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# **Chemical characteristics of Pacific tropospheric air in the region of the Intertropical Convergence Zone and South Pacific Convergence Zone**

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**Abstract. The Pacific Exploratory Mission (PEM)-Tropics provided extensive aircraft data to study the atmospheric chemistry of tropospheric air in Pacific Ocean regions, extending from Hawaii to New Zealand and from Fiji to east of Easter Island. This region, especially the tropics, includes some of the cleanest tropospheric air of the world and, as such, is important for studying atmospheric chemical budgets and cycles. The region also provides a sensitive indicator of the global-scale impact of human activity on the chemistry of the troposphere, and includes such important features as the Pacific "warm pool," the Intertropical Convergence Zone (ITCZ), the South Pacific Convergence Zone (SPCZ), and Walker Cell circulations. PEM-Tropics was conducted from August to October 1996. The ITCZ and SPCZ are major upwelling regions within the South Pacific and, as such, create boundaries to exchange of tropospheric air between regions to the north and south. Chemical data obtained in the near vicinity of the ITCZ and the SPCZ are examined. Data measured within the convergent zones themselves are not considered. The analyses show that air north and south of the convergent zones have different chemical signatures, and the signatures are reflective of the source regions and transport histories of the air. Air north of the ITCZ shows a modest urban/industrialized signature compared to air south of the ITCZ. The chemical signature of air south of the SPCZ is dominated by combustion emissions from biomass burning, while air north of the SPCZ is relatively clean and of similar composition to ITCZ south air. Chemical signature differences of air north and south of the zones are most pronounced at altitudes below 5 km, and, as such, show that the ITCZ and SPCZ are effective low-altitude barriers to the transport of tropospheric air. At altitudes of 8 to 10 km, chemical signatures are less dissimilar, and air backward trajectories (to 10 days) show cross-convergent-zone flow. At altitudes below about 5 km, little cross-zonal flow is observed. Chemical signatures presented include over 30 trace chemical species including ultrafine, fine, and heated-fine (250øC) aerosol.** 

### **1. Introduction**

**The Pacific Exploratory Mission (PEM)-Tropics provided extensive aircraft data to study the atmospheric chemistry of** 

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**tropospheric air in the central and eastern Pacific Ocean regions. This region of the Pacific extending from Hawaii to New Zealand (20øN to 45øS latitude) and from Fiji to east of Easter Island (175øE to 110øW longitude) is a data sparse region in terms of tropospheric air chemistry data. Existing data are limited to the Global Atmospheric Measurements Experiment on Tropospheric Aerosols and Gases (GAMETAG) aircraft missions of 1977 and 1978 [Davis, 1980], the Stratospheric Ozone Experiment (STRATOZ III) mission [Marenco, 1988], scattered ozone sonde releases from selected sites (e.g., Samoa and New Zealand), and intermittent land-based or ship-based surface observations. This region, especially the tropics, is one of the cleaner regions of the world and, as such, is important for studying atmospheric chemical budgets and cycles. It also provides a sensitive indicator of the global-scale impact of human activity on the chemistry of the troposphere. The region includes such features as the Pacific "warm pool," the Intertropical Convergence Zone, the South Pacific Convergence Zone, and important Walker Cell circulations.** 

The PEM-West A and B missions conducted in 1991 and **1994 (Journal of Geophysical Research, 101, 1641-2147, 1996; 102, 28,223-28,671, 1997) studied the northern Pacific Ocean regions with a focus on understanding the importance of Asian** 

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Figure 1. PEM-Tropics study area and major meteorological features.

outflow to tropospheric chemistry during periods of maximum impact from Asian outflow (northern hemisphere winter and PEM-West B) and less dominant outflow (summer, PEM-West A). Results show that North Pacific region is no longer pristine and free of major influences from anthropogenic emissions and that the impact of Asian outflow on tropospheric air is sizable and dominant. PEM-West A and B and PEM-Tropics data are in the public domain and are available from the NASA Langley Research Center via the Distributed Active Archive Center (DAAC) and from the Global Tropospheric Experiment (GTE) Wide World Web site at html://www-gte.larc.nasa.gov.

The PEM-Tropics experiment was conducted during the southern hemisphere winter/spring (August, September, and October, 1996) and utilized both the NASA DC-8 and P3-B aircraft in a coordinated effort to study the South Pacific region. The aircraft operated from multiple staging areas: Tahiti (both aircraft); Easter Island (both); Christmas Island (P3-B); Guayaquil, Ecuador (P3-B); Fiji (DC-8); and Christ Church, New Zealand (DC-8). Both aircraft ferried to the region via Hawaii. Typically, each aircraft flew two to three missions ( $\approx$ 7 hour duration) from a staging area and included flight altitudes to about 13 km (DC-8) and 8 km (P3-B) above mean sea level. The PEM-Tropics program objectives, participants, suite of instruments, and experimental approaches are discussed in the overview paper by Hoell et al. [this issue]. A meteorological overview of the mission is given by *Fuelberg et al.* [this issue]. Figure 1 shows the PEM-Tropics study area and illustrates the meteorological features important to regional air flow during the southern hemisphere winter/spring. The shaded circles in the figure indicate the aircraft staging areas. The two features of importance to our analyses are the Intertropical Convergence Zone (ITCZ) and the South Pacific Convergence Zone (SPCZ).

