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# Biomass burning influences on the composition of the remote South Pacific troposphere: analysis based on observations from PEM-Tropics-A

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## Abstract

Airborne, in situ measurements from PEM-Tropics-A (September/October 1996) are analyzed to show the presence of distinct pollution plumes in the middle-tropical troposphere of the remote South Pacific (10–30°S). These elevated plumes cause a relative maximum at about 5–7 km altitude in the vertical distribution of primary and secondary species characteristic of fuel combustion and biomass burning (CO, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>6</sub>, CH<sub>3</sub>Cl, PAN, O<sub>3</sub>). Similar plumes were also observed at mid-latitudes in the middle troposphere during three flights east of New Zealand (40–45°S). In all, pollution plumes with CO larger than 100 ppb were observed 24 times on seven separate flight days south of the equator. The observed plumes were frequently embedded in very dry air. Ten-day back trajectory analysis supports the view that these originated from the biomass burning regions of South Africa (and South America) and were transported to the South Pacific along long-distance subsiding trajectories. The chemical composition of the southern Pacific troposphere analyzed from the PEM-Tropics-A data is compared with data from the tropical regions of the northern Pacific (PEM-West-A) and southern Atlantic (TRACE-A) during the same Sept/Oct time period. Sizable perturbations in the abundance of ozone and its key precursors, resulting from the transport of pollution originating from biomass burning sources, are observed in much of the Southern Hemispheric troposphere. © 1999 Elsevier Science Ltd. All rights reserved.

**Keywords:** Trace chemicals; Biomass burning pollution; Remote atmospheres

## 1. Introduction

The NASA Global Tropospheric Experiment (GTE) aims to elucidate the composition and chemistry of the

earth's atmosphere through a series of airborne field experiments (McNeal et al., 1983). The tropical marine regions of the atmosphere are thought to be relatively free of direct influences from man-made pollution. Significant industrial growth in Asia and biomass burning pollution in Africa and South America have the potential to greatly impact these pristine tropical regions. The environmental consequences of such pollution are diverse and include emission of acidic gases leading to

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acidic precipitation, release of particles that can impact cloud formation and precipitation, and release of photochemical precursors (hydrocarbons and  $\text{NO}_x$ ) that can result in widespread production of ozone in the troposphere (Crutzen and Andreae, 1990; Fishman et al., 1991; Lacaux, 1992; Jacob et al., 1996; Davis et al., 1996). As part of GTE, a field experiment [Pacific Exploratory Mission (PEM)-Tropics-A] was performed in September/October 1996 to explore the south central Pacific region in search of nearly unpolluted air. PEM-Tropics-A complemented two previous GTE missions conducted during the same months in the western Pacific (PEM-West-A; 1991) and South Atlantic transport and atmospheric chemistry near the Equator–Atlantic (TRACE-A) 1992). While PEM-West-A explored the industrial outflow from Asian countries, TRACE-A largely concentrated on sampling biomass burning pollution from Africa and South America. In all cases an instrumented NASA DC-8 aircraft was utilized for airborne measurements to 12-km altitudes.

The geographical coverage of the PEM-Tropics-A missions is shown in Fig. 1. The NASA DC-8 flew 14 missions in the South Pacific covering areas from the equator to about  $45^\circ\text{S}$  and from  $155^\circ\text{E}$  to  $110^\circ\text{W}$ . An exploratory flight to the Antarctic was also conducted. Distributions of important trace chemicals observed in the troposphere are analyzed with a focus on the potential impact of biomass burning pollution. Measurements obtained during PEM-West-A, TRACE-A, and PEM-Tropics-A are compared. Experimental and meteorological details of PEM-West-A (Hoell et al., 1999), TRACE-A (Fishman et al., 1996) and PEM-Tropics-A (Hoell et al., 1999; Fuelberg et al., 1999) can be found in the available literature.

### 1.1. PEM-Tropics-A comparisons with PEM-West-A and Trace-A

Fig. 2 shows mean latitudinal profiles of select species characteristic of primary and secondary products of

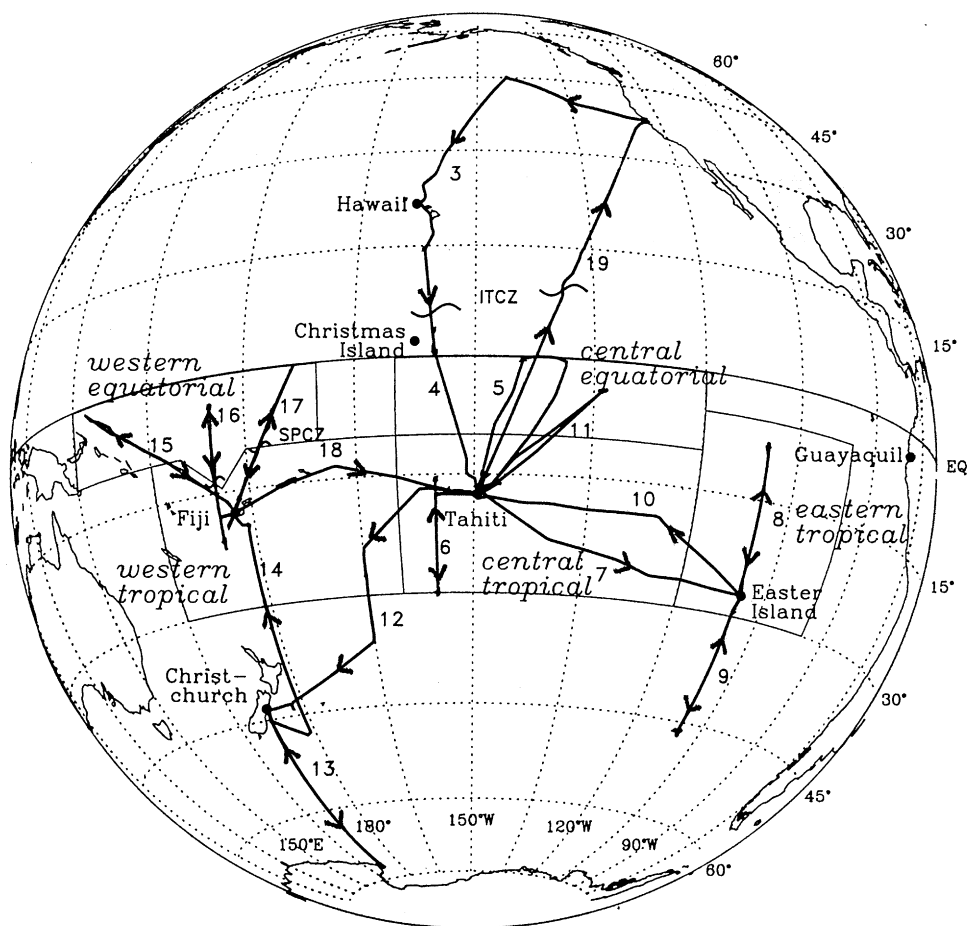


Fig. 1. Geographical coverage of the PEM-Tropics-A airborne expedition. This experiment took place from 30 August to 6 October 1996. Mission numbers are indicated.

