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Global mercury emissions to the atmosphere from anthropogenic and natural sources

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Abstract. This paper provides an up-to-date assessment of global mercury emissions from anthropogenic and natural sources. On an annual basis, natural sources account for 5207 Mg of mercury released to the global atmosphere, including the contribution from re-emission processes, which are emissions of previously deposited mercury originating from anthropogenic and natural sources, and primary emissions from natural reservoirs. Anthropogenic sources, which include a large number of industrial point sources, are estimated to account for 2320 Mg of mercury emitted annually. The major contributions are from fossil-fuel fired power plants ($810 \, \mathrm{Mg \, yr^{-1}}$), artisanal small scale gold mining ($400 \, \mathrm{Mg \, yr^{-1}}$), non-ferrous metals manufacturing ($310 \, \mathrm{Mg \, yr^{-1}}$), cement production ($236 \, \mathrm{Mg \, yr^{-1}}$), waste disposal ($187 \, \mathrm{Mg \, yr^{-1}}$) and caustic soda production ($163 \, \mathrm{Mg \, yr^{-1}}$). Therefore, our current estimate of global mercury emissions suggests that the overall contribution from natural sources (primary emissions + re-emissions) and anthropogenic sources is nearly 7527 Mg per year, the uncertainty associated with these estimates are related to the typology of emission sources and source regions.

1 Introduction

Advances achieved during the last decade on mercury emissions from major man-made and natural sources have contributed to better constrain the assessment of the impact of atmospheric mercury deposition on terrestrial and aquatic environments (Pirrone et al., 2001a,c; Hedgecock et al., 2006; Dastoor and Davignon, 2009; Jaeglé et al., 2009; Jung et al., 2009; Seigneur et al., 2009; Travnikov and Ilyin, 2009; Bullock and Jaeglé, 2009).

Policy makers have also taken the advantage of improved information on emissions to assess the effectiveness of measures aimed to reduce the impact of this highly toxic contaminant on human health and ecosystems. For example, following the preparation of the EU Position Paper on Ambient Air Pollution by Mercury (Pirrone et al., 2001b) and the first assessment of mercury contamination on global scale (Global Mercury Assessment Report, GMA) (UNEP, 2002), the European Union adopted the European Mercury Strategy, which is aimed to phase out the use of mercury in goods and industrial applications and to reduce, to the extent possible, mercury emissions to the atmosphere from fossil-fuel power plants and industrial facilities. As follow up of the GMA report publication and the adoption of the European Mercury Strategy, a number of activities have been developed in order to support the achievement of the objectives set by the

UNEP Governing Council (decisions 23/9 in 2005, 24/3 in 2007 and 25/4 in 2009) aiming to elaborate possible strategies and mechanisms addressed to phase out the use of mercury in a wide range of products and reduce emissions from industrial plants.

Earlier studies of global mercury emissions were aimed primarily to assess the contributions from anthropogenic sources (Nriagu and Pacyna, 1988; Pirrone et al., 1996, 1998; Pacyna et al., 2003, 2006), particularly from coal, oil and wood combustion as well as from solid waste incineration and pyrometallurgical processes. Several studies have estimated emissions from volcanoes (Nriagu and Becker, 2003; Pyle and Mather, 2003; Ferrara et al., 2000), artisanal small scale gold mining (Lacerda, 1995; Veiga et al., 2006), re-emission from oceans and surface waters (Pirrone et al., 2001a; Mason and Sheu, 2002; Hedgecock et al., 2006), top soil and vegetation (Gustin et al., 2000) and forest fires (Friedli et al., 2003; Cinnirella and Pirrone, 2006; Ebinghaus et al., 2007; Wiedinmyer and Friedli, 2007). More recently, assessments of mercury emissions to the global atmosphere have included the contribution of the most important anthropogenic and natural sources (AMAP/UNEP, 2008; Pacyna et al., 2010; Pirrone et al., 2009).

The evaluation of global emissions presented in this paper differs from previous published assessments because i) some new sources have been included in the estimate (e.g. vinyl chloride monomer production, coal-bed fires); ii) others have been updated (e.g. biomass burning, cement production), and iii) some regional estimates particularly with reference to coal combustion have been improved (i.e China, India).

2 Mercury emissions from natural sources

The estimate of mercury emissions from natural sources include the contribution from primary natural sources and re-emission processes of historically deposited mercury over land and sea surfaces. The mercury emitted from volcanoes, geothermal sources and topsoil enriched in mercury pertains to primary natural sources, whereas the re-emission of previously deposited mercury on vegetation, land or water surfaces is primarily related to land use changes, biomass burning, meteorological conditions and exchange mechanisms of gaseous mercury at air-water/top soil/snow-ice pack interfaces (Pirrone et al., 2001b; Mason, 2009).

The contribution of volcanoes varies over time depending whether they are in a degassing or eruption phase. The Hg/SO$_2$ mass ratio is generally adopted to estimate mercury emissions, though this approach is very controversial because of the paucity of relevant data and their variability (Nriagu and Becker, 2003; Pyle and Mather, 2003). The Hg/SO$_2$ ratios of 10^{-4} for explosive volcanoes, 10^{-4}–10^{-6} for passive degassing volcanoes and 10^{-6}–10^{-7} for ash rich plumes have been used in several previous evaluations

(Nriagu and Becker, 2003; Pyle and Mather, 2003; Ferrara et al., 2000; Bagnato et al., 2009a). Mercury emissions from calderas may also represent an important natural source of mercury; the Phlegrean fields (Pozzuoli, Italy) show fluxes of mercury, as Hg-S complexes, in the range of 0.9 to 19 g day^{-1} (Ferrara et al., 2000; Bagnato et al., 2009b). On average, volcanoes and geothermal activities release about 90 Mg yr^{-1} of mercury to the atmosphere (Mason, 2009), accounting for nearly 2% of the total contribution from natural processes. Hereafter, GEb indicates the percentage calculated with reference to Global Emission, AEb the percentage calculated with reference to total Anthropogenic Emission and NAb the percentage calculated with reference to NAtural sources.

Several studies suggest that the evasion of elemental mercury from surface waters is primarily driven by (i) the concentration gradient of mercury between the top-water microlayer and air above the surface water, (ii) solar irradiation which is responsible for the photo-reduction of oxidized mercury in the top-water microlayer, and (iii) the temperature of the top-water microlayer and air above the surface water (air-water interface) (Pirrone et al., 2003, 2005; Hedgecock et al., 2006). The evasion of mercury from lake surfaces is generally higher than that observed over the sea. In general, internal waters show a maximum net evasion of 2.39 ng m^{-2} h^{-1}. Over the open sea, mercury emission rates were found to be in the range of 1.16–2.50 ng m^{-2} h^{-1}, though dissolved mercury concentrations in the top-water microlayer (6.0 ng L^{-1}) were very similar to those observed in unpolluted coastal areas. On average, coastal waters and the Mediterranean Sea have the highest evasional flux, 1.83 and 1.96 ng m^{-2} h^{-1}, respectively. (Pirrone et al., 2003; Hedgecock et al., 2006). Mason (2009) reports recent estimates of total mercury evasion from ocean basins and lakes, which account for 2778 Mg yr^{-1} (37% GEb) of net gaseous mercury evasion to the atmosphere.

