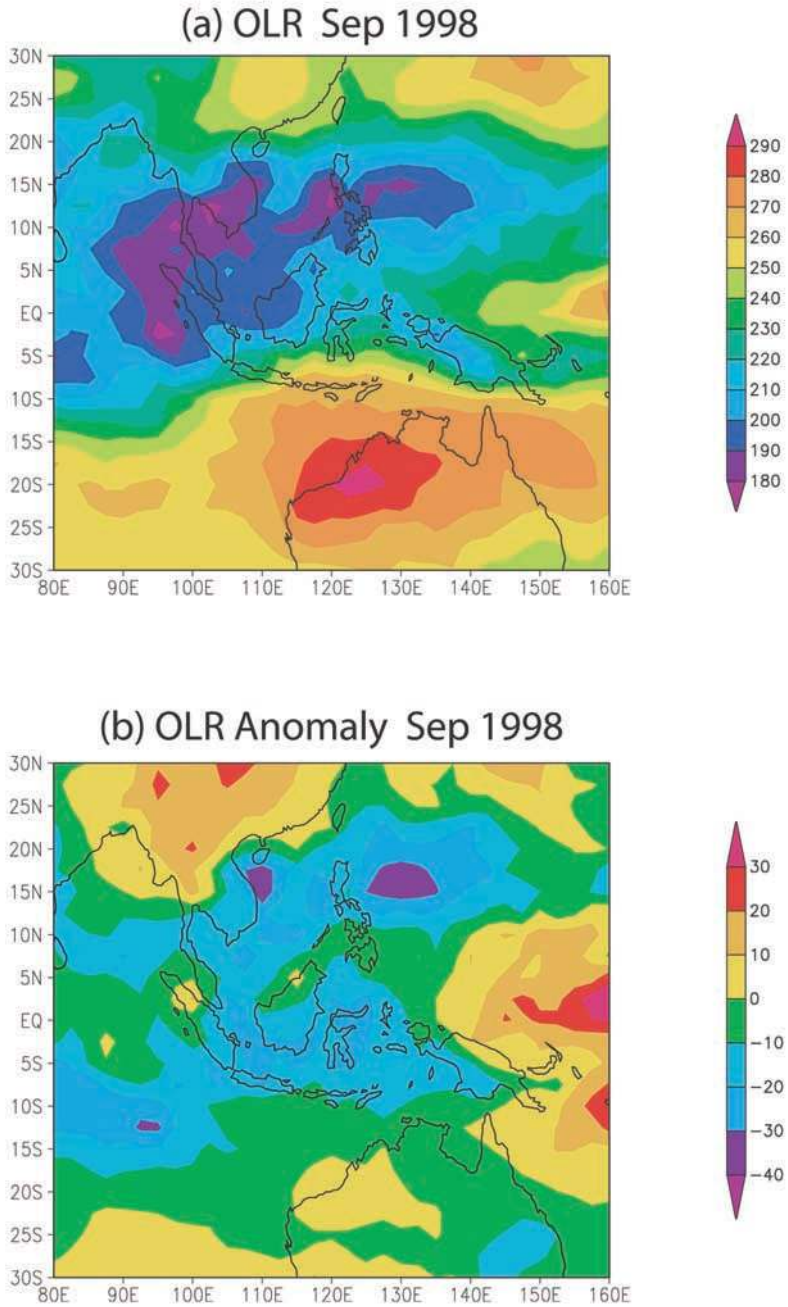

Figure 2. (a) Outgoing longwave radiation (OLR) field in September 1998 (temperature in K). (b) Deviation of the OLR value in September 1998 from the average values in years between 1979 and 1995.

BIBLE A tropical air masses were also likely to have been strongly affected by outflow from convection over the ocean. Although the NO_x values in BIBLE A Indonesian air masses were about a factor of two smaller than in the PEM-West B high-NO_x regime, they show a similar tendency.

[14] Measurements of reactive nitrogen were also made over the South Pacific in September–October 1996 during PEM-Tropics A. In Figure 3c, median values of NO (instead of NO_x) in air masses sampled at north and south of the SPCZ at latitudes between 0° and 22°S [*Gregory et al.*, 1999] are also shown for comparison. At altitudes above 5 km, the NO values in SPCZ south air are much larger than the BIBLE A NO_x values. The chemical characteristics indicate that these air masses had been affected by biomass burning, consistent with air mass trajectories which passed over Australia, Indonesia, and/or Africa where biomass burning was occurring [*Gregory et al*, 1999]. On the other hand, SPCZ north air had been transported by easterly wind and confined for many days to an equatorial Pacific. However the NO level in these air masses at 9 km is comparable with NO_x values in BIBLE A Indonesian air masses.

[15] The NO_x/NO_y ratios were generally similar in all three air masses, although they were slightly higher in