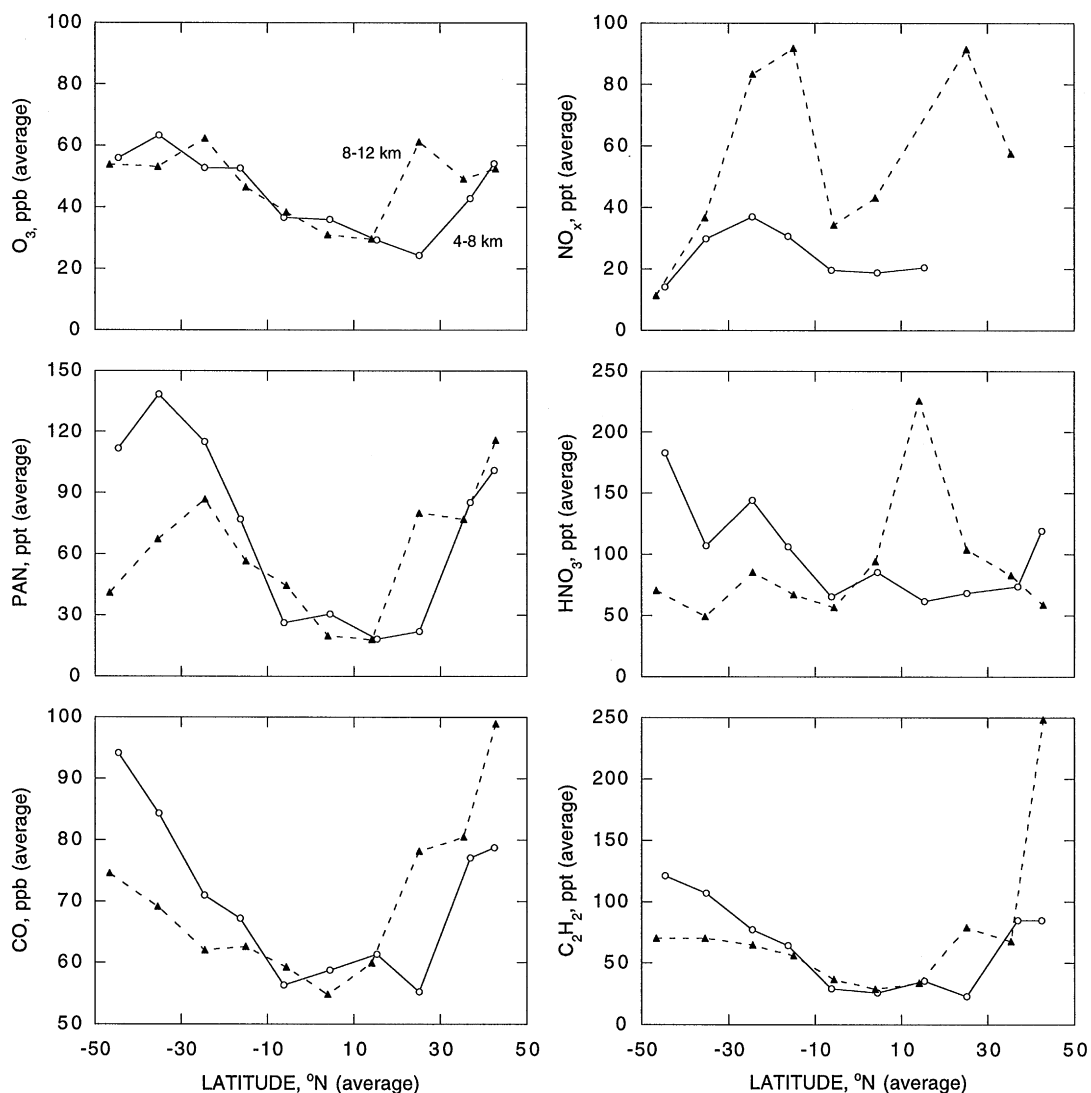


Fig. 2. Average latitudinal distribution ( $10^\circ$  bins) of selected reactive nitrogen species and primary tracers measured during PEM-Tropics-A.

continental and biomass combustion ( $\text{CO}$ ,  $\text{C}_2\text{H}_2$ ,  $\text{NO}_x$ , PAN,  $\text{HNO}_3$  and  $\text{O}_3$ ) in the free troposphere.  $\text{NO}_x$  is defined as the sum of the measured  $\text{NO}$  and  $\text{NO}_2$ . It has been shown that during PEM-Tropics-A there was good agreement between measured  $\text{NO}_2$  and that derived from measured  $\text{NO}$  (Bradshaw et al., 1999). The profiles are obtained by averaging data from all PEM-Tropics-A missions shown in Fig. 1 for the middle (4–8 km; 600–400 mb) and upper (8–12 km; 400–200 mb) troposphere over  $10^\circ$  latitude bins. As is evident from Fig. 1, the density of data was the greatest between 0 and  $30^\circ\text{S}$  with more limited coverage elsewhere. The most salient feature of these latitudinal profiles (Fig. 2) is the enhancement in the mixing ratio of key chemicals in the Southern

