Review

Factors influencing concentrations of dissolved gaseous mercury (DGM) and total mercury (TM) in an artificial reservoir

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Long-term in-situ monitoring of TM and DGM concentrations with various factors was executed in a large artificial reservoir in this study.

ABSTRACT

The effects of various factors including turbidity, pH, DOC, temperature, and solar radiation on the concentrations of total mercury (TM) and dissolved gaseous mercury (DGM) were investigated in an artificial reservoir in Korea. Episodic total mercury accumulation events occurred during the rainy season as turbidity increased, indicating that the TM concentration was not controlled by direct atmospheric deposition. The DGM concentration in surface water ranged from 3.6 to 160 pg/L, having a maximum in summer and minimum in winter. While in most previous studies DGM was controlled primarily by a photo-reduction process, DGM concentrations tracked the amount of solar radiation only in winter when the water temperature was fairly low in this study. During the other seasons microbial transformation seemed to play an important role in reducing Hg(II) to Hg(0). DGM increased as dissolved organic carbon (DOC) concentration increased (p-value < 0.01) while it increased with a decrease of pH (p-value < 0.01).

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1. Introduction

Mercury differs from other heavy metals in that it continuously goes through the deposition and re-emission cycle in the environment because of its high vapor pressure (Poissant et al., 2000). It is emitted from both natural sources (including volcanic eruptions, wild fires, and the ocean surface) and anthropogenic sources (including coal-fire power plants, waste incinerators, metal smelters, and mercury-related industrial facilities; Pacyna et al., 2003; UNEP, 2002). Most of mercury emitted exists in the atmosphere as inorganic mercury, including elemental mercury (Hg^0), reactive gaseous mercury (RGM; Hg^{2+}), and particulate mercury ($Hg(p)$; Hg^{2+}). Elemental mercury is the predominant species in ambient air since it has a long residence time (0.5–1 yr), while RGM and $Hg(p)$ have relatively high deposition velocities resulting in low concentrations in ambient air (Lai et al., 2007) It is well known that RGM and $Hg(p)$ are very important with respect to mercury deposition (Lindberg and Stratton, 1998; Pan et al., 2008). Once Hg^{2+} is deposited into the aqueous phase, it can be: re-emitted after reduction to elemental mercury, become attached to existing particles followed by sedimentation, or be converted to methyl mercury (MeHg) which is a highly toxic form (Kotnik et al., 2002; Park et al., 2008).

Atmospheric mercury species do not constitute a direct public health risk at the current levels of exposure (Driscoll et al., 2007). However, after it is transformed into MeHg it readily bioaccumulates in aquatic food chains resulting in adverse health effects in humans and wildlife that consume contaminated fish (Hammerschmidt and Fitzgerald, 2006). Therefore knowing the amount of atmospheric Hg deposition to water bodies is critical for understanding its transformation into other Hg forms such as MeHg in water ecosystems. Many researchers have shown that atmospheric Hg deposition is a major source of Hg for the production MeHg in lake ecosystems (Hammerschmidt and Fitzgerald, 2006; Orihel et al., 2006; Wiener et al., 2006). However biological Hg hotspots are not restricted to areas of high Hg deposition because lake characteristics including the amount of drainage area, oxidation–reduction conditions, hydrologic flow paths, and nutrient loading greatly influence Hg transport and transformation (Driscoll et al., 2007). While much is still not known regarding the Hg linkage from air to water to fish and other biota, studies generally suggest that there are three main routes of mercury loss from a lake, including outflows, evasion to the atmosphere, and sedimentation (Mason and Sullivan, 1997).

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In water mercury can exist as a truly dissolved form (dissolved gaseous mercury: DGM), as inorganic Hg(II) associated with dissolved substances, inorganic Hg(II) bound to particles, and methylated Hg. In water mercury continuously changes its form through various abiotic and biotic reactions (O'Driscoll et al., 2005). Dissolved gaseous mercury (DGM) that predominantly exists as Hg^0 can readily volatilize to the atmosphere (loss from a lake; Morel et al., 1998). Its concentration is known to be dependent on various factors such as the amount of dissolved organic carbon (DOC), solar radiation, and/or pH (Matthiessen, 1996,1998; Zhang and Lindberg, 2001; Siciliano et al., 2002; Kelly et al., 2003).

Much research has been conducted in order to identify the factors influencing Hg transport and transformation in freshwater lakes (Amyot et al., 1994; Mason et al., 1995; Poissant et al., 2000; Garcia et al., 2005; Fleming et al., 2006), however most of these studies centered on shallow natural lakes. In addition effects of various factors on reduction and oxidation pathways of Hg have been tested in controlled systems at the laboratory scale by several researchers (Waite et al., 2002; Kelly et al., 2003; O'Driscoll et al., 2004). General effects of lake characteristics on Hg concentrations have also been characterized by many researchers, however they mostly focused on the geographic distribution of Hg concentrations (Amyot et al., 1994,1997; O'Driscoll et al., 2004; Driscoll et al., 2007). In this study Hg cycling in an artificial reservoir, Lake Soyang, which has the largest surface area and pondage in Korea was investigated. The main objectives of this study were to identify the temporal and spatial characteristics of total mercury (TM) and DGM concentrations, and to clarify the effects of the various factors including solar radiation, turbidity, DOC, and pH on DGM and TM concentrations using in-situ measurements made over three years.

