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Author(s)	MATSUNAGA, Katsuhiko
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## Oceanic Residence Time of Mercury

Katsuhiko MATSUNAGA\*

### Abstract

The annual flux of mercury to the oceans was estimated by using the mercury concentrations in river water, precipitation, and by estimating the amount of mercury released into the environment from chlor-alkali production. The annual fluxes from precipitation, river water and human activities are  $5 \times 10^8$  g,  $1.2 \times 10^8$  g and  $0.3 \times 10^8$  g, respectively. The annual mercury deposition rate was estimated by using the mercury concentration in marine sediments and the sedimentation rate, resulting in a flux of  $8.0 \times 10^8$  g of mercury to marine sediments. From these results the residence time of mercury in the oceans was calculated to be ca.  $1 \times 10^4$  years.

### Introduction

Klein and Goldeberg<sup>1)</sup> have estimated that human activities add  $4-5 \times 10^9$  g of mercury a year to the aquatic environment which is nearly equivalent to that added by continental weathering. Weiss et al.<sup>2)</sup> have reported that the annual mercury flux into the oceans from degassing of crustal materials was  $2.5 \times 10^{10}$ – $1.5 \times 10^{11}$  g, which is ten to one hundred times greater than from both continental weathering and human activities. They estimated the mercury flux into the aquatic environment by using values of mercury concentrations in both river water and precipitation and by using data for mercury consumption. However, the reported mercury concentrations in river water, precipitation and sea water have not been correct. One source of error is to be found in the use of polyethylene bottles used for sample storage. Matsunaga et al.<sup>3)</sup> found that nanogram amounts of mercury were leached out of the plastic into the samples, whose mercury concentrations increased by a factor of 2–100, even if the bottles had been washed out with nitric or hydrochloric acid before use.

Using all glass bottles which do not release mercury during sample storage, Matsunaga et al.<sup>4)</sup> and Matsunaga<sup>5)</sup> have reported that the mercury concentrations in the Kuroshio and Oyashio regions, the Japan sea, and in the northern North Pacific Ocean and the Bering Sea are  $5 \pm 0.5$  and  $4.2 \pm 0.4$  ng/l, respectively. Recently Mukherji and Kester<sup>6)</sup> reported that the mercury concentration in the Gulf Stream was  $4.1 \pm 1.0$  ng/l. These values are at least one twentieth smaller than those previously reported in the literature.

The value 5 ng/l will be used as the average mercury concentration in the open ocean. If the mercury of the flux was  $4 \times 10^9$  (Klein and Goldberg) or  $1.5 \times 10^{11}$  g/y (Weiss et al.), the calculated residence time of mercury becomes 40–2000

\* *Laboratory of Marine Chemistry, Faculty of Fisheries, Hokkaido University*  
(北海道大学水産学部海洋化学講座)

years. This value seems too short to explain the uniformity of the mercury concentration in sea water.

In this paper, the correct mercury flux into the oceans from rivers, precipitation and human activities was calculated and also a mercury deposition rate was estimated.

### Estimation of mercury Flux into the oceans

#### *Atmospheric flux*

Matsunaga and Goto<sup>7)</sup> have reported that the residence time of mercury in the atmosphere by measuring its concentration in air and precipitation. The concentration in precipitation was constant regardless of rainfall,  $1 \pm 0.2$  ng/l. By assuming that worldwide precipitation is 1000 mm/y (Budyko et al.<sup>8)</sup>) the atmospheric mercury flux is calculated to be  $5 \times 10^8$  g/y.

#### *River flux (Continental weathering)*

Matsunaga<sup>9,10)</sup> has reported that the average mercury concentration in Japanese river waters which do not receive waste discharge from mines or factories is  $3 \pm 2$  ng/l. The flux of river water into the oceans is  $4 \times 10^{16}$  l/y (Budyko et al.<sup>8)</sup>). Thus, the mercury flux from rivers becomes  $1.2 \times 10^8$  g/y.

#### *Anthropogenic flux*

The amount of mercury produced throughout the world is about  $1 \times 10^{10}$  g/y. A substantial loss of mercury into the environment occurs in chlor-alkali production, and about one-third of the mercury consumed annually in the United States, as well as in Japan was used in that process. The mercury loss in the process is by volatilization to the atmosphere, deposition in piping, sorption on saline mud and direct discharge into the aquatic environment. However, the amount of mercury vaporized into the atmosphere has not been well quantified, and it is not clear whether or not the mercury concentration of the atmosphere has increased. Mercury which is released into the atmosphere by chloralkali production and subsequently dissolved in precipitation is included in the estimated mercury flux from precipitation. However, the amount of mercury released into the atmosphere per year, even if all of mercury used in chlor-alkali production was vaporized into the atmosphere, is comparable to the total amount of mercury in the atmosphere,  $2.9 \times 10^9$  g (Matsunaga and Goto<sup>7)</sup>).

