

Mixing between a stratospheric intrusion and a biomass burning plume

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Ozone, carbon monoxide, aerosol extinction coefficient, acetonitrile, nitric acid and relative humidity measured from the NOAA P3 aircraft during the TexAQ/GoMACCS 2006 experiment, indicate mixing between a biomass burning plume and a stratospheric intrusion in the free troposphere above eastern Texas. Lagrangian-based transport analysis and satellite imagery are used to investigate the transport mechanisms that bring together the tropopause fold and the biomass burning plume originating in southern California, which may affect the chemical budget of tropospheric trace gases.

Measurements & modelling

NOAA P3 measurements of aerosol extinction coefficient (σ_{ep}) and acetonitrile can be used as biomass burning tracers. Positive correlations between CO , σ_{ep} and acetonitrile indicate the biomass burning origin of CO enhancements. HNO_3 can be used as a stratospheric tracer. Positive correlations between HNO_3 and ozone indicate a stratospheric origin of ozone enhancements.

To identify the origin of the polluted plume and simulate air pollution transport over North America, we use the FLEXPART Lagrangian particle dispersion model (version 6.2). We conducted forward simulations of long-range and mesoscale transport of anthropogenic pollution tracers from North America and emissions from BB to assess their impact on observed CO concentrations over Texas and the Gulf of Mexico (url: <http://esrl.noaa.gov/csd/metproducts/flexpart/>)

FLEXPART was driven by model-level data from ECMWF	91 vertical levels, global resolution of $1^\circ \times 1^\circ$ with nested wind at $0.36^\circ \times 0.36^\circ$ (108°W - 27°W , 9°N - 54°N)
anthropogenic CO emissions from Canada and Mexico	EDGAR 2000 inventory
anthropogenic CO emissions from USA	EPA 1999 inventory
biomass burning (BB) CO emission	We use the MODIS fire detection data and link it to land cover types to calculate emission factors (Andrea and Merlet (2001). Area burned is assessed with reports of forest fires. Injection height between 0 and 5km.
Stratospheric ozone tracer	Initialized within the model ($\text{PV} > 2\text{pvu}$) and continually released at the model boundaries (130°W - 55°W , 10°N - 61°N) with a linear relationship $\text{ozone}/\text{PV} = 50$ ppbv/pvu. The stratospheric ozone tracer distribution in the troposphere is only due to transport from the stratosphere

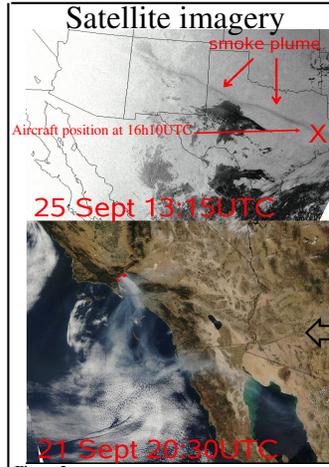


Figure 3. Top: Visible channel of the GOES East imager, 13-15 UTC September 25. Dark areas represent clouds or aerosol plumes and light areas represent clear air or the night time region, as the sun was rising in the east at this time. The red cross represents the aircraft position at 16:10 UTC. Bottom: Southern California and northern Baja California as depicted by the visible channel of MODIS on the Aqua satellite, at 20:30 UTC, September 21. The red dots represent active fires.

Figure 3 (top) presents the visible channel of the GOES East satellite at 13:15 UTC, September 25, 3 hours before the aircraft sampled the BB plume. A narrow plume is visible north of the aircraft position at 16:10 UTC (red cross), and extends from eastern Texas to western New Mexico. This narrow plume is only visible at sunrise when the sun is low in the sky and the underlying surface is not bright.

FLEXPART backward calculations were used to locate the source region of the BB CO that contributed to the two P3 CO peaks. Figure 4 presents the percentage of surface contribution to the BB CO mixing ratios, scaled according to the maximum surface contribution encountered in the domain output (red pixel). It shows that the main source is located north of Los Angeles.

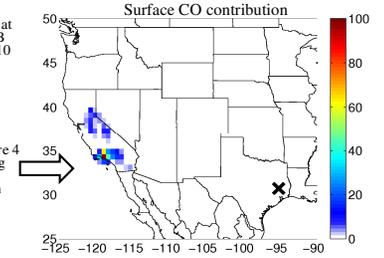
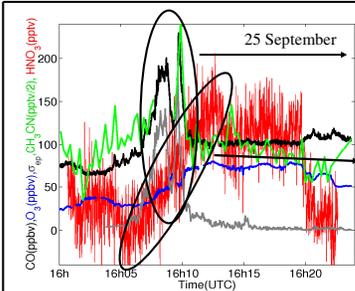


Figure 4. Surface CO contribution of the CO peak measured at about 16h10UTC due to biomass burning in the main domain output, colored by the percentage of contribution relatively to the maximum encountered.

On 21 September 20:30 UTC (Fig. 3 bottom), the visible channel of the MODIS satellite instrument shows a large forest fire located in the Los Padres National Forest north of Los Angeles at the same location as the BB contribution simulated by FLEXPART. The NASA Earth Observatory reports that this fire began on September 4 and burned with varying intensity until the end of September.



At 4km altitude, the enhancements of CO (up to 220ppbv), σ_{ep} and acetonitrile and their enhancement ratios to CO_2 (not shown) suggest a **biomass burning origin** rather than anthropogenic pollution.

The enhancement of ozone (from 30 to 80 ppbv) is positively correlated with the nitric acid (from 30 to 150 pptv) and negatively correlated with relative humidity. The HNO_3 to ozone slope is $\text{HNO}_3 = 0.0022 \times \text{O}_3 - 0.055$, and is consistent with previous measurements obtained in the **lower stratosphere** where the typical HNO_3 mixing ratio at the tropopause is 300pptv in September (Neuman et al., 2001).