2. Experimental section

2.1. Sampling

Lake Soyang, the biggest reservoir in Korea, is located in Chuncheon and has an average surface area of 44.4 km² and average water storage of 2160 m³ (Table 1) The watershed is mainly forested (87%) and agricultural (6%). Three-day intensive DGM and TM samples were obtained at three locations onboard a boat for each season from 2006 to 2008 (Fig. 1). DGM samples were obtained 1 m below water level three times a day (morning, midday, afternoon). In addition there were short-term measurements made every 2 h for 3 days in order to identify diurnal variations in DGM concentrations. Vertical profiles of TM concentrations were measured in samples obtained at 1, 10, 50, and 80 m (right above the sediment surface) below the lake surface in September 2006 and February 2007. FTPE bottles (coated by Teflon) were used for both DGM and TM to limit absorption of Hg^0 onto the walls. After sampling bottles were tightly sealed, placed in a portable cooler, and transported to the laboratory within 30 min after sampling. Meteorological parameters, dissolved organic carbon (DOC), pH, DO (dissolved oxygen), water temperature, and turbidity were also measured in this study.

2.2. Analysis

Within 30 min of sampling, DGM was analyzed using a gas sparging method described in Lindberg et al. (2000). In summary collected lake water was poured into

Table 1
Hydrological characteristics of the drainage basin of Lake Soyang (Kim and Kim, 2004).

Maximum surface area (km²)	70
Average surface area (km²)	44.4
Maximum water capacity (m³)	29×10^9
Average water storage (m³)	20×10^9
Mean hydraulic residence time (yr)	0.75
Mean depth (m)	33.8
Maximum length (km)	57
Average surface water level (as elevation) (m)	176
Watershed area (km²)	2703
Urban area in drainage basin (km²)	1
Forest and mountain area (km²)	2524
Agricultural area (km²)	159

a Pyrex bottle equipped with a glass impinger attached to a four-way Teflon stopcock. Ultra High Purity (UHP) air then flowed into the bubbler so that DGM was stripped and transported into a Tekran 2537A. Purging continued until the Hg level read by the Tekran 2537A dropped below 0.01 ng/m³. A soda-lime trap was placed between the Pyrex bottle and Tekran 2537A to remove humidity. During analysis the Pyrex glass was covered by aluminum foil to limit photo-reactions. All glassware and Teflon tubing in the analytical system were purged by ultra high purity air before the analysis in order to remove remaining Hg. Also a gold sand trap was placed between ultra high purity air tank and the analytical system to remove any Hg in the high purity air.

For TM samples 0.08 M HCl was added to the sample in the field (0.5% of the sample volume) to limit volatilization, followed by the addition of BrCl to limit Hg adsorption onto the surface of the bottle and to convert all mercury forms to Hg^{2+}. $SnCl_2$ was added right before analysis to reduce all Hg^{2+} to Hg^0, followed by analysis with a Tekran 2600. Detailed analytical methods for DGM and TM can be found in: US EPA (1994) and Lindberg et al. (2000). Water temperature was measured using a YSI model 44 temperature probe, and pH was monitored by AP 62 (Fisher Sci.). Water samples for DOC and POC (particulate organic carbon) analyses were filtered through pre-combusted (450 °C) Whatman GF/F filters. The filters were dried for 1 h at 50 °C before measuring POC concentration using gas thermal conductivity (Flash EA1112, ThurmerFisher). The filtrate, used for DOC, was measured by HTCO method using a Shimadzu TOC-5000A total organic carbon analyzer equipped with Pt catalyst on quartz wool. All glassware and Teflon tubing were cleaned with an 11-day acid cleaning procedure before sampling and analysis (US EPA, 1994).

2.3. QA/QC

Overall precision of DGM measurements from duplicate samples was calculated to be $9.62 \pm 4.31\%$ (geomean ± geostandard deviation) ($N = 69$). The Wilcoxin rank sum test showed that two duplicate sets of samples were not statistically different (p-value > 0.1). Field blanks were routinely measured since DGM samples were transported from the boat to the laboratory, possibly resulting in contamination during transport. Distilled water that was previously purged by ultra high purity air was placed in pre-cleaned bottle, and transported between the boat and the laboratory along with samples. The field blank values were 0.36 ± 0.75 pg/L ($N = 9$), and 7 out of 9 field blanks were less than instrumental detection limits.

For TM, overall precision from duplicate samples was calculated to be $8.46 \pm 2.61\%$ (geomean ± geostandard deviation), and there was no statistical difference between two sets (p-value (p-value > 0.1) ($N = 49$)). Blank values were negligible (<1% of sample value).

3. Results

3.1. Factors related to TM concentration

Intensive sampling for TM was performed in May, September, and November of 2006, and February, June, and October of 2007. Samples were collected three times in a day (morning, noon, and before sunset) to identify the daily variation of TM at three sites (Fig. 1). The mean concentration of TM in surface water during the whole sampling period was 1.19 ± 1.06 ng/L. There was no diel variation (p-value = 0.2, Wilcoxon test) or spatial variation among the three sites (p-value > 0.1). However TM concentrations varied temporally, having approximately three to six times higher concentrations in November 06 and February 07 than in the other periods (Fig. 2).

Vertical TM concentration profiles were also measured (at 0, 10, 50, and 80 m) in September 2006 and February 2007. TM vertical patterns were quite different between the two seasons, in February TM ranged from 2.1 (10 m below the water surface) to 4.9 ng/L (at the bottom), showing a similar pattern with previous research (Amyot et al., 1994). However the maximum concentration appeared at 50 m of depth in September. This will be discussed in more detail below.