Hemisphere. In virtually all cases (e.g.  $\text{CO}$ ,  $\text{C}_2\text{H}_2$ , PAN,  $\text{HNO}_3$ ) these enhancements are larger in the middle troposphere than in the upper troposphere. In the case of  $\text{NO}_x$  the larger mixing ratios were encountered in the upper troposphere where twin peaks at  $25^\circ\text{N}$  and  $16\text{--}25^\circ\text{S}$  were observed (Fig. 2). The  $\text{NO}_x$  peak near  $25^\circ\text{N}$  is similar to that observed during PEM-West-B (Feb/Mar, 1994) in the western Pacific at  $20\text{--}30^\circ\text{N}$ . (Singh et al., 1998). Brunner (1998), based on sampling from instruments aboard commercial aircraft, reports the presence of persistent high  $\text{NO}_x$  mixing ratios in the upper troposphere throughout the NH. No ready explanation can be given for the observed  $\text{NO}_x$  maximum in the SH upper troposphere. However, meteorological

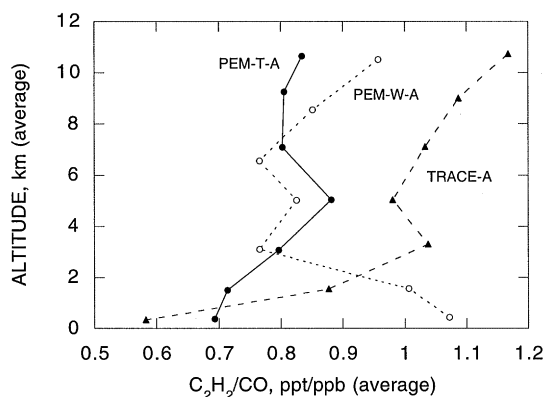


Fig. 3. Vertical profiles for the tropical ocean areas PEM-Tropics-A, TRACE-A, and PEM-West-A of  $C_2H_2/CO$  identifying tropospheric layers of aged air (low values) and freshly polluted air (high values  $> 1$ ).

analysis supports the possibility that these air masses encountered lightning during transport to the sampling region (Fuelberg et al., 1999). The mixing ratios of  $HNO_3$  during Mission 4 just south of Hawaii at about  $12^\circ N$  were high. The concurrent presence of extremely low  $NO$  (10–15 ppt) suggests that this may have involved an aged plume in which most of the  $NO_x$  had been oxidized to  $HNO_3$ .

To compare the vertical profiles of chemical data for the tropical South Pacific (PEM-Tropics-A;  $0\text{--}30^\circ S$ ,  $100^\circ W\text{--}160^\circ E$ ), central/western tropical North Pacific (PEM-West-A;  $0\text{--}20^\circ N$ ,  $160^\circ W\text{--}120^\circ E$ ), and tropical South Atlantic (TRACE-A;  $0\text{--}30^\circ S$ ,  $10^\circ E\text{--}30^\circ W$ ), it is instructive to examine Fig. 3 which shows profiles of the ratio of  $C_2H_2/CO$  for the three tropical ocean regions. This ratio is a measure of the relative processing (age) of the air masses sampled (Smyth et al., 1996). It is evident from Fig. 3, that both the free troposphere of TRACE-A and the boundary layer of PEM-West-A have values of this ratio approaching and exceeding 1.0. Such high values are suggestive of relatively recent contact with fresh pollution sources. During TRACE-A biomass burning emissions were the most likely sources of pollution (Talbot et al., 1996; Chatfield et al., 1996; Singh et al., 1996; Thompson et al., 1996), while during PEM-West-A it was the outflow of pollution from the Asian continent (Dibb et al., 1996; Talbot et al., 1996). The air sampled during PEM-Tropics-A on the whole is associated with generally low values of the  $C_2H_2/CO$  ratio, that suggest an air mass aged with respect to input from fresh pollution sources. A very similar picture also emerges from a detailed analysis of back trajectories and 3-D model simulations (Fuelberg et al., 1999; Chatfield et al., 1999).

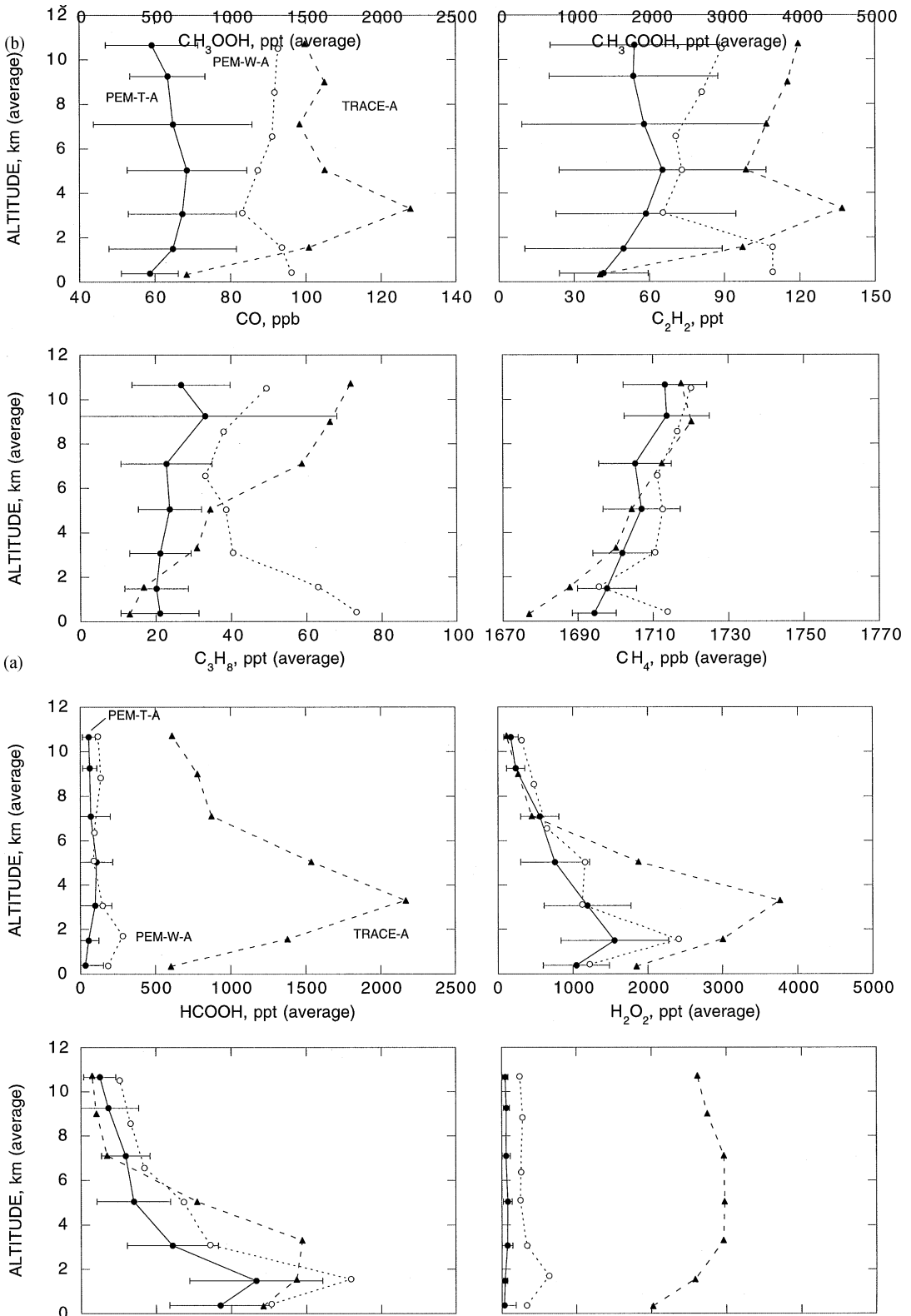
Shown in Fig. 4a and b are the vertical distributions of some primary and secondary tracers of varying solubility and lifetimes for the selected tropical marine regions.  $CO$ ,