Mercury emissions from top soils and vegetation are significantly influenced by meteorological conditions, historical atmospheric deposition and the type of vegetation and top soil. Mercury fluxes from unaltered or background sites in North America have been found to be in the range of −3.7 to 9.3 ng m^{-1} hr^{-1}, and are similar to those observed in other background areas. In altered geologic sites the mean mercury flux was 15.5±24.2 ng m^{-1} hr^{-1} on average, and highest values up to 3334 ng m^{-1} hr^{-1} were found where calcine waste had been disposed off (Nacht and Gustin, 2004). Mercury emissions from vegetation depend upon several factors, including mercury uptake from the atmosphere, atmospheric deposition to foliage and mercury uptake from roots (Rea et al., 2002); however, the proximity of vegetation to natural or anthropogenic sources (hot spots or contaminated sites) may increase its mercury content (Lodenius, 1998; Lodenius et al., 2003). Recent studies show that most of the mercury found in foliage tissue originates from the atmosphere (Ericksen et al., 2003; Ericksen and Gustin, 2004). Summing

**Table 1.** Global mercury emissions by natural sources estimated for 2008.

Source	Mercury ($Mg\,yr^{-1}$)	Contribution (%)
Oceans	2682	52
Lakes	96	2
Forests	342	7
Tundra/Grassland/Savannah/ Prairie/Chaparral	448	9
Desert/Metalliferous/Non-vegetated Zones	546	10
Agricultural areas	128	2
Evasion after mercury depletion events	200	4
Biomass burning	675	13
Volcanoes and geothermal areas	90	2
TOTAL	5207	100

up all the net evasional fluxes from all regions and media (Forests, Tundra/Grassland/Savannah/Prairie/Chaparral, Desert/Metalliferrous/Non-vegetated Zones and Agricultural areas) the total net global mercury evasion is $1464\,Mg\,yr^{-1}$ (Mason, 2009).

Mercury emissions from biomass burning have only recently been considered in regional and global estimates (Friedli et al., 2003; Cinnirella and Pirrone, 2006; Wiedinmyer and Friedli, 2007; Cinnirella et al., 2008; Friedli et al., 2009a,b). The most recent estimate suggests that on a global scale nearly 675 Mg of mercury is released to the atmosphere from biomass burning every year (annual average for the period 1997-2006), which accounts for about 13% of the total contribution from natural sources (Friedli et al., 2009a). The highest contributing regions (Fig. 1) are equatorial Asia (28%), boreal Asia (15%) and Southern Hemisphere South America (14%). The part of Africa located in the Northern Hemisphere represents 12% of the global contribution, followed by Southern Hemisphere Africa (9%), southeast Asia (8%), central America (4%) and Australia (3%). The contribution from temperate North America (1%), boreal North America (3%), central Asia, Northern Hemisphere South America, Europe and Middle East combined (2%) is minor.

The current estimate of mercury emissions from natural processes (primary mercury emissions + re-emissions), including mercury depletion events, is estimated to be $5207\,Mg\,yr^{-1}$ (Table 1), which represent nearly 70% of the global mercury emission budget. Oceans are the most important sources (36% GEb) followed by biomass burning (9% GEb), deserts, metalliferous and non-vegetated zones (7% GEb), tundra and grassland (6% GEb), forests (5% GEb) and evasion after mercury depletion events (3% GEb).

Overall, the relative contribution of terrestrial surfaces is $2429\,Mg\,yr^{-1}$ (47% NAb) and that from surface waters is $2778\,Mg\,yr^{-1}$ (53% NAb). On an area basis, emissions from
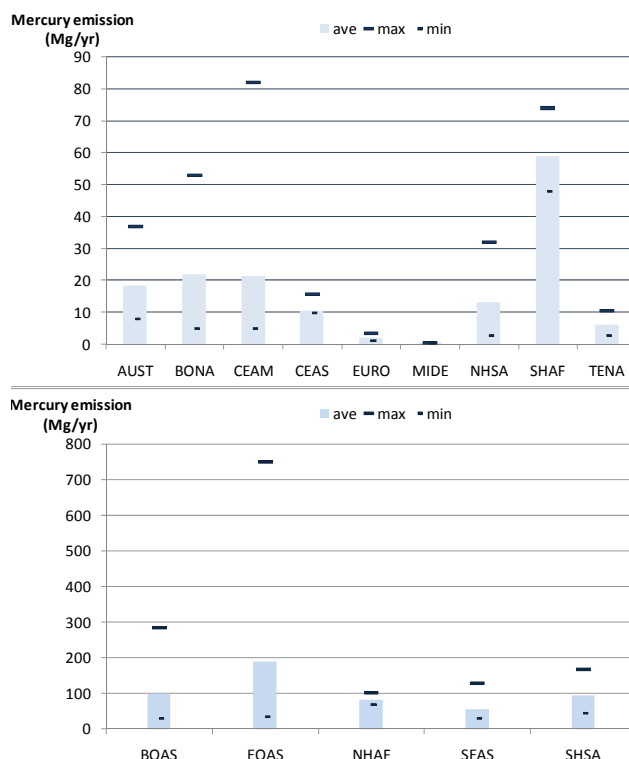
Fig. 1. Mercury emissions from biomass burning in Australia (AUST), boreal Asia (BOAS), boreal North America (BONA), central America (CEAM), central Asia (CEAS), Equatorial Asia (EQAS), Europe (EURO), Middle East (MIDE), Northern Hemisphere Africa (NHAF), Northern Hemisphere South America (NHSA), southeast Asia (SEAS), Southern Hemisphere Africa (SHAF), Southern Hemisphere South America (SHSA), temperate North America (TENA) (original data from (Friedli et al., 2009a)).

land (surface 1.46×10^8 km^2) are higher than those from the ocean (surface 3.49×10^8 km^2). Biomass burning represents 28% of emissions from land, whereas desert and non-vegetated zones represent 23% of the total, followed by tundra and grassland with 18% and forest with 14% of the total emission from land. Primary natural sources account for about 4% of current terrestrial outputs.

3 Mercury emissions from anthropogenic sources

Mercury is released to the atmosphere from a large number of man-made sources, which include fossil-fuel fired power plants, ferrous and non-ferrous metals manufacturing facilities, caustic soda production plants, ore processing facilities, incinerators for urban, medical and industrial wastes, cement plants and chemicals production facilities.

