500 years of mercury production: global annual inventory by region until 2000 and associated emissions

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Abstract

Since pre-industrial times, anthropogenic emissions of Hg have at least doubled global atmospheric Hg deposition rates. In order to minimize environmental and human health effects, efforts have been made to reduce Hg emissions from industries and power plants, while less attention has been paid to Hg mining. This paper is a compilation of available data on primary Hg production and associated emissions with regional and annual resolution since colonial times. Globally, approximately one million tons of metallic Hg has been extracted from cinnabar and other ores during the past five centuries, half already before 1925. Roughly half has been used for mining of gold and silver, but the annual Hg production peaked during a short period of recent industrial uses. Comparison with total historic Hg deposition from global anthropogenic emissions (0.1–0.2 Mtons) suggests that only a few percent of all mined Hg have escaped to the atmosphere thus far. While production of primary Hg has changed dramatically over time and among mines, the global production has always been dominant in the region of the mercuriferous belt between the western Mediterranean and central Asia, but appears to be shifting to the east. Roughly half of the registered Hg has been extracted in Europe, where Spanish mines alone have contributed one third of the world's mined Hg. Approximately one fourth has been mined in the Americas, and most of the remaining registered Hg in Asia. However, the Asian figures may be largely underestimated. Presently, the dominant Hg mines are in Almadén in Spain (236 t of Hg produced in 2000), Khaydarkan in Kyrgyzstan (550 t), Algeria (estimated 240 t) and China (ca. 200 t). Mercury by-production from mining of other metals (e.g. copper, zinc, gold, silver) in 2000 includes 48 t from Peru, 45 t from Finland and at least 15 t from the USA. Since 1970, the recorded production of primary Hg has been reduced by almost an order of magnitude to approximately 2000 t in the year 2000. Mining is thus still of similar magnitude as all current anthropogenic Hg emissions to the atmosphere, and mined Hg may account for more than one third of these emissions. Also before use, mercury is emitted from Hg mines locally during the mining and refining processes and from mining waste. Global direct emissions to the atmosphere amount to 10–30 t per year currently (up to 10 at Almadén alone), and probably exceed 10 000 t historically. Termination of Hg mining will reduce associated local emissions to the atmosphere and biosphere. Since several economically viable Hg-free

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alternatives exist for practically all applications of Hg, the production and use of Hg can be further reduced and all primary production of Hg other than by-production terminated.

1. Introduction

Mercury (Hg) is a globally spread pollutant due to characteristics such as low melting and boiling points, conversions between chemical forms and participation in biological cycles. As a result of anthropogenic emissions, the global atmospheric Hg deposition rate is approximately three times higher than in pre-industrial times and has increased by a factor of 2–10 in and around the most industrialised regions, (e.g. Meili, 1995; Bergan et al., 1999; Martínez-Cortizas et al., 1999; Lamborg et al., 2002). In order to reduce environmental input and human exposure, efforts have been directed at reducing Hg emissions from industries, waste incinerators and coal-fired power plants. Mercury emissions from cement kilns, roasting of sulfide ores for production of sulfuric acid and smelters processing sulfide ores (i.e. in the production of metals such as gold, copper, iron, lead and zinc) have been well documented, but local emissions from primary Hg production are generally missing in Hg emission inventories (Nriagu and Pacyna, 1988; Pacyna et al., 2001; MSC-E, 2001; Pacyna and Pacyna, 2002).

The quantities of primary Hg produced and its potential for emission within shorter or longer time frames has also been insufficiently considered to date. While a fraction of Hg present in municipal waste is of natural origin, the dominating part originates from anthropogenic sources, mainly from mined Hg. This has been demonstrated in Uppsala, Sweden, where the Hg concentration in municipal waste has been reduced more than fivefold during a 20-year period after legislation to reduce the use of Hg (from >2.5 g t^{-1} w.w to <0.5 g t^{-1} w.w; Hylander et al., 2002).

The aim with this study is to quantify the mobilization of Hg by mining, both historically and at present, and to relate the role of Hg mining to the primary uses of mined Hg and to anthropogenic Hg emissions. In addition, alternatives to the use of Hg are indicated.

A prerequisite for estimating present and historic, direct and indirect emissions of Hg is to collect the best possible records of Hg primary production, preferably with regional and annual resolution. Therefore, we made an attempt to compile all available data on primary Hg production and associated emissions since colonial times. The extraction of Hg has been carefully recorded for centuries in many mines, due to the economic significance of Hg in old times. This, in combination with the comparatively small number of Hg mines, has contributed to good documentation of metallic Hg production in Europe and the Americas, while production figures for Asia, the former USSR countries and Africa are incomplete.

2. Materials and methods

Data on mined quantities of Hg were compiled country by country from different sources, such as statistical yearbooks and scientific articles. Practically all North American production and a large part of global production during the last century and a half were obtained from Mineral Resources of the United States (1885–1934), USGS Minerals Yearbook (1943–1987) and Jasinski (1994), and USGS Mineral Commodity Summaries/Minerals Yearbook, Mercury (2002). These data were compared and complemented by data from Metallgesellschaft and other sources, as shown in Table 1. Data on early Spanish, South and Central American production were mainly collected from Bakewell (1971) and Fisher (1977), and Bethell (1984). Data on early Hg production in the Idrija mine (also spelled Idria) in present-day Slovenia were obtained in Mlakar (1974). The aim was to include all primary (virgin) production including by-production from mining of other metals, but to exclude secondary production from metal recy-

Table 1
Global, historical primary production (metric tons) of Hg until 2000, by dominating producer regions and characteristic epochs[a]. See Fig. 1 for annual resolution

