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Transport of biomass burning smoke to the upper troposphere by deep convection in the equatorial region

M. O. Andreae[1], P. Artaxo[2], H. Fischer[1], S. R. Freitas[7], J.-M. Grégoire[3], A. Hansel[4], P. Hoor[1], R. Kormann[1], R. Krejci[5], L. Lange[1], J. Lelieveld[6], W. Lindinger[4], K. Longo[2], W. Peters[6], M. de Reus[6], B. Scheeren[6], M. A. F. Silva Dias[7], J. Ström[5], P. F. J. van Velthoven[8], and J. Williams[1]

Abstract. During LBA-CLAIRE-98, we found atmospheric layers with aged biomass smoke at altitudes >10 km over Suriname. CO, CO_2, acetonitrile, methyl chloride, hydrocarbons, NO, O_3, and aerosols were strongly enhanced in these layers. We estimate that 80-95% of accumulation mode aerosols had been removed during convective transport. Trajectories show that the plumes originated from large fires near the Brazil/Venezuela border during March 1998. This smoke was entrained into deep convection over the northern Amazon, transported out over the Pacific, and then returned to South America by the circulation around a large upper-level anticyclone. Our observations provide evidence for the importance of deep convection in the equatorial region as a mechanism to transport large amounts of pyrogenic pollutants into the upper troposphere. The entrainment of biomass smoke into tropical convective clouds may have significant effects on cloud microphysics and climate dynamics.

1. Introduction

Vegetation fires, particularly in the tropics, are a major source of atmospheric pollution, affecting large areas of the globe. Most fires occur in the dry tropics, where large-scale subsidence prevents deep convection and associated rainfall, and thus allows the vegetation to become dry enough to burn. Since deep convection, which is needed to transport smoke to the upper troposphere, also produces rainfall and thus precludes fires, it was often assumed that the influence of biomass burning would be mostly limited to the lower troposphere. However, observations showed large amounts of pollutants such as CO and O_3 in the upper troposphere over the tropics [*Reichle et al.*, 1990; *Watson et al.*, 1990], which could only be explained as resulting from vegetation fires. Some of the lofting of smoke to the upper troposphere may be taking place in episodic frontal convective events [*Pickering et al.*, 1996]. A more general mechanism was proposed to explain the transport of smoke to high altitudes, which entailed transport by the trade wind circulation towards the ITCZ, followed by convective transport to the middle and upper troposphere [*Crutzen and Andreae*, 1990]. Modeling analysis of airmass transport and chemical processing, applied to results from several campaigns, was able to explain the injection of smoke into the tropical upper troposphere on regional to continental scales [*Chatfield et al.*, 1996; *Jacob et al.*, 1996; *Jonquières and Marenco*, 1998; *Schultz et al.*, 1999]. However, because of the heterogeneous distribution of vegetation fires and the long transport paths involved in this mechanism, it has hitherto not been possible to validate it by linking plumes observed in the upper troposphere to specific fires detected by remote sensing.

1) Max Planck Institute for Chemistry, Mainz, Germany
2) Instituto de Fisica, Universidade de Sao Paulo, Brazil
3) Environment Institute, JRC/CEC, Ispra, Italy
4) Institute of Ion Physics, Innsbruck University, Austria
5) Inst. Applied Environm. Research, Stockholm University, Sweden
6) Inst. for Marine and Atmospheric Research, Utrecht, Netherlands
7) Dept. Ciencias Atmosfericas, Universidade de Sao Paulo, Brazil
8) Royal Netherlands Meteorological Institute, De Bilt, Netherlands

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2. Results and Discussion

During LBA-CLAIRE-98 (Large-scale Biosphere-Atmosphere Experiment in Amazonia - Cooperative LBA Airborne Regional Experiment '98) we conducted research flights over Suriname, in the northern part of Amazonia. While the campaign was taking place, large fires were burning in the savanna/forest transition region in northern Brazil and adjacent portions of Venezuela and Guyana. These fires had begun in late February and were most intense during 16-30 March 1998. AVHRR data showed the fires to be concentrated in an area bounded by 1°-5°N, 59°-62°W. The smoke was evident on maps of the distribution of UV-absorbing aerosol prepared from TOMS sensor [*Herman et al.*, 1997] data in real time during the campaign, and covered areas several 100 km in diameter. Since the lower tropospheric circulation in the region is dominated by northeasterly trade winds, we did not expect to encounter smoke from these fires in our study area over Suriname, some 500 km NE (upwind) of the fires.