The ITCZ and SPCZ are major upwelling regions within the

South Pacific troposphere and, as such, create boundaries to exchange of tropospheric air between regions to the north and south. Based upon the dynamics of formation of the ITCZ and SPCZ, a convergence of low-altitude air from different source areas, differences in the chemical composition of tropospheric air north and south of each zone are anticipated. However, the chemical composition of air, the relative magnitude and persistence of concentration differences for major trace chemical species, and the importance of natural emissions and human activities on the chemical composition of air north and south of the ITCZ and SPCZ are not well known. In addition, the effective vertical height of the ITCZ and SPCZ in terms of limiting mass transport is not known. Our analyses address these issues. In the study we examine the chemical composition of tropospheric air as a function of altitude on each side (north versus south) of the convergent zones using data from portions of DC-8 flights which crossed the ITCZ and SPCZ. In addition, back trajectories for air sampled are examined to identify potential source areas. The degree to which the chemical composition of air north and south of the ITCZ and SPCZ differs is an indicator of the effectiveness of the zone as a transport barrier. Substantially different chemical signatures not only indicate an effective barrier, but also suggests that the air north and south of the zone are from different sources. The data used in the study are restricted to measurements in the nearvicinity of the zones and excludes data measured within the ITCZ and SPCZ.

#### **ITCZ and SPCZ Meteorology** 2.

The ITCZ and SPCZ are discussed by numerous authors including Wallace and Hobbs [1977], Merrill [1989], Hastenrath [1991], Holton [1992], Vincent [1994], and Fuelberg et al. [this issue]. Our discussion is a brief synopsis of the literature high-



Figure 2. The 1996 PEM-Tropics flight tracks: NASA DC-8 aircraft. Circled flight numbers identify data used in the analyses.

lighting points important to our analyses. Discussions are referenced to Figure 1.

## 2.1. ITCZ

The ITCZ is an intense band of convection which provides a meteorological boundary between northern and southern Pacific tropospheric air. Meandering between 2° and 12°N latitude, the ITCZ roughly parallels the equator as it transverses the Pacific Ocean. The convergence of clockwise flow around the North Pacific subtropical high at 30°N and counterclockwise flow around the South Pacific subtropical high at 30°S produces a persistent easterly flow along both sides of the ITCZ. North of the ITCZ, the prevailing winds frequently exhibit a northerly component, while south of the ITCZ, they exhibit a southernly component. The degree of northerly or southernly component depends upon the location and strength of the subtropical highs. Vertically, the ITCZ is well-identified at altitudes up to 3 or 4 km, becoming less distinct at higher altitudes. Although the major airflow component is easterly along the ITCZ, cross-zonal flow does occur. Offshore Asian airflow will penetrate deep into Australia during January, while in July, Australian airflow will penetrate as far north as southern Asia.

In the North Pacific  $(30^{\circ}$  to  $50^{\circ}$ N), tropospheric air is basically flow from the west in which transport of continental emissions from Asia reach the tropics by the westerlies as flow around the eastern side of the North Pacific subtropical high. Once air comes under the influence of the subtropical high, general subsidence causes the air to descend as it travels toward the tropics and the ITCZ. A heterogeneous airflow is not uncommon and exists when a component of North American emissions is also added on the eastern side of the high.

### 2.2. SPCZ

The SPCZ is a northwest-southeast oriented zone of convergence terminating in the south central Pacific near 30°S and 130°W and tends to merge with the ITCZ near the equator at 140°E. It is maintained by the intrusions of midlatitude air masses from Australia and Antarctic during cyclogenesis to the east of Australia colliding with the persistent easterly trades coming across the Pacific Ocean. The portion of the SPCZ from New Guinea to just west of Tahiti contains the more established, persistent, and expansive cloud bands as compared to the portion east of Tahiti. This established portion of the SPCZ is the focus of our analyses. North of the SPCZ, easterly flow dominates from the surface to several kilometers altitude, above which the flow switches to westerly. South of the SPCZ, lower and midtropospheric air flow is from the west and, at times, exhibits a southernly component having passed over Indonesia, New Zealand, and/or Australia.

#### 3. **Approach to Analyses**

The purpose of the analyses is to examine and compare the chemical composition of tropospheric air on the north and south side of the ITCZ and SPCZ, respectively. Only data from the DC-8 flights are used in the analyses. Figure 2 shows the PEM-Tropics DC-8 flight tracks where data from circled flight numbers are used in the analyses. Flights 4, 5, and 19 provide data for the ITCZ analyses, and flights 15, 16, and 17 provide data for the SPCZ analyses. Figures 3 and 4 show the DC-8 flight tracks for the ITCZ and SPCZ flights, respectively. The flight tracks are overlaid on surface (1000 hPa level) winddivergence maps which are used to locate the ITCZ and SPCZ. relative to the aircraft track. The wind-divergent maps are computed from grid point data supplied by the European Centre for Medium-Range Weather Forecasts (ECMWF). The locations of the ITCZ and SPCZ used in the analyses are indicated on the figures. Flights 4 and 19 (ferry flights) crossed the ITCZ. Flight 5 (staged from Tahiti) did not cross the ITCZ; however, data from the northern leg are included in the south ITCZ case. The three SPCZ flights were staged from Fiji to cross the SPCZ and provide altitude data on each side of the SPCZ.