Year	Europe[b]				America			Former USSR[k]	Asia[b]		Africa Algeria[n]	Other countries[o]	Globally mined
	Spain[c]	Slovenia[d]	Italy[e]	Other[f]	North[g,h]	S&C[g,i]	Mexico[j]		China[l]	Other[m]			
1501–1550	2601[p]	2449	0	0	0	0	0	0	>0?[q]	0	0	0	5050
1551–1600	4138	3931	0	0	0	8091	0	0	>0?	0	0	1	16 161
1601–1650	8740	6062	0	0	0	11 255	0	>0?	>3	0	0	0	26 060
1650–1700	5372	4111	0	0	0	11 628	0	>0?	>2	0	0	0	21 113
1701–1750	14 437	4241	0	0	0	9460	0	>0?	>0?	0	0	0	28 138
1751–1800	32 708	14 815	0	0	0	9887	0	>0?	>0?	0	0	0	57 410
1801–1850	35 544	11 299	0	0	266	1968	0	>0?	>0?	0	0	0	49 077
1851–1860	9330	2371	4	0	8464	>0?	0	>0?	>0?	0	0	0	20 169
1861–1870	10 879	2841	55	0	14 603	>0?	>0?	>0?	>0?	0	0	0	28 378
1871–1880	11 228	4327	744	25	17 975	>0?	>0?	>0?	>0?	0	0	93	34 392
1881–1890	17 434	5249	2566	279	12 794	>0?	>0?	892	1000	0	0	398	40 612
1891–1900	15 785	5521	2457	0	10 193	>0?	983	3253	6000	0	0	011	44 203
1901–1910	11 428	5740	4812	164	9400	>0?	905	2267[r]	1758	0	0	1136	37 610
1911–1920	10 627	6309	9373	693	8099	>10	398	79[r]	1241	136	0	2063	39 028
1921–1930	14 801	5955	11 719	626	4190	621	679	703[s]	314[r]	241	24	1962	41 835
1931–1940	12 086	3810	12 082	817	6547	699	2338	2049	549	344	99	1057	42 477
1941–1950	16 729	3972	16 058	635[s]	11 946	747	5575	400	769	1235	239	2280	60 585
1951–1960	16 226	4821	19 180	294	8041	934	5962	5949	4348	3164	21	858	69 798
1961–1970	20 601	5517	18 170	898	10 126	1806	6634	13 578	8992	5030	49	147	91 548
1971–1980	14 782	3409	6842	3309	8386	694	4717	21 300	8100	3236	7150	0	81 925
1981	1560	7	252	296	962	3	240	1700	800	204	30	0	6054
1982	1540	7	159	275	888	2	295	1200	800	246	386	0	5798
1983	1416	58	0	209	864	4	221	1200	850	162	828	0	5812
1984	1520	78	0	232	657	2	384	1220	800	182	586	0	5661
1985	1539	7	0	288	570	1	394	1200	800	226	801	80	5906
1986	1471	82	0	315	470	0	185	2250	700	262	764	0	6499
1987	1553	74	0	308	100	0	124	2300	700	211	756	0	6126
1988	1499	77	0	303	379	0	345	2300	940	97	662	0	6602
1989	967	57	0	291	428	0	651	2300	880	202	587	0	6363
1990	962	44	0	266	460	0	735	2100	800	60	639	0	6066
1991	52	16	0	149	58	0	340	1900	700	25	431	0	3671
1992	36	14	0	145	64	0	21	1900	392	5	476	0	3053
1993	636	7	0	148	70	0	12	1190[s]	520[s]	0	459	0	3042
1994	386	12	0	133	70	4	12	534	470	0	414	0	2035
1995	1497	11	0	140	70	13	15	520	780	0	292	0	3338
1996	1024[t]	5	0	108	30	13	15	709	510	0	368	0	2782
1997	389[t]	5	0	83	15[u]	15	15	725	835	0	447	0	2529
1998	474[t]	0	0	74	15[u]	24	15	725	225	0	224	220	1996

Table 1 (*Continued*)

Year	Europe[b]				America			Former USSR[k]	Asia[b]		Africa Algeria[n]	Other countries[o]	Globally mined
	Spain[c]	Slovenia[d]	Italy[e]	Other[f]	North[g,h]	S&C[g,i]	Mexico[j]		China[l]	Other[m]			
1999	433	0	0	40	15[u]	33	15	705	200	0	240	380	2061
2000[v]	236[t]	0	0	45	15[u]	48	25	640	200	0	240	400	1849
Sum	304 666	107 311	104 473	11 588	137 230	57 962	32 250	77 788	45 978	15 268	17 212	11 086	922 812
% of total	33.0	11.6	11.3	1.3	14.9	6.3	3.5	8.4	5.0	1.7	1.9	1.2	100

