

**Worldwide trend of  
atmospheric mercury  
since 1995**

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# Worldwide trend of atmospheric mercury since 1995

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Received: 8 January 2011 – Accepted: 13 January 2011 – Published: 21 January 2011

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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## Abstract

Concern about the adverse effects of mercury on human health and ecosystems has led to tightening emission controls since the mid 1980s. But the resulting mercury emissions reductions in many parts of the world are believed to be offset or even surpassed by the increasing emissions in rapidly industrializing countries. Consequently, concentrations of atmospheric mercury are expected to remain roughly constant. Here we show that the worldwide atmospheric mercury concentration have decreased by about 20 to 38% since 1996 as indicated by long term monitoring at stations in the Southern and Northern Hemispheres combined with intermittent measurements of latitudinal distribution over the Atlantic Ocean. The total reduction of the atmospheric mercury burden of this magnitude within 14 yrs is unprecedented among most of atmospheric trace gases and is at odds with the current mercury emission inventories with nearly constant emissions over the period. It suggests a major shift in the biogeochemical cycle of mercury including oceans and soil reservoirs. Decreasing reemissions from the legacy of historical mercury emissions are the most likely explanation for this decline since the hypothesis of an accelerated oxidation rate of elemental mercury in the atmosphere is not supported by the observed trends of other trace gases. Consequently, models of the atmospheric mercury cycle have to include soil and ocean mercury pools and their dynamics to be able to make projections of future trends.

## 1 Introduction

Gaseous elemental mercury has an atmospheric lifetime of about 1 yr and is thus transported within a hemisphere and even globally (Lindberg et al., 2007). Consequently, mercury can be deposited after its oxidation to less volatile and more soluble compounds in areas far away from where it was originally emitted. Part of the deposited mercury is then transformed to highly toxic methylmercury which bioaccumulates in the aquatic food chain and, at its end, reaches concentrations which are adverse to human

ACPD

11, 2355–2375, 2011

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health and ecosystems (Mergler et al., 2007; Scheuhammer et al., 2007). Because of these concerns, mercury emissions of developed countries have been substantially reduced by control measures introduced in the late 1980s (Pirrone et al., 2009). However, as the emissions due to the rapid industrialization in many parts of the world offset the reductions (Streets et al., 2009; Pacyna et al., 2010), a constant or even slightly increasing mercury concentration in the air is expected.

Mercury concentrations in air and wet deposition have been monitored since 1995 at an increasing number of sites in the Northern Hemisphere (Temme et al., 2007; Prestbo and Gay, 2009; Cole and Steffen, 2010) but only at one site in the Southern Hemisphere (Slemr et al., 2008). At a majority of sites with records long enough to establish significant trends, the mercury concentrations in air and mercury wet deposition decreased with time. Recently, Ebinghaus et al. (2011) analysed the baseline mercury concentrations of mercury measured since 1996 at Mace Head, Ireland, and they found a downward trend of  $0.028 \pm 0.010 \text{ ng m}^{-3} \text{ yr}^{-1}$ , representing a decrease of 1.6–2.0% per year. A somewhat smaller downward trend of  $0.015 \pm 0.003 \text{ ng m}^{-3} \text{ yr}^{-1}$  was observed at Cape Point, South Africa, between 1996 and 2004 without any data selection (Slemr et al., 2008), representing a decrease of 1.0–1.5% per year. Assuming these trends to be representative for the pertinent hemispheres, which is not unreasonable given the atmospheric mercury lifetime of about 1 yr (Lindberg et al., 2007) and an intrahemispheric mixing time of about 3 months (Warneck, 1988), trends of this magnitude would accumulate to a decrease of 15–30% of atmospheric mercury burden over a period of 15 yrs. Taking into account that anthropogenic emissions represent only about one third of all mercury emissions (Selin et al., 2007), such decrease would be intriguing and clearly at odds with the current emission inventories.

