In-situ observations of mid-latitude forest fire plumes deep in the stratosphere

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[1] We observed a plume of air highly enriched in carbon monoxide and particles in the stratosphere at altitudes up to 15.8 km. It can be unambiguously attributed to North American forest fires. This plume demonstrates an extra-tropical direct transport path from the planetary boundary layer several kilometers deep into the stratosphere, which is not fully captured by large-scale atmospheric transport models. This process indicates that the stratospheric ozone layer could be sensitive to changes in forest burning associated with climatic warming. *INDEX TERMS:* 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 3362 Meteorology and Atmospheric Dynamics: Stratosphere/troposphere interactions. **Citation:** Jost, H.-J., et al. (2004), In-situ observations of mid-latitude forest fire plumes deep in the stratosphere, *Geophys. Res. Lett.*, *31*, L11101, doi:10.1029/2003GL019253.

1. Introduction

[2] Episodic emissions from mid and high latitude forest fires affect tropospheric concentrations of trace gases, such as carbon monoxide (CO), ozone (O_3), volatile organic compounds (VOC), and aerosol several thousand kilometers away [*Forster et al.*, 2001; *Wotawa and Trainer*, 2000]. Additionally, emissions from boreal fires lofted by convection could substantially alter upper tropospheric and lowermost stratospheric radiation balance and chemistry [*Cofer et al.*, 1996; *Waibel et al.*, 1999]. An increase in boreal fire activity and severity has been observed, and further, climate change-induced increase is expected [*Stocks et al.*, 2003]. In current conceptual models of stratosphere-troposphere exchange, upward transport into the mid-latitude stratosphere is thought to be through synoptic scale storm systems or convection, but limited to a relatively shallow mixing zone above the tropopause and well below the 380 K isentropic surface [*Stohl et al.*, 2003]. Because isentropic surfaces below 380 K intersect the tropopause in the subtropics, air injected to these levels can either be mixed back down into the troposphere by isentropic transport, or moved downward across isentropic surfaces by the average radiative cooling at these levels. It is currently believed that tropospheric air reaches the region above a potential temperature of ≈380 K exclusively in the tropics. We present a combined, correlated in-situ dataset from a suite of instruments that support a mid-latitude transport path to these altitudes and discuss the likely injection process.

2. Observations

[3] On the July 7 and 9 2002 flights of the Cirrus Regional Study of Tropical Anvils and Cirrus Layers - Florida Area Cirrus Experiment (CRYSTAL-FACE) mission operating out of Key West, Florida, unusually high CO volume mixing ratios (VMR) [*Loewenstein et al.*, 2002] were measured aboard the NASA WB-57 (Figure 1a). Up to 193 ppb CO was present in the stratosphere at 15.37 km ($\theta = 382$ K), about 1.3 km above the local tropopause as determined by the microwave temperature profiler [*Denning et al.*, 1989], while typical values at these altitudes are less than 50 ppb. Layers of enhanced CO were traversed several times on both flights between 14.7 km and 15.8 km ($\theta = 368$ and 393 K), up to 1.7 km above the local tropopause, and the observations of high CO were horizontally separated by up to 311 km on July 9. Particle number concentrations [*Jonsson et al.*, 1995] of up to 140 cm^{-3} were observed in the size range of 90–2000 nm (Figure 1b), more than 5 times higher than typical stratospheric background and more typical for tropospheric air. Carbon dioxide (CO_2) [*Daube et al.*, 2002] was also enhanced by about 1 ppm and the ratio of $\Delta CO/\Delta CO_2$ was approximately 0.05, which is consistent with a biomass burning source. There was no enrichment in nitric oxide (NO), but total reactive nitrogen (NO_y) [*Weinheimer et al.*, 1998] was enhanced compared to the surrounding air indicating an aged pollution source where NO was already converted to NO_y. Water vapor [*Weinstock et al.*, 1994] was enhanced from typical stratospheric VMR of 5 ppm to 15 ppm. Ozone [*Proffitt and McLaughlin*, 1983] was slightly lower than typical stratospheric VMR observed

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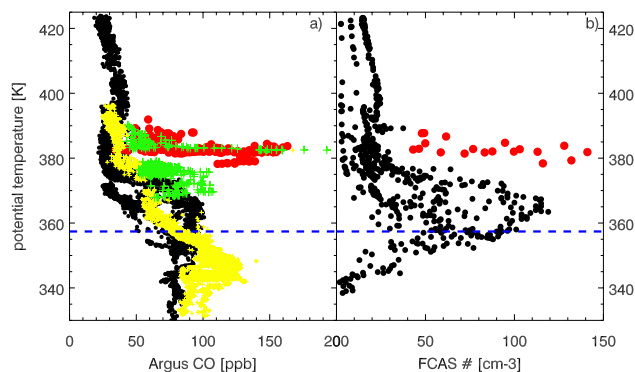[11]University of Denver, Denver, Colorado, USA.