It is also difficult to estimate accurately the discharge of mercury into the oceans. It was reported by Nakanishi et al.<sup>11)</sup>, that the total amount of mercury lost from chlor-alkali production around Tokuyama Bay, Japan, during the period from 1952 to 1972 was 508 tons, and that the amount of mercury discharged into the bay was 14 tons, of which about 4 tons dissolved in sea water during that period, corresponding to about 1% of the total amount of mercury lost. In the world, the anthropogenic flux of mercury is calculated to be  $3 \times 10^7$  g/y.

#### *Estimation of mercury deposition rate into marine sediments*

The mercury concentration in marine sediments is scattered ranging from 0.06–0.8 ppm, however, there is little variation in the mercury concentration of

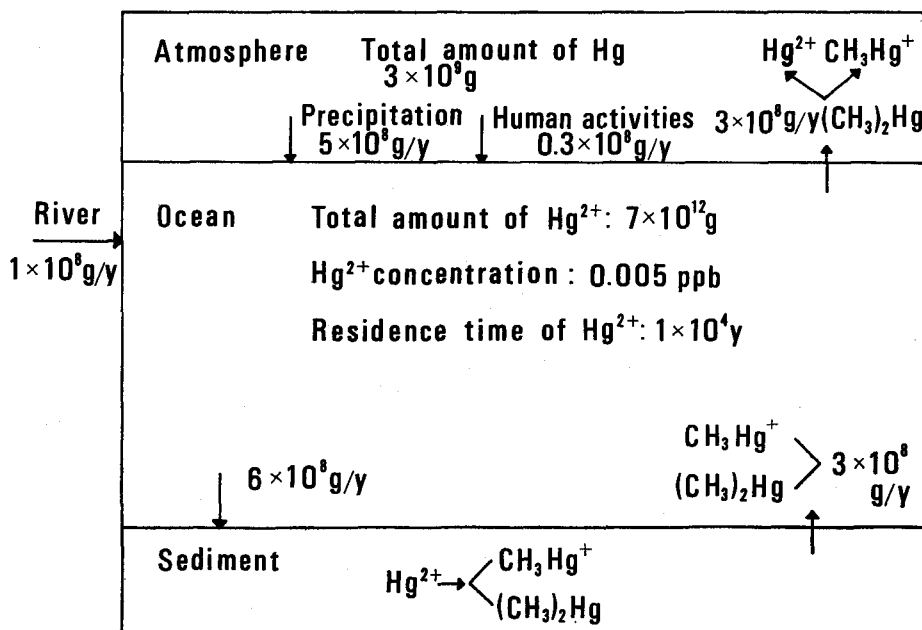


Fig. 1 Mercury budget in the oceans

pelagic sediments (0.1–0.4 ppm). Although the average mercury concentration in marine sediments has not been established, it is estimated that most deep sea sediments have mercury concentrations of about 0.2 ppm.

The average sedimentation rates in the Pacific and Atlantic Ocean are 0.3 and  $1.2 \times 10^{-3} \text{g/cm}^2$ , respectively (Turekian<sup>12</sup>). The rate in the Indian Ocean is on the order of the Pacific Ocean sedimentation rate. Although the other oceans have higher sedimentation rates than the Atlantic Ocean, the sedimentation rate in the other oceans was regarded as the order of the Atlantic Ocean. Thus, the mercury deposition rate is calculated to be  $1.4 \times 10^8 \text{g}$  in the Pacific and Indian Ocean, and  $7.2 \times 10^8 \text{g}$  per year in the Atlantic and the other oceans, respectively. The mercury deposition rate in the ocean becomes  $8.6 \times 10^8 \text{g/y}$ , which is on the same order as the estimated mercury flux into the oceans. From these results, the residence time of mercury in sea water is calculated to be ca.  $1 \times 10^4$  years.

### Results and Discussion

Mercury budget was shown in Fig. 1. Matsunaga et al.<sup>13</sup> reported that the mercury concentration in marine cores decreased from the surface to a nearly constant value below 20–50 cm depth. These results mean that mercury in marine sediments may be released into interstitial waters and eventually lost from the sediments by forming soluble mono- or dimethylmercury. It is generally assumed that methylmercury exists in natural waters and that aquatic organisms concentrate it, because it has been detected in all aquatic organisms.

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