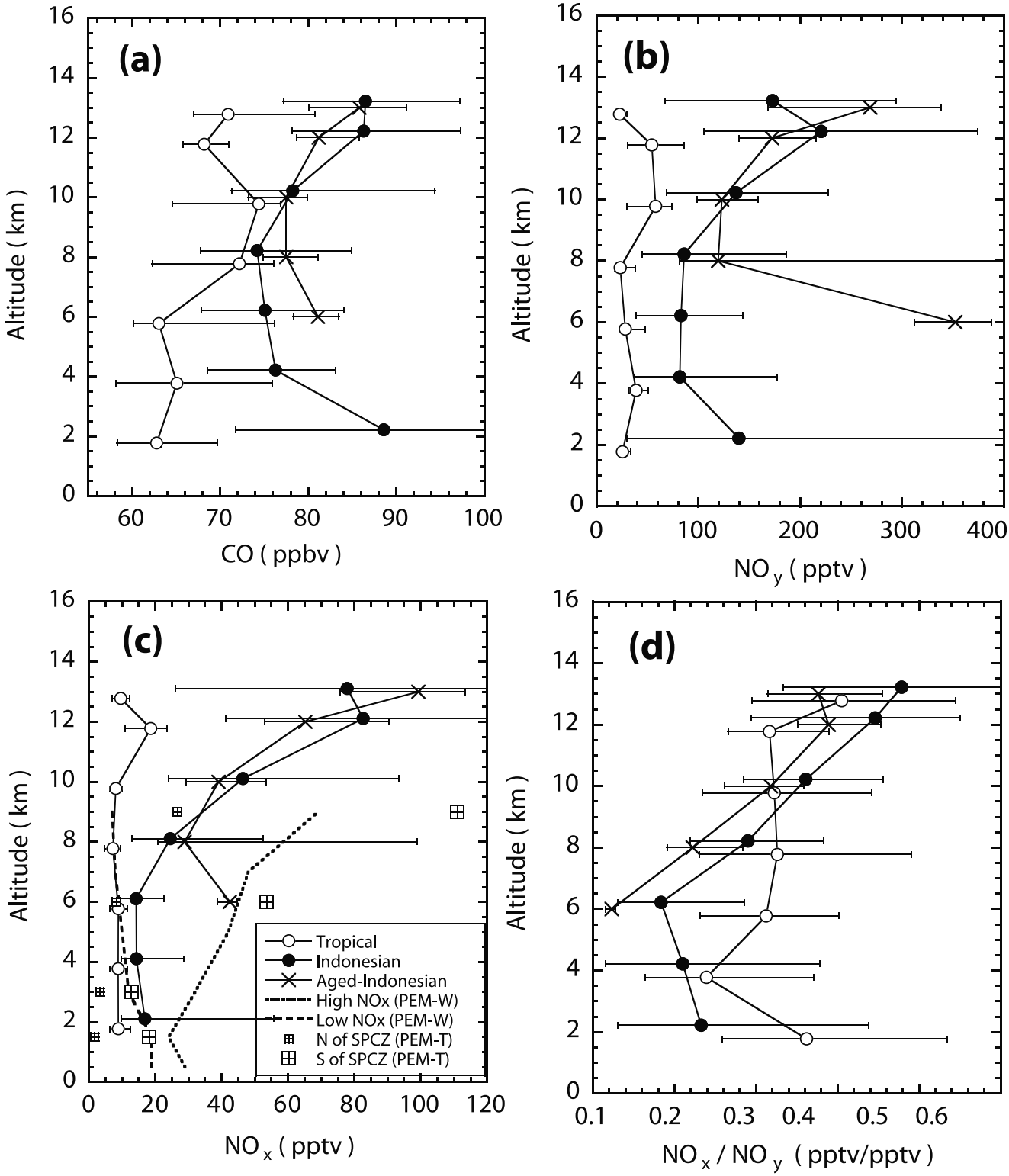

Figure 3. Vertical profile of the median values of the CO, NO_y, NO_x, and NOx/NO_y ratio in Indonesian, tropical, and aged Indonesian air masses. The bars indicate the central 67% range. In Figure 3c, values obtained during the PEM-West B (PEM-W) and PEM-Tropics A (PEM-T, values of NO instead of NO_x) experiments are also shown.

Indonesian air masses in the UT. The photochemical lifetime of NO_x was estimated using three-dimensional model calculations [*Bey et al.*, 2001]. At an altitude of 12 km at the equator, it was 2.4 days in September. It was systematically shorter than that in midlatitudes primarily because of stronger solar radiation. Consequently, the similarity in the NO_x/NO_y ratios within the three air masses in the UT was likely because NO_x and its reservoirs, such as HNO_3, were

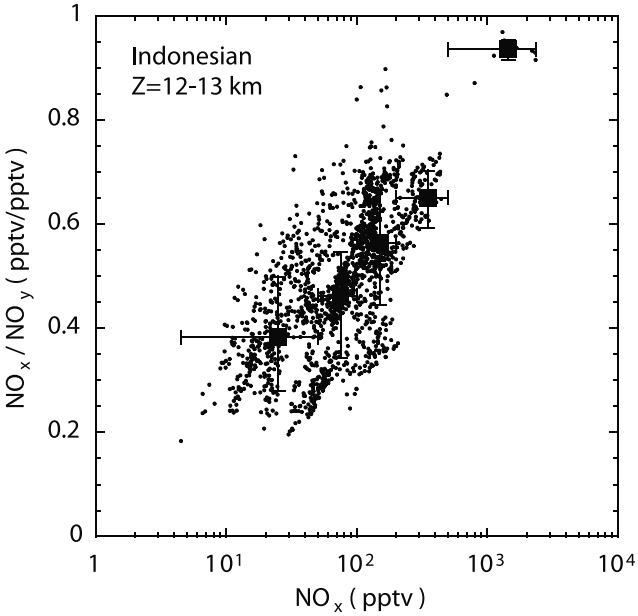

Figure 4. Scatterplot between NO_x mixing ratio and NO_x/NO_y ratio at altitudes between 12 and 13 km in Indonesian air masses. Median value of NO_x/NO_y ratio in each NO_x range is also shown.

generally in photochemical equilibrium. Slightly higher ratios in Indonesian air masses were considered to be due to recent input of NO by lightning production, as described below.

[16] In Figure 4, the relationship between the NO_x/NO_y ratio and NO_x mixing ratio is shown for Indonesian air masses. In Figure 4, only data obtained at altitudes between 12 and 13 km were used, to minimize the altitude dependence of the NO_x/NO_y ratio caused by the change in the lifetime of NO_x. As seen in Figure 4, NO_x/NO_y ratios and NO_x values have a positive correlation. When NO_x was lower than 50 pptv, the median value of the NO_x/NO_y ratio was 0.38, which was close to the value in the tropical air masses (Figure 3d) and was considered to be close to the photochemical equilibrium ratio. On the other hand, a range of the median values of the NO_x/NO_y ratio was 0.56–0.94, when NO_x was higher than 100 pptv. As described below, air masses in which a degree of influence from biomass burning was largest were sampled at altitudes below 3 km. In addition, air masses influenced by urban sources were sampled during take-off and landing at the Bandung airport. In these air masses (CO > 100 ppbv), NO_x/NO_y ratios were only 0.15 and 0.20, respectively. Consequently, transport of NO_x in these air masses could not account for the observed high NO_x/NO_y ratios in the UT especially in air masses in which NO_x values were high. These results indicated that the observed high NO_x values in the Indonesian air masses in the UT were largely due to recent lightning NO production. Quantitative estimation of the contribution is given below.

4.2. Contributions From Surface Sources and Lightning NO Production

4.2.1. Approach

[17] Air masses classified as Indonesian are used to infer the contributions from surface sources of reactive nitrogen and lightning NO production over the Indonesian islands. In the LT, Indonesian air masses are expected to have higher NO_x and NO_y concentrations than those in tropical air masses because of influences from biomass burning and other surface emissions from land area. CO is a good indicator of influence from biomass burning and fossil fuel combustion. We used the Indonesian air masses in the LT to establish a reference correlation between CO and other species (CH_3Cl, C_2H_2, NO_x, and NO_y). If this air mass is transported to the UT with no other modification, the measured CO and other species in the UT should follow the same correlation. Mixing of transported air masses in the UT with ambient air, which is tropical air masses, would not change the correlation greatly because the lowest levels of various species in Indonesian LT air masses were generally similar to the average levels in tropical UT air masses, as shown below. Our analyses below will show that the correlations for CH_3Cl and C_2H_2 suggest the similarities in air mass characteristics between Indonesian UT air masses and LT air masses sampled in nonurban areas. In contrast, the measured NO_x and NO_y are larger than can be expected from the correlation established in the LT. We infer that the enhancements resulted from lightning NO production.

4.2.2. Reference Relationships in the LT

[18] Biomass burning activity in the Indonesian islands was quite weak in 1998 during the BIBLE A campaign as compared to other years, in contrast to the situation in September and October 1997 when very severe biomass burning took place under El Niño conditions [e.g., *Tsutsumi et al.*, 1999; *Fujiwara et al.*, 1999]. The weak biomass burning in 1998 was due primarily to higher precipitation rates than those in normal years, owing to La Niña, and also because a considerable amount of biomass had already been burned in 1997 and early 1998. Locations of high-temperature spots (hot spots), indicating biomass burning fires, detected by the Along Track Scanning Radiometer (ATSR) satellite sensor in September and October 1998 are shown in Figure 5. The number of the hot spots detected in the area

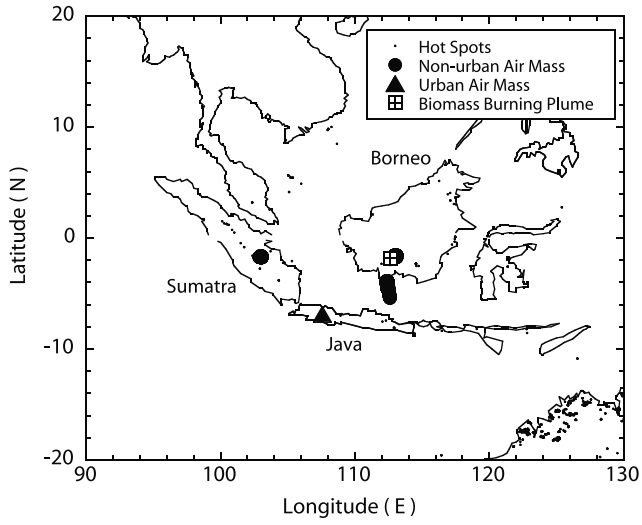

Figure 5. Location where Indonesian air masses in the LT (Z < 3 km) were sampled. Hot spots detected by the ATSR satellite sensor in September and October 1998 are also shown.

within 10°N–10°S and 95°E–125°E was 101 during that period. It was a factor of about 100 smaller than that in September and October 1997.

