Emissions of gaseous mercury from biomass burning in South America in 2005 observed during CARIBIC flights

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[1] Plumes of biomass burning effluents were observed during CARIBIC flights between São Paulo and Santiago de Chile on August 31 and October 5, 2005, as well as during the last part of the flight from Frankfurt to São Paulo on October 4, 2005. Total gaseous mercury (TGM) correlated with CO on August 31 and October 4 yielding a TGM/CO emission ratio of $(1.2 \pm 0.2) \times 10^{-7}$ and $(2.4 \pm 1.0) \times 10^{-7}$ mol/mol, respectively. No significant TGM/CO correlation was observed on October 5 probably because of variable background concentrations of both gases. The TGM/CO emission ratios observed here over South America fall within the rather narrow range of $(0.67–2.4) \times 10^{-7}$ mol/mol reported hitherto for sites geographically as different as South Africa, Canada, and the U.S.A. A total average emission of 437 Tg CO/yr from biomass burning over the years 1996–2000 implies an average TGM emission of 210–750 t/yr from biomass burning, representing 3–11% of all mercury emissions. TGM emissions from biomass burning are likely to be larger than anthropogenic emissions in the southern hemisphere during the burning season in August–October.
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1. Introduction

[2] Mercury is emitted into the atmosphere from a number of natural as well as anthropogenic sources. Long-range atmospheric transport, the transformation to highly toxic methylmercury and subsequent bioaccumulation in the aquatic food chain may result in human exposure to hazardous levels even in ecosystems remote from local sources [*National Research Council*, 2000].

[3] Worldwide mercury emissions by anthropogenic activities such as fossil fuel combustion, metal and cement production, as well as waste incineration, are presumed to be 2235 t/yr in 1995 [*Pacyna and Pacyna*, 2002] and 2190 t/yr in 2000 [*Pacyna et al.*, 2006], representing about one third of total mercury emissions of about 6600 t/yr [*Mason and Sheu*, 2002, and references therein]. One third of the emissions is attributed to natural emissions from land and oceans and the rest to re-emission of anthropogenic mercury [*Mason and Sheu*, 2002]. However, emission of mercury from biomass burning, an important source of many other pollutants [e.g., *Andreae and Merlet*, 2001] is not mentioned in the studies. In this paper we report observations of mercury emissions from large scale biomass burning in South America. The biomass burning plumes were observed during regular CARIBIC (*C*ivil *A*ircraft for *R*egular *I*nvestigation of the Atmosphere *B*ased on an *I*nstrumented *C*ontainer, available at www.caribic-atmospheric.com) flights to South America in 2005.

[4] Substantial emissions of mercury from biomass burning have been reported only recently. *Brunke et al.* [2001] observed a Hg/CO emission ratio of 2.1×10^{-7} mol/mol during a fire of mixed vegetation at Cape Point in South Africa. *Friedli et al.* [2001, 2003a, 2003b] reported Hg/CO emission ratios of 0.67×10^{-7} mol/mol for a forest wildfire in Washington (USA) and 2.04×10^{-7} mol/mol for a fire burning black spruce in a boreal forest in Quebec (Canada). Hg/CO emission ratios of 1.46×10^{-7} and 1.57×10^{-7} mol/mol for biomass burning in Pacific Northwest and Alaska, respectively, were derived recently from observations at Mt Bachelor observatory [*Weiss-Penzias et al.*, 2007]. With a total CO emission of 437 Tg/yr from biomass burning [*Duncan et al.*, 2003], these emission ratios imply mercury emissions ranging from 209 to 656 T/yr, a substantial part of the total emissions mentioned above.

[5] Here we report Hg/CO emission ratios derived from aircraft observations over South America which appear to be comparable to those mentioned above and thus underline the global scale importance of mercury emissions from biomass burning.

2. Experimental

[6] The new CARIBIC container [*Brenninkmeijer et al.*, 2007] holds automated analyzers for total gaseous mercury (TGM), CO, O_3, NO, NOy, CO_2, O_2, total and gaseous H_2O, oxygenated volatile organic compounds, fine aerosol particles (three counters for particles with diameters >4 nm, >12 nm, and >18 nm), larger particles (>150 nm, optical particle counter). Also a differential optical absorption system (for HCHO, BrO, and others) is present. Furthermore, air and aerosol particle samples are taken in flight and subsequently analysed for greenhouse gases, hydrocarbons, halocarbons, and elemental composition of aerosols, respectively. The TGM instrument (Tekran®) is based on enrichment of mercury (sample air is first passed through a 0.2 μm