Three main parameters are used when estimating mercury emission from different industrial processes: the bulk material amount, the mercury content of the material and the

Table 2. Emission factors adopted to estimate the emissions (from (Pacyna et al., 2006; Streets et al., 2009b)).

Source category	Unit	Emission factor
Coal combustion power plants	g Mg^{-1}	0.04-0.3
Coal combustion in residential and commercial boilers	g Mg^{-1}	0.1–0.5
Oil combustion	g Mg^{-1}	0.058
Biofuel combustion	g Mg^{-1}	0.02
Cu smelting	g Mg^{-1}	5.0-6.0
Pb smelting	g Mg^{-1}	3.0
Zu smelting	g Mg^{-1}	7.5-8.0
Cement production	g Mg^{-1}	0.065-0.1
Pig iron and steel production	g Mg^{-1}	0.04
Municipal wastes incineration	g Mg^{-1}	1.0
Sewage sludge wastes	g Mg^{-1}	5.0
Mercury production (Primary)	kg Mg^{-1}	0.2
Gold production (Large scale)	g g^{-1}	0.5

Table 3. Median mercury removal efficiency (%) for some technologies and different categories (from (USEPA, 1997, 2002a,b; Wang et al., 2010))

Technology	Coal	Cement	Waste	Soda	Battery
ESP	32	25			
FF	42	50			75
FGD	34 (18–97)	–			
SDA+ESP	67(23–83)	–			
SDA+FF	30 (6–97)	–			
AC			50–95		
GSC				90	
ME				90	
AAC				90	

ESP=Electrostatic precipitators; FF=Fabric Filter; FGD=Flue Gas Desulfurization; SDA=Spry Drier Absorber; AC=Activated Carbon; GSC=Gas Stream Cooling; ME= Mist Eliminators; AAC=Adsorption on Activated Carbon.
ESP or FF installed before.
or sodium sulfide.

technology adopted to reduce emissions (abatement technology). The combination of mercury concentration in the material and the type and efficiency of abatement technology lead to the emission factor. Most used emission factors derived from literature are reported in Table 2. These emission factors have been used in our estimates for the assessment of mercury emission from each source category.

The installation of BATs in industrial plants plays a fundamental role in the emission control as most technologies can reduce mercury emissions up to 95% (USEPA, 1997, 2002a,b; Wang et al., 2010). Removal efficiency depends from adopted technology and production process (Table 3). Fossil fuels-fired power plants are the largest point sources of mercury released to the atmosphere, though other emission sources (e.g. artisanal gold mining) provide an important contribution to the global atmospheric budget (Pirrone et al., 2009).

World coal consumption in 2006 was 6118 Tg, representing the primary fuel used in electrical power generation facilities (42%) and accounts for about the 27% of world's energy consumption (EIA, 2009). Although it is very difficult to generalize the mercury concentration in coal, the literature indicates that the mercury content in coal varies between 0.01 and 1.5 g per Mg (Toole-O'Neil et al., 1999; Mukherjee et al., 2008; Pirrone et al., 2009) (Table 4). The concentration of mercury is somewhat lower in lignite coals than in bituminous and sub-bituminous coals. However, the lower heating values of lignite coals relative to bituminous and sub-bituminous coals suggest that the amount of lignite burned per MW of energy produced is higher compared to other coal types (Tewalt and Finkelman, 2001). Moreover, concentrations of mercury within the same mining field may vary by one order of magnitude or more (Mukherjee et al., 2009).

Table 4. Mercury concentration (μg g^{-1})) in coals from different geographic regions (Mg yr^{-1}).

Country/region	Hg in coal	Hg in coal fly ash	Reference
Australia	0.01–1.0	0.34	(1)
China	0.19–1.95	–	(1),(2)
Guizhou Province	0.52	–	(1)
Europe	0.01–1.5	0.23	(1)
India	0.11–0.80	0.007-0.28	(1)
Japan	0.045	-	(1)
Korea	0.012–0.048	–	(1)
Russia	0.02–0.9		(1)
South Africa	0.01–1.0	0.56–0.64	(1)
Argentina	0.021–0.96		(3)
Brazil	0.041–0.778		(3)
Colombia	0.020–0.17		(3)
Peru	0.041–0.63		(3)
Venezuela	0.030–0.280	0.268	(3)
USA	0.17 (mean)		(4)
World	0.02–1.0	0.62	

References: (1) (Mukherjee et al., 2008); (2) (Wang et al., 2000); (3) (Karlsen et al., 2006); (4) (Toole-O'Neil et al., 1999).

In developing countries and in countries with economies in transition (i.e., India) wood waste is primarily used to produce heat in the industrial sector, while wood is used in fireplaces and wood stoves in the residential sector with no emission control technology. Insufficient data are available, however, to estimate the typical mercury content of wood and wood wastes (Mukherjee et al., 2009).

Mercury emission from oil burning, as part of the fossil fuels category, represents a minor contribution compared to

Table 5. Global emissions of total mercury from major anthropogenic sources (Mg yr^{-1}).

	SC[a]	NFMP	PISP	CP	CSP	MP	GP	WD	O	T	Reference year	Reference[b]
S. Africa	32.6	0.3	1.3	3.8	0.0	0.0	0.3	0.6	1.3	40.2	2004	(1)
China	268.0	203.3	8.9	35.0	0.0	27.5	44.7	14.1	7.6	609.1	2003	(2)
India	124.6	15.5	4.6	4.7	6.2	0.0	0.5	77.4	7.5	240.9	2004	(3)
Australia	2.2	11.6	0.8	0.9	0.0	0.0	0.3	0.2	0.6	16.6	2005	(4)
Europe	76.6	18.7	0.0	18.8	6.3	0.0	0.0	10.1	14.7	145.2	2005	(5)
Russia	46.0	5.2	2.6	3.9	2.8	0.0	4.3	3.5	1.5	69.8	2005	(5)
N. America	65.2	34.7	12.8	15.1	10.3	0.0	0.0	13.0	1.7	152.8	2005	(6)
S. America	8.0	13.6	1.8	6.4	2.2	0.0	16.2	0.0	1.5	49.7	2005	(5)
Total	623.2	302.9	32.8	88.6	27.8	27.5	66.3	118.9	36.4	1324.3		
Rest of the world	186.8	7.1	10.4	147.1	135.1	22.5	333.7	68.5	28.2	939.4	2006	(7)
Total	810.0	310.0	43.2	235.7	162.9	50.0	400.4	187.4	64.6	2319.7[c]		

[a] SC, Stationary combustion; NFMP, Non-ferrous metal production; PISP, Pig iron and steel production; CP, Cement production; CSP, Caustic soda production; MP, Mercury production; GP, Gold production; WD, Waste disposal; CB, Coal-bed fires; VCM, Vinyl chloride monomer production; O, Other; T, Total.

