

Exchange flux of total gaseous mercury between air and natural water surfaces in summer season

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Abstract The exchanges of mercury between surface and air are of significance in the biogeochemical cycling of Hg in the environment, but there are still few reliable data on air/surface exchange in aquatic systems. Field measurement campaigns over seawater surface at Kristineberg Marine Research Station (KMRS) and over Hovgårdsån River surface at Knobesholm in southwestern Sweden were conducted to measure mercury flux using a dynamic flux chamber technique coupled with automatic mercury vapor-phase analyzers. Both sites show net emissions during summer time. Mercury fluxes measured over both river and seawater surfaces exhibit a consistently diurnal pattern with maximum fluxes during the daytime period and minimum fluxes during the nighttime period. At freshwater site, mercury flux is strongly correlated with the intensity of net solar radiation, and negatively correlated with relative humidity. A typical exponential relationship between mercury flux and water temperature was observed at freshwater measurement site. At seawater site, a strong correlation between mercury flux and intensity of solar radiation was obtained. The driving force of mercury emission from water surface to air is the super-saturation of dissolved gaseous mercury in aqueous phase.

Keywords: flux chamber, mercury, DGM, biogeochemical cycling, exchange flux.

Due to relatively high volatility of mercury and most of its compounds, it cycles in the environment with atmosphere as the transportation medium as a result of natural and anthropogenic activities^[1]. The amount of mercury mobilized and released into the atmosphere has increased since the beginning of the industrial age^[2]. The worldwide anthropogenic mercury emissions currently were estimated to be around 4000 tons per year^[3]. Coal combustion, waste incineration, chlorine-alkali production as well as metal smelting, refining and manufacturing are major source categories. Human activities emit both elemental mercury (Hg^0) with a long life-time in the atmosphere and reactive gaseous mercury (RGM) and particulate mercury, which are shortly lived in the air^[1,2]. Mercury is also released into the atmosphere by a number of natural processes, including outgassing of the earth's mantle/crustal material, evasion from surficial soils^[4-10], water bodies (both fresh-water and salt-water)^[7, 11-15], vegetation surfaces^[16-18], wild fires, volcanoes, and geothermal sources. Natural emission of mercury includes the re-emissions of mercury that is previously deposited to the earth surface. Mercury is believed to be released from

natural sources mainly as Hg^0 vapor^[1,2]. However, there is a rather large uncertainty on the estimation of mercury emissions from natural sources due to lacking of sufficient field emission measurement data and so far a range from 2500 to 30000 tons per year has been estimated^[19].

The mercury exchange between air and water surfaces (including fresh water and sea water) is recognized to be of significance in the global biogeochemical cycling of mercury^[1,20]. Mercury emission from water surfaces (especially from sea water) is considered as one of the major atmospheric sources^[20]. On the other hand, the mercury emission reduces the mercury burden in the water and may limit the methylmercury production and accumulation in fish^[14]. This de-toxification process can thus serve to ameliorate the risks associated with mercury contamination especially in fresh water systems. So far the estimation of mercury exchange flux between air and seawater surface has been accomplished by means of computation from measurement results of dissolved gaseous mercury (DGM) concentration in seawater^[11]. It is normally difficult to obtain mercury exchange flux with high time resolution and a continuous way. For freshwater, the exchange flux of mercury between air and water surfaces can be achieved by both calculation from measurement of DGM concentration in water which is used in seawater studies^[1,4] and mostly by direct on-site measurement methods^[7,11–13,15]. Owing to limited analytical approaches, the time resolution of these direct methods were not good, and it was difficult to procure continuous measurement results in a relatively long period. In this paper, we measured mercury fluxes over seawater and fresh water surfaces by using a dynamic flux chamber method coupled with high time resolution atmospheric mercury analysis techniques in southwestern Sweden.

