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Worldwide atmospheric mercury measurements: a review and synthesis of spatial and temporal trends

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Abstract

A large number of activities have been carried out during the last decade in different regions of the world, including polar regions, aiming to assess the level of mercury (Hg) species in ambient air and in precipitation observing their variation over time and with changing meteorological conditions. Following the discovery of atmospheric Hg depletion events (AMDEs) in Polar Regions several studies have indeed been conducted in order to assess the chemical-physical mechanisms related to AMDEs occurred in polar atmospheres with special attention to the consequences of these phenomena in terms of contamination of polar environment due to the rapid conversion of atmospheric gaseous Hg (Hg^0) into reactive and water-soluble forms that may potentially become bioavailable. The understanding of the way in which mercury released to the atmosphere is eventually incorporated into biota is of crucial importance not only for the polar regions but also for the marine environment in general. The world's oceans and seas are in fact both sources and sinks of Hg and although it appears that the atmosphere is the major transport/distribution medium for Hg, because most Hg emissions are to the atmosphere, oceans and seas also play an important role. Currently, however, a coordinated observational network for Hg does not exist. There are a number of state and national programs that are collecting atmospheric Hg data but the parameters monitored, the locations of the monitoring sites and the methods employed may prohibit their utility in assessing Hg long-trend variations. The large increase in mercury emissions in fast developing countries (i.e., China, India) over the last decade due primarily to a sharp increase in energy production from the combustion of coal are not currently reflected in the long-term measurements of total gaseous mercury in ambient air and in precipitation data at several continuous monitoring sites in North Europe and North America. The discrepancy between observed gaseous mercury concentrations (steady or decreasing) and global mercury emission inventories (increasing) is not yet clear however, could be at least in part accounted by the increasing in the potential oxidation of the atmosphere recently documented. Therefore, measurements

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of other key atmospheric constituents at the global monitoring sites are necessary for us to develop a better understanding of the global redistribution of Hg and to further refine model parameterizations of the key processes. The sharing of data from this network, allowing, in fact, access to comparable and long-term data from a wide array of locations for understanding temporal and spatial patterns of Hg transport, deposition and re-emission process producing thus data that will support the validation of regional and global atmospheric Hg models. This paper presents a detailed overview of atmospheric mercury measurements conducted in the Northern and Southern Hemispheres at several terrestrial sites (industrial, rural and remote) during the last decade as well as measurements performed over the world's ocean and seas and in Polar Regions with reference to the monitoring techniques and location of monitoring sites in most of the continents.

1 Introduction

Mercury is emitted into the atmosphere from a variety of anthropogenic (i.e., power generation facilities, smelters, cement production, waste incineration and many others) (Pirrone et al., 1996, 1998, 2001) and natural sources (i.e., volcanoes, crustal degassing, oceans) in different chemical and physical forms (Pacyna et al., 2001; Carpi, 1997). In the troposphere the most important species are gaseous elemental mercury (Hg^0), divalent reactive gaseous mercury, Hg^{II}, which consists of various oxidised compounds, and particle-bound Hg, Hg-p, which consists of various Hg compounds. It should be noted that information on the speciation/fractionation of these different chemical and physical forms is largely operationally defined. Conversions between these different forms provide the basis of Hg's complex distribution pattern on local, regional and global scale. Hg cycling between different environmental compartments depends on the rate of different chemical and physical mechanisms (i.e., dry deposition, wet scavenging) and meteorological conditions as well as on the anthropogenic variables and forcing which affect its fate in the global environment. Experimental field data and

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model estimates indicate that anthropogenic mercury emissions are at least as great as those from natural sources, contributing together to the global atmospheric pool. It was in fact observed from analysis of lake sediments, ice cores and peat deposits from both hemispheres, a threefold increase of mercury deposition since pre-industrial times (Engstrom and Swain, 1997; Bindler et al., 2001; Biester et al., 2002; Lamborg et al., 2002; Lindberg et al., 2007 and references therein). Recent studies have highlighted that in fast developing countries (i.e., China, India) mercury emissions are rapidly increasing in a dramatic fashion due primarily to a sharp increase in energy production from the combustion of coal. Recent emission estimates highlighted that the Asian emissions are considered to be of global importance. However, potentially increased Asian emissions are neither reflected both in the long-term measurement of Total Gaseous Mercury (TGM) and precipitation data in Europe and North America. The reason for this is not yet clear however, it was hypothesized that atmospheric mercury cycling is possibly going on a faster rate than previously thought. Currently it is extremely difficult to derive the long-term global mercury increasing due to the lack of complete spatio-temporal data sets. Regional differences, temporal trends and potential sources and source regions can be identified by monitoring, especially when carried out in networks. Although atmospheric Hg monitoring stations have increased in the past decade, the database is sparse, especially in remote locations and above all in the Southern Hemisphere. General scientific consensus exists about the current global background concentration of airborne mercury which is taken as ca. 1.5 to 1.7 $\mathrm{ng\,m^{-3}}$ in the Northern Hemisphere and ca. 1.1 to 1.3 $\mathrm{ng\,m^{-3}}$ in the Southern Hemisphere (Lindberg et al., 2007). The atmosphere provides the main environmental pathway for redistribution of Hg around the globe, and therefore, quantifying the transfer of Hg from the air to the earth's surface via wet and dry deposition is critically important. Like atmospheric Hg, there is currently not a globally coordinated network of atmospheric Hg deposition sites. There are a few coordinated networks in certain regions of the world including: North American Mercury Deposition Network (MDN) that was initiated in the early 1990s as part of the National Atmospheric Deposition

Program (NADP); EMEP in Europe; and networks in Japan as well as other parts of Asia. Initial efforts are underway to develop such monitoring capabilities for Hg in the Northern Hemisphere. Long-term monitoring of Hg in the atmosphere would indeed provide valuable information about the impact of emission controls on the global budget of atmospheric Hg, and their observance (Fitzgerald, 1995). In addition, systematic Hg assessment could also provide an insight into the global Hg cycle, especially into the ratio of anthropogenic and natural emissions which is currently poorly defined (Ebinghaus et al., 1999). The need for such measurements was recognized in the mid-1990s (Fitzgerald, 1995) and several monitoring stations have been brought into operation since (i.e., Schroeder et al., 1998; Slemr and Scheel, 1998; Ebinghaus et al., 2002a). A complementary approach to measurements at a few stationary sites for long periods are campaign measurements from moving platforms such as ships or aircraft and no-permanent sampling sites distributed on global scale. Slemr et al. (2003) attempted to reconstruct the worldwide trend of atmospheric Hg (TGM) concentrations from long-term measurements of known documented quality at 6 sites in the Northern Hemisphere, 2 sites in the Southern Hemisphere, and multiple ship cruises over the Atlantic Ocean made since 1977. The authors throughout this reconstruction suggested that the TGM concentrations in the global atmosphere had been increasing since the first measurements in 1977 to a maximum in the late 1980s, after which Hg concentrations decreased to a minimum in 1996 and then remained constant at a level of about 1.7 ng m^{-3} in the Northern Hemisphere. In contrast, Lindberg et al. (2007) have pointed out a number of reasons to support the null hypothesis (i.e. there has been little change since 1977). Another complementary approach is to use Hg^0 in the interstitial air of firn (perennial snowpack) in the Greenland icecap to retrieve an history of atmospheric Hg^0 at middle and high northern latitudes. From this record, it has been shown that anthropogenic emissions caused a two-fold rise in boreal atmospheric GEM concentrations before the1970s, which likely contributed to higher deposition of mercury in both industrialized and remotes areas (Faïn et al., 2008, 2009).

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TGM results for the Southern Hemisphere do not suggest that there has been much change in TGM levels in the global remote atmosphere over the past 15–20 years (De Mora et al., 1993; Sprovieri and Pirrone, 2000; Sprovieri et al., 2002; Temme et al., 2003). In the USA, there is currently an effort underway to expand the MDN network to include measurements of Hg speciation in air, and for estimating dry deposition (http://nadp.sws.uiuc.edu/mdn/). This paper describes a detailed overview of atmospheric measurements performed in the Northern and Southern Hemisphere at several stationary sites (industrial, rural and remote) and complementary measurements performed within campaign measurements from moving platforms such as ships or aircraft covering large areas of the globe. Occasional shipboard measurements should thus be a part of the global monitoring network for atmospheric Hg. TGM measurements on board ships proved to provide valuable complementary information to measurements from the ground based monitoring network. This information consists of a snapshot of large-scale geographical distribution. With proper quality control to ensure comparability and a relatively low measurement uncertainty, the combination of intermittent shipboard and long-term ground measurements can provide information about the worldwide distribution and trend of atmospheric Hg. Ebinghaus et al. (1999) have shown, that good agreement of the atmospheric mercury concentrations determined by different laboratories using different techniques makes a combination of data sets from different regions of the world feasible.