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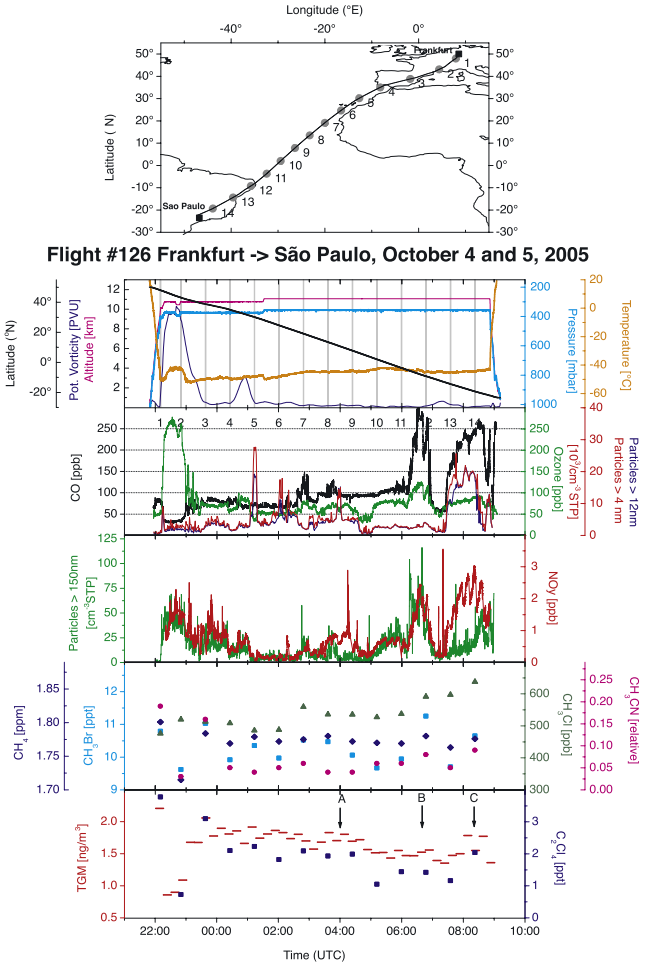

Figure 1. CARIBIC flight on October 4, 2005, from Frankfurt to São Paulo. Fourteen whole air samples were taken at locations shown in the uppermost panel and analyzed for greenhouse gases, halocarbons, and some other trace gases. Selected results of these analyses are shown in the two bottom panels.

PTFE filter) on a gold collector followed by detection by atomic fluorescence upon its thermal desorption. The instrument was modified for automatic operation at air pressures down to 200 hPa. During the flights reported here it was operated with 15 min long sampling intervals with a measurement uncertainty of about 0.05 ng/m^3. The TGM concentration is given in ng/m^3(STP, 1013.25 hPa, 273.15 K). CO is measured by a fast VUV fluorescence detector with a time resolution of 1 s and a measurement uncertainty of about 1 ppb. For details of the instruments and the aircraft inlet system see *Brenninkmeijer et al.* [2007]. For correlations with TGM the CO data were averaged over the sampling interval of the TGM instrument.

[7] The CARIBIC container is operated monthly onboard a Lufthansa Airbus 340–600 during flights to South America or Southeast Asia. The flights reported here were made on the route Frankfurt- São Paulo- Santiago de Chile and back.

3. Results and Discussion

[8] Figure 1 gives an overview of the flight from Frankfurt to São Paulo on October 4–5, 2005. With the exception of two shorter intervals of intercepting stratospheric air (October 4, 22:15–23:30, and October 5, 0:30–1:15, as indicated by elevated PV and O_3 concentrations) the flight (cruising altitude about 11 km) took place in the troposphere. The pronounced minimum in O_3 concentrations October 5, 4:50 UTC coincided with the crossing of the ITCZ at latitude of about 6N, as was found from analysis of satellite observations of clouds and of meteorological analysis of ECMWF data. Apart from the lower concentrations in the stratosphere, TGM concentrations in the lowermost panel of Figure 1 show a pronounced latitudinal gradient with lower concentrations in the southern hemisphere (SH). The latitudinal TGM distribution in the upper troposphere is not well known because of lack of data but the concentrations of 1.70–2.21 ng/m^3 observed in the northern hemisphere (NH) lie within the range of 1.44–2.38 ng/m^3 measured at the ground in the years 1995–2002 [*Slemr et al.*, 2003]. This is in agreement with the absence of pronounced vertical TGM gradients reported so far [*Ebinghaus and Slemr*, 2000; *Banic et al.*, 2003]. In the SH only the lowest TGM concentrations of 1.35 and 1.36 ng/m^3 fall within the range of 1.14–1.37 ng/m^3 measured at the ground in 1995–2002 [*Slemr et al.*, 2003]. Opposite to the sharp TGM gradient at the ITCZ over the surface of the Atlantic Ocean [*Slemr et al.*, 1985], only a gradual decrease from the higher TGM concentrations in the NH to the lower ones in the SH is expected in the upper troposphere in analogy to the tropospheric CO distribution [*Seiler*, 1974].