Figure 1. In-situ measurements aboard the NASA WB-57 aircraft during the July 7 (yellow/green +) and July 9, 2002 (black/red ●) flights. (a) Carbon monoxide. The layer is defined by high CO values standing out against a background profile and corresponding points are shown in red/green. (b) FCAS particle number concentrations, Blue dashed line represents tropopause height at CO peak VMR.

during early July, suggesting either substantial ozone production above tropospheric values, or considerable dilution of a tropospheric plume, or a combination of both.

[4] The key support that forest fires are the combustion source of the CO comes from the Particle Analysis by Laser Mass Spectrometry (PALMS) instrument [*Thomson et al.*, 2000] which records mass spectra of individual particles during flight. Two particle mass spectra obtained during the CRYSTAL-FACE mission within and out of the plume are presented in Figures 2b and 2c, respectively. Figure 2a shows a typical particle mass spectrum recorded when the same PALMS instrument intercepted a fresh forest fire plume in the troposphere, aged 2 hours, on a previous mission [*Hudson et al.*, 2004]. Both this and the mass spectrum of Figure 2b, contain varying amounts of mass 12, carbon, and mass 39, potassium. These particles also contain organics (mass 24, 26, and 28) and mass 30, NO^+, which represents a nitrogen-containing compound such as ammonium or nitrate. The similarity of the known fire plume particle to the CRYSTAL-FACE particle implies that the plume on July 9
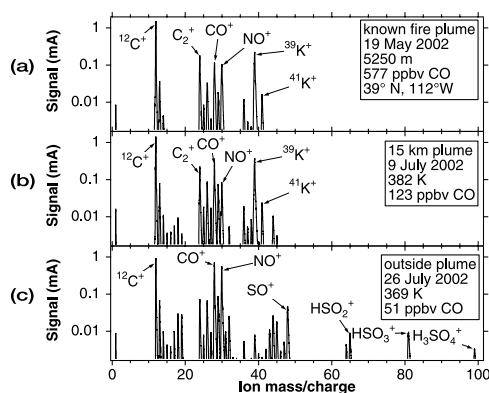

Figure 2. Single particle positive ion mass spectrum recorded by the PALMS instrument. (a) Mass spectrum of a particle of known, 2 hours old forest fire plume. (b) Mass spectrum of a particle in the high CO layer. (c) Mass spectrum of a representative, stratospheric sulfate particle.
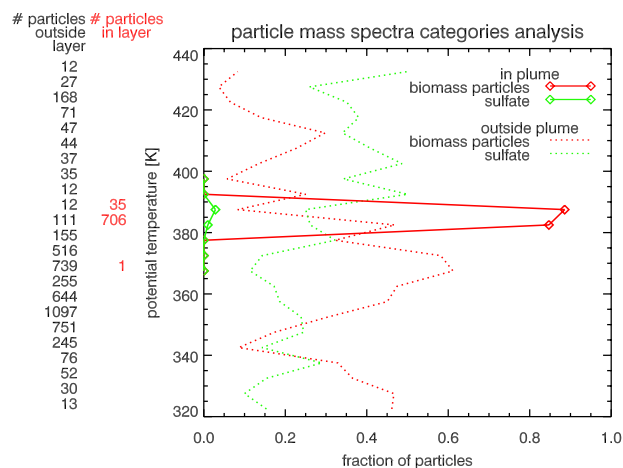

Figure 3. Fraction of biomass burning (red) or mixed sulfates and organics with NO^+ (green) particles measured by PALMS. See Figure 2 for representative examples of these particles. The solid line is the fraction of the total number of particles observed in the CO layer on July 7 and 9 and the dashed line the fraction during the rest of the mission (12 flights in July 2002). The numbers on the left side of the figure give the corresponding number of particles observed inside and outside the layer.

originated from biomass burning. Furthermore, the biomass burning particle mass spectra lack species such as sulfate and iodine which are typical components of stratospheric particles. The mass spectrum of a particle obtained outside of the plume (Figure 2c) is markedly different from that of either Figures 2a or 2b. This is representative of a sulfate particle with organic constituents. In order to find particles similar to those in Figure 2a, the mass spectra were categorized by a hierarchical cluster analysis that combines particles into categories based on the presence and relative areas of all peaks in a spectrum as described in detail in *Murphy et al.* [2003]. Particles within the category identified as biomass burning particles are all greater than 80% correlated to the mean of the category. As can be seen in Figure 3, the most common particle types found above $\theta = 390$ K are sulfate organic mixtures. However, within the plume itself, as shown in Figure 3, mass spectra as in Figure 2b represent about 90% of the particles.

