Extreme CO concentrations in the upper troposphere over northeast Asia in June 2003 from the in situ MOZAIC aircraft data

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Received 4 April 2005; revised 11 May 2005; accepted 16 June 2005; published 20 July 2005.

[1] This paper analyses 320 MOZAIC (Measurements of Ozone aboard Airbus in-service airCraft) flights between Europe and East Asia in the year 2003. Carbon monoxide (CO) measurements in the upper troposphere (UT) clearly show the influence of plumes from boreal forest fires burning around Lake Baikal between April and July. On many flights, CO concentrations were above 300 ppbv over several hundred kilometers, with values above 500 ppbv over 50 kilometers and peaks up to 800 ppbv. To our knowledge, such high UT CO concentrations have never been reported before. A case study reveals that biomass burning plumes were rapidly transported by a warm conveyor belt. On the regional and seasonal scale, MOZAIC monthly-mean concentrations were above 150 ppbv on average, i.e. 30% above the northern hemisphere background as determined over Europe.

Citation: Nedelec, P., V. Thouret, J. Brioude, B. Sauvage, J.-P. Cammas, and A. Stohl (2005), Extreme CO concentrations in the upper troposphere over northeast Asia in June 2003 from the in situ MOZAIC aircraft data, *Geophys. Res. Lett.*, *32*, L14807, doi:10.1029/2005GL023141.

1. Introduction

[2] Carbon monoxide (CO) is a key trace gas in tropospheric photochemistry. Since the reaction with CO has a strong control on the hydroxyl radical (OH) concentrations in the unpolluted atmosphere [*Logan et al.*, 1981], the oxidizing capacity in the troposphere is influenced by the level of CO. Changes in OH oxidation, in turn, could perturb the growth rates of many greenhouse gases such as CH_4 and O_3 [*Thompson and Cicerone*, 1986]. CO_2 is directly perturbed by the oxidation of CO to CO_2. Due to its long lifetime (several months), CO is also a very good tracer for atmospheric transport processes, particularly for vertical transport. Thus, a better understanding of atmospheric CO and the processes influencing it is important.

[3] CO is mainly emitted in the atmosphere via incomplete combustion of biomass and fossil fuels. According to *Wotawa et al.* [2001], 63% of the CO variability in the extra-tropical Northern Hemisphere in summer can be explained by boreal forest fires in North America and Russia. Historically, CO concentrations have been measured globally at many monitoring stations in the planetary boundary layer [*Novelli et al.*, 1998, and references therein]. Since the year 2000, the MOPITT (Measurement of Pollution in the Troposphere) instrument delivers a global picture of CO [*Edwards et al.*, 2004, and references therein], showing CO maxima over biomass burning regions (e.g., over Africa) [*Edwards et al.*, 2003; *Bremer et al.*, 2004] and over northeast Asia [*Kar et al.*, 2004]. While aircraft, satellites and balloons were used for measurements of global CO, in situ data in the upper troposphere are still relatively sparse and the satellite data have relatively poor resolution, particularly in the vertical. The MOZAIC program [*Marenco et al.*, 1998; see Web site for recent details http://www.aero.obs-mip.fr/mozaic/] routinely performs CO measurements onboard five commercial Airbus A340 aircraft since December 2001 [*Nedelec et al.*, 2003]. This paper presents a large set of measurements over northeast Asia that reveals very high CO concentrations in the upper troposphere in 2003 (April to August). During this period, intense biomass burning occurred in this area, leading to long-range transport of Siberian fire plumes [*Jaffe et al.*, 2004].

2. Data

[4] We are using 320 MOZAIC flights between Europe and northeast Asia performed in 2003. Data are recorded from aircraft take-off to landing, providing vertical profiles and cruise data between 8 and 12.5 km altitude. In this study only cruise data taken above 8 km will be used. Table 1 gives the number of flights available for every month in 2003. Figure 1 displays the routes between Europe and northeast Asia in June 2003 (30 flights), superposed on a map of fire hot spots for the same month from Along Track Scanning Radiometer (ATSR). Note the large number of hot spots around Lake Baikal (100–120°E, 50–60°N). The orange box shows the northeast Asia region used for the climatological study at the end of the paper. CO measurements were performed using an improved infrared correlation instrument, with 30 seconds time resolution (7.5 km at cruise altitude) and a precision of ±5 ppbv ±5%, assumed to be the absolute accuracy.