2 Results and discussion

2.1 Monitoring network needs and mercury measurements on a global scale

Hg concentration measurements in ambient air of documented and accepted quality are available since the mid 1970 and concentration data are available for both hemispheres. Long-term monitoring of atmospheric mercury with high time resolution has been started at Alert, Canada (January 1995) and Mace Head, Ireland

(September 1995), followed by numerous other sites since then. In 1995, Fitzgerald's initiative for the installation of a global AMNET (Fitzgerald et al., 1995) has partly been accomplished on a regional scale within the Canadian Atmospheric Mercury Network (CAMNet) that was established in 1996 to provide accurate, long-term measurements of TGM concentration and the Hg deposition in precipitation (wet deposition) across Canada. Programs such as the World Meteorological Organization's Global Atmosphere Watch US and Canadian Monitoring sites, and UN-ECE's European Monitoring and Evaluation Programme (EMEP) sites have made substantial efforts to establish data centers and quality control programs to enhance integration of air quality measurements from different national and regional networks, and to establish observational sites in under-sampled, remote regions around the world. Similarly, the International Global Atmospheric Chemistry project (of the International Geosphere-Biosphere Programme) has strongly endorsed the need for international exchange of calibration standards and has helped coordinate multinational field campaigns to address a variety of important issues related to global air quality. The value of long-term atmospheric mercury monitoring and the need for additional sites is important in order to provide a dataset which can give new insights in the mercury cycling on different temporal and spatial scales, due to "surprising discoveries", such as Atmospheric Mercury Depletion Events (AMDEs) as a prominent example. A coordinated observational network for mercury (Hg) could be used by the modeling community for establishing recommendations for protecting human and environmental health on a global scale. Current networks are, in fact, inadequate because of lacking of a number of key point relating to (a) observations of all forms of Hg in the ambient air and in both wet and dry deposition; (b) long-term measurements of Hg and other air pollutants; (c) comprehensive monitoring sites in the free-troposphere; and (d) measurement sites that permit a careful investigation of inter-hemispheric transport and trends in background concentrations.

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2.2 Mercury measurements in Europe

2.2.1 Monitoring networks and trends

Continuous monitoring data sets exist for the time period 1998 to 2004 for two coastal background sites, Mace Head, west-Irish Atlantic coast and Zingst peninsula on the southern shore-line of the Baltic Sea. Automated total gaseous mercury (TGM) measurements have been carried out and an evaluation of the two data sets has been published by Kock et al. (2005). Between 1998 and 2004 the annually averaged TGM concentrations measured at Mace Head (1.74 ng m^{-3}) and Zingst (1.64 ng m^{-3}) remained fairly stable. For both stations higher concentrations were detected during the winter months and lower concentrations during summer, respectively. Since Mace Head is located at the European inflow boundary and therefore considered to be less influenced by continental emissions an unexpected west to east gradient was observed. Since no local anthropogenic mercury sources exist near the Mace Head station, it was concluded that enhanced emission from the sea provide the most probable explanation for the observed differences. Extensive evaluation of mercury measurements in air and precipitation at EMEP or OSPAR stations, respectively has been carried out by Wangberg et al. (2007). These data were obtained at coastal sites around the North Sea and originate from Ireland, Netherlands, Germany, Norway and Sweden. The observation period is 1995 to 2002 and a reduction in deposition (10–30%) has been observed, probably due to the decrease for the emission controls in Europe. In contrast, no decreasing trend in TGM data could be observed during the same time periods. The authors suggest that a plausible explanation is that European TGM emission reductions may be over-compensated by increasing emissions in other northern hemispheric regions. The project "Mercury species over Europe" (MOE) was aimed at identifying sources and atmospheric Hg-species behaviour (Pirrone et al., 2001; Munthe et al., 2003). Table 1 shows the comparison between average of Hg-species observed during MOE-MAMCS and the next EU-MERCYMS projects whereas the locations of

the measurement sites chosen during these projects are reported in Fig. 1. TGM concentrations at Mace Head is higher than those observed at the two Swedish stations Rörvik and Aspvreten and more similar to the levels at Zingst on the German Baltic Sea coast. The results from Neuglobsow, Zingst, Rörvik and Aspvreten follow a slightly decreasing trend, which is in line with the location of the main European source areas. There are no local sources of Hg at Mace Head and the slightly elevated concentrations are most likely caused by re-emissions from the sea surface (Pirrone et al., 2003; Munthe et al. 2003). Schmolke et al. (1999) reviewed regionally different background concentrations of TGM in North Central Europe and from the most southern sampling site to the northern most site median values of 1.93, 1.78, 1.53 and 1.54 ng m^{-3} TGM were detected. The Total Particulate Mercury (TPM) North-western European distribution pattern has been measured during the MOE project as well showing, as for the TGM, a south-to-north declining TPM gradient (Table 1). Since no direct emissions of particulate mercury were found, a possible explanation for the clear gradient is that TPM is formed after emissions and the measured fractions are actually secondary TPM (i.e. formed in the air mass during transport). A plausible mechanism is adsorption of Reactive Gaseous Mercury, Hg^{II} on existing particles which is an operationally defined gaseous Hg fraction present in ambient air. A comparison of the MOE data with results of an EU funded project (MAMCS) focusing on Southern European sites around the Mediterranean Sea have been carried out. Four synchronized seasonal field campaigns were performed at five coastal sites around the Mediterranean and in North Europe from 1998 to 1999 (Fig. 1). TGM, TPM and Hg^{II} were simultaneously measured at all sites along with meteorological parameters and compared. The results indicates that the TGM is slightly but significantly higher in the Mediterranean area than in North Europe, as well one of the major findings was that TPM and Hg^{II} concentrations were higher in the Mediterranean area than over Northern Europe in spite of the higher density of industrial and urban centres in Northern compared to Southern Europe (Pirrone et al., 2001; Wangberg et al., 2001; Munthe et al., 2001). The most probable interpretation is that higher emission rates and/or more active atmospheric

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transformation processes in the Mediterranean basin occurred. Photochemical processes in the marine boundary layer (MBL) lead to enhanced oxidation of elemental mercury vapour which would lead to increased concentrations of Hg^{II} and TPM via gas-particle interactions (Wangberg et al., 2001; Hedgecock et al., 2003; Sprovieri et al., 2003; Pirrone et al., 2003). The enhanced re-emission fluxes of mercury from the sea surface are, in fact, partly governed by sunlight and temperature and the warmer climate in the Mediterranean basin. High Hg^{II} concentrations have also been observed in the MBL of the North Atlantic (Bermuda) (Mason et al., 2001) and the Pacific (Laurier et al., 2003). The importance of Hg exchange processes between the atmosphere and surface waters has been highlighted in recent studies related to the Mediterranean region (Sprovieri et al., 2003; Hedgecock and Pirrone, 2004; Horvat et al., 2001, 2003; Gardfeldt et al., 2003; Kotnik et al., 2007; Sprovieri and Pirrone, 2008); the lack of knowledge of the magnitude of these exchange mechanisms is one of the main factors affecting the overall uncertainty associated with the assessment of net fluxes of mercury between the atmospheric and marine environments in the Mediterranean region. Deposition rates of mercury species from the atmosphere to receptor bodies depend on the chemical and physical properties of the species involved and their cycling, from speciated emission, transport, deposition, interaction with biota and possible re-emission to the atmosphere. Therefore, there is the need of a coordinated monitoring network in Northern and Southern Europe where the full range of measurements are made (i.e., atmospheric Hg speciation and dry deposition estimation, event-based wet deposition and flux, and the measurement of required ancillary parameters and detailed meteorology).

2.3 Over water Hg measurements and air/water exchange

The following section provides an overview of where and when measurements of atmospheric mercury and its compounds have been made in the marine environment on global scale. These measurements along with flux estimates have been made on

the Mediterranean, Atlantic, Pacific, Arctic oceans, North and Baltic Seas. Most results often show that Hg^0 concentrations appeared to be saturated relatively to the atmosphere thus the resultant flux of Hg^0 is from the ocean to the atmosphere.

2.3.1 Mediterranean

In the framework of the MED-OCEANOR project (Pirrone et al., 2003; Sprovieri et al., 2003, 2008, 2010) an in-depth investigation was carried out from 2000 to 2007 by several research groups to quantify and possibly explain spatial and temporal patterns of Hg-species concentrations in air, surface and deep water samples, and gaseous Hg exchange rates at the air–water interface along paths of a 6000 km cruise routes around the Sea basin (Sprovieri et al., 2003; Sprovieri and Pirrone, 2008; Gardfeldt et al., 2003; Horvat et al., 2003; Hedgecock et al., 2005; Pirrone et al., 2003; Kotnik et al., 2007). Figure 1 shows the routes followed by the research vessel (RV) during different seasons and covering the two sectors of the Mediterranean basin. Mediterranean Sea has been sorted into two regional sectors: western and eastern Mediterranean sectors. A statistical summary of the overwater Hg species observed during the Mediterranean cruises are reported in Table 2. Table 2 also reports a summary of TGM measurements performed over the Atlantic Ocean throughout several cruise campaigns by several research groups from 1977 to 2000 sorting the Atlantic Ocean into Northern and Southern Hemisphere (Slemr and Langer, 1992; Lamborg et al., 1999; Mason et al., 2001; Temme et al., 2003; Laurier and Mason, 2007). The transformations of Hg and its compounds which take place in marine water are of crucial importance to the understanding of the way in which mercury released to the atmosphere is eventually incorporated into biota, thereby becoming a risk to human and ecosystem well being. The major components of total mercury (Hg-tot) in seawater are mercuric chloride complexes, mercuric ions associated with dissolved organic carbon (DOC) (Munthe, 1991) and suspended particles. Some of these Hg forms can be reduced to Hg^0 both through biotic (Mason et al., 1995) and abiotic processes (Allard and Arsenie, 1991; Xiao et al., 1995; Costa and Liss, 1999) contributing to the super

