Total gaseous mercury distribution in the UT/LS observed onboard the CARIBIC passenger aircraft

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Introduction

Total gaseous mercury (TGM) was observed between May 2005 and March 2007 on the routes Frankfurt – São Paulo – Santiago and Frankfurt – Guangzhou – Manila shown in Fig. 1. The data provide first insights in the distribution and seasonal variation of TGM in the UT/LS region and in the mechanism of TGM depletion In the LS.

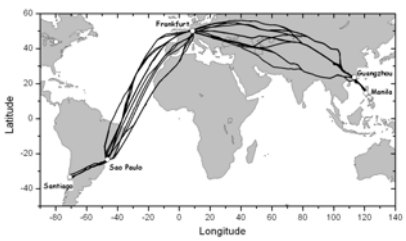

Fig. 1: CARIBIC flights from May 2005 to March 2007.

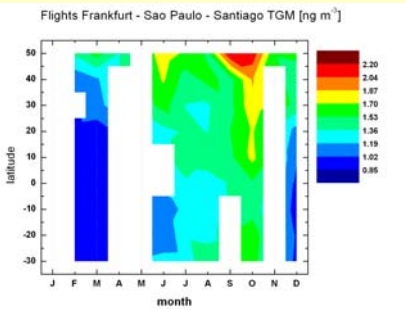

Fig. 2: Time latitudeTGM distribution in the UT during the flights from Frankfurt to South America.

TGM in the upper troposphere

- Hg emissions from biomass burning observed during the burning season over South America (Ebinghaus et al., Geophys. Res. Lett. 34, L08813, doi:10.1029/2006GL028866 – these emissions are partly responsible for the seasonal variation of TGM in the UT of the southern hemisphere see Fig.2
- enhanced TGM concentrations observed over Europe and Southeast Asia – see Figs. 2 and 3 - the Hg/CO emission ratios in the plumes observed over Southeast Asia correspond to the values reported by others
- TGM correlates with CO outside of the plumes – the Hg/CO slopes show a pronounced seasonal variation which reflects the different lifetimes of TGM and CO – see Fig. 4 - the wintertime Hg/CO correlation slope of about 3 ng m^{-3} ppb^{-1} is smaller than that of industrial emissions

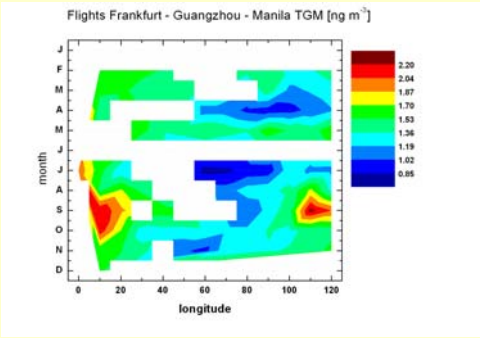

Fig. 3: Time longitude TGM distribution in the UT during the flights to Guangzhou and Manila

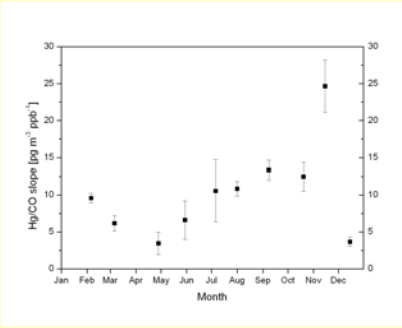

Fig. 4: Seasonal variation of Hg/CO slopes in the UT outside of the plumes.

TGM in the lower stratosphere (LS)

- TGM decreases with increasing PV, O_3, NOy and decreasing CO, acetone, water – see Fig. 5
- TGM depletion was always observed – Hg/CO correlation slopes of about 11 ng m^{-3} ppb^{-1} in both hemispheres and without any seasonal variation
- the rate of TGM depletion is about 0.4 ng m^{-3} yr^{-1} – based on dating by SF_6 during two flights
- stratospheric end point TGM concentrations (i.e. at CO < 30 ppb and O_3 > 380 ppb) decrease with increasing particle number concentration – see Fig. 6
- mercury as an element cannot disappear – depleted gaseous mercury has to reappear as a particulate mercury – this conclusion is supported by qualitative Hg measurements on aerosols by a particle mass spectrometer made by Murphy et al.
- the failure to detect by PIXE mercury on aerosol samples taken during the CARIBIC flights suggests that the more volatile $HgBr_2$ and $HgCl_2$ are more likely to be the products of the mercury oxidation than the less volatile HgO
- zero TGM concentrations have never been observed suggesting an equilibrium between gaseous and particulate mercury

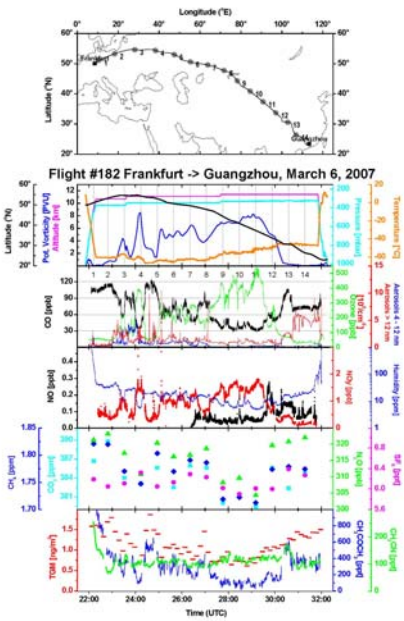

Fig. 5: Overview of the flight #182.

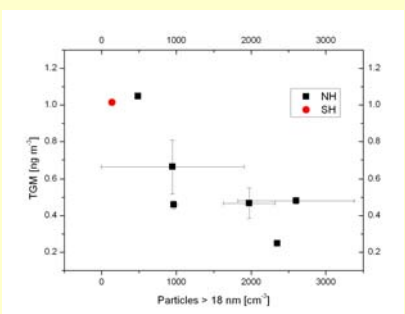

Fig. 6: The relation of stratospheric end point TGM concentrations to particle number concentrations.

Conclusions

TGM is directly or more likely via a semivolatile bivalent Hg compound converted to particulate mercury in the lower stratosphere. The conversion rate is about 0.4 ng m^{-3} yr^{-1}. Models of atmospheric mercury have thus to incorporate gas-to-particle conversion processes. This has not been usually made so far.

Acknowledgements

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