Response to reviewer #2

We thank for the review.

General
I have some questions regarding how the measurements were performed. To me, the methods shed uncertainty on the results. I think the authors are making sweeping hand-waving analysis of these large data sets. To me, the answer(s) lies in very detained analysis of all the data sets. This would present a more convincing argument for their theories. The calculated lifetime just seems unreasonably long.

The techniques used in obtaining our data are described in detail by Slemr et al. (2016) and discussed additionally in our reply to Seth Lyman´s review. As discussed there, the speciation technique using quartz wool was operated within the range of conditions tested by Lyman and Jaffe (2010) and the data should thus be of comparable quality.

As far as the "unreasonably" long stratospheric lifetime concerns: the stratospheric lifetime is according to Volk et al. (1997) and SPARC report (Ko et al., 2013, lifetime definitions are summarized on page 2-20) defined as the atmospheric burden (of the whole atmosphere) divided by the stratospheric sink. For gases without tropospheric sinks, such as N_2O, the stratospheric lifetime is equal to the atmospheric one. For gases with tropospheric sinks, such as mercury, the definition implies a longer stratospheric lifetime than the atmospheric one: the larger the tropospheric sinks are the longer will be the stratospheric lifetime. For methane, e.g., the atmospheric lifetime is ~ 10 yr (Table 6.2, Ko et al., 2013) and the stratospheric one ~160 yr according to Table 3 (Ko et al., 2013). As explained on page 2-7 of the SPARC report, long stratospheric lifetimes are governed by the rate of delivery of air with the species to the loss region (Ko et al., 2013).

Line 54 – if the lifetimes are really 70 some years, why are the mixing ratios so small? It seems to me that TM and GEM must be removed faster than this.

We do not understand this question because it is not clear which of the lifetimes the reviewer means: atmospheric, tropospheric, or stratospheric? Lifetime is defined as the abundance of a species divided by its emission or destruction rate. The mixing ratio of a species in steady state thus depends on its emission rate as well as on its destruction rate.

Line 64 – do we really know that its oxidation is slow? I do not think so.

"Slow" is here meant relatively to the transport times. If the oxidation were much faster mercury would not be as evenly distributed over the globe as it is (Sprovieri et al., 2010). The text has been modified.

Line 92 – Conflicting information? Or, differences in meteorology and atmospheric conditions?

"Conflicting" is perhaps a too strong word – we replaced it with "differing".

Line 120 – "to figure out" I would reword to "in an attempt to unravel and decipher the chemical cycling of GEM in the UT/LS." Experimental

Done.

Line 140 – temperature controlled inlet line. What temperature is maintained? This is critical to maintaining the integrity of the mercury phase partitioning.

Ambient mercury phase partitioning (specifically: GOM vs PBM) at ~-50°C at the tropopause is surely not conserved in CARIBIC sampling because the air warms up to ~30°C before it arrives at the instrument. The integrity issue is discussed in detail by Slemr et al. (2016).

Line 144 – why is the sampling rate 0.5 SLPM? What not 1 SLPM? This would improve your sensitivity, especially with a sampling time of 5 minutes.

To get the sampling flow rate of 0.5 SLPM at an inlet pressure of ~ 250 hPa we had to add an additional pump to aid the internal Tekran one (Neuberger Model KNF UN89 KTDC, Slemr et al., 2016) and even with it the sampling flow rate of 0.5 SLPM could not be kept at higher altitudes. We would very much appreciate an offer of a more efficient pump within the operational constraints, i.e. of about the same size, weight, and comparable DC power consumption.

Line 150 – I personally would not use quartz wool. This non-descriptive material is unpredictable in how in acts under varying atmospheric conditions. It just adds unnecessary uncertainty in your analytical scheme.

As mentioned in our response to Seth Lyman´s review, many methods of separating GOM from GEM have been proposed over the last two decades without having reached consensus on the accurate and reliable one (Gustin et al. 2015). The use of quartz wool may now seem obsolete but was not considered as such in 2014 when we started using it. In addition, its use may still provide useful data under UT/LMS conditions as detailed in our response to Seth Lyman´s review and in an added supporting information.

