Tropical Deep Convection and Ozone Formation

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Abstract

Theoretical studies, aircraft and space-borne measurements show that deep convection can be an effective conduit for introducing reactive surface pollutants into the free troposphere. The chemical consequences of convective systems are complex. For example, sensitivity studies show potential for both enhancement and diminution of ozone formation. Field observations of cloud and mesoscale phenomena have been investigated with the Goddard Cumulus Ensemble and Tropospheric Chemistry models. Case studies from the tropical ABLE 2, STEP and TRACE-A experiments show that free tropospheric ozone formation should increase when deep convection and urban or biomass burning pollution coincide and decrease slightly in regions relatively free of ozone precursors (often marine). Confirmation of post-convective ozone enhancement in the free troposphere over Brazil, the Atlantic and southern Africa was a major accomplishment of the September-October 1992 TRACE-A (Transport and Atmospheric Chemistry near the Equator - Atlantic) aircraft mission. A flight dedicated to cloud outflow showed that deep convection led to a factor of 3-4 increase in upper tropospheric ozone formation downwind. Analysis of ozonesondes during TRACE-A was consistent with 20-25% of seasonally enhanced ozone over the south Atlantic being supplied by a combination of biomass burning emissions, lightning and deep convection over South America. With the tropics the critical region for troposphere-to-stratosphere transfer of pollutants, these results have implications for the total ozone budget. Cloud-scale analyses will guide the development of more realistic regional and global chemical-transport models to assess the full impact of deep convection on atmospheric chemical composition.

1. Introduction

The importance of deep convection in atmospheric chemistry was first suggested by Crutzen and coworkers [Gidel, 1983; Crutzen and Gidel, 1983; Chatfield and Crutzen, 1984; Chatfield and Delany, 1990]. Deep convection can remove pollutants from the lower atmosphere and inject them rapidly into the middle and upper troposphere, and occasionally into the stratosphere. Tropopause penetration occurs in ~ 10% of tropical convective events (B. Carlson, personal communication, 1996). Pollutants transported vertically in core updrafts may exit clouds wherever there is detrainment, including from cloud anvils in the upper troposphere. One of the first observations of this phenomenon was made during PRE-STORM flights over Oklahoma in 1985 [Dickerson et al., 1987]. In the vicinity of convective anvils, concentrations of carbon monoxide (CO) at 9-11 km were similar to CO concentrations in the rural boundary layer (Fig. 1). Calculations suggest that over the central US in summertime, approximately half the CO entering the boundary layer is transported upward by deep convection [Thompson et al., 1994]. A benefit is that convection acts as a chimney, preventing concentrations of CO and other pollutants at the surface from reaching unhealthy levels. However, once aloft, CO is advected long distances where it contributes to ozone production. Photochemical calculations using the Goddard Tropospheric Chemistry model constrained by data from PRESTORM flights (1985) showed significant enhancements in ozone production downstream of convective events [Pickering et al., 1990]. A global chemistry-transport model indicates that up to 50% of ozone exported off the northeast United States during summertime could originate from post-convection ozone formation [Jacob et al., 1993].

Although ozone in the boundary layer ("smog") threatens agriculture and human health, ozone in the middle and upper troposphere acts as a greenhouse gas. Thus, concerns about the link between deep convection and ozone formation are global. The tropics are of particular interest because of the coexistence of deep convection and widespread pollution from biomass burning and rapid industrial development. The tropical impact of deep convection can be seen from space. The MAPS (see Acronym List at end of Article) instrument on the Space Shuttle [Reichle et al., 1986; Fishman, 1991; http:// stormy.larc.nasa.gov/press.html] detects mid-tropospheric CO over the Atlantic (and Indian Oceans) far from its source in savanna burning regions of South America and Africa (and southeast Asia).

The net effect of deep convection on free tropospheric ozone awaits global chemical-transport models with realistic treatments of convection and other processes that affect trace gas chemistry. Simulations must capture downdrafts as well as updrafts, stratosphere-troposphere exchange -- all sub-grid scale -- as well as chemical reactions that can occur on and inside aerosols and hydrometeors [Lelieveld and Crutzen, 1990]. The required simulations are not yet available. Neither are global maps of tropospheric ozone, CO and NO_2 available for model input and validation, although improved sensors for these constituents are forthcoming from satellite instruments like the EOS MOPITT and TES.

