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ENHANCEMENT OF FREE TROPOSPHERIC OZONE PRODUCTION BY DEEP CONVECTION

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ABSTRACT

It is found from model simulations of trace gas and meteorological data from aircraft campaigns that deep convection may enhance the potential for photochemical ozone production in the middle and upper troposphere by up to a factor of 60. Examination of half a dozen individual convective episodes show that the degree of enhancement is highly variable. Factors affecting enhancement include boundary layer NO_x mixing ratios, differences in the strength and structure of convective cells, as well as variation in the amount of background pollution already in the free troposphere.

1. INTRODUCTION

It has been observed that deep convective clouds are a major means of transporting insoluble trace species such as CO, NO_x, O_3, and hydrocarbons from the boundary layer to the middle and upper troposphere (Dickerson et al., 1987). Ozone precursor gases, once detrained from a convective cloud, can react and produce O_3 photochemically in the free troposphere downwind from a convective system. Ozone produced in the free troposphere has a much longer lifetime than O_3 produced photochemically in the boundary layer, thereby allowing it to be transported large distances from the precursor source region. Another consequence of deep convection is that O_3 production in the boundary layer may actually become more efficient following dilution of polluted boundary layer air with cleaner air descending from aloft in convective downdrafts. More ozone may be produced in the boundary layer, thereby enhancing the ozone production potential of the entire tropospheric column.

These conclusions are based on a series of model analyses to estimate the downwind perturbation in tropospheric O_3 production in convective outflow using airborne trace gas observations from convective events that occurred during NASA- and NSF/NOAA-sponsored field programs. This paper describes the analytical method and major results from this series of model simulations. We present a summary of O_3 production enhancements for six studies completed to date and a discussion of how chemical and dynamical characteristics contributed to the degree of enhancement in each event.

2. METHOD

The analysis procedure consists of three steps: (1) The convective system is simulated with a two-dimensional cloud dynamical and microphysical model (e.g., Tao et al., 1991) initialized with a sounding of temperature, moisture, and winds.
(2) The wind fields produced in this simulation are used in a tracer transport model to redistribute trace gases measured from aircraft that can, as a first order approximation, be considered conserved during rapid convective transport (O_3, NO_x, CO, and many nonmethane hydrocarbons).
(3) Profiles representing "undisturbed" and "cloud-processed" trace gas composition are selected from the resulting 2-D trace-gas fields and used to constrain a one-dimensional photochemical model (Thompson and Cicerone, 1986; Pickering et al., 1990; 1991) to calculate O_3 production rates for the 24-hour period following the convective event.

These 24-hour averaged rates are designated the "ozone production potential". We have defined a "convective enhancement factor" (CEF) which is the ratio of column ozone production potential in cloud-processed air to that in undisturbed air. The column is chosen based on the vertical extent of the free tropospheric cloud-outflow layer in each convective event.

3. RESULTS AND DISCUSSION

Table 1 lists the convective events analyzed with their convective enhancement factors. The results and discussion are grouped by geographic region.

Oklahoma - PRESTORM

We have simulated two vigorous thunderstorm events from the Oklahoma PRESTORM experiment of June 1985 (Studies 1 and 5). The O_3 production rates for the June 15 mesoscale convective complex were calculated (Pickering et al, 1990) with only the photochemical model using direct aircraft observations to distinguish undisturbed and cloud-processed air (ie., without using the cloud simulation), whereas the June 10 squall line was simulated (Pickering et al, 1992a) with the full procedure described in the previous section. The June 15 cloud-processed observations (Dickerson et al., 1987) reflect the effects of convective redistribution and lightning on a typical rural central U. S. NO_x profile; a CEF of ~4 was computed. Rural NO_x mixing ratios also characterized the June 10 event, but the O_3 production calculations were performed with the simulated NO_x redistribution with no additional NO_x from lightning. The resulting CEF was 2.5. The June 10 storm later passed over the Oklahoma City region, entraining the urban plume. When measured trace gas mixing ratios characteristic of the Oklahoma City plume (50 km downwind of the city) were redistributed by the simulated

Table 1. Convective Ozone Production Enhancement

Study	Description	CEF	Reference
1	Okla. PRESTORM, 15 June 85	4	Pickering et al (1990)
2	ABLE 2A, 3 August 85 $P(O_3)$	- -- +	Pickering et al (1991)
2A	ABLE 2A, 3 August 85 dynamics		
	Savanna burning	53	Pickering et al
	Forest burning	59	(1992b)
3	ABLE 2B, 6 May 87	~1	Scala et al (1990)
4	ABLE 2B, 26 April 87		
	Unpolluted	2.5	Pickering et al
	Urban plume	35	(1992a)
5	Okla. PRESTORM, 10 June 85		
	Rural	2.5	Pickering et al
	Urban plume	3.9	(1992a)
6	STEP/EMEX, 2 Feb. 87	< 1	Pickering et al (1992d)

storm, the CEF increased to 3.9.

