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Anthropogenic mercury emissions to the atmosphere in Brazil: The impact of gold mining

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Abstract

Emissions of Hg to the atmosphere in Brazil were estimated using consume and production parameters for each sector and technologies actually operating in the country. However, whenever no data were available, international parameters were used. Total estimated emissions reach approximately 116 $t \cdot yr^{-1}$. Chlor-alkali production presently emits only 12 $t \cdot yr^{-1}$ of Hg (10.1% of the total), although it had been the major source of Hg to the environment in Brazil up to the 1970's. Paint and dye industry, electro–electronics manufacturers, and energy production from fossil fuel and biomass burning, respond to less than 0.5% of the total input. Pyrometallurgy of Pb, Zn and Cd mainly, responds with 3.9% of the total with 4.6 $t \cdot yr^{-1}$, whereas steel and iron production may emit 12 $t \cdot yr^{-1}$ (10.4%). Burning of natural vegetation to produce agriculture lands or pastures may represent an important diffuse source of Hg to the atmosphere in Brazil and may reach 8.7 $t \cdot yr^{-1}$, (7.5%). Gold mining is presently the major source of Hg to the atmosphere in the country with an average input of 77.9 $t \cdot yr^{-1}$ (67.3%). Mercury emissions from gold mining are practically restricted to the Amazon region which results in a very high emisson per area of approximately 16 $g \cdot km^{-2}$, and atmospheric deposition rates larger than in industrial areas.

Keywords: mercury; atmospheric emission; industry; mining; Brazil

1. Introduction

Mercury emissions to the atmosphere are from natural sources; such as wind-borne soil particles, sea spray, volcanoes, forest fires, and degassing from soil and water surfaces; and from various anthropogenic sources. The anthropogenic emissions are either in concentrated local discharges as industrial effluent and waste disposal, or as diffuse discharges associated to energy production and waste incineration. Natural emissions of Hg to the atmosphere reaches ca. 2,500 $t \cdot yr^{-1}$, whereas anthropogenic emissions may reach up to 3,500 $t \cdot yr^{-1}$ (Nriagu and Pacyna, 1988). These estimates however are from data bases of industrialized countries, implicitly assuming small inputs from developing countries (Lacerda et al., 1995).

Diverse chemical forms of Hg are released into the atmosphere from anthropogenic sources. The dominant species is Hg^0 (vapor) (Pacyna and Münch, 1991), other important species, depending on source type, are Hg^{2+}, Hg-particle and HgO (Hall et al., 1991; Marins and Tonietto, 1995). Methylmercury, the most toxic relevant Hg species is not present in the atmospheric emission, and can only be produced by bacterial activity on Hg^{2+}. However, since atmospheric chemical reactions can convert nearly any form of Hg into the Hg^{2+}, all Hg releases to the

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atmosphere can eventually supply the bacterial methylation process with a continuous source of Hg^{2+} (Mason et al., 1994).

Local releases of Hg to the environment have been related to harm to human health. Although atmospheric emissions have not been directly implicated as a human health concern, fish from remote areas, which receive Hg solely from atmospheric deposition, show elevated Hg levels. Since levels of Hg that are toxic to man are not far above background levels, a moderate increase in the amount of Hg in the global atmospheric circulation could enhance exposure of man to Hg on a wide geographical scale, at least for populations consuming large quantities of fish, bringing intakes to near toxic levels for some individuals. Detailed studies of the Hg atmospheric emissions are therefore important to assess the consequences of changes in anthropogenic emission rates (Lindqvist et al., 1991).

Nowadays, point sources of Hg to the environment are being effectively controlled and Hg release to the environment from these sources is being controlled. Diffuse discharges however, have increased with increasing combustion of fossil fuels, and more recently, with the increase in Hg utilization in gold mining in tropical countries, which can contribute with 500 to 600 $t \cdot yr^{-1}$ of Hg to the atmosphere and have not been considered in previous data bases (Lacerda, 1997). Therefore, data bases including developing countries should be produced urgently in order to continuously update the global data base on Hg emissions to the atmosphere and its potential risk for ecosystem and human health.

