Impacts of Anthropogenic and Boreal Forest Fire Emissions in the Central North Atlantic Lower Free Troposphere: Summertime Observations at the PICO-NARE Observatory

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1. INTRODUCTION

We present measurements of CO, O_3, aerosol Black Carbon (BC) made over the central North Atlantic lower Free Troposphere (FT) during the summers of 2001-2004 along with measurements of nitrogen oxides (NO_x and NO_y) made during the summer of 2004 (ICARTT period) and non-methane hydrocarbons (NMHCs) made during the winter 2004-spring 2005.

Main Findings: Anthropogenic and boreal wildfire emissions dominate impacts on CO, O_3, NO_x, NO_y, BC, and NMHCs over the North Atlantic lower FT during summer seasons.

2. STATION OVERVIEW

The PICO-NARE station is located in the Azores Islands, Portugal (2225 m asl), which is an ideal location for sampling North American pollution, forest fire emissions, and clean marine background air [Fig. 1].

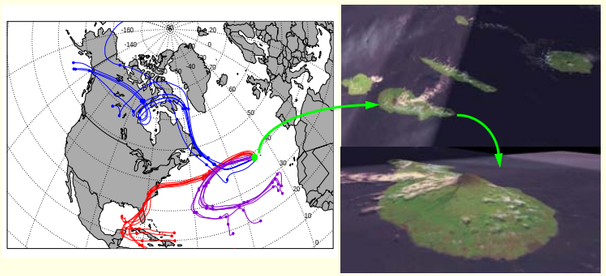

FIGURE 1. Location of the PICO-NARE station in the Azores Islands. Also shown are typical flow pathways that carry boreal fire emissions (blue), U.S. anthropogenic emissions (red) and clean marine background air (purple) to the station. Dots indicate each 2 days upwind. Also shown are a simulated view of the central Azores Islands and a view of the northwest side of Pico Island from an altitude of 6 km (Nasa Worldwind).

Free Tropospheric Air Sampling?

The station altitude is well into the FT in all seasons [Figs. 2-3]. However, marine boundary layer (MBL) air occasionally reaches the station due to daytime buoyant uplift (periods of weak winds) and mechanically driven uplift [1] (periods of strong winds) [Figs. 3-4].

FIGURE 2. View of the Pico mountain and view of the station near the cliff at the summit. MBL-capping clouds are visible beyond. Also visible is the ITOP BAE 146 airplane during an intercomparison flyby.

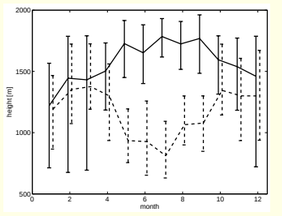

FIGURE 3. Annual cycle of MBL height (dashed) based on FNL data. Also shown is the dividing streamline height (solid), which is the minimum height of air transported to the summit by mechanical orographic uplifting. Lines connect medians, and error bars span from 1st to 3rd quartiles.

3. LARGE IMPACTS OF BOREAL WILDFIRES

3a. Strong Variability in CO, O_3, NO_x, NO_y and Aerosol BC

Major fires in Alaska and western Canada repeatedly impacted our station during the summer 2004. Extreme enhancements of CO, O_3, NO_x, NO_y, BC, and NMHCs (not shown) levels above background occurred during these periods [Fig. 4].

- $d[O_3]/d[CO]$ was similar to some previous reports for well-aged forest fire plumes, although in some cases O_3 production was suppressed, especially in the most concentrated part of the plume [Fig. 5a].
- $d[NO_y]/d[CO]$ was a significant fraction of the estimated NO_x/CO ratio [2] and only moderately smaller than previous measurements much closer to fires [Fig.5b], indicating limited NO_y removal during transport to the site and an additional potential for O_3 formation downwind [3].

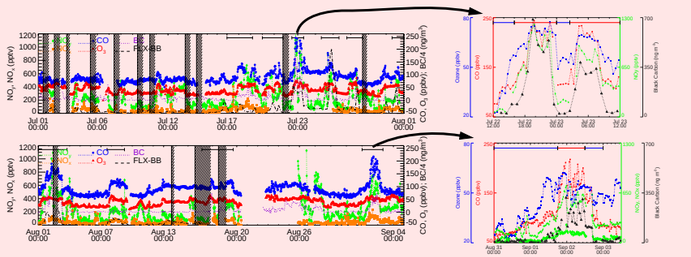

FIGURE 4. CO, O_3, NO_x, NO_y, BC, and Flexpart boreal fire CO tracer during summer 2004. Periods marked by bars along the top of the plot are events attributed to boreal fire emissions (based on analyses of back trajectories, MODIS images, and correlations among the species). Hatched areas show periods when buoyant and mechanically driven upslope flow may have impacted the site; these periods are not considered in the analyses. Also shown are two events in more detail. Blue and red bars on top of the plots indicate some of the periods plotted in Fig. 5.

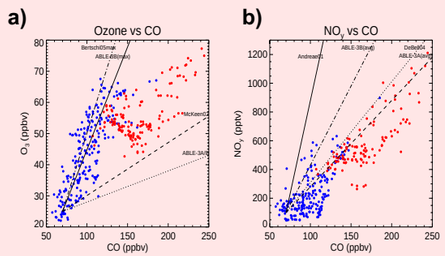

FIGURE 5. Scatter plots of O_3 and NO_y against CO during the summer 2004 boreal fire events. Periods with apparently differing $d[O_3]/d[CO]$ are in red. Also shown are O_3-CO and NO_y-CO enhancement ratios from previous observations [2,4-8].