[19] During BIBLE A, although we tried to sample air masses strongly influenced by biomass burning using the real-time hot spot information, only one biomass burning plume was sampled, and the sampling lasted for only 15-s. Within this plume, the CO mixing ratio was as high as 300 ppbv. The gradient of NO_x and NO_y increase relative to the increase in CO (dNO_x/dCO and dNO_y/dCO) above the levels of these species in the surrounding air (CO = 110 ppbv, NO_x = 25 pptv, NO_y = 180 pptv) were calculated to be 11.5 and 24.0 pptv/ppbv, respectively. These values were close to the average values (11.8 and 19.2) derived from a much larger data set obtained in the northern part of Australia during the BIBLE B campaign carried out in October 1999. The dNO_x/dCO value was also within a range of values (although on the lower side) found in earlier studies obtained over Indonesia, Brazil, North America, and Africa (8 to 110 pptv/ppbv) [*Sawa et al.*, 1999, and references therein].

[20] During BIBLE A, Indonesian air masses at altitudes between 0.5 and 3 km were sampled at three locations in addition to those sampled during take-off and landing at the Bandung airport (Figure 5). In each case, measurements were made at a constant altitude level for 10 minutes. In the middle of Sumatra Island and south of Borneo Island were locations where biomass burning was relatively intense during the BIBLE A period, as seen in Figure 5. In the third case, air masses were sampled over the ocean between Java and Borneo Islands. Thus the air masses we sampled at these three locations were considered to be representative of those in which the degree of influence from biomass burning was highest among air masses over Indonesia during the BIBLE A period (from now on, we will call them nonurban air masses). Typical relationships between CO and NO_x and between CO and NO_y in these air masses in the LT were calculated using data obtained at these three locations at altitudes below 3 km (Figure 6). Although the data obtained in the biomass burning plume were excluded, those obtained just outside the plume were used in this calculation. As a result, dNO_x/dCO and dNO_y/dCO values were calculated to be 0.74 (r^2 = 0.78) and 4.3 (r^2 = 0.57) pptv/ppbv, which were lower than the values in the biomass burning plume by factors of 15 and 6. In this calculation, the CO value at the intercept for NO_x = 0 or NO_y = 0 was determined to be 63–65 ppbv, which was close to the median CO value in tropical air masses of 63 ppbv, where NO_x = 10 and NO_y = 25 pptv (Figures 3a–3c), consistent with the trajectory analysis described above.

[21] In October 1997, when biomass burning activity was quite high around Indonesia, NO_x values of 1–4 ppbv were observed at 1–4 km over Borneo Island [*Tsutsumi et al.*, 1999]. In contrast, the NO_x and NO_y mixing ratios observed during BIBLE A in the LT were only 10–60 pptv and 30–600 pptv, respectively, even just outside the plume. These results indicate that in general, air masses in the LT were not strongly affected by biomass burning during BIBLE A. Assuming that convective transport occurred at various locations randomly, air masses that were transported to the UT should have chemical characteristics typical of those observed in air masses sampled outside the biomass burning plume, although the impact is expected to be greater when a plume is transported. This is because the relative frequency of the vertical transport of a biomass burning plume is, to a first approximation, proportional to the biomass burning area, which was quite small in 1998. In other words, to estimate the influence from the convective transport of LT air masses to the UT, the typical relationships of $CO\text{-}NO_x$ and $CO\text{-}NO_y$ in nonurban air masses, rather than the relationships in the biomass burning plume, should be used.

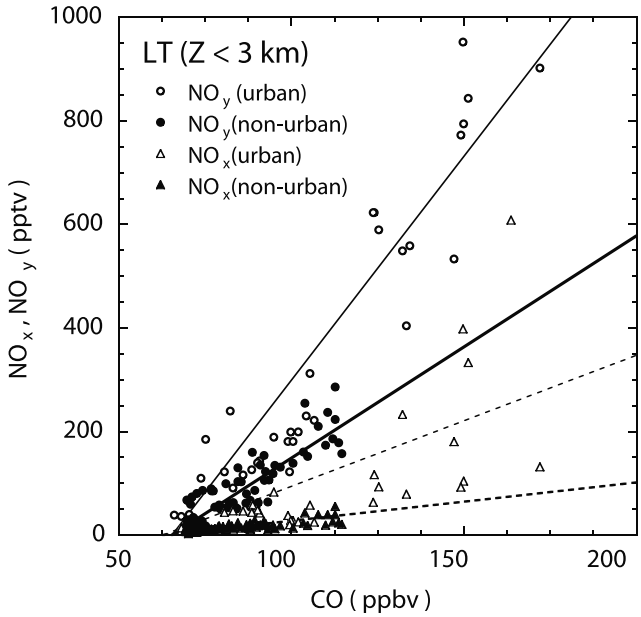

Figure 6. The $CO\text{-}NO_y$ and $CO\text{-}NO_x$ relationships in the LT in Indonesian (nonurban and urban) air masses. Closed and open symbols denote data obtained in nonurban and urban air masses, respectively. Thick and thin lines denote linear regression relationships for these data.

[22] Urban air masses were sampled during take-off and landing at the airport in Bandung, which is about 130 km from Jakarta and is the third largest city in Indonesia. In these air masses CO mixing ratios of up to 170 ppbv were observed below 3 km. When urban data are used, dNO_x/dCO and dNO_y/dCO gradients of 2.5 and 8.7 pptv/ppbv were obtained, shown in Figure 6. In these calculations, a regression line was forced to have the same background (intercept) value for CO as that of nonurban air masses. Because the horizontal extent of influence from urban sources in the LT was not identified in this study, the importance of urban sources on the reactive nitrogen levels in the UT could not be estimated quantitatively. However, the urban area in Indonesia was still limited and the relative importance was assumed to be insignificant.

4.2.3. Chemical Characteristics of UT Air Masses

[23] The differences in the chemical characteristics of Indonesian air masses from those of tropical air masses in the UT indicate the influence of vertical transport of air masses from the LT and MT. In fact, analyses using air mass trajectories and Geostationary Meteorological Satellite (GMS)-derived cloud height data indicate that most Indonesian air masses sampled in the UT, had encountered clouds over the Indonesian islands during the previous 3 days, as described later in section 4.3. Thus one would

expect that Indonesian air masses in the UT had chemical characteristics similar to those of nonurban air masses sampled in the LT, although mixing with ambient air masses transported from the upper stream (namely tropical air masses) might have changed the absolute values of various species in the UT.

