

## VARIATIONS OF STABLE CARBON ISOTOPE COMPOSITIONS OF DISSOLVED INORGANIC AND PARTICULATE ORGANIC CARBON IN SURFACE WATERS OF TWO LAKES, NORTHEASTERN TAIWAN

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### ABSTRACT

Stable carbon isotope ratios of total dissolved inorganic carbon (DIC) and particulate organic carbon (POC) for surface waters of the Mei Hwa Lake (MHL; 50 m a.s.l.) and the Shuang Lian Pond (SLP; 470 m a.s.l.), northeastern Taiwan collected monthly during 2003-2004, were investigated. The TOC/TN ratios of POC for both lakes are less than 10, which indicate aquatic algal photosynthesis is the main source for the POC.  $\delta^{13}\text{C}$ -POC values range between -21.1‰ to -28.9‰, as colder seasons showed relative depletions. Moreover, the time-series of  $\delta^{13}\text{C}$ -POC values for both lakes show parallel decreasing trend for the isotope composition through time. Generally, similar depleted trends for values of  $\delta^{13}\text{C}$ -DIC in MHL and SLP were recorded except from March to August in 2003. The  $\delta^{13}\text{C}$ -DIC values in MHL tend to be enriched by 1-3‰. Such heavier  $\delta^{13}\text{C}$  signatures in DIC of MHL are likely caused by a local drought episode during the early 2003.

**Key words:** dissolved inorganic carbon (DIC), lake, particulate organic carbon (POC), stable carbon isotope, Taiwan

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## 1. INTRODUCTION

Several characteristics of lakes, such as short residence time of element recycling, quick response of biota, and high sedimentation rate, make them advantageous for monitoring climatic changes on local ecosystems. Particularly,  $\delta^{13}\text{C}$  values of dissolved inorganic carbon (DIC), particulate organic carbon (POC), and sedimentary organics have been demonstrated very useful for tracing the transportation and fate of carbon in lacustrine ecosystems (e.g., Quay *et al.*, 1986; Meyers, 1997; Cole *et al.*, 2002; Street-Perrott *et al.*, 2004). Using such isotope tracer, Scartazza *et al.* (2004) indicated that stable carbon isotope ratio of photosynthetic products and respiratory  $\text{CO}_2$  in an ecosystem would reflect the seasonal climate change.

The  $\delta^{13}\text{C}$ -DIC within lake water is mediated by many factors, e.g., geochemical compositions (e.g., DIC, pH, alkalinity), morphometric factors (lake size and shape), biological activities (gross primary production, respiration), and others (see Bade *et al.*, 2004 and reference therein for a review). Using a model processed with data from 104 lakes, Bade *et al.* (2004) concluded that ecosystem metabolism induce substantial variation in  $\delta^{13}\text{C}$ -DIC around the potential value that is set by geochemical factors of the watershed. However, works of McKenzie (1985), Talbot (1990) and Leng & Anderson (2003) indicated the importance of isotopic exchange of carbon isotopes between aqueous bicarbonate and atmospheric  $\text{CO}_2$  which resulted in greater  $\delta^{13}\text{C}$  values in aqueous bicarbonate ion in lacustrine environment. This process occurs potentially in a close-basin lake with large surface area, long residence time and high evaporation (Leng & Anderson, 2003). Differentiating the long-term trend in  $\delta^{13}\text{C}$ -DIC from seasonal variation and other specific factors of an individual lake is crucial for judging the principal mechanism. Because the stable carbon isotope composition of lake water is influenced by temperature effect, kinetic fractionation, lipid content of phytoplankton cell and carbon source (see Descolas-Gros & Fontugne, 1990 and references therein for a review).

For deciphering the influence of major factor on variations of the stable carbon isotope composition in lake water in the western Pacific East Asian Monsoon region, two adjacent lakes at different altitudes in Ilan County (Figure 1), northeastern Taiwan were chosen to investigate the  $\delta^{13}\text{C}$  variations in organic and inorganic carbons during a two-year period from 2003 to 2004. The Mei Hwa Lake (MHL) is located at 24.64°N, 121.72°E at elevation of 50 m, and has an area of 18.2 ha with ~ 1 m of water depth. The Shuang Lian Pond (SLP), 4.6 ha in size and <2 m in water depth, is located nearby in a hilly endorreic basin at 24.76°N, 121.63°E at 470 m above sea level (a.s.l.). Both lakes receive mainly precipitation and little supply of streamlet water along the slope of surrounding highland, and have only one outlet, respectively.