2. Database generation

To estimate total inputs to the atmosphere we created a database established by using *Emission Factors* (EF's) from each sector of Hg sources. Most EF's were based on the technological processes actually occurring in Brazil. Therefore emission parameters from chlorine, alkali, electric–electronic and paint and dye industries, were calculated specifically for the Brazilian situation and may not be applied indiscriminately elsewhere (Ferreira and Appel, 1991; Laborão, 1991). Mercury emission from coal combustion were calculated using the composition of Brazilian coals, relatively poor in Hg, and the production process in Brazilian energy plants (MME, 1992; Martins and Zanella, 1990). On the other hand, Hg emission from oil-fired plants were estimated based on emission factors from the international literature (Chu and Porcella, 1995), since Brazilian oil-fired plants and the oil burnt in them are similar to other countries. Industrial fuel–oil combustion was not included in the data base, since its use has been reduced due to substitution by electricity and natural gas combustion.

The input from iron and steel production were calculated using a world average emission factor (Annema et al., 1993) and a relatively high emission factor from Hutton and Symon (1986), since scrap metal is being extensively used in production plants. Emissions from pyrometallurgical processes were calculated using international EF's (Nriagu and Pacyna, 1988; Pacyna and Münch, 1991; Pacyna, 1995), since the technological processes as well as the ore used in Brazil, do not differ significantly from other countries (DNPM, 1992). Emission parameters from biomass burning were based on biomass data based on tropical forests and other vegetation types submitted to seasonal burning, particularly in the Amazon region where approximately 11,000 km^2 are burnt every year (INPE, 1992). Mercury concentrations in the forest biomass and other tropical vegetation, recently reviewed by Lacerda (1995) was used to estimate emission factors. Annual deforestation rates due to burning followed the most recent reviews published by the Brazilian and USA space agencies (INPE, 1992).

Emission parameters from gold mining using Hg amalgamation have been intensively studied by many authors, since this source of Hg is becoming of global importance. We used here the compilation of emission parameters published by Lacerda and Salomons (1991), Pfeiffer and Lacerda (1988) and Lacerda et al. (1995). Mercury emissions from this activity is being carefully monitored in Brazil since the 1980's. Summaries of these estimates from Lacerda and Salomons (1991) and Ferreira and Appel (1991) were used in the present calculations.

Some potential Hg sources were not included in this data base. For example, urban waste incineration is a significant Hg source in many countries (Nriagu and Pacyna, 1988). However, this practice has been banned by federal law in 1977. Spontaneous combus-

tion may and do occur in landfills. This unpredictable Hg source is practically impossible to estimate. Incineration of medical wastes has also been suggested as a significant source of Hg to the atmosphere (EPA, 1993) and is a common practice in Brazilian hospitals. Unfortunately no study up to now is available on Hg fate in such wastes in Brazil and this potential source was not included in our estimate. Notwithstanding such limitations, we feel that the present data base includes the majority of Hg significant sources to the atmosphere in Brazil.

3. Results and discussion

Consume and emission parameters and the total atmospheric Hg input for the major emission sectors in Brazil are presented in Table 1. Sectors such as pharmaceutical industry, dentistry and chemical industry, although emitting Hg to the environment, have not been considered, due to their low or non existent emission to the atmosphere. Practices such as refuse and waste incineration, which are particularly important due to high emission factors, are not common in Brazil, and have also not been included. The following sectors were included in the data base:

3.1. Chlorine and alkali industry

This industry has been the major consumer of Hg in Brazil up to 1979, when it was responsible for nearly 60% of the total Hg consumed in the country (ca. 100 $t \cdot yr^{-1}$) (Ferreira and Appel, 1991). Mercury cells were the basis for the production technol-

Table 1
Mercury emissions to the atmosphere from all industrial sources and gold mining in Brazil