Here we present new data obtained at Cape Point since March 2007 and onboard ships over the Atlantic Ocean in 2008 and 2009. By a combination of these recent data with the older data we will show that mercury is quite evenly distributed in the Southern Hemisphere and thus the trend observed at Cape Point can be considered to be representative for the Southern Hemisphere. In the Northern Hemisphere, the trend

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displayed by baseline mercury measurements at Mace Head is substantially smaller than the one suggested by measurements onboard ships, but it is still large and negative. The overall large negative trend of atmospheric mercury burden is at last discussed in terms of changes in anthropogenic inventories, oxidation capacity, and of a legacy of past mercury emissions.

## 2 Experimental

### 2.1 Techniques, monitoring sites and cruises

The Cape Point station (34°21' S, 18°29' E) in South Africa is operated as one of the World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) stations. The station is located at the southern tip of the Cape Peninsula on top of a peak 230 m a.s.l., ca. 60 km south of Cape Town. Gaseous mercury was measured there since September 1995 until December 2004 by a manual double amalgamation technique (Slemr et al., 2008). Apart from 1995, 37–173 samples, each taken over a period of 3 h, were taken annually. Since March 2007 gaseous mercury has been continuously monitored by a Tekran analyser (Model 2537A, Tekran Inc., Toronto, Canada), which represents essentially an automated single amalgamation version of the aforementioned technique.

Mace Head, in County Galway on the west coast of Ireland, at 53°20' N, 9°54' W is also operated as one of the WMO GAW stations. Gaseous mercury has been continuously measured since September 1995 using a Tekran instrument (Ebinghaus et al., 2002, 2011).

At Neumayer station in Antarctica, a Tekran analyzer has been operated from 23 December 1999, to 5 February 2001. The station is located at 70°39' S, 80°15' W, on the Ekströmisen, 8 km from Atka Bay (Temme et al., 2003a).

Since 1979 gaseous mercury has been measured during 10 ship cruises during the crossings of Atlantic Ocean from Germany to different ports in Argentina (Buenos

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Aires, Mar del Plata, Punta Quilla) or Chile (Punta Arenas). Only the R/S *Meteor* in January/February 1979 and R/S *Polarstern* cruises in December/February 1999/2000 and in November 2008 went to the Gulf of Guinea and to Cape Town, respectively. During 6 cruises until 1994, the manual technique used onboard ships was the same as that at Cape Point (Slemr et al., 2008). Usually about 100 samples (each over a period of 2–3 h) were taken in each hemisphere (Slemr et al., 2003). The cruises since 1996 were carried out using the automated Tekran analyzer.

The precision of individual measurements made by the Tekran analyzer was  $\pm 0.05$  and  $\pm 0.1 \text{ ng m}^{-3}$  with 30 min (cruise in 1996, Cape Point, Mace Head) and 15 min (cruise in 1999/2000) sampling times, respectively. The cruises in 2008 and 2009 were dedicated to measurements of air-sea fluxes, and the Tekran analyzer was run with a 5 min sampling time and a correspondingly worse precision of about  $0.15 \text{ ng m}^{-3}$  of the individual measurement. Averaged over 15 or 30 min the precisions given above are reached. The Tekran instruments were usually automatically calibrated every 25 h using the internal permeation source. The permeation rate of the source was in turn calibrated by injections of known mercury amount from a vapour saturation device. This calibration was made before and after a cruise and approximately every 3 months at the stations (Ebinghaus et al., 2011).

The precision of individual manual measurements was  $\pm 5.8\%$  at Cape Point, and for ship cruises. Each manual measurement is calibrated using the injection of known mercury amount from a vapour saturation device (Slemr et al., 1985). Thus the precision given above includes the precision of the individual calibrations. All concentrations in this paper are given in  $\text{ng m}^{-3}$  (STP) with the standard temperature of 273.16 K and pressure of 1013 mbar.