In the meantime, researchers are using smaller-scale models and aircraft data to evaluate the chemical consequences of deep convection. This paper summarizes tropical studies that use the cloud-resolving Goddard Cumulus Ensemble (GCE) model with the Goddard Tropospheric Photochemical model. Initial studies based on data from ABLE 2 (1985 and 1987 experiments), in some cases, suggested substantial enrichment of upper tropospheric ozone following deep convection; however, post-convective upper tropospheric observations were lacking (Section 2). A flight from Brasília dedicated to convective outflow during the GTE/TRACE-A field campaign in September 1992 confirmed the convection-ozone link and better defined the magnitude of the impact (Section 3).

2. PRE-TRACE-A Tropical Studies

The Goddard Cumulus Ensemble (GCE) model (Tao and Simpson, 1993) has been used to simulate the evolution of cloud systems observed in the field. Both two- and three- dimensional versions of the GCE have been used. Initialization is obtained from a sounding of temperature, winds, and water vapor. The model is nonhydrostatic, using a stretched vertical grid (finer resolution near the surface) up to ~20 km; the horizontal domain is typically 250-1000 km. The model uses a detailed microphysical scheme, with 2-phase liquid water and a three-phase ice scheme, and appropriate radiation. Simulations are usually made for a 6-8 hour period.

Winds from the 2D and 3D simulations are subsequently used in tracer transport calculations. Cloud dynamics are easily visualized by running trajectories periodically from a given location or simultaneously from multiple locations.

The pathways of air entrained at the bottom of the leading edge of the cloud, as simulated for a system observed in Amazonia, Brazil in April 1987 during the GTE/ABLE 2B expedition, are traced in Fig. 2a. Along some parcel trajectories, transport from 0.3 to 15 km took 30 minutes or less. Fifty percent of trajectories reached 12 km or more. Significant detrainment from the rear of the cloud at lower altitudes (25% of trajectories) and rotor circulation involved 15% of the trajectories. Back trajectories from the anvil outflow region in an ABLE 2A episode (3 August 1985; Fig. 2b) show that all air parcels detrained at 10 km originated in the boundary layer. The pre-convection equivalent potential temperature profiles of the two ABLE events (Fig. 2c) explain some of the difference in their dynamical structure. The wet season event (26 April 1987), with a less pronounced minimum in the profile, developed weaker downdrafts and updrafts than the 3 August 1985 ABLE 2A event. Studies of five tropical episodes with the GCE (summarized in Pickering et al., 1994) show that the complexity illustrated in Fig. 2 is typical. Highly variable dynamical patterns preclude simple generalizations about chemical transport. An ABLE 2B event less than two weeks after the 26 April 1987 episode showed even greater lower-level detrainment and over-turning [Scala et al., 1990].

Tracer simulations of the 26 April 1987 ABLE 2B event (Fig. 3) show substantial alteration in the vertical distributions of chemical constituents. Fig. 3 shows a two-dimensional profile of NO_x ($NO_x = NO + NO_2$) after 4 hours of simulation. The initial profile was taken from NO measurements to 6 km made on the NASA Electra aircraft and includes measurements of the Manaus, Brazil, urban plume. The normal gradation (highest NO_x near surface and in upper troposphere) is altered as successive clouds form along the squall line, giving rise to "cloud redistribution" of the trace gas.

Nitric oxide, which forms in the boundary layer from combustion (auto exhaust and biomass burning are primary tropical sources) usually has the greatest impact on ozone formation. Lightning is an important middle and upper tropospheric NO source, with cloud-to-ground lightning possibly a boundary layer source as well. Ozone, O_3, forms from the following sequence of reactions:

$NO + HO_2 \text{-->} NO_2 + OH$ $\qquad\qquad$ $NO + RO_2 \text{-->} NO_2 + RO$

$$NO_2 + h\nu \text{ -->} O + NO$$

$$O + O_2 + M \text{-->} O_3 + M$$

where the right-hand NO to NO_2 conversion is initiated by free radicals, RO_2, formed from hydrocarbons.