saturation of Dissolved Gaseous Mercury (DGM) found in natural waters and thus to the evasion of Hg to the atmosphere (Schroeder and Munthe, 1998). The efficiency of the evasion process depends upon the intensity of the solar radiation, ambient temperature of the air above the seawater and the water temperature. Table 3 summarized mercury evasion data observed from oceans and sea waters. The evasional flux observed by Ferrara et al. (2000) over the Tyrrhenian Sea during 1998 showed a typical daily trend, being highest at midday when the ambient temperature and solar radiation were at a maximum, and lowest, near to zero, during the night, suggesting that solar radiation is one of the major driving factors affecting the release of Hg^0 from surface waters. In addition, a seasonal trend was also observed, with minimum values during the winter period and maximum values during the summer, probably due to higher water temperature that may have facilitated biotic and abiotic processes in the water column. The average Hg evasion value for the Tyrrhenian Sea, calculated by Ferrara et al. (2000), is consistent with a suggested gradient from west to south-east. Mason et al. (1994a) estimated the emission (mostly Hg^0) from the water surfaces to account for 30% (2000 tons per year) of the total emission of Hg to the atmosphere. The total global emission from the sea surface has been re-evaluated (Mason and Sheu, 2002) to 2600 tons per year. The average evasion from the western Mediterranean Sea was lower than the eastern sector, probably due to the higher mean degree of Hg^0 saturation in the east compared to the west (Table 3). Past or present tectonic activity may contribute to the high DGM concentrations found at these areas and therefore to major Hg evasion processes from the sea (Horvat et al., 2003). The Hg^0 evasion reported by Gardfeldt et al. (2003) from the western Mediterranean and the Tyrrhenian Sea is of the same order of magnitude as that estimated both by Ferrara et al. (2000) and Cossa et al. (1997). DGM data combined with an empirical gas-exchange model (Wanninkhof, 1992) data suggested that about 66 tonnes of Hg^0 are released to the atmosphere from the Mediterranean Sea during the summer (Gardfeldt et al., 2003). This emission is considerable in comparison to European anthropogenic emissions and should thus be taken into account in regional atmospheric modelling and assessment.

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The degree of saturation observed by Andersson et al. (2007) in the Mediterranean sectors shows variations between the seasons, explained by differences in water temperature. As for the saturation degree, the wind speed and the flux also varied between the different Mediterranean sections. The estimated summer flux results comparable to those estimated by Gårdfeldt et al. (2003) (Table 3), calculated using the gas exchange model (GEM) developed by Wanninkhof (1992). The total Hg^0 evasion from the Mediterranean Sea surface was calculated to be 77 tons per year (Gardfeldt et al., 2003; Andersson et al., 2007). Considering the estimations of the total oceanic evasion by Mason and Sheu (2002) (2600 tons per year) and Mason et al. (1994a) (2000 tons per year) the Mediterranean Sea would account for approximately 3–4% of the total oceanic evasion. Taking into account that the Mediterranean Sea represents 0.8% of the oceanic surface, the evasion from the Mediterranean Sea is almost 4–5 times higher compared to the global average sea surface evasion. However, there are currently great uncertainties in the estimation of the global Hg emissions from sea surfaces (Mason and Sheu, 2002).

2.3.2 Atlantic Ocean

The first measurements made on board ships during north–south traverses of the Atlantic Ocean were made between 1977–1980 (Slemr et al., 1981, 1985) and repeated in 1990 and 1994 (Slemr and Langer, 1992; Slemr et al., 1995). In 1996, Lamborg et al. (1999) performed Hg measurements in the South and Equatorial Atlantic Ocean from Montevideo, Uruguay to Barbados. The tracks of all cruises performed over the Atlantic are reported in Fig. 1. The open-ocean samples recorded a distinctive inter-hemispheric gradient, which is consistent with a long-lived trace gas emitted to a greater extent in the Northern than in the Southern Hemisphere (Lamborg et al., 2002). The results of all cruises made over the Atlantic Ocean are summarised in Table 2 in statistical terms. In the Northern Hemisphere (NH) TGM mean values are almost always higher than those obtained in the Southern Hemisphere. All cruises show a pronounced concentration gradient between the hemispheres. In addition, a rather

homogeneous distribution of TGM in the Southern Hemisphere (SH) was observed during the previous cruises (Slemr et al., 1981, 1985, 1995; Slemr and Langer, 1992). When latitude is taken into account, the TGM concentrations measured on board a ship are also comparable to measurements at remote coastal sites such as Mace Head (Ireland), Cape Point (South Africa), and Lista (Norway) (Ebinghaus et al., 2002; Baker et al., 2002). The agreement shows that combination of long-term measurements at several sites with snapshots of latitudinal distribution obtained by ship measurements is feasible and may provide information about the worldwide trends of atmospheric Hg. The gradient and the higher variability observed in the Northern Hemisphere suggest that the majority of emissions and re-emissions are located in the Northern Hemisphere. The inter-hemispherical gradient with higher TGM concentrations in the Northern Hemisphere remained nearly constant over the years. Measurements of Hg-species over water have also been performed during the BATS cruises by Mason et al. (2001) and on a follow-up cruise in August 2003 made by Laurier and Mason (2007). Hg^{II} measurements by Mason et al. (2001) represented the first reported measurements in the remote ocean marine boundary layer. Hg^{II} showed a clear and consistent diurnal cycle with maxima ranged up to 27 pg m^{-3} and the minima were often at the instrument detection limit, suggesting that the processes leading to a Hg^{II} build-up are likely photo-chemical. Similarly, a diurnal fluctuation in Hg^{II} was found during the North Pacific cruise (Laurier et al., 2003), although concentrations were much higher, up to 100 pg m^{-3}. For the subtropical North Pacific, the average Hg^{II} concentration was 11.8 pg m^{-3}, which was somewhat higher than that found for the North Atlantic, but showed the same degree of variability. In the Atlantic Ocean, only a few of these studies focused on measurements of DGM at offshore sites (Mason et al., 1998; Mason and Sullivan, 1999; Temme et al., 2005; Andersson, 2008). Mason et al. (1998) carried out measurements in the North Atlantic Ocean, while Mason and Sullivan (1999) measured DGM in the Equatorial and South Atlantic Ocean. The average concentration measured was 130±80 pg L^{-1} and the flux estimated was 15.9±10.8 ng m^{-2} h^{-1} (Table 3). In 1996, higher DGM concentrations have been measured by Mason and

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Sullivan (1999) in the Equatorial and South Atlantic Ocean than those in the north and consequently the flux estimated was higher (Table 3). It is suggested that the concentrations reflect a net accumulation of Hg^0 in the surface water. The flux was extrapolated to an annual flux of 700 ng m^{-2} yr^{-1}, a value that could not balance the atmospheric input. It was thus suggested that the DGM cycling in the surface was not in balance with the atmospheric input (Lamborg et al. 1999). Temme et al. (2005) measured DGM continuously in the North Atlantic Ocean in 2004, giving comparable DGM results with Andersson et al. (2008). An estimated flux, according to Nightingale et al. (2000), of 750 ng m^{-2} $month^{-1}$ has been estimated by Temme et al. (2005). Andersson (2008) continuously measured high resolution DGM concentrations in the North Atlantic in summer 2005 and the average flux was calculated to be 0.42±0.36 ng m^{-2} h^{-1}. An annual flux was estimated to be 460 ng m^{-2} $month^{-1}$ using the NOAA data base for the annual wind speed and water temperature, 9.5 m s^{-1} and 281 K, respectively.

2.3.3 Pacific Ocean

Measurements over the Pacific Ocean started in the early 1980s. There are insufficient data to extrapolate little change in concentrations over time for the North Pacific (Lamborg et al., 2002; Laurier and Mason, 2007) and/or differences between seasons and in their latitudinally distribution. More recent studies have included speciation measurements, particularly during a cruise between Japan and Hawaii in 2002 (Fig. 1) (Laurier et al., 2003). As for the Atlantic speciation data, there is clear evidence for a diurnal trend in Hg^{II} concentration, especially in the latter part of the cruise where the ship was in a lower ozone region, and there were higher UV radiations and higher temperatures, and often lower wind. All these factors would enhance the photochemical production of Hg^{II} and also lead to an increase in the atmospheric concentration. Measurements of DGM in ocean waters were first made in the Equatorial Pacific Ocean (Kim and Fitzgerald, 1988; Fitzgerald, 1995). Across the Equator, DGM varied from 10 to 27 pg L^{-1}, and waters were saturated relative to the atmosphere. In contrast, lower concentrations have been observed in the North Pacific region. Measurements of DGM

were also obtained in the equatorial region during a cruise in 1990/91 (Mason et al., 1991; Mason and Fitzgerald, 1993). For the Equatorial Pacific Ocean, the estimated evasion rates ranged between 12 and 230 ng m^{-2} day^{-1}. These evasion rates exceed the rate of atmospheric deposition estimated for the region (16 ng m^{-2} day^{-1}, Mason et al., 1994b) and another source of inorganic Hg to the mixed layer is required to maintain the estimated average evasion. It has been suggested that equatorial upwelling of Hg into the thermo cline provides this additional Hg source and this Hg is supplied to the equatorial thermo cline by southern circulation of Hg deposited at mid-latitudes (Mason and Fitzgerald, 1993; Mason et al., 1994b). From more recent DGM data, collected on a cruise in the North Pacific in May/June 2002, the evasional flux estimated (based on the average wind speed, surface water temperatures and the gas exchange equation of Wanninkhof, 1992) is higher in the tropical water and particularly during high wind speed events (Laurier et al., 2003).