Table 1 summarizes the bounds for the data groupings. Using the 1000 hPa surface wind-divergent maps and the super-



**Figure 4. SPCZ analyses: surface (1000 hPa level) winddivergence maps. The location of the ITCZ and SPCZ (dashed lines) are shown on each map as well as the DC-8 flight track (solid line). (a) Flight 15 (September 26, 1996), (b) flight 16 (September 28, 1996), and (c) flight 17 (October 1, 1996).** 

**Longitude** 

**Longitude** 

**160W** 

**ITCZ** 

**SPCZ** 

**160E** 

**160E 160W 120W Longitude** 

**ITCZ** 

**120W** 

**SPCZ** 

**SPCZ** 

**Figure 3. ITCZ analyses: surface (1000 hPa level) winddivergence maps. The location of the ITCZ and SPCZ (dashed lines) are shown on each map as well as the DC-8 flight track (solid line). (a) Flight 4 (August 31, 1996), (b) flight 5 (September 5, 1996), and (c) flight 19 (October 5, 1996).** 





**imposed flight track (i.e., data of Figures 3 and 4), the location of the aircraft at crossing of the ITCZ and SPCZ are determined. Using these locations, UV-DIAL [Browell et al., 1996] remotely sensed aerosol data from the DC-8 are used to estimate the extent of the ITCZ and SPCZ cloud zones which, in turn, are taken to represent the boundaries of the ITCZ and**  SPCZ after allowing for a margin of uncertainty of 1<sup>°</sup> of lati**tude. Data north and south of the zones are binned for chemical analyses. For the north ITCZ case a northern boundary**  limit (each flight) of 7° north of the northern boundary of the **ITCZ is used. For the south ITCZ data a southern boundary limit is placed at 5øS latitude. For the north SPCZ group, data from flight 15 are excluded as that portion of the flight paralleled the SPCZ (see Figure 2). For the south SPCZ group, data**  south of 20°S are excluded as well as 30 min of data associated **with takeoffs and landings at Fiji.** 

**The results of the chemical analyses are given in Tables 2 and 3. Results are binned by altitude and reported as median and quartile values (see footnotes of the tables). Gaseous specie data are reported in "per volume units" as, for example, parts-per-trillion by volume (pptv). Only entries with 3 or more observations are included, and missing entries are coded as -9. Altitude bins are 0 to 1, 1 to 2, 2 to 4, 4 to 8, 8 to 10, 10 to 12, and >12 km. For the SPCZ analyses the 0 to 1 km bin is subdivided into 0 to 0.5 km and 0.5 to 1 km bins. The altitude bins of 10 to 12 km and >12 km provide limited data and are not included in the tables. Species (identified by generic and chemical formula names) are grouped according to overall general interest or type of compound (see table footnotes). The second column entry of each table is a code which indicates whether the north (N) or south (S) data exhibits the higher concentration for the species. It was not necessary that an increase occur in all altitude bins for the code to be assigned. In general, assignment of the code was weighted to the altitude bins of 1-2 and 2-4 km. A blank entry in the code column means that no definitive conclusion between the groups are made, and thus for our purposes, specie concentrations are judged to be similar. Species not included in the table (see Hoell et al. [this issue] for a complete listing of DC-8 measurements) either (1) do not have sufficient observations for our altitude-binned analyses, (2) are data at or near measurement detection limits, or (3) do not include data for both a north and south group. Generally, the shorter-lived (e.g., hours) compounds like propene and ethene fall into the lower detection limit category. The reader is directed to Hoell et al. [this issue] for a description of the measurement techniques and measurement accuracies. Instrument and measurement details are also found in companion papers which discuss specific chemical species. Discussion in the body of the paper focus only on a few of the key species of Tables 2 and 3.** 

**Before discussing the results, two comments are necessary. First, the data used for the analyses are only those from the DC-8 flights, are restricted to locations in the near vicinity of the ITCZ and SPCZ, and represent about 10% of the DC-8 data. Larger chemical data bases can be constructed which may be more representative of the overall chemical signature of regional air. For example, south of the SPCZ a larger database (particularly for the acids, peroxides, and sulfur species) can be constructed which includes additional data from portions of flights 6, 12, 14, and possibly 18. Companion papers [e.g., O'Sullivan et al., this issue; Talbot et al., this issue; Thornton et al., this issue; Vay et al., this issue; E. Atlas et al., unpublished manuscript, 1998; K. B. Bartlett et al., unpublished manuscript, 1998] address regional chemical issues for many chemical tracers and use larger databases, including combining DC-8 and P3-B data.** 

**Second, while not critical to our conclusions, we cite chemical ratios to provide an approximate "age" of air from emission sources. These ratios should only be used qualitatively as it is recognized that "atmospheric processing" and "photochemical age" both affect these ratios [McKeen et al., 1996], and the two processes are not separated in the analyses. In selecting and using chemical ratios we considered the decay rate of various chemical ratios. For example, an emission ratio of 10 at the source for ethyne/carbon monoxide decays to 4 (after 3 days), 1.5 (after 6 days), and <0.5 (after 10 days), assuming ethyne and carbon monoxide half-lives are approximately 2 days and 1 month, respectively. Considering (1) the above decays, (2) the ocean location of the sampling (removed from immediate land sources), and (3) the information from references [e.g., Greenberg and Zimmerman, 1984; Greenberg et al., 1990; Singh and Zimmerman, 1992], we approximate the photochemical lifetimes in the tropics for ethene and propene (hours), ethyne and propane (days), and ethane and carbon monoxide (weeks) and broadly use the ethyne/carbon monoxide (hereafter, ethyne/CO) ratio in units of pptv/ppbv as an emission source "age" indicator. Ratios of about <0.5, 1, and 3 are interpreted as representative of air aged approximately 10 days, 5 to 7 days, and a few days, respectively, from an emission source.** 

