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Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft mission: Design, execution, and first results

Daniel J. Jacob,¹ James H. Crawford,² Mary M. Kleb,² Vickie S. Connors,² Richard J. Bendura,² James L. Raper,² Glen W. Sachse,² John C. Gille,³ Louisa Emmons,³ and Colette L. Heald¹

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[1] The NASA Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft mission was conducted in February–April 2001 over the NW Pacific (1) to characterize the Asian chemical outflow and relate it quantitatively to its sources and (2) to determine its chemical evolution. It used two aircraft, a DC-8 and a P-3B, operating out of Hong Kong and Yokota Air Force Base (near Tokyo), with secondary sites in Hawaii, Wake Island, Guam, Okinawa, and Midway. The aircraft carried instrumentation for measurements of long-lived greenhouse gases, ozone and its precursors, aerosols and their precursors, related species, and chemical tracers. Five chemical transport models (CTMs) were used for chemical forecasting. Customized bottom-up emission inventories for East Asia were generated prior to the mission to support chemical forecasting and to serve as a priori for evaluation with the aircraft data. Validation flights were conducted for the Measurements Of Pollution In The Troposphere (MOPITT) satellite instrument and revealed little bias ($6 \pm 2\%$) in the MOPITT measurements of CO columns. A major event of transpacific Asian pollution was characterized through combined analysis of TRACE-P and MOPITT data. The TRACE-P observations showed that cold fronts sweeping across East Asia and the associated warm conveyor belts (WCBs) are the dominant pathway for Asian outflow to the Pacific in spring. The WCBs lift both anthropogenic and biomass burning (SE Asia) effluents to the free troposphere, resulting in complex chemical signatures. The TRACE-P data are in general consistent with a priori emission inventories, lending confidence in our ability to quantify Asian emissions from socioeconomic data and emission factors. However, the residential combustion source in rural China was found to be much larger than the a priori, and there were also unexplained chemical enhancements (HCN, CH₃Cl, OCS, alkylnitrates) in Chinese urban plumes. The Asian source of CCl₄ was found to be much higher than government estimates. Measurements of HCN and CH₃CN indicated a dominant biomass burning source and ocean sink for both gases. Large fractions of sulfate and nitrate were found to be present in dust aerosols. Photochemical activity in the Asian outflow was strongly reduced by aerosol attenuation of UV radiation, with major implications for the concentrations of HO_x radicals. New particle formation, apparently from ternary nucleation involving NH₃, was observed in Chinese urban plumes.

INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; *KEYWORDS:* atmospheric chemistry, air pollution, China, Asia, climate, MOPITT

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1. Introduction

[2] The Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft mission was conducted in February–April 2001 by the NASA Global Tropospheric Experiment (GTE) to observe the chemical outflow from Asia to the Pacific and relate it quantitatively to its sources.

It used two aircraft, a DC-8 and a P-3B, based in Hong Kong and at Yokota Air Force Base (AFB) near Tokyo. It was the latest in a 20-year series of GTE aircraft missions focused on the chemistry of the global troposphere [McNeal *et al.*, 1998]. Industrialization of the Asian continent is expected to be a major driver of future global change in atmospheric composition [Intergovernmental Panel on Climate Control (IPCC), 2001]. Most of the outflow from the Asian continent is to the Pacific. GTE had previously conducted a series of Pacific Exploratory Missions (PEM) including PEM-West A and B (August–September 1991 and February–March 1994) over the NW Pacific [Hoell *et al.*, 1996, 1997], and PEM-Tropics A and B (September–October 1996 and March–April 1999) over the tropical and South Pacific [Hoell *et al.*, 1999; Raper *et al.*, 2001]. The PEM-West missions offered a first characterization of Asian outflow to the Pacific. The task of TRACE-P was to go beyond PEM-West in providing a quantitative understanding of the outflow, its origin, and its evolution.

[3] TRACE-P was organized around two objectives. The first was “to determine the chemical composition of the Asian outflow over the western Pacific in spring in order to understand and quantify the export of chemically and radiatively important gases and aerosols, and their precursors, from the Asian continent”. This objective defined a broad chemical scope for the mission encompassing long-lived greenhouse gases, aerosols and their precursors, and oxidants and their precursors. It responded to the need for an integrated multispecies approach toward addressing climate change and global air pollution issues [Hansen *et al.*, 2000]. It also aimed to take advantage of the improved constraints on sources offered by concurrent measurements of a large number of chemical species with overlapping origins.

[4] The second objective of TRACE-P was “to determine the chemical evolution of the Asian outflow over the western Pacific in spring and to understand the ensemble of processes that control this evolution”. This objective recognized the importance of near-field chemical aging in determining global Asian influence on ozone, aerosols, and other relatively short-lived species. It responded to the need to improve understanding of radical chemistry and aerosol dynamics in the complex Asian outflow characterized by high concentrations of aerosols [Dentener *et al.*, 1996; Zhang and Carmichael, 1999], mixing of fossil fuel, bio-fuel, and biomass burning influences [Bey *et al.*, 2001], and interweaving of polluted and stratospheric air [Carmichael *et al.*, 1998].

[5] Three-dimensional chemical transport models (CTMs) played a central role in guiding the design and execution of TRACE-P. This represented a major departure from previous GTE missions, whose objectives were more exploratory. It was recognized at the outset of TRACE-P that CTMs would play a critical role in addressing quantitatively the mission objectives. We wished to optimize the value of the data set for constraining and evaluating these models. Several CTMs were engaged during mission design to predict the dominant source regions and processes contributing to chemical outflow to the Pacific. These predictions guided the selection of operational sites and flight plans. The same CTMs were then engaged during the mission to provide daily chemical forecasts, which were used together with meteorological

forecasts for day-to-day flight planning. Customized “bottom-up” emission inventories for East Asia in 2000, based on best available knowledge of socioeconomic data and emission factors, were generated by Streets *et al.* [2003] as input to the CTMs for the above exercises. These inventories were then used after the mission as a priori knowledge to be tested with the “top-down” constraints provided by the aircraft observations.

[6] Another distinguishing element of TRACE-P was the extensive linkage with observations from other platforms. Joint flights were conducted with the ACE-Asia aircraft mission based out of Iwakuni (Japan) and focused on characterizing the chemical and radiative properties of Asian aerosols over the western Pacific [Huebert *et al.*, 2003; A. D. Clarke *et al.*, Size distributions and mixtures of black carbon and dust aerosol in Asian outflow: Physico-chemistry, optical properties, and implications for CCN, unpublished manuscript, 2003, hereinafter referred to as Clarke *et al.*, unpublished manuscript, 2003]. Joint CTM forecasts were used to support the PHOBEA-II aircraft mission based out of Seattle (U.S. west coast) and focused on transpacific transport of pollution [Jaeglé *et al.*, 2003; Price *et al.*, 2003]. The TRACE-P aircraft data were integrated with satellite observations, in particular the MOPITT measurements of carbon monoxide (CO), for improved characterization of Asian outflow (C. L. Heald *et al.*, Trans-Pacific transport and chemical evolution of Asian pollution observed from satellite and aircraft, submitted to *Journal of Geophysical Research*, 2003, hereinafter referred to as Heald *et al.*, submitted manuscript, 2003). Validation profiles for MOPITT were conducted as part of TRACE-P and results are below.

2. Design

2.1. Seasonal Timing

[7] The early spring timing of TRACE-P was motivated by several reasons. Early spring is the season of strongest outflow from Asia to the western Pacific, due to frequent wave cyclones and associated cold fronts and warm conveyor belts (WCBs) that lift Asian air to the free troposphere where it is then picked up by the prevailing westerlies [Yienger *et al.*, 2000; Bey *et al.*, 2001; Stohl, 2001]. In winter, lifting to the free troposphere is suppressed by the Siberian High and the Asian outflow is directed south (winter monsoon). In summer, the outflow is dominated by deep convection (summer monsoon) and is often above the ceiling of conventional aircraft. In addition, part of that upper tropospheric outflow in summer is transported west toward the Middle East rather than east toward the Pacific [Liu *et al.*, 2002]. Fronts in the fall are further south than in spring, with less entrainment of the outflow into the midlatitude westerly circulation [Liu *et al.*, 2003].

[8] A second factor for the choice of early spring was the timing of the biomass burning season in southeast Asia. The season typically lasts from February to April and peaks in March [Chan *et al.*, 2003a; Duncan *et al.*, 2003]. Burning extends from northeast India to southern China, and is strongest in Burma and Thailand. Prior to TRACE-P, only fragmentary information on biomass burning outflow from Asia was available. Ozone profiles over Hong Kong had shown a seasonal enhancement from biomass burning

Table 1. Operational Characteristics of the DC-8 and P-3B Aircraft During TRACE-P

	DC-8	P-3B
Vertical range, km	0.15–12	0.15–7
Ground speed, knots	240–480	270–330
Endurance, hours	10	8
Payload, tons	20.0	7.0

[Chan et al., 1998; Liu et al., 1999]. Strong biomass burning influence had been found in Asian combustion plumes over the northeast Pacific during PEM-Tropics B [Blake et al., 2001; Staudt et al., 2001]. Biomass burning had received little attention during PEM-West B (February–March 1994), although a CTM analysis by Bey et al. [2001] suggests that it made a large contribution to Asian outflow of CO and ozone during that mission. Better understanding of the biomass burning component of the Asian outflow was a high priority for TRACE-P.

[9] Additional factors contributed to the selection of early spring. Photochemistry in Asian outflow is already active in that season, as previously demonstrated by PEM-West B [Crawford et al., 1997]. Stratospheric intrusions are at their seasonal maximum and are particularly frequent along the Asian Pacific rim due to cyclogenesis and the unusually strong jet stream over Japan (“Japan jet”) [Austin and Midgley, 1994; Stohl, 2001]. Carmichael et al. [1998] had previously found that these intrusions lead to complicated interleaving of stratospheric and pollution influences in the Asian outflow. Finally, spring is the dust season in eastern Asia, and there was strong interest in better understanding the chemical modification of the Asian outflow due to

reactions on dust particles [Dentener et al., 1996; Zhang and Carmichael, 1999].

2.2. Aircraft Payloads

[10] The characteristics of the DC-8 and P-3B aircraft are listed in Table 1. The general strategy in deploying the two aircraft was to maximize the geographical coverage of Asian outflow. The DC-8 had a higher ceiling (12 km) than the P-3B (7 km), a greater range, and a greater payload capacity. Because of overheating, it could not operate below 3 km altitude for more than about one hour before having to climb to above 8 km altitude to cool off. The P-3B was the platform of choice for sampling of the lower troposphere, although the DC-8 also provided frequent vertical profile information down to 0.15 km altitude.

[11] The aircraft payloads are shown in Figures 1a and 1b, and the instruments are listed in Table 2. There was much commonality between the two aircraft but also some differences. The DC-8 included remote sensing of ozone and aerosols (both zenith and nadir) with the Differential Absorption Lidar (DIAL) [Browell et al., 2003]. It included measurements of peroxides, carbonyls, and other oxygenated organics that were not aboard the P-3B. The P-3B included a more extensive payload for aerosol microphysics, and a fast 3-D air motion sensing system for vertical flux measurements [Thornhill et al., 2003].