[24] In Figures 7a and 7b, relationships of CO to CH_3Cl and CO to C_2H_2 in Indonesian UT air masses are compared with those in LT nonurban and urban air masses. The median value in the UT in tropical air masses is also shown for reference. CH_3Cl is known to be a good indicator of the influence from biomass burning [*Blake et al.*, 1996, and references therein]. In fact, the slope dCH_3Cl/dCO is greater in nonurban air masses than in urban air masses. Although the correlation is not very similar in the UT, data generally follow a mixing line between nonurban air mass and tropical air masses. C_2H_2 is produced both by biomass burning and fossil fuel combustion. The CO-C_2H_2 pair had one of the best correlations both in the LT and UT ($r^2 = 0.70$) during BIBLE A [*Kita et al.*, 2002]. The gradient (dC_2H_2/dCO) was shallower in nonurban air masses than in urban air masses, likely due to aging processes, namely photochemistry and mixing (Figure 7b). Data in the UT generally followed the nonurban relationship. The results shown here suggest that UT air masses generally had chemical characteristics similar to those of nonurban air masses sampled in the LT. In this study, the typical relationships in nonurban air masses were used to estimate the contribution from lightning NO production, and the relationships in urban air masses were utilized to estimate uncertainties of the present analyses.

4.2.4. Contribution From Lightning NO Production

[25] Scatterplots of CO and NO_x and CO and NO_y in the UT are shown in Figures 8a and 8b, together with the typical relationships in nonurban air masses in the LT. The relationship in urban air masses and the median value in the tropical UT air masses are also shown for reference. Although a positive correlation was generally found between CO and NO_x and between CO and NO_y, the NO_x and NO_y concentrations were systematically higher than the corresponding concentrations predicted by the measured CO and the typical relationship in nonurban air masses. These results indicate that simple vertical transport of air masses in the LT cannot account for the observed high NO_x and NO_y values in the UT. Consequently, reactive nitrogen sources in the free troposphere, most likely lightning NO production, are strongly suggested.

[26] The positive correlation observed between CO and NO_x and between CO and NO_y in the UT (Figures 8a and 8b) was reasonable because lightning activity was generally caused by convection over land, which can transport air masses near the surface to higher altitudes. Air masses with enhanced mixing ratios of CO and NO_y were considered to have been affected more by surface sources and lightning NO production, while air masses with lower values were considered to have been transported from over the ocean with less influence from surface sources. This was confirmed by an event observed near the coast of Papua New Guinea during flight 13 made on October 9. In this case, the NO_x mixing ratio reached 2,500 pptv and the NO_x/NO_y ratio was close to unity, suggesting that it was due to very recent lightning NO production. Even in this case, a positive correlation between NO_y and CO was seen in air masses, in

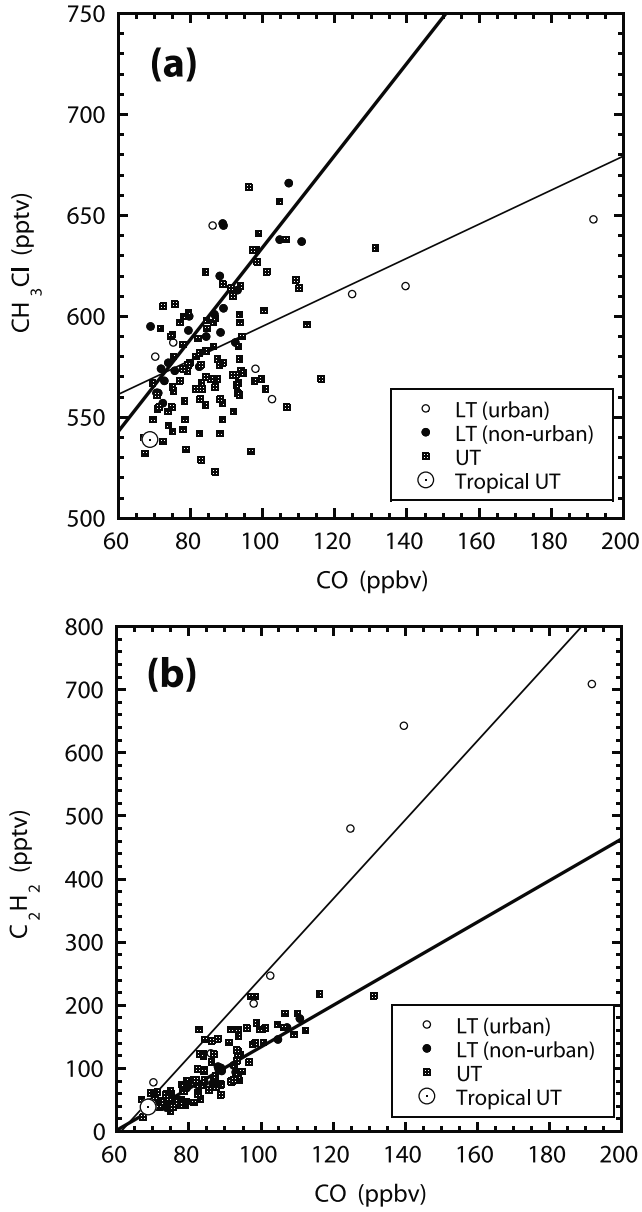

Figure 7. Scatterplot of CO-CH_3Cl and CO-C_2H_2 in Indonesian urban and nonurban air masses in the LT and that in the UT. Thick and thin lines denote linear regression relationships for LT data. Median value in the UT in tropical air masses is also shown.

which the NO_y and CO mixing ratios were 50–250 pptv and 50–65 ppbv, respectively (not shown). The NO_y values observed in this event were larger than suggested by the typical CO-NO_y relationship in the LT, consistent with lightning NO production. During the Transport and Atmospheric Chemistry near the Equator-Atlantic (TRACE-A) experiment, enhancements of NO_x due to lightning production in association with deep convection that transported trace gases from biomass burning were also observed in the equatorial South Atlantic [*Pickering et al.*, 1996; *Smyth et al.*, 1996].

[27] As seen in Figures 6, 8a and 8b, the lowest values of CO, NO_x, and NO_y in Indonesian air masses were similar

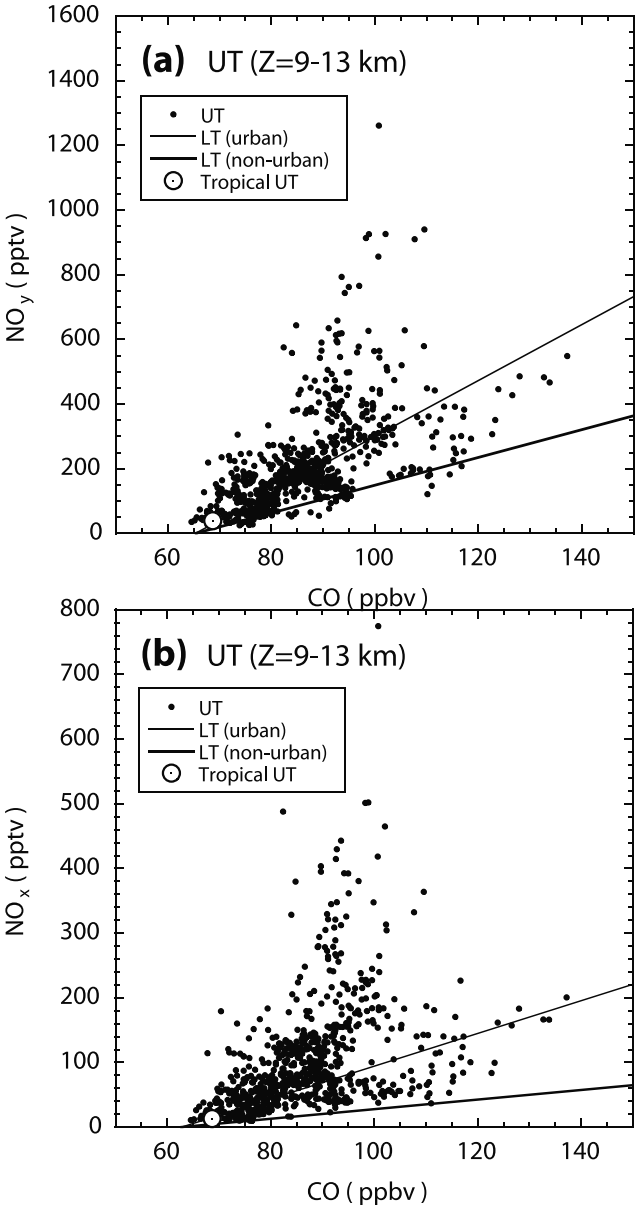

Figure 8. Scatterplot of CO-NO_y and CO-NO_x in the UT in Indonesian air masses. Relationships obtained in the LT (Indonesian nonurban and urban air masses) and median value in the UT in tropical air masses are also shown.