[a] Unless otherwise indicated, data are from Mineral Resources of the United States (1885–1934), USGS Minerals Yearbook (1943–1987), Jasinski (1994) and Kuo (2001) and USGS Mineral Commodity Summaries/Minerals Yearbook, Mercury (2002). Data for the last 50 years were cross-checked with data from Metallgesellschaft (1939–1998), Crowson (1999) and Roskill's (2000); t denotes metric tons; [b] excluding former USSR countries; [c] mainly Almadén, data for 16th–19th centuries mainly from Bethell (1984); [d] Idrija, data for 16th–mid 20th centuries mainly from Cigale (1997) and Mlakar (1974); the mine(s) belonged to Austria/Hungary 1867–1918, Italy 1919–1945, Yugoslavia 1945–1991; [e] mainly Mt Amiata, Hg produced at Idrija 1919–1945 is listed under Slovenia; [f] Austria 47 t, Czechoslovakia 5 883 t, Finland 2 090 t, Germany 2 081 t, Hungary 1 160 t, Ireland 264 t, Rumania 104 t; Sweden 1 t; [g] excluding Mexico; [h] mainly New Almadén and several other mines in California, a few in Nevada, Virginia; Canada 6 124 t; [i] mainly Huancavelica, Peru; data for 16th–mid 19th centuries from Bakewell (1971), Bethell (1984), Blanchard (1989), Brading and Cross (1972) and Fisher (1977); Yanacocha 1993–2000 from UNEP Chemicals (2002); [j] small, not reported quantities of Hg produced during colonial time; [k] mainly Nikitovka in Ukraine, Russia, Khaydarkan in Kyrgyzstan, and Jijikrut in Tajikistan (an antimony-Hg mine); [l] mainly in south-western China; data estimated if not otherwise indicated; 17th century Hg exports to Spanish America according to Bakewell (1971); 1890–1914 according to Bain (1933); [m] Japan (mainly Hokkaido) 6 810 t, Philippines (Palawan) 2 138 t, Turkey 6 179 t; [n] Incl. Tunisia 109 t, South Africa 9 t; [o] Australia 101 t, New Zealand 41 t, the rest is estimates for not identified countries; [p] (Blanchard, 1989; Mlakar, 1974); [q] Hg produced but not quantified; [r] exported quantity; [s] Metallgesellschaft (1939–1998); former USSR countries 1700 t, China 468 t for 1993 in Metallgesellschaft, 1996; [t] MAYASA, Almadén, pers. comm. 1999; 2002; [u] (Roskill's, 2000); [v] estimated quantity in reference.
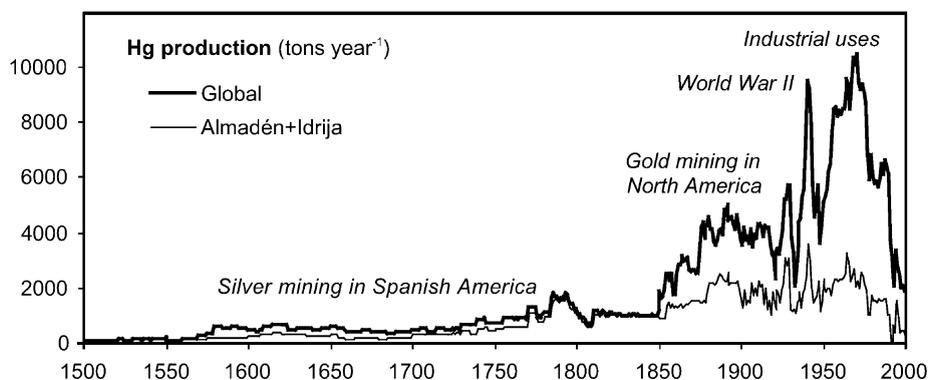
Fig. 1. Global, historic primary production of Hg, and the total contribution from the two main mining sites in Europe, Almadén in Spain and Idrija in present-day Slovenia. See Table 1 for data sources and production of Hg by regions.

cling, flue gas cleaning, etc. Reported data were in some cases exported quantities and in other cases they were estimates. In cases where reported data did not agree between different data sources, the lower value was used unless it was apparent that it was wrong. Missing data from a few years within a data series of continuous production were estimated by calculating the expected trend based on preceding and succeeding years. Missing data in the beginning and end of time series have not been corrected unless otherwise stated in Table 1. Data reported in flasks (generally 34.507 kg) and in quintals (46.0095 kg) have been transformed so that all data in this article are reported in metric tons.

Local emissions from mining and processing of primary Hg were calculated by using data and estimates from the mine and smelter in Almadén, Spain, where measurements were made in 1993 and 1994 (Ferrara et al., 1998).

3. Results and discussion

3.1. Total historic production of mercury and usage of mercury in gold and silver mining

Globally, a total production of close to one million tons of metallic Hg is reported throughout history (Table 1, Fig. 1). The Hg mines in Almadén, Spain, have alone contributed one third to the total reported world production. For most of the 500-year period, one of the mines in Almadén had the largest annual production among individual mines, only temporally surpassed by the Santa Bárbara mine in Huancavelica, Peru, or by New Almadén in the US (Mineral Resources of the United States, 1883; Fisher, 1977). Cinnabar has been mined in Almadén for over 2000 years (Allan, 1979; EBO, 2001), with temporally intensified activity under the Arabic Empire when 1000 laborers were employed there in the early 12th century (Goldwater, 1972), but the Hg production rate was unusually low until 500 years ago. This long history of mining and associated Hg emissions to the environment is reflected in the natural Hg record of a Spanish peat core (Martínez-Cortizas et al., 1999).

Mercury has been used in China for approximately 2000 years as an amalgamation agent (Liu, 2000). Cinnabar was used already more than 1000 years ago as a drug and preservative and as a pigment for the bright red/orange color in temples and palaces and on seals stamped on paper money (Liu, 2000; Rugoff, 1964). Quantities of cinnabar mined in China for these purposes are not available. The cinnabar production in Idrija (present-day Slovenia) for similar purposes during a 500-year period corresponded to 7628 t of metallic Hg, indicating a potential source of Hg pollution (Cigale, 1997). Data on Hg production in China before the 20th century are scarce, but Bakewell (1971) reported that at least 5 t of Hg was shipped from China to South America in the 17th century. Mines

producing cinnabar, as well as metallic Hg, have been reported in Tibet during the 19th century (Mineral Resources of the United States, 1883). Mined Hg quantities reported from China in the beginning of the 20th century are exported quantities (59 t in 1926), while the official production in 1926 was 128–470 metric tons (Mineral Resources of the United States, 1928). The production of just a few hundred tons mined yearly in the 1950s appears to be an underestimate considering the amount of Hg needed for chlor-alkali plants and gold mining (He et al., 2000; Qi et al., 2000).