[12] New to the GTE program were measurements of oxygenated organics aboard the DC-8 by Gas Chromatography and Mass Spectroscopy (GC-MS) [Apel et al., 2003], measurements of HCHO aboard the DC-8 by Tunable Diode Laser Spectroscopy (TDLS) [Fried et al., 2003], measurements of peroxy radicals aboard the P-3B

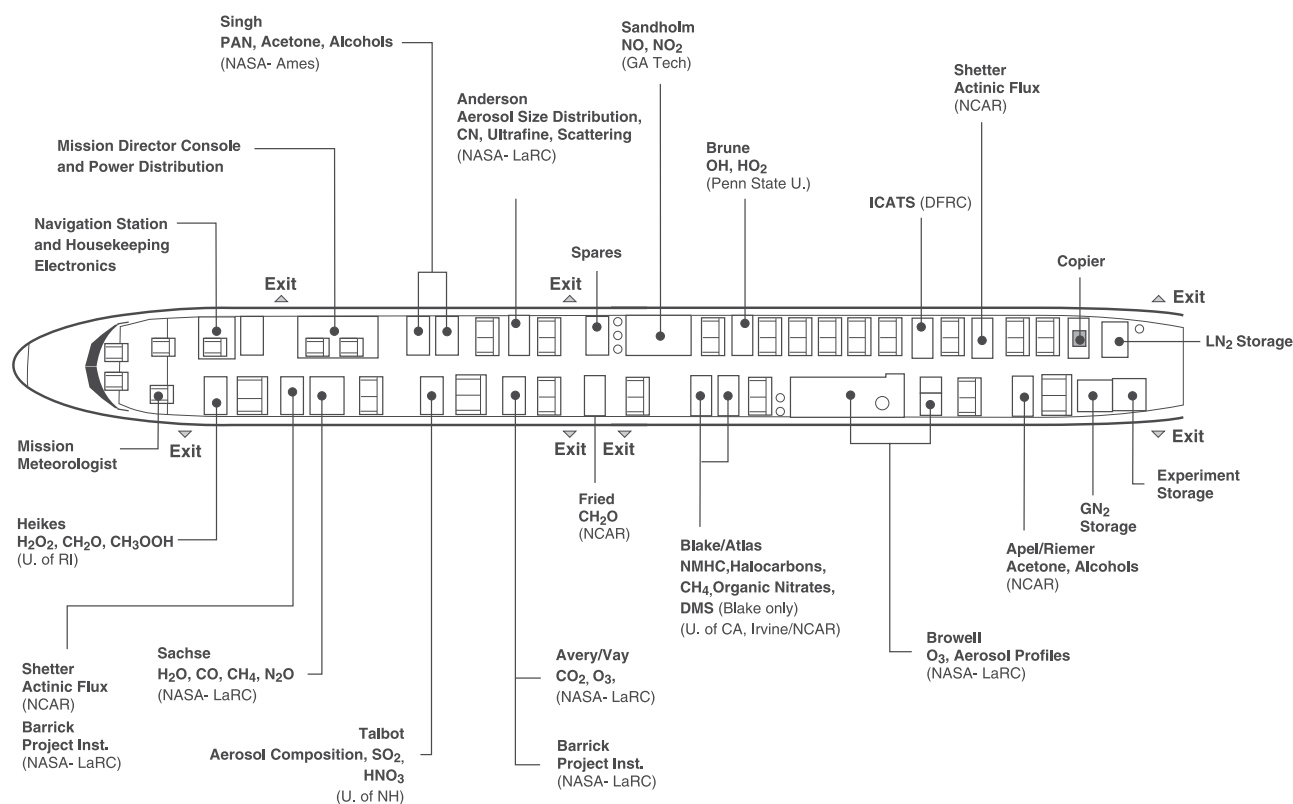


Figure 1a. (a) DC-8 aircraft payload in TRACE-P.

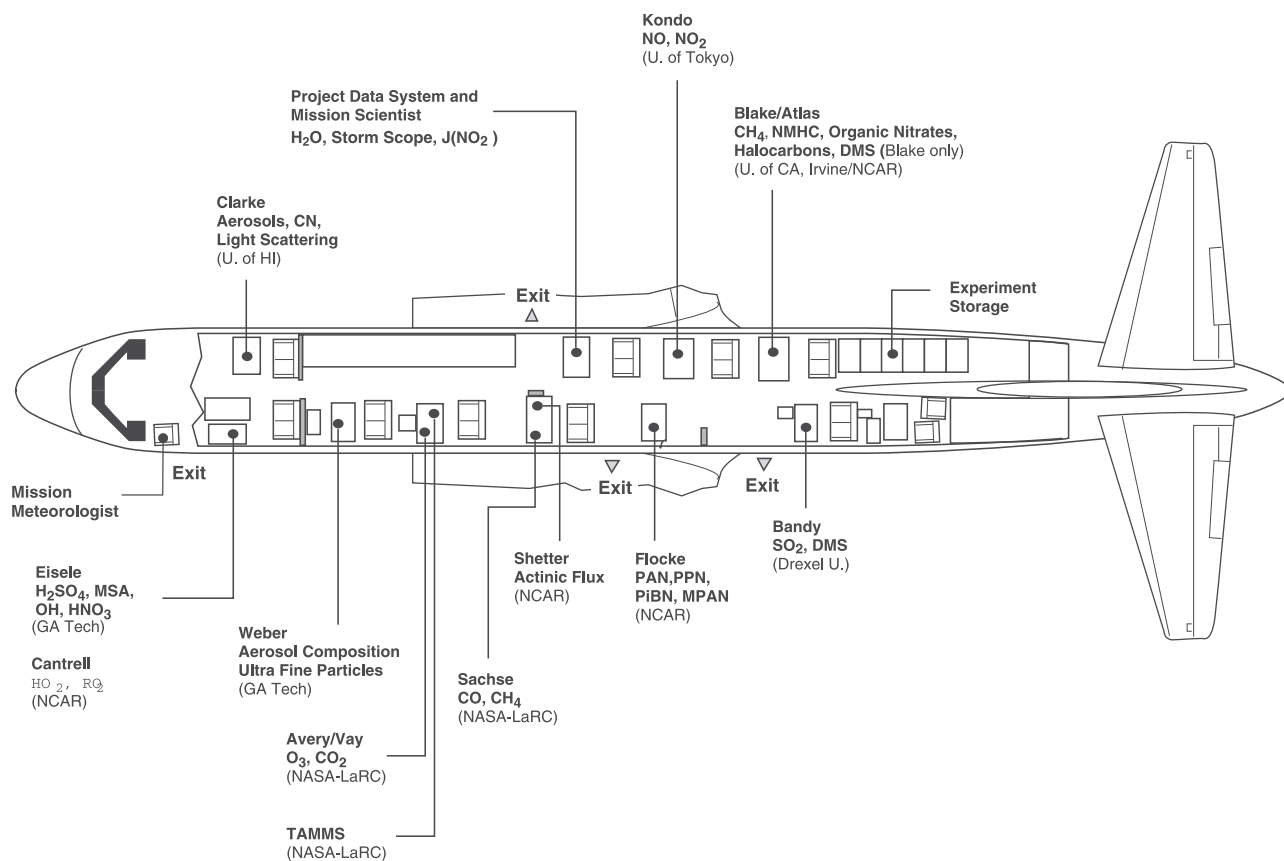


Figure 1b. P-3B aircraft payload in TRACE-P.

by Chemical Ionization Mass Spectroscopy (PerCIMS) [Cantrell *et al.*, 2003], fast measurements of bulk aerosol composition aboard the P-3B by the Particle-Into-Liquid Sampler (PILS) [Ma *et al.*, 2003], fast measurements of SO_2 by Atmospheric Pressure Ionization Mass Spectrometry (APIMS) [Tu *et al.*, 2003], and measurements of PAN by GC-MS aboard the P-3B (F. Flocke). New capabilities were also added to existing instruments, e.g., GC measurements of HCN and CH_3CN [Singh *et al.*, 2003] and CIMS measurements of HNO_3 [Zondlo *et al.*, 2003].

[13] Intercomparison of measurements aboard the two aircraft was conducted on three flight legs over the course of the mission with the aircraft flying side by side. Results from these intercomparisons are presented by Eisele *et al.* [2003].

2.3. Operational Sites

[14] The two main operational bases were Hong Kong (22°N , 114°E) for the first half of the mission (until 16 March) and Yokota Air Force Base in Fussa, a Tokyo suburb (36°N , 140°E) for the second half (Figure 2). These two sites were chosen to enable sampling of a wide range of pathways for Asian outflow. No overflights of the Asian continent were authorized except for Japan. Okinawa (26°N , 128°E) was also considered as an operational site but did not have the facilities necessary. It was used as an overnight site. Kona, Hawaii (19°N , 156°W) and Guam (13°N , 145°E) were transit sites for both the DC-8 and the P-3B, and Kona was also used for a DC-8 sortie. Additional transit sites for the P-3B were Palmdale, California (35°N ,

118°W), Wake Island (19°N , 167°E), and Midway Island (28°N , 178°W). A P-3B sortie was conducted out of Midway. The aircraft home bases were Dryden Flight Research Center (DFRC), California (35°N , 118°W) for the DC-8 and Wallops Flight Facility (WFF), Virginia (38°N , 75°W), for the P-3B.

2.4. Ancillary Observations and Forecasts

[15] Supporting observations during TRACE-P included ozonesonde profiles from a number of sites (Figure 2): Trinidad Head, California (40°N , 124°W); Hilo, Hawaii (20°N , 155°W); Sapporo, Japan; (43°N , 141°E), Tateno, Japan (36°N , 140°E); Cheju Island, South Korea (34°N , 127°E); Kagoshima, Japan (32°N , 131°E); Naha, Japan (26°N , 128°E); Taipei, Taiwan (25°N , 121°E); Hong Kong, China (22°N , 114°E). Lin'an, China (30°N , 120°E), and Kunming, China (25°N , 103°E). Ozonesonde launches were generally made 1–3 times a week. Daily measurements were made at Lin'an and Kunming from 3 to 13 March [Chan *et al.*, 2003b]. Continuous column measurements of CO , C_2H_2 , C_2H_6 , and HCN by solar IR absorption were made by M. Koike (University of Tokyo) at three sites in Japan: Moshiri (44°N , 142°E), Rikubetsu (43°N , 144°E), and Tateno.

[16] Table 3 lists the ensemble of forecast products used during the mission to guide day-to-day flight planning. Five different CTMs (two global, three regional), driven by different meteorological products, delivered chemical forecasts continuously during the mission to supplement the more standard meteorological forecasts. These same CTMs

Table 2. Aircraft Measurements in TRACE-P

Measurement	DC-8	P-3B	Method	Investigator
Meteorological and radiative variables, including fast vertical winds (P-3B)	X	X	J_{NO_2} filter radiometer, chilled-mirror hygrometer, Turbulent Air Motion Measurement System (TAMMS)	NASA Langley (J. D. Barrick and L. Thornhill)
Photolysis frequencies	X	X	actinic flux spectrometer	R. E. Shetter, NCAR
Aerosol size distribution and optical properties, black carbon (BC), volatile and nonvolatile condensation nuclei (CN)	X		optical sizing probes, CN counter, nephelometer, absorption photometer	B. E. Anderson, NASA Langley
Aerosol size distribution and optical properties, BC, volatile and nonvolatile CN, ultrafine aerosol, cloud liquid water content		X	optical sizing probes, CN counter, nephelometer, absorption photometer, differential mobility analyzer	A. D. Clarke, University of Hawaii
Aerosol chemical composition, ultrafine aerosol		X	particle-into-liquid sampler (PILS), ultrafine CN counter	R. J. Weber, Georgia Institute of Technology
Ozone and aerosol vertical profiles	X		differential absorption lidar (DIAL)	E. V. Browell, NASA Langley
Ozone	X	X	chemiluminescence	M. A. Avery, NASA Langley
OH, HO ₂	X		laser-induced fluorescence (LIF)	W. L. Brune, Pennsylvania State University
OH, H ₂ SO ₄ , methanesulfonic acid, HNO ₃		X	chemical ionization mass spectroscopy (CIMS)	F. L. Eisele, NCAR
Peroxy radicals		X	CIMS	C. A. Cantrell, NCAR
Aldehydes, ketones, alcohols	X		GC/MS	E. C. Apel, NCAR
HCHO	X		tunable diode laser absorption spectrometer (TDLAS)	A. Fried, NCAR
H ₂ O ₂ , CH ₃ OOH, HCHO	X		HPLC, flow injection analysis (FIA)	B. G. Heikes, University of Rhode Island
CO, CH ₄ , H ₂ O	X	X ^a	TDLAS	G. W. Sachse, NASA Langley
CO ₂	X	X	IR spectrometer	S. A. Vay, NASA Langley
NO, NO ₂	X		two-photon LIF (TP-LIF)	S. T. Sandholm, Georgia Institute of Technology
NO, NO ₂ , NO _y		X	chemiluminescence	Y. Kondo, University of Tokyo
Peroxyacynitrates		X	GC	F. Flocke, NCAR
Peroxyacynitrates, aldehydes, ketones, alcohols, nitriles, alkyl nitrates	X		GC	H. B. Singh, NASA Ames
HNO ₃ , SO ₂ , aerosol chemical composition	X		Mist chamber-ion chromatography, bulk aerosol filter	R. W. Talbot, University of New Hampshire
SO ₂		X	Atmospheric pressure ionization mass spectrometry (APIMS)	A. Bandy, Drexel University
Alkyl nitrates, halocarbons, OCS	X	X	GC/MS	E. Atlas, NCAR
Hydrocarbons, halocarbons, alkyl nitrates, OCS, CS ₂ , dimethylsulfide	X	X	GC-ECD/FID/MS	D. R. Blake, University of California, Irvine

^aExcept for water vapor, which was measured by TDLAS only aboard the DC-8.

had been used previously in the mission design stage, through simulation of earlier meteorological years, to identify the dominant Asian outflow pathways to be sampled in TRACE-P. The CTM forecasts used anthropogenic Asian emission inventories compiled by *Streets et al.* [2003] for spring 2000. Biomass burning emissions were climatological estimates and varied between the models. Most models focused on forecasts of CO as a tracer of Asian pollution outflow; real-time observations of CO were available aboard the aircraft to check the quality of the forecasts during flight. The LaRC forecast also included a stratospheric ozone tracer and the CFORS forecast included a large suite of chemical and aerosol tracers [*Carmichael et al.*, 2003a].