Amazon Basin - ABLE 2A and 2B

Studies 2 through 4 were conducted for convective events that occurred during the NASA Global Tropospheric Experiment Amazon Boundary Layer Experiment (ABLE 2A and ABLE 2B) in Brazil. The dry season case (August 3, 1985 - Study 2) included observations of low NO_x and hydrocarbons (primarily isoprene), causing a net loss of O_3 in the undisturbed free troposphere (Pickering et al., 1991). Following the convective event, the cloud-processed air contained sufficient NO_x vented from the boundary layer to allow net O_3 production to occur in the free troposphere. The integrated ozone production potential in the 5-13 km cloud-outflow layer changed from net destruction to net production as a result of convective transport. The simulation of the August 3 squall line shows efficient transport of boundary layer air to the upper troposphere in relatively undilute core updrafts (a "hot tower" as shown by the trajectories in Figure 1).

In a sensitivity study of ozone production potential (Pickering et al., 1992b), the August 3, 1985, cloud dynamics were assumed to apply to regions in Brazil affected by intense pollution from biomass burning (Study 2A). When the simulated squall line redistributed the trace gases (Figure 2) over these regions, pollution plumes were introduced into a pristine free troposphere, enhancing the ozone production potential by a factor of between 50 and 60. The CEF for redistribution of forest burning emissions is slightly higher than that for savanna burning because of the presence of isoprene over the forest. Figure 3 shows the profile of ozone production potential in cloud processed and undisturbed air for the forest burning case.

We have extrapolated our cloud-scale CO convective transport results to the regional-scale for an area intensely polluted by biomass burning. Using the convective transport in the August 3 event as a prototype, along with satellite-derived convective cloud climatology and areal extent of biomass burning pollution, we have estimated (Pickering et al., 1992c) that 10 - 40% of CO emissions from biomass burning may be rapidly transported to the free troposphere

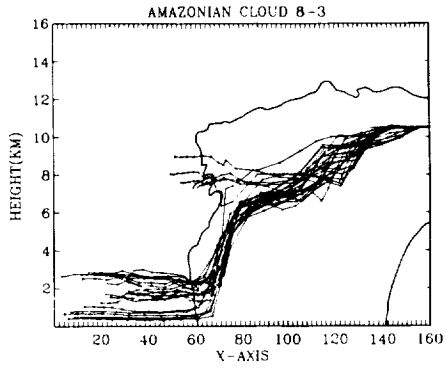

Fig. 1. Set of back trajectories simultaneously arriving at 26 horizontal positions at 10.5 km at the rear of the August 3, 1985 Amazonian squall line. Horizontal scale shows grid points at 0.5-km intervals. Heavy line is approximate outline of cloud (after Pickering et al. (1991), copyright American Geophysical Union).

over the major region of deforestation in the Amazon Basin. This result suggests that a significant fraction of the O_3 precursors generated from forest burning in Brazil may actively generate O_3 in the free troposphere where O_3 has a much longer lifetime than in the boundary layer.

In the wet season over the Amazon Basin, NO_x concentrations in unpolluted air are even lower than in the dry season and the ozone production potential was very close to zero (May 6, 1987 event - Study 3, Scala et al, 1990). Convective transport to the free troposphere was considerably less than in the August 3, 1985, dry season case, due to much overturning and midlevel detrainment of air from the storm. The well-mixed moist troposphere in which the squall line developed may have hindered strong updraft and downdraft development. Parcel trajectory analyses show that more than 50% of the air transported to the anvil region of the storm originated above 6 km, not from the boundary layer via undilute cores, as in the dry season event. The resulting CEF

