Large-scale Impacts of Anthropogenic and Boreal Fire Emissions Apparent in Multi-year Free Tropospheric Observations in the Azores

Richard Honrath: reh@mtu.edu

R. E. Honrath[1], M. Val Martin[1], J. Kleissl[1], R. C. Owen[1], K. Lapina[1], P. Fialho[2], D. Helmig[3], and D. Tanner[3]

[1]Department of Civil and Environmental Engineering, Michigan Technological University, U.S.
[2]Grupo de Quimica e Fisica da Atmosfera, Universidade dos Acores, Portugal
[3]Institute of Arctic and Alpine Research, University of Colorado, U.S.

1. INTRODUCTION

Pico mountain in the Azores Islands provides an ideal location for studies of the central N. Atlantic lower free troposphere. The PICO-NARE station has operated there since summer 2001. Here, we present key findings from summertime measurements, during 2001-2004.

Main Findings: Anthropogenic and boreal wildfire emissions dominate variations in CO and have major impacts on O_3, nitrogen oxides, non-methane hydrocarbons (NMHCs) and black carbon.

2. STATION OVERVIEW

The PICO-NARE station is located at 2225 m asl in the Portuguese Azores Islands. Fig. 1 shows the location and example flow pathways bringing clean marine air, N. American pollution, and subarctic air potentially containing boreal fire emissions.

Measurements at the station include: CO (2001-present), O_3 (2001, 2003-present), NO, NO_2, and NO_y (2002-2005), black carbon (2001-present), and NMHCs (2004-present). The station was recently shut down for the first time, for the 2005-06 winter season. Pending new funding, it will reopen in spring 2006 with new measurements of aerosol size distribution, CCN, and CO_2, and resumption of NO_x and NO_y measurements.

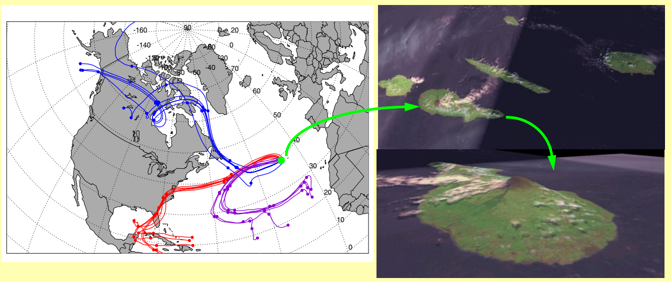

FIGURE 1. Location of the PICO-NARE station in the Azores Islands. Top right: The Azores Islands central group. Bottom right: a simulated view of the northwest side of Pico Island from an altitude of 6 km (NASA Worldwind).

Free Tropospheric Air Sampling

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

FIGURE 2. View of Pico mountain and of the station near the cliff at the summit.

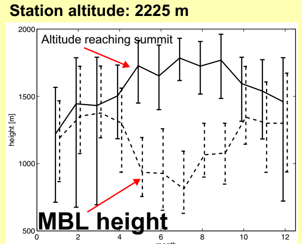

FIGURE 3. Annual cycle of MBL height (dashed) based on FNL data. Also shown is the dividing streamline height (solid), which is the minimum height of air transported to the summit by mechanical orographic uplifting. Lines connect medians, and error bars span from 1st to 3rd quartiles.

3. IMPACTS OF DISTANT BOREAL FIRES ARE LARGE

3a. Fire-plume enhancements in CO, O_3, NO_x, NO_y and black carbon

Major fires in Alaska and western Canada repeatedly impacted our station during summer 2004. Extreme enhancements of CO, O_3, NO_x, NO_y, BC, and NMHCs (not shown) 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 emission 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 likely O_3 formation downwind [3].

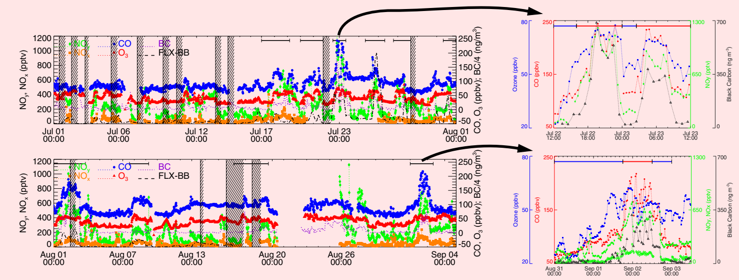

FIGURE 4. CO, O_3, NO_x, NO_y, BC, and FLEXPART boreal fire CO tracer during summer 2004. Periods marked by bars along the top of the plot are events attributed to boreal fire emissions (based on analyses of back trajectories, MODIS images, correlations among the species, and FLEXPART simulations [A. Stohl, see Acknowledgements]). Data during these periods are plotted in Fig. 5. 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.