[17] Several satellite instruments provided near real-time data during the TRACE-P mission to guide flight planning. CO column data from MOPITT were produced by the MOPITT Science Team specifically for TRACE-P operations and were delivered as imagery within 48 hours of data collection. Other satellite products included TOMS (tropospheric ozone, absorbing aerosols), AVHRR (fire counts), and LIS (lightning). The TOMS and LIS products were

combined with forecast forward trajectories to predict the spatial distributions of dust, biomass burning, and lightning influences on a day-to-day basis (Table 3).

2.5. Bottom-up Asian Emission Inventories

[18] *Streets et al.* [2003] used best available knowledge of socioeconomic data and emission factors to generate “bottom-up” gridded emission inventories of Asian emissions for 2000 in support of TRACE-P. Preliminary versions of these inventories were used during the mission as input to the CTM chemical forecasts. The inventories provide gridded emissions of long-lived greenhouse gases (CO₂, CH₄), ozone precursors (NO_x, reactive organic gases, CO), and aerosols and their precursors (black carbon, organic carbon, SO₂, NH₃), all treated in a consistent manner (i.e., on the basis of the same data). They include uncertainty estimates, for each species and source region, in order to assess consistency with the “top-down” constraints from the TRACE-P aircraft observations and in particular to provide the error characterization needed for inverse modeling of sources.

[19] Anthropogenic emissions in East Asia include a large contribution from residential combustion sources, in partic-

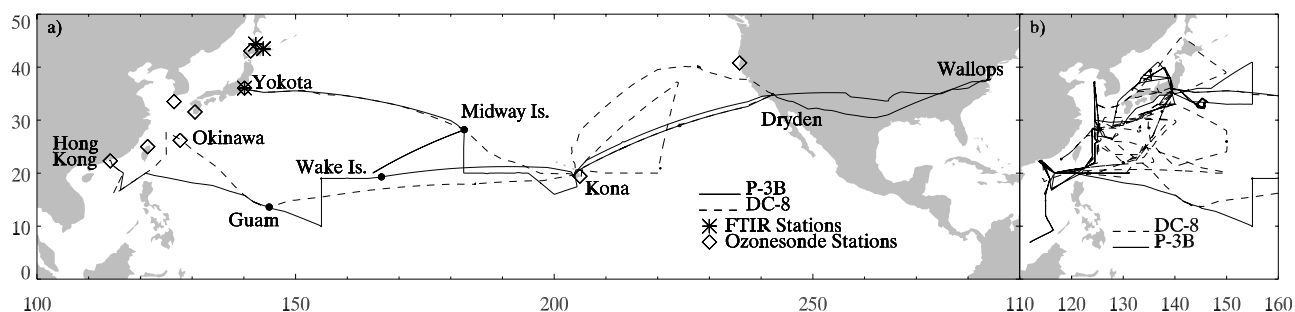


Figure 2. TRACE-P operational sites and flight tracks. (a) Transit flights. (b) Sorties out of Hong Kong, Yokota AFB, and Okinawa.

ular from biofuels. Biofuel emissions of CO for China in the *Streets et al.* [2003] inventory are $45 \pm 32 \text{ Tg yr}^{-1}$, as compared to $64 \pm 27 \text{ Tg yr}^{-1}$ for fossil fuel emissions. *Streets et al.* [2003] estimated a factor of 2 seasonal variation in residential energy use in China with a peak in January and a minimum in June. March–April values are near the annual mean.

[20] One issue in TRACE-P planning was to determine what changes to expect relative to the 1994 PEM-West B mission, conducted in the same region and roughly in the same season (February–March). *Streets et al.* [2003] estimated that anthropogenic emissions in East Asia had changed from 1994 to 2000 by -17% for SO_2 , $+12\%$ for NO_x , $+11\%$ for nonmethane volatile organic compounds (NMVOCs), $+16\%$ for NH_3 , $+1.1\%$ for CO_2 , $+6.4\%$ for CH_4 , and -26% for black carbon (BC), largely determined by trends in China. The large decrease in BC emissions was attributed to reduction in residential coal use. The relatively small changes for other species implied that no major emission-driven change in the chemical composition of the outflow would be expected between the PEM-West B and TRACE-P data sets.

3. Execution

3.1. Meteorological Conditions

[21] *Fuelberg et al.* [2003] give an overview of the meteorological environment during TRACE-P, and *Liu et al.* [2003] describe the outflow pathways for Asian pollution. These two papers also put the mission in a seasonal and interannual context. The mission took place during a transition period from weak cold to neutral ENSO condi-

tions. Deep convection over SE Asia was stronger than normal and cold fronts were more frequent than normal. There were no major anomalies in the mean flow and precipitation patterns over the North Pacific [*Fuelberg et al.*, 2003; *Price et al.*, 2003].

[22] Cold fronts swept across East Asia every 6 days on average during TRACE-P [*Liu et al.*, 2003]. These fronts extended southward from wave cyclones forming in the area around northern Japan and following the storm tracks toward the Aleutians [*Fuelberg et al.*, 2003]. The movement of the fronts followed a northwesterly track across China and the Pacific rim. As the fronts progressed south to $\sim 20^\circ\text{N}$ over the Pacific their orientation gradually changed from SW-NE to W-E, and they became indistinct as they merged into the tropical air. Asian outflow to the Pacific associated with the cold fronts took place (1) ahead of the front in southwesterly WCBs lifting boundary layer air to the free troposphere and (2) behind the front in boundary layer advection capped at 1–2 km altitude by dry subsiding air. Two detailed case studies are presented by *Hannan et al.* [2003].

[23] Deep convection during TRACE-P was largely confined to southeast Asia, although it migrated northward toward China as the mission progressed [*Fuelberg et al.*, 2003; *Miyazaki et al.*, 2003]. The convective plumes in the upper troposphere were carried by westerlies over the NW Pacific and were sampled on several flights.

[24] *Cho et al.* [2003] examined the turbulent flow characteristics during TRACE-P using the fast vertical wind measurements aboard the P-3B. They found that the free troposphere was usually stable, and that most of the instability there was the result of wind shear. In the MBL,

Table 3. Forecast Products Used for Flight Planning

Type	Product	Underlying Meteorological Model	Investigator
Meteorological forecast	synoptic maps, trajectories	ECMWF	H. E. Fuelberg, Florida State University
Satellite-based source influence forecasts	forward trajectories initialized from near-real-time satellite observations of fires and dust (TOMS) and lightning (LIS)	GEOS	A. M. Thompson, NASA GSFC
Global CTM forecasts			
ECHAM	CO	ECMWF	M. G. Schultz, MPI Hamburg
GEOS-CHEM	CO	GEOS	D. J. Jacob, Harvard University
Regional CTM forecasts			
CFORS	CO, ozone and precursors, aerosols and precursors, tracers of sources	RAMS	G. R. Carmichael, University of Iowa
RAQMS	CO, ozone tracers	NCEP	R. B. Pierce, NASA Langley
Meso-NH	CO	Meso-NH	C. Mari, LA Toulouse

Table 4a. DC-8 Flights

Flight ^a	Day ^b (UT)	Location	Features ^c
4	26 Feb.	Dryden-Kona	trans-Pacific Asian pollution [<i>Blake et al.</i> , 2003; Heald et al., submitted manuscript, 2003], MOPITT validation
5	27 Feb.	Kona-Guam	trans-Pacific Asian pollution [<i>Blake et al.</i> , 2003; Heald et al., submitted manuscript, 2003; Newell et al., unpublished manuscript, 2003], MOPITT validation
6	3 March	Guam-Hong Kong	frontal crossing and convective outflow [<i>Browell et al.</i> , 2003], intercomparison with P-3B ^d
7	7 March	Hong Kong (sortie 1)	frontal crossing, frontal and postfrontal outflow [<i>Carmichael et al.</i> , 2003a; <i>Crawford et al.</i> , 2003; <i>Liu et al.</i> , 2003]
8	9 March	Hong Kong-Okinawa	chemical aging of frontal outflow [<i>Carmichael et al.</i> , 2003a]
9	10 March	Okinawa-Hong Kong	boundary layer outflow from China to Yellow Sea, frontal and convective outflow downwind of south China [<i>Browell et al.</i> , 2003]
10	13 March	Hong Kong (sortie 2)	aged postfrontal boundary layer outflow, upper tropospheric convective outflow
11	17 March	Hong Kong-Okinawa	frontal outflow [<i>Browell et al.</i> , 2003; <i>Lefer et al.</i> , 2003], MOPITT validation
12	18 March	Okinawa-Yokota	boundary layer outflow from China to Formosa Strait; multiple outflows (frontal, convective, dust), stratospheric intrusions
13	20 March	Yokota (sortie 1)	Shanghai plume [<i>Simpson et al.</i> , 2003; <i>Talbot et al.</i> , 2003; <i>Vay et al.</i> , 2003], frontal crossings and WCB [<i>Hannan et al.</i> , 2003; <i>Mari et al.</i> , manuscript in preparation, 2003], dust outflow, stratospheric air, MOPITT validation
14	23 March	Yokota (sortie 2)	convective outflow, stratospheric intrusions, intercomparison with P-3B, MOPITT validation
15	26 March	Yokota (sortie 3)	frontal crossing and WCB [<i>Lefer et al.</i> , 2003; <i>Tang et al.</i> , 2003a; <i>Mari et al.</i> , manuscript in preparation, 2003], convective outflow [<i>Liu et al.</i> , 2003], dust outflow, stratospheric intrusions
16	29 March	Yokota (sortie 4)	sunrise photochemistry, ship plumes, MOPITT validation
17	31 March	Yokota (sortie 5)	WCB, stratospheric intrusions in cyclonic system, Miyake-jima volcanic plume [<i>Carmichael et al.</i> , 2003a], overpass of ACE-Asia Twin Otter
18	3 April	Yokota-Kona	WCB outflow and stratospheric influence [<i>Miyazaki et al.</i> , 2003; <i>Wild et al.</i> , 2003], marine boundary layer chemistry
19	6 April	Kona (sortie)	North Pacific background, MOPITT validation
20	9 April	Kona-Dryden	stratus chemistry, aged Asian outflow, intercomparison with P-3B

^aFlights 1–3 were test flights out of Dryden AFB.

^bDate at takeoff. Most flights took off between 0900 and 1100 LT and were 8–10 hours in duration.

^cPublications presenting case studies for specific flights are indicated.

^dCorresponding to P-3B flight 8; the DC-8 took off at 2350 UT on 3 March, and the P-3B took off at 0030 UT on 4 March.

the air was less stable and convection was a more important source of turbulence. They found large vertical gradients of water vapor in the MBL even under unstable conditions, suggesting fast vertical transport by shallow convective cells.

3.2. Flight Tracks and Sampled Air Masses

[25] Individual flights for each aircraft are listed in Table 4, and the corresponding flight tracks are shown in Figures 3a and 3b (the ensemble of flight tracks is shown in Figure 2). The DC-8 and P-3B conducted 17 and 21 research flights, respectively, in addition to three test flights (numbered 1–3) at the beginning of the mission. The flights lasted typically 8–10 hours for the DC-8 and 7–8 hours for the P-3B. Takeoff was usually between 0900 and 1100 LT, so that observations were during sunlit hours. Two flights, one each for the DC-8 (flight 16) and the P-3B (flight 22), were “sunrise” flights with take off well before dawn. Prominent features of each flight are identified in Table 4, and papers using particular flights as case studies are referenced there.