The reversal of summer southwest and winter northeast monsoon in northeastern Taiwan causes seasonal fluctuation on precipitation (Hsu and Chen, 2002). Thus, the cold and warm seasons in this work are defined as the interval of dominated winter and summer monsoon, respectively (cf. Peng *et al.*, 2002). The monthly mean precipitation and air temperature during the investigation period for the studied areas are shown in Table 1. We expect that the  $\delta^{13}\text{C}$  signals of these two lakes would reflect seasonal fluctuation due to local temperature and precipitation effect, as well as caused by their differences in lake size, altitude and climatic responses.

## 2. METHOD AND MATERIALS

Surface lake waters for stable isotope analyses were sampled monthly from Feb. 2003 to Dec. 2004. In addition, rainfall samples were also collected on the 21<sup>st</sup> of Feb. 2005 for comparison. Sample collection and preparation procedures for DIC, POC and water oxygen isotope analyses are the same as described in Peng (1995), Kao and Liu (2000), and Peng *et al.* (2002), respectively.

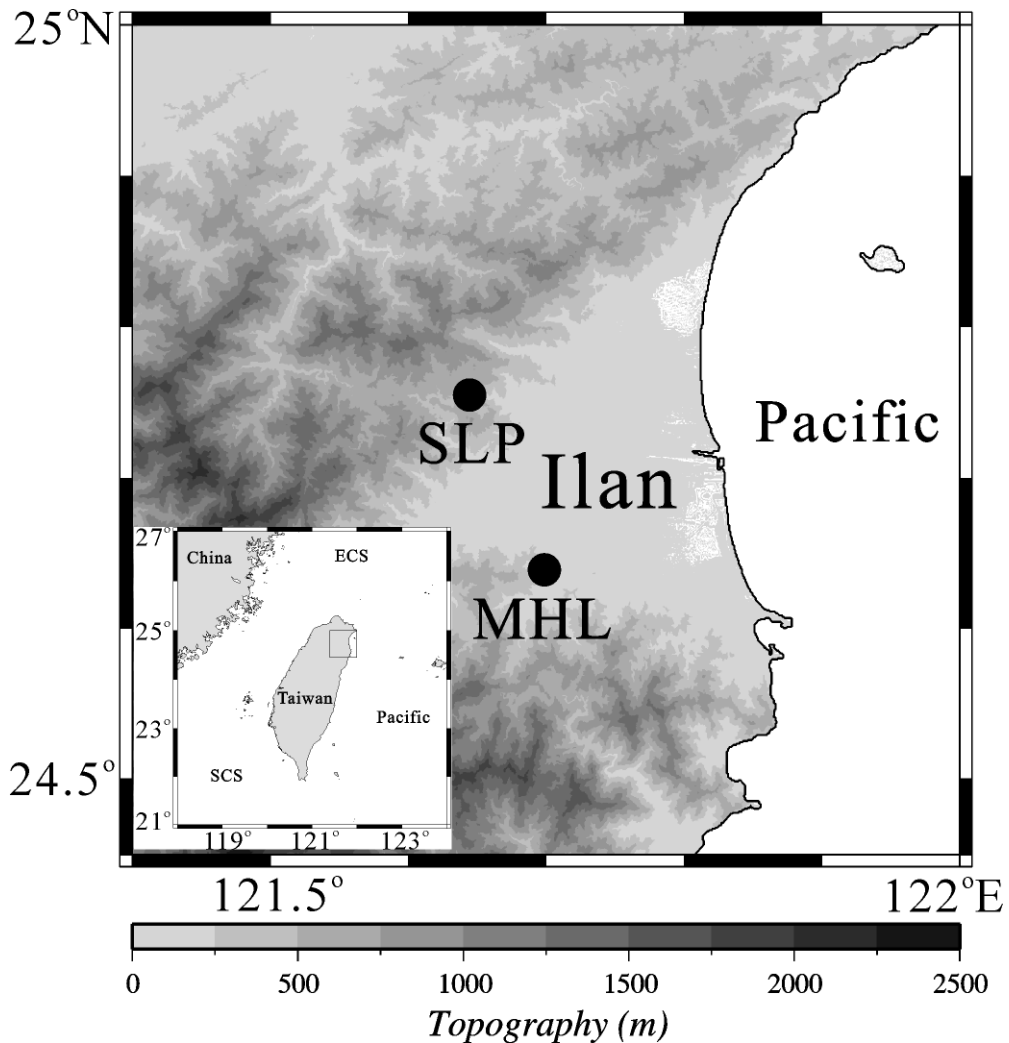


Figure 1. Topographic map shows locations of the Mei Hwa Lake (MHL) and Shung Lian Pond (SLP) in the Ilan County, northeastern Taiwan. Inset: Taiwan is situated between the East China Sea (ECS), Pacific, and South China Sea (SCS). The small rectangle indicates the area of Ilan County.