3.1.1. Turbidity

Summer is the rainy season in Korea, when approx 2/3 of the annual precipitation occurs. Since the watershed of Lake Soyang consists mainly of forest (87%) and agricultural area (6%) under cultivation, runoff causes soil erosion to occur during periods of heavy rain (Choi et al., 2001). During these periods the turbidity of the lake increases significantly. In the summer of 2006 severe rain
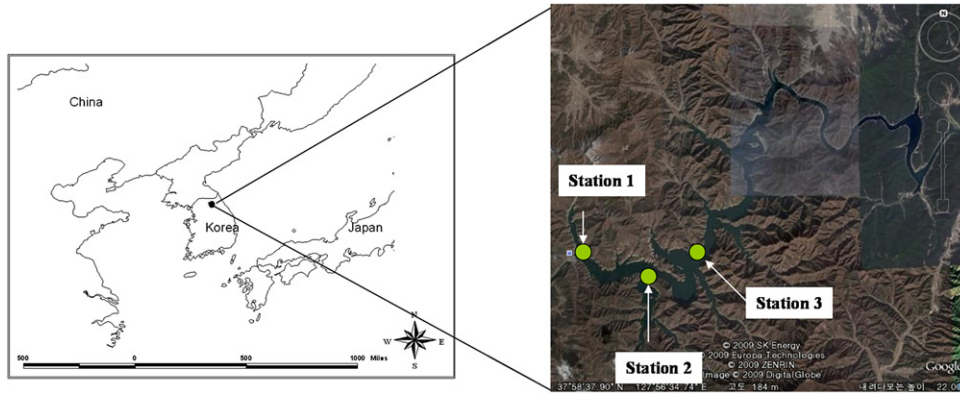
Fig. 1. Locations of monitoring sites for TM, DGM, pH and DOC.

caused a large amount of turbid water from tributaries to flow into Lake Soyang increasing the turbidity down to the mid-depth immediately above the thermocline (upper panel of Fig. 3). However after early winter turn-over the turbid water mixed up to the surface of the lake, increasing turbidity and TM concentrations in the surface water (lower panel of Fig. 3). In September 2006 turbidity was the highest at mid-depth, however as turn-over occurred turbidity in surface water increased more than 7 times (31.3NTU) over that observed in September (4.4NTU). TM concentrations had exactly the same vertical pattern as turbidity during both periods (Fig. 3).

Seasonal variation of TM in the surface water followed the variations in turbidity and particulate organic carbon (POC) with high concentrations in November 06 and February 07 (Fig. 2). These results suggest that most of mercury in the water column exists as particulate mercury similar to previous work (Amyot et al., 1994; Scherbatskoy et al., 1998). Episodic mercury accumulation events occurred during the rainy season as turbidity increased. Taken together these results suggest that TM concentrations in the water column were not controlled by direct atmospheric Hg deposition but by Hg present in surface soil previously deposited to the watershed of Lake Soyang.

3.2. Factors affecting DGM concentration

3.2.1. Solar radiation

Intensive sampling for DGM was performed from May 2006 to July 2008 three times a day (morning; around 8 am, noon; around 1 pm, and before sunset; around 5 pm) at three locations. Spatially there was no statistical variation between sites. The mean concentration during the complete sampling period was 51.1 ± 34.5 pg/L (3.6–160 pg/L). Correlation between DGM and TM concentrations was not found. DGM made up 0.7% of the TM in winter and up to 18.9% in summer (9.5% on average). DGM concentrations varied with the season (Table 2) whereas TM concentrations varied with turbidity (p-value $= 0.02$, Fig. 4).

Previous researchers found that the DGM concentration was controlled primarily by a photo-reduction process initiated by solar radiation (Zhang and Lindberg, 2001; Siciliano et al., 2002). However in this work the diel variation in DGM did not follow the
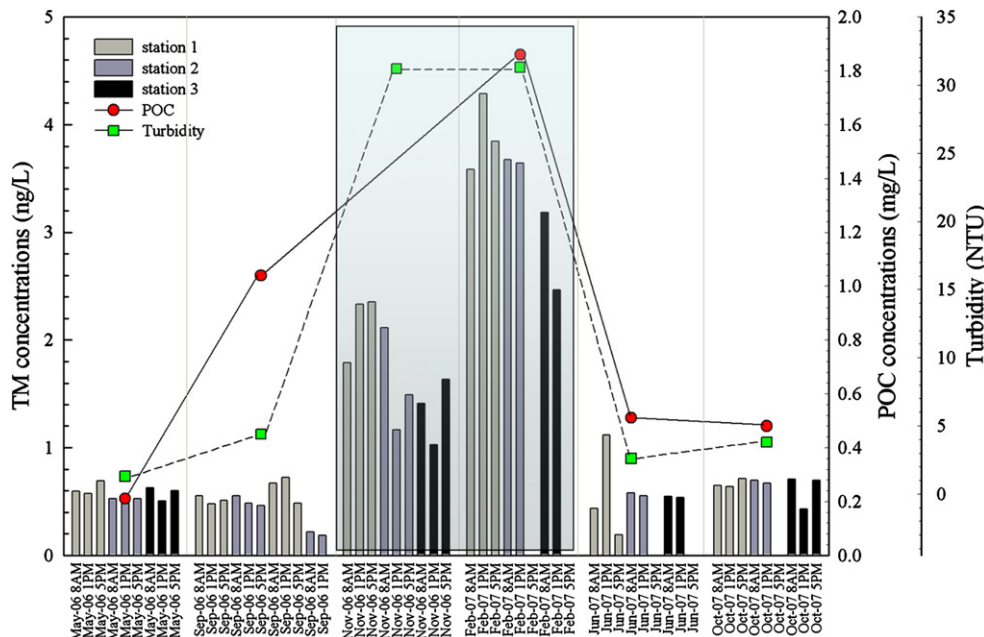