Table 1. Number of Flights Between Europe and Northeast Asia in 2003

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Number	19	28	33	29	31	30	26	28	26	27	26	17

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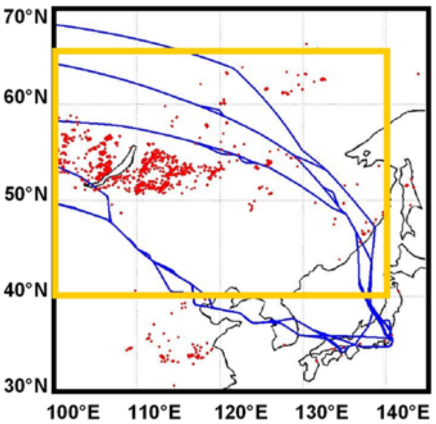

Figure 1. MOZAIC routes in June 2003 between Europe and northeast Asia (30 flights), superposed on a fire hot spot map for June 2003 from ATSR (http://dup.esrin.esa.it/ionia/wfa/index.asp).

Details on the measurement technique can be found in *Nedelec et al.* [2003].

3. Results

[5] From the MOPITT results over Asia presented in *Kar et al.* [2004], maximum CO concentrations in the UT over China were about 130 ppb in June 2000. Even though the global picture shows a maximum over Asia, the concentration is not strongly elevated above the background level, i.e. 110 ppbv as estimated from a preliminary climatological analysis of MOZAIC data. The nearly daily MOZAIC flights over northeast Asia yield a unique UT in situ CO data set that will be used in this paper to show that monthly-mean UT CO concentrations were extremely high in the boreal forest fire season (April to August) of 2003.

[6] Figure 2 shows the CO measurements between 40 and 65°N for 8 days in June 2003, where very high CO concentrations (>300 ppbv) were measured over very large areas (200–1000 km). This is the case for about half of the MOZAIC flights in June 2003. This suggests that Siberian biomass burning plumes may be regularly transported up to the UT. To our knowledge, these MOZAIC examples present the most striking cases in terms of CO concentrations and horizontal extent of biomass burning plumes reported so far. Asian plumes with 200–250 ppbv CO concentrations have been reported during TRACE-P [*Heald et al.*, 2003], 320–380 ppbv CO concentrations were found by Japanese commercial aircraft in November 1997 [*Matsueda and Inoue*, 1999]. *Damoah et al.* [2004] reported that in May 2003, the Siberian forest fires also studied here resulted in smoke plumes circling the Northern Hemisphere. Other authors reported high CO concentrations in altitude in different regions: over the Mediterranean Sea [*Fischer et al.*, 2002] or due to North American fires [*Jost et al.*, 2004; *Bertschi and Jaffe*, 2005].

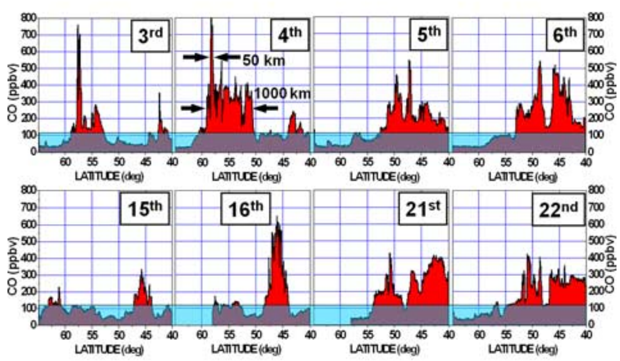

Figure 2. CO concentrations measured above 8 km over Asia between 40 and 65°N, for 8 days in June 2003. The light blue shading indicates the "background" CO level of 110 ppbv.