### **4. Results**

### **4.1. Intertropical Convergence Zone (ITCZ)**

**Figures 5-7 and Table 2 summarize the ITCZ chemical results. Data in the figures are plotted at the midpoint of the altitude bin and the boundaries of the boxes are at the lower and upper quartile values. For clarity of presentation, south data boxes are offset 0.25 km in altitude. Lines drawn between the boxes are at the medians of the data; the solid line is for the** 



Table 2. PEM-Tropics ITCZ Comparison: North Versus South



Group	Code	Specie		0 to 0.5 km Altitude				0.5 to 1 km Altitude			
			Zone	Median	<b>UQuar</b>	LOuar	${\bf N}$	Median	<b>UQuar</b>	LQuar	N
$\mathbf{1}$		North latitude, deg	North South	$-1.1$ $-21.5$	$-0.5$ $-21.3$	$-4.6$ $-21.8$	2355 729	$-1.4$ $-21$	$-0.1$ $-21$	$-4.2$ $-22$	405 134
		East longitude, deg	North South	186.5 175	186.8 175	175 175	2355 729	186.3	187	175.1	405
		Wind speed, kt (from	North	13	17	11	2098	175 $-9$	175.1 $-9$	175 $-9$	134 0
		$DC-8$ Wind direction, deg (from	South North	16 78	17 94	15 68	659 2098	$-9$ $-9$	$-9$ $-9$	-9	0
		$DC-8$	South	92	96	89	659	-9	$-9$	$-9$ $-9$	$\boldsymbol{0}$ $\bf{0}$
		Air temperature, °C	North South	24 19	25.6 19.2	23.8	2355	21.1	22	20.6	405
		Dew point temperature, °C	North	$-9$	$-9$	18.8 -9	729 $\bf{0}$	15.1 $-9$	15.8 $-9$	14.1 -9	134 $\boldsymbol{0}$
$\overline{c}$	S	(project composite)	South	14.8	15.3	14.5	729	13.5	14.1	13.2	134
		Ozone $(O3)$ , ppbv	North South	13.7 38.4	14.7 38.9	12 38	2355 729	13.7 39.7	16.3 40.2	11.2 38.9	405 134
	S	Carbon monoxide (CO),	North	62.1	63	55.1	385	61.4	65.2	54.9	80
		ppby Methane (CH4), ppbv	South North	83.1 1704.6	83.8 1705.9	82 1701.1	116 385	83.9 1702.3	84.2 1705.6	82.7 1700.1	28 80
			South	1701.1	1701.6	1700.5	116	1702.9	1703.3	1699.7	28
		Carbon dioxide (CO2), ppmv	North South	361.5 360.8	361.5 360.8	361.4 360.7	345 116	361.5 360.9	361.5 360.9	361.2 360.7	80
3	S	Ethane (C2H6)	North	271.3	300.4	234.1	12	240.6	274.5	229.4	28 $\overline{7}$
		Ethene (C2H4)	South North	403	423.1	400.4	$\overline{\mathbf{4}}$	407.2	418.5	199.8	3
			South	3.3 3.1	3.8 3.3	2.5 2.8	12 4	3 2.6	3.8 3	2.3 1.3	$\overline{\mathcal{I}}$
	S	Ethyne (C2H2)	North	13	15.3	11.1	12	15.4	17.5	10.6	$\frac{3}{7}$
	S	Propane (C3H8)	South North	93.3 10.9	95 13.2	86.4 9.6	4 12	93.4 12.9	98.9 13.4	41.6 4.9	$\frac{3}{7}$
4			South	29.7	39	25.7	4	30.6	32	14.2	$\frac{3}{7}$
	S	Propane/ethane, pptv/pptv	North South	0.042 0.074	0.046 0.092	0.034 0.064	12 $\overline{\mathbf{4}}$	0.051 0.075	0.053 0.08	0.021 0.033	3
	S	Ethyne/CO, pptv/ppbv	North	0.216	0.23	0.206	9	0.248	0.31	0.192	$\boldsymbol{7}$
		Ethene/CO, pptv/ppbv	South North	1.116 0.057	1.15 0.06	1.048 0.046	$\overline{\mathbf{4}}$ 9	1.091 0.055	1.152 0.064	0.507 0.039	3 $\overline{7}$
			South	0.036	0.039	0.034	4	0.032	0.036	0.015	3
		Ethane/CO, pptv/ppbv	North South	4.612 4.9	4.729 5.1	4.339 4.8	9 4	4.405 4,9	4.644 4.9	4.177	$\boldsymbol{7}$
5		CFC-22 (C H Cl F2)	North	120.1	123.9	117.2	12	124.1	126.8	2.4 119.7	3 $\overline{7}$
		CFC-12 (C Cl2 F2)	South North	118.2 528.2	123.5 529.5	114.5	4	117	119.1	56.5	3
			South	526.2	527.4	524.6 525.7	12 4	528 525.7	529.7 527.2	522.2 262.1	$\overline{7}$
		CFC-11 (C Cl3 F)	North	266.3	268	265	12	266	267	263	$\frac{3}{7}$
		CFC-113 (C Cl2 F	South North	264.6 83	267.6 83.5	264.3 82.4	4 12	264 82.7	265 83.2	131.3 82.3	3 $\overline{7}$
		C CI F2)	South	82.8	83.2	82.5	$\overline{4}$	82.9	83.1	41.3	3
		CFC-114 (C CI F2 C CL F2)	North South	14.1 14. I	14.2 14.2	14 14	12 4	14.1 14.1	14.1 14.1	13.9 7	7 3
		Tetra-chloro-ethylene	North	1.2	1.3	1.2	7	1.2	1.3	1.1	$\overline{\mathcal{L}}$
	S	(C2 Cl4) Methyl chloride (CH3 Cl)	South North	1.4 547	1.5 560	1.4 535	$\overline{4}$ 12	1.4 557	1.4 567	0.6 545.5	3 $\boldsymbol{7}$
			South	570.5	578	565	4	572	573	284.5	3
6	S	Nitric oxide (NO)	North South	1.7 1.7	1,8 $\overline{2}$	1.5 1.3	15	1.6 2.2	1.6	1.5	5
	N	Sulfur dioxide (SO2)	North	56.9	84.8	54.5	4 6	-9	3.9 $-9$	1.8 -9	$\mathfrak{Z}$ $\overline{\mathbf{c}}$
	N	Dimethyl sulfide (CH3	South	25.7	25.7	12.8	2	$-9$	-9	$-9$	$\boldsymbol{0}$
		SCH3	North South	37 26	90.8 26.5	35.1 25.5	6 $\overline{c}$	$-9$ $-9$	-9 $-9$	-9 $-9$	$\overline{\mathbf{c}}$ 0
	S	PAN (CH3 CO O2 NO2)	North	$\mathbf{1}$	$\mathbf{I}$	1	$\overline{7}$	$-9$	-9	$-9$	$\mathbf{1}$
	N	Methyl nitrate (CH3	South North	-9 33.5	-9 40.5	$-9$ 25.7	$\overline{2}$ 12	$-9$ 26.1	-9 29.5	$-9$ 22.9	$\pmb{0}$ $\boldsymbol{7}$
		ONO <sub>2</sub> )	South	7.4	8.2	6.9	4	6.9	7.3	3.5	3
	N	Ethyl nitrate (C2H5 ONO <sub>2</sub> )	North South	11.7 2.3	14.6 2.5	7.9 2.1	12 4	7.8 2.2	10.3 2.3	6.7	$\boldsymbol{7}$
7	S	Nitric acid (HNO3)	North	14	15.5	10	Ħ	-9	$-9$	$\mathbf{1}$ $-9$	$\mathfrak{Z}$ $\sqrt{2}$
	S	Formic acid (HCOOH)	South North	24 14	25.5 15.5	12 11	3	$-9$	-9	$-9$	$\mathbf{1}$
			South	32	37	15.5	$\mathbf{1}$ 3	-9 -9	$-9$ -9	$-9$ $-9$	$\sqrt{2}$ $\mathbf{1}$
	${\bf S}$	Acetic acid (CH3 COOH)	North	13	15 <sup>5</sup>	10.5	11	-9	-9	-9	$\sqrt{2}$
			South	27	32.5	12.5	3	-9	-9	$-9$	$\mathbf{1}$