[b] References: (1) (Leaner et al., 2009); (2) (Feng et al., 2009; Streets et al., 2009a); (3) (Mukherjee et al., 2009); (4) (Nelson, 2007); (5) (AMAP/UNEP, 2008); (6) (USEPA, 2005; Canada, 2008; CEC, 2001); (7) (Feng et al., 2009; Streets et al., 2009b).

[c] This sum considers also CB and VCM estimates, which account for 32.0 Mg yr^{-1} and 24 Mg yr^{-1} respectively. Totals for countries do not include these values.

that emitted from coal combustion. The list of the top five consumers of oil for power generation facilities include the USA, Japan, Russia, China and Germany. Relatively large volumes of distillate and residual oils are burned each year in the world. These fuels are used by electric utilities, commercial and industrial boilers (which, depending on their size, may be fired by either residual or distillate oils or a combination thereof) and residential boilers as well. Fuel oils contain mercury with concentrations that vary with crude oil type (Wilhelm, 2001). These values range from 0.007 to 30 g Mg^{-1}, with a typical value being 3.5 g Mg^{-1} (Wilhelm, 2001; Mukherjee et al., 2009). It is expected that mercury concentrations in residual oils are higher than those found in distillate oils, the latter being produced at an earlier stage in oil refineries. Heavier refinery fractions, including residual oils, contain higher quantities of mercury.

Natural gas may contain small amounts of mercury but the element is normally removed from the raw gas during the recovery of liquid constituents as well as during the removal of hydrogen sulfide. Therefore, it is assumed that mercury emissions from natural gas combustion are not significant when compared to those from other sources (Pirrone et al., 1996, 2001b).

Mercury emissions from stationary combustion facilities are certainly affected by the type and efficiency of control equipment, such as electrostatic precipitators (ESPs) and fabric filters (FFs) that are now commonly used as abatement measures in major electric power plants and central heating plants worldwide. Also flue gas desulfurization (FGD) units are used to control emissions but they are not common, especially in countries with economies in transition.

The combustion of fossil fuels (primarily coal) in stationary combustion (SC) facilities represents the most important anthropogenic source of mercury released to the global atmosphere annually (35% AEb) accounting for about 810 Mg yr^{-1}, with an important contribution from Asian countries (nearly 50% of the total) (Table 5). Global mercury emissions were also estimated on the basis of world coal consumption in fossil fuel-fired power plants (3400 Tg) and mercury emission factors in the range of 0.1–0.3 g^{-1} (EEA, 2009); the global mercury emission of 747 Mg^{-1} (min 374 Mg^{-1} and max 1121 Mg^{-1}) represents the majority (>90%) of mercury released to the atmosphere from SC facilities. On average, the uncertainty associated with these estimates is ±25%, as suggested by Swain et al. (2007) and Pacyna et al. (2009).

Mercury appears as an impurity of copper, zinc, lead and nickel ores as well as in gold ores (emissions from gold mining are discussed in the following section). Smelting processes to obtain these metals are known to be large sources of mercury released to the atmosphere, especially in developing countries (UNEP, 2002; Telmer and Veiga, 2009). Increasing trends in non-ferrous metal production by different processes, especially in new emerging countries, are leading to an increase of mercury releases to the atmosphere. Combustion temperatures in boilers, furnaces and roasters are key parameters affecting the amount of mercury released into the atmosphere though the chemical form and particle size distribution, and emission control technologies play an important role (Pirrone et al., 1996, 2001b). It is very difficult to discuss the average content of mercury in the copper, zinc, lead, nickel and gold ores as very little information

is available in the literature. On the basis of a US Geological Survey, best estimates of mercury emitted from non-ferrous ore processing are about 310 Mg yr^{-1} (USGS, 2004) with consistent differences with previous estimates (Pacyna et al., 2006; Hylander and Herbert, 2008; Pacyna et al., 2010) due to a substantial contribution from China which is about 203 Mg yr^{-1} (Streets et al., 2005, 2009a) (Table 2). The global annual contribution of about 276 Mg yr^{-1} reported by Hylander and Herbert (2008) includes a small release of mercury (83 Mg yr^{-1}) from smelting processes in China.

Emissions of mercury from primary and secondary pig iron and steel manufacturing plants are very much related to the overall production and the efficiency of emission control measures. Nearly 43 Mg (2% AEb) of mercury per year are released to the environment and no major changes have been reported for this sector during the 1990's and early 2000's (Pirrone et al., 2001a; Pacyna et al., 2006), whereas changes in local economies have led to changes in emissions at country level (e.g. Asia with a 10 Mg yr^{-1} increase) (Table 2).

In cement kilns, coal combustion is a significant source of mercury emissions. Mercury measurements in flue gases of cement kilns are very limited, therefore, the collection of new emissions data could be important for this source category. The evaluation of mercury emissions on the basis of emission rates should be performed keeping in mind that large differences may occur in cement kiln technology, which substantially affect the emission rates. Our estimate of mercury emissions from this particular source is based on an emission factor of 0.1 g per Mg of cement produced (Pacyna et al., 2006) and an annual cement production of 2315 Gg (2005), which leads to 236 Mg yr^{-1} of mercury emitted to the atmosphere (10% AEb). The uncertainty in this estimate is ±30% as suggested by Streets et al. (2009b) (Table 5).

Approximately 135 chlor-alkali plants using mercury cell technology were in operation worldwide in 2007, though in Europe after the approval of recent legislation most of plants have phased out the use of such technology and have been converted to membrane technology (WCC, 2007; Mukherjee et al., 2004). Due to the process characteristics, mercury can be emitted/released from the mercury cell process through emissions to the air, discharge of waste waters and solid wastes. The total mercury emission in Western Europe was 9.5 Mg in 1998, ranging from 0.2–3.0 g of mercury per Mg of chlorine capacity at the individual plants (EC, 2001a, 2002). In the literature, significant discrepancies can be found between the amount of emissions reported and the amount of mercury purchased to replace mercury in cells. This missing amount of mercury is in the range of 0.069 to 0.35 kg per Mg of NaOH produced; however, very different figures have been found for new emerging countries (i.e. India) where this amount is 25 times higher than that used to derive the global best estimate. Our estimate (Table 5) of mercury emissions from this industrial sector is about 163 Mg yr^{-1} (Mukherjee et al., 2009; Streets et al., 2009a).

Primary mercury production is another source of mercury released to the atmosphere. The official data on mercury production from mining is very uncertain because most countries do not report their mercury production in official statistical yearbooks. At present, productive primary mercury mines are located in Algeria, the People's Republic of China, Kyrgyzstan and Spain, whereas Italy, Mexico, Slovakia, Slovenia and Turkey retain significant reserves as a consequence of previous mining activities. In 2000, nearly 1800 Mg of mercury were produced (Maxson, 2006) which led to a global mercury emissions of nearly 50 Mg yr^{-1} (Table 5), this estimate is considered to be very conservative because of the large uncertainty associated with both the amount of mercury and the emission factors used (Pirrone et al., 2009).