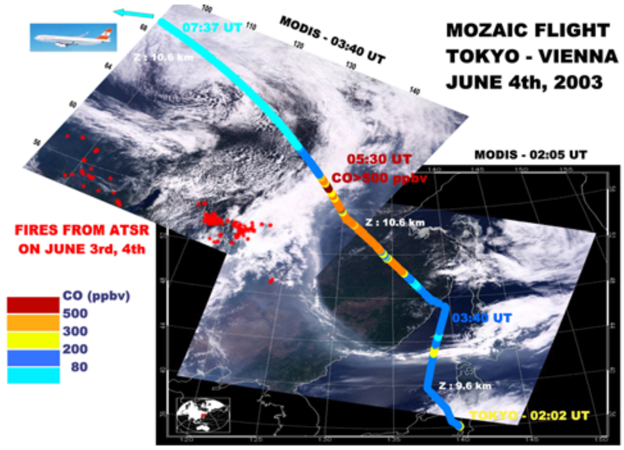

Figure 3. MOZAIC flight track color-coded with CO concentration (ppbv) from Tokyo to Vienna on June 4th, 2003, plotted over a composite of two MODIS satellite images taken on the same day at 02:05 and 03:40 UT. Time and altitude of the aircraft are plotted along the flight route. ATSR fire spots are plotted for June 3rd and 4th.

[7] A detailed analysis of the MOZAIC flight performed on the 4th of June 2003 (Figure 3) is supported by almost synchronous MODIS images (Moderate-Resolution Imaging Spectrometer, http://terra.nasa.gov/About/MODIS/). Below the northeast part of the aircraft route, a deepening surface low has its associated cold front that has swept across Lake Baikal the day before. In the UT the aircraft crosses two large smoke plumes, firstly north of Japan where CO peaks to 225 ppbv, and secondly over the continent with more than 300 ppbv over a distance exceeding 1000 km. Around 05:30 UTC within an area of 50 km, CO concentrations of more than 500 ppbv were detected, with peak values in excess of 850 ppbv. As far as we know, such high UT CO concentrations have never been reported before. The latter peak is recorded above the warm conveyor belt (WCB) clouds (measured relative humidity 65–70%) moving ahead of the surface cold front. *Cooper et al.* [2004] described the importance of WCB in this area for intercontinental transport. Two days before the inflow to the WCB was located directly above the hot spots. The daily sequence of aerosol index as seen by the Total Ozone Mapping Spectrometer (TOMS) instrument (Figure 4) confirms how aerosols from biomass fires on both sides of Lake

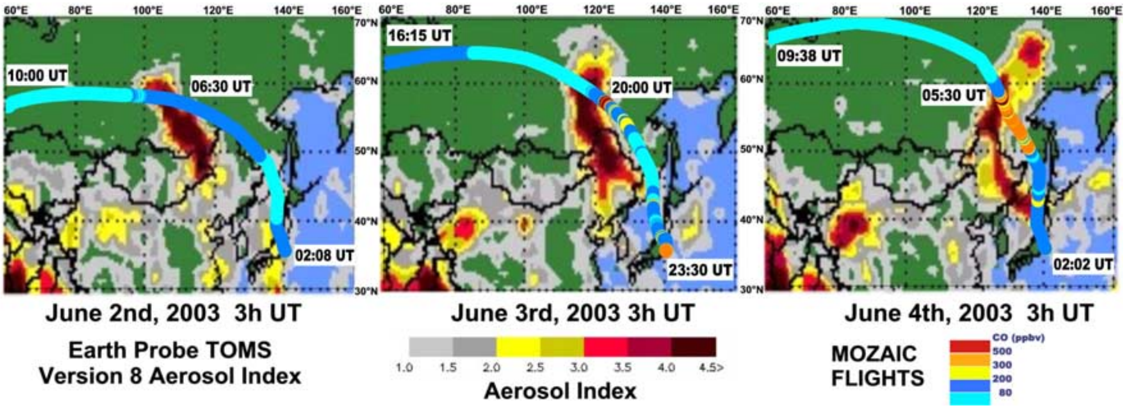

Figure 4. Daily aerosol index maps from Earth Probe TOMS (http://toms.gsfc.nasa.gov/eptoms/ep.html) with MOZAIC routes color-coded with CO concentrations (ppbv). TOMS orbits are at local noon and hours (UTC) are indicated along the aircraft paths.