Sector		Consumption/production parameters	Emission parameters to atmosphere	Emission ($t\,Hg \cdot yr^{-1}$)	% of total
Chlorine industry		$25.7\ t\,Hg \cdot yr^{-1}$ [a]	45% [b]	11.65	10.1
KOH production		$125\ g\,Hg \cdot t^{-1}$ KOH [c]	45% [b]	0.37 [d]	0.3
Paint and dye industry		$34.3\ t\,Hg \cdot yr^{-1}$ [a]	1% [b]	0.34	0.3
Electric industry		$9.1\ t\,Hg \cdot yr^{-1}$ [a]	0.2% [b]	0.02	< 0.1
Coal combustion		$27.2 \times 10^9\ MJ \cdot yr^{-1}$ [e]	$0.13\ \mu g\,Hg \cdot MJ^{-1}$ [f]	0.004	
Oil combustion		$28.3 \times 10^9\ MJ \cdot yr^{-1}$ [e]	$0.07–0.6\ \mu g\,Hg \cdot MJ^{-1}$ [g]	0.01 [h]	All energy production
Wood combustion		$4.8 \times 10^6\ MJ \cdot yr^{-1}$ [e]	$0.01–0.05\ g \cdot t^{-1}$ [i]	0.12 [h]	0.1
Pyrometallurgy	Pb	$62.023\ t \cdot yr^{-1}$ [j]	$2–4\ g\,Hg \cdot t\,Pb^{-1}$ [j]	0.19 [h]	
	Zn	$163.000\ t \cdot yr^{-1}$ [j]	$8–45\ g\,Hg \cdot t\,Zn^{-1}$ [j]	4.30 [h]	3.9
	Cd	$197\ t \cdot yr^{-1}$ [j]	$8–45\ g\,Hg \cdot t\,Cd^{-1}$ [j]	0.05 [h]	
Iron and steel production		$15 \times 10^7\ t \cdot yr^{-1}$ [k]	$0.04–0.12\ g\,Hg \cdot t^{-1}$ [k]	12 [h]	10.4
Slash and burn agriculture		$11.100\ km^2 \cdot yr^{-1}$ [l]	$7.8\ g\,Hg \cdot ha^{-1}$ [m]	8.7 [m]	7.5
Gold mining		$87\ t\,Au \cdot yr^{-1}$ [n]	$0.92\ t\,Hg \cdot t\,Au^{-1}$ [o]	77.9	67.3
Total		–	–	115.7	100

[a] Total consumption of mercury in sector production in 1989 after Ferreira and Appel (1991).
[b] Percentage of mercury consumed emitted to the atmosphere from sector after Bezerra (1991).
[c] Consume factor of sector after Bezerra (1991).
[d] Based on an annual KOH production of 6,500 tons after Bezerra (1991).
[e] Production figures from MME (1992).
[f] Emission factors for mercury from coal combustion based on Candiota Power plant (Martins and Zanella, 1990).
[g] Emission factors from Chu and Porcella (1995).
[h] Average values.
[i] Range of mercury concentrations in tropical wood assuming 100% loss during burning after Lacerda (1995).
[j] Production figures from DNPM (1992) and emission factors from Nriagu and Pacyna (1988).
[k] Iron and steel production from World Resources Institute (1992); emission factors from Hutton and Symon (1986) and Annema et al. (1993).
[l] Deforestation rates after INPE (1992) and Lacerda (1995).
[m] Emission factor and total mercury input from slash-and-burn agriculture after Lacerda (1995).
[n] Average gold production using mercury amalgamation (1985–1991) after Lacerda and Salomons (1991) and Ferreira and Appel (1991).
[o] Emission factors from Pfeiffer and Lacerda (1988) and Lacerda et al. (1995).

ogy, responding for nearly 98% of all producing plants (Bezerra, 1991). From 1980 onward however, strictly emission control policies and improving or substituting of technologies, resulted in a decrease of Hg consume in this sector to less than 26 $t \cdot yr^{-1}$ (Ferreira and Appel, 1991). Presently, Hg cells respond to less than 36% of chlorine production in Brazil (Bezerra, 1991). Using emission and consume factors for the sector in Brazil, chlorine and alkali production emits approximately 12 $t \cdot yr^{-1}$ of Hg to the atmosphere in the country, corresponding to 10.1% of the total Hg atmospheric emission.

3.2. Paint and dye industry and electro–electronics

These sectors although presenting a small, but continuous increase in Hg consume in Brazil, are still only minor contributors to the direct atmospheric emission. Together they respond to less than 0.5% of the total emission, from a yearly input of less than 0.5 tons. Major emissions from these sectors are to soils, waters and waste deposits, through disposal of fluorescent bulbs and batteries. Some emission of Hg to the atmosphere may occur through degassing from solid waste disposal sites, but no attempt have yet been made to estimate this source. Since 1977 no industrial waste is allowed to be incinerated in Brazil, decreasing significantly its importance as a source of trace metals to the atmosphere (Barcellos and Lacerda, 1994).

3.3. Energy generation

Energy generation from fossil fuel burning is a major source of Hg to the global atmosphere (Pacyna, 1995). In Brazil however, this source of energy is of minor importance, since over 93% of the total energy production in the country come from hydroelectric power plants (MME, 1992). Coal combustion, the most important Hg source in Europe (Pacyna, 1995), is insignificant in Brazil. It is restricted to a few plants in the South, where coal deposits are abundant. Also Hg content of Brazilian coals is very low, resulting in small EF's from those plants (Martins and Zanella, 1990). As a result, Hg emission to the atmosphere from coal-fired plants is very low and contributes to less than 0.1% of the total emission.