2.3.4 Arctic Ocean

Sommar et al. (2007) reports measurements of DGM in Kongsfjorden in May 2002. A clear diurnal variation was observed with concentrations as high as 70 pg L^{-1} during day-time and concentrations as low as 12 pg L^{-1}. All waters measured were found to be supersaturated with respect to Hg, and evasion between 0.1 and 7 ng m^{-2} h^{-1} was calculated by the flux model developed by Wanninkhof (1992). It was suggested that this evasion was overestimated due to the fjord being partly ice covered thus hindering the wave field. St. Louis et al. (2007) carried out measurements of DGM at two ice-covered locations off of Ellesmere Island. The average concentration was 129±36 pg L^{-1}, which corresponds to super-saturation. The average flux was calculated according to Wanninkhof and McGillis (1999) to be 5.4±1.2 ng m^{-2} h^{-1}. The authors suggest that an even higher evasion would be expected during Hg depletions events, when more Hg is deposited into the Arctic Ocean and during ice-break-up and melt each spring. Andersson et al. (2008) carried out continuous measurements of DGM along the west coast of Greenland, into the Canadian archipelago, along the Alaskan coast into Russia around

the Wrangel Island and finally crossing the Arctic Ocean, from Barrow, Alaska across the North Pole to Spitsbergen. The overall average concentration measured in the water sampled was 45±22 pg L^{-1}, with a wide range of concentrations measured (from 5 pg L^{-1} in the Canadian archipelago to 134 pg L^{-1} north of Alaska). Measurements were carried out in both ice-covered and non-ice-covered areas, and the DGM concentration increased up to 80% between non-ice-covered and ice-covered areas. During transit through ice-covered areas, enhanced TGM concentrations were observed and it was speculated that the sea ice may act as a barrier for the evasion of Hg^0 from the sea surface, however when the ship broke the ice evasion became possible. Since most of the waters sampled were in ice-covered areas, the evasion of Hg^0 may be restricted. The authors, however, calculated according to Nightingale et al. (2000) the Hg^0 flux for the open waters and an Hg^0 evasion of 98 ng m^{-2} h^{-1} was estimated.

2.3.5 North Sea

Only few studies have been carried out for Hg speciation in the North Sea both at offshore and at coastal stations (Coquery and Cossa, 1995; Baeyens and Leermakers, 1998) (Table 3). These studies include measurements of DGM in the surface water and estimation of the flux from the area. Most samples demonstrated that the water was super-saturated of Hg giving a net evasion from the sea surface.

2.3.6 Baltic Sea

Six expeditions have been carried out in the Baltic Sea. Wangberg et al. (2001b) conducted two expeditions in summer 1997 and winter 1998 in the southern area (Fig. 1). Higher flux, calculated using the flux model by Wanninkhof (1992) was observed during the summer than the winter (Table 3). Kuss and Schneider (2007) carried out continuous measurements in the south of the Baltic Sea during all seasons in 2006. The lowest DGM concentrations were measured during winter and autumn whereas the highest concentrations were observed during summer and spring (Table 3). Fluxes

were calculated using the flux model by Weiss-Penzias et al. (2007). The lowest flux was observed during the winter season, when deposition of Hg was observed. The highest flux was calculated in the summer. The authors state that the annual evasion calculated for the Baltic Sea cannot be compensated by deposition of mercury.

2.4 Mercury measurements in North America

Long-term monitoring of atmospheric Hg with high time resolution has been started at Alert, Canada (January 1995). During the same year, Fitzgerald argued for and defined the basic requirements of an Atmospheric Mercury Network (AMNET) (Fitzgerald et al., 1995). Recognizing in fact, that TGM and Hg in wet deposition are spatially heterogeneous, several studies have aimed to set up monitoring networks in order to compare trends between sites in the same region, between regions, and to determine the influence of local and regional emissions sources. These needs have partly been accomplished on a regional scale within the Canadian Atmospheric Mercury Network (CAMNet) (CAMNet, www.msc.ec.gc.ca/arqp/camnet_e.cfm) that was established in 1996. Wet deposition is measured at the CAMNet sites as part of the Mercury Deposition Network (MDN), which includes sites in the United States, Canada and Mexico (http://nadp.sws.uiuc.edu/mdn/). A statistically significant decreasing trend for TGM concentrations at several rural CAMNet sites was seen for the time period 1995 to 2005 (Table 4). TGM concentrations at all the CAMNet sites were similar to or slightly lower than those observed at European background sites. Seasonal variations of TGM concentrations are observed for all sites. More recently, some sites within CAMNet have been measuring atmospheric Hg-species concentrations in addition to TGM. Table 5 presents an overview of the Hg-species measurements that were occurred in the USA since high-precision measurements have been made (since early 1990s). Concerning the Hg deposition measurements, the largest, most ambitious network of sites is the MDN. Some of these sites are co-located with Nation Trends Network (NTN) sites where concentrations of the major ions in precipitation were measured. The NADP-MDN network has been operating since 1996 (http://nadp.sws.uiuc.edu/mdn/),

with measurements made in Canada as part of CAMNet. The spatial pattern of Hg concentrations observed in wet deposition across some sites seem not be correlated very well with the spatial distribution of known mercury sources. In particular it was observed low Hg concentrations in wet deposition close to coal-fired power plants (i.e., Pennsylvania and Ohio) and high Hg values where there are few mercury sources (i.e., Florida). This means that removal processes are important in determining Hg deposition patterns (oxidation/scavenging) along with meteorological conditions. The MDN is essential for understanding the spatial and temporal patterns of Hg in wet deposition, but without at least TGM measurements and speciated Hg measurements in conjunction with precipitation measurements, little can be inferred about the processes responsible for controlling Hg deposition and temporal change of Hg concentrations in wet deposition. The few studies that have both speciated Hg measurements as well as Hg in wet deposition suggest that Hg in deposition can vary by 2-fold between an urban and a rural site, TGM might vary by 50–75%, and PM may vary 5–10 fold between sites. Since Canadian measurements began in 1995, mercury levels in the atmosphere have shown only a slight decline throughout most of Canada. The largest decreases in TGM were seen between 1996 and 2005 on the north shore of Lake Ontario, near Toronto and at St. Anicet, near Montréal, where levels fell by −13%. This is in good agreement with the overall trend in Hg-tot concentrations in precipitation observed within the comparable NADP-MDN sites, indicating that these changes are most likely driven by local or regional changes in mercury emissions. While wet deposition networks are currently in-place for Hg, the measurements challenges involved in quantifying Hg dry deposition have far prevented these measurements from becoming routine. Dry deposition measurement techniques have been developed (Keeler and Dvonch, 2005) using techniques that measure the various forms of Hg in the atmosphere as well as meteorological parameters to model the dry deposition flux at the measurement site (Driscoll et al., 2007). The key for estimating the dry deposition flux should be accurate measurement of atmospheric Hg in the gaseous forms and on size-fractionated particulate matter.

2.5 Mercury measurements in South America

Relatively few observations of atmospheric Hg have been carried out in South America or Mexico mostly of them carried out near to, or downwind of major sources (i.e., mining, industrial facilities, biomass fires). All South American observations reported Hg^0 concentrations substantially greater than the accepted global background level. However at present, there is no information in South America or Mexico that can be used to establish long-term trends. TGM concentrations observed in urban and rural areas were up to 10 ng m^{-3} (Hachiya et al., 1998) whereas Adjacent to mining areas concentrations up to 16 ng m^{-3} were found (Higueras et al., 2005). De la Rosa et al. (2004) sampled at four sites in Mexico. High TGM values and high variability were found suggesting strong nearby sources. Mean Hg values at Zacatecas were very high at 71.7 ng m^{-3}, whereas the Mexico City site was not as elevated (9.8 ng m^{-3}). At two rural sites, mean TGM values were near accepted global background concentrations (1.46 and 1.32 ng m^{-3}). Extreme TGM concentrations, up to nearly 100 µg m^{-3} were observed at some gold recovery operations (milling and amalgamation) (Higueras et al., 2005). From the above data, it is clear that past and current gold mining in South America represents a large source of Hg to the atmosphere. Lacerda (1997) estimated global Hg emissions to the environment from gold mining of 460 Mg yr^{-1} globally released to the environment, 300 Mg or 65%, of this is released to the atmosphere. Of this total, nearly 60% is released in South America. The atmospheric emissions of Hg in South America by gold mining calculated by Lacerda (1997) is nearly twice the total Hg emissions from all sources in South America estimated by Pacyna et al. (2006). However it should be noted that the Pacyna et al. (2006) inventory does not quantify Hg emissions from South American gold mining nor does it attempt to quantify Hg emissions from illegal gold mining activities. Thus, while emissions of Hg from gold mining in South America are clearly a substantial source to the global atmosphere, there is a significant uncertainty in the actual emissions. Future work on Hg emissions in South America should focus on reducing the large uncertainty in the emissions.

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2.6 Mercury measurements at altitude (including aircraft)