Briefly, an aliquot of the water sample was filtered on site immediately using a Whatman Cellulose membrane (5  $\mu\text{m}$  in pore size, 47 mm in diameter) for preparation of  $\delta^{13}\text{C}$ -DIC analysis. Then, barium hydroxide was put into the filtrate to precipitate enough amount of dissolved inorganic carbon in the form of precipitate with chemical composition of  $\text{BaCO}_3$ . The purified  $\text{BaCO}_3$  was then treated with orthophosphoric acid to release  $\text{CO}_2$  in a vacuum system under  $50^\circ\text{C}$ . The released  $\text{CO}_2$  was then introduced into a mass spectrometer for stable carbon isotope analysis.

Table 1. Monthly data of stable carbon and oxygen isotope ratios (‰) of lake surface waters of Meihwa Lake (MHL) and Shuang-Lian Pond (SLP), and mean precipitation (MP, cm) and air temperature (AT,  $^\circ\text{C}$ ) on studied area during 2003-2004. The monthly MP and AT at MHL and SLP are from Suao Station, CWB, and from the Fu Shan Experimental Forest TERN (Taiwan Ecological Research Network), respectively. DIC: dissolved inorganic carbon; POC: suspended particulate organic carbon.

2003	MHL					SLP				
	$\delta^{13}\text{C}$ -DIC	$\delta^{13}\text{C}$ -POC	$\delta^{18}\text{O}$	MP	AT	$\delta^{13}\text{C}$ -DIC	$\delta^{13}\text{C}$ -POC	$\delta^{18}\text{O}$	MP	AT
Jan				7.4	16				5.3	10
Feb				6.5	18	-13.9	-23.8	-4.3	6.0	13
Mar	-12.6	-22.5	-1.5	11.1	18				19.9	13
Apr	-13.2	-23.4	-0.8	24.6	23	-14.6	-22.9	-3.3	38.7	19
May				3.5	25				20.0	20
Jun	-12.9	-24.4	-2.1	31.0	27	-15.7	-21.1	-4.3	57.8	22
Jul				0.4	30				7.5	25
Aug	-11.8	-24.5	1.0	11.2	29	-14.8	-24.6	-3.5	40.2	24
Sep				23.8	27				54.5	22
Oct	-15.5	-25.7	-2.8	33.1	24	-16.2	-26.6	-4.5	56.9	18
Nov				104.8	22				38.9	16
Dec	-15.2	-28.3	-2.5	12.5	17	-15.4	-28.7	-4.0	4.4	12
2004										
Jan	-14.6	-26.7	-2.3	6.0	16	-15.9	-26.3	-4.0	6.1	11
Feb	-14.4	-23.8	-2.5	31.9	17	-15.7	-25.3	-4.1	25.1	12
Mar	-15.2	-23.6	-2.1	22.6	18	-14.4	-25.3	-3.7	22.4	13
Apr	-14.7	-26.3	-1.7	6.5	21	-16.3	-24.1	-3.7	3.4	19
May	-14.6	-26.1	-0.6	13.8	25	-16.1	-23.5	-3.7	40.1	21
Jun	-14.1	-26.1	-2.0	18.4	26	-15.8	-24.2	-4.3	25.8	22
Jul	-13.9	-27.1	-2.2	20.8	28	-14.7	-24.8	-4.6	26.4	24
Aug	-14.7	-25.6	-0.7	22.1	28	-15.5	-27.5	-4.1	119.0	24
Sep	-14.4	-23.6	-4.2	62.9	27	-15.0	-24.5	-6.5	53.6	22
Oct	-14.4	-28.9	-3.1	26.9	23	-14.0	-25.4	-5.5	33.7	17
Nov	-14.3	-25.9	-3.5	42.3	21	-14.9	-27.8	-5.1	34.4	16
Dec	-14.2	-27.5	-4.2	46.6	19	-14.1	-26.8	-5.5	80.1	14

Another aliquot of water was also filtered on site as quickly as possible using a precombusted (450°C) Whatman glass fiber filter (GF/F, 0.7  $\mu\text{m}$  in pore size, 47 mm in diameter). Membrane with suspended particulate organic matters covering on it was dried, acidified and delivered for total organic carbon (TOC), nitrogen (TN), and carbon isotope measurements.