The recovery and uses of mercury have been described since antiquity, possibly since the early 2nd millennium BC in Egypt, and the mining and recovery of cinnabar were certainly described in the 4th century BC. Alchemists in China are believed to have used mercury in trying to convert base metals to gold as early as in the 2nd century BC, and the Roman writer Pliny the Elder wrote in the 1st century about extraction of mercury by distillation and condensation, and its use in mining for amalgamation, all forerunners of modern methods of metallurgical treatment. Pliny also described the trade in elemental mercury and cinnabar between Spain and Rome, as well as mercury poisoning in mines (The New Encyclopedia Britannica, 1995; EBO, 2001). Similar to the Chinese, the Romans used the red colored ore as a pigment in paints and dyes (MAYASA, 2001). However, mined quantities of Hg remained small until the 16th century (Fig. 1), when large quantities of Hg began to be shipped from Europe to Central and South America for use in silver mining (Nriagu, 1993).

This onset of Hg pollution on a global scale is interestingly linked to Japan, the host of the 6th International Conference on Mercury as a Global Pollutant held in 2001 in Minamata, and also the site of the most well-known environmental disaster caused by Hg intoxication (this volume). Christopher Columbus had read that Marco Polo described Cipangu (Japan, also spelled Cipango, Cypangu, Zipangu) as a country with unlimited resources of gold, where the emperor's palace was covered by gold and the floor consisted of gold plates (Rugoff, 1964). This was one of Columbus' main arguments

to set out for his journey in 1492, after Queen Isabella I of Spain had promised to support him (Madariaga, 1944; EBO, 2001). It was by then already known that the earth was globe-shaped, and during his service with the Portuguese king, Columbus had even found a global map drawn in 1474 by Toscanelli (Fig. 2, cf. Madariaga, 1944). Based on several erroneous assumptions by Marco Polo, Toscanelli and Columbus, Columbus regarded the distance across the Atlantic ocean from the Canary Islands to the island of Cipangu (Japan), presumably located up to 1500 nautical miles from the mainland of Cathay (China), to be only 2400 instead of 10 600 nautical miles (Madariaga, 1944; EBO, 2001). According to this reckoning, Japan would have lain on the meridian of the Bermuda and Virgin Islands, easternmost China in Florida, and India not far from China (Fig. 2). Moreover, Columbus really encountered land confusingly close to where he had expected Japan to be situated, which added to the difficulties of accepting the 'discovery' of a new continent.

The Spaniards' arrival to Central and South America resulted in a systematic looting of gold and silver. Consequently, practically all gold and silver wares, which had been accumulated for generations, were shipped to Europe until 1540 (Bethell, 1984). Once these stocks were depleted, the focus shifted towards producing new silver and gold from ores. The production of silver in South America remained low for the first decades, when a smelter technique similar to the one used in Europe was employed. Around 1555, the Hg amalgamation process used in the Saxon mining districts of Europe was introduced to New Spain (present-day Mexico) by a German named Lomann and a Spanish merchant, Bartolomé de Medina (Cowen, 1999). By adding both Hg and copper sulfate to the ore, the method was subsequently adapted to the silver sulfide ores of Central and South America (Bethell, 1984). This drastically reduced the consumption of firewood, which had become scarce around the mines, and made silver extraction profitable even in the abundant silver ores with a metal content lower than those extracted in Europe.

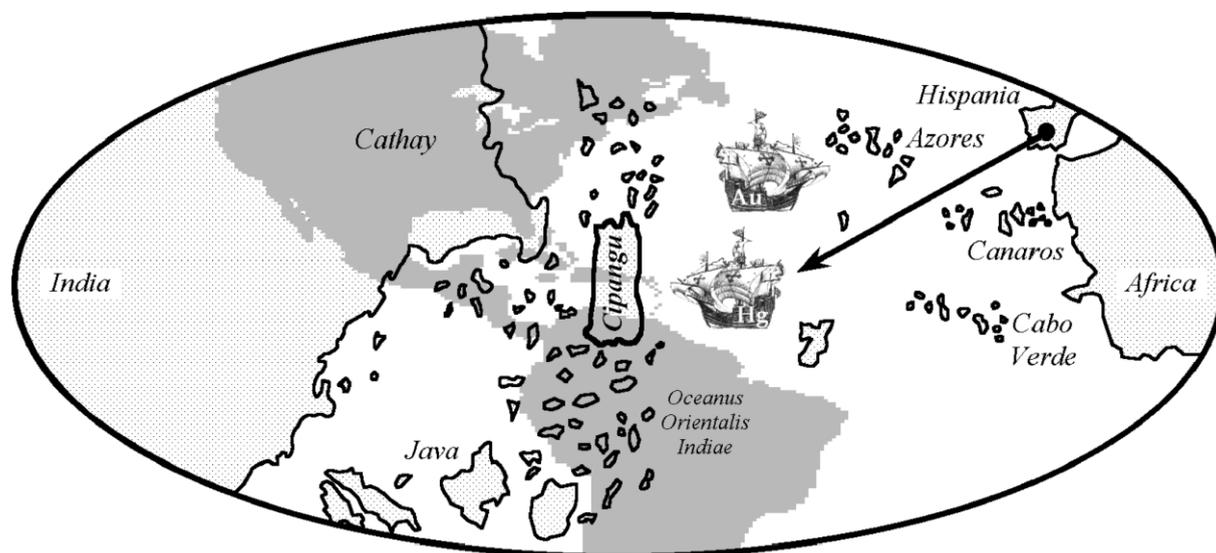Shipments of Hg from Spain for use in silver mining in the Spanish colonies in America lasted