In an effort to understand the relative importance of anthropogenic and natural emissions of airborne Hg, several studies have attempted to calculate Hg fluxes from source regions. This could be in addition done by correlating Hg enhancements in plumes to other tracers whose emissions are known (i.e., CO). Ebinghaus et al. (2007) observed enhanced CO and TGM on two CARIBIC (Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrumented Container) flights between São Paolo and Santiago de Chile in 2005. The measured TGM/CO ratio on these two flights, (1.2×10^{-7} and 2.4×10^{-7} mol/mol), respectively, were similar to previous reports of biomass burning plumes. From these ratios the authors estimate global emissions of TGM from biomass burning in the range of 210–750 $\mathrm{Mg\,yr^{-1}}$. In two recent studies, TGM and CO were measured in 22 pollution transport "events" at Mount Bachelor (2800 m a.s.l.) between 2004 and 2005 (Weiss-Penzias et al., 2007). East Asian industrial events yielded a TGM/CO enhancement ratio of $\sim$0.005 $\mathrm{ng\,m^{-3}}$ ppbv, whereas plumes from Western USA anthropogenic sources and from biomass burning in the Pacific Northwest and Alaska gave a ratio of $\sim$0.001 $\mathrm{ng\,m^{-3}}$ ppbv. Thus, the TGM/CO ratio is an important distinguishing feature of Asian long-range transport. Scaling these ratios with estimated emissions of CO from China and global biomass burning, an emission of 620 $\mathrm{Mg\,yr^{-1}}$ is calculated for total Hg emissions from Chinese anthropogenic sources and 670 $\mathrm{Mg\,yr^{-1}}$ for global biomass burning. The Hg^0/CO molar enhancement ratio was observed in pollution plumes at Okinawa, Japan and produced a value of 6.2×10^{-7} mol/mol (0.0056 $\mathrm{ng\,m^{-3}}$ ppbv) (Jaffe et al., 2005). These plumes were identified to have originated from the industrialized region of eastern China and they produced a similar ratio to those observed at Mt. Bachelor. However, recent Chinese emissions inventories are a factor of two lower than the ratio in the plumes. Likely explanations for this discrepancy are (1) Chinese Hg emissions have been underestimated and (2) there are large natural sources of Hg that are not accounted for. Faïn et al. (2009b) report data of speciated atmospheric mercury levels at a high-altitude

research station, Storm Peak Laboratory (3220 m a.s.l.) in the Rocky Mountains in Colorado. They showed a very regular occurrence of high Hg^{II} levels in the dry troposphere that were not related to pollution events, but showed signs of atmospheric reduction, likely in the free troposphere or possibly over the Pacific Ocean. These observations provide evidence that the free tropospheric pool of mercury is enriched in divalent mercury compared to the boundary layer where high Hg^{II} levels are mainly related to local and regional pollution. Most air quality monitoring networks rely entirely upon ground-based sites that sample within the boundary layer. Addressing global air quality problems such as Hg contamination, however, will require observations that are made at higher altitudes above the boundary layer. Studies have shown that transport of pollution including Hg between Asia and the United States occurs primarily through the middle and upper troposphere, and because of the highly episodic nature of this transport, there can be significant inhomogeneity in the air masses reaching the continental United States. Thus, networks that only sample air masses within the boundary layer would not allow a quantitative determination of long-range pollutant fluxes. While sampling with aircraft can provide detailed information about Hg in the upper atmosphere (Banic et al., 2003; Ebinghaus et al., 2000; Friedli et al., 2004; Swartzendruber et al., 2006), in terms of long-term monitoring, the use of aircraft ahs obvious limitations. The preferred approach it to use mountain-top monitoring sites, that are frequently in the free-troposphere, and which located around the globe are essential to understanding the global transport of Hg and other pollutants (Jaffe et al., 2003). Currently, there are a number of such sites in existence, including Mt. Batchelor in the western USA (Jaffe et al., 2003), Mona Loa in Hawaii (Landis et al., 2005), Wank Mt. in Germany (Slemr et al., 2003), and the Lulin station in Taiwan (Sheu et al., 2007).

2.7 Mercury measurements in Asia

Kim et al. (1996) reported TGM concentrations from 13 remote mountainous sampling stations in Korea, from 1987 to 1993. Table 6 reports a statistical summary of the mean concentrations of atmospheric Hg determined from a number of monitoring

sites located in the East Asian countries. TGM concentrations ranged between 1.48 to 8.00 ng m^{-3}. Kim et al. (1996) concluded that the observed Hg levels and the wide spreadness of the observed data suggests that Hg pollution in the Korean atmosphere may result in generally enhanced levels compared to other northern hemispheric regions. This finding is supported by Sohn et al. (1993) who reported rural concentrations in Korea to be between 1.0 to 7.0 ng m^{-3} (mean 3.8 ng m^{-3}) for the years 1988–1989. TGM monitoring data for Korean GAW station (An-Myun Island) have been published by Nguyen et al. (2007). Measurements were routinely recorded between December 2004 and April 2006. The mean TGM concentration was 4.61±2.21 ng m^{-3} with a range of 0.10–25.4 ng m^{-3}. Analysis of the seasonal patterns indicated TGM concentration levels generally peaked in spring, while reaching a minimum in summer. Nguyen et al. (2007) concluded that Hg concentration levels at An-Myun Island can be affected intensively by trans-boundary input processes over certain periods of time and that its springtime dominance hence suggests combined effects of various local source processes and the meteorological conditions favourable for the massive air mass transport phenomenon (i.e., Asian Dust storms). Urban data from Beijing, China, show a similar distribution between summer and winter. Liu et al. (2002) and Wang et al. (2007) give winter concentration ranges between 8 and 25 ng m^{-3}, and lower summer values between 5 and 13 ng m^{-3}, with autumn and spring concentration in between. Feng et al. (2004) have reported TGM concentration data for Guiyang city in 2001 with a mean value of 8.40 ng m^{-3} on the basis of one year observation. The authors concluded that TGM concentrations in Guiyang are significantly elevated compared to the continental global background values and that coal combustion from both industrial and domestic uses is probably the primary atmospheric source. Similar data were obtained earlier (Feng et al., 2003) during 4 measurement campaigns in 2000 and 2001 in Guiyang. Hg-species are continuously measured from 2007 as well as hazardous heavy metals in particles and in precipitation at Cape Hedo Atmosphere and Aerosol Monitoring Station (CHAAMS) located on the north end of the island of Okinawa and used for many years to study the outflow of pollution from East Asia and the Asian continent. Monthly

mean concentrations of Hg^0 from October 2007 to January 2008 were approximately 1.3 to 1.7 ng m^{-3}, which were slightly lower than the spring observation in 2004.

2.8 Mercury concentrations in Africa

2.8.1 Monitoring network and trends

The Cape Point observations constitute the only long term data set of atmospheric TGM in the Southern Hemisphere. The monitoring of TGM was established at the Cape Point Global Atmospheric Watch (GAW) station in September 1995. Baker et al. (2002) presented the first data obtained until June 1999. Atmospheric Hg concentrations were found to be fairly homogeneous fluctuating between 1.2–1.4 ng m^{-3}. Whilst no significant diurnal variation is detectable, a slight seasonal variation with a TGM minimum in March–May and maximum in June–August was observed. A minimum annual TGM concentration was detected in 1997. The existing Cape Point TGM data base comprises both manual measurements with low temporal resolution as well as automated measurements with a resolution of 15 min. Good agreement exists between the manual analysis method and the automated measurements (Ebinghaus et al., 1999). The most prominent feature of the highly resolved TGM data is the frequent occurrence of events with almost complete mercury depletion which have so far not been observed at any other non-polar stations (Brunke et al., 2009). The Cape Point GAW station was found to constitute a suitable site for monitoring TGM concentrations in the Southern Hemisphere (SH).

2.9 Atmospheric mercury concentrations in Polar Regions

Polar Regions used to be considered pristine environments. Indeed, the Arctic is relatively far from industrial sities located at mid-latitude in the Northern Hemisphere and is less populated than other parts of the world. In the Southern Hemisphere, Antarctica is even less populated and impacted by anthropogenic activities, except on a local scale

by a few scientific stations. However, due to a combination of long-range transport associated with a specific climatology, the Arctic and, to a lesser extent the Antarctic, are affected on a large scale by pollutants originating from the mid-latitudes of the Northern Hemisphere. The substantial different geographical distribution of landmasses around both poles influences the Hg^0 annual mean concentration observed in the Arctic (~1.6 ng m^{-3}) and Antarctica (~1.0 ng m^{-3}). Research papers and reviews (Ebinghaus et al., 2002; Sprovieri et al., 2002, 2005a, b; Temme et al., 2003; Steffen et al., 2008; Dommergue et al., 2010; Nguyen et al., 2009) provide a comprehensive assessment of the state of the Hg science in the context of Atmospheric Mercury Depletion Events (AMDEs) in Polar Regions since 1995.

2.9.1 AMDEs in the Arctic

A surprising discovery that provided a great impetus for Arctic atmospheric chemistry research in several nations was the observing of an unusual phenomenon called Atmospheric Mercury Depletion Events (AMDEs) in the atmospheric boundary layer of the Arctic and sub-arctic regions. During the 3-month period following polar sunrise, it has been identified for the first time at Alert in the Canadian High Arctic (Schroeder et al., 1998), an atmospheric mechanism by which gaseous elemental mercury (Hg^0) may be converted to reactive and water-soluble forms, Hg^{II} and/or Particulate Mercury, Hg-p, that deposit quickly thus increasing the mercury fluxes and deposition processes in the fragile ecosystems of the Arctic and/or Antarctica. Springtime AMDEs first noted in the Arctic have also been observed in Antarctica (Ebinghaus et al., 2002). AMDEs occur at the same time as tropospheric ozone depletion events suggesting that both species were removed by similar unknown homogeneous and/or heterogeneous chemical reactions involving reactive halogen species (such as Br and BrO) across open waters and polynas. Several field experiments have been performed at different Arctic and sub-arctic locations. A comprehensive review of measurements performed in the Arctic regions is reported in Steffen et al. (2008). The Arctic is currently undergoing rapid and dramatic changes including warming which is changing the timing and extent of sea ice

and its coverage (Serreze et al., 2002) and it is affecting the seasons with winter coming later and spring melt coming earlier. As well, coal and fossil fuel combustion in Asia, a major global source of Hg, is expected to increase up to 350% between 1990 levels and 2020 (van Aardenne et al., 1999). The effects of these increasing emissions on AMDEs processes and the long term deposition of Hg to the Polar Regions will only be discernible if long term measurements are collected at numerous locations.