Mercury released from artisanal and small scale gold mining activities (ASGM) is one of the most critical environmental issues, because almost all activities are in developing countries and countries with economies in transition. Current estimates are derived from governments data on mercury and gold exports/imports, field reports and analysis of the production and technology used. As result, ASGM is active in 70 countries, with 1000 Mg yr^{-1} of mercury released to the environment from this particular source. Nearly 400 Mg yr^{-1} (17% AEb) is the amount of mercury released to the atmosphere from ASGM , which includes 350 Mg yr^{-1} from amalgam burning and 50 Mg yr^{-1} from tailings (Telmer and Veiga, 2009) (Table 5).

Hazardous or non-hazardous waste generation is strictly related to the consumption of goods and the recycling processes adopted in the region or country. Maxson (2004) estimated that mercury use from 1994-2000 for all products and processes production has averaged 3600 Mg per year. A recent assessment for 2005 shows that the mercury supply is in the range of 3000–3800 Mg yr^{-1} (UNEP, 2006), which is quite different from that reported by Maxson (2006). Major uses of mercury are in small-scale artisanal gold mining, vinyl chloride monomer (VCM) and chlor-alkali production, as well as in batteries, dental amalgam, electronic devices and fluorescent lamps manufacturing (Fig. 2). Mercury is also emitted through cremation, agricultural practices and other minor uses (UNEP, 2002; Maxson, 2004). The amount of mercury in solid waste depends upon the mercury content in products, the products' lifetime and waste disposal mechanisms. Knowledge of mercury in different types of wastes is scarce and this implies also that the mercury emission estimate from waste disposal practices (i.e., incinerators, landfills) is affected by a large uncertainty.

Mercury in industrial wastes originates mostly from the phasing out of mercury from industrial processes and mercury-containing products. The most important source that generates wastes containing mercury is the chlor-alkali industry. Waste from the chlorine industry contains 10 to 17 g of mercury per Mg of chlorine capacity (EC, 2001b). Chlor-alkali production based on mercury cells represents
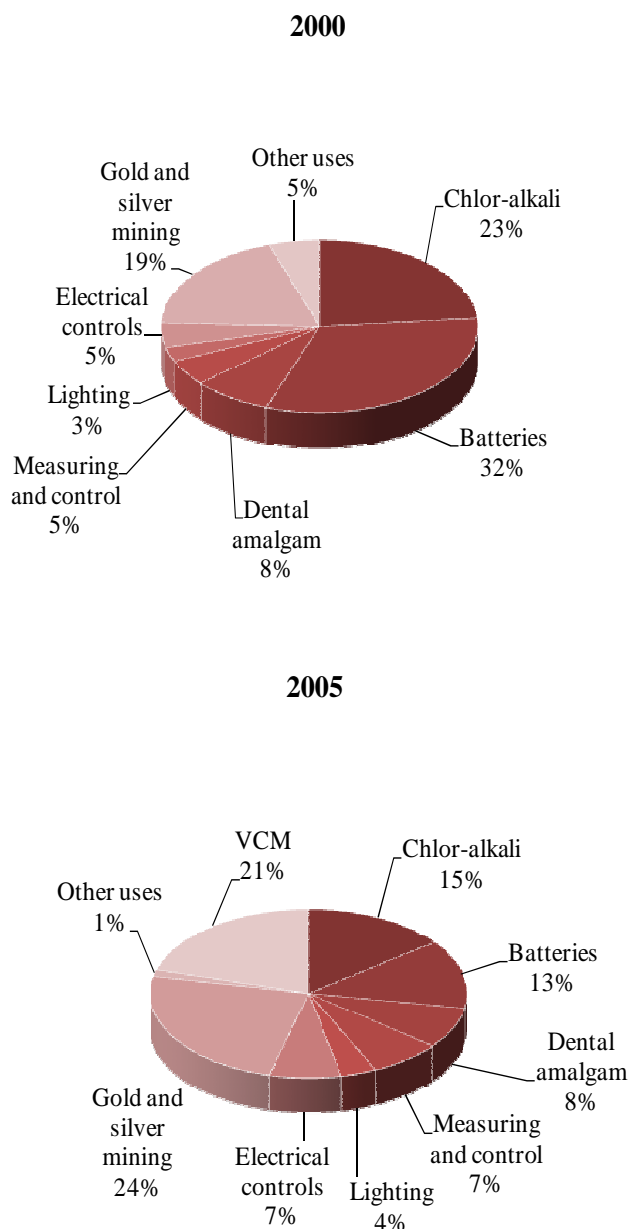
2000

2005

Fig. 2. Percentages of global mercury demand by use category in 2000 **(a)** and 2005 **(b)**. Global demand was 3386 Mg and 3415 Mg, respectively in 2000 and 2005 (from Maxson (2004) and UNEP (2006)).

21% of the total world capacity. Based on the chlorine production capacity by mercury cells (12 Tg) and the progressive reduction in the use of mercury cell technology ($10\% \, yr^{-1}$), it is estimated that in 2008 mercury waste from chlor-alkali plants was between 12 and 20 Mg with an average value of 18 Mg. Metal smelting is an additional source of waste containing mercury. The current estimate gives 9.4 Mg as upper boundary with a very low uncertainty (lower value is 8.4) (Pacyna et al., 2010). In the near future a large

amount of equipment phased out from industrial processes as well as mercury-containing products are expected to become mercury-containing waste.

Mercury in municipal waste is primarily related to consumer products. Mercury is used in batteries, dental applications, measurement and electronic devices, lamps and other minor applications. The estimate of mercury release from municipal solid wastes was based on the distribution coefficients pertaining to disposal (i.e. release by breaking, incineration, landfilling, recollection) and to related emission coefficients. For some European Countries, Mukherjee et al. (2004) estimated that 240 Mg per year is the amount of mercury in waste related to consumer products, however, mercury emissions from this sector have not been estimated yet. Slag produced from waste incineration processes could be a mercury source. It is mostly used for road construction, noise barriers, concrete production or landfill material. Slag has mercury concentration from 0.02 to $7.75 \, mg \, kg^{-1}$. Assuming a mercury content of $4 \, mg \, kg^{-1}$, the total mercury mobilized in slag from waste incinerators in Europe (EU-15 + 3 non-EU countries) varies between 24 and 54 Mg, which is partially released to the atmosphere (Mukherjee et al., 2004).

Mercury in medical waste has not been yet estimated in many countries, as a consequence a global assessment of mercury emissions from this particular source has not been made yet, and emissions are often lumped in the overall waste incinerators estimate (UNEP, 2002). In the United States, about 5000 medical waste incinerators are in operation with most of them releasing mercury that is 50 times higher than that released by a municipal solid waste incinerator (USEPA, 2008).