Fig. 2. TM concentrations in surface water of Lake Soyang. The shaded area indicates the period when turbidity was higher than 30 NTU.
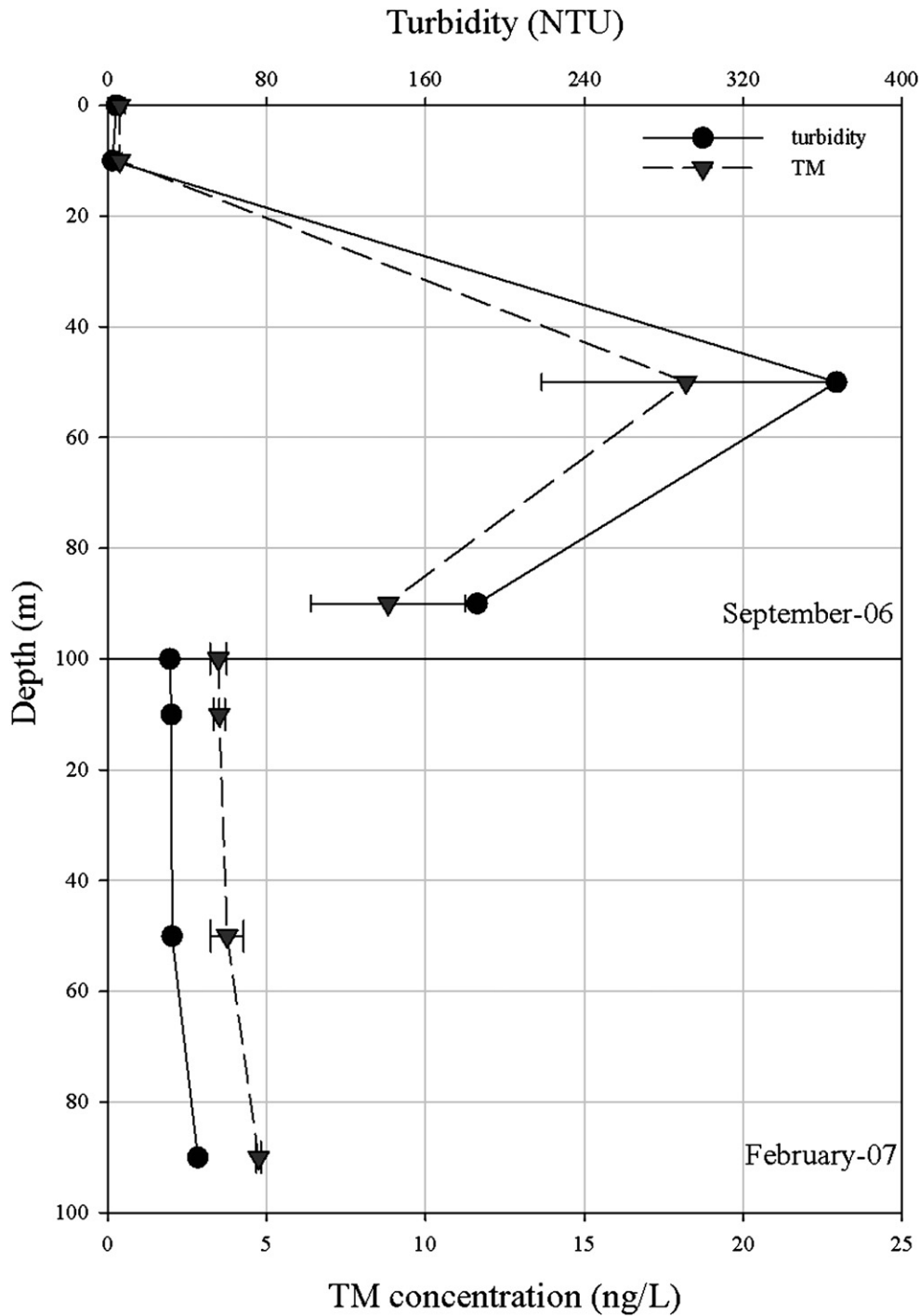
Fig. 3. Vertical pattern of TM concentration and turbidity in water column of Lake Soyang. Error bars indicate one standard deviation.

Table 2
Statistical result (p-value) for seasonal variation in DGM concentration.

	Spring	Summer	Fall	Winter
Spring		0.007	0.103	0.01
Summer	0.007		0.369	0.00001
Fall	0.399	0.369		0.001
Winter	0.01	0.00001	0.001	

Note: Data were tested using independent sample t-test. p-Value lower than 0.05 indicate that there was a statistical difference on DGM concentration between seasons.

measured radiation pattern. For example the largest DGM concentration (160 pg/L) was measured in November 2006 early in the morning during a period of weak solar radiation (Fig. 4). In general solar radiation peaked around 12 pm, however the DGM concentration did not follow the pattern of radiation except in February 2007.