To quantify the effects of deep convection on free tropospheric ozone, net ozone formation rates pre- and post-convection are computed. These are calculated with the one-dimensional Goddard Tropospheric Chemistry model, using profiles derived from the tracer model. A 24-hour averaged rate of net O_3 production, referred to as the "Ozone Production Potential" (OPP), is computed. Ozone formation requires NO and hydrocarbons. Net O_3 formation is given by:

$$P(O_3) = k_1[NO][HO_2] + \Sigma\, k_i[NO][R_iO_2]$$
$$- [O_3]\{k_3[OH] + k_4[HO_2]\} - k_5[O(^1D)][H_2O],$$

where the symbol R_iO_2 signifies free radicals derived from hydrocarbon oxidation. The last three terms in the equation represent ozone losses. Tracer model output, as in Fig. 3, is used for chemical model profiles for O_3, NO_x, CO, H_2O, and hydrocarbons, usually from initial (undisturbed) conditions and after 4-6 hours of convection (post-convection). The free radicals are calculated self-consistently using the mechanism described in Pickering et al [1990; update in Thompson et al., 1996b].

Even when the net effect of convection is to redistribute the original amount of material within a convective cell, more ozone may form after convection than under non-convective conditions. The reason is that the net formation of ozone, as shown in the expression for $P(O_3)$, is a non-linear process with respect to NO_x concentrations. At extremely low concentrations of NO (< 10-20 parts per trillion), as in the boundary layer or free troposphere far from sources, $P(O_3)$ is negative, ie ozone is destroyed. As NO_x concentrations increase up to ~1 ppbv, ozone formation is positive. An opposing tendency, however, is that the ozone-forming efficiency per molecule decreases at the same concentration range at which ozone forms. This is because at higher ozone levels, more OH and HO_2 radicals are produced, leading to loss (first negative terms in the $P(O_3)$ expression).

In the case of the urban plume sampled during the 26 April 1987 episode, the contrast in mid-tropospheric undisturbed and post-convection NO_x in Fig. 3 leads to increased ozone production in the mid- to upper troposphere. A comparison of ozone formation rates in the free troposphere (Fig. 4) shows more rapid production of ozone with cloud-perturbed conditions.

The ratio of O_3 production potential with and without convection is designated the Convective Enhancement Factor (CEF):

$$\frac{\mathrm{OPP}^{24\text{-hr}}{}_{\text{Post-convection}}}{\mathrm{OPP}^{24\text{-hr}}{}_{\text{No convection}}} = \mathrm{CEF}$$

Because of the nonlinear effects of NO_x on ozone, convective redistribution of ozone and its precursors can lead to CEF greater or less than unity. In Fig. 4, a comparison of columnintegrated O_3 formation from 4-15 km yields CEF = 35. This is expected to be an upper limit because low NO conditions observed below 6 km, were extended upward in the model. If lightning, for example, or NO_x-rich layers from previous storms had increased upper tropospheric ozone, the CEF would have been lower. A reduction in upper tropospheric O_3 production (CEF < 1.0) was estimated for an episode sampled near Darwin, Australia, during STEP (2 February 1987) in which boundary layer air with very low NO_x was transported upward. This is probably representative of vast unpolluted marine regions as suggested by Lelieveld and Crutzen [1994].

A summary of CEFs for five tropical systems analyzed with the Goddard models is given in Table 1. Both chemical and dynamical variability contributed to a wide range of CEFs. In general:

1) The pre-convection equivalent potential temperature profile is a good predictor of up and downdraft structure and outflow characteristics. More vigorous up- and downdrafts result when the θ_e profile has a pronounced mid-tropospheric minimum.

2) Maximum outflow in the middle troposphere tends to be most effective for enhanced O_3 formation because the middle tropospheric NO_x photochemical lifetime is increased over its lifetime in the boundary layer. Ozone forms during transport downwind from the convective event. Overturning to the boundary layer (extensive in the ABLE 2B event of 6 May 1987) and detrainment in the (cold) upper troposphere suppress net O_3 formation.

3) All events simulated were limited in O_3 formation by the NO_x concentration; maximum CEF results from the greatest increase in post-convective mid-tropospheric NO_x compared to NO_x concentrations prior to convection. This occurs when emissions from urban or biomass burning activities are injected into a previously clean troposphere.

