North American Boreal Wildfire and Anthropogenic Emission Impacts in the Lower Free Troposphere over the North Atlantic Region: Observations at the PICO-NARE station

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OVERVIEW

We present analyses of CO, O_3, nitrogen oxides (NO_x and NO_y) and aerosol black carbon (BC) measurements made in the lower free troposphere (FT) over the North Atlantic region during summers 2004 (ICARTT period) and 2005.

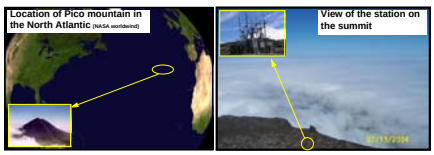
Main Findings:

1. **Boreal wildfires in 2004 dramatically impacted CO, BC, NO_x and NO_y and significantly impacted O_3. These wildfires resulted in very large-scale impacts on tropospheric BC, NO_x and O_3.**
2. **North American anthropogenic emissions during summer 2005 also impacted levels of CO, O_3, NO_x and NO_y although to a lesser extent than wildfire emissions in 2004.**

STATION LOCATION

The PICO-NARE station is located on the summit of Pico mountain (2.2 km asl) in the Azores Islands. The station was established in July 2001 and has been proven to be useful to study long-range transport of pollution in the FT [1,2].

The PICO-NARE station is currently in transition to become a permanent Global Atmospheric Watch station under Portuguese control with international participation.

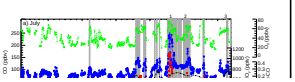


IMPACT OF BOREAL WILDFIRE EMISSIONS

1. Frequent impact of wildfire emissions during summer 2004 [Fig. 1]

Major fires in Alaska and Canada repeatedly impacted the PICO-NARE station during summer 2004. Fire-impacted periods accounted for 16% of the measurement time from July to early September, 2004.

Fig. 1. **Summer 2004 time series of CO, BC, NO_x, NO_y and O_3 with fire-CO fraction simulated by MOZART. Fire-impacted periods are indicated by hatched areas. Enumerated periods are analyzed in Fig. 2. Note the enhancements of the species coinciding with peaks in fire-CO simulated by MOZART.**



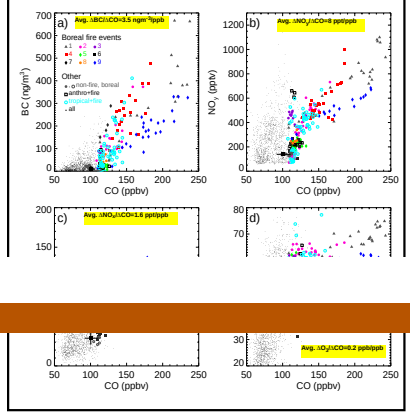
2. Extreme impacts on BC, NO_y and NO_x [Fig. 2 a-c]

- Levels of BC, NO_x and NO_y during fire-impacted outflow were well above typical summertime background at the site.
- Avg. $\Delta BC/\Delta CO$ (4 ngm^{-3}/ppb; Fig. 2a) and $\Delta NO_y/\Delta CO$ (8 ppt/ppb; Fig. 2b) were a significant fraction of estimated BC/CO (6 ngm^{-3}/ppb [3]) and NO_y/CO (12-26 ppt/ppb [4,3]) emission ratios from boreal wildfires. Avg. $\Delta NO_y/\Delta CO$ was also within the range of observed $\Delta NO_y/\Delta CO$ closer to fires (6-14 ppt/ppb [5]). This indicates the efficient long-range transport of BC and NO_y.
- Large $\Delta NO_y/\Delta CO$ (Fig. 2c) indicates that decomposition of PAN to NO_x was an important source of NO_x. High levels of NO_x imply continuing O_3 formation in these highly aged plumes.
- Large variability in $\Delta NO_y/\Delta CO$ and $\Delta BC/\Delta CO$ was observed, attributed to a combination of removal during transport and variation of fire types and emissions.

3. Complex behavior of O_3 [Fig. 2 d]

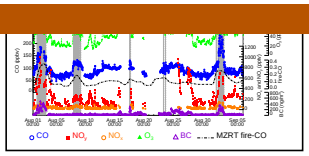
- Significant O_3 enhancements above background were observed in all but one plume. This indicates that O_3 photochemical production occurred in the fire plumes during
- Low $\Delta O_3/\Delta CO$ and negative O_3-CO relationships observed in some plumes imply suppression of O_3 production, likely due to reduced OH and limited NO_x or removal of O_3 due to reaction with organic aerosols or nighttime chemistry. This behavior is not well understood yet and deserves further study.

Fig. 2. **Relationship of BC, NO_x, NO_y and O_3 to CO in fire plumes. Colored symbols show fire periods unaffected by recent anthropogenic emissions or tropical air. Numbers correspond to those in Fig. 1.**



4. Large-scale implications

- North American wildfires in 2004 resulted in a significant source of BC, NO_y, NO_x and O_3 to the North Atlantic lower FT. Since our observations were made 6-15 days downwind from the wildfires, this suggests very large-scale impacts on tropospheric NO_x and O_3 budgets and on direct radiative forcing by BC.
- We estimate that the 2004 North American wildfires could result in a source as large as 40% of the typical summer NO_y exported from the U.S. and 10% of the summer net photochemical O_3 production in northern high latitudes. Likewise, total wildfires in the Northern hemisphere (including Siberia) in a typical year could result in as much as 80% and 20%, respectively.