In order to investigate the impact of solar radiation on DGM concentration the DGM concentration was measured during short time periods (every 2 h) for 24 h. Interestingly the DGM concentration was fairly constant during each measurement period and did not follow the variation in solar radiation except possibly the
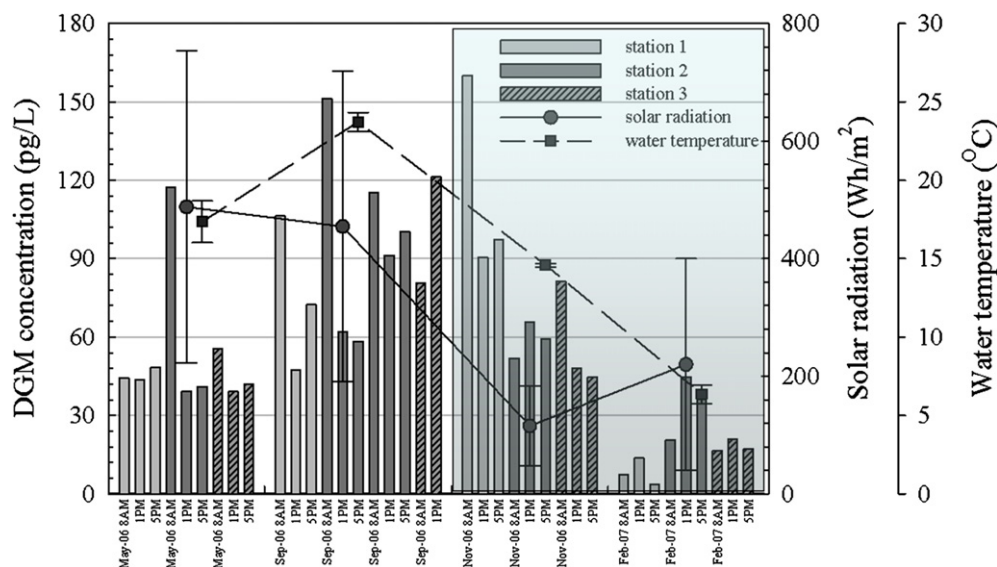
Fig. 4. Relationship of DGM concentrations with solar radiation and water temperature in surface water. Radiation symbols denote averaged hourly values, and shaded area indicates the period when turbidity was higher than 30 NTU. Error bar represents one standard deviation.

March 2008 sample (Fig. 5). In most of the previous research diurnal patterns in DGM concentrations were observed with highs occurring at midday and lows at midnights, closely following the radiation pattern (Amyot et al., 1994, 1997; O'Driscoll et al., 2003, 2007; Dill et al., 2006; Zhang et al., 2006).

3.2.2. Dissolved organic carbon (DOC)

DOC is often acknowledged to be important in the photo-reduction of mercury (Matthiessen, 1996, 1998), although results about how DOC impacts DGM production are contradictory. Watras et al. found that an increasing DOC concentration was related to an exponential decrease in the ratio of DGM to TM (Watras et al., 1995), however other studies (Xiao et al., 1995; Costa and Liss, 1999; Ravichandran, 2000) showed that DGM production increased significantly in the presence of DOC. On the other hand, Matthiessen (1998) observed no reduction of mercury by humic substances. Some studies suggested that DOC structure and composition affect to the DGM production rate (O'Driscoll et al., 2004; Garcia et al., 2005), and also that small change in DOC concentration did not correspond to a change in DGM production rate (Peters et al., 2007). The contradictory results on the relationship between DGM and DOC in different lakes might be due to different DOC structures and the narrow range in DOC concentrations found in previous studies.

In this study a consistent relationship between DOC and DGM production was not observed. However during the November 06 and February 07 sampling periods the lake was very turbid. Excluding the data collected during the periods of high turbidity (>30 NTU) increasing DOC concentration was related to an increase in DGM concentration (p-value < 0.001; Fig. 6). Other researchers (Zepp et al., 1987; Cooper et al., 1989; O'Driscoll et al., 2004) have also suggested that DOC was one of the factors affecting mercury reduction as DOC absorbed solar radiation and emitted electrons (eq. (1)) which were then available to reduce mercury (eq. (2)).

$$[DOC] + h\nu \rightarrow [DOC^{\bullet+}] + e^-(aq) \qquad (1)$$

$$2e^-(aq) + [Hg^{2+}] \rightarrow [Hg^0] \qquad (2)$$
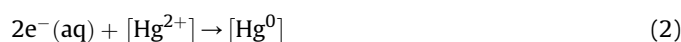
In addition to above reactions, Hg(II) can be directly reduced by humic substances via ligand metal charge transfer (Allard and Arsenie, 1991; Spokes and Liss, 1995). Also photolysis of DOC can form reactive intermediate reductants, such as $HO_2^\bullet$, that reduces Hg(II) (Voelker et al., 1997; Zhang and Lindberg, 2001).

An increase of DGM with increasing DOC showed that DOC played an important role in the reduction of Hg(II) in this lake, however when the water was turbid solar radiation penetration was restricted, and light attenuation by suspended particles would decrease the photo-reduction of Hg(II) (Fig. 6). Garcia et al. also found that DGM was produced at a higher rate in clear water than in turbid water. As described above a significant mercury (TM) load occurred during the rainy season in Lake Soyang as turbid water entered the lake (Fig. 2), however during these periods photo-reduction was limited by high turbidity, thereby decreasing DGM production.

3.2.3. pH

The effect of pH on fish mercury levels has been extensively studied, generally showing that fish in low pH lakes have higher mercury concentrations than fish in higher pH lakes (Driscoll et al., 2007; Kamman et al., 2004). However the effect of pH on DGM production is not well understood in natural aquatic systems. Metthiessen found that increases in pH resulted in an increase in DGM production (Matthiessen, 1998).