Summing up all contributions from the incineration of urban, medical and industrial wastes the global mercury emissions to the atmosphere from this emission source category is $187 \, Mg \, yr^{-1}$ (Table 5).

Coal-bed fires have occurred since prehistoric times and were initiated by natural causes including spontaneous combustion, lightning strikes and forest fires. However, they have proliferated worldwide since the Industrial Age, primarily as a consequence of anthropogenic activities (Stracher and Taylor, 2004). Today, tens of thousands of uncontrolled coal-bed fires are active in the world, which emit mercury among other compounds (Stracher, 2007). Hundreds of fires are currently active in China and the United States (Fig. 3). In China, there may be 200 coal-bed fires and in the United States more than 140, while there may be as many as 10 000 small coal-bed and peat fires in Indonesia (A. Whitehouse, personal communication, 2004). Taking into account the mean of the estimates for the amount of coal consumed annually by uncontrolled coal-bed fires (200 million Mg of which 112.5 million Mg in China and 87.5 million Mg in the rest of the World) and considering 0.16 g per Mg of coal as the average mercury content in coal, the amount of mercury released annually to the atmosphere by uncontrolled coal-bed fires is 32 Mg ($\sim$ 1% AEb) (Table 5).
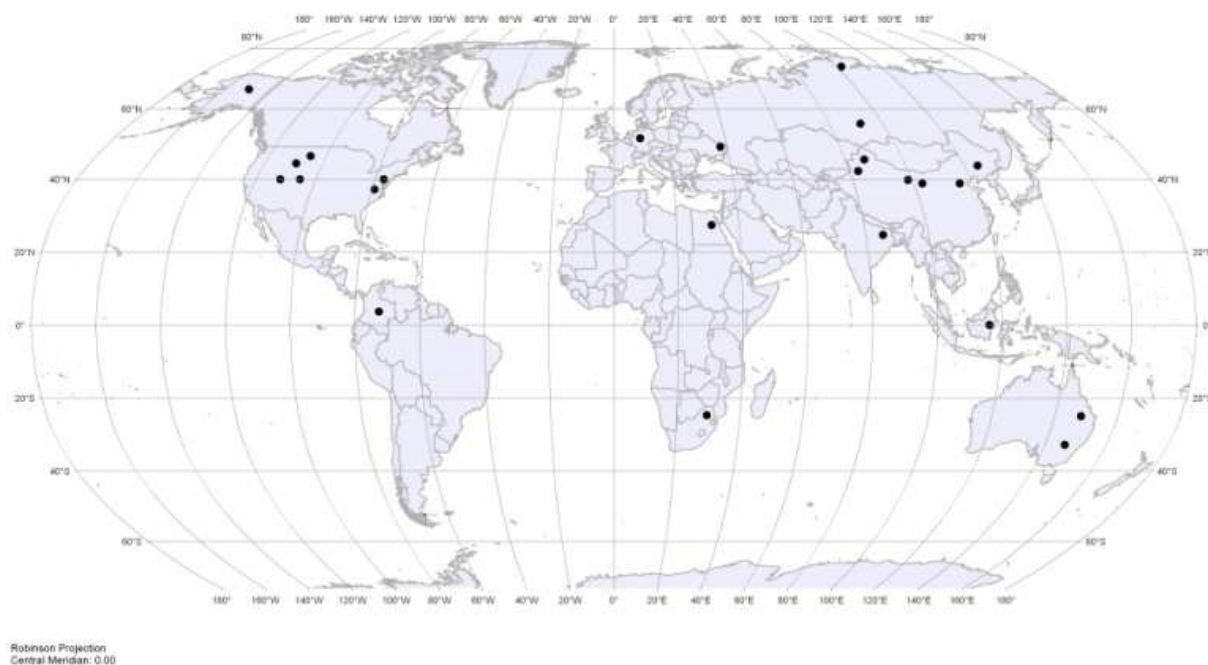
Fig. 3. Coal-bed fires distribution around the World (from http://www.gi.alaska.edu/~prakash/coalfires/global_distribution.html).

Vinyl chloride monomer (VCM) is an intermediate feedstock in the production of polyvinyl chloride (PVC). Among the two processes used to manufacture vinyl chloride, the acetylene process uses mercuric chloride on carbon pellets as a catalyst, while the other does not use mercury. In order to estimate mercury emission from PVC production, information on the precise amount of PVC resin produced by the acetylene technologies and mercury consumption/wastage per megagram of produced VCM is required. Global production of VCM in 2007 was almost 40 Tg. From 2004 to 2007, global consumption of VCM grew by about 5.5% per year as a result of strong demand for PVC, mainly for construction end-uses (Linak, 2009). Actual data on mercury consumption associated with catalyst for VCM production is fragmented. Nevertheless an investigation and calculation shows that the Hg/PVC ratio is in the range of 0.12–0.20 kg mercury per Mg of PVC produced and the amount released to the atmosphere is a small fraction (0.01) (Tsinghua University, 2009). Following the methodology proposed by the Tsinghua University group (Tsinghua University, 2009), mercury emissions have been estimated from global production of PVC, which in 2007 was approximately 34 Tg. Total PVC production involving mercury catalyst was near 12 Tg (35%) accounting for 24 Mg of mercury released to the atmosphere (Table 5).

Previous studies have not paid too much attention to mercury emissions from mobile sources. Recent estimates in the United States indicate that the overall emissions are relatively small compared to other emission source categories. For example, in the USA National Emissions Inventory (NEI) for 2002, it is reported that less than 1 Mg of mercury per year is emitted from mobile sources (highway vehicles, construction vehicles, recreational boats and aircrafts), which is less than 1% of the total mercury emission of the country (USEPA, 2005). Nevertheless, a significant effort has been made to assess mercury emissions from vehicular traffic (Conaway et al., 2005; Landis et al., 2007). A very conservative global assessment of mercury emissions from petroleum fuel consumption for 2000 was made recently by considering emission factors reported in literature and the world consumption of petrol and diesel (Pirrone et al., 2009). Petrol combustion contributed with $238 \, \text{kg yr}^{-1}$ ($121–281 \, \text{kg yr}^{-1}$) of mercury emissions, while diesel contributed $140 \, \text{kg yr}^{-1}$ ($71–209 \, \text{kg yr}^{-1}$). The total mercury emission was around $378 \, \text{kg yr}^{-1}$ ($192–564 \, \text{kg yr}^{-1}$) with a growing trend due to the increase of gasoline and diesel consumption. Region by region, North America released 156 kg, followed by Asia (94 kg) and Europe (80 kg). The global contribution from petroleum fuels combustion represented 0.00013% on AEb and can be neglected in our global assessment, however, our current estimate does not consider the contribution from fuel consumption in the shipping, aviation and military sectors and it does not account for the contribution related to the combustion of biodiesels.