$C_2H_2$ ,  $C_3H_8$ , and the long-lived  $CH_4$  are insoluble tracers that are released during combustion and have atmospheric lifetimes that may range from weeks to years (Blake et al., 1999; Mauzerall et al., 1998). In contrast, organic acids ( $CH_3COOH$  and  $HCOOH$ ) are more soluble largely primary pollutants while peroxides ( $H_2O_2$  and  $CH_3OOH$ ) are soluble secondary pollutants. Hydrogen peroxide ( $H_2O_2$ ) is highly soluble and along with methylhydroperoxide ( $CH_3OOH$ ) provides a good indication of the degree of wet chemical removal. While the profiles of tropospheric temperature and water vapor content (not shown) were similar for the three areas and characteristic of the tropical atmosphere, some significant differences in the chemical profiles are apparent. The most profound of these is the difference in the mixing ratios of tracers observed during PEM-Tropics-A and TRACE-A. Air masses sampled in the South Atlantic during TRACE-A were greatly influenced by the presence of nearby biomass burning activity in Africa and by transport of the resulting pollution to the sampling region within 3–5 d (Singh et al., 1996). The PEM-West-A data collected over the central/western tropical North Pacific represent the higher concentrations of tracers typically found in the NH (Blake et al., 1999). The continental Asian outflow (Talbot et al., 1996) caused the marine boundary layer of the tropical North Pacific to be the most polluted area of the troposphere for the primary insoluble tracer species (Fig. 4a).

Hydrogen peroxide,  $CH_3OOH$  are secondary products of photochemistry and their high concentrations in the tropics reflect the greater availability of sunlight and moisture in this region (Heikes et al., 1996; O'Sullivan et al., 1999). The photochemical source is most effective in the lowest troposphere where the largest concentrations are found. Unlike the peroxides, the organic acids are directly released by biomass burning emissions and the TRACE-A region was greatly influenced (Fig. 4b). Meteorological analysis supports the possibility that air masses arriving at the PEM-Tropics-A sampling region likely encountered clouds and rain during transport (Fuelberg et al., 1999).

Fig. 5 shows vertical profiles of the reactive nitrogen species ( $NO_x$ , PAN and  $HNO_3$ ) and  $O_3$ . These species have complex source and removal processes (Singh et al., 1996; Jacob et al., 1996). The impact of pollution sources on these chemicals is clearly different than in the case of the biomass tracers. A comparison of these data clearly shows that significant perturbations in the ozone and

Fig. 4. Comparison between average vertical profiles of chemical tracers characteristic of continental combustion and biomass burning for the tropical ocean areas of PEM-Tropics-A (1996), PEM-West-A (1991), and TRACE-A (1992) (Sept/Oct period). For clarity the standard deviations for PEM-T-A are indicated only.



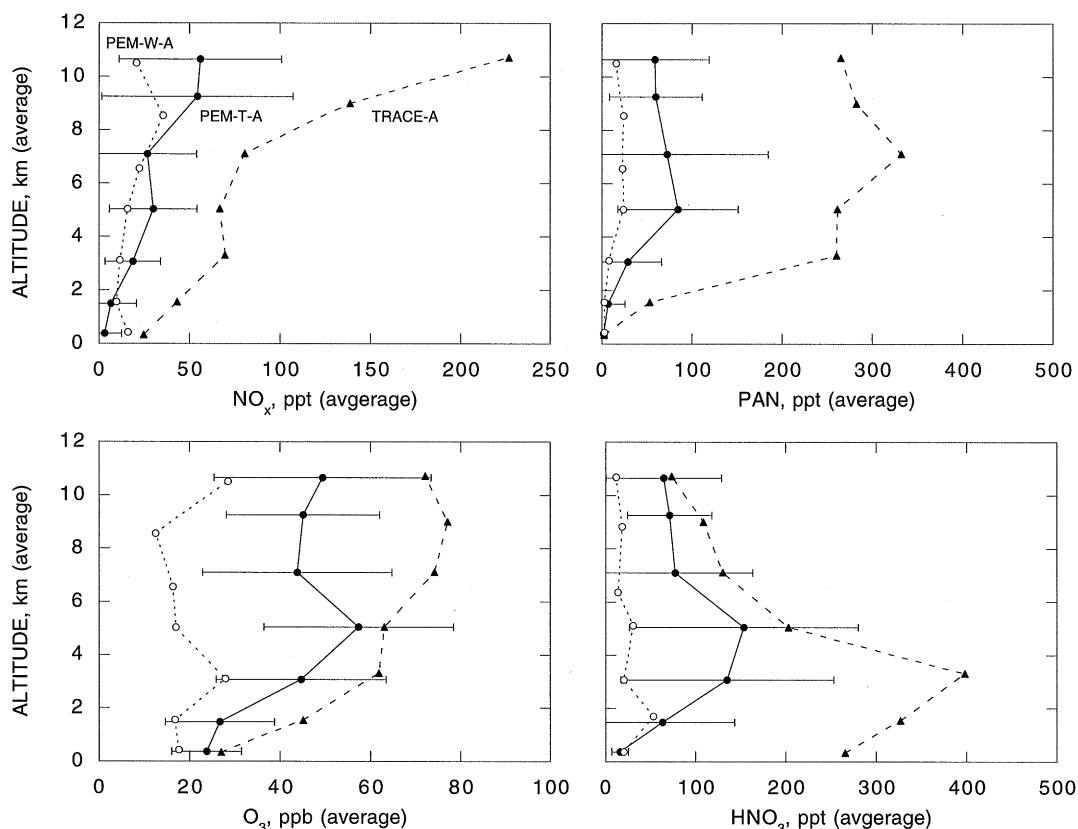


Fig. 5. Comparison between average vertical profiles of selected reactive nitrogen species and ozone for the tropical ocean areas of PEM-Tropics-A, TRACE-A, and PEM-West-A (Sept/Oct period).