Gaseous atmospheric mercury consists predominantly of almost insoluble and volatile gaseous elemental mercury (GEM). Less volatile and more soluble gaseous mercury compounds, termed reactive gaseous mercury (RGM) and defined by the analytical procedures for their detection, usually represent only a few percent of the total gaseous mercury concentrations at ground stations (Ebinghaus et al., 2009). Although

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RGM was found to be transmitted by the inlet tubing under conditions of extremely low humidity and low temperature encountered at the Neumayer site in Antarctica (Temme et al., 2003a), this is considered to be an exception. Under humid conditions with inlet tubing and PTFE filter loaded with sea salt (the Tekran instrument is usually run with a 0.2 or 0.45  $\mu\text{m}$  PTFE filter to protect the gold collectors and the inner plumbing of the instrument from contamination by aerosols), RGM will not be transmitted to the gold collectors. Thus the measurements at Cape Point, Mace Head and onboard ships are believed to represent only GEM whereas those at Neumayer represent total gaseous mercury (TGM). The difference between TGM and GEM in marine atmospheres is usually only a few percent and thus small in comparison with the trends discussed later (Ebinghaus et al., 2009). The uncertainty in the attribution of the measured species will thus not influence substantially the results of this paper.

## 2.2 Comparability of the data

Ideally we would have continued the manual measurements some time after the installation of the Tekran instrument at Cape Point to validate the continuity of the data. For reasons beyond our control this was not possible and thus we have to assess the comparability of the data until 2004 and since 2007 in a different way. In the context of this paper, the comparability of the data has two aspects: analytical comparability and comparability of data with different temporal coverage.

The manual double amalgamation technique used at Cape Point and cruises until 1994 was found to provide results undistinguishable from those obtained by Tekran instrument during an international field intercomparison at Mace Head carried out in 1995 (Ebinghaus et al., 1999). In addition, the manual measurements at Cape Point were found to be comparable with those made with Tekran onboard R/V *Polarstern* in December 1999 and January 2000 when the ship was near Cape Point (Temme et al., 2003b). This demonstrates that both techniques provide the comparable results. A generally good agreement of different techniques for measurement of GEM and/or TGM was found in other intercomparisons (Schroeder et al., 1995; Munthe et al., 2001)

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Automated measurements by Tekran instrument provide an almost complete data coverage; time lost by calibration and maintenance of the instrument represents usually only a few percent loss of a given measurement period. Manual collection of some 150 samples at Cape Point per year covers only 450 h, i.e. only about 4.5% of the year. The coverage disparity is further aggravated by selection of baseline data: a data filter using concentrations of  $^{222}\text{Rn} < 100 \text{ mBq m}^{-3}$  yielded only 18% of baseline data (Brunke et al., 2004) which would leave only some 30 manual measurements a year. Table 1 presents the average concentrations and medians of filtered and unfiltered Tekran data at Cape Point in the years 2007–2009. The average of unfiltered data was higher than that of filtered ones by 0.055, 0.012 and  $0.005 \text{ ng m}^{-3}$  in 2007, 2008, and 2009, respectively. The difference is statistically significant in 2007 and 2008 but even then it is small in comparison with the trends discussed later. Consequently, the unfiltered Cape Point data can be used for further evaluation. Table 1 also shows a generally small difference between medians and averages suggesting that the data at Cape Point are nearly normally distributed. Altogether, we conclude that randomly distributed manual sampling over the whole year provides a good approximation (within a few  $0.01 \text{ ng m}^{-3}$ ) of the annual means and medians measured by a continuous instrument.

### 3 Results and discussion

Figure 1 summarize the data from the Southern and Northern Hemisphere (NH and SH), respectively. The data from the GAW stations Cape Point (CPT), South Africa, and Mace Head (MH), Ireland, are presented as annual medians to suppress the influence of occasional high mercury concentrations due to episodes of local or regional pollution such as by biomass burning at CPT (Brunke et al., 2001). The annual medians for MH were calculated from baseline data only (Ebinghaus et al., 2011).

In the SH the land-based measurements at CPT and Neumayer agree well with measurements made during ship cruises over the southern Atlantic Ocean. The

homogeneity of the mercury concentrations in the SH is not surprising given that point anthropogenic emissions constitute only a small fraction of all emissions in the SH (Slemr et al., 2008). The SH sources are dominated by emission from oceans, the import from the NH and for a few months by emission from biomass burning of which only the last one is confined to certain but still large areas (Slemr et al., 2008).