**Table 3. PEM-Tropics SPCZ Comparison: North Versus South** 







**Values are in pptv unless noted. Definitions: median, midpoint of the observations (50% of the observations are below the median, and 50% are above); LQuar, 25% of the observations are below this value; UQuar, 25% of the observations are above this value; N, number of independent observations. Group definitions: 1, meteorological and location parameters; 2, overall general interest species; 3, major hydrocarbon species; 4, hydrocarbon ratios that provide an indicator as to the age of the emissions; 5, halocarbons useful for identifying urban or biomass burning emissions; 6, sulfur, nitrates, and other species of photochemical interest; 7, acids and peroxides; 8, aerosols; 9, other halocarbons. Missing/invalid data code is -9.** 

**north of ITCZ data, and the dashed line is for the south of the ITCZ data.** 

**Figure 5 represents the major species associated with combustion and plots ozone, carbon monoxide, carbon dioxide, ethane, methane, and ethyne. Figure 6 plots tetra-chloro**ethylene  $(C_2Cl_4)$ , methyl chloride (CH<sub>3</sub>Cl), the ethyne/CO ra**tio, and the fine aerosol ratio (0.015 to 1 micron diameter**  aerosols; ratio equal to nonvolatiles at 250°C/total). Tetra**chloro-ethylene and methyl chloride are relatively long-lived traces of urban and biomass burning emissions, respectively [Blake et al., 1996, 1997; Rasmussen et al., 1980]. The fine aerosol ratio is particularly sensitive to continental-source air showing an increase in air enriched in soot, nitrates, fine crustal materials, and other nonvolatile aerosols. However within the marine boundary layer, interpretation of the ratio is more difficult as a result of oceanic processes. Figure 7 represents species which show a boundary layer source for air south of the ITCZ, namely, dimethyl sulfide, methyl nitrate, and ethyl nitrate.** 

