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Atmospheric Chemistry and Physics Discussions

Increased Northern Hemispheric carbon monoxide burden in the troposphere in 2002 and 2003 detected from the ground and from space

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Abstract

Carbon monoxide total column amounts in the atmosphere have been measured in the High Northern Hemisphere (30°–90° N, HNH) between January 2002 and December 2003, based on the analysis of infrared solar spectra recorded with spectrometers of high and moderate resolution. They are compared to ground-level CO mixing ratios and to total column amounts measured from space by the Terra/MOPITT instrument. In comparison to the unperturbed 2000–2001 period, all these databases reveal increased CO abundances in 2002–2003 summer-autumn times, with maximum anomalies observed in September 2002 and August 2003. Using a simple two-box model, the corresponding annual CO emission anomalies have been found equal to 98 Tg in 2002 and 142 Tg in 2003, thus close to those for 1996 and 1998. It is most likely that strong boreal forest fires in the HNH induced the increased CO burdens.

1. Introduction

The importance of biomass burning for atmospheric composition, including the concentrations of greenhouse gases on global and hemispheric scales, is widely recognized (Andreae and Merlet, 2001; Galanter et al., 2000 and references therein). According to Holloway et al. (2000), during a normal year (i.e. without catastrophic wildfires) biomass burning contributes 748 Tg CO/year globally (including 68 Tg CO/year emitted by extratropical forest fires, Andreae and Merlet, 2001). This is much larger than the global contribution from the combustion of fossil fuel (300 Tg/year) and is comparable to contributions from biogenic hydrocarbons (683 Tg/year) or from methane oxidation (760 Tg/year). The CO emission from HNH boreal forest fires experiences significant interannual variations (Wotawa et al., 2001).

In a recent paper (Yurganov et al., 2004), the CO tropospheric burden anomaly in the High Northern Hemisphere (HNH) between 1996 and 2001 was determined from total column spectrometric measurements combined with surface *in situ* data (Novelli et al.,

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2003). Using a simple box model, the 1998 CO emission anomaly (in comparison to the period 1996–2001, without 1998) was estimated at 38 Tg/month in August and 96±29 Tg for 1998, close to the inventory calculated by Kasischke et al. (submitted)[1] but higher than most other estimates. The upper limit of the error due to possible changes in CO sinks was estimated at ±20%.

This paper presents updated results of surface-based measurements of CO burdens supplemented by new satellite data (Edwards et al., 2004). The CO burdens in summer-autumn of 2002 and 2003 were found to be higher than in 2000 and 2001, and the corresponding CO emission anomalies have been estimated in a way similar to that used by Yurganov et al. (2004).

2. Observational stations and methods

In this work we present CO total column amounts derived from infrared solar observations performed between January 2002 and December 2003 with Fourier transform spectrometers (FTIR) operated at seven stations located in the Arctic, Scandinavia, Western Russia, the European Alps, and the subtropical Atlantic (Table 1). The high resolution spectra (better than 0.005 cm^{-1}) were fitted with calculated spectra, and CO vertical profiles were retrieved. However, only total column amounts have been used in this paper. The spectra recorded by a Russian home-made grating spectrometer with a resolution of 0.2 cm^{-1} just allowed determination of total column amounts of CO (Yurganov et al., 2002). Rinsland et al. (1998) estimated the random and systematic error of a single total column measurement of CO as ±2–3% and ±5%, respectively. Details of retrieval procedures can be found in Hase et al. (2004) and Yurganov et al. (2004). These data (as well as others, see below) were used for a calculation of the CO burden in the reservoir 30° N–90° N between 0 and 10 km altitude.

[1] Kasischke, et al., "Influences of boreal fire emissions on Northern Hemisphere atmospheric carbon and carbon monoxide", Global Biogeochem. Cycles, submitted, 2004.

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Air samples collected in the surface atmospheric layer by the NOAA/CMDL Cooperative Air Sampling Network and by other programs (Table 2) were analyzed for local CO concentrations. Most of these measurements (which are accessible at the WDCGG archive, WMO, 2004) were made by gas chromatography/HgO reduction detection using instruments from Trace Analytical, Inc. (Novelli et al., 2003). At some stations, CO mixing ratios were measured continuously using non-dispersive gas correlation IR instruments (WMO, 2004).