[26] The transit flights across the Pacific (DC-8 flights 4–5 and 18–20, P-3B flights 4–7 and 20–23) had two objectives: (1) to observe the transpacific transport of Asian pollution plumes and (2) to characterize the background air composition over the North Pacific against which to reference the perturbation from Asian outflow. The outbound transit flights, from California to Kona and from Kona to Wake Island and Guam, observed extensive Asian pollution layers with CO concentrations approaching 300 ppbv and ozone approaching 80 ppbv (Heald et al., submitted manuscript, 2003; R. E. Newell et al., Following potential vorticity and

pollution across the Pacific-spring 2001. 1: Mainly layers in pollution, unpublished manuscript, 2003, hereinafter referred to as Newell et al., unpublished manuscript, 2003). The return flights sampled a major WCB outflow event over the NW Pacific (DC-8 flight 18 and P-3B flight 20 [*Hannan et al.*, 2003]), and then background air further east as a blocking ridge over the central Pacific deflected Asian outflow to the Arctic.

[27] The sorties out of Hong Kong focused principally on frontal outflow [*Browell et al.*, 2003] and the chemical aging of Asian air masses. Deep convective continental outflow was also sampled on several DC-8 flights (Table 4). Frontal outflow flights typically sampled (1) the clean marine air ahead of the front, (2) the WCB within the front, and (3) the boundary layer outflow behind the front. Chemical aging flights (DC-8 flights 8 and 10, P-3B flight 10) revisited continental air masses in the lower troposphere after they had evolved for a few days over the Pacific behind decaying fronts.

[28] The move of the aircraft to Japan on 17 March for the second half of the mission signaled a change in the outflow regimes sampled by the aircraft, as discussed by *Fuelberg et al.* [2003]. Less priority was given to frontal outflow, which was still present but had been sampled extensively out of Hong Kong. Operations out of Japan were closer to the center of cyclonic activity [*Fuelberg et al.*, 2003]. They afforded more opportunity to examine the WCBs associated with these cyclones [*Hannan et al.*, 2003]. This was a major theme of two of the flights (DC-8 flight 13, P-3B flight 19). Convective outflow in the upper troposphere was also sampled more frequently than in the flights out of Hong

Table 4b. P-3B Flights

Flight ^a	Day ^b (UT)	Location	Features ^c
4	Feb. 24	Wallops-Palmdale	stratospheric intrusion over western United States
5	Feb. 26	Palmdale-Kona	trans-Pacific Asian pollution
6	Feb. 27	Kona-Wake Island	trans-Pacific Asian pollution (Newell et al., unpublished manuscript, 2003)
7	1 March	Wake Island-Guam	tropical-subtropical chemical gradient
8	4 March	Guam-Hong Kong	Asian outflow to South China Sea, intercomparison with DC-8 ^d
9	7 March	Hong Kong (sortie 1)	postfrontal boundary layer outflow [Carmichael et al., 2003a], frontal cloud profiling [Crawford et al., 2003]
10	9 March	Hong Kong (sortie 2)	aged frontal outflow with anthropogenic and biomass burning layers [Carmichael et al., 2003a; Ma et al., 2003; Clarke et al., unpublished manuscript, 2003]
11	10 March	Hong Kong (sortie 3)	South China Sea under easterly flow, Manila Plume
12	13 March	Hong Kong (sortie 4)	aged boundary layer outflow
13	17 March	Hong Kong-Okinawa	lower free tropospheric outflow to Yellow Sea [Tu et al., 2003]
14	18 March	Okinawa-Yokota	Chinese urban plumes over Yellow Sea [Blake et al., 2003; Carmichael et al., 2003a; Ma et al., 2003; Tu et al., 2003; Weber et al., 2003], volcanic plume
15	21 March	Yokota (sortie 1)	frontal and postfrontal cloud profiling [Hannan et al., 2003]
16	23 March	Yokota (sortie 2)	intercomparison with DC-8, lower tropospheric outflow north and south of Japan
17	26 March	Yokota (sortie 3)	air-sea exchange, Miyake-jima volcanic plume [Carmichael et al., 2003a; Lefer et al., 2003; Tang et al., 2003a, 2003b]
18	30 March	Yokota (sortie 4)	Asian outflow to Sea of Japan, intercomparison with ACE-Asia C-130
19	2 April	Yokota (sortie 5)	Seoul plume [Ma et al., 2003; Weber et al., 2003; Simpson et al., 2003; Clarke et al., unpublished manuscript, 2003], Sea of Japan frontal crossing, intercomparison with ACE-Asia C-130 [Thornhill et al., 2003]
20	3 April	Yokota-Midway	WCB outflow [Hannan et al., 2003; Miyazaki et al., 2003]
21	6 April	Midway (sortie)	aged Asian outflow, stratospheric influence
22	7 April	Midway-Kona	sunrise photochemistry, stratospheric influence
23	9 April	Kona-Dryden	intercomparison with DC-8, stratospheric intrusion
24	10 April	Dryden-Wallops	tropical influence in upper troposphere

^aFlights 1–3 were test flights out of Wallops Flight Facility.

^bDate at takeoff. Most flights left between 0900 and 1100 LT and were 7–8 hours in duration.

^cPublications presenting case studies for specific flights are indicated.

^dCorresponding to DC-8 flight 6; the DC-8 took off at 2350 UT on 3 March, and the P-3B took off at 0030 UT on 4 March.

Kong due to the building summer monsoon [Miyazaki et al., 2003]. Stratospheric intrusions were frequently sampled, and the DC-8 aircraft flew occasional long legs in the stratosphere (e.g., flight 13). Identifiable dust plumes were sampled on DC-8 flights 12 and 13, although the largest Asian dust plumes in the spring of 2001 actually occurred after the end of the TRACE-P mission and were sampled by the ACE-Asia and PHOBEA-II missions [Huebert et al., 2003; Price et al., 2003]. The volcanic plume from the Miyake-jima volcano in southern Japan was sampled on DC-8 flight 17 and P-3B flight 17 [Carmichael et al., 2003a]. This volcano was a major source of SO₂ to the NW Pacific during the TRACE-P mission, comparable to that from China [Tang et al., 2003a; Carmichael et al., 2003b].

[29] The WCB outflow was typically sampled at 2–6 km altitude in the flights out of Hong Kong, and occasionally at higher altitudes in the flights out of Japan [Carmichael et al., 2003a; Hannan et al., 2003; Liu et al., 2003; Singh et al., 2003]. The WCBs typically originated from SE Asia and southern China [Miyazaki et al., 2003] and had chemical signatures from both anthropogenic pollution and biomass burning. WCBs were also found in some instances to lift clean maritime tropical air to the free troposphere and dilute the continental outflow (C. Mari et al., The effect of clean warm conveyor belts on the export of pollution from East Asia, manuscript in preparation, 2003, hereinafter referred to as Mari et al., manuscript in preparation, 2003). Boundary layer outflow behind the cold fronts generally contained high levels of anthropogenic pollution

and was devoid of biomass burning influence [Carmichael et al., 2003a; Liu et al., 2003]. Convective outflow plumes sampled in the upper troposphere during the Hong Kong flights were affected by biomass burning in SE Asia but were largely devoid of anthropogenic influence [Li et al., 2003; Russo et al., 2003].

[30] Chinese urban plumes sampled in the boundary layer outflow behind cold fronts, typically 1–2 days downwind of their sources and strongly capped by subsidence inversions based at 1–2 km altitude, contained the highest pollutant concentrations observed during TRACE-P. Such plumes were observed over the Yellow Sea (DC-8 flight 9, P-3B flight 14), in the Formosa Strait (DC-8 flight 12), and downwind of Shanghai (DC-8 flights 13 and 16) and Seoul (P-3B flight 19). The highest pollution levels were observed in the ~18-hour old Shanghai plume sampled on DC-8 flight 13; CO and O₃ concentrations exceeded 1.2 ppmv and 140 ppbv, respectively. Discussions of the chemical composition of these plumes, and their relations to sources, are given, for example, by Carmichael et al. [2003b], Ma et al. [2003], Russo et al. [2003], Simpson et al. [2003], and Singh et al. [2003].

[31] No evident European anthropogenic plumes or African biomass burning plumes were observed during TRACE-P, although these sources obviously contributed to the background. Fuelberg et al. [2003] point out that back trajectories calculated along the aircraft flight tracks almost never had a direct European origin. A CTM analysis by Liu et al. [2003] shows weak European pollution plumes (Δ CO

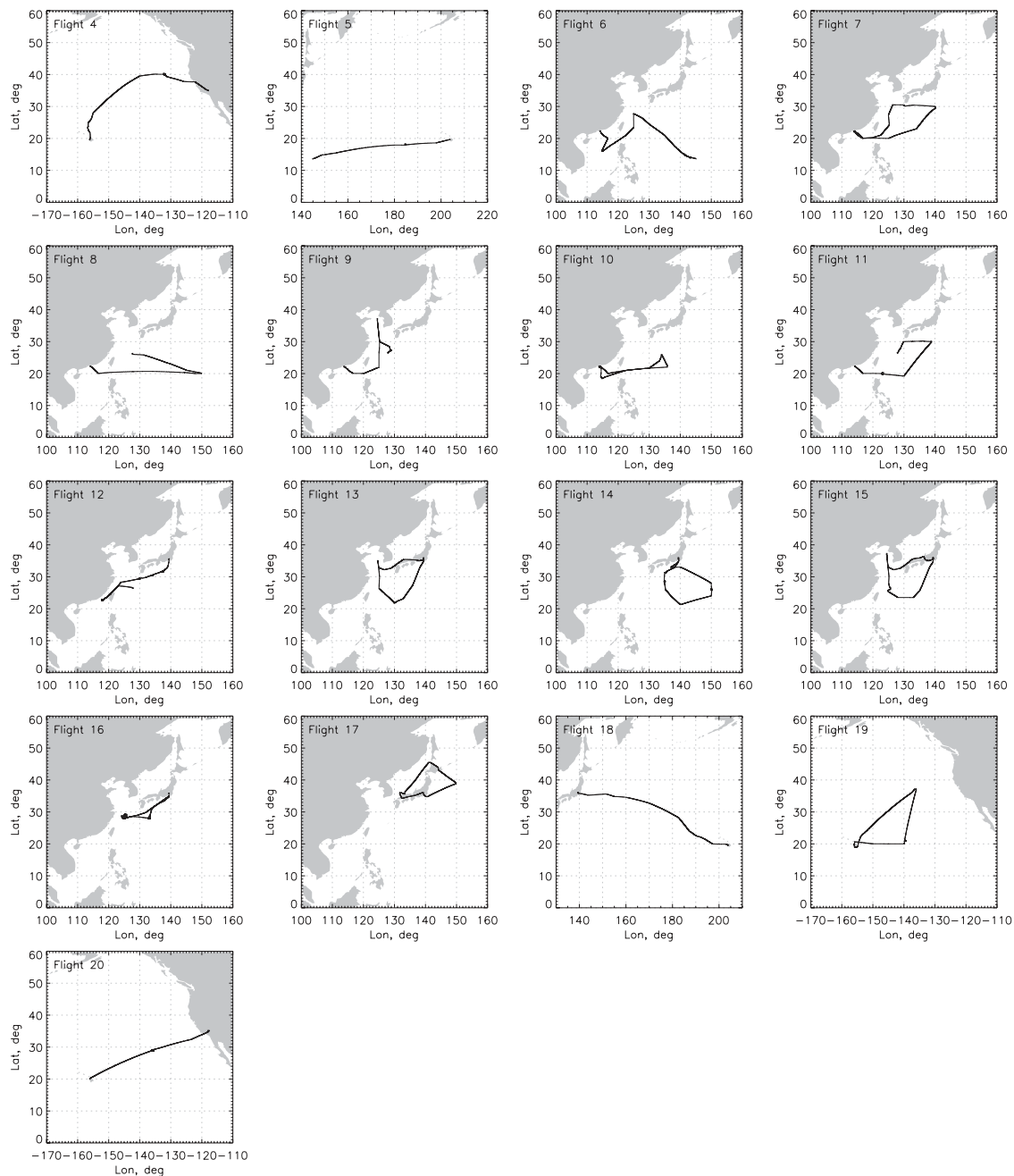


Figure 3a. DC-8 flights during TRACE-P. Also see Table 4a.