Oil burning is particularly important as energy generation process in the northern region of the country where over 27 small to medium size plants are responsible for the production of over 70% of the region's electricity. Oil burning is however, of minor importance due to the very low emission factors to the atmosphere. In fact, Hg emission factors from this sector has only recently been published due to the analytical difficulties in determining the very low Hg concentrations in oil and in fuel gases. Chu and Porcella (1995), recently reviewed U.S.A. figures and reached emission factors ranging from 0.07 to 0.6 $\mu gHg \cdot MJ^{-1}$. Since the technological processes are very similar in the two countries, we used the same emission factors, reaching a total input of approximately 0.01 $t \cdot yr^{-1}$ ($< 0.1\%$ of the total).

Finally, biomass burning, although involving only small amounts of wood (ca. 500,000 $t \cdot yr^{-1}$) compared to oil consumption (MME, 1992), has a relatively high emission factor, resulting in a Hg emission ten times higher than oil or coal combustion (0.12 $t \cdot yr^{-1}$). Even combining all energy generation processes, Hg inputs from this source is very small, reaching less than 1% of the total.

3.4. Pyrometallurgy

Pyrometallurgy of Pb, Zn and Cd is the fourth most important contributor to the atmospheric Hg emission in Brazil. The sector responds to nearly 4.0% of the total emission with 4.6 $t \cdot yr^{-1}$. The importance of this sector is likely to augment in future years due to the new cycle of accelerated industrial growth in the country (DNPM, 1992; CACEX, 1988).

3.5. Iron and steel production

Steel production using open furnace–electric arc is the major production process in the country. Large variability in Hg emissions is expected due to ore composition, but highest emission factors occur when scrap metal is used. Emission factors from this sector may range from 0.04 to 0.12 $g\,Hg \cdot t^{-1}$ (Hutton and Symon, 1986; Annema et al., 1993). Using an average emission factor, Hg input from iron and steel production can reach 12 $t \cdot yr^{-1}$, corresponding to 10% of the total atmospheric input.

3.6. Slash-and-burn agriculture

Burning of natural vegetation to produce agriculture lands or pastures is a major practice in Brazil, in particular in the Amazon region, where an estimated area of 11,000 km^2 is burnt annually for this purpose (INPE, 1992). Although Hg content in the tropical vegetation is very small, the large area involved and the high biomass of the forest, significantly contributes with the emission of this metal to the atmosphere (Lacerda, 1995). This sector represents the fourth most important direct emission of Hg to the atmosphere in Brazil, responding for 7.5% of the total, with an annual emission of 8.7 tons.

3.7. Gold mining

The use of Hg amalgamation as a major process of gold production in Brazil started in the late 1970's, when this process was responsible for the yearly production of less than 5 tons of gold. By 1990 this technology was producing nearly 90 tons of gold per year (Lacerda and Salomons, 1991). Emission factors of Hg to the atmosphere from gold mining are the highest ever reported for any other sector, reaching 0.92 $t Hg \cdot t Au^{-1}$ produced (Pfeiffer and Lacerda, 1988). This activity presently involves over 600,000 prospectors mostly located in the Amazon region. Therefore, it is not a surprise that gold mining using Hg amalgamation is the most important source of Hg to the atmosphere with a total annual input of 77.9 tons, responding for over 67% of the total Hg emissions to the atmosphere in Brazil. Nearly all Hg from gold mining is emitted into the Amazon region. This results in a relatively high input to area ratio of 16 $g Hg \cdot km^{-2}$, which is equivalent to the ratio reported for the entire U.S.A. industrial park of around 35 $g Hg \cdot km^{-2}$ (Fitzgerald, pers. commun., 1994). High input to area ratios are highly significant to the accumulation of Hg by the biota.

Total yearly Hg emission to the atmosphere in Brazil are in the same order of magnitude of U.S.A. industrial emissions, estimated as 303 tons (EPA, 1993) and with Europe's of 726 tons (Pacyna, 1995). As in other countries, Hg emissions from diffuse sources, such as biomass burning, are becoming more important as a Hg source than direct point sources such as chlor–alkali industry. However, whereas most worldwide data bases of Hg emissions show that energy production is the major source of Hg to the atmosphere, in Brazil this source is very minor, being replaced in importance by gold mining.