The satellite-borne MOPITT (Measurements Of Pollution In The Troposphere) instrument is a thermal IR nadir-viewing gas correlation radiometer described in detail by Drummond (1992) and Deeter et al. (2003). MOPITT uses a cross-track scan, which allows for almost complete coverage of the Earth's surface in about 3 days, with individual pixels of 22×22 km horizontal resolution.

3. Results and discussion

Monthly mean CO total column (TC) amounts above 7 sites are plotted in Fig. 1. Ny Ålesund (Spitsbergen), Kiruna (Sweden), Harestua (Norway) and Zvenigorod (near Moscow, Russia) are low altitude stations and the measured total column amounts represent concentrations integrated over the entire atmosphere, thus including the boundary layer (BL), the free troposphere (FT), and the stratosphere. Total columns measured at the Alpine mountain sites Zugspitze, Jungfraujoch, and Izaña (Canary Isles) are CO amounts integrated between the altitude of the station and the top of the atmosphere. Monthly mean values in Fig. 1 (triangles) may be compared to values averaged over the reference period between March 2000 and February 2002 (heavy blue lines). These raw data clearly indicate higher than normal CO total columns during summer-autumn periods of 2002 and 2003 at all sites. A similar effect can be found in the MOPITT data also presented in Fig. 1.

Record high CO column amounts were measured at Zvenigorod in summer-autumn time 2002 with a maximum in September. According to the International Forest Fire

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News (IFFN, 2003), there was smoke pollution from peat and forest fires in the region around Moscow between July and September, 2002. This caused a dramatic reduction in visibility, to less than 100 m in the city, and also had detrimental impacts on the health of the Muscovite population. The fires reached a peak on 6 September. The observed CO spikes (up to 7.3 E18 molecules/cm^2 on 10 September 2002) reflected intrusions of this polluted air (there were no visible fires around the station itself). The horizontal extent of the highly polluted area remains uncertain and in this paper the days with these spikes were omitted. There were nine days in July–September, 2002 with CO columns that exceeded the summer time 2002 lowest daily value (2.02 E18 molecules/cm^2) by more than 4 standard deviations, i.e. above 2.9 E18 molecules/cm^2. Specifically, there were 1 (12), 1 (6), and 7 (10) such days in July, August, and September, respectively (numbers in brackets indicate the total observational days for each month).

The relative anomalies in CO abundance (monthly means divided by the values measured during the reference period) are plotted in Fig. 2. The anomalies in CO total columns for all data sets have two maxima, in September-October, 2002, and in July-August, 2003. The CO anomalies at the southernmost station Izaña (28° N) are lower than for those at other sites. This supports the assumption that northern mid-latitude wildfires play a decisive role in the anomaly in hemispheric CO burdens (c.f. Yurganov et al., 2004 conclusion for 1998).

The total column anomaly measured by the MOPITT and integrated over the HNH, has a similar temporal shape (Fig. 2). Edwards et al. (1994) analyzed the MOPITT data in conjunction with simultaneously measured aerosol optical depths (AOD) from the Moderate-resolution Imaging Spectroradiometer (MODIS). Fire counts from MODIS allowed estimating locations of fires. In September 2002 enhanced CO and AOD were observed over the European part of Russia. In 2003 forest fires started burning in May and were most intense in Siberia.

The boundary layer measurements at 16 stations located in the HNH (for coordinates see Table 2) are in general accordance with the total columns, but the maximum anomaly in 2002 is higher than that for the total column amounts (2003 data are still not

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ready for publicizing). A similar effect was observed in 1998 (Yurganov et al., 2004); a surface location of the CO source may explain this difference.

We assume that the interannual variations of CO column amounts occur mainly in the troposphere. This assumption may be justified by the fact that about 90% of CO column amount in the NH resides below 12 km (Zhao et al., 2002). Moreover, the time constant for the vertical exchange between troposphere and stratosphere is more than one year (Brasseur et al., 1999, p. 6).