3. TRACE-A Flight 6

For the pre-1990 tropical cases studied with the Goddard models, there were very few measurements of ozone precursors or ozone in upper tropospheric convective outflow. Aircraft measurements in cloud outflow layers over a pollution source were sought to confirm convective enhancement of ozone. In particular, it was hoped that the GTE/TRACE-A DC-8 aircraft mission would give a flight of opportunity. TRACE-A (Transport and Atmospheric Chemistry near the Equator - Atlantic), as the name suggests, was an experiment designed to determine the interaction of chemistry and dynamics in causing a seasonal maximum in tropospheric ozone that appears each September and October over the south Atlantic Basin [Fishman et al., 1996]. A motivating hypothesis for TRACE-A was that widespread savanna burning over southern Africa causes photochemical ozone formation over Africa and the adjacent Atlantic. Because of the ABLE 2 analyses [Pickering et al., 1991; 1992a], it seemed likely that during the early wet season, deep convection over Brazilian savanna burning could further enhance the south Atlantic ozone burden.

A nearly perfect coincidence of biomass burning and the development of a series of mesoscale convective complexes indeed occurred in late September 1992 in Brazil during TRACE-A [Pickering et al., 1996]. A series of convective events (22-28 September 1992) began in southern Brazil, associated with a cold frontal system pushing northeastward from the midlatitudes of the southern hemisphere. By 25 September, little thermal contrast remained across the front at the surface, but a strong zone of low-level convergence persisted and advanced rapidly northward. Large mesoscale convective systems developed on 26 September and persisted through the early morning of 27 September. TRACE-A Flight 6, on 27 September 1992, and Flight 7 on 28 September 1992, both staged from Brasília, were designed to look at post-convection trace gas distributions and to characterize the biomass burning source, respectively. Fig. 5a shows the flight track for TRACE-A Flight 6 superimposed on the previous day’s fire counts, as determined from AVHRR, with locations of 26-27 September cloud tops, derived from satellite imagery (see also Fig. 5b).

A typical photo from the 27 September 1992 flight (Flight 6; Fig. 6) shows that fires in the sampling region were quite isolated. Concentrations of ozone, CO and NO_x in the lower troposphere were only sporadically elevated above a moderately polluted background in plumes from individual fires. Some more heavily polluted layers were observed between 3 and 4 km. However, deep convection was very effective in getting pollutants out of the boundary layer to the upper troposphere.

Two sections of the 27 September flight were made at cloud outflow level: the first at 9.5 km and the second at 11.3 km. Carbon monoxide (CO) concentrations at these altitudes represent a mixture of background upper tropospheric air (CO ~90 ppbv) and boundary-layer fire-exposed air masses (> 300 ppbv CO). Nitric oxide mixing ratios in the cloud outflow layer are at least 2-4 times higher than "background" conditions at these altitudes without deep convective mixing. Fig. 7 shows this elevated NO. The CO mixing ratios along the flight tracks are also elevated and the NO/CO ratio is approximately constant over much of each flight segment in cloud outflow. Seven three-minute periods during which the NO/CO ratio jumps by a factor of 1.5-3 indicate lightning NO production. In Fig. 7 one of these instances shows the 3-min averaged NO mixing ratio exceeding 1 ppbv. Detailed analysis of the NO/CO ratio in cloud outflow layers suggests that at least 30-40% of upper tropospheric NO_x on the TRACE-A flight derived from lightning [Pickering et al., 1996].

Fig. 8 shows a surface of CO with 200 ppbv mixing ratio computed from a three-dimensional GCE tracer simulation of one cell in the 26-27 September event. The effects of deep convection are widespread. This is also evident in a tracer study that uses winds from a mesoscale model (nonhydrostatic version of the Penn State/NCAR MM5 model). Convective redistribution of CO from the mesoscale convective complex of 26-27 September 1992 covered a large section of northeast Brazil [Wang et al., 1996]. A regional study of biomass burning emissions, cloud dynamics and ISCCP data showed that 10-40% of CO emitted by biomass fires in a typical burning season over Brazil would be vented to the free troposphere by deep convection [Pickering et al., 1992b].