In the last decades a considerable amount of research has been done to improve mercury emission inventories at country level, including those countries with economies in transition (Feng et al., 2009; Streets et al., 2009a). In Europe,

mercury emissions from anthropogenic sources in the year 2005 were near 145 Mg, with the highest contribution from stationary combustion sources (52%). The second contributing category consisted of several industrial sectors, including chlor-alkali production, ferrous and non-ferrous metal production and cement production (38%), whereas other sources, including waste incinerators and emissions from various mercury uses, account for about 10% of the total.

Coal combustion and the incineration of solid waste account for most mercury emissions in the United States (USEPA, 2005), whereas smelters for non-ferrous metal production accounts for most of the mercury emissions in Canada and Mexico (CEC, 2001; Canada, 2008). The total anthropogenic mercury emission from North America is estimated to be $153 \, \text{Mg yr}^{-1}$, which is lower than that reported in the previous regional and worldwide estimates (that assumed 1996 as the reference year) in which North American emissions ranged from $240 \, \text{Mg yr}^{-1}$ to $333 \, \text{Mg yr}^{-1}$ (Pirrone et al., 1996, 1998) (Table 5).

According to the official data, the total emission of mercury from Russian facilities was 2.9 Mg in 2001. Besides this information, a significant amount of mercury is released from area sources and from processes in which mercury is present as a natural impurity in the raw materials. The total Russian anthropogenic emissions are estimated to be $70 \, \text{Mg yr}^{-1}$, with 77% being the contribution from processes where mercury is mobilized as an impurity (ACAP, 2005) (Table 5).

Mercury emissions in China were estimated to be 609 Mg in 2003, with a large fraction (44%) due to coal combustion, which in China includes three major subcategories: coal-fired power plants, industrial boilers and residential uses. Emissions from these categories increased from 202 Mg in 1995 to 334 Mg in 2005 (with the largest contribution from power plants and manufacturing industries) (Streets et al., 2009a). As China is the largest coal producer and consumer in the world, mercury emissions in China have been increasing rapidly in recent years and are receiving increasing attention (Wu et al., 2006; Wang et al., 2010). By 2007, coal consumption by power generation in China increased to 1.49 billion tons, indicating a even higher annual growth rate during 2004–2007 (5.9%) (Wu et al., 2006). In addition, approximately 33% of the mercury is released from non-ferrous metals smelters (Feng et al., 2009). The emissions from cement production facilities (6%) and mercury mines (5%) represent a minor contribution. The mercury emissions from biomass burning was nearly $14 \, \text{Mg yr}^{-1}$, whereas spontaneous burning in coal mines accounts for $3 \, \text{Mg yr}^{-1}$ as reported by Streets and colleagues (2005; 2009a) (Table 5).

In Australia, the total mercury emission from anthropogenic sources is $16.6 \, \text{Mg yr}^{-1}$ with coal-fired power plants ($2.2 \, \text{Mg yr}^{-1}$) and non-ferrous metal smelters ($11.6 \, \text{Mg yr}^{-1}$) representing the major emission sources (Nelson, 2007). This estimate is larger than that reported in the National Pollution Inventory ($1.1 \, \text{Mg yr}^{-1}$), and significantly lower than that reported in the GMA ($97 \, \text{Mg yr}^{-1}$) (UNEP, 2002), however, it is in relatively good agreement with the earlier estimate of $6.3–8.6 \, \text{Mg yr}^{-1}$ reported in Pirrone et al. (1996) for the period of 1983 to 1992 (Table 5).

Mercury contamination is widespread in India and a recent study (Mukherjee et al., 2009) has dealt with industrial emissions of mercury from coal combustion, the iron and steel industry, non-ferrous metallurgical plants, chlor-alkali plants, the cement industry, waste disposal and others minor sources (i.e. brick manufacturing). No information was found in the literature for the pulp and paper industry or for the oil and petrochemical industry in India. The highest contributing source categories are coal combustion (52%) and waste disposal through incineration (32%). Industrial mercury emissions in India have decreased from 321 Mg in 2000 to 241 Mg in 2004. The Ministry of Environment and Forest in New Delhi has reported that 86% of mercury-cell chlorine plants have been converted to membrane technology (Mukherjee et al., 2009). This change suggests that mercury emissions have decreased from 132 Mg in 2000 to 6.2 Mg in 2004 (Table 5).

Limited information is available for African countries in relation to emissions from anthropogenic sources and mercury content in products, however, several studies have been carried out in South Africa in developing an emission inventory for major anthropogenic sources (Leaner et al., 2009). Nevertheless most of mercury released in the environment originates from artisanal gold mining activities (Telmer and Veiga, 2009). The country is a primary producer of important and strategic metals (e.g. gold, platinum, lead, zinc) and is a major producer and consumer of coal in Africa. Although the production facilities of these minerals and materials are known for their contribution to mercury pollution, detailed mercury emission inventories for these sources are not yet fully developed (Leaner et al., 2009). Leaner and colleagues critically revised previous estimates, giving an estimates for the country of about $40 \, \text{Mg yr}^{-1}$ (Leaner et al., 2009). Most of the mercury emissions are related to electric power generation facilities that account for 81% of the total national emission (Dabrowski et al., 2008). The coal gasification process accounts for 4% of the total, whereas coal combustion in cement kilns and producing clinker is the major source of mercury in cement production, representing 9% of the total emission (Table 5).

In Brazil, the amount of mercury entering the environment was estimated to be about $200 \, \text{Mg yr}^{-1}$ (Trade and Environment Database (TED) case 132). Gold recovery is performed by removing sediments from river bottoms and adjacent areas and feeding them through a number of mercury-coated sieves. Roughly 1.0 kg of mercury enters the environment for every kilogram of gold produced by artisans (Telmer and Veiga, 2009). Another estimate in the Alta Floresta area, Brazil, shows that a typical month's gold production of 230 kg emitted 240 kg of mercury to the atmosphere as elemental mercury vapor and 60 kg of mercury into rivers. In

