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Apparent rates of production and loss of dissolved gaseous mercury (DGM) in a southern reservoir lake (Tennessee, USA)

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ABSTRACT

Apparent rates of dissolved gaseous mercury (DGM) concentration changes in a southern reservoir lake (Cane Creek Lake, Cookeville, Tennessee) were investigated using the DGM data collected in a 12-month study from June 2003 to May 2004. The monthly mean apparent DGM production rates rose from January (3.2 pg L^{-1}/h), peaked in the summer months (June–August: 8.9, 8.0, 8.6 pg L^{-1}/h), and fell to the lowest in December (1.6 pg L^{-1}/h); this trend followed the monthly insolation march for both global solar radiation and UVA radiation. The monthly apparent DGM loss rates failed to show the similar trend with no consistent pattern recognizable. The spring and summer had higher seasonal mean apparent DGM production rates than the fall and winter (6.8, 9.0, 3.9, 5.0 pg L^{-1}/h, respectively), and the seasonal trend also appeared to closely follow the solar radiation variation. The seasonal apparent DGM loss featured similar rate values for the four seasons (5.5, 4.3, 3.3, and 3.9 pg L^{-1}/h for spring, summer, fall, and winter, respectively). Correlation was found of the seasonal mean apparent DGM production rate with the seasonal mean morning solar radiation ($r=0.9084$, $p<0.01$) and with the seasonal mean morning UVA radiation ($r=0.9582$, $p<0.01$). No significant correlation was found between the seasonal apparent DGM loss rate and the corresponding afternoon solar radiation ($r=0.5686$ for global radiation and 0.6098 for UVA radiation). These results suggest that DGM production in the lake engaged certain photochemical processes, either primary or secondary, but the DGM loss was probably driven by some dark processes.

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1. Introduction

Air/water exchange plays an important role in global biogeochemical cycle of mercury (Hg) (Fitzgerald et al., 1991; Schroeder and Munthe, 1998). Mercury exchange at air/water interface of an aquatic system such as a lake depends on concentration of dissolved gaseous mercury (DGM), which in turn is controlled by its photochemical redox transformation in the water body (Amyot et al., 1994; Gardfeldt et al., 2001; Nriagu, 1994; Zhang, 2006; Zhang and Lindberg, 2001). To understand the photochemodynamics of freshwater Hg, one may quantify Hg evasion flux and determine DGM production separately in controlled simulation systems. Alternatively, one can gain inference by inspecting in situ temporal change of DGM concentration (O'Driscoll et al., 2003). This registers both Hg evasion and redox transformation in a lake, serving as a comprehensive index in a sense. The in situ DGM concentration change rates are valuable particularly for modeling aquatic Hg photochemodynamic cycle and verifying the models. The rate information can also provide useful insights into the mechanisms of aquatic photochemical redox cycle of Hg, especially the chemical nature of daily and seasonal rise and fall of DGM levels in a lake (e.g., Dill et al., 2006; Zhang et al., 2006). The rate information can also assist to assess the relative significance of chemical transformation vs. air/water exchange in the Hg cycle in a lake. Despite their importance

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and value, the data of DGM concentration change rates has remained deficient in the literature, especially out of long-term studies.

Changes of DGM concentrations in an open lake system occur inevitably under ever-changing environmental conditions (e.g., solar insolation, water temperature, wind speed, humidity, etc.). As a result, only apparent kinetic rates are readily obtainable, which may be empirically collected by the following approach:

$$R_a = \Delta[\mathrm{DGM}]/\Delta t = \left([\mathrm{DGM}]_2 - [\mathrm{DGM}]_1\right)/(t_2 - t_1) \tag{1}$$

where R_a is the apparent DGM concentration change rate, and $([\mathrm{DGM}]_2 - [\mathrm{DGM}]_1)$ is the DGM concentration change over the time interval of $(t_2 - t_1)$. The positive rate values thus represent an apparent net production of DGM while the negative ones indicate an apparent net loss of DGM in a lake. Hence, the value and sign of the DGM kinetic rates jointly provide valuable chemodynamic information.