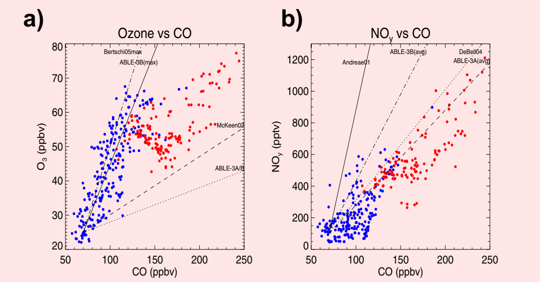

FIGURE 5. Scatter plots of O_3 and NO_y against CO during the summer 2004 boreal fire events. Colors highlight periods with differing O_3 enhancements. Also shown are O_3-CO and NO_y-CO enhancement ratios from selected previous studies [2,4-8].

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]. The CO and BC increases are attributed to enhanced fires in Quebec in 2002, and much enhanced fires in Siberia in 2003 and in Alaska and western Canada in 2004 [9,12].

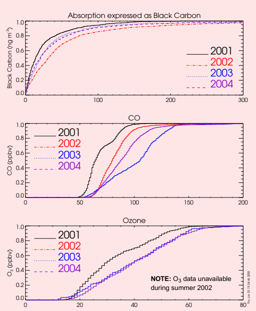

FIGURE 6. Cumulative distributions of all hourly average observations of BC, CO and O_3 during the summers of 2001-2004.

3c. Impact on summer O_3 background

To distinguish fire impacts on O_3 from effects of interannual variations in flow pathways, we compare O_3 observations in 2 data subsets each year: Measurements in air flowing from N of 50N but with low CO (low fire emissions) and those in air from N of 50N but with elevated CO (likely fire emissions). The fire-impacted data exhibit ~20 ppbv higher median O_3, implying significant O_3 impacts even after 7-10+ days transport [10].

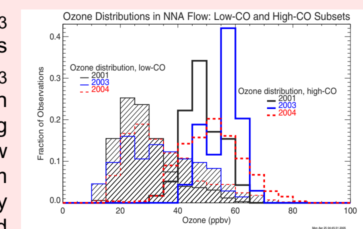

FIGURE 7. Frequency distributions of O_3 observations in northern North America flow only, divided into high-CO and low-CO subsets.

4. SIGNIFICANT IMPACTS OF N. AMERICAN 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).

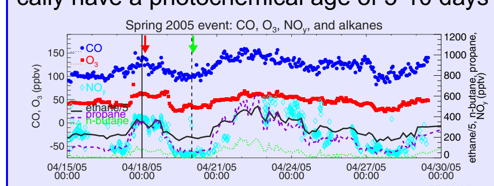

FIGURE 8. Observations during a spring 2005 apparent U.S. export event. Arrows identify periods plotted in Fig. 9.

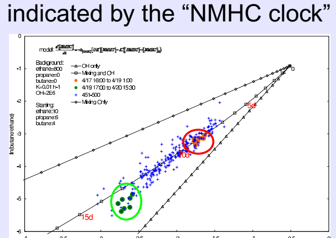

FIGURE 9. Apparent photochemical age determined from HC/HC ratios. Blue symbols show all winter 2004--spring 2005 HC observations; the red circles show data during the period of maximum mixing ratios around 4/18 and the green circles show data during the period of minimum mixing ratios around 4/20 [Fig. 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 significant O_3 production downwind of N. America

In 2001, few fires occurred, and nearly all periods of elevated CO and O_3 occurred during U.S. outflow. The $d[O_3]/d[CO]$ slopes during these periods were significantly higher than those reported previously near the N. American coast, even after accounting for CO loss in transit 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.

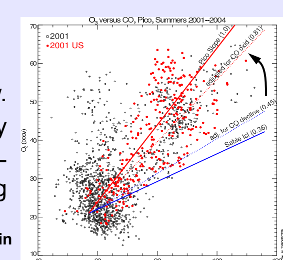

FIGURE 10. Comparison of the 2001 Pico $d[O_3]/d[CO]$ to slopes near North America in the 1990s [12-14].