The pH measured in Lake Soyang ranged from 5.95 to 8.67. In general DGM concentration increased significantly as the pH decreased (Fig. 7, p-value < 0.001). This finding can be explained by the fact that almost all Hg(II) is bound with organic ligands such as DOC or with inorganic ligands such as Cl_2 (Aiken et al., 2003). At low pH, ligands such as DOC in natural waters will be less negatively charged and therefore less likely to complex Hg(II) (Kelly et al., 2003), making it more available photo-reduction either by abiotic or biotic processes. DOC is an important factor for DGM production in Lake Soyang (Fig. 6), therefore the ability of DOC to bind Hg(II) may influence mercury reduction rates. Increases of H^+ ion can limit Hg(II)-DOC ligand formation, making Hg(II) more available for reduction (eqs. (1) and (2)). Reducibility of Hg bound with DOC has been assumed to be dependent on DOC structure (O'Driscoll et al., 2004; Garcia et al., 2005).
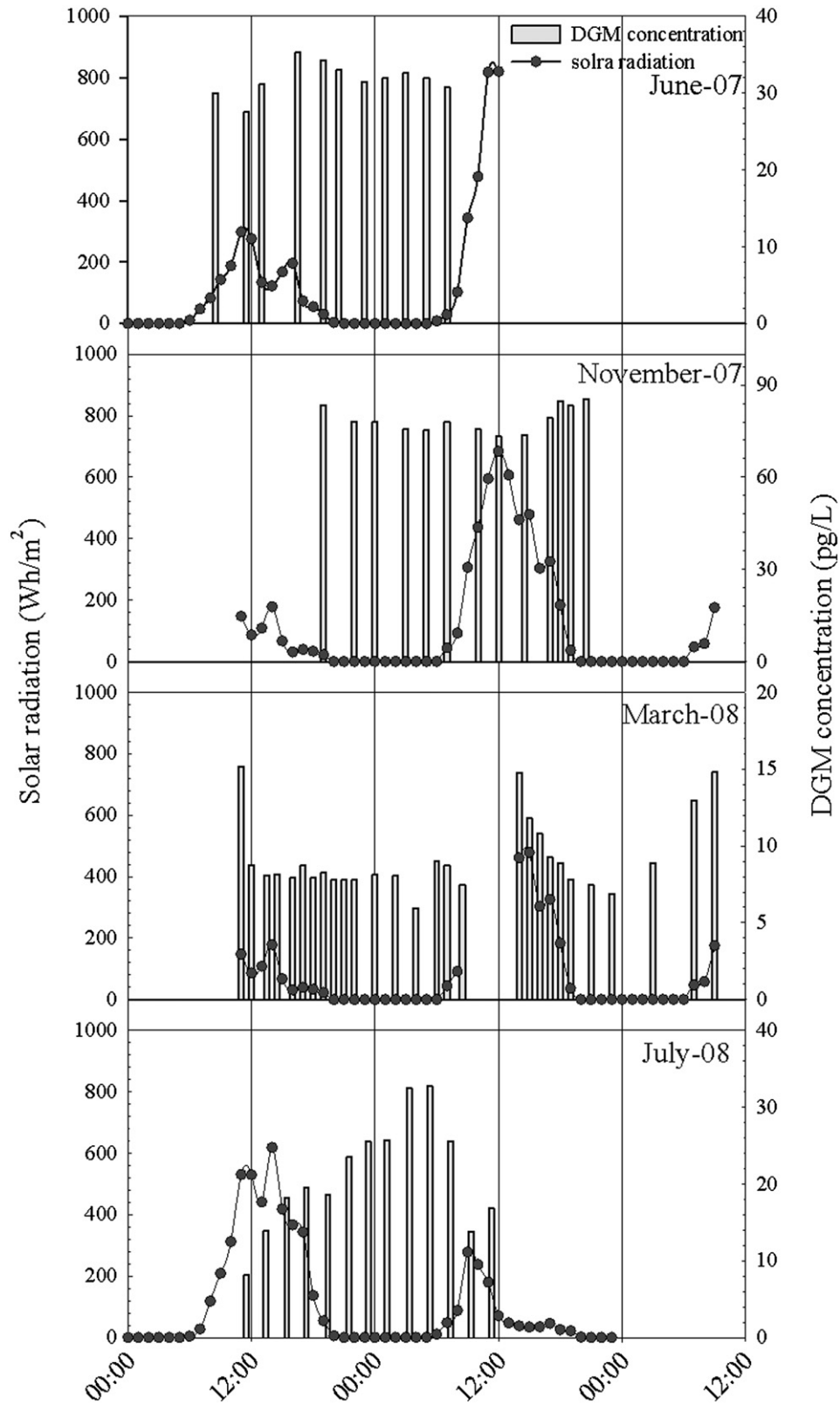
Fig. 5. Variation of DGM concentrations with solar radiation in four different periods. Average hourly radiation is shown.

4. Discussion

Strong diel fluctuations in DGM concentrations have been observed in most lakes with levels of DGM peaking at noon