Baikal have been lifted up by the progression of the cold front. A MOZAIC aircraft flies above the plume (no CO enhancement measured) on June 2nd, and two other flights cross the plume on the following two days (the spatial mismatch on June 3rd is due to the time lag between TOMS and the aircraft). As explained by *Fromm and Servranck* [2003] and *Fromm et al.* [2005], very high TOMS aerosol index values often indicate elevated smoke plumes, as the instrument is more sensitive to absorbing aerosols at greater altitudes. Hence variations in aerosol index are probably due to aerosol uplifting as the cold front swept across Lake Baikal.

[8] The FLEXPART model [*Stohl et al.*, 2003], based on ECMWF analyses, was run backward in time from small boxes generated along the aircraft route where measured CO concentrations were above 300 ppbv to calculate a sensitivity function to emission input. The column-integrated sensitivity up to 60 hours back (Figure 5) shows the largest values around Lake Baikal, indicating that fire emissions introduced into the atmospheric column around Lake Baikal would have had a strong influence on the measured concentrations. The highest sensitivities are found in the middle and/or upper troposphere, suggesting that the fire emissions were effectively released at altitude (GOES09 infrared images indicate deep pre-frontal convective cells from June 3rd 12:00 UTC to June 4th 00:00 UTC along 120°W and between 50 and 60°N on the eastern edge of the fires, auxiliary material[1]). Results from this backward modeling rule out southeast China as an emission source contributing to the large CO concentrations and, thus, confirm the boreal forest fires as the source.

[9] To further discuss these extreme events at the seasonal time scale, Figure 6 gives the seasonal cycle of CO in the UT over the year 2003 shown as monthly "box charts" for Europe and North East-Asia. To discriminate between troposphere and stratosphere we have used an ozone threshold corresponding to its monthly mean concentration at the tropopause as recently established by *Thouret et al.* [2005]. The strongest average CO concentration enhancements over Asia were found in June when as many as 75% of the measurements were above 110 ppbv and 5% were greater than 375 ppbv. However, extreme values were occasionally recorded in all months between April and August. This period corresponds exactly to the burning season in the vicinity of Lake Baikal in 2003 (see the ATSR web site for seasonal averages). The CO seasonal cycle over Europe does not exhibit such a strong variability, even though it was likely also influenced to some extend by the Siberian forest fires emissions [*Wotawa et al.*, 2001; *Damoah et al.*, 2004]. Spring maxima do not exceed 150 ppbv while fall minima are about 80 ppbv. Monthly-mean differences between the two regions

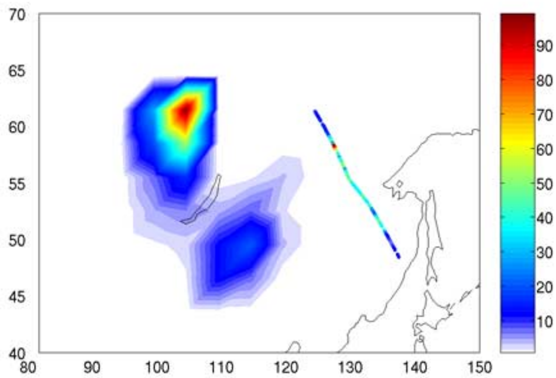

Figure 5. Emission sensitivity distribution in the whole atmospheric column for trajectory particles arriving in small receptor boxes (0.5° latitude and longitude, 500 m thickness) along the MOZAIC aircraft route on June 4th, 2003 within a 1-hour time interval. Particles were released only from boxes where the measured CO exceeded 300 ppbv. Residence times were calculated from arrival until 60 hours back and values are given as percentages of the maximum. The MOZAIC aircraft trajectory is color-coded with a percentage of the 800-ppbv maximum CO concentration.

[1] Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2005GL023141.