The anomalies in CO burden in the reservoir 30° N–90° N, 0–10 km were calculated using three data sets (Fig. 3, top panel and left scale). Firstly, the anomalies in CO total column amounts at four low altitude stations, Harestua, Kiruna, Zvenigorod, and Ny Ålesund were averaged and multiplied by the surface area of the reservoir (open circles in Fig. 3). Secondly, the average anomalies in CO mixing ratios in the BL (available until December 2002) were multiplied by the number of air molecules in the lowest 1.5 km layer of the chosen reservoir. The density-altitude stratification was taken from the 1976 US Standard Atmosphere. These CO burden anomalies were added to the FT anomalies derived from in situ (5 mountain stations listed in Table 2 and indicated by *) and column measurements at the Jungfraujoch and Zugspitze stations (rectangles in Fig. 3). The data from the Izaña mountain station were not considered here, because the station is located outside the chosen reservoir. And thirdly, the CO burden anomalies for the same reservoir were calculated from the global MOPITT data (full green triangles). All data have similar shapes and are very close in their absolute values.

A box model was used to estimate the monthly mean anomalies in the CO source in the HNH taking into account transport to the Low Northern Hemisphere (LNH, 0° N–30° N, 0–10 km of altitude) and chemical removal.

$$P'_{HNH} = dM'_{HNH}/dt + L'_{trans} + L'_{chem} \quad (1)$$

$$L'_{trans} = (M'_{HNH} - M'_{LNH})/TAU_{trans} \quad (2)$$

$$L'_{chem} = M'_{HNH}/TAU_{chem} \quad (3)$$

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$$TAU_{chem} = 1/k\,[OH] \quad (4)$$

$$k = 1.5^{*}E{-}3(1+0.6^{*}p)\ cm^3\ mol^{-1}\ s^{-1} \quad \text{(Demore et al., 1997)}, \quad (5)$$

where P_{HNH} is the CO emission rate (in Tg/month), M_{HNH}, and M_{LNH} are CO tropospheric burdens in the HNH and LNH reservoirs (in Tg); dM_{HNH}/dt is the change in the tropospheric CO burden during the month, L_{trans} and L_{chem} are loss terms due to the transport between the semi-hemispheres and OH-consumption, respectively; p is air pressure in atm; the prime ($'$) designates the anomaly, i.e. the deviation from the average over the reference period between March 2000 and February 2002.

The interannual variations of the sink processes (in other words, TAU'_{chem} and TAU'_{trans}) were neglected. TAU_{trans} was calculated using the GEOS-CHEM global 3D CTM with assimilated 1998 meteorology (Bey et al., 2001; Yurganov et al., 2004). TAU_{chem} was calculated using OH seasonal/latitudinal/altitudinal field, derived by Spivakovsky et al. (2000). Yurganov et al. (2004) estimated that halving [OH] leads to 22% lower values of P' in August.

Burden anomalies for LNH (M'_{LNH}) were calculated from the surface measurements by Novelli et al. (2003) (1996–1999), from the MOPITT measurements (2002–2003), and were taken as the average of the two in 2000 and 2001.

Estimates of the CO emission anomalies in the HNH using the three data sets described before are plotted on the middle panel of Fig. 3 (the reference period is indicated by the shaded area and is different from that used by Yurganov et al. (2004)). Results presented in Figs. 1, 2, and 3 are given also in a tabular form in the Supplement (http://www.copernicus.org/EGU/acp/acpd/4/4999/acpd-4-4999-sp.pdf). All three data sets clearly reveal abnormally high CO emissions in 2002 and 2003. The monthly emission anomalies were summed annually and presented in Table 3. Since 1996 there have been four years with high CO emission of similar magnitude (1996, 1998, 2002, and 2003) and four years with low emission (1997, 1999, 2000, and 2001).