between LT and UT and they were 60–70 ppbv, 0–20 pptv, and 10–50 pptv, respectively. These values were considered to be background values and are also close to the median values in tropical air masses as seen in Figures 8a and 8b, consistent with the trajectory analysis described above. Similarity in the background values of these species between air masses in the LT and UT allowed us to make a direct comparison of the CO-NO_x or CO-NO_y relationships between the two air masses. An excess of NO_x (ΔNO_x) and NO_y (ΔNO_y) was defined as the difference between the observed values of NO_x or NO_y in the UT and the value expected from the typical relationships in nonurban air masses in the LT. A median value of ΔNO_x and ΔNO_y was calculated in every 2-km interval and shown in Figure 9a. Using these values and median values of NO_x

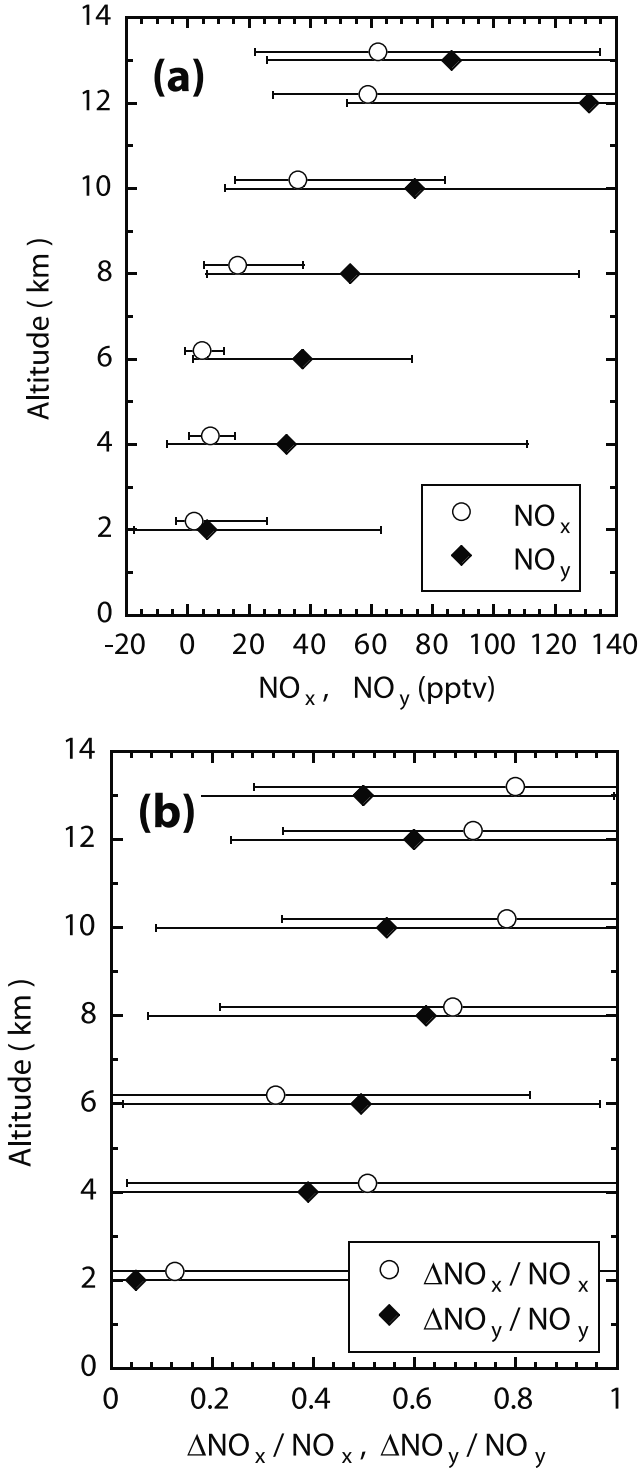

Figure 9. (a) The excess of NO_x (ΔNO_x) and NO_y (ΔNO_y) defined as the difference between the observed values of NO_x or NO_y in Indonesian air masses and the value expected from the typical nonurban relationships in the LT. Median value and 67% range are shown. (b) The ratio between ΔNO_y and the median value of the observed NO_y in Indonesian air masses. The ratio between ΔNO_x and the median value of the observed NO_x is also shown.

and NO_y shown in Figures 3b and 3c, $\Delta NO_x/NO_x$ and $\Delta NO_y/NO_y$ were also calculated and shown in Figure 9b. The $\Delta NO_x/NO_x$ and $\Delta NO_y/NO_y$ values suggest the relative contribution from lightning NO production to the observed levels of NO_x and NO_y. As seen in Figures 9a and 9b, ΔNO_x and ΔNO_y values increased with altitude, suggesting an increasing importance of a contribution from lightning NO production. In the UT, ΔNO_x and ΔNO_y values reached 40–60 pptv and 70–100 pptv, respectively. This result indicates that 80% and 50% of the observed levels of NO_x and NO_y were likely due to lightning NO production over the Indonesian islands. Because NO_y values were lower in the MT than in the UT, the relative importance of lightning NO production ($\Delta NO_y/NO_y$) was comparable between the two altitude ranges. It should be noted that NO_x recycling from HNO_3 could also cause the increase in ΔNO_x values in the UT. This effect will be discussed later.

[28] As described in section 4.1, observed differences in NO_y values between Indonesian and tropical air masses in the UT (Figure 3b) were likely due to inputs which had been made when Indonesian air masses passed over the Indonesian islands. The difference in the NO_y values was 80–170 pptv with an average value of 130 pptv in the UT. On the other hand, in the UT the ΔNO_y values (Figure 9a) were 70–100 pptv with an average value of 85 pptv. These results suggest that about 65% of the difference in NO_y between Indonesian and tropical air masses was likely due to lightning NO production. The rest of the difference could be attributed to the influence from surface sources.

4.2.5. Uncertainties in the Estimations

[29] Various uncertainties in the estimation of the contribution from lightning NO production were examined. First, influence from urban sources is considered. In this study, the contribution from lightning NO production was estimated using CO-NO_x and CO-NO_y relationships in nonurban air masses. When the relationships in urban air masses are used instead, ΔNO_x and ΔNO_y values become negative at altitudes below 7 km and from one-third to one-fourth above 9 km (UT). Although the negative value in the MT does not make sense, our method could have overestimated the contribution from lightning in the UT by ignoring the urban data. Unfortunately, we are unable to provide a quantitative estimate of the relative influence between urban and nonurban air masses. Furthermore, mixing ratios of light alkane species such as ethane (C_2H_6) and propane (C_3H_8) in a part of air samples in the UT exceeded the values in urban and nonurban air masses [*Kita et al.*, 2002]. This result suggests that the chemical characteristics of Indonesian air masses in the UT was not fully accounted for a simple mixing of urban, nonurban, and tropical air masses, resulting an additional uncertainty in the estimations of this study.