1 Site and methods

1.1 Site and measurement of meteorological parameters

The estate of Knobesholm, which is located near the village of Asige, 40 km east from the Swedish coastline, 120 km south of Göteborg, was chosen as the sampling site for the fresh water flux measurement site. The measurements were performed on the small river of Hovgårdsån, which is a runoff from the lake Borrsjön. The water depth at Hovgårdsån was 1 to 1.40 m. The meteorological parameters, such as air temperatures inside and outside the flux chamber, water temperature, solar irradiation and relative humidity were measured by a mini meteorological station (Skye Datahog 2).

Kristineberg Marine Research Station (KMRS) was chosen as the seawater measurement site, and it is located on the Swedish west coast at the mouth of Fjord Gullmaren, 120 km north of Göteborg. The sea measurements were performed in the coastal area of the open sea, in the basin of a hexagonal shaped platform. The platform is placed 1 km from the shore and the water depth is 9–10 m. The KMRS is equipped with the standard meteorological instruments for air temperature, water temperature, wind speed, and intensity of UV_A (320–400 nm) part of the solar radiation.

1.2 Method for flux measurement

The mercury flux measurement was conducted with a dynamic flux chamber of FEP

TeflonTM constructed at ORNL^[6,21]. Field flux chamber allows for measurement of gas flux from a small surface area and provides a sensitive means of measuring small fluxes^[22]. Because the field chamber from which gas flux is being measured is constrained, relationships with environmental variables such as temperature, light are relatively easily established. It is portable and relatively inexpensive. Xiao et al.^[12] and Schroeder et al.^[13] firstly introduced chamber method to measure mercury exchange flux between air and water and soil surfaces. It makes mercury exchange flux measurement be done in a more precise and high time resolved way possible that the development of automatic atmospheric mercury analyzers (i.e. Tekran 2537A and Gardis 1A) and of flux chambers constructed from low mercury blank materials such as Teflon and Quartz. The rectangular, open-bottom chamber (60 cm×20 cm×20 cm) is supported on an external aluminum frame. A piece of polystyrene block wrapped with Teflon sheet was fastened on each side of the chamber to make the chamber to float on the water surface. Mercury flux from the water surface exposed in the chamber was calculated using eq.(1)^[7,8]:

$$F = \frac{(C_o - C_i)}{A} \times Q, \quad (1)$$

where F is the flux of total gaseous mercury, which consists of mostly $\text{Hg}^{0[2]}$ in $\text{ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$; C_o and C_i are concentrations of Hg at the outlet and inlet of the chamber in $\text{ng} \cdot \text{m}^{-3}$, respectively; A is the surface area of bottom surface area of the chamber in m^2 (0.12 m^2); and Q is the flushing flow rate through the chamber in $\text{m}^3 \cdot \text{h}^{-1}$ ($0.45\text{--}0.47 \text{ m}^3 \cdot \text{h}^{-1}$). The inlet and outlet mercury concentrations were monitored alternately by the mercury analyzer (either Gardis or Tekran) in a time interval of 10 min. A magnetic valve was used to switch the sampling port of mercury analyzer between inlet and outlet (fig. 1). The mercury analyzers were calibrated by injection a volume of mercury saturated air with known concentration. The Teflon tubing for both the inlet and outlet measurements was slightly heated by heating bands to a temperature of $10 \text{ }^\circ\text{C}$ above ambient air temperature. Two heating fans, which were fastened at the corners of the aluminum frame, were gently heating the flux chamber during the measurement campaign to prevent water condensation on the inner wall of the flux chamber.

The blank of the chamber was measured by sealing the chamber bottom to a large, clean piece of FEP TeflonTM and the results showed that usually negligible blanks were detected.