Hot spots detected by the Along Track Scanning Radiometer (ATSR) may be used as an independent indicator of wildfires (Arino and Plummer, 2001). The bottom panel

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of Fig. 3 illustrates the interannual variations of total numbers of hot spots after July 1996. The monthly anomalies of hot spots were calculated similarly to CO anomalies. 2002 and 2003 were characterized by positive anomalies of hot spots with maxima in August, 2002 and May–July, 2003. Positive anomalies of hot spots were observed also in 1996 and 1998. However, a positive anomaly in 2000 and negative anomalies of 1999 and 2001 disagree with anomalies of CO emissions during those years. This may be explained by varying nature of wildfires (e.g. contributions of low-temperature peat fires or high-temperature forest fires) and differences in CO emission factors between the years.

The average anomalies for all three data sets used were converted into absolute emission assuming some "normal" emission seasonal cycle. 2000 biomass burning emission rates (50.7 Tg CO/year) calculated for the MOZART-2 CTM (Schultz and Granier, personal communication, 2003; Schultz, 2002; Olivier et al., 1996) were used as normal (Fig. 4). In 1996 and 1998 maximum emissions were observed in August. In 2002 emissions had a broad maximum in July–August. In 2003 the emissions in August were also high, but lower than in June and July.

4. Conclusions

Spectroscopic measurements of carbon monoxide total column amounts from ground based stations in the Arctic and Europe reveal increased CO abundance in summer and autumn time of 2002 and 2003 in comparison with the previous two years. Similar increases were observed in 1996 and 1998 (Yurganov et al., 2004). Increased CO concentrations near the surface were also observed by the CMDL and GAW networks (16 stations) in summertime of 2002. Moreover, space-based MOPITT measurements with a full HNH coverage reveal a similar pattern in 2002 and 2003. An inversion of monthly burden anomalies into a source anomaly (assuming a stable sink) has been done using a simple box model. The annual emission anomalies in 2002 and 2003 were similar in magnitude to those in 1996 and 1998, in the range 100–140 Tg CO/year. The anoma-

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lies in the CO emission rates correlate well with anomalies of fire counts, detected by a satellite-based radiometer. It is most likely that strong wildfires are responsible for the hemispheric CO build up.

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Table 1. Sites of total column measurements and characteristics of spectrometers.

Site	Coordinates	Altitude, m a.s.l.	Type of spectrometer	Typical resolution, cm^{-1}	Typical number of spectra per day
Ny Ålesund, Spitsbergen	78.92° N 11.94° E	20	Bruker IFS 120 HR	0.005	2–3
Kiruna, Sweden	67.84° N 20.41° E	419	Bruker IFS 120 HR	0.005	2–3
Harestua, Norway	60.22° N 10.75° E	596	Bruker IFS 120 M	0.005	12±6
Zvenigorod, Russia	55.70° N 36.80° E	200	Grating, home-made	0.18–0.23	17±6
Zugspitze, German Alps	47.42° N 10.98° E	2964	Bruker IFS 120 HR	0.0045	8±4
Jungfraujoch, Swiss Alps	46.55° N 8.00° E	3580	Bruker IFS 120 HR	0.0028 and 0.0044	6
Izaña, Spain	28.3° N 16.48° W	2367	Bruker IFS 120 M	0.005	1–3

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Table 2. Surface CO monitoring locations.

Name	Agency	Latitude	Longitude	Altitude, m
Alert, Nunavut, Canada	CMDL	82.45	−62.50	210
Ny-Ålesund, Spitsbergen, Norway	CMDL	78.90	11.88	475
Barrow, Alaska, USA	CMDL	71.32	−156.60	11
"M" Ocean Station, Norway	CMDL	66.00	2.00	7
Heimaey, Vestmannaeyjar, Iceland	CMDL	63.25	−20.15	100
Shetland Island, UK	CSIRO	60.17	−1.17	30
Cold Bay, Alaska, USA	CMDL	55.20	−162.72	25
Mace Head, Galway, Ireland	CMDL	53.33	−9.90	25
Shemya Isl. Alaska, USA	CMDL	52.72	174.10	40
Vancouver, Estevan Pt., Canada	CSIRO	49.38	−126.54	39
Zugspitze, Germany (*)	IMK-IFU	47.42	10.98	2964
Jungfraujoch, Switzerland (*)	EMPA	46.55	7.98	3578
Park Falls, Wisconsin, USA	CMDL	45.93	−90.27	868
Rishiri Isl, Japan	NIES	45.07	141.12	35
Sary Taukum, Kazakhstan	CMDL	44.45	77.57	412
Kaz_mount, Plateau Assy, Kazakhstan (*)	CMDL	43.25	77.88	2519
Niwot Ridge, Colorado, USA (*)	CMDL	40.05	−105.58	3475
Wendover, Utah, USA (*)	CMDL	39.90	−113.43	1320
Ryori, Japan	JMA	39.03	141.83	230
Azores, Terceira Island, Portugal	CMDL	38.77	−27.38	40
St. Davids, Bermuda, UK	CMDL	32.37	−64.65	30