2.9.2 AMDEs in Antarctica

A comprehensive review of measurements in Antarctica is presented in a parent paper (Dommergue et al., 2010). The first extended baseline data for the concentration and speciation of atmospheric mercury in Antarctica were reported by De Mora et al. (1993). The measurements reported by Ebinghaus et al. (2002) comprise the first annual time series of ground-level TGM concentrations in the Antarctic to investigate the occurrence of possible AMDEs in south polar regions. The study also provides high-resolution data that can be compared with existing data sets of AMDEs in the Arctic revealing similarities. The TGM series measured at Neumayer showed several Hg depletion events during Antarctic springtime (between August and November) 2000; TGM and O_3 were strongly positively correlated as seen in the Arctic boundary layer after polar sunrise. Simultaneous measurements of Hg^0 and Hg^{II} were performed Terra Nova Bay from November 2000 to January 2001. Hg^{II} concentrations during the measurement period were surprisingly high and comparable with those at sites directly influenced by significant anthropogenic Hg sources. Recent studies performed in the Arctic (Lindberg et al., 2001) also report very high Hg^{II} concentrations between polar dawn and snowmelt, suggesting that there are specific mechanisms and/or characteristics of polar environments that at certain times, and apparently in the presence of surface snow are extremely favorable to the production of Hg^{II}. Comparable Hg^{II} results have been reported by Temme et al. (2003) at Neumayer during Antarctic summertime suggesting that the snow-pack is directly involved in maintaining

high Hg^{II} concentrations. Further studies are necessary to explain the reaction mechanism and the kinetics of the AMDEs and the Hg^{II} production in the Antarctic (Ebinghaus et al., 2002; Sprovieri et al., 2002; Temme et al., 2003). Long term measurements of Hg^0 and other atmospheric Hg species in the Polar Regions are very limited and need to be increased. These types of measurements can yield critical information to better understand the processes involved in the cycling of Hg in the polar atmosphere and thus the deposition of this pollutant to this fragile environment.

3 Conclusions and research needs

A monitoring Hg network on a global scale should leverage its efforts by collocating with other existing monitoring programs such as the World Meteorological Organization's Global Atmosphere Watch sites, US and Canadian Monitoring sites, and UN-ECE's European Monitoring and Evaluation Programme (EMEP) sites. The principal goals of a global Hg monitoring network consists in (a) to study the temporal and spatial variability of atmospheric Hg and atmospheric composition; (b) to provide long-term monitoring of changes in the physical and chemical state of Hg in the lower atmosphere and thus discern and understand the causes of such changes; (c) to establish the links between changes in atmospheric Hg, tropospheric chemistry and climate; (d) to support intensive field campaigns focusing on specific Hg processes occurring at various latitudes and seasons. Therefore, the over-arching benefit of a coordinated global Hg monitoring network would clearly be (e) the production of an universal high-quality measurement data sets that are currently desperately and the validation of models on different spatial and temporal scales. Currently it has become clear that while atmospheric Hg models have had some success in predicting the levels and trends in ambient Hg levels, the scarcity of global measurement data available for the comparisons make the exercise and results less significant. There is, therefore, a critical need for a coordinated global mercury monitoring network incorporating existing long-term atmospheric Hg monitoring stations with a number of additional sites to obtain a globally representative

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picture of atmospheric Hg in the troposphere along with trace gases, particles, and physical parameters at strategic sites that are globally distributed. A successful network, in fact, would consist of a relative small number of "intensive" sites, where the full range of measurements are made (i.e., Hg speciation in ambient air and dry deposition estimation, event-based wet deposition and flux, and ancillary parameters and detailed meteorology) and a larger number of "cluster" sites where only weekly wet deposition is collected. The cluster sites would allow for integration between the intensive sites, and examine the effects of local and regional conditions, while the intensive sites would provide the detailed information needed to calibrate and test global and regional Hg models (Driscoll et al., 2007). This approach is one model of how such a network would be constructed. Effective coordination among programs with related observational needs can avoid redundant data collection efforts or data gaps that occur when individual programs lack the resources to adequately support continued observational efforts. In addition, having more than one laboratory striving for the same goal is all the most effective way to assure that the highest quality observations will be made. The models would benefit from measurements at surface based sites performed as part of the coordinated network but would also benefit from closely linked intensive aircraft studies. Taking into account the needs for model development, the Hg network would be necessary integrated by (a) coordinated monitoring Hg wet deposition at global background sites far from anthropogenic sources as well as sites strategically located in/downwind of various source areas; (b) surface sites for continuous monitoring of Hg-species along with fundamental gas-phase species (i.e., CO, O_3, particulate and NO_x, SO_x) including remote locations for tropospheric background condition (i.e., Mt. Bachelor), sites suitable for measuring Asian outflow; locations where it will possible to examine the reactions in the MBL and the reactions occurring in polar regions, where oxidation of Hg^0 is enhanced. In addition, aircraft-based studies are essential both to identify the vertical distribution of Hg and correlations between Hg and other atmospheric species to evaluate the evolution of Hg^0, Hg^{II} and Hg in plumes downwind of major emission sources. Aircraft-based studies needs also for evaluating the effect

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of cloud processing on ambient Hg, as a basis for assessing the possible reduction of Hg^{II} through aqueous reactions. The need, therefore of a development of a coordinated global Hg monitoring program requires a high capacity and willingness to transfer knowledge and technology in order to support national and international programs and conventions (i.e., UNEP, UNECE-LRTAP, EU Mercury Strategy) in developing policy tools for forecasting the evolution of mercury pollution on regional and global scales with changing meteorological conditions and mercury emissions to the atmosphere, in order to reduce the impact on human health and ecosystems caused by the releases of mercury to aquatic and terrestrial ecosystems.

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Table 1. TGM, RGM and TPM average values observed at the selected coastal sites in the Mediterranean during the four seasonal campaigns of the MOE, MAMCS and MERCYMS projects. "D and N" mean "Day and Night", respectively.

Sampling campaigns	Sites	Coordinates	Fall			Winter		
			TGM ($ng\,m^{-3}$)	RGM ($pg\,m^{-3}$)	TPM ($pg\,m^{-3}$)	TGM ($ng\,m^{-3}$)	RGM ($pg\,m^{-3}$)	TPM ($pg\,m^{-3}$)
MOE	Neuglobsow, Germany	53°8.6′ N 13°02′ E	2.22	–	98.83	2.14	19.86	21.00
	Zingst, Germany	54°26.2′ N 12°43.5′ E	1.60	–	70.93	1.67	37.48	21.65
	Rörvik, Sweden	57°24.8′ N 11°56′ E	2.69	15.30	18.58	1.40	19.19	4.78
	Aspvreten, Sweden	58°48′ N 17°23′ E	1.68	–	12.37	1.31	11.13	9.99
	Mace Head, Ireland	53°20′ N 9°54′ W	2.03	28.59	3.99	1.72	25.68	3.51
MAMCS	Mallorca, Spain	39°40′ N 2°41′ E	3.16	1.88	34.40	3.08	99.59	86.12
	Calabria, Italy	39°25′ N 16°0.0′ E	1.30	40.18	26.32	1.86	24.84	28.55
	Sicily, Italy	36°40′ N 15°10′ E	1.34	90.14	5.57	2.37	46.39	8.46
	Antalya, Turkey	36°28′ N 30°20′ E	1.68	–	14.66	8.71	10.44	14.39
	Haifa, Israel	32°40′ N 34°56′ E	1.83	–	115.39	0.90	36.14	27.30
MERCYMS	Cabo de Creus, Spain	42°19.2′ N 3°18.9′ E	1.60	2.20	9.60	1.50	0.24	9.10
	Thau Lagoon, France	43°25′ N 3°35′ E	1.60	8.60	3.00	2.90	41.90	82.00
	Piran Marine, Slovenia	45°32.9′ N 13°33′ E	–	4.50	–	0.80	1.00	18.70
	Calabria, Italy	39°25′ N 16°0.0′ E	1.30	1.60	1.00	1.90	4.20	6.10
	Haifa, Israel	32°40′ N 34°56′ E	D 1.19 N 0.78	33.00	89.00	D 0.80 N 0.50	2.20	3.90

Sampling campaigns	Sites	Coordinates	Spring			Summer		
			TGM ($ng\,m^{-3}$)	RGM ($pg\,m^{-3}$)	TPM ($pg\,m^{-3}$)	TGM ($ng\,m^{-3}$)	RGM ($pg\,m^{-3}$)	TPM ($pg\,m^{-3}$)
MOE	Neuglobsow, Germany	53°8.6′ N 13°02′ E	1.98	27.23	46.17	1.58	27.94	30.93
	Zingst, Germany	54°26.2′ N 12°43.5′ E	1.47	54.61	23.81	1.69	9.15	22.48
	Rörvik, Sweden	57°24.8′ N 11°56′ E	1.54	18.24	7.94	1.39	17.41	7.61
	Aspvreten, Sweden	58°48′ N 17°23′ E	1.46	13.65	7.00	1.27	9.25	7.48
	Mace Head, Ireland	53°20′ N 9°54′ W	1.62	31.01	10.18	1.45	27.13	10.58
MAMCS	Mallorca, Spain	39°40′ N 2°41′ E	3.85	76.02	44.11	4.15	–	33.56
	Calabria, Italy	39°25′ N 16°0.0′ E	1.42	46.74	22.71	1.09	35.47	45.55
	Sicily, Italy	36°40′ N 15°10′ E	1.89	77.49	11.02	2.18	29.48	9.11
	Antalya, Turkey	36°28′ N 30°20′ E	1.34	21.00	25.25	–	–	65.25
	Haifa, Israel	32°40′ N 34°56′ E	1.45	34.81	97.89	–	–	4.19
MERCYMS	Cabo de Creus, Spain	42°19.2′ N 3°18.9′ E	1.60	2.20	9.60	2.10	1.20	11.20
	Thau Lagoon, France	43°25′ N 3°35′ E	1.60	8.60	3.00	3.30	191.00	662.00
	Piran Marine, Slovenia	45°32.9′ N 13°33′ E	–	4.50	–	4.00	15.40	9.40
	Calabria, Italy	39°25′ N 16°0.0′ E	1.30	1.60	1.00	1.60	–	–
	Haifa, Israel	32°40′ N 34°56′ E	D 1.19 N 0.78	33.00	89.00	D 1.24 N 1.21	8.30	22.70

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Table 2. Over waters TGM, Hg^0, Hg^{II} and Hg-p data observed in the Atlantic Ocean and Mediterranean sea during seasonal campaigns performed from 1977 to 2007.