1.3 Methods for DGM, reactive and total mercury measurement in water

500 to 2000 mL sampled water was immediately after collection transferred

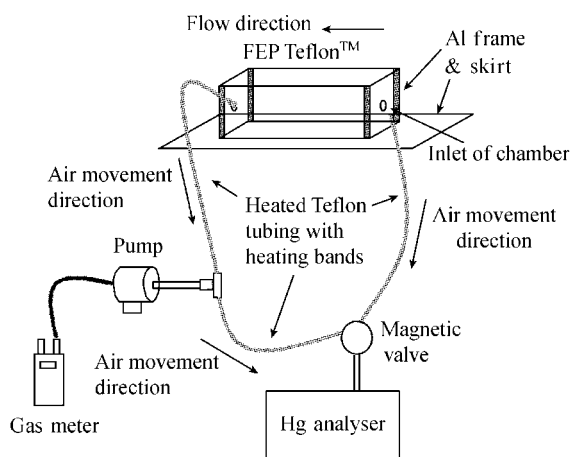


Fig. 1. The set-up of the dynamic flux chamber for measuring mercury flux over water surfaces.

into an extensively cleaned borosilicate glass impinger, and purged with mercury free argon with a flow rate of $300 \text{ mL} \cdot \text{min}^{-1}$ for 30 min and the DGM was collected on a gold trap in the field. Mercury collected on the gold traps were analyzed using dual-stage amalgamation coupled with AFS detection¹⁾. 50 mL water samples were stored in pre-cleaned Teflon bottles with addition of 0.5% HNO_3 and preserved at low temperature for reactive mercury and total mercury. Reactive mercury was analyzed by direct reduction of SnCl_2 and dual-stage amalgamation coupled with AFS detection, and total mercury was done by BrCl oxidation followed with reduction of SnCl_2 and dual-stage amalgamation coupled with AFS detection^[23].

2 Results and discussion

2.1 Measurements over river surface

Four water samples were collected from the river. The total mercury concentration in the water is $2.36 \text{ ng} \cdot \text{L}^{-1}$, and reactive Hg (Hg_r) in the water consists of 74% of total mercury (Hg_T), which is in the range of those of background natural waters^[1].

The measurement campaign of mercury flux over river surface was carried out on August 5–16, 1999. The mercury fluxes measured over the river surface with the dynamic chamber as well as the TGM concentrations are given in table 1. The average TGM concentration in the air 20 cm

Table 1 Statistical summary of the measurement results over the river surface

	Average	Median	S.D.	CV(%)	Min.	Max.	<i>n</i>
TGM/ $\text{ng} \cdot \text{m}^{-3 \text{ a)}$	2.61	2.39	1.95	74.7	0.90	7.04	792
Evasion/ $\text{ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$	11.06	6.69	11.83	106.9	0.50	88.92	788
Deposition/ $\text{ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$	1.14	0.74	0.99	86.8	0.27	2.48	4

a) TGM was measured in the air 20 cm above water surface.

above the water surface was $2.61 \text{ ng} \cdot \text{m}^{-3}$ throughout the campaign. Except for 4 occasions of

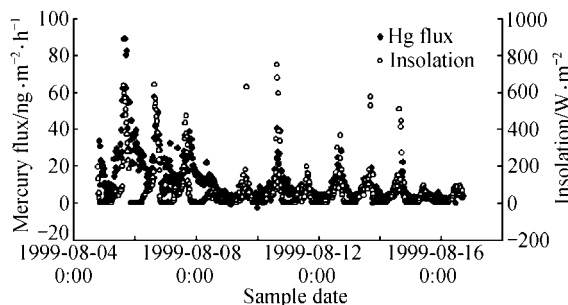


Fig. 2. Mercury flux measured over river surface at Knobesholm as well as the net solar radiation.

deposition event that occurred during night, evasion of mercury from the water surface dominated the exchange processes. A clear diurnal pattern is shown in fig. 2 with mercury evasion during the mid-day reaching the maximum followed by reduced fluxes and usually reached the minimum at the beginning of sunrise, occasionally giving deposition during the nighttime period.

The mercury fluxes tracked the intensities of solar radiation very well with a linear correlation coefficient (*r*) of 0.61. The mercury flux from the river is negatively lineally correlated with

1) Feng, X. B., Sommar, J., Lindqvist, O. et al., Occurrence, emissions and deposition of mercury during coal combustion in the province Guizhou, China, *Water Air Soil Pollut.*, in press.

relative humidity ($r = 0.67$) (fig. 3).