[30] Second, the influence from the removal of HNO_3 by wet deposition during the convective transport is examined. Because HNO_3 is highly soluble in water (Henry's law constant at room temperature is 2.1×10^5 mol L^{-1} atm^{-1} [*Schwartz and White*, 1981]), a considerable amount of HNO_3 could be removed within the convection system. As a result, a smaller amount of NO_y could be transported to the UT as compared to the amount expected from the CO-NO_y relationship in the LT. Thus we could have underestimated the influence of lightning NO production. In the LT, the median values of NO_x/NO_y ratios were about 0.2, suggesting an upper limit of HNO_3/NO_y ratios of 0.8. During PEM-Tropics A, HNO_3/NO_y ratios of about 0.5 were observed over the South Pacific at altitudes below 2 km both inside and outside air masses influenced by combustion [*Talbot et al.*, 2000]. During PEM-West B, an HNO_3/NO_y ratio of 0.8 was found in the tropical marine boundary layer [*Kawakami et al.*, 1997; *Kondo et al.*, 1997b]. Assuming that 50% of NO_y was removed during convective transport, the ΔNO_y values between 10 and 12 km would be 100–140 pptv. This would mean that the contribution from lightning could be 80% of NO_y rather than 50%.

[31] Third, a contribution to increase ΔNO_x values from recycling of NO_x from HNO_3 by photolysis and reaction with OH is examined. Because of the short NO_x recycling time, 2–3 days in the tropical UT, NO_x production from HNO_3 could result in an increase in ΔNO_x even when there was no net increase in ΔNO_y due to lightning. If this is the case, the estimation of influences from lighting NO production using ΔNO_x values might be overestimated. The contribution of recycled NO_x depends on the amount of HNO_3 in the LT and the removal rate of HNO_3 during convective transport. The differences in NO_x values between Indonesian and tropical air masses in the UT (Figure 3c) were 40–70 pptv, with an average value of 60 pptv. If air parcels were in photochemical equilibrium, 65% of this difference (40 pptv) would have been due to lightning NO production (see NO_y discussion above). This value is close to a ΔNO_x of 40–60 pptv (average was 50 pptv) in the UT, suggesting that recycling of NO_x would not result in a significant overestimation of lightning influences. The increase in NO_x that could result from NO_x recycling through the HNO_3 transport was possibly suppressed by the removal of HNO_3 during convective transport. Note that recycling of NO_x is still very important to keep a high NO_x level in the tropical UT. Lightning NO production increases the NO_x level and recycling keeps its level high. Both processes are essential for O_3 production, as discussed later.

4.3. Lightning NO Production and Deep Convection

[32] Both the Lightning Image Sensor (LIS) onboard the tropical Rainfall Measuring Mission (TRMM) satellite and Optical Transient Detector (OTD) onboard the MicroLab-1 satellite [*Nesbitt et al.*, 2000] showed that there was considerable lightning activity around the Indonesian islands during the BIBLE A period (Figure 10). Most lightning flashes were detected over islands, consistent with the earlier results from the global scale satellite measurements, which show that most lightning occurs over land [e.g., *Turman and Edgar*, 1982]. Because of La Niña, deep convection around the Indonesian islands during the BIBLE A period was generally stronger than in normal years, as seen in Figures 2a and 2b. As a result, deep convection events were frequently seen over the Indonesian islands, even though the BIBLE A experiment was carried out during the dry season. The total number of lightning flashes detected by the LIS sensor in the region 10°S–10°N and 95°E–115°E in September and October 1998 was comparable to those in other months and those in September and October 1999 (Figure 11). Thus the amount of lightning during the BIBLE A period was typical and so was the amount of NO_x produced by lightning activity.

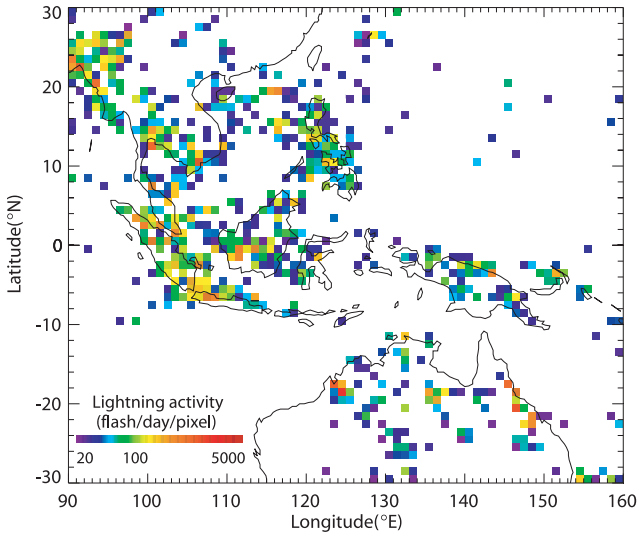

Figure 10. Lightning flashes rate detected by the LIS satellite sensor during the BIBLE A period. A pixel size for these statistics is 1° × 1° in latitude and longitude.

[33] To investigate the relationship between the observed NO_x levels and lightning activity, we examined each air mass for lightning influence along back trajectories. Unfortunately, because the LIS and OTD sensors looked at one specific area only a short time period each day, we could not find a case in which lightning flashes were coincidentally detected on a back trajectory of an observed air mass. Instead of using lightning data, we investigated the relationship between the observed NO_x level and deep convective activity along a back trajectory, which might have caused lightning over land. For this analysis we used hourly blackbody temperature (Tbb) data obtained by the Geostationary Meteorological Satellite (GMS-5). When the temperature of an air mass along its trajectory was higher than the Tbb of a cloud, we assumed that the air mass was affected by convection. Furthermore, to discriminate cumulus clouds caused by deep convection from cirrus clouds, we selected only the cases in which two IR channels, channel 1 (10.5–11.5 μm) and channel 2 (11.5–12.5 μm), agreed to within 2.3 K. This was because cumulus clouds were generally optically thick and the difference in Tbb in these two channels was small [*Inoue*, 1989]. In Figure 12, NO_x mixing ratios in the UT are plotted versus the difference in time between the measurement and an encounter with the most recent deep convection. In Figure 12, different symbols are used for Indonesian air masses when they encountered clouds over the Indonesian islands or over the ocean. In addition to Indonesian air mass data, all of the data obtained in the UT at latitudes south of 10°N were also plotted.

[34] In Figure 12, several important results are illustrated. First, most Indonesian air masses sampled in the UT (89%) encountered clouds within 3 days, and in most of these cases (79%) they encountered clouds over the Indonesian islands. In other words, most air masses that passed over Indonesian islands were affected by convection over the islands.