reactive nitrogen fields are possible in regions far away from pollution sources. Indeed one could conclude that ozone perturbations of a factor of two or three are possible in the SH from the influence of these pollution sources and these regions can be more polluted than the tropical NH regions sampled in PEM-West-A. Another typical feature of these distributions is that the lowest mixing ratios are found in the marine boundary layer. The origin of tropospheric ozone and  $\text{NO}_x$  over the tropical south Pacific has been further analyzed by Schultz et al. (1999) with the help of a photochemical model. They emphasize the important photochemical source of  $\text{O}_3$  and  $\text{NO}_x$  from westerly transport into the region associated with biomass burning in Africa and South America and conclude that  $\text{NO}_x$  in the lower troposphere is maintained by the PAN reservoir.

## 2. Pollution plumes and their origin during PEM-Tropics-A

Specific PEM-Tropics-A missions observed very distinct pollution plumes in the middle and upper tropo-

sphere. Pollution plumes defined as  $\text{CO} > 100$  ppb were observed 24 times on seven separate missions south of the equator. During M12, a long flight from Tahiti to New Zealand, plumes were observed for a total cumulative time period of 1.5 h. One plume had a vertical thickness of nearly 2 km while another covered a horizontal distance of about 200 km. Large plumes were observed also on M13 east and just south of New Zealand. On other flights, narrow distinct plumes were detected for periods of up to 20 min. Most frequently, these plumes were present in the mid-troposphere (4–8 km), however, upper tropospheric pollution was also seen.

Fig. 6a shows an example of a CO pollution plume (maximum  $\text{CO} = 113$  ppb) observed during M6 (out of Tahiti) when the DC-8 flew southward along  $155^\circ\text{W}$  longitude at 10 km altitude (see Fig. 1). The plume, encountered at  $26^\circ\text{S}$  when the plane emerged from cirrus clouds, was embedded in very dry air (80 ppm water vapor). Coincident with the dramatic CO increase and water vapor decrease,  $\text{O}_3$  increased from 30 to 90 ppb while NO, PAN, and the NMHCs (not shown) increased similarly. The wind speed (not shown) increased to a maximum of 105 knots while the ambient temperature

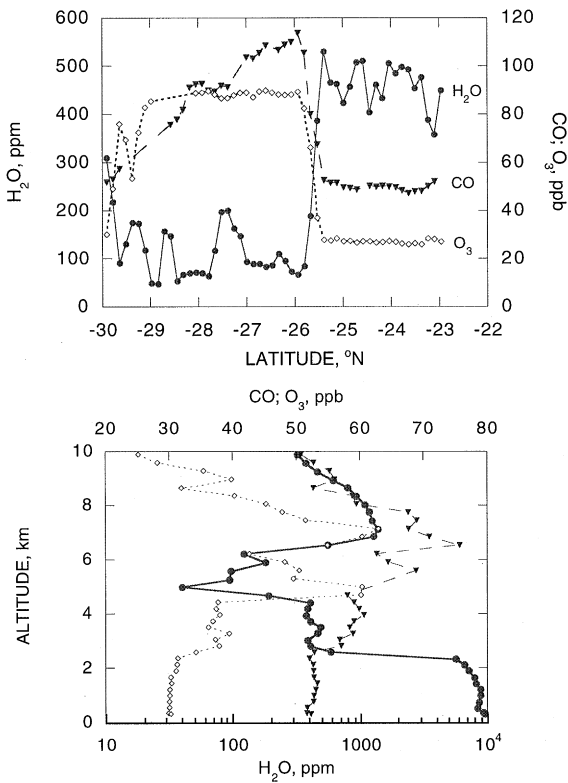


Fig. 6. Pollution plumes observed during PEM-Tropics-A Mission 6 (Tahiti), 5 September 1996. (top) Horizontal flight at 10 km altitude (23–30°S, 155°W); (bottom) Descent at 30°S, 155°W.

which had been steady at  $-42^{\circ}$  to  $-43^{\circ}\text{C}$  began a slight decrease. The observed gradients in wind speed, temperature and humidity at 25–26°S identified a minor airmass boundary. Trajectories relevant to the data of M6 published by Fuelberg et al. (1999) suggest that this airmass boundary existed between two sets of trajectories that (1) carried moist air and cirrus clouds from possible thunderstorms in the equatorial areas near and north of the Fiji Islands toward the area of M6 (equatorward of 25°S) and (2) carried dry (subsiding) air parcels toward the area of longitude 155°W and south of 25°S along strong westerly winds from across Australia and the Indian Ocean. The latter trajectories have been shown to be associated with biomass-burning pollution plumes (Chatfield et al., 1999) such as shown in Fig. 6a starting at 26°S.