However, back-trajectories available for planning of flight CLAIRE-08 (26 March 1998) indicated that airmasses were arriving at altitudes ~10 km over Suriname, which had originated in a deep convection region downwind of these fires. This region was clearly evident on GOES imagery, and the associated precipitation was seen in the data from the TRMM (Tropical Rainfall Measuring Mission) satellite (data available at http://lake.nascom.nasa.gov/data/dataset/TRMM/). The plume travel time estimated from the trajectory calculations was 9-10 days. We decided to investigate the hypothesis that smoke from these fires was entrained into deep convection, carried aloft and transported to our study region by the large-scale circulation around a high-level anticyclone. For this purpose, vertical soundings and a transect at ~10 km were flown over southern Suriname. Our instrumentation allowed the determination of CO_2, CO, O_3, NO, several hydrocarbons, acetone, acetonitrile (CH_3CN), methyl chloride (CH_3Cl), and aerosol particles [*Crutzen et al.*, 2000].

The predicted presence of smoke plumes in the upper troposphere was confirmed by in-situ measurements. The vertical profiles (Figure 1) show a layer of enhanced CO and O_3 at the altitudes predicted by the trajectory calculations, above clean air that originated in the central North Pacific. To elucidate the mechanisms responsible for the transfer of pyrogenic pollution to the upper troposphere, we conducted forward and backward trajec-

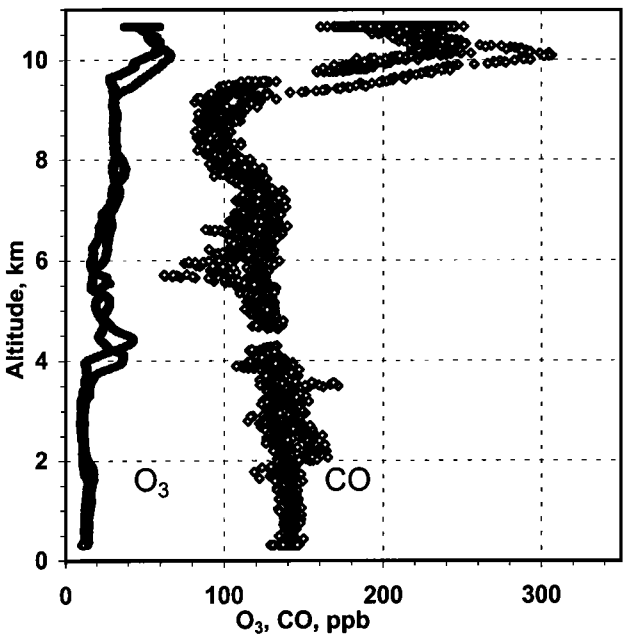

Figure 1. Vertical profiles of CO and O_3 sampled during Flight 08 of LBA-CLAIRE-98 over southern Suriname

tory analyses using the USP (University of São Paulo) convective model [*Freitas et al.*, 2000] forced with NCEP analysis, and the KNMI model with ECMWF data in the backward mode (Figure 2). The USP model (Figure 2a) uses a detailed convective scheme and applies a relatively high resolution over South America, making it highly suitable for forward and backward trajectory calculations in regions with deep convection. The KNMI model (Figure 2b), on the other hand, uses kinematic vertical velocities from the ECMWF data set and a coarser resolution, and is less suitable for resolving convective activity. Although the ECMWF model reproduces the ITCZ, convection is not necessarily simulated in agreement with the time and location in the real atmosphere. Both models predict transport from the fires in the Roraima/Venezuela region to convective areas in the ITCZ, lofting of the smoke-laden air to the 10-12 km level, and transport around a high-altitude anticyclone to the Guyana/Suriname region. In spite of the large uncertainties typically associated with trajectory calculations running over 10 days, the excellent agreement between two entirely different models, using different data sets, and different directions of computation, lends strong support to the validity of the transport mechanism proposed here. In summary, this mechanism is characterized by the following sequence of events: 1) Smoke emission from fires in the dry regions under the subsiding branch of the Hadley cell, 2) Transport by the trade wind circulation into the Amazon Basin, 3) Lofting to the upper troposphere by deep convection, 4) Eastward transport by the high-altitude circulation.