[9] Superposed on this general gradient are three local TGM maxima on October 5 at about 4:00 UTC (arrow A), 6:45 (arrow B), and 8:30 (arrow C). All these TGM maxima are accompanied by maxima of NO_y but only the last two by pronounced CO maxima. Eight day backward trajectories [*Scheele et al.*, 1996] (calculated at 1 min intervals, i.e. 15 trajectories per TGM measurement) for the first maximum (A) suggest the significant mercury emissions to have originated in southern West Africa. However, lack of enhanced concentrations of tracers for biomass burning, such as CO, CH_3CN, CH_3Cl, and CH_3Br in this plume preclude emissions from this source [*Andreae and Merlet*, 2001].

[10] In contrast to plume A, the two plumes (B and C) intercepted few hours before landing at São Paulo bear all characteristic features of biomass burning: enhanced concentrations of CH_3CN, CH_3Cl, and CH_3Br in addition to those of NO_y, CO, and aerosol particles [*Andreae and Merlet*, 2001]. Higher concentrations of O_3 and of large aerosol particles (>150 nm) together with a lack of smaller particles (<150 nm) in plume B and the opposite characteristics of the plume C suggest that the plume C is a fresh one and the plume B has already been photochemically processed. Eight day backward trajectories for the plume B show that the intercepted air mass had surface contact about 6 days before in the area of Central Brasil, south of the Amazon river (southern Pará and Mato Grosso). Most of the backward trajectories for the plume C point to a long range high altitude transport (above 250 hPa) of air from above the Pacific Ocean or Peru and Ecuador. Only a few of the trajectories had contact with surface during the 7th or 8th day in the area of Colombia, Venezuela, and the Brazilian states of Roraima and Amazonas, of which only the last one had a few fire counts. The observed pollution thus had to be