Fig. 2. Reconstruction of Toscanelli's map of 1474 (after e.g. Ålund 1892) which lended support to Columbus' venture in 1492 of sailing west to Japan (Cipangu), 'the country glimmering in gold' east of China (Cathay); the subsequent trade with the New World (superimposed) entailed that mercury became an early global pollutant (see Fig. 1), triggered by the Spanish access to the unique mercury resources in Almadén (indicated).

for more than 250 years, resulting in more than 60 000 t of Hg sent overseas, mainly to present-day Mexico (Fig. 1, Table 1), and exchanged for silver and gold. Between 1621 and 1645 (2633 t; Bethell, 1984) and after 1786 (quantities not known), Hg was also shipped to Spanish America from the Hg mine in Idrija, Slovenia, which had been discovered in 1490. Until 1820, more than 50 000 t of Hg were produced in the Huancavelica Hg mines in Peru, discovered in 1570 (Table 1).

Between 1571 and 1660, 17 300 t of silver were imported to Spain from America (Brading and Cross, 1972). Approximately 25 000 t of Hg were used during this period (Table 1). This suggests a Hg to silver ratio of 1.5:1. Considering that part of the silver was produced by smelting, the ratio may have been somewhat larger, but likely in the same range as the Hg to gold ratios in present-day gold mining activities in Brazil (generally approx. 0.7–2.0; Mallas and Benedicto, 1986; Pfeiffer and Lacerda, 1988; Farid et al., 1990). Mercury supplies to Central America were drastically reduced during the second half of the 17th century due to exhaustion of Hg resources in Almadén and inter-

ruption of Hg supplies from Idrija in 1645 because of difficulties to pay for the Hg by the Spanish crown (Bethell, 1984). Mercury outputs from Huancavelica were also reduced, due to exhaustion of Hg resources and difficulties bringing laborers into the 'mita' system (system of drafted laborers, which the Spaniards took over from the Incas, Bethell, 1984). As a consequence, the amalgamation process was largely replaced by smelting until a new ore was discovered in Almadén in 1698, and Hg supply became abundant again (Bethell, 1984). Silver mining in Central and South America declined in the beginning of the 19th century due to disruption of the Hg supply after 1805 because of the Napoleon wars in Europe and insurrections in 1810–1811 in Central America, where more than two thirds of the American silver had been produced by then (Bethell, 1984).

Records on Hg production at Huancavelica after independence are rare. Nriagu (1994) estimated that 70 000 t of Hg were used for silver production in Central and South America between 1820 and 1900, considering that an estimated 99 400 t of silver were produced and assuming that 70% of

this silver was produced by amalgamation. Most Hg was probably imported from Spain (Table 1). Mercury was used also in gold production, especially in New Granada (Spanish viceroyalty that included present Colombia, Panama (after 1751), Ecuador, and Venezuela (Blanchard, 1989; EBO, 2001). However, Hg quantities used for gold production before the second half of the 19th century were limited, since most gold was discovered in Brazil, at that time a Portuguese colony without any Hg mines and without legal access to Spanish Hg (Blanchard, 1989).

In North America, gold deposits were discovered already in 1799 in North Carolina, but it was the discovery of gold in California in 1847 that initiated the gold rushes in North America, which culminated half a century later in Klondike in western Canada and Alaska (Nriagu and Wong, 1997). Extraction of local Hg deposits began just before 1850. The largest North American Hg mine was named New Almadén and another one New Idria after the European mines (Mineral Resources of the United States, 1883; Ganguli et al., 2000). The adverse effects on the environment because of the widespread use of Hg in gold amalgamation are still manifest in locally elevated Hg contents in soils and sediments, and possibly in regionally elevated Hg levels in fish leading to restrictions on fish consumption in several regions of the USA (Nriagu and Wong, 1997; Alpers and Hunerlach, 2000; Ganguli et al., 2000; USGS Mineral Commodity Summaries/Minerals Yearbook, Mercury, 2002). Nriagu (1994) estimated that 61 400 t of Hg were dispersed in the North American environment between 1850 and 1900. Cyanide leaching was introduced approximately 1880 (Mineral Resources of the United States, 1883) and became widely used in approximately 1905 (Nriagu, 1994), but a substantial use of Hg in gold mining continued until 1920, as recorded in sediment profiles (Pirrone et al., 1998). Amalgamation still takes place in Western Canada and Alaska, although on a small scale (Callahan et al., 1994; Lacerda, 1997). Although ores rich in Hg were present and the production and use of Hg was quite intense in North America for a century and a half, only 15% of the global Hg production has been mined there (Table 1).

An increased gold price during the 1980s boosted a new gold mining boom using Hg for amalgamation. This time the boom took place in the Amazon, Central Africa and South East Asia. An estimated 2000 t of Hg have been dispersed in the Amazon alone (Barbosa et al., 1995) during this latest gold rush, which still continues although at reduced scale. Large but not quantified amounts of Hg have also been used for gold extraction in China, Mongolia and Russia, especially Siberia, using Hg from local mines (He et al., 2000; Laperdina et al., 1996; Tumenbayar, pers. comm. 2001, Eco-Minex International Co., Ltd. Mongolia, E-mail: tumenba@magicnet.mn). Amalgamation has officially been banned in China since 1985 (He et al., 2000) and in Russia since 1990 (Laperdina et al., 1996). In spite of the bans, Hg consumption for gold amalgamation in China was estimated at 80 t in 1995, of which up to 75% may have been recycled (He et al., 2000), while Sukhenko and Vasiliev (1996) report 30 t of annual Hg emissions from amalgamation in Siberia in the beginning of the 1990s. Nine tons of Hg emissions from the amalgamation process were officially reported in the former USSR for 1988 (Laperdina et al., 1996).