Cruise	TGM Range ($ng\,m^{-3}$)	TGM Mean ($ng\,m^{-3}$)	TGM SD ($ng\,m^{-3}$)	Hg^0 Range ($ng\,m^{-3}$)	Hg^0 Mean ($ng\,m^{-3}$)	Hg^0 SD ($ng\,m^{-3}$)	Hg^{II} Range ($pg\,m^{-3}$)	Hg^{II} Mean ($pg\,m^{-3}$)	Hg^{II} SD ($pg\,m^{-3}$)	Hg-p TGM ($pg\,m^{-3}$)	Hg-p RGM ($pg\,m^{-3}$)	Hg-p TPM ($pg\,m^{-3}$)
Atlantic Northern Hemisphere[a,b]												
Oct 1977[c]	1.0–3.6	1.8	0.4	–	–	–	–	–	–	–	–	–
Nov–Dec 1978	1.4–2.7	1.9	0.3	–	–	–	–	–	–	–	–	–
Jan–Feb 1979	1.6–3.1	2.2	0.4	–	–	–	–	–	–	–	–	–
Oct–Nov 1980	1.4–3.4	2.1	0.4	–	–	–	–	–	–	–	–	–
Oct–Nov 1990	1.4–3.4	2.3	0.4	–	–	–	–	–	–	–	–	–
Oct–Nov 1994	1.3–3.2	1.8	0.4	–	–	–	–	–	–	–	–	–
Oct–Nov 1996	0.4–16.0	2.1	1.0	–	–	–	–	–	–	–	–	–
Dec 1999–Jan 2000	1.4–3.7	2.0	0.3	–	–	–	–	–	–	–	–	–
Atlantic Southern Hemisphere[a,d]												
Oct 1977[c]	0.8–1.7	1.2	0.3	–	–	–	–	–	–	–	–	–
Nov–Dec 1978	0.9–1.9	1.4	0.2	–	–	–	–	–	–	–	–	–
Jan–Feb 1979	1.1–2.1	1.3	0.2	–	–	–	–	–	–	–	–	–
Oct–Nov 1980	1.1–1.9	1.5	0.2	–	–	–	–	–	–	–	–	–
Oct–Nov 1990	0.9–2.4	1.5	0.3	–	–	–	–	–	–	–	–	–
Oct–Nov 1994	0.8–2.1	1.2	0.2	–	–	–	–	–	–	–	–	–
Oct–Nov 1996	1.0–2.3	1.4	0.1	–	–	–	–	–	–	–	–	–
Dec1999–Jan 2000	0.5–1.8	1.3	0.1	–	–	–	–	–	–	–	–	–
Feb–Mar 2000	0.2–1.3	1.0	0.1	–	–	–	–	–	–	–	–	–
Jan–Feb 2001	0.8–1.4	1.1	0.1	–	–	–	–	–	–	–	–	–
Mediterranean Eastern sector[e]												
Jul–Aug 2000	0.2–9.5	1.9	0.5	–	–	–	1.1–8.6	3.8	2.0	1.9–16.4	7.3	4.5
Aug 2003	0.7–15.7	1.6	0.5	0.2–11.4	1.3	0.7	2.8–22.5	9.1	5.3	0.04–10.1	1.8	2.0
Mar–Apr 2004	1.0–2.0	1.6	0.2	1.1–1.9	1.6	0.1	0.6–9.7	3.9	2.5	1.9–5.7	3.6	1.1
Oct–Nov 2004	0.7–4.0	1.60	0.50	0.7–4.0	1.5	0.4	0.1–6.3	6.7	12.0	0.04–51	4.5	8.0
Jun 2005	–	–	–	0.1–5.4	2.00	0.70	0.8–40	8.2	8.1	0.04–9.1	2.9	2.0
Sep 2006	–	–	–	0.4–2.8	1.2	0.5	0.4–76	14.4	16.4	0.1–14.8	4.4	2.6
Mediterranean Western sector[e]												
Jul–Aug 2000	0.1–11.1	1.7	0.8	–	–	–	0.2–30.1	11.6	9.8	4.8–17	9.6	3.2
Aug 2003	0.1–32	2.2	1.5	0.8–2.8	1.2	0.2	1.0–13.1	6.3	4.4	0.3–7.1	1.4	1.7
Mar–Apr 2004	1.0–8.6	1.8	0.3	0.5–4.4	1.7	0.3	0.1–25.3	6.2	5.5	0.2–11.9	2.6	2.0
Jul 2007	–	–	–	0.2–116.9	2.20	4.00	0.1–97.8	8.2	10.4	0.4–77.5	11.2	10.1

[a] Measurements made during Walther Herwing cruises in 1977 and 1978; Meteor cruises 1979 and 1980; Polarstern cruises in 1990, 1994, 1996, and 1999, 2001. [b] Nord of ITCZ. [c] Corrected for systematic error in the analytical method used during the Walther Herwing cruise in 1977. [d] South of ITCZ. [e] Measurements made during Medoceanor cruises from 2000 to 2007.

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Table 3. Mercury evasion from seawater surface of some aquatic environments.

Location	Flux[a] ($ng\,m^{-2}\,h^{-1}$)	Method[b]	References
Open waters			
Baltic Sea autumn average	0.8–2.1	GEM	Kuss and Schneider (2007)
Baltic Sea winter average	0.8	GEM	Wangberg et al. (2001b)
Baltic Sea spring average	1.0–2.1	GEM	Kuss and Schneider (2007)
Baltic Sea spring average	1.6	GEM	Wangberg et al. (2001a)
Baltic Sea spring average	3.1–6.2	GEM	Kuss and Schneider (2007)
North Sea	1.6–2.5	GEM	Cossa et al. (1997)
North Sea	0.49–9.25	GEM	Baeyens and Leermakers (1998)
North Atlantic	15.9±10.8	GEM	Mason et al. (1998)
Equatorial and Southern Atlantic	80	GEM	Mason and Sullivan (1999)
The mid-Atlantic Bight adjacent the East Coast of North America	2.5	GEM	Mason et al. (2001)
Open waters, Mediterranean sites			
North West Mediterranean sites	1.2	GEM[c]	Cossa et al. (1997)
Western Mediterranean Sea	2.5	GEM	Gardfeldt et al. (2003)
Western Mediterranean Sea summer average	5.2	GEM	Andersson et al. (2007)
Tyrrenian Sea	4.2	GEM	Gardfeldt et al. (2003)
Tyrrenian Sea	1.8	FC	Ferrara et al. (2000)
Tyrrenian Sea autumn average	3.2	GEM	Andersson et al. (2007)
Tyrrenian Sea spring average	0.7	GEM	Andersson et al. (2007)
Tyrrenian Sea summer average	4.1	GEM	Andersson et al. (2007)
Ionian Sea autumn average	3.8	GEM	Andersson et al. (2007)
Ionian Sea spring average	1.0	GEM	Andersson et al. (2007)
Ionian Sea summer average	2.8	GEM	Andersson et al. (2007)
Adriatic Sea autumn average	5.4	GEM	Andersson et al. (2007)
North Adriatic Sea autumn average	19.0	GEM	Andersson et al. (2007)
Strait of Sicily	2.3–40.5[d]	GEM	Gardfeldt et al. (2003)
Strait of Sicily spring average	0.7	GEM	Andersson et al. (2007)
Strait of Sicily summer average	3.5	GEM	Andersson et al. (2007)
Strait of Messina summer average	12.4	GEM	Andersson et al. (2007)
Strait of Otranto autumn average	2.1	GEM	Andersson et al. (2007)
Strait of Otranto spring average	0.8	GEM	Andersson et al. (2007)
Strait of Otranto summer average	3.3	GEM	Andersson et al. (2007)
Eastern Mediterranean Sea	7.9	GEM	Gardfeldt et al. (2003)
Coastal waters			
Skagerack part the North Sea summer average	0.8	FC	Gardfeldt et al. (2001)
Atlantic water at the Irish west coast	2.7	GEM	Gardfeldt et al. (2003)
The Tyrrenian Sea, polluted coastal zone	6.8	FC	Ferrara et al. (2000a)
Mediterranean Sea, near shore sites round Sardinia	3.8	FC	Gardfeldt et al. (2003)

[a] Average values from the corresponding sampling period. [b] GEM: Gas Exchange Model, FC: Flux Chamber. [c] Not based on DGM measurements due to a high detection limit. [d] Range from the corresponding sampling period.

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Table 4. Statistical summary of TGM measurements at CAMNet sites. R-W means RURAL-WEST (RFL, EST, FCH, BRL); R-E means RURAL-EAST (KEI, STA); R-A means RURAL-AFFECTED (WBZ, PPT, EGB); R-C means RURAL-CENTRAL (BNT).