Gold mining has never been assumed in any published data base as an important contribution to the global atmospheric Hg load. However, recent reports from China, Venezuela, the Philippines, Indonesia and other important developing countries, are suggesting similar results to the Brazilian case (Lacerda et al., 1995). Therefore, present evaluations of Hg inputs to the atmosphere may be significantly underestimated. Also, millions of tons of Hg-rich tailings are left behind gold mining operations. Degassing from such tailings may be significant but has never been quantified.

Atmospheric deposition rates are a highly environmentally significant measurement, since Hg emitted to the atmosphere is eventually transformed into Hg^{2+}. The atmosphere is a major source of Hg^{2+} to terrestrial and aquatic ecosystems, at least in a re-

Table 2
Atmospheric mercury deposition from gold mining and industrial areas in Brazil, compared with pristine environments and other industrialized areas

Location	Method	Deposition rate ($\mu g \cdot m^{-2} \cdot yr^{-1}$)	Reference
Central Brazil gold mines	bulk deposition	67–151	von Tümpling et al. (1996)
Central Brazil gold mines	sediment core	90–120	Lacerda and Salomons (1991)
Alta Floresta, South Amazon gold mines	sediment core	40–210	Gomes et al. (1996)
Sepetiba Bay, SE Brazil, industrialized	bulk deposition	76	Marins et al. (1996)
Southern USA, industrialized	bulk deposition	50	Mason and Morel (1993)
Pristine continental areas between 10°N and 10°S	bulk deposition	12.7	Mason et al. (1994)
Remote Norway	sediment core	9–30	Steines and Andersson (1991)

gional scale, after atmospheric oxidation reactions (Mason et al., 1994). Table 2 compares Hg deposition rates estimated for industrial and gold mining areas in Brazil, as well as comparative data from other countries published in the literature.

Atmospheric deposition of Hg over pristine areas were estimated by Mason et al. (1994). Deposition flux between latitude 10°N and 10°S averaged 12.7 $\mu g \cdot m^{-2} \cdot yr^{-1}$. This is nearly 10 times lower than the estimate for Central Brazil gold mining sites, which range from 67 to 151 $\mu g \cdot m^{-2} \cdot yr^{-1}$ (von Tümpling et al., 1996) and 20 times lower than maximum deposition rates measured in Southern Amazon of up to 210 $\mu g \cdot m^{-2} \cdot yr^{-1}$ (Gomes et al., 1996). Notwithstanding these very high values, the existing results on Hg concentration and distribution in the Amazon atmosphere affected by gold mining are preliminary, hampering the development of a reasonable model for its behavior in such an important environmental compartment. The high deposition rates suggested by the few studies however, indicate that atmospheric Hg may be the major Hg^{2+} source for methylation in Amazon ecosystems.

Deposition rates from industrial areas are very poorly documented in Brazil. The only consistent estimate is for Sepetiba Basin, SE Brazil, a significant and typical industrial area in the State of Rio de Janeiro, harboring over 400 industries in 2,500 km^2. Atmospheric Hg deposition was estimated as 76 $\mu g \cdot m^{-2} \cdot yr^{-1}$ (Marins et al., 1996), which is similar to other industrialized areas in the world (Mason and Morel, 1993). Deposition rates from gold mining areas are similar and sometimes much higher than those in industrialized areas.

4. Conclusions

The estimates presented here show that Brazil has witnessed a shift in major Hg sources to the environment in the last two decades. The implications of this phenomenon are many. Most Brazilian legislation regarding the control of Hg emissions were made for point sources, such as chlor–alkali plants. For example, Hg is seldom included in monitoring programs for many industrial areas because they lack point sources, although in many such areas Hg contamination is clearly building up. Contrary to the industrial emissions, generally located in the populated south and eastern portions of the country, where legislation enforcement and fiscalization are stronger. On the other hand, Hg emission from gold mining occurs particularly in the complex and pristine Amazon region. This solely, creates an enormous logistic problems to assess and monitor these sources. The amount of people involved in this activity is so large and representative for the region that, control and fiscalization is rather a social-economic problem than an environmental issue alone. Finally, the ignorance of Hg cycling and fate in Amazonian ecosystems strengthens the environmental significance of this pollution process, compared to more traditional and better understood industrial processes. This may result in unexpected impacts upon the environment and on public health of region.

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