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An uncertainty estimate of global mercury emissions using the Monte Carlo technique

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Abstract. In recent years, a substantial amount of work has been done to evaluate uncertainty associated with major industrial source emissions. Yet, little has been done to assess uncertainty associated with natural source emissions. Importantly, uncertainty estimates continue to be particularly relevant in the assessment of potential regulatory options, as confidence in emissions can lead to different cost-benefit assessments. To address this problem, we employed the Monte Carlo technique to improve uncertainty estimates associated with mercury emissions from both natural and anthropogenic sources. Results demonstrate that uncertainties, as they are understood in the existing literature, are overestimated. While we are aware that a probabilistic approach like the Monte Carlo technique has certain limitations (it does not consider the accuracy of available input data, for example) it still is useful in crafting a better assessment of mercury emission uncertainty.

Key words: Uncertainty, stochastic simulation, error natural sources, anthropogenic sources

Introduction

Over the last few years, a number of newly published papers have provided revised global assessments of mercury emissions from both anthropogenic (Pacyna et al., 2010; Pirrone et al., 2010) and natural sources (Friedli et al., 2009a; Friedli et al., 2009b; Pacyna et al., 2010; Streets et al., 2009a).

These global assessments have improved information on mercury emissions from both anthropogenic sources and natural sources, provided new global and regional assessments of atmospheric mercury transport and deposition patterns, highlighted major issues related to the definition of source-receptor relationships, and provided new scenarios of future mercury emissions (AMAP/UNEP, 2008; Pirrone et al., 2009; Streets et al., 2009b; Pacyna et al., 2010; Pirrone et al.,2010).

In addition, a substantial amount of past work has been done to evaluate the uncertainty associated with major industrial sources (Lindberg et al., 2007; Pacyna et al., 2003; Streets et al., 2005; Swain et al., 2007; Wu et al., 2009; Wu et al., 2006), while little was done to assess the uncertainty associated with natural source emissions. Although estimates of current anthropogenic emissions for many other pollutants are often cited with a greater precision, a general uncertainty of ±30% for major industrial sources of mercury is widely accepted.

Uncertainties are particularly relevant in the assessment of potential regulatory options, as confidence in emissions numbers can lead to different cost-benefit assessments and, therefore, drive environmental policies at national and international levels.

To improve the uncertainty estimates associated with mercury emissions from both natural and anthropogenic sources, the Monte Carlo technique has been adopted.

Materials and Methods

The Monte Carlo technique is a practical way to evaluate uncertainty.By employing a stochastic simulation technique, the Monte Carlo technique bases its estimate on the generation of random values from specified density functions (Buslenko et al., 1966; Hammersley and Handscomb, 1979). The simulation randomly generates thousands of iterations of data,to account for the uncertainty and performance variation associated with a particular variable.

In our case, we generated anuncertainty estimate

Table 1. Upper and lower bounds of mercury emissions (Mg/yr) based on several published papers and international reports.

Natural	Avg	Min	Max	Anthropogenic	Avg	Min	Max
Atlantic Ocean	840	220	1680	Coal & oil combustion	845	810	880.1
Pacific and Indian Ocean	1700	520	3400	Non-ferrous metal production	226	141.4	310.02
Antarctic Ocean	12	4	22	Pig iron and steel production	44	43.2	45.3
Mediterranean	70	8	80	Cement production	213	189.3	235.7
Coastal waters	60	30	100	Caustic soda production	105	46.8	162.9
Lakes	96	57,7	194	Mercury production	29	8.84	50
Forest	342	103	425	Gold production	256	111.3	400.02
Tundra/Grassland/Savannah/Prairie/Chaparral	448	248,8	895	Waste disposal	111	35	187
Desert/Metalliferrous/ Non-vegetated Zones	546	302	1353	Coal bed fires	32	16	48
Agricultural areas	128	68	258	VCM	24	12	36
Evasion after MDE	200	100	300	Other	45	25.54	65
Volcanoes/Geothermal	90	60	600	Gasoline, diesel, kerosene	0.19	0.378	0.57
Biomass burning	675	435	915				
Total	**5206**	**2157**	**10222**		**1930**	**1440**	**2421**

Table 2. Key outputs of the Monte Carlo simulation

	Natural sources				Anthropogenic sources			
Error (%)	5	10	20	30	5	10	20	30
Iterations (#)	1019	255	64	28	155	39	10	4
Average (Mg/yr)	6165	6204	6162	6126	1935	1935	1925	1873
Median (Mg/yr)	6148	6130	5957	5899	1937	1929	1924	1924
Standard deviation (Mg/yr)	1029	1038	1062	1132	111	117	99	91
True error (Mg/yr)	96.67	195.07	399.14	638.47	26.64	56.24	95.18	131.90

a) b)