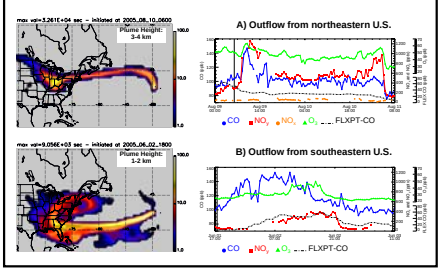
More information and references in Val Martin et al. [2006], Significant enhancements of NO_x, NO_y, BC and O_3 in the North Atlantic lower FT resulting from North American boreal wildfires, accepted, *J.Geophys. Res.*, doi:10.1029/2006JD007530.

IMPACT OF NORTH AMERICAN ANTHROPOGENIC EMISSIONS

1. Examples of impact of U.S. emissions [Fig. 3]

During summer 2005, we identified three periods with outflow from the northeastern U.S. and one period with very low altitude outflow from the southeastern U.S. These periods present clear outflow from the U.S. with no evidence of mixing with fire emissions.

Fig. 3. **Time series of CO, O_3, NO_x, NO_y and FLEXPART CO U.S. tracer during U.S. outflow (right). An example FLEXPART retroplume is also shown for each period (left). Although the time of FLEXPART is slightly off relative to the time of the observations, note the enhancements of the species associated with increases in U.S. CO simulated by FLEXPART.**



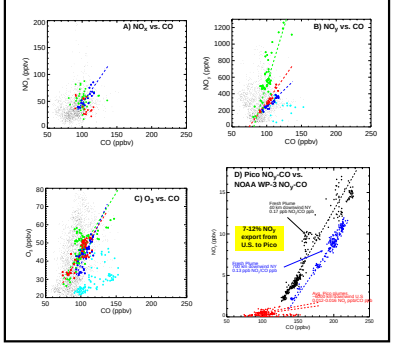
2. NO_x and NO_y in U.S. plumes [Fig. 4 a-c]

- NO_y and NO_x were significantly enhanced above background in all but one event. NO_y was highly correlated with CO, suggesting transport of NO_y in the plumes to the PICO-NARE station.
- Avg. NO_y-CO slope (0.012 ppb/ppb all events; 0.016 ppb/ppb well correlated events) was significantly lower than that observed in fresh plumes downwind from the eastern U.S. (0.13-0.17 ppb/ppb [6]), indicating the important removal of NO_y in urban plumes during transit. However, a fraction, 7-12% of NO_y is transported to the central North Atlantic.
- Exported NO_y and observed NO_x levels in the range of 50-90 ppt imply that additional potential O_3 formation may occur during transport.

3. O_3 versus CO in U.S. plumes [Fig. 4 d]

- O_3 was enhanced and well correlated with CO in all but one event, suggesting significant export of O_3 in these plumes.
- Observed O_3-CO slopes in the well correlated U.S. events (0.6-0.9 ppb/ppb) were larger than those observed in plumes up to 2 days downwind from the eastern U.S. during ICARTT [Andy Neuman, NOAA, personal communication]. This suggests that significant O_3 formation may occur during ~5-10 days transport to the PICO-NARE site.
- Avg. O_3-CO slope in well correlated events (0.7 ppb/ppb) during summer 2005 was somewhat smaller than that reported at Pico for similar outflow in 2001 and 2003 (~1 ppb/ppb [2]). This needs further evaluation.

Fig. 4. **Relationship of NO_x, NO_y and O_3 to CO in anthropogenic plumes. A, B and C: Observations at Pico. Plotted are all summer observations (small black dots), 3 events from northeastern U.S. (green, blue and red circles) and 1 event from southeastern U.S. (cyan circles). Regression line plotted when $r^2 > 0.5$. Data in Fig. 3A are green dots and in Fig. 3B are cyan circles. D: Comparison of Pico NO_y-CO (aged plumes) to NOAA-WP3 NO_y-CO (fresh plumes) (from [6]).**



CONCLUDING NOTES

- Our observations demonstrate that the summer 2004 North American boreal wildfires resulted in an important source of BC, NO_y, NO_x and O_3 in the North Atlantic lower FT, 6-15 days downwind from the fires. This suggests the very large-scale impacts of the wildfires both on tropospheric NO_x and O_3 budgets and on direct radiative forcing by BC, and supports previous conclusions that boreal wildfires impact the O_3 background over the North Atlantic region during high fire years [7].
- North American U.S. outflow during summer 2005 resulted in significant impact on NO_x, NO_y and O_3 in the lower FT of the Azores region, 5-10 days downwind from the eastern U.S. However, this impact was small relative to that resulting from fire-impacted boreal region outflow in summer 2004.
- Significant removal of NO_y occurred in the urban plumes during transit to the PICO-NARE station. However, export of NO_y and significant levels of NO_x (>50 ppt) in the urban plumes suggest additional O_3 formation may occur during transport. We are expanding the analysis of the impact of anthropogenic emissions on nitrogen oxides and O_3 to other seasons and regions (Asia and Europe).

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