Notes: 1) (*) indicate mountain stations.
2) Monitoring agencies: CMDL = Climate monitoring and Diagnostics Laboratory, Boulder, Colorado, USA; CSIRO = Commonwealth Science and Industry Research Organization, Canberra, Austaralia; JMA = Japan Meteorological Agency, Tokyo, Japan; NIES = National Institute of Environmental Studies, Tsukuba, Japan; IMK-IFU = IMK-IFU, Forschungszentrum Karlsruhe, Garmisch-Partenkirchen, Germany; EMPA = Swiss Federal Laboratories for Materials Testing and Research, St. Gallen, Switzerland.

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Table 3. Annual CO emission anomalies in Tg CO/year for the HNH retrieved from the low-altitude FTIR (P′(FTIR)); in situ BL, combined with mountain FTIR and in situ data (P′(BL+FT)); and satellite measurements (P′(MOPITT)) using a box model. The average of the three estimates is also given.

Year	P′(FTIR)	P′(BL+FT)	P′(MOPITT)	Average	Accuracy, Tg/yr
1996	166.6	80.8		123.7	40
1997	14.8	2.5		8.7	6
1998	149.2	121.3		135.2	20
1999	39.3	18.8		29.1	10
2000	−4.7	3.8	1.8	0.3	4
2001	9.7	−4.6	−0.9	1.4	7
2002	120.5	99.4	73.5	97.8	30
2003	165.3		118.0	141.6	30

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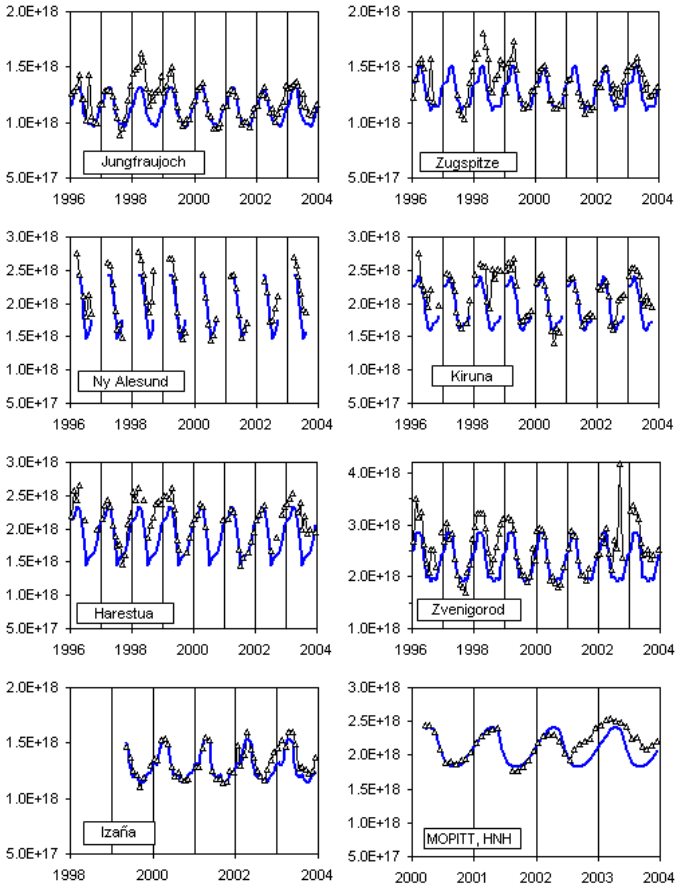

Fig. 1. Monthly mean column amounts (triangles; in molecules/cm^2) of carbon monoxide above various sites. Heavy blue lines correspond to the monthly means, averaged over the period between March, 2000, and February, 2002.