In the plume layer, pyrogenic trace gases were elevated and showed strong mutual correlations. The enhancements are shown in Table 1 as ratios $\Delta X/\Delta CO_2$ or $\Delta X/\Delta CO$, where Δ denotes the enhancement of a species over its background level. Background values were obtained using data from clean air just below the polluted layer for all species except the hydrocarbons, where we used upper tropospheric values from preceding and subsequent flights. Analysis of trace gas and aerosol enrichments in this layer shows it to be composed of several sub-layers or plumes with slightly different compositions. The variability between these layers is reflected in the ranges and standard deviations given in Table 1.

The observed enrichment of CH_3CN and CH_3Cl, both tracers for biomass burning, and the absence of elevated anthropogenic halocarbons, are clear evidence that the plumes came from biomass fires and not from urban/industrial sources. The $\Delta CO/\Delta CO_2$ ratios in our plumes are in the range typical of savanna fires (0.04-0.07) and below those of forest fires (0.08-0.10) [*Andreae et al.*, 1996a]. Because of the 10-day age of the plumes, however, it has probably decreased by some 20-30% from its initial value [*Mauzerall et al.*, 1998], which would make it consistent with the origin of the smoke from the savanna/forest transition zone. For a semi-quantitative analysis of the effect of plume aging, we have included in Table 1 the estimated "initial" values of the emission ratios, i.e., the values that would have evolved to the observed ones after 2 days of transport at ~2 km and 7.5 days at ~10 km. For most species, these back-extrapolated values fall within the

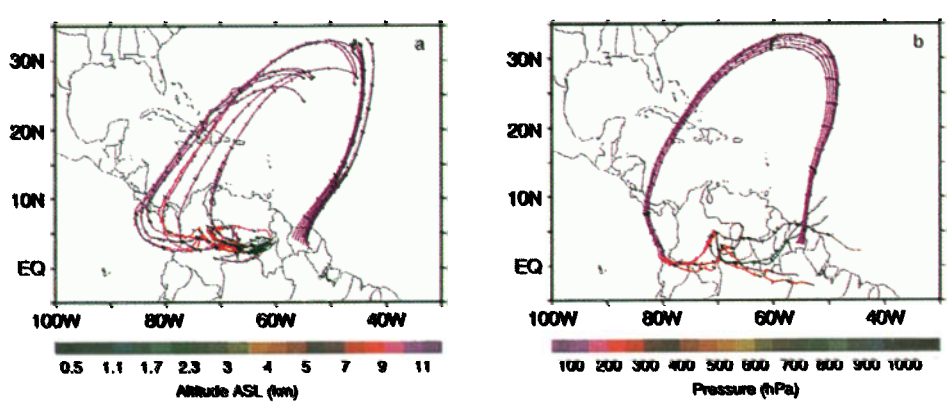

Figure 2. a) Combined 8-day forward trajectories from the fire region in Brazil/Venezuela and 2-day back-trajectories from the flight track, computed with the USP convective model. The forward trajectories were initialized on 16-3-98 2000UTC at 2 km a.s.l. in the region with the most active fires, the back-trajectories near the transect over Suriname (near 4°N 56°W) at the measurement time and altitude. Convective lifting is evident just north of the Equator near 70°W. The forward and backward trajectory bundles come together on 24-3-98 in a region near 30°N 45°W. Small arrows on the trajectories show the direction of airmass travel; black dots indicate 24 h of travel time. b) 10-day back-trajectories from the flight track computed with the KNMI model. The trajectories were initialized at the exact time, altitude (~10 km) and position of the aircraft during the transect (near 4°N 56°W). Several of the trajectories indicate airmass origins near the surface in the region of active fires, and convective lifting in the same region as the forward trajectories in Figure 2a. Trajectory labeling is as in Figure 2a; the color-coding of pressure levels corresponds to the altitude scale in Figure 2a.

Table 1: Comparison of literature data on chemical characteristics of fresh savanna fire smoke and aged plumes with the high-altitude plumes observed during CLAIRE-98 (mole ratio * 10^3).