To measure the oxygen isotope composition of water, lake water was preserved in situ within a bottle by addition of mercury chloride, and was equilibrated with  $\text{CO}_2$  in a vacuum system in thermostat at 30°C for two hours in laboratory. The equilibrated  $\text{CO}_2$  was subsequently extracted to a mass spectrometer for measuring its stable oxygen isotope composition.

Carbon isotopic ratio of DIC and oxygen isotope composition of lake waters were measured using a VG SIRA-10 triple collector mass spectrometer at the Institute of Earth Sciences, Academia Sinica. Carbon isotope ratios of POC were analyzed using a Finnegan Delta Plus mass spectrometer at the Department of Geosciences, National Taiwan University. The results are expressed in  $\delta$ -notation with per mil (‰) relative to VPDB and VSMOW for carbon and oxygen (Coplen, 1996), respectively. The precisions of the measurements were  $\pm 0.06\%$ ,  $\pm 0.10\%$  and  $\pm 0.10\%$  for  $\delta^{13}\text{C}$ -DIC,  $\delta^{13}\text{C}$ -POC and  $\delta^{18}\text{O}$ - $\text{H}_2\text{O}$ , respectively.

Total organic carbon and nitrogen content of POM analyses follow the procedures described in Ku *et al.* (2005). The decalcified sample was combusted using a ThermoQuest EA1110 elemental analyzer to get the TOC and TN contents, and thus the calculated TOC/TN ratio. The precisions of measurement for TOC and TN were both  $\pm 0.2$  wt%.

### 3. RESULTS AND DISCUSSION

#### 3.1 Characteristics of $\delta^{13}\text{C}$ -DIC

Time-series of  $\delta^{13}\text{C}$ -DIC values in both lakes showed seasonal variation with values ranging between -11.80‰ and -15.46‰ for MHL, and -13.94‰ to -16.30‰ for SLP (Figure 2). The mean  $\delta^{13}\text{C}$ -DIC value of MHL (-14.14‰) was heavier than that of SLP (-15.16‰). The  $\delta^{13}\text{C}$ -DIC values of rainwater in winter season of 2004/2005 at the MHL and SLP were -16.27‰ and -16.55‰ (Figure 2), respectively. Which are much depleted than the value -8‰ of air source (Langenfelds *et al.*, 2002). The mean  $\delta^{13}\text{C}$ -DIC values of both lakes are close to the rainwater sources, especially for the SLP, indicating that lake waters source from rainwater and are little affected by isotopic exchange between lake water and air  $\text{CO}_2$ . Moreover, photosynthesis of phytoplankton also affect the  $\delta^{13}\text{C}$ -DIC of lake water, as evidently by decoupled trends between values of  $\delta^{13}\text{C}$ -DIC and  $\delta^{13}\text{C}$ -POC in the warm seasons during the monitoring period.

On the other hand, our data also reveal the impact of climate change on the  $\delta^{13}\text{C}$ -DIC of lake waters. Heavier value from MHL during local drought episode from March-August, 2003 indicates the occurrence of enrichment of carbon isotope (Figure 2). During this drought period, the mean monthly precipitation dropped while the  $\delta^{18}\text{O}$  of lake waters became heavier (Figures 3a; 4a).

Apparently,  $\delta^{13}\text{C}$ -DIC decreases with altitudes as indicated by the relative depleted value in  $^{13}\text{C}$  -15.16‰ in SLP (alt. 470 m), as compared to -14.14‰ in MHL (alt. 50 m) (Figure 2), which is similar to the trend of  $\delta^{18}\text{O}$  in both lakes (Figure 4b). That is, therefore, more likely due to the larger lake size for the MHL would lead to stronger evaporation, and more extensive isotopic exchange between water and air while the DIC pool is more enriched in  $^{13}\text{C}$ . Bade *et*

al. (2004) witnessed that the  $\delta^{13}\text{C}$ -DIC of larger lake tend to be near isotopic equilibrium with the atmosphere, owing to an increase in the gas-water exchange rate resulted from high wind speed. Moreover, the  $\delta^{13}\text{C}$ -DIC values in the SLP tend to show lighter values than that in the MHL during the warm seasons in 2003 and 2004. We consider this as a result of a much enhanced primary production deduced from  $\delta^{13}\text{C}$ -POC values in the SLP during the intervals (Figure 2).