Altogether, the concentration of gaseous mercury in the SH seems to have decreased by from about  $1.35 \text{ ng m}^{-3}$  around 1996 to about  $0.9 \text{ ng m}^{-3}$  around 2008. The trend calculated from the least-square fit of the Cape Point data only is  $-0.034 \pm 0.005 \text{ ng m}^{-3} \text{ yr}^{-1}$  ( $r^2 = 0.8677$ ,  $n = 10$ ). The years 1995, 1997, and 1997 were not considered in this calculation because of their uneven seasonal coverage (Slemr et al., 2008). The trend calculated from Cape Point and ship data is with  $-0.035 \pm 0.006 \text{ ng m}^{-3} \text{ yr}^{-1}$  ( $r^2 = 0.7439$ ,  $n = 15$ ) comparable. Both trends are substantially larger than  $-0.015 \pm 0.003 \text{ ng m}^{-3} \text{ yr}^{-1}$  calculated from the manual measurements at Cape Point until 2004 (Slemr et al., 2008). As discussed before, this difference can be explained neither by the different measurement techniques nor by the different data coverage. Figure 1 suggests that the downward trend accelerated in the second half of the measurement period in both hemispheres. Starting from the concentration of about  $1.35 \text{ ng m}^{-3}$  around 1996 the trend of  $-0.035 \text{ ng m}^{-3} \text{ yr}^{-1}$  translates to a decrease of  $0.46 \text{ ng m}^{-3}$  in 14 years until 2009, i.e. a decrease of 34% since 1996. No other long-term observations of gaseous mercury concentrations in the SH have been reported for this time interval so far.

The baseline gaseous mercury concentration at MH decreased from about  $1.75 \text{ ng m}^{-3}$  in 1996–1999 to about  $1.4 \text{ ng m}^{-3}$  in 2009. As shown by composite back attribution plots for MH in Fig. 1 of Ebinghaus et al. (2011), the baseline MH data are representative for a large area of Northern Atlantic including Greenland, Canada, and parts of the Arctic Ocean. The data from ship cruises, measured predominantly south of  $50^\circ \text{ N}$ , suggest an even larger decrease within this period. Please note that gaseous mercury concentrations of about  $1.1 \text{ ng m}^{-3}$  measured over the northern Atlantic Ocean in 2008 and 2009 are smaller than the annual median

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concentrations observed at CPT in mid 1990s. Figure 1 of this paper also suggests a larger downward trend after about 2002, in agreement with the observations in the SH. The least-square fit of the baseline annual medians at Mace Head provides a trend of  $-0.024 \pm 0.005 \text{ ng m}^{-3} \text{ yr}^{-1}$  ( $r^2 = 0.6398$ ,  $n = 14$ ) for the 1996–2009 period which is comparable to  $-0.028 \pm 0.010 \text{ ng m}^{-3} \text{ yr}^{-1}$  calculated from the baseline monthly averages (Ebinghaus et al., 2011). Inclusion of the median concentrations from the ship measurements in 1994–2009 leads to a somewhat larger trend of  $-0.035 \pm 0.007 \text{ ng m}^{-3} \text{ yr}^{-1}$  ( $r^2 = 0.5915$ ,  $n = 19$ ). The least-square fit of the 1994–2009 ship data only is with  $-0.051 \pm 0.017 \text{ ng m}^{-3} \text{ yr}^{-1}$  ( $r^2 = 0.7576$ ,  $n = 5$ ) not significant.

The different trends calculated from the MH data only and from data including the ship measurements point to an inhomogeneous distribution of mercury within the NH. This is most likely due to emissions which are dominated by anthropogenic sources mostly located within the latitude band of 40–60° N and shifting from Europe and North America to South and East Asia (Pacyna et al., 2010). Despite the inhomogeneous distribution of the sources, Slemr et al. (2003) showed that measurements at different remote stations in the NH provide comparable results. The rather narrow spread of the annual medians and similar changes with time shown by Slemr et al. (2003) are the result of the short air mixing intrahemispherical time of about 3 months (Warneck) relatively to the tropospheric lifetime of about 1 yr of elemental mercury. Almost all of the long-term gaseous mercury (CAMNet) and mercury wet deposition (MDN) monitoring sites are also located within the latitude band of 40–60° N (Temme et al., 2007; Cole and Steffen, 2010; Prestbo and Gay, 2009). The average downward trend of gaseous mercury concentrations observed on 6 CAMNet sites with a long enough monitoring period (1996–2005) was  $0.57 \pm 1.09\% \text{ yr}^{-1}$  with large differences between the individual locations (Temme et al., 2007). The data from Alert did not provide a significant trend for the period 1995–2005 (Temme et al., 2007), but their reanalysis after including the data from 2006 and 2007 also show a decrease by 0.6% per year ( $0.0086 \pm 0.0014 \text{ ng m}^{-3} \text{ yr}^{-1}$ ) over a period of 13 yrs (Cole and Steffen, 2010). This is comparable to the decrease observed at MH. Despite large differences between the