The ITCZ data suggest that the chemical composition of air **north and south are different, particularly at the lower altitudes. Air north of the ITCZ shows evidence of combustion and urban emissions common to northern hemisphere air, while south of the ITCZ the air is relatively clean and of more southern hemisphere composition (e.g., carbon monoxide at 50 ppbv). Specifically and for the north of the ITCZ data, carbon monoxide, methane, ethane, ethyne, the ethyne/CO ratio,**  CFC-22, CFC-12, and C<sub>2</sub>Cl<sub>4</sub> are mostly elevated (compared to **south), while carbon dioxide (large continental uptake in the**  northern hemisphere) is lower. As discussed by *Vay et al.* [this **issue], the Pacific "warm pool" is a source of carbon dioxide.** 

Their data place the "warm pool" between 8°N and 8.5°S with a zone of intensity from 6.5°N to 1°S. This colocates the "warm **pool" with our south of the ITCZ data and provides additional support for elevated carbon dioxide south of the ITCZ.** 

**The northern hemisphere signature for the north of the ITCZ data is, however, modest. For example, north of the ITCZ (2- to 4-km bin) carbon monoxide is 62 ppbv (51 ppbv south), ethyne is 25 pptv (19), and CFC-22 is 122 pptv (116). This modest increase is not surprising as the majority of back trajectories indicate that the air sampled north or south of the ITCZ is basically outflow from either the North or South Pacific subtropical high (discussed with Figure 1) and, as such, air was over ocean for many days prior to measurement. The "aged" status of the air is further supported by the chemical data as (1) the short-lived hydrocarbons like ethene or propene are at detection limits in both groups (thus, not in Table 2), (2) the ethyne/CO ratio of <0.5 suggests well-aged air, and (3) the fine aerosol ratio is about 0.1 or less for altitudes above the marine boundary layer, indicating a modest continental component.** 

**For the most part, concentration differences between north and south of the ITCZ air are nonexistent at higher altitudes. For example at 8 to 10 km, carbon monoxide is 57.9 (north) versus 58.1 ppbv (south); ethyne, 30.0 versus 30.8 pptv; ethyne/CO ratio, 0.527 versus 0.533; and CFC-22, 120.2 versus**  122.3 pptv. These observations suggest that the ITCZ is not a **barrier to transport at higher altitude. This is supported by the back trajectory data of Figure 8. Low and midaltitude trajectories (Figures 8b and 8c) indicate that air sampled either north or south of the ITCZ remained north or south of the ITCZ prior to measurement. However, the higher-altitude tra-** 



**jectories of Figure 8a show evidence of cross-ITCZ flow whether the source of the air was from the west or east.** 

A noteworthy observation in the south of the ITCZ data is **the abundance of dimethyl sulfide, methyl nitrate, and ethyl nitrate near the ocean surface. These species have a sizeable oceanic source within the warm southern hemisphere tropical ocean [Thornton etal., this issue; Atlas et al., 1993; this issue]. The lower-altitude increases in sulfur dioxide (see Table A-i) which tend to track the dimethyl sulfide enhancements south of the ITCZ are the result of dimethyl sulfide to sulfur dioxide conversion [Thornton et al., this issue].** 

**The acids and peroxides of group 7 are, for the most part, elevated in the north of the ITCZ air as compared to south. However, these species, their concentrations and ratios, are highly dependent upon the degree of atmospheric processing which has occurred as the result of mixing, rain out, cloud processing, etc. Since air from both sides of the ITCZ was, prior to measurement, over the ocean for many days and in regions of intense atmospheric processing and sunlight, it is likely that these processes have influenced the results. The analyses do not account for these processes. It is noted that Talbot et al. [this issue], in their binned analyses, show similar acid results. (Compare our north of the ITCZ acid data with**  Talbot's data at  $0^\circ$  to 15°N, 120 to 170°W.) O'Sullivan et al. [this **issue] and D. Cohan et at. (Sources and lifetimes of peroxides in the tropical upper troposphere, submitted to Geophysical**  Research Letters, 1997, hereinafter referred to as Cohan et al., **submitted manuscript, 1997) discuss the sources and lifetimes of the peroxides within the troposphere.** 

### **4.2. South Pacific Convergence Zone (SPCZ)**

**Table 3 and Figures 9 through 11 summarize the SPCZ chemical results. Formats are identical to those presented pre**viously with the exception that the 0 to 1 km altitude bin has **been subdivided into 0 to 0.5 and 0.5 to 1 km bins. The data show that the chemical composition of air north and south of the SPCZ is substantially different, especially at altitudes below 4 km. Air south of the SPCZ is strongly influenced by combustion sources, while air north of the SPCZ is relatively clean. The 2 to 4 km median results from Table 3 of (1) elevated levels of ozone (63 ppbv for air south of the SPCZ compared to 21 ppbv north), carbon monoxide (91 compared to 59 ppbv), ethane (486 compared to 283 pptv), and ethyne (103 compared to 18 pptv); (2) little or no increase between north and south values for the shorter-lived hydrocarbons (e.g., ethene at 2.3 pptv); and (3) a south ethyne/CO ratio of 1 to 1.5 suggest that the air, at the time of measurement, is of the order of several days to a week in transient time from combustion sources. A combustion source for air south of the SPCZ is not surprising based upon PEM-Tropics field observations, namely, all flights south of the SPCZ, most flights staged from Tahiti, and many from Easter Island encountered numerous atmospheric layers and plumes at altitudes of 2 to 8 km. These layers were enriched in ozone, carbon monoxide, and various combustion hydrocarbons with hydrocarbon ratios indicative of plume ages that ranged from several days to longer than a week [see Vay et al., this issue; J. Logan et al., unpublished manuscript, 1998].** 

