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Gaseous mercury distribution in the upper troposphere and lower stratosphere observed onboard the CARIBIC passenger aircraft

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Abstract. Total gaseous mercury (TGM) was measured onboard a passenger aircraft during monthly CARIBIC flights (Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrumented Container) made between May 2005 and March 2007 on the routes Frankfurt–São Paulo–Santiago de Chile and back and Frankfurt–Guangzhou–Manila and back. The data provide for the first time an insight into the seasonal distributions of TGM in the upper troposphere and lower stratosphere (UT/LS) of both hemispheres and demonstrate the importance of mercury emissions from biomass burning in the Southern Hemisphere. Numerous plumes were observed in the upper troposphere, the larger of which could be characterized in terms of Hg/CO emission ratios and their probable origins. During the flights to China TGM correlated with CO in the upper troposphere with a seasonally dependent slope reflecting the longer lifetime of elemental mercury when compared to that of CO. A pronounced depletion of TGM was always observed in the extratropical lowermost stratosphere. TGM concentrations there were found to decrease with the increasing concentrations of particles. Combined with the large concentrations of particle bound mercury in the stratosphere observed by others, this finding suggests either a direct conversion of TGM to particle bound mercury or an indirect conversion via a semivolatile bivalent mercury compound. Based on concurrent measurements of SF₆ during two flights, the rate of this conversion is estimated to 0.4 ng m⁻³ yr⁻¹. A zero TGM concentration was not observed during some 200 flight hours in the lowermost stratosphere suggesting an equilibrium between the gaseous and particulate mercury.



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