~ 50 ppbv) in the boundary layer outflow over the Pacific Rim north of 35°N , but these would be masked by the larger outflow of Asian pollution taking place under the same meteorological conditions. The same analysis shows African biomass burning influence in the upper troposphere south of 25°N but the corresponding structure ($\Delta\text{CO} \sim 10$ ppbv) would be too weak to be detected from the aircraft.

[32] Three in-flight intercomparisons between the DC-8 and the P-3B were conducted during TRACE-P (Table 4) [Eisele et al., 2003]. Extensive collaboration with ACE-Asia had been planned prior to the mission, but mechanical problems delayed the deployment of the ACE-Asia aircraft and restricted collaborative flights to the last two TRACE-P sorties out of Japan. The P-3B aircraft

conducted two joint flights (flights 18 and 19) with the ACE-Asia C-130 aircraft, allowing intercomparison of measurements aboard the two aircraft [Thornhill et al., 2003; Clarke et al., unpublished manuscript, 2003]. The DC-8 aircraft (flight 17) provided overhead aerosol DIAL data for the ACE-Asia Twin Otter as the latter conducted a column radiative closure experiment offshore of southern Japan.

3.3. MOPITT Validation

[33] Observations of CO from MOPITT offered continuous mapping of Asian outflow during TRACE-P (Heald et al., submitted manuscript, 2003). The MOPITT instrument [Drummond, 1992; Edwards et al., 1999] flies on the Terra satellite in a Sun-synchronous orbit with local overpass

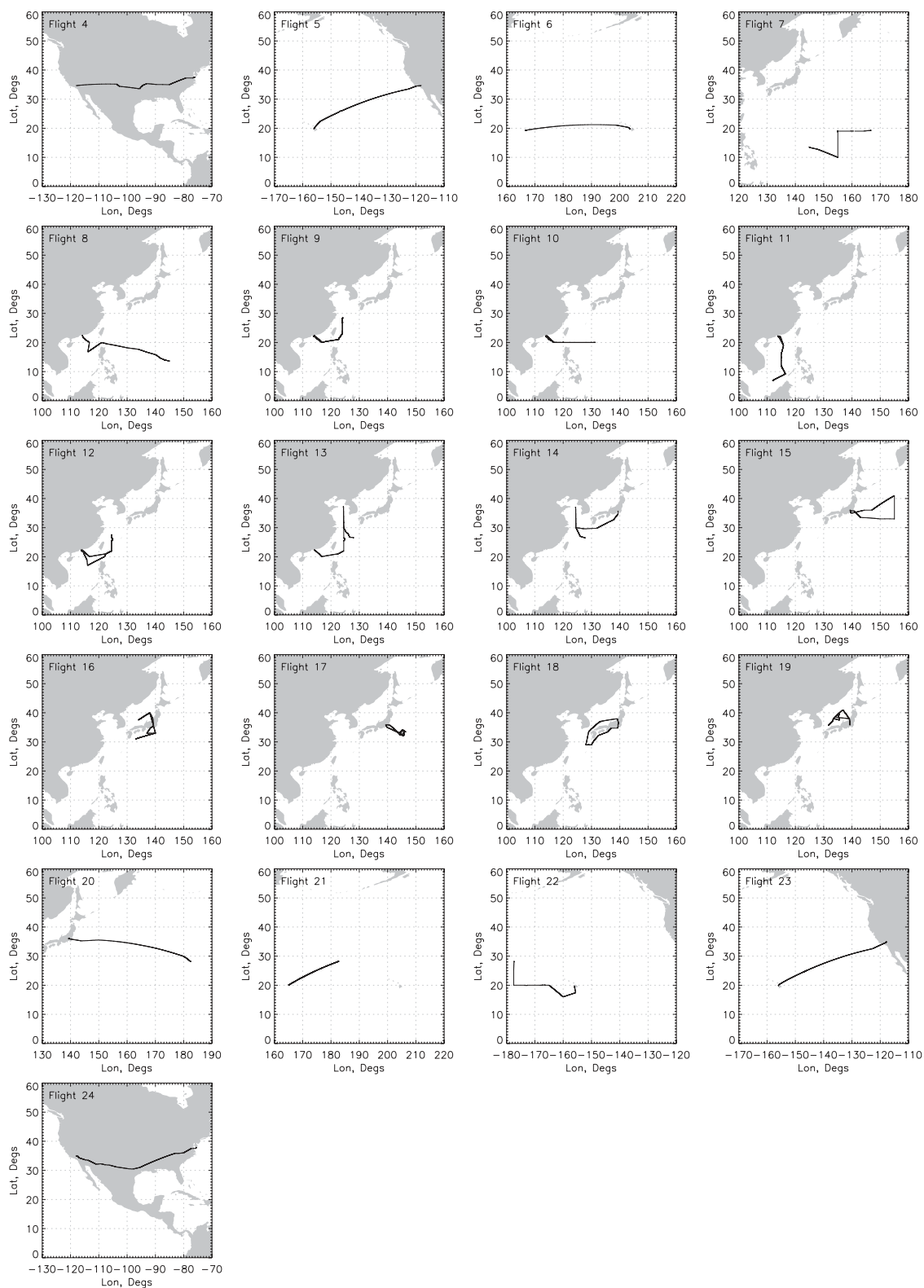


Figure 3b. P-3B flights during TRACE-P. Also see Table 4b.

times of 1045 and 2245. The instrument measures upwelling radiation around the CO absorption band at 4.6 μm . The instrument field of view is $22 \times 22 \text{ km}^2$, with cross-track scanning extending 320 km on each side of the orbit track, resulting in quasi-global coverage of the Earth every 3 days

excluding cloudy scenes. MOPITT reports CO concentrations for seven vertical levels, but there is in fact little more than one independent piece of information in the MOPITT retrieval (Heald et al. (submitted manuscript, 2003); also see averaging kernels in Figure 4). MOPITT thus effectively

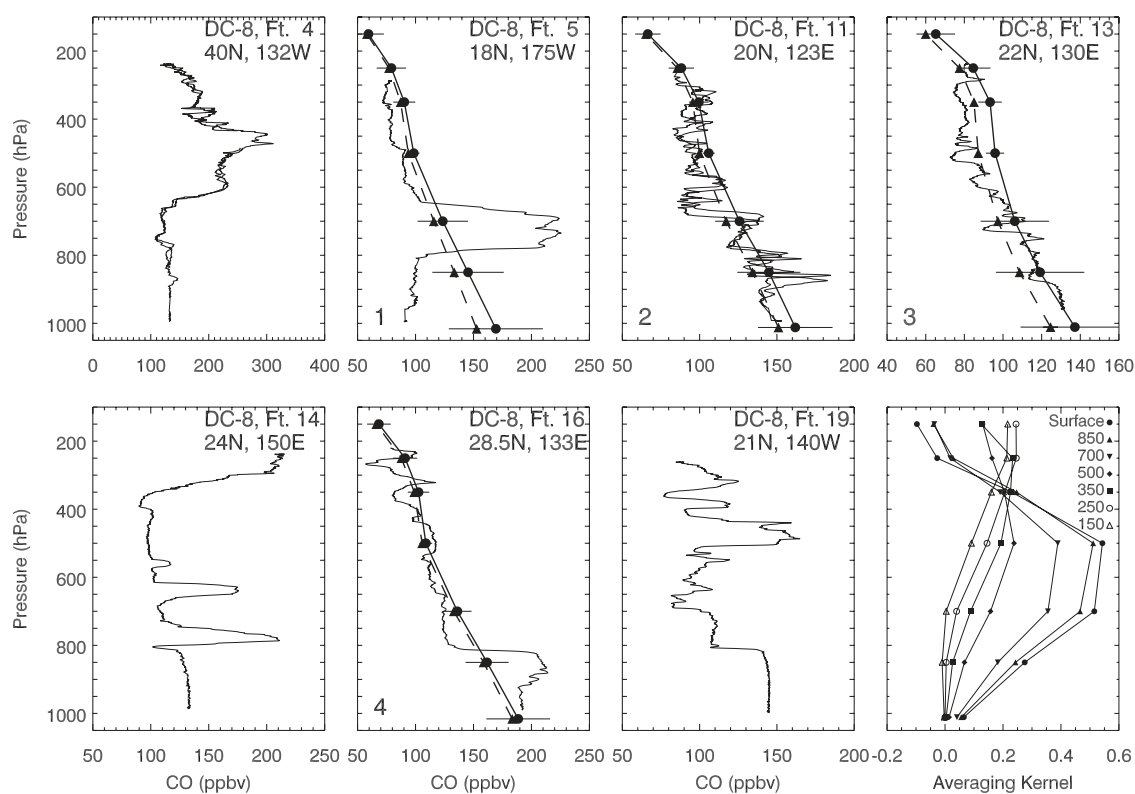


Figure 4. MOPITT CO validation profiles during TRACE-P. Thin lines show observed CO concentrations from the DC-8 spirals (double lines on flights 4 and 11 indicate double spirals; see text). The bottom right panel shows the averaging kernels for the standard pressure levels (hPa) of the MOPITT retrieval, for a typical scene over the North Pacific. Thick dashed lines show the aircraft observations convolved with the MOPITT averaging kernels (equation (1), with $G\epsilon=0$). Thick solid lines show the MOPITT data. No MOPITT data are yet available for the vertical profiles of flights 4, 14, and 19 because of complications from cloud effects.

provides a column measurement of CO weighted toward the middle and upper troposphere.

[34] A goal of TRACE-P was to enable integrated analysis of the aircraft and MOPITT data toward characterization of the Asian outflow. Aircraft validation of the MOPITT data was an essential step toward that goal. Vertical profiles for MOPITT validation were conducted on seven DC-8 aircraft flights (Figure 4). The profiles exhibited a variety of structures. Some featured large Asian plumes in the free troposphere (flights 4, 5, 14), while others were more representative of background but still had substantial structure (flights 11, 13, 16, 19). The profiles were conducted as spirals at precisely the location and time of the satellite overpass. The spirals extended from 0.15 km altitude up to the aircraft ceiling, and used a spiral diameter of 20 km matching the MOPITT field of view. Because of the midmorning timing of the satellite overpass, most validation profiles were conducted early in the flights, and fuel load limited the DC-8 ceiling to 9–11 km altitude. In addition to the vertical profiles, a 1600 km curtain was conducted on DC-8 flight 19 along the satellite orbit track to validate the horizontal gradients observed by MOPITT.

[35] Decision on whether or not to conduct a MOPITT validation profile on a given DC-8 flight was made on the day before the flight by considering the location of the MOPITT orbit in relation to the flight objectives and the

cloud forecasts. Favorable conditions were found on about half the flights, and decision to conduct a validation profile was made on half of those. Satellite cloud maps were consulted just before take off to adjust the location of the validation spiral; the broad cross-track swath of MOPITT made it easier to find clear areas. Five of the validation profiles were made under clear-sky or scattered-cloud conditions, while two (flights 4 and 19) were made deliberately over solid stratus decks. The validation curtain on DC-8 flight 19 was conducted in a subsiding air mass northeast of Hawaii and began with a vertical spiral at (21°N, 140°W), coincident with the MOPITT overpass, followed by continuous in-progress descents and ascents along the MOPITT orbit track to (37°N, 136°W).

[36] All validation profiles were done in temporal coincidence with the satellite overpass. Some temporal mismatch was unavoidable because MOPITT measures the profiles essentially instantaneously, while the DC-8 spirals take about 20 min to complete. To demonstrate that this 20-min mismatch does not introduce significant representation error, we conducted two of the validation profiles (flights 4 and 11) as double spirals at the satellite overpass point. The first spiral was initiated 20 min before the satellite overpass time, was completed close to the overpass time, and was immediately followed by a reverse spiral over the same point. We found that the vertical

profiles on the two successive spirals were highly reproducible, even in the presence of substantial structure, as shown in Figure 4. Vertical structures have sufficient persistence in the troposphere, at least outside strongly convective regions, that satisfactory temporal coincidence between satellite and aircraft can be effectively achieved.