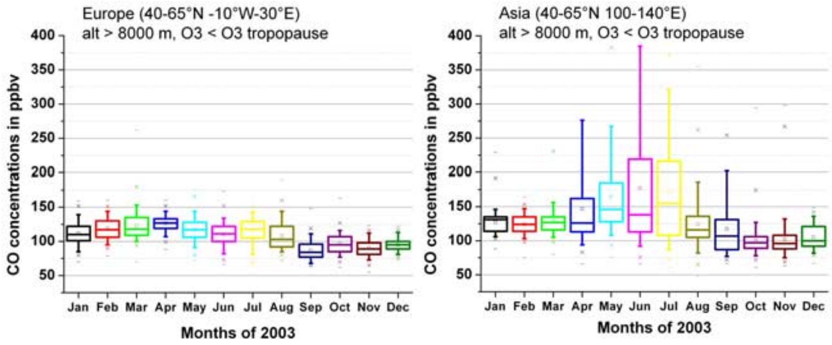

Figure 6. "Box charts" of the CO monthly frequency distributions for altitudes over 8000 m over Europe (left panel) and over northeast Asia (right panel) for 2003. Horizontal bars in the boxes give the median, while the boxes themselves extend from the 25% for the 75% percentiles. The small squares inside the boxes give the mean value, and the vertical bars give the 5% and 95% percentiles. Crosses give the 1% and 99% percentiles. Small dashes give the minimum and the maximum.

range from 25 to 60 ppbv during April–August, i.e., when the Siberian forest fires burned.

[10] Finally, the regional and seasonal CO anomaly over northeast Asia, attributed here to the transport of biomass burning products into the UT, calls for an assessment of the impact on ozone formation. Generally, no elevated ozone concentrations were seen in the plumes sampled in UT. However, the relative freshness of biomass fire plumes studied here prevents any preliminary assessment; this will require further studies.

4. Conclusion

[11] The analysis of 320 MOZAIC flights between Europe and Asia in 2003 shows that CO concentrations at 8–12.5 km altitude may be as high as 800 ppbv over a few tens of kilometers above Siberia. Concentrations of 300 to 500 ppbv were frequently sampled over meso-scale distances with MOZAIC aircraft during the boreal forest fire season. On the regional and seasonal scale, monthly-mean concentrations in the upper troposphere over northeast Asia from May to July 2003 were above 150 ppbv, i.e. 30% above the background determined from MOZAIC data over Europe (110 ppbv).

[12] Further studies are in preparation to better use the MOZAIC database in terms of quasi-global distribution, in terms of the chemical composition of the UT region and in terms of a comparison with satellite data.

[13] **Acknowledgments.** The authors acknowledge for their strong support the European Communities, EADS, Airbus and the airlines (Lufthansa, Austrian, Air France) who carry free of charge the MOZAIC equipment and perform the maintenance since 1994. We acknowledge the European Space Agency ESA/ESRIN for providing the ATSR World Fire Atlas and NASA for providing MODIS and TOMS free-access images.

References

Bertschi, I. T., and D. A. Jaffe (2005), Long-range transport of ozone, carbon monoxide, and aerosols to the NE Pacific troposphere during the summer of 2003: Observations of smoke plumes from Asian boreal fires, *J. Geophys. Res.*, *110*, D05303, doi:10.1029/2004JD005135.

Bremer, H., et al. (2004), Spatial and temporal variation of MOPITT CO in Africa and South America: A comparison with SHADOZ ozone and MODIS aerosol, *J. Geophys. Res.*, *109*, D12304, doi:10.1029/2003JD004234.

Cooper, O. R., et al. (2004), A case study of transpacific warm conveyor belt transport: Influence of merging air streams on trace gas import to North America, *J. Geophys. Res.*, *109*, D23S08, doi:10.1029/2003JD003624.

Damoah, R., N. Spichtinger, C. Forster, P. James, I. Mattis, U. Wandinger, S. Beirle, T. Wagner, and A. Stohl (2004), Around the world in 17 days-hemispheric-scale transport of forest fire smoke from Russia in May 2003, *Atmos. Chem. Phys.*, *4*, 1311–1321.

Edwards, D. P., et al. (2003), Tropospheric ozone over the tropical Atlantic: A satellite perspective, *J. Geophys. Res.*, *108*(D8), 4237, doi:10.1029/2002JD002927.

Edwards, D. P., et al. (2004), Observations of carbon monoxide and aerosols from the Terra satellite: Northern Hemisphere variability, *J. Geophys. Res.*, *109*, D24202, doi:10.1029/2004JD004727.