Four DGM measurements were carried out on August 16, 1999. The average DGM concentration was $0.56 \text{ ng} \cdot \text{L}^{-1}$, while the saturation concentration of DGM should be $9.0 \times 10^{-3} \text{ ng} \cdot \text{L}^{-1}$ with the average air Hg concentration of $2.6 \text{ ng} \cdot \text{m}^{-3}$ and Henry's Law coefficient of 0.29 for Hg^0 (at 20°C). Obviously, the water was super-saturated in terms of DGM. It is interesting to notice that GM concentration changed significantly with time while total mercury concentration in water kept stable (table 2). Noteworthy, DGM concentration in water is significantly correlated to the intensity of net solar radiation (fig. 4). Fig. 5 shows a very good correlation ($r = 0.77$) between the reciprocal absolute temperature of the river water and the $\ln \text{Hg}$ flux.

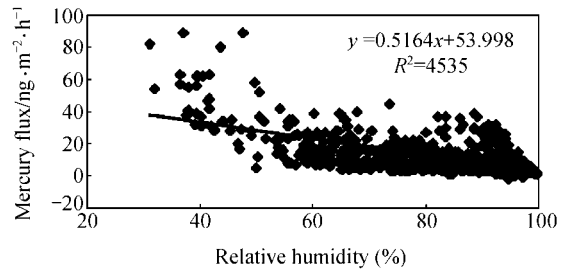


Fig. 3. Relationships between mercury flux measured over the river surface and the relative humidity.

Table 2 DGM and total mercury concentrations in water

Sampling Time	DGM/ $\text{ng} \cdot \text{L}^{-1}$	Total Hg/ $\text{ng} \cdot \text{L}^{-1}$
11:40	0.84	2.54
13:30	0.52	2.21
15:45	0.43	2.31
17:15	0.46	2.34
Average	0.56	2.35

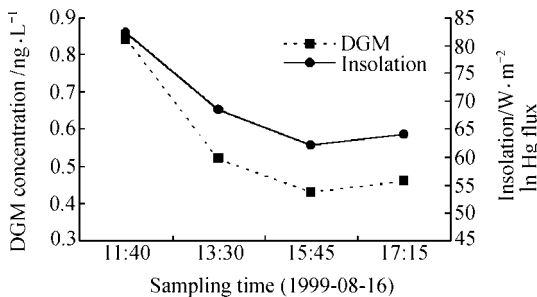


Fig. 4. DGM concentrations in river water and the intensity of net solar radiations at the sampling time. The sampling was conducted on August 16, 1999.

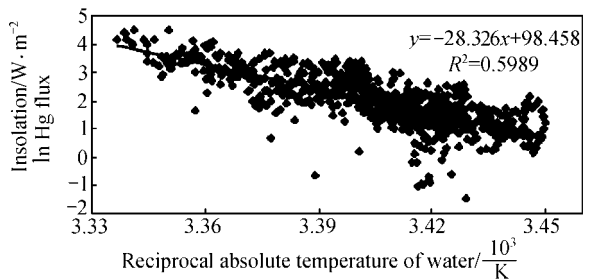


Fig. 5. The temperature dependence of mercury flux over the river surface.

During the measurement course the average mercury flux over the river surface was $+11.0 \text{ ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$, which is comparable with the average Hg flux ($+11.2 \text{ ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$) from lake Stora Låresbovatten in Sweden during summer measured by Xiao et al. [12].

2.2 Measurements over sea surface

Three flux measurement campaigns were carried out at KMRS from June 23 to July 2 of 1997, August 6–12 of 1997, and June 17–23 of 1998, respectively. For the first campaign we

also collected data of air temperature, water temperature, wind speed and UV_A intensity of solar radiation from KMRS. For the third campaign, only data on UV_A intensity of solar radiation were collected from KMRS. The average TGM in the air 20 cm above sea surface was $2.81 \text{ ng} \cdot \text{m}^{-3}$ throughout the three campaigns. The measurement results are listed in table 3. During all sampling campaigns, the water column was sampled for dissolved gaseous mercury (DGM) during daytime. DGM concentrations in water varied from 0.04 to $0.1 \text{ ng} \cdot \text{L}^{-1}$. It is obvious that during daytime seawater is supersaturated in terms of DGM since the saturated DGM concentration is about $9.7 \times 10^{-3} \text{ ng} \cdot \text{L}^{-1}$.