After encountering the pollution plume at 10 km near 26°S the DC-8 continued southward. Ozone and CO remained above 80 ppb over a horizontal distance of about 220 km (26–28°S) after which mixing ratios began to decrease reaching values of 20–30 ppb for  $\text{O}_3$  and 50–60 ppb for CO near 30°S. At 30°S 155°W, the DC-8 started a descent into the marine boundary layer. The

descent profiles of  $\text{O}_3$ , CO, and water vapor, based on 1-min data, are shown in Fig. 6b. A cloud layer associated with a local weather system was present from 10 km to about 7 km. An  $\text{O}_3$  plume of 1 km thickness and 65 ppb peak mixing ratio was observed near the cloud base embedded in a broad CO plume of 77 ppb peak mixing ratio. A layer of dry air (100 ppm water vapor) was subsequently observed from 6 to 4 km associated with a 70 ppb secondary CO plume and a lamina of 60 ppb  $\text{O}_3$  just below. Thermal lapse rate revealed a stable troposphere. At lower altitudes, more clouds and a moist boundary-layer were associated with a marked decrease in CO and  $\text{O}_3$  to near background levels (50 and 30 ppb, respectively). The nadir-looking DC-8 DIAL identified the location and magnitude of the two  $\text{O}_3$  peaks, one near 5 km and the other at 7 km (Fenn et al., 1999). Widespread biomass burning activity was found to be present in South America, Central Africa and Australia (Olson et al., 1999). The broad CO plume of Fig. 6b extending from about 5–8 km with mixing ratios exceeding 65 ppb has been further analyzed by Chatfield et al. (1999). Their computer simulations suggest that it was associated with long-range trajectories that originated near the southern coast of Africa and as far as the southern tip of Brazil. Although the mid and upper troposphere have a complex layered structure and CO and  $\text{O}_3$  are not identical tracers of biomass burning, it is concluded that the plumes evident in the data of Fig. 6b are primarily the result of distant biomass burning. The marine boundary layer shown in Fig. 6b is characterized by low mixing ratios of  $\approx 30$  ppb  $\text{O}_3$  and  $\approx 50$  ppb CO. In general, the boundary layer was only minimally influenced by pollution intrusions because of the presence of a strong temperature inversion.

Fig. 7 shows vertical profiles of the mean mixing ratio of several principal species using the average of all data from M6 and data from M12 and M14 between 15°S and 30°S. These flights were clearly influenced by the presence of pollution plumes. A relative maximum in the mixing ratios of many species at 4–8 km altitude is evident. Methylchloride, a tracer of biomass burning pollution, is elevated along with other less specific tracers such as CO. Sharp maximum was also present for secondary photochemical species such as PAN,  $\text{O}_3$  and  $\text{HNO}_3$ . Chemicals such as  $\text{C}_2\text{Cl}_4$  (characteristic of urban pollution) were at their expected background levels.

Another property of the observed plumes was that they were most frequently embedded in very dry air (Fig. 6). From a total of 100 CO measurements (1-min average) larger than 100 ppb collected in the area from the equator to 30°S, 73% were associated with dry air containing less than 1000 ppm water vapor. Out of these 73 observations, 46 observations (63%) were associated with very dry air of less than 200 ppm water vapor. These data are in accordance with the subsiding nature of the long-range trajectories of the pollution plumes. The



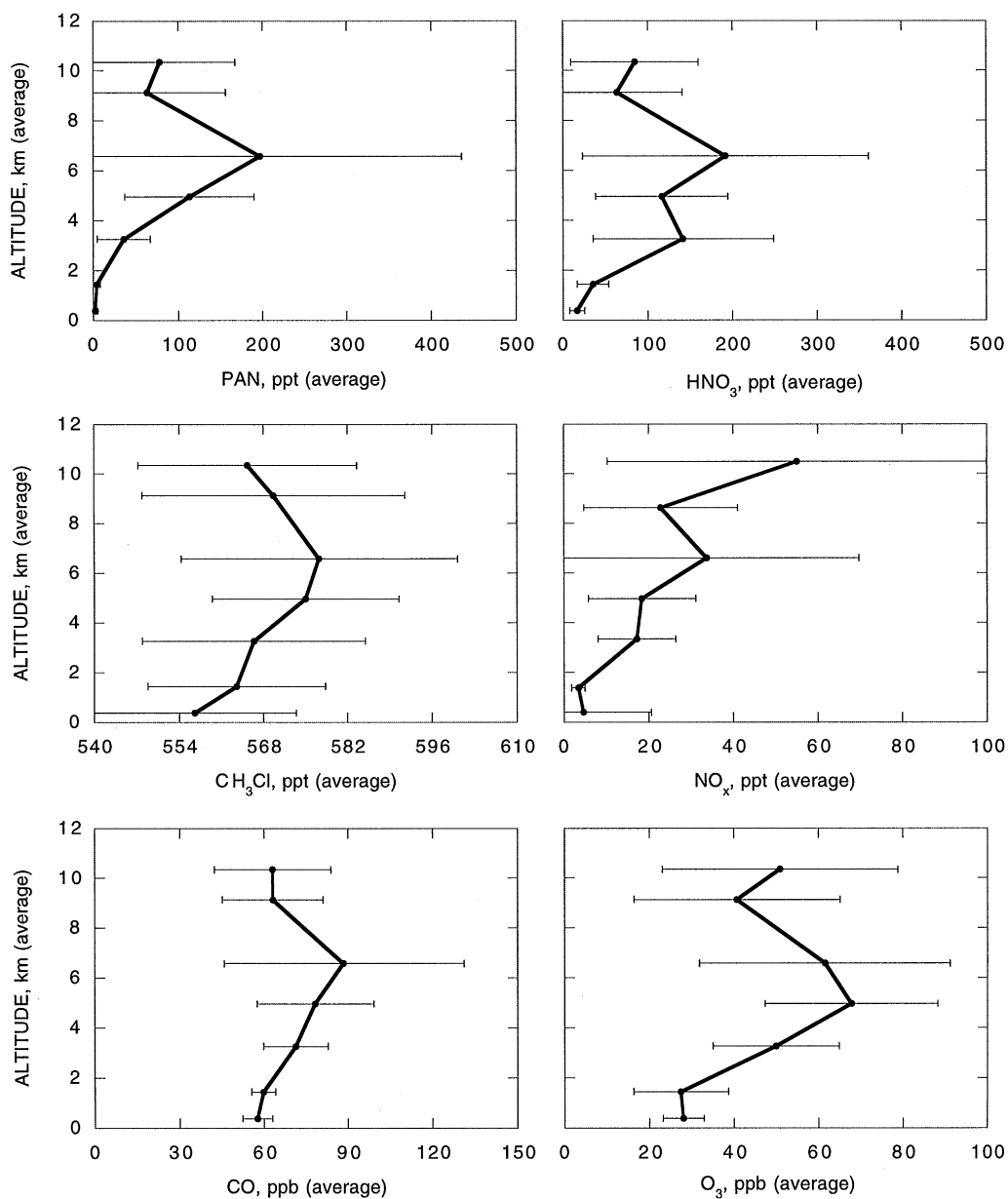


Fig. 7. Vertical profiles of tracers and reactive nitrogen species using the average of all data from PEM-Tropics-A Missions 6, 12, and 14 for the tropical troposphere between 15°S and 30°S.

large, extremely dry plumes should have unique photochemical and radiative properties that deserve further study.