### 3.2 Characteristics of $\delta^{13}\text{C}$ -POC

The  $\delta^{13}\text{C}$ -POC values in these two lakes were similar and fluctuated seasonally (Figure 2). The  $\delta^{13}\text{C}$ -POC values during warm seasons are heavier than those in the cold seasons. The seasonal trend is apparently corresponding to thermal effect of  $\text{CO}_2$  solubility. Dissolubility of gaseous  $\text{CO}_2$  in surface water decreases with elevated air temperature. The  $\delta^{13}\text{C}$  value of dissolved  $\text{CO}_2$  is more negative, while bicarbonate usually has a less depleted in  $^{13}\text{C}$  of carbon

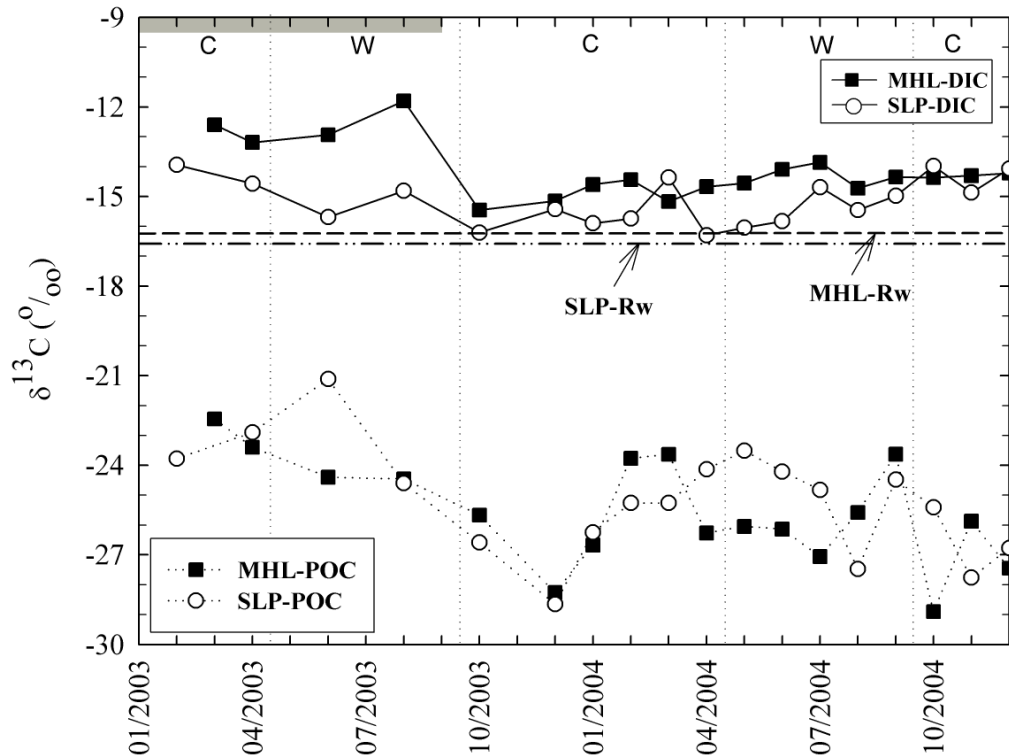


Figure 2. Monthly variation of  $\delta^{13}\text{C}$  values of dissolved inorganic carbon (DIC, solid lines) and particulate organic carbon (POC, dotted lines) in lakes MHL (filled squares) and SLP (open circles) between Feb. 2003 and Dec. 2004. The  $\delta^{13}\text{C}$  value of DIC in rainwater of Feb. 2005 is indicated with dash and dash-dot-dot line for the MHL (MHL-Rw) and SLP (SLP-Rw), respectively. C and W indicate cold and warm interval in time, respectively. Horizontal grey bar marks the drought period in 2003.