[37] Comparisons with MOPITT retrievals for four of the seven validation profiles are shown in Figure 4. The MOPITT profiles were generated with version 3 of the MOPITT retrieval algorithm. They are averages for all pixels within a 200 km radius centered at the spiral point; this averaging was done to improve the signal-to-noise ratio. Retrievals for the other three profiles (flights 4, 14, 19) have not been attempted yet because the algorithm is not equipped to deal with cloud albedos. The MOPITT retrieval uses the maximum likelihood technique, which incorporates a priori information on the CO profile [Pan *et al.*, 1998]. A single a priori profile is used for all retrievals with concentrations decreasing monotonically from 120 ppbv at the surface to 80 ppbv at 10 km and 50 ppbv at 16 km [Deeter *et al.*, 2003]. The retrieved CO vertical profile (concentration vector $\hat{\mathbf{x}}$) is a weighted average of the true profile (\mathbf{x}) and the a priori profile (\mathbf{x}_a):

$$\hat{\mathbf{x}} = \mathbf{A}\mathbf{x} + (\mathbf{I} - \mathbf{A})\mathbf{x}_a + \mathbf{G}\boldsymbol{\varepsilon}, \quad (1)$$

where \mathbf{A} is the averaging kernel matrix (Figure 4), \mathbf{I} is the identity matrix, and $\mathbf{G}\boldsymbol{\varepsilon}$ is the retrieval error [Rodgers, 2000]. We show in Figure 4 the original aircraft profiles and the profiles smoothed by the MOPITT averaging kernels, using equation (1) with $\mathbf{G}\boldsymbol{\varepsilon} = \mathbf{0}$ (most likely value) and applying the a priori concentrations above the aircraft ceiling. As would be expected, essentially all of the vertical structure is lost in the smoothing. We choose therefore to focus the comparison on the CO columns obtained by integrating the vertical profiles over the depth of the atmosphere [Rodgers and Connor, 2003]. Results are shown in Figure 5. We find that MOPITT reproduces the absolute values and variability of the aircraft CO columns measured on the four validation profiles with a mean positive bias of $6 \pm 2\%$ and $R^2 > 0.99$. The accuracy of the aircraft data is 1%, as determined by NOAA/CMDL standards. Mean columns and standard deviations for the four profiles are $2.25 \pm 0.19 \times 10^{18}$ molecules cm^{-2} in the MOPITT data and $2.12 \pm 0.23 \times 10^{18}$ molecules cm^{-2} in the aircraft data with the averaging kernels applied. To demonstrate that this close agreement does not reflect bias introduced by application of the MOPITT averaging kernels to the aircraft data, we also show in Figure 5 the actual 950–300 hPa aircraft columns with no averaging kernels applied. The mean and standard deviation for these aircraft columns is $1.58 \pm 0.19 \times 10^{18}$ molecules cm^{-2} s^{-1} , and the correlation with the MOPITT columns is $R^2 = 0.98$. At least for these four validation profiles, we see that MOPITT preserves the variability in the original CO columns and does not induce significant bias or dampening of variability through the averaging kernels.

4. Overview of First Results

[38] We present here a brief overview and synthesis of the results presented in the first collection of TRACE-P papers

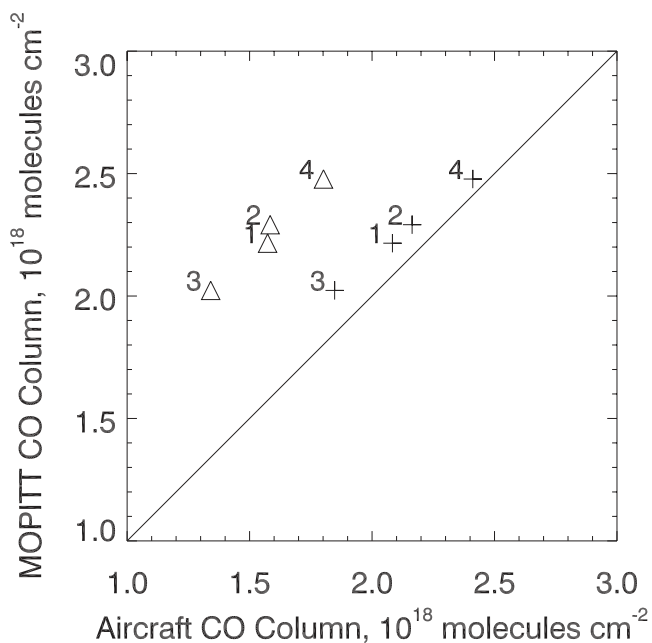


Figure 5. Scatterplot of MOPITT versus TRACE-P aircraft CO columns for the four numbered validation profiles of Figure 4. Results are shown for the aircraft data with MOPITT averaging kernels applied (pluses) and for the aircraft 950–300 hPa columns with no averaging kernels applied (triangles). The latter columns are lower because they do not include CO below 950 hPa or above 300 hPa.

assembled in this special section of *Journal of Geophysical Research*. The reader is referred to the specific papers for more information.

4.1. Instrument and Model Intercomparisons

[39] Eisele *et al.* [2003] present the ensemble of in-flight instrument intercomparisons conducted during the mission. Intercomparisons between the two aircraft showed agreement within 1–2% for ozone, CO, CO₂, CH₄, and photolysis frequencies; those measurements were made on board both aircraft by the same instrument and investigator team. Agreement within 10–20% was found for NO, PAN, and HNO₃; Zondlo *et al.* [2003] give a detailed discussion of the HNO₃ intercomparison. Measurements of NO_x, PAN, and HNO₃ aboard the P-3B accounted together for 86% of concurrently measured NO_y, indicating good closure for the NO_y chemical family [Miyazaki *et al.*, 2003]. Poorer agreement, but within stated instrument accuracies (~50%), was found for OH and HO₂. Yet poorer agreement, with discrepancies outside stated instrument accuracies and unsatisfactory correlations, were found for NO₂, SO₂, PPN, and oxygenated organics. As Eisele *et al.* [2003] point out, the discrepancies between the OH, HO₂, HCHO, and NO₂ measurements aboard the two aircraft represent a significant gap in our ability to test models of HO_x and NO_x chemistry. Fried *et al.* [2003] found that better agreement is found in the HCHO intercomparison when ensemble means rather than individual measurements are used.

[40] Thornhill *et al.* [2003] intercompared the fast 3-D air motion systems aboard the P-3B and the ACE-Asia NCAR C-130 aircraft during a wingtip-to-wingtip formation flight

from 0.3 to 3 km altitude (P-3B flight 19). This was the first ever such intercomparison between air motion systems. Results show good agreement in the variances and spectra of 3-D winds and temperature, and in the associated covariances, lending confidence in the ability of these systems to measure micrometeorological and chemical vertical fluxes.

[41] *Kiley et al.* [2003] intercompared results from seven different CTMs (four global, three regional) applied to the simulation of TRACE-P CO observations. All models used the same anthropogenic and biomass burning sources of CO. Large differences were found between the simulations, particularly in the free troposphere. Simulation of boundary layer outflow was more consistent between models, but there was general difficulty in reproducing the locations and strengths of pollution plumes. Model skill, as quantified by the correlation coefficient r between simulated and observed CO concentrations, was similar for all models ($r = 0.5-0.7$).

4.2. Asian Outflow Pathways

[42] TRACE-P demonstrated that wave cyclones and the associated cold fronts and WCBs are the principal mechanism for export of Asian pollution to the Pacific in spring. Convective outflow was less important and mainly restricted to SE Asia [*Miyazaki et al.*, 2003]. *Hannan et al.* [2003] present a detailed analysis of the mechanisms by which the wave cyclones ventilated Asian pollution to the troposphere during TRACE-P. The general mechanism involves (1) lifting of air ahead of the cold front in a southeasterly WCB and (2) boundary layer outflow behind the front capped at about 2 km by a strong subsidence inversion [*Carmichael et al.*, 2003a; *Liu et al.*, 2003]. The WCB lifting during TRACE-P took place mostly over SE Asia and China, spanning a wide range of latitudes from 15° to 42°N, and leading to ventilation of both biomass burning and anthropogenic effluents [*Miyazaki et al.*, 2003]. Mountainous terrain in SE Asia and southern China played an important role in facilitating the lifting [*Carmichael et al.*, 2003a; *Liu et al.*, 2003]. The WCB outflow was associated with considerable cloudiness, and *Crawford et al.* [2003] showed that CO concentrations in cloudy air masses during TRACE-P were on average 30% higher than in clear sky. Occasional WCBs were found to originate offshore over the NW Pacific, and *Mari et al.* (manuscript in preparation, 2003) show that these injections of marine air led to dilution and fragmentation of the continental outflow in the free troposphere. However, *Hannan et al.* [2003] found that the air lifted in these marine WCBs could also be heavily polluted by postfrontal boundary layer outflow from the previous cyclone. Downwelling of stratospheric air on the back side of the cyclones was observed [*Browell et al.*, 2003; *Miyazaki et al.*, 2003]. Such interleaving of air masses of different origins in the free troposphere resulted in great complexity in interpreting the observed relationships between species [*Talbot et al.*, 2003].

[43] Pervasive mixing of urban, biofuel, and agricultural or forest biomass burning influences was found in the continental WCBs sampled in TRACE-P. This mixing was manifest from observations of the biofuel and biomass burning tracers HCN, CH₃CN, CH₃Cl, C₂O₄²⁻, and K⁺, in combination with the urban tracers C₂Cl₄ and sulfate [*Dibb et al.*, 2003; *Ma et al.*, 2003; *Miyazaki et al.*, 2003;

Singh et al., 2003]. It was reproduced in CTM simulations of the TRACE-P data [*Carmichael et al.*, 2003a; *Liu et al.*, 2003]. *Ma et al.* [2003] used observed K⁺/sulfate ratios to infer that 30% of total aerosol mass in Asian pollution plumes was of biofuel or biomass burning origin. Biofuel and biomass burning influences were difficult to separate on the basis of chemical tracers, although there was some indication that CH₃CN is more specific for biomass burning [*Li et al.*, 2003]. The best separation was on a geographical basis, as boundary layer outflow below 2 km was essentially devoid of biomass burning influence [*Liu et al.*, 2003] but yet contained biofuel influence mixed with other anthropogenic effluents.

[44] *Koike et al.* [2003] estimated export efficiencies of NO_y and SO_x from Asia to the western Pacific by using observed correlations with CO and comparing to NO_x/CO and SO₂/CO emission ratios from *Streets et al.* [2003]. They found export efficiencies to the marine boundary layer and to the free troposphere of 20–40% and 15% for NO_y and 25–45% and 10–20% for SO_x, respectively. Most of the NO_y enhancement in the outflow was as PAN (only 0.5% as NO_x) and most of the SO_x was as sulfate. A focused analysis of WCB outflow by *Miyazaki et al.* [2003] found similar NO_x and NO_y export efficiencies. The measurements of NO_y and SO_x did not include coarse nitrate and sulfate particles, possibly causing a low bias in the estimated export efficiencies particularly for SO_x.

[45] *Heald et al.* (submitted manuscript, 2003) combined analyses of TRACE-P and MOPITT data with back trajectories and a global CTM simulation to investigate the transpacific transport of Asian pollution. They found that the Asian pollution layers sampled on the outbound transit DC-8 flights 4 and 5, over a 22° range of latitude (18°N to 40°N), were the remnants of a single Asian plume lifted in a WCB over East Asia four days earlier and split by a blocking High over the central Pacific. The northern plume continued its transport to North America and was sampled on flight 4, while the southern plume subsided over the tropical Pacific and was sampled on flight 5. Chemical tracer data in both plumes indicated a mixture of fossil fuel, biofuel, and biomass burning influences [*Blake et al.*, 2003]. Most of NO_y in the northern plume was present as PAN, consistent with the Asian outflow observations of *Koike et al.* [2003], and no significant ozone enhancement was observed in that plume. In the southern plume, by contrast, PAN decomposition in the subsiding tropical environment led to a large ozone enhancement. *Heald et al.* (submitted manuscript, 2003) found that MOPITT observations successfully tracked this transpacific pollution transport event. They went on to identify three other major events of transpacific transport of Asian pollution observed by MOPITT during the TRACE-P period. The other events were sampled over the west coast of the United States during the PHOBEA-II aircraft mission or at the Cheeka Peak ground station [*Jaffe et al.*, 2002; *Jaeglé et al.*, 2003].