(Amyot et al., 1997; Krabbenhoft et al., 1998; Siciliano et al., 2002). However in this study DGM concentrations did not follow the solar radiation pattern. Irregular diel fluctuations were observed indicating that other factors in addition to solar radiation must be
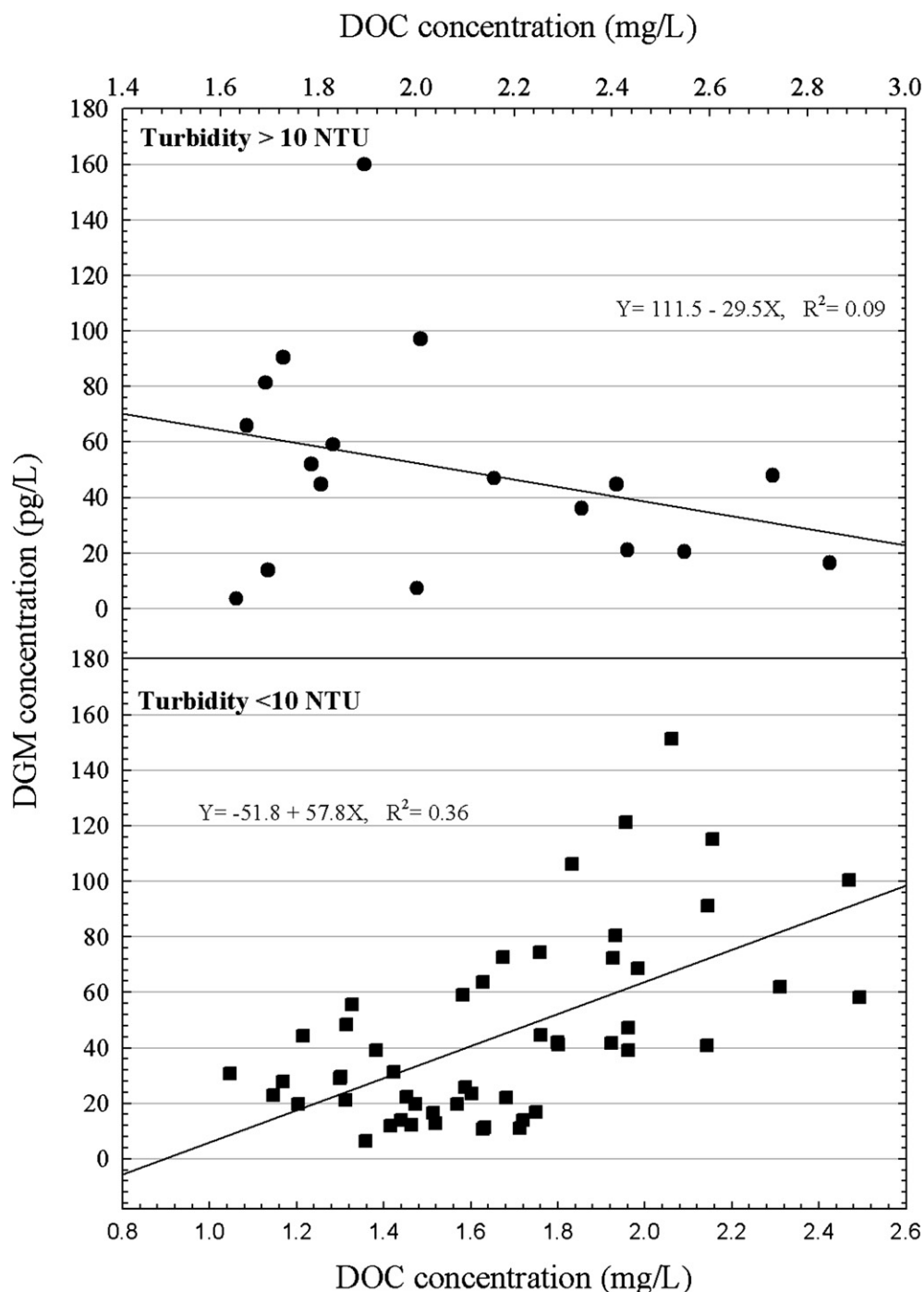
Fig. 6. Relationship between DGM and DOC concentrations in surface water.

influencing the oxidation–reduction of mercury in this lake. A few previous studies have shown that both abiotic and microbial transformations reduce Hg(II) to Hg(0) (Mason et al., 1995; Siciliano et al., 2002), but the role of microbial activity has been largely assumed to be less important than abiotic photo-reduction.

In freshwater it has been postulated that heterotrophic bacteria may play a role in DGM production (Nazaret et al., 1994; Siciliano et al., 2002; Wiatrowski et al., 2006). We observed that DGM concentrations were correlated with radiation only in February 07 and March 08 (Figs. 4 and 5) when the water temperature was fairly low (average water temperatures were 23.0 °C, 20.4 °C, 6.6 °C, and 27.5 °C in June 07, November 07, March 08, and July 08,

respectively). However in warm seasons DGM concentrations did not track radiation possibly because microbial transformation reducing Hg(II) to Hg(0) was more active in the warmer season than in the colder season.

Siciliano et al. (2002) measured microbial mercury reductase activity over a 3 day period in Brookes Bay, Jack's Lake, and they observed that microbial reductase activity consistently peaked around 10 am, reached the lowest point at 3 pm, and increased again during midnight (7pm–3am). This pattern is out of phase with solar radiation and may explain the fairly constant DGM concentration in warmer seasons. In this study we often observed that DGM levels increased in the early morning (Fig. 4), and also
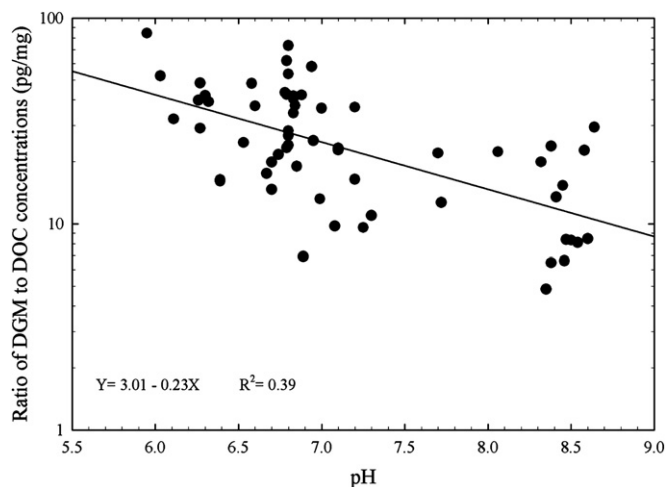
Fig. 7. Relationship between DGM concentration and pH.

that DGM did not steadily decrease at night in the warm season (Fig. 5). In addition monthly averaged DGM concentrations correlated with water temperature better than with solar radiation.