Africa has large Hg reserves in Algeria, but less than 2% of globally mined Hg has been produced there thus far, including smaller quantities in Tunisia and South Africa (Table 1). Although some African quantities may not have been reported, the outputs from Africa are small because infrastructure is poor, mining temporarily affected by war conditions, and geographical location off the main markets. India has no reported production of Hg. This is mainly because of its location outside of the mercuriferous belt, and consequently, only limited extractable resources. Quantities of Hg possibly extracted and used in local gold amalgamation are limited, if any at all (Ghose, 1994). However, India is nowadays one of the major consumers of Hg (Hylander, 2001), importing large quantities for use in chlor-alkali and thermometer factories with subsequent large emissions (Corpwatch, 2001).

The total global production of primary Hg has been approximately 2000 t annually in recent years (Table 1). This is a dramatic decline since World

War II and the 1960s and 1970s, when production peaked at approximately 10 000 t per year (Fig. 1), mainly because of intense industrial use.

3.2. Industrial and related usage of mercury, and mercury-free alternatives

Global Hg production increased markedly in the 20th century, due to industrial use other than for gold and silver mining. One of the main consumers was and still is the chlor-alkali industry, where Hg cells are used for production of chlorine and sodium hydroxide by electrolysis of a brine solution. In 1996, approximately 1344 t of Hg, 40% of all Hg produced, was consumed by the world's chlor-alkali industry (Sznopek and Goonan, 2000). The chlor-alkali industry in the USA still accounts for 35% of the domestic Hg consumption, approximately 136 t per year 1996 and close to 50% in 2000 (Sznopek and Goonan, 2000; USGS Mineral Commodity Summaries/Minerals Yearbook, Mercury, 2002). Most of the Hg cells are located in Europe. Chlor-alkali plants in Western Europe have reduced their Hg consumption markedly during recent years but are still consuming approximately 100 t per year according to Euro Chlor (1999). It is not clear whether the apparent consumption is kept low, due to recycling of Hg from decommissioned Hg cells. While 60% of the chlorine and alkali production in Europe is done with Hg cells, the corresponding figure for the USA is 13%, and Japan has been producing all chlorine and alkali with Hg-free technologies for the past 15 years (USGS Mineral Commodity Summaries/Minerals Yearbook, Mercury, 2002; Euro Chlor, 1999). In the PARCOM Decision 90/3 of 14 June 1990, on 'Reducing Atmospheric Emissions from Existing Chlor-Alkali Plants', contracting parties agreed to phase out Hg-cell chlor-alkali plants, stating that 'the objective is that they should be phased out completely by 2010'. It may socially, as well as economically, be feasible to convert all chlor-alkali factories in EU to Hg-free technology already by 2007 (Cadiou and Sørup, 2001). Converting from Hg cells to the membrane technology, the best available alternative, results in several advantages including eliminated emissions of mercury and lower energy consumption (IPPC, 2001). Approx-

imately 12 000 t of metallic Hg is currently in use as electrodes in West European chlor-alkali plants (Lindley, 1997). Considerable amounts of Hg are apparently lost or stored in waste in and around the chlor-alkali plants, since the fate of nine tenths of Hg bought by West European chlor-alkali plants is not reported (Euro Chlor, 1999).

The disposal of this Hg after conversion to Hg-free technology needs to be investigated to reduce negative effects to health and environment in the future. In order to remove Hg from the society and to minimize emissions to the environment, an expert commission has proposed to the Swedish government that the growing stockpile of metallic Hg and Hg-containing wastes in Sweden be permanently stored in a deep rock repository in Sweden (SOU, 2001). An abandoned mine with suitable geological and hydrological characteristics may be an acceptable repository (SOU, 2001). One possibility is to return Hg waste from European chlor-alkali factories back to its main origin, the mines of Almadén. In this way, the flow of Hg would be reversed in the current millennium, approximately 500 years after anthropogenic activities initiated the large-scale release of Hg from the earth's crust. This would be a significant decision symbolically as well as environmentally.

The war industry and factories producing explosives were previously other large consumers of Hg, since Hg fulminate [$Hg(ONC)_2$] was used as detonators in bombs and other explosives, including firecrackers. This use contributed to the large production of Hg during World War I and II and in the 1970s for the Vietnam War (Fig. 1). Another part of the US National Defence stockpile of Hg, presently making up 4435 t, was intended for production of hydrogen bombs (USGS Mineral Commodity Summaries/Minerals Yearbook, Mercury, 2002).

Industrial use of Hg has decreased during the latest decades due to increased awareness about the toxicity of Hg. In practically all applications, Hg can be substituted by less harmful elements or alternative techniques. Mercury in thermometers and other measuring devices can be replaced by ethanol and other liquids or by digital thermometers and electronic measuring devices, respectively (Rein and Hylander, 2000). Mercury-containing