Fischer, H., et al. (2002), Deep convective injection of boundary layer air into the lowermost stratosphere at mid-latitudes, *Atmos. Chem. Phys.*, *3*, 739–745.

Fromm, M., and R. Servranck (2003), Transport of forest fire smoke above the tropopause by supercell convection, *Geophys. Res. Lett.*, *30*(10), 1542, doi:10.1029/2002GL016820.

Fromm, M., R. Belavicqua, R. Servranck, J. Rosen, J. P. Thayer, J. Herman, and D. Larko (2005), Pyro-cumulonimbus injection of smoke to the stratosphere: Observations and impact of a super blowup in northwestern Canada on 3–4 August 1998, *J. Geophys. Res.*, *110*, D08205, doi:10.1029/2004JD005350.

Heald, C. L., D. J. Jacob, P. I. Palmer, M. J. Evans, G. W. Sachse, H. B. Singh, and D. R. Blake (2003), Asian outflow and trans-Pacific transport of carbon monoxide and ozone pollution: An integrated satellite, aircraft, and model perspective, *J. Geophys. Res.*, *108*(D24), 4804, doi:10.1029/2003JD003507.

Jaffe, D. A., I. Bertschi, L. Jaeglé, P. Novelli, J. S. Reid, H. Tanimoto, R. Vingarzan, and D. Westphal (2004), Long-range transport of Siberian biomass burning emissions and impact on surface ozone in western North America, *Geophys. Res. Lett.*, *31*, L16106, doi:10.1029/2004GL020093.

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.

Kar, J., et al. (2004), Evidence of vertical transport of carbon monoxide from Measurements of Pollution in the Troposphere (MOPITT), *Geophys. Res. Lett.*, *31*, L23105, doi:10.1029/2004GL021128.

Logan, J. A., M. J. Prather, S. C. Wofsy, and M. B. McElroy (1981), Tropospheric chemistry: A global perspective, *J. Geophys. Res.*, *86*, 7210–7254.

Marenco, A., et al. (1998), Measurement of ozone and water vapour by Airbus in-service aircraft: The MOZAIC airborne program, an overview, *J. Geophys. Res.*, *103*, 25,631–25,642.

Matsueda, H., and H. Y. Inoue (1999), Aircraft measurements of trace gases between Japan and Singapore in October of 1993, 1996, and 1997, *Geophys. Res. Lett.*, *26*, 2413–2416.

Nedelec, P., J. P. Cammas, V. Thouret, G. Athier, J. M. Cousin, C. Legrand, C. Abonnel, F. Lecoeur, G. Cayez, and C. Marizy (2003), An improved infra-red carbon monoxide analyser for routine measurements aboard commercial Airbus aircraft: Technical validation and first scientific results of the MOZAIC program, *Atmos. Chem. Phys.*, *3*, 1551–1564.

Novelli, P., K. A. Masarie, and P. M. Lang (1998), Distributions and recent changes of carbon monoxide in the lower troposphere, *J. Geophys. Res.*, *103*, 19,015–19,033.

Stohl, A., C. Forster, S. Eckhardt, N. Spichtinger, H. Huntrieser, J. Heland, H. Schlager, S. Wilhelm, F. Arnold, and O. Cooper (2003), A backward modelling study of intercontinental pollution transport using aircraft measurements, *J. Geophys. Res.*, *108*(D12), 4370, doi:10.1029/2002JD002862.

Thompson, A. M., and R. J. Cicerone (1986), Possible perturbations of atmospheric CO, CH_4 and OH, *J. Geophys. Res.*, *91*, 10,853–10,864.

Thouret, V., J.-P. Cammas, B. Sauvage, G. Athier, R. Zbinden, P. Nedelec, P. Simon, and F. Karcher (2005), Tropopause referenced ozone climatology and inter-annual variability (1994–2003) from the MOZAIC programme, *Atmos. Chem. Phys. Discuss.*, in press.

Wotawa, G., P. C. Novelli, M. Trainer, and C. Granier (2001), Inter-annual variability of summertime CO concentrations in the Northern Hemisphere explained by boreal forest fires in North America and Russia, *Geophys. Res. Lett.*, *28*, 4575–4578.

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