Ozone formation following the 27 September 1992 TRACE-A event was computed for cloud outflow layers with a box model version of the Goddard Tropospheric Chemistry model. The box model starts with a composition derived from observed mixing ratios at cloud outflow level. The evolution of ozone in parcels at the two levels sampled is shown in Table 2. At the balloon ozone site at Natal, Brazil (6°S, 35°W), approximately 3 days downwind, ozone in the upper troposphere increased between 28 and 30 September 1992 at the rate shown in Table 2 (Fig. 9). What was the CEF for the 26-27 September 1992 event? Taking ratios of ozone formation computed by the box model for the altitudes of cloud outflow with and without lightning and convective NO and CO enrichment gives CEF = 3-3.5. This is much lower than the estimates for the urban-influenced ABLE 2B event (26 April 1987) and the sensitivity studies based on an ABLE 2A episode (3 August 1985). In the urban plume event (26 April 1987 ABLE 2B), there was a greater pre- and post-convective mid-tropospheric NO contrast than was observed on TRACE-A. Higher NO concentrations were observed in the undisturbed upper troposphere in the TRACE-A event than were assumed for the sensitivity studies with the 3 August 1985 ABLE 2A event.

4. Regional Consequences of Post-convective Ozone Formation

The case studies over Brazil are suggestive of larger scale impacts of deep convection over the south Atlantic Basin. A major discovery of TRACE-A [Fishman et al., 1996] was that throughout the south Atlantic (the mission covered eastern South America to southern Africa), upper tropospheric NO concentrations were high enough to maintain net ozone formation [Jacob et al., 1996; Thompson et al., 1996a]. This was even the case in regions for which the mid- and lower troposphere were NO-poor regions of ozone loss. Although lightning is a strong candidate for upper tropospheric NO_x enrichment, other NO sources are also plausible: convective injection of biogenic or fire-derived organic nitrogen compounds [Smyth et al., 1996]. These species would subsequently be photolyzed to the more reactive species NO or NO_2, initiating the ozone-forming reactions shown above.

Other effects of deep convection are suggested by trajectory studies. Fig. 10 shows the frequency of deep convection over South America taken from ISCCP (available from http://eosdis.larc.nasa.gov) superimposed on a grid of AVHRR-derived fire counts for Sept. 1992. Lower altitude winds are east to west or northeast-to-southwest, so that emissions from fires in northeast Brazil can readily be introduced into regions of most concentrated deep convection.

A climatology of upper tropospheric forward trajectories from convectively active regions of South America during the TRACE-A period (from 25 September-20 October 1992) showed a general distribution of parcel locations over 5-8 days' transit over the south tropical Atlantic and Africa [Thompson et al., 1996a]. This suggests a substantial impact for post-convective ozone formation over the south Atlantic in the latter part of the 1992 biomass burning season.

Further support for this mechanism is found in back trajectories that were run on each day for which there were ozone soundings during TRACE-A. Ozonesondes were launched at seven sites during TRACE-A, including Natal (Brazil), Ascension Island, and three locatations in southern Africa (Fig. 11a). Back trajectories (8-day runs with the Goddard isentropic trajectory model; Schoeberl et al., 1992) were initialized at each sounding site at pressure levels corresponding to ozone peaks in the lower (0-5 km), middle (5-10 km) and upper (10-15 km) troposphere. Within each of these three layers, the amount of ozone in excess of background profiles for the non-biomass burning season (in Dobson Units) was computed. An origin based on the trajectories -- South America, the Atlantic, or Africa -- was assigned to each peak and weighted with the amount of ozone to show the chemical impact of the source. Fig. 11b shows an example of this calculation for an Ascension Island (8°S, 15°W) ozone sounding on 27

September 1992. At 10-15 km, the back trajectories corresponded to air parcels that passed over a convectively active area of South America 5 days earlier. The 10-15 km layer had an ozone column amount of 12.4 DU (1 Dobson Unit = 2.69 x 10^{16} cm^{-2}), which was 5.8 DU higher than during the non-burning season. The total column excess for the 27 September Ascension sounding was 13.6 DU. Thus, if we are correct about the post-convective origin of excess ozone in this sounding, 43% of the excess ozone at 10-15 km was due to this mechanism. To remove ambiguity about the origins of the upper tropospheric ozone would require simultanteous measurements of surface tracers, especially hydrocarbons, and CO, along with the sounding.