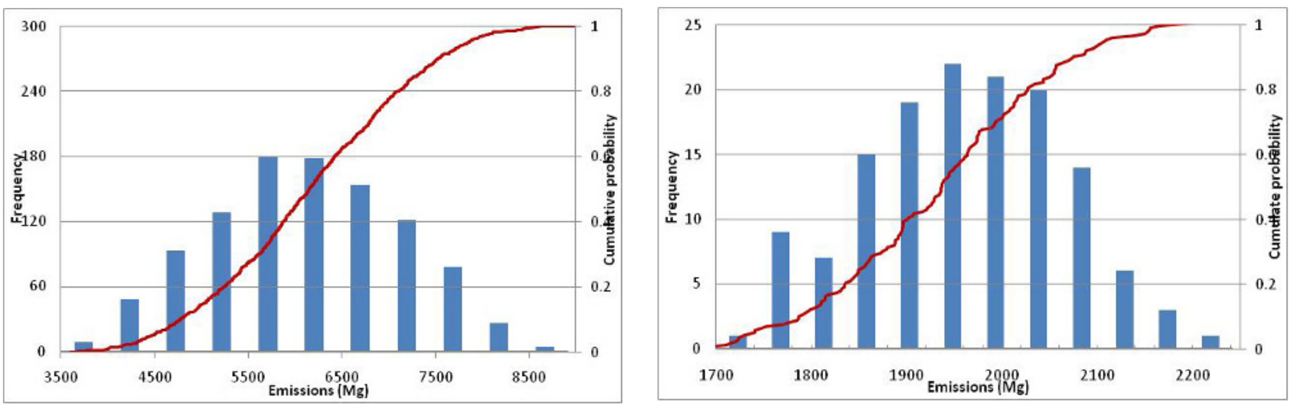


Fig. 1. Frequency and cumulative distribution curves of total simulated emissions from natural sources **a)**. and anthropogenic sources **b)**. Both figures are based on the 95% confidence interval, which led to 1019 and 138 iterations, respectively.

associated with mercury emissionsby considering the following equation for cumulative probability:

$$\Pr\left\{\left|\frac{1}{N}\sum_{N}\xi-\mu\right|<\frac{3\sigma}{\sqrt{N}}\right\}\approx 0.997$$

where N is the number of iterations, ξthe variable, μ the average and the standard deviation.

We established upper and lower bounds for both natural and anthropogenic sources on the basis of several published papers and international reports (i.e., Sunderland and Mason 2007, Mason 2009, Pirrone et al. 2009, Pacyna et al., 2010; Pirrone et al., 2010).

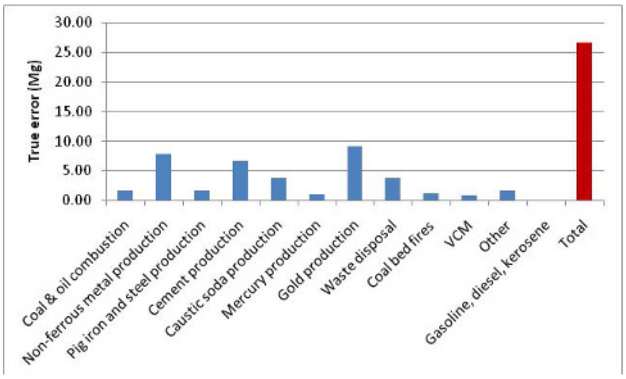
For each category, we assumed a normal distribution. Therefore, the total emissions becomes anormally-distributed random variable with a value between the minimum and the maximum.

The total error given by $\varepsilon=\frac{3\sigma}{\sqrt{N}}$ was simulated at 5, 10, 20 and 30%.

Results and Discussion

Table 1 reports the upper and lower boundaries, as well as the averages for each category, revealing a rather wide amplitude for natural sources ranging from -141% to +96%; whereas, for anthropogenic sources, the range is between -34% to +25%.

a)



b)

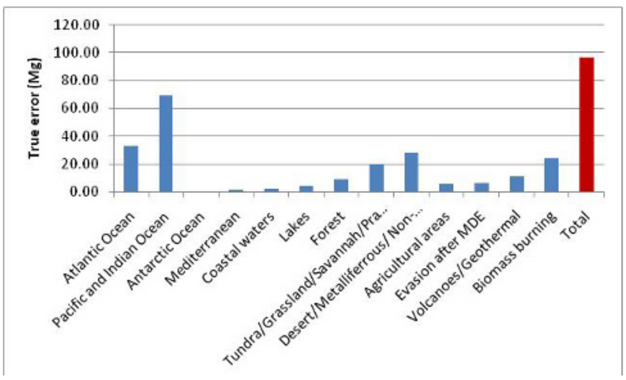


Fig. 2. True error estimated for each source category of natural **a)**, and anthropogenic **b)** emission.

Calculated without uncertainty, the emission from total natural sources averaged 5206 Mg y^{-1}; whereas it averaged 1930 Mg y^{-1} for anthropogenic sources.

Using the Monte Carlo approach with a 5% confidence interval, emissions where 6165 and 1935 Mg y^{-1} for natural and anthropogenic sources, respectively. The standard deviation was 1029 in the former, and 111 in the latter (Table 2). Results indicate a really close overlap with a Gaussian density distribution, despite the resampled mean reflecting a small bias in the resampling procedures (less than 3% in this case) (Figure 1). In addition to running a Monte Carlo simulation for total emissions, we also calculated each error source alone. With regard to anthropogenic emissions, we obtained a true error value that is low compared to the estimations by Streets et al. (2005), indicating that an overestimation of uncertainty exists prior to any consideration of statistical methods (Figure 2a). Non-ferrous smelters, cement, and gold industrial plants are associated with the largest uncertainty estimates. In addition, natural emissions have a high global uncertainty (Figure 2b). Emissions from oceans, as expected, have the largest uncertainty.

Conclusion

In the existing literature, estimations of both natural and anthropogenic emissions are associated with large uncertainties. Our simulations using the Monte Carlo technique demonstrated that, on the contrary, these uncertainties are overestimated. Though, it is important to note that this probabilistic approach does not consider the accuracy of the available input data, but is instead based on a range of values whose calculations cannot be analysed here.

For a more reliable calculation of the components of the overall uncertainty in computing mercury emissions into atmosphere, it is best to use a number of different techniques (e.g. the Ishikawa cause and effect diagram), so that one can obtain a qualitative analysis of all the factors involved.

As employed here, the Monte Carlo technique produces an estimate of overall uncertainty in calculating atmospheric mercury emissions, While its main components have been simplified,, it continues to improve our assessment of mercury emission uncertainty.

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