electric switches in cars, industry, and building constructions can be replaced by mechanical or electronic ones. Alternatives to Hg-containing alkaline dry cells and Hg-zinc batteries include zinc-air dry cells and lithium button cells available (USGS Mineral Commodity Summaries/Minerals Yearbook, Mercury, 2002). Nickel–cadmium dry cells are also Hg-free, but should be avoided because of the content of cadmium. Fluorescent tubes with reduced Hg content are on the market and Hg-free white bulbs are currently being developed and put into production (LightLab, 2002). In the past, organic Hg compounds were widely used as fungicides added to seeds, paper pulp and wall paints, but this is now prohibited in many countries. Mercury has long since been abandoned in mirror production (Svidén and Jonsson, 2001). The use of Hg for gilded roofs could be halted by gluing the gold to the copper plates with varnish, and by the electroplating method or by using a thicker gold layer when gilding irregular wares, such as candleholders (Svidén and Jonsson, 2001). Mercury has been used in medicine for centuries, e.g. for treatment of syphilis, and is still used as a preservative for vaccines (Svidén and Jonsson, 2001). The use of Hg in dentistry continues to result in significant environmental emissions in many countries (Hylander, 2001; Rein and Hylander, 2000; Munthe et al., 2001). Composite resins, glass ionomer cements, ceramics and gold, are substitutes to amalgam in dentistry. Consequently, there is an agreement to not use amalgam fillings for children and youth (<20 years) in Sweden. Despite the absence of laws restricting amalgam use for dental fillings in Japan, Hg amalgams were used in only 3.7% of the 70 million fillings in 1999 (Welfare Statistical Society, 2001).

3.3. Mines producing mercury at present

Production of Hg in Europe and North America has decreased during the past decades, accordingly preceding the peak and decline in atmospheric emissions (Pirrone et al., 1996). The last primary Hg mine in the USA, the McDermitt mine, closed in 1990 (USGS Mineral Commodity Summaries/ Minerals Yearbook, Mercury, 2002) and Idrija closed in 1995 (Cigale, 1997). The only primary Hg mine still in operation in the West is situated in Almadén, Spain. It is run by Minas de Almadén y Arrayanes S.A. (MAYASA), a state-owned company. The production was more than 1000 t of Hg per year in 1995 and 1996, but was reduced to approximately 400 t of Hg per year during 1997– 2000, mainly due to the declining demand for Hg (Table 1, Fig. 1). In addition, Hg is extracted as a by-product at mines with other metals as the primary product. For example, Outokumpu Mining Oyj in Finland dominated production in central and northern Europe with 40–90 t of Hg per year in 1994–2000 (Table 1, Kuo, 2001). Extraction of Hg as a by-product of gold production is estimated to 15 t per year in the USA during recent years (Table 1; Metallgesellschaft, 1939–1998; Roskill's, 2000). The Yanacocha Gold Mine in Peru produced 48 t of Hg in 2000 and has reported reserves for two more decades of operation (UNEP Chemicals, 2002).

Besides Almadén, the most important Hg mine in the world is currently the Khaydarkan mining complex in Kyrgyzstan. The reported Hg production of Kyrgyzstan has been approximately 600 t per year during 1996–1999, while Russia, Tajikistan and Ukraine produced approximately 50, 35 and 20 t per year, respectively, taken together approximately 700 t per year in the former USSR (Table 1).

Quantities of Hg produced in the other two main producing countries, Algeria and China, are uncertain. Algeria produced approximately 200–500 t per year for the latest decade (Table 1). China produced an estimated 835 t of Hg in 1997 and the production for 1999 and 2000 is estimated to be 200 t (Table 1), which may be underestimated (see Section 3.4). Mercury is also produced in North Mongolia (Tumenbayar, 2001, pers. comm.) and many other countries, but quantities have not been reported and are probably minor.

3.4. Relationship between reported and mined quantities

Historically, Hg has been an exclusive element. Accordingly, Hg production was carefully registered and its further handling was accompanied by rigorous rules to avoid thefts or other losses (Lang,

1994). For several centuries, Spanish Hg was mainly used within the empire and reporting routines were well established. Consequently, reported quantities for Spain and Spanish America for the colonial era have a high degree of accuracy. Quantities reported for other parts of Europe and America during the industrial period are also expected to agree with quantities actually mined. Deviations might have been caused by incorrect conversion of weight units, or quantities forgotten or not reported for other reasons, e.g. for Algeria, Chile, Czechoslovakia and Germany during some years of World War II. Average values given for the years reported before and after these missing data are a conservative estimate, since Fig. 1 indicates that the production of Hg peaked during World War II.

Our compilation suggests that only 5–6% of registered Hg production originates in Asia (mainly China), excluding the former USSR countries, and slightly more than 8% in the former USSR countries (Table 1). It should be noted, however, that the Asian figures may be largely underestimated, since much information is missing in the historic databases, and registered figures often represent exported quantities, which do not include domestic consumption in China, Russia and neighbouring producer countries. Also, present day information from China is uncertain, and numbers are estimates rather than measured quantities, partly because Hg is produced at several places, sometimes illegally. Estimates also vary considerably between different sources for certain years (Table 1).

In Russia, Hg has been widely used for gilding roofs, (e.g. churches) and in gold mining, especially in Siberia (Laperdina et al., 1996). However, as in the case in Europe, the largest consumers of Hg are the chlor-alkali factories. In the early 1990s, the chlor-alkali factories in Siberia alone were emitting more than 200 t of Hg per year, while the gold mines dispersed approximately 30 t per year (Sukhenko and Vasiliev, 1996). The quantities of mined Hg reported in Table 1 are insufficient to cover the requirements of chemical and war industries in the former USSR.

In conclusion, reported quantities should be regarded as a lower limit for mined Hg, while actual quantities may be considerably larger. Taking into consideration that rich Hg ores have been found in some Asian countries and that Hg has been used in many of the Asian countries for centuries, a reasonable assumption is that the Asian (including former USSR countries) figures should be doubled or tripled.