4.3. Quantification of Asian Sources

[46] Several papers discuss the top-down constraints provided by the TRACE-P observations on anthropogenic sources in East Asia. The a priori bottom-up inventory of *Streets et al.* [2003] is an important starting point for that purpose. Emission estimates from that inventory were

evaluated with the TRACE-P data for a number of species by *Carmichael et al.* [2003a, 2003b] using a regional CTM simulation and by *Russo et al.* [2003] using species-species correlations. An inverse model analysis by *Palmer et al.* [2003a] combined the bottom-up inventory and the top-down constraints from the TRACE-P data, with their errors, to derive an optimized emission inventory for CO.

[47] Overall, the TRACE-P data indicate remarkable consistency with the *Streets et al.* [2003] inventory for hydrocarbons, NO_x, and SO₂. However, *Carmichael et al.* [2003a] find that CO, black carbon (BC), and other combustion gases (ethane, acetylene) are underestimated in boundary layer outflow originating from central regions of China. They attribute this underestimate to emissions from the domestic combustion sector in rural China, in particular residential coal use, and find that an increase in Chinese domestic combustion emissions by a factor of 3–5 from the bottom-up estimates would be needed to reconcile model results with observations. *Palmer et al.* [2003a] find from their inverse model that total anthropogenic CO emissions from China in the *Streets et al.* [2003] inventory are too low by 30%. A global CTM simulation of CO conducted with Chinese anthropogenic emissions 66% higher than *Streets et al.* [2003] shows no significant bias when compared with TRACE-P, MOPITT, or PHOBEA-II data [*Jaeglé et al.*, 2003; *Li et al.*, 2003; Heald et al., submitted manuscript, 2003]. Previous inverse model analyses using sparse surface observations from the NOAA-CMDL network had suggested even higher anthropogenic CO emissions from East Asia [*Kasibhatla et al.*, 2002; *Pétron et al.*, 2002].

[48] Relating the biomass burning influence observed in TRACE-P to bottom-up emission estimates requires time-specific information on fires. *Heald et al.* [2003] constructed a daily global biomass burning inventory for the TRACE-P period by using AVHRR satellite fire data to scale the *Duncan et al.* [2003] climatological biomass burning inventory. Biomass burning emissions during TRACE-P were highest in Thailand, Cambodia, and northeast India. There was little burning in China and all of it was confined to the southern end of the country. Burning in SE Asia in February–April 2001 was close to the climatological average defined by the TOMS Aerosol Index. CTM simulations of the TRACE-P and MOPITT data using the *Duncan et al.* [2003] climatological inventory with day-to-day scaling from *Heald et al.* [2003] indicate that SE Asian biomass burning emissions of CO in that inventory are overestimated, perhaps by 50% or more [*Palmer et al.*, 2003a; Heald et al., submitted manuscript, 2003].

[49] *Palmer et al.* [2003b] used the halocarbon-CO correlations measured in TRACE-P, combined with back trajectories, to infer the sources of different halocarbons from East Asian countries. They derived an East Asian source of CCl₄ of 18.5 Gg yr⁻¹, considerably larger than government estimates [*United Nations Environmental Program (UNEP)*, 2002]. Sources of CH₃CCl₃ were mainly from central China and the Seoul area and were consistent with government estimates. Halon-1211 had a strong source from the Shanghai area and no apparent sources from elsewhere. *Blake et al.* [2003] identified large point sources of CH₃Br from Seoul and Tokyo.

[50] TRACE-P provided extensive data on the Asian outflow of CO₂ and CH₄. *Vay et al.* [2003] found that the Asian continent was a net source of CO₂ during TRACE-P, as would be expected for that season. By combining the observed CO₂ enhancements and vector winds, they derived a net CO₂ source from the Asian continent of 14 Tg d⁻¹ for the period. The *Streets et al.* [2003] inventory estimates a corresponding Asian source of CO₂ of 6 Tg d⁻¹ from anthropogenic activities and biomass burning, and *Vay et al.* [2003] attributed the remaining 8 Tg d⁻¹ to respiration. *Bartlett et al.* [2003] examined the constraints offered by the TRACE-P data on the Asian sources of CH₄. Livestock and rice paddies provide the principal source in current inventories [*Streets et al.*, 2001], but TRACE-P was conducted in the rice planting season when that source is minimum. *Bartlett et al.* [2003] found that CH₄ in TRACE-P had a strong and consistent relationship with C₂H₆, and to a lesser extent with CO, suggesting a dominant urban source (possibly from fossil fuel use or landfills) which might however be correlated with livestock.

[51] *Singh et al.* [2003] and *Li et al.* [2003] used the TRACE-P HCN and CH₃CN measurements aboard the DC-8 to constrain sources and sinks for these two gases. These were the first in situ HCN measurements in the troposphere and were shown by *Singh et al.* [2003] to be consistent with previous spectroscopic measurements of HCN columns at Japanese sites. The TRACE-P data demonstrated a dominant biomass burning source and ocean sink (lifetime of 5–6 months) for both gases. Urban air masses sampled in southern California during test and transit flights showed no enhancements, confirming previous laboratory findings that the sources from fossil fuel combustion are negligible. However, Chinese urban plumes showed large HCN enhancements without concurrent CH₃CN enhancements, an observation that *Singh et al.* [2003] attributed tentatively to residential combustion. A global CTM budget analysis by *Li et al.* [2003] shows that a combination of biomass burning and residential combustion sources, together with an ocean sink, allow a good simulation of the HCN and CH₃CN observations in TRACE-P, of their correlations with CO, and also of previous HCN column and CH₃CN aircraft measurements.

4.4. Aerosol Outflow

[52] Aerosol outflow from Asia during TRACE-P was mostly below 2 km altitude, although there were also major Asian aerosol influences in the middle troposphere [*Browell et al.*, 2003; *Jordan et al.*, 2003b]. The largest aerosol enhancements were associated with postfrontal boundary layer outflow from China. Soot accounted on average for 2% of total aerosol mass in Chinese outflow and 12% in outflow from SE Asia [*Jordan et al.*, 2003b].

[53] Even though no major Asian dust events occurred during TRACE-P, *Dibb et al.* [2003] found that Ca²⁺ (a dust tracer) was a major contributor to the total cation loading in Asian outflow, and *Jordan et al.* [2003b] found that dust accounted for 77% of total aerosol mass in the low-altitude Chinese outflow. *Clarke et al.* (unpublished manuscript, 2003) and *Jordan et al.* [2003a] showed that the dust was generally mixed with pollution and that a large fraction

of sulfate and nitrate was taken up by dust particles, presumably through displacement of carbonate. Sulfate was typically in excess of ammonium, but the high dust concentrations kept the total aerosol in a near-neutral state [Dibb *et al.*, 2003]. Dust led to $\text{NO}_3^-/\text{HNO}_3(\text{g})$ ratios in excess of unity in Chinese outflow, in contrast to much lower ratios in background air [Jordan *et al.*, 2003a]. Clarke *et al.* (unpublished manuscript, 2003) point out that dust reduces the radiative effects of anthropogenic Asian aerosols by promoting their collection and the condensation of aerosol precursors on the large dust particles.

[54] Weber *et al.* [2003] found that new particle formation took place in Chinese and Korean urban plumes sampled during TRACE-P, but not in biomass burning and volcanic plumes. They show evidence that ternary $\text{NH}_3\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation was responsible for the new particle formation in urban plumes. The submicron aerosol in these plumes was principally a neutralized $\text{H}_2\text{SO}_4\text{-HNO}_3\text{-NH}_3\text{-H}_2\text{O}$ mixture, implying the presence of gas-phase NH_3 to drive the ternary nucleation process.

4.5. Chemical Evolution of Asian Outflow

[55] Better understanding of radical photochemistry in the Asian outflow was a major objective of TRACE-P. Cantrell *et al.* [2003] compared their peroxy radical measurements aboard the P-3B to standard photochemical model calculations constrained with the ensemble of aircraft observations including photolysis frequencies. They found good agreement over the range of conditions encountered in the mission. There were no apparent discrepancies associated with concentrated plumes or high aerosol levels. The good agreement extended to relationships of the peroxy radicals with their precursors, in particular NO_x . Fried *et al.* [2003] found general agreement between their HCHO measurements and photochemical model calculations, although there were some major disagreements for portions of the data set and also some apparent difficulties in the simulation of CH_3OOH concentrations under low- NO_x conditions. Fried *et al.* [2003] also found evidence of significant uptake of HCHO by clouds.

[56] The above results offer some encouragement that standard photochemical models may be used for describing HO_x and NO_x chemistry in Asian outflow. However, a persistent concern for model evaluation is the disagreement between HO_x and HCHO measurements by different instruments (section 4.1). In addition, Lefer *et al.* [2003] and Tang *et al.* [2003a] show that proper accounting of the optical effects of clouds and aerosols is essential. Aerosol extinction coefficients at 550 nm during TRACE-P were typically $\sim 10^{-4} \text{ m}^{-1}$ in the boundary layer, dropping to $\sim 10^{-6} \text{ m}^{-1}$ in the upper troposphere, and the corresponding total aerosol optical depths in the Asian outflow were typically in the range 0.2–0.6. Mean single-scattering albedos were 0.76 in dust events, 0.80 in outflow from SE Asia, and 0.84–0.93 in air masses of other origins [Jordan *et al.*, 2003b; Clarke *et al.*, unpublished manuscript, 2003]. The aerosol was sufficiently absorbing that it decreased photolysis frequencies on average by 35–40% near the surface and by about 20% in the free troposphere relative to aerosol-free conditions [Lefer *et al.*, 2003]. The radiative effects of aerosols caused significant decreases in HO_x concentrations (OH down by 40% in the boundary layer) relative to model

calculations assuming aerosol-free conditions [Tang *et al.*, 2003a].

[57] The TRACE-P observations indicate fast chemical aging of NO_x and reactive organic gases in the Asian outflow. Talbot *et al.* [2003] found that NO_x concentrations in Chinese urban plumes sampled 1–2 days downwind of their sources were generally less than 100 pptv, and NO_x/NO_y concentration ratios were typically less than 10%. Rapid depletion of NO_x was also observed in the WCB outflows, with NO_x amounting to only 3% of NO_y on average [Koike *et al.*, 2003; Miyazaki *et al.*, 2003]. Even in the fresh Shanghai plume (~ 18 hours old) sampled on DC-8 flight 13, NO_x accounted for only 15% of total NO_y [Talbot *et al.*, 2003]. Simpson *et al.* [2003] found that ethylnitrate/ethane and 2-propylnitrate/propane concentration ratios were much higher than would be expected from ethane and propane chemistry, with no such discrepancy found for other alkyl nitrates. Oxygenated organic emissions missing from current inventories might be responsible for the missing sources of ethylnitrate and 2-propylnitrate; such emissions could possibly have important implications for explaining the high concentrations of acetaldehyde and acetone previously reported in the remote troposphere by Singh *et al.* [2001].

[58] Rapid ozone production was observed in the SE Asian biomass burning plumes sampled during TRACE-P [Tang *et al.*, 2003b]. This biomass burning enhancement was also apparent in the Chinese ozonesonde data from Hong Kong and Kunming [Chan *et al.*, 2003b] and in the ensemble of TRACE-P DIAL ozone data south of 28°N [Browell *et al.*, 2003]. High ozone (up to 140 ppbv) was observed in the Shanghai urban plume [Russo *et al.*, 2003]. However, boundary layer outflow from China, Korea, and Japan was found in general not to be associated with elevated ozone, presumably because of weak UV fluxes at that time of year. Dibb *et al.* [2003] saw little correlation between ozone and either ^7Be or ^{210}Pb , suggesting that neither the stratosphere nor regional photochemistry over Asia were major sources of ozone in the Asian outflow. It appears that the overall ozone budget during TRACE-P mostly reflected the hemispheric-scale buildup of tropospheric ozone during spring, rather than processes taking place on the regional scale. A CTM simulation by Pierce *et al.* [2003] reproduces the observed seasonal buildup in the TRACE-P region and attributes it to increasing photochemical production, but finds little net outflow of ozone from the region.

[59] Considerable structure was found for ozone in the free troposphere during TRACE-P [Wild *et al.*, 2003]. An analysis of the high-ozone layers by Newell *et al.* (unpublished manuscript, 2003) indicates two principal sources: Asian outflow lifted in low-latitude WCBs and stratospheric influence on the back side of cyclones. Ozonesonde observations at Lin'an show strong stratospheric influence in the upper troposphere, in contrast to Kunming or Hong Kong which show no such influence [Chan *et al.*, 2003b]. A CTM simulation by Wild *et al.* [2003] reproduces many features of the observed stratospheric influence on ozone in the upper troposphere.