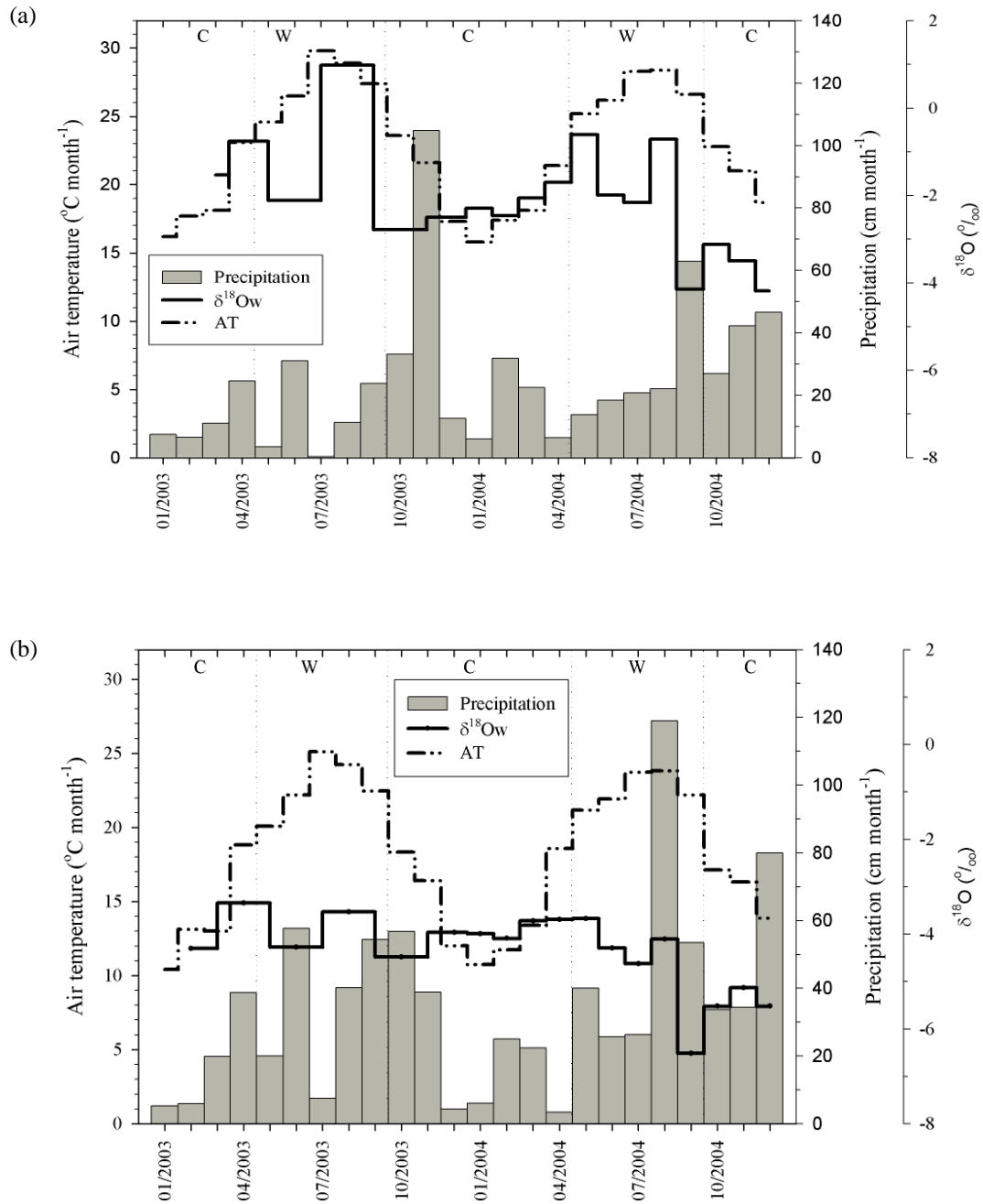


Figure 3. Time-series of  $\delta^{18}\text{O}$  values of lake surface waters of (a) MHL and (b) SLP, and monthly mean precipitation (MP) and mean air temperature (AT) during 2003-2004 in the studied area, respectively.