Comparison of the halocarbon data for air north and south **of the SPCZ provides information as to the source of combus**tion. Note that  $C_2Cl_4$  (Figure 10) and the other urban halo**carbons of group 5 show little differences north and south of**  the SPCZ, while CH<sub>3</sub>Cl, our biomass burning tracer, exhibits **higher concentrations south of the SPCZ (574 compared to 549 pptv). These findings (as well as others) support a biomass burning source as opposed to urban emissions. Figure 12, in the same format as Figure 8, represents trajectories for air sampled north and south of the SPCZ and indicates the gen** 









**Figure 6.** ITCZ chemical data for  $C_2Cl_4$  (an urban tracer), CH<sub>3</sub>Cl (a biomass burning tracer), ethyne/CO  $\overline{C_2Cl_4}$  (an urban tracer),  $\overline{C_1Cl_4}$  (and  $\overline{C_2Cl_4}$ ). The format of the figure ratio (an emission age indicator), and fine aerosol ratio (nonvolatile at 250°C/total). The format of the figure<br>. **is the same as Figure 5.** 



**Altitude, km 10 2 •'""•"'E"....... \_ 0**  50 **0 25 Methyl Nitrate, pptv** 

**Figure 7. ITCZ chemical data for species which indicate an oceanic source south of the ITCZ. The format of the figure is the same as Figure 5.** 



Figure 8. Back trajectories of air sampled north and south of the ITCZ. Arrows within each panel indicate the major pathways of tropospheric air into the sampling region. Air was sampled at altitudes (a) > 10 km, (b) 4 to 8 km, and (c) < 2 km. Each panel includes approximately 40 to 50 trajectories and includes only trajectory data which correspond to the time periods of the data of Table 2.



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**Figure 10.** SPCZ chemical data for C<sub>2</sub>Cl<sub>4</sub> (an urban tracer), CH<sub>3</sub>Cl (a biomass burning tracer), ethyne/CO ratio (an emission age indicator), and fine aerosol ratio (nonvolatile at 250°C/total). The format of the figure **is the same as Figure 5.** 

**eral location of the biomass burning. For the south of the SPCZ air and at altitudes above the marine boundary layer, most trajectories (Figures 12b and 12c) indicate westerly air flow with passage over Australia (some Indonesia) several days prior to sampling. When trajectories are extended beyond Australia, in some cases the air passed over Africa 7 to 10 days earlier (H. E. Fuelberg, private communication, 1998). Evidence of biomass burning during this time of year and in the regions of Australia, Indonesia, and Africa is well documented from satellite databases [e.g., Justice et al., 1996].** 

**The gradients in trace species across the SPCZ are sizeable as opposed to the modest gains observed across the ITCZ. This is attributed to the substantially different types (and ages) of air south and north of the SPCZ. North of the SPCZ air is aged and highly processed, having been over the ocean for many days (not that dissimilar to south ITCZ air, see next section); south of the SPCZ air is relatively fresh (several days), direct outflow from continental and biomass burning sources. However, and as was seen with the ITCZ results, chemical signature differences between north and south SPCZ air tend to de**crease at higher altitude (e.g., see Table 3, 8- to 10-km altitude, **and ethyne and the ethyne/CO ratio). Similarly to the ITCZ, high altitude trajectories (Figure 12a) show evidence of cross-SPCZ air flow, mainly from south to north, while the lower altitude trajectories (Figures 12b and 12c) show little evidence of cross-SPCZ flow. The indication of cross-SPCZ flow and the reduction of contrast in the chemical signatures at the higher altitudes indicate that the effectiveness of the SPCZ as a bar-** **rier to transport decreases at the higher altitudes. When using a different altitude-binning scheme for the data between 4 and 8 km altitude, results suggest that chemical signature differences begin to diminish as altitudes begin to approach 5 or 6 km.** 

**The aerosol data above 2 km altitude are particularly interesting. As expected based on the above discussions, nonvolatile**  (heated to 250°C) fine aerosol number densities for air mea**sured south of the SPCZ are considerably larger then values measured in air north of the SPCZ. However, north of the SPCZ, total fine (unheated) and ultrafine (0.004 to 1 micron diameter) aerosol number densities are the larger. We account for this observation by suggesting that air measured north of the SPCZ, having been confined for many days to an equatorial region bounded by the SPCZ and ITCZ and having been processed via clouds, contains a sizeable fraction of aqueous compounds. This observation is further supported in noting that the ratio of heated to unheated fine aerosol for air north of the SPCZ (Figure 10, above 2 km) shows that 95% of the aerosols are volatile at 250øC. Air measured south of the SPCZ indicates that less than 50% of the fine aerosol are volatile. Also important is a source of new-growth aerosols (ultrafine) for air north of the ITCZ, the result of the transport of oceanic emissions aloft and subsequent in situ production of aerosols via heterogenous processes. As indicated in Table 3 and Figure 11, there is an abundance of dimethyl sulfide, methyl nitrate, and ethyl nitrate in the marine boundary layer for air measured north of the SPCZ.** 