[5] Back trajectories provide a probable link between the WB-57 in-situ observations and a region of enhanced aerosol extinction observed by the POAM III satellite instrument on June 29, 2002. We tracked the area of estimated extent of the plume that the aircraft encountered back in time for the 10 days prior to the measurements (Figure 4). On June 29 at 01:57 UTC when the trajectories indicate that this area extends from Hudson Bay down to South of Lake Huron (Figure 4), POAM III observed enhanced aerosol extinction more than 2 km above the tropopause over James Bay (54° N, 102° W) (Figure 5). This is at the same potential temperature and very close to the advected extent of the plume. The extinction is moderately high compared to other stratospheric aerosol enhancements observed by POAM on different occasions [*Fromm et al.*, 2000].

[6] It is very likely that a convective system injected the smoke from forest fires into the stratosphere on the previous
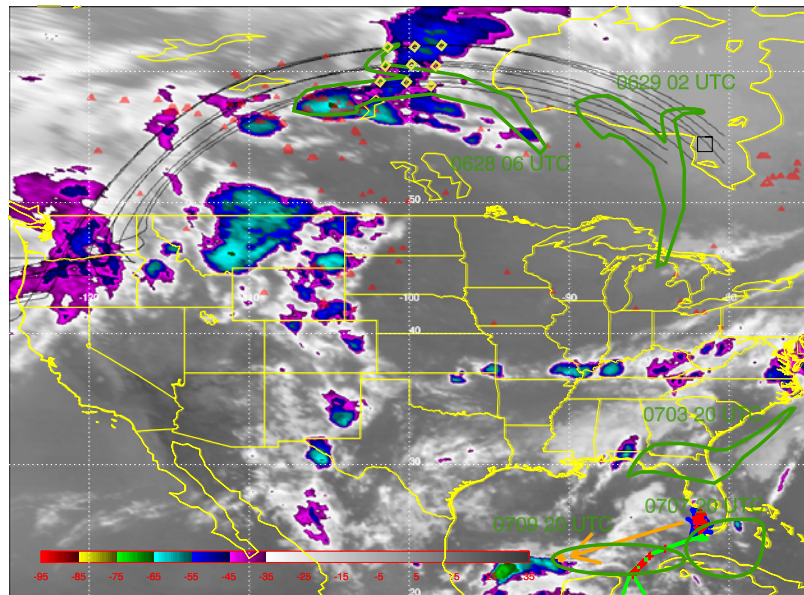
Figure 4. Cloud infrared brightness temperature from GOES weather satellite from June 28, 2002 05:45 UTC, showing at the top center the convective system likely responsible for the pumping up of the smoke plume. We estimate a minimal extent of the plume by advecting (indicated by dark yellow arrow) the highest CO points (red +) measured on the flight of July 7 (blue trace off SW Florida) to July 9 (dark yellow +), combining them with the measured points (red +) from July 9 (green trace) and assuming the plume to be contiguous within the green ellipse. This ellipse was then advected backward isentropically (θ = 382 K) in time to June 28 (shapes are labeled in MMDD HH format). The black lines are back trajectories initialized at and around the POAM footprint (white square), and the yellow diamonds mark the position at the time of the GOES image. Red triangles corresponds to MODIS fire locations in the period June 25 through June 29 above 40°N.

day. From June 26 to June 28, heavy smoke plumes are apparent on satellite images (see e.g., http://cimss.ssec.wisc.edu/goes/misc/020628/020628.html) resulting from extreme fire activity mostly in the Canadian province of Saskatchewan (over 950 hot spots on June 26 represented in Figure 4). In the area of heavy smoke a mesoscale convective system started to develop on June 27, 23:45 UTC near 59°N, 106°W. Both the back trajectories from the aircraft observations as well as trajectories initialized at the POAM III footprint extend over the cold cloud tops on June 28, 05:45 UTC (Figure 4). The lowest cloud top temperatures

of this system are below −65°C, similar to or lower than the tropopause temperatures of the close-by radiosondes. This indicates that the system had the potential to penetrate into the stratosphere. In fact, similar convective systems have been observed to inject air into the upper troposphere and lower stratosphere [*Fromm and Servranckx*, 2003]. There are at least three mechanisms that could be responsible for the plume observations at these high altitudes: (a) convective system overshooting its level of neutral buoyancy due to its inertia, and mixing at the top [*Adler and Mack*, 1986; *Wang*, 2003]; (b) additional energy input by the fires in form of heat and/or water vapor; (c) radiative self heating and subsequent lofting of the injected plume. The Total Ozone Mapping Spectrometer (TOMS) aerosol index which is very sensitive to upper tropospheric and lower stratospheric aerosol is supporting the high altitude observations: It shows an area of very strong enhancement first on June 28 in the area of the injection and it moves southeast through the POAM location over the next days (see http://jwocky.gsfc.nasa.gov/aerosols/today_plus/yr2002/images_2002.html).