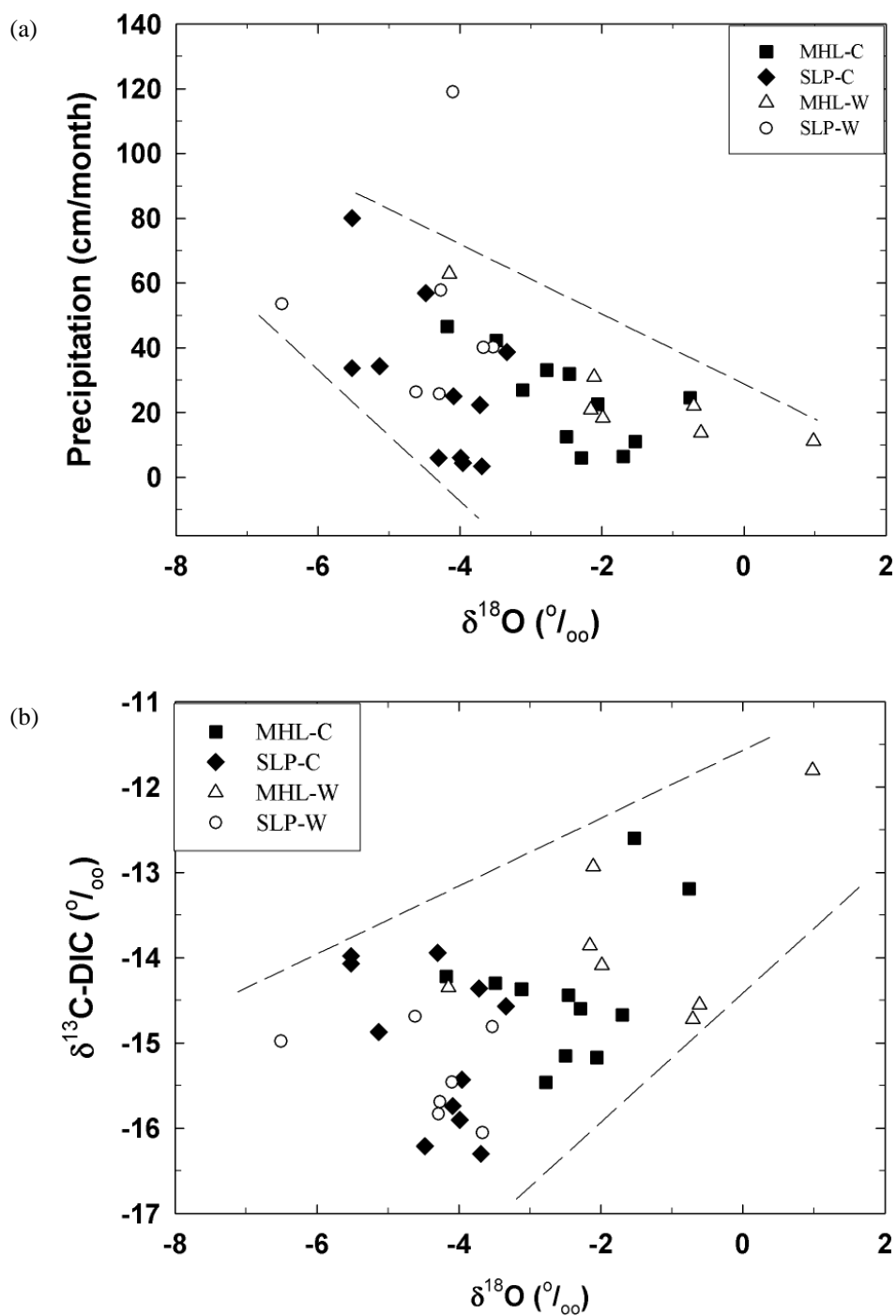


Figure 4. Plots of  $\delta^{18}\text{O}$  against (a) monthly mean precipitation amount, and (b)  $\delta^{13}\text{C-DIC}$  in MHL and SLP during years 2003 and 2004. (C: cold season; W: warm season).



isotope value (Mook *et al.*, 1974). Their work revealed that the discrepancy of  $\delta^{13}\text{C}$  value between these two carbon pools is around 7.9‰ at 25°C. A relative depleted  $\delta^{13}\text{C}$ -POC value in surface water, therefore, reflects the increase of incorporation of dissolved  $\text{CO}_2$  into organic products during algae photosynthesis in cold period. This temperature-dependent phenomenon has also been verified by Sackett *et al.* (1965). Fontugne & Duplessy (1981) and Rau *et al.* (1982) showed that the phytoplankton  $\delta^{13}\text{C}$  values in sea surface waters become more depleted in  $^{13}\text{C}$  when the sea surface temperatures were lower. The greater discrimination in  $\delta^{13}\text{C}$ -POC at high concentration of  $\text{CO}_2$  dissolved in water than at low  $\text{CO}_2$  were also respectively documented by Estep *et al.* (1978) and Sharkey and Berry (1985).

However, it is suggested that  $\delta^{13}\text{C}$ -POC more enriched in  $^{13}\text{C}$  in the SLP than in MHL is a result of enhanced primary production in the SLP during the warm intervals in years 2003 and 2004 (Figure 2). The growth rate of phytoplankton in SLP might increase in association with the rising temperature or strengthening sunlight in the early spring (Table 1), resulting in less selectivity on carbon sources between dissolved  $\text{CO}_2$  and bicarbonate ion, and little discrimination on their  $^{12}\text{C}$  and  $^{13}\text{C}$ . In a review work, Meyers (1997) noted that smaller lakes are generally more productive than larger ones. Furthermore, Hollander and McKenzie (1991) and Lehmann *et al.* (2004) also observed the same trend as our data: that the POM enriched in  $^{13}\text{C}$  as primary productivity increased during spring in Lakes Greifen and Lugano, Switzerland, respectively. The enhanced primary production simultaneously impacted on a much depleted  $^{13}\text{C}$  for the DIC pool in the SLP than that in MHL during the warm seasons (Figure 2; Table 1).