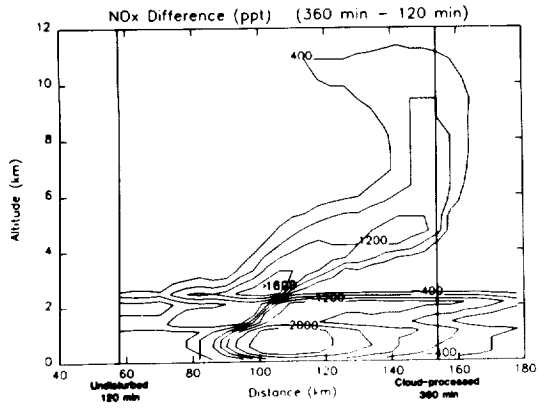

Fig. 2. Difference in tracer mixing ratios computed by subtracting undisturbed fields from cloud-processed fields, showing the magnitude of trace gas redistribution when biomass burning pollution encounters a convective storm such as the August 3, 1985, event. NO_x difference, contour interval 400 pptv (after Pickering et al. (1992b), copyright Kluwer Academic Publishers).

was very close to 1.

In a second wet season case (April 26, 1987 - Study 5), the NO_x was slightly greater and the convection more vigorous, producing a CEF of about 2.5 (Pickering et al., 1992a). This squall line also entrained the urban plume from the city of Manaus, containing similar NO_x, CO, and hydrocarbon levels to those in the Oklahoma City plume (Study 5). Because of the much cleaner free troposphere over Brazil, the CEF for this case was 35, compared to only 3.9 for convective redistribution of the Oklahoma City urban plume. This contrast demonstrates the importance of the amount of background pollution already in the free troposphere in determining the degree of convective enhancement. The CEF computed for the entire tropospheric column in the redistributed Manaus plume shows that convection increased column ozone production potential by almost 50%. In addition to the greater O_3 production in the middle and upper troposphere, O_3 production efficiency per molecule of NO in the boundary layer increased by 60% as cleaner air descended to dilute the more polluted air (Figure 4).

Marine tropics - STEP/EMEX

Simulation of a convective event off the northern Australian coast (Study 6 - February 2, 1987; Pickering et al., 1992d) showed relatively weak vertical transport. This event occurred during the Stratosphere-Troposphere Exchange Project (STEP) and the Equatorial Mesoscale Experiment (EMEX). Low NO_x marine air was transported upward from the lower troposphere, resulting in a CEF < 1 in the cloud outflow. Both observations and the simulation showed that the cloud was relatively deep (15-16 km), but vertical velocities did not exceed 10 m s^{-1}. Forward trajectories (Figure 5) constructed from model-computed wind fields showed boundary layer air not reaching altitudes higher than 12 km with substantial mid-level detrainment.

As in the Study 3 wet season ABLE 2B event (May 6, 1987), the troposphere was already quite well mixed prior to the event, as evidenced by the profile of equivalent potential temperature shown in Figure 6. The convective transport characteristics of the STEP/EMEX case and of Study 3 are in sharp contrast to those of the Amazon dry season event

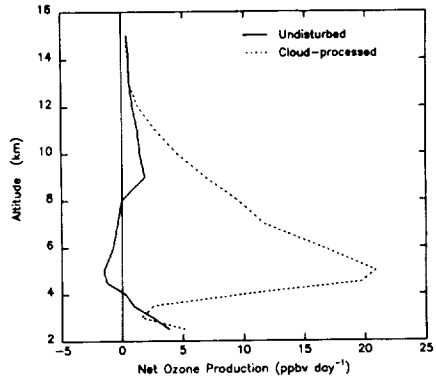

Fig. 3. Diurnally-averaged ozone producton potential for the 24 hours following the redistribution of pollution from forest burning by the August 3, 1985, convective event (after Pickering et al. (1992b), copyright Kluwer Academic Publishers).

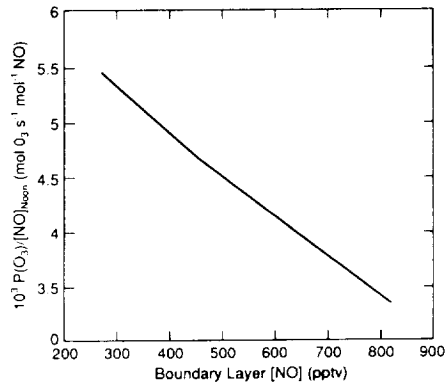

Fig. 4. Ozone production efficiency in the boundary layer as a function of NO for the April 26, 1987, ABLE 2B convective event. The highest value of NO existed in the undisturbed urban air and the lower values are representative of the cleaner air that descended into the boundary layer during the storm (after Pickering et al. (1992a), copyright American Geophysical Union).