Figure 1 and 2: The 5-day BB tracer shows that the positive correlation between ozone and CO (from 30 ppbv of ozone and 100 ppbv of CO to 50 ppbv of ozone and 180 ppbv of CO) is related to mixing between relatively clean background air and the biomass burning plume.

The stratospheric ozone tracer indicates that an air mass with a significant stratospheric component (100 ppbv of CO and 80 ppbv of ozone) mixed with both a relatively clean tropospheric airmass (115 ppbv of CO and 55 ppbv of ozone) and the BB plume (200 ppbv of CO and 50 ppbv of ozone).

The anticorrelations between the BB CO and stratospheric ozone tracers, P3 ozone and CO , and P3 relative humidity and CO , reinforce the hypothesis of mixing between a stratospheric intrusion and a biomass burning plume.

Figure 1. Top: NOAA P3 time series of CO (black line, ppbv), ozone (blue line, ppbv), σ_{ep} (gray line, Mm^{-1}), acetonitrile (green line, pptv/2) and nitric acid (red line, pptv) on September 25 between 16:00 and 16:25 UTC. Bottom: NOAA P3 time series of altitude (red solid line, km) and relative humidity (green solid line, %, right vertical axis). Also shown are the FLEXPART biomass burning CO tracer (black dashed line, ppbv), anthropogenic CO tracer (red dashed line, ppbv) and stratospheric ozone tracer (blue dashed line, ppbv).

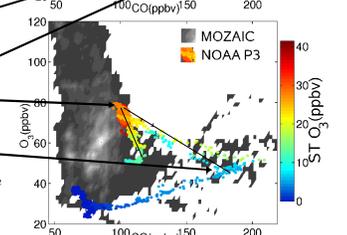
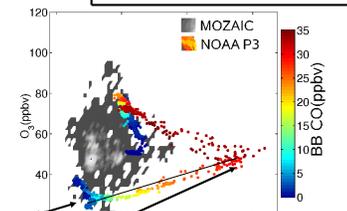


Figure 2. NOAA P3 ozone and CO mixing ratios on September 25, 16:00-16:25 UTC, colored by (top) the modeled biomass burning CO tracer and (bottom) by the stratospheric ozone tracer. The grey background represents the distribution of MOZIAC ozone and CO measurements in (top) the free troposphere (defined by 300hPa -pressure- $<800\text{hPa}$) and (bottom) the entire troposphere/lowermost stratosphere (pressure- $>300\text{hPa}$) of September. Lighter (darker) shading indicates a greater (lesser) number of measurements.

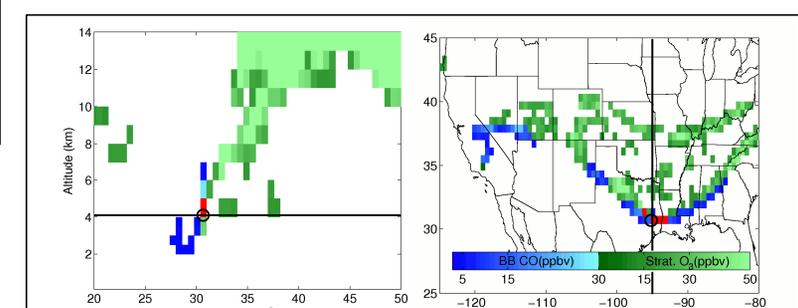


Figure 5. (Left) Vertical and (Right) horizontal cross sections of the FLEXPART stratospheric ozone tracer (green) and the biomass burning tracer (blue) at 16:00 UTC, September 25. Grid cells containing both tracers are shaded red. The black circles indicate the aircraft position at 16:10 UTC and the black lines indicate the altitude of the horizontal cross section (Left) and the longitude of the meridional cross section (Right).

Figure 5 (Right) presents a horizontal cross section of the FLEXPART stratospheric ozone tracer at 4 km altitude and the mean BB CO tracer between 3 and 5 km altitude. The narrow BB plume lies next to the tropopause fold from Texas to the Great Lakes region. The narrow BB plume seen in the GOES image (Fig. 3, top) is well simulated by FLEXPART (Fig. 5, Right). Figure 5 (Left) presents a meridional vertical cross section at the longitude of the aircraft position at 16:10 UTC (black circle in Fig. 5, Right). A potential mixing region (red pixels) is identified between 3 and 5 km, using a threshold of 10 ppbv for the BB CO tracer and 15 ppbv for the stratospheric ozone tracer, in the vicinity of the NOAA P3 aircraft (black circle).

Conclusion

This Lagrangian-based study confirms that mixing does occur between biomass burning plumes and stratospheric intrusions in the free troposphere. To assess the frequency of these events, we performed a statistical analysis using the FLEXPART forward simulations from August 1 to October 1, 2006 above the Texas and Gulf of Mexico region. We used the same FLEXPART simulations for stratospheric tracers and biomass burning, assuming an area burned of 180 ha per fire detection (Stohl et al., 2007). When a stratospheric intrusion (ozone tracer > 30 ppbv) was present in this region, some portion of it was co-located with biomass burning plumes (BB CO tracer > 10 ppbv) 65% of the time at the surface (2 stratospheric intrusions events), 74% between 100m and 4km of altitude (6 events), 28% between 4 and 8km (9 events) and 11% above 8km. The percentage decreases with altitude because the biomass burning plumes are less frequent in the free and upper troposphere. These results imply that mixing between stratospheric intrusions and biomass burning plumes can influence tropospheric chemistry and may have implications for surface air quality.