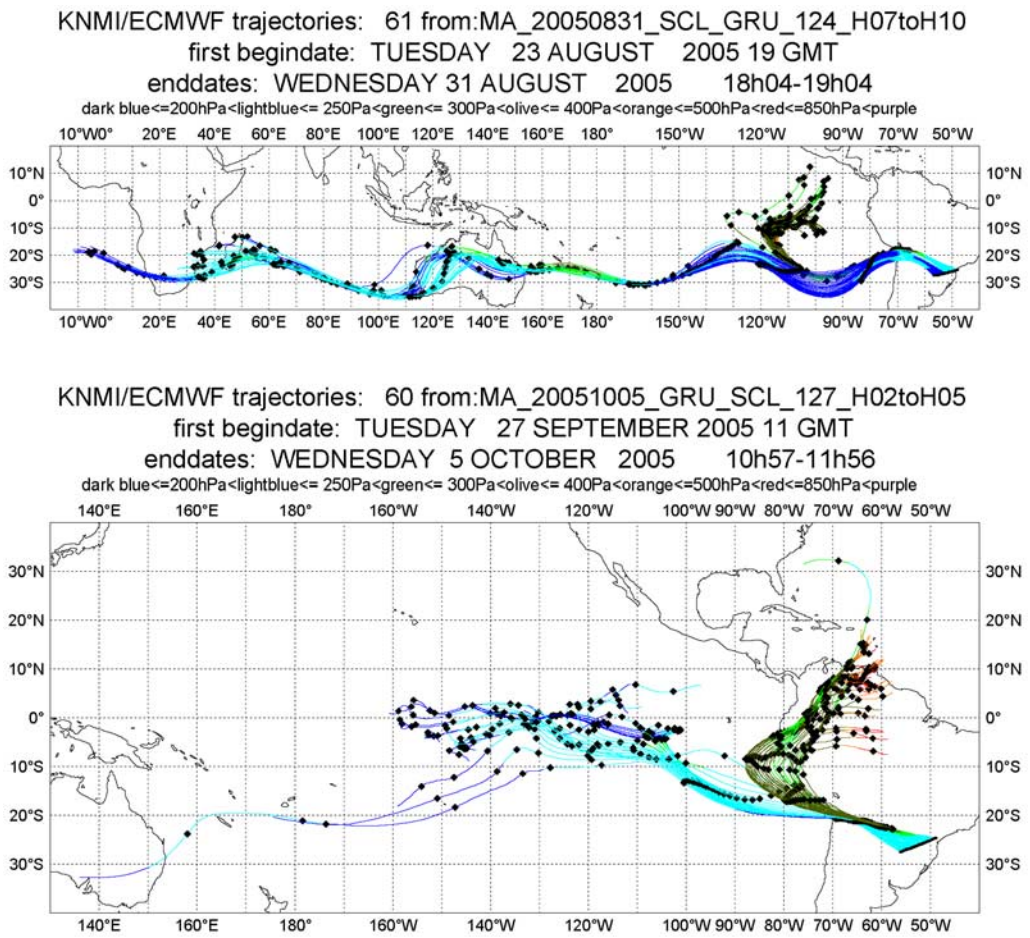

Figure 2. Eight day backward trajectories for the air encountered during the tropospheric sections of (top) the return flight from Santiago de Chile to São Paulo on August 31, 2005 and (bottom) the outgoing flight from São Paulo to Santiago on October 5, 2005. The trajectories for the tropospheric sections of the outgoing flight on August 31 and return flight on October 5 were similar and are thus not shown.

injected by convective transport into the air masses crossing the continent between 20° and 30°S. Convective transport of polluted air to the flight level from a region of south-west Brazil (Mato Grosso do Sul) is suggested by satellite pictures showing a pronounced cloud band in this region. Furthermore, satellite pictures (AQUA/MODIS, TERRA/MODIS, GOES-12) witness numerous fires on October 4 in Brazil and Paraguay with highest density in Bahia, Goiás, Tocantins, Minas Gerais, Mato Grosso, and Pará.

[11] Pronounced biomass burning plumes with enhanced TGM and CO concentrations were also observed during the short flights from São Paulo to Santiago and back on August 31 and October 5, 2005 (not shown). All four 3-h-flights are very similar with a tropospheric section for about 1 h after the take off or before landing at São Paulo and a stratospheric one for the rest of the flight (with exception of the descent to Santiago). The tropospheric section is characterized by very high CO concentrations of up to 480 ppb on August 31 and 350 ppb on October 5. The data on concentrations of O_3, NO_y, aerosol particles, CH_3CN, and halocarbons, although not as complete as those shown above for the flight on October 4, all indicate plumes of biomass burning with some photochemical processing between the two extremes observed on October 4. As shown in Figure 2 (top) all backward trajectories for the tropospheric part of the two flights on August 31 indicate a high altitude long range transport (above 300 hPa) from above southern Pacific Ocean. Consequently, the observed pollution must have been brought to the flight level by deep convection, most probably in a cloud band extending from states Paraná and São Paulo in Brazil to northern Bolivia as shown by satellite pictures and ECMWF analyses. On August 31 the strongest fire activity observed by satellites was located in Pará, Mato Grosso, Amazonas, and Rondonia, but substantial activity was also detected in Tocantins, Minas Gerais, São Paulo, as well as in Peru and Paraguay.

[12] On October 5 the backward trajectories for the tropospheric part of the flight, shown in the Figure 2 (bottom), indicate a slow transport of air masses from northern South America that "originated" near the surface in a large region encompassing the Amazon basin, Peru, Ecuador, Colombia, and Venezuela. A cloud band with the same position as on August 31 suggests additional convection of polluted air in the states of Parana and São Paulo. In summary, the observed air pollution on August 31 was likely injected by a deep convection of polluted air masses from southern Brazil into homogeneous southern hemispheric background air masses whereas on October 5

Table 1. Overview of Hg/CO Emission Ratios in Plumes From Biomass Burning Reported So Far

Region	Hg/CO Emission Ratio, mol/mol	Reference
South America, Aug. 31, 2005, flights #123+ 124	$(1.17 \pm 0.15) \times 10^{-7}$, $R^2 = 0.924$, n = 7, 99.9%	This work
South America, Oct. 5, 2005, flight #126	$(2.39 \pm 0.99) \times 10^{-7}$, $R^2 = 0.540$, n = 7, 95%	This work
Cape Point, South Africa	$(2.10 \pm 0.21) \times 10^{-7}$	*Brunke et al.* [2001]
Quebec, Canada	2.04×10^{-7}	*Friedli et al.* [2003a]
State Washington, USA	$(0.67 \pm 0.04) \times 10^{-7}$	*Friedli et al.* [2003b]
Pacific Northwest, USA	$(1.46 \pm 0.90) \times 10^{-7}$	*Weiss-Penzias et al.* [2007]
Alaska, USA	$(1.57 \pm 0.67) \times 10^{-7}$	*Weiss-Penzias et al.* [2007]

convective transport took place into air masses which were already preloaded by emissions from a large area of northern South America. These air masses were transported southward and upward by the South American low level jet [*Marengo et al.*, 2004] and by convection triggered by this jet, likely through interaction with the upper level jet.