Parameter	Fresh smoke[1]	Aged plumes[2] (2-6 days)	Highly Aged Plumes (>6 days)[3]	CLAIRE High-Altitude Plumes[5]	Back-extrapolated to emission[6]
$\Delta CO/\Delta_{CO2}$	40-80	20-90	16±2	55 (48-73)	74 (64-98)
$\Delta CH_3Cl/\Delta CO$	0.8-1.1	1.2±0.12	1.4±0.1	1.1±0.2	0.8±0.2
$\Delta CH_3CN/\Delta CO$	0.4-2.5	---	---	1.2±0.3	0.9±0.2
$\Delta C_2H_2/\Delta CO$	1-10	2.6±0.2	2.8±0.2	0.9±0.3	1.3±0.4
$\Delta C_2H_6/\Delta CO$	4-9	7.1±1.1	8.0±0.7	13±2	12±2
$\Delta C_2H_4/\Delta CO$	5-12	---	---	≤0.3	---
$\Delta C_3H_8/\Delta CO$	0.9-2.0	1.2±0.4	---	2.0±0.5	3.8±0.9
$\Delta C_6H_6/\Delta CO$	1.1-1.5	0.48±0.05	---	0.42±0.10	1.3±0.3
$\Delta(CH_3)_2CO/\Delta CO$	1.5-12	---	---	19.5±1.6	---
$\Delta NO/\Delta CO_2$	1-3	<0.1	0.05±0.01[4]	0.06±0.01	---
$\Delta_{O3}/\Delta CO$	---	0.3-0.7	0.74±0.90	0.25 (0.2-0.29)	---
$\Delta N_{>0.1}/\Delta CO$	10-30	5±3	4.6±2.4	1.2 (0.6-4.5)	---
$\Delta N_{Aitken}/\Delta CO$	---	8-18	---	4.9 (2.7-6.4)	---
$\Delta N_{ultrafine}/\Delta CO$	---	---	---	8.1 (1.4-10.1)	---

[1]Data from *Anderson et al.* [1996], *Andreae et al.* [1996a, 1996b], *Hao et al.* [1996], *Mauzerall et al.* [1998], *Holzinger et al.* [1999]
[2]Data from *Andreae et al.* [1994], *Mauzerall et al.* [1998]
[3]Data from *Mauzerall et al.* [1998]
[4]NO_x
[5]Means and ranges for continuously measured data, mean and analytical uncertainty for canister data
[6]) Assuming 2 days transport at 2 km and 7.5 days at 10 km; lifetimes from *Mauzerall et al.* [1998]

range observed for fresh biomass smoke, except for ethane and propane where they are slightly higher. The alkenes, which in fresh emissions are more abundant than the alkanes, are present only at or below the detection limits. Surprisingly, acetone is also enriched in comparison to fresh smoke, in contrast to what would be expected from its OH lifetime and the production from propane. We have observed similar enrichment in other plumes [D. Sprung, unpublished data, 2000], suggesting the possibility of additional production mechanisms.

$\Delta NO/\Delta CO_2$ was much lower than in fresh smoke, and similar to values in aged plumes [*Andreae et al.*, 1994; *Chatfield et al.*, 1996; *Mauzerall et al.*, 1998]. NO concentrations in the layer were ~200-300 ppt. Unfortunately, due to instrument limitations we did not measure other nitrogen species, such as NO_2 or HNO_3. The $\Delta O_3/\Delta CO$ ratio, indicative of the amount of photochemical O_3 formation that has taken place in the plumes, is near the lower end of the range observed in mature plumes in the lower troposphere. This ratio is dependent on the initial NO_x content of the plume and on the chemical evolution during photochemical processing in the atmosphere. The fairly low value observed on CLAIRE-08 suggests that NO_x species were removed early in the evolution of the plume. Lightning activity during cumulonimbus convection could have re-supplied reactive NO_x to the plumes and led to additional photochemical ozone production.

The low aerosol concentration in the plumes shows the importance of wet removal of particles during convection, consistent with the remote-sensing observations of cumulonimbus and rainfall. In fresh smoke, the ratio $\Delta N_{>0.1}/\Delta CO$, i.e., the enhancement in particles with diameters >0.1 μm normalized to the enhancement of CO, varies around 20 cm^{-3} (STP) ppb^{-1} [*Le Canut et al.*, 1996]. After an initial phase, when particle number concentrations decrease rapidly by coagulation, the particle enhancement ratio decreases very slowly with plume age, and lower tropospheric plumes with ages of up to a week still have $\Delta N_{>0.1}/\Delta CO$ around 5 cm^{-3} ppb^{-1} [*Andreae et al.*, 1994; *Mauzerall et al.*, 1998]. In contrast, our plumes had much lower $\Delta N_{>0.1}/\Delta CO$ (~1 cm^{-3} ppb^{-1}), with the exception of one sub-layer that had a $\Delta N_{>0.1}/\Delta CO$ of 4.5 cm^{-3} ppb^{-1}. This implies that, in most of the plumes, number concentrations of accumulation mode particles had decreased to about 5% of the initial value, and suggests that deep convection and the associated rain production mechanisms are efficient at removing atmospheric aerosol in this size fraction. If we accept the range of 5-20 cm^{-3} ppb^{-1} as an estimate of enhancement ratios before the onset of convection, our results suggest that some 80-95% of the accumulation mode particles are removed during deep convection.