Station	N	Mean ($ng\,m^{-3}$)	Median ($ng\,m^{-3}$)	Min ($ng\,m^{-3}$)	Max ($ng\,m^{-3}$)	Lower quartile ($ng\,m^{-3}$)	Upper quartile ($ng\,m^{-3}$)	SD ($ng\,m^{-3}$)
Alert	3603	1.55	1.58	0.03	3.12	1.45	1.73	0.37
Kejimkujik	3168	1.45	1.46	0.54	2.30	1.31	1.59	0.21
St. Andrews	2774	1.42	1.40	0.74	2.46	1.26	1.57	0.23
St. Anicet	3164	1.64	1.60	0.92	16.31	1.44	1.79	0.40
Point Petre	3275	1.78	1.73	0.80	4.26	1.55	1.93	0.34
Egbert	3207	1.67	1.66	0.95	6.90	1.50	1.80	0.27
Burnt Island	2680	1.58	1.58	0.99	2.48	1.43	1.72	0.21
Bratt's Lake	1424	1.53	1.52	0.79	2.68	1.38	1.64	0.24
Esther	878	1.65	1.65	1.19	2.14	1.54	1.75	0.15
Fort Chipewyan	305	1.36	1.35	0.95	1.77	1.28	1.47	0.15
Reifel Island	1642	1.67	1.67	0.91	2.92	1.56	1.79	0.19
Category (median of stations)								
R-W	2612	1.60	1.60	0.91	2.56	1.48	1.71	0.20
R-E	3263	1.43	1.43	0.88	2.09	1.31	1.56	0.19
R-A	3342	1.68	1.67	1.11	2.99	1.52	1.81	0.22
R-C	2680	1.58	1.58	0.99	2.48	1.43	1.72	0.21
ALL	3959	1.58	1.58	0.21	2.75	1.48	1.68	0.17

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Table 5. Summary of Hg^0, RGM and Hg-p measurements made at remote, rural and urban locations in the United States. NR means "not reported".

Location	Duration of study	Hg^0 mean ($ng\,m^{-3}$)	RGM mean ($pg\,m^{-3}$)	Hg-p mean ($pg\,m^{-3}$)	References
Remote Sites					
Mount Bachelor, Oregon	1.5 yr, 4 mth	1.4–1.8	39–60	4.4	Weiss-Penzias et al. (2007), Swartzendruber et al. (2006)
Cheeka Peak, Washington	1 yr	1.45–1.55	0–2.7	0–2.9	Weiss-Penzias et al. (2003)
Ship, between Bermuda and Barbados	2 mth	1.63±0.08	5.9	NR	Laurier and Mason (2007)
Rural Sites					
Chesapeake Bay Laboratory, Maryland	7 mth	1.7–1.8	6–13	NR	Laurier and Mason (2007)
Look Rock, Tennessee	2 mth	1.65	5	7	Valente et al. (2007)
Salmon Creek Falls Reservoir, Idaho	1.25 yr	1.3–1.6	1–10	NR	Abbott et al. (2007)
Great Mountain Forest, Connecticut	5 yr	1.4–1.6	NR	NR	Sigler and Lee (2006)
Cove Mountain, Tennessee	40 days	3.2	16.4	9.7	Gabriel et al. (2005)
Dexter, Michigan	4 mth, 6 mth	1.49–1.51	2–3	12±5.2	Lynam and Keeler (2005a)
Potsdam, Stockton, and Sterling, New York	3 yr	1.84–2.59	NR	NR	Han et al. (2004)
Pompano Beach, Florida	1 mth	1.6–2.0	1.6–4.9	3.5±2.8	Malcom et al. (2003)
Stillpond, Maryland	1 yr	1.7±0.5	21±22	42±50	Sheu et al. (2002
Underhill, Vermont	1 yr	2	NR	NR	Burke et al. (1995)
Walker Branch Watershed, Tennessee	6 studies over 3 yr	2.2	92±60	NR	Lindberg and Stratton (1998)
Urban, Industrial, Mining, or Fire Sites					
North-central Nevada	1 mth	2.5–3.0	7–13	9–13	Lyman et al. (2008)
Detroit, Michigan	1 yr	2.2±1.3	17.7±28.9	20.8±30.0	Liu et al. (2007)
Detroit, Michigan	2 mth	NR	NR	1–39	Lynam and Keeler (2005b)
Desert Research Institute, Reno, Nevada	3 yr, 3 mth	2.1–2.5	37±28	7±9	Stamenkovic et al. (2007)
Chicago, Illinois	15 mth	3.6±2.9	NR	70±67	Landis et al. (2002)
Baltimore, Maryland	2 yr	4.4±2.7	89±150	74±197	Sheu et al. (2002)
Athens, Georgia	5 days	3.9–8.7	9–129	NR	Landis et al. (2004)
Tuscaloosa, Alabama	1 mth	4.05	16.4	16.4	Gabriel et al. (2005)
Earlham College, Richmond, Indiana	6 studies over 3 yr	4.1	104±57	NR	Lindberg and Stratton (1998)

Table 6. Statistical summary of the mean concentrations of atmospheric Hg determined from a number of monitoring sites located in the East Asian countries.

Countries	Sites	City/Province	Study period	Hg mean ($ng\,m^{-3}$)	References
Korea					
1	Residential	Kwa Chun	99~00	5.25	Kim and Kim (2001b)
2	Han Nam	Seoul City	99~00	5.34	Kim and Kim (2001b)
3	13 Mountains	Nationwide, Korea	87~93	4.47	Kim and Kim (1996)
4	Yang Jae	Seoul City	99~00	5.26	Kim and Kim (2001a)
5	3 Terminals	Seoul City	Mar 98	6.54	Kim and Kim (2001c)
6	2 Mountains	Korea	97–98	7.03	Kim and Kim (2001c)
7	Hari	Kang Hwa Island	01–02	3.15	Kim et al. (2003)
Japan					
8	Urban	Chiba and two others	91~96/94	31.6	Nakagawa and Hiromoto (1997)
9	Urban areas	Chiba and three others	91~96/95	10.3	Nakagawa and Hiromoto (1997)
10	Suburban	Kushiro and two others	91, 94, 95	5.73	Nakagawa and Hiromoto (1997)
11	Oceans	Japan sea/Pacific	91	3.4	Nakagawa and Hiromoto (1997)
12	Rural city	Hayama and two others	91~96, 95, 96	16.5	Nakagawa and Hiromoto (1997)
13	Farmland	Tukui and two others	95/93	42.4	Nakagawa and Hiromoto (1997)
China					
14	Shijingshan	Beijing	Feb and Sep 98	6.75	Liu et al. (2002)
15	Xuanwu	Beijing	Jan and Sep 98	16.7	Liu et al. (2002)
16	Tiananmen Sq.	Beijing	Feb and Sep 98	10.5	Liu et al. (2002)
17	Two rural sites	Beijing	Feb and Sep 98	3.75	Liu et al. (2002)
18	Mountain sites	Guizhou	Unreported	3.35	Tan et al. (2000)
19	residential	Beijing	Jan and Feb 98	8.47	Liu et al. (2002)

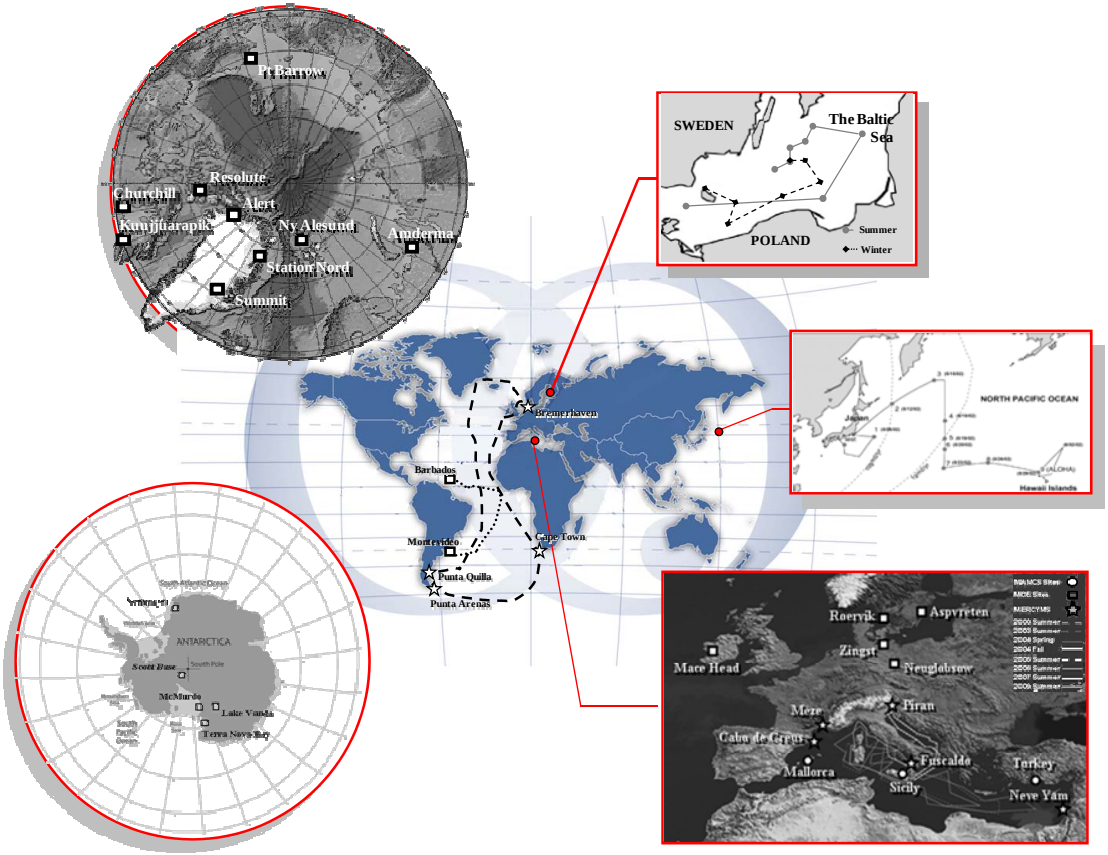

Fig. 1. Map of Measurement sites for mercury species in the Northern and Southern Hemisphere.

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