When excess ozone for all the TRACE-A Ascension soundings is averaged together, 21% of excess ozone is ascribed to this mechansim. For four of the five sites shown in Fig. 11a, 20-30% of total excess ozone in the troposphere was from South America at the altitudes of maximum convective outflow (Fig. 11c). The early onset of the 1992 wet season in Brazil that gave rise to the coincidence of biomass fires and deep convection shown in Figure 10 was exceptional. Precipitation at central Brazil sampling locations in September 1992 was 2-4 times higher than the mean of the previous 10 years [Kirchhoff et al., 1996]. Thus, upper tropospheric ozone production downwind of convective outflow may have been greater during TRACE-A than in a typical September-October period.

5. Summary

Model simulations of trace gas observations and dynamics during tropical convective episodes have shown that ozone formation can be enhanced when deep convection injects boundary layer pollutants into the free troposphere. Lightning is an additional promoter of tropospheric ozone formation through the production of NO_x within convective clouds. A dedicated cloud outflow flight during TRACE-A put the rate of tropospheric ozone formation at 3-4 times its value in the absence of convection. The TRACE-A ozonesonde network showed that the impact of upper tropospheric ozone due to convective injection from South American pollution and/or lightning acccounted for 20-25% of the seasonal tropospheric ozone maximum in the south Atlantic basin.

A version of the Goddard Cumulus Ensemble is now coupled with the Goddard Tropospheric Chemistry model (Fig. 12). The coupled model will available for interpretation of aircraft trace gas data to be taken on future convective sampling missions. Despite the confirmation of post-convective ozone formation in TRACE-A, those observations represented a single flight of opportunity. A more definitive evaluation of the interaction of deep convection and ozone formation requires systematic sampling downwind of deep convective systems. This would include a consistent use of tracers, sampling at cloud base and cloud outflow layers as part of a sampling strategy that scaled from single cloud to mesoscale and beyond. A hierarchy of coupled models with consistent transport is required to integrate the results to the regional scale. Studies of convective systems like those on TRACE-A are critical for assessing the global impact of deep convection. Besides defining the magnitude of local chemical effects, they provide dynamical and chemical variables to validate the parameterizations used in mesoscale and global models.

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Acronym Guide

ABLE 2 = Amazon Boundary Layer Experiment (A= 1985, B= 1987)
EOS = Earth Observing System
GCE = Goddard Cumulus Ensemble Model
GTE = Global Tropospheric Experiment
ISCCP = International Satellite Cloud Climatology Project
MAPS = Measurement of Atmospheric Pollution from Space
MM5 = Mesoscale Model (NCAR/Penn State), Version 5
MOPITT = Measurement of Pollution in the Troposphere
STEP = Stratospheric-Tropospheric Exchange Project (1987)
TES = Tropospheric Emission Spectrometer
TRACE-A = Transport and Atmospheric Chemistry near the Equator - Atlantic (1992)

Table 1. Convective Enhancement Factors for O_3 production.

Study	Tropical Event	CEF
1	ABLE 2A, 3 August 1985	P(O_3) increases from slightly negative to positive
1A	ABLE 2A, 3 August 1985 dynamics*	
	Savanna burning	53
	Forest burning	59
2	ABLE 2B, 6 May 1987	~1
3	ABLE 2B, 26 April 1987	
	Unpolluted	2.5
	Urban plume	35
4	STEP/EMEX, 2 Feb. 1987	< 1
5	TRACE-A, 26-27 Sept. 1992	3-3.5

* Dynamics from 3 Aug. 1985 event with chemical composition appropriate for savanna or forest burning based on 1980 Brazil campaign [Delany et al, 1985].

Table 2. Cumulative O_3 Formation from Cloud Outflow Following 26-27 September 1992 Mesoscale Event

Layer	Cumulative Increased Ozone at End of Day (ppbv)				
	1	2	3	4	5
9.5 km	5.6	12	18	24	30
11.3 km	8.2	16	22	27	32

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Figure Captions

Fig. 1 Schematic of cloud dynamics and transport of ozone precursors (CO, NO and HC = hydrocarbons). Ozone is initially reduced in upper troposphere when unpolluted air is pumped upward. Soluble gases like nitric acid (HNO_3), hydrogen peroxide (H_2O_2), and sulfuric acid modify chemical composition of precipitation. Nitric and sulfuric acids are chief components of acid rain.