[13] Correlations of TGM with CO have been used to derive information about the TGM/CO emission ratio of biomass burning [*Andreae and Merlet*, 2001; *Brunke et al.*, 2001; *Friedli et al.*, 2003a, 2003b; *Weiss-Penzias et al.*, 2007] and their theoretical justification has been discussed by *Jaffé et al.* [2005]. We use orthogonal regression [*York*, 1966] with TGM and CO uncertainties set to 0.05 ng/m^3 and 1 ppb, respectively. Significant correlations of Hg with CO were observed on August 31 (flights 123 and 124, significance level 99.9%, $R^2 = 0.924$, TGM range 1.36–1.65 ng/m^3) and for the fresh plume on October 4 (flight 126, 95%, $R^2 = 0.540$, TGM range 1.35–1.78 ng/m^3), but not for the plume observed on October 5 (flights 127 and 128). The above meteorological analysis and the difference in backward trajectories suggest that the lack of correlation on October 5 was most probably due to inhomogeneous TGM and CO concentrations upwind of the source area. Nearly constant TGM and CO concentrations upwind of the source area are one of the major conditions for the calculation of Hg/CO emission ratios [*Jaffe et al.*, 2005].

[14] The TGM/CO emission ratios are summarized in Table 1 together with other ratios reported for biomass burning. Despite of very different regions (boreal and tropical forests) and types of vegetations (fynbos shrubs, pines, tropical vegetation) they all fit a rather narrow range of $(0.67 - 2.4) \times 10^{-7}$ mol/mol. A total average emission of 437 Tg CO/yr from biomass burning over the years 1996–2000 [*Duncan et al.*, 2003] then implies an average mercury emission of 210–750 t/yr, representing 3–11% of total mercury emissions. Based on the geographical distribution of the CO emissions [*Duncan et al.*, 2003], almost all of the mercury will be emitted in the tropics (30°N–30°S). Emission of TGM from biomass burning will vary strongly from year to year, and may reach as much as 257–917 t/yr in years with anomalously high biomass burning such as 1997/1998 [*Langenfelds et al.*, 2002]. As for other gases, the interannual variation of TGM concentrations caused by interannual variation of biomass burning of this magnitude should be revealed by long term high precision monitoring of TGM.

[15] The emission of mercury from biomass burning is also strongly dependent on the season. Based on CO emission of 188 Tg/yr [*Duncan et al.*, 2003] about 90–321 t/yr of TGM will be emitted in the SH between June and November with a peak of about 63 t/month in September. Taking into account the geographic distribution given by *Wilson et al.* [2006], about 418 t/yr of mercury are emitted in the SH. The seasonal amplitude of anthropogenic emissions is only about 12% as can be roughly estimated from the seasonal amplitude of 18% for coal burning [*Rotty*, 1987], representing 65% of anthropogenic Hg emissions [*Pacyna et al.*, 2006], and no seasonal variation for all other emissions (cement production, ore processing, alkali production, and waste incineration). Consequently, during the burning season the monthly emission from biomass burning is comparable to or even surpasses the monthly anthropogenic emissions of 35 t/month ± 12% in the SH.

4. Conclusion

[16] Plumes of biomass burning with enhanced TGM concentrations were observed over South America during the CARIBIC flights on August 31, October 4 and October 5, 2005. TGM/CO emission ratios of $(1.2 \pm 0.2) \times 10^{-7}$ and $(2.4 \pm 1.0) \times 10^{-7}$ mol/mol were derived from the observations on August 31 and October 4, respectively. These are the first TGM/CO emission ratios reported for tropics where 90% of all CO from biomass burning is emitted [*Duncan et al.*, 2003]. The TGM/CO emission ratio observed in the tropics fall in a rather narrow range of emissions ratios determined elsewhere lending confidence in estimates of worldwide TGM emissions from biomass burning. With 3–11% of all mercury emissions, biomass burning represents one of the major mercury sources which should be included in global and regional models of atmospheric mercury.

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