1990

1995

2000

2007
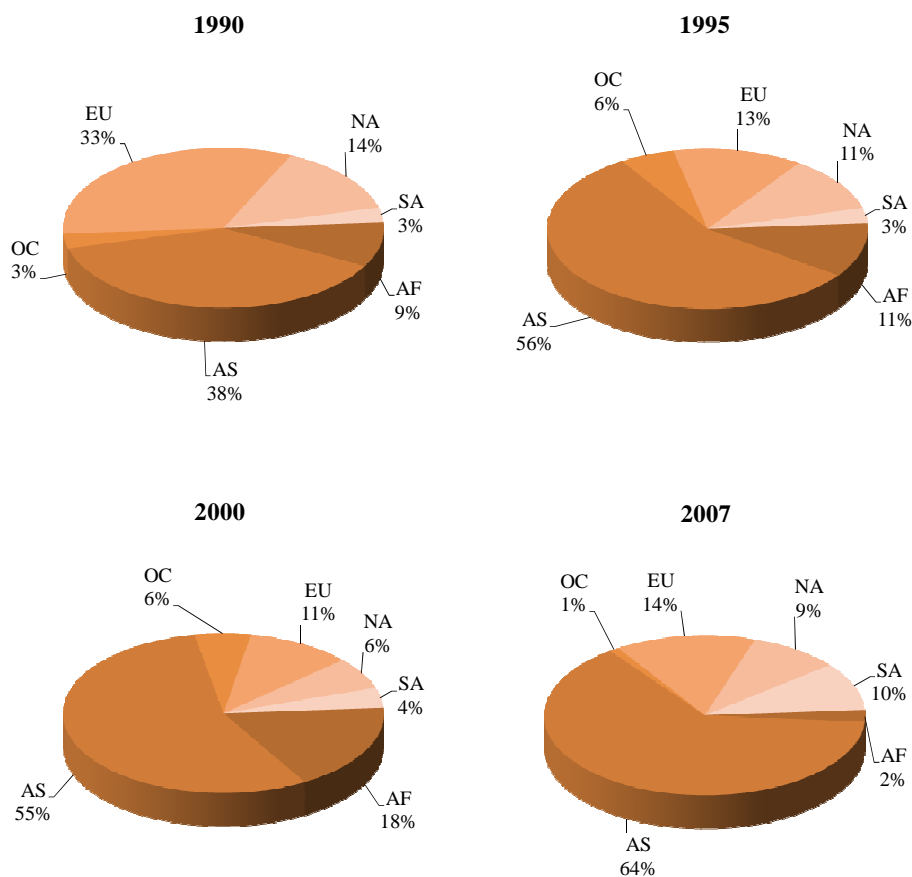
Fig. 4. Trends of global anthropogenic emissions by region based on Pirrone et al. (1996) **(a)**, Pacyna et al. (2003) **(b)**, Pacyna et al. (2006) **(c)** and this work **(d)**. Data reported in Fig. 3d are for most contributing countries as reported in Table 2 AF, Africa; AS, Asia; EU, Europe; NA, North America; OC, Oceania; SA, South America.

addition, emissions of mercury from coal fired power plants is about $5.6 \, \text{Mg yr}^{-1}$ (emission factor $0.2 \, \text{mg kg}^{-1}$) with a coal consumption of about $28 \, \text{Tg yr}^{-1}$ (Mukherjee et al., 2009) (Table 5).

Our current estimate suggests that summing up the contributions from anthropogenic sources, nearly 2320 Mg of mercury is released annually to the global atmosphere (31% GEb) (Table 6). The present assessment shows that the majority of mercury emissions originate from combustion of fossil fuels (11% GEb), followed by artisanal small scale gold mining (5% GEb), non-ferrous metal production (4% GEb), cement production (3% GEb), caustic soda production (2% GEb), waste incineration (2% GEb) and pig-iron production (1% GEb).

A comparison of our estimates with those reported in the literature (Fig. 4) suggests that Europe and North America are reducing their contribution to the global mercury burden, whereas emissions in Asia are increasing, the latter is primarily driven by the upward trend of energy demand that in the last decade has grown at a rate of 6 to 10% per year.

Table 6. Global mercury emissions from anthropogenic sources.

Source category	Hg emission (Mg yr^{-1})	Reference
Coal and oil combustion	810	This work
Non-ferrous metal prod.	310	(USGS, 2004)
Pig iron and steel prod.	43	(Pirrone et al., 2001b; Pacyna et al., 2006)
Cement production	236	This work
Caustic soda production	163	This work
Mercury production	50	This work
Artisanal gold mining prod.	400	(Telmer and Veiga, 2009)
Waste disposal	187	This work
Coal bed fires	32	This work
VCM production	24	This work
Other	65	This work
TOTAL	2320	

As widely recognized, mercury emission estimates are subjected to uncertainty (Pacyna et al., 2003; Streets et al., 2005; Lindberg et al., 2007; Wu et al., 2006; Swain et al., 2007; Wu et al., 2010). The primary methodology used for the uncertainty assessment was described in the work of Streets et al. (2003). Although estimates of current anthropogenic emissions for many other pollutants are cited with a greater precision, an uncertainty of $\pm 30\%$ for major industrial sources of mercury is widely accepted (Pacyna et al., 2010). Mercury emission estimates are directly related to the emission factors and activity levels ascribed to major anthropogenic activities, and the uncertainties in these two factors have an additive influence on the overall uncertainty associated with emission estimates (Pirrone et al., 2010). In detail, uncertainty for stationary fossil fuel combustion is 25% while that for non-ferrous metal, iron and steel and cement production is 30% (Pacyna et al., 2010).

In addition, a specific concern is for regions that are inadequately described in terms of point sources or exhibit unusually high uncertainties. Pacyna et al. (2010), suggested that emission estimates can be 27% for North America, 30% for Australia and Europe and 50% for Africa and South America.

4 Further research

The uncertainty of anthropogenic emission estimates is mostly related to rapid economic development in emerging economies, particularly in South and South-East Asia in which the impact of fossil fuel use in energy production is twofold. Firstly because fossil fuel power plants are the single most important anthropogenic mercury emission to the atmosphere, secondly because the other pollutants emitted as a result of fossil fuel combustion such as NO_x and SO_2 play an important role in the atmospheric chemistry of mercury and influence its local deposition patterns. A specific concern is for regions that are inadequately described in terms of point sources (Africa, South America) or exhibit unusually large uncertainties (Asia). These uncertainties affect model development, environmental policy and human welfare.

Atmospheric mercury models developed in recent years for assessing the relationship between emission source regions and receptor regions show a limited accuracy. The ability to determine the accuracy of current models is severely limited by the lack of a unified global emission inventory that includes an improved emission source characterization related to fossil fuel power plants in fast developing countries where energy demand is projected to continue to follow an upward trend in the next decade.

The improvement of the mercury emission inventory on a global scale, with special attention to fossil fuel-fired power plants in countries characterized by fast economic growth (i.e., China, India) will lead to a better assessment of the impact of different energy production strategies foreseen in ma-

jor environmental outlooks elaborated by leading institutions such as the UNEP, World Bank, Worldwatch Institute and International Energy Agency. Detailed mercury emission inventories at regional and global scales may help nations to shape future energy management strategies that, among others, will lead to a better assessment of countries' potential for renewable and non-renewable energy production; this is in agreement with recommendations and requirements of major international conventions and programs aimed to reduce the impact of anthropogenic pressures on ecosystems and human health.

The emission of mercury to the atmosphere driven by natural processes represents an important part of the global atmospheric mercury budget and is a dominant part of the global mercury cycle. However, while there is an on-going and continued effort to quantify these fluxes, the magnitude of their extent, including both primary and secondary (recycled) sources, is still poorly constrained.

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