Table 3 Statistical summary of the measurement results over the sea surface^{a)}

		Average	Median	S.D.	CV (%)	Max.	Min.	<i>n</i>
All three campaigns	TGM ^{a)} /ng · m ⁻³	2.81	2.75	0.69	24.6	5.36	1.62	827
1997-06-23—	evasion	0.42	0.30	0.40	94.5	1.91	0	114
1997-08-06	deposition	0.35	0.16	0.37	107.2	1.20	0.01	38
1997-08-01—	evasion	1.00	0.77	0.82	82.5	3.97	0	249
1997-08-06	deposition	0.36	0.26	0.25	69.2	1.03	0.06	44
1998-06-20—	evasion	1.01	0.46	1.33	131.9	8.84	0	179
1998-06-25	deposition	0.57	0.24	0.67	117.3	2.72	0.02	55

a) TGM concentrations in the air were measured at 20 cm above sea water. Fluxes are expressed in $\text{ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$.

The measured mercury fluxes over seawater surface in all campaigns indicated evasion as well as dry deposition process (fig. 6). The maximum evasion of mercury measured during the three campaigns was $8.8 \text{ ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ whereas the maximum deposition was $2.7 \text{ ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ (table 3). The average mercury flux of $+0.61 \text{ ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ obtained during the summer season from the studies is quite similar to the literature values of mercury flux at coastal areas calculated by Costa and Liss^[24]. As observed in river measurement, evasion over seawater reached the maximum during mid-day and deposition occurred during nighttime. At the first and the third campaigns, it is clear that the mercury fluxes mimicked the UV_A intensities ($r = 0.63$ at the first campaign).

Calculation shows that there is weak linear correlation between mercury flux and air temperature ($r = 0.27$), and wind speed ($r = 0.22$). The exponential relationship between mercury flux and air temperature observed in river measurements is, however, not seen in the seawater studies.

2.3 Mechanisms for DGM production

The DGM, which is mainly in form of Hg^0 , could be formed in many processes in the water system. Demethylation^[25], bacterial reduction from water^[25] and sediment^[26], reduction by humic and fulvic acid in water^[27], and photo-induced reduction in water^[28–33] are so far the possible processes suggested to be responsible for the formation of DGM in aquatic system. Although several of these processes are well known, it was not until quite recently that photo-induced mercury reduction and subsequent volatilization became the subject of theoretical^[33], laboratory^[34]

– 37] and field^[28–31] investigations. The conclusions obtained from these studies are that certain mercury species such as reactive mercury could be photo-reduced to Hg^0 and organic matter such as humic and fulvic acids do enhance the reaction rate.

It is indicated by our measurement data that photo-induced reduction processes play a crucial role in the formation of DGM in both river and sea water system in the study areas, even though we cannot exclude the minor contributions from other processes. In the river studies, mercury flux is strongly correlated with solar radiation (fig. 2), and the DGM concentration in river water during daytime periods is also strongly correlated with solar radiation (fig. 4). The river is a runoff from the lake Borrsjön, which serves as a large mercury reservoir and stably supplies mercury to the river. That makes the total mercury concentration in river fairly stable. Although the total mercury concentration in river water is not very high (about $2.3 \text{ ng} \cdot \text{L}^{-1}$), the main portion of total mercury is in reactive forms

(74%), which are available for being involved in the photo-reduction processes. There is, therefore, still a huge reservoir in terms of mercury that are available for participating in the reduction processes in the river system. Moreover, the high concentration of organic matter (TOC concentration is up to $9.0 \text{ mg} \cdot \text{L}^{-1}$) in the river may facilitate the photo-reduction processes to a great extent. All these facts may easily explain the high DGM concentration in the river during daytime and high mercury evasion flux measured from the river surface. After sunset occurs the intensity of net solar radiation declines drastically to nothing, however, the mercury evasion flux decreases slowly to the minimum at sunset (fig. 2). Obviously there is no net production of DGM in the river system during nighttime periods. The DGM pool in the river at sunset is large enough to explain the decreasing mercury evasion flux measured during night periods.