Line 159 – No in-flight checks on instrument operation are performed? It seems to be that this is not a great strategy and leaves uncertainty in the results.

See response to Seth Lyman´s review.

Line 165 – 167 This is all highly speculative adding more uncertainty.

See response to Seth Lyman´s review.

Line 181 – 183 This too is all highly speculative, adding uncertainty.

See response to Seth Lyman´s review.

Line 227 – 228 I would not say that CH_4 has large emission from biomass burning. CO yes of course. I think this discussion on biomass burning could be shortened as it detracts from the main focus of this paper.

CH_4 emissions from biomass burning are surely not the major CH_4 source but they are substantial enough (Andreae and Merlet, 2001) for using methane as a tracer for biomass burning, in combination with other tracers.

Line 259 – You might want to add how the altitude of the thermal tropopause was determined.

As mentioned in lines 292 -300 we use our ozone measurements in combination with ozone sonde data to determine the thermal tropopause. The method is described in detail by Sprung and Zahn (2010).

Line 330 – This is another reason not to use quartz wool.

See our response before.

Line 363 – 365 These data should be interrogated to try and determine why the difference is greatest in these air masses. This could provide important information on mercury cycling.

The interpretation of this difference is not straightforward. It could be ascribed to large scale subsidence from the stratospheric "overworld". If that were the only reason then the difference in summer with higher proportion of tropospheric air should be smaller which is at odds with the observations. Photochemistry could thus also play a role. We think that the data base is too tenuous to discuss this point in detail.

Lines 387 – 401 All of these investigators are top scientists. These data set need to be interrogated independently to try to decipher differences in what was measured. The instruments are all operated with different sensitivities and LODs. These need to be factored in to the analysis.

Our understanding of the differences is discussed in lines 405 to 455.

Stratospheric Lifetime
Although the method used in this section is valid, I have a gut feeling that it is wrong. Mercury and N_2O and SF_6 are chemically very different. I just can't believe that lifetime is 70 + years.

See paragraph at the beginning of our reply.

References

Andreae, M.O., and Merlet, P.: Emission of trace gases and aerosols from biomass burning, Global Biogeochem. Cycles, 15, 955-966, 2001.

Gustin, M.S., Amos, H.M., Huang, J., Miller, M.B., and Heidecorn, K.: Measuring and modeling mercury in the atmosphere: a critical review, Atmos. Chem. Phys., 15, 5697-5713, 2015.

Ko, M.K.W., Newman, P.A., Reimann, S., and Strahan, S.E., eds.: Lifetimes of stratospheric ozone-depleting substances, their replacements, and related species, SPARC Report No. 6, WCRP-15/2013, December 2013.

Lyman, S.N., and Jaffe, D.A.: Formation and fate of oxidized mercury in the upper troposphere and lower stratosphere, Nature Geosci., 5, 114-117, 2012.

Slemr, F., Weigelt, A., Ebinghaus, R., Kock, H.H., Bödewadt, J., Brenninkmeijer, C.A.M., Rauthe-Schöch, A., Weber, S., Hermann, M., Becker, J., Zahn, A., and Martinsson, B.: Atmospheric mercury measurements onboard the CARIBIC passenger aircraft, Atmos. Meas. Tech., 9, 2291-2302, 2016.

Sprovieri, F., Pirrone, N., Ebinghaus, R., Kock, H., and Dommergue, A: A review of worldwide atmospheric mercury measurements, Atmos. Chem. Phys., 10, 8245-8265, 2010.

Sprung, D., and Zahn, A.: Acetone in the upper troposphere/lowermost stratosphere measured by the CARIBIC passenger aircraft: Distribution, seasonal cycle, and variability, J. Geophys. Res., 115, D16301, doi:10.1029/2009JD012099, 2010.

Volk, C.M., Elkins, J.W., Fahey, D.W., Dutton, G.S., Gilligan, J.M., Loewenstein, M., Podolske, J.R., Chan, K.R., and Gunson, M.R.: Evaluation of source gas lifetimes from stratospheric observations, J. Geophys. Res., 102, D21, 25543-25564, 1997.