[35] Second, when NO_x mixing ratios were higher than 100 pptv, most air masses had encountered convective clouds within 48 hours. Air masses with especially high NO_x values (>200 pptv) had encountered clouds within 24 hours. These results indicate that high NO_x values were mostly due to recent convective events. As described above,

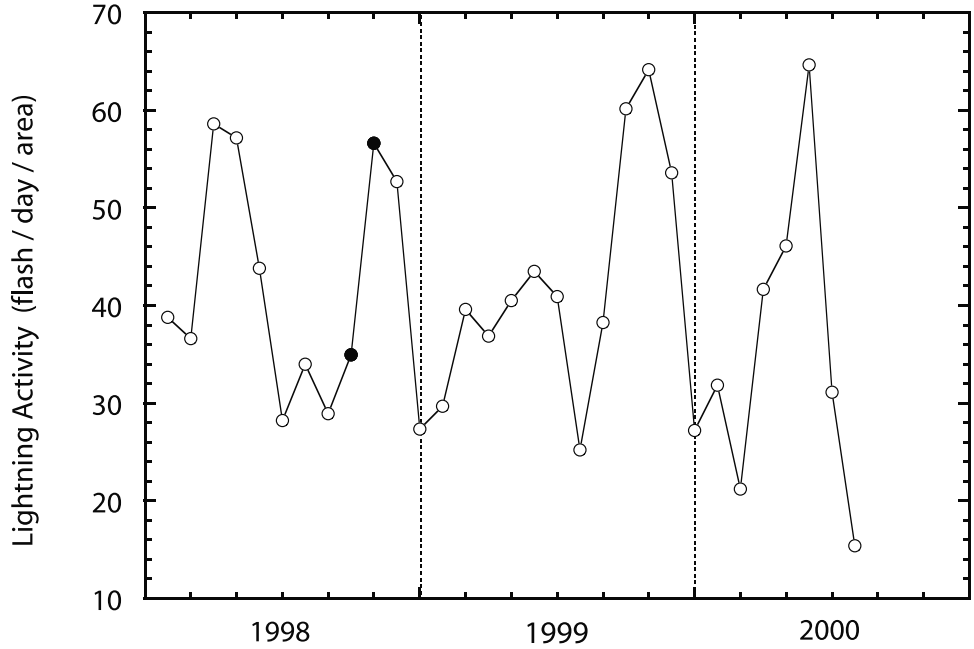

Figure 11. Monthly average lightning flash rate detected by the LIS satellite sensor in the region 10°S–10°N and 95°E–155°E between January 1998 and July 2000. Data obtained in September and October 1998 are denoted by solid circles. A pixel size for these statistics is 1° × 1° in latitude and longitude. Because solar local time of the measurement at one particular location changed by 24 hours within 40 days, diurnal variation of the lightning activity is considered to be negligible in this plot.

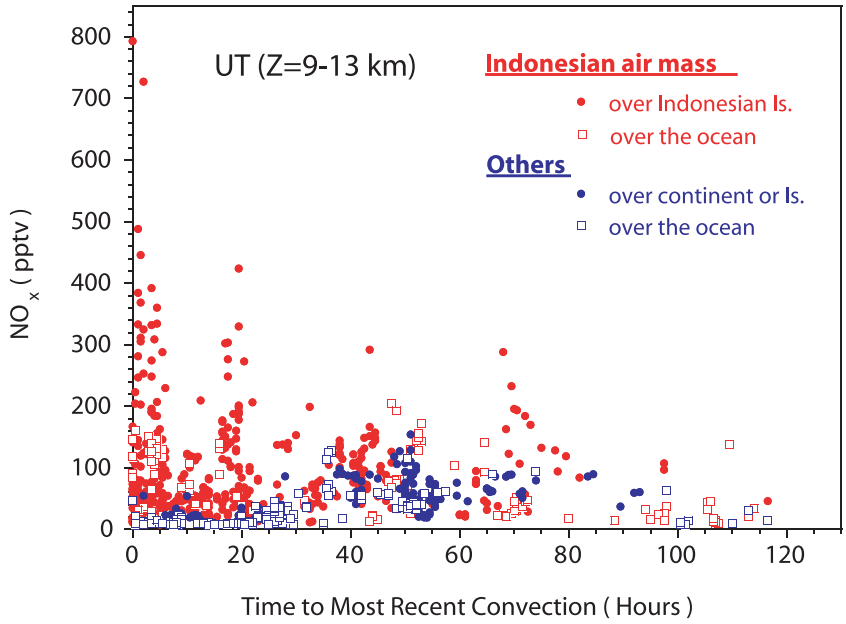

Figure 12. The NO_x mixing ratio as a function of the time difference between the measurement and most recent encounter with a deep convection. All the data obtained in the UT at latitudes south of 10°N are shown.

the NO_x and NO_x/NO_y values had a positive correlation in the UT and the range of the median value of the NO_x/NO_y ratios was 0.56–0.94 in air masses in which NO_x values were higher than 100 pptv (Figure 4). These results further confirm that the observed high NO_x values in the Indonesian air masses in the UT was largely due to lightning NO production that took place over Indonesia within a few days prior to the measurement.

[36] Third, when air masses other than Indonesian air masses encountered clouds over the ocean, NO_x values were generally low, 5–20 pptv. This was quite likely due to convective transport of air masses from the marine boundary layer where NO_x mixing ratios were quite low. Most of these air masses were those classified as tropical air masses, and they were generally affected by deep convection in association with large scale convective activity, as seen in Figures 2a and 2b. The results obtained here suggest strongly that convection over the land, which is generally accompanied by lightning activity, enhanced the NO_x level, while convection over the ocean generally lowered it. This is a first attempt to show the statistical relationship between the NO_x level and deep convection activity over the western Pacific. The results presented here did not change significantly when clusters of trajectories were used instead of a single trajectory.

[37] One of the high-NO_x events observed during flight 11 made on October 7 is examined here. During this roundtrip flight, air masses over the west coast of South Sumatra were sampled at the same horizontal locations twice but at different altitudes, 11 and 13 km. Mixing ratios of NO_x and NO_y reached 200–800 and 300–1000 pptv, respectively, with NO_x/NO_y ratios of 0.4–0.7. Air masses with these high NO_x values extended about 300 km. The correlation between CO and NO_x mixing ratios was weak, suggesting that contributions from NO_y sources at the surface were relatively small. Based on back trajectory analysis of air masses and GMS Tbb data, the air mass with the highest NO_x/NO_y ratio observed on this day had probably encountered convective cloud within several hours prior to the measurement. Although the LIS and OTD sensors did not make measurements at the location where these air masses encountered clouds on that day, lightning activities were occassionally observed in that region.

4.4. Impact on Ozone

[38] Diurnally averaged photochemical production rates and loss rates of O_3 were calculated by the AER photochemical box model [*Ko et al.*, 2002] using observed values of atmospheric temperature, pressure, O_3, NO, CO, CH_4, H_2O, NMHCs, and $J(NO_2)$. Acetone mixing ratios were calculated from the observed values of CO using the reported CO-acetone correlation [*McKeen et al.*, 1997]. Median values of the net photochemical O_3 production rate (production minus loss) is shown for Indonesian, tropical, and aged Indonesian air masses in Figure 13. The net production rate was generally higher at higher altitude primarily due to the lower H_2O concentration in the UT resulting in a lower O_3 loss rate through the $O(^1D) + H_2O$ reaction [*Ko et al.*, 2002]. In tropical air masses, although positive net production rates were observed above 9 km, they were less than 0.5 ppbv/day primarily because of the low NO_x mixing ratios (Figure 3c). In contrast, positive net O_3 production of 1.5–2 ppbv/day was calculated for the UT in Indonesian air masses, as a result of high NO_x mixing ratios. In addition, other O_3 precursor species, such as CO, CH_4, and NMHCs were also generally higher in these air masses due to convective transport over the Indonesian islands as compared to those in the tropical air masses, which also contributed to higher O_3 production rates [*Kita et al.*, 2002]. The net production rate obtained during the PEM-West B experiment in high-NO_x and low-NO_x regimes given by *Crawford et al.* [1997] are also shown