3.5. Contribution of mined Hg to global mercury emissions

Global efforts are currently made to assess and control Hg emissions to the environment, including the quantification of atmospheric emissions and their fate worldwide (MSC-E, 2001; Pacyna and Pacyna, 2002; Lamborg et al., 2002; UNEP Chemicals, 2002). Losses during as well as after mining should be included in emission inventories to determine the total quantities of Hg at risk of entering the biological cycle. Measurements and estimates at Almadén (Ferrara et al., 1998) suggest that up to 10 t per year may be lost to the atmosphere during roasting operations at a production of up to 1500 t of Hg per year. This exceeds the total emissions of many European countries (Pacyna et al., 2001) and yields a direct emission factor on the order of 0.5–1%. Adopting this emission factor, the recent global atmospheric Hg emissions from Hg mines can be estimated to approximately 10–30 t per year, and the accumulated historic emissions at roughly 10 000 t. This includes emissions only to the atmosphere and implies using present-day industrial technology, although emission factors were most likely higher than 1% in old times. Many Hg mines in the USA had condenser losses as high as 40% of the input before 1900 (Jasinski, 1994). In 1918, the losses were reduced to 5% at two mines studied and a corresponding reduction of emissions had been accomplished throughout the American Hg mines around 1930 (Jasinski, 1994). Mercury condenser losses at the last primary Hg mine in operation in the USA were less than 1% (Jasinski, 1994), which is in parity with present emissions at Almadén (Ferrara et al., 1998).

Losses to air, soil, and water during the lifetime of the Idrija mine, one of the major Hg mines in the world, have been estimated to 40% of the Hg

production reportedly sold (Miklavčič, 1998). In the period of 1968–1972, 7–10 t of Hg per year was emitted to the atmosphere from the mine and roaster facility in Idrija (Dizdarevič, 2001). Atmospheric Hg emissions from Hg mines are suggested as a possible source of contamination occurring already before 1800 in Europe (Martínez-Cortizas et al., 1999) as well as in the Arctic (Bindler et al., 2001), but have not been reported in previous historic (Nriagu, 1994; Pirrone et al., 1998) or present-day Hg emission inventories (Pacyna et al., 2001; Pirrone et al., 1996, 2001).

Current anthropogenic Hg emissions to the global atmosphere are estimated to approximately 2000 t per year (Pacyna and Pacyna, 2001, 2002). Coal combustion is currently the main source of these emissions, contributing between 750 (Pirrone et al., 1996) and 1500 t per year (Pacyna and Pacyna, 2002), whereby estimates for China alone range between 200 (Wang et al., 2000) and 500 t per year (Pacyna and Pacyna, 2001). Worldwide, even larger amounts of Hg are mobilized through mining (Table 1) and may ultimately be transformed into biologically available species. Part of the mined Hg is lost to the atmosphere at waste incineration. Pacyna and Pacyna (2002) reported approximately 100 t of Hg emissions from waste incineration in 1995. However, this is stated to be largely underestimated and may be five times larger, since only few countries report quantities of waste incinerated, and even fewer measure Hg emissions from waste incinerators. Even less is known about Hg emissions from wastes disposed in landfills, where atmospheric emissions are enhanced by intentional or unintentional open fires. Other sources of atmospheric Hg emissions are gold amalgamation, which possibly still contributed 300–325 t of Hg in 1995 (Lacerda, 1997; Pacyna and Pacyna, 2001), and emissions of mined Hg from chlor-alkali and other industries contributing approximately 100–200 t per year (cf. Pacyna and Pacyna, 2002). Taken together, atmospheric emissions of mined Hg may be of similar magnitude as the Hg emissions from coal combustion and may account for more than one third of current global Hg emissions to the atmosphere.

The global environmental impact of all historically mined Hg (1 Mton) can be estimated by comparison with historic atmospheric deposition of anthropogenic emissions. The natural Hg deposition is approximately 2–3 μg m^{-2} $year^{-1}$, based on a reevaluation of natural archives (Meili, 1995) as well as global modelling (Lamborg et al., 2002). A steady increase during approximately one century to a threefold deposition (ibid.) on the earth (5×10^{14} m^2) suggests a total historic deposition of Hg from anthropogenic sources of approximately 0.1–0.2 Mtons, not accounting for deposits on land and emissions to waters. If one third of this deposition has originated from mined Hg, this accounts for only a few percent of all mined Hg. This implies that a minor but significant fraction of mined Hg has escaped from the technosphere and contaminated the global environment. This also implies that mined Hg is likely to contribute to future emissions unless it is immobilized in natural and man-made deposits.

4. Conclusions and prospects

Roughly half of all Hg mined globally during history has been used for mining of gold and silver, although the annual production of Hg peaked during the 20th century as a result of industrial and other uses (Fig. 1). While production has changed dramatically over time and among mines, the global production of primary Hg has always been dominant in the region of the mercuriferous belt between the western Mediterranean and central Asia, but dominance appears now to be shifting from the western to the eastern part of this belt (Table 1).

Since 1970, reported and estimated primary production of Hg has been reduced by almost one order of magnitude, but may still account for approximately one third of the global anthropogenic Hg emissions to the atmosphere. The production and use of Hg can be further reduced, and probably eliminated, since viable Hg-free alternatives exist for most current Hg applications. Termination of Hg mining will reduce associated direct and indirect emissions to the atmosphere and biosphere.

There is a large stock of Hg in products and processes in use in the technosphere, including

tens of thousands of tons of metallic Hg used in chlor-alkali plants. Transformation to Hg-free technology must include a suitable plan of how to dispose off this stock without risk of export to countries with inadequate legislation and practises resulting in emissions. A better alternative is to return excess Hg back to its main origin, the mines of Almadén or Idrija, for final disposal. In this way, the flow of Hg would be reversed with the turn of the millennium, approximately 500 years after its entry as a significant global pollutant.

Acknowledgments

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