Additionally, the ratios of total organic carbon to total nitrogen (TOC/TN) in particulate organic matter are always smaller than 10 (Figure 5), indicating an algal origin (Meyers, 1994). The  $\delta^{13}\text{C}$ -POC values indicate that phytoplanktons in both lakes photosynthesize by the  $\text{C}_3$  pathway. The lack of correlation between  $\delta^{13}\text{C}$ -POC and TOC/TN (Figure 5) suggests that the photosynthesis products are composed of lipids, proteins and sugars (Degens *et al.*, 1968), and the composition of primary producers might vary through the monitoring period.

#### 4. CONCLUSIONS

This study shows that the  $\delta^{13}\text{C}$  values of DIC were generally heavier in the plain lake MHL compared to the hilly lake SLP. We interpret that larger lake size, higher evaporation and less photosynthesis activity in MHL are the principal controlling factors for the differences. A more enhanced primary production revealed by  $\delta^{13}\text{C}$ -POC in the SLP than the MHL during both warm seasons in 2003 and 2004 is suggested. The  $\delta^{13}\text{C}$ -POC lighter trends in both studied lakes from warm to cold time reflect the strong thermal effect on amount of dissolution of  $\text{CO}_2$  during winter season, and the selective utilization of dissolved  $\text{CO}_2$  immediately after replenishment as well.

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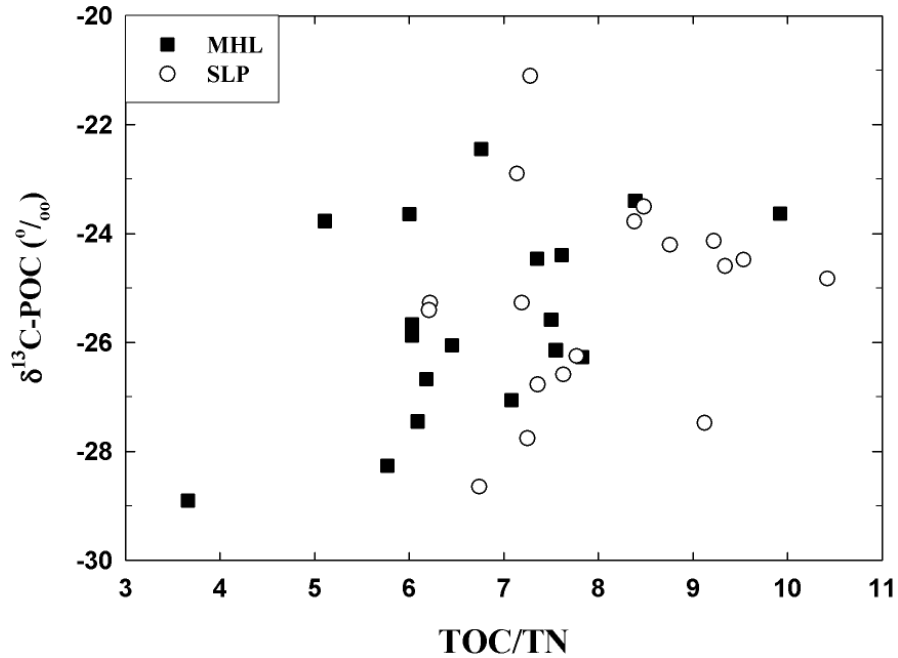


Figure 5. Plot of TOC/TN ratios against  $\delta^{13}\text{C-POC}$  of lake surface waters in MHL and SLP during years 2003 and 2004.

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## 台灣東北部兩湖泊表層水穩定碳同位素比值變化

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### 摘 要

2003年至2004年間，針對台灣兩湖泊每月採集表層水進行分析溶解態無機碳及顆粒態有機碳的穩定碳同位素比值。該兩湖泊位於台灣東北部，分別為處於海拔高度五十公尺及四百七十公尺的梅花湖與雙連埤。兩湖泊表層水顆粒態有機物之碳氮比值與浮游植物所具有的比值相近(<10)，顯示所採得懸浮顆粒有機物組成來自藻類生成，其碳同位素比值介於-21.1‰和-28.9‰之間，相對較負比值則出現於冷季。此外，在兩年連續觀測之時間序列變化，兩湖泊之顆粒態有機碳同位素比值成相互平行，且隨時間而逐步變輕。而溶解態無機碳同位素比值之時間序列變化，兩湖泊也呈現如顆粒態有機碳同位素比值的變化趨勢。例外的情形為梅花湖2003年三月至八月期間，溶解態無機碳同位素比值顯現較富集1-3‰。此一溶解態無機碳同位素比值較重訊號，可能與2003年初該地區發生乾旱有關。

關鍵詞：溶解態無機碳、湖泊、氧同位素、顆粒態有機碳、穩定碳同位素、台灣