5. CONCLUSIONS AND FINA… …EMARKS

- **Large boreal wildfires** in 2002-2004 … **…ngly impacted atmospheric composition over the ce… …North Atlantic lower FT**, and dominated the interannua… …bility of CO, O_3 and aerosol BC over 2001-2004. The fre… …distribution of O_3 shifted toward higher levels, **sug… …g significant impacts on the summertime O_3 backgr…** …over the region.
- **Nitrogen oxides levels** during fire-im… …periods were very high for such a remote region, sugg… …significant continuing O_3 production in these well-aged b… …ire plumes.

** This deserves further study since little… …own about the impact of boreal wildfires on the O_3 le… …the Northern Hemisphere. Boreal wildfire activity is e… …d to increase in the future due to an increase in te… …ures resulting from global climate change [15].*

- **O_3 enhancements during U.S. outflo…** …sported to the site over 5-10 days were significantly… …r than those reported from previous observations near… …America.

**Further work is needed to determine w… …this indicates larger O_3 impacts from U.S. emissions… …currently estimated.*

- The **PICO-NARE station** has been pro… …be a valuable platform for observations of the regiona… …ground and of U.S. and boreal fire impacts. It is also i… …d by European and African emissions, although less freq…

**Additional climate-relevant measureme… …O_2, aerosols, and CCN) are planned beginning in 200… …ding new support). Future PICO-NARE CO_2 and O_3… …ations will be incorporated into the NOAA-CMDL reco…*

**The Portuguese Met. Inst. and the Reg… …overnment of the Azores are making significant prog… …ward converting the station into a permanent GAW o… …ory in ~2008.*

REFERENCES

[1] Kleissl J. et al.,Analysis and application of Sheppard's airflow model to predict me… …raphic lifting, JAM, in preparation.
[2] Andreae, M. O. et al., Emission of trace gases and aerosols from biomass burnin… …001.
[3] Val Martin, M. et al. Observations of enhanced nitrogen oxides levels from borea… …e North Atlantic lower free troposphere. GRL, in preparation.
[4] Bertschi I.T and Jaffe D.A. Long-range transport of O_3, CO, aerosol to the NE… …sphere. during the summer of 2003: Observations of smoke plumes from Asian boreal fires. JGR, 110, 2005.
[5] Mauzerall, D. L. et al., Origin of tropospheric ozone at remote high northern latitu… …r. JGR, 101,1996.
[6] McKeen, S.A. et al., Ozone production from Canadian wildfires during June and… …JGR, 10.1029/2001JD000697, 2002.
[7] Wofsi S. C., et al., Atmospheric chemistry in the Arctic and subarctic: Influenc… …res, industrial emissions, and stratospheric inputs. JGR, 97, 1992.
[8] DeBell, L. J., et al. A major regional air pollution event in the northeastern Unite… …ed by extensive forest fires in Quebec, Canada. JGR, 109, 2004.
[9] Kasischke, E. S. et al., Influences of boreal fire emissions on Northern Hemisph… …ic carbon and carbon monoxide. GBC, 19, 2005.
[10] Lapina, K. et al., Impact of large-scale boreal fires on levels of tropospheric o… …orthern Hemisphere. GRL, in preparation.
[11] Owen, R.C. et al., Mechanism of North American export and transport to the… …Atlantic lower FT during July 2003. JGR, in preparation.
[12] Honrath, R.E et al., Regional and hemispheric impacts of anthropogenic and… …ng emissions on summertime CO and O_3 in the North Atlantic lower FT. JGR, 119, 10.1029/2004JD005147, 2004
[13] Parrish, D.D. et al., Relationship between O3 and CO at surface sites in the Nor… …ion. JGR, 103, 1998.
[14] Parrish, D.D. et al., Decadal change in carbon monoxide to nitrogen oxide ratio… …ar emissions. JGR, 2002.
[15] Stocks, B.J., et al., Large forest fires in Canada, 1959-1997. JGR, 108, 2003.

ACKNOWLEDGEMENTS

We thank Dr. Andreas Stohl for sharing simulations of the F… …model. Technical and logistical support was provided by Mike Dziobak (MTU). Additio… …sistance was provided by Forca Aerea Portuguesa, and Bombeiros Voluntarios do M… …o Pico. Jessica Strane (MTU) assisted with the analysis of the airflow events. This wo… …pported by NOAA/OGP grant NA030AR4510002 and NA16GP1658, and NSF grant AT… …3.