(August 3, 1985, Studies 2 and 2A) which showed little entrainment or detrainment in the middle troposphere. The equivalent potential temperature profile for the August 3 event (also in Figure 6) shows a very pronounced midtropospheric minimum. These findings agree with the conclusion reached by Scala et al. (1990) that showed that the up and downdraft structure of tropical convective storms may depend largely on the shape of the equivalent potential temperature profile prior to the storm.

4. CONCLUSIONS

The analyses summarized in Table 1 are the beginning of a global climatology of the effects of deep convection on tropospheric chemistry. The major factors affecting the degree of enhancement of O_3 production by convection are the available boundary layer NO_x, strength and structure of the convective cells, and the amount of background pollution in the free troposphere.

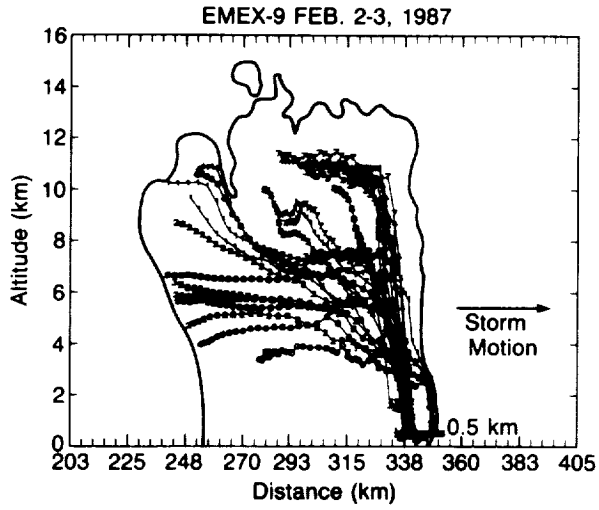

Fig. 5. Set of forward trajectories simultaneously beginning at 26 horizontal positions at an altitude of 0.5 km at the forward edge of the February 2, 1987, STEP/EMEX convective system.

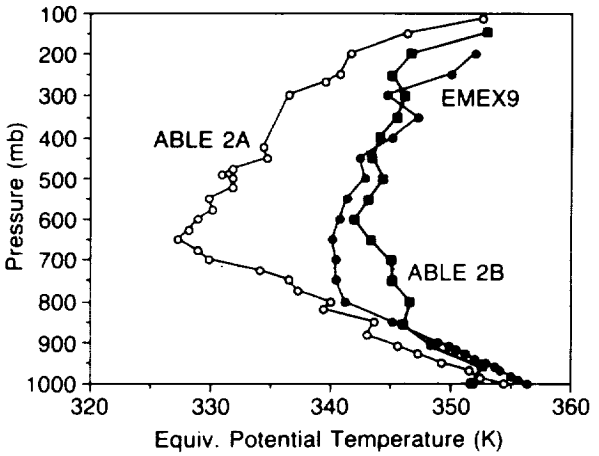

Fig. 6. Profiles of equivalent potential temperature observed prior to the August 3, 1985, ABLE 2A squall line and the February 2, 1987, STEP/EMEX convective event.

Major conclusions that we have reached from our series of analyses are as follows:

(1) The maximum convective enhancement of O_3 production in the free troposphere occurred with the redistribution of intense boundary layer biomass burning pollution into a pristine free troposphere by a storm with undilute updrafts. At the other extreme a small decrease in the ozone production potential of the free troposphere has been computed for a marine low NO_x event with very weak vertical transport.

(2) Convective transport characteristics appear to correlate with how well mixed the troposphere is prior to a convective event. Therefore, the efficiency of vertical transport of ozone precursors to the middle and upper troposphere might be predictable from the equivalent potential temperature profile prior to a storm.

(3) In addition to pumping O_3 precursors into the free troposphere, a convective system may also bring air with lower NO_x mixing ratios into a polluted boundary layer. The efficiency of O_3 production per molecule of NO_x in the boundary layer increases, and the total tropospheric column ozone production potential is enhanced.

(4) When considered on a regional basis over a season, deep convection may vent a significant fraction of biomass burning emissions to the free troposphere over parts of the Amazon Basin, enhancing the free tropospheric ozone production potential over broad areas. Our results show that it is critical that parameterizations of deep convective transport be improved in 3-D global and regional chemical models.

The CEF's for ozone production in cloud outflow are derived from model calculations initialized with observations in undisturbed air. More research flights are needed to confirm our model predictions of convective transport and of O_3 production rates in cloud-processed air. Measurements of ozone precursors are required in outflow layers immediately downwind of storms, as well as measurements of O_3 in the first several hundred kilometers of downstream transport.

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