At sea site, a clearly diurnal pattern of mercury fluxes was observed in all three campaigns, and mercury flux is significantly correlated with the UV_A intensity of the solar radiation. It is quite

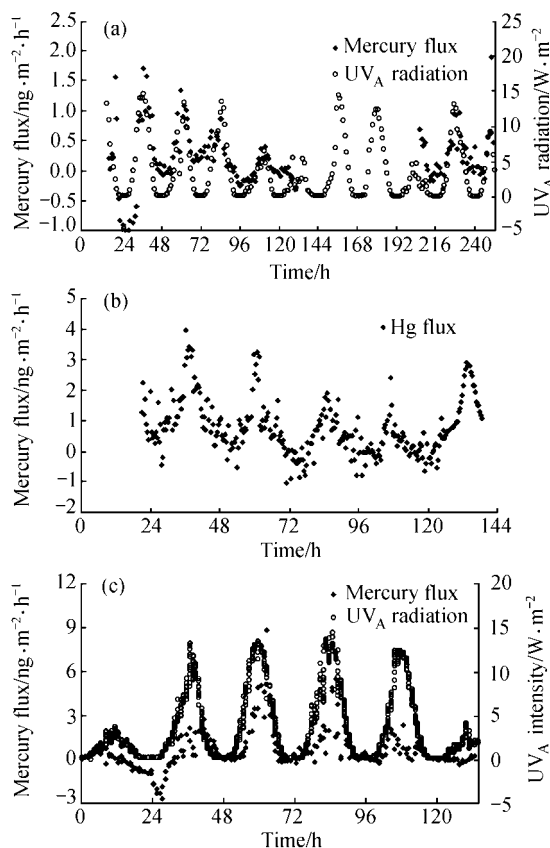


Fig. 6. Mercury flux measured over seawater surface at Kristineberg marine research station (KMRS) as well as the net solar radiation. (a) The first campaign starting on June 23, 1997; (b) the second campaign starting on August 6, 1997; (c) the third campaign starting on June 17, 1998.

reasonable to assume that the intensity of UV_A measured at any place on earth is significantly linearly correlated with the total intensity of solar radiation measured at the same site. During daytime, therefore, DGM in seawater is produced mainly by photo reduction. Studies show that during nighttime GDM in seawater will be oxidized to divalent mercury with the existence of Cl^- ^[28]. Hence, the observed diurnal pattern of mercury flux over sea surface can be explained by the formation and evaporation of DGM by solar radiation during daytime and the oxidation of DGM in the dark to Hg (II) in the presence of chloride, which may cause the decrease of mercury evasion flux and even the dry deposition of Hg to water surface due to the depletion of DGM in seawater.

3 Conclusions

(1) Mercury fluxes measured over both river and seawater surface exhibit a consistently diurnal pattern with maximum fluxes during the daytime period and minimum fluxes during the nighttime period. Both sites are net emission sources during summer. The average mercury emission rate at the river and seawater surfaces are $+11.0$ and $+0.61 \text{ ng} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$, respectively.

(2) At freshwater site, mercury flux is strongly correlated with the intensity of net solar radiation, and negatively correlated with relative humidity. A typical exponential relationship between mercury flux and water temperature was observed at freshwater measurement site. At seawater site, a strong correlation between mercury flux and intensity of solar radiation was obtained.

(3) Solar radiation plays an important part in the formation of DGM in both river and seawater. The DGM formed during daytime is depleted during night by means of evasion at the river site, and due to the high production of DGM during daytime; there is still strong evasion flux measured during nighttime. At seawater, DGM photo-reduced from Hg(II) during daytime will be depleted by the evasion and re-oxidation to Hg (II) in the presence of chloride in water. Therefore, DGM will be rapidly depleted and dry deposition process occurs frequently after sunset.

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