4.6. Comparison to PEM-West B

[60] Comparisons of the TRACE-P observations with those from PEM-West B, conducted in the same region in

February–March 1994, are presented in several papers. The bottom-up inventory of *Streets et al.* [2003] indicates little difference between 1994 and 2001 anthropogenic emissions in East Asia (section 2.5), reflecting the leveling off of emissions in the late 1990s due to the Chinese economic downturn, industrial restructuring, and environmental controls [*Streets et al.*, 2001]. Springtime biomass burning emissions in SE Asia were of comparable magnitude in 1994 and 2001 according to the TOMS Aerosol Index [*Heald et al.*, 2003]. PEM-West B was conducted under warm-phase ENSO (El Niño) conditions, whereas TRACE-P was conducted under cold-phase to neutral ENSO conditions. *Liu et al.* [2003] report a much greater frequency of cold frontal passages during TRACE-P than during PEM-West B, reflecting the difference in ENSO phase. Weather in Hong Kong was sunnier and drier in TRACE-P than in PEM-West B [*Wang et al.*, 2003]. Another factor of difference for photochemically active species is the seasonal phase lag, as TRACE-P was shifted ~ 3 weeks later in spring than PEM-West B. Finally, differences in flight strategies between the two missions could be a complicating factor. PEM-West B had other objectives besides the characterization of Asian outflow, and this affected in particular the tropical flights; however, the priority for the extratropical flights was the sampling of Asian outflow (D. D. Davis, personal communication, 2002).

[61] *Blake et al.* [2003] and D. D. Davis et al. (Trend in western North Pacific ozone photochemistry as defined by observations from NASA's PEM-West B (1994) and TRACE-P (2001) field studies, manuscript in preparation, 2003, hereinafter referred to as Davis et al., manuscript in preparation, 2003) found no significant difference in CO concentrations between PEM-West B and TRACE-P, suggesting that the two missions indeed provided similar general characterizations of the Asian outflow. They found lower concentrations of C₂–C₆ hydrocarbons in TRACE-P; relative differences correlated with species lifetimes and could thus be explained by the seasonal phase lag between the two missions. Ozone concentrations were on average 15 ppbv higher in TRACE-P, a large trend that Davis et al. (manuscript in preparation, 2003) investigated in detail through comparative analysis with ozonesonde records and DIAL observations, and concluded could be explained by the seasonal phase lag. *Wang et al.* [2003] also found a 14 ppbv increase in ozone at Hong Kong between the PEM-West B and TRACE-P sampling periods. *Browell et al.* [2003] found a large increase in potential vorticity over the NW Pacific between PEM-West B and TRACE-P, and concluded that both increased stratosphere-troposphere exchange and photochemical production contributed to the higher ozone in TRACE-P.

[62] *Blake et al.* [2003] and *Russo et al.* [2003] reported a decline in the concentrations of most halocarbons between PEM-West B and TRACE-P, qualitatively consistent with the expected decline in anthropogenic emissions. Halon-1211, a gas used in fire-fighting equipment and for whom China is thought to account for 90% of global production, increased by 50% from PEM-West B to TRACE-P and was a useful marker for air masses of Chinese origin [*Blake et al.*, 2003]. No significant trend was found for CH₃Cl, which has a dominant source from biofuels and biomass burning [*Blake et al.*, 2003]. Methane concentrations in TRACE-P

were higher than in PEM-West B, reflecting the trend in the background, but a detailed analysis by *Bartlett et al.* [2003] indicates that the Asian enhancement over background was similar in both missions, implying no detectable change in Asian emissions.

[63] The largest differences between PEM-West B and TRACE-P were found for aerosols and soluble gases in the boundary layer. Comparisons of DIAL vertical cross sections indicated much stronger boundary layer aerosol outflow in TRACE-P than in PEM-West B [*Browell et al.*, 2003]. In situ measurements indicated much higher dust influence in TRACE-P [*Dibb et al.*, 2003]. *Dibb et al.* [2003], *O'Sullivan et al.* [2003], and *Talbot et al.* [2003] found that concentrations of NO_y species, SO₂, and water-soluble aerosol in the Asian boundary layer outflow were 2–5 times higher in TRACE-P than in PEM-West B. These results are consistent with a greater frequency of frontal passages in TRACE-P [*Liu et al.*, 2003], delivering polluted Asian air to the marine boundary layer in the postfrontal outflow with minimal scavenging. Concentrations of H₂O₂ were 2.5 times lower in TRACE-P than in PEM-West B, possibly because of stronger outflow of SO₂ in TRACE-P providing a sink for H₂O₂ through reaction in clouds [*O'Sullivan et al.*, 2003].

5. Conclusions

[64] The NASA TRACE-P two-aircraft mission in February–April 2001 was designed to observe the Asian outflow of environmentally important gases and aerosols to the Pacific, to relate this outflow quantitatively to its sources, and to understand its near-field chemical evolution over the Pacific. Quantifying the contributions from large geopolitical source regions to global atmospheric composition is presently a major issue for atmospheric chemistry research. A priori information from bottom-up emission inventories must be tested and improved using top-down constraints from atmospheric observations. In the case of short-lived species (ozone, aerosols, and their precursors), the role of continental boundary layer processing in determining the outflow must be understood. It is increasingly recognized that an integrated observational strategy is needed involving satellites, aircraft, ground stations, and chemical transport models (CTMs). TRACE-P aimed to contribute to the development of such a strategy.

[65] The aircraft payload in TRACE-P included extensive instrumentation for measurements of greenhouse gases, ozone and its precursors, aerosols and their precursors, related species, and chemical tracers. Planning prior to the mission identified the major Asian outflow pathways and chemical signatures to be expected and investigated. This planning involved the construction of a customized a priori emission inventory for East Asia in 2000 based on socioeconomic data and emission factors, and the use of five different CTMs to provide hindcasts of Asian chemical outflow for previous meteorological years. During the mission, the same CTMs provided chemical forecasts to guide the aircraft toward regions of outflow and thus provide the best data for testing both the a priori emission inventories and the models of chemical evolution. After the mission, results from the CTMs were intercompared to quantify the range of model error. A biomass burning

inventory with daily resolution was constructed from satellite fire counts to provide further constraints on emissions.

[66] The mission thus provided a detailed characterization of the pathways for Asian outflow to the Pacific. It showed that wave cyclones and the associated cold fronts are the principal mechanism for export of Asian pollution to the Pacific in spring. This mechanism involves both warm conveyor belts (WCBs) rising in the free troposphere ahead of the fronts and postfrontal boundary layer outflow capped at ~ 2 km by strong subsidence inversions. Frontal passages were unusually frequent in spring 2001. Fuel and industrial pollution from East Asia, biomass burning effluents from Southeast Asia, and tropical maritime air were mixed in the WCB outflow and resulted in complicated chemical signatures. In contrast, the postfrontal boundary layer outflow was dominated by fuel and industrial pollution and contained the highest concentrations observed in the mission (up to 1.4 ppmv CO and 140 ppbv O₃ in the Shanghai plume). Deep convection during TRACE-P was largely confined to Southeast Asia and the associated upper tropospheric plumes had dominant biomass burning influences. Long-range transport of European and African (biomass burning) effluents did not produce detectable enhancements in the Asian outflow. Export efficiencies of NO_y and SO_x from the Asian continent to the free troposphere were estimated to be 10–20%.

[67] The observations from TRACE-P generally confirmed the high quality of the a priori inventories of Asian anthropogenic emissions generated by *Streets et al.* [2003]. These inventories cover a wide range of species including CO₂, CO, CH₄, higher hydrocarbons, reactive nitrogen species, sulfur oxides, and carbonaceous aerosols. The largest discrepancies appeared to reflect domestic emissions in rural China (notably residential coal combustion), with the TRACE-P observations implying much higher values than the a priori estimates. Previous top-down estimates of CO emissions from East Asia, obtained by inverse modeling of surface observations from the NOAA/CMDL network, appear to be too high in light of the TRACE-P observations. Current biomass burning emission inventories for SE Asia also appear to be too high. The TRACE-P observations in Chinese urban plumes indicate high concentrations of a number of compounds (including CH₃Cl, COS, HCN) that are not normally found in urban plumes of the developed world. They could possibly originate from domestic combustion sources but more information is needed. Halocarbon-CO correlations measured in TRACE-P indicate that the CCl₄ source from East Asia is much higher than government estimates. Large sources of halon-1211 (China) and CH₃Br (Japan, Korea) were also identified. Observations of HCN and CH₃CN demonstrated a dominant biomass burning source and ocean sink for both.

[68] Intercomparison of measurements of HO_x radicals and HCHO aboard the TRACE-P aircraft indicated some major discrepancies that limit the extent to which photochemical model calculations can be evaluated. Within these uncertainties, it was found that current photochemical models can reproduce the HO_x chemistry observed in Asian outflow, although proper consideration of aerosol attenuation of UV radiation is critical. Aerosol optical depths in Asian outflow during TRACE-P were typically in the range 0.2–0.6, and single-scattering albedos were relatively low, result-

ing in major attenuation of photolysis frequencies by aerosols. Both dust and soot contributed to aerosol absorption.

[69] Photochemical ozone enhancements in TRACE-P were observed in biomass burning plumes and occasionally in urban plumes but were otherwise weak, reflecting the early spring timing of the mission. Significant buildup of ozone over the course of the TRACE-P mission appeared to reflect the general behavior of tropospheric ozone at extratropical northern latitudes, rather than regional production. TRACE-P observations on the outbound transit flights indicated no significant ozone production in Asian pollution plumes transported across the Pacific at northern midlatitudes. However, large ozone production driven by PAN decomposition was observed in subsiding plumes transported to the tropics. Considerable layering of ozone was observed in the upper troposphere in association with stratospheric intrusions.

[70] Although no major dust events were sampled during TRACE-P, dust was a pervasive component of the aerosol in Asian outflow. A large fraction of total sulfate and nitrate in the outflow was associated with dust particles. New particle formation during TRACE-P was found to take place in urban plumes but not in biomass burning or volcanic plumes. The submicron aerosol in urban plumes was found to consist principally of ammonium sulfate and ammonium nitrate, and the formation of new particles was ascribed to ternary NH₃-H₂SO₄-H₂O nucleation.

[71] Comparison of observations from TRACE-P and PEM-West B (February–March 1994) indicated similar CO concentrations, and trends in ozone and hydrocarbons consistent with the seasonal phase lag between the two missions. The *Streets et al.* [2003] inventory reports only small trends (<20%) in Asian anthropogenic emissions between 1994 and 2001; the TRACE-P data are consistent with this view. Concentrations of most halocarbons decreased from PEM-West B to TRACE-P, consistent with the reductions mandated by the Montreal Protocol. However, concentrations of aerosols and water-soluble gases (SO₂, NO_x) sampled in the lower troposphere were several-fold higher in TRACE-P than in PEM-West B. This could possibly reflect the greater frontal activity in TRACE-P, with postfrontal boundary layer transport delivering water-soluble species over the western Pacific with minimum scavenging.

[72] Satellite observations are becoming increasingly important as a source of information for tropospheric chemistry, and TRACE-P provided a test bed to explore the integration of aircraft and satellite observations toward addressing mission objectives. Of particular interest were the MOPITT measurements of CO aboard the Terra polar-orbiting satellite. Validation profiles for MOPITT were conducted on several TRACE-P flights, taking advantage of the high ceiling of the DC-8 aircraft. Comparison of CO columns measured on four of these profiles indicates high correlation between TRACE-P and MOPITT ($R^2 > 0.99$, $n = 4$) and only a small high bias ($6 \pm 2\%$) in the MOPITT data. The MOPITT data support the TRACE-P findings that a priori estimates of anthropogenic emissions from CO were too low while biomass burning emissions were too high. Extensive transpacific transport of Asian pollution encountered on the TRACE-P outbound transit flights was also observed by MOPITT.

[73] The TRACE-P observations provide a remarkably rich data set for investigating chemical outflow from the Asian continent. The results presented in this special TRACE-P section of the *Journal of Geophysical Research* represent only a first pass at mining the data. We look forward to further analyses of the data by the atmospheric chemistry research community over the coming years. (The data are available at <http://www-gte.larc.nasa.gov>.)

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