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

Hans-Jürg Jost,1 Katja Drdla,2 Andreas Stohl,3,4 Leonhard Pfister,2 Max Loewenstein,2 Jimena P. Lopez,2 Paula K. Hudson,5,6 Daniel M. Murphy,5 Daniel J. Cziczo,5,6 Michael Fromm,7 T. Paul Bui,2 J. Dean-Day,8 Christoph Gerbig,9 M. J. Mahoney,10 Erik C. Richard,4,6 Nicole Spichtinger,3 Jasna Vellovic Pittman,9 Elliot M. Weinstock,9 James C. Wilson,11 and Irène Xueref9

Received 10 December 2003; revised 29 January 2004; accepted 5 March 2004; published 2 June 2004.

[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 extratropical 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 (O3), 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 forest 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 (θ ≈ 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 (θ = 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 (CO2) [Daube et al., 2002] was also enhanced by about 1 ppm and the ratio of ΔCO/ΔCO2 was approximately 0.05, which is consistent with a biomass burning source. There was no enrichment in nitric oxide (NO), but total reactive nitrogen (NOy) [Weinheimer et al., 1998] was enhanced compared to the surrounding air indicating an aged pollution source where NO was already converted to NOY. 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...
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 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
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

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 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.

[10] Acknowledgments. This work was supported by the NASA ESE Radiation Science Program. We wish to thank the NASA WB-57 pilots and crew, B. Ridley, A. Weinheimer, D. Knapp, D. Montzka, F. Grahek for providing NO and NOy data, and Marion Legg and Oanh Nguyen for help with satellite data. We thank ECMWF and DWD for data access. MODIS data are distributed by the Land Processes Distributed Active Archive Center (LP DAAC), located at the U.S. Geological Survey’s EROS Data Center http://LPDAAC.usgs.gov. Work at the Jet Propulsion Laboratory, California Institute of Technology, was carried out under contract with the National Aeronautics and Space Administration.

References


J. Dean-Day, San José State University, One Washington Square, San José, CA 95192, USA.

M. Fromm, Computational Physics, Incorporated, 8001 Braddock Road, Suite 210, Springfield, VA 22151, USA.

C. Gerbig, J. V. Pittman, E. M. Weinstock, and I. Xueref, Harvard University, 12 Oxford Street, Cambridge, MA 02138, USA.

H.-J. Jost, Bay Area Environmental Research Institute, 560 Third Street West, Sonoma, CA 95476, USA. (hjost@mail.arc.nasa.gov)

M. J. Mahoney, Jet Propulsion Laboratory, 4800 Oak Grove Drive, Pasadena, CA 91109, USA.

N. Spichtinger, Technical University Munich, Am Hochanger 13, D-85354 Freising, Germany.

A. Stohl, CIERES, University of Colorado/NOAA Aeronomy Laboratory, 216 UCB, Boulder, CO 80309, USA.

J. C. Wilson, Department of Engineering, University of Denver, 2390 South York Street, Denver, CO 80208, USA.