We here report an investigation of the apparent rates of DGM concentration changes in a southern reservoir lake, Cane Creek Lake at Cookeville (Tennessee, USA). An analysis was conducted of the apparent rates of DGM concentration changes in the lake obtained in a 12-month consecutive study from June 2003 to May 2004. The main purposes of this investigation were (1) to provide a rate profile of DGM concentration changes over a one-year period in a southern reservoir lake, (2) to quantify the apparent rates of the DGM concentration changes on various temporal scales, and (3) to compare the rates in the morning DGM production phase and afternoon DGM loss phase so as to gain insights into the possible underlying mechanisms of photochemical redox cycling of aquatic Hg in the lake. The primary focus of this article is the pertinent rate analysis and mechanistic inference. The daily and seasonal changes of DGM levels in the lake were characterized in details elsewhere (Dill, 2004; Dill et al., 2006; Zhang et al., 2006).

2. Site and methods

The apparent rates of DGM production and loss were obtained using the data of the DGM concentrations collected during a 12-month study on temporal variations of DGM concentration conducted at Cane Creek Lake (Cookeville, Tennessee, USA). A detailed description of the basic characteristics of the lake and the sampling site is available elsewhere (Dill, 2004; Dill et al., 2006).

Lake water was sampled each month from June of 2003 to May of 2004, usually twice a month; more intensive sampling campaigns were conducted in the summer of 2003. Sampling went generally from early morning till late evening, as frequently as possible (approximately one sample per hour). The DGM data reported here include those generally spanning the period of ~8:00 am–~7:30 pm (~9:00 am–~5:30 pm for winter time). The analysis of the results was thus based on the data sets obtained within the timeframe studied.

Fresh water samples were taken by surface grab primarily at a nearshore master site. Upon each sampling, field measurements were also carried out in situ to follow water temperature, global solar radiation (R_g), and UVA radiation. The water samples were promptly transferred to our laboratory on campus where the DGM in a sample was immediately purged (generally within ~0.5 h after sampling) and collected on a gold–sand trap, and then the trap was analyzed for total Hg by means of cold vapor atomic fluorescence spectroscopy (CVAFS). The detailed procedures for the field operations and laboratory analyses were documented elsewhere (Dill, 2004; Dill et al., 2006).

All the apparent kinetic rates were computed using Eq. (1). with the DGM data obtained in the field study. It is no surprise that the DGM levels in the lake were slightly or mildly fluctuating from time to time along the general temporal variation trends because of natural change or operational error, resulting in occasional DGM data inconsistent with the general trends, e.g., occasional negative rates in the morning and positive rates in the afternoon. To facilitate the rate analysis aimed at inspecting the general kinetic trends on various temporal scales and inferring the possible mechanisms of aquatic photochemical redox cycling of Hg in the lake, a simplification was adopted by appropriately excluding the irregular rates from the rate data pool (i.e., excluding the negative rate data for the morning phase and positive rate data for the afternoon phase). Consequently, all the trend descriptions and mechanistic speculations were drawn based on the data set thus appropriately processed to remove the fluctuation noise. The trade-off of this practice is a slight reduction in the size of the rate data set, but refined rate analysis and better inference can be obtained. In addition, the lack of a sufficient number of the rate data for November 2003 and February 2004 led us to exclude these two months in the analysis of the monthly apparent DGM kinetics. Nevertheless, a solid monthly rate analysis based on the data of the 10 months resulted with a satisfied data set.

3. Results and discussion

3.1. *Daily trends of apparent rates of DGM production and loss*

Distinct changes of DGM concentration in Cane Creek Lake were observed (Dill et al., 2006; Zhang et al., 2006), exhibiting the diurnal patterns in agreement with the previous findings for other freshwater aquatic systems (e.g., see Amyot et al., 1997; Krabbenhoft et al., 1998; O'Driscoll et al., 2003; Vette, 1998; Zhang and Lindberg, 2000, 2002; Zhang et al., 2002). Fig. 1 provides representative examples of the daily rate profiles of DGM concentration changes in Cane Creek Lake in various seasons. A general feature is the positive apparent rates indicating net DGM production in the morning phase and the negative rates indicating net DGM loss in the afternoon phase post the around-noon peak of DGM level. This pattern parallels the trends of insolation variation in the morning and afternoon, respectively. However, in any particular morning or afternoon phase, the trend of the rate distribution shows no clear pattern, reflecting the variable nature of the DGM kinetics on a small time scale (e.g., hourly). These kinetic characteristics seemed quite prevalent (data not shown).

On the daily scale, interestingly, both the daily mean morning apparent DGM production (DMAP) rates and the daily mean afternoon apparent DGM loss (DMAL) rates varied

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