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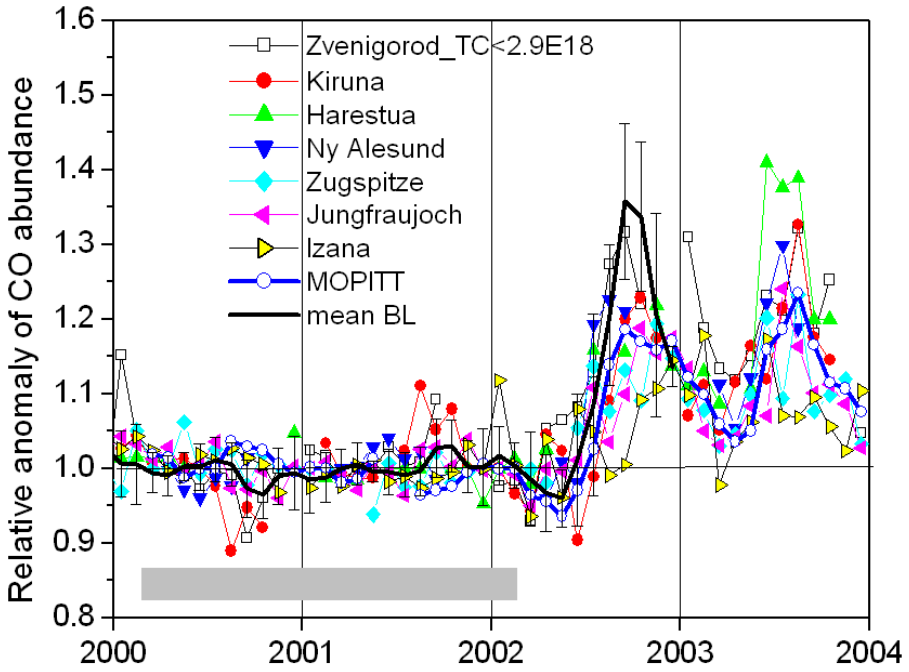

Fig. 2. Anomalies of CO total column amounts and surface mixing ratios (measured monthly means divided by the averages over the period between March 2000 and February 2002, grey rectangle). MOPITT data are averaged over the HNH, mean BL is the average over the data of 16 low-level stations with standard deviations. Extreme daily values exceeding 2.9 E18 molecules cm^{-2} in Zvenigorod in July–August, 2002, were omitted (see text).

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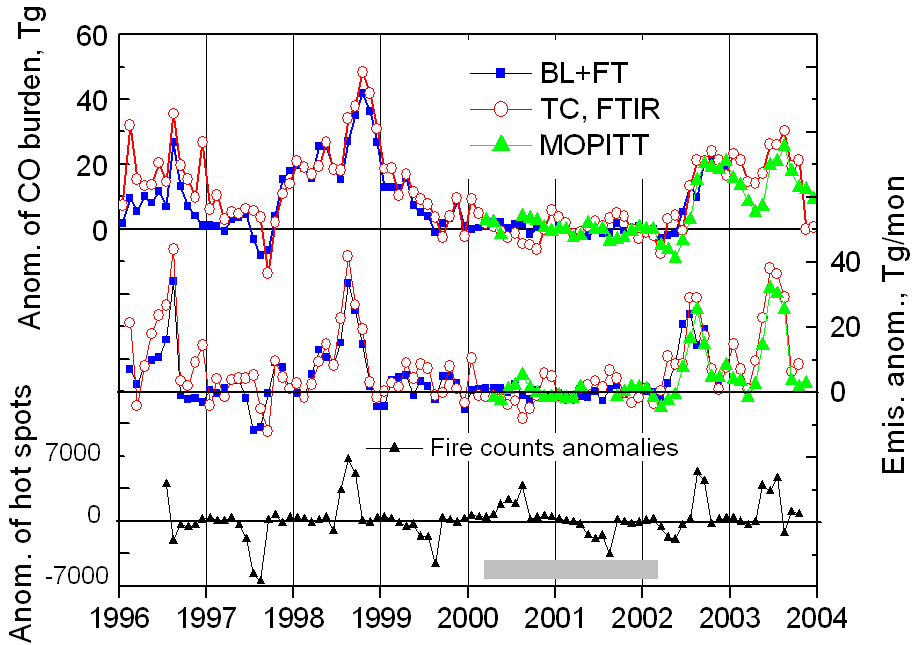