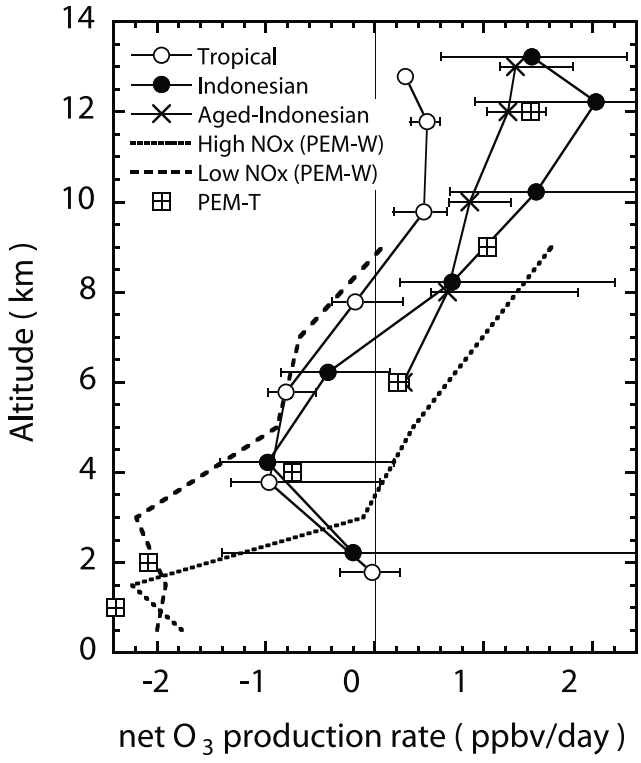

Figure 13. Median values of the net O_3 production rate (production minus loss). Values obtained during the PEM-West B (PEM-W) and PEM-Tropics A (PEM-T) experiments are also shown.

in Figure 13. Consistent with the NO_x profiles (Figure 3c), similar tendencies were found between the low-NO_x and tropical air masses and between high-NO_x and Indonesian air masses. The net production rate obtained during the PEM-Tropics A experiment in the region of 0°–30°S and 170°E–120°W reported by *Schultz et al.* [1999] is also shown. Air masses sampled both north and south of the SPCZ were included for statistical reasons. The net production rates in Indonesian air masses are generally close to those in air masses sampled over the equatorial-tropical South Pacific, which had been partly affected by biomass burning in Australia, Indonesia, Africa, and South America.

[39] Aged Indonesian air masses had generally crossed over Indonesian islands within 5 to 10 days prior to the measurements, as described above. Consistent with these air mass trajectories, median values of CO at 5–13 km were similar between Indonesian and aged Indonesian air masses (Figure 3a), suggesting similar characteristics of the two types of air masses. Consequently, lightning NO production in addition to the vertical transport of NO_y was likely a source of NO_x in the Aged Indonesian air masses in the UT. Because of the rapid recycling of NO_x in the tropical UT, NO_x/NO_y ratios were not greatly different between Indonesian and aged Indonesian air masses (Figure 3d), and therefore NO_x levels were also similar between the two types of air masses. As a result, net photochemical O_3 production rates in aged Indonesian air masses were as high as those in Indonesian air masses along the trajectories (Figure 13). aged Indonesian air masses could have been further affected by deep convection after leaving the Indonesian islands and NO_x levels could have been altered by these events. However, observed results suggest that once NO_x molecules were produced by lightning over the Indonesian islands, their concentration generally remained high, allowing for efficient O_3 production during transport. Considering the high HO_x concentration due to the convective transport of precursor species and high solar radiation intensity in the tropical UT, NO_x production by lightning should have a significant impact on the O_3 budget over the western Pacific. The observed O_3 levels and photochemical O_3 production processes are discussed in more detail by *Kita et al.* [2002].

5. Summary

[40] Measurements of NO and NO_y were made over the tropical western Pacific during the BIBLE A aircraft campaign conducted in September and October 1998. Using ten-day back trajectories, we defined three air mass types sampled at latitudes south of 20°N: Indonesian air masses (having crossed over Indonesia within 3 days prior to the measurement), tropical air masses (originating in the central Pacific and transported without crossing any continent or islands for 10 days), and aged Indonesian air masses (having passed over Indonesia five to ten days prior to the measurement). Because 60% of Indonesian air masses in the UT had originated from the central Pacific before they crossed over the Indonesian islands, the difference in mixing ratios between Indonesian and tropical air masses was likely due to processes that occurred while air masses were over the Indonesian islands.

[41] Mixing ratios of CO, NO_x, and NO_y in Indonesian air masses were systematically higher than those in tropical air masses. In Indonesian air masses, the NO_x and NO_y mixing ratios at 12 km were as high as 80 and 200 pptv, respectively. Biomass burning and fossil fuel combustion were likely the sources of CO over Indonesian islands, however lightning NO production was an important source of NO_x and NO_y.

[42] To assess the influences of surface sources on NO_x and NO_y levels in the UT in Indonesian air masses, CO-NO_x and CO-NO_y relationships in the LT were studied. Typical gradient of NO_x and NO_y to CO (dNO_x/dCO and dNO_y/dCO) in nonurban air masses was much lower than that in the biomass burning plumes and urban air masses encountered over Indonesia, suggesting the influence of these two sources were averaged over a large area and generally not significant in the LT. The relationships between CO and CH_3Cl and CO and C_2H_2 are generally similar in UT air masses and nonurban air masses sampled in the LT, suggesting they had similar chemical characteristics through the efficient convective transport over the islands. When the CO-NO_x and CO-NO_y relationships in the UT were compared to the typical relationships in the LT, the NO_x and NO_y values were higher by 40–60 pptv (80% of NO_x) and 70–100 pptv (50% of NO_y), respectively. Furthermore, there was a positive correlation between NO_x and the NO_x/NO_y ratio at 12–13 km, and the range of median NO_x/NO_y ratios was 0.56–0.94 when NO_x was higher than 100 pptv. A contribution from the lightning NO production was suggested as a significant source of NO_x and NO_y in the UT over the Indonesian region. This result indicates that

on average, 65% (85 pptv) of the difference in NO_y between Indonesian and tropical air masses in the UT (130 pptv) was likely due to lightning NO production. The influence from lightning could have been overestimated if uncertainties in the contribution from urban and other unidentified sources are considered. On the other hand, it is likely underestimated if removal of HNO_3 during convective transport is not considered.

[43] Influence from deep convection on the NO_x level in the UT was examined using air mass trajectories and GMS satellite-derived cloud height data. Most Indonesian air masses were found to have been influenced by deep convection over the Indonesian islands. When NO_x mixing ratios were higher than 100 or 200 pptv, they had generally been affected by deep convection over the Indonesian islands within 48 and 24 hours, respectively. On the contrary, when air masses other than Indonesian air masses encountered clouds over the ocean, NO_x values were generally as low as 0–20 pptv. These results are a strong indication that convection over land, which was usually accompanied by lightning activity, increased the UT NO_x values, while convection over the ocean generally lowered the NO_x level due to lack of lightning activity.

[44] Air mass trajectories and similarity in the CO mixing ratios suggested that aged Indonesian air masses had been influenced by the surface sources and lighting NO production, similar to Indonesian air masses. The NO_x/NO_y ratio and NO_x mixing ratios were generally similar between Indonesian air masses and aged Indonesian air masses, likely because rapid NO_x recycling in the tropical UT kept the NO_x values at high levels. As a consequence, net photochemical O_3 production rates of 1–2 ppbv/day were found in both air masses. These results suggest that NO_x production by lightning over the Indonesian islands has a significant impact on the O_3 budget over the western Pacific.

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