In a recent study, Mauzerall et al. (1998) examined the chemical characteristics of biomass burning plumes observed during TRACE-A. They calculated an enhancement ratio (ER),  $\Delta Y/\Delta X$  where  $Y$  is a tracer of interest and  $X$  is a reference tracer such as CO or CO<sub>2</sub>, as a function of time to determine plume age (fresh, recent, aged, and

old). For example, their mean ER for the regression of C<sub>2</sub>H<sub>2</sub> versus CO based on the analysis of a large number of plumes is 2.8 (ppt/ppb) for old plumes and 4.2 for a fresh plume. The decrease of ER with time is the result of the greater dilution with time for the selected tracer of interest than for the long-lived reference tracer CO. We calculated similar ER for a number of selected chemicals measured in PEM-Tropics-A. Fig. 8 shows an example of the linear relationship found between C<sub>2</sub>H<sub>2</sub> and PAN

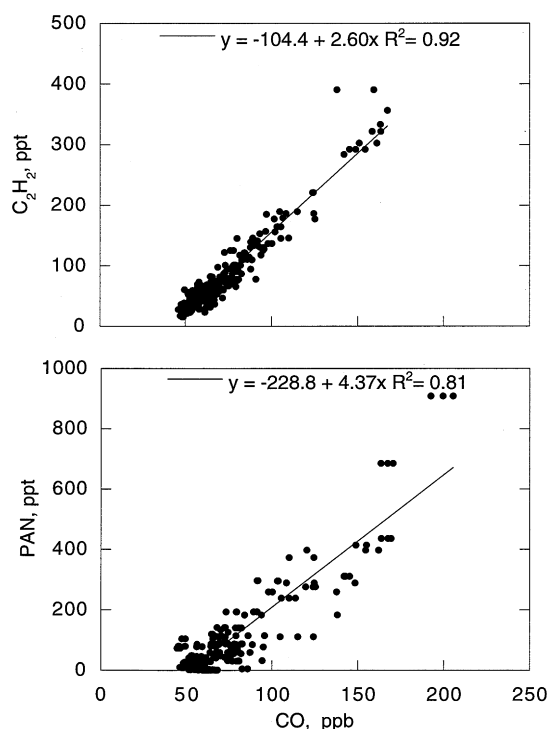


Fig. 8. Examples of linear correlation between selected tracers and the reference tracer CO using plume-specific data from PEM-Tropics-A missions 6, 12, and 14 (10–30°S). Regression slopes identify enhancement ratios.

with that of CO using specific plume-related tracer data from M6, 12, and 14 between 15°S and 30°S. Table 1 summarizes the ER (slope of the regression curves) calculated from correlations such as shown in Fig. 8 and also presents the values determined by Mauzerall et al. (1998) for characteristic old biomass-burning plumes for the TRACE-A region. The ER values for several species

such as CH<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>6</sub>, CH<sub>3</sub>Cl, PAN, O<sub>3</sub> are positive and comparable to those determined by Mauzerall et al. (1998), further suggesting that these plumes had the characteristics of emissions from biomass burning sources. There is no linear relationship seen between the abundance of peroxides (H<sub>2</sub>O<sub>2</sub>, CH<sub>3</sub>OOH) and CO in the PEM-Tropics-A data. This is likely due to their complex photochemical production and destruction mechanisms along with their high selective removal via wet chemical processes during long-range transport. The ER for NO<sub>x</sub> (Table 1) is also quite different from that obtained by Mauzerall et al. (1998) for old biomass-burning plumes. This is expected as NO<sub>x</sub> in the PEM-Tropics-A study region was greatly influenced by additional sources such as lightning and by recycling from PAN and HNO<sub>3</sub> reservoir species (Schultz et al., 1999).

### 3. Conclusions

Chemical tracers of continental combustion and biomass burning collected during PEM-Tropics-A are analyzed to show that distinct pollution plumes were frequently present in the upper and middle troposphere of the remote tropical South Pacific. The observed pollution plumes originated from the biomass burning areas of South Africa and South America. It appears that biomass burning pollution is easily lofted into the free troposphere in and near its source areas and subsequently transported throughout the Southern Hemisphere in the form of relatively broad plumes. While ozone and tracer levels in the free troposphere are strongly impacted, the marine boundary layer is only minimally influenced because of the strong temperature inversion. Many of the plumes were extremely dry and may have unique photochemical and radiative properties that should be studied further.

Table 1  
Enhancement ratios (ER) derived from PEM-Tropics-A and TRACE-A measurements

Enhanced ratios	PEM-Tropics-A ER ( $R^2$ ) <sup>a</sup>	TRACE-A ER $\pm$ SE <sup>b</sup>
$\Delta$ O <sub>3</sub> / $\Delta$ CO (ppb/ppb)	0.75 (0.56)	0.74 $\pm$ 0.90
$\Delta$ CH <sub>4</sub> / $\Delta$ CO (ppb/ppb)	0.33 (0.39)	0.44 $\pm$ 0.12
$\Delta$ CH <sub>3</sub> Cl/ $\Delta$ CO (ppt/ppb)	0.59 (0.46)	1.43 $\pm$ 0.13
$\Delta$ NO <sub>x</sub> / $\Delta$ CO (ppt/ppb)	0.47 (0.18)	3.3 $\pm$ 1.4
$\Delta$ PAN/ $\Delta$ CO (ppt/ppb)	4.37 (0.81)	6.0 $\pm$ 0.3
$\Delta$ H <sub>2</sub> O <sub>2</sub> / $\Delta$ CO (ppt/ppb)	1.97 (0.01)	20.8 $\pm$ 17.8
$\Delta$ CH <sub>3</sub> OOH/ $\Delta$ CO (ppt/ppb)	-2.27 (0.02)	10.6 $\pm$ 7.1
$\Delta$ C <sub>2</sub> H <sub>2</sub> / $\Delta$ CO (ppt/ppb)	2.60 (0.92)	2.8 $\pm$ 0.2
$\Delta$ C <sub>2</sub> H <sub>6</sub> / $\Delta$ CO (ppt/ppb)	8.08 (0.88)	8.0 $\pm$ 0.7

<sup>a</sup>Data from missions 6, 12 and 14 (< 30°S).