**Figure ll. SPCZ chemical data for species which indicate an oceanic source north of the SPCZ. The format of the figure is the same as Figure 5.** 

#### **4.3. South ITCZ Versus North SPCZ Air**

**Before concluding our discussions we briefly compare the chemical signatures of air measured south of the ITCZ and north of the SPCZ. From (1) the pictorial of Figure 1, (2) meteorological discussions, (3) the chemical data which indicate that both the ITCZ and SPCZ are effective transport barriers at low to midtropospheric altitudes, and (4) the consideration that the region between the two zones have similar oceanic sources for dimethyl sulfide, methyl nitrate, and ethyl nitrate as well as similar photochemical tendencies; the chemical signatures of air south of the ITCZ and north of the SPCZ are expected to be similar. Table 4, constructed from Tables 2 and 3, compare median values for some of the major species. Noting that both data sets include measurements for only 3 days and the longitude of the measurements are different (ITCZ, 138 ø to 155øW and SPCZ, 160øE to !75øW), we surmise that the air south of the ITCZ and north of the SPCZ are of similar chemical signature. Small differences in the chemical signatures most likely reflect differences in the "age" of the air and the amount of atmospheric processing as opposed to significantly different source/sink histories. Our analyses suggests that air south of the ITCZ and north of the SPCZ can be combined into a single data set and be representative of air between the ITCZ and SPCZ.** 

### **5. Concluding Remarks**

**We have examined PEM-Tropics DC-8 chemical data obtained in the near vicinity of the Intertropical Convergence**  **Zone (ITCZ) and the South Pacific Convergence Zone (SPCZ). The analyses show that air north and south of the convergent zones have different chemical signatures and the signatures are reflective of the source regions and transport histories of the air. Air north of the ITCZ shows a modest urban/industrialized signature compared to air south of the ITCZ. The chemical signature of air south of the SPCZ is dominated by combustion emissions from biomass burning, while air north of the SPCZ is relatively clean and of similar composition to ITCZ south air. Chemical signature differences of air north and south of the zones are most pronounced at the altitudes below 5 km and, as such, show that at these altitudes the ITCZ and SPCZ are effective barriers to the transport of tropospheric air. At higher altitudes, chemical signatures become less dissimilar. For example, air sampled at 8- to 10-km altitude north and south of the zones have similar chemical signatures. Back trajectories for air sampled at the higher altitudes show cross-zonal flow; whereas, at altitudes below about 5 km, little cross-zonal flow is observed.** 

**Specifically for the ITCZ and for air sampled at 2- to 4-km altitude, north of the ITCZ carbon monoxide is 61 ppbv (52 ppbv south), methane is 1726 ppbv (1707), ethyne is 25 pptv (20), and CFC-22 (urban source) is 122 pptv (116). The modest enhancements are attributed to the age of the urban/ industrialized emissions and extensive dilution during transport. Back trajectory analyses and the chemical data are consistent in identifying north of the ITCZ air as "aged outflow" from the North Pacific Subtropical High and south of the ITCZ air as outflow from the South Pacific Subtropical High.** 



Figure 12. Back trajectories air sampled north and south of the SPCZ. Arrows within each panel indicate the major pathways of tropospheric air into the sampling region. Air was sampled at altitudes (a) >10 km, (b) 4 to 8 km, and (c) <2 km. Each panel includes approximately 40 to 50 trajectories and includes only trajectory data which correspond to the time periods of the data of Table 2.





**Values are in pptv unless noted.** 

**Marine boundary layer air sampled south of the ITCZ show enhancements of dimethyl sulfide, methyl nitrate, and ethyl nitrate and are consistent with known production regions within the warm tropical South Pacific Ocean.** 

**For the South Pacific Convergence Zone and at 2- to 4-km altitude, south of the SPCZ ozone is 63 ppbv (21 ppbv north), carbon monoxide is91 ppbv (59), ethane is 489 pptv (283),**  ethyne is 103 pptv (18), and CH<sub>3</sub>Cl (a biomass burning tracer) **is 574 pptv (549). The magnitude of these chemical differences show the SPCZ to be a significant barrier to tropospheric**  transport. Urban and industrialized tracers like C<sub>2</sub>Cl<sub>4</sub> and the **CFCs show no or little enhancement inthe south SPCZ air.**  Enhancements of the short-lived hydrocarbons (e.g., ethene, **propene) are also not observed, or are at modest levels, indicating that the biomass burning emissions are "aged" several days. Trajectory analyses and the chemical data are consistent with transport times of several days to a week from biomass burning regions inAustralia, New Zealand, Indonesia, and to a lesser degree from as far as Africa. Air sampled north of the SPCZ is relatively clean and at the low altitudes and within the**  marine boundary layer show enhancements in dimethyl sulfide, **methyl nitrate, and ethyl nitrate. A comparison of the air sampled south of the ITCZ and north of the SPCZ suggests their chemical signatures are similar and can be combined to represent air between the ITCZ and SPCZ.** 

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