Fig. 3. Top panel and left axis. Deviations of CO burden in the HNH from "normal" values averaged over the period indicated by the grey rectangle. "TC, FTIR" curve represents the average anomaly measured at four low altitude stations. "BL+FT" curve corresponds to the BL network (sixteen stations) for lower 1.5 km, the in situ data of six mountain stations, and two Alpine FTIR (until December, 2002). "MOPITT" curve is the anomaly of CO total column amounts measured by MOPITT and integrated over the HNH. Middle panel and right axis. Anomalies of CO emission rates in the HNH derived from three data sets for CO burden (top panel), using the box model (see text). Bottom panel and left axis. Anomalies of monthly ATSR fire-counts integrated over HNH.

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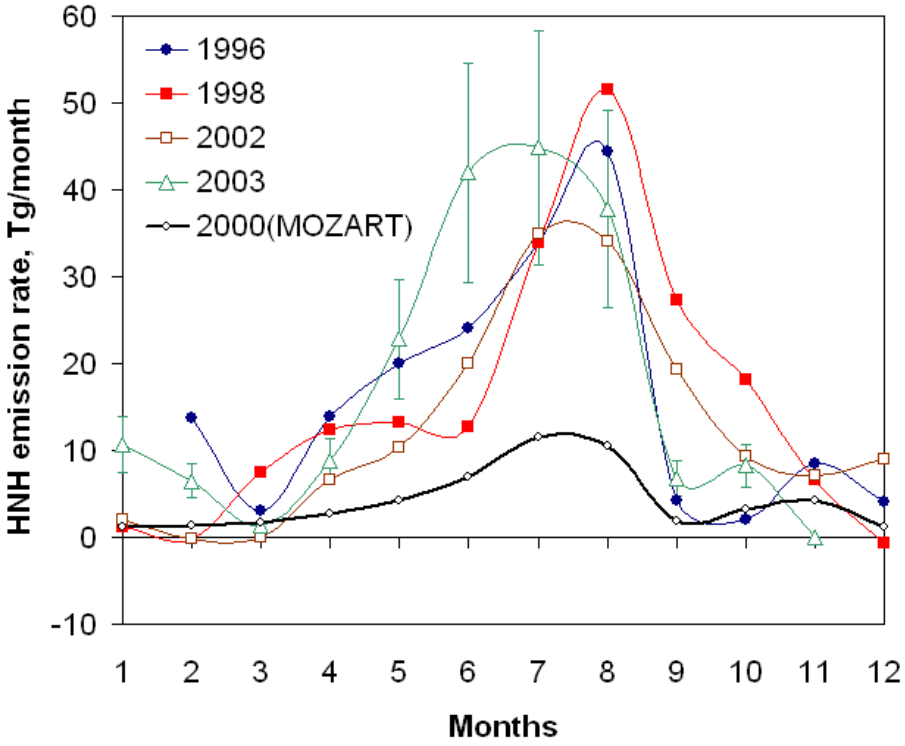

Fig. 4. Seasonal variations of CO emission from the biomass burning in the HNH calculated from a top-down estimate of the anomaly (Fig. 3) plus the normal seasonal cycle (MOZART-2 inventory for 2000). Error bars correspond to 30% accuracy (Yurganov et al., 2004).

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