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23 MDN sites with significant trends, mercury wet deposition decreased on average by  $1.74 \pm 0.74\% \text{ yr}^{-1}$  between 1996 and 2005. Neglecting the possible trends in the distribution of precipitation, this decrease is about three times as large as observed at CAMNet sites and compares well with the trend suggested by the ship measurements over the northern Atlantic Ocean.

Figure 2 shows the NH/SH gaseous mercury concentration ratios over the Atlantic Ocean measured during 10 cruises between 1977 and 2009, and for comparison the ratios of the concentrations at MH to those at CPT (MH baseline annual medians/CPT annual medians). The ratios measured during ship cruises remained with  $1.49 \pm 0.10$  ( $n = 10$ ) rather constant over the last 30 yrs. The MH/CPT ratios since 1996 are with  $1.47 \pm 0.13$  ( $n = 12$ ) comparable. Please note that the seasonal variations of gaseous mercury concentrations in both hemispheres are roughly synchronous (Slemr et al., 2008) making the ratio measured intermittently onboard ships rather insensitive to season. The nearly constant NH/SH concentration ratio of 1.48 over such a long period is important since it shows that the distribution of the sources between the two hemispheres has not changed substantially over the last 30 yrs.

The NH/SH mercury concentration ratio can remain nearly constant only if the relative trends in both hemispheres are nearly equal. We consider the trend in the SH to be better defined because of the more homogeneous distribution of mercury concentrations. The absolute trend of  $-0.035 \pm 0.006 \text{ ng m}^{-3} \text{ yr}^{-1}$  in the SH and the initial concentration of about  $1.3 \text{ ng m}^{-3}$  at Cape Point in 1996 yield a relative negative trend of 2.7% per yr. The initial concentration at MH in 1991–1999 is about  $1.75 \text{ ng m}^{-3}$ . The relative negative trend of 2.7% per yr would then yield an absolute trend in the NH of  $-0.047 \text{ ng m}^{-3} \text{ yr}^{-1}$  which is comparable to the trend suggested by ship measurements only but is substantially larger than the significant trends both from MH data only and from combined MH and ship data. Based on the initial concentration of  $1.75 \text{ ng m}^{-3}$ , the relative negative trends calculated from the MH data only and MH and ship data correspond to 1.4 and 2.0% per yr, respectively.

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In summary, there is an overwhelming evidence of a downward trend of mercury concentrations in the air of both hemispheres over the last 14 yrs. Using the CPT, MH and ship data we conclude that the atmospheric mercury burden decreased between 1996 and 2009 with a relative rate somewhere between 1.4 and 2.7% per yr in NH and SH, respectively, i.e. by 20 to 38% since 1996. Such decrease is unprecedentedly large for almost any trace gas and it suggests a substantial shift in the global biogeochemical cycle of mercury. Below we discuss three possible explanations: (a) the decrease of the anthropogenic emissions, (b) the acceleration of the oxidation of mercury in the atmosphere, and (c) the decrease of emissions from the legacy of historical mercury use and emissions.