3b. Interannual Variability

Summertime CO, BC and O_3 levels were higher in 2002 than in 2001, and much higher in 2003-2004 [Fig. 6]. This increase is attributed to enhanced fires in Quebec in 2002, and much enhanced fires in Siberia in 2003 and in Alaska and western Canada in 2004 [9].

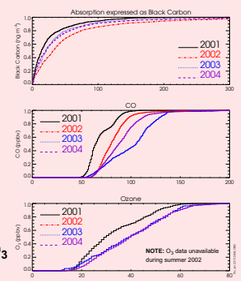

FIGURE 6. Cumulative distributions of all hourly average observations of BC, CO and O_3 during the 2001-2004 summers.

3c. Increase in Summertime O_3 Background?

To minimize the effect of transport differences in the assessment of boreal fire impacts on O_3, we compare observations in 2 subsets of each summer data: air that travelled north of 50ºN, but low CO, and air that travelled north of 50ºN with relatively high CO (indicating likely fire emissions) [Fig.7]. The high-CO subset exhibit ~20 ppbv higher median O_3, implying significant O_3 production from fire-emitted precursors [10].

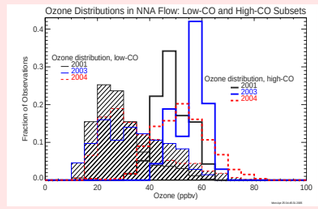

FIGURE 7. Frequency distributions of O_3 observations in northern North America flow only, divided into high-CO and low-CO subsets.

4. IMPACTS OF U.S. ANTHROPOGENIC EMISSIONS

Enhancements of CO, O_3, BC, NO_x, NO_y and NMHCs levels above background also occur during flow from the U.S. [Fig. 8]. Most of these events travel in the lower FT in a route governed by the Azores-Bermuda High and transient northerly lows [11], and typically have a photochemical age of 5-10 days as indicated by the "NMHC clock" [Fig. 9] and backward trajectories (not shown).

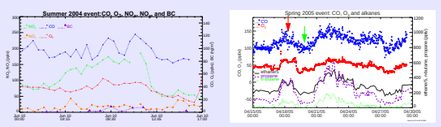

FIGURE 8. Observations of CO, O_3, NO_x, NO_y, BC, and NMHCs during two possible U.S. airflow events in summer 2004 and spring 2005. Arrows identify periods plotted in Fig. 9.

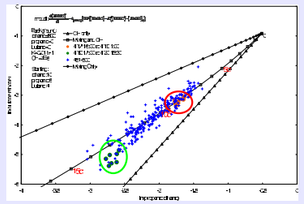

FIGURE 9. Apparent photochemical age determined from HC/HC ratios. Blue symbols show all winter 2004--spring 2005 HC observations; the red circles show data during the period of maximum mixing ratios around 4/18 and the green circles show data during the period of minimum mixing ratios around 4/20 [Fig. 8]. The lower and upper lines show the expected trend on this plot resulting from pure photochemical aging (reaction with OH) or mixing with background air. Approximate ages since emission are written along the center line, which reflects a combination of mixing and photochemical aging.

Evidence of Increased O_3 Export from the U.S.?

In 2001, few fires were observed and nearly all periods of elevated CO and O_3 occurred during U.S. outflow. The $d[O_3]/d[CO]$ slope during these periods was unexpectedly higher than those reported previously over the North Atlantic region, even after accounting for CO loss and for declining North American CO emissions [Fig. 10] [12]. This suggests an addition of significant amount of O_3 to air reaching Pico in 2001, relative to air near North America in the early 1990s.

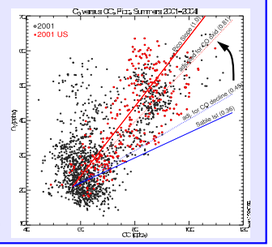

FIGURE 10. Comparison of the 2001 Pico $d[O_3]/d[CO]$ to a-priori expectations [12-14].

5. CONCLUSIONS AND FINAL REMARKS

- **Large boreal wildfires** in 2002-2004 **strongly impacte air quality over the central North Atlantic** lower FT, and inated the interannual variability of CO, O_3 and aerosol BC 2001-2004. The frequency distribution of O_3 shifted to higher levels, suggesting **impacts on the summertim background** over the region.
- **Nitrogen oxides levels** during fire-impacted periods **extremely high** for such a remote region (*e.g.*, NO_x > pptv), suggesting that significant **additional O_3 produ** must have occurred in these well-aged boreal fires plumes.

* *This deserves further study since little is known about impact of boreal wildfires on the O_3 levels in the North Hemisphere. Boreal wildfire activity is expected to incre in the future due to an increase in temperatures resu from global climate change* [15].

- **O_3 enhancements during U.S. outflow** transported to site over 5-10 days were significantly higher than t reported from previous observations.

Further work is needed to determine whether this indica **larger O_3 impacts from U.S. emissions that simulated Global Chemical Transport Models.*

- The **PICO-NARE station** has been proved to be a **valu platform** for observations of the regional background and and boreal fires impacts. (It is also impacted by European African emissions, although less frequently).

Additional climate-relevant measurements (CO_2, aeros and CCN**) are planned beginning in 2006 (support pendi PICO-NARE CO_2 and O_3 observations will be incorpora into the NOAA-CMDL record.*

We are working to convert the station into a perman Portuguese **GAW observatory over the next ~3 years.*

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