3. Model Calculations

[7] FLEXPART is a Lagrangian transport model [*Stohl et al.*, 1998] based on meteorological analyses from the European Centre for Medium-Range Weather Forecasting (ECMWF) with 0.5 degree resolution on analyzed wind fields and includes a subgrid scale convective transport scheme. Daily emissions of a passive CO tracer were estimated based on fire reports and hot spot locations
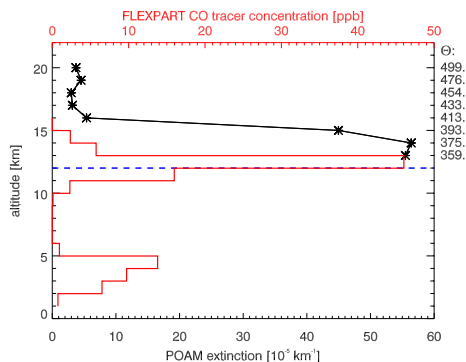

Figure 5. POAM III satellite extinction measurements above James Bay (54°N, 102°W) on June 29 at 01:57 UTC. Dashed blue line represents tropopause at POAM location. The red line represents predicted CO at the POAM location from the FLEXPART model.
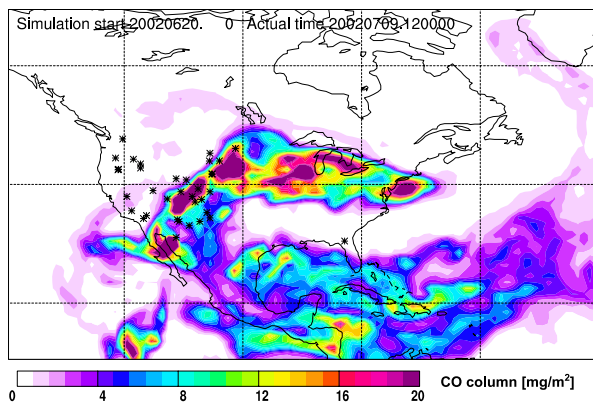
Figure 6. FLEXPART predicted columns from 12–17 km of a forest fire CO tracer originating from forest fires burning in the United States for July 9, 2002 at 12 UTC. The run was started on June 20, 2002. Asterisks mark positions of major fires. Animations showing the period from 24 June–12 July 2002 can be found as auxiliary material[1].

(Figure 6), assuming an emission factor of 4500 kg CO per hectare burnt [*Forster et al.*, 2001]. Comparison of the simulated CO total tracer columns with TOMS aerosol index data showed qualitatively good agreement. Vertical transport occurred in several mesoscale convective systems close to the fire locations. Once having reached the subtropical upper troposphere and lower stratosphere, transport was slow and the CO tracer remained there for a few weeks. Figure 6 shows that the upper troposphere and lowermost stratosphere (12–17 km) over the Gulf of Mexico was polluted mostly by US fires, in qualitative agreement with the high percentage of forest fire smoke particles seen in PALMS measurements during July 2002 (Figure 3). Since the CO source strength of the fires has large uncertainties, the absolute CO values should not be taken as exact.

[8] However, if we compare FLEXPART CO predictions at the POAM location with POAM extinction measurements (Figure 5) it becomes evident that the model does not inject high enough. The altitude difference of about 2 km between the FLEXPART model results and the POAM profile is highly significant both in terms of our understanding of how mesoscale convective systems penetrate the tropopause and the lifetime of the pollutants in the stratosphere.

4. Conclusions

[9] We present first-time in-situ observations of smoke plumes from boreal fires above 380 K. Our measurements suggest a pathway for airmasses to reach above 380 K at mid-latitudes that is not currently reproduced by large-scale models. Due to a lack of data close to the fires for the case presented, the details of the injection process and whether the fires or smoke triggered or enhanced the convection and lofting still need further investigation. The plumes observed from either the WB-57 or POAM were obvious because of

their high CO or particulate content. More research is needed to determine if high latitude convection makes more frequent but less obvious injections of tropospheric air into the stratosphere. Further studies are required to understand the mechanisms causing vertical transport of boreal forest fire plumes and the large-scale impacts of the injected material on the stratosphere.

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[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2003GL019253.

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