In contrast to the strong depletion of accumulation mode particles in the high-altitude plumes, smaller particles in the Aitken and ultrafine modes were still quite abundant. Aitken mode particles (here defined as the size class 18<D<120 nm) were at concentrations of around 1000, and ultrafine particles (7-18 nm dia.) were present at 500-1000 cm^{-3} (STP). The few available datasets from moderately aged plumes show $\Delta N_{Aitken}/\Delta CO$ values in the range of 8-18 cm^{-3} ppb^{-1}, about twice the value found here [*Andreae et al.*, 1992, 1994]. This is consistent with less efficient removal of particles <100 nm during convection. The presence of a substantial number of ultrafine particles shows that new particle formation must be occurring in the plumes, since these particles are too short-lived to have originated in the fires. The removal of most of the accumulation mode particles actually favors nucleation of new particles, because it sharply reduces the surface area available for the condensation of low-volatility vapors. This suggests that aerosol-depleted fire plumes are active regions of new particle formation in the upper troposphere, and that deep convective transport of biomass smoke may make a substantial contribution to the aerosol particle burden of the upper troposphere.

The differential effects of convective processing on the constituents of smoke plumes can explain the marked difference in aerosol abundance between smoke layers sampled at different altitudes in previous investigations [*Andreae et al.*, 1994; *Anderson et al.*, 1996]. While the levels of O_3 and CO in these layers are similar below and above the trade wind inversion, aerosols are much less abundant above [*Anderson et al.*, 1996; *Browell et al.*, 1996]. The smoke layers below the trade wind inversion are usually lofted by relatively shallow convection without rain production, and thus without significant removal of particles. Deep convection, on the other hand, will deplete soluble constituents, including aerosols, without affecting the concentrations of poorly soluble gases such as CO, NMHC, NO and O_3. This removal process is particularly effective for aerosol particles that contain enough soluble material to nucleate cloud droplets, and leads to a low abundance of remaining accumulation mode aerosol.

The entrainment of biomass smoke into deep convective clouds has profound implications for cloud microphysics and climate dynamics. The addition of cloud condensation nuclei (CCN) from biomass burning leads to an increase in cloud droplet concentrations and a reduction in droplet size, which suppresses rain formation in the lower, warmer portions of convective clouds [*Roberts et al.*, 2000; *Rosenfeld*, 1999]. This forces a shift of rain generation to higher altitudes where precipitation forms through the ice processes, with far-reaching consequences for global circulation dynamics [*Graf et al.*, 2000].

While the results from CLAIRE-08 are a case study limited to a particular instance, the mechanisms demonstrated here are of general validity for pyrogenic emissions in the tropics and tropical atmospheric circulation. Together with data from other cam-

paigns, our data show that biomass smoke is an important contributor to the chemical dynamics in the upper troposphere. Long-range transport of biomass smoke reaches even the most remote regions of the troposphere, as shown by measurements over the Tropical and South Pacific [*Gregory et al.*, 1999]. Since net upward motion across the tropopause takes place in the tropics, some of the biomass burning effluents may even reach into the stratosphere [*Holton et al.*, 1995].

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M. O. Andreae, H. Fischer, P. Hoor, R. Kormann, L. Lange, and J. Williams, Max Planck Institute for Chemistry, PO Box 3060, 55020 Mainz, Germany

P. Artaxo, K. Longo, S. R. Freitas, and M. A. F. Silva Dias, University of São Paulo, São Paulo, Brazil

J.-M. Grégoire, Environment Institute, JRC/CEC, Ispra, Italy

A. Hansel and W. Lindinger, Institute of Ion Physics, Innsbruck University, Technikerstr. 25, A-6020 Innsbruck, Austria

J. Ström and R. Krejci Inst. Applied Environm. Research, Stockholm University, Stockholm, Sweden

J. Lelieveld, W. Peters, M. de Reus and B. Scheeren, Inst. for Marine and Atmospheric Research, Princetonplein 5, Utrecht, Netherlands

P. F. J. van Velthoven, Royal Netherlands Meteorological Institute, P.O. Box 201, De Bilt, Netherlands

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