In colder seasons when microbial activity often decreases, the photo-reduction pathway may become dominant resulting in DGM concentrations that track radiation (February 07 and March 08). There is also a possibility that the lack of diurnal DGM concentration variation in response to changes in solar radiation was, in part, due to the lack of sufficient time resolution (~ 2 h) used for DGM measurement in this study since large variations in DGM concentrations can occur over short timeframes. However, other researchers who used continuous analysis systems (Amyot et al., 2001; O'Driscoll et al., 2003) found strong diel patterns in DGM concentrations along with solar radiation that would have been seen at the sampling time resolution used in this study, suggesting that solar radiation was not the primary factor driving DGM concentration variations.

Besides solar radiation and bacteria, this study showed that other chemical factors such as DOC and pH affect DGM production. We observed that DGM concentrations increased with an increase of DOC and with a decrease of pH (Figs. 6 and 7). There are three pathways by which DOC can affect DGM production. First, DOC absorbs solar radiation and emits aqueous electrons, which are available to reduce Hg(II) (O'Driscoll et al., 2004). Second, the formation of stable Hg-humic complexes can lead to a subsequent direct reduction to Hg(0) by an intra molecular electron transfer (Allard and Arsenie, 1991). Third, reactive intermediate reductants formed via the photolysis of DOC can reduce Hg(II). According to Haitzer et al. (2002, 2003) Hg-thiol group complexes are a preferred form to Hg-carboxylic group complexes at a low ratio of Hg/DOM (<1 μg-Hg/mg-DOM). Therefore we hypothesized that Hg–DOC complexes in this lake mainly were Hg-thiol group complexes rather than Hg-carboxylic group complexes due to the low ratio of Hg/DOM. Since Hg binding to reduced sulfur groups is many orders of magnitude stronger than for Hg-carboxylic groups (Hintelmann et al., 1997; Haitzer et al., 2003), Hg-sulfur group complexes limit the availability of photo-reducible Hg. Even though Hg–DOC complexes mainly existed as Hg-thiol DOC is still an important factor in Hg(II) reduction through the first and third pathways described above. Xiao et al. (1995) also reported that Hg(II) bound with inorganic ligands such as Hg(OH)$_2$ and HgCl$_2$ were more easily reduced in the presence of DOC.

When the pH decreases there is a significant increase in proton competition for Hg(II) binding sites. Therefore the decrease in pH

decreased the binding of Hg(II) with thoil group, making it more available photo-reduction. On the other hand, Hg-carboxylic group which is a more photo-reducible form than thiol group has lower pKa values ($=4.3$; Perdue, 1985), indicating that the carboxylic groups are predominantly negatively charged in this lake, and pH changes appeared in this study were unlikely to significantly alter the amount of Hg binding by carboxylic groups.

If this was the case in Lake Soyang, there would be synergetic effect on DGM production when DOC concentration was high and pH was low. To investigate this the DGM concentration data were divided into four groups 1) low pH and high DOC, 2) low pH and low DOC, 3) high pH and high DOC, and 4) high pH and low DOC. The highest DGM concentration was observed when the pH was lower than 7 and DOC was higher than 1.7 mg/L, and the lowest DGM concentration occurred at high pH and low DOC concentrations. In addition to the effect on Hg–DOC complexes a few studies showed that bacteria uptake of both charged and uncharged Hg(II) species increased at low pH (Kelly et al., 2003), which probably resulted in more active reduction by biotic process.

5. Conclusions

In this study long-term monitoring of TM and DGM concentrations found several interacting factors were important in controlling their concentrations in a large artificial reservoir. The mean concentration of TM and DGM in surface water were 1.19 ± 1.06 ng/L and 51.1 ± 34.5 pg/L, respectively, showing that DGM made up on average only 4.3% of the TM. Temporal variations in TM levels tracked variations in turbidity and particulate organic carbon. These results suggest that mercury exists mainly as particulate mercury in Lake Soyang.

Seasonal variation in DGM concentrations in general tracked water temperatures rather than solar radiation. Hourly DGM concentrations tracked radiation only during cold seasons, and in other seasons there was no relationship with solar radiation. This result contradicts many previous studies that observed that DGM concentrations were primarily controlled by a photo-reduction process. Since seasonal concentration of DGM generally tracked water temperature we hypothesis that heterotrophic bacterial transformations reducing Hg(II) to Hg(0) were generally the dominant reduction process. However in cold seasons when microbial activity often decreases photo-reduction becomes more important.

DOC concentrations were found to be positively related with DGM concentration while DGM increased with a decrease of pH. An increase in DGM with increasing DOC suggests that DOC played a positive role in the reduction of Hg(II) in the lake. Several studies suggested that DOC structure affects the amount of reducible Hg, however we hypothesized that DOC structure remained consistent within this lake although clearly it may differ between different lakes and that the binding of Hg to DOC is controlled by reactive thiol functional group at the low ratio of Hg/DOC found. With high H$^+$ ions concentrations Hg(II)-thiol ligands are suppressed, producing more reducible Hg(II) because H$^+$ competes for the Hg(II) binding sites. In this study we observed the higher DGM concentrations when DOC concentrations were high and pH was low. In the future it will be worthwhile to intensively characterize the interaction of DGM with various factors including DOC structure and bacterial types.

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