Fig. 2 (a) Forward trajectories initialized at 3-min intervals with winds derived from GCE. Simulation of tropical event during ABLE 2B (26 April 1987). Simulated cloud top at ~ 16 km, agrees with GOES IR imagery. (After Pickering et al, 1992a); (b) Back trajectories from anvil outflow for event toward end of dry season in ABLE 2A (3 August 1985; Pickering et al, 1991); (c) Equivalent potential temperature from soundings for events sampled in (a) and (b). After Pickering et al, 1994.

Fig. 3 Tracer simulation of NO_x from 2D version of GCE. Units - pptv. Simulation for ABLE 2B event, 26 April 1987. Cross-section at 6 hours is shown, with initial values (near left) at 2 hour point in simulation. Based on NASA Electra NO measurements below 6 km of pollution from Manaus, Brazil, plume (see Pickering et al, 1992a).

Fig. 4 Ozone production rate for undisturbed (no cloud) and post-convection conditions: 26 April 1987 ABLE 2B event. 24-hr average is illustrated. Chemical composition assumed at start of chemistry model run based on tracer profiles, eg NO_x in Fig. 3 (no cloud at left; post-convection in cell). From Pickering et al, 1992a.

Fig. 5 (a) Sampling location on TRACE-A Flight 6 (27 Sept. 1992), cloud locations from METEOSAT imagery of 26-27 September, superimposed on AVHRR-derived fire counts for 26 September 1992. (Fire counts from J. Kendall and C. Justice, NASA/GSFC).

Fig. 5 (b) Satellite imagery shows cloud top heights reached up to 16 km. METEOSAT-4 imagery, 1830 UT on 26 September 1992.

Fig. 6 Photo of Brazilian fires taken from NASA/DC-8 during Flight 6 of TRACE-A, 27 September 1992. (Photo courtesy of K. E. Pickering).

Fig. 7 NO mixing ratios in regions of cloud outflow sampled on TRACE-A Flight 6. Legs at 9.5 and 11.3 km shown. Highest NO mixing ratios at each altitude assumed to include some lightning contribution.

Fig. 8 CO 3D GCE tracer simulation for one convective cell in 26-27 Sept 1992 event in TRACE-A sampling over NE Brazil. 200 ppbv isosurface shown. Initialization based on DC-8 and INPE aircraft CO profiles. Plot is at 150 mins. into simulation.

Fig. 9 Sounding on 30 Sept. 1992 was 2-4 days downwind from TRACE-A Flight 6 sampling. Note 25-30 ppbv O_3 increase from 28 to 30 Sept. in selected layers. March-April-May (M-A-M) is mean ozone during non-biomass burning season. Back trajectories initiated at Natal show middle and upper level O_3 peaks on 30 Sept. originating from cloud outflow regions sampled on 27 Sept. flight. Lower level air mass origins for 28 and 30 Sept. 1992 soundings are African [Thompson et al, 1996a].

Fig. 10 - Sept. 1992, mean number of fires/day in Brazil from AVHRR (courtesy of A. Setzer, INPE, Brazil) with ISCCP-derived frequency of deep convection. Low-level winds are from east and/or northeast, which promotes transport from regions of biomass burning toward active convection.

Fig. 11 - (a) TRACE-A and SAFARI (Southern African Fire Atmospheric Research Initiative) ozone sounding sites in September-October 1992. (b) Excess ozone illustrated for typical sounding at Ascension Island, 27 September 1992. Origin of excess ozone determined by running back trajectories in clusters from lower (0-5 km), middle (5-10 km) and upper tropospheric (10-15 km) ozone peaks; (c) Composite of excess ozone origins for five sounding sites in (a). Fractions determined by weighting origins by amount of excess O_3 in each layer. Excess ozone referenced to March-April-May ozonesondes except for

Okaukuejo, which was determined by averaging March-April-May soundings for other African sites.

Fig. 12 Schematic of coupled transport-chemistry model using GCE.