<sup>b</sup>Mean ER and standard error (SE) for old plumes from Mauzerall et al. (1998).

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## References

- Blake, N.J., Blake, D.R., Wingenter, O., Anderson, B., Gregory, G., Sachse, G., Kok, G., Rowland, F.S., 1999. Influence of southern hemispheric biomass burning on the south Pacific mid-troposphere: results from PEM-Tropics A and ACE-1. *Journal of Geophysical Research* 104, 16213–16232.
- Bradshaw, J., et al., 1999. Photofragmentation two-photon laser-induced fluorescence detection of NO<sub>2</sub> and NO: comparison of measurements with model results based on airborne observations during PEM-Tropics-A. *Geophysical Research Letters* 26, 471–474.
- Brunner, D.W., 1998. One-year climatology of nitrogen oxides and ozone in the tropopause region—results from B-747 aircraft measurements. Diss. ETH No. 12556, the Swiss Federal Institute of Technology, Zurich, 1998, submitted.
- Chatfield, R.B., Guo, Z., Sachse, G.W., Blake, N., Blake, D., Browell, E., 1999. Mechanism of the Southern Global Plume: how tropical burning plumes affect the remote Pacific. *Journal of Geophysical Research*, submitted for publication.
- Chatfield, R.B., Vastano, J., Singh, H., Sachse, G., 1996. A general model of fire emissions and chemistry African/oceanic plumes (O<sub>3</sub>, CO, PAN, smoke) in TRACE A. *Journal of Geophysical Research* 101, 24279–24306.
- Crutzen, P.J., Andreae, M.O., 1990. Biomass burning in the tropics: Impact on atmospheric chemistry and biogeochemical cycles. *Science* 250, 1669–1678.
- Davis, D.D., et al., 1996. An assessment of the photochemical ozone tendency in the western north Pacific. *Journal of Geophysical Research* 101, 2111–2134.
- Dibb, J., et al., 1996. Asian influence over the western north Pacific during the fall season: inferences from Pb-210, soluble species and ozone. *Journal of Geophysical Research* 101, 1779–1792.
- Fenn et al., 1999. Ozone and aerosol distribution and air mass characteristics over the south Pacific during the burning season. *Journal of Geophysical Research* 104, 16197–16212.
- Fishman, J., et al., 1991. Identification of widespread pollution in the southern hemisphere deduced from satellite analysis. *Science* 252, 1693–1696.
- Fishman, J., et al., 1996. The NASA GTE TRACE-A experiment (September–October, 1992): overview. *Journal of Geophysical Research* 101, 23865–23879.
- Fuelberg, H.E., Newell, R.E., Longmore, S., Zhu, Young, Westberg, D.J., Browell, E.V., Blake, D.R., Gregory, G.L., Sachse, G.W., 1999. A meteorological overview of the PEM-Tropics period. *Journal of Geophysical Research* 104, 5585–5622.
- Heikes, B., et al., 1996. Hydrogen peroxide and methylhydroperoxide distributions related to ozone and odd hydrogen over the north Pacific in the fall of 1991. *Journal of Geophysical Research* 101, 1891–1905.
- Hoell, J.M., Davis, D., Jacob, D., Rodgers, M., Newell, R., Fuelberg, H., McNeal, R., Raper, J., Bendura, R., 1999. Pacific exploratory mission in the tropics; September 1996. *Journal of Geophysical Research* 104, 5567–5583.
- Jacob, D.J., 1996. Origin of ozone and NO<sub>x</sub> in the tropical troposphere: A photochemical analysis of aircraft observations over the south Atlantic basin. *Journal of Geophysical Research* 101, 24235–24250.
- Lacaux, J., 1992. Precipitation chemistry in the Mayombe forest of equatorial Africa. *Journal of Geophysical Research* 97, 6195–6206.
- Mauzerall, D.L., Logan, J., Jacob, D., Anderson, B., Blake, D., Bradshaw, J., Heikes, B., Sachse, G., Singh, H., Talbot, R., 1998. Photochemistry in biomass burning plumes and implications for tropospheric ozone over the tropical south Atlantic. *Journal of Geophysical Research* 103, 8401–8423.
- McNeal, R.J., Mugler Jr., J.P., Harriss, R.C., Hoell Jr., J.M., 1983. NASA global tropospheric experiment. *Eos Transactions AGU* 64, 561–562.
- Olson, J.R., Baum, B.A., Cahoon, D.R., Crawford, J.H., 1999. Frequency and distribution of forest, savanna, and crop fires over tropical regions during PEM-Tropics A. *Journal of Geophysical Research* 104, 5865–5876.
- O'Sullivan, D.W., Heikes, B.G., Lee, M., Chang, W., Gregory, G.L., Blake, D.R., Sachse, G.W., 1999. Distribution of hydrogen peroxide and methylhydroperoxide over the Pacific and South Atlantic ocean. *Journal of Geophysical Research* 104, 5635–5646.
- Schultz, M., et al., 1999. Origin of Tropospheric Ozone and NO<sub>x</sub> Over the Tropical South Pacific. *Journal of Geophysical Research* 104, 5829–5843.
- Smyth, S., et al., 1996. Comparison of free tropospheric western Pacific air mass classification schemes for the PEM-West A experiment. *Journal of Geophysical Research* 101, 1743–1762.
- Singh, H.B., et al., 1996. Impact of biomass burning emissions on the composition of the south Atlantic troposphere. *Journal of Geophysical Research* 101, 24203–24219.
- Singh, H.B., et al., 1998. Latitudinal distribution of reactive nitrogen in the free troposphere over the Pacific ocean in late winter/early spring. *Journal of Geophysical Research* 103, 28237–28246.
- Talbot, R., et al., 1996. Chemical characteristics of continental outflow from Asia to the western Pacific ocean during September–October 1991: results from PEM-West A. *Journal of Geophysical Research* 101, 1713–1726.
- Thompson, A., et al., 1996. Ozone over southern Africa during SAFARI-92/TRACE A. *Journal of Geophysical Research* 101, 23739–23807.