The steady-state budgets of atmospheric mercury assume that the primary anthropogenic sources with biomass burning represent only about one third of the total mercury emissions (Selin et al., 2007). The inventories of anthropogenic mercury emissions yield a total anthropogenic emission of about  $2000 \text{ t yr}^{-1}$  (uncertainty is assumed to be less than  $\pm 30\%$  (Pacyna et al., 2010)) which is believed to have changed only by about  $\pm 15\%$  between 1995 and 2006 (from  $2128 \text{ t yr}^{-1}$  in 1996 to  $2480 \text{ t yr}^{-1}$  in 2006 (Streets et al., 2009) and from  $2317 \text{ t yr}^{-1}$  in 1995 to  $1930 \text{ t yr}^{-1}$  in 2005 (Pacyna et al., 2010)). It is obvious that rather small changes in anthropogenic emissions representing only about one third of total emissions cannot explain the observed decrease of the atmospheric mercury burden. The discrepancy would remain substantial even if the anthropogenic emissions were the only mercury emissions, but such assumption would not be consistent with the constant NH/SH concentration ratio and the simultaneous 30% decrease of the atmospheric mercury burden with emissions located and reduced mostly in the NH.

Despite more than 20 yrs of research on the atmospheric mercury cycle and its crucial importance to mercury cycling in the atmosphere, the oxidation mechanism of gaseous elemental mercury to compounds amenable to removal by dry and wet deposition remains elusive (Lin et al., 2006; Lindberg et al., 2007). Three major pathways are proposed and their importance disputed: reaction of  $\text{Hg}^0$  with OH,  $\text{O}_3$  and Br/BrO

(Lin et al., 2006; Holmes et al., 2010). OH is the major oxidant for the majority of atmospheric trace gases and its global increase on the order of 30% within 14 yrs would induce large decreasing trends in the concentrations of many long lived substances, such as CH<sub>4</sub>, which has not been observed (Clerbaux and Cunnold, 2007). In fact, a recent study shows that OH concentrations remained nearly constant within the period discussed here (Montzka et al., 2011). Ozone concentrations have increased in some regions of the troposphere and decreased in others (Oltmans et al., 2006). Despite the regional trend differences and some inconsistencies in the data (Clerbaux and Cunnold, 2007), an overall increase of tropospheric ozone concentrations on the order of 30% since 1995 is not supported by the observations. The tropospheric organic bromine reservoir consists to a larger part of long-lived compounds of purely (halones) or partly (CH<sub>3</sub>Br) anthropogenic origin and to a smaller part of short-lived compounds mostly of natural origin. The measurements of the latter ones suggest that their emission remains constant (Clerbaux and Cunnold, 2007). Controls imposed by the Montreal Protocol on emissions of halones and anthropogenic emissions of CH<sub>3</sub>Br has led to a peak of stratospheric BrO concentrations at around 2000 and a slight decrease since (Clerbaux and Cunnold, 2007). In summary, the hypothesis of an accelerating oxidation of elemental mercury in the atmosphere by the amount suggested here is not supported by observations.

The atmospheric mercury burden and the mercury deposition are believed to have increased by a factor of  $3 \pm 1$  since pre-industrial times (Mason and Sheu, 2002; Lindberg et al., 2007). The human activity has also substantially increased the soil and ocean mercury reservoirs and the fluxes between them and the atmosphere (Mason and Sheu, 2002; Lamborg et al., 2002; Fitzgerald and Lamborg, 2003; Strode et al., 2007; Smith-Downey et al., 2010). The atmospheric residence time of about 1 yr is somewhat shorter than the mercury residence time of 1.2–4 yr in ocean mixed layer (depending on the assumed mixed layer depth) (Lamborg et al., 2002; Strode et al., 2007) and both are much shorter than the residence time of 80–100 yr in soils (Fitzgerald and Lamborg, 2003; Smith-Downey et al., 2010). Consequently, the temporal response of

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atmospheric and surface ocean mercury reservoirs to changing mercury release into the environment will be controlled by the slowly responding soil reservoir. With nearly constant anthropogenic emissions in the last 15 yrs (Streets et al., 2009; Pacyna et al., 2010), the atmospheric mercury burden will thus follow the changing emissions from the soil which partly contain mercury from a legacy of historical anthropogenic mercury pollution (Fitzgerald and Lamborg, 2003; Smith-Downey et al., 2010). As the historical anthropogenic mercury release into environment is poorly documented, one can only speculate about the past decrease of emissions leading to the observed trend. The phasing out of mercury-containing products and the increasing control of manufacturing processes with mercury involvement started in the late 1980s (Hylander and Meili, 2003) but mercury emissions from coal burning, one of the largest anthropogenic sources, might have started to decrease already in the late 1970s and early 1980s as a corollary of regulations limiting the emissions of other pollutants from coal fired power plants (Pirrone et al., 2009). The sediment, peat, and ice core studies suggest that the mercury deposition peaked between 1950 and 1990 at a majority of sites (Schuster et al., 2002; Biester et al., 2007; Muir et al., 2009; Mast et al., 2010), although it is still increasing at some locations (Fitzgerald et al., 2005). The firm study by Fain et al. (2009) suggests that the mercury concentration in air peaked at around 1970. The decreasing emissions from the legacy of historical mercury use and emissions thus appear to be the most plausible explanation for the observed trend.

## 4 Conclusions

The measurements presented here provide strong evidence that the global atmospheric mercury burden has decreased by about 30% since 1995. Consequently, the conclusion reached by the Panel on Source Attribution of Atmospheric Mercury (Lindberg et al., 2007) that “there has been no discernible net change in the size of the atmospheric pool of Hg in the Northern Hemisphere since the mid-1970s” has to be updated.

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We further conclude that the observed decrease in the atmospheric mercury burden is most likely due to the decreasing emissions from soil reservoir whose mercury burden originates partly from a legacy of historical anthropogenic release of mercury into the environment. As the response of the atmospheric mercury burden to changing emissions will essentially depend on mercury residence time in soil reservoir, we consider soil to be an essential part of any model trying to predict the response of the atmospheric burden to changing anthropogenic emissions in future. And last but not least: more extensive mercury monitoring especially south of the 40–60° N is needed to confirm the reported trend of the atmospheric mercury burden and to follow its future development.

*Acknowledgements.* Our thanks go to a multitude of people who helped to carry out the ship measurements at the R/S *Walther Herwig* and the R/S *Polarstern* and enabled to sustain the long-term TGM measurements at Cape Point and Mace Head. Alistair Manning and Richard Derwent provided the classification of baseline air masses arriving at the Mace Head station. We also thank Chris Holmes for valuable comments on the draft. This work was partly supported by the Deutsche Forschungsgemeinschaft and the Irish Environmental Protection Agency.

The service charges for this open access publication have been covered by the Max Planck Society.

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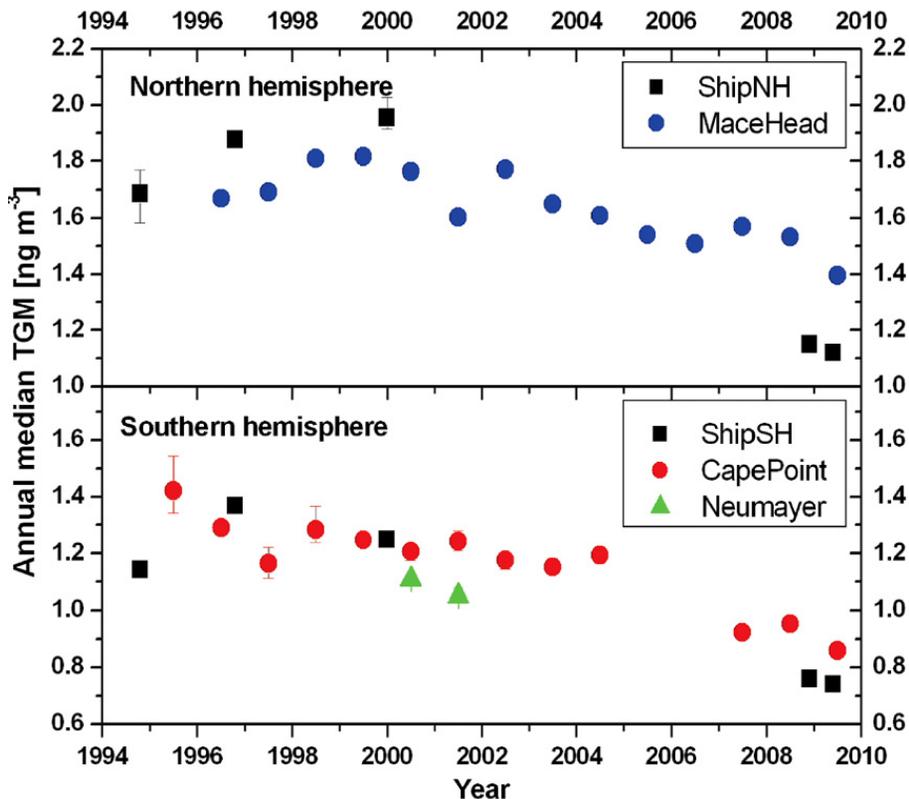
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**Table 1.** Average mercury concentrations and medians of all data measured by the continuous technique and data measured at concentrations of  $^{222}\text{Rn} < 100 \text{ mBq m}^{-3}$ . The standard deviations and number of measurements are also given.

Year	GEM concentrations at $^{222}\text{Rn} < 100 \text{ mBq m}^{-3}$ [ $\text{ng m}^{-3}$ ]		GEM concentrations, all data	
	Average	Median	Average	Median
2007	$0.873 \pm 0.090$ (90)	0.879	$0.928 \pm 0.181$ (13036)	0.935
2008	$0.921 \pm 0.106$ (858)	0.948	$0.933 \pm 0.132$ (14418)	0.953
2009	$0.867 \pm 0.092$ (1579)	0.861	$0.872 \pm 0.139$ (12958)	0.870

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**Fig. 1.** Trends of GEM concentrations in the Northern and Southern Hemispheres. The points and bars represent the annual medians and the 95 % confidence intervals of the medians, respectively. The annual medians of automated TGM measurements at Cape Point were calculated using daily mean concentrations. The annual medians at Mace Head were calculated from baseline hourly mean concentrations (Ebinghaus et al., 2010). The median confidence intervals for the continuous measurements are smaller than the symbols.

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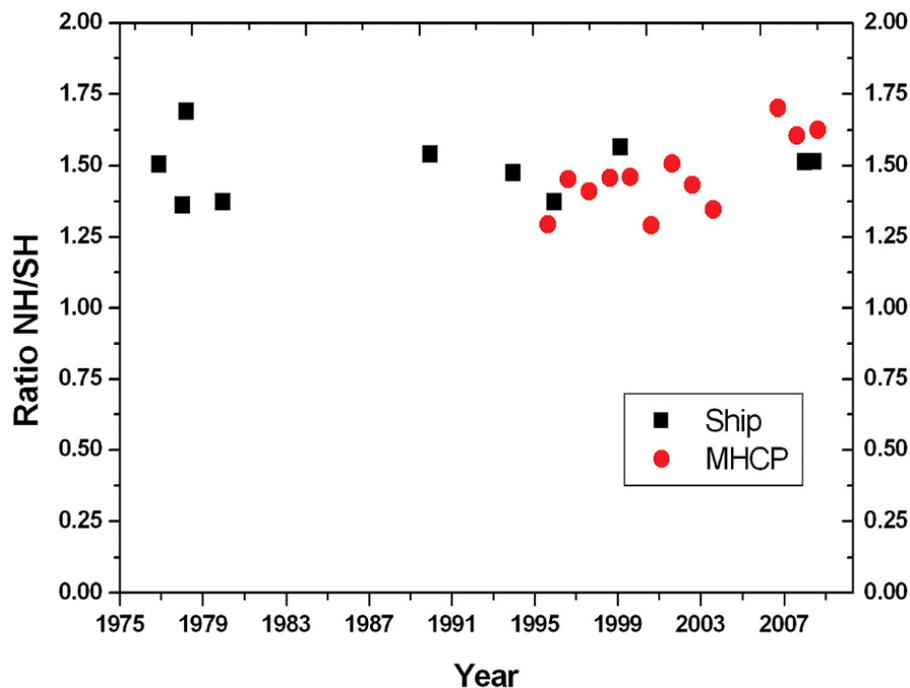
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**Fig. 2.** Ratios of northern to southern hemispheric median concentrations measured over the Atlantic Ocean since 1977 and ratios